POSITRON INDUCED IONIZATION PROCESSES

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ABSTRACT

In this work, a monoenergetic positron beam obtained from a radioactive source and guided by a magnetic field has been used to study ionization processes, such as positronium formation and annihilation, induced by positron collisions with atoms and molecules.

Precise total ionization cross-sections for Ne and Xe have been performed in the course of the present work in order to aid the extraction of the positronium formation cross-sections by subtracting direct ionization cross-sections. In the case of Xe, the energy dependence of the total ionization and positronium formation cross-sections has been scrutinized. No evidence of a structure around 80eV, predicted by a theoretical calculation, has been found. However, the positronium formation cross-sections in the noble gases have revealed a double-peak structure, which might be related to the positronium formation in its various quantum states. The double peak structure has also been confirmed by direct measurements for Ar and Xe described in this thesis. This type of investigation has also been extended to He. The finding of a simple scaling formula for positron- and electron-impact ionization cross-sections, also described in this thesis, has enabled the estimation of the contribution of the excited state positronium formation in the noble gases. These estimates suggest that excited state positronium might occur much more abundantly than previously thought.

Despite a comparatively poor energy resolution, low beam intensity and an uncertainty in the absolute energy calibration of the positron beam, the energy dependence of the annihilation cross-sections below the positronium formation threshold in Ne, Xe and O\textsubscript{2} has been investigated by measuring coincidence yields between $\gamma$-rays and ions. This study has been prompted by the observation of a residual ion yield in the total ionization cross-sections for Ne below the positronium formation threshold. Whilst in the case of Ne and O\textsubscript{2}, the annihilation cross-sections suggest some increase significantly below the positronium formation threshold, for Xe the increase is ambiguous.

It is anticipated that this work will stimulate further experimental and theoretical investigations.
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To My Tam
CHAPTER 1
INTRODUCTION

1.1. Historical background

The existence of the positron, the antiparticle of an electron, was predicted by Dirac (1930a) through the negative solution to his relativistic wave equation describing the motion of free electrons in the presence of an electromagnetic field. In his “hole theory”, Dirac assumed the vacuum to consist of an infinite sea of electrons occupying all negative energy states (from $-mc^2$ to $-\infty$) in accordance with the Pauli exclusion principle. If an electron from the “negative energy sea” were excited into a positive energy level, it would leave a hole behind, which would act exactly like a positively charged electron with a positive energy. Initially, Dirac believed that this positive particle was a proton, however it was soon shown by Weyl (1931) that the masses of particle and antiparticle had to be equal.

Experimental confirmation of the existence of the positron followed swiftly. It was first observed in cosmic-ray tracks in a cloud chamber by Anderson (1932a, 1932b, 1933). Later, Blackett and Occhialini (1933) confirmed that the mass-to-charge ratio for a positron was of the same magnitude as that of an electron, as predicted by Weyl (1931).

The discovery of the positron was followed by the prediction of Mohorovicic (1934) of a bound state of a positron and an electron, called positronium (Ps). The Bohr spectroscopic structure of Ps was qualitatively discussed by Ruark (1945), whilst its binding energy and lifetimes were calculated by Wheeler (1946). The first observation of positronium was made by Deutsch (1951) through the measurement of positron lifetimes in gases.

Subsequent advancements in the understanding of the interactions of positrons with matter have resulted in a plethora of applications of positrons in numerous branches of science. In solid-state physics, positrons may be used as a non-destructive bulk and surface probe of many materials, especially in the detection of structural defects (for a review see Schultz and Lynn, 1988; Coleman, 2002). In atomic and
molecular physics, comparative studies of positron, electron, proton and antiproton scattering from various gaseous targets can provide valuable information on the effect of mass, charge, electron exchange, polarisation and electron-capture processes (for a review see e.g. Knudsen and Reading, 1992). In astrophysics, positron-electron annihilation radiation is an important probe of energetic events in our Galaxy since it provides unique information about the environment of the annihilation region and the sources of positrons (Brown and Leventhal, 1987; Kinzer et al, 2001). In medicine, positron emission tomography is a well-established technique to image functioning organs, which can be used to detect amongst other conditions, coronary heart diseases, tumours and brain seizures (Ott, 2003).

Recently, cold antihydrogen atoms (bound states of positrons and antiprotons) have been produced and detected by two independent groups (Amoretti et al, 2002 and Gabrielse et al, 2002). From the CPT theorem, which requires that the laws of physics are invariant under the combined operations of charge conjugation, parity and time reversal, antihydrogen is expected to have spectroscopic properties identical to that of atomic hydrogen. The latest advances in cold antihydrogen production bring the stringent tests of CPT, such as comparison of frequencies for the electronic transition from the ground-state to the first excited state in antihydrogen and atomic hydrogen, a step closer.

The following sections present some of the fundamental properties of the positron and the Ps atom, an overview of their production and application as well as the motivation for this work.

1.2. Fundamental properties of positrons and positronium

1.2.1. Positron

The positron is stable in vacuum with lifetime of $2 \times 10^{22}$ years (Bellotti et al, 1983). Within the experimental limits, it has the same mass as the electron $(511.0034 \pm 0.0014)\text{keV/c}^2$ (Cohen and Taylor, 1973) and the same spin, but opposite charge and thus magnetic moment. A positron can annihilate with an electron resulting in an emission of a number of photons. The total energy of the photons, in the rest-frame, is equal to the total mass energy of the positron-electron pair, $1.022\text{MeV}$. The total number of photons is ruled by the charge parity, $P_c$, of the annihilating system and must be conserved throughout the interaction. Charge parity for a single photon is $-1$ and so $P_c$ for a system of $n$ photons can be written:
Chapter 1  Introduction

\[ P_c = (-1)^n \]  
\[ (1.1) \]

By symmetry arguments Yang (1950) has shown that the charge parity for a positron-electron system is given by:

\[ P_c = (-1)^{L+S} \]  
\[ (1.2) \]

where \( L \) and \( S \) are the total orbital angular momentum and total spin of the system, respectively. Hence, it can be seen that the number of photons into which a positron-electron pair will annihilate is dictated by the quantum state of the system prior to annihilation. In figure 1.1, the Feynman diagrams for annihilation into one to four photons are shown. In the case of the single quantum emission the presence of a nearby third body is required to conserve momentum. The cross-section of a given decay mode is directly proportional to \( \alpha^n \), where \( m \) is the number of photon interactions, which corresponds to the number of vertices in the Feynman diagrams, and \( \alpha = e^2 / 2\varepsilon_0 \hbar c = 1/137 \) is the fine structure constant. Here, \( e \) is the electron charge, \( \varepsilon_0 \) is the vacuum permittivity, \( h \) is the Planck constant and \( c \) is the speed of light. Therefore, the most probable decay mode, if allowed by the charge parity conservation, is mode (b) with only two vertices. All the modes presented in figure 1.1 have been observed ((a) Palathingal et al, 1991; (b) Klemperer, 1934; (c) Chang et al, 1982 and 1985; (d) Adachi et al, 1990).

The cross-sections for 2-\( \gamma \) annihilation in the non-relativistic limit of a positron and an electron in a singlet state was expressed by Dirac (1930b) as:

\[ Q_{2\gamma} = \frac{m r_0^2 c}{v} \]  
\[ (1.3) \]

where \( v \) is the velocity of the positron with respect to the electron and \( r_0 = e^2 / 4\pi\varepsilon_0 m_0 c^2 \) is the classical electron radius, where \( m_0 \) is the mass of the electron.

Figure 1.1. Feynman diagrams for: (a) one; (b) two; (c) three and (d) four photon decay modes.
When applied to annihilation with atomic or molecular targets, this equation is usually expressed in terms of the dimensionless parameter $Z_{\text{eff}}$ that represents the effective number of electrons per atom or molecule available for annihilation:

$$Q_{2\gamma} = \frac{\pi^2 \alpha^2 Z_{\text{eff}}}{v}.$$  \hspace{1cm} (1.4)

In the energy range typically employed for positron scattering measurements, Bransden (1969) determined this cross-section to be of the order of $10^{-26}$ m$^2$. Thus, direct annihilation may, in general, be assumed to be negligible as compared with other scattering processes such as ionization or Ps formation. We shall return to this process in chapter 4, where investigations of annihilation below the positronium formation threshold for Ne, Xe and O$_2$ are discussed.

### 1.2.2. Positronium

Positronium (Ps) is the bound state of a positron and an electron. The ground singlet-state (para-Ps or p-Ps) has a vacuum lifetime of approximately 125ps and decays predominantly into two $\sim$511keV $\gamma$-rays emitted back-to-back. The lifetime of the $1^3S_1$ state (ortho-Ps or o-Ps) is approximately 142ns and it decays predominantly into three coplanar $\gamma$-rays. The characteristics of the two spin states of ground-state Ps are given in table 1.1.

The continuous energy distribution arising from o-Ps decay was first predicted by Ore and Powell (1949). This is shown in figure 1.2, where it is compared with the more recent calculation of Adkins (1983) and the measurement of Chang et al (1985).

Recently, rare positronium decay modes into four- and five-photon final states have been measured using an efficient detector (Vetter and Freedman, 2002). No events violating charge-conjugation symmetry have been observed, thus setting new limits on

<table>
<thead>
<tr>
<th>Name</th>
<th>State</th>
<th>Sub-state</th>
<th>Ground-state decay rates ($\mu$s$^{-1}$)</th>
<th>No. and energy of annihilation quanta</th>
</tr>
</thead>
<tbody>
<tr>
<td>p-Ps</td>
<td>$1^1S_0$</td>
<td>m=0</td>
<td>7990.9 $\pm$ 1.7$^a$</td>
<td>even ($2 \times 511$ keV)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>m=0, ±1</td>
<td>7.0398 $\pm$ 0.0029$^c$</td>
<td>odd (3, 0-511keV)</td>
</tr>
<tr>
<td>o-Ps</td>
<td>$1^3S_1$</td>
<td>m=0</td>
<td>7989.5$^b$</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>m=0, ±1</td>
<td>7.0420$^d$</td>
<td></td>
</tr>
</tbody>
</table>

branching ratios for o-Ps decaying into four $\gamma$'s ($< 3.7 \times 10^{-6}$) and p-Ps decaying into five $\gamma$'s ($< 2.7 \times 10^{-7}$).

Ps is structurally equivalent to the hydrogen atom with half the reduced mass, binding energy (6.8eV) and, hence, twice the Bohr radius. The large magnetic moment of the positron (657 times larger than that of a proton) makes the magnetic spin-spin interaction in Ps comparable with the spin-orbit interactions thus removing the distinction between the fine and the hyperfine structure observed in the H atom. The energy levels for Ps (calculated by Fulton and Martin, 1954) and H are compared in figure 1.3.

**Figure 1.2.** The energy distribution of $\gamma$-rays emitted in o-Ps decay.

**Figure 1.3.** Comparison of the energy level diagrams for hydrogen and positronium.
1.3. Positron sources and interaction with solids

Fast positrons ($\beta^+$) may be obtained from the nuclear decay of radioactive isotopes (such as those listed along with their properties in table 1.2) or from pair production at particle accelerators. As illustrated in figure 1.4 for $^{58}$Co, positrons are produced with a large energy spread, which limits their use in controlled experimentation. However, as also shown in figure 1.4, moderation allows a compression of phase space. The slowing down process, which leads to this through the emission of positrons with a few eV of energy, is discussed briefly below.

When a positron from a radioactive source enters a solid, it rapidly loses its kinetic energy (~10ps) to inelastic processes until it reaches its thermal energy. Most of the positrons from radioactive sources typically reach equilibrium within 0.1 to 1mm in a defect-free solid. A significant number of positrons thermalize near the surface, with

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life</th>
<th>$\beta^+$ Fraction (%)</th>
<th>Max. activity (Ci$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{58}$Co</td>
<td>71 days</td>
<td>15</td>
<td>16.6</td>
</tr>
<tr>
<td>$^{22}$Na</td>
<td>2.6 yrs</td>
<td>90</td>
<td>1.25</td>
</tr>
<tr>
<td>$^{64}$Cu</td>
<td>12.8 hrs</td>
<td>19</td>
<td>600</td>
</tr>
<tr>
<td>$^{68}$Ge</td>
<td>288 days</td>
<td>88</td>
<td>6</td>
</tr>
<tr>
<td>$^{11}$C</td>
<td>20 min</td>
<td>99</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 1.2. Properties of some $\beta^+$ emitting isotopes (from Dupasquier and Zecca, 1985).

![Figure 1.4. The energy distribution of positrons emitted from a W(110) moderator in comparison to those emitted from a $^{58}$Co radioactive source.](image-url)
~1% of positrons stopping within 1500Å of the surface (Schultz and Lynn, 1988). This is comparable to the average distance that a thermalized positron will diffuse in a defect-free solid during one mean lifetime. After thermalization, the positron motion continues as a quantum diffusion process during which the positron scatters off host atoms and effectively behaves like a propagating wave in the solid. The typical time scales for positrons are summarized in Table 1.3.

<table>
<thead>
<tr>
<th>Process</th>
<th>Time (s)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermalization</td>
<td>(~10^{-12})</td>
<td>Perkins and Carbotte (1970)</td>
</tr>
<tr>
<td>Trapping (specific rate per atom)</td>
<td>(~10^{-15})</td>
<td>Hodges (1970)</td>
</tr>
<tr>
<td>Freely diffusing</td>
<td>(~1 \times 10^{-10})</td>
<td>West (1973)</td>
</tr>
<tr>
<td>Monovacancy trapped</td>
<td>(~2 \times 10^{-10})</td>
<td>West (1973)</td>
</tr>
<tr>
<td>Multivacancy trapped</td>
<td>(~4 \times 10^{-10})</td>
<td>Hautojärvi (1979)</td>
</tr>
<tr>
<td>Ps lifetimes</td>
<td>(\leq 10^{-9})</td>
<td>Dupasquier (1981)</td>
</tr>
</tbody>
</table>

Table 1.3. Time scales for positrons and Ps (from Schultz and Lynn, 1988).

The possible fates of positrons that have diffused to a metal surface are shown schematically in figure 1.5. An incident positron may return to the surface as a thermal or epithermal positron or it may be annihilated within the metal. Thermal positrons that are ejected from a metal surface may contribute to a slow positron beam. Although the epithermal positron emission from a tungsten surface at incident positron beam energy of 3keV has been found to be negligible (Goodyear et al, 1994), generally, slow positron beams may contain small amounts of unmoderated positrons. Their contribution to the measured spectra may be significant as discussed further in chapter 4 of this work.

In figure 1.6, a one-dimensional representation of the single-particle potential energy for a positron near a metal surface is shown in the case of a negative positron work function (\(\phi_+\)). Thus, thermal positron emission from a solid into a vacuum is
Figure 1.5. Simplified illustration of the interaction of positrons at a metal surface (from Mills, 1983).

Figure 1.6. The single-particle potential for a thermalized positron near a metal surface. \( V_{corr} \) is due to correlation with the conduction electrons and \( V_0 \) is due to the ion cores.
energetically allowed. The work function of a particular surface has two contributing factors, the relevant chemical potential ($\mu_c$) and the surface dipole ($\Delta \varphi$). The positron work function can be written, following Tong (1972), as $\phi_+ = -\Delta \varphi - \mu_c$. The chemical potential contains terms due to the electron and positron interactions with the other electrons and with the ion cores. The surface dipole is primarily caused by tailing of the electron distribution into the vacuum for a distance $\sim 10^{-16} \text{m}$. The fact that the surface dipole is attractive for positrons enables $\phi_+$ to be negative for certain materials.

Clean single crystal moderators with known lattice orientations were first studied by Mills et al (1978). It was found that the variation of the temperature of the moderator resulted in the change of the slow positron beam yield. Muray and Mills (1980) measured the positron moderation efficiencies of Al and Cu for various values of $\phi_+$ by controlling the temperature, surface coverage of S atoms and the orientation of the crystal lattice. Their results are shown in figure 1.7, where it can be seen that as the work function becomes more negative, the positron yield increases.

Dale et al (1980) found that annealing (described in more detail in chapter 2) of the moderator increased the moderation efficiency. The best result was obtained with chemically etched and heated to 2200°C polycrystalline W vanes. This was attributed to the fact that the heat treatment reduced the number of defects in the lattice, which act as traps, allowing more positrons to diffuse back to the surface.

![Figure 1.7](image.png)

**Figure 1.7.** The variation of the slow positron yield from Al and Cu, with changing positron work function, $\phi_+$ (Murray and Mills, 1980).
Gullikson and Mills (1986) reported that rare gas solids (RGS) reveal unusually high yields of epithermal positrons. This was attributed to the large diffusion length of the hot positrons in RGS and, thus, the inability to thermalize completely. In contrast to metals, there are no free electrons near the surface so the dipole potential contribution is smaller and $\phi_e$ is positive. However, many of the positrons reach the surface epithermally and thus can overcome the positive $\phi_e$ barrier and be emitted into the vacuum.

The application of RGS-moderated positrons in conjunction with an appropriately modified Penning-Malmberg trap has resulted in high-quality, cold positron beams of 25meV energy resolution (Greaves et al, 1994; Greaves and Surko, 2002). In figure 1.8, a schematic illustration of a buffer accumulator and the variation of the electrical potential and pressure along its three stages are shown. Neon-moderated positrons are confined along the trap by an axial magnetic field of typically 1000-1500 Gauss. They enter the trap passing over the electrostatic potential into region I, which contains N$_2$ at a pressure of around 10$^{-3}$Torr. In this region, positrons are trapped by inelastic collisions, ‘A’, with buffer gas molecules. Subsequently, positrons filter to lower energies by collisions ‘B’ and ‘C’ and as a result are confined in region III, which is maintained at low pressure by differential pumping. In region III positrons cool rapidly to 300K by a combination of vibrational and rotational excitations. It was found that although N$_2$ is the most efficient for positron trapping, it is an inadequate choice for positron cooling (Greaves and Surko, 2002). This problem was overcome by introducing a small amount of cooling gas, such as CH$_4$, directly into the third stage of the trap.

**Figure 1.8.** Schematic illustration of the buffer accumulator of Greaves and Surko (2002), showing the three stages of differential pumping and the electrostatic potential.
1.4. Positron interactions with atoms and molecules

In the last decade, there has been much progress in positron-atom/molecule collisions. The present experimental status of positron collisions with atomic and molecular targets is summarized in table 1.4. Recently, slow positron beam interactions with atoms and molecules have been reviewed by Laricchia and Charlton (1999), Charlton and Humberston (2001) and Laricchia (2002).

<table>
<thead>
<tr>
<th>Interaction</th>
<th>Experimental status</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.4.1. Total cross-section</td>
<td>$Q_i$ for many atomic and molecular targets including H, He, alkali and Mg. Some $Q_{el}$ and $dQ_{el}/d\Omega$.</td>
</tr>
<tr>
<td>$e^+ + A \rightarrow all$</td>
<td></td>
</tr>
<tr>
<td>1.4.2. Elastic scattering</td>
<td>$Q_{Ps}$ for $1 &lt; E &lt; 100 eV$ including H, alkali, Mg; some $dQ_{Ps}/d\Omega$.</td>
</tr>
<tr>
<td>$e^+ + A \rightarrow e^+ + A$</td>
<td>First state-resolved $Q_{el}$ for electronic and vibrational excitations. $Q_i^{+\ast}$ for $1 &lt; E &lt; 1000 eV$</td>
</tr>
<tr>
<td>1.4.3. Positronium formation</td>
<td>including H, some $dQ_i^{+\ast}/d\Omega$, $d^2Q_i^{+\ast}/d\Omega_i dE_i$ and first $d^3Q_i^{+\ast}/d\Omega_i d\Omega_j dE_i$.</td>
</tr>
<tr>
<td>$e^+ + A \rightarrow Ps + (m-1)e^- + A^{m\ast}$</td>
<td>First energy dependent measurements.</td>
</tr>
<tr>
<td>1.4.4. Target excitation</td>
<td>Only PsH observed – failed search for PsCl and PsF.</td>
</tr>
<tr>
<td>$e^+ + A \rightarrow e^+ + A^\ast$</td>
<td></td>
</tr>
<tr>
<td>1.4.5. Direct ionization</td>
<td></td>
</tr>
<tr>
<td>$e^+ + A \rightarrow e^+ + me^- + A^{m\ast}$</td>
<td></td>
</tr>
<tr>
<td>1.4.6. Annihilation</td>
<td></td>
</tr>
<tr>
<td>$e^+ + A \rightarrow 2\gamma + A^\ast$</td>
<td></td>
</tr>
<tr>
<td>1.4.7. Formation of compounds</td>
<td></td>
</tr>
<tr>
<td>$e^+ + A \rightarrow (PsC) + D^\ast$</td>
<td></td>
</tr>
</tbody>
</table>

*Table 1.4. Status on experimental collision studies with positrons (Laricchia, 2002).*

1.4.1. Total cross-sections

The total scattering cross sections ($Q_i$) have been extensively studied (for reviews see Kauppila and Stein, 1990; Zecca *et al.*, 1996). Among the first atoms treated experimentally were the noble gases since they exist in atomic form at room temperature. In figure 1.9 and 1.10, interesting features of $Q_i$ by positron- and
electron-impact for the inert atoms are evident. In the case of He, it can be seen that \( Q_{e^-}(e^-) \) is significantly larger than \( Q_{e^+}(e^+) \) at low energies. This is due to partial cancellation of the static (repulsive) and polarization (attractive) interactions in the case of a positron projectile. A similar behaviour is found in the total cross-section for other noble gases, except that \( Q_{e^-}(e^-) \) for Ar, Kr and Xe exhibit narrow deep minima at low energies (<1eV) where only elastic scattering occurs. In this region \( Q_{e^+}(e^+) \) exceeds \( Q_{e^-}(e^-) \). These minima were first observed by Ramsauer (1921, 1923), Townsend and Bailey (1922) and Ramsauer and Kollath (1929) and referred to as Ramsauer-Townsend effects. They arise from quantum mechanical effects associated with an attractive polarization interaction between the incident projectile and the target atom. In the case of positron scattering, as seen in figures 1.9 and 1.10 that the Ramsauer-Townsend

![Figure 1.9. Total scattering cross-section by positron- and electron-impact on the noble gases (from Kauppila and Stein, 1982). The arrows indicate thresholds for positronium formation, excitation and ionization in order of increasing energy.](image-url)
minima exist for He, Ne and possibly Ar between 1-3eV, where they are due to the zero in the s-wave phase shift at such low energies. In the case of electrons, Ramsauer-Townsend effects occur because the s-wave shift passes through $\pi$ radians (or its multiple).

