ERRATA

Page, line:
1, 16. delete "between", insert "among"
1, 22. the equation should read
   \[ H^{i+} + A^0 \rightarrow A^{g^+} + H^{f+} + (f + g - 1)e \]
1, 26. delete "of projectile".
10, 8. to support preferentially
10, 13. determined only by
11, 4. data join
11, 5. remain
11, 6. Branscomb
26, 14. maximum
30, 6. \[ \sigma_{11} \text{ N}_- \] not \[ \sigma_{1\overline{1}} \text{ N}_- \]
35, 18. Keithley
40, 23. eliminate "that" after "expect".
68, 26. insert "in Ne". after "Barnett's values".
69, 3. insert "in Ne" after "values".
82, 17. Branscomb
83, 18. Branscomb
107, 10. Bayly
128, 14. Boltzmann
128, 18. delete "ion" insert "molecule in the"
131, 10. delete "2\overline{4}" insert "infinity"
133, 10. indicate
146, 16. delete "is" insert "are"
150, 13. insert "\gamma increases" after "that"
153, 19. delete "is" insert "are"
162, 21. delete "beam", insert "curve"

Diag. 18, 19, 20. The graph labelled "Gryzinski, 1953" should be labelled "calculated from Gryzinski, 1963".

Diag. 33. The legends for Fogel, 1958, and Stier and Barnett, 1956, should be interchanged.

Additions to Bibliography.


This thesis entitled

"An Experimental Investigation of Charge Changing Collisions of the Hydrogen Atomic and Molecular Ions, $H_1^+, H_2^+, H_3^+$ and $H_1^-$ and Hydrogen Atoms, $H_1^0$, of Energies 2 to 50 Kev, in Collision with Hydrogen and the Inert Gases."

has been submitted for the degree of

Doctor of Philosophy

in the Australian National University

by J.F. Williams

Enrolled: 1 January, 1962
Submitted: 29 January, 1965
The work presented in this thesis has been an individual effort and has not been carried out in conjunction with any other workers. The supervisor for the study was Professor D.N.F. Dunbar.

J. F. WILLIAMS
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11. Acknowledgements

12. References
1.1 Formulation of the Problems

At the commencement of the investigations described in this thesis there had been no previous work in the field of atomic collisions within the author's laboratory. This fact strongly influenced the decision to select for study some comparatively simple problems which required simple unsophisticated apparatus. From the many reviews of the experimental investigations of atomic collision phenomena by Massey and Burhop (1952), Branscomb (1957), Allison (1958) and Hasted (1960), but in particular from the review by Allison, it was apparent that such a study could be readily founded upon the numerous discrepancies which existed between the measurements of the total charge changing cross sections for even the simplest of elements, hydrogen.

Table 1 presents a list of the possible charge changing collisions between hydrogen atoms and ions and a neutral gas atom A. The cross section for a collision of the type $H^+ + A^0 \rightarrow A^+ + (f + g - i)e$ maybe represented by the notation $\sigma(\text{io}/fg)$ or briefly $\sigma_{\text{if}}$. By confining our attention to inert gas target atoms, which do not form stable negative ions, the electron loss (or sometimes called detachment, stripping or ionization of projectile)
## Table 1

### Possible Charge Changing Collisions

for Fast Hydrogen Beams

<table>
<thead>
<tr>
<th>Collision</th>
<th>Process</th>
<th>Notation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. $H_1^+ + A \rightarrow H_1^0 + A^+$</td>
<td>One electron capture (charge transfer)</td>
<td>$\sigma^{10}$</td>
</tr>
<tr>
<td>2. $H_1^+ + A \rightarrow H_1^- + A^{++}$</td>
<td>Two electron capture (charge transfer)</td>
<td>$\sigma^{11}$</td>
</tr>
<tr>
<td>3. $H_1^0 + A \rightarrow H_1^- + A^+$</td>
<td>One electron capture (charge transfer)</td>
<td>$\sigma^{01}$</td>
</tr>
<tr>
<td>4. $H_1^0 + A \rightarrow H_1^+ + e + A$</td>
<td>One electron loss (no charge transfer)</td>
<td>$\sigma^{01}$</td>
</tr>
<tr>
<td>5. $H_1^- + A \rightarrow H_1^0 + e + A$</td>
<td>One electron loss (no charge transfer)</td>
<td>$\sigma^{-10}$</td>
</tr>
<tr>
<td>6. $H_1^- + A \rightarrow H_1^+ + 2e + A$</td>
<td>Two electron loss (no charge transfer)</td>
<td>$\sigma^{-11}$</td>
</tr>
</tbody>
</table>
processes do not include charge transfer from projectile to target. The electron capture collision is necessarily a charge transfer process. For all six collisions of Table 1 the target atom, after collision, may be left in an excited state or a more highly ionized state.

Hydrogen atoms and ions are the simplest possible projectiles. There are only three possible charge states, $H^+$, $H^0$ and $H^-$ and the ions have no stable electronically excited states. The positively charged hydrogen atomic ion is a bare proton. The negatively charged hydrogen ion, $H^-$, has such a small binding energy for its outer electron in the ground state (0.74 ev) (Massey, 1950) that it is most unlikely that any stable discrete excited states exist with appreciable binding energy. For the hydrogen atom there are any number of excited states as all quantum numbers (as allowed by the selection rules) are possible.

A summary of charge changing cross section measurements for fast hydrogen atoms and ions is presented in Table 2 which is based on a similar table by Allison (1962). The experimental methods, which are described in the review articles mentioned above, are noted for each measurement. Only the method used by Fogel, that of mass analysis of the projectile beam to measure the rate of growth of new charge states which result from single collisions, determines a definite total charge changing cross section for a projectile in which three or more
### Table 2
Summary of Charge Changing Cross Section Investigations with Hydrogen Beams

<table>
<thead>
<tr>
<th>Investigation</th>
<th>Quantity measured</th>
<th>Energy range kev</th>
<th>Target gases</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Goldman (1931)</td>
<td>$\sigma_{10}$</td>
<td>1-4</td>
<td>H$_2$</td>
<td>Target ion collection</td>
</tr>
<tr>
<td>Keene (1949)</td>
<td>$\sigma_{10}$</td>
<td>3-30</td>
<td>H$_2$</td>
<td>Target ion collection</td>
</tr>
<tr>
<td>Ribe (1951)</td>
<td>$\sigma_{10}$</td>
<td>35-150</td>
<td>H$_2$</td>
<td>Attenuation in transverse magnetic field</td>
</tr>
<tr>
<td>Kasner (1951)</td>
<td>$\sigma_{01}, \sigma_{10}$</td>
<td>30-300</td>
<td>Air</td>
<td>Attenuation in transverse magnetic field</td>
</tr>
<tr>
<td>Hasted (1952)</td>
<td>$\sigma_{10}$</td>
<td>0.025-4</td>
<td>Ar</td>
<td>Target ion collection</td>
</tr>
<tr>
<td>Whittier (1954)</td>
<td>$\sigma_{10}, \sigma_{01}$</td>
<td>4-50</td>
<td>H$_2$</td>
<td>Attenuation in transverse magnetic field</td>
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<tr>
<td>Fogel et al. (1955a)</td>
<td>$\sigma_{10}$</td>
<td>12.3-36.7</td>
<td>H$_2$</td>
<td>Target ion collection</td>
</tr>
<tr>
<td>Stedeford (1955)</td>
<td>$\sigma_{10}, \sigma_{10}$</td>
<td>3-40</td>
<td>H$_2$, He, Ne, Ar, Kr, Xe</td>
<td>Target ion collection cont./over</td>
</tr>
<tr>
<td>Investigation</td>
<td>Quantity measured</td>
<td>Energy range kev</td>
<td>Target gases</td>
<td>Method</td>
</tr>
<tr>
<td>-----------------------------</td>
<td>-------------------</td>
<td>------------------</td>
<td>----------------------------------</td>
<td>---------------------------------------------</td>
</tr>
<tr>
<td>Hasted (1955)</td>
<td>$\sigma_{\text{10}}, \sigma_{\text{10}}$</td>
<td>0.1-4.0</td>
<td>$\text{H}_2, \text{He}, \text{Ne}, \text{Ar, Kr, Xe}$</td>
<td>Target ion collection</td>
</tr>
<tr>
<td>Gilbody &amp; Hasted (1956)</td>
<td>$\sigma_{\text{10}}$</td>
<td>0.01-40</td>
<td>$\text{N}_2, \text{CO}, \text{NH}_3$</td>
<td>Target ion collection</td>
</tr>
<tr>
<td>Stier &amp; Barnett (1956)</td>
<td>$\sigma_{\text{01}}, \sigma_{\text{10}}$</td>
<td>3-30</td>
<td>$\text{H}_2, \text{He}, \text{N}_2, \text{O}_2, \text{Ne, Ar}$</td>
<td>Attenuation in an electric field</td>
</tr>
<tr>
<td>Afrosimov et al. (1958a)</td>
<td>$\sigma_{\text{10}}$</td>
<td>5-180</td>
<td>$\text{Ar}$</td>
<td>Target ion collection and e/m analysis</td>
</tr>
<tr>
<td>Barnett &amp; Reynolds (1958)</td>
<td>$\sigma_{\text{10}}$</td>
<td>250-1000</td>
<td>$\text{H}_2, \text{He}, \text{N}_2, \text{Ar}$</td>
<td>$\sigma_{\text{01}}$, plus beam equilibrium</td>
</tr>
<tr>
<td>Ilin et al. (1959)</td>
<td>$\sigma_{\text{10}}$</td>
<td>5-180</td>
<td>$\text{Air, N}_2$</td>
<td>Target ion collection and e/m analysis</td>
</tr>
<tr>
<td>Fogel et al. (1955)</td>
<td>$\sigma_{\text{11}}$</td>
<td>13-30</td>
<td>$\text{H}_2$</td>
<td>$\frac{\text{d}F_{\gamma}}{\text{d}\pi}$ at low $\pi$-values</td>
</tr>
<tr>
<td>Fogel et al. (1956)</td>
<td>$\sigma_{\text{11}}$</td>
<td>15-30</td>
<td>$\text{H}_2$</td>
<td>$\frac{\text{d}F_{\gamma}}{\text{d}\pi}$ at low $\pi$-values</td>
</tr>
<tr>
<td>Fogel &amp; Mitin (1957)</td>
<td>$\sigma_{\text{11}}$</td>
<td>10-30</td>
<td>$\text{O}_2$</td>
<td>($\frac{\text{d}F_{\gamma}}{\text{d}\pi}$) at low $\pi$-values</td>
</tr>
<tr>
<td>Fogel et al. (1959)</td>
<td>$\sigma_{\text{11}}$</td>
<td>3-65</td>
<td>$\text{H}_2, \text{He}, \text{N}_2, \text{Ne, Ar, Kr, Xe}$</td>
<td>$\frac{\text{d}F_{\gamma}}{\text{d}\pi}$ at low $\pi$-values</td>
</tr>
</tbody>
</table>

cont./over
<table>
<thead>
<tr>
<th>Investigation</th>
<th>Quantity measured</th>
<th>Energy range (keV)</th>
<th>Target gases</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kozlov et al. (1963)</td>
<td>( \sigma_{11} )</td>
<td>2-50</td>
<td>( \text{H}_2, \text{Ar}, \text{Kr} )</td>
<td>( \frac{dF_1}{d\pi} ) at low ( \pi )-values</td>
</tr>
<tr>
<td>McClure (1963)</td>
<td>( \sigma_{11} )</td>
<td>6-50</td>
<td>( \text{H}_2 )</td>
<td>( \frac{dF_1}{d\pi} ) at low ( \pi )-values</td>
</tr>
<tr>
<td>Montague (1951)</td>
<td>( \sigma_{01} )</td>
<td>40-250</td>
<td>( \text{H}_2 )</td>
<td>Attenuation in transverse magnetic field</td>
</tr>
<tr>
<td>Barnett &amp; Reynolds (1958)</td>
<td>( \sigma_{01} )</td>
<td>250-1000</td>
<td>( \text{H}_2, \text{He}, \text{N}_2, \text{Ar} )</td>
<td>Attenuation in transverse electric field</td>
</tr>
<tr>
<td>Fogel et al. (1958a)</td>
<td>( \sigma_{01}, \sigma_{01} )</td>
<td>5-40</td>
<td>( \text{H}_2, \text{He}, \text{Ne}, \text{N}_2, \text{O}_2, \text{Ar}, \text{Kr}, \text{Xe} )</td>
<td>( \frac{dF_1}{d\pi} ); ( \frac{dF_1}{d\pi} ) at low ( \pi )-values</td>
</tr>
<tr>
<td>Curran &amp; Donahue (1959)</td>
<td>( \sigma_{01} )</td>
<td>5-40</td>
<td>( \text{H}_2 )</td>
<td>Attenuation; Target ion collection</td>
</tr>
<tr>
<td>Fogel et al. (1960)</td>
<td>( \sigma_{01} )</td>
<td>5-10</td>
<td>( \text{Kr} )</td>
<td>( \frac{dF_1}{d\pi} ) at low ( \pi )-values</td>
</tr>
<tr>
<td>Rose et al. (1958)</td>
<td>( \sigma_{10} )</td>
<td>0.4-2000</td>
<td>( \text{H}_2, \text{O}_2, \text{Ar}, \text{CO}_2 )</td>
<td>Attenuation in transverse field</td>
</tr>
<tr>
<td>Fogel et al. (1957)</td>
<td>( \sigma_{11} )</td>
<td>5-40</td>
<td>( \text{H}_2, \text{He}, \text{N}_2, \text{O}_2, \text{Ne}, \text{Ar}, \text{Kr}, \text{Xe} )</td>
<td>( \frac{dF_1}{d\pi} ) at low ( \pi )-values</td>
</tr>
<tr>
<td>Bartels (1932)</td>
<td>( \sigma_{10} )</td>
<td>5-30</td>
<td>( \text{H}_2 )</td>
<td>Beam equilibrium</td>
</tr>
<tr>
<td>Meyer (1938)</td>
<td>( \sigma_{10} )</td>
<td>30-100</td>
<td>( \text{H}_2 )</td>
<td>Beam equilibrium</td>
</tr>
</tbody>
</table>
DIAG. 1.

PROJECTILE ENERGY – KEV.

CROSS SECTION – CM$^2$/ATOM

- $H_1^+$
- $H_2$
- $\sigma_{10}$

STIER 1956
RIBE 1951
STEDFORD 1955
KASNER et al 1957
KEENE 1949
GOLDMAN 1931
HASTED 1955
GRYZINSKI 1963
charge states are possible. This method has been used in the present investigation. The "multiple collision" or beam equilibrium method yields extremely complicated combinations of the six possible cross sections $\sigma_{if}$ for hydrogen, while the methods of attenuation of the primary beam in a transverse electric or magnetic field and the collection of slow target ions and electrons both measure a sum of cross sections. The determination of a single cross section involves either (i) further measurements or (ii) the assumption that some cross sections are negligibly small.

1.11 Fast Protons

Almost half of the measurements in Table 2 are for $\sigma_{10}$, the single electron capture. The results of Hasted and Stedeford (1955) and Stier and Barnett (1956) in the five inert gases and hydrogen agree within 5%; in both absolute value and energy dependence. Their results are generally near the mean of the results of other workers. Diag. 1 shows the results of $\sigma_{10}$ in hydrogen. Nearly every worker in the field of atomic collisions has measured $\sigma_{10}$ for $H_1^+$ in hydrogen gas with the result that this cross section can be reasonably well defined for its energy dependence but there exists a considerable spread of absolute values at any given energy. (The logarithmic scale of diag. 1 appears to minimize this spread.) Despite this fact a mean value of $8.2 \times 10^{-16}$ cm$^2$/molecule at 10 Kev proton
energy appears to be well established. The difficulties of absolute cross section determination which arise mainly from target gas number density measurements, have led several workers (Fite, 1960 and McClure, 1963) to determine relative cross sections and then to standardize their values against the most well known cross section value, which has been $\sigma_{10}$. This procedure has also been followed by the present author who also has considered that agreement between the shape of his experimental determination of $\sigma_{10}$ versus energy curve and the mean curve of $\sigma_{10}$ from the results of Table 2 is reasonable confirmation of the satisfactory operation of the present apparatus and general measurement techniques. In the inert gases there is agreement within 5% between the results of Hasted and Stier and Barnett.

The single electron capture by protons is fundamentally unsuitable for a standardization value since it is a sum of all the partial cross sections of capture into excited states of the atom and, for a hydrogen target, of possible vibrational excitation or dissociation complications. A process free from all these objections is double electron capture by protons in hydrogen and helium gas:

$$H^+_1 + H_2 \rightarrow H^-_1 + 2H^+_1$$

$$H^+_1 + He \rightarrow H^-_1 + He^{++}$$

where all particles are in their ground states. Fogel has


**DIAG. 2.**

**PROJECTILE** $H^+_1$

**TARGET** $H_2$

**CROSS SECTION** $\sigma_{11}$

---

**FOGEL 1959**

**1955**

**1956**

**1957**

---

**CROSS SECTION — CM$^2$/ATOM**

---

**PROJECTILE ENERGY — KEV.**
DIAG. 3.
TWO ELECTRON CAPTURE CROSS SECTIONS $\sigma_{1T}^{-1}$
PROJECTILE $- H^+$
TARGETS $- He, Ne, Ar, Kr, Xe$. 

PROJECTILE VELOCITY — $10^6$ cm/sec.
measured $\sigma_{11}$ in hydrogen on four different occasions (1955, 1956, 1957 and 1959) and has obtained four vastly different results for both the absolute magnitude and the energy dependence as shown in diag. 2. At 15 Kev where his measurements possibly indicate a maximum cross section his values differ by a factor of five. Possible reasons for such inaccuracies are suggested in Chapter 4 where the accurate measurement of this cross section is reported.

Fogel (1959) has also measured $\sigma_{11}$ in He and the other inert gases; these cross sections are shown in diag. 3. The cross section in He has a peak value of $7.10^{-19} \text{ cm}^2$/atom at 35 Kev and rapidly decreases at other energies. The small value and the uniqueness of this reaction therefore offers a good test of an apparatus sensitivity and accuracy of measurement. The only theoretical predictions of $\sigma_{11}$ are for $H_1^+$ in He. The perturbed stationary state (P.S.S.) calculations of Rosentsveig (1955) for $H_1^+$ energies less than 10 Kev and the Born approximation calculations of Gerasimenko (1962) above 100 Kev are compared with the present accurate measurements of $\sigma_{11}$ in He in Chapter 4. Fogel's measurements (1959) of $\sigma_{11}$ in Ne show a broader single peaked function than for He but a maximum value of only $6.10^{-19} \text{ cm}^2$/atom which is less than that for He - a surprising result. All the other inert gases show a double peaked function. Fogel proposes that the higher energy peak, which for all three gases has
a larger maximum value than the lower energy peak, is due to the process $H_1^+ + Xe \rightarrow H_1^- + Xe^{++} + e$; i.e. double electron capture with simultaneous ionization of the target atom. It seems unlikely that this process should be more probable than simple two electron charge transfer. In view of the wide discrepancies between Fogel's 1959 data in $H_2$ and his three earlier papers and the lack of any other experimental evidence to preferentially support his 1959 data, there is a definite need to re-measure accurately the $\sigma_{11}$ cross section in all the inert gases.

1.12 Fast $H_1^-$

The one electron loss (detachment) cross section from $H_1^-$ has only been determined by methods which use the assumption that the simultaneous two electron loss process is a most unlikely event. In Hasted's (1952) and Hasted and Stedeford's (1955) experiments, the loss cross section is measured by the collection of the current of detached electrons. As the double electron loss process also occurs their experiments measure $(\sigma_{10} + 2 \sigma_{11})$. The only other reported measurements are those of Stier and Barnett (1956) who determined $\sigma_{10}$ by the attenuation of a beam of atoms in a gas of sufficient pressure either to equilibrate the charge distribution or to allow only single collisions to occur. However it is the change in the composition of the beam which is studied and not the currents of slow ions or electrons formed in the target gas. Their experiments
measure \((\sigma_{10} + \sigma_{11})\). Hence differences between the measured cross sections of the two workers might indicate that the \(\sigma_{11}\) process should not be neglected. Diag. 4 shows that, in Argon, Stier and Barnett's data joins fairly smoothly onto Hasted's 1952 data but remains considerably below Hasted's 1955 data. As Branscombe (1957) has suggested, it appears that this difference could be due to an onset in the value of \(\sigma_{11}\) around 3 Kev. The \(\sigma_{10}\) data in Kr and Xe show a similar large increase beginning around 3 Kev, while in He and Ne (not shown in diagram) there is no such increase in \(\sigma_{10}\) values. The status of these previous measurements is indicated in Chapter 4 of this thesis in which:

(a) the simultaneous measurements of the growth from single collisions of \(H^0\) and \(H^+\) from \(H^-\) ions has enabled both \(\sigma_{10}\) and \(\sigma_{11}\) to be determined in \(H_2\) and the inert gases,

(b) small increments of 0.5 Kev in the \(H^-\) projectile energy enable the accurate form of \(\sigma_{10}\) to be determined over the doubtful 2 to 6 Kev region.

The only previous measurements of \(\sigma_{11}\) for negative hydrogen ions are those of Fogel (1957) whose experimental accuracy is only 30% and whose measurements of \(\sigma_{11}\), which were obtained with the same apparatus and method as was used for his \(\sigma_{11}\) measurements, are shown in Chapter 4 of this thesis to be in error. There is considerable need
then to repeat the measurements of $\sigma_{11}$.

The present measurements of $\sigma_{10}$, which are free of any contribution from $\sigma_{11}$, now permit a true comparison to be made of the experimental values of $\sigma_{10}$ with the Born approximation calculations of Sida (1955) for the loss process in helium

$$H_1^-(1s)^2 + He(1s)^2 \rightarrow H(1s) + e + He(1s)^2$$

and with McDowell and Peach's (1959) calculations for

$$H_1^-(1s)^2 + H(1s) \rightarrow H(1s) + e + \sum_{n,l} H(nl)$$

where $\sum (nl)$ indicates a summation over all excited states. Comments are made on (a) the energy range of validity of the Born approximation, (b) the expected contributions of excited states to the measured cross section and (c) the equivalence of two hydrogen atoms and a hydrogen molecule for the electron loss process $\sigma_{10}$.

1.13 Fast Hydrogen Atoms $H_1^0$

The single electron loss from $H_1^0$ is the only "ionization" process investigated in this thesis. Fogel (1960) and Stier and Barnett (1956) have measured $\sigma_{01}$ in He, Ne and Ar. Their results agree well in shape but Fogel's results are consistently, at all energies from 3 to 40 Kev, about 25% lower than Stier and Barnett's data. Fogel's measurements in Kr and Xe are surprising. He finds that Kr and Xe are less efficient in removing the electron from the H atom than is Ar at energies less than 15 Kev. At all energies less than 40 Kev, Kr is less
Diagram 5.

PROJECTILE — $H_1^0$

TARGET — $H_2^0$

CROSS SECTION — $\sigma_{01}$
efficient than Ar while below 20 Kev, Xe is less efficient than Kr. No reason for this behaviour is given and intuitively it does not appear to be reasonable. These measurements have been repeated and the results presented in Chapter 4.

The first and only evidence of a fine structure for charge changing cross sections of hydrogen beams has been presented by Curran and Donahue (1960) for the process $\sigma_{01}$ in hydrogen gas. They suggest that possibly the energy increments used by other workers have been too large to show this structure, which is shown is diag. 5. This possible explanation of the failure of other data to show the fine structure has been investigated in Chapter 4.

Diag. 5 also shows the results of Fogel above 10 Kev are about 25% lower than Stier and Barnett's results, but are in good agreement with the general shape of Curran and Donahue's results. However below 10 Kev a marked divergence of the data into two rapidly diverging groups occurs. Curran and Donahue's results decrease rapidly to a value of $0.9 \times 10^{-17}$ cm$^2$/atom at 4 Kev while Stier and Barnett's curve shows a subsidiary broad maximum at about 5 Kev of $4.10^{-16}$ cm$^2$/atom. Stier and Barnett suggest (1956) that the lower energy peak corresponds to the H$_2$ molecule target remaining in the ground state while the higher energy peak corresponds to the H$_2$ molecule being left in excited or ionized states. Curran and Donahue
DIAG. 6.

PROJECTILE \( \rightarrow H^0 \)

TARGET \( \rightarrow H_2 \)

CROSS SECTION \( \rightarrow \sigma_{01} \)

\begin{tikzpicture}

\begin{axis}[
    xlabel=PROJECTILE ENERGY–KEV.,
    ylabel=CROSS SECTION–CM\(^2\)/ATOM,
    ymin=0.3,
    ymax=5,
    ymode=log,
    xmode=log,
    xmin=2,
    xmax=100
    ]

\addplot[\solid,mark options={solid}]
coordinates {
(2,0.5)
(4,1.2)
(6,2.5)
(10,3.0)
(20,2.8)
(40,2.2)
(60,1.8)
(100,1.5)
};
\addlegendentry{STIER & BARNETT 1956}

\addplot[\dashed,mark options={solid}]
coordinates {
(2,0.8)
(4,1.5)
(6,2.2)
(10,2.8)
(20,2.4)
(40,1.9)
(60,1.5)
(100,1.2)
};
\addlegendentry{FOGEL 1958}

\addplot[\dotted,mark options={solid}]
coordinates {
(2,1.0)
(4,1.8)
(6,2.5)
(10,3.0)
(20,2.8)
(40,2.2)
(60,1.8)
(100,1.5)
};
\addlegendentry{CURRAN & DONAHUE 1960}

\addplot[\dashed-dotted,mark options={solid}]
coordinates {
(2,0.7)
(4,1.4)
(6,2.1)
(10,2.7)
(20,2.3)
(40,1.8)
(60,1.4)
(100,1.1)
};
\addlegendentry{WHITTIER 1954}

\end{axis}
\end{tikzpicture}
suggest that their fine structure results from the capture of the electron, removed from the fast H atom, by the $H_2^-$ molecule to form $H^-_2$ which immediately dissociates into $H^-_1 + H^0_1$. Their suggestions are examined.

In an attempt to understand the vast differences between the two sets of data, the presence of non negligible populations of excited states, in particular the metastable $2S$ state, in the primary $H^0_1$ beam is considered and the possible effects of such states upon the measured cross section $\sigma_{01}$ is investigated. Accurate measurements of $\sigma_{01}$ have been made especially in the region below 10 Kev and possible competing collision processes are discussed in their relationship to the measurements of $\sigma_{01}$. The Born approximation calculations of Bates and Williams (1957) for the electron loss processes

$$H(1s) + H(1s) \rightarrow H^+_1 + e + \leq H(nl)$$

$$He(1s)^2 \leq He(n' l', n'' l')$$

are compared with experimental values. Similarities are noted in the Born approximation predictions for the loss cross sections $\sigma_{10}^{-}$ and $\sigma_{01}^{-}$.

The one electron capture by H atoms presents some problems in nearly all the gases studied. In hydrogen the measurements again fall into two groups as shown in diag.6. Stier and Barnett used an experimental method similar to Curran and Donahue and Whittier yet their results are
quite different and they agree well with Fogel's results. There appears to be no obvious connection between the experimental methods and the division of experimental results into two distinct groups. Curran and Donahue obtained evidence of a fine structure which is less prominent than similar structure shown in their measurements of $\sigma_{01}$. The present measurements of $\sigma_{01}$ have included a careful search for any such fine structure.

Fogel (1958) has measured $\sigma_{01}$ in all five inert gases, however his results are graphed on such a small linear scale together with the cross section $\sigma_{01}$, which is almost an order of magnitude larger than $\sigma_{01}$, that it is most difficult to ascertain his values within a factor of two. Stier and Barnett (1956) have measured $\sigma_{01}$ in He, Ne and Ar. In He and Ne their results are in satisfactory agreement above 11Kev, but below this energy there is a marked discrepancy as Stier and Barnett's values decrease with decreasing energy down to 8 Kev far more quickly than Fogel's values. The present values extend down to 2 Kev and give a clear indication of the manner in which $\sigma_{01}$ depends on energy. In Argon the cross sections suggest a subsidiary maximum at about 30 Kev, however neither Fogel or Stier and Barnett comment on its presence. The present measurements confirm its existence and attribute its origin to the removal of an additional electron from the target atom simultaneously with the single electron capture
by the primary atom, that is to a $00_1^/-2e$ collision.

Chapter 4 reports the present measurements of $\sigma_{01}$ in $H_2$ and the inert gases over the energy range 2 to 50 Kev. There are no theoretical predictions for $\sigma_{01}$.

Of the six charge changing cross sections discussed above only $\sigma_{10}$ has been measured with satisfactory agreement between different workers for all the inert gases and hydrogen. For the other five cross sections of the set $\sigma_{1f}$ it has been shown that there are numerous discrepancies, generally of unknown origin, between the results of different workers. The present investigation undertakes extensive testing of the consistency and correctness of the apparatus, measurement technique and results. An apparatus similar to that used by Fogel has been selected to measure the cross sections by the method of the growth with pressure of those primary beam particles which have changed charge in single collisions. A unified set of measurements of the six possible charge changing cross sections, $\sigma_{1f}$, for hydrogen beams has been determined to better than 3% relative accuracy. These measurements are shown to be of considerable value in the development of empirical relations to describe electron capture and loss processes and in the testing of available theoretical calculations.
1.2 Theoretical Predictions.

Quantum mechanical studies of charge changing collisions have been confined generally to collision systems of atomic hydrogen and helium by the availability of accurate and simple wave functions. Even for these simple systems the solutions to the scattering problem are only made by approximation methods of which detailed descriptions have been given by Hott and Massey (1950) and Bates (1962). This chapter presents a very brief account of the difficulties and the successes of the theoretical attempts to predict charge changing cross section values in agreement with experimental values.

In the high velocity region, generally above $10^8$ cm/sec, the approximation is made in which the perturbation causing the transition is taken to be the interaction potential. The electron loss processes generally do not involve an electron exchange and so have been treated quite successfully (Bates, 1962) by the Born approximation in a similar manner to that used for simple excitation processes. The electron capture process however, requires an electron exchange which makes the high velocity approximation physically and mathematically complicated.

Two facts cause concern in the use of the Born approximation for electron capture by protons in atomic hydrogen: (a) the interaction potential to be used and (b) initially no account had been taken of the non-
Electron capture cross section $\sigma_{10}$ for protons in hydrogen gas as a function of incident proton energy.

- the average experimental values for $\sigma_{10}$ in hydrogen gas from diag. 1.

