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IONIZATION OF CESIUM ATOMS IN COLLISIONS WITH ATOMIC OXYGEN

by

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This report is identical to a thesis submitted to the University of Colorado in partial fulfillment of the requirements for the Ph.D. degree in physics.

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ABSTRACT

Woodward, Benjamin Weston (Ph.D., Physics)

Ionization of Cesium Atoms in Collisions with Atomic Oxygen

Thesis directed by Lecturer Alan C. Gallagher

A new technique has been used to measure the absolute cross section for the electron transfer reaction, \( O + \text{Cs} \rightarrow O^- + \text{Cs}^+ \), for both the \(^3\text{P}\) and \(^1\text{D}\) states of the oxygen atom. The \( O \) atoms are obtained by photodetachment from a 0.2 to 1.8 keV negative oxygen ion beam using high power pulsed lasers. By a suitable choice of laser wavelength, \( O \) atoms are produced either entirely in the ground \(^3\text{P}\) state or with a known fraction in the metastable \(^1\text{D}\) state. The fraction of negative ions undergoing photodetachment is great enough that the absolute neutral flux can be determined by measuring the loss in the negative ion beam with a gated detector. The cesium beam is a collimated thermal beam produced in an effusive source. The reaction products, both charged particles, are electrostatically separated from the neutral atoms and accelerated to approximately 5 keV for detection by open electron multipliers. Individual events are counted in both channels, and delayed coincidence is employed to provide a high signal-to-noise ratio. The cross section for electron transfer with \( O(^3\text{P}) \) atoms is of the order of \( 10 \times 10^{-16} \text{ cm}^2 \) across the energy range studied; and the total cross section for ionization of cesium is less than 30% larger. The electron
transfer cross section using $^1S_0$ states is significantly different only at 1 keV, where the cross section is much smaller than that for $0(^3P)$ atoms. In addition, the total absolute cross section for ionization of Cs in collisions with $0(^3P)$ is found to be of the order of $4 \times 10^{-16}$ cm$^2$ over the same energy range.
ACKNOWLEDGEMENTS

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# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>CHAPTER</th>
<th>PAGE</th>
</tr>
</thead>
<tbody>
<tr>
<td>I. INTRODUCTION</td>
<td>1</td>
</tr>
<tr>
<td>II. THEORETICAL CONSIDERATIONS</td>
<td>7</td>
</tr>
<tr>
<td>A. &quot;Curve Crossing&quot; Model</td>
<td>7</td>
</tr>
<tr>
<td>B. Results of Similar Experiments</td>
<td>14</td>
</tr>
<tr>
<td>III. DESCRIPTION OF EXPERIMENTAL APPARATUS</td>
<td>18</td>
</tr>
<tr>
<td>A. O⁻ Beam</td>
<td>16</td>
</tr>
<tr>
<td>B. Photodetachment of O⁻</td>
<td>26</td>
</tr>
<tr>
<td>C. Pulsed O Atom Beam</td>
<td>26</td>
</tr>
<tr>
<td>D. Cesium Atom Beam</td>
<td>31</td>
</tr>
<tr>
<td>E. Interaction Region</td>
<td>34</td>
</tr>
<tr>
<td>F. Detection of Charged Particles</td>
<td>36</td>
</tr>
<tr>
<td>G. Signal Processing</td>
<td>39</td>
</tr>
<tr>
<td>IV. THEORY OF THE EXPERIMENT</td>
<td>41</td>
</tr>
<tr>
<td>A. Crossed Beam Experiment</td>
<td>42</td>
</tr>
<tr>
<td>B. Measuring Collision Event Rates</td>
<td>44</td>
</tr>
<tr>
<td>C. Noise Sources in O Atom Measurements</td>
<td>52</td>
</tr>
<tr>
<td>V. EXPERIMENTAL PROCEDURE</td>
<td>55</td>
</tr>
<tr>
<td>A. Data Collection Procedure</td>
<td>55</td>
</tr>
<tr>
<td>B. Calibration of the Absolute O Atom Detector</td>
<td>56</td>
</tr>
<tr>
<td>C. Calibration of the Relative O Atom Detector</td>
<td>57</td>
</tr>
<tr>
<td>D. Measurement of Cesium Flux and Form Factor</td>
<td>59</td>
</tr>
</tbody>
</table>
### TABLE OF CONTENTS (continued)

<table>
<thead>
<tr>
<th>CHAPTER</th>
<th>PAGE</th>
</tr>
</thead>
<tbody>
<tr>
<td>V. (continued)</td>
<td></td>
</tr>
<tr>
<td>E. Charged Particle Detection Efficiencies</td>
<td>66</td>
</tr>
<tr>
<td>F. Arrival Time Spectrum of Cs$^+$ Ions</td>
<td>76</td>
</tr>
<tr>
<td>G. Miscellaneous Monitors</td>
<td>78</td>
</tr>
<tr>
<td>H. Tests for Validity of Data</td>
<td>79</td>
</tr>
<tr>
<td>VI. DISCUSSION OF ERRORS</td>
<td>84</td>
</tr>
<tr>
<td>A. Errors in Signal Determination</td>
<td>84</td>
</tr>
<tr>
<td>B. Errors in O Atom Measurements</td>
<td>88</td>
</tr>
<tr>
<td>C. Errors in Cs Atom Measurements</td>
<td>88</td>
</tr>
<tr>
<td>D. Errors in $^1$D Cross Sections</td>
<td>92</td>
</tr>
<tr>
<td>E. Summary</td>
<td>93</td>
</tr>
<tr>
<td>VII. RESULTS</td>
<td>95</td>
</tr>
<tr>
<td>A. $^1$D Fraction with N$_2$ Laser</td>
<td>95</td>
</tr>
<tr>
<td>B. Cross Sections</td>
<td>96</td>
</tr>
<tr>
<td>C. Discussion</td>
<td>104</td>
</tr>
<tr>
<td>D. Secondary Electron Yield on Be-Cu Surfaces</td>
<td>105</td>
</tr>
<tr>
<td>VIII. IONIZATION OF CESIUM IN COLLISIONS WITH NEGATIVE IONS</td>
<td>107</td>
</tr>
<tr>
<td>A. Description of Experiment</td>
<td>107</td>
</tr>
<tr>
<td>B. Theoretical Considerations</td>
<td>108</td>
</tr>
<tr>
<td>C. Discussion of Results</td>
<td>110</td>
</tr>
<tr>
<td>IX. CONCLUSIONS AND SUGGESTIONS FOR FUTURE EXPERIMENTS</td>
<td>119</td>
</tr>
<tr>
<td>A. Collision Processes</td>
<td>119</td>
</tr>
<tr>
<td>B. Photodetachment Studies</td>
<td>120</td>
</tr>
<tr>
<td>CHAPTER</td>
<td>PAGE</td>
</tr>
<tr>
<td>-------------------------------</td>
<td>------</td>
</tr>
<tr>
<td>BIBLIOGRAPHY</td>
<td>125</td>
</tr>
<tr>
<td>APPENDIX</td>
<td></td>
</tr>
<tr>
<td>A NITROGEN LASER</td>
<td>128</td>
</tr>
<tr>
<td>B ION SOURCE</td>
<td>142</td>
</tr>
<tr>
<td>C TIME OF FLIGHT</td>
<td>146</td>
</tr>
<tr>
<td>D ABSOLUTE O ATOM DETECTOR DETAILS</td>
<td>152</td>
</tr>
<tr>
<td>E CALCULATION OF CROSS SECTIONS FROM DATA</td>
<td>155</td>
</tr>
</tbody>
</table>
LIST OF TABLES

<table>
<thead>
<tr>
<th>Table</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>Laser Summary</td>
<td>26</td>
</tr>
<tr>
<td>II</td>
<td>Reaction Parametric Dependence</td>
<td>45</td>
</tr>
<tr>
<td>III</td>
<td>Signal and Noise Distributions</td>
<td>47</td>
</tr>
<tr>
<td>IV</td>
<td>1 keV Cross Section Ratios</td>
<td>96</td>
</tr>
<tr>
<td>V</td>
<td>Cross Sections for $O + Cs \rightarrow O^- + Cs^+ (\sigma)$ and $O + Cs^+ + e^- (\sigma')$</td>
<td>97</td>
</tr>
<tr>
<td>VI</td>
<td>$N_2$ Laser/Dye Laser Cross Section Ratios</td>
<td>99</td>
</tr>
<tr>
<td>VII</td>
<td>Ratios of Secondary Electron Yields</td>
<td>105</td>
</tr>
<tr>
<td>VIII</td>
<td>Cesium Ionization Cross Sections at 1 keV</td>
<td>115</td>
</tr>
<tr>
<td>IX</td>
<td>Candidates for Electron Affinity Measurements</td>
<td>124</td>
</tr>
<tr>
<td>X</td>
<td>YK - 217 Coaxial Cable</td>
<td>130</td>
</tr>
<tr>
<td>XI</td>
<td>Nitrogen Laser Infrared Output</td>
<td>141</td>
</tr>
<tr>
<td>XII</td>
<td>$O$ Atom/$O^-$ Ion Times of Flight</td>
<td>147</td>
</tr>
<tr>
<td>XIII</td>
<td>$Cs^+$ Time of Flight Program</td>
<td>151</td>
</tr>
<tr>
<td>XIV</td>
<td>Extraction of Collision Event Rates from Data</td>
<td>157</td>
</tr>
<tr>
<td>XV</td>
<td>Form Factor and Cesium Flux Program</td>
<td>160</td>
</tr>
<tr>
<td>XVI</td>
<td>Cesium and Oxygen Beam Profile Data</td>
<td>162</td>
</tr>
</tbody>
</table>
# LIST OF ILLUSTRATIONS