In general, $Q_t(e^-)$ change very smoothly as different inelastic channels open, whereas $Q_t(e^+)$ reveal a sudden increase at the positronium formation threshold, $E_{Ps}$, (indicated by first arrows in figure 1.9 and a blue arrow in figure 1.10) suggesting that this process plays a significant role in the positron scattering by inert atoms. Indeed, the minimum at low energies and the sharp rise at $E_{Ps}$ are also typical features observed in positron scattering from many molecular targets. As the impact energy is increased, the dominance of the static interaction ultimately should lead to merging of $Q_t$ for the two projectiles, as illustrated in figure 1.10 for positron-He scattering where merging occurs at around 200eV. No such merging has been observed for heavier targets up to 800eV (Kauppila et al, 1981; Dababneh et al, 1982). On the other hand however, Kimura et al (2000) reported on the merging of $Q_t$ by positron and electron impact at energies as low as 50eV in molecular targets such as C$_2$H$_2$, C$_2$H$_4$, SiH$_4$ and CCl$_4$. Recent investigations of total cross-sections in Ar and Kr at energies between 250eV and 5keV.
of Karwasz et al (2002) indicated an asymptotic convergence of cross-sections by positron and electron projectiles in the case of Ar above 2keV. The results for Kr across this energy range were inconclusive.

Recently, the total cross-section for H has been measured by Zhou et al (1997) and a good agreement found between the experiment and available theories, except at low energies where experimental data lie below the theoretical results, as depicted in figure 1.11.

![Figure 1.11. Comparison of the experimental and theoretical total cross-sections for $e^-+H$ scattering (from Zhou et al, 1997).](#)

**1.4.2. Elastic scattering**

In the case where annihilation is relatively improbable as compared to other processes, $Q_o$ is approximately equal to $Q_{el}$ below the first inelastic threshold. Campeanu et al (1987) deduced the behaviour of $Q_{el}$ for He by adding together the positronium formation and the single ionization cross-sections of Fromme et al (1986), fitting a curve to these points and then subtracting it from the total cross-section of Stein et al (1978). This resulted in an indication that a cusp-like feature may be present in $Q_{el}$ at the positronium formation threshold. However, neither the results of Coleman et al (1992) nor Moxom et al, (1993) confirmed such a feature of the size implied by Campeanu et al (1987). Indeed, measurements of $Q_{el} + Q_{es}$ for He display very smooth
variation of $Q_{el}$ across $E_{Ps}$ for this system (Moxom et al, 1993). This is in agreement with near-threshold measurements of Moxom et al (1994) and the Kohn variational calculation of Van Reeth and Humberston (1997).

A more sensitive method of investigating elastic cross-sections is through differential cross-sections. Measurements of such cross-sections for positron scattering by Ne, Ar, Kr and Xe revealed diffraction-like structure below the relevant $E_{Ps}$, that disappears as the incident positron energy is increased (Smith et al, 1990; Dou et al, 1992; Kauppila et al, 1996). This was attributed to the absorption effect of the positronium formation channel on the elastic one. In figure 1.12, the relative differential cross-sections for positron-methane quasielastic scattering (the rotational and vibrational excitations could not be resolved due to large energy resolution of the projectile beam) of Przybyla et al (1997) are compared to theory of Jain and Gianturco (1991) and Germano and Lima (1993). These differential cross-sections are remarkably similar to those of Ar (Smith et al, 1990). The fact that the structure at 6eV and 10eV is consistent with the calculation of Jain and Gianturco (1991) and is seen to be washed out at 30eV and inconsistent with the calculation at small angles above 50eV suggests that positronium formation (not included in the calculation) for this target has an important absorption effect on the inelastic channel. Similar structures have been observed for other symmetric molecules such as N$_2$, O$_2$, CO and SF$_4$.

Figure 1.12. Relative differential cross-section for $e^-$-CH$_4$ elastic scattering (from Kaupilla et al, 2002).
1.4.3. Positronium formation

Positronium formation cross-sections ($Q_{\text{Ps}}$), integrated over all quantum states of Ps and the ion, have been measured for all the inert atoms (Moxom et al., 1993, 1994; Overton et al., 1993; for compilation of earlier data see Charlton and Laricchia, 1990), some molecules (Moxom et al., 1993; Laricchia and Moxom, 1993; Laricchia et al., 1993), atomic hydrogen (Zhou et al., 1997 and referenced therein), alkali atoms (Zhou et al., 1994; Surdutovich et al., 1996, 2002) and Mg (Stein et al., 1996). $Q_{\text{Ps}}$ for H, H$_2$ and the inert atoms are discussed in detail in chapter 3. Briefly, good convergence exists among recent theories and experimental $Q_{\text{Ps}}$ in the case of atomic hydrogen (Zhou et al., 1997). For He, good agreement between the experiment (Overton et al., 1993) and the theory (Campbell et al., 1998) exists at the intermediate energy range, however discrepancies still remain both near threshold (Van Reeth and Humberston, 1999) and at high energies (Campbell et al., 1998). In the case of He, the coupled-pseudostate calculation of Campbell et al. (1998) suggest that the ground-state Ps formation dominates the cross-section with only ~20% of Ps formed in an excited state. In the case of the alkali atoms, Ps formation is an exothermic process so it is expected that $Q_{\text{Ps}} \rightarrow 0$ as $E \rightarrow 0$. Calculations of Walters et al. (1995) predicted a collapse of ground-state Ps formation in going from Li to Cs with a simultaneous increase in the excited state Ps formation. Interestingly, a trend has been revealed in the estimates of the excited state positronium formation, which increases from Ne to Xe, as discussed further in chapter 3. In figure 1.13, $Q_{\text{Ps}}$ for potassium and sodium are presented together with various theories (Zhou et al., 1994) in the energy ranges from 1eV to 100eV for K and 1eV to 20eV in the case of Na. These energy ranges were extended down to around 0.2eV and measurements were performed for lithium and sodium (Surdutovich et al., 2002). From figure 1.13, it can be noticed that there is a larger difference between the upper and lower limits in the case of K than in the case of Na. For these targets the two sets reveal opposite trends below approximately 5eV with the lower limit measurements exhibiting a striking decrease and the upper limit values continuing to rise. The theory suggests that the lower limit results are closer to the true $Q_{\text{Ps}}$ and the divergence between the upper and lower limits has been attributed to the incomplete collection of the scattered positrons.

The first experimental investigations of $dQ_{\text{Ps}}/d\Omega$ were stimulated by theories which indicated that a high degree of forward scattering of the outgoing positronium
Figure 1.13. Positronium formation cross-sections for K and Na (from Zhou et al., 1994).

is present and hence by a prospect of being able to produce positronium beams. In figure 1.14, the results of Laricchia et al (1987) for the variation of the fraction ($F_{Ps}$) of positrons scattered by He and emitted as positronium at small forward angles is compared to the corresponding predictions obtained from the theory of Mandal et al (1979). It was concluded by Laricchia et al (1987) that approximately 4% of the scattered positrons could be detected as o-Ps atoms collimated in a 6° cone about the incident positron direction.

Figure 1.14. Fraction $F_{Ps}(\theta)$ of positrons scattered by He and emitted as positronium within 5, 10 and 20° angular ranges about the incident positron direction. The experimental points – Laricchia et al (1987); theory – Mandal et al (1979).
Very recent investigations of the efficiency for conversion of a positron beam into a positronium beam by charge exchange from N\textsubscript{2} have indicated that although H\textsubscript{2} remains the best converter at low energies (Garner et al, 1996), N\textsubscript{2} becomes more efficient from 90eV upwards (Leslie et al, 2002). In section 1.5 of this chapter, recent advances in positronium scattering are briefly described.

1.4.4. Target excitation

Recently, the development of buffer-gas trapping and magnetized beam formation has provided positron beams with much smaller energy resolutions ($\Delta E \leq 25$ meV), which in turn, has enabled scattering experiments at low energies with significantly improved selectivity for excitation experiments (Sullivan et al, 2002a).

In figure 1.15, the apparatus used for the scattering measurements is depicted. Positrons are released from the accumulator, described in section 1.3, with a well-defined energy and energy spread in pulses of about $2 \times 10^4$ positrons each. The pulsed positron beam is then passed through a scattering gas cell containing the gas under study and subsequently through a retarding potential analyzer (RPA). The detection of the positron beam is achieved by measuring the 511keV $\gamma$-rays from the annihilation of positrons on the metal collector plate at the end of the RPA. Scattering cross-sections were obtained by exploiting the properties of the positron motion in high magnetic fields. The total energy ($E_i$) of a positron can be separated into two components parallel ($E_{par}$) and perpendicular ($E_{per}$) to the vector of the magnetic field, since $E_i = E_{par} + E_{per}$. Elastic and inelastic scattering results in a redistribution or

![Figure 1.15. Schematic diagram of the apparatus and the associated potential profile used for measurements of elastic scattering, vibrational and electronic excitations and positronium formation (Sullivan et al, 2002a).](image-url)
transfer of energy between $E_{\text{par}} = E_s \cos^2(\theta)$ and $E_{\text{per}} = E_s \sin^2(\theta)$, $\theta$ being the positron scattering angle and the positron energy after scattering $E_s = E_{in}$ or $E_s = E_{in} - E_{es}$ ($E_{in}$ is the incident positron energy and $E_{es}$ is the energy lost due to target excitation). In a slowly varying magnetic field the quantity $E_{\text{par}}/B$ is an invariant. If the magnetic field ($B_j$), where the transmitted positrons are analyzed, is much smaller than that where they are scattered ($B_s$), then the beam is parallelised (according to $B_j / B_s \propto \sin^2 \theta_j / \sin^2 \theta_s$) and most of $E_{\text{per}}$ is converted into $E_{\text{par}}$. Thus the scattered positron beam has a distribution of $E_{\text{par}}$ values and this is reflected in the signal measured by the RPA.

Absolute cross-sections for the vibrational excitations of CO, CO$_2$ and H$_2$ were measured in the energy range from 0.5eV to several eV by Sullivan et al (2001b). In figure 1.16, the absolute vibrational cross-section for the excitation of the $v=1$ mode of H$_2$ is shown and compared to available theories. The agreement with calculations of Gianturco and Mukherjee (2001) and Sur et al (1985) seems reasonable, whilst the prediction of Bailie and Darewych (1974) is lower at energies <2eV.

In the recent work of Sullivan et al (2001a) the first state-resolved measurements of electronic excitations by positron impact on Ar, H$_2$ and N$_2$ were made. In figure 1.17,

![Graph showing cross-section values vs energy](image)

**Figure 1.16.** Absolute integral cross-sections for the excitation of the $v=1$ vibrational mode of H$_2$. Experimental data of Sullivan et al (2002a) compared to various theories: long-dashed curve – Sur et al (1985); short-dashed curve – Bailie and Darewych (1974); solid line – Gianturco and Mukherjee (2001). From Sullivan et al (2002a).
Figure 1.17. Absolute integral cross-sections for the excitation of the 3p^3(^3P_{3/2,1/2})4s(J=1) states of Ar from threshold to 30eV compared to theories. Solid symbols refer to cross-sections by positron impact, whereas open symbols to the electron impact (from Sullivan et al, 2002a).

the absolute integral cross-sections for the excitation of the 3p^3(^3P_{3/2,1/2})4s(J=1) states in Ar are shown. These are two out of the four lowest-lying 4s excited states that can be excited by positrons (those with total angular momenta J=1,2 are metastable). A structure is evident near 15eV in the state coupled to the ^2P_{1/2} level. Although the cross-sections are small, they are comparable in magnitude to their electron scattering counterparts.

1.4.5. Direct ionization

Single direct ionization cross-sections by positron impact are discussed in more detail in chapter 3. For reviews see e.g. Charlton and Humberston (2001), Laricchia (2002) and Laricchia et al (2003).

Kővér and Laricchia (1998) reported the first triply differential cross-section for positrons at an incident energy of 100eV in collisions with H_2. The scattered positron and the ejected electron were both detected around 0°, with the electron also being energy-analyzed. The ejected-electron spectrum is shown in figure 1.18a. A small peak close to 42eV present in the experimental data was attributed to electron-capture-to-the-continuum (ECC), a phenomenon that arises from the Coulomb attraction between the two outgoing particles in the final state. This work was further extended to the positron incident energy of 50eV by Kővér et al (2001), as shown in figure 1.18b. The
experimental results are compared to the theory of Fiol et al. (2001) and as can be seen in the figure, the agreement between the data would be fair were the theoretical data shifted by 1.6eV towards lower energies. It was suggested by Kövér et al. (2002) that a physical effect, which could give rise to the shift, would be the occurrence of ionization simultaneous to another energy loss such as vibrational excitation or molecular dissociation of the target.

1.4.6. Annihilation

Positron annihilation on atomic and molecular targets is very topical at present. It is described in greater detail in chapter 4. Here it suffices to mention that, until very recently, the majority of annihilation measurements were performed using lifetime techniques over a restricted temperature range (for reviews see Heyland et al., 1982; Charlton, 1990).

Energy-resolved measurements (discussed further in section 4.2) have now been performed using an apparatus shown in figure 1.19 (Gilbert et al., 2002). The pulsed positron beam was derived from the trap, already discussed in section 1.3, and guided by a magnetic field through a cylindrical gas cell containing the gas under investigation. The positron beam energy was defined by adjustable voltages $V_c$ on the gas cell and $V$ on the exit of the trap. Annihilation events were detected from a 10cm region along the gas cell by a specially designed CsI detector. The low level of the annihilation signal required practically a perfect shielding from stray $\gamma$-rays. This was achieved by
shielding the detector and the gas cell with a thick layer of lead. As shown in figure 1.19, signals from γ-rays that would arise from positrons hitting the collector plate at the end of the beam-line were prevented by keeping positrons in flight by means of a reflector electrode while the annihilation signal was recorded.

1.4.7. Formation of compounds

There has been much theoretical and experimental interest in studying the possibilities of forming Ps-containing molecules with \( Ps^- \) being observed by Mills (1981). PsH, PsF, PsCl, PsBr and Psl have all been predicted to be chemically stable (Wheeler, 1946; Simons, 1953; Ore, 1951; Cade and Farazdel, 1977; for a recent review see Mitroy et al, 2002). The formation of positronium hydride (PsH) in collisions between positrons and methane was reported by Schrader et al (1992). The signature for the formation of PsH was a detection of \( CH_3^+ \) ion below the threshold (7.55 eV) for the production of \( CH_3^+ + H + Ps \). The small step in the \( CH_3^+ \) yield between 6 eV and 8 eV, depicted in figure 1.20, was interpreted as direct evidence of the PsH formation. From these measurements a value of \( (1.1 \pm 0.1) \text{ eV} \) for the binding energy of PsH was obtained, which is in broad agreement with theoretical calculations (e.g. Ho, 1986; Yan and Ho, 1999).

More recently, an investigation to study fragmentation patterns of the halomethanes such as CCl\(_2\)F\(_2\), CCl\(_3\), CF\(_4\) and CHF\(_3\) from threshold up to 50 eV was undertaken by Moxom et al (2000). The yields of the fragment ions were measured as a
function of positron energy. If Ps compounds such as PsF and PsCl were formed during the collisions, thresholds for relevant fragmentation processes would be reduced by their binding energies. Within the experimental limitations, no evidence of compound formation has been observed.

1.5. Positronium scattering

A positronium beam can be produced by neutralizing a monoenergetic positron beam in a gaseous target (Laricchia et al, 1987; Laricchia and Zafar, 1992). The beam produced in such a way includes only o-Ps, since the lifetime of p-Ps is three orders of magnitude shorter and is untransportable at atomic velocities. The Ps energy is expected to be given, to the first approximation, by \( E = E_+ - \left( E_i + 6.8eV/n^2 \right) \), and is tunable via the incident positron energy \( E_+ \), \( E_i \) is the target ionization energy and \( 6.8eV/n^2 \) is the Ps binding energy in a state of principal quantum number \( n \). The total cross-sections for Ps scattering have been measured for He, Ar, O$_2$ and H$_2$ (Garner et al, 1996; Garner et al, 1998; Garner et al, 2000). These cross-sections are difficult to measure due to very small fluxes of Ps beams. In figure 1.21, the total scattering cross-section for He is shown (Garner et al, 1996). The characteristic feature, namely a rapid increase of the cross-section, a broad peak and a slow decrease at higher energies, present in the total
Figure 1.21. Positronium scattering cross-sections for He.

Figure 1.22. Longitudinal energy spreads of the positrons released from positronium break-up (from Armitage et al, 2002).
cross-section of He is also exhibited by Ar and H₂. Recently, the absolute cross-section for the fragmentation (or break-up) of Ps in collisions with He was measured by Armitage et al (2002) and is also shown in figure 1.21. It can be seen in the figure that a good agreement is found with the theory of Blackwood et al (1999). However, in the case of the total cross-section, a significant discrepancy exists between the same theory and the data of Garner et al (1996). This might be suggestive of the elastic scattering cross-section being underestimated in the calculation and/or a presence of target inelastic effects in Ps-He scattering.

In figure 1.22, the longitudinal energy distributions of the positrons resulting from the break-up of Ps are shown. Peaks in these distributions are situated just below half the residual energy \( E_r = E - 6.8 \text{ eV} \), \( E \) is the incident Ps energy) as indicated by red arrows in the graphs. This suggests that both a positron and an electron travel in the forward direction with the same velocity, which might indicate the occurrence of electron loss to the continuum.

1.6. Aim and motivation of the present work

In this chapter, a general background to positron physics and positron interactions in various media has been given. Outstanding problems in the study of positron scattering off atomic and molecular targets include unsatisfactory agreement between available experimental results and the theoretical calculations for the positronium formation in the noble gases. This has been a motivation for performing, in the course of this work, additional total ionization cross-section measurements for Ne and Xe in the energy range from a few to around 120eV. These results supplement the data of Laricchia et al (2002) and, from the two sets of measurements, positronium formation cross-sections have been extracted by subtraction of the direct ionization cross-sections. However, the present set of data was primarily used to investigate the existence or otherwise of a structure predicted by McAlinden and Walters (1992) in the vicinity of 80eV. No evidence of such a structure has been observed experimentally neither in the total nor positronium formation cross-sections.

A conspicuous feature in the form of a double peak structure has been found in the positronium formation cross-section in the noble gases by Laricchia et al (2002), the second peak being attributed to the positronium formation in an excited state. In this work, an attempt has been made to estimate the contribution from the excited state positronium to the total positronium formation cross-sections following the discovery of
a simple scaling formula for positron- and electron-impact ionization cross-sections (Szliwińska et al, 2002).

Later in the present work, the energy dependence of the positronium formation cross-sections for Ar, Xe and He was measured directly by detecting coincidence signals between γ-rays from Ps annihilations and remnant ions. The present results reproduce the double-peak structure found by Laricchia et al (2002) very well. An analysis has also been carried out in order to obtain the dependence of the Ps detection efficiency on the energy for the present geometry of the gas cell. These investigations are described in chapter 3.

Another process that can lead to ionization is annihilation. Until now, annihilation near the positronium formation threshold has remained unexplored experimentally but has initiated a heated theoretical debate. The annihilation measurements in the present work have been prompted by the fact that positive residual ion yields have been found in the total ionization cross-sections in the case of Ne below the positronium formation threshold. Measurements have also been extended to Xe and O₂ and are described in chapter 4.
CHAPTER 2
THE EXPERIMENTAL APPARATUS

2.1. Introduction

The positron beam used throughout the present work was produced by moderating fast $\beta^+$ particles from a commercially obtainable radioactive source. The fast $\beta^+$ particles were moderated in energy by annealed tungsten meshes and formed into a beam by means of simple electrostatic optics and axial magnetic field. The positron beam traversed the interaction region, which consisted of a differentially pumped gas cell, before being detected at the end of the beam-line.

In this chapter, components and features of the apparatus common to all measurements performed during the course of this study are described. Any modifications added throughout the work will be subsequently mentioned in the relevant chapters.

2.2. The positron beam production

The decay rates and branching ratios for $\beta^+$ emission and electron capture for $^{22}\text{Na}$ are shown in figure 2.1. This radioactive source, used throughout the work, was supplied by DuPont Pharmaceutical and came within a capsule in the form of a 4mm spot deposited on a 4mm thick platinum disc with a diameter of 18mm. The source was sealed with a 10$\mu$m titanium window. It was estimated by Massoumi et al (1988) that around 40% of the $\beta^+$ particles liberated from the source are allowed to escape in the
Forward direction. $^{22}$Na has a half-life of 2.6 years approximately, which gives a range of source activities from 9 to 5.2mCi during the experimentation period.