- experimental values for $\sigma_{10}$ for atomic hydrogen. Fite (1960).

- Brinkman and Kramers (1930) using $v^{ep}$.

- Bates (1953). $\sigma_{10}$ for capture into the ground state only using $v^{ep} + pp$.

- Jackson and Schiff (1953). $\sigma_{10}$ for capture into all excited states using $v^{ep+pp}$.

- McCarron (1961). $\sigma_{10}$ for capture into all excited states using $v^{orthog}$.

orthogonality of initial and final wavefunctions. The influence of such facts upon the theoretical predictions and their agreement with experimental values is shown in diag. 7. Oppenheimer (1928) and Brinkman and Kramers (1930) used only the electron-proton interaction, $V^{ep}$, and neglected the proton-proton interaction, $V^{pp}$, since physically the proton-proton interaction determines only the path of the incident proton and should not influence the electron capture process. Later Jackson and Schiff (1953) and Bates and Dalgarno (1953) included $V^{pp}$ on the basis that its inclusion partially corrected defect (b) above and also gave better agreement with experimental values. Bates (1960), and later Bassel and Gerjuoy (1962) in a simpler treatment, proposed an effective interaction potential, $V^{\text{orthog}}$, which appears to correct for both the above defects (a) and (b). McCarroll (1961) has calculated the $H^+_1 + H_1^0$ electron capture cross section with interaction potentials, $V^{ep}$, $V^{pp+ep}$ and $V^{\text{orthog}}$. The cross section using $V^{\text{orthog}}$ tends to the cross section using $V^{ep+pp}$ at low energies, ~50 Kev, while at 1 Mev it tends to the cross section using only $V^{ep}$. Unfortunately there are no experimental data in atomic hydrogen to test these predictions for energies above 50 Kev, where the Born approximation is expected to be valid. Prior to 1960 it was common practice to assume that for high energy electron capture a hydrogen molecule was equivalent to two hydrogen atoms. On this
basis the theoretical predictions above have been compared in diag. 7, with the experimental data of Stier and Barnett (1956).

Tuan and Gerjuoy (1960) and Gerjuoy (1960) have noted that, while there is apparent agreement between the experimental data in molecular hydrogen and the theoretical predictions in atomic hydrogen, the comparison is improper for fundamental reasons. The equivalence of a hydrogen molecule to two hydrogen atoms for other charge changing processes is investigated in Chapter 4.

Electron capture by protons in helium is free from the complications due to the molecular nature of hydrogen. The interaction potential, \( V(ep+pp) \), has been used to calculate \( \sigma_{10} \) for capture into the ground state of hydrogen (Bransden, 1954) and into all excited states (Mapleton, 1961), the latter calculation agreeing well with the experimental values shown in diag. 17.

As the relative velocity of the colliding particles becomes less than about \( 10^8 \) cm/sec. the validity of the Born approximation for \( \sigma_{10} \) processes becomes doubtful due to an increase in the number of scattering matrix elements which become important. Attempts to take account of these elements are made in the distortion and second Born approximations but neither is remarkably successful in improving the agreement with experimental values at lower velocities.
CROSS SECTION-CM^2/ATOM

PROTON ENERGY - KEV.

CROSS SECTION FOR ONE ELECTRON CAPTURE IN COLLISION BETWEEN PROTONS AND HYDROGEN ATOMS.

- BRINKMAN & KRAMERS - BORN APPROX.
- BATES & DALGARNO
- GRYZINSKI - CLASSICAL APPROX.
- FERGUSON 1961 - without
- FERGUSON 1961 - with ALLOWANCE FOR ELECTRON MOMENTUM CHANGE - P.S.S. METHOD.
For these slower collisions considerable success has been achieved with the perturbed stationary state (P.S.S.) method in which transitions are considered to occur between states of the pseudo molecule formed by the projectile and target and the perturbation is taken to be the relative kinetic energy (Mott, 1931; Massey and Smith 1933). For the special case of the symmetric resonant one electron capture process for protons in atomic hydrogen, diag. 8 shows the failure of the Born approximation and the success of the P.S.S. method in obtaining good agreement with the experimental values of Fite et al (1960). At high velocities the rapid \( v^{-10} \) decrease of \( \sigma_{10} \) with velocity is attributed to the difficulty of transferring appreciable momentum to the "stationary" electron in the target atom from the fast projectile. In the P.S.S. approximation Ferguson (1961) has made allowance for this change which becomes noticeable at energies as low as 5 Kev.

For more complex atoms approaches have been made in terms of Slater orbitals (Gurnee and Magee, 1957), hydrogenic orbitals (Rapp and Ortenburger, 1960) and semi-empirical orbitals (Dalgarno, 1958; Iovitsu, 1960; Rapp and Francis, 1962), all of which yield a form for the symmetric resonant cross section, \( \sigma_{10} \) of

\[
\frac{\sigma_{10}}{a} = a_1 \ln v + a_2
\]

where \( a_1 \) and \( a_2 \) are functions of the ionization potentials of the colliding atoms. This function gives good
Diagram 9. \( \sigma \) for \( H^+ \) in inert gases.
agreement with experimental data (Rapp, 1962).

The extension of the P.S.S. method to the asymmetric non-resonant one electron capture process has not been too successful because of (a) the approximate substitution of atomic orbitals for molecular orbitals in the expansion of the total wave function and (b) the requirements of the principle of detailed balance and statistical weight considerations of possible transition states (Takayanagi, 1955; Gurnee and Magee, 1957; Rapp and Francis, 1962). The best success of such approaches is seen in diag. 9 where the calculations of Rapp and Francis are compared with Hasted's (1952, 1956) data. The theory has only partial success in predicting (a) the qualitative slopes of the $\sigma(v) \rightarrow E$ curves, (b) the approximate positions of the cross section maxima and (c) the rough absolute values of the cross sections. In Chapter 4 the present values of $\sigma_{10}$ are compared with the predictions of Rapp and Francis (1962) and the general form of their cross section versus energy curves is compared with that for other capture cross sections $\sigma_{11}$ and $\sigma_{01}$.

The simplest approximation of an inelastic collision is that of a classical binary encounter between two charged impenetrable spheres (J.J. Thomson, 1912). Gryzinski (1957, 1959, 1963) utilized the similarities between Coulomb and gravitational fields to apply the general dynamical description of classical binary
encounters between planetary bodies, as developed by Chandrasekhar (1941), to inelastic atomic collisions. His calculations have given surprisingly good agreement with experimental values for processes such as electron and proton ionization of inert gases, and the stopping of protons in gases. However his predictions of the one electron capture cross sections in H, H₂ and He generally show poor agreement with experimental values (see diagrams 7 and 8) and are quite inferior to the more accurate quantum mechanical predictions. Gryzinski's calculations are extended to all the inert gases in Chapter 4 of this thesis. The main asset of his predictions are their ease of calculation.
1.3 Factors entering into the Behaviour of Collision Cross Sections.

It has been noted that reasonably accurate theoretical predictions are restricted to H and He collision systems. The extension of the P.S.S. method to the asymmetric non-resonant one electron capture process for protons in the inert gases yields little better than an empirical description of the process. The predictions for other charge changing collisions are almost non-existent. As a result recourse is taken to qualitative descriptions of such processes. The framework into which these qualitative descriptions are most often put is that of the adiabatic theory which was first suggested by Massey (1949) to describe the qualitative aspects of collisions in the very low energy region.

The Adiabatic Theory.

If in an atomic collision the projectile particle approaches a target slowly enough for the electronic motion to adjust itself to the perturbation, then a transition will be unlikely. The characteristic time of the collision can be written as \( \frac{a}{v} \) where "a" is a distance of atomic dimensions and v is the relative velocity. The time involved in the electronic transition is \( \frac{h}{\Delta E} \) where h is Planck's constant and \( \Delta E \) is the change in internal energy. The above condition for a transition to be unlikely can then be written as the inequality,
\[
\frac{a}{v} \gg \frac{\hbar}{\Delta E} \quad \text{or} \quad \frac{\hbar v}{a \cdot \Delta E} \ll 1 \quad \ldots \ldots \ldots 1.31
\]

which in the familiar statement of the adiabatic criterion and defines the adiabatic region.

As the velocity rises out of the adiabatic region and the inequality approaches an equality, a transition becomes relatively likely. This increase gives rise to the often used adiabatic maxima rule which says that the cross section will be in the vicinity of its maximum when the collision times and transition times are comparable, a statement represented by the approximate equality:

\[
\frac{\hbar v}{a \cdot \Delta E} \simeq 1 \quad \text{or} \quad E_m \simeq 3 a^2 M (\Delta E)^2 \quad \ldots \ldots 1.32
\]

where \(M\) is in proton masses

"\(a\)" is in Angstrom units

\(E_m\) and \(\Delta E\) are in electron volts.

There is an alternative way of viewing these statements. Suppose one makes a Fourier expansion of the time-dependent energy of interaction of the projectile and target particles. If the amplitude of the Fourier component, \(\omega_\xi\), defined by \(\hbar \omega_\xi = \Delta E\), is small, then the probability of a transition is small. As the frequency dependence of the interaction potential is changed by changing \(v\), and the amplitude of the \(\omega_\xi\) component reaches its maximum, then the transition probability will also reach its maximum.

The peak of the cross section often can be observed and equation 1.32 used in such cases to find the quantity
"a" which is often referred to as the interaction distance for the reaction. This name is consistent with the manner in which it was introduced, and it seems reasonable to ascribe to it the properties characteristic of the collision process. In categorizing the great amount of data available on charge exchange, ionization etc. it has been found that "a" calculated in the manner above tends to be constant for a large number of collision processes and is therefore independent of the participating members of the collision. Thus according to Hasted (1960), a \( \approx 7^{\circ}\text{A} \) for single charge exchange collisions, while Fogel (1959) finds a \( \approx 1.5^{\circ}\text{A} \) for double charge exchange. Hasted (1962) recently showed that a more general grouping can be made for these processes. He considers the energy defect at \( R = 0.92 \left( Z_1 + Z_2 \right)^{1/3} \) taking into account Coulomb or polarization energies of the product particles for reactions of the type \((n + 1) \rightarrow (n + 1 - m)m\). Using the data of many workers for processes of the electron capture types 10/01, 00/11 and 10/12, he showed that the product "ma" is approximately a constant equal to \( 7^{\circ}\text{A} \).

In order to gain further insight into the meaning of the interaction distance Drukarev (1960) has presented an argument which associates it with the momentum imparted in the collision. By making use of the relation, \( \frac{P_0^2 - P_1^2}{2m} = \Delta E \), and observing that \( |P_0 - P_1| \ll P_0 \) for most atomic
collisions he obtains for the momentum imparted at the angle $\theta$

$$q(\theta) = |P_0 - P_1| = \left[\left(\frac{\Delta E}{v}\right)^2 + 4P_0^2 \sin^2\left(\frac{\theta}{2}\right)\right]^\frac{1}{2} \quad \ldots \ldots 1.33$$

where $P_0$ and $P_1$ are the momenta in the centre of mass system before and after the collision respectively. This becomes for small angle forward scattering, $q(\theta) = \frac{\Delta E}{v}$. In particular if $v = v_m$, then $q_m = \frac{\Delta E}{v_m}$ where $v_m$ is the value of $v$ for maximum cross section. Combining this with the adiabatic maximum rule, equation 1.32, he obtains

$$a = \frac{h}{q_m} \quad \ldots \ldots \ldots \ldots \ldots 1.34$$

Thus he suggests that one may consider processes having the same "a" as processes having a characteristic momentum transfer in the forward direction.

In applying the adiabatic maxim rule (equation 1.32) or envisioning the qualitative behaviour of the cross section one must choose a value of $\Delta E$ that is really characteristic of the reaction. If one selects it to be $\Delta E_\infty$, that is the value of $\Delta E$ at infinite separation of the colliding particles, the data for a given process will usually seem consistent with the adiabatic theory. However, when an attempt is made to categorize the many reactions studied and to look for systematic consistencies, there is an indication that $\Delta E$ must be chosen at the separation at which the transition occurs or is most likely. This is not unreasonable in view of the manner in which $\Delta E$ was introduced into the theory. Thus, the interaction energies
DIAG. 10. POSSIBLE INITIAL AND FINAL INTERACTION POTENTIALS FOR PARTICLES IN AN INELASTIC COLLISION.
may appear similar to one of the plots in Fig. 10 and may be either larger or smaller than $\Delta T_\infty$. In view of the fact that few data are available for interaction energies as a function of $R$, the internuclear separation, it has been customary to use $\Delta T_\infty$.

The functional behaviour of the cross section in the adiabatic region is not forthcoming from these qualitative considerations, but one might expect the cross section to depend on the adiabatic parameter $k \frac{1}{a \Delta E}$ explicitly. Massey (1949) suggested a power law dependence or an exponential of the form

$$\sigma = \sigma_0 \exp \left[ -\frac{k a \Delta E}{\hbar \nu} \right] \cdots 1.35$$

A large number of charge exchange cross sections have been successfully fitted to this functional form, and examination of the form of curves from calculations using the P.S.S. method also suggests the exponential behaviour in the adiabatic region. The preceding equation can be written in the form

$$\sigma = \sigma_0 \exp \left[ -\frac{K}{E^2} \right] \cdots 1.36$$

where $K = ka(\Delta E m^2)$.

For those processes which exhibit exponential behaviour a semi-log plot of $\sigma$ versus $E^{-\frac{1}{2}}$ will give a straight line of slope $(-K)$. Hasted (1960) has made such plots for many cases of charge exchange. He has plotted the slopes obtained from those graphs as a function of $(\Delta E m^2)$ and found $(ka)$ is a constant for the cases plotted i.e. he obtained a straight line. Hasted (1961) has
recently used the exponential rate of rise for some cross sections, coupled with the fact that \((ka)\) was found to be nearly constant, to calculate the interaction distance "a" for some processes. One can reason that if \((ka)\) is a constant for some reactions and if "a" is constant for those same reactions, then \(k\) is a constant. Then for other reactions, assume \(k\) is the same constant and use measured values of \((ka)\) to compute "a". This, then, has been used as an alternative to the adiabatic maximum rule for obtaining values of "a".

Gerjuoy and Krall (1961) have recently presented a time-dependent calculation for the excitation of \(H\) atoms by protons. Their theory predicts that the maximum cross section should occur at an incident velocity \(v_m = \left(\frac{6 \cdot e^2 a_0 \omega}{\hbar}\right)^{\frac{1}{2}}\). This result, which makes \(v_m\) proportional to the square root of \(E = \hbar \omega\), is in direct conflict with the adiabatic maximum rule (equation 1.32), which requires that \(v_m\) be proportional to \(\Delta E\). An unpublished classical theory due to Fan is mentioned by Hasted (1960), which also has \(v_m\) proportional to \((\Delta E)^{\frac{3}{2}}\).

Data of Fogel and co-workers (1959) on double charge exchange show multiple maxima when their ion source is operated in a fashion which will yield metastable ions, but show only one maximum for known ground state ions. If the known energies of the metastable ions are used and if the interaction distance "a" is assumed to be independent
of the excitation of the ion, then the adiabatic criterion (equation 1.32) correctly predicts the relative position of the maxima. These seem to be the only data which substantiate one type of dependence of $v_m$ on $\Delta E$ above another, since application of equation 1.32 to individual processes leaves open the choice of "a". Even in the above case involving metastable ions, the assumption of equal "a"'s may be unjustified. One must also note that $\Delta E_\infty$ was used in Fogel's comparisons.

The above description of the results and formulations of the adiabatic hypothesis has been detailed because it forms the framework for the qualitative analysis of the results of this thesis.
Chapter 2. The Experimental Method.

2.1 Principle of the Measurement.

The composition of an atomic hydrogen beam, which has three possible charge states, as it passes through a target gas is given by the set of equations

\[
\frac{dN^+}{d(\text{nl})} = -(\sigma_{\text{f}0} + \sigma_{\text{t}1})N^+ + \sigma_{\text{f0}}N_0 + \sigma_{\text{t1}}N^- \quad \ldots \quad (a)
\]

\[
\frac{dN_0}{d(\text{nl})} = + \sigma_{\text{t0}}N^+ -(\sigma_{\text{f0}} + \sigma_{\text{t1}})N_0 + \sigma_{\text{f0}}N^- \quad \ldots \quad (b)
\]

\[
\frac{dN^-}{d(\text{nl})} = + \sigma_{\text{t1}}N^+ + \sigma_{\text{f0}}N_0 -(\sigma_{\text{f0}} + \sigma_{\text{t1}})N^- \quad \ldots \quad (c)
\]

where \(N^+\), \(N_0\) and \(N^-\) are the numbers of protons, neutral hydrogen atoms and negative hydrogen ions in the projectile beam at any point in the traversal of a target gas.

\(n\) is the number of target gas atoms/cm\(^3\)

\(l\) is the beam path length in the target gas

\(\sigma_{\text{if}}\) is the cross section for the collision in which a particle of charge \(i\) is changed to a particle of charge \(f\).

Allison (1958) has fully discussed these equations and their solutions, from which it is readily shown that

\[
\frac{N_f}{N_i} = \sigma_{\text{if}}(\text{nl}) + a_{\text{if}}(\text{nl})^2 + b_{\text{if}}(\text{nl})^3 + \text{higher powers of (nl)}
\]

and then

\[
\frac{d}{d(\text{nl})} \frac{N_f}{N_i} = \sigma_{\text{if}} + 2a_{\text{if}}(\text{nl}) + 3b_{\text{if}}(\text{nl})^2 + \ldots \ldots \ldots (2)
\]

where the \(a_{\text{if}}\) and \(b_{\text{if}}\) are complicated functions of the six possible cross sections \(\sigma_{\text{if}}\). The method of measuring the cross section, \(\sigma_{\text{if}}\), that follows from equation (2) is to
EINZEL LENS
ACCELERATING LENS
GLASS INSULATING COLUMN

EINZEL LENS
ACCELERATING LENS

GLASS INSULATING COLUMN

4" DIFFUSION PUMP
250 L/sec.

IONIZATION GAUGE

BEAM DEFINING APERTURES AND FARADAY CUPS

DIAG.II.
BEAM FOCUSING AND ACCELERATING SECTION.
DIAG. 12. PROJECTILE BEAM ANALYSING MAGNET.
study the dependence of the growth of $N_f$, from a constant $N_i$, on the quantity $(n\ell)$. This dependence is seen to be linear, that is the change of charge of the projectile results only from a single collision, when $\frac{2a_{if}(n\ell)}{\sigma_{if}} \ll 1$. This condition of linearity of $\frac{dN_f}{d(n\ell)}$ is dependent upon the nature of the projectile and target atoms and the projectile energy. Therefore for every individual cross section measurement a graph of the growth of collision products with increasing target gas number density, $(n\ell)$, must be drawn to separate the linear and curved components. Examples of such graphs for each $\sigma_{if}$ are given in the Chapter 4 after a discussion is given of the apparatus required for their determination.

2.2 APPARATUS

The above equation (2) implies that for the accurate measurement of a given cross section one must construct an apparatus consisting of the following component parts:

(1) a beam of ions (or atoms) which is of known charge to mass ratio, intensity, direction and energy.

(2) a target gas of known purity and number density.

(3) a means of analysis and detection to determine the extent to which the projectile beam properties mentioned in (1) are changed by (2).

Diagrams 11, 12 and 13 show the apparatus which was constructed for this investigation. The characteristics
of the electrodeless discharge type ion source and its extracted positively charged ion beam have been extensively investigated and are reported separately in chapter 7. A maximum beam current of 280 μA is contained within a 10° angle of divergence for an ion extraction energy from the source of 2.5 Kev. The basic requirement of the focusing system was to form the ions into a parallel beam of 2mm. diameter at a distance of 60 cm. from the source and, at this image position, to permit the ions to arrive with known but controllable energy within the range 2 to 50 Kev.

The acceleration of a 2.5 Kev beam to 50 Kev can readily be accomplished by a single lens formed by two cylindrical electrodes having a voltage ratio of 20/1 (Spangenburg, 1948). At 50 Kev potential difference, this electrode configuration presents a very strong lens to a 2.5 Kev beam. However by positioning the lens such that the beam originates from an object just beyond the first focal point, the lens focuses the ions to form an image at the required distance on the far side of the lens. At ion energies less than 50 Kev the lens is weaker and no longer brings the ion beam to the same image point. To accommodate this change in focal properties with voltage ratio, the object distance from the ion source to the lens has to be increased. This can be accomplished by either physically moving the position of the accelerating lens.
or by interposing another lens system between the ion source and the accelerating lens which has the property of effectively increasing this distance.

The first alternative was rejected because of the difficulty of physically moving the lens from outside the vacuum system. The second alternative was readily set up by using an Einzel lens (Liebmann, 1949) preceding the accelerating lens. The central electrode potential of the Einzel lens is variable from 0 to -20 Kv relative to the reference potential of the ion source canal. The 2.5 Kev ions from the source are thus focussed so that they appear to the accelerating lens to originate from a point beyond the ion source. At energies greater than 50 Kev the accelerating lens has a very short focal length and the Einzel lens has to be removed from the system. For energies less than 8 Kev it was convenient for ease of focussing to reduce the initial energy of the ions extracted from the source. Aberrations of the beam image were greatly reduced by restricting the beam divergence from the ion source to 6 degrees.

The image position of this lens system coincided with the object position of the first momentum analysing magnet. This magnet was designed as an inflection type, second order direction focusing instrument (Kerwin, 1949). Hintenberger (1948) has shown that a first order focusing field can be improved to give perfect focusing by the
shaping of the magnetic field boundary. However by drawing a tangent through the inflection point of the perfect focusing boundary a straight line boundary is obtained which gives second order focusing. In practice the fringing field of the magnet restricted the attainment of other than first order focusing. The design and performance details of this electromagnet and its power supply are given in appendix 2. With beam defining apertures of 1mm. diameter at the object and image positions of this magnet, an energy resolution of \( \frac{1}{10^8} \) was obtained.

The beam was further collimated by another 1.0 mm. diameter aperture placed 50 mm. in front of the entry canal to the collision cell. A pair of electrostatic deflector plates (see diagram 13) placed immediately in front of the entry canal enabled the neutral and charged particle contents of the projectile beam which entered the collision cell to be determined. The collision cell was geometrically 10.04 cm. long with cylindrical entry and exit canals of 1.5 and 2.0 mm. diameter respectively and 5.0 mm. long.

The projectile beam emerging from the collision cell was momentum analysed by a 10 cm. radius of curvature, deflection magnet, whose design and performance details are given in appendix 2. The object and image positions of this magnet coincided with the exit canal of the
collision cell and the entrance aperture of the detectors respectively. The beam optics were checked at many points between the ion source and the detectors by (a) visual observation of the image formed on an earthed zinc sulphide screen and by (b) the considerably more accurate method of the comparison of the beam profiles as measured by a Faraday cup with a fixed entrance aperture. These measurements are fully discussed in the next chapter.

The neutral particles were detected by secondary electron emission from an aluminium surface. The characteristics of the neutral particle detectors are discussed fully in chapter 10. The positively and negatively charged particles were collected in Faraday cups. The outputs from the three charge state detectors were amplified by three electrometers; an Ecko vibrating reed electrometer, model N616 A, provided a full scale deflection (F.S.D.) of $3.10 \times 10^{-15}$ amps with noise less than $0.7 \times 10^{-15}$ amp; a Keithly electrometer, model 603, with F.S.D. of $2.5 \times 10^{-14}$ amps with noise less than $0.1 \times 10^{-14}$ amps and a General Radio electrometer, model 1230 A, with a F.S.D. of $3.0 \times 10^{-13}$ amps with noise less than $0.5 \times 10^{-13}$ amp. The noise limitations were determined by the effectiveness of the shielding for the radio frequency fields from the ion source. Therefore with eight orders of magnitude difference between the projectile beam current ($10^{-6}$ amp) and the limit of detection of the
 electrometers even the most improbable collisions with a cross section of the order of $10^{-20}$ cm$^2$ should be readily detectable.

Liquid air cooled surfaces (of areas about 80 cm$^2$) were located inside the collision cell and in the pump baffle immediately below the collision cell. Three baffled oil diffusion pumps of speeds 250 l/sec. were adequate for gas throughputs from the ion source of approximately 200 l/sec and 230 l/sec from the collision cell and were able to maintain an overall system pressure better than $1 \times 10^{-6}$ mmHg.

Relative pressure measurements were made with A.W.A. type AV26 ionization gauges and Edwards Penning type gauges. The control circuit constructed for the ionization gauge, is shown in appendix 2.

The ion beam accelerating and source extraction voltages were obtained from Hursant 50 Kev and Fluke 5 Kev supplies, both stabilised to better than 0.1%. The beam energy is equal to the sum of these accelerating and extraction voltages plus the source plasma potential. The latter has been determined in Chapter 7. The former were determined by measuring the voltage drop across a resistor in a long chain of 100x1 M$\Omega$ with a Fluke differential voltmeter. The resistor values were measured to better than 1% with the G.R. electrometer.

The method of preparation of the $H^0$ and $H^-$ beams is a
slight variation on the usual methods. The $H_1^0$ atoms were formed by single electron capture by protons in collision with the residual gas atoms along the beam drift space between the exit focus of the 30 cm. magnet and the deflector plates $P_1$ and $P_2$ (see diag. 13). The original $H_1^+$ currents were of the order of $3 \times 10^{-6}$ and the resultant $H_1^0$ flux about $10^9$ atoms/sec.

The $H_1^-$ ions were formed by double electron capture by protons in collision with the residual gas atoms along the drift space between the main accelerating lens and the entry focal point of the 30 cm. magnet. The 1.0 mm. beam defining aperture, placed at the entry focal point of the magnet, was held in a large disc which completely sealed the 4" vacuum tubes and so the residual gas pressure prior to this aperture could be raised to give larger $H_1^-$ beam currents $\sim 10^{-10}$ amps. for all ion energies.

It is shown in Chapters 3 and 4 that this method of preparation of the $H_1^0$ and $H_1^-$ beams and the resulting small beam currents gave cross sections as accurate as those determined using the much larger proton currents.
Chapter 3

Accuracy and Validity of the Measurements

The accuracy and validity of the cross section measurements have been investigated in the following comprehensive experiments, several of which have been partly based on, or initiated by, considerations such as those given by Allison (1958), Fogel (1960) and Stier and Barnett (1956). The apparatus and symbols referred to below are those shown in diag. 13.

3.1 Detector Aperture.

A detector entrance aperture diameter of 10 mm. has been shown to be sufficiently large to ensure the collection of at least 99.9% of all the beam particles. This result is derived from the observation of the beam profiles as measured by the detector response which results from sweeping the beam across the detector aperture by variation of the magnet field B (the 10 cm. radius of curvature magnet of diag. 13). From the geometry of the beam path through the magnet B, a change in magnetic field strength can be related to the image movement and to the width of the beam at any given fraction of its maximum amplitude. For all the tests described below the width of the beam profile at 0.1% of its maximum amplitude was less than 10 mm. and the beam profile always showed a flat top, which indicated that at least 99% of the beam was collected.
by the detector. Whilst a detailed analysis of the beam profile may yield further information on the scattering process this was not attempted as the present purpose was solely to ensure adequate collection of the beam particles.

(a) Identical beam profiles were obtained at positions \( D_3 \) and \( D_0 \) by variation of the electric field between plates \( P_1 \) and \( P_2 \) with the entry and exit canals of the collision cell removed. Hence any scattering of the beam by collision with the residual gas molecules along the beam path was shown to have a negligible broadening effect on the primary beam profile.

(b) Identical beam profiles were obtained at the \( D_1 \) and \( D_2 \) positions by reversing the direction of the magnetic field \( B \). Interchange of detectors \( D_1 \) and \( D_2 \) had no effect on the beam profiles. The beam profiles at positions \( D_1 \) and \( D_2 \) were slightly wider than that at the \( D_0 \) position due to the defocusing action of the magnetic field.

(c) Test (b) above was performed at beam energies of 2, 10 and 50 Kev, which represent the upper and lower energy limits used for cross section measurements and also the standardization energy (10 Kev) for calibration of the experimental values (see Chapter 4). The beam optics were such that a parallel beam was produced for all the above energies. The results of test (b) above were applicable over the entire energy range from 2 to 50 Kev.

(d) The positioning of the entry and exit canals of
diameters 1.5 and 2.0 mm. respectively on the collision cell had no detectable effect on the beam profile. Such effects may have arisen from further collimation of the beam or defocusing of the beam due to a build up of charge on the canal walls.

(e) As the collision cell gas pressure was increased from its minimum value of $2.10^{-6}$ mm.Hg. to $1.10^{-4}$ mm.Hg whilst the gas pressure external to the cell was maintained below $1.10^{-6}$ mm.Hg., the $H_1^+$ beam profile was broadened by about 10% at 0.1% of its peak value. The $H_1^-$ beam, formed by the double electron capture by the $H_1^+$ beam in the collision cell, had a profile width 25% greater than that of the incident $H_1^+$ beam at 0.1% of the peak beam current. But this width was then only about 3mm. These beam profiles are shown in diagram 14.

(f) Tests (a) to (e) were repeated for projectile beams of $H_1^-$ and test (e) was repeated on the collision products from $H_1^0$ primary beams. In all cases the beam profile width at 0.1% of the peak beam current was less than 10 mm.

Examination of the profiles of $H_1^0$ beams would require a movable detector. This was not attempted. However one may expect that $H_1^0$ beam profiles to be no broader than $H_1^-$ beam profiles when both beams are formed from the same parent $H_1^+$ beam by the capture of one and two electrons, respectively, because in the latter process strong Coulomb
forces are operative between the collision products.

3.2 Impurities in the Projectile Beam.

The primary $H_1^+$ beam always contains some impurity $H_1^0$ and $H_1^-$ particles, whose possible origins are as follows.

(a) The protons may gain either one or two electrons by collision with the edges of the beam defining apertures $A_2$ and $A_3$ or with the walls of the entry and exit canals. There appears to be no reported investigation of the reflected primary particles with energies greater than 2 Kev from a metal surface. However Massey and Burhop (1952) report that for low energy (400 ev) protons incident on a mercury surface (i) the coefficient for production of $H_1^-$ is about $10^{-5}$ per incident proton and (ii) the energy of the emergent $H_1^-$ is appreciably lower than that of the incident proton. If one assumes that similar results were applicable to the present situation then result (i) would have serious implications since, for the present cross section $\sigma_{it}$ measurements, one is required to measure $H_1^-$ currents of the order of $10^{-5}$ of the incident $H_1^+$ current. But result (ii) implies that these surface-formed $H_1^-$ would most probably not enter the detectors after their momentum analysis by magnet B.