<table>
<thead>
<tr>
<th>Figure</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Schematic diagram of triple crossed beam experiment</td>
<td>5</td>
</tr>
<tr>
<td>2</td>
<td>Coulomb potentials for the lowest lying states of the (0 + Cs) and (0^- + Cs^+) systems</td>
<td>8</td>
</tr>
<tr>
<td>3</td>
<td>Approximate adiabatic potentials for the lowest lying states of the (0 + Cs) and (0^- + Cs^+) systems</td>
<td>10</td>
</tr>
<tr>
<td>4</td>
<td>Results of Landau-Zener approximation calculation for (0 + Cs + 0^- + Cs^+)</td>
<td>11</td>
</tr>
<tr>
<td>5</td>
<td>Non-adiabatic Coulomb potentials for the (0 + Cs) and (0^- + Cs^+) systems</td>
<td>12</td>
</tr>
<tr>
<td>6</td>
<td>Results of measurements of the cross section for (H + Mg \rightarrow H^- + \ldots)</td>
<td>17</td>
</tr>
<tr>
<td>7</td>
<td>Hot cathode ion source for production of negative ions</td>
<td>19</td>
</tr>
<tr>
<td>8</td>
<td>Schematic diagram of experimental apparatus</td>
<td>21</td>
</tr>
<tr>
<td>9</td>
<td>Experimental and theoretical cross sections for photodetachment of (0^-) ions</td>
<td>22</td>
</tr>
<tr>
<td>10</td>
<td>Arrangement of nitrogen and dye lasers with respect to the (O^-) beam</td>
<td>25</td>
</tr>
<tr>
<td>11</td>
<td>Schematic diagram of the absolute (O) atom detector</td>
<td>28</td>
</tr>
<tr>
<td>12</td>
<td>Schematic diagram of the relative (O) atom detector</td>
<td>30</td>
</tr>
<tr>
<td>13</td>
<td>Detail of the cesium reservoir and oven, interaction region, and (Cs^+) lens system</td>
<td>32</td>
</tr>
<tr>
<td>14</td>
<td>Detail of the interaction region and extraction lenses</td>
<td>35</td>
</tr>
<tr>
<td>15</td>
<td>Schematic diagram of the signal processing equipment</td>
<td>40</td>
</tr>
<tr>
<td>16</td>
<td>A typical calibration measurement for the absolute (O) atom detector</td>
<td>58</td>
</tr>
<tr>
<td>Figure</td>
<td>Title</td>
<td>Page</td>
</tr>
<tr>
<td>-------</td>
<td>-----------------------------------------------------------------------</td>
<td>------</td>
</tr>
<tr>
<td>17</td>
<td>Demonstration of the plateau effect in the Cs$^+$ current as a function of the current in the tungsten hot wire</td>
<td>61</td>
</tr>
<tr>
<td>18</td>
<td>Demonstration of the plateau effect in the Cs$^+$ current as a function of bias potential</td>
<td>62</td>
</tr>
<tr>
<td>19</td>
<td>O atom and Cs atom beam profiles in the interaction region</td>
<td>63</td>
</tr>
<tr>
<td>20</td>
<td>Comparison of the Cs$^+$ current expected and that actually measured</td>
<td>65</td>
</tr>
<tr>
<td>21</td>
<td>Calibration of the scanning slit and hot wire motion</td>
<td>67</td>
</tr>
<tr>
<td>22</td>
<td>Pulse height distributions from O$^-$ multiplier</td>
<td></td>
</tr>
<tr>
<td></td>
<td>a. &quot;Good&quot;</td>
<td>70</td>
</tr>
<tr>
<td></td>
<td>b. &quot;Bad&quot;</td>
<td></td>
</tr>
<tr>
<td>23</td>
<td>Demonstration of the plateau in Cs$^+$ count rate as a function of the potential on the repeller plates</td>
<td>73</td>
</tr>
<tr>
<td>24</td>
<td>Demonstration of the flat dependence of the Cs$^+$ count rate on the Cs$^+$ lens 1 potential</td>
<td>74</td>
</tr>
<tr>
<td>25</td>
<td>Demonstration of the flat dependence of the Cs$^+$ count rate on the Cs$^+$ lens 2 potential</td>
<td>75</td>
</tr>
<tr>
<td>26</td>
<td>Arrival time spectrum for Cs$^+$ produced in electron transfer events</td>
<td>77</td>
</tr>
<tr>
<td>27</td>
<td>Demonstration of the linearity of the deduced event rate with measured O atom flux</td>
<td>81</td>
</tr>
<tr>
<td>28</td>
<td>Demonstration of the linearity of the deduced event rate with the measured column density of Cs atoms</td>
<td>82</td>
</tr>
<tr>
<td>29</td>
<td>Absolute cross sections for electron transfer events as a function of O atom energy</td>
<td>101</td>
</tr>
<tr>
<td>30</td>
<td>Absolute cross sections for electron transfer events as a function of O atom velocity</td>
<td>102</td>
</tr>
<tr>
<td>Figure</td>
<td>Title</td>
<td>Page</td>
</tr>
<tr>
<td>--------</td>
<td>----------------------------------------------------------------------</td>
<td>------</td>
</tr>
<tr>
<td>31</td>
<td>Comparison of theoretical and experimental results for the electron transfer cross sections</td>
<td>103</td>
</tr>
<tr>
<td>32</td>
<td>Velocity dependence of the secondary electron yield enhancement obtained with nitrogen laser detached O atoms over that obtained with dye laser detached O atoms</td>
<td>106</td>
</tr>
<tr>
<td>33</td>
<td>Non-adiabatic Coulomb potentials for the O− - Cs systems</td>
<td>109</td>
</tr>
<tr>
<td>34</td>
<td>Demonstration of the linearity of Cs+ signal rate with O− current</td>
<td>111</td>
</tr>
<tr>
<td>35</td>
<td>Demonstration of the linearity of Cs+ signal rate with cesium column density</td>
<td>112</td>
</tr>
<tr>
<td>36</td>
<td>Total cross section for ionization of cesium by O− impact</td>
<td>113</td>
</tr>
<tr>
<td>37</td>
<td>Total cross sections for ionization of cesium by O atom and by O− ion impact</td>
<td>114</td>
</tr>
<tr>
<td>38</td>
<td>Demonstration of the linearity of the measured quantity ε1/0 with the value for ε2 obtained from the O− atom - Cs experiment</td>
<td>117</td>
</tr>
<tr>
<td>39</td>
<td>Relative cross section for photodetachment of S− as a function of photon energy</td>
<td>122</td>
</tr>
<tr>
<td>40</td>
<td>Switching circuit used for the pulsed nitrogen laser</td>
<td>129</td>
</tr>
<tr>
<td>41</td>
<td>Example of the narrow channel (flat) N2 laser (cross section view)</td>
<td>131</td>
</tr>
<tr>
<td>42</td>
<td>Example of the round channel N2 laser (cross section view)</td>
<td>132</td>
</tr>
<tr>
<td>43</td>
<td>Output spectrum from a small N2 laser of the round channel type</td>
<td>134</td>
</tr>
<tr>
<td>44</td>
<td>N2 laser output power as a function of measured capacitance in bank</td>
<td>135</td>
</tr>
<tr>
<td>45</td>
<td>N2 laser output power as a function of number and type of coaxial cables</td>
<td>136</td>
</tr>
<tr>
<td>Figure</td>
<td>Title</td>
<td>Page</td>
</tr>
<tr>
<td>--------</td>
<td>----------------------------------------------------------------------</td>
<td>------</td>
</tr>
<tr>
<td>46</td>
<td>N\textsubscript{2} laser output power as a function of applied voltage</td>
<td>137</td>
</tr>
<tr>
<td>47</td>
<td>N\textsubscript{2} laser output power as a function of N\textsubscript{2} pressure</td>
<td>138</td>
</tr>
<tr>
<td>48</td>
<td>Temporal differences in UV and IR outputs from the N\textsubscript{2} laser</td>
<td>140</td>
</tr>
<tr>
<td>49</td>
<td>Circuit diagram for the filament current regulator</td>
<td>144</td>
</tr>
<tr>
<td>50</td>
<td>Potential plot obtained for the interaction region y-z plane with a modeled screen</td>
<td>148</td>
</tr>
<tr>
<td>51</td>
<td>Potential plot obtained for the interaction region y-z plane without screen</td>
<td>149</td>
</tr>
<tr>
<td>52</td>
<td>Assumed Cs acceleration potentials</td>
<td>150</td>
</tr>
<tr>
<td>53</td>
<td>Circuit diagram of Gaussian filter amplifier</td>
<td>153</td>
</tr>
<tr>
<td>54</td>
<td>Circuit diagram of gated integrator</td>
<td>154</td>
</tr>
</tbody>
</table>
CHAPTER I

INTRODUCTION

Reliable atomic collision cross sections depend on knowledge of the kinetic energies and the energy states of the interacting particles. It is the purpose of this paper to introduce a technique suggested in 1965\textsuperscript{1} for the study of atomic collision cross sections at intermediate energies, a technique which allows identification of both the kinetic energy and electronic state of one of the species in the collision process. To demonstrate the feasibility of this technique, a reaction was selected for which a large cross section might be expected. The reaction $O + Cs \rightarrow O^- + Cs^+$ is only 2.4 eV endothermic and has the advantage that both reaction products are charged particles, which can be easily detected. The absolute cross section for this reaction has been measured at O atom energies from 200 eV to 1.8 keV and for two atomic states of the O atom: the ground $^3P$ state and the metastable $^1D$ state. The cesium atoms are in the ground state since they are present in the form of a thermal beam. This paper gives the details of these measurements and establishes the feasibility of this technique as a general method for obtaining a variety of atom-atom cross sections.