The source and the moderator arrangement are placed in the middle of a specially designed vacuum chamber, shown in figure 2.2, which consists of two concentric stainless steel cylinders with an axis perpendicular to that of the beam-line. The source is mounted on the end of the retractable shaft enabling the source to be

![Figure 2.1. The decay schemes and branching ratio of $^{22}$Na.](image)

**Figure 2.1.** The decay schemes and branching ratio of $^{22}$Na.

![Figure 2.2. A cutaway schematic of the housing chamber for the source and the moderator assembly.](image)

**Figure 2.2.** A cutaway schematic of the housing chamber for the source and the moderator assembly.
withdrawn from the centre of the chamber and the inner cylinder to be rotated. In this way the lead plug, on which the moderator assembly is mounted, can be safely removed from the system. The high level of radiation from the source is greatly reduced by partial filling of the inner cylinder with lead. Further reduction of the background radiation is achieved by shielding the outside of the chamber with lead bricks.

The source holder and the moderator assembly are shown in figure 2.3. Typically, the moderator consists of three, superimposed 90% transmission, annealed tungsten (W) meshes, with a diameter of approximately 14mm. The annealing process, which is described fully by Zafar et al (1988 and 1989), is carried out within a vacuum chamber in an “oven”, shown in figure 2.4. This is made from two pieces of 99.95% pure tungsten sheet, typically of dimensions 2cm by 5cm. Two W-alloy clamps hold the strips of tungsten, outwardly bowed in the centre, such that the prepared meshes can be placed in the middle and heated by the current passing through the walls of the oven. The temperature of the meshes can be monitored through a pyrex window by means of a pyrometer. The annealing process consists of repeatedly raising the temperature of the oven to around 2000°C for a few seconds in a pressure of less than 10^{-4} Torr. At the beginning of the annealing process, the increase in temperature is accompanied by an increase in pressure in the vacuum chamber. This is caused by the impurities being released from the moderator and the oven itself. The annealing is carried out for up to several hours, until no increase of pressure in the chamber is seen. Later, the meshes are allowed to cool down to room temperature under vacuum and are then transferred in air to the e^-beam system. Despite the fact that the W meshes are pretty robust, any additional handling increases the likelihood of stress and thus damage to the crystal structure.

The moderator holder consists of a PTFE sleeve of diameter 20mm and length 9mm, into which annular PTFE washers and brass rings are inserted to provide support and insulation for the grids, as shown in figure 2.3. The electrical contacts to the grids are made through brass rings. The source is held at the same potential as the moderator. The moderator meshes are placed as close as possible to the β^+ source, in order to increase the solid angle and thus to improve the positron beam intensity. The slow positrons are emitted perpendicularly to the surface of the moderator if no inelastic processes at the surface take place. The potential applied to the moderator meshes (V_m) determines the energy, E, of the slow positrons emerging from it, according to:
Figure 2.3. A schematic diagram of the $^{22}$Na source and the moderator holder (to scale).

Figure 2.4. The moderator annealing oven.
\[ E = eV_m + \phi_+ \]  

(2.1)

where \( \phi_+ \) is of the order of a few eV and arises from the positron work function of the moderator and any possible contact potentials. For a clean W surface, \( \phi_+ \) equals 2.8 eV (Jacobsen et al, 1990).

2.3. The vacuum system and the positron beam transport

The vacuum system and the beam transport are shown schematically in figure 2.5. The system is maintained in vacuum of the order of \( 10^5 \text{Torr} \) by means of four diffusion pumps. The source and moderator chamber are evacuated by an Edwards E02 diffusion pump (pump 1 in the figure) backed by a Varian SD-300 rotary vane pump. The regions prior to and after the scattering gas cell, as well as the ion detector chamber, are evacuated by three Edwards E04 diffusion pumps backed by two rotary vane pumps. Diffusion pump 2, as shown in figure 2.5, is backed by a Leybold Trivac D4B rotary vane pump, and pumps 3 and 4 by a Leybold Trivac D16B.

As well as backing the diffusion pumps, the rotary pumps are used to evacuate the system from atmospheric pressures to around \( 10^{-2} \text{Torr} \), the level required prior to switching on the diffusion pumps. The pressure in the backing lines is monitored by means of pirani gauges. Any failure in the vacuum system would cause an increase in the backing pump pressure, which if higher than \( 10^{-4} \text{Torr} \), would trigger a system protection device. This would shut the power to all the diffusion pumps and high voltage supplies, as well as closing the magnetic valves. Similarly, a failure in the flow of cooling water for the diffusion pumps would also switch on the trip mechanism.

The positron beam is guided along the experimental axis by a longitudinal magnetic field of strength indicated in figure 2.5. The magnetic field is produced by current-carrying coils of 20cm internal diameters positioned along the beam-line, as shown in figure 2.5. Two 30cm diameter coils are placed around the source and moderator chamber, from where positrons are guided to the interaction region via a 1m long solenoid with a 15° bend, a positron- (R1) and an electron-repeller (R2) and an \( \mathbf{E} \times \mathbf{B} \) filter. The bend in the solenoid and the \( \mathbf{E} \times \mathbf{B} \) filter serve to reduce the number of fast particles, such as energetic positrons and electrons, from the beam as well as to prevent a line-of-sight between the source and the interaction region. Additional lead shielding, obtained by inserting lead collimators at one end of the solenoid and in the source chamber as well as lead bricks placed around the bend of the solenoid, allowed a
Figure 2.5. Schematic of the vacuum system and the beam transport (not to scale).
further reduction in the γ-rays emitted by the source from reaching the interaction region.

The $E \times B$ filter, sometimes referred to as a Wien filter or a trochooidal monochromator, is shown in figure 2.6a, in its commonly used geometry with two parallel planar electrodes of length ($L$). The electric field ($E$) is applied in the direction perpendicular to that of an axial magnetic field ($B$) which causes the gyration centre of the positrons to drift with a constant velocity given by:

$$v = \frac{E \times B}{B^2}.$$  \hspace{1cm} (2.2)

The vertical beam displacement ($D$) can be written as:

$$D = \frac{|E \times B| \cdot L}{B^2 \cdot v_z}.$$ \hspace{1cm} (2.3)

where $v_z$ is the constant axial speed of the positrons. The displacement is thus inversely proportional to the positrons speed and the slow particles experience a deflection orders of magnitude greater than the fast particles emitted by the source. In this way, the slow positron beam can be transmitted through an aperture whilst the fast particles may be blocked. Following Hutchins et al (1986) a cylindrical plate geometry, depicted in figure 2.6b, has been employed to eliminate the possible distortion of the positron beam profile. The Wien filter used in the present work comprises two parallel, 120mm long, deflection plates (parts of cylindrical surfaces) separated by a distance of 10mm and having 8mm apertures at each end. The approximate radii of the cylinders are 20cm. The deflection plates are covered with graphite in order to reduce possible secondary

\textbf{Figure 2.6.} a) Planar geometry $E \times B$ filter; b) Distortion of the positron beam profile passing through a planar (left) and cylindrical geometry (right) filter (from Hutchins et al, 1986).
electron emission from that region. The displacement (D), the distance between the axes of the collimating apertures before and after the Wien filter, is 10mm. The potential applied to the deflecting electrodes was coupled with that of the moderator via a "square rooter", which supplied positive and negative input to the plates corresponding to the square root of the moderator voltage. In this way, voltages applied to the deflecting plates would be appropriately modified when the moderator voltage was ramped.

Two brass repeller tubes (R1 and R2) are inserted inside the solenoid, as shown in figure 2.5. R1 is used to bias off positrons while R2 serves to remove secondary electrons from the positron beam by applying a potential of -400V. This is different in comparison to the system used by Moxom et al (1995), where R1 served to remove background electrons and R2 to bias off a low energy portion of the beam. Secondary electrons come from metal surfaces such as the moderator holder and associated grids, the source and even from R1 itself. A positive potential is applied to R1 in order to bias off a low energy portion of the positron beam, thus reducing its energy spread. The effect of the retarding potential ($V_{R3}$) on the energy distribution and the intensity of the positron beam is discussed in chapter 4.

The tuning and optimisation of the positron beam was achieved bearing in mind that if a charged particle passes from a region where the magnitude of the guiding magnetic field is $B_1$ to another of magnetic field strength $B_2$, then provided that the magnetic field strength changes gradually, the pitch angles of the particle in these two regions are related by:

$$\frac{B_1}{B_2} \propto \frac{\sin^2 \alpha_1}{\sin^2 \alpha_2}$$

(2.4)

where $\alpha_1$, $\alpha_2$ are the pitch angles in the regions where the magnetic field strengths are $B_1$ and $B_2$. Thus, when $B_1 > B_2$ it follows than $\alpha_1 > \alpha_2$. This may be used to parallelise the positron beam as it traverses the interaction region, if the magnetic field strength is arranged to be lower than at the positron source. However, the reduced pitch angle leads to an increase in the positron Larmor radii and, hence, in the diameter of the beam. The low magnetic field in the interaction region may also fail to confine scattered positrons.

In figure 2.7a, the circuit used to monitor the intensity of the positron beam is shown. The potential applied to the moderator is ramped whilst the pulses from a positron detector are amplified tenfold, fed to a constant fraction discriminator (CFD) and counted on a multichannel scaler (MCS). The MCS is controlled by an external ramp providing the advance pulse for increasing the voltage of the moderator in steps.
Figure 2.7. a) The circuit used to monitor the intensity of the positron beam; b) Typical variation of the positron beam intensity versus the moderator voltage.

and, correspondingly, of the $E \times B$ filter via a square rooter. In figure 2.7b, an example of the measured intensity of the positron beam against the moderator voltage is depicted.

2.4. The interaction region

In figure 2.8a, the scattering gas cell employed throughout the present work is shown. It was initially designed by Otto (1996), during the course of his Master's degree project, with a view to performing measurements of the excited-state positronium ($Ps^*$) formation cross-sections through detection of the prompt Lyman-α photon (from the 2P-1S decay of $Ps^*$) in coincidence with the remnant ion. The
scattering gas cell is differentially pumped and ions, produced in the interaction region, are extracted via a weak negative electric field. The inside of the aluminium hemispherical electrode (with a diameter of 90mm) of the gas cell is polished in order to increase the photon reflectivity. Two 8mm apertures on the sides of the cell allow the positron beam to transverse the interaction region and define its dimension. The apertures are positioned such that the centre of the positron beam passes 14mm from the base of the gas cell.

A weak non-uniform electric field is produced by a cylindrical electrode with length of 3mm and a diameter of 0.5mm placed in the hole at the centre of the bottom electrode, as shown in figure 2.8b. The electrode is insulated from the walls of the cell by means of a PTFE disc. After passing through the extracting electrode, ions are accelerated towards a channeltron, placed in a separate vacuum chamber with its own

![Diagram](image)

**Figure 2.8.** a) General view of the scattering gas cell; b) Schematic of the scattering gas cell and the ion detector (to scale).
diffusion pump, as shown in figure 2.5, to ensure vacuum conditions $<10^5$Torr necessary for its efficient operation. The cone of the channeltron is held at approximately -3kV to ensure that ions are detected efficiently as they hit the cone. For a similar type of detector, manufactured by Philips, the detection efficiency for protons and positive ions at 3kV impact energies is 50%.

The target gas is leaked to the middle of the gas cell through a LV10K Fine Control Leak Valve (manufactured by Edwards) and the pressure is monitored by means of a MKS 127 Baratron capacitance manometer.

2.5. Computer simulation of the performance of the ion extractor

The performance of the ion extractor was simulated using the Simion 6.0 computer program. It allows positron trajectories, time of flights, extraction efficiencies and other parameters such as the electric potential along the central axis of the gas cell to be characterized, so that the performance of the gas cell can be optimised.

Since the extractor was designed to be used with a DC voltage, a maximum value for the extracting lens voltage had to be determined, such that the trajectories of the positrons traversing the gas cell were not perturbed. In order to achieve this, the trajectories of positrons inside the gas cell were simulated and the potentials resulting from different voltages ($V_{\text{lens}}$) applied to the extracting lens were calculated. In figure 2.9, the dependence of the potential on the distance from the centre of the extracting lens is depicted for $V_{\text{lens}}$ equal to -500 and -800V. It can be seen in the inset of the figure that in the region traversed by the beam the absolute value of the potential arising from $V_{\text{lens}} = -800\text{V}$ increases more rapidly, on average by a factor of 1.6, as compared to $V_{\text{lens}} = -500\text{V}$. It has been concluded that the maximum voltage applied to the lens during measurements should not exceed $-500\text{V}$. Subsequently, it has been estimated that, in this case, the electric field perturbation results in the positron beam being accelerated on average by 0.2V, with less than 4% of the positron beam being accelerated on average by 0.4V, the highest value possible for the present geometry being 0.54V. Of course, the time of flights and time spreads of the ions detected depend on the value of the voltage applied to the extracting lens and the geometry of the gas cell.

In figure 2.10, the trajectories of Xe$^+$ ions inside the gas cell are shown. These results were obtained by performing simulations, with ions created isotropically within
Figure 2.9. The dependence of the potential on the distance from the centre of the extracting lens along the central axis of the gas cell.

Figure 2.10. Trajectories of Xe⁺ as simulated with Simion 6.0.
the region of the positron beam overlap. Only some of the trajectories are shown for clarity. Ions, which originate from the centre of the gas cell are transmitted through the lens and detected on the channeletron with comparatively short times of flight, whilst the ones originating somewhat outside the central axis significantly increase the time spread. It has also been found that ions are extracted from only half of the volume of the gas cell. The simulated arrival times and time spreads, which were found to agree well with the measured ones, are shown in figure 2.11 for Ar$^+$. For these targets, extraction efficiencies of around 50% have been obtained from the simulation.

It has also been found that for heavier targets, such as Xe, the extraction efficiency, $\varepsilon$, is not influenced by the strength of the magnetic field and, indeed, the magnetic field was not included in the simulations described above. It should be noted, however, that $\varepsilon$ is affected by the strength of the magnetic field in the case of the lighter Ne ions. In figure 2.12, the dependence of the extraction efficiency of Ne$^+$ versus the intensity of the magnetic field is shown. These findings will be used in chapter 4.

**Figure 2.11.** Arrival times and time spreads for Ar$^+$.2+ and Xe$^+$.2+ as simulated with Simion 6.0. The peak heights have been normalized for comparison of the respective widths.
2.6. Summary

The main common components of the apparatus used throughout the present work have been described. Particular attention has been given to the features which allow the system to be used to study the ionization phenomena. The scattering cell, which has been built with a view to perform Ps* formation cross-section measurements, has been characterised with respect to possible perturbations on the positron beam within the gas cell, the time of flight distributions of ions as well as their extraction efficiencies.

Figure 2.12. Dependence of the Ne⁺ extraction efficiency upon the magnetic field intensity.
CHAPTER 3

POSITRON IMPACT TOTAL IONIZATION CROSS-SECTIONS FOR NE AND XE AND ESTIMATES OF THE POSITRONIUM FORMATION CROSS-SECTIONS FOR THE INERT ATOMS

3.1. Introduction

In this chapter, experimental and theoretical results for positronium formation and direct ionization cross-sections \( Q_{Ps} \) and \( Q^+ \), respectively) for the noble gases are reviewed and compared. Results for hydrogen are also included because of their fundamental interest. Discrepancies are discussed and, where possible, the need for further investigations is identified. A method recently employed to extract Ps formation cross-sections by subtracting \( Q^+ \) from the total ionization cross-sections \( Q^+ \) is also discussed. In the present work, \( Q^+ \) which include a contribution from positronium formation, for Ne and Xe have been measured with high precision using the apparatus discussed in chapter 2. In contrast with a theoretical calculation, no evidence of a structure has been found in either \( Q^+ \) or the extracted \( Q_{Ps} \) in the vicinity of 80eV for Xe. Furthermore, an empirical scaling for positron- and electron-impact ionization cross-section has been found and enabled estimates for the contribution of excited-state positronium (Ps*) to the total Ps formation cross-section in the noble gases.

3.2. Experimental and theoretical overview

3.2.1. Positronium formation

Positronium in the ground or higher states can be formed when a positron collides with an atom or molecule \( (A) \), as described by the following capture reaction:

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

(3.1)

where the remnant ion may itself be left in an excited state. The threshold energy is defined by \( E_{Ps}=E_i-6.8eV/n^2 \), where \( E_i \) is the first ionization potential of the target atom/molecule, 6.8eV is the ground-state binding energy of Ps and \( n \) is the principal quantum number.
The first direct determinations of relative positronium formation cross-sections for He, Ar, H\textsubscript{2} and CH\textsubscript{4} were reported by Charlton \textit{et al} (1980) in the energy range from around the threshold to around 15eV above the threshold. The method relied on passing a beam of slow positrons through a scattering cell and detecting Ps formed by counting the simultaneous coincidence of the 3\gamma from the decay of o-Ps. These measurements were further extended to energies up to 150eV for all the noble gases by Charlton \textit{et al} (1983) with an improved version of the apparatus. However, it was later concluded (see Charlton and Laricchia, 1990) that these results were subject to considerable systematic errors, e.g. loss of signal due to the quenching of o-Ps and its decay into 2\gamma-rays resulting in the cross-sections in some instances being up to a factor of 5 lower than values acquired by Fornari \textit{et al} (1983).

Fornari \textit{et al} (1983) measured absolute $Q_{ps}$ from He, Ar and H\textsubscript{2} up to 76.3eV by measuring positrons lost in the scattering cell due to Ps formation. The method employed the simultaneous measurement of a time-of-flight (TOF) spectra and the intensity of the beam transmitted through a cell with and without gas. The idea was that all the positrons that did not form Ps should be confined by a uniform magnetic field (of ~100 Gauss) and reach the detector. The fraction ($F$) of the incident beam, which underwent any form of scattering, was extracted from the attenuation of the TOF spectra and the fraction ($f$) of the incident beam, which failed to be detected, was determined from the fractional decrease in the detector count rate. The Ps formation cross-section was then determined from $f Q_{tot} / F$, where the total positron cross-section, $Q_{tot}$, was taken from literature.

The work of Fornari \textit{et al} (1983) was extended to higher energies by Diana \textit{et al} (1986a and b) and to other inert atoms by Diana \textit{et al} (1985a, 1987, 1989). Their results agree reasonably between each other within the overlapping energy range, however the latter cross-sections contain unexplained structure above 100eV.

Fromme \textit{et al} (1986, 1988) measured the ionization of He and H\textsubscript{2} using a method based on the detection of ions produced in time correlation with the scattered positrons. The number of detected ions was a measure of the sum $Q_i^+ + Q_{ps}$, whilst the number of time-correlated ions was a measure of $Q_i^+$. The positronium formation cross-section obtained for He agrees well with the results of Fornari \textit{et al} (1983), however it does not show the structure exhibited by the data of Diana \textit{et al} (1986a) at high energy.
An experiment to measure $Q_{ps}$ for He performed by Overton et al. (1993) was essentially an improved continuation of a method employed by Diana et al. (1986a). The new cross-section was found to fall more rapidly with an increasing energy than that of Diana et al. (1986a) and, like the cross-section of Fromme et al. (1986), not to exhibit the structure present in $Q_{ps}$ of Diana et al. (1986a).

In figure 3.1, the experimental results for Ps formation in He are compared with the distorted-wave approximation of Mandal et al. (1979), the polarized-orbital method of Khan and Ghosh (1983), the close-coupling approximation of Hewitt et al. (1992), the second-order Born calculation of Sarkar et al. (1992) and the coupled-state calculation of Campbell et al. (1998). The theoretical calculations of Khan and Ghosh (1983), Khan et al. (1985) and Hewitt et al. (1992) contain contributions from ground and first excited state of Ps, while the values obtained by Mandal et al. (1979) and Sarkar et al. (1992) contain only contribution from the ground state formation. The more recent calculation for positron scattering by He of Campbell et al. (1998) includes a set of 27 pseudo-states and accounts for positronium formation in all excited states using the $n^3$ scaling for $\text{Ps}(n>3)$. This theory is in accord with most of experimental results up to 60eV and with the data of Fornari et al. (1983) and Diana et al. (1986a) up to 90eV. Unlike the measurements of Fornari et al. (1983) and Diana et al. (1986a), the cross-section obtained by Overton et al. (1993) follows the theoretical curve of Campbell et al. (1998) between 60eV and 120eV.

![Figure 3.1. Comparison of experimental and theoretical results for $Q_{ps}$ in He.](image_url)
although it lies 30% higher in this energy range. These measurements followed the earlier classical-trajectory Monte Carlo calculations of Schultz and Olson (1988), not shown in the graph, which indicate that above ~100eV the charge-transfer cross-section should fall off as $E^{-3.5}$, a result typical for charged-particle capture, however the experimental results follow the energy dependence closer to $E^{-1} - E^{-1.5}$. It was suggested that the discrepancy could be due to experiments failing to completely confine positrons scattered at large angles, thus overestimating $Q_{ps}$.