To minimize such effects the beam defining apertures were machined with a knife edge to minimize the surface area on which the proton beam could capture electrons and continue without appreciable angular deviation from the
(b) The primary proton beam may capture one or two electrons in collisions with the residual gas molecules along the beam path between the first beam defining aperture $A_2$ (after which point the beam follows a linear path) and the entry canal of the collision cell. The residual gas pressure outside the collision cell does not change within the limits of detection (less than $1 \times 10^{-6}$ mm. Hg.) when the collision cell gas pressure is increased to its maximum value used in the cross section measurements of 2 $\times 10^{-4}$ mm. Hg.

The numbers of $H^0_1$ and $H^-_1$, formed in the manner described above in paragraphs (a) and (b), are independent of the collision cell gas pressure. Effectively all of the $H^0_1$ and $H^-_1$ so formed will pass through the collision cell, irrespective of the gas pressure, into the detectors because, in the present "single collision" experiments, not more than 1% of the primary beam undergoes a collision in the gas cell. Further, since the method of the determination of a cross section is to measure the linear rate of growth of the collision products with pressure, the numbers of $H^0_1$ and $H^-_1$, which are produced in the above manner and which are pressure independent, will appear as a constant term which does not influence the slope of the collision product versus pressure graph. For the measurement of very small cross sections the total increase
of the number of collision products over a given pressure range may not be large compared with the number of impurity \( H_1^0 \) and \( H_1^- \) particles, which, therefore, make the determination of the slope of the collision product growth curve more difficult.

3.3 Pressure Dependent Effects arising from \( H_1^0 \) and \( H_1^- \) Impurities.

The impurity \( H_1^0 \) and \( H_1^- \) particles, discussed in the previous section 3.2, will traverse the collision cell and so may collide with the target gas atoms to give rise to linearly pressure dependent collision products.

(a) The errors arising from those neutral atoms, which are formed in the residual gas or on the edges of the beam defining apertures, but not those from the walls of the entry canal, are simply determined by electrostatic deflection of all charged particles from the beam prior to their entry to the gas cell. Thus the remaining "primary" beam is now a neutral atom, \( H_1^0 \), beam from which the collision products \( H_1^+ \) and \( H_1^- \) may be observed as a function of the gas pressure. The fraction of such \( H_1^- \) impurities as compared with those \( H_1^- \) ions produced from the \( H_1^+ \) will thus depend upon (i) \( \sigma_{10} \) for \( H_1^+ \) in the residual gas of the vacuum system and (ii) the relative values of \( \sigma_{17} \) and \( \sigma_{67} \) in a given target gas, whilst each cross section depends upon the projectile energies. This type of investigation has been made for every cross section measurement. Table 3
<table>
<thead>
<tr>
<th>Target Gas Pressure</th>
<th>$P_1 \cdot 10^{-5}$ mmHg</th>
<th>$H^+ \cdot 10^{-10}$</th>
<th>$H_1 \cdot 10^{-13}$</th>
<th>Detector currents - amps</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>0</td>
<td>9.9 \cdot 10^{-7}</td>
<td>1.9</td>
<td>17.0</td>
</tr>
<tr>
<td>(b)</td>
<td>2.9 \cdot 10^{-14}</td>
<td>2.9 \cdot 10^{-14}</td>
<td>0.65</td>
<td>0.02</td>
</tr>
<tr>
<td>(c)</td>
<td>8.93 \cdot 10^{-7}</td>
<td>8.93 \cdot 10^{-7}</td>
<td>61</td>
<td>313</td>
</tr>
<tr>
<td>(d)</td>
<td>5.61 \cdot 10^{-14}</td>
<td>5.61 \cdot 10^{-14}</td>
<td>0.65</td>
<td>1.6</td>
</tr>
</tbody>
</table>

Table 3

<table>
<thead>
<tr>
<th>Deflection Voltage between plates $P_1$ and $P_2$</th>
<th>$V$ volts</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>0</td>
</tr>
<tr>
<td>(b)</td>
<td>250</td>
</tr>
<tr>
<td>(c)</td>
<td>0</td>
</tr>
<tr>
<td>(d)</td>
<td>250</td>
</tr>
</tbody>
</table>
shows typical values of the detected currents for the case of 10 Kev $H^+$ in hydrogen gas, for which the neutral atom impurities discussed above give rise to a 0.5% error in $\sigma^{-}$. 

(b) The negative ions, formed in the primary beam before the electrostatic deflector plates, may lose electrons to form $H^0$ or $H^+$. A method of determination of this error is not possible in the manner of paragraph (a) because the electric field will deflect the $H^-$. However the fraction of $H^-$ present in the beam is so small (in table 3 it is $2.10^{-5}$ of the primary $H^+$ beam) that, even if all these $H^-$ changed charge to either $H^0$ or $H^+$, the increase in the fractions of $H^+$ or $H^0$ would not be detectable.

(c) An estimate of any errors arising from those neutral atoms which may be formed on the entry canal walls was attempted by increasing the diameter of the entry canal from 1.5 to 2.0, then 3.0 mm. (for a beam diameter of 1.0 mm.). The minimum gas pressure within the cell was unchanged by increasing the entry canal diameter since the cell had a separate pumping line. This increase of entry canal diameter had no detectable effect upon the readings presented in Table 3. Therefore an upper limit may be placed upon the number of neutrals formed on the entry canal walls of 2% of the number of $H^0$ formed by collision of the primary $H^+$ with the residual gas along the beam path. Less than 1% of these surface-formed $H^0$ atoms will
suffer a collision with the target gas so that the fraction of H$^-_1$ ions formed from these H$^0$ will be negligible compared with that formed from the H$^+_1$ primaries in collision with the target gas at its minimum pressure.

3.4 Other Considerations.

The use of liquid air cooled surfaces, both inside and outside the collision cell, to reduce the minimum gas pressures resulted in substantial reductions in the number of particles which had changed charge. This observation supports the belief that, for the possible errors discussed above, the contributions from gaseous collisions are much greater than those from surface collisions.

The beam profile study of section 3.1 indicated that the collision with the walls of the exit canal was most unlikely for those primary particles which had changed their charge by collision with the target gas. However for completeness of this study the possibility was investigated by increasing the exit canal diameter from 2.0 to 2.5, 3.5 and 10 mm. For a constant minimum gas pressure in the collision cell, these changes in the exit canal diameter had no detectable (i.e. less than 0.5%) effect on the collected currents of all charge states.

For a 10 Kev H$^+_1$ beam emerging from the collision cell (in which the gas pressure was about 10$^{-4}$ mm.Hg.) identical beam currents were collected by a Faraday cup placed alternately at (a) position D$_3$, which was 4 cm. after the
exit canal of the collision cell and (b) at the detector $D_0$ (see diag. 13). There is therefore a negligible loss of primary beam particles between the exit canal and the detectors. This result may naturally be extended to the beams of collision products which emerge from the collision cell. The gas pressure in the region between the collision cell and the detectors was measured to be the same as that pressure near the beam path before the gas cell. Table 3 shows that, for the case of 10 Kev $H^+$ traversing the background gas, for which the total charge changing cross section is of the order of $10^{-15}$ cm$^2$/atom, there is approximately a 0.01% loss of protons from the primary beam. Diag. 13 shows that the number of gas molecules/cm$^2$ of beam path before the gas cell is approximately $\frac{1}{4}$ of the same quantity after the gas cell. The largest total charge changing cross section is estimated to be of the order of $5.10^{-15}$ cm$^2$/atom for $H^-$ for which 0.2% of the beam would be lost. This small loss from the beam would not be detectable.
Experimental Results and Discussion.

4.1 Gross Section Determination.

It has been shown in Chapter 2 that the cross section, $\sigma_{\text{if}}$, is determined from the relationship

$$\frac{d}{d(N_l)} = \sigma_{\text{if}} + a_{\text{if}}(N_l) + b_{\text{if}}(N_l)^2 + \ldots \ldots \ldots (2)$$

The $N_i$ and $N_f$ are measured absolutely with electrometers which had been calibrated against a Fluke differential voltmeter. The quantity $(N_l)$ is found as follows. It is assumed that $l$ remains constant during the increase of $n$ which is made for the cross section measurement. Proof of this assumption requires the measurement of the gradient of $n$ along the length, and in the vicinity, of the entry and exit canals. This measurement would be made difficult by the baffling action of the beam defining aperture and the electrostatic deflection plates (see diag. 13) on the pumping speed in the vicinity of the entry canal. However the assumption of a constant value for $l$ is well supported by the growth of collision product, $N_f$, with $(N_l)$ relationship (shown below) which, at low $(N_l)$ values for all target gases used, was linear within 2%. When the contributing errors to this 2% figure are considered below it is seen that $l$ would vary by far less than 2%.

The number of gas atoms/cm$^3$ along the beam path, $n$, was not measured absolutely. Only relative values, $n'$,
Diagram 15(a). Growth of $H_0$ from a 10 KeV $H_1^+$ beam in hydrogen.

Diagram 15(b). Growth of $H_1^0$ from 2 KeV $H_1^+$ beam in argon.
were measured with an ionization type gauge. Thus from
the linear region of the relationship (2) above,

\[ \sigma_{ef} = \frac{1}{N_i} \cdot \frac{dN_f}{d(nl)} \cdot \frac{1}{c} \]

\[ \text{(3)} \]

where \( c = \frac{\Delta(nl)}{\Delta n} \)
and is a constant of the present apparatus for a given
target gas.

Diag. 15(a) shows one of the best examples from the
present work of the growth of \( N_f \) with (nl), in particular
of the growth of \( H_1^0 \) from a 10 KeV \( H_1^+ \) beam incident upon a
hydrogen gas target. Such a large number of experimental
points was taken only during the early part of this study
and the first few measurements of a given \( \sigma_{ef} \) in each
target gas. Diag. 15(b) shows a more typical growth of
collision product graph for the case of the growth of \( H_1^0 \)
from 2 KeV \( H_1^+ \) in argon. For all \( \sigma_{ef} \) determinations, the
ratio \( N_f/N_i \) rarely exceeded 1% and \( n' \) was varied by at
least a factor of 50. The error in determining the slope
of the linear relationship of \( \Delta \left( \frac{N_f}{N_i} \right) \) to \( \Delta n' \) in diag. 15
is seen to be approximately 2%. This 2% is composed of
(a) an error of less than 0.7% in measuring \( N_i \), \( N_f \) and \( n' \),
and (b) an error of less than 1% in fitting a straight
line to the experimental points. The exceptionally high
stability of 0.1% in both current and position of the
primary beam was mainly responsible for the almost zero
scatter of the experimental points about the linear
CROSS SECTION — CM$^2$/ATOM

DIAG. 16.

PROJECTILE $H_1^+$
TARGET $H_2$
CROSS SECTION $\sigma_{10}$

PRESENT DATA
STIER 1956
RIBE 1951
STEDEFORD 1955
KASNER et al. 1957
KEENE 1949
GOLDMAN 1931
HASTED 1955
GRYZINSKI 1963

CROSS SECTION — CM$^2$/ATOM

PROJECTILE ENERGY — KEV.
The constant, \( c \), in equation (3) above, was determined by the measurement of the conversion of \( \text{H}_1^+ \) to \( \text{H}_1^0 \) by one electron capture in each target gas used and assuming the cross section value for \( \sigma_{10} \) at a given energy. The energy of 10 Kev was selected for this standardization procedure because, as shown in diag. 16, there are a large number of experimental measurements by other workers which appear to have their best agreement at that energy. Further, the majority of the accuracy tests described in Chapter 3 were performed at 10 Kev, which fact gives most confidence to the present experimental results at that energy. The value of \( 8.2 \times 10^{-16} \, \text{cm}^2/\text{gas molecule} \) was selected for \( \sigma_{10} \) at 10 Kev in hydrogen as that figure is a mean value of all those experimental results shown in diag. 16. If this value of \( \sigma_{10} \) should be shown to be in error in the future, all of the present measurements (in hydrogen) can be corrected accordingly by applying the same multiplicative factor to all cross section values in hydrogen. An alternative standardization procedure would be to position the present experimental curve such that the best agreement in shape over as wide an energy range as possible was obtained with the mean shape of the results of diag. 16. It is seen that this procedure leads to the same results as the first procedure.

A similar standardization procedure was followed in
CROSS SECTION — CM/GAS ATOM
PROJECTILE $H^+$
TARGET He
CROSS SECTION $\sigma_{10}$

-- MAPLETON 1961
-- BRANSDEN 1954
-- GRYZINSKI 1963

• • • PRESENT DATA

STIER & BARNETT 1956

PROJECTILE ENERGY — KEV.
CROSS SECTION — $cm^2/gas\ atom$

$10^{-17}$
$10^{-16}$
$10^{-15}$

1 8 10 100
DIAG. 18.

PROJECTILE $H^+$
TARGET $Ne$
CROSS SECTION $\sigma_{10}$

CROSS SECTION — $cm^2$/gas atom

- • • • PRESENT DATA
- STIER & BARNETT 1956
- • • • GRYZINSKI 1963

PROJECTILE ENERGY — keV.

$10^{-17}$ $10^{-16}$ $10^{-15}$
PROJECTILE $H^+$
TARGET $Ar$
CROSS SECTION $\sigma_{10}$

CROSS SECTION — $cm^2/gas$ atom

PROJECTILE ENERGY — keV.

- • • • PRESENT DATA
- STIER & BARNETT 1956
- • • • GRYZINSKI 1963
DIAG. 20. TARGET CROSS SECTION

PROJECTILE ENERGY - KEV

PRESENT DATA
STER & BARNETT 1956
CALCULATED from GRYZINSKI THEORY 1963.

PROJECTILE CROSS SECTION

H\textsuperscript{+} \quad Kr \quad \sigma_{10} \quad 10^6

CROSS SECTION - CM\textsuperscript{-2}/ATOM

10^{-15} \quad 10^{-16}
the five inert gases He, Ne, Ar, Kr and Xe. Diagrams 17 to 21 show the excellent agreement in energy dependence between the present $\sigma_{10}$ experimental curves and those of Stier and Barnett (1956) when 10 Kev has been selected as the standardization energy. Only in Kr is there any variation when the present values become larger than that of Stier and Barnett at 8 Kev until, at 2 Kev, the present values are 20% higher. This generally good agreement has been regarded as the final test of the accuracy and validity of the present apparatus and measurement techniques and of the confidence which may be placed in the present results.

There is currently some interest in the classical binary collision treatment of single electron charge transfer by Gryzinski (1959, 1963), who has calculated $\sigma_{10}$ for H$^+$ in hydrogen and helium. As shown in diagrams 16 and 17 his predicted values are in poor agreement with experimental values. His calculations have been extended to all the inert gases (see appendix 3) except Xenon as his theory fails badly when the ionization potential of the target atom is less than that of the projectile atom. These calculations are compared with present experimental values in diagrams 18, 19 and 20. In all cases the agreement with experiment is poor, particularly above a projectile velocity, $v$, of approximately $2 \times 10^8$ cm/sec. when the predicted values decrease far more rapidly than
the experimental values.

It may be seen from appendix 3 that Gryzinski's formula for the capture cross section consists of two parts - the first is non-velocity dependent (proportional to $\frac{U_i^B}{U_i^A}$) and determines the magnitude of the cross section while the second part, $\left[ G_c \left( \frac{U_i^B}{U_i^A} ; \frac{v_{\text{projectile}}^B}{v_{\text{electron}}^A} \right) \right]$, which is a function of only the ionization potentials of the colliding atoms A and B and the velocities of the struck electron, $v_{\text{electron}}$, and the projectile atom B, $v_{\text{proj}}^B$, determines the dependence of the cross section on the projectile velocity. This function $G_c$ is shown in diag. 99 of appendix 3. Attempts have been made to fit this extremely simple function to the present experimental data, however there is only a vague resemblance between them. This is only to be expected because of the inadequacy of the concept of a binary collision theory to describe the collision of a proton with a many electron atom. The main justification for use of Gryzinski's theory is that for gases other than $\text{H}_2$ and He, there are, at present, no other predictions available. However in $\text{H}_2$ and He, the use of his simple formula is a poor alternative to the better approximate quantal calculations which are available.

There is also current interest in the production of $\text{H}_1^0$ in highly excited states by 10/01 processes in gases. The present experimental measurements are nominally of the total electron capture into all excited states of the
hydrogen atom. All those $H^0_1$ atoms formed by electron capture must pass undeviated through the charged particle magnetic analyzing field to reach their detector. The equivalent electric field of the $\mathbf{V} \times \mathbf{B}$ field will, however, ionize those $H^0_1$ in highly excited states. At the lowest $H^0_1$ velocity of $0.66 \times 10^8$ cm/sec$^{-1}$ the $H^0_1$ atom must pass through a $B$ field of 645 gauss. The equivalent $\mathbf{V} \times \mathbf{B}$ field of $426 \, \text{V cm}^{-1}$ will then ionize those $H^0_1$ excited states of principal quantum number $N \geq 25$ (Hiskes, 1962). At the highest $H^0_1$ velocity of $3.1 \times 10^8$ V cm$^{-1}$, the equivalent $\mathbf{V} \times \mathbf{B}$ field is $3.1 \times 10^8 \times 3.22 \times 10^3 = 1.0 \times 10^4$ V cm$^{-1}$ which is sufficient to ionize those $H^0_1$ excited states with $N \geq 16$. Those atoms which are ionized by the $\mathbf{V} \times \mathbf{B}$ field will then be deflected from the beam path and fail to enter any detector. The experiments of Sweetman (1963) and the theory of Butler (1964) both indicate that the numbers of $H^0_1$ with values of $N$, as shown above, which may be ionized in this manner, is less than $10^{-4}$ of the total $H^0_1$ beam population and thus their effect on the present measurements is negligibly small. It is also noticed that if this effect were not negligible then one would expect different values of $\sigma_{10}$ to result from those experiments which used magnetic analysis of the projectile beam after collision than from those which used electrostatic analysis of the beam because the electric fields used in the latter type experiments would be much
**Diag. 22(a)** Growth of $\frac{H^-}{H^+}$ with $n'$ for 2 keV $H^+$ in helium.

**Diag. 22(b)** Growth of $H^-$ from a 10 keV $H^+$ beam in hydrogen.
PROJECTILE $H_1^+$
TARGET $He$
CROSS SECTION $\sigma_{1T}$

 DIAG. 23.

![Graph showing cross section vs. projectile energy](image-url)

- **PRESENT DATA**
- **FOGEL 1959**
- **ROSENTSVEIG**
- **GERASIMENKO**

CROSS SECTION — cm$^2$/ATOM

PROJECTILE ENERGY — keV.
smaller than the $V \times B$ fields created by the magnetic analysis.

4.2 The Electron Capture Process $\sigma_{\text{ir}}$

As mentioned in Chapter 1.1, special interest is found in the collision $\sigma_{\text{ir}}$ for $H_1^+$ in He and $H_2$ because of the absence of excited states in both the projectile and target atoms, both before and after collision. Diag. 22(a) shows the growth of the ratio $H^-_1/H_1^+$ with the number of target gas atoms/cm² of beam path, $n'$, for 2 Kev $H_1^+$ in He. This example was selected because (a) it gave the smallest cross section measured in this thesis and (b) it had the greatest scatter of any experimental points from which $\sigma_{\text{ir}}$ was determined. Other curves of $H^-_1/H_1^+$ against $n'$ showed less scatter as $\sigma_{\text{ir}}$ increased, which is shown in diag. 22(b) for the growth of $H^-_1$ from a 10 Kev $H_1^+$ beam in hydrogen.

Diag. 23 shows the present experimental results of $\sigma_{\text{ir}}$ for $H_1^+$ in He. Two cross section determinations are made at each energy, one with the collision cell pressure increasing and the other decreasing. At 2 Kev, values of $\sigma_{\text{ir}}$ of 3.5 and $1.5 \times 10^{-20}$ cm²/atom are obtained. This was by far the largest variation ever obtained for any cross section, but it is not unreasonable in view of the very small value of the cross section. The spread of these two measurements is seen to steadily decrease till, at 35 Kev, there is only a 3½ difference between values of about $7.1 \times 10^{-19}$ cm²/atom.
The agreement of the present results with those of Fogel (1959) is excellent above 20 Kev. Both curves show the same maximum cross section value of $7.1 \times 10^{-19} \text{ cm}^2/\text{atom}$ at the same proton energy of 35 Kev. Such good agreement illustrates well the suitability of the use of this collision as a standard cross section measurement. However as the proton energy decreases below 20 Kev the present results become increasingly smaller than Fogel's values. An examination of Fogel's paper reveals a possible explanation of his larger values. He used entry and exit canals of 50 mm. in length and 5.0 mm in diameter relative to a beam diameter determined by two apertures of 3.0 and 2.0 mm. so positioned that the angular collimation of the beam may have permitted the beam to strike the walls of either the entry or exit canals. The primary $H^+$ may then be neutralized and these neutrals may then capture electrons to form $H^-$. This error may be quite large.

The neutralization coefficient for $H^+$ as a function of $H^+$ energy is not known, however $\sigma_{\text{e}}$ is known as a function of energy. At 3 Kev the ratio of $\sigma_{\text{e}}$ to $\sigma_{\text{i}}$ equals $1.1 \times 10^{-18}/0.025 \times 10^{-18} = 44$ while at 30 Kev the ratio $\sigma_{\text{e}} / \sigma_{\text{i}} = 6.3 \times 10^{-18}/0.65 \times 10^{-18} = 9.7$. Therefore at 3 Kev if 2% of the total number of particles entering the collision cell from the entry canal were neutral
hydrogen atoms then equal numbers of $H^-_1$ ions would be produced from the primary $H^+_1$ and the $H^0_1$ atoms by electron capture with the target gas. This $H^-_1$ production from the $H^0_1$ atoms becomes less important as the beam energy increases.

Mention must also be made of the fact that Fogel has used such large values of $(nl)$ (see equation (2)), that his "growth of collision product with $(nl)$" graph is mainly a parabolic relationship from which there is considerable uncertainty in determining the linear component. However for both of the above suppositions, there is insufficient data in Fogel's paper to make more precise conclusions as to the reasons for his larger cross section values.

The Born approximation predictions of Gerasimenko (1962) for proton energies above 100 Kev are clearly orders of magnitude too large and of quite a different energy dependence to that shown by the experimental results. Possible reasons for the disagreement of his predictions from experimental results may lie in the facts that Gerasimenko has (a) used non-orthogonal initial and final state wave functions, (b) neglected the change in the translational motion of the two captured electrons and (c) used a hydrogen-like wave function for the Helium atom. It has been shown by many workers (see Chapter 1.2) that each of these factors exerts an appreciable effect on the
PROJECTILE $H^+_1$
TARGET $H_2$
CROSS SECTION $\sigma_{IT}$

- PRESENT DATA
- FOGEL 1959
- McClure 1963
- FOGEL 1957
- FOGEL 1956
- FOGEL 1955
one electron charge transfer cross section calculations and so it is not unreasonable to expect a similar effect of these factors upon the double electron transfer cross section.

In the energy region from 35 to 50 Kev where the cross section begins to decrease as the proton energy increases the present results agree well with Fogel's values which extend to 60 Kev to indicate that $\sigma \propto E^{-10}$, while Gerasimenko's $\sigma \propto E^{-3}$. The following argument indicates that Fogel's result is the more reasonable one. For single electron capture by protons in helium (see chapter 1.2) the Born approximation predicts that $\sigma \propto E^{-5.5}$ at high energies. If the double electron capture by protons in helium were to be considered as two successive single electron captures from a helium atom in which there was no correlation between its two electrons, then $\sigma_{ij}$ may be proportional to $E^{-11}$, which is close to the experimental dependence of $E^{-10}$.

The perturbed stationary state (P.S.S.) calculations of Rosentsveig (1955) over the energy region from 2 to 10 Kev show closer agreement with experimental values than the high energy predictions of Gerasimenko, however they are still so very different in both magnitude and energy dependence that no success can be claimed in the application of the P.S.S. method to the problem.

Diag 24 shows $\sigma_{ij}$ for $H_1^+$ in hydrogen and compares
the present results with the four attempts of accurate measurement by Fogel. Fogel's 1959 data, his latest attempt, shows good agreement with the present data only in obtaining a maximum cross section value of $1.02 \times 10^{-17}$ cm$^2$/mol. at an energy of 20 Kev. At other energies the agreement is not so good and shows variations of up to 100% at 3 Kev. The use of a logarithmic scale makes the agreement between the various results appear to be much better than it actually is. The earlier Fogel data of 1955, 1956 and 1957 would appear to be in serious disagreement with the latest (1959) data of Fogel who gives no explanation of the errors of his earlier measurements.

The recent data of McClure (1964) gives excellent agreement with the present results above 10 Kev, but below 10 Kev his values become increasingly smaller than the present results. Whilst criticism may be levelled at Fogel's work on the basis of the experimental considerations and accuracy such as discussed in Chapter 3 of this thesis, no such criticism may be made of McClure's work. No explanation is obvious for his lower $\sigma_{17}$ values at low energies.

The wide range of the discrepancies between the many results demonstrates the difficulty of accurate measurement of such small cross section values. Possible errors in these cross section values may be attributed only to
PROJECTILE \( H^+ \)
TARGET \( Ne \)
CROSS SECTION \( \sigma_{11} \)

**PRESENT DATA**

**FOGEL 1959**

**GRAPH:**
- X-axis: Projectile Energy - keV
- Y-axis: Cross Section - \( \text{cm}^2 / \text{gas atom} \)

- Present Data
- Fogel 1959
PROJECTILE $H_1^+$
TARGET $K_T$
CROSS SECTION $\sigma_{1T}$

**DIAG. 27.**

![Graph showing cross section versus projectile energy.](image)

- Present Data
- Fogel 1959

CROSS SECTION $\text{cm}^2 / \text{gas atom}$

PROJECTILE ENERGY — KEV.
Diag. 28.

Projectile: $H_1^+$
Target: $Xe$
Cross section: $\sigma_{II}$

![Graph showing cross sections with present data and Fogel 1959 data. The x-axis represents projectile energy in keV, and the y-axis represents cross section in $cm^2/gas$ atom on a logarithmic scale.]
instrumental origins as there are no excited states possible in any of the collision products to complicate the collision process.

Diagrams 25, 26, 27 and 28 show $\sigma_{11}$ for $H_1^+$ in Ne, Ar, Kr and Xe respectively and compare those values with the only other data available - that of Fogel (1959). In Ar, Kr and Xe the two sets of data agree within 20% both in energy dependence and magnitude, but in Ne the present data is from 1.5 to 2 times higher.

4.21. Discussion of the Cross Section $\sigma_{11}$

The measured cross section $\sigma_{11}$ in hydrogen and helium is a single-peaked function of the projectile velocity and this peak is unambiguously associated with the collision, for example in helium,

$$H_1^+ + He \rightarrow H_1^- + H_{e^+}^+ + \Delta E^1$$

where all particles, both before and after collision, are in their ground state. The energy defect, $\Delta E^1$, for the collision is calculated (after Hasted, 1962) from the relation

$$|\Delta E^1| = |\Delta E_\infty| + E_{Coul}$$

$$= (V_H^i + S_H) - (V_{He}^i + V_{He^+}^i) + E_{Coul}$$

where $V^i$ is the ionization energy of a given atom

$S$ is the electron affinity

$E_{Coul}$, the coulomb attraction energy between the collision products,

$$= \frac{3}{2} \times e^2 \times \frac{r_1^2 - r_2^2}{r_1^3 - r_2^3} \times (n - m)m \times 27.2 \text{ ev for a}$$
collision of the type \( \frac{n_0}{(n - m)m} \)

\[
r_{1,2} = k_1,2\left\{(Z_A - n)^{2/3} + Z_B^{2/3}\right\}^{1/2}
\]

\[k_1 = 1.44\]

\[k_2 = 0.48\]

\(Z_{A,B}\) = atomic number of atom A or B.

Table 4 lists all those values required for the above quantities together with the results of present calculations which are referred to in the following discussion. Using the adiabatic criterion of Chapter 1.3, equation 2, it is found that, for \( \sigma_{17} \), \( \ma_{\text{corr}} \) is 9.5 \(^{0}\)A for He and is 7.3 \(^{0}\)A in \( \text{H}_2 \). For the other many electron targets of He to Xe, each target atom must lose a minimum of two electrons. Therefore the smallest \( \Delta E \) for a given collision must correspond to the target atom being left in the ground state of its doubly charged ion. The adiabatic criterion predicts that as \( |\Delta E| \) decreases then \( E_{\text{max}} \), the projectile energy at which the maximum value of the cross section occurs, also decreases. As the atomic number of the target atom increases from He to Xe, \( \Delta E \) decreases. Thus the lower velocity peak in the cross sections in He, Ar, Kr and Xe is associated with the process 10/12 with the doubly charged ion in its ground state. This is supported by the calculations of \( \ma_{\text{corr}} \) for each of the gases. Column 8 of Table 4 shows that the values of \( \ma_{\text{corr}} \) obtained are approximately 7\(^{0}\)A. This result supports the finding of Hasted (1962) that \( \ma_{\text{corr}} \approx 7^{0}\)A.
### TABLE 4

**Description of Collision 10/7 n**

<table>
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<tr>
<th>Target Atom</th>
<th>Atomic Number</th>
<th>Ionization Potentials</th>
<th>$\Delta E_{\infty}$</th>
<th>$E_{\text{Coul}}^2$</th>
<th>$E_{\text{max}}$</th>
<th>$m_{\text{corr}}$</th>
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<td>Z</td>
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<td>$1V_i$ - ev, $2V_i$ - ev</td>
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<td>-64.7</td>
<td>41.8</td>
<td>36</td>
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<td>21.56</td>
<td>41.07</td>
<td>-48.34</td>
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<td>13.5</td>
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<td>15.76</td>
<td>27.6</td>
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<td>5</td>
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<td>-24.27</td>
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<td>Kr</td>
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<td>2.5</td>
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<tr>
<td>Xe</td>
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<td>-</td>
<td>-35.75</td>
<td>41.8</td>
<td>18</td>
</tr>
<tr>
<td>H$_2$</td>
<td>2</td>
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<td>12.2</td>
<td>14.9</td>
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<td>24.2</td>
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<td>7.3</td>
<td>52.9</td>
<td>42.9</td>
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<td>18.0</td>
<td>27.2</td>
<td>28</td>
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<tr>
<td>(decreed)</td>
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<td>111.84</td>
<td>63.5</td>
<td>36.6</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Va</td>
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</tbody>
</table>

TABLE 4 (cont.)
for electron capture processes 00/11, 10/01 and 10/12, irrespective of the atomic number of the colliding particles.