The basic idea is that the oxygen atoms are produced by photo-detachment ($O^- + hv \rightarrow O + e^-$) from a monoenergetic focused beam of $O^-$ ions. The signal-to-noise ratio that one calculates for the
brightest cw light sources is extremely low, since the atom production is considerably smaller than that from stripping reactions on the background gas. We have employed pulsed lasers of very high peak power to obtain a high signal-to-noise ratio, although at very low signal rates.

One of the motivations for this experiment was the analogy with a low energy reaction sequence of importance in re-entry wake ionization in the upper atmosphere.\textsuperscript{2,3} The reaction $O + Na \rightarrow O^- + Na^+$ followed by collisional\textsuperscript{2} and/or associative\textsuperscript{4} detachment of the $O^-$ ion was of considerable interest, but the rate for the $O + Na$ reaction could only be guessed. The $O + Cs$ reaction at high energy is taken as a prototype of the lower energy re-entry problem.

The remainder of this chapter gives an over-view of the essential features of this experiment. Chapter II reviews theoretical considerations and the results of other experiments to try to make predictions regarding the cross section to be expected. The description of the apparatus and the experimental procedures are separated by a treatment of the theory of the experiment, because the details of that theory depend on an understanding of the apparatus and because the procedure is in turn dependent on the theory of the experiment. Discussions of errors and results are followed by a chapter on another experiment performed on ionization of cesium. We conclude by describing other experiments to which some of these techniques might be applied.

The principal experiment to be described in this paper is a crossed beams measurement of the absolute cross sections for the
processes

\[ \begin{align*}
\text{O}^{(3P)} + \text{Cs} & \rightarrow \text{O}^- + \text{Cs}^+ \\
\text{O}^{(1D)} & \end{align*} \]  (1)

(which we term electron transfer) over the energy range from 180 to 1600 eV in the center of mass. Since the c.m. energy is 39 % of the O atom energy, the energies referred to in the rest of this paper will be the energy of the oxygen atom in the laboratory. Data were taken at five values of the O atom energy: 200, 300, 500, 1000, and 1800 eV. The cross sections for the process

\[ \text{O} + \text{Cs} \rightarrow \text{O} + \text{Cs}^+ + \text{e} \]  (2)

are also studied in these experiments since the O⁻ and Cs⁺ are detected independently. The oxygen atoms are produced by photodetachment from the O⁻ ion as mentioned above. The ion beam is a mass-analyzed focused beam of the order of 30 nA. Since there is a negligible momentum transfer in low energy photon collisions, the product neutral atoms retain the momentum distribution characteristic of the negative ions at the point of detachment. State selection in the O atoms is achieved by using two pulsed lasers operating at different wavelengths. The shorter wavelength laser, a molecular nitrogen laser, produces photons energetic enough to leave a large fraction of the resulting neutral O atoms in the metastable \( ^1\text{D} \) state; whereas the longer wavelength dye laser photons are not energetic enough to make other than ground state neutrals.

After detachment, the continuous ion beam is electrostatically deflected into a Faraday cup, while the pulses of neutral atoms continue along a straight path into an electrically and thermally
shielded interaction region where they cross a thermal cesium beam (see Fig. 1). $0^-$ ions formed there continue downstream with negligible momentum transfer and are electrostatically separated from the neutral atoms. These product $0^-$ ions are then accelerated into an electron multiplier where individual counts are detected. The Cs$^+$ ions formed in the $0^-$-Cs collision are extracted from the interaction region by an electric field which does not affect the $0^-$ products, and the Cs$^+$ ions are also accelerated into an electron multiplier. Counts from these two multipliers are gated according to the time of flight of each, and coincidences are sought in these gates.

In order to obtain absolute cross sections, the fluxes of the reacting neutral beams must also be measured. Rather than depend on the absolute value of the $0^-$ photodetachment cross section, photometry, and a knowledge of the spatial overlap of the light beam with the $0^-$ beam, we measure the $0$ atom flux absolutely by integrating the "notches" or holes in the ion beam due to the pulsed detachment. The Faraday cup at point S in Figure 1 is then the absolute atom "detector" in this experiment. It is an absolute measurement because the electronic system can be calibrated by reducing the ion beam a known amount by pulsing it off repetitively. The cesium beam flux is measured using a surface ionization detector which can be scanned across the entire Cs beam.

Because this is a triple crossed beam experiment, the signal rates are very low; but, because the atom pulses are very short (~1 usec), the gate widths in the $0^-$ and Cs$^+$ detection channels
Figure 1. Schematic diagram of the triple crossed beam experiment.
can also be very short, in principle giving high signal-to-noise ratios. In addition, the use of the coincidence technique allows a further improvement in the signal-to-noise ratio for the electron transfer process.
CHAPTER II

THEORETICAL CONSIDERATIONS

The emphasis of this chapter is to provide a basis for predicting the order of magnitude to be expected for the electron transfer cross section. The first part is a review of the theoretical situation; and in the second part the results of other experiments are considered with respect to the size of electron transfer cross sections in other systems.

A. "Curve Crossing" Model

Because a complete treatment of collision processes such as the ones considered in this paper involves an infinite set of coupled differential equations corresponding to all open channels, these problems have frequently been treated theoretically using a semiclassical approach termed the "Landau-Zener-Stueckelberg approximation" (LZS). This treatment is concerned with only two states, the initial and final ones. If one plots only the Coulomb potentials of the initial and final states of the oxygen-cesium system just for the ground states, there is an apparent crossing of the curves at an internuclear separation of about .6 nm. Because these two states are coupled, however, the adiabatic potential curves do not cross but join smoothly, giving some minimum separation between them, thus the term "pseudo crossing." For example, the adiabatic
Figure 2. Coulomb potentials for the lowest lying states of the O + Cs and O^- + Cs^+ systems. No other interatomic forces are included.
curves might appear as shown in Figure 3. The LZS approximation assumes that all of the probability of reaction between these systems occurs in the neighborhood of the "crossing point"; i.e., the internuclear separation for minimum separation between the adiabatic curves. Van den Bos has applied this approach to the two state system just described with the result shown in Figure 4.

The limitations of the LZS approximation have been discussed extensively in the literature. One of the more glaring faults is the high energy behavior where the cross section falls in proportion to $E^{-1/2}$ instead of $E^{-1}$. The experiments reported here probably do not cover an energy range sufficiently large to consider that defect the most serious. In Figure 5, we show energy levels for a number of the low lying oxygen and cesium states, where for this purpose, again only Coulomb interactions are considered. The multiplicity of states is larger even than shown, because spin-orbit splittings are ignored; furthermore, the interaction between the ionic and atomic states near the crossings splits the magnetic sublevels. Not all of the curves so formed are interacting, because of molecular symmetry considerations; nevertheless, there are even more interacting states than are shown in Figure 5. It is doubtful, then, that any two-state approximation can adequately predict the cross section for the reactions considered. It may be useful to use the curve crossing approach to make some plausibility arguments regarding the direction of error. It is assumed that the probability for changing curves at the "crossing" is independent of direction; i.e., it is the same coming in as going out. Let that probability be $p_c$. Then, we must have the probability for a reaction to occur,
Figure 3. Approximate adiabatic potentials for the lowest lying states of the $O + Cs$ and $O^- + Cs^+$ systems. Only Coulomb and some inter-system interactions are included.
Figure 4. Results of Landau-Zener approximation calculation for $O + Cs \rightarrow O^- + Cs^+$. (See Reference 7).
Figure 5. Non-adiabatic Coulomb potentials for the $O + Cs$ and $O^- + Cs^+$ systems, showing some of the excited state levels.
P = 2 p_c (1 - p_c) because the crossing must occur only once, but either coming in or going out. Now P has a maximum for p_c = \frac{1}{2} and becomes small for small or large p_c. First consider the case \( ^3P \rightarrow Cs + 0^- + Cs^+ \). Van den Bos's calculation will be valid only if p_c is very small for each of the outer crossings on the 0^- - Cs^+ curve. Since p_c is a value of order \( \frac{1}{2} \) for the desired crossing, it seems unreasonable to suppose it is much much smaller, say of order 10^{-2}, for the outgoing crossings; therefore, it seems at least plausible to expect a lower theoretical value for the cross section than the two state calculation predicts.

The case for \( ^1D \rightarrow Cs + 0^- + Cs^+ \) has not been computed at all; however, it would be a still cruder assumption to use a two state model in this case where there are a number of outgoing channels with nearly the same energy. Again, if one tries to use a succession of two state calculations, one for each crossing, each of the several p_c must be of order \( \frac{1}{2} \) in order to observe the charged reaction products.

The situation is further complicated by the possibility of a crossing with the potential well due to \( 0^- - Cs^+ (5p^5 6\sigma) \); i.e., \( 0^- \) and the first excited state of \( Cs^+ \). If that is an accessible state, then it will have many crossings with neutral-neutral potential curves. The depth of the potential well required and the instability of the component species seem to make this possibility rather remote.

Coffey, Lorents, and Smith consider the effects of a large number of crossings in the He^+ - Ne system in which oscillations
are observed experimentally in the cross sections for inelastic scattering. These oscillations are attributed to interference effects between competing channels. The period of the oscillations is energy dependent, but it is as much as several hundred eV.