Zhou et al. (1997) carried out experiments investigating total and positronium formation cross-sections for $H$ and $H_2$. The experimental approach involved setting lower (LL) and upper limits (UL) on $Q_{ps}$ in conjunction with the total cross-section measurements. The lower limits were obtained using two photomultiplier tubes placed on opposite sides of a scattering cell and detecting coincidences between the 511keV annihilation γ-rays produced by the decay of p-Ps and by the interaction of o-Ps with the walls of the scattering cell. An important consideration was the magnitude of the magnetic field, which had to be sufficiently high in order to prevent positrons scattered without forming Ps from reaching the inner surface of the interaction region. The upper limits were obtained by performing beam attenuation measurements with the aim of detecting all bare scattered projectiles. The experimental and theoretical results for $Q_{ps}$ in $H_2$ are compared in figure 3.2. The lower and upper limits for $Q_{ps}$ of Zhou et al. (1997) determine a range of the cross-section values that is in agreement with the results of Fornari et al. (1983), Diana et al. (1986a) and Fromme et al. (1988) at all energies. The UL of Zhou et al. (1997) and the results of Fornari et al. (1983) were taken using essentially the same technique, but disagree by up to 18%. The results of Charlton et al. (1983), although affected by systematic errors, are also depicted since they are in fair agreement with other results at energies up to 12eV. The theoretical results of Bussard et al. (1979) were obtained semi-empirically below the ionization and excitation thresholds by extracting $Q_{ps}$ from the total cross-sections and above 50eV by using the theoretical data of Sural and Mukherjee (1970). This resulted in a cross-section, which overestimates the experimental data by ~30% at the maximum, but reproduces fairly well the experimental trends. The calculations performed by Sural and Mukherjee (1970) incorporated the first Born approximation and they agree well with the calculations of
Ray et al (1980), which were based on the molecular Jackson-Schiff approximation, in the range from 50-1000eV. Both the calculations of Sural and Mukherjee (1970) and those of Ray et al (1980) lie below the experimental results of Fornari et al (1983) and Diana et al (1986a). Calculations of Biswas et al (1991), also depicted in the figure, use the first Born approximation to predict the cross-sections, and unlike the earlier results, include a contribution from excited-state Ps(n=2). These are in fair agreement with the experimental values of Fornari et al (1983), Diana et al (1986a) and Fromme et al (1988) above 70eV. More recent investigations of Biswas and Ghosh (1997) of ground state positronium formation in H₂, using a simplified second order Born approximation, are believed to be more accurate since they include second-order perturbative terms which are a more appropriate way of treating a two-step process such as capture.

A similar comparison of the experimental and the theoretical results as for He and H₂ can be made in the case of H. In figure 3.3, the earlier discussed measurements of $Q_{Ps}$ of Zhou et al (1997) are compared to the experimental results of Weber et al (1994) and Hofmann et al (1997). Whilst there is a degree of consistency among the experimental results (Zhou et al, 1997 over the whole energy range and Weber et al, 1994 above 15eV) and the theoretical calculations, the data of Hofmann et al (1997) fail to reproduce the energy dependence of any of the recent theoretical predictions. The measurements of Zhou et al (1997) agree above 8eV with the Kohn variational calculation of Brown and Humberston (1985), which is believed to be one of the most
accurate in the Oe gap. Below this energy the calculations of Brown and Humberston (1985) are lower than the experimental results of Zhou et al (1997). The calculation of Janev and Solov’ev (1999) agrees very well with the experimental results of Zhou et al (1997) in the common energy range. The hyperspherical coordinate method employed by Igarashi and Toshima (1994) is in accord with experimental results of Zhou et al (1997) particularly in the energy range from 18-30eV and agrees well at low energies with other theoretical calculations, namely Brown and Humberston (1985), Kernoghan et al (1996) and Mitroy (1996). The close-coupling approach employed by Mitroy et al (1996) included a large L² basis of 28 positron-hydrogen states supplemented by the Ps(1s), Ps(2s) and Ps(2p) states. Within these calculations both Ps formation and ionization cross-sections for H were computed. A calculation performed on the same system using the coupled-state formalism with 33 states was performed by Kernoghan et al (1996). Both approaches agree reasonably well with experimental results of Zhou et al (1997). However, the calculations of Kernoghan et al (1996) exceed those of Mitroy et al (1996) at the maximum, as well as all other theoretical calculations shown, but decrease somewhat faster than the calculations of Mitroy et al (1996) at intermediate energies and merge with them at around 75eV. Both calculations include contributions from excited state Ps(n>2) assuming the n³ scaling law (Kernoghan et al, 1996). The theoretical calculations of Higgins and Burke (1993) and Igarashi and

![Figure 3.3. Comparison of experimental and theoretical results for Q_p in H.](image-url)
Toshima (1994) include only the ground state Ps formation and are consistent with the results of Zhou et al (1997) at energies between 18eV and 35eV.

The most recent and comprehensive investigation of relative total ionization cross-sections from the threshold up to 1000eV and total positronium formation cross-sections in the noble gases have been carried out by Laricchia et al (2002). The experimental set-up was very similar to that described in chapter 2. The experimental method relied on measuring the total ion yield (Y) as a function of energy:

\[ Y = \frac{1}{p} \frac{N_I - B_i}{N_{e+} - B_{e+}} \]

where \( N_I \) and \( N_{e+} \) refer to the time-normalized ion and incident positron counts, respectively, \( B_i \) and \( B_{e+} \) correspond to the associated backgrounds and \( p \) is the target gas pressure in the scattering gas cell. The total ion yields are directly proportional to the total ionization cross-sections defined by:

\[ Q'_i = Q_{Ps} + Q'_i + \sum HO \]

where \( Q_{Ps} \) and \( Q'_i \) are the cross-sections for positronium formation and direct single ionization, respectively and \( \sum HO \) represents the sum over all other processes contributing to ionization, such as annihilation, transfer ionization and multiple ionization, and is usually small.

The \( Q'_i \) were measured with a pulsed electric field using a gas cell described in detail by Moxom et al (1995) with a reduced positron beam spread (\( \Delta E \leq 0.4 \) eV) up to energies of 40eV. However, in the case of Ne and Xe, \( Q'_i \) obtained in the course of the present work and presented in the following sections of this chapter were also included, providing an additional set of data measured with low statistical uncertainties. The high precision of the present results is due to the use of a weak DC electric field to extract ions and the full-spread (\( \Delta E \approx 1.5 \) eV) positron beam.

The \( Q'_i \) of Laricchia et al (2002) were extended to energies high enough such that the absolute scale could be set by normalizing to corresponding cross-sections by electron impact. Subsequently, the positron data for both total and direct ionization were renormalized to new high-precision electron results of Sorokin et al (1998 and 2000), as discussed in detail by Van Reeth et al (2002). Previously, the normalization had been made to the data of Krishnakumar and Srivastava (1988). In order to evaluate \( Q_{Ps} \), the values of \( Q'_i \), obtained by a fit to the renormalized cross-sections of Kara et al (1997)
for Ne, Kr and Xe and Moxom et al (1996) for Ar (see section 3.2.2), were subtracted from $Q'$, according to:

$$Q_n = Q' - \sum HO$$

(3.4)

Whilst the higher order term $\sum HO$ includes all ionization processes of degree >1, inner shell ionization and annihilation, the available total double ionization cross-sections of Bluhme et al (1999a and 1999b) were included. Since the total double ionization cross-section contribution is small (in Xe up to 8% at energies between 100 and 250eV), they were not renormalized.

In figure 3.4, $Q_n$ for Ne, Ar, Kr and Xe of Laricchia et al (2002) are compared with previous experimental results as well as theoretical calculations of McAlinden and Walters (1992) and Gilmore et al (2002). Overall, the agreement amongst the available data is not satisfactory. There is, however, a broad agreement among the experimental results at low energies; in particular, in the case of Ne and Ar between the results of Laricchia et al (2002) and those of Jin et al (1994) in the common energy range. In almost all cases, the results of Laricchia et al (2002) lie below all other experimental data (Stein et al, 1998 UL; Diana et al, 1985a, 1986b, 1987, 1989 and Fornari et al, 1983). The only exception being the cross-sections of Charlton et al (1983) which, as already discussed, are thought to have been affected by systematic errors. These resulted in a progressively lower detection efficiency for increasingly faster positronium atoms. As a consequence, the second peak in those cross-sections indicates the opening of one or more channels for positronium formation, which could give rise to slower positronium atoms. It might be surprising that the data of Laricchia et al (2002) lie in the intermediate energy range below LL of Stein et al (1998), since their $Q_{ps}$ for H are in very good agreement with other experimental and theoretical calculations, as shown in figure 3.3. However, it is possible that in the case of heavier noble gases scattering at large angles occurs, which would lead to overestimation of $Q_{ps}$ in the case of LL of Stein et al (1998). The results of Laricchia et al (2002) do not reveal the structures in the cross-sections observed by Diana et al (1985a, 1986b, 1987, 1989) at higher energies. In the case of Kr, there is some agreement with the data of Diana et al (1987, 1989) at low energies. In the case of Xe, the upper limit measurements of Stein et al (1998) display a similar position for the first peak and show evidence of a structure at somewhat higher energies than the second peak in the data of Charlton et al (1983).
Figure 3.4. Comparison of $Q_{PS}$ for the noble gases (from Laricchia et al., 2002).
A conspicuous feature of the results of Laricchia \textit{et al} (2002) is the double-peak structure in $Q_{Ps}$ for Ar, Kr and Xe. The first peaks appear at roughly twice the threshold energy for ground state positronium formation. The maxima of the second peaks appear roughly at twice the value of the threshold energy for excited-state positronium formation. They have been attributed to the excited state positronium formation. However, an earlier interpretation of Stein \textit{et al} (1998) suggested that the second peak in $Q_{Ps}$ of Ar, Kr and Xe measured by Charlton \textit{et al} (1983) (and a similar structure in Xe in their own data) could be due to the ground-state Ps formation with an inner-shell electron. Stein \textit{et al} (1998) noted that the thresholds for the Ps formation from the $ns$ subshell (29.2, 27.5, 23.4eV for Ar, Kr and Xe respectively) were in close proximity to the position of the second peaks in the cross-sections of Charlton \textit{et al} (1983). However, the peak in $Q_{Ps}$ of Charlton \textit{et al} (1983) for Ne appears clearly below the 2s threshold (48.8eV). More convincingly, as already shown in figure 3.1, $Q_{Ps}$ of Charlton \textit{et al} (1983) for He also reveals a second peak, even though no inner shell-processes can be attributed to this target.

Very recent results of Sullivan \textit{et al} (2002b) for the Ps formation cross-section of Ar, also shown in figure 3.4, were obtained by performing a beam attenuation measurement with a positron beam of 25meV energy resolution, shown in figure 1.15. Buffer-gas trapping has been discussed in more detail in section 1.3. It can be seen that although there is a good agreement on the magnitude and the energy dependence of the first peak between these results and those of Laricchia \textit{et al} (2002), the data of Sullivan \textit{et al} (2002b) begin to drop below those of Laricchia \textit{et al} (2002) above 20eV.

The coupled static calculations of McAlinden and Walters (1992) for ground state positronium formation, in general, overestimate the magnitude of the cross-sections of Laricchia \textit{et al} (2002) at low energies and underestimate them at higher energies. In the case of Xe, a reasonable agreement is found between this experiment and the theory. For Ar, Kr and Xe the theoretical calculations hint to a possible structure in the cross-sections at around 100eV. Since the most pronounced structure is present in the case of Xe, a study has been undertaken as part of the present work to confirm or discount it and will be described in section 3.3.

The distorted wave Born approximation calculations of Gilmore \textit{et al} (2002) includes Ps formation in 1s, 2s, 2p, 3s, 3p, 3d states and uses the $n^3$ scaling rule to estimate Ps formation in the higher states. Allowance is also made for the capture of an
electron from both the outermost and the first inner shell of the atom. As indicated in figure 3.4, the theory overestimates the experimental cross-sections by roughly a factor of 2 in the case of Ne and Ar and roughly a factor of 3 in the case of Kr and Xe. The double peak structure in Ar and the shoulders in Kr and Xe are reproduced and seem to confirm that they arise from Ps formation in the excited state. However, the calculated widths of the formation curves are much smaller than those measured in the experiment.

3.2.2. Direct ionization

Single direct ionization by positron impact with a threshold energy $E_i$ can be defined by the following reaction:

$$e^+ + A \rightarrow A^+ + e^+ + e^- \quad \text{(3.5)}$$

where the remnant ion may be left in an excited state.

Fromme et al (1986, 1988) measured $Q_i^+$ simultaneously with $Q_{ps}$ for He and H$_2$, as discussed in 3.2.1. The $Q_i^+$ were obtained from background-subtracted TOF spectra. Measurements of $Q_i^+$ by electron impact with the same apparatus allowed the determination of the energy at which these two cross-sections merge and the relative cross-sections were normalized above 750eV to the data of Montague et al (1984). Their results show that $Q_i^+$ by positron impact exceeds that by electron impact at intermediate energies.

The energy loss experiments of Mori and Sueoka (1994) were extended to energies of approximately 100eV and the normalization method employed total ionization cross-sections obtained by other experiments. Measurements relying on detection of coincidences between correlated ions and scattered positrons (e.g. Kara et al, 1997; Moxom et al, 1996; Jacobsen et al, 1995a and 1995b; Knudsen et al, 1990) were obtained typically up to 1000eV where normalization can be made to the known electron cross-sections. In the case of the results of Kara et al (1997) for Ne, Kr and Xe, the electron data of Krishnakumar and Srivastava (1988) were used for normalization.

In figure 3.5, the experimental and theoretical results of $Q_i^+$ for He are compared, including the experimental data of Mori and Sueoka (1994), Diana et al (1985b), Fromme et al (1986), Knudsen et al (1990), Jacobsen et al (1995b) and Moxom et al (1996). There is a broad agreement amongst these data. The data of Fromme et al (1986) and Knudsen et al (1990) are above the data of Jacobsen et al (1995b) and
Figure 3.5. Comparison of experimental and theoretical results for \( Q^+ \) in He.

Moxom et al (1996) at low energies. The data of Mori and Sueoka (1994) are in broad agreement with the other experimental results in the common energy range, though they have large uncertainties. Similarly, the results of Diana et al (1985b) are characterised by large uncertainties and do not follow a smooth curve as suggested by the other experimental results.

The experimental data are also compared with recent theoretical results. The calculation of Campbell et al (1998) has already been discussed with respect to positronium formation in He in section 3.2.1 and is seen to be in fair agreement with most experiments. The coupled state calculation of the positron ionization cross-section for He was performed by Chen and Msezane (1994) from 60eV to 600eV. Although, this calculation exceeds the experimental values in the intermediate energy range, it reproduces the energy dependence well at higher energies. The continuum optical potential model calculation of Ratnaveluv (1991) is in good agreement with experimental data over the entire energy range. The most recent distorted-wave model of Campeanu et al (2002) yields a cross-section that is lower than most of the other experimental and theoretical data in the intermediate energy range. It is up to 20% lower in the region of the peak than the results of Moxom et al (1996) but tends to converge with them above 250eV.
Figure 3.6. Comparison of experimental and theoretical results for $Q_e^+$ for the noble gases (Van Reeth et al., 2002).
In figure 3.6, the renormalized (see Van Reeth et al., 2002) experimental data of $Q_i^*$ Kara et al. (1997) and Moxom et al. (1996) for Ne, Ar, Kr and Xe are shown together with the experimental results of Mori and Sueoka (1994), Jacobsen et al. (1995b) and the distorted wave calculations of Moores (1998) and Campeanu et al. (2002). In general, there is a broad agreement between the experimental data and the calculation of Campeanu et al. (2002) from the region of the peak up to 500eV, although this was not the case for He. The cross-section of Moores (1998) peak at an energy close to the experimental data but are up to 60% larger, the disagreement being greatest for the heavier targets. There is good agreement in the shape of the cross-sections measured by Kara et al. (1997) and Moxom et al. (1996) with those of Jacobsen et al. (1995b). In the case of Ar, the data also agree in magnitude but they do not in the case of Ne. This, however, may be fully accounted for by the different data used for normalization in the case of the two measurements. The data of Mori and Sueoka (1994) have large uncertainties but, within those, they agree with the data of Kara et al. (1997) and Moxom et al. (1996).

### 3.3. Positron impact total ionization cross-sections for Ne and Xe and the extracted positronium formation cross-section for Xe

#### 3.3.1. Overview

Measurements of $Q_{p_i}$ are experimentally difficult due to problems associated with discriminating between direct ionization and positronium formation, as both processes result in an ion in the final state. With regards to this, another useful quantity, yet much easier to measure, is the total ionization cross-section, already defined by eq. 3.3. Much important information may be extracted from the total ionization cross-section, even though it includes more than one process. Above the Ps formation threshold, $E_{Ps}$, annihilation may be neglected and hence positronium formation becomes the first channel to open. Below the ionization threshold, $E_i$, $Q_i^t = Q_{p_i}$ and $Q_i^t$ can be used to study the near threshold behaviour of $Q_{p_i}$. At higher positron impact energies, $Q_{p_{ps}}$ becomes small such that $Q_i^t \approx Q_i^+$, therefore $Q_i^t$ can be used to study the merging of $Q_i^+$ by positron and electron impact, as predicted by the Born approximation. Since $Q_i^+$ for a variety of targets are available, as discussed in section 3.2.2, $Q_{p_{ps}}$ may be
obtained from eq. 3.4 by subtracting \( Q'_i \) from \( Q_i \). This is relevant since significant discrepancies exist among the experimental results of \( Q_{ps} \) for the inert atoms, as reviewed in section 3.2.1.

In this work, the total ionization cross-sections have been measured with high precision for Ne and Xe in the intermediate energy range using a gas cell incorporating a DC ion extraction electric field and a full-energy spread positron beam. The choice of targets was dictated by a few factors. Firstly, detailed measurements of the total ionization cross-section for Xe by Laricchia et al (2002) hinted to a possible structure in the vicinity of 80eV, as was predicted in the theoretical cross-sections of McAlindien and Walters (1992). Secondly, very precise measurements of \( Q'_i \) provided additional data for Xe and Ne later used for normalization and, subsequently, extraction of \( Q_{ps} \), as discussed in section 3.2.1. Lastly, measurements performed with Ne, where no hint of a structure in the theoretical prediction was found, gave a necessary check in order to rule out any perturbation of the weak electric field on the positron beam.

3.3.2. The experimental method and systematic effects

The experimental arrangement has already been discussed in detail in chapter 2 and is shown in figure 2.5.

The experimental method followed previous works by Moxom et al (1995) and Laricchia et al (2002) and relied on measuring total ion yields already defined by eq. 3.2. These ion yields are directly proportional to the total ionization cross-section, as given by eq. 3.3.

Figure 3.7 depicts the electronic circuit used to measure ion yields. Signal from the ion detector, amplified tenfold, was processed by the Ortec 584 constant fraction

**Figure 3.7. Schematic diagram of the electronic circuit used to measure ion yields.**
Chapter 3  Positron impact $Q_1$ for Ne and Xe and estimates of $Q_1$, for the inert atoms

discriminator and collected on a multichannel scaler (MCS), set to operate in a multiple sweep mode with a dwell time of 100 seconds. This minimised instrumental drifts and fluctuations in the gas pressure for the runs which lasted for ~70ks. The MCS was controlled by an external ramp, which provided an advance pulse in order to increase the voltage of the moderator in 2V steps and correspondingly the potential applied to the ExB plates via the square rooter.

Tests were carried out to ensure that at all energies the signal increased linearly with the target gas pressure. In order to maintain single collision conditions, the pressure range was arranged so that no more than approximately 10% of the incident positron beam was attenuated by the target gas.

A significant finding of Laricchia et al (2002) with respect to that of Moxom et al (1995) was the observation that, for Ar and the heavier inert atoms, the ion yield decreased at intermediate energies with increasing magnetic field in the scattering region. This was interpreted as being related to the production of secondary electrons (and ions from these) from unconfined scattered projectiles. The ion yield was found to be insensitive to further increase in the magnetic field for a given geometry at around 140 Gauss. In the present work, the magnetic field in the scattering region was set at ~150 Gauss.

In order to eliminate any possible contributions from fast particles in the beam, both the incident beam and the ion count rate were measured with the slow positron beam biased on and off. In each case, the two time-normalized spectra were subtracted from each other according to eq. 3.2.

A very important consideration for the present measurements was the influence of the weak DC electric field, produced by a ~500V voltage applied to the extracting lens, on the positron beam. Although, as discussed in chapter 2, computer simulations have been carried out in order to determine the maximum possible voltage, which could be applied to the lens without perturbing the positron beam, experimental tests have also been performed by measuring total ion yields for Ne. These yields were then normalized both in energy and magnitude to the data of Laricchia et al (2002). It has been found that in order to match the energy dependence of the results of Laricchia et al (2002), 1.6eV needed to be added to the present moderator voltage. In figure 3.8, the total ionization cross-section obtained for Ne is compared to that of Laricchia et al (2002). Very good agreement is found between the two sets. No effect is observed in the present data due to possible perturbations of the weak static electric field on the collisions.
However, a deviation of \( \approx 5\% \) in the energy range 20-40eV between the present results and those of Laricchia et al (2002) has been attributed to the different ion extraction methods employed. The use of the full-intensity positron beam resulted in an increased data collection rate, such that a statistical uncertainty of around 1% has been achieved above the positronium formation threshold. The error bars on the present \( Q_i' \) in figure 3.8 lie within the size of the experimental points.

In the case of Ne, a residual ion yield below the positronium formation threshold has been observed in this work. An investigation has been carried out in order to understand this phenomenon and the results are presented in chapter 4 of this work. Since this residual yield amounts to no more than 1% of the ion yield 5eV above \( E_{ps} \), its average value was determined from the experimental points below the positronium formation threshold and subtracted from the total ionization cross-section.