The higher velocity peak in the cross section must occur from a collision of the type

\[ \text{He}^+ + \text{Xe} \rightarrow \text{He}^{-} + (\text{Xe}^{++})_{\text{excited}} + \text{El} \]

Table 4 shows that if this peak, and not the low velocity peak, were due to the Xe\(^{++}\) ion being formed in the ground state, then \( m_{\text{corr}} \sim 30 ^0\text{A} \), which is contrary to all other experimental evidence. The energy defect of the above collision is

\[ \Delta E^{1\text{ll}} = \Delta E^{1} + E_{\text{ex}} \]

where \( E_{\text{ex}} \) is the excitation energy of the doubly charged target ion. To determine the value of \( E_{\text{ex}} \), and hence the particular process, from which the high velocity peak originates, \( m_{\text{corr}} \) may be assumed equal to \( 7^0\text{A} \) and then that value of \( E_{\text{ex}} \) found which predicts a maximum cross section at that velocity which is observed experimentally. Column 12 of Table 4 shows the values of \( E_{\text{ex}} \) so obtained. These values do not correspond to any known excited levels of Ar\(^{++}\), Kr\(^{++}\) or Xe\(^{++}\). However if it is then assumed that the target ion may be triply charged and due allowance is made for the increased coulomb interaction (\( 3E_{\text{Coul}} \), which is now the interaction between a single negative charge and three positive charges), values of \( E_{\text{ex}} \) are obtained which agree well (see columns 13 and 15 of Table 4) with
the ground state of a triply charged target ion. Therefore the higher velocity peak in the cross section function in Ar, Kr and Xe results predominantly from a collision of the type 10/13, that is
\[ \text{H}_1^+ + \text{Xe} \rightarrow \text{H}_1^- + \text{Xe}^{+++} + e. \]
If we were to assume this result and work backwards then we find (column 16 of Table 4) that values of \( \text{ma}_{\text{corr}} \) of 5.7, 6.8 and 7.2 \( \text{Å} \) are obtained, in good agreement with that found for 10/12 collisions.

Fogel (1959) has arrived at the same conclusions as above by the use of an adiabatic hypothesis argument in which he used a value of "a" of 1.5 \( \text{Å} \) and neglected the coulomb interaction between the charged collision products. His values of \( E_{\text{ex}} \) however are up to 20% below the third ionization potential of the target atom, which does leave some doubt about his deduction. The present calculation makes proper allowance for \( E_{\text{Coul}} \) and uses \( \text{ma} = 7 \text{ Å} \) and, as shown above, then obtains values of \( E_{\text{ex}} \) within 4% of the third ionization potential of the target atom.

Even though the above analysis indicates a plausible explanation of the experimental results by involving only the ground states of the doubly and triply charged target ion, the experimental cross section is in fact the superposition of all the cross sections for double electron capture to leave the target ion in the ground and all of its excited states. It is probable then that the cross sections in which the target ion is left in an excited
state is not large compared with that in which the target ion is left in the ground state.

Two facts indicate why a second peak is not observed in the cross section $\sigma_{17}$ in Ne. First the adiabatic hypothesis predicts that the maximum cross section for $\text{IO}/13$ should occur at a proton energy of 95 Kev which is above the present range. Secondly diagrams 26, 27 and 28 show that, as the atomic number of the target decreases, the height of the higher velocity peak relative to that of the lower velocity peak decreases whilst the height of the lower velocity peak itself decreases, i.e. the maximum values of the cross sections for $\text{IO}/12$ and $\text{IO}/13$ have a different dependence on atomic number. The above relative decrease is such that when the atomic number is decreased to ten, i.e. that of neon, the high velocity peak may well be too small to be seen above that of the $\text{IO}/12$ process.

The dependence of the cross section on projectile energy has been examined by Hasted (1961) who has fitted a large number of $\sigma_{10}$ values to the exponential form of equation 1.36, i.e. $\sigma = \sigma_0 \exp\left(-\frac{ka}{\hbar \nu}\right)$, and subsequently obtained from that relationship a value of the interaction distance "a" which is in good agreement with the value of "a" determined from the adiabatic maximum criterion. Fogel (1959) has applied Hasted's approach to $\text{IO}/12$ collisions for $\text{Na}^+$ and $\text{Li}^+$ ions in the inert gases but has found that "ka" of equation 1.36 is not a constant and is
CROSS SECTION $- \text{cm}^2 / \text{Atom}$

$10^{-17}$

$10^{-18}$

$10^{-19}$

$10^{-20}$

$\frac{1}{\sqrt{10^8 \text{cm/sec.}}}$

SLOPE $= -0.63$

SLOPE $= -0.56$

SLOPE $= -0.55$

△ △ △ HYDROGEN
○ ○ ○ HELIUM
□ □ □ NEON

DIAG.29. GRAPH OF $\log \sigma_{ii}$ Versus (PROTON VELOCITY)$^{-1}$
 DIAG. 30  

PLOT OF \( \frac{\sigma_{IT}}{\sigma_{MAX}} \) \( \rightarrow \) \( \frac{V}{V_{MAX}} \)
dependent upon the type of target atom. The following paragraph tries the same approach for $H^+$ ions in He, Ne and hydrogen. Diag. 29 shows the relationships of $\ln \sigma_{ij}$ against $\lambda$. Away from the maximum cross section all curves show approximately the same slope and hence, from equation 1.36, "$ka$" cannot be constant since the $\Delta E$'s for all target gases (see Table 4) are quite different. It is well established (Hasted, 1960) that "$a$" should be constant for a given collision process and so "$k$" must depend upon the nature of the target gas atom. This conclusion agrees with that found by Fogel for $10/12$ collision of $Na^+$ and $Li^+$ in the inert gases.

This apparent independence of slope of the graphs of diag. 29 on $\Delta E$ suggests a modification of equation 1.36 to exclude $\Delta E$, which may be achieved from the adiabatic criterion that $\sigma_m$ occurs when $a \cdot \frac{\Delta E}{h \cdot v_{max}} \approx 1$. Then,

\[ \frac{\sigma}{\sigma_{max}} = \frac{\sigma_0}{\sigma_{max}} \cdot \exp \left[ - k \cdot \frac{v_{max}}{v} \right] \]...

Diag. 30 shows a plot of $\frac{\sigma}{\sigma_{max}}$ against $\frac{v}{v_{max}}$ for $\sigma_{ij}$ for hydrogen and the inert gases. In Ar, Kr and Xe the low velocity peak only has been plotted. The form of this cross section function, both above and below the cross section maximum, is seen to be independent of the type of target atom and energy defect, $\Delta E$. Since the form of equation 4.22(i) has no sound theoretical basis, it may be used as a prototype for further empirical development to
DIAG 31. GROWTH OF $H_1^-$ FROM 4 KeV $H_1^0$ IN ARGON.
SECTION - CM/GAS ATOM

PROJECTILE $H_1^0$
TARGET $H_2$
CROSS SECTION $\sigma_{0T}$

PRESENT DATA
FOGEL 1958
STIER & BARNETT 1956
CURRAN & DONAHUE 1959

CROSS SECTION $\text{cm}^2$/GAS ATOM

PROJECTILE ENERGY - KEV

$(10^{-18}, 10^{-17}, 10^{-16})$
describe the dependence of the cross section $\sigma_{17}$ on energy for a wider variety of projectile and target atoms.

4.3 The Electron Capture Cross Section $\sigma_{17}$

Diag. 31 shows the growth of $H_1^-$ from a 4 Kev $H_1^0$ beam traversing Argon at values of $n'$ (see equation 4.2) sufficiently small that less than 0.5% of the primary beam captures an electron to form $H_1^-$. The growth of $H_1^-$ with $n'$ is linear over the entire range of $n'$. The error in fitting a straight line to the experimental points is less than 2% and the maximum error of the cross section $\sigma_{17}$ relative to the 10 Kev standardization cross section $\sigma_{10}$ for all target gases is not greater than 4%. Diag. 31 is typical of the growth of collision product ions with $n'$ graphs from which $\sigma_{17}$ has been determined. In no case did the ratio $H_1^-/H_1^0$ exceed 1%.

The experimental measurements of $\sigma_{17}$ for $H_1^0$ incident upon hydrogen and the inert gases are shown, and compared with all published values, in diagrams 32 to 37. There are no theoretical predictions for $\sigma_{17}$.

Hydrogen. Diag. 32 shows that there is no support in the present values for the enhanced cross section in the region of the maximum value as obtained by Curran (1960) nor is there any evidence of the multiple peaks in the cross section as obtained by Curran. Curran has suggested that possibly other workers have missed this fine structure because of too large an energy increment between their
CROSS SECTION — CM/GAS ATOM

PROJECTILE $H_1^0$

TARGET He

CROSS SECTION $\sigma_{GT}$

---

**Diagram 33.**

- Present Data
- Fogel 1958
- Stier & Barnett 1956

**Axes:**
- **CROSS SECTION — CM²/GAS ATOM**
- **PROJECTILE ENERGY — KEV.**
PROJECTILE $H_1^0$
TARGET $Ne$
CROSS SECTION $\sigma_{\text{OT}}$

**DIAG. 34.**

- **PRESENT DATA**
- STIER & BARNETT 1956

![Graph showing cross section vs. projectile energy](image-url)

**CROSS SECTION — cm$^2$/gas atom**

**PROJECTILE ENERGY — KEV.**
measurements. This reason has been discounted in the present investigation where (a) an energy increment of 1.0 Kev has been used in the range from 2 to 10 Kev and (b) the energy resolution of the beam by the 30 cm. analysing magnet was better than 1%.

The velocity at which the maximum cross section, $\sigma_{\text{max}}$, occurs cannot be explained in terms of the adiabatic criterion. Such an anomalous and unexplained position of $\sigma_{\text{max}}$ is also well known for the single electron capture cross section $\sigma_{10}$ for $\text{H}_1^+$ in hydrogen (Hasted, 1955).

Helium. The energy range has been extended from 10 Kev down to 2 Kev as shown in diag. 33. The cross section values in this range support an extrapolation of Stier and Barnett's (1956) data in contrast to the very sharp decrease suggested by Fogel's data (1958).

Of the inert gases only He and Ne (see diagrams 33 and 34) show a maximum cross section in the present energy range. However Hasted (1962) has shown that for all the inert gases the velocity at which the maximum cross section occurs agrees well with that predicted from the adiabatic criterion when an interaction distance \( "a" = 7^{0}\text{A} \) is used. This success of the adiabatic criterion gives confidence to the use of equation 1.36 to determine independently the constant \( "a" \) from the rate of rise of the cross section in the low energy region for both the present values and Stier and Barnett's values. These values of \( "a" \) may then
DIAG. 35

PROJECTILE $H_1^0$

TARGET Ar

CROSS SECTION $\sigma_{o-T}$

CROSS SECTION $\text{cm}^2$/gas atom

PROJECTILE ENERGY — KEV.
CROSS SECTION - CM^2/GAS ATOM

PROJECTILE \( H_1^0 \)
TARGET \( Kr \)
CROSS SECTION \( \sigma_{0T} \)

 DIAG. 36.

\[
\begin{align*}
\text{CROSS SECTION} & \quad \text{CM}^2/\text{GAS ATOM} \\
\text{PROJECTILE ENERGY} & \quad \text{KEV} \\
10^{-16} & \quad 10^{-17} \\
10^{-18} & \quad 10^{-19} \\
1 & \quad 10 \\
100 & \quad 100
\end{align*}
\]

- - - PRESENT DATA
DIAG. 37.

PROJECTILE $H_1^0$
TARGET $Xe$
CROSS SECTION $\sigma_{OT}$
the collision
\[ H_1^0 + Ar \rightarrow H_1^- + Ar^+ \]  \( \ldots \ldots \) 4.4(i)
where all collision participants are in their ground states. The adiabatic criterion indicates that if \( H_1^0 \) is in an excited state the energy defect \( |\Delta E| \) is lowered and the maximum cross section occurs at a velocity lower than that of the ground state \( H_1^0 \) collision. Similarly if the \( Ar^+ \) ion is formed in an excited state, \( |\Delta E| \) is raised, and hence the maximum cross section occurs at a velocity higher than that of the ground state collision. With the assumption that the projectile \( H_1^0 \) is in the ground state, the subsidiary maximum at 25 Kev could arise from the \( Ar^+ \) ion being formed in an excited state. By the use of an argument, based on the adiabatic criterion, identical with that used in Chapter 4.3 for the analysis of the \( \sigma_{11} \) data, the value of 29.9 ev is obtained for the excitation energy of the \( Ar^+ \) ion. This value is within 10\% of the second ionization potential of Argon. Hence the subsidiary maximum of the measured \( \sigma_{01} \) curve is predominantly due to the process
\[ H_1^0 + Ar \rightarrow H_1^- + Ar^{++} + e \]
Krypton and Xenon. The experimental results are shown in diagrams 36 and 37. The decrease of cross section with increasing \( H_1^0 \) velocity is slow and \( \sigma_{01} \) may well be a compound cross section as discussed above for Argon, however there is no definite evidence of any subsidiary
Dependence of $\sigma_{\text{max}}$ on $\Delta E_\infty$ for electron capture cross sections.

$\Delta E_\infty$ for $\sigma_{\text{max}}$ and $\sigma_0$.
4.4 General Discussion of the Electron Capture Cross Sections \( \sigma_{10}, \sigma_{11}, \) and \( \sigma_{01} \).

Several interesting comparisons and similarities may be made and observed in the electron capture charge changing collisions \( \sigma_{10}, \sigma_{11}, \) and \( \sigma_{01} \).

(a) The simple observation is made that \( \sigma_{10} > \sigma_{01} > \sigma_{11} \) for a given target gas. The inequality that \( \sigma_{10} > \sigma_{11} \) appears to be simply related to the difficulty of capturing two electrons compared with one electron. The inequality that \( \sigma_{10} > \sigma_{01} \) may be related to the different binding energies of the atom and the ion but perhaps, more likely, to the different perturbing electric fields which exist between the target atom and the incident atom or ion.

(b) The adiabatic criterion gives no indication of the absolute or relative values of the peaks in the cross section versus velocity functions. Nor does there appear to be any published information thereon. However diag. 39 reveals that the same functional form relates each of the maximum values of the capture cross sections \( \sigma_{10}, \sigma_{01}, \) and \( \sigma_{11} \) to the energy defect \( \Delta E \) and to the target atomic number \( Z \) for each of the targets of hydrogen and the inert gases. There is only one exception to the above empirical relationship, that of \( \sigma_{10} \) for \( H^+ \) in Xe and this collision is the only collision which is exothermic. It is readily
\[ \frac{\sigma}{\sigma_{\text{MAX}}} \text{ IN BLACK} \]
\[ \frac{\sigma_{17}}{\sigma_{\text{MAX}}} \text{ IN RED} \]
\[ \frac{\sigma_{19}}{\sigma_{\text{MAX}}} \text{ IN GREEN} \]

- • • • HYDROGEN
- × × × HELIUM
- ○ ○ ○ NEON
- △ △ ARGON
- □ □ KRYPTON

**Diag. 39(a).** Plot of \( \frac{\sigma}{\sigma_{\text{MAX}}} \rightarrow \frac{v}{v_{\text{MAX}}} \)
seen that $\sigma_{\text{max}}$ increases as $\Delta E$ decreases and/or $Z$ increases.

(c) There does not appear to be any direct relationship of the empirical results mentioned in the previous paragraph (b) with any theoretical predictions. However it is noted that each of the cross sections $\sigma_{10}$, $\sigma_{01}$, and $\sigma_{11}$ (neglecting the contribution from the process $10/13e$) has a similar dependence upon the projectile velocity. The form of this cross section versus velocity function is vaguely similar to that predicted for $10/01$ collisions by Gryzinski (diag. 99 in appendix 3) and by Rapp and Francis in Diagram 9; however the simplest empirical function which gives the best fit for all these electron capture processes is that used by Hasted (1960) for $10/01$ collisions, and used in diag. 30 for $10/12$ processes, namely $\sigma/\sigma_{\text{max}} = \sigma_{\text{01}}/\sigma_{\text{max}} \times \exp(-k.v_{\text{max}}/v)$. In diag. 39(a) the graph of $\sigma/\sigma_{\text{max}} \rightarrow v/v_{\text{max}}$ reveals an approximately similar distribution of the single electron capture $\sigma_{01}$ and $\sigma_{10}$ values, while the double capture $\sigma_{11}$ values have a narrower distribution. However the scatter of the experimental data is large.

(d) The cross section $\sigma_{11}$ clearly demonstrates the dominating influence at high velocities of the process $10/13e$ over the process $10/12$. This was not surprising since it is well established (Hasted, 1960) that the cross section for ionization (without capture) of a target atom by a proton attains a maximum value at a higher velocity than a simple capture cross section. An appreciable contribution to the $\sigma_{01}$ in argon has been attributed to an $00/12e$ collision. It can be anticipated then that
DIAG. 40. GROWTH OF $H_1^+$ FROM 3 Kev $H_1^0$ IN Xe.
capture with additional ionization of the target atom occurs simultaneously with simple electron capture for \( \sigma_{01} \) in other target gases and may also occur with the cross section \( \sigma_{10} \). However in paragraph (a) above it was shown that \( \sigma_{10} > \sigma_{11} > \sigma_{01} \) which presumably explains the masking of the process type 10/02e in \( \sigma_{10} \) and 00/12e in \( \sigma_{01} \).

(c) One further observation for which no explanation can be offered is (i) that \( \sigma_{10} \) in He is greater than \( \sigma_{10} \) in Ne at proton energies greater than 30 keV and (ii) that \( \sigma_{01} \) in He is greater than \( \sigma_{01} \) in Ne at \( H^0 \) energies greater than 35 keV. For all target gases and for both \( \sigma_{10} \) and \( \sigma_{01} \) at all energies none of the electron capture cross section versus projectile energy curves cross except for the above anomalous cases. This exception is also well supported by the data of Stier and Barnett (1956).

4.5 The Electron Loss Cross Section \( \sigma_{01} \).

Diag. 40 shows the growth of the collision product \( H^+ \) with increasing target gas number density for 3 keV \( H^0 \) incident upon Xe. This example was selected because it shows (a) the shortest linear region from which any \( \sigma_{01} \) was determined and (b) that, while the primary \( H^0 \) beam currents were only of the order of \( 10^{-10} \) amp, there was no loss of experimental accuracy or no greater scatter of experimental points than was obtained for the much larger proton currents (\( \sim 10^{-6} \) amp) used in Chapter 4.1.

Much interest has been aroused in the cross section
CROSS SECTION — CM$^2$/GAS ATOM

PROJECTILE $H^0$
TARGET $H^2$
CROSS SECTION $\sigma_{01}$

Diagram 41.

PROJECTILE ENERGY — KEV.

CROSS SECTION — CM$^2$/GAS ATOM

10$^{-16}$

10$^{-17}$

10$^{-18}$

+ + + PRESENT DATA — 2S STATE NOT QUENCHED

• • • PRESENT DATA — 2S STATE QUENCHED

FOGEL 1958
STIER & BARNETT 1956
CURRAN & DONAHUE 1959
RIBE 1951
\( \sigma_{01} \) in hydrogen as there are two distinct groups of results, particularly at energies less than 10 Kev, as shown in diag. 41. The present measurement indicate values of \( \sigma_{01} \) 10\% lower than those of Stier and Barnett above 10 Kev. Below 10 Kev the present values are 25\% lower than, but of a similar energy dependence to, those of Stier and Barnett which is in marked contrast to those of Curran (1960) which are 300\% lower at 4 Kev. The subsidiary maximum at about 5.5 Kev is supported by the present measurements.

No evidence in favour of any fine structure in \( \sigma_{01} \) has been obtained. The comments on the small energy increments and high energy resolution for \( \sigma_{01} \) measurements in hydrogen in Chapter 4.4 apply equally well here. Curran has also associated, in part, the amplitude of his fine structure with the presence of water vapour in the target gas. Present measurements have shown that condensable vapours (predominantly water and diffusion pump oil vapours) have no detectable influence on \( \sigma_{01} \). The present apparatus was equipped with liquid air cooled baffles (-150°C) both inside and immediately below (outside) the collision cell. The use of these baffles necessitated recalculation of the relative pressure measurements by remeasurement of \( \sigma_{10} \) at the gas temperature as determined by the liquid air baffle because the method of determination of the absolute value of the cross sections.
(Chapter 4.1) requires that both "n" and "l", the target gas number density and the beam path length in the target gas respectively, be independent of the gas temperature during relative cross section measurements.

It is possible that the incident \( H_1^0 \) beam contains a large population of excited state atoms. The possible effect of such atoms upon the measured cross sections involves the following considerations:

(a) A knowledge of the cross sections for formation of \( H_1^0 \) with given quantum numbers \( n \) and \( l \) relative to that of the ground state. This information is not available. The mode of formation of \( H_1^0 \) varies from the present method of electron capture by protons in the residual gas of the vacuum system to Fogel's (1958) method at low energies (\( \sim 5 \text{ KeV} \)) of the charge transfer and dissociation of \( H_3^+ \) ions in collision with mercury vapour. Comparison of the \( H_1^0 \) beam excited state populations at their time of formation is therefore most difficult.

(b) The relative excited state populations of the \( H_1^0 \) beam at the time of collision with the target gas will further depend upon (i) the time of flight, \( t \), of a given atom from the place of formation to the place of collision and (ii) the life time, \( T \), of the given excited state. In the present experiment \( t \) lies in the range 8.3 to 83.10\(^{-8}\) sec at 2 KeV \( H_1^0 \) energy and in the range 1.6 to 16.10\(^{-8}\) sec at 50 KeV. From the life times of hydrogen given by Bethe
and Salpeter (1957) it is seen that a 2 Kev $H_1^0$ beam would now contain approximately all the ns states, no 2\ell, 3\ell or 4\ell ($\ell \neq s$) states (as they would have decayed to the ground state) and all other higher nl states.

(c) However the $H_1^0$ beam necessarily passes through a uniform electric field of strength $F$ (V cm $^{-1}$) prior to entry into the collision cell. What is the effect of this field upon the excited state populations? Consider a specific state, the $2S_{\frac{1}{2}}$. In the absence of an electric field transitions from the $2S_{\frac{1}{2}}$ to the lower energy states $1S$ and $2P_{\frac{3}{2}}$ are forbidden. The $2S$ state lifetime $\sim 0.15$ sec while the $2P$ lifetime is $1.6.10^{-9}$ sec. In an external static electric field the atom will experience an asymmetric field which will mix some of the $2P$ state wave functions into the wave function describing the atom, and this part of the wave function can "connect" with the ground state in dipole radiation. The lifetime, $T_{2S}$, of the $2S$ state is given by $T_{2S} \approx \left(\frac{475}{F}\right)^2 T_{2P}$ (Bethe and Salpeter, 1957). When $F = 475$ Volt cm $^{-1}$, $T_{2S}$ is equal to $T_{2P} \sim 1.6.10^{-9}$ sec. For higher ns states, the critical field strength ($\frac{475}{F}$) decreases very rapidly as $n^{-4.5}$ and is approximately $\frac{58}{F}$, $\frac{12}{F}$ and $\frac{1.7}{F}$ for $n = 3, 4$ and 6. These np state atoms then have the normal np state lifetimes.

The following simple experiment was performed to investigate the possible influence of such excited states upon the measured cross section $\sigma_{01}$. The results shown
Table 5

$F_{2S}$, the fraction of $H_1^0$ metastable 2S states not quenched by the electrostatic field.

<table>
<thead>
<tr>
<th>$H_1^0$ Energy</th>
<th>$F_{2S}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kev</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0.86</td>
</tr>
<tr>
<td>3</td>
<td>0.72</td>
</tr>
<tr>
<td>4</td>
<td>0.57</td>
</tr>
<tr>
<td>5</td>
<td>0.48</td>
</tr>
<tr>
<td>6</td>
<td>0.39</td>
</tr>
<tr>
<td>8</td>
<td>0.31</td>
</tr>
<tr>
<td>10</td>
<td>0.20</td>
</tr>
</tbody>
</table>
in diag. 41 were obtained with an electric field in excess of 475 V cm\(^{-1}\) between the deflector plates. If one assumes that the excited state population of the incident \(H_1^0\) beam is predominantly the 2S state before the deflector plates, then this field is sufficient to entirely quench the 2S state and the beam entering the collision cell is a ground state \(H_1^0\) beam.

It was necessary to move the deflector plates from their previous position such that the smallest possible field strength could be used between the deflector plates to give minimum quenching but yet be sufficient to cause that deflection which would prevent any positively charged particles within the \(H_1^0\) beam from just entering the entry canal of the collision cell. It is noted that even if the unlikely event occurred that a small fraction of the protons did pass into the collision cell, their numbers would be pressure invariant and hence would not contribute any error to the cross section \(\sigma_{01}\).

Knowing that a given state population decays exponentially and the decay rate, \(D\), is the inverse of \(T\), the expression for the fraction, \(F_{2S}\), of 2S states not quenched by the field is obtained, that \(F_{2S} = \exp\left[-\left(\frac{F}{475}\right)^2 \cdot \frac{1}{T_{2F}} \cdot t\right]\) where \(t\) is the time of flight through the deflector plates. Table 5 shows the fraction \(F_{2S}\) predicted by this relation at energies from 2 to 10 Kev. Diag. 41 shows the values of \(\sigma_{01}\) measured under the above experimental
Diag. 42.

PROJECTILE \( H^0 \)
TARGET \( He \)
CROSS SECTION \( \sigma_{01} \)

CROSS SECTION — \( \text{cm}^2/\text{gas atom} \)

- Present data
- Fogel 1958
- Stier & Barnett 1956
- Bates & Williams 1957

PROJECTILE ENERGY — KEV.
PROJECTILE $H_1^0$
TARGET $Ne$
CROSS SECTION $\sigma_{01}$

**Diagram 43**

Cross section in $cm^2/gas atom$ vs. projectile energy in keV for $H_1^0$ on Ne target. Three data sets are shown:
- Present data
- Fogel 1958
- Stier & Barnett 1956
CROSS SECTION - CM/GAS ATOM

PROJECTILE $H_1^0$
TARGET $Ar$
CROSS SECTION $\sigma_{01}$

DIAG. 44:

\begin{axis}[
    xlabel={PROJECTILE ENERGY — KEV.},
    ylabel={CROSS SECTION — $\text{cm}^2/\text{gas atom}$},
    xmode=log,
    ymode=log,
    xmin=1,
    xmax=100,
    ymin=1e-17,
    ymax=1e-15,
    legend entries={PRESENT DATA, FOGEL 1958, STIER & BARNETT 1956, SOLOVIEV 1962},
    legend style={at={(0.5,0.12)},anchor=north},
]

% Data points

% Legend

\end{axis}
PROJECTILE $H^0$
TARGET $KT$
CROSS SECTION $\sigma_{01}$

**DIAG. 45.**

- **PRESENT DATA**
- **FOGEL 1958**
- **SOLOVIEV 1962**

CROSS SECTION — $cm^2$/gas atom

PROJECTILE ENERGY — kev.
DIAG 46

PROJECTILE $H^0$
TARGET $Xe$
CROSS SECTION $\sigma_{01}$

Graph showing the relationship between projectile energy (in keV) and cross section (in $cm^2/gas\ atom$). The graph includes data points labeled "PRESENT DATA" and a line labeled "FOGEL 1958."
conditions. (Each point is the average of two determinations. In no case is the increase in $\sigma_{01}$ greater than 8\%. However all measurements do show an increase in $\sigma_{01}$ with surprising consistency.

It is concluded that the contribution of any 2S metastable states of $H_1^0$ present in the primary beam to the measured cross section is small. Such an explanation cannot account for the large difference between the present values and those of Curran and Donahue.

**Helium.** Diag. 42 shows the two separate determinations of $\sigma_{01}$ in He both of which show two maxima, one at 9.5 Kev and the other at about 35 Kev. The basic collision thought to be studied here is

$$H_1^0 + He \rightarrow H_1^+ + e + He + \Delta E$$

where $\Delta E = 13.57$ ev. $\Delta E$ is then independent of the nature of the target gas and the adiabatic theory predicts the maximum cross section to occur at the same velocity for all gases. Diagrams 43 to 46 show $\sigma_{01}$ for the inert gases Ne, Ar, Kr and Xe. It is clearly seen that $\sigma_m$ does not occur at the same velocity in all gases. Hence the adiabatic criterion cannot be applied to the process $\sigma_{01}$, and in particular cannot be used to determine possible origins of the dual maxima in $\sigma_{01}$ in He.

There is an enormous difference between the present values and those of Fogel (1958) for $\sigma_{01}$ in Kr and Xe. There is insufficient data in Fogel's paper to permit an
DIAG. 47. DEPENDENCE OF $\log \sigma_{01}$ ON $E^{-1/2}$

BATES (1957) BORN APPROX. IN He

$\sigma_{01} \cdot \text{cm}^2 / \text{atom}$

$10^{15}$

$10^{16}$

$10^{17}$

$E^{-1/2}$ $10^{-2} \text{ ev.}$

0 1 2 3
investigation of beam geometry to determine probable errors and to enable a comparison to be made with the present geometry. The author's attention has recently been drawn to the measurements of $\sigma_{01}$ in Ar and Kr by Soloviev (1962). His values are in good agreement with the present values. At all energies in the range 2 - 50 Kev both the present and Soloviev's data show $\sigma_{01}$ values which increase with increasing atomic number of the target gas. This is in marked contrast to Fogel's results which show $\sigma_{01}$ decreasing with increasing atomic number for $H^0_1$ energies less than 20 Kev. It is difficult to find a reasonable explanation of Fogel's result.

Despite the inability of the adiabatic criterion to predict the velocity at which the $\sigma_{01}$ cross section maximum occurs, some success has been achieved in fitting the growth of cross section with $H^0_1$ energy, to an exponential form (equation 1.36). Diag. 47 shows a plot of $\log \sigma_i$ against $E^{-\frac{1}{2}}$ for all the inert gases. The slope of each plot increases with increasing atomic number except for Xe, for which the slope is the same as in Ar. An indication of the significance of these graphs may be obtained from their comparison with the only available theoretical prediction for $\sigma_{01}$, that of Bates and Williams (1957) in He. Their Born approximation calculations, shown in diag. 42, agree fairly well with experimental values above 50 Kev. At lower energies their predictions
Diagram 49.