B. Results of Similar Experiments

The electron transfer mechanism has been discussed in connection with low energy reactions of the type

\[ M + X_2 \rightarrow MX + X \]  \hspace{1cm} (3)

for which the "harpooning" model proposes the intermediate complex \( M^+X^- \), where \( M \) represents an alkali atom, and \( X_2 \) a halogen molecule. Cross sections for a number of reactions of the type (3) have been measured and are quite large. Since the energy defects of the \( MX_2 \) systems are small, the potential curves of \( M + X_2 \) and \( M^+ + X^- \) cross at a large internuclear separations, suggesting large cross sections for formation of the intermediate. One complication over the simpler atomic case is some uncertainty in the electron affinity to use in computing the crossing point, because the vertical affinity is not the same as the adiabatic one. Our purpose here, however, is to make order of magnitude arguments, so that question can be neglected. If, as postulated in the "harpooning" model, the cross section for electron transfer is principally dependent only on the radius of the crossing point, then the results should be valid for the \( Cs + O \) case, providing the large difference in kinetic energy of the reactants can be ignored. This assumption may be a gross simplification.
In the preceding section we have just argued that we should expect a smaller cross section than the Landau-Zener one due to the competition in outgoing channels. In the case

\[ M + X_2 \rightarrow M^+X^-_2 \rightarrow MX + X \]  

(4)

the departure of the halogen atom surely occurs before the \( M^+ \) and \( X^-_2 \) are widely separated, so the \( MX \) exists on the lowest lying adiabatic curve. In other words, the reaction products are competitive with the appearance of \( M^+ \) and \( X^-_2 \), so that the apparent electron transfer cross section would be quite low for this case.

For comparison purposes, Brodhead et al. report the cross section for \( Cs + I_2 \rightarrow CsI + I \) as \( 180 \pm 25 \times 10^{-16} \text{ cm}^2 \).

Some work has been reported\(^{14,15} \) on the relative cross sections for alkali ionization in low energy collisions of the type (5):

\[ M + X_2 \rightarrow M^+ + \text{products} \]  

(5)

Data for \( Cs - Br_2 \) and \( Na - O_2 \) show maxima at approximately 6 and 8 eV respectively in center-of-mass, and fall off approximately linearly with energy for energies greater than 10 eV. In the case of \( Na - O_2 \) collisions, the total cross section for sodium ionization was calculated to be \( 0.1 + .035, -.05 \times 10^{-16} \text{ cm}^2 \) at a center of mass energy of 10 eV.\(^{14} \)

Finally, we note a recent experimental result for the reaction \( H + Mg \rightarrow H^- + \text{products} \)\(^{16} \) where one concludes that \( Mg^+ \) is the principal additional product formed at the lower energies. The total cross section for formation of \( H^- \) is given, so it is at worst an
upper limit for the process \( H + Mg \rightarrow H^- + Mg^+ \). The cross section at 5 keV energy is \( 10^{-16} \text{ cm}^2 \) and falls exponentially with energy over the first 20 keV (see Fig. 6).

To summarize the results of other experiments, it does not appear to be possible to predict from them an order of magnitude for the electron transfer cross section in \( O - Cs \) collisions near 1 keV. The theoretical prediction of van den Bos is subject to rather large uncertainties in view of the approximations used.
Figure 6. Results of measurements of the cross section for \( H + Mg \rightarrow H^- + \ldots \). (See Reference 16).
CHAPTER III

DESCRIPTION OF EXPERIMENTAL APPARATUS

This chapter is intended to provide a thorough discussion of the apparatus used in this experiment. The atomic beams and the detectors are all contained inside a high vacuum system differentially pumped in four distinct stages. The pressure in the ion source is not measured but is estimated to be of the order of a few Torr. The next two chambers, in which the ion optics are contained, normally are operated at $2 \times 10^{-5}$ and $5 \times 10^{-7}$ Torr respectively. The photodetachment and collision chambers are ultra-high vacuum quality with metal gaskets everywhere except at the present photodetachment window. The pressure there ranges from $6$ to $60 \times 10^{-8}$ Torr during operating conditions.

The procedural details involving this apparatus are reserved to a later chapter.

A. $O^{-}$ Beam

The negative oxygen ions are formed in a hot cathode discharge source that has been used in previous negative ion experiments. A diagram of the present version of this source is shown in Figure 7. The filament is .051 cm diameter thoriated tungsten wire; external magnets (not shown) produce an axial magnetic field of about 200 Gauss between the soft ion anode and cathode plates.
Figure 7. Hot cathode ion source for production of negative ions.
The discharge is run in carbon monoxide gas, producing \( O^- \) and \( C^- \) by dissociative attachment and/or by ion pair formation as well as a number of impurity ions. The current obtainable at the cesium beam is energy dependent, ranging from more than 50 nanoamperes at 1.8 keV to 10 nA at 200 eV. The beam energy is identical to the anode voltage since the beam is at ground potential in the photodetachment region.

The energy spread from this source is estimated to be approximately 10 eV, when operated with 200 volts across the arc. This estimate is taken from the width of mass peaks and from the arrival time of \( N_2 \) laser photodetached \( O \) atoms at the relative atom detector.

The series of cylindrical and quadrupole lenses used to define a focused ion beam is not shown in the over-all apparatus schematic, Figure 8. The \( 90^\circ \) mass spectrometer has a resolution of less than 1 mass unit (up to at least mass 32) with the exit slit set at about 0.3 cm, a typical value for this experiment. The potential of the mass spectrometer region is held positive with respect to ground to gain the focusing advantage of higher energy ions. The effect of ion energy on the focusing properties of the beam is strong, with beam size increasing with decreasing beam energy.

B. Photodetachment of \( O^- \)

1. Cross Section

Figure 9 shows the cross section for photodetachment of \( O^- \) as a function of photon energy. The open circles are aggregates of
ELECTRON TRANSFER APPARATUS

$[O + Cs \rightarrow O^- + Cs^+]$
Figure 9. Experimental and theoretical cross sections for photodetachment of O$^-$ ions. (See References 1 and 18 for experimental values; Reference 19 for theoretical result)
experimental values,
and the solid curve is the most recent theoretical determination. The threshold at 3.43 eV corresponds to onset of production of the metastable \(^1D\) state of the neutral O atom, which has a lifetime of \(\approx 100\) sec. For photon energies less than 3.43 eV, photodetachment of \(O^-\) into any but the ground state \((^3P)\) of the neutral is energetically forbidden; whereas, above that value a certain fraction of the resulting neutrals will be in the \(^1D\) state. This fraction is estimated at 0.37 from the measured photodetachment cross section by a short extrapolation of the \(^3P\) part of the curve out to 337.1 nm, the wavelength of the nitrogen laser.

2. Molecular Nitrogen Laser

The pulsed nitrogen laser used here is based on ideas and designs developed elsewhere with some features incorporated to obtain maximum energy per pulse. The laser operates on the \(0 \rightarrow 0\) bandhead transition at 337.1 nm from the \(^3\Pi_u\) state to the \(^3\Pi_g\) state of molecular nitrogen. This is a self-terminating transition because the lifetime of the \(B\) state is much longer than the lifetime of the \(C\) state. Pumping is by direct electron impact in a pulsed dc discharge; the risetime of the current pulse must be short with respect to the 40 nsec spontaneous emission lifetime of the \(C\) state. A description of the relevant circuitry and performance data are given in Appendix A.

The electrodes in the laser channel are parallel to the direction of the laser beam, with a length which can be as short as 30 cm
and as long as 150 cm. This arrangement is generally known as a "crossed-field" geometry, since the electric field is perpendicular to the light direction. An important feature of this laser is its very high gain per unit length, a fact which leads to saturation of output power in a small fraction of the total length. This saturation gives rise to an effect termed "super-radiance", meaning that the laser gives stimulated emission output without the interferometer cavity employed in most lasers. The $N_2$ laser used in this experiment operates in a way which resembles a cavity in appearance only. Figure 10 shows a mirror at each end of the $N_2$ laser channel. One of the mirrors serves to provide a unidirectional output, and the other merely doubles the light intensity through the $0^-$ beam. A number of measurements with partially transmitting mirrors in interferometric alignment showed no detectable change in power at 337.1 nm as the mirror was tuned in and out of alignment.

3. Organic Dye Laser

For this experiment a $5 \times 10^{-4}$ M solution of Rhodamine 6G in ethanol is flashlamp pumped using a linear flashlamp in an elliptical cavity. The flashlamp is fired using the same type of pulse discharge circuit used for the nitrogen laser. The light output peaks near 595 nm with a spectral halfwidth of approximately 17 nm when no wavelength narrowing is attempted. The shape is nearly trapezoidal with the output flat over about 12 nm. For all of the absolute data collected, the output mirror is simply a fused silica plate with a reflection of just over 4% per surface. The optical cavity does not contain the $0^-$ beam due to space limitations.
Figure 10. Arrangement of nitrogen and dye lasers with respect to the $O^-$ beam.
Table I
Laser Summary

<table>
<thead>
<tr>
<th>Property</th>
<th>$N_2$ Laser</th>
<th>Dye Laser</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>337.1 nm</td>
<td>590 ± 10 nm</td>
</tr>
<tr>
<td>Photon Energy</td>
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<td>2.07 ± 0.03 eV</td>
</tr>
<tr>
<td>Pulse Width</td>
<td>10 nsec</td>
<td>300 nsec</td>
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<tr>
<td>Energy/pulse</td>
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<td>5 mJ</td>
</tr>
<tr>
<td>Photons/pulse</td>
<td>$5 \times 10^{15}$</td>
<td>$1.5 \times 10^{16}$</td>
</tr>
<tr>
<td>Photodetachment Cross Section</td>
<td>$9 \times 10^{-18}$ cm$^2$</td>
<td>$6 \times 10^{-18}$ cm$^2$</td>
</tr>
</tbody>
</table>

C. Pulsed O atom Beam

1. Separation from $O^-$

The parallel plates, D2 in Figure 8, electrostatically deflect the remaining negative ions into the shielded Faraday cup, which is positioned above the beam axis. The burst of neutral atoms, some 3000 per laser pulse, proceeds into the interaction region with the same energy and momentum that characterized the $O^-$ beam in the photodetachment region. The O atom slug is 2 to 4 cm long corresponding to a width in time from 0.2 to 0.8 μsec, depending on the beam velocity and laser type.
2. Absolute 0 Atom Detector

Absolute measurement of the neutral atom flux is accomplished by integration of the laser induced "notches" in the $0^-$ beam with a calibrated detector. Figure 11 is a schematic diagram of the detection circuitry. The laser induced notch in the $0^-$ beam is capacitively coupled into the preamplifier which produces a voltage step whose height is proportional to the input charge. This step is differentiated and amplified again prior to the first gate. This signal is too small and the baseline too unstable to be integrated directly; consequently, the Gaussian filter amplifier and FET switch are used to provide a longer, high amplitude signal and a quiet zero baseline respectively. The output of the integrator is a voltage proportional to the accumulated charge missing from the $0^-$ beam. The background subtraction using a comparison gate 0.2 msec after the laser detachment signal is a modification to the original system. This change has allowed substantial improvement in the 0 atom statistics. Detailed circuits are given in Appendix D.