### 3.3 Results and discussion

Having verified the reliability of the present method with Ne, the total ionization cross-section for Xe has been measured. The results are presented in figure 3.9 where a very good agreement can be seen between the present data and those of Laricchia et al.
(2002). However, the results for total single ionization, $Q_{i}^{+}$, of Bluhme et al (1999a) are very different from the present ones up to 100eV. The statistical uncertainties of the current results are approximately 1.5%. While the previous data of Laricchia et al (2002) exhibit a greater scatter above 30eV, with statistical uncertainty of more than 3% and hint to a possible structure at around 80eV, the present data decrease very smoothly with increasing energy.

The present $Q_{i}^{+}$ and those of Laricchia et al (2002) for Xe have been normalized to the corresponding cross-sections by electron impact of Sorokin et al (1998 and 2000) at high energies, as already discussed in section 3.2.1. The positronium formation cross-section for Xe has been obtained using eq. 3.4 by subtracting the values of $Q_{i}^{+}$ of Kara et al (1997) obtained by a fit to the renormalized cross-section (discussed in detailed by Van Reeth et al, 2002) and the total double ionization cross-section ($Q_{i}^{2+}$) of Bluhme et al (1999a) from the present $Q_{i}^{+}$ and those of Laricchia et al (2002). This is illustrated in figure 3.10, where the present experimental results and those of Laricchia et al (2002) are compared to the renormalized $Q_{i}^{+}$ of Kara et al (1997) and $Q_{i}^{2+}$ of Bluhme et al (1999a). A fit to $Q_{i}^{+}$ of Kara et al (1997) and the ionization cross-section by electron impact of Sorokin et al (2000) are also shown.

![Figure 3.9. Total ionization cross-section of Xe.](image-url)
Figure 3.10. Ionization cross-sections of Xe.

In figure 3.11, the positronium formation cross-section for Xe, obtained in the manner described above, is compared with the theory of McAlinden and Walters (1992). No evidence of a structure in the vicinity of 80 eV is found, as shown in the enlarged inset of the figure.

The experimental data reveal two maxima in the cross-section, one at around 10 eV and the other just above 20 eV. The theoretical calculations predict a peak at a somewhat lower energy in comparison with the first peak found in the experimental data, although its magnitude is comparable. The second peak appears only in the experimental results and, as discussed in section 3.2.1, might be associated with positronium formation in excited states (Laricchia et al, 2002), which is not included in the theory.
3.4. An empirical scaling and estimates for the excited-state positronium formation cross-sections of the inert atoms

3.4.1. Introduction

It has been shown in the case of electron impact that scaling laws and empirical and semiempirical methods can be powerful tools in estimating sufficiently accurately absolute values of direct ionization cross-sections (for a review see Younger and Märk, 1985; for more recent developments see Kim et al., 1998 and Deutsch et al., 1995). This is particularly valuable when there are no experimental data available and rigorous theoretical treatments are not feasible. Some of the existing approaches are sophisticated and include the effect of the binding energy from different electron shells in a target atom (e.g. Lotz, 1967, 1968 and 1970). All of them, however, assume the Born dependence, \((\ln E/E)\), of the high-energy limit of the cross-section. In the case of positron impact, such a parametrization would prove particularly helpful since the available data are much more limited as compared to the electron ones.
One of the attempts to predict ionization cross-sections by positron and electron impact was that reported by Rost and Pattard (1997). It was found that the ionization cross-sections, $Q_x$, for a given projectile had a common shape when plotted as $Q_x/(Q_x)_{\text{max}}$, where $(Q_x)_{\text{max}}$ is a maximum value of a cross-section, versus the scaled energy $E/E_{\text{max}}$, $E_{\text{max}}$ being the energy at which the maximum value of the cross-section occurred. With this formula, several cross-sections by either positron- or electron-impact were found to fit a universal-shape function.

Recently, strong correlation between cross-sections and threshold energies for positronium formation and direct ionization were revealed (Van Reeth et al, 2000 and 2001; Humberston et al, 2001) for the noble gases and the alkali atoms. The cross-sections for a given process, $Q_x$, of atoms belonging to the same column of the periodic table were found to decrease exponentially with a relevant threshold energy, $E_{th}$, and were expressed as:

$$Q_x(E') = a(E') \exp[-b(E')E_{th}]$$ (3.6)

where coefficients $a$ and $b$ depend both on the excess energy, $E' = E - E_{th}$, and are the same for a given column of the periodic table. This formula was used, for instance, to predict a value of the triple ionization of Xe at 3eV excess energy and was found to agree reasonably well with that subsequently measured by Moxom (2000). Although, the investigated correlations extend over a few orders of magnitude, its specific exponential form is not yet understood. However, the fact that data relating to several processes fit so well to eq. 3.6 is suggestive of a significant underlying physical cause.

3.4.2. Excited-state positronium formation

An attempt to estimate lower (LL) and upper (UL) limits for the excited state positronium (Ps*) formation cross-section has been prompted by the results of Laricchia et al (2002), where a second peak in $Q_{Ps}$ in Ar, Kr and Xe has been attributed to the possible formation of positronium in the excited states, as discussed in section 3.2.1.

The evaluation of the UL follows the finding of a simple scaling formula which factors out the main target dependence of the shape and magnitude of the positron- and electron-impact ionization cross-sections. More specifically, cross-sections for positronium formation or direct single ionization are found to fall broadly on a common curve when scaled in terms of their corresponding threshold energy ($E_{th}$) as follows:
\[ \frac{Q_x^A \left( \frac{E}{E_{th}^A} \right)}{(Q_x^A)_{\text{max}}} = \frac{Q_x^B \left( \frac{E}{E_{th}^B} \right)}{(Q_x^B)_{\text{max}}} \]  

(3.7)

where \( x \) refers to a particular process (e.g. Ps formation or direct ionization) for a given projectile (e.g. positrons or electrons) and \( A, B \) refer to any two atoms. \( E \) is the incident projectile energy and \((Q_x^A)_{\text{max}}\) the maximum value of \( Q_x^A \).

In figure 3.12a, the ground-state positronium formation cross-sections, \( Q_{ps}^1(1s) \), for H and He (Kernoghan et al, 1996 and Campbell et al, 1998) are plotted in the scaled units \( Q_{ps} (Q_{ps})_{\text{max}} \) versus \( E/E_{th} \). Although the scaled cross-sections for He are slightly shifted to higher values of \( E/E_{th} \), the shapes of the cross-sections for both atoms are very similar. A good correspondence is also observed for the cross-sections for positronium formation in the first excited states \( Q_{ps}^2(2s) \), as shown in figure 3.12b and somewhat more poorly for \( Q_{ps}^2(2p) \), as depicted in figure 3.12c. It should be remarked that the sum of the theoretical data for the various quantum states has been found to be in good agreement with measurements of \( Q_{ps}(all\ n) \) for both targets, as shown in figures 3.1 and 3.3.

In figure 3.13, the scaled experimental results of Laricchia et al (2002) for \( Q_{ps}(all\ n) \) for Ne, Ar, Kr and Xe are shown together with the theoretical calculations of \( Q_{ps}^1(1s) \) for He (Campbell et al, 1998). These cross-sections have been scaled to the

**Figure 3.12.** Scaled positronium formation cross-section \( Q_{ps} / (Q_{ps})_{\text{max}} \) versus \( E/E_{th} \) for He and H. Here \( E_{th} \) is the Ps formation threshold energy. Ground state positronium formation cross-section a), excited states positronium formation cross-sections b) 2s and c) 2p (from Szluńska et al, 2002).
magnitude of the first peak and the energy has been scaled to the threshold energy for positronium formation into the ground state. The results for Ar, Kr and Xe are seen to overlap up to the position of the first maximum (at \( E/E_{th} \approx 1.8 \)) and somewhat beyond it. For Ne, a broader peak is found at \( E/E_{th} \approx 2.3 \). Although slightly shifted to a higher value of \( E/E_{th} \), the scaled \( Q_{Ps}(1s) \) of He displays a similar shape to the first peak of the heavier inert atoms.

With reference to figure 3.14, illustrating the case for all the inert atoms, the upper limits for \( Q_{Ps}(n>1) \) have been estimated by subtracting, from the measured \( Q_{Ps}(n) \) (all \( n \)), \( Q_{Ps}(n=1) \) obtained by empirically scaling the energy dependence of the He cross-section with an additional parameter \( \alpha \), according to:

\[
E^A = \alpha \left( E^{He} / E^{He}_{Ps} \right) E^{A}_{Ps}
\]

where the superscript \( A \) refers to a given atom. In order to reproduce the low-energy data and the correct position and width of the first peak, values of \( \alpha = 0.88, 0.85, 0.84 \) and 0.82 have been determined for Ne, Ar, Kr and Xe, respectively. In order to obtain
Figure 3.14. Positronium formation cross-sections for Ne, Ar, Kr and Xe (see caption).
the high-energy tail of the scaled $Q_{p_s}(n=1)$ of He, an exponential function has been fitted to the decrease of this peak, as shown in figure 3.14.

The lower limits have been obtained by extrapolating the energy dependence from the decrease of the first peak to the high-energy tail, the latter reduced by 30% as for the high-energy $Ps^*$ contribution calculated for He by Campbell et al (1998). The absence of the distinct peaks in Ne does not make the evaluation of the lower limit possible for this target. It should be noted that in both estimation procedures for UL and LL, the first peak is assumed to be entirely due to $n=1$ positronium. This might lead to an underestimation of $Ps^*$ at low energies. This is particularly evident in the case of Ne, where as shown in figure 3.14, the UL appears approximately 15eV above the actual threshold for the excited-state positronium.

In figure 3.15, both the LL and UL fractional contributions of $Ps^*$ to the total positronium formation cross-section estimated for Ne, Ar, Kr and Xe are shown together with the theoretical calculation for He of Campbell et al (1998) from around threshold to 60eV. Above this energy, an insufficient number of experimental points as well as large associated uncertainties prevent meaningful deductions. The estimated LL contributions of $Ps^*$ increase from $\sim(25 \pm 12)\%$ for Ar and Kr to $\sim(50 \pm 13)\%$ for Xe at the intermediate energy range. The UL are considerably larger $(40 \pm 8)\%$ and $(100 \pm 13)\%$ for Ne and Xe respectively, at intermediate energies. Whilst direct theoretical and experimental investigations of $Ps^*$ formation from these targets are undoubtedly needed, the present estimates suggest that $Ps^*$ formation might occur much more abundantly than previously thought (Van Reeth et al, 2000 and Humberston et al, 2001).

![Figure 3.15. Energy dependence of the relative contributions from $Ps^*$ to the total positronium formation cross-section (from Szluńska et al, 2002).](image)
3.4.3. Further work

In section 3.4.2, the application of the simple scaling formula has been discussed with reference to positronium formation cross-sections for the noble gases and hydrogen. This procedure was also extended to direct ionization cross-section by positron- and electron-impact. In the case of electron collisions, data are available not only for H and the inert atoms but also for atoms belonging to the last six columns of the periodic table. As demonstrated by Szűnrı́ska et al (2002), the scaled cross-sections by electron impact for atoms belonging to the IV, V, VI columns, the halogens and the noble gases fall on a common curve when atoms belonging to the second row (i.e. C, N, O, F and Ne) are omitted. These, however, scale within themselves. The reason why they differ from the rest is not yet understood and requires further investigation. Despite the differences among various atoms, it seems that there exists a common shape, 

\[ Q_x \left( \frac{E}{E_{th}} \right) \left( \alpha \right)_{max} \] 

which describes the probability, \( P_x \), for the occurrence of a particular process \( x \) by a given projectile.

The parametrization expressed by eq. 3.7 depends on two parameters, namely \( (Q_x)_{max} \) and \( E_{th} \), for each atom but can be further simplified to depend merely on one parameter since \( (Q_x)_{max} \) for atoms belonging to the same column of the periodic table were found to correlate very well with \( E_{th} \), as depicted in figure 3.16. For a specific process \( x \), the maximum in the cross-section, \( (Q_x)_{max} \), can be approximated as:

\[ (Q_x)_{max} = \alpha \exp(-\beta E_{th}) \]  (3.9)

where \( \alpha \) and \( \beta \) are constants, which are the same for all atoms belonging to a given column of the periodic table. Substitution of (3.9) into (3.7) leads to a simple formula in terms of \( P_x \):

\[ Q_x \left( \frac{E}{E_{th}} \right) = P_x \exp(-\beta E_{th}^x). \]  (3.10)

Therefore, this formula, with only one input parameter, can be used to estimate a cross-section for a given atom and process if the maxima in the cross-sections of any another two atoms belonging to the same column of the periodic table are known. The mechanism responsible for such a factorisation has yet to be understood.

It is interesting to note that the scaling formula described by eq. 3.7 is reminiscent of that developed by Rost and Pattard (1997), the connection being that, for a given process and projectile, the position of the maximum in the cross-sections appears at
Figure 3.16. Maximum value of the cross-section plotted versus relevant threshold energy, $E_{th}$: (a) direct ionization of the inert atoms by positron impact; (b) Ps formation for the inert atoms; (c) single ionization by electron impact for the inert atoms, halogens and atoms belonging to the IV, V and VI columns (from Szluińska et al, 2002).

approximately the same multiple value of $E_{th}$. The formula described by eq. 3.10 may seem similar in form to the correlations discussed in section 3.4.1 and expressed by eq. 3.6. However there is not a direct link between the two since, as already discussed above, coefficients $a$ and $b$ in eq. 3.6 are dependent on the excess energy, whereas $\alpha$ and $\beta$ in eq. 3.7 are constants that are the same for atoms belonging to the given column of the periodic table.

3.5. Summary

In this chapter, an overview of the experimental and theoretical results for positronium formation and direct ionization cross-sections has been given. High precision measurements of the total ionization cross-section, in the intermediate energy range, for Ne and Xe have been presented. The total ionization cross-section for Ne was used to test the system, which employed an ion extractor with a weak static electric field. A good agreement with the results of Laricchia et al (2002), which employed a pulsed electric field, was found in the common energy range. The total ionization cross-sections for Xe were thus measured, and together with the results for Ne provided an additional set of data, which were used to extract $Q_{ps}$. It has been found that both $Q_{i}'$ and $Q_{ps}$ for Xe decrease smoothly with increasing energy and no structure has been observed at around 80eV, which the theoretical calculations of McAlinden and Walters

Prompted by the recent results of \( Q_{p^0} \) for the noble gases of Laricchia et al (2002), the lower and upper limits for the contribution of the excited state positronium formation for these targets has been estimated. The evaluation of the UL follows the finding that plots of \( Q_{p^0} / (Q_{p^0})_{\text{max}} \) versus \( E / E_{th} \) broadly yield a common curve for a variety of targets ((\( (Q_{p^0})_{\text{max}} \) being the peak value of the cross-section and \( E_{th} \), in this case, is the Ps formation threshold energy). The results presented for LL and UL suggest that the Ps* might occur much more abundantly than previously thought (Van Reeth et al, 2000; Humberston et al, 2001).

It is worth mentioning that in the case of the total ionization cross-section for Ne a positive residual signal below \( E_{p^0} \) has been observed. The origin of this signal has been a motivation for work subsequently undertaken and described in the next chapter.
4.1. Introduction

The annihilation of positrons in collision with atoms and molecules is presently very topical. Together with positronium formation and direct ionization, it constitutes a process by which an atom or molecule may be ionized.

In this chapter, work undertaken to determine the energy dependence of the annihilation cross-sections \( Q_{\text{ann}} \) for Ne, Xe and \( \text{O}_2 \) below the positronium formation threshold \( E_{\text{ps}} \) is presented. The energy resolution of the present positron beam is much inferior to that from a positron trap and, therefore, structures like that reported by Gilbert et al (2002) (discussed further in section 4.2) would be washed out by the present experimental resolution. However, having found an ion signal below \( E_{\text{ps}} \) in the total ionization cross-sections for Ne, discussed in Chapter 3, we were encouraged to attempt the present investigation. In comparison to the San Diego method, which uses single \( \gamma \)-ray counts to detect annihilation events (e.g. Sullivan et al, 2002b), the present work has the advantage of measuring coincidences between \( \gamma \)-rays and mass-selected ions. Such a signal is characteristic of annihilation and positronium formation in positron-gas collisions and is less prone to background counts.

4.2. Theoretical and experimental overview

The basic principles governing positron annihilation have been described in section 1.2.1, where the free annihilation cross-section was given by eq. 1.4 in terms of the annihilation-rate parameter \( Z_{\text{eff}} \) as \( Q_{\gamma \gamma} = \frac{\pi n_0^2 c Z_{\text{eff}}}{\nu} \).

For almost 40 years, positron annihilation has been studied primarily by employing the lifetime method. In figure 4.1, an example of the schematic of a positron lifetime spectrometer is shown. In essence, a lifetime experiment measures the time
interval between the introduction of an energetic positron into the gas under investigation and its subsequent annihilation. The positron emission from the source is followed swiftly by a 1.28MeV γ-ray, which when detected by a photomultiplying tube starts a timing sequence. This sequence is stopped upon the detection of a γ-ray from annihilations in surrounding gas. Measurements are repeated for a large number of positrons in order to build up a lifetime spectrum. Details of this technique have been given by Coleman et al. (1974) and Griffith and Heyland (1978). Moreover, using the lifetime method Heyland et al. (1982) provided one of the first pieces of experimental evidence of large $Z_{eff}$ for molecules such as alkanes.

Following the technique used by Coleman et al. (1974) and Griffith and Heyland (1978), the positron lifetime parameters in $\text{H}_2$, $\text{CO}_2$ and $\text{CH}_4$ (Wright et al., 1983) and in $\text{Kr}$ and $\text{Xe}$ (Wright et al., 1985) were investigated. A typical lifetime spectrum and the method of analysis are shown in figure 4.2. The spectrum exhibits characteristic features such as the prompt $t=0$ peak, corresponding to events due to positron annihilation in the source holder and chamber walls as well as p-Ps annihilation, followed by fast components (shown in greater detail in the inset of the figure). The free-positron shoulder, caused by annihilation prior to thermalization, is present in all positron-inert
Figure 4.2. Positron lifetime spectrum for Xe at 9.64 Amagat and $T=297K$ with 0.109ns per channel. The inset shows the fast component (from Wright et al, 1985).

atoms lifetime spectra. Finally, positrons and $\alpha$-Ps reach thermal equilibrium with the gas and are each characterized by a decay constant.

A fundamentally different approach to the study of positron annihilation in gases has been developed by Surko and co-workers. The method, discussed in detail in chapter 1, uses a trapping technique to obtain a monoenergetic positron beam of 25meV resolution, which is then confined in a region of low-density gas (Greaves et al, 1994).

Measurements of the equilibrium annihilation rate parameter have been made for many gases over a wide density range. It has been found that although for small molecules $Z_{\text{eff}}$ is roughly of the order of $Z$, the annihilation rates for the larger organic molecules are much higher (Paul and Saint-Pierre, 1963; Surko et al, 1988; Iwata et al, 1995) than expected from a simple collision (Heyland et al, 1982; McEachran et al, 1977 and 1978). In table 4.1, the theoretical and experimental values of $Z_{\text{eff}}$ for the noble gases are shown. Traditional lifetime experiments have found $Z_{\text{eff}}$ to be density-dependent. The experimental data, derived from Coleman et al (1975), and theoretical results, extracted from McEachran et al (1978, 1979 and 1980) who performed a systematic calculation for the noble gases using the polarized-orbital approximation, are
in reasonable agreement. Van Reeth et al (1996) obtained the most accurate theoretical value up-to-date of 3.88 for He, which is in good agreement with the result reported by Coleman et al (1975). The value of $Z_{\text{eff}} = 320 \pm 5$ for Xe, obtained by Wright et al (1985), is in good accord with the results of Coleman et al (1975). However, it was suggested by Wright et al (1985) that this value might be underestimated due to incomplete thermalization of the positrons in this gas and high probability of epithermal annihilation. Although in the case of Ar, the values of $Z_{\text{eff}}$ measured using the traditional lifetime method and the positron trap method are in good agreement, Iwata et al (1995) found $Z_{\text{eff}} = 90$ for Kr, compared to $Z_{\text{eff}} = 64.6$ of Coleman et al (1975) and $Z_{\text{eff}} = 65.7$ of Wright et al (1985). This value is also higher than that obtained theoretically (McEachran et al, 1980).

<table>
<thead>
<tr>
<th>Gas</th>
<th>$Z_{\text{eff}}$ Experiment</th>
<th>$Z_{\text{eff}}$ Theory</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>$3.94 \pm 0.02^a$</td>
<td>$3.88 \pm 0.01^b$</td>
</tr>
<tr>
<td>Ne</td>
<td>$5.99 \pm 0.008^a$</td>
<td>$7.0 \pm 0.3^c$</td>
</tr>
<tr>
<td>Ar</td>
<td>$26.77 \pm 0.09^a$</td>
<td>$27.6^d$</td>
</tr>
<tr>
<td>Kr</td>
<td>$64.6 \pm 0.08^a$</td>
<td>$57.6 \pm 2.9^e$</td>
</tr>
<tr>
<td>Xe</td>
<td>$320 \pm 10^a$</td>
<td>$217 \pm 11^e$</td>
</tr>
</tbody>
</table>

Table 4.1. Experimental and theoretical values of $Z_{\text{eff}}$ for the noble gases at low densities and room temperature ($^a$ Coleman et al, 1975; $^b$ Van Reeth et al, 1996; $^c$ McEachran et al, 1978; $^d$ McEachran et al, 1979; $^e$ McEachran et al, 1980).