- Projectile: $H_1$
- Target: Ne, Ar
- Cross Section: $\sigma_{T1}$

The diagram shows the projectile energy on a logarithmic scale ranging from 1 to 100 keV. The cross section is plotted on a logarithmic scale ranging from $10^{-17}$ to $10^{-16}$ cm$^2$/atom. The data points are labeled as 'PRESENT DATA' for Ar and Ne. The line labeled 'FOGEL 1957' connects the data points for Ar.
are considerably smaller than experimental values and this behaviour is attributed to the anticipated failure of the Born approximation at low energies. But if one plots their predicted values in this low energy range on a semilog scale, as in diag. 47, their values conform to the relation $\sigma = 1.2 \exp(-32.3E^{-\frac{1}{2}})$. The slope of this graph is seen to agree, within 10%, with that found experimentally for He namely $\sigma = 3.8 \exp(-35.4E^{-\frac{1}{2}})$. This agreement may well be fortuitous because, in the energy region where the relationship is valid, the Born approximation is expected to fail. Further implications of this agreement are discussed after the other electron loss cross sections $\sigma_{10}$ and $\sigma_{11}$ are considered.

4.6 The Double Electron Loss Cross Section $\sigma_{11}$

Diagrams 48 and 49 show (a) the present experimental values of $\sigma_{11}$ for H$^-$ in H$_2$, He, Ne and Ar and (b) the only published values of $\sigma_{11}$ which were obtained by Fogel (1957). The agreement of the two sets of measurements is generally only fair, but it is within the experimental accuracy of $\pm40\%$ of Fogel's values. The present values are accurate to within 5%. (Chapter 4.1)

There are notable similarities between $\sigma_{11}$ and $\sigma_{01}$.

(a) For a given target gas both $\sigma_{11}$ and $\sigma_{01}$ have a similar energy dependence. For example, in He, both $\sigma_{11}$ and $\sigma_{01}$ have the same shape with a maximum value at the same projectile energy of 10 Kev. (b) At a given
projectile energy, the value of $\sigma_{11}$ in any target gas relative to the value of $\sigma_{11}$ in any other target gas is the same as the similar ratio for $\sigma_{10}$. Generally, at any projectile energy, both $\sigma_{11}$ and $\sigma_{01}$ increase as the atomic number of the target atom increases except for the anomalous position of He in which both $\sigma_{11}$ and $\sigma_{01}$ are larger than $\sigma_{11}$ and $\sigma_{01}$ in Ne at energies below about 10 Kev.

However the main interest in $\sigma_{11}$ arises from the possible effect of the process $10/10ee$ upon the measurements of $\sigma_{10}$ by Hasted (1955) and Stier and Barnett (1956).

### 4.7 The Single Electron Loss Cross Section $\sigma_{10}$

It was mentioned in Chapter 1.1 that Hasted (1952, 1955 and 1956) has measured ($\sigma_{10} + 2\sigma_{11}$) while Stier and Barnett (1956) and Whittier (1954) have measured ($\sigma_{10} + \sigma_{11}$). Branscombe (1956) has suggested that the difference between their results in the noble gases could be due to large values of $\sigma_{11}$ compared with $\sigma_{10}$. In principle one could obtain values for $\sigma_{11}$ by merely subtracting Stier's measurements from Hasted's. This method is subject to great inaccuracy as the difference between their results is often less than the error of their measurements; in particular in diag. 51 for $\sigma_{10}$ in He at energies less than 6 Kev Stier's values of ($\sigma_{10} + \sigma_{11}$) are greater than Hasted's ($\sigma_{10} + 2\sigma_{11}$).
DIAG. 50(a). GROWTH OF $H_i^0$ WITH $n^1$ FOR 15 KeV $H^-_T$ IN NEON.

DIAG. 50(b). GROWTH OF $H_i^0$ WITH $n^1$ FOR 45 KeV $H^-_T$ IN KRYPTON.
CROSS SECTION — CM / ATOM

PROJECTILE — H
TARGET — He
CROSS SECTION — $\sigma_{10}$

Diagram 51.

Present data $\sigma_{10}$

$\sigma_{10} + 2\sigma_{11}$

Stier & Barnett 1956
Hasted & Steedford 1955
Hasted 1952
Sida 1954 – Born Approx.

Cross Section – CM$^2$/Atom

Present data $\sigma_{10}$

$\sigma_{10} + 2\sigma_{11}$

Stier & Barnett 1956
Hasted & Steedford 1955
Hasted 1952
Sida 1954 – Born Approx.

Projectile Energy – KEV.
PROJECTILE: $H_1$
TARGET: Ne
CROSS SECTION: $\sigma_{1o}$

**Diagram 52.**

- Present Data
- $\sigma_{10} + 2 \sigma_{T1}$
- STIER & BARNETT 1956
- HASTED & STEDEFORD 1955
- HASTED 1952

**Axes:**
- **X-axis:** Projectile Energy - KeV
- **Y-axis:** Cross Section - $cm^2/atom$

**Legend:**
- Present Data
- $\sigma_{10} + 2 \sigma_{T1}$
- STIER & BARNETT 1956
- HASTED & STEDEFORD 1955
- HASTED 1952
PROJECTILE - $H^-_1$
TARGET - Ar
CROSS SECTION - $\sigma_{10}$

**Diagram 53.**

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

- ○ ○ ○ PRESENT DATA
- " " $\left[\sigma_{T0} + 2 \sigma_{T1}\right]$
- STIER & BARNETT 1956
- HASTED & STEDELFORD 1955
- HASTED 1952

**Axes:**
- **Y-axis:** CROSS SECTION - $\text{cm}^2/\text{atom}$
- **X-axis:** PROJECTILE ENERGY - KEV.

**Scale:**
- Y-axis: $10^{-16}$ to $10^{-15}$
- X-axis: 1 to 100 KEV.

**Note:** The graph illustrates the relationship between projectile energy and cross section for different targets and projectile types.
This chapter now presents the first direct measurements of \( \sigma_{10} \). Sample curves of the growth of collision product with increasing target gas number density are shown in diag. 50(a) for 15 Kev \( H_1^- \) in Ne and in diag. 50(b) for 45 Kev \( H_1^- \) in Kr. Example (a) was a typical growth-of-current curve while example (b) shows the largest value of the ratio \( \frac{H_0}{H_1} \) which was used for a \( \sigma_{10} \) measurement.

Diagrams 51 to 55 show the cross section \( \sigma_{10} \) as a function of \( H_1^- \) energy for the target gases of He, Ne, Ar, Kr and Xe respectively. In all five inert gases the present values of \( \sigma_{10} \) are 10% lower than Hasted's 1952 data and Stier's 1956 data. When \( \sigma_{11} \) and \( 2\sigma_{11} \) are added to the present values the agreement with Stier's and Hasted's (1952) data, respectively, is quite good.

There is support for Hasted's 1955 data only in He. The present values of \( \frac{\sigma_{10}}{10} + 2\sigma_{11} \) become increasingly smaller than Hasted's values as the target gas atomic number increases. Let us examine Branscombe's (1957) suggestion that the sharp increase in Hasted's values around 8 Kev could be due to a sharp increase in the \( \sigma_{11} \) values. Diagrams 48 and 49 show that \( \sigma_{11} \) does increase sharply in the energy region 7 to 10 Kev which is precisely where Hasted's \( \frac{\sigma_{10}}{10} + 2\sigma_{11} \) values increase sharply. However, as seen in diagrams 52 and 53 from the present values of \( \frac{\sigma_{10}}{10} + 2\sigma_{11} \) for Ne and Ar, the values of \( \sigma_{11} \) are not large enough and do not increase quickly enough
DIAG. 54.

PROJECTILE — \( H^+ \)
TARGET — \( Kr \)
CROSS SECTION — \( \sigma \)

\[ \text{CROSS SECTION} = \frac{\text{CM}^2}{\text{ATOM}} \]

\[ \sigma = 10^{-14} \]

\[ \sigma = 10^{-15} \]

\[ \sigma = 10^{-16} \]

PROJECTILE ENERGY — KEV.

- ○ ○ ○ PRESENT DATA
- " " \( \sigma \) \( \left[ \frac{1}{10} + 2 \sigma_{II} \right] \) (FOGEL)
- HASTED & STEDEFORD 1955
- HASTED 1952
CROSS SECTION - CM\(^2\)/ATOM

PROJECTILE — H\(^+\)
TARGET — Xe
CROSS SECTION — \(\sigma_{10}\)

DIAG. 55.

PRESENT DATA

\[ \sigma_{10} + 2 \sigma_{11} \] (FOGEL)

HASTED & STEDEFORD 1955
HASTED 1952

PROJECTILE ENERGY — KEV.
CROSS SECTION - CM / ATOM

PROJECTILE — $^1H$
TARGET — $H_2$
CROSS SECTION — $\sigma_{10}$

PRESENT DATA
HASTED & SMITH 1956
STIER & BARNETT 1956
WHITTIER 1954
HUMMER 1960
MCDOWELL 1959

PROJECTILE ENERGY — KEV.
CROSS SECTION — CM$^2$/ATOM
with increasing projectile energy to account satisfactorily for Hasted's 1955 values. Similar conclusions are readily reached in Kr and Xe by comparing the sum \( (\sigma_{10} + 2\sigma_{11}) \), in which the present values of \( \sigma_{10} \) and Vogel's 1957 values for \( \sigma_{11} \) are used, with Hasted's data. A possible explanation of this discrepancy may lie in the following facts. (a) Hasted used the same method, but a different apparatus, for his cross section measurements in 1952 and 1955. (b) Hasted's actual experimental points have been plotted on his data in diagrams 51 to 55 to reveal that he has generally only one or two points to support the controversial low energy (5 to 10 Kev) region of his results. Therefore some doubt exists about his values in this region.

**Hydrogen.** Diag. 55 shows the present measurements of \( \sigma_{10} \). When \( \sigma_{11} \) from diag. 48 is added to \( \sigma_{10} \) the agreement with Stier and Barnett's (1956) measurements of \( (\sigma_{10} + \sigma_{11}) \) is excellent to all energies. It should not be forgotten that the present values of \( \sigma_{10} \) have been standardized against Stier and Barnett's values of \( \sigma_{10} \) at 10 Kev and hence good agreement between the values of \( \sigma_{10} \) is expected. Hasted and Smith's (1956) values of \( (\sigma_{10} + 2\sigma_{11}) \) are 20% lower than the present values of \( \sigma_{10} \) while Whittier's (1954) values of \( (\sigma_{10} + \sigma_{11}) \) are 20% higher than the present values.

There is considerable interest in \( \sigma_{10} \) in \( \text{H}_2 \) because
(a) Humber (1950) has measured $\sigma_{10}^-$ in atomic hydrogen and (b) McDowell and Peach (1959) have calculated $\sigma_{10}^-$ in atomic hydrogen.

Humber (1950) has measured a total cross section $\sigma_{10}^-$ which includes the cross sections for the processes $\tilde{\sigma}/(CC)e$ and $\tilde{\sigma}/Oleee$. From diag. 56 it is seen that $\sigma_{10}^-$ in atomic hydrogen is roughly twice $\sigma_{10}^-$ in molecular hydrogen when the cross section is expressed in units of cm$^2$/atom, i.e. for single electron detachment from $H_1$ a molecular hydrogen and an atomic hydrogen target are roughly equivalent in the energy range from 2 to 50 Kev.

The Born approximation calculations by McDowell and Peach (1959) of $\sigma_{10}^-$ for $H_1^-$ in atomic hydrogen and by Sida (1955) of $\sigma_{10}^-$ for $H_1^-$ in helium may be discussed and compared on two aspects (a) the shape of the cross section versus energy relationship and (b) the absolute values of the cross section. These two aspects may be discussed independently since the energy variation of the cross section is not sensitive to the choice of wave function which determines the value of the cross section.

Both Sida (1955) and McDowell's (1959) predictions for $\sigma_{10}^-$ have the same energy dependence as the experimental values for $H_1^-$ energies above 5 Kev. Below 5 Kev the theoretical curves both decrease with decreasing energy only slightly more rapidly than the experimental curves. McDowell suggests that his predictions are
reasonably accurate down to about 50 Kev, when, due to the failure of the Born approximation, the predicted values should considerably overestimate the true $\sigma_{10}^-$. This suggestion is contrary to fact, when, as is seen in diag. 56, the predicted cross section is, at all energies, less than the measured cross section and, when the predicted values departs from the experimental values, the predictions underestimate the true values. A similar observation is made between theory and experiment for $\sigma_{10}^-$ in He of diag. 51. It appears then that the Born approximation may be valid for $H^{-}$ energies as low as 5 Kev for $10/00e$ processes.

McDowell (1959) has shown that the absolute value of the cross section is quite sensitive to the wave functions used. For ease of computation he made several simplifying assumptions which probably prevented any close agreement of theory with experimental values. His predictions shown in diag. 56 were obtained by using simple hydrogenic orbitals for $H^{-}$ and representing the ejected free electron as a plane wave. The use of any other simple wave functions reduced his predicted values and so gave poorer agreement with the experimental values.

The experimental data in $H_2$ or the inert gases does not show any subsidiary maxima or other suggestions of excitation of the target atom simultaneous with electron loss from $H^{-}$. However the predictions of McDowell (1959),
Diagram 57. Born approx. for $\sigma_{\text{total}}$ partial cross sections.
shown in diag. 57, indicate that the cross section $\sigma_{10}$ in which the target H atom is left in a 1s state has its maximum at lower velocities than do those in which the H atom is left in the 2p state. As seen from diag. 57, this results in a much slower fall off of $\sigma_{10}$ with increasing energy than would otherwise be expected. Further, for any given state of the target atom, the major contribution to $\sigma_{10}$ comes from single transitions of the $H^-$ ion, that is, from electron loss from $H^-$ without excitation of the residual projectile H atom. The sum of all these partial cross sections for capture into a given state is seen to give the best agreement with the present experimental data.
SECTION B.
Charge Changing and Dissociation Cross Sections of Diatomic and Triatomic Hydrogen Molecular Ions, \( H_2^+ \) and \( H_3^+ \), in Collisions with Hydrogen and the Inert Gases.

Chapter 5
Introduction.

5.1 The Need-to-Know.

Much of the recent interest in collisions involving fast \( H_2^+ \) and \( H_3^+ \) ions has been stimulated by the need to know the collision cross section values in the fields of thermonuclear energy production and, to a much lesser extent, in high voltage glow discharges.

One possibility of producing a plasma of sufficiently high density and temperature for a thermonuclear reaction is the Oak Ridge D.C.X. Experiment (Snell, 1959) in which a preheated, or accelerated, beam of \( H_2^+ \) ions is injected into a region where the \( H_2^+ \) ions are dissociated and the resulting \( H_1^+ \) ions are trapped, or contained, by a magnetic field. For the optimization of this process a knowledge is required of the cross sections for the production and loss of the molecular and atomic ions.

High voltage (\( \sim 100 \text{ Kv} \)) glow discharges, maintaining currents up to 10 amps for several microseconds in tubes filled with deuterium and using deuterium loaded cathodes, have been found to produce neutron yields of the order of \( 10^5/\text{sec} \) (Kelley, 1961). Existing glow discharge theories
do not seem to apply (McClure, 1961) to the condition of voltage, current and pressure under which such high voltage discharges operate. The mechanism of these discharges is thought (McClure, 1961) to be strongly dependent on the loss processes for \(D_2^+\) ions, for which the cross sections were not known. However the interest of this thesis in collisions involving fast \(H_2^+\) ions is of a fundamental nature. The \(H_2^+\) molecular ion is the simplest of all non-monatomic molecules and it introduces into the charge changing processes the possibilities of dissociation and vibrational and rotational excitation which are not present in atomic beam collisions.

5.2 The Collisions.

For a complete description of any collision it would be necessary to determine the charge state, the velocity, the direction of travel and all the quantum numbers necessary for a unique description of a given particle for all the particles, and photons, involved both before and after the collision. The attempts to measure the charge changing and dissociation cross sections for the collision of \(H_2^+\) ions with gases will be discussed on the basis of (a) their success in obtaining part of the above information and (b) how the molecular nature of the projectile, \(H_2^+\), influences the consideration of (i) the method of cross section measurement and particle detection,
(ii) the beam geometry and the impact of projectile ions on surfaces.

(iii) the method of production of the $H_2^+$ and $H_3^+$ ions.

The complete description of a collision is far from being realized. The technique of crossed beams (Fite, 1960), which enables the momentum analysis of both projectile and target particles, both before and after collision, when coupled with a coincidence counting method, would provide a link between the older methods of analysis of either the fast projectiles or stationary target particles. However, the crossed beam method has been used with $H_2^+$ ions only in the photodissociation experiments of Dunn (1962) and the analysis of the slow target atoms has been made by Hasted only for electron capture by $H_2^+$ in hydrogen gas. The following discussion is then concerned with the collision cross sections for the change of charge-to-mass ratio for fast $H_2^+$ projectiles with little consideration being given to the target particle except to regard it as providing the electrons to maintain a charge balance for the collision. The possible processes which must be considered for the $H_2^+$ ion are as follows:

1. $H_2^+ \rightarrow H_2^0$ - ordinary charge transfer with the capture of a single electron.

2. $H_2^+ \rightarrow H_1^0 + H_1^+$ - dissociation without electron transfer

3. $H_2^+ \rightarrow 2H_1^+$ - " with one electron loss.
4. \( H_2^+ \rightarrow 2H^0 \) \{ dissociation with one \\
5. \( H_2^+ \rightarrow H_1^+ + H_1^- \) \}
6. \( H_2^+ \rightarrow H_1^O + H_1^- \) \{ two electron capture \\
7. \( H_2^+ \rightarrow 2H_1^- \) \{ three \\

These dissociative collisions of the \( H_2^+ \) ion are simpler than those with other molecules since the dissociation products \( H_1^+ \) and \( H_1^- \) have no excited states (see Section A, Chapter 1.1). Only one estimate of \( H_1^- \) ion production has been published. Fedorenko (1959) has shown that the total \( H_1^- \) production cross section, i.e. the sum \( (\sigma_5^- + \sigma_6^- + 2 \sigma_7^-) \), where \( \sigma_n^- \) is the cross section for process \( n \) above, is approximately \( 2 \times 10^{-18} \) cm\(^2\)/mol. for 12 Kev \( H_2^+ \) ion incident on argon. Other investigators appear contented to show that \( H_1^- \) production presents a negligible loss process from the primary beam. Chapter 8 of this thesis presents the results of what is believed to be the first systematic measurement of the total \( H_1^- \) production cross section for \( H_2^+ \) incident on hydrogen and the inert gases.

The determination of the cross sections for the individual collision processes requires an electric or magnetic field to separate the \( H_2^+ \), \( H_1^+ \) and uncharged particles followed by either particle scintillation counters (Sweetman, 1960; Guidini, 1961) or gas proportional counter (Schmid, 1961; McClure, 1963) to distinguish double events from single events for particles
of the same e/m. To distinguish the $2H_1^0$ event from the $H_2^0$, a fine slit-scanning technique (Sweetman, 1960) has been used. Coincidence counting has been used by Guidini to record simultaneous events at the different e/m counters and thus determine a cross section such as $\sigma_2$.

Using these particle counting techniques the above workers have obtained excellent agreement in the measurement of neutral particle production cross sections, $\sigma_{H_2^0}$, $\sigma_{H_1^0}$ (equal to $\sigma_2 + 2 \sigma_4 + \sigma_6$) and $\sigma_{2H_1^0}$, for $H_2^+$ ions incident upon hydrogen gas as shown in diag. 58. Sweetman (1960) appears to have satisfactorily explained the angular distribution of the fast neutral collision products and the relatively high value (70%) for the dissociation of the fast $H_2^0$ formed by single electron capture of the $H_2^+$ projectile. In view of this satisfactory agreement in the measurements of neutral particle production those cross sections have not been measured in this thesis. Also the secondary electron emission detectors, used for neutral atom, $H_1^0$, detection in Section A, do not permit the separation of $H_1^0$ and $H_2^0$.

Many measurements have been made of the total proton production cross section, ($\sigma_2 + 2 \sigma_3 + \sigma_5$), mainly because of the simplicity of using an integrated charge collection method with large beam currents. Such a detection method has the inherent source of error in that it must be demonstrated that all secondary electron effects.
have been eliminated. Despite the claims that adequate precautions against such effects have been made, there exists a much greater spread of cross section values from such detectors than from those experiments using energy sensitive detectors. These discrepancies are shown in diag. 59 for $H_2^+$ ions incident upon hydrogen gas. McClure (1963), Schmid (1961) and Guidini (1961) used energy sensitive detectors while all other workers used Faraday cups.

However whilst there exist marked differences in the absolute values of the cross section above 25 Kev, there is good agreement in the energy dependence of the cross section. This fact suggests that the discrepancies between values may have their origin in an energy independent aspect of the measurement, for example in the absolute determination of the target gas pressure which is not easy to measure within 1% with a McCleod Gauge in the pressure region around $10^{-4}$ mm Hg. Below 25 Kev the curves of diag. 59 do not agree in value or energy dependence and a more basic source of error appears likely.

Some doubt exists on the accuracy of Fedorenko's results. He appears to have been unable to measure both his $H_2^+$ and $H_1^+$ beams simultaneously and to have used different detection techniques for the two beams. However it is not clear what exact experimental method he used. He appears to have measured the primary beam with an electrode
within the target chamber and consequently considerable difficulty must have been experienced in preventing spurious currents from ions and electrons produced within the gas from reaching this detector. Such difficulties tend to place more reliability on direct particle counting techniques.

In spite of this evidence against Faraday cups their use in Section A of this thesis gave cross section values in excellent agreement with those determined by McClure (1963) using energy sensitive detectors for $\sigma_{11}$ for $H_1^+$ incident upon hydrogen. The apparatus described in Chapter 2 will enable the cross sections for total $H_1^+$ and total $H_1^-$ production to be measured in the energy range from 2 to 50 Kev where diag. 59 has shown that a wide spread of results exists.

The importance of aperture size relative to beam size, which was discussed at length in Chapter A.3. for atomic beams, does not appear to have received due consideration in the above literature. Consideration must be given to the facts that (i) molecular ions may dissociate into atomic ions by glancing collisions with the metal edges of collimating apertures (Hagstrum, 1953) and that (ii) internal energy of the molecule may be converted to a transverse translational velocity component and thus the beam profile is broadened. Chapter 6 of this thesis investigates the effect of both of these facts upon the
beam profiles of the projectile ions, both before and after collision, and hence upon the measured cross sections, at several energies within the range 2 to 50 Kev.

5.3 Ion Beam Composition.

In most of the early work it appears to be assumed that the projectile H$_2^+$ ions were predominantly in the same (ground) state before a collision; or, if some of the H$_2^+$ ions were in an excited state, then either their percentage population of the total beam was small or their cross section for a given process was not much different from that for ground state ions. However at the commencement of the present investigation (June, 1963) there were becoming available some experimental results which indicated that some sources of H$_2^+$ ions produced these ions in various electronically and vibrationally excited states, each state probably having its own particular cross section for a dissociative collision.

The most direct approach to this problem was made by Riviere and Sweetman (1961) and Kaplan et al. (1961) who determined the populations of the two uppermost vibrational levels ($v = 17$ and 18) of H$_2^+$, extracted from an R.F. source, and a P.I.G. source, respectively, by the dissociation from these levels of H$_2^+$ by electric fields and the equivalent $V \times B$ fields of the order of $10^5$ V/cm. The relative populations of these levels of H$_2^+$ from both sources were found to be approximately independent of the
source operating conditions of pressure and extraction voltage, which were varied over an unspecified range. The populations of these \( v = 17 \) and \( 18 \) levels represented less than 0.1% of the total beam current and their experiments did not give any information about any other excited states in the beam.

Barnett and Ray (1963) have measured total proton production cross sections from \( H_2^+ \) ions obtained from an R.F. source and found no dependence of the cross section on the source operating conditions of R.F. power and gas pressure. However contradictory evidence was found by McClure (1963) who showed that \( \sigma_{H_1^+} \), from \( H_2^+ \) formed in P.I.G. source, depended markedly (up to 20%) upon the source parameters of pressure and extraction voltage. Unfortunately there is no really sound basis for comparison between the above four experiments as there are too many unknown factors, in particular the relative populations of all the excited states and the source parameters which determine such populations are not known.

This thesis reports, in Chapters 7 and 8, a detailed investigation of the effect of a wide variety of operating variables of an R.F. ion source upon the total \( H_1^+ \) and \( H_1^- \) production cross sections.

Riviere and Sweetman (1961) and Kaplan (1961) have independently found that the \( v = 17 \) and \( 18 \) vibrational levels of \( H_2^+ \) are about five times more populated when the
H$^+_2$ ions are formed from the dissociation of H$_3^+$ in collisions with a hydrogen gas target than when it is formed in an ion source. Using the H$^+_2$ formed in this manner Riviere (1961) found, in the energy range 280 - 670 Kev, that $\sigma(H^+_1 + H^0_1)$ increased by 7 ± 4% and $\sigma_2H^+_1$ increased by 4 ± 4%. The following comments may be made on their results.

(a) The populations of the $v = 17$ and 18 vibrational levels did not exceed more than 0.3% of the total beam current and no information was available on the relative populations of the other vibrational levels. Hence the increases in their cross sections are not necessarily due to the increases in the $v = 17$ and 18 vibrational level populations.

(b) Riviere used an R.F. ion source in which some H$^+_2$ ions are formed from the collision of H$_3^+$ with H$^0_2$ and H$^0_1$ which process is strongly dependent upon the gas pressure. As he was unable to investigate his ion source operating conditions or to measure the gas pressure it may be expected that source operating conditions may be found which will provide much larger relative populations of the $v = 17$ and 18 vibrational levels than obtained by Riviere.

5.4 Theoretical Considerations.

The hydrogen molecular ion, H$^+_2$, as the simplest of all molecules, has been extensively investigated theoretically (Hirschfelder et al., 1955). With the approximation of
stationary nuclei the Schrödinger equation is exactly solvable for the wave function and electronic energy eigenvalues. The availability of wave functions and the relative simplicity of the system then invites using it as a prototype in calculations of important molecular collisions.

Calculations have been made of the cross sections for photoionization (Dares, 1953), dissociative recombination (Sauer, 1956; Stabler, 1952), dissociation by proton impact (Gerjuoy, 1955) and electron impact (Kerner, 1953; Ivens, 1958). Only the photodissociation calculation by Gibson (1956) may be compared with experiment. Dunn (1963) has measured the photodissociation of $H_2^+$ for wavelength in the range 3000°A to 9000°A. He obtains cross section of the order of $3.10^{-18}$ cm² which are an order of magnitude smaller than the predictions of Gibson.

The dissociation of $H_2^+$ in collisions with gases $H_2$, Ar, and $N_2$ has been studied by Salpeter (1950) using the Born approximation. Excitation of $H_2^+$ to the $\Sigma_u^+$ state and the double proton state are considered. In order to make the problem calculable he considered the probability of transfer of momentum to the electron of the projectile ion, $H_2^+$, by a stationary hydrogen atom. The total proton production cross sections were shown to be dependent on the inverse square of the projectile energy. Above 300 Kev, where the Born approximation is expected to be valid,
the agreement between theory and experiment in hydrogen (Sweetman, 1960) is fair but at lower energies the Born calculations are far too high. In argon and nitrogen the experimental data (Guidini, 1961) does not extend above 150 kev and cannot be accurately compared with Salpeter's predictions which would appear to be almost an order of magnitude too high.

5.5 The Inert Gases.

Total proton production cross sections have been measured (a) in Ne and Ar by Fedorenko (1954) over the energy range 5 to 25 kev, (b) in He by Fedorenko (1959) from 10 to 150 kev and (c) in He and Ar by Guidini (1961) from 30 to 250 kev. Chapter 8 of this thesis presents the measurement of total proton and total H\textsubscript{1} production cross sections from 2 to 50 kev for all five inert gases.
Chapter 6

Apparatus and Experimental Accuracy.

The apparatus, as described in Chapter A.2, was used without alteration, except for the collision cell exit canal and detector entrance diameters which are discussed below, for the measurements involving molecular ion projectiles. The R.F. ion source supplied ample currents of $\text{H}_2^+$ and $\text{H}_3^+$ ions of about $1\mu\text{A}$ in a 1 mm. diameter. Metallic deposits which tended to form on the walls of the discharge vessel were not cleaned off. The recombination of atomic hydrogen on the walls was thus enhanced, with a subsequent increase in the $\text{H}_2^+$, and then $\text{H}_3^+$, molecular ion content of the extracted beam.

The method of cross section measurement by the determination of the linear rate of growth of the collision products with target gas number density, as discussed in Chapter A.2, is applicable to any type of projectile particle, whether atomic or molecular in nature and irrespective of the number of possible different $\frac{e}{m}$ charge changed products that may result from collisions. Also, as for the atomic projectiles, the fact, that the extent of linearity of the collision product growth curve is dependent upon the nature of the target and the projectile molecules, the projectile energy and a complicated function of all the possible $\sigma_{if}$, necessitates that a graph of $\Delta \left( \frac{N_s}{N_i} \right)$ against $\Delta(n)$ (see Chapter 4.1) must be
GROWTH OF $H_1^+$ WITH $n'$ FOR 6 KeV $H_2^+$ IN HYDROGEN.
drawn to establish the extent of the linear relationship for every individual cross section measurement. Diag. 61 shows the growth of $H_1^+$ with target gas number density ($n'$ of equation 2 of Chapter 4.1) from 6 Kev $H_2^+$ incident upon hydrogen gas. All growth of current curves used to determine cross sections possessed only a small parabolic component, if any, and the ratio of the fast collision products to the original projectile beam current rarely exceeded 1%.

6.1 Accuracy and Validity of the Measurements.

The investigation of experimental accuracy has been even more thorough for molecular ion beams than for atomic ion beams. All those considerations discussed in Chapter A.3 have been re-examined for $H_2^+$ and $H_3^+$ projectile beams. In this chapter only those considerations are discussed in which significant differences occurred from the results for atomic ion beams.

6.11 Collision Cell Exit Canal Diameter.

For the atomic hydrogen beam work of Section A, the beam diameter was 1.0 mm., the collision cell entry canal diameter 1.5 mm. and the exit canal diameter was 2.0 mm. For molecular ion beams the possibility arises that the internal energy of the molecules may change in a collision to give rise to a transverse velocity component to its constituent atoms and hence broaden the beam. The broadening effect of any given transverse velocity
RELATIVE BEAM AMPLITUDES

MAGNETIC FIELD GAUSS.