The 0 atom detector is as complex as it is due to the high frequency noise on the ion beam which is well in excess of shot noise. This noise is so great that the notch in the ion beam cannot be observed on a single pulse basis, for example, on an oscilloscope. The integrator output for typical laser powers, $0^-$ beam intensities, and noise levels very little resembles the ideal of a "ramp", due to the very high noise level. The sum of 10 - 15 runs of 1000 pulses each, however, repeatedly provides a statistically meaningful mean, whose deviation can be as small as 7%.
Figure 11. Schematic diagram of the absolute O atom detector.
3. Relative O Atom Detector

At the very end of the apparatus (Fig. 8) is positioned an electron multiplier operated in a low gain mode (2 to 2.5 kV across this 15 stage multiplier produces a gain of about $10^4$). This detector observes the continuous beam of stripped neutrals as well as the burst of laser-detached neutrals. The burst of 3000 O atoms in a time of the order of 1 μsec gives a high signal-to-noise ratio signal which is processed as shown in Figure 12.

This is essentially the same basic scheme used in the absolute detector, but here the noise is only the stripped neutral background rather than the many times larger negative ion beam. This detector has a real time readout available (A in Fig. 12) which is used in optimization of photodetachment probability by careful adjustment of the laser beam overlap with the ion beam. This adjustment is normally accomplished simply by adjusting the height and position of the cylindrical focusing lens (Fig. 10) while observing the signal from the relative atom detector at point A.

The other function of this detector is to obtain a high signal-to-noise readout of accumulated O atom signals. The advantage is that the average relative O atom flux can be determined for a data run of even 100 pulses, rather than 100 times that much. In addition, this detector can be calibrated absolutely (for a given multiplier gain) in terms of the absolute detector (refer to Chapter V). The relative atom detector is also used in measuring the O atom beam profile, where absolute flux determinations are not necessary.
Figure 12. Schematic diagram of the relative # atom detector.
D. Cesium Atom Beam

1. Source

Figure 13 shows the essential details of the Cs reservoir and oven assembly finally used. This assembly is entirely of stainless steel and sealed with a gold wire gasket. The disadvantage of using stainless steel with its low heat conductivity is offset by the fact that this unit does not leak cesium metal (except through the oven slit). A previous all-copper assembly proved extremely difficult to seal adequately, with a number of serious consequences:

a. Cs on multipliers eventually forms CsOH which is extremely deliquescent; and water reduces the multiplier gains sharply.

b. Cs on insulators gives high leakage currents.

c. Cs on surfaces near detectors gives rise to higher background counting rates, and in the $0^-$ channel, unacceptably large.

The reservoir and oven sections are separately heated; the thin-walled steel tube between the oven and reservoir provides considerable thermal isolation. The reservoir section temperature establishes the vapor pressure, the functional dependence $^{26}$ given by

$$\log_{10} p = 11.053 - 1.35 \log_{10} T - \frac{4041}{T}$$

(6)

where $p$ is the partial pressure of Cs in Torr and $T$ the temperature in °K. The oven temperature determines only the mean velocity of the beam, and is kept hotter than the reservoir to prevent condensation of cesium there.
Figure 13. Detail of the cesium reservoir and oven, interaction region, and Cs\(^+\) lens system in the plane perpendicular to the O atom beam. B: Cs beam flag. S: screen. R: repeller plates. W: tungsten hot wire.
Since the vapor pressure is an exponential function of temperature, it is necessary to keep the reservoir temperature fixed quite closely. Using Equation 6 to calculate the change in vapor pressure as a function of a temperature change:

$$\frac{\Delta P}{P} = -1.35 \frac{\Delta T}{T} + \frac{4041}{\log_{10}e} \cdot \frac{\Delta T}{T^2}$$

which, for $T = 150^\circ C$, is 4.9 % per degree temperature change. As a result of such extreme sensitivity to temperature changes, a temperature controller is used to keep the reservoir temperature fixed. The device, an Artronix 5301, uses a platinum resistance wire as the temperature sensing element and is in principle capable of regulating the temperature to within .01$^\circ C$.

Iron-constantan thermocouples sense the temperatures of the reservoir and oven. These thermocouples are used in conjunction with a pyrometer which has an internal temperature reference. The pyrometer has a nominal accuracy of 2 % (full scale).

2. Cs Beam Properties

The exit aperture from the superheated oven is a .01 by 0.5 cm slit at the end of a wedge shaped canal. At the pressures used in these experiments, this aperture results in approximately an ideal effusive source, for which $I_c(\theta) = I_c \cos \theta$ describes the intensity of the cesium beam as a function of the angle $\theta$ away from the normal to the slit plane (see Fig. 13). Collimating slits imbedded in the bottom of the interaction region are 0.25 cm across by 0.5 cm long so that the intensity profile across the useful beam should be
nearly flat, as indeed it is found to be (see Fig. 19). All evidence indicates that the beam size is just what the geometrical projections predict. Typical densities at the 0 atom beam are from $1$ to $3 \times 10^{10}$ atoms/cm$^3$.

The unused fraction of the Cs beam is collected on the bottom of the interaction region and in the upper copper block, which is chilled by contact with the liquid nitrogen dewar (see Fig. 13). One is concerned with the chance that cesium might not stick with unity probability and migrate to other surfaces. At 80°K, the vapor pressure of cesium is negligible; and in practice the background contamination due to cesium appears to be much greater from other sources, notably leaks in the reservoir-oven assembly, and reflection from the warmer beam flag when it interdicts the beam. The cesium beam is detected in the interaction region, which is discussed in the next section.

E. Interaction Region

The interaction region is the heart of the apparatus, and a number of different functions are carried out there. It is a thick-walled copper box with entrance and exit holes for reacting atoms and product ions. It is in close thermal contact, through the upper Cu block, with a liquid nitrogen dewar; the temperature of the interaction region is found to be less than 100°K. Diagrams are shown in Figures 13 and 14.

Inside the interaction region, there are three individual elements used in measuring the beam profiles in the z-direction. These
Figure 14. Detail of the interaction region and extraction lenses in the plane perpendicular to the Cs beam. C: cesium beam collimating slits. S: screen. R: repeller plates. W: tungsten hot wire.
elements are the slit scanner (L), the hot wire (W), and repeller plates (R) (see Fig. 14). The slit is .051 by 1.0 cm, vertically, in a grounded stainless steel housing. The tungsten wire, .025 cm in diameter, is heated to approximately 1500 K to detect the neutral cesium beam by surface ionization (Langmuir – Taylor method). A ceramic rod supports the hot wire and is attached to the slit housing. The slit and wire are moved from outside the vacuum system by a mechanical feed-through with a micrometer adjustment. The motion is coupled directly into forward and backward movement of the scanning slit and hot wire by a lever arm which engages a sapphire rod connected to the slit housing. Although the slit and hot wire scan simultaneously, the wire is offset from the slit so as not to interfere with particles passing through it. The repeller plates are divided at the center to allow the tungsten wire to pass behind the plates. One function of these plates is to act as collector for Cs\(^+\) ions formed at the hot wire. In the second mode, a positive potential is applied to the repeller plates to push Cs\(^+\) ions formed in the oxygen-cesium reaction out through the exit hole. A screen is used at the position S in some of the experiments.

F. Detection of Charged Particles

1. O\(^-\) Ions

The old procedure for Cs\(^+\) extraction allowed a small electric field inside the interaction region to affect the O\(^-\) reaction product; however, at the 1 keV energy used, this field gives a negligible deflection. In most of the experiments, there is no electric
field inside the interaction region until the $0^-$ ions formed there have left.

To first order it is assumed that momentum transfer in the interactions is small, so that within this limitation the $0^-$ ions have the same trajectory as did the neutrals entering the interaction region. To check on momentum transfer in the collisions, we measured the $\text{Cs}^+$ arrival time spread (refer to Chapter V) and found it to indicate a very small momentum transfer.

A cylindrical tube after the interaction region operates as a refocusing lens (lens 3) for the lower energy ions which are more spread out in space. To separate the reaction product $0^-$ ions from the neutral atoms, a $90^\circ$ analyzer is used. The entrance and exit apertures of the analyzer are large (1.6 cm by 2.2 cm) since the beam is rather large at this point and high transmission is desired rather than energy analysis. The transmission of the analyzer assembly generally lies near 90%.