The temperature dependence of positron annihilation rates in the noble gases was first investigated by Kurz et al (1996) over a wide energy range from 0.025eV to 0.6eV. The experimental technique relied on positrons first being accumulated in a trap at room temperature, and then raising their energy by applying radio-frequency noise to one of the trap electrodes. The annihilation rate in a specific gas and the positron temperature were measured with time as the positrons cooled in the gas. In figure 4.3, the results for the annihilation rate in the noble gases are shown together with the theoretical predictions of McEachran (1977, 1978, 1979 and 1980). In general, there is a good
Figure 4.3. Positron annihilation rates on the noble gases, normalized to unity at room temperature: o, He; ●, Ne; ■, Ar; ▲, Kr; ○, Xe. Solid line, calculation of McEachran et al (1977, 1978, 1979 and 1980) for Ne, Ar, Kr, Xe and the dashed line for He.

agreement between the experimental results and the theoretical calculations, although deviations are noticeable in the case of Ar and Kr above about 0.15eV. For Xe, the experimental results are in good accord with the theory, however they stop earlier, at approximately 0.3eV, than was the case for the other noble gases. A trend is revealed in the relative variation of $Z_{\text{eff}}$, whilst for Xe $Z_{\text{eff}}$ drops by a factor of 5 over the energy range investigated, the corresponding variations for the other noble gases continue to decrease with their atomic number. The results of Van Reeth et al (1996) for He cannot be distinguished from experiment at the level of accuracy displayed by the experimental results of Kurz et al (1996).

The positron trap-technique was used to measure the annihilation rates of thermalized positrons in a variety of substances, including the noble gases, simple inorganic molecules, hydrocarbons, substituted hydrocarbons and aromatics (Iwata et al, 1995). $Z_{\text{eff}}$ values of several orders of magnitude greater than the number of electrons in the molecule, for example $Z_{\text{eff}} = 18000$ for benzene and greater than $10^6$ for anthracene, were observed. The data exhibited a number of chemical trends and illustrated the importance of electronic structure, symmetries and vibrational modes in
the annihilation process. The very high annihilation rates were thought to arise from pseudo-bound-states or resonances with the molecule, in which the positron is trapped in the vicinity of a molecule for much longer than the usual collision time.

It was discovered by Murphy and Surko (1991) that data for nonpolar molecules without double or triple bonds followed the empirical rule:

\[ \ln Z_{eff} = \frac{A}{E_i - 6.8eV} + B \]  

(4.1)

where 6.8eV is the binding energy of positronium and \( E_i \) is the ionization potential of the molecule. \( A \) and \( B \) are constants. Eq. 4.1 was interpreted by Murphy and Surko (1991) as suggesting a process in which a highly correlated positron-electron pair moved in the field of a positive ion.

From the theoretical point of view, it was shown that in order to describe positron-atom interactions accurately, the effect of virtual positronium formation had to be included in the theoretical description (Amusia et al, 1976; Basu et al, 1989 and 1990; Dzuba et al, 1993; Higgins and Burke 1993; Mitroy, 1994 and Gribakin and King, 1994). The fact that inclusion of virtual processes, such as positronium formation and electronic/vibrational excitations, into the positron-atom correlation potential greatly improves the agreement between the experimental and theoretical values for annihilation rates was investigated by Silva et al (1996), Dzuba et al (1996) and Gianturco et al (1999). Gribakin and King (1996) showed, using many body theory, that in the case of Mg virtual positronium formation terms, when added perturbatively to the polarization potential, could give sufficient extra attraction to support bound and resonant states of positrons with this atom. Dzuba et al (1996) suggested that the possible existence of such bound states and vibrational compound resonances might explain the observed high annihilation rates. However, Iwata et al (1997) concluded, from their annihilation rate measurements for deuterated and protonated alkanes, that the vibrational modes of molecules were not the dominant factors in determining the annihilation rates. It was also suggested by Iwata et al (1997) that low-energy positrons did not penetrate deep into the atom and mostly annihilate with the valence electrons.

The above arguments and the empirical scaling rule of Murphy and Surko (1991), described by eq. 4.1, prompted Laricchia and Wilkin (1997) to develop an alternative positron-trapping model based on the increasing significance of virtual processes as the threshold energy for real processes is approached. Specifically, it was assumed that a positron may form a virtual positronium atom below threshold and be trapped in the
vicinity of the target system for a time given by the uncertainty principle 
\( \Delta t \approx \hbar / |E - E_i + 6.8eV| \), during which it may annihilate with one of the electrons. The

value of \( Z_{\text{eff}} \) is then given by:

\[
Z_{\text{eff}} = \frac{Q_v}{\pi R_e^2 c} \left( \gamma \left[ 1 - \exp(-\lambda_d t_c) \right] + (1 - \gamma) \left[ 1 - \exp\left( -\frac{\Delta t}{t_c} \right) \left( \lambda_{sa} + \lambda_{po} \right) \right] \right)
\]  

(4.2)

where \( Q_c \) is the elastic scattering cross-section, \( \lambda_d \) and \( \lambda_{po} \) are the direct and pick-off annihilation rates, \( \lambda_{sa} \) is the spin-averaged annihilation rate of positronium and \( \gamma = \exp(-\Delta t / t_c) \), where \( t_c \) is the collision time.

Using crude estimates for some of the unknown variables, the model could reproduce the measured variation of \( Z_{\text{eff}} \) over several orders of magnitudes, its saturation with decreasing positronium formation threshold and the correct general trends found among different molecular families (Laricchia and Wilkin, 1997). It also predicted a quasiresonant enhancement of the annihilation probability as a function of the incident positron energy in the vicinity of energy thresholds for inelastic processes (Laricchia and Wilkin, 1998). This is illustrated qualitatively in figure 4.4, where the

![Figure 4.4. Qualitative illustration of the variation of the ratio \( R = Q_{\text{ann}}/Q_e \) of the annihilation to elastic collision cross-sections plotted versus the incident positron for H, He, H\(_2\), Xe and C\(_{14}\)H\(_{10}\). Also shown are the contributions to \( Q_{\text{ann}} \) arising from virtual positronium formation and in-flight annihilation for He (from Laricchia and Wilkin, 1998).](image-url)
contributions arising from direct annihilation and virtual positronium formation are plotted relative to the elastic collision cross-section in the case of He. Whilst the direct annihilation probability is seen to decrease as the incident positron energy is increased due to the reduced interaction time, as expected, the annihilation probability via the occurrence of a virtual process increases as the relevant threshold is approached. The maximum is reached at the threshold itself. The prediction of the model of Laricchia and Wilkin (1997 and 1998), namely the enhancement of the annihilation probability as the positron approaches the positronium formation threshold, was confirmed qualitatively by the theoretical calculations for H and He of Van Reeth and Humberston (1998), although the latter results reached infinity at $E_{p^+}$.

In figure 4.5.a, the effective number $Z_{\text{eff}}$ of electrons available for annihilation in $e^+ (E) + H(1s)$ process below $E_{p^+} (1s)$, as recently calculated by Igarashi et al (2002) using a hyperspherical close-coupling approximation is compared to other available theories. Below $E_{p^+}$, the results of Igarashi et al (2002) agree very well with the variational calculation of Van Reeth and Humberston (1998) and the momentum-space Lippmann-Schwinger calculation of Ryzhikh and Mitroy (2000). However, unlike the latter two theoretical treatments, that of Igarashi et al (2002) gives a finite $Z_{\text{eff}}$ at the threshold. As before, the agreement with the data of Laricchia and Wilkin (1997) is only qualitative. Figure 4.5b compares the partial-wave contributions to the annihilation cross-sections very close to the threshold. The dominant contribution to $Q_{p^+}$ comes from the S wave and there is only a minute contribution from L>0. As can be seen in the figure, the $Q_{\text{ann}}(S)$ bridges smoothly over $E_{p^+} (1s)$ to $Q_{p^+} (S)$. The result of Igarashi et al (2002) is in agreement with the calculation of Gribakin and Ludlow (2002) in the extremely narrow energy range shown in figure 4.5b, but not beyond it. It is concluded by Igarashi et al (2002) that direct annihilation dominates over the indirect annihilation via virtual positronium formation well below $E_{p^+}$ and that near $E_{p^+}$ it is meaningless to separate positronium formation from direct annihilation.

An alternative explanation to high annihilation rates for large molecules, which implies the existence of long-lived Feshbach-resonances, was proposed by Gribakin (2000). Two possible mechanisms of low-energy positron annihilation in binary collisions with molecules were considered, namely the direct and resonant annihilation. The direct annihilation described positron annihilation with small atoms or molecules,
as well as with molecules which did not form bound states with positrons. In such cases, values of $Z_{\text{eff}}$ of up to $10^3$ could be produced. The resonant annihilation applied when a positron formed a temporary bound state with a molecule with a positive positron affinity. The positron capture was considered as a resonant process where the energy of the positron was transferred into vibrational excitations of the positron-molecule complex. It was estimated that the resonant process could produce annihilation rates of up to $10^8$.

The recent measurements of Gilbert et al. (2002), as well as their continuation by Barnes et al. (2003), have been interpreted as being generally consistent with the model of Gribakin et al. (2000) and are believed to provide the first direct evidence that the large observed values of annihilation rates are due to excitations of long-lived Feshbach resonances of the positron-molecule complex. In figure 4.6, the annihilation rates for
butane, propane and ethane are shown as a function of incident positron energy. The prominent features of C$_4$H$_{10}$, C$_3$H$_8$ and C$_2$H$_6$ are the large peaks with $Z_{\text{eff}} = 23000$, 10500 and 900 respectively, occurring at energies some 20-30meV below the C-H stretching mode. The shift of peaks away from the vibrational threshold is considered to provide experimental evidence of the occurrence of Feshbach resonances. To confirm that the observed peaks were due to the excitations of the C-H vibrational stretch mode, measurements were taken with deuterated butane, which has the same electronic structure as butane. As shown in figure 4.6a, the entire spectra for butane and d-butane agree well when the d-butane spectrum is scaled by the square root of the ratio of the reduced masses associated with C-D stretch modes. The annihilation rate for 2,2-difluoropropane (C$_3$H$_6$F$_2$) is shown in figure 4.6b and illustrates that fluorination results in a lack of the resonant C-H peak.

**Figure 4.6.** Positron annihilation rate, $Z_{\text{eff}}$, as a function of positron energy for: (a) butane, (b) propane, and (c) ethane. Vertical bars along the x-axis indicate the strongest infrared-active vibrational modes. Arrows on the y-axis indicate $Z_{\text{eff}}$ for Maxwellian distribution of positrons at 300K (from Gilbert et al, 2002).
Since the shifts in the energy, that are believed to demonstrate the occurrence of Feshbach resonances, for butane and propane are small (up to 30meV and in the case of ethane the peak occurs at the threshold itself), the above interpretation relies strongly on a correct energy determination of the positron beam. Although Gilbert \textit{et al} (2002) did not provide details on the energy calibration method used, several approaches dealing with this problem were discussed by Sullivan \textit{et al} (2002b). They included relative energy determination methods, such as using a gas cell as an analysing element, and an absolute one, which relied on measuring the time-of-flights of positrons in the gas cell. These different calibration techniques resulted in a discrepancy of 60meV, which was taken by the authors as an estimate of the uncertainty on the absolute energy of the positron beam.

4.3. Determination of the energy dependence of the annihilation cross-section for Ne, Xe and O\textsubscript{2} below positronium formation threshold

4.3.1. Modifications to the experimental apparatus and data acquisition

The apparatus described in chapter 2 has been employed in this study with an additional $\gamma$-ray detector placed on the opposite side of the scattering gas cell. A schematic of the apparatus is depicted in figure 4.7, where additional lead shielding placed underneath and around the $\gamma$-ray detector, as well as prior to the gas cell and at the position of the positron detector to prevent the stray radiation is also shown.

In figure 4.8, the electronic circuit used for the present measurements is shown. Signals from both ion and $\gamma$-ray detectors were amplified tenfold and fed to two Ortec 584 constant fraction discriminators. A conventional delayed-coincidence circuit was

\textbf{Figure 4.7.} The apparatus used for the measurement of the annihilation cross-sections.
set up between the γ-ray (start) and delayed ion (stop) signals using an Ortec 467 time-to-pulse height converter. In the case of Ne, such coincidence spectra were recorded directly on a multichannel analyser (MCA) for a given energy of the positron beam. In order to allow for ion mass selection and to reduce further the background a window was set on the MCA around the γ-ion coincidence peak and this was fed to a multichannel scaler (MCS 1). An external ramp was used to control the MCA, providing the advance pulse for increasing in steps the voltage of the moderator and, correspondingly, of the Wien filter. In this way, the energy dependence of the signal could be determined across a range of energies in a single run. In the case of Xe and O₂, \( Q_{\text{ann}} \) below \( E_{Pz} \) was obtained in this manner. For Ne, the measurements were repeated with this method to check the agreement between the two determinations. The ion count rate was also measured simultaneously with the coincidences using a second multichannel scaler (MCS 2).

**Figure 4.8.** Electronic circuit used for the measurement of the annihilation cross-sections.
4.3.2. Experimental method and systematic effects

The measurement of the energy dependence of the annihilation cross-section below and in the vicinity of $E_{ps}$ poses a number of difficulties arising from possible systematic effects, which need to be considered and suitably accounted for. Firstly, the fact that the positronium formation cross-section increases very rapidly at the Ps formation threshold means that the annihilation signal may be swamped out by the positronium formation due to poor beam energy resolution near $E_{ps}$, as the signal measured for annihilation is identical to that resulting from Ps formation (i.e. detection of a γ-ray and an ion in coincidence). Thus a high-energy resolution (i) as well as an absolute energy determination (ii) are required. With a weak DC electric field present in the gas cell, any accelerating effect on the positron beam needs to be evaluated (iii). Furthermore, in the present work, diffusion pumps are used for system evacuation and a signal may arise from residual gases and contaminants in the vacuum system (iv), especially since oil vapours are known to have very high annihilation rates (Iwata et al, 1995 and references therein). It is also important to note that, in general, slow positron beams contain a small number of epithermal positrons\(^1\) (v), which can themselves produce a non-negligible background via positronium formation. Lastly, a signal might also arise due to the release of secondary electrons, and ions from those, from positron impact annihilation on the gas cell apertures. A similar effect would also be expected if positrons elastically scatter from the target gas and hit the walls of the gas cell. Hence, the effect of the strength of the magnetic field in the gas cell on the measured coincidence count rate was studied (vi). The systematic effects (i - vi) are described in detail in the following sections.

(i) Energy resolution of the positron beam

Figures 4.9(a) and (b) show the energy distributions of the positron beam with a moderator voltage ($V_m$) of 3V and 10V, respectively. These were measured at the end of the beam-line by applying a retarding voltage to grids R3 (shown in figure 4.7) in front of the positron detector. In order to improve the energy resolution, successive portions of the beam were biased off by applying different voltages ($V_{R1}$) to the positron repeller (R1): $V_{R1} = V_m + \alpha$, where $\alpha = 1.5\text{V}$ or $2\text{V}$, corresponds to the slow positron beam

\(^1\) In this work, no distinction is made between epithermal (not fully moderated) and fast (unmoderated) positrons.
Figure 4.9. The measured energy distribution of the positron beam. $V_m$ and $V_{R3}$ are the potentials applied to the moderator and R3, respectively. The red arrow indicates a cut-off voltage.

being “switched on” and $V_{R1} = V_m + 3$ corresponds to the slow positron beam being stopped at R1 (i.e. only fast positrons are transmitted). From the energy distributions depicted in figure 4.9a, the position of the peak ($U_{\text{max}}$) and the energy spread ($\Delta E$) of the positron beam were determined. Whilst the distribution for $V_{R1} = V_m + 2$ was found to be approximately symmetric about $U_{\text{max}}$, that for $V_{R1} = V_m + 1.5$ was skewed towards higher values of the voltage differences. The cut-off voltage ($V_{\text{co}}$), obtained from more precise measurements shown in figure 4.9b, was defined as the voltage above which the contributions from differences of $(V_{R1} = V_m + \alpha) - (V_{R1} = V_m + 3)$ added up to zero within errors. As indicated by the arrow in the inset, $V_{\text{co}}$ was determined to be at $(3.2 \pm 0.1)$V above the value of $V_m$. From figure 4.9a, the position of the peak relative to the cut-off voltage is found to be $V_{\text{co}} - U_{\text{max}} = (0.6 \pm 0.1)$ eV for $V_{R1} = V_m + 1.5$ and $V_{\text{co}} - U_{\text{max}} = (0.5 \pm 0.1)$ eV for $V_{R1} = V_m + 2$. These values will be used, in part (iii) of this section, in the final determination of the positron beam energy.

In the case of $V_{R1} = V_m + 1.5$, $\Delta E \approx 1$ eV at full width at half maximum (FWHM) whilst for $V_{R1} = V_m + 2$, $\Delta E \approx 0.5$ eV. Since reducing the beam energy spread decreases the intensity of the positron beam (e.g. from $\Delta E \approx 1$ eV to $\Delta E \approx 0.5$ eV by over a factor of 3), a compromise was sought between an acceptable energy resolution of the beam
and its intensity. For Ne, where the Ps formation threshold is relatively high (14.76eV) and a positive coincidence signal was detected some eV below $E_{Ps}$, initially only a few experimental points with high precision were obtained. Thus a larger step of 3.7V in $V_m$ and a positron beam with $\Delta E \approx 1$ eV were chosen. In the case of Xe and O$_2$, the Ps formation thresholds (5.33eV and 5.27eV, respectively) are significantly lower than for Ne. Furthermore, in the case of O$_2$ there is an electronic excitation threshold ($E_{ex} = 4.48$eV) at 0.8eV below $E_{Ps}$. The proximity of these two inelastic thresholds required a smaller energy spread for O$_2$ than in the case of Ne. Because of these considerations, and the fact that in the case of Xe and O$_2$ the initial measurements did not show strong evidence of positive coincidence signal until fairly close to $E_{Ps}$, a beam of $\Delta E \approx 0.5$ eV was used.

(ii) Absolute energy calibration of the positron beam

In order to assign an absolute energy scale to the present measurements, a time-of-flight technique was used. For this purpose, an additional grid was installed in front of the moderator, to which a square-wave pulse was applied, to chop the positron beam. All the potentials between this grid and the positron beam detector were grounded. The grid was held at $(V_m + 3.5)V$ in order to stop the beam. The chopped signals were obtained from a pulse generator to trigger a rapid drop of 3V on the grid potential, for a period of ~0.1µs every 7µs, and to simultaneously start a time-to-amplitude converter (TAC). The TAC was subsequently stopped by the detection of positrons arriving at the detector at the end of the beam-line. The MCA channel (C), multiplied by the time per channel ($\tau$), in which the signal began to rise above the background level was taken to correspond to the flight time ($t$) of the fastest positrons:

$$C \ast \tau = t = t_0 + \frac{l \sqrt{m}}{\sqrt{2E}} + \frac{l \sqrt{m}}{\sqrt{2(eV_m + \phi_s)}} = t_0 + \frac{a}{\sqrt{(eV_m + \phi_s)}}$$

(4.3)

where $E$ and $m$ are the energy and mass of the positron, respectively, and $l$ is the unknown length of the flight path. $(eV_m + \phi_s)$ is the upper limit of the energy distribution, where $\phi_s$ is the magnitude of the positron work function corrected for any contact potential effects and is an unknown quantity. The time zero ($t_0$) was obtained by chopping the beam just in front of the positron detector at the highest $V_m$ considered in

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order to reduce the transit time to a minimum and $a = t\sqrt{m}/\sqrt{2}$. From eq. 4.3 it follows:

$$\frac{1}{(t-t_0)^2} = \frac{eV_m}{a^2} + \frac{\phi_s}{a^2}. \quad (4.4)$$

By measuring $t$ for various values of $V_m$ and fitting the results to eq. 4.4, $\phi_s$ was found to be equal to $(2.4 \pm 0.3)$eV. This value corresponds to the flight path of $(1.9 \pm 0.1)$m, which is approximately consistent with the physical length of the apparatus.

(iii) Beam perturbation due to weak static electric field

During normal operation, the positron beam within the scattering gas cell is perturbed by the weak static electric field produced by the ion-extracting lens. A computer simulation has been used to obtain the mean perturbation voltage on the positron beam and its energy distribution within the gas cell. More specifically, for simplicity, the beam cross-section has been considered to be a square divided into 25 elements, as shown schematically in figure 4.10. The chamfered corners of the squares represent the fact that these elements have been given a weight of 0.5 in an attempt to approximate a circular beam cross-section. The electric potential due to the extracting lens has been calculated at the centre of each element for differing positions of the cross-section along the y-axis, as indicated in the figure 4.10. The obtained values of the electric potential have been divided into 0.5V bins and summed, thus providing the

![Figure 4.10](image)

**Figure 4.10.** Schematic representation of the positron beam cross-section used to simulate the mean energy of the beam within the scattering gas cell with a potential applied to the ion-extracting lens (not to scale).
perturbation on the positron beam in the gas cell due to the accelerating voltage applied to the lens, as shown in figure 4.11. The ion extraction efficiency and the trajectories of ions within the gas cell were also simulated, as discussed in chapter 2. It was found that ions are extracted from only the central half volume of the gas cell. This effect was taken into consideration in the calculation of the mean energy and distribution of the positron beam within the scattering gas cell. It is remarked that had the whole cell been considered, the estimated perturbation on the positron beam would have been smaller.