- $H_1^+$
- $H_2^+$

$2.0 \times 10^{-6} \text{ mm.Hg.}$
$1.1 \times 10^{-4} \text{ mm.Hg}$
component on an ion beam will naturally be largest when the ion is travelling at its slowest forward velocity which occurs for the 2 Kev $H_2^+$ beam in this present work. Therefore the currents of collision products $H_1^+$ and $H_1^-$ from a 2 Kev $H_2^+$ beam traversing Xenon were examined as a function of the exit canal diameter. Canal diameters of 2.0, 2.5, 3.0, 5.0 and 10.0 mm. were used. The collected $H_1^+$ and $H_1^-$ beam currents were independent of canal diameters which were equal to or larger than 3.0 mm. Therefore an exit canal diameter of 3.0 mm. has been used for all $H_2^+$ charge changing cross section measurements.

6.12 Detector Aperture Diameter.

Diag. 60 compares the detector responses as the following beams are swept across the detectors for (a) a 10 Kev primary $H_2^+$ beam at pressures of $2 \times 10^{-6}$ and $1.1 \times 10^{-4}$ mm. Hg. in the collision cell and (b) the $H_1^+$ and $H_1^-$ collision product beams resulting from beam (a). The detector responses are shown as values relative to the maximum beam currents detected for each beam of a given $e/m$. Similar graphs were obtained for primary $H_2^+$ energies of 2 and 50 Kev. A detector aperture diameter of 12 mm. was found to be adequate to collect all the collision products of both $H_1^+$ and $H_1^-$. It is noted that for the present measurements of total charge changing cross sections for a given $e/m$ product, a high momentum resolution of the product beam is not required. Therefore the detector
<table>
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Table 6

Deflection Voltage
Target gas pressure
aperture may be made very large, much larger than the necessary minimum size, provided of course that the necessary potential well may be maintained across the detector entrance to ensure 100% containment efficiency.

6.13 Impurities in the Primary Beam

Table 6 gives the results of the current measurements to the $H_1^+$, $H_1^-$ and neutral particle detectors for (i) maximum and minimum target gas pressures (ii) for a large voltage between the deflector plates $P_1$ and $P_2$ of diag. 13 prior to the entry canal with a 10 KeV $H_2^+$ primary beam incident on hydrogen gas.

In Table 6, column 5, the title $\geq (\gamma H^0)$ represents the sum of $\gamma(H_2^0)N(H_2^0) + \gamma(H_1^0)N(H_1^0)$ where $N(H_2^0)$ and $N(H_1^0)$ are the numbers of uncharged hydrogen molecules and atoms, respectively, entering the neutral atom detector per second. By assuming $\gamma(H_2^0)$ equals twice $\gamma(H_1^0)$ (at the same particle velocity) one may obtain a rough indication of the number of neutral particles/second formed in the beam.

From Table 6, line (b) it is readily seen that not more than 0.01% of the primary $H_2^+$ ions were neutralized or dissociated on the edges of the beam defining apertures $A_2$ and $A_3$ (of diag. 13). Such a percentage would have a negligible effect upon the measured cross sections.

Those error sources which depend upon cross section values (as mentioned in Chapter 3) will naturally change when the projectile changes from atomic hydrogen to the
H$^+$ molecular ion and the number of possible $\sigma_{if}$ increases from 3 to 5. However, Table 6 shows that such errors in $\sigma_{H_1^+}$ are negligible and in $\sigma_{H_1^-}$ amount to $\frac{6}{262-10} \cdot 100\%$ or 2.4%. This error in $\sigma_{H_1^-}$, which is due to impurity atoms in the primary H$^+$ beam, has been estimated and allowed for in all $\sigma_{H_1^-}$ measurements since it is a simple procedure to apply the deflection voltage, V, between plates P$_1$ and P$_2$.

All other possible errors may be accounted for in a similar manner to that used in Chapter 3 for primary proton beams. The total percentage error in any cross section value should be less than 5% which is composed of (a) $\pm$ 1% in the measurement of each of the beam currents and the target gas number density, n$l$, and (b) 2% in the measurement of the slope of the growth of collision product curves.
The Electrodeless Discharge Ion Source

7.1 Introduction. The electrodeless discharge type ion source, of which reviews have been made by Thoneman (1953) and Francis (1960), was selected originally for the study of Section A of this thesis because of its promise of providing a hydrogen ion beam which has a high proton percentage with a low energy spread. The work described in this chapter originated from the process of optimization of the source operating conditions and is presented at this stage of the thesis because of the complex dependence of the dissociation cross sections of the extracted $\text{H}_2^+$ and $\text{H}_3^+$ molecular ions upon the source operating conditions by way of (a) the mean beam energy and (b) the populations of the vibrationally excited states of the molecular ions.

It is convenient to consider three phases in the production of an ion beam: (a) the establishment of a high degree of ionization, (b) the extraction of a large number of ions from the discharge and (c) the mean energy and the energy spread of the extracted ions. These three phases will be considered in turn.

(a) High ion densities are readily, and most usually, attained by increasing either the gas pressure or the R.F. excitation power. The former is limited by electrical breakdown in the extraction region or by the tolerable limit of neutral gas flow from the source. The latter is
readily achieved. However the present investigation has sought to attain a higher ion density by using static magnetic fields to cause an equivalent increase in gas pressure (von Engel, 1934). Neuert (1954) and Swann (1952) reported a resonance between the R.F. excitation frequency and the electron cyclotron frequency for a transverse magnetic field with an enhanced ion density. Under such conditions a much larger ion current was obtained with the use of a transverse, rather than a longitudinal, field. On the contrary Hall (1943) and Bailey (1948) have obtained larger ion currents by the use of axial (longitudinal) magnetic fields. The present study has sought to find an optimum static magnetic field at various gas pressures and excitation frequencies.

(b) The problem of optimum extraction of ions from the discharge has been studied by Eubank (1954), Moak (1951) and Thoneman (1953). The latter author has discussed this problem in a similar manner to that used for designing electron guns of high perveance. The problems involved are similar in that the ion flow in space charge limited and the ions are emitted from an equipotential "surface" with negligible initial velocities. The present study systematically determines an optimum electrode geometry.

Unfortunately the two phases (a) and (b) have been shown by Eubank (1954) to be interdependent. For given dimensions of the discharge tube and extraction electrodes,
DIAG. 62. EXTRACTION ELECTRODE GEOMETRY
the focusing characteristics of the plasma boundary depend upon the extraction field strength and the plasma density, which varies with the gas pressure and the R.F. power input to the discharge, which in turn varies with the plasma density. As a consequence of these complex relationships it is not possible to examine the source characteristics as a function of only one of these variables. However it is possible to note some general patterns in the source and extracted beam characteristics and from these to draw a qualitative picture of the processes governing beam extraction.

(c) The determination of the mean beam energy and the energy spread of the extracted ion beam had received little attention in the literature at the commencement of this investigation in 1961. The present investigation determines these beam characteristics as a function of the source parameters mentioned above.

Source Characteristics

7.2 Optimum Electrode Geometry.

Diag. 62 shows the basic electrode geometry which is similar to that used by Bailey (1948). The 60 watt output R.F. oscillator and other ion source circuitry is given in appendix 2. The electrodes were raised above the level of the brass base plate to (a) reduce eddy current losses in the metal flanges, (b) permit axial movement of the axial magnetic field coil so as to vary the convergence of the
EXTRACTION CANAL LENGTH — mms.

Q, HYDROGEN GAS CONSUMPTION — CC./HR. AT 675 mm. Hg.

$I_F$, TOTAL BEAM CURRENT — $\mu$A

DIAG. 63.
flux lines onto the extraction aperture. The simple electrode shapes of diag. 52 were used since Thoneman (1953) had shown that the effect of electrode shape on the ion trajectories is of small importance compared with the effect caused by the curvature of the plasma boundary. The efficiency of a given electrode configuration in extracting ions from the source is investigated by maintaining the gas pressure and the R.F. excitation output power, and hence the plasma density, constant and adjusting the extraction voltage to give the maximum total extracted positive ion current. The basic electrode configuration of diag. 52 was investigated for the following dimensions: for $d_c = 2.0$ mm and $l_c = 8.0$ mm, $d_e$ was varied from 2 to 6 mm. in steps of 1 mm. and, for each $d_e$, $l_ec$ was varied from 0.2 to 3.2 mm. in steps of 0.5 mm. Optimum relative dimensions are $d_e = 1.5 \, d_c$ and $l_ec = 0.6 \, d_c$ within $\pm 10\%$ accuracy. These relationships were found to hold within 10% for $d_c = 1.0$ and 3.0 mm. These optimum relative dimensions agree closely with those values given by Eubank (1954) and Thoneman (1953).

Diag. 63 shows the measured dependence of the extracted beam current on the extraction canal length, $l_e$, for the value of $d_c = 2.0$ mm., and the estimated values of neutral gas flow from the source as a function of the canal length. A value of $l_c = 8$ mm. was selected as a reasonable compromise between high beam current and low gas flow.
OSCILLATOR FREQUENCY — 16.7 Mc/sec.

TOTAL BEAM CURRENT — \( \mu \text{A} \)

20\( \mu \)  
8\( \mu \)  
6\( \mu \)  

2\( \mu \) = GAS PRESSURE

(TRANSVERSE MAGNETIC FIELD) COIL CURRENT — AMPS

DIAG. 64.
7.3 Static Magnetic Fields.

The influence of a static magnetic field on the source characteristics is shown in diag. 54 and 65. Diag. 54 shows the manner in which the extracted positive ion current depends upon the transverse magnetic field over the range of gas pressures from 2 to 20 \( \mu \) Hg. A similar curve for 30 \( \mu \) Hg gas pressure was obtained but it was almost coincident with that obtained at 20 \( \mu \) Hg pressure. For all gas pressures above 4 \( \mu \), there is, at a transverse field of 6 gauss, a pronounced peak in the extracted ion current. This peak may be explained by an enhancement of the plasma density by a resonance in the electron energy since the electron gyrofrequency, around a field of 6 gauss, is exactly equal to the excitation frequency of 16.7 Mc/sec.

This explanation requires, for the present discharge which is excited within the tank coil of the R.F. oscillator, that the axial electrostatic force due to the voltage difference between the ends of the coil, rather than the electromagnetic force, which is normal to the coil axis, be the dominant force on the electrons. This requirement was tested as follows. A cylindrical electrostatic shield was placed between the R.F. coil and the discharge vessel. The shield was constructed from thin copper strips 2 x 70 mm. which were placed parallel to the discharge axis and spaced 5 mm. apart and joined at
one end. When the discharge was operating under the same conditions as were used for diag. 63, it was found that earthing the electrostatic shield caused a 20% reduction in beam current at a gas pressure of 20 microns Hg. This reduction in the beam current became greater as the gas pressure was reduced until, for pressures less than 5μHg, the discharge could not be maintained. These observed reductions in beam current must be attributed only to the reductions in the electrostatic field which, therefore, appears to be of increasing importance in maintaining the discharge as the gas pressure decreases until for pressures less than 5μHg, the electrostatic field alone appears to maintain the discharge.

It appears then that, at low gas pressures in particular, the electrons move in crossed steady magnetic and alternating electric fields. For such electron motion, Francis (1960) has shown that the average energy, ΔE, gained by an electron between collisions is given by the expression

\[
\Delta E = \frac{e X^2}{4m} \left\{ \frac{1}{(w - w_H)^2 + \nu_e^2} + \frac{1}{(w + w_H)^2 + \nu_e^2} \right\}
\]

where \( w_H = \) The electron gyrofrequency = \( \frac{e H}{2\pi m c} \)

\( \nu_e = \) electron collision frequency

\( \approx 5.9 \times 10^9 P \) (Brown, 1959)

and P is the gas pressure (mm.Hg)

\( \omega = \) R.F. excitation frequency
\[ X_0 = \text{amplitude of the electric field (e.s.u/cm)} \]

For diag. 64, \( \omega = 10^8 \text{ c/sec.} \) When \( P = 1 \mu \), \( \gamma_e = 6 \times 10^6 \ll \omega \)

When \( P = 20 \mu \), \( \gamma_e \approx \omega \). From the above expression, when \( \omega_H = \omega \), \( \Delta E \) shows a resonance which is large when \( \gamma_e \ll \omega \) and which is negligible when \( \gamma_e \gg \omega \). When such a resonant increase in the mean electron energy occurs, one may expect a corresponding increase in the ion density provided that the cross section for ion production is still increasing as the electron energy increases. Goodyear (1961) has shown that the most probable process for proton production is

\[ H_1^0 + e \rightarrow H_1^+ + 2e \]

which has a maximum cross section at an incident electron energy of 40 ev. Therefore one may expect the largest increase in ion density when the mean electron energy is increased to above 40 ev.

This behaviour of \( \Delta E \) with pressure is seen to be very similar to the behaviour of the extracted positive ion current with pressure as shown in diag. 64. The ion current resonance is very large at a pressure of 6 \( \mu \)Hg but it becomes obscured at pressures above 20 \( \mu \)Hg when the electron motion is interrupted by collision with the gas molecules. At a pressure of 6 \( \mu \)Hg, a second, smaller resonance is obtained when the electron gyrofrequency is twice the applied frequency. The completely different shape, and much decreased value, of the curve obtained at
TOTAL BEAM CURRENT

EXTRACTION VOLTAGE ADJUSTED FOR MAXIMUM BEAM CURRENT
OSCILLATOR FREQUENCY — 16.7 Mc/sec.

(TOTAL BEAM CURRENT — μA)

GAS PRESSURE

(AXIAL MAGNETIC FIELD) COIL CURRENT — AMPS

DIAG. 65.
a pressure of 2\(\mu\) Hg may be explained by the fact that, at this pressure, the mean free path is 6.5 cm which is larger than the discharge vessel diameter of 5.0 cm. Thus the electrons will collide predominantly with the discharge vessel walls from which must originate further electrons to maintain the discharge.

The results shown in diag. 64 are sufficient to suggest possible reasons for the discrepancies between the results of those workers mentioned in Chapter 7.1(a) concerning the magnitude of the enhancement of the extracted ion current as a function of the transverse magnetic field. None of those workers investigated the pressure dependence of the resonance which has been shown here to have a marked effect on the magnitude of the resonance. Equation(7.1) also suggests that the R.F. input power will have a large effect on the size of the resonance however this has not been investigated here. At pressures above 20\(\mu\), where the discharge is maintained predominantly by the electromagnetic field of the exciting coil, the transverse magnetic field will act mainly to reduce electron diffusion to the ends of the discharge.

Diag. 65 shows the dependence of the extracted ion current on the static axial magnetic field for several gas pressures in the range from 4 to 20\(\mu\) Hg. The magnetic field is now parallel to the direction of the electrostatic force. The magnetic field acts now to reduce
the diffusion of the electrons to the walls in a direction perpendicular to the field (Francis, 1960). The effects of reduced electron diffusion to the walls on the plasma density are seen to be large for the present discharge dimensions where the electron mean free path is of the order of the discharge vessel dimensions, and where there are several collisions of the electrons with the gas atoms per oscillation of the electric field. At gas pressures above $20\mu\text{Hg}$, the dependence of the extracted ion current on the static magnetic field is similar for both transverse and axial directions. This similarity confirms the result that for high gas pressures the transverse magnetic field merely acts to reduce electron diffusion to the discharge walls. There is no resonance as the electron motion is interrupted by collisions and the electrostatic field within the gas is greatly reduced due to the increased conductivity.

Diagram 65 shows that the extracted ion current, and hence the ion density within the discharge, is enhanced by the axial magnetic field to an extent which depends markedly on the gas pressure. As the plasma density is determined by both the gas pressure and the R.F. excitation power, it is probable that for R.F. power inputs, greater than 50 watts, to the discharge that the effects of the transverse magnetic field upon the extracted ion current may be quite different from the
GAS PRESSURE = 4 \times 10^{-3} \text{ mm. Hg}

B AT RESONANCE

\omega = 10^8 \text{ c/sec.}

TOTAL BEAM CURRENT — µAMPS

R.F. INPUT POWER—WATTS

BEAM EXTRACTION VOLTAGE — KEV
<table>
<thead>
<tr>
<th>Ion</th>
<th>Transverse</th>
<th>Exxt</th>
<th>Exxt</th>
<th>Tr</th>
<th>Gas consumption</th>
<th>Gas pressure</th>
<th>Water pressure</th>
<th>Power input</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.3</td>
<td>T</td>
<td>-</td>
<td>-</td>
<td>T</td>
<td>10</td>
<td>4</td>
<td>60</td>
<td>60</td>
<td>300</td>
</tr>
<tr>
<td>0.7</td>
<td>T</td>
<td>-</td>
<td>-</td>
<td>T</td>
<td>10</td>
<td>4</td>
<td>60</td>
<td>60</td>
<td>300</td>
</tr>
<tr>
<td>1.5</td>
<td>15</td>
<td></td>
<td></td>
<td>15</td>
<td>4</td>
<td>4</td>
<td>60</td>
<td>60</td>
<td>300</td>
</tr>
</tbody>
</table>

Table 7
present results. For this reason it is difficult to compare the results of Swann (1952) and Neuert (1954) with the present results as they did not investigate either the pressure or possible input power dependence of their results.

Diss. 6 shows the dependence of the extracted ion current on the extraction voltage for various R.F. power inputs at a gas pressure of $\mu$ and with the transverse magnetic field adjusted for resonance. The peak extracted ion current, $i$, is approximately proportional to the $3/2$ power of the extraction voltage at each power input. If a simple Pierce electron gun model is applied to the ion source, as suggested by Thoneman, (1953), the constant value of the ratio $1/V_e$, the perveance, may be interpreted as indicating that the emitted ion current is space charge limited and the emitting surface has the same curvature for optimum ion extraction conditions for a given electrode geometry.

The results of this chapter have been used to select those optimum operating conditions for the extraction of the maximum positive ion current from the discharge. Such an ion source is seen in Table 7 to compare favourably with other similar sources described in the literature.

7.4 Beam Characteristics

The extracted positive ion energies were measured by decelerating the ions in the region between the extraction
<table>
<thead>
<tr>
<th>CURVE</th>
<th>PRESSURE (10^3) mm Hg</th>
<th>TRANSVERSE MAG. FIELD</th>
<th>MEAN BEAM ENERGY (EV)</th>
<th>BEAM ENERGY SPREAD (EV)</th>
<th>OSCILLATOR FREQUENCY</th>
</tr>
</thead>
<tbody>
<tr>
<td>O-----</td>
<td>25</td>
<td>RES. VALUE</td>
<td>80</td>
<td>50</td>
<td>16.7 MHz</td>
</tr>
<tr>
<td>******</td>
<td>25</td>
<td>ZERO</td>
<td>145</td>
<td>36</td>
<td></td>
</tr>
<tr>
<td>X-----</td>
<td>5</td>
<td>RESONANCE</td>
<td>175</td>
<td>34</td>
<td></td>
</tr>
<tr>
<td>————-</td>
<td>5</td>
<td>ZERO</td>
<td>220</td>
<td>20</td>
<td></td>
</tr>
<tr>
<td>三角万</td>
<td>22</td>
<td>RESONANCE</td>
<td>23</td>
<td>25</td>
<td></td>
</tr>
</tbody>
</table>

"Shielded Discharge"

**Diagram 67**

**Total Beam Current — μA**

**Deceleration Voltage Minus Extraction Voltage — Volts**
OSCILLATOR FREQUENCY = 16.7 mc/sec.

\[ \text{GAS PRESSURE} \times 10^{-3} \text{ mm. Hg.} \]

\[ \text{MEAN BEAM ENERGY (EV)} \]

\[ \text{SHIELDED DISCHARGE} \]

\[ \text{H = RESONANCE VALUE} \]

\[ \text{H = 0} \]

\[ \text{DIAG. 68.} \]
EXTRACTION VOLTS 2850.
OSCILLATOR FREQUENCY 16.7 mc/sec.

GAS PRESSURE = $25 \times 10^{-3}$ mm Hg

COIL CURRENT-AMPS

DIAG. 69.
canal and the Faraday cup with the voltage difference of $V_e + \Delta V$, where $\Delta V$ is that voltage above the extraction voltage, $V_e$, which is necessary to decrease the Faraday cup current to zero. The experimental apparatus and precautions are discussed in appendix 4. Typical deceleration curves are given in diag. 67 where the extracted ion current is graphed against $\Delta V$. By differentiating such curves, the energy distribution function of the ions may be obtained, however the simpler process of estimating both the mean beam energy and the beam energy spread directly from the deceleration curves has been used. A systematic variation of all the source operating conditions was not made during this investigation. Only the gas pressure and the transverse magnetic field were varied. These parameters were those which the author, in chapter 8 of this thesis on the dissociation cross sections of $H_2^+$ and $H_3^+$ ions extracted from this ion source, had found to influence those cross sections.

Diagrams 68 and 69 show that, over the ranges indicated, the mean beam energy decreases and the beam energy spread increases as either the gas pressure or the transverse magnetic field increases, respectively.

A simple "Fierc model" of an electrodeless discharge ion source as proposed by Thoneman (1953) predicts a mean beam energy equal to the sum of the extraction voltage, $V_e$,
and the plasma potential, $V_p$. The plasma potential is given by $V_p = \frac{kT}{2e} \ln\left(\frac{m_p}{2\pi m_e}\right)$. If one assumes an electron temperature of $10^5$ °C as typical of a low pressure discharge in hydrogen, then $V_p = 25$ volts. However the present results have indicated much larger values of $V_p$ up to 220 volts. The origin of such large values of $V_p$ is indicated by the use of an earthed electrostatic shield placed inside the R.F. exciting coil. Diagrams 68 and 69 show that, for given source operating conditions, the shield reduces the mean beam energy (in excess of the extraction voltage), $\bar{E}$, from 100 ev to 15 ev and the beam energy spread, $\Delta E$, from 35 ev to 25 ev. The two quantities, $\bar{E}$ and $\Delta E$, are found to be only very slightly dependent upon the gas pressure when the shield is in position. The large values of $\bar{E}$ are thus shown to be directly attributable to the electrostatic field of the coil.

For the unshielded discharge Ero (1956, 1958) and Levitski (1958) have attributed the excess beam energy to a build up of the plasma potential due to a rectifying action on the electrons exposed to the R.F. field between the plasma and the extraction aperture and between the plasma and the walls. The rectifying action of the R.F. field on the electrons is evident in a uniform velocity due to the field of $\frac{eX_0}{m_w} \cos \phi$. For the most favourable phase $\phi = 0$, $\omega = 10^8$ rad./sec, $X_0 = 300$ V/cm, then this velocity $\approx 3 \times 10^9$ cm/sec. This is approximately the same
velocity as possessed by an electron of 300 ev energy.

The distribution of $X_0$ over the discharge could not be measured, but the experimental results suggest the following model. Below $\mu$ gas pressure the gas conduction is so low that the electromagnetic fields cannot produce appreciable currents in the gas. As the gas pressure and the transverse magnetic fields increase, electromagnetically derived currents increase and the plasma density and hence gas conduction increase.

The maximum amplitude of the electrostatic field between the ends of the R.F. coil is constant and so, as the gas conductivity changes, the electrostatic field must be redistributed. Large electrostatic fields are then expected to develop between the ends of the coil and the plasma, in particular between the plasma and the walls, that is, in the region which determines the plasma potential. An increase in the electron velocity due to the increasing electrostatic field follows, which means an increase in the plasma potential and then, an increase in the mean beam energy. The magnitude of $\bar{E}$ estimated in this manner from the above expression is of the same order of magnitude to that shown in diag. 59.

For the shielded discharge the plasma density is maintained by the electromagnetically induced electric field which is everywhere normal to the discharge axis and hence to the direction of extraction of the ions. One may
then expect plasma potentials of the order of 20 ev which are calculated from plane probe theory for electron temperatures of $10^5$ $^0K$.

The observed values of the beam energy spread, $\Delta E$, in diag. 69 are an order of magnitude greater than would result from thermal ion velocities in the plasma or from the effect of R.F. fields acting directly on the ions. The only measurements of $\bar{E}$ and $\Delta E$ available for comparison with the present results are those of Cook (1962) which were published after the present results were obtained. His discharge operating conditions were similar to those of this thesis except that the R.F. power input was much higher at 350 to 500 watts. Cook obtained values of $\bar{E}$ from 250 to 450 ev and of $\Delta E$ from 80 to 180 ev. The dependence of both $\bar{E}$ and $\Delta E$ on gas pressure is similar to that obtained in this thesis. Cook has proposed that the observed large values of $\Delta E$ may arise from a time variation of the plasma potential of the form $V_p(\tau) = V_o + \Delta V \cos(\omega \tau + \phi)$.

He derives an expression for $\Delta E = \frac{H e_\Delta V}{\omega \cdot d} \left(\frac{e}{2 m_i}\right)^{1/2}$, (7.2)

where $d = \text{space charge sheath width} = 2.2 \cdot 10^{-4} \left(\frac{V^{1/2}}{e}\right)^{1/2} \text{cm}$.

$V_o = \text{steady component of the plasma potential} = \bar{E}$

$\Delta V = \text{amplitude of the time varying component of } V_p$

$= \text{a maximum value of } V_p$

For his discharge, $\Delta E(\text{calculated}) \approx 65 \text{ ev which is}$
considerably lower than his experimental values. However, for the present discharge \( d \sim 0.4 \text{ cm}, \omega = 10^8 \text{ rad/sec}, \) \( V_p = 220 \text{ volts (maximum)} = \Delta V \) which gives a values of \( \Delta E \) of 32 ev. To this value of \( \Delta E \) should be added that value of \( \Delta E \) which is measured when the electrostatic shield surrounds the discharge, i.e. 15 ev, as it may be assumed that this value of \( \Delta E \) originates from within the plasma independently of any time varying plasma potential. The total value of \( \Delta E \) is now 47 ev. which is in good agreement, even though fortuitous, with the maximum experimental value shown in diag. 69.

In spite of this agreement the above expression for \( \Delta E \) appears to predict that \( \Delta E \) should increase as the plasma potential increases and this is contrary to the observations. Other measurements have also shown that \( \Delta E \) is independent of \( \omega \) and \( V_e \) and this is contrary to the predictions of the above expression for \( \Delta E \). An explanation of these anomalies is not obvious and is not attempted here because of the crudity of the above model and of the complex interdependencies of many of the discharge parameters. The basis has been established for more detailed work in this field.
Chapter B.8

H\textsuperscript{+}\textsubscript{2} Projectiles

8.1 Hydrogen Gas Target

8.1.1 The Ion Source Effect

Tables 8 and 9 show the dependence of the cross sections for the production of protons, negative hydrogen ions and neutral hydrogen atoms upon the ion source operating conditions of gas pressure, transverse magnetic field, R.F. oscillator input power and frequency and ion extraction voltage. H\textsuperscript{+}\textsubscript{2} energies of 6, 20 and 50 kev have been used. The parameters of \(\omega\), \(X_0\) and \(V_s\) were restricted to small ranges by the limitations imposed by the R.F. oscillator and a reasonable beam current; however within these ranges, \(\omega\), \(X_0\) and \(V_s\) had no marked (i.e. less than 5\%) effect upon the cross sections. At all projectile energies the cross sections were increased as either the gas pressure or the transverse magnetic field were decreased. At 6 Kev the maximum variation that could be found in \(\sigma_{H_2^+}\) was 30\% and this variation decreased to about 5\% at 50 Kev. The variations in \(\sigma_{H_2^+}\) were comparable with those for \(\sigma_{H^+}\) for all tests.

These experiments clearly demonstrate that the cross sections depend upon the ion source operating conditions and this is possible through their only common factor, the populations of the various electronic vibrational and rotational excited states of the projectile H\textsuperscript{+}\textsubscript{2} ion.
TABLE 8

Dependence of $H^+ \, _2$ dissociation cross sections on the R.F. ion source operating conditions.

All cross sections are given in units of $\text{cm}^2$/molecule.

$\sigma_{H^+_1}$ is calculated on the basis that $\sigma_{H^+_2}$ is independent of source operating conditions and $\sigma_{H^+_2} = 16.65 \times 10^{-16}$ $\text{cm}^2$/mol.

Table 8.1

Projectile $H^+_2$ Energy 6 Kev.
Source gas pressure = $20.10^{-3}$ mm.Hg.

8.11 Dependence of cross section on transverse magnetic field, $B$.

| $w$ = 22.5 Mc/sec. | $X_0$ = 160 watts. | $V_s$ = 3 kv. |

<table>
<thead>
<tr>
<th>B. gauss</th>
<th>$\sigma_{H^+_1} \cdot 10^{16}$</th>
<th>$\sigma_{H^-_1} \cdot 10^{19}$</th>
<th>$\sigma_{H^+_0} \cdot 10^{16}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>1.46</td>
<td>2.16</td>
<td>4.48</td>
</tr>
<tr>
<td>resonance $\sqrt{2}$</td>
<td>1.32</td>
<td>1.01</td>
<td>4.22</td>
</tr>
<tr>
<td>resonance</td>
<td>1.20</td>
<td>1.16</td>
<td>4.02</td>
</tr>
</tbody>
</table>

8.12 Dependence of cross section on $V_s$, the ion source extraction voltage.

| $w$ = 22.5 Mc/sec. | $X_0$ = 160 watts. | $B$ = resonant value. |

<table>
<thead>
<tr>
<th>$V_s$ volts</th>
<th>$\sigma_{H^+_1} \cdot 10^{16}$</th>
<th>$\sigma_{H^-_1} \cdot 10^{19}$</th>
<th>$\sigma_{H^+_0} \cdot 10^{16}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1000</td>
<td>1.22</td>
<td>0.33</td>
<td>4.22</td>
</tr>
<tr>
<td>2000</td>
<td>1.19</td>
<td>1.20</td>
<td>4.06</td>
</tr>
<tr>
<td>3000</td>
<td>1.23</td>
<td>0.98</td>
<td>4.10</td>
</tr>
</tbody>
</table>
8.13 Dependence of cross section on $X_0$, the oscillator input power.

$w = 22.5$ Mc/sec.
$B = $ resonant value.
$V_s = 3000$ volts.

<table>
<thead>
<tr>
<th>$X_0$ . watts</th>
<th>100</th>
<th>130</th>
<th>160</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{H_1}^+ . 10^{16}$</td>
<td>1.18</td>
<td>1.24</td>
<td>1.22</td>
</tr>
<tr>
<td>$\sigma_{H_1}^- . 10^{19}$</td>
<td>0.87</td>
<td>1.04</td>
<td>1.03</td>
</tr>
<tr>
<td>$\sigma_{H_1}^0 . 10^{16}$</td>
<td>4.14</td>
<td>4.20</td>
<td>4.08</td>
</tr>
</tbody>
</table>

8.14 Dependence of cross section on $w$, the R.F. excitation frequency.

$B = $ resonant value.
$V_s = 3000$ volts.
$X_0 = 160$ watts.