After the analyzer, the negative ions are accelerated into the first dynode of a fifteen stage (or seventeen stage in some experiments) electron multiplier of the venetian blind type. There is information in the literature \cite{27,28} regarding the efficiency and other properties of these multipliers. In general they are regarded to be high efficiency low noise detectors of both positive and negative ions with energies of several keV. Details of how these efficiencies are estimated are given in Chapter V.
2. Cs$^+$ Ions

The weak electric field mentioned above is to accelerate the Cs$^+$ ions formed in collisions to the aperture in the interaction region, where they are extracted by Cs$^+$ lens 1 (see Figs. 13, 14). The potential on the repeller plates is $+\frac{4}{3}$ V to $+6$ V; and the potential distribution inside the interaction region has been mapped with two dimensional electrostatic models. Outside the interaction region the ions are accelerated through the cylindrical lenses and plane parallel plates, shown in Figure 8, up to the potential on the first dynode of the multiplier, typically 6 - 8 kV. The multiplier is of the same type discussed above for detection of $0^-$ ions.

In order to obtain a high signal-to-noise ratio in the Cs$^+$ channel, it is necessary to make reasonably good time of flight calculations for the Cs$^+$ ion. To keep computations as simple as possible, the potentials are approximated by segments that are either uniform or of constant gradient. The calculations are programmed to be performed on the time-sharing computer. (Diagrams of the potential plots and approximations and the time of flight program are given in Appendix C). Calculations of time of flight were made for different acceleration voltages and various trajectories in the interaction region. These variations made slight differences in the over-all time of flight. The effect of different initial velocities is more significant, however, particularly for velocities opposite to the direction of extraction.
G. Signal Processing

The result of an electron transfer event is the appearance of a burst of charge at the collector of each electron multiplier, which is at an elevated potential. Figure 15 is a schematic diagram of the signal processing from that point. The first three elements in the signal processing train are standard commercial units, the result of which is a positive 0.5 μsec pulse 5 volts in amplitude. Of course the arrival times of the product ions at the two multipliers are different. The times of flight of Cs⁺ and O⁻ are calculated and measured to set the gate delays relative to initiation of a laser firing trigger pulse. For each cycle the coincidence logic unit puts out a pulse if there is at least one pulse in each channel during the gate. The gate width in the O⁻ channel is set at 1 to 3 μsec, and the Cs⁺ channel gate is set at 5 to 10 μsec.
Figure 15. Schematic diagram of the signal processing equipment.
CHAPTER IV

THEORY OF THE EXPERIMENT

Because the relationship of measured experimental parameters to
the absolute cross sections for ionization of cesium is intimately
connected with the experimental apparatus, this chapter is inserted
between a description of the apparatus and the experimental proce-
dures. The measured parameters are the following: count rates in
each of the three channels -- $O^-$, $Cs^+$, and coincidence; detection
efficiencies in the $O^-$ and $Cs^+$ channels; oxygen atom flux;
cesium atom flux; oven temperature; and the form factor or beam
overlap integral. The succeeding sections discuss the extraction of
the cross section in terms of these quantities. An important inter-
mediate step is the determination of the actual rate for $Cs$ ion-
ization events from the count rates and detection efficiencies. This
extraction depends directly on a full treatment of all the processes
occurring which give rise to signal, spurious signals, and noise.
The latter two sources of counts are generally referred to together
as noise in this discussion. Since there is a substantial noise
contribution in the $O$ atom determination, the sources of that noise
are also discussed in this chapter.
A. Crossed Beam Experiment

The advantages of the crossed beam technique for measuring cross sections include knowledge of the velocities of the interacting particles. The biggest disadvantage is that one must pay close attention to the spatial overlap of the two beams. These factors are contained in the expression for the rate $S$ of any collision event in the interaction volume.\(^{29}\)

$$
S = \sigma \frac{(v_c^2 + v_o^2)^{1/2}}{v_c v_o} \int_{-\infty}^{\infty} \int \int f_c(y', z') f_o(x', z') \, dx' dy' dz' \, \text{sec}^{-1} \quad (8)
$$

where $\sigma$ is the cross section for the processes with rate $S$.

$v_c$ is the cesium atom velocity

$v_o$ is the oxygen atom velocity

$f_c$ is the cesium atom flux ($#/\text{cm}^2 \cdot \text{sec}$)

$f_o$ is the total oxygen atom flux ($#/\text{sec}$).

The primed co-ordinates are those of the moving (O atom system) in the rest frame of the slower (Cs) atoms. Following the usual simplification,\(^{29}\)

$$
S = \sigma \frac{(v_c^2 + v_o^2)^{1/2}}{v_c v_o} \int f_c(z) f_o(z) \, dz \quad (9)
$$

where, since $z = z'$,

$$
f_c(z) = \int f_c(y', z') \, dy'
$$

and

$$
f_o(z) = \int f_o(x', z') \, dx'
$$
The integral can be rewritten in terms of total atom fluxes (#/sec) and a geometrical factor only, called the form factor:

\[ \int f_c(z) f_o(z) \, dz = F_c F_o f \]  

(10)

where

\[ F_m = \int f_m(z) \, dz \]

and

\[ f = \frac{\int f_c(z)f_o(z)dz}{\int f_c(z)dz \int f_o(z)dz} \]

We have then,

\[ S = \sigma \frac{(v_c^2 + v_o^2)^{\frac{1}{2}}}{v_o v_c} \frac{F_c F_o f}{F_c F_o f}. \]  

(11)

In this experiment, the largest cesium velocities are \(5 \times 10^4\) cm/sec, whereas \(v_o\) (minimum) is \(5 \times 10^6\) cm/sec. In that case

\[ (v_c^2 + v_o^2)^{\frac{1}{2}} = v_o \]  

(12)

and

\[ S = \frac{\sigma F_c F_o f}{v_c}. \]  

(13)

Of course, the cesium atoms have a velocity distribution, which for an effusive beam is given by

\[ f(v) = f_c \frac{2v^3}{\alpha^4} e^{-v^2/\alpha^2} \quad \text{where} \quad \alpha^2 = \frac{2kT_o}{m}. \]  

(14)

The mean of that distribution is

\[ \frac{\langle v \rangle}{v_c} = \frac{3}{4} \sqrt{\frac{2\pi k T_o}{m_c}}. \]  

(15)
It will be useful to define the cesium column density, $D_c$, as the number of Cs atoms per cm$^2$ through the full 0 atom path. Here, the column density,

$$D_c = \frac{F_0^f}{V_c};$$

therefore

$$S = \frac{\sigma F_0^f}{V_c} = \sigma F_0 D_c.$$ (17)

A test of the validity of the data is to demonstrate the linearity of the experimentally determined event rate, $S$, with $F_0$ and $D_c$.

B. Measuring Collision Event Rates

1. Sources of Signal and Noise

The separation of signal from noise requires an examination of the sources of unwanted counts. Some of these are due to additional atomic collision processes taking place, and others are due to electronic effects. In the succeeding examination, most of the possible noise sources are small or even unobservable.

In the following table these symbols are defined:

- $O$: laser produced 0 atoms
- $O_s$: 0 atoms formed by 0$^{-}$ stripping
- $B$: background gas
- $S$: surface
- $M$: an arbitrary particle other than Cs
**Table II**

Reaction Parametric Dependence

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Operational Dependence</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$O^-$ Beam</td>
</tr>
<tr>
<td>1. $O + Cs \rightarrow O^- + Cs^+$ (signal)</td>
<td>x</td>
</tr>
<tr>
<td>2. $O + Cs \rightarrow O + Cs^+ + e^-$ (signal)</td>
<td>x</td>
</tr>
<tr>
<td>3. $O_s + Cs \rightarrow O^- + Cs^+$</td>
<td>x</td>
</tr>
<tr>
<td>4. $O_s + Cs \rightarrow O + Cs^+ + e^-$</td>
<td>x</td>
</tr>
<tr>
<td>5. $O + Cs_2 \rightarrow O^- + Cs^+ + Cs$</td>
<td>x</td>
</tr>
<tr>
<td>6. $O + Cs_2 \rightarrow O + Cs^+ + Cs + e^-$</td>
<td>x</td>
</tr>
<tr>
<td>7. $O + B \rightarrow O^- + B^+$</td>
<td>x</td>
</tr>
<tr>
<td>8. $O + B \rightarrow O + B^+ + e^-$</td>
<td>x</td>
</tr>
<tr>
<td>9. $O + e \rightarrow e^- (\pm M^+)$</td>
<td>x</td>
</tr>
<tr>
<td>10. $Cs^+$ in beam</td>
<td>x</td>
</tr>
<tr>
<td>11. $h\nu + e \rightarrow e^-$</td>
<td>x</td>
</tr>
<tr>
<td>12. &quot;dark&quot; counts (dark counts, ions from pumps, corona)</td>
<td>x</td>
</tr>
<tr>
<td>13. laser induced electrical noise</td>
<td>x (but not light dependent)</td>
</tr>
</tbody>
</table>


It is apparent that we cannot distinguish between O atom reactions with Cs and those with Cs₂ which dissociatively ionize the dimer. The dimer concentration in the source, however, is estimated to be smaller than 0.15 % for the most severe operating conditions. We assume the cross section for the spurious reactions listed are small enough (< 10⁻¹⁴ cm²) that these contributions are negligible. One can put limitations on the masses of spurious positive ions, B⁺, M⁺ according to the gate width used in the Cs⁺ channel. The range can be rather broad, but it certainly excludes such candidates as O⁺ and Cs₂⁺. The table of reactions ignores second order effects such as Os + B.