Due to the linear magnification \( M \) of the centre of the orbital motion \( M = \sqrt{B_i / B_f} \), where \( B_i, f \) are the initial and final magnetic field magnitudes (Kruit and Read, 1983), the radius of the positron beam as it enters the scattering gas cell is estimated to reduce from 8mm (the diameter of the apertures) to 7.2mm for \( B_f = 90 \) Gauss and to 6mm for \( B_f = 130 \) Gauss. Thus, allowing for different magnetic fields strengths and corresponding beam radii, it is estimated that the electric field perturbation results in the positron beam being accelerated on average by \( V_{per} = 0.2 \) V and its energy further broadened by less than 0.1 V with less than 4% of the positron beam being accelerated on average by 0.4V, the highest value possible for the present geometry being 0.54V.

Finally, combining the results discussed above, the energy calibration is given by

\[
E = eV_m + e\left[\phi_e - (V_{co} - I_{max})\right] + V_{per}
\]

where \( e \) is the charge of an electron, \( V_m \) is the
potential applied to the moderator, $\phi_r$ is the magnitude of the work function obtained in section (ii), $(V_{oc} - I_{max})$ gives the position of the peak in the positrons beam distribution relative to the cut-off voltage and $V_{per}$ is the perturbation voltage due to the ion-extracting lens. Therefore, the energy equals $(V_m + 2.1)eV$ in the case of $V_r = V_m + 2$ and $(V_m + 2)eV$ for $V_r = V_m + 1.5$, with an uncertainty of ±0.3eV.

(iv) Annihilation signal from residual gases and gas contaminants

With a system base pressure of $3 \times 10^{-6}$ Torr and the use of diffusion pumps for evacuation, the possible signal arising from annihilation from residual gases in the vacuum chambers (e.g. oil vapours) was explicitly measured and subtracted. It was observed that the contribution from the net signal due to annihilation of slow positrons on residual gas was in most cases zero within errors and that the energy dependence of the gross signal was flat, thus considerably different from the net signal obtained in gas, especially close to relevant $E_{p_r}$. In principal, a signal could also arise from annihilations on contaminants present in the high-purity gases (99.994%, 99.993% and 99.9995% for Ne, Xe and O$_2$, respectively) investigated. However, as discussed in detail in the appendix, this has been estimated to be at least two orders of magnitude smaller than the signal observed below $E_{p_r}$ and thus considered to be negligible.

(v) Epithermal positron contribution

The contribution to the beam from epithermal positrons, extracted from the measurements of positrons emitted from a tungsten re-moderator at 3kV incident energy (Goodyear et al, 1994), has been found to be less than 1% and is expected to be negligible for $\beta^+$ particles emitted from a $^{22}$Na source (Hamilton et al, 1958). Nevertheless, in the present work, the contribution from epithermal positrons was measured explicitly with $V_{R1} = V_m + 3$ both in gas and vacuum. These contributions were then subtracted from associated measurement in gas and vacuum with $V_{R1} = V_m + \alpha$.

More specifically, time-normalized coincidence count rates by slow positrons in gas, $S_g(E)$, and vacuum, $S_v(E)$, were obtained with a voltage of $V_{R1} = V_m + \alpha$, $S_{g,v}(\alpha)$ and $V_{R1} = V_m + 3$, $S_{g,v}(3)$, applied to R1, where $\alpha = 1.5$ in the case of Ne and $\alpha = 2$ in the case of Xe and O$_2$, as follows:
\[ S_g(E) = S_g(\alpha) - S_g(3) \frac{P_aN_g^a(3)}{P_3N_g^3(3)} \quad (4.5a) \]

\[ S_v(E) = S_v(\alpha) - S_v(3) \frac{N_v^a(3)}{N_v^3(3)} \quad (4.5b) \]

where \( N_g^a(3) \) and \( N_v^3(3) \) correspond to the time-normalized intensities of epithermal positrons \textit{measured in vacuum} with \( V_{R1} = V_m + 3 \), associated with \( S_g(\alpha) \) and \( S_g(3) \) runs, respectively. Here \( p_a \) and \( p_3 \) are the target gas pressures associated with \( S_g(\alpha) \) and \( S_g(3) \) runs, respectively. Subsequently, the coincidence yields, \( Y(E) \), were obtained according to:

\[ Y(E) = \frac{1}{p_a} \left( \frac{S_g(E)}{N_g} - \frac{S_v(E)}{N_v} \right) \quad (4.6) \]

where \( N_g = [N_g^a(\alpha) - N_g^a(3)] \) are the slow positron beam intensities, \textit{measured in vacuum}, associated with \( S_g(\alpha) \) and \( S_v(\alpha) \), respectively. They were obtained by subtracting the positron beam intensities measured with \( V_{R1} = V_m + 3 \) (the whole slow positron beam being stopped at R1) from those measured with \( V_{R1} = V_m + \alpha \). The coincidence yield, as given by eq. 4.6, below \( E_{ps} \) is directly proportional to the annihilation cross-section and above \( E_{ps} \), to the sum of the positronium formation and annihilation cross-sections.

\( \text{(vi) Dependence of the coincidence yields on the strength of the magnetic field} \)

The possible dependence of the coincidence yields upon the strength of the magnetic field \( (B) \) has been investigated since insufficient positron confinement after scattering could lead to impact annihilation on the cell apertures, which if accompanied by any secondary electron emission and consequent production (and extraction) of ions, could have contributed to the measured coincidence yield, as discussed by Laricchia \textit{et al} (2002). Thus, in this work for the targets under investigations, different magnetic fields strength were used. In the case of Ne, three different values of the magnetic field \( B = 40, 67 \) and 93 Gauss were employed whilst for \( O_2 \) \( B = 67 \) and 93 Gauss and for Xe \( B = 93 \) and 130 Gauss, as summarized in table 4.2. It was expected that as the magnetic field was increased, the coincidence count rate would decrease if annihilation on the cell apertures was a significant effect. Therefore, extrapolation to infinite magnetic field...
strength would give the value of the coincidence yields due to annihilation with the target gas alone. However, no decrease in the coincidence count rates or a change in its energy dependence was found as a function of the $B$ field strength. This can be explained by the fact that, as mentioned above, ions are only extracted from the central half volume of the gas cell. Therefore, ions produced by impact annihilation near the apertures would not be detected. It has thus been concluded that, for all the magnetic field strengths considered and in the energy region investigated, no signal arose due to positron annihilation on the cell apertures and the weighted means of all the runs were taken in the final analysis.

In the case of Ne, it was noticed that the strength of the magnetic field changed the ion extraction efficiency. The dependence of the extraction efficiency on the strength of the magnetic field was simulated using the Simion 6.0 program, as discussed in chapter 2. Figure 2.12 shows the extraction efficiency progressively decreasing by approximately 20% over the range from 0 to 100 Gauss, rapidly falling above 100 Gauss to reaching a few per cent at approximately 200 Gauss. Thus the highest magnetic field strength employed in the case of Ne was 93 Gauss and a correction was applied to data measured at each energy with different magnetic fields to account for this effect. In the case of Xe and O$_2$, the correction for runs with different $B$ was not necessary as, at each $B$ field, all energies were measured in one scan and the extraction efficiency is not dependent on the positron beam energy. For Ne, measurements were also repeated in the same manner as it was done for Xe and O$_2$ and a good agreement was found between the two methods. These results will be discussed in the following section.

### Table 4.2. Magnetic field strengths used for Ne, Xe and O$_2$.

<table>
<thead>
<tr>
<th></th>
<th>Magnetic Flux Density (Gauss)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ne</td>
<td>40, 67 and 93</td>
</tr>
<tr>
<td>Xe</td>
<td>93 and 130</td>
</tr>
<tr>
<td>O$_2$</td>
<td>67 and 93</td>
</tr>
</tbody>
</table>
4.3.3. Results and discussion

In figures 4.12a and 4.12b, $Q_{p^+}$ of Laricchia et al (2002) for Ne and Xe (black hollow squares), measured with an energy resolution of $\Delta E \leq 0.4\,\text{eV}$ and a pulsed electric field, are shown. These data have, respectively, been fitted with third- and fifth-order polynomials, also shown in the figures as black curves. Laricchia et al (2002) corrected their energy scale for contact potential effects by imposing the threshold law of Wannier (1953) and choosing the intercept on the abscissa at $E_{p^+}$. These results, as well as all the other previous measurements, assume any signal below $E_{p^+}$ to be background.

The polynomial fits of $Q_{p^+}$ for Ne and Xe serve a dual purpose: firstly, they are used to normalize the present coincidence yields; secondly, they will be used to obtain the expected near-threshold behaviour of $Q_{p^+}$, by convoluting them with our energy resolution, in order to deduce the influence of positronium formation on the signal apparently below $E_{p^+}$. In the polynomial fits, it has been assumed that $Q_{p^+}$ is equal to zero at the threshold. However, there is negligible change when either the s-wave dependence of $Q_{p^+}$ near $E_{p^+}$ is imposed according to the Wannier threshold law or when the threshold value is set to non-zero, as suggested by the hyperspherical close-coupling calculations of Igarashi et al (2002) for H. Two threshold values were used in this test, namely $5.6 \times 10^{-20}\,\text{cm}^2$ from Igarashi et al (2002) and, ten times this value for Xe (Gribakin and Ludlow, 2002).

Looking at figure 4.12b, the coincidence yields for Xe (black solid circles) have been normalized at 7.4eV to the fit of $Q_{p^+}$ of Laricchia et al (2002). However, it was noted that the energy dependence of $Q_{p^+}$ of Laricchia et al (2002), if shifted in energy by +0.2eV (blue hollow squares), agrees considerably better with the present measurements, again normalized at 7.4eV (blue solid circles). This suggests the possibility that the present calibration might have overestimated the absolute energy of the positron beam. Therefore, we have examined the consequence of this possibility also in the case of Ne, as shown in figure 4.12a. Whilst for Ne the two determinations are rather similar near $E_{p^+}$, for Xe they differ significantly: the fit to the actual data of Laricchia et al (2002) is much steeper near the threshold than the fit to the same data shifted in energy by +0.2eV.

\footnote{Comparison should be made between data points and curves of the same colour.}
Figure 4.12. Normalization of the present $Q_{ann}$ below $E_R$, and above it, of the sum of $Q_{ann} + Q_{ps}$.
In the case of $O_2$, a different normalization procedure has been employed. The present total ion yields (measured simultaneously to the coincidence yields) have been chosen for normalization, since a sufficient number of points is available above $E_{Ps}$ and, more importantly, since the energy calibration is (obviously) identical for each data set. The absolute scale for the present ion yields has been assigned by calculating the average value of the points at 6.7eV and 6.8eV and normalizing it to the value of the point at 6.7eV from $Q'_p$ of Laricchia et al (1993), which were obtained with an energy resolution of $\Delta E \approx 1$ eV. The coincidence yields have then been normalized to the fit of total ion yields. The solid line represents a third-order polynomial fit to the preset total ion yields with the threshold value set to zero. It is noted that $Q'_p$ are shifted with respect to the present total ion yields by $+0.2$eV, with the sign of the shift being opposite to that observed for Ne and Xe. However, taking into account that the energy calibration for the cross-section of Laricchia et al (1993) ignored contact potential effects, only the present energy calibration will be considered in the discussion that follows.

The near-threshold behaviours of the positronium formation cross-sections for the various targets have been determined by convoluting the energy profiles of the beam with the corresponding fits to $Q_{Ps}$ shown in figure 4.12. In figure 4.13, the two field-free energy distributions of the positron beam (obtained after subtracting the background) can be seen to be described fairly well by a Gaussian and by a five-parameter Weibull fit. The final positron energy profile, used in the convolution, combines the Gaussian distribution for Xe and $O_2$ and the Weibull distribution for Ne with the perturbed profile within the scattering cell due to the ion-extraction lens.

Figure 4.14 displays our final results for the energy dependence of $Q_{ann}$ (below $E_{Ps}$) and the sum of $Q_{Ps}$ and $Q_{ann}$ (above $E_{Ps}$), together with the expected behaviours of $Q_{Ps}$ for Ne, Xe and $O_2$. The error bars combine the uncertainties on the weighted

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3 As discussed in subsection v, the contribution from epithermal positrons is measured and subtracted explicitly.

4 $y = y_0 + a \left( \frac{c-1}{c} \right)^{1+c} \frac{x-x_0}{b} + \left( \frac{c-1}{c} \right)^{c-1} e^{-\left[ \frac{x-x_0}{b} \left( \frac{c-1}{c} \right)^c \right]} + \frac{c-1}{c}$, $x>x_0$ and $a, b, c>0$

5 Again, comparison should be made between data points and curves of the same colour.
means of different measurements, the statistical errors arising from the counting techniques and the uncertainties on the target gas pressure. Tests were also carried out to verify that at all energies the signal increased linearly with the target pressure, thus confirming single collision conditions. Moreover, errors of ±7%, ±8% and ±20% are assigned in the absolute scale for Ne, Xe and O₂ respectively, due to the uncertainty of ±0.3eV in the present absolute energy calibration, as discussed in subsection (iii). It should be mentioned that all of the present \(Q_{\text{ann}}\) might be underestimated by up to 40% below the Ps formation threshold due the different counting efficiencies below and above \(E_{Ps}\), since for positronium formation 75% of the annihilations give rise to three \(\gamma\)-rays (from o-Ps), as opposed to the two \(\gamma\)-rays below \(E_{Ps}\) from direct annihilation. The correction due to this change in the detection efficiency is difficult to apply due to its unknown variation near \(E_{Ps}\), where the positronium formation signal is smeared by the experimental energy resolution. However, as discussed below, the uncertainties
Figure 4.14. The energy dependence of $Q_{ann}$ below $E_{Ps}$ and $Q_{Ps} + Q_{ann}$ above $E_{Ps}$. The vertical dotted lines indicate the positronium formation thresholds ($E_{Ps}$) and the vertical dot-dashed line the electronic excitation threshold ($E_{ex}$) for $O_2$. 
associated with the different normalizations do not affect the conclusions, which can be
drawn from the present results and, indeed, the possible energy overestimation and
magnitude underestimation below $E_{ps}$, imply that the present results are conservative
estimates of $Q_{ann}$ below $E_{ps}$.

In the case of Ne, for which the two expected $Q_{ps}$ curves exhibit the same
behaviour near $E_{ps}$, a positive signal is clearly seen from as much as 9eV below $E_{ps}$.
As shown in figure 4.15, these results have been confirmed by measurements performed
with a constant magnetic field (in the same manner as for Xe and O$_2$) to check the
correction applied for the magnetic field dependence of the ion extraction efficiency (as
discussed in section 2.5). For Xe, taking into consideration the uncertainty in the energy
calibration and the discrepancy between the fits representing the two $Q_{ps}$, there is little
evidence of an increase in the cross-section, as shown in figure 4.14b. In the case of O$_2$,
as shown in figure 4.14c, the cross-section increases at $(4.5 \pm 0.3)$eV, close to the
electronic excitation threshold. From around $(0.7 \pm 0.3)$eV below $E_{ps}$ onwards, the
present results lie significantly above those expected from the convoluted fit of $Q_{ps}$. It

![Figure 4.15. The energy dependence of $Q_{ann}$ below $E_{ps}$ and, above it, of $Q_{ann} + Q_{ps}$ for Ne.](image)

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is remarked that the adoption of the energy scale of the data of Laricchia et al (1993) would result in the onset at $E_x$ being shifted to $(0.5 \pm 0.3)$ eV below $E_p\!$. This would still be significantly below the positronium formation threshold but noticeably above the electronic excitation threshold for this target. On the other hand, the consequence of a possible overestimation of the present energy scale, as suggested by the data of Laricchia et al (2002) for Xe, has been examined also for O$_2$. In such a case, a more pronounced increase would be observed starting at approximately $(0.9 \pm 0.3)$ eV below $E_p\!$.

Two possible concerns about the increases of $Q_{\text{ann}}$ below $E_p\!$, in the case of Ne and O$_2$, might be that they could be due to an underestimation of the accelerating effects of the positrons by the electric field of the lens within the gas cell and/or of epithermal positrons in the high-energy tail of the beam distribution. The latter was tested by simulating what positron beam profile with a non-zero high-energy tail would be required to explain the increase in $Q_{\text{ann}}\!$. This beam profile was then convoluted with relevant $Q_{p\!}$. In the case of O$_2$, it was found that a tail extending up to 1 eV above the measured cut-off energy of the positron beam would provide an explanation for an increase in $Q_{\text{ann}}\!$, whereas for Ne a tail extending up to 9 eV is required. These findings are inconsistent since the high-energy tail (and thus the contribution from epithermal positrons) would not change with different beam energy resolutions. Similarly, the perturbation of the positron beam is dependent upon the lens potential (kept constant throughout these measurements) and not the gas investigated and should, therefore, result in the same effect for all targets, which is in contradiction with observation. Nevertheless, work with a pulsed electric field is planned in order to remove the additional uncertainty that the weak static electric field in the scattering gas cell introduces to the present absolute energy calibration.

Gribakin and Ludlow (2002) have predicted an enhancement of $Z_{\text{eff}}$ in Xe at 0.1 eV below $E_p\!$ of approximately 10 times that in H. Our $Q_{\text{ann}}$ cannot verify an increase at such close proximity to the threshold due to insufficient energy resolution. However, there is an indication of an increase in $Q_{\text{ann}}$ of Xe at approximately 5 eV but this is confined just to one datum and is, therefore, viewed with caution. Ludlow and Gribakin (2002) have also investigated the effect of the finite energy-spread of the positron beam on the threshold behaviour of the annihilation cross-section and
suggested that major increases in resolution ($\Delta E < 20 \text{ meV}$) are needed in order to resolve near-threshold enhancements of the annihilation cross-sections. The present results for Ne and O$_2$ indicate that this might not be necessary at least for these targets. The rise of $Q_{\text{ann}}$ for O$_2$ in the vicinity of the threshold for the electronic excitation raises the possibility that this channel might be contributing to the annihilation cross-section below $E_{p_s}$. It is noted, however that, in such a case, the rise would be expected to occur from below $E_{e_x}$, as discussed by Laricchia and Wilkin (1997 and 1998) and by the recent work of Yamanaka et al (2003), which predicts an enhancement in the direct annihilation cross-section for H well above $E_{p_s}$, due to temporary positron capture simultaneous to an electronic excitation. In view of the uncertainties in our energy scale, further investigations in this region are needed.

4.4. Conclusions

In this chapter, the energy dependence of the annihilation cross-sections below $E_{p_s}$ for Ne, Xe and O$_2$ has been investigated by measuring the coincidences between $\gamma$-rays and ions. Despite the relatively wide energy-spread of the positron beam and the uncertainty in the absolute energy calibration, there is evidence of a non-negligible annihilation cross-section for Ne significantly below $E_{p_s}$. In the case of Xe, no clear increase below $E_{p_s}$ is apparent, although more data points with a finer step in this region would be useful. For O$_2$, where the electronic excitation threshold precedes the positronium formation threshold, an onset in $Q_{\text{ann}}$ is observed around $E_{e_x}$. Undoubtedly further experimental investigations of annihilation performed with a smaller energy spread are required, especially in the case of atomic targets where complications arising from the presence of the vibrational excitations would be avoided. In this respect, measurements with a He target would prove particularly desirable, since several theoretical results for this target are available.
CHAPTER 5
DIRECT MEASUREMENTS OF THE
POSITRONIUM FORMATION CROSS-SECTIONS IN HE, AR AND XE

5.1. Introduction

The status of the determination of positronium formation cross-sections ($Q_{Ps}$) for the noble gases has been discussed in detail in chapter 3. An encouraging degree of convergence has been achieved for $Q_{Ps}$, especially in the case of Ne and Ar between the data of Laricchia et al (2002) and Jin et al (1994) and also, in the case of Ar, on the position and the magnitude of the first peak between the data of Laricchia et al (2002) and Sullivan et al (2002b). The very recent determinations of $Q_{Ps}$ by Laricchia et al (2002) have revealed an interesting feature, in the form of a two-peak structure, with the second peak being attributed to the positronium formation in an excited state. An indication of a similar structure has been observed in $Q_{Ps}$ of Gilmore et al (2002), when an excited positronium formation channel had been added to the calculation.

In this chapter, the work performed to obtain $Q_{Ps}$ for He, Ar and Xe is described. This includes a detailed analysis of the energy dependence of the positronium detection efficiency for the geometry of the present gas cell.

The experimental approach relies on measuring coincidences between $\gamma$-rays from direct/positronium annihilation and the target ions. This signal is unique of either (i) direct annihilation ($e^+ + A \rightarrow 2\gamma + A^+$) or (ii) positronium formation ($e^+ + A \rightarrow Ps + A^+$). As described in detail in chapter 4, below $E_{Ps}$, the experimental set-up allows the study of (i). Above it, information on the sum of processes (i) and (ii) can be obtained. It is assumed that above $E_{Ps}$, positronium formation is dominant and thus detecting the coincidence yields is a direct method for obtaining positronium formation cross-sections.
5.2. Experimental method and systematic effects

The experimental method and the apparatus have been described in detail in chapter 4. Measurements of the total ion yields have been measured simultaneously with the coincidence yields, $Y_{\text{coi}}$, which are expressed as:

$$Y_{\text{coi}} = \frac{1}{p} \frac{N_{\text{coi}} - B_{\text{coi}}}{N_{e^*} - B_{e^*}}$$

(5.1)

where $N_{\text{coi}}$ and $N_{e^*}$ refer to the time-normalized coincidence and incident beam counts respectively, $B_{\text{coi}}$ and $B_{e^*}$ correspond to their associated backgrounds and $p$ is the target gas pressure in the scattering gas cell. In the present determination, $B_{\text{coi}}$ have been obtained by calculating the average coincidence signal below the positronium formation threshold and subtracting it from the data above $E_{p_s}$. In other words, as in all other experimental determinations (see summary in e.g. Laricchia et al, 2002), the annihilation cross-section is assumed to be negligible above $E_{p_s}$. As discussed in chapter 4, whilst in the case of Xe no appreciable annihilation coincidence signal has been found below $E_{p_s}$, its contribution in Ne, as compared to the positronium formation cross-section 1eV above $E_{p_s}$, is of the order of 8%.