<table>
<thead>
<tr>
<th>$w$ . Mc/sec</th>
<th>22.5</th>
<th>17.1</th>
<th>14.6</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{H_1}^+ . 10^{16}$</td>
<td>1.24</td>
<td>1.30</td>
<td>1.28</td>
</tr>
<tr>
<td>$\sigma_{H_1}^- . 10^{19}$</td>
<td>0.67</td>
<td>1.09</td>
<td>0.96</td>
</tr>
<tr>
<td>$\sigma_{H_1}^0 . 10^{16}$</td>
<td>4.20</td>
<td>4.04</td>
<td>4.12</td>
</tr>
</tbody>
</table>

8.21 Dependence of cross section on transverse magnetic field.

Source gas pressure = $4.10^{-3}$ mm.Hg.

$w = 22.5$ Mc/sec.
$X_0 = 160$ watts.
$V_s = 3000$ volts.

<table>
<thead>
<tr>
<th>$B$ . gauss</th>
<th>0</th>
<th>resonance $\sqrt{2}$</th>
<th>resonance</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{H_1}^+ . 10^{16}$</td>
<td>1.88</td>
<td>1.68</td>
<td>1.40</td>
</tr>
<tr>
<td>$\sigma_{H_1}^- . 10^{19}$</td>
<td>1.92</td>
<td>1.56</td>
<td>1.53</td>
</tr>
<tr>
<td>$\sigma_{H_1}^0 . 10^{16}$</td>
<td>5.22</td>
<td>4.82</td>
<td>4.45</td>
</tr>
</tbody>
</table>
8.3 $\text{H}_2^+$ energy = 20 Kev

$w = 22.5$ Mc/sec.
$X_0 = 160$ watts.
$V_B = 3000$ volts

8.31 Source gas pressure = $20 \cdot 10^{-3}$ mm.Hg.

<table>
<thead>
<tr>
<th>$B$</th>
<th>$0$</th>
<th>resonance $\sqrt{2}$</th>
<th>resonance $\sqrt{2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{H_1}^+ \cdot 10^{16}$</td>
<td>2.13</td>
<td>2.03</td>
<td>1.92</td>
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<tr>
<td>$\sigma_{H_1}^- \cdot 10^{19}$</td>
<td>7.8</td>
<td>8.3</td>
<td>4.2</td>
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<tr>
<td>$\sigma_{H_1}^0 \cdot 10^{16}$</td>
<td>7.2</td>
<td>6.9</td>
<td>6.65</td>
</tr>
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</table>

8.32 Source gas pressure = $4 \cdot 10^{-3}$ mm.Hg.

<table>
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<tr>
<td>$\sigma_{H_1}^+ \cdot 10^{16}$</td>
<td>2.33</td>
<td>2.27</td>
<td>2.12</td>
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<tr>
<td>$\sigma_{H_1}^- \cdot 10^{19}$</td>
<td>9.7</td>
<td>9.4</td>
<td>6.9</td>
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<tr>
<td>$\sigma_{H_1}^0 \cdot 10^{16}$</td>
<td>8.4</td>
<td>8.0</td>
<td>7.6</td>
</tr>
</tbody>
</table>
TABLE 9

Dependence of cross sections upon transverse magnetic field for H$_2^+$ projectile energy of 50 kev.

$w = 22.5$ Mc/sec
$X_0 = 160$ watts.
$V_s = 3000$ volts.

9.11 Source gas pressure $= 20.10^{-3}$ mm.Hg.

<table>
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<td>$\sigma_{H_1^+} \cdot 10^{-16}$</td>
<td>2.08</td>
<td>2.08</td>
<td>2.06</td>
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<tr>
<td>$\sigma_{H_1^-} \cdot 10^{-19}$</td>
<td>8.4</td>
<td>9.0</td>
<td>7.6</td>
</tr>
<tr>
<td>$\sigma_{H_1^0} \cdot 10^{-16}$</td>
<td>6.1</td>
<td>6.1</td>
<td>6.0</td>
</tr>
</tbody>
</table>

9.12 Source gas pressure $= 4.10^{-3}$ mm.Hg.

<table>
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</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{H_1^+} \cdot 10^{-16}$</td>
<td>2.24</td>
<td>2.20</td>
<td>2.12</td>
</tr>
<tr>
<td>$\sigma_{H_1^-} \cdot 10^{-19}$</td>
<td>9.5</td>
<td>7.9</td>
<td>8.2</td>
</tr>
<tr>
<td>$\sigma_{H_1^0} \cdot 10^{-16}$</td>
<td>6.3</td>
<td>6.28</td>
<td>6.28</td>
</tr>
</tbody>
</table>
Consider now some possible origins of this "ion source effect".

Chapter 7 reported the results of investigation of the energy characteristics of the positive ion beam extracted from the R.F. source. It was established that both the mean beam energy, $\overline{E}$, and the beam energy spread, $\Delta E$, depended upon $B$ and $\rho$ and not upon $\omega$ and $V_s$. Further it was shown that $\overline{E}$ increases as both $B$ and $\rho$ decrease and $\Delta E$ decreases as both $B$ and $\rho$ decrease. Hence the measured cross sections increases occur when either $\overline{E}$ increases or $\Delta E$ decreases. Unfortunately at the time of measurement of $\overline{E}$ and $\Delta E$ it was not possible to readily perform the deceleration measurements on the separate charge to mass ratio components of the positive ion beam. Subsequent magnetic analysis of the beam revealed that $\frac{e}{m}$ components of 1, $1/2$ and $1/3$ were present in inconsistent amounts for identical settings of those ion source controls external to the source. The percentage of protons varied between 86\% and 46\% while $H_2^+$ and $H_3^+$ ions were invariably present in the ratio of 4/1. It is then not possible to specify the percentage content of $H_2^+$ in the positive ion beam extracted from the source. Using a simple plasma model Thoneman (1953) has shown that the expected mean beam energy, $\overline{E}$, is equal to the plasma potential which is proportional to the logarithm of the mass of the extracted ion. The manner in which the plasma potential varies for
a beam composed of comparable fractions of different ion masses is not known. Hence one is forced to assume that the measured mean beam energies are characteristic of the $H_2^+$ ions.

In chapter 7 only partially successful attempts were made to relate $\bar{E}$ and $\Delta E$ of the positive ions to $\overline{E}$ and $\overline{\Delta E}$ of the electrons within the ion source. However it is reasonable to expect that $\overline{E}$ of the positive ions varies in a similar manner as $\overline{E}$ for electrons. More successful attempts have been made to relate the electron energy to the population of the vibrationally excited states of $H_2^+$ formed by electron bombardment of gas molecules.

The gas temperature within the ion source $\sim 300^0 K$. Assuming a Boltzmann distribution of the excited states it is seen that less than 1% of the molecules are in states other than $v = 0$. It is a reasonable assumption that the population of the $v'$ level of $H_2^+$ formed by electron bombardment of $H_2$ ion $v = 0$ level is given by the square of the overlap of the appropriate vibrational wave functions

$$P_{v, v'} = \left| \int \psi_0(R) \psi_{v'}(R) dR \right|^2$$

where $\psi_0(R)$ is the vibrational wave function for the ground state molecule

$\psi_{v'}(R)$ is the vibrational wave function for the $H_2^+$ ion.

Harmonic oscillator functions of the form $\psi_v(R) = R_n(\sqrt{\beta} R) \psi_{v}^{\sqrt{\beta}}(\frac{\beta R^2}{2})$ to fit a parabolic potential for $H_2^0$ and $H_2^+$ have been used
<table>
<thead>
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<th>( P_{ov} )</th>
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<tbody>
<tr>
<td>0</td>
<td>4.3</td>
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<tr>
<td>1</td>
<td>10.1</td>
</tr>
<tr>
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<td>14.7</td>
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<td>4</td>
<td>13.5</td>
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<td>8</td>
<td>4.5</td>
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<td>12</td>
<td>0.95</td>
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<td>16</td>
<td>0.2</td>
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<tr>
<td>17</td>
<td>0.1</td>
</tr>
<tr>
<td>18</td>
<td>0.02</td>
</tr>
</tbody>
</table>
Relative populations of the various vibrational levels of the $^2\Sigma^+$ state of the $H_2^+$ ion for various energies of the ionizing electrons. The curve for each electron energy shown is normalized to one for the most populous level.
as given by Herzberg. $H_n(\sqrt{E/R})$ is the Hermite polynomial of the $n^{th}$ degree. The harmonic oscillator functions and the parabolic potential are good approximations to the $H_2^+$ potential as given by Herzberg for the lowest vibrational levels, but are probably quite erroneous for the upper vibrational levels. However they lend themselves to simple calculations. The percentage populations of several vibrational levels of $H_2^+$ is shown in Table 10.

These calculations were not pursued as more accurate estimates were published by McGowan (1964). He used more accurate wave functions (Cohen 1959) for $H_2^+$ and made allowance for the ionizing electron energies. His results are shown in diag. 70. For electron energies greater than 24 ev, McGowan's results do not differ greatly from those calculated using the much simpler model. Diag. 70. predicts that the upper vibrational levels of $H_2^+$ are increasingly populated as the electron energy rises from threshold, about 16 ev, to 24 ev after which the relative populations of the vibrational levels are not sensitive to electron energy.

From the above discussion there can be seen a similar behaviour pattern relating an increasing population of the $H_2^+$ uppermost vibrational levels to an increasing electron energy, which, in turn, increases the mean energy of the $H_2^+$ projectile ions. The increases in these quantities are determined by decreases of the ion source parameters of
gas pressure and transverse magnetic field. Through this sequence of events an increasing dissociation cross section is related to an increase in the upper vibrational levels of the $H_2^+$ molecular ion.

The inadequacies of the ion source prevent more definite statements. The $H_2^+$ beam extracted from the source will contain a relative population of vibrationally excited states determined, not from a delta function of electron energies, but by integration over that part of the electron energy distribution between 16 and 24 ev. The present measurements of cross section are then further integrated by measuring the sum of the cross section for each vibrational state multiplied by the relative populations of those states. Furthermore the source operates in a gas pressure region of several microns where deactivation of the vibrational levels by collision with the gas molecules is not negligible.

8.12 The Collision

It was shown in the previous section that those source parameters which produced a maximum variation of 30% in $\sigma_{H_2^+}$ at a projectile energy of 6 kev were also responsible for the maximum variation in $\sigma_{H_1^+}$ of 5% which could be obtained at 50 kev. Diag. 71 shows the cross section $\sigma_{H_1^+}$ as a function of $H_2^+$ energy for these source parameters.

The measured total $H_1^+$ production cross section is the
sum \( (\sigma_1 + 2\sigma_2) \). The various ways in which protons may be produced from \( H_2^+ \) is seen from diag. 72 which shows the potential energy curves for known \( H_2 \) electronic states. The electron loss dissociation occurs through a transition to the double proton state of which the two protons would separate with about 18 ev centre of mass energy. One would expect that this mechanism may not be influenced to a large degree by the degree of vibrational excitation of the \( H_2^+ \) ion since the energy difference between the \( v = 0 \) and \( v = 18 \) levels of 2.4 ev is not large compared with the 36 ev energy required for a transition to the double proton state.

The collision induced dissociation with the production of \( H_1^+ \) and \( H_1^0 \) may proceed in several ways.

(a) Excitation of the lowest \( ^2\Xi \) state of \( H_2^+ \) from the ground \( ^2\Xi_g \) state. This leads to dissociation since the \( ^2\Xi \) state is repulsive. The resulting proton will have about 5 ev centre of mass kinetic energy.

(b) A quasi-elastic collision of the target particle with either an H atom or \( H^+ \) ion of the \( H_2^+ \) molecule in which sufficient energy is given to the atom or ion to break the molecular bond. This may be a vertical, or if Frank-Condon condition do not apply, diagonal transition to the top of the \( ^2\Xi_g \) potential well or to the continuum above it. The resulting proton will have low energy.

(c) A vertical (Frank-Condon) transition from the bound \( ^2\Xi_g \) state to an electronic state higher than the \( ^2\Xi \).
The known electronically bound states (the $2p\eta_u$ and $3d\sigma_g$ are very weakly bound with 0.25 ev and 1.35 ev respectively) have minima greatly displaced from the minima of the $H_2$ molecule and $H_2^+$ ion ground states. Application of the Frank Condon principle indicates that electronic excitation of $H_2^+$ would then lead directly to dissociation. All other electronically excited states are repulsive. The protons from this type of dissociation will have at least 10 ev centre of mass kinetic energy.

The data contained in this latter paragraph indicates the fact, not previously mentioned, that the projectile ion $H_2^+$ is most unlikely to have emerged from the ion source in either of its two bound electronically excited states. Hence any dependence of the cross section on the state of excitation of $H_2^+$ must occur through the vibrational and rotational excited states of $H_2^+$.

From the potential energy diagram it is seen that the energy separation between the $2\Xi_g$ and $2\Xi_u$ states is a rapidly varying function of the nuclear separation. In highly excited vibrational states the two protons of the molecular ion spend a relatively large portion of a vibration period at their widest separation and in this region the electron energy difference between the $2\Xi_g$ and $2\Xi_u$ states is very small. It may therefore be reasonably expected that the transition probability to the repulsive $2\Xi_u$ state is greatly increased by an increased
DIAG. 73. ELECTRON PROB. DENSITY DISTRIBUTION

FOR $^2\Sigma_g^+$ AND $^2\Sigma_u^+$ STATES OF $H_2$
population of the upper vibrational levels of the state.

It is instructive to look at the electron probability density distribution for the \( ^2\Sigma_g \) and \( ^2\Sigma_u \) states as shown in diag. 73. The bonding nature of the \( ^2\Sigma_g \) orbital is due to the absence of a nodal surface in the eigenfunction in the region between the nuclei and this implies a high electron density and therefore a contribution to the attraction of the nuclei. The \( ^2\Sigma_u \) eigenfunction has a nodal surface and hence low electron density between the nuclei with a subsequent antibonding orbital. As the vibrational excitation of the nuclei increases, the protons move further apart and the electron probability density distributions of the two states approach a similar limit - that of the separated proton and atom. The dissociation processes (a) and (b) above are then somewhat similar as the nuclear separation becomes large.

8.13 Comparison of the Present Results with those of Other Workers.

The measurements by Guidini (1961) of the partial proton production cross sections, \( \sigma_1 (H_1^+ + H_0^0) \) and \( \sigma_2 (2H_1^+) \), permits an explanation of the shape of the total proton production cross section, \( \sigma_{H_1^+} \) \( (=\sigma_1 + 2\sigma_2) \) versus energy curve in diag. 71. For \( H_2^+ \) energies less than 15 Kev, \( \sigma_2 \) constitutes less than 5% of \( \sigma_{H_1^+} \). Hence the 30% variation in \( \sigma_{H_1^+} \), that occurs when the source gas pressure
is changed from 4 to $20.10^{-3}$ mm.Hg., is almost entirely a 30% change in $\sigma_i$. That is to say the measured cross section $\sigma_{H^+}$ is the cross section for the "collision induced" dissociation of the fast $H_2^+$ ion into $(H_1^+ + H_1^0)$ without the capture or loss of an electron.

For higher $H_2^+$ energies, $\sigma_i$ gradually increases in value relative to $\sigma_i$ until at 40 Kev, $\sigma_i$ is equal to $2\sigma_2$. Then the observed 5% variation in $\sigma_{H^+}$, with change of source gas pressure, may have originated from either $\sigma_i$ or $\sigma_2$.

The result that the "ion source effect" variation of $\sigma_{H^+}$ decreases as the $H_2^+$ projectile energy increases permits a correlation to be made of the somewhat contradictory results of several workers. Firstly Barnett and Ray's failure to find any dependence of $\sigma_{H^+}$ on the R.F. ion source parameters of gas pressure and R.F. input power for $H_2^+$ energies from 25 to 250 Kev is not inconsistent with the present results since the accuracy of their measurements was $\pm 10\%$ in the region where the present results show the variation of $\sigma_{H^+}$ is only $5 \pm 3\%$.

Sweetman's result, that $\sigma_{H^+}$ was increased by at most $7 \pm 4\%$ for an increase in the relative populations of the $v = 17$ and 18 levels from 0.13 to 0.3% of the total beam population in the energy region from 280 to 670 Kev, is readily explained. Diag. 70 indicates that such a small change in the relative populations of the $v = 17$ and 18
levels does not indicate a drastic change in the relative populations of the lower vibrational levels. In fact Kerwin's results have shown that such a change in the $v = 17$ and 18 level populations would produce a change of less than 10% in the cross section $\sigma_{H_i^+}$ at an energy as low as 2 Kev. With a simple physical model it is seen that for energies of several hundred Kev, an $H_2^+$ ion takes about $2 \times 10^{-16}$ sec. to traverse the 8°A "effective collision distance" (Hasted, 1952) and this time is less than $10^{-2}$ of a nuclear vibration period. Although the nuclei tend to spend more time near the ends of their vibrational paths the probability of the target particle seeing the projectile nuclei at their widest separation should decrease as the collision transit time becomes progressively smaller than the nuclear vibration period. One may then expect the increased populations of the upper vibrational levels to cause larger increases in the $H_2^+$ dissociation cross sections as the $H_2^+$ velocity decreases and the effective collision time becomes comparable to the nuclear vibrational period. However for $H_2^+$ energies of 2 Kev the collision transit time is still small of the order of $2 \times 10^{-15}$ sec. Thus the percentage changes found by Kerwin and Sweetman are not incompatible with one another and with the present results.
8.2 Inert Gas Targets.

8.21 The Ion Source Effect.

The total proton production cross section, \( \sigma_{H^+} \), is measured for 6 Kev \( H_2^+ \) ions incident upon helium for \( H_2^+ \) ions extracted from the R.F. ion source in which gas pressures of 4 and \( 20 \times 10^{-3} \) mm.Hg. are used. All other ion source parameters are fixed at those values used in Chapter 8.1 to produce the largest \( \sigma_{H^+} \) in hydrogen. For \( 4 \mu \text{Hg} \) source pressure \( \sigma_{H^+} \) is \( 1.05 \times 10^{-15} \) cm\(^2\)/atom and for \( 20 \mu \text{Hg} \) source pressure \( \sigma_{H^+} \) is \( 0.72 \times 10^{-16} \) cm\(^2\)/atom, that is a decrease of 35%. This "ion source variation" of 35% for \( \sigma_{H^+} \) in helium is approximately the same as that produced in hydrogen of 30%. It appears that the "ion source effect" may then be independent of the target gas and is truly only of an ion source origin. The dependence of \( \sigma_{H^+} \) on ion source parameters other than gas pressure has not been investigated as the nature of the ion source effect has been well demonstrated.

8.22 Further Measurements of \( \sigma_{H^+} \) in the Inert Gases.

Previous chapters have indicated that the present apparatus is limited in its applications by the production of a molecular ion beam whose population is spread over all the vibrational levels and by the measurement of a total charge production cross section averaged over the ion beam population. In spite of such limitations further cross section measurements were made for the following
PROJECTILE \( \text{H}_2^+ \)

TARGET \( \text{He} \)

CROSS SECTION Total proton production

---

**Diagram 74.**

**Cross Section - \( \text{cm}^2/\text{atom} \)**

**Projectile Energy - Kev.**

- **FEDORENKO 1959**
- **GUIDINI 1961**
- **GUIDINI - 1961**
- **PRESENT DATA**
reasons.

(1) Only Fedorenko (1954, 1959) and Guidini (1961) have measured $\sigma_{H^+}$ over portions of the energy range 5 - 50 Kev for He, Ne, Ar and nitrogen. Their values are only in fair agreement. No measurements of $\sigma_{H^+}$ have been reported for Kr and Xe. The present apparatus has been shown to collect all the wide angle scattering collision products and to be capable of a high accuracy of 3%. The opportunity was available to use the present apparatus for a limited time; hence it was decided to obtain a unified set of measurements of $\sigma_{H^+}$ in nitrogen and the inert gases.

(2) By operating the ion source in the low pressure region the $H_2^+$ ion beam will have a relative population of the vibrational levels in the "saturation" region (see diag. 70) such as that predicted from electron bombardment ion sources with electron energies greater than 24 ev. The measured cross sections are then expected to be the maximum cross sections obtainable from $H_2^+$ ions extracted from present day ion sources for all operating conditions.

8.23 Helium.

Diag. 74 shows $\sigma_{H^+}$ as a function of $H_2^+$ energies from 2 to 50 Kev for $H_2^+$ ions incident upon helium gas. There is good support for the shape of the cross section versus energy curve as obtained by Fedorenko (1959). Each cross section value of diag. 74 is the average of two separate
PROJECTILE $H_2^+$
TARGET Ne
CROSS SECTION Total proton production

Graph showing the cross section vs. projectile energy for $H_2^+$ incident on Ne target. The graph includes data from Fedorenko 1954 and present data, plotted on a logarithmic scale from $10^{-17}$ to $10^{-15}$ cm$^2$/atom against projectile energy from 1 to 100 keV.
PROJECTILE $H^+_2$
TARGET Ar
CROSS SECTION Total proton production

![Graph showing cross-section as a function of projectile energy. The graph includes data from FEDORENKO 1954, FEDORENKO 1959, GUIDINI 1961, and SWEETMAN 1960. The present data is also shown.](image)
PROJECTILE $- \text{H}_2^+$

TARGET $- \text{Kr, Xe}$

CROSS SECTION $-$ Total $\text{H}_1^+$

Diagram 77

-agram of projectile energy vs. cross section for different targets.

Axes:
- Cross section in $\text{cm}^2/\text{atom}$
- Projectile energy in keV

Legend:
- Kr
- Xe

Graph shows a trend where cross section increases with projectile energy for both Kr and Xe targets.
determinations except those values at 30, 40 and 50 Kev which are the average of four separate determinations. The first cross section maximum at about 28 Kev and the suggestion of a further maximum value above 50 Kev are supported by the present data. The partial cross section measurements of Guidini (1961) suggest that the maximum in $\sigma_{H^+}$ at 28 Kev is predominantly a maximum in $\sigma_1$, i.e. from a collision induced dissociation without an electron capture or loss.

8. Ne, Ar, Kr and Xe.

Diagrams 75, 76 and 77 show $\sigma_{H^+}$ as a function of energy for $H^+$ ions incident upon Ne, Ar, Kr and Xe. The double maxima in the cross section curves in H$_2$ and He gases are not present in the other inert gases. The partial cross section measurements of $\sigma_1$ and $\sigma_2$ by Guidini (1961) in argon suggest that the absence of the dual maxima is due entirely to the much larger values of $\sigma_1$ compared to $\sigma_2$ over the energy range 2 to 50 Kev.

The present values of $\sigma_{H^+}$ agree within several percent with the values of Sweetman (1960) at 50 Kev. However as the $H^+$ projectile energy decreases below 20 Kev the present values of $\sigma_{H^+}$ become progressively larger than Fedorenko's values (1954 and 1959). This discrepancy is thought to originate from the present efficient collection of all the large angle scattered fast collision products. For this same reason the measurement of $\sigma_{H^+}$ in nitrogen is
DIAG. 78

PROJECTILE: $H^+_2$
TARGET: $N_2$

CROSS SECTION: Total proton production

PROJECTILE ENERGY — KEV.

CROSS SECTION — CM$^2$/ATOM

GUIDINI 1961
SWEETMAN 1960
FEDORENKO 1954
PRESENT DATA
interesting. Diag. 78 shows that there are large differences between the results of all workers. From this discrepancy between the present values and those of Fedorenko (1954) which has occurred in H₂, Ne, Ar and N₂, it would appear that Fedorenko has failed in his 1954 measurements to collect those fast collision products which have been scattered through large angles.

8.25 Discussion

It has been shown, by the use of the partial cross section measurements of Guidini, that the measured cross section \( \sigma_{H_2^+} \), for \( H_2^+ \) energies less than about 20 Kev, is predominantly that for the simple "collision induced" dissociation of the \( H_2^+ \) ion into a proton and a hydrogen atom. At any given energy \( \sigma_{H_2^+} \) increases with the size of the target atom.

Unsuccessful attempts have been made to explain the above cross section measurements for primary \( H_2^+ \) ions by the use of the adiabatic hypothesis (Chapter 1.3). The velocities at which the cross section maxima occur are not consistently explained by the adiabatic and the rate of rise of the cross section at low velocities does not adhere to a consistent exponential form for the various target gases. Dunn et al. (1962) have also attempted to explain their Lyman \( \alpha \) radiation production cross sections for \( H_2^+ \) ions incident upon hydrogen and helium by the use of the adiabatic hypothesis. They found values of the interaction
DIAG. 79. PROJECTILE $H_2^+$
TARGET $H_2^0$
CROSS SECTION — Total $H_1^-$ production

[Graph showing cross section versus $H_2^+$ projectile energy with ion source pressure levels 4 μ and 20 μ indicated.]
distance, a, which varied from 1.4 to 7.5 Å; there being considerable discrepancy between the values of "a" found from the adiabatic hypothesis and from those cross section values which had been fitted to an experimental rate of rise. The above inconsistencies are not surprising in view of the expected complications from the various excited states, and their variable populations, of the molecular hydrogen ion and also of the target atoms.

8.26 The Total $H_1^-$ Production Cross Section, $\sigma_{H_1^-}$.

Diag. 79 shows the total $H_1^-$ production cross section, $\sigma_{H_1^-}$, for $H_2^+$ ions incident upon hydrogen gas for $H_2^+$ ion source gas pressures of 4 and 20 microns Hg. The dependence of $\sigma_{H_1^-}$ on the source pressure over the energy range 2 - 50 KeV and the explanation of this dependence is readily seen to be similar to that for $\sigma_{H_1^+}$ which has been discussed at length earlier in this chapter. However that dependence is partly obscured by the scatter of the experimental points which is inexplicably large for the negative hydrogen ions. Whilst each cross section measurement was considered accurate to within 3%, the same cross section showed variations of up to 20% when measured at different times over a period of six months. This behaviour was not observed for the proton production cross sections which have always been measured simultaneously with the negative hydrogen ion production cross sections.

Diag. 80 shows $\sigma_{H_1^-}$ for the inert gases and hydrogen
PROJECTILE — $\text{H}_2^+$

TARGET — $\text{H}_2$, He, Ne, Ar, Kr, Xe.

CROSS SECTION — $\text{H}_1^-$ Production (total).

DIAG. 80. $\text{H}_2^+$ PROJECTILE ENERGY — KEV.
for $\text{H}_2^+$ energies from 2 to 50 Kev for $\text{H}_2^+$ ions extracted from the ion source at a pressure of 4 $\mu\text{Hg}$. The only published measurement of $\sigma_{\text{Hi}}^-$ is that of Fedorenko (1959) who finds a value of $1.2 \times 10^{-18}$ cm$^2$/atom for $\sigma_{\text{Hi}}^-$ for 12 Kev $\text{H}_2^+$ ions incident upon argon. This value is more than twice the present value. There is a general similarity between the shapes of the $\sigma_{\text{Hi}}^-$ curves of diag. 80 and those curves of diagrams 24 to 28 for the double electron capture by protons in the inert gases. In particular the cross section maxima for each of those cross sections in $\text{H}_2$, Ne, Ar and Kr occur at the same projectile velocities. This result is not surprising since $\sigma_{\text{II}}^-$ is the cross section for a simple double electron capture while $\sigma_{\text{Hi}}^-$ is the cross section for, basically, a single or a double electron capture and subsequent dissociation.
GROWTH OF $H_2^+$, $H_1^+$ and $H_1^-$ FROM 10 KEV $H_3^+$ IN HYDROGEN.
Chapter 9.

The Triatomic Hydrogen Molecular Ion, $H_3^+$.

9.1 Hydrogen Gas Target.

This investigation of the charge changing and dissociation cross sections of $H_3^+$ ions is basically a repetition of the investigation of Chapter 8 for $H_2^+$ ions. The effect of those ion source parameters, which caused the greatest variation in the $H_2^+$ dissociation cross sections, is investigated for $H_3^+$ ions. The cross sections for total $H_2^+$ production, $\sigma_{H_2^+}$; total proton production, $\sigma_{H^+}$, and total $H_1^-$ production, $\sigma_{H_1^-}$, are measured for 2 to 50 Kev $H_3^+$ ions incident upon $H_2^+$, He, Ne and Ar.

Diag. 81 shows a typical growth of fast collision products $H_2^+$, $H_1^+$ and $H_1^-$ with target gas number density, $n_l$, for 10 Kev $H_3^+$ in hydrogen gas. The slopes of the $H_2^+$ and $H_1^+$ growth curves are determined within $2\%$. There is a much greater spread of experimental points in the $H_1^-$ growth relationship and there is up to $5\%$ error in fitting a straight line to the data. As there was only one Faraday cup for the collection of positively charged ions, the following two methods have been used to determine the growth of $H_2^+$ and $H_1^+$ with $(n_l)$. (a) The $H_2^+$ and $H_1^+$ beams are alternately switched into the Faraday cup for each setting of $(n_l)$. (b) Only one of the positively charged beams is collected while $(n_l)$ is varied over its normal range. The
equivalence of these methods is shown indirectly by also determining the growth, with \( n1 \), of the response of the secondary electron emitting detector to the uncharged dissociation products during each of the above methods. The primary \( H_3^+ \) beam is measured in the same Faraday cup both before and after a cross section run, but during a run it is monitored by the current collected by the aperture \( A_3 \) (see diag. 13) prior to the collision cell. The \( H_3^+ \) beam was constant in position, energy and current amplitude to better than 0.5\% over a one hour period.

Table 11 shows the dependence of \( \sigma_{H_3^+} \), \( \sigma_{H_2^+} \) and \( \sigma_{H^-} \) at primary \( H_3^+ \) energies of 20, 12 and 5 Kev, upon the \( H_3^+ \) ion source gas pressure and transverse magnetic field. By comparing these results with those for the similar study of the \( H_2^+ \) dissociation cross sections shown in Tables 8 and 9 (Chapter 8.11), it is readily seen that a given variation of one of the ion source parameters causes similar changes to occur in both the \( H_2^+ \) and \( H_3^+ \) primary ion dissociation cross sections. An explanation of this phenomenon may be readily given in terms similar to those used previously in Chapter 8.