2. Detection

In analyzing the manner in which the above reactions are detected, consider that there are eight combinations of on-off relations for the three beams. In addition we consider a ninth: the difference between the laser light blocked and the laser not firing, when both other beams are off. This case is necessary to check for counts due to electrical noise associated with laser firing. The following table demonstrates the appearance of signal and noise counts from the various reactions in each of the three counting channels as a function of the nine beam configurations. The numbers tabulated refer to the reactions listed in Table II. It is desired to express these relations in terms of signal and noise. As remarked previously, the reaction with Cs₂ cannot be separated here, but one is confident that contribution can be ignored. In that case, one
<table>
<thead>
<tr>
<th>Run</th>
<th>Channel</th>
<th>0⁻</th>
<th>Cs⁺</th>
<th>Coincidence</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>all off</td>
<td>12</td>
<td>12</td>
<td></td>
</tr>
<tr>
<td></td>
<td>no firing</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>a</td>
<td>all off laser (L) firing</td>
<td>12,13</td>
<td>12,13</td>
<td>13</td>
</tr>
<tr>
<td>b</td>
<td>L on</td>
<td>11,12,13</td>
<td>12,13</td>
<td>13</td>
</tr>
<tr>
<td></td>
<td>0⁻ off</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cs off</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>c</td>
<td>L off</td>
<td>12,13</td>
<td>12,13</td>
<td>13</td>
</tr>
<tr>
<td></td>
<td>0⁻ on</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cs off</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>d</td>
<td>L on</td>
<td>7,8,9,11,12,13</td>
<td>7,8,9,12,13</td>
<td>7,13</td>
</tr>
<tr>
<td></td>
<td>0⁻ on</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cs off</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>e</td>
<td>L off</td>
<td>12,13</td>
<td>10,12,13</td>
<td>13</td>
</tr>
<tr>
<td></td>
<td>0⁻ off</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cs on</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>f</td>
<td>L on</td>
<td>11,12,13</td>
<td>10,12,13</td>
<td>3,13</td>
</tr>
<tr>
<td></td>
<td>0⁻ off</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cs on</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>g</td>
<td>L off</td>
<td>3,12,13</td>
<td>3,4,10,12,13</td>
<td>3,13</td>
</tr>
<tr>
<td></td>
<td>0⁻ on</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cs on</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>h</td>
<td>all on</td>
<td>1,3,5,7,8,9,11,12,13</td>
<td>1 to 10,12,13</td>
<td>1,3,5,7,13</td>
</tr>
</tbody>
</table>
finds the same functional relationship for the noise in each of the three counting channels; it is the combination \((d + g - c)\). This result is just the sum of counts obtained with 0 atoms off and with Cs atoms off minus the number of counts obtained when both beams are off. The net signal is then given by \((h + c - d - g)\). This fortunate circumstance considerably simplifies the data collection process; and, if certain contributions are small, further simplifications are possible. There may, of course, be accidental coincidences counted. These and other corrections have not yet been taken into account but will be treated in the following section.

The pressure dependent reactions are indicated in Table II because of the importance background pressure plays in most crossed atomic beam experiments. In this experiment the pressure dependent sources of noise can be completely accounted for in an appropriate group of background runs. This circumstance is in distinct contrast to the situation in ac modulated beams, in which the signal must be measured as a function of pressure and extrapolated to zero pressure.\(^{32}\)

3. **Signal Extraction**

Because the output pulse of the linear amplifier (Fig. 15) has a basewidth of some 2 μsec, the integral discriminator can generate only one output pulse in a similar time interval. This interval is a large fraction of the gate width in each channel; in addition, the coincidence logic can produce a count only once per cycle independent of gate width. The result of these electronic details is that, to
first order, we may expect at most one count per cycle in each channel regardless of signal arrival rate. To minimize the size of the corrections, data is collected at low rates. It is still necessary to analyze these corrections for two reasons: one, in order to be able to put an upper limit on the correction terms which are not considered; and two, to properly reduce all of the data, some of which is taken at higher rates to enhance the signal-to-noise ratio.

In general, the background counting rates are of the same order of magnitude as the signal rates, so that in the following discussion terms of the same order in signal × noise are grouped together.

- **a. 0^- Channel.** Let $p_0$ be the probability for exactly one electron transfer event to occur per laser pulse; let $\varepsilon_1$ be the probability for the scaler to register a count if one such event occurs (efficiency); let $a_0$ be the noise counting rate (per pulse) and $a_1$ be the signal + noise counting rate; then, because the probability for $m$ events per pulse is $p_0^m$,

\[
a_1 = a_0 + \varepsilon_1 p_0 \quad \text{(1st order)}
\]

\[
+ \varepsilon_1 (2 - \varepsilon_1) p_0^2 - \varepsilon_1 a_0 p_0 \quad \text{(2nd order)} \tag{18}
\]

\[
+ \varepsilon_1 (3 - 3\varepsilon_1 + \varepsilon_1^2) p_0^3 - \varepsilon_1 (2 - \varepsilon_1) a_0 p_0^2 \quad \text{(3rd order)}
\]

\[
+ \cdots
\]

and the over-all signal rate per pulse is given by

\[
s_0 = p_0 + 2 p_0^2 + 3 p_0^3 + \cdots \tag{19}
\]
In this channel, the probability of obtaining more than one count per cycle is assumed to be zero because of the very narrow gate used (~2 μsec).

b. \textbf{Cs}⁺ \textbf{Channel.} In this instance, the gate is open for an interval from 5 to 10 μsec. We introduce two parameters: \( \alpha \), an estimate of the probability of counting the second of two un-correlated pulses (signal + noise), and \( \beta \), the probability of counting the second of two correlated pulses (both signal). The arrival time spectrum of the \textbf{Cs}⁺ indicates that the probability of counting three signal events is negligible. Let \( p_1 \) be the probability for exactly one cesium ionization event to occur per pulse, \( p_0 \) the probability for exactly one electron transfer event per pulse, \( b_0 \) the noise count rate (per pulse), \( b_1 \) the signal + noise count rate, and \( \varepsilon_2 \) the \textbf{Cs}⁺ channel efficiency. Then

\[
\begin{align*}
b_1 &= b_0 + \varepsilon_2(p_0 + p_1) \\
&+ \varepsilon_2(2-\varepsilon_2 + \beta \varepsilon_2) \cdot (p_0^2 + p_1^2 - p_0 p_1) - \alpha b_0 \varepsilon_2(p_0 + p_1) \\
&+ \left[ \varepsilon_2(3-3\varepsilon_2 + \varepsilon_2^2) + \varepsilon_2^2 \beta(3-\varepsilon_2 - \beta \varepsilon_2) \right] \cdot \left( p_0^3 - p_0^2 p_1 - p_0 p_1^2 + p_1^3 \right) \\
&- \alpha b_0 \varepsilon_2(2 - \varepsilon_2 + \beta \varepsilon_2) (p_0^2 + p_1^2 - p_0 p_1) \\
&+ \cdots \\
\end{align*}
\]

(20)

and

\[
s_1 = p_1 + 2 p_1^2 + 3 p_1^3 + \cdots
\]

(21)

is the over-all signal rate (per pulse) for cesium ionization (only) events.
c. Coincidence Counter. In the coincidence channel, it is impossible to register more than one count per cycle, so that the treatment is rather like the 0\(^{-}\) channel alone. Using the previous definitions and letting \( c \) be the counting rate in the coincidence register,

\[
c = \varepsilon_1\varepsilon_2 p_0 + \varepsilon_1(1 - \varepsilon_2) (b_0 + \varepsilon_2 p_1) p_0 + \varepsilon_2(1 - \varepsilon_1) a_0 p_0^2 \\
+ \varepsilon_1\varepsilon_2 (2 - \varepsilon_1) \cdot (2 - \varepsilon_2) p_0^2 \\
+ a_0\varepsilon_2(2-\varepsilon_2) p_1^2 + \varepsilon_1(1-\varepsilon_2)^2 \cdot (b_0 + \varepsilon_2 p_1) p_0^2 + \varepsilon_2(1-\varepsilon_1)^2 a_0 p_0^2 \\
+ \varepsilon_1\varepsilon_2(3 - 3\varepsilon_1 + \varepsilon_1^2) \cdot (3 - 3\varepsilon_2 + \varepsilon_2^2) p_0^3 - \varepsilon_1\varepsilon_2 p_0 (b_0 + \varepsilon_2 p_1) a_0 \\
+ \ldots
\]  

(22)

d. Solutions. There are three equations in two unknowns; consequently, one could solve any two and use the third as a redundancy check. It happens, however, that the Cs\(^{+}\) efficiency, \( \varepsilon_2 \), is not well known; therefore, we treat it as a parameter to give the most reasonable solutions. To be specific, Equation 18 is solved directly for \( p_0 \), (giving \( s_0 \)). Then we treat Equations 20 and 22 as simultaneous equations in two variables, redesignating \( p_0 = p_2 \) to distinguish it from the result of Equation 18. Then

\[
s_2 = p_2 + 2 p_2^2 + 3 p_2^2
\]

(23)

which is to be compared with \( s_0 \). Now suppose the second order terms are small with respect to first order ones. The ratio
\[
\frac{s_0}{s_2} = \varepsilon_2 \left[ \frac{a_1 - a_0}{c} + \text{higher order terms} \right]
\] (24)

must be unity because there is no mechanism for producing \( O^- \) without a Cs ionization event. It is possible but tedious to compute \( \varepsilon_2 \) directly; so we use the simplicity of Equation 24 to iterate rapidly for a best fit to all data runs taken under the same conditions.

C. Noise Sources in O Atom Measurements

In the case of the oxygen atom flux measurement it is worthwhile to divide the sources of noise into two categories: one, the fluctuations in the ion beam, laser power and resulting atom flux; and two, noise introduced in the O atom detector itself.