Tests were carried out to ensure that a single collision condition was maintained throughout the measurements. For Ar and Xe, pressures were adjusted so that not more than 10% of the incident positron beam was attenuated in the gas cell. In principle, the small cross-section of He preserves single collision conditions to much higher pressures than in the case of Ar and Xe, however due to the limitations of the system to pump He atoms effectively, maximum attenuations of approximately 5% of the positron beam were achieved.

In figure 5.1, the present results for the total ionization cross-section for Ar, obtained by normalizing the present total ion yields at the highest measured energies to the cross-section of Laricchia et al (2002), is shown. The present measurements were performed with the full-energy spread of the positron beam ($\Delta E \leq 2.5\text{eV}$) and those of Laricchia et al (2002) with a reduced energy-spread ($\Delta E \leq 0.4\text{eV}$) for energies below 40eV. The ion extraction method employed in the two measurements were also different: the present results were obtained with a DC field and those of Laricchia et al (2002) with a pulsed electric field. A consensus is found between the two sets of data with differences being not larger than 5%. A good agreement is also found in the

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present total ionization cross-sections and those of Laricchia et al (2002) for Xe and Moxom et al (1995) for He. The quality of this agreement gives confidence in the performance of the system.

The strength of the magnetic field in the gas cell is an important factor in this type of measurement because of the possibility that, at higher incident energies, the magnetic field might fail to confine positrons elastically scattered at large angles. If these positrons hit the walls of the gas cell, the release of secondary electrons (which could ionize the target gas) and γ-rays from annihilation could result in coincidences not related to positronium formation. Therefore, the variation of the energy dependence of the coincidence yield with different strengths of the magnetic field was investigated for He and the results, normalized in the peak region, are shown in figure 5.2. It can be seen that the coincidence yield obtained with the lowest magnetic field of 110 Gauss starts rising above the ones obtained with the higher magnetic fields at approximately 70eV. However, the energy dependencies of the coincidence yields measured with magnetic fields of 130 and 150 Gauss appear to agree over the common energy range. This indicates that for He, within the present experimental limitations on the strength of the magnetic field, a converged energy dependence of the positronium formation
cross-section can be obtained up to 70eV above $E_{Ps}$. This energy is expected to shift to lower values with increasing static interaction for heavier targets, which is expected to result in a progressively broader angular distribution of the scattered projectiles (Paludan et al, 1997).

5.3. Energy dependence of the positronium detection efficiency

A detailed analysis has been performed to investigate the energy dependence of the positronium detection efficiency for the geometry of the present gas cell. For the purpose of the present analysis, it has been assumed that p-Ps decays instantaneously emitting 2 $\gamma$-rays and has a detection efficiency $\varepsilon_{2\gamma}$. If o-Ps reaches the walls of the gas cell, it is assumed to quench. Defining $P$ as the probability of o-Ps reaching the walls of the gas cell, the probability of o-Ps decaying in flight before reaching the walls is $(1-P)$. Whilst quenching on the walls is associated with the same detection efficiency of $\varepsilon_{2\gamma}$ as p-Ps, o-Ps decaying in flight has a detection efficiency of $\varepsilon_{3\gamma}$. It has been assumed that:
to account for the number of $\gamma$-rays emitted in each mode. The probability $P$ was obtained from the exponential decay law:

$$P = \frac{1}{I_0} = \exp(-\lambda t)$$ (5.3)

where $\lambda = 7.04 \mu s^{-1}$ and the time $t$, needed for $o$-Ps of certain energy to reach the walls of the cell, was computed using the Simion 6.0 ion optics simulation program.

Figure 5.3 illustrates schematically the trajectories of Ps atoms originating from three positions along the centre of the beam axis (i.e. at $x = 0$mm). As mentioned in section 2.5, ions can only be extracted from half of the volume of the gas cell. Thus, in this simulation only trajectories of Ps atoms originating in 5mm intervals from $y = -25$mm to $y = 25$mm were taken into consideration. The time needed for Ps atoms, emitted isotropically at varying angles, to travel from the point of origin to the walls of the gas cell was recorded. Both the azimuthal ($\theta$) and the polar ($\phi$, not shown in the graph) were stepped in $10^\circ$ intervals, where $0^\circ < \theta < 360^\circ$ and $0^\circ < \phi < 180^\circ$. Overall, a

$$\epsilon_{3y} = \frac{3}{2} \epsilon_{2y}$$ (5.2)

**Figure 5.3. Illustration of Ps trajectories originating along the centre of the beam axis.**

For clarity only three starting positions are shown, the central and the two most extreme ones (to scale).
total of 6754 trajectories were simulated. The extracted times where fed in to eq. 5.3, from which an average probability of o-Ps, at a given energy, reaching the walls of the gas cell was calculated. This procedure was repeated for several energies of Ps atoms in the range from 1eV to 80eV. The coordinates of the position where o-Ps hit the walls were recorded simultaneously with the time, therefore it was possible to correct $P$ for the solid angle subtended by the $\gamma$-detector to that point.

The present model is limited by the assumption that Ps atoms are emitted with equal probabilities at all angles. This poses a problem for example at higher positron impact energies, where Ps formation is expected to be forward collimated (Laricchia et al., 1987) and, therefore, Ps atoms may escape undetected through the apertures of the gas cell. This effect would tend to decrease the total Ps detection efficiency with increasing energy.

In figure 5.4, the energy dependence of the probability of o-Ps reaching the walls of the gas cell is shown (blue curve). At low energies, it increases rapidly from approximately 0.55 at 1eV to almost 0.9 at around 30eV. The shape of the function is not modified considerably when the correction is made for the solid angle, the largest change being approximately 2% at the highest energies considered. The solid angle correction for the o-Ps portion that decays in flight in the gas cell before reaching the

![Figure 5.4. The energy dependence of the probability of o-Ps reaching the walls of the gas cell.](image-url)
walls should be even smaller and is therefore ignored.

The energy dependence of the total detection efficiency of positronium ($\varepsilon_{Ps}$) for the present geometry of the gas cell can be written as:

$$\varepsilon_{Ps}(E) = 0.75P(E)\varepsilon_{2\gamma} + (1 - P(E))\varepsilon_{3\gamma} + 0.25\varepsilon_{2\gamma}.$$  (5.4)

where the coefficients of 0.75 and 0.25 take into account that the relative population of p-Ps:o-Ps is 1:3. Substitution of eq. 5.2 into eq. 5.4 with $\varepsilon_{2\gamma} = 1$ leads to:

$$\varepsilon_{Ps}(E) = 0.75P(E) + 3/2(1 - P(E)) + 0.25.$$  (5.5)

In figure 5.5, this energy dependence is shown normalized to 1 at 1eV. It decreases rapidly with increasing energies but the reduction between 1eV and 80eV is only approximately 12%. This analysis demonstrates that although the probability of o-Ps quenching on the walls of the gas cell is greater than its probability to decay in flight, this latter mode dominates the energy dependence of the detection efficiency due to the emission of three rather than two $\gamma$-rays.

The positronium formation cross-sections for He, Ar and Xe, presented in the next section of this chapter, have been divided by $\varepsilon_{Ps}$ shifted by the appropriate Ps threshold energy in order to obtain the correct energy dependence.

**Figure 5.5.** The energy dependence of the positronium detection efficiency for the geometry of the present gas cell.
5.4. Results and discussion

In figure 5.6, the direct measurement of the positronium formation cross-section in He is compared to experimental results of Overton et al (1993) and the elaborate calculation of Campbell et al (1998), both already discussed in chapter 3. The present results have been normalized to the data of Campbell et al (1998) in the region of the peak and errors bars of up to 6.5% arise primarily from the uncertainty in the normalization procedure. The present $Q_{ps}$ agrees with the energy dependence of the measurements of Overton et al (1993) but rises above the theoretical data from approximately 50eV onwards by up to 30% at 80eV. Due to the fact that in the energy range from 60eV to 120eV the results of Overton et al (1993) lie approximately 20% above the calculation of Campbell et al (1998), it had been speculated whether they might still have been affected by incomplete confinement of scattered positrons. However, in the light of the present measurements, obtained with a different method to that of Overton et al (1993) and believed to be converged with respect to scattered projectile confinement in the energy range shown in the graph (see discussion section 5.2.), further theoretical investigations would be desirable.

Figure 5.6. Positronium formation cross-section for He.
In figure 5.7, the present $Q_{Ps}$, expressed in terms of the excess energy $E' = E - E_{Ps}$, $E$ being the incident positron energy, is compared with the near threshold experimental results of Moxom et al (1994) and the Kohn variational calculation of Van Reeth and Humberston (1997). The present results seem to follow the same energy dependence of the cross-section as the data of Moxom et al (1994), which were measured with a better positron-beam energy-resolution ($\Delta E \approx 0.8$ eV FWHM) and a pulsed electric field for ion extraction. The theoretical calculation of Van Reeth and Humberston (1997), although displaying a similar slope, are lower than the data of Moxom et al (1994) by approximately 30%. However, whilst the theoretical calculation is believed not to be fully converged and thus has an uncertainty of approximately 20%, the experimental result of Moxom et al (1994) has uncertainties in the energy calibration and approximately a 20% error due to the normalization procedure. An alternative normalization procedure, relying on the normalization of the ionization cross-section to the electron scattering cross-section, as was done for the other noble gases by Laricchia et al (2002), may aid in removing some of the outstanding uncertainties.

Figure 5.7. Positronium formation cross-section for He near $E_{Ps}$.
In figures 5.8 and 5.9, the present $Q_{ps}$ from Ar and Xe measured with a magnetic field strength of 130 Gauss are shown. These results were normalized to the data of Laricchia et al (2002) in the region of the first peak. The error bars comprise only the statistical uncertainty. The present $Q_{ps}$ for Ar follow the energy dependence of the data of Laricchia et al (2002) up to 40eV. They reproduce the position and the relative height of the second peak but rise above the data of Laricchia et al (2002) at approximately 30eV above $E_{ps}$, as would be expected from an increased failure to confine scattered projectiles. The recent measurements of Sullivan et al (2002b), performed with an energy resolution of 25meV, are also shown in figure 5.8. They agree reasonably well with the present $Q_{ps}$ on the energy dependence and the magnitude of the first peak but not beyond it.

For Xe, the present $Q_{ps}$ display the same energy dependence of the first peak (up to 10eV above $E_{ps}$) as the data of Laricchia et al (2002), however the maximum of the second peak lies approximately 10% above the cross-section of Laricchia et al (2002). Again, this is believed to be due to the increasing static interaction between incoming positrons and target atoms of higher atomic number.

![Figure 5.8. Positronium formation cross-section for Ar.](image-url)
As discussed in section 5.3, the forward collimation of positronium is expected to rise with increasing positron impact energy and to result in an overestimation of the positronium detection efficiency due to Ps atoms escaping from the gas cell. It has been estimated from the theoretical differential calculations of McAlinden and Walters (1994) that the fraction of Ps atoms emitted from the centre of the gas cell within the solid angle subtended by the aperture does not vary significantly with energy. Furthermore, for Ar and Xe this fraction only contributes approximately 4% and 2%, respectively to the total Ps formation. Consequently, the overall effect on the present total Ps detection efficiency is negligible.

It is worthy to note, that in the case of Ar and Xe, the increases in the present cross-sections at approximately 22eV and 15eV respectively, are significantly below the inner-shell excitation thresholds for these targets (indicated by vertical dashed lines in the graphs) and therefore are thought to support the suggestion of Laricchia et al (2002) that the second peaks could be due to excited-state positronium formation.

![Figure 5.9. Positronium formation cross-section for Xe.](image)
5.5. Comparison of the direct and the extracted $Q_{ps}$ for He

In the case of He, the positronium formation cross-section has been extracted by subtracting the direct ionization cross-section from the total ionization cross-section as was done by Laricchia et al (2002) (see section 3.2.1). In figure 5.10, the present total ionization cross-section of He is shown normalized to the cross-section of Moxom et al (1995). The uncertainties of approximately 5% in the present results are mainly due to large error bars in $Q_i$ of Moxom et al (1995), which in turn were normalized to the data of Fromme et al (1986). Subsequently, as shown in figure 5.11, the present $Q_i$ were used to obtain $Q_{ps}$ according to eq. 3.4 by fitting a fifth-order polynomial to the direct ionization cross-section of Moxom et al (1996) and subtracting it from $Q_i$. As shown in the figure 5.11, the agreement between the extracted $Q_{ps}$ and that measured directly is reasonable considering the uncertainties arising from the present normalization procedure. The difference in magnitudes between the two $Q_{ps}$ is approximately 10% at 45eV.

![Figure 5.10. Total ionization cross-section for He.](image-url)
5.6. Conclusions

In this chapter, direct measurements of the positronium formation cross-section for He, Ar and Xe have been presented. They were obtained by counting coincidences between the target ions and γ-rays from positronium annihilation. Above $E_{Ps}$, such coincidences arise predominantly from positronium formation.

An analysis has also been carried out to determine the energy dependence of the detection efficiency of positronium for the geometry of the present gas cell. This shows that whilst a large proportion of o-Ps reaches the walls of the gas cell, where it is assumed to quench, in flight annihilation of o-Ps dominates the energy dependence of the positronium detection efficiency due to the emission of three rather than two γ-rays. As a result, the total positronium detection efficiency decreases rapidly with increasing energies, which has been accounted for in the present $Q_{Ps}$.

Present $Q_{Ps}$ for Ar and Xe agree well with those of Laricchia et al (2002) in that they reproduce a double-peak structure. In the case of Ar a very good agreement has
been found with the results of Laricchia et al (2002) up to 30eV above $E_{p_i}$. For Xe, for which the second peak is higher than that in the data of Laricchia et al (2002), this is true only up to 10eV above $E_{p_i}$. It is believed that the increasing static interaction between the incident positrons and targets of a higher atomic number ($Z$) progressively reduces the excess energy up to which a converged cross-section can be obtained. The lowest-$Z$ atom considered was He, for which a converged $Q_{p_i}$ has been obtained up to 70eV above $E_{p_i}$. This cross-section agrees well with the experimental results of Overton et al (1993), which had been thought to be affected by unconfined positron scattering at higher energies, and exceeds the theoretical calculations of Campbell et al (1998) from 50eV onwards. It is hoped that the present result will prompt further investigations. Furthermore, an analysis to obtain $Q_{p_i}$ for He by subtracting the direct ionization cross-section from the total ionization cross-section has also been performed. Given the considerable uncertainties in the normalization procedure, the agreement between the two determinations of $Q_{p_i}$ is satisfactory.
CHAPTER 6
CONCLUSIONS AND OUTLOOK

In this work, a monoenergetic positron beam obtained from a radioactive source and guided by a magnetic field was used to study ionization processes, namely annihilation and positronium formation, induced by positron collisions with atoms and molecules. The experimental apparatus comprised an efficient ion detector and a γ-detector. Whilst measuring total ion yields has allowed total ionization cross-sections to be obtained, measuring coincidences between γ-rays and target ions has enabled the study of annihilation below $E_{Ps}$ and, above it, the sum of annihilation and positronium formation.

The total ionization cross-sections for Ne and Xe have been used together with the data of Laricchia et al (2002) to extract positronium formation cross-sections, as described in Chapter 3. It has been found that both $Q'$ and $Q_{Ps}$ for Xe vary smoothly at intermediate energy and no evidence of the structure at around 80eV predicted by the theoretical calculation of McAlinden and Walters (1992), and hinted to by the results of Laricchia et al (2002), has been observed.

The extracted positronium formation cross-sections have been supplemented with the direct measurements for He, Ar and Xe, described in Chapter 5. These results, corrected for the energy dependence of the positronium detection efficiency for the geometry of the present gas cell, have been found to confirm the double-peak structure exhibited by the results of Laricchia et al (2002) in the case of Ar and Xe. The second peak in $Q_{Ps}$ of Laricchia et al (2002) for Ar, Kr and Xe has been interpreted as arising from the formation of positronium in its various quantum states. In the case of He, the present $Q_{Ps}$ values agree very well with the experimental data of Overton et al (1993), but exceed the elaborate theoretical calculation of Campbell et al (1998) from 50eV onwards. Further investigations, both experimental and theoretical, would be needed to clarify the existing discrepancy at intermediate energies. Moreover, $Q_{Ps}$ for He, obtained by subtracting the direct ionization cross-section of Moxom et al (1996) from the present total ionization cross-section, has been compared to $Q_{Ps}$ measured directly. A satisfactory agreement between the two experimental determinations has been found.
It is anticipated that an alternative normalization procedure, relying on the electron ionization cross-sections, as was done for the other noble gases by Laricchia et al. (2002), may aid in removing some of the outstanding uncertainties. It would also be very interesting to measure the positronium formation cross-section for H$_2$O, for which available data are scarce and whose biological relevance is paramount.

The positronium formation cross-sections of Laricchia et al. (2002) for the noble gases have stimulated the estimation of lower and upper limits for the contribution of excited state positronium formation for these targets. The evaluated limits suggest that positronium formation may occur more abundantly than previously thought. An experiment is planned to test this hypothesis by measuring the Lyman-α photon of 5.1eV energy, emitted through the 2P-1S transition, in coincidence with the remnant ion. Such an investigation would provide the first direct cross-sections for the excited-state positronium formation.

As described in Chapter 3, the evaluation of the upper limit for the contribution of excited state positronium formation followed the finding of an empirical scaling formula which factors out the gross target dependence of single ionization cross-sections by positron- and electron-impact. The formula can be used to estimate cross-sections for any atom, if the cross-section maxima for any other two atoms belonging to the same column of the periodic table are known. Further analysis of data, for these and other combinations of projectiles and targets, is intended in order to understand the physical cause underlying these factorizations.

The investigation of the energy dependence of the annihilation cross-sections below $E_{ps}$ for Ne, Xe and O$_2$, described in Chapter 4, has been prompted by the finding of a positive residual signal below $E_{ps}$ in the total ion yields for Ne. Despite the relatively wide energy spread of the positron beam and the uncertainty in the absolute energy calibration, there is evidence of non-negligible positive values of the annihilation cross-section for Ne significantly below $E_{ps}$. Whilst in the case of Xe, the increase below $E_{ps}$ might be attributed to the resolution of the beam, for O$_2$, where the excitation threshold precedes the positronium formation threshold, an onset in $Q_{ann}$ is observed around $E_{ex}$. It is hoped that further measurements with a pulsed ion-extracting field will help to eliminate remaining uncertainties in the present energy calibration. Clearly, further experimental investigations of annihilation cross-sections performed with a smaller energy spread are warranted, especially in the case of atomic targets.
where complications arising from the presence of the vibrational excitations would be avoided. In this respect, measurements with a He target would prove particularly desirable, since theoretical results obtained with a variety of approaches for this target are available.
Appendix

**Estimation of the contribution from contaminants in the gas cylinders used in chapter 4**

Below $E_{ps}$, a signal arising from annihilations on contaminants, present in the high-purity gases, has been estimated in order to assess whether the increase in the measured $Q_{ann}$ could have originated from these.

For Ne (for which a non-negligible signal has been found some eV below $E_{ps}$), using the concentration values from the table below* and the total cross-sections of Kauppila and Stein (1990) for N$_2$ and Sueoka et al (2000) for C$_6$H$_5$CH$_3$ (chosen as an example of a hydrocarbon), the signal associated with these main contaminants has been estimated to contribute no more than 0.2% and 2%, respectively, to the measured $Q_{ann}$ at approximately 10eV. Similarly for Xe, the possible contribution from Kr amounts to no more than 0.2% and that from N$_2$ to 0.03% at 5eV. In the case of O$_2$ gas, contributions from N$_2$ and Ar at 5eV together would not exceed 0.1% of the observed signal. It is also worth noting that the ion-mass selection method, employed in these measurements and discussed in 4.3.1, would serve to reduce yet further the signal contribution from these impurities.

<table>
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<th>Concentration (ppm)</th>
<th>$E_{ps}$(eV)</th>
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</thead>
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<td>17.79</td>
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<tr>
<td>N$_2$</td>
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<td>8.78</td>
</tr>
<tr>
<td>O$_2$</td>
<td>2</td>
<td>5.27</td>
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<tr>
<td>Total hydrocarbons</td>
<td>3</td>
<td>&gt;2eV</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>2</td>
<td>5.82</td>
</tr>
</tbody>
</table>

Ne purity 99.994%; $E_{ps}$=14.76eV

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<td>O$_2$</td>
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<tr>
<td>Total hydrocarbons</td>
<td>3</td>
<td>&gt;2eV</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>5</td>
<td>5.82</td>
</tr>
</tbody>
</table>

Xe purity 99.993%; $E_{ps}$=5.33eV

<table>
<thead>
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<th>Contaminants</th>
<th>Concentration (ppm)</th>
<th>$E_{ps}$(eV)</th>
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<tr>
<td>Ar</td>
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<tr>
<td>Total hydrocarbons</td>
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<tr>
<td>CO and CO$_2$</td>
<td>0.05</td>
<td>7.21; 6.98</td>
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</table>

O$_2$ purity 99.9995%; $E_{ps}$=5.27eV

*Gas purities and contaminant concentrations as quoted by BOC Gases, UK.


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