Diagrams 82 and 83 show the dependence of \( \sigma_{H_2^+} \) and \( \sigma_{H_3^+} \), respectively, upon \( H_3^+ \) energy in the range 2 to 50 Kev for primary \( H_3^+ \) ions incident upon hydrogen gas and for those ion source gas pressures of 4 and 20 \( \mu \)Hg, which have caused the largest variations in the dissociation cross sections.
**TABLE 11**

Dependence of $H_3^+$ dissociation cross sections on the R.F. ion source operating conditions.

All cross section values are in units of cm$^2$/molecule.

Fixed ion source operating conditions are:

- $X_0 = 160$ watts.
- $V_S = 3$ Kv.
- $w = 16.7$ Mc/sec (Transverse magnetic field $\sim 6$ gauss for electron cyclotron resonance effect).

Table 11.1 Source gas pressure = $4 \times 10^{-3}$ mm.Hg.

<table>
<thead>
<tr>
<th>$H_3^+$ Energy Kev</th>
<th>20</th>
<th>12</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>$B$ gauss</td>
<td>0</td>
<td>4.2</td>
<td>6</td>
</tr>
<tr>
<td>$\sigma_{H_2^+} \cdot 10^{17}$</td>
<td>15.0</td>
<td>14.1</td>
<td>13.5</td>
</tr>
<tr>
<td>$\sigma_{H_1^+} \cdot 10^{17}$</td>
<td>13.8</td>
<td>12.8</td>
<td>12.0</td>
</tr>
<tr>
<td>$\sigma_{H^-} \cdot 10^{17}$</td>
<td>13.0</td>
<td>8.0</td>
<td>9.8</td>
</tr>
</tbody>
</table>

Table 11.2 Source gas pressure = $20 \times 10^{-3}$ mm.Hg.

<table>
<thead>
<tr>
<th>$H_3^+$ Energy Kev</th>
<th>20</th>
<th>12</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>$B$ gauss</td>
<td>0</td>
<td>4.2</td>
<td>6</td>
</tr>
<tr>
<td>$\sigma_{H_2^+} \cdot 10^{17}$</td>
<td>13.1</td>
<td>12.5</td>
<td>12.7</td>
</tr>
<tr>
<td>$\sigma_{H_1^+} \cdot 10^{17}$</td>
<td>11.8</td>
<td>11.0</td>
<td>10.4</td>
</tr>
<tr>
<td>$\sigma_{H^-} \cdot 10^{17}$</td>
<td>7.4</td>
<td>11.5</td>
<td>9.5</td>
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</table>

<table>
<thead>
<tr>
<th>$H_3^+$ Energy Kev</th>
<th>20</th>
<th>12</th>
<th>5</th>
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<tbody>
<tr>
<td>$B$ gauss</td>
<td>0</td>
<td>4.2</td>
<td>6</td>
</tr>
<tr>
<td>$\sigma_{H_2^+} \cdot 10^{17}$</td>
<td>10.0</td>
<td>9.5</td>
<td>9.3</td>
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<tr>
<td>$\sigma_{H_1^+} \cdot 10^{17}$</td>
<td>7.5</td>
<td>7.1</td>
<td>6.8</td>
</tr>
<tr>
<td>$\sigma_{H^-} \cdot 10^{17}$</td>
<td>6.8</td>
<td>5.5</td>
<td>4.5</td>
</tr>
<tr>
<td>$\sigma_{H^-} \cdot 10^{17}$</td>
<td>3.1</td>
<td>2.0</td>
<td>2.35</td>
</tr>
</tbody>
</table>
DIAG. 84. TARGET He
DIAG. 86. TARGET ARGON

PROJECTILE ENERGY — KEV.

H⁺³ PROJECTILE ENERGY — CM²/ATOM

10⁻¹⁶

10⁻¹⁷

1 10 100

σ⁺⁻⁻ FEDORENKO 1954

σ⁺⁻⁻ PRESENT

σ⁺⁻⁻ FEDORENKO 1954

σ⁺⁻⁻ PRESENT
TARGET $H_2^0$

CROSS SECTION—Total $H_1^-$ production

$$\text{CROSS SECTION} - \text{CM}^2/\text{MOL.}$$

$P = 4 \mu$

$P = 20 \mu$

$H_3^+$ PROJECTILE ENERGY — KEV.
PROJECTILE — $H_3^+$
TARGET — $H_2$, $He$, $Ne$, $Ar$
CROSS SECTION — $H_1^-$ Production (total).

Graph showing cross section vs. $H_3^+$ projectile energy (keV) for different targets.
9.2 Inert Gas Targets.

Diagrams 84, 85 and 86 show $\sigma_{H_3^+}$ and $\sigma_{H_3^+}$ for (i) primary $H_3^+$ ions incident upon He, Ne and argon respectively (ii) an $H_3^+$ energy range of 2 to 50 Kev (iii) $H_3^+$ ions extracted from the electrodeless discharge ion source whose operating conditions are such as to cause the largest values of $\sigma_{H_2^+}$ and $\sigma_{H_3^+}$ to occur as was determined from Table 11. The published results of McClure (1963) and Fedorenko (1954) are compared with the present data.

9.3 The Total $H_3^-$ Production Cross Section, $\sigma_{H_3^-}$

Diag. 87 shows the dependence of $\sigma_{H_3^-}$ upon the ion source gas pressure and projectile energy for $H_3^+$ projectiles incident upon hydrogen gas. The large scatter of the experimental points, which was observed for $\sigma_{H_3^+}$ from $H_2^+$ ions, is again evident. However, for $H_3^+$ energies less than 10 Kev, the higher pressure data is clearly becoming smaller than the low pressure data and hence, in accordance the results of Table 11 and the discussion of Chapter 8, the change in populations of the vibrationally excited states of the $H_3^+$ ion has a definite influence on $\sigma_{H_3^-}$.

Diag. 88 shows $\sigma_{H_3^-}$ as a function of energy for $H_3^+$ primary ions incident upon He, Ne and Ar for those ion source conditions used in section 9.2 above. The scatter of the experimental points is inexplicably large, particularly in Ne.
9.4 Discussion.

The inefficiency of Fedorenko's 1954 apparatus in collecting those fast collision products, which are scattered through wide angles, is again evident particularly at low energies. This feature of his 1954 work was apparent for those dissociation cross sections from \( \text{H}_2^+ \) ions in Chapter 8.

Riviere and Sweetman (1961) have shown that the \( \text{H}_3^+ \) ion is more readily dissociated by electric fields into \( \text{H}_1^+ + \text{H}_2^0 \) than into \( \text{H}_2^+ + \text{H}_1^0 \) as the latter process requires an additional 1.8 ev. Therefore if \( \text{H}_3^+ \) were to dissociate in a collision in the manner indicated for \( \text{H}_2^+ \) ions at low velocities, i.e. by a "collision induced" dissociation without an electron capture or loss, one may expect \( \sigma_{\text{H}_1^+} \) to be larger than \( \sigma_{\text{H}_2^+} \). However in \( \text{H}_2^+ \), \( \sigma_{\text{H}_2^+} \) is larger than \( \sigma_{\text{H}_1^+} \) and in the inert gases, He, Ne and Ar, \( \sigma_{\text{H}_1^+} \) is only slightly larger than \( \sigma_{\text{H}_2^+} \). An explanation of this behaviour may then be as follows. The velocity dependence of the electron loss cross sections considered in this thesis for \( \text{H}_1^0 \), \( \text{H}_1^- \) and \( \text{H}_2^+ \) ions all have small values at velocities less than \( 1.10^8 \) cm/sec. In contrast the electron capture cross sections, particularly in argon, generally have large values in that velocity region. One may then expect the capture process to contribute significantly to \( \sigma_{\text{H}_1^+} \) and/or \( \sigma_{\text{H}_2^+} \). These capture processes are

\[
\text{H}_3^+ + e \rightarrow \text{H}_3^0 \quad \text{(unstable)} \rightarrow \text{H}_2^0 + \text{H}_1^0 \rightarrow \text{H}_1^+ + \text{H}_1^-.
\]
The dissociation mode to \( H_2^+ + H^- \) cannot be large as diag. 88 shows the total \( H^- \) production cross section is at least an order of magnitude smaller than the observed \( \sigma_{H_1^+} \) and \( \sigma_{H_2^+} \). Hence the electron capture process is not expected to make any significant contribution to \( \sigma_{H_1^+} \) and \( \sigma_{H_2^+} \) at low velocities.

The electron loss process is

\[
\text{H}_3^+ - e \rightarrow \text{H}_3^{++} \text{ (unstable)} \rightarrow \text{H}_1^+ + \text{H}_2^+ \rightarrow \text{H}_1^+ + \text{H}_1^0
\]

The production of \( H_2^+ \) from the loss process thus includes an equal number of \( H_1^+ \). These simple considerations have failed to account for the result that \( \sigma_{H_1^+} \) is greater than \( \sigma_{H_2^+} \) in hydrogen and is larger than expected in the other target gases. Further discussion is not made since (a) little is known about the structure of the \( \text{H}_3^+ \) molecular ion and its possible excited states and (b) the possible large number of excited states for the \( \text{H}_3^+ \) ion, its dissociation products and those of the target atoms probably have a complex combination to form the presently observed total charge production cross sections.

Since there is generally some difficulty in ascertaining with any certainty many of the cross section values which appear in the literature, the present values for the dissociation cross sections of the \( \text{H}_3^+ \) ion are tabulated in Table 14 of Appendix 10.7.
Chapter 10

Appendices

10.1 Secondary Electron Emission from Metal Surfaces

Bombarded by Fast Hydrogen Atoms $H_1^0$ and Ions, $H_1^+, H_2^+$ and $H_3^+$.

The present study of secondary electron emission from Al, Ni and Cu-Be surfaces arose from the need of a simple neutral atom detector to measure incident atom fluxes in the range $10^4$ to $10^{12}$ atoms/sec. Throughout the course of the charge changing cross section measurements the atomic and molecular ions, $H_1^+, H_2^+$ and $H_3^+$ and the neutral atoms $H_1^0$ were incident upon the prepared emitting surface of the secondary electron detector. In this way a knowledge of the secondary electron yields for hydrogen atoms and ions was obtained.

10.11 Introduction.

When a beam of ions is incident upon a metal surface it may give rise to a number of phenomena. These include (a) emission of secondary electrons, (b) sputtering, (c) reflection and scattering of the ion beam, (d) emission of neutral atoms or negative ions. In the present work the effects (b), (c) and (d) are assumed to be at least two orders of magnitude smaller than effect (a) in accordance with the findings of Fogel, Slabospitskii and Rastrepin (1960). The secondary electron emission coefficient $\gamma$ is defined as the number of ejected electrons per incident ion or
atom. In the energy range 10 to 130 KeV the work of Allen (1939) indicated that the surface conditions of the target influenced $\gamma$ considerably. Since then the experiments of Murdoch (1955), Bourne (1955), Telkovskii (1956) and Large (1962) have confirmed these observations. Large (1962) has shown the cleaning processes of polishing, chemical etching, vacuum baking at 450°C and "flashing" the target at a temperature just below the melting point may, in turn, successively reduce $\gamma$ by as much as a factor of five. The latter process is claimed to produce, at a pressure of better than $5 \times 10^{-8}$ mm.Hg, a surface which has no more than one monolayer of absorbed gas. For such a surface Large finds (i) that for $H_1^+$, $H_2^+$ and $H_3^+$ increases by only about 50% as the atomic number of the target increases from 20 to 80, and (ii) that the average ratio of the yields for $H_1^+$, $H_2^+$ and $H_3^+$, namely $\gamma_{H_2^+)/\gamma_{H_1^+}$ and $\gamma_{H_3^+)/\gamma_{H_1^+}$, decrease from 2.77 and 2.07, at ion velocities of $1.5 \times 10^8$ cm/sec to 2.60 and 1.87, respectively, at ion velocities of $3.5 \times 10^8$ cm/sec. The result (ii) is independent, within 10%, of the atomic number of the target. Results similar to (i) and (ii) are reported by Telkovskii (1956) for atomically clean surfaces.

Contrary to the above results, both Chambers (1964) and Schwirzke (1960) have found for Cu-Be surfaces, cleansed only by polishing, that $\gamma_{H_3^+)/\gamma_{H_1^+} \approx 2.09$ and
ALUMINIUM FOIL

COPPER BLOCK

DIAG.89 SECONDARY ELECTRON EMITTING DETECTOR.
\[ \frac{N_{H_2}}{N_{H_1}} \simeq 1.46 \text{ within } 10\% \] and that these ratios are independent of ion velocities in the range 0.5 to 2.0 \times 10^8 \text{ cm/sec}. The above two groups of experimental conditions appear to differ only in the nature of the emitting surface. The present investigation has used surface conditions similar to that of Schwirzke to provide additional data on the above anomaly. The present investigation uses polished Cu-Be and clean Al foil and Ni foil surfaces, which are similar to those used by Chambers and Schwirzke, to provide further data on the conflicting results discussed above.

10.12 The Detector.

The basic design of the neutral atom detector is shown in diag. 89. The instrument is a combination of a Faraday cup and secondary electron emission detector (Montague, 1951) and a fast foil differential thermocouple similar to that described by Gardon (1953). The thickness of the nickel foil was 0.00005 inch, which yielded sensitivities of up to 10 microvolts per milliwatt with a time constant of about seven seconds. The sensitivity appeared to be limited by the diameter of the central copper wire. The foil and the central wire were extremely fragile. The temperature of the relatively massive copper cylindrical block was assumed independent of the beam because of the good conduction of its supporting structure. An aluminium-iron thermocouple was found to be less
successful than the nickel-copper structure.

An alternative thermal detector, consisting of a 0.001 inch (approximately) copper-constantan wire thermocouple soldered to aluminium foils 0.0005 thick, gave identical responses to within 2%, the limit of accuracy of the measurements, to the faster thermal detector. The time constant of this alternative thermal detector was about five minutes.

The target could be switched to either an electrometer when the net current was read, or to ground when the secondary electron current was read as shown in the diagram. Alternatively the electron collecting plate could be earthed and the target connected to an electrometer to record a current equal to \((1 + \gamma)\) times the incident ion current. The measurement procedure was to simultaneously record the secondary electron current and the thermocouple response for a given hydrogen atom beam. The corresponding proton beam current was adjusted to give the same thermocouple response and then the secondary electron current was read. Assuming that equal numbers of protons and hydrogen atoms per unit area per second give the same thermocouple response, the ratio \(\frac{\gamma_{H_1^+}}{\gamma_{H_1^0}}\) is immediately obtained.

Normal precautions were taken to ensure that the measurements were not invalidated by (i) insufficient potential difference between target and collector or
DIAG. 90.

PROJECTILE ENERGY—KEV
Diag. 92. Projectile Energy vs. Secondary Electron Coefficient

- **H**
- **H**
- **H**
- **H**

Present Data

Chambers 1964
(ii) inadequate detector entrance aperture or (iii) escape of electrons from the detector or (iv) impurity atoms in the proton beam.

10.13 Results.

The ratio $\gamma_{H_1^0}/\gamma_{H_1^+}$ over the energy range 2 - 50 Kev is shown in diag. 90. The present results, for a nickel surface inclined at $60^\circ$ to the beam, are seen to agree closely with the data of Stier (1954) for a nickel surface inclined at $0^\circ$ to the beam and also with the data of Chambers for a copper-beryllium surface inclined at $60^\circ$ to the beam. These results are not inconsistent with the view that the ratio $\gamma_{H_1^0}/\gamma_{H_1^+}$ is independent of the angle of inclination of the target to the beam.

The secondary electron yields, $\gamma$, as a function of the ion energy for the hydrogen ions $H_1^+$, $H_2^+$ and $H_3^+$ on Al and Cu-Be targets are shown in diagrams 91 and 92 respectively. No results are available for comparison with the present results for aluminium. Schwirzke's data on Cu-Be is not shown as it is almost identical with that of Chambers. The present results agree well with Chamber's results only for incident protons, while for $H_2^+$ and $H_3^+$ the two sets of data differ considerably in absolute value and energy dependence. An explanation of this discrepancy may be sought in the fact that the targets may have had a different composition and surface preparation. However, Large (1962) obtained a similar shape and energy dependence.
of his yields for widely varying methods of surface preparation even though the absolute values of $\gamma$ changed considerably. This result suggests that another, but not obvious, experimental factor must be responsible for the above disagreement of results.

The secondary electron yields for the three different ions $H_2^+$, $H_2^+$, and $H_3^+$ may be compared at the same ion velocity. Diag. 93 shows the ratios $\gamma_{H_2^+}/\gamma_{H_1^+}$ and $\gamma_{H_3^+}/\gamma_{H_1^+}$ for Al and Cu-Be surfaces for ion velocities in the range 0.6 to $1.8 \times 10^8$ cm/sec. For both surfaces at ion velocities less than $1.0 \times 10^8$ cm/sec, $\gamma_{H_3^+}/\gamma_{H_1^+} = 2.82$ and $\gamma_{H_2^+}/\gamma_{H_1^+} = 2.02$. These figures are (a) in good agreement with those of Large who used atomically clean surfaces whilst the present surfaces were only polished, and (b) in poor agreement with the results of Chambers who used polished surfaces prepared similarly to the present ones. The results of diag. 93 clearly show that, for velocities less than $1.0 \times 10^8$ cm/sec, molecular ions behave similarly to the appropriate number of constituent atomic ions at the same velocity. The slight difference in the yield of atomic and molecular ions may be accounted for by the fact that the cross section for ionization by $H_1^+$ is not necessarily half that by $H_2^+$ or one third that by $H_3^+$ at the same velocity. This is certainly the case when the target is a gas (Afrosimov, 1958,b) for then $\gamma_{H_2^+}/\gamma_{H_1^+} < 2$. The
ratio of molecular to atomic ion yields decreases as the ion velocity increases.

However above a velocity of \(1.0 \times 10^8\) cm/sec the values of the ratios \(\gamma_{\text{H}_3^+} / \gamma_{\text{H}_1^+}\) and \(\gamma_{\text{H}_2^+} / \gamma_{\text{H}_1^+}\) for aluminium become progressively less than those for the Cu-Se surface. Such an effect was suspected of being "instrumental" in origin, but its source was not discovered. A possible dependence of \(\gamma\) on the vibrationally excited states of the molecular ions was sought for, but not found. Any such dependence was certainly less than the 2% error in the determinations for that variety of ion source operating conditions discussed in Chapter 7.
Appendix 10.2

Further Information on the Apparatus

10.21 The 30 cm. Radius of Curvature Magnet

The magnet design is essentially a modified form of that used by Kerwin (1949). To obtain 90° deflection the equivalent first order "inflection" sector angle is 144 degrees with the pole edges inclined at 27 degrees to the normal incident beam direction. The inflection type magnet was selected because (a) for a given deflection angle it requires less iron than the normal 90° type magnet and (b) theoretically it should give second order focusing. However this improved focusing was not achieved in practice because of the fringing fields (see diag. 12) at the entry and exit pole edges. The theoretical energy resolution for an inflection type magnet is 

\[ \frac{E}{\Delta E} = \frac{R}{(2S + a)} \cdot \frac{0.165}{\theta} \]

where 

- \( R \) = radius of curvature = 30 cm., 
- \( S \) = width of entry and exit slits = 2 mm., 
- \( a \) = aberration which is estimated from beam profiles to be 2 mm. and \( \theta = \frac{1}{8} \) angle of divergence at the exit slit \( \approx \) not more than 3 degrees. Then \( \frac{E}{\Delta E} = 157 \).

The experimental energy resolution was approximately 108.

Diag. 94(a) shows a sketch of this magnet which was cast commercially from a low carbon steel.

A beam image stability of better than 0.1 mm. required a magnetic field stability of 0.02% which was readily achieved by a current stability of 0.01%. The energizing coils produced negligible heating of the iron core.
3 AMP. CONSTANT CURRENT SUPPLY.
The 30 cm. Magnet Power Supply

The nominally 3A, 30V constant current power supply, shown in diag. 94, was constructed to energize the 30 cm. electromagnet. The supply is basically a standard and simple design. The two series ASZ 18 control transistors were each mounted on blackened copper heat sinks of total area 220 cm$^2$ to restrict their temperature rise to 40 centigrade degrees above ambient. To limit the power dissipation in these transistors the series control resistors $R_1$ to $R_{10}$ were gauged with the input Variac transformer. The lowering of the input voltage caused unsatisfactory operation of the differential amplifier formed by the two OC 26 transistors which were then connected to a constant voltage collector supply of 55 volts. The 500 $\Omega$ resistance in parallel with the Zener diode OAZ 210, was a 10 turn helipot, which permitted changes of less than 1 ma to be made in an output current of 3 Amps. The coarse current control resistors $R_1$ to $R_{10}$ were wound from manganin wire.

Performance data

For an output current of 3 amps the current drift over 10 minutes was 0.01% and over two hours was 0.05% maximum fluctuation. Ripple across the 10 ohm magnet was 3 millivolts peak to peak. All these figures were improved at smaller output currents.

Diag. 97 shows a typical mass scan of the hydrogenic ions emerging from the R.F. ion source at a pressure of
Diag. 95. AV 26 Ionization Gauge Control Circuit.
DIAG. 96. ELECTRON COUPLED HARTLEY OSCILLATOR.
2.10^{-3} \text{mm.Hg. The high yield of } H_1^+ \text{ relative to } H_2^+ \text{ is obtained only when the internal glass surfaces of the source are clean. The } H_3^+ \text{ yield is strongly dependent on the gas pressure. The peak at an apparent mass of } \frac{1}{3} \text{ is due to protons with energy } \frac{E}{2} \text{ formed from the dissociation of } H_2^+ \text{ ions of energy } E.

10.23 \textbf{The Ionization Gauge Control Circuit}

The circuit of diag.95 was constructed to amplify the ionization currents from the AV 26 ionization gauge, whose sensitivity was approximately 1000 \mu A ionization current per $10^{-3}$ mm.Hg of hydrogen per 20 ma emission current. Full scale meter deflections corresponding to $10^{-5}$, $10^{-4}$, $10^{-3}$, and $10^{-3}$ mm.Hg were obtained. The operation of the circuit is evident from the diagram. The response of both the amplifier and the gauge were shown to be linear.

10.24 \textbf{The R.F. Oscillator}

Diag.96 shows the electron coupled Hartley type R.F. oscillator circuit which was selected originally to give an oscillation frequency, in the range 5 to 60 Mc/sec, which was practically independent of the load impedance to be coupled into the tank coil. A screen grid tube, an 807, was selected as it could generate a frequency practically independent of the plate supply voltage by adjusting experimentally the ratio of plate to screen voltage.
MAGNET constructed from 1" m.s. plates

MACHINED STUDS 1/2" diam.

WINDOW (2" x 2")

COILS (2) of approx 5000 t. (one only shown)

SIDE PLATES to compensate for window

DIAG. 98  4" RADIUS MAGNET
The complete R.F. oscillator and the electrodeless discharge type ion source were enclosed in an earthed wire gauze cage. All leads emerging from this cage were doubly shielded. The shielding was sufficiently effective to permit the operation of electrometers detecting currents of the order of $10^{-15}$ amps. only ten feet away from the R.F. oscillator.

10.25 The 10 cm. Collision Product Analysing Magnet

Diag.98 shows the collision product analysing magnet, which has a 10 cm. radius of curvature and is a normal $90^\circ$ deflection type magnet. The ion beam entered the field at the centre of the 6" pole gap so that both the positively and negatively charged components of the beam could be simultaneously momentum analysed. In order that the uncharged beam components could pass through the 2" x 2" hole in the magnet yoke the two energizing coils were placed as shown in the diagram. The beam geometry is similar to that for a standard $90^\circ$ deflection magnet. The magnet was fabricated from 6 x 1" thick low carbon iron sheets. A plot of magnetic field strength, B, against proton energy, E, showed that protons were deflected according to the relationship $E = 4.95 B^2$ (Kev/K gauss) ± 0.5%. The magnetic field strength was measured with a Hall effect gaussmeter.
FIG. 99  THE GRAPH OF THE FUNCTION WHICH DETERMINES THE CAPTURE PROCESS.
Appendix 10.3

Calculation of $\sigma_{10}$ for Protons in the Inert Gases.

Gryzinski (1957, 1959, 1963) shows that the capture cross section, $\sigma_{10}$, for a fast ion $B$ colliding with a stationary atom $A$ is given by

$$\sigma_{10} = \frac{k \cdot U_i^B}{(U_i^A)^3} \cdot G_c$$

where $k = 2\pi e^4 q_B^2$ \text{in atomic units}$

$e =$ electronic charge
$q_B =$ charge of the projectile ion $B$
$U_i^A, U_i^B =$ ionization potential of the target atom $A$ and the projectile atom, $B$, respectively.

$G_c =$ the one electron capture cross section function which is a function of the $U_i^A$ and $U_i^B$ and $v_B$ and $v_A$

$v_B =$ velocity of the projectile ion $B$
$v_A =$ orbital velocity of the captured electron of atom $A$.

The function $G_c$, has been calculated from a lengthy expression given by Gryzinski and is plotted in diag. 99 for several values of the parameters $U_i^B/U_i^A$ and $v_B/v_A$.

Sample calculations for one electron capture by a fast proton in various target gases are given below.

For a helium atom target

$$\sigma_{10} = 2 \cdot 4 \cdot 10^{-16} \ G(0.55; v_{H_1^+}/2.95 \cdot 10^8) \ \text{cm}^2/\text{atom}$$
For a neon target

\[ \sigma_{10} = 3.6 \times 10^{-16} \text{ } G(0.63; \nu_{H^+}/2 \cdot 75 \cdot 10^8) \text{ cm}^2/\text{atom} \]

For an argon target

\[ \sigma_{10} = 3.0 \times 10^{-15} \text{ } G(0.87; \nu_{H^+}/2 \cdot 4 \cdot 10^8) \text{ cm}^2/\text{atom} \]

For a krypton target

\[ \sigma_{10} = 1.31 \times 10^{-15} \text{ } G(0.97; \nu_{H^+}/2 \cdot 2 \cdot 10^8) \text{ cm}^2/\text{atom} \]

For an xenon target his theory fails badly since the capture process is exothermic. The predicted shape of \( \sigma_{10} \) is seen from diag. 99 for ratios of \( U_i^B/U_i^A \) greater than unity and this shape is readily seen to vary greatly from the experimental energy dependence of \( \sigma_{10} \).
Appendix 10.4

Beam Deceleration Measurements

Deceleration techniques for measuring beam energies have been discussed at length by Hagstrum (Symposium on Mass Spectrometry at National Bureau of Standards, 1951) and Fox (Rev. Sci. Inst. 19, 462, 1948). Diag. 100 shows the electrode arrangement. The beam defining slit was the base plate of the R.F. ion source since all the ions emerging from the source were to be collected. The suppressor guard confines the ion suppression (or deceleration) field to the vicinity of the suppressor plates. The equipotential lines were plotted on an electrostatic field plotter to check the efficiency of these guards and to estimate the beam defocusing. Great care was taken to ensure that defocusing did not destroy the efficiency of the collector.

Diag. 101 shows a typical deceleration curve in which the ion current, collected by the Faraday cup is plotted against the deceleration voltage in excess of that voltage which is used to extract the positive ions from the source. The differential of such a curve gives the distribution of energies of the ions in the beam. The width of the beam is the beam energy spread, $\Delta E$, and the voltage at which the peak of the distribution occurs is taken as the mean beam energy, $\bar{E}$. In general both $\Delta E$ and $\bar{E}$ may be readily estimated without the necessity of differentiating the deceleration curves.
Electron Capture and Loss Cross Section Values  
Projectiles - H\(^+\), H\(^0\), H\(^-\) Target - He  
All cross sections are in units of cm\(^2\)/atom

<table>
<thead>
<tr>
<th>Projectile Energy</th>
<th>$\sigma_{10}$</th>
<th>$\sigma_{17}$</th>
<th>$\sigma_{01}$</th>
<th>$\sigma_{07}$</th>
<th>$\sigma_{10}$</th>
<th>$\sigma_{11}$</th>
</tr>
</thead>
<tbody>
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Electron Capture and Loss Cross Section Values
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Electron Capture and Loss Cross Section Values
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All cross section values are in units of cm$^2$/atom

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Electron Capture and Loss Cross Section Values

Projectiles $H^+$, $H^0$, $H^-$ Target Krypton

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Electron Capture and Loss Cross Section Values
Projectiles $H^+, H^0, H^- -$ Target Xenon

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**Appendix 10.6**

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**Table 13**

Cross Section Values for total proton production, $\sigma^+_H$, and total negative hydrogen ion production, $\sigma^-_H$, for the collision of H$_2$ ions with the inert gases and nitrogen.

All cross section values are given in units of cm$^2$/atom.

<table>
<thead>
<tr>
<th>Target</th>
<th>He</th>
<th>Ne</th>
<th>Ar</th>
<th>Kr</th>
<th>Xe</th>
<th>N$_2$</th>
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</thead>
<tbody>
<tr>
<td>H$_2$ Energy/Kev</td>
<td>$\sigma^+_H$ (10$^{-17}$)</td>
<td>$\sigma^-_H$ (10$^{-20}$)</td>
<td>$\sigma^+_H$ (10$^{-16}$)</td>
<td>$\sigma^-_H$ (10$^{-19}$)</td>
<td>$\sigma^+_H$ (10$^{-16}$)</td>
<td>$\sigma^-_H$ (10$^{-18}$)</td>
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<tr>
<td>2</td>
<td>3.28</td>
<td>0.50</td>
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<td>0.82</td>
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<td>1.26</td>
<td>5.4</td>
<td>1.72</td>
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<td>2.5</td>
<td>1.40</td>
<td>5.0</td>
<td>1.93</td>
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Table 14
Cross section values for total $H_2^+$ production, $\sigma_{H_2^+}$; total $H_1^+$ production, $\sigma_{H_1^+}$; and total $H_1^-$ production for the collision of $H_3^+$ ions with He, Ne and Ar.

All cross section values are in units of cm$^2$/atom.

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<tr>
<th>Target</th>
<th>$H_3^+$ Energy Kev</th>
<th>$\sigma_{H_2^+}$ 10$^{-17}$</th>
<th>$\sigma_{H_1^+}$ 10$^{-17}$</th>
<th>$\sigma_{H_1^-}$ 10$^{-20}$</th>
<th>$\sigma_{H_2^+}$ 10$^{-17}$</th>
<th>$\sigma_{H_1^+}$ 10$^{-17}$</th>
<th>$\sigma_{H_1^-}$ 10$^{-19}$</th>
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Chapter 11.

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