1. Fluctuation in O Atom Production

First, consider the ideal case of a shot noise limited ion beam in photodetachment. The number of electrons detached per laser flash is given by

\[
\mu = \sigma_{\text{pd}} \frac{W I_{O^-}}{E \nu} \tag{25}
\]

assuming spatially uniform beams of the same height, \( w \) and no saturation effects,

where \( W \) is the laser energy per pulse

\( E \) is the photon energy

\( \sigma_{\text{pd}} \) is the cross section for photodetachment of \( O^- \)

at the photon energy, \( E \)
$I_0$ is the $0^-$ current
$v$ is the $0^-$ velocity.

Using the $N_2$ laser and a 1 keV ion beam, we have

\[ n = 130 I_0 (\text{nA}); \]  

(26)

therefore one expects of the order of $10^3$ or more neutral $0$ atoms per pulse even for small currents, giving a shot noise limit of only 3%. Moreover, after $10^3$ pulses, shot noise contributes 0.1% uncertainty, in nearly the worst cases.

The situation is considerably worse than this due to two factors: one, the high frequency noise on the ion beam is considerably in excess of shot noise; and two, the pulse to pulse stability of the pulsed lasers is not good. The result of these factors is that the number of $0$ atoms produced per pulse fluctuates widely from one shot to the next. The ion beam noise varies roughly as the square root of the ion current, giving some improvement in signal-to-noise with lower ion currents. We estimate the degree of white noise up to several MHz on the ion beam at 20 to 70% depending on ion current and source conditions. The pulse to pulse power fluctuations in the lasers are not well established, but the standard deviation is probably less than 40% of the mean in the more severe case, the dye laser, and 20% of the mean for the nitrogen laser. It should be remarked that these fluctuations are small enough that a good mean atom signal can be determined from the relative $0$ atom detector after the order of 100 laser pulses.
2. Absolute O Atom Detector

In detecting the number of neutrals produced by looking at the charge lost in a much larger, noisy ion beam, one is faced with the problem of measuring a signal in a situation in which the signal-to-noise ratio is very small. The solution is to integrate long enough to reduce the uncertainty to an acceptable value. This procedure is viable only if all the noise is completely random.

The output of the atom detector is a voltage which is assumed to be proportional to the charge missing from the ion beam during a short time interval, the gate width. One important test of this assumption is to check the detector output for linearity with input in the calibration procedure (refer to Chapter V); we require that it be linear at least over the device's operational range. Another test is the repeatability of the output for a given input; i.e., to determine that the gain of the system is constant with time. Finally, it is important to check that the baseline or zero of the detector is time independent over the duration of a few runs with the light on and off. The absolute detector did not completely satisfy this last criterion over some of the data collection; however, it happens that the fluctuations in the zero are random within a reasonable approximation. Thus the net effect of this contribution is to broaden the distribution of data points for atom number, occasionally leading to very large uncertainties.
CHAPTER V

EXPERIMENTAL PROCEDURE

The validity of the data obtained is primarily dependent on the details of experimental procedure and on the satisfaction of a number of consistency checks. This chapter discusses these details and shows the results of the tests that are necessary to insure the highest possible confidence in the data.

A. Data Collection Procedure

In the previous chapter it is shown that the background counts in each channel are given by the letter designations \((g + d - \sigma)\). These experiments regularly allow a simplification in the data collection procedure. Recall that the difference between a \(g\) and \(\sigma\) run in the \(0^-\) channel is only that due to signal from stripped neutrals. Even in the coincidence channel this contribution is almost never observed, so we have taken the background counting rate in the \(0^-\) channel \((a_0)\) to be the \(d\) run alone; i.e., the counts obtained with only the Cs beam off.

In the \(Cs^+\) channel, the counting rates were never significantly different in \(d\) and \(\sigma\) runs; moreover, the time of flight spectra indicate a minimum (unobservable) collection rate of positive ions of masses outside the range 110 to 160 amu. Consequently, the background rate in the \(Cs^+\) channel has been taken to be that given
by the $g$ run; i.e., that with the laser light blocked, so no ions are photodetached.

The data collection procedure is as follows: We record the counting rates in each of the three channels as well as the atom signals from both the absolute and relative detectors for the sequence of runs $g$, $h$, $d$. The number of laser flashes per run is either 1000 or 2000, taken at a repetition rate of 5, 10, or 20 per second. We number each set of three runs ($g$, $h$, $d$) sequentially for each opening of the vacuum system (group). Subgroups of data runs are designated for sets of conditions which are approximately uniform.

E. Calibration of the Absolute O Atom Detector

Calibration of the absolute O atom detector is performed in a straightforward manner. A short 50 V pulse is applied to deflection plates near the photodetachment region by a pulser capable of delivering up to $10^5$ pulses per second. If these pulses are approximately $1/2$ usec wide, the total $O^-$ beam, also measured at the shielded Faraday cup, is reduced by about 5%, and the decrease in $O^-$ beam is measured directly using a Keithley Electrometer, model 610 B. The next step is to reduce the pulse rate to 10 sec$^{-1}$ and record the 0 atom detector output for a certain number of pulses. This establishes a definite ratio between charge lost and integrator output:

$$\frac{\Delta I}{e} = \text{number of charges lost per } 10^5 \text{ pulses} \tag{27}$$

Let charges lost be called "atoms", 
\[ \frac{\Delta I}{1.6 \times 10^{-14}} = \text{number of atoms per pulse}, \]  

but \( \beta \frac{V}{N} \) = number of atoms per pulse where \( \beta \) is the calibration constant, and \( V \) is the integrator output in volts for \( N \) pulses on \( D_1 \).

If \( \Delta I \) in nA, \( \beta = \frac{N \Delta I}{16V} \times 10^6 \)

It is essential to determine the linearity of this rather complex device, since the input signal and noise vary over a wide range.

The \( 0^- \) beam is attenuated at the exit slit of the mass spectrometer, and the above procedure is repeated. Figure 16 shows a typical result, proving linearity within the measurement uncertainties. This test is repeated with the current fixed but making changes in \( \Delta I \) by altering the width of the deflecting pulse. A similar result is obtained.

C. Calibration of the Relative 0 Atom Detector

The absolute and relative 0 atom detectors have been operated simultaneously for nearly all of the data runs; however, in some cases the signal from the absolute detector is too noisy to obtain good absolute cross sections. In addition, frequently the 0 atom signal fluctuates markedly over the span of a data session. These facts make necessary the calibration of the multiplier detector relative to the absolute detector.

The calibrations are almost trivial. The net signals from the absolute detector are compared with the net signals from the relative multiplier detector for 10 to 20 runs of 2000 pulses. Net
Figure 16. A typical calibration measurement for the absolute O atom detector.
signals are those obtained after corrections have been added to account for the laser induced electrical noise in the prompt gate and for any other background signals.

D. Measurement of Cesium Flux and Form Factor

The scanning slit and hot tungsten wire described in Chapter III are used in measuring the cesium and O atom profiles in the z direction, from which information the cesium flux and form factor are calculated. Typically the following operations are carried out simultaneously, but that is not necessary.

1. O Atom Profile

It is significant to note that the profile of the neutral atom beam is scanned and not the ion beam, even though one assumes the two would be nearly identical in the interaction region. The relative atom detector integrates some 500 atom pulses (with a factor 10 larger gain than is used for the full beam) for several slit positions across the beam. Results of typical O atom beam scans are shown in Figure 19.

2. Cs Atom Profile

Since the work function of tungsten exceeds the ionization potential of cesium, one expects 100% ionization at any wire temperature which maintains a clean surface.\textsuperscript{33} We have obtained a temperature - current comparison using an optical pyrometer; however, it is more significant to measure the current collected as a function
of wire heating current. One can be certain that the ionization is complete when the collector current is flat as a function of hot wire current. Such a plot is shown in Figure 17.

Because the interaction region is designed primarily as a reaction chamber, collection of the \( \text{Cs}^+ \) current is far from optimum. Probably due to the open slot in the collector, it takes an unusually large collection field to obtain a plateau in ion current as a function of detector bias (see Fig. 18). When the \( \text{Cs}^+ \) current has reached a plateau for hot wire current and collection field at all positions in the cesium beam it is assumed that the collection is 100% efficient. In general the cesium beam is nearly constant over approximately 0.5 cm in the interaction region, dropping very sharply at the edges. Figure 19 shows the superposition of oxygen and cesium beams.

The integrals required are computed numerically on the time sharing computer using linear interpolation between the measured points. The form factor depends only on the relative positions of the two beams, but the absolute cesium flux is obtained from the cesium current data. The current measured,

\[
I_c = e \int_{a}^{b} \lambda w,
\]

where \( \int_{a}^{b} \lambda \) is the conventional flux,

\( \lambda \) is the length of the beam, and

\( w \) is the diameter of the hot wire.

The total cesium flux, \( F_c = \int l \int_{a}^{b} dz \)

so that \( F_c = \frac{1}{ew} \int I_c dz \) atoms per second

(30)
Figure 17. Demonstration of the plateau effect in the Cs\(^+\) current as a function of the current in the tungsten hot wire.
Figure 18. Demonstration of the plateau effect in the Cs\textsuperscript{+} current as a function of bias potential on the repeller plates.
Figure 19. O atom and Cs atom beam profiles in the interaction region.
The wire diameter is measured before and after these experiments, giving

\[ F_c = 2.50 \times 10^{14} \int I_c \ dz \quad \text{Cs atoms/sec.} \quad (33) \]

To complete the relationship between cesium beam and source temperatures, the values of detected Cs\(^+\) current are shown as a function of indicated reservoir temperature (Fig. 20). For comparison the solid line gives the values computed assuming an ideal effusive source and the vapor pressure vs temperature relationship from the Handbook of Chemistry and Physics.\(^{34}\) These values lie between those of Taylor and Langmuir\(^{26}\) and of Stull and Sinke.\(^{31}\) The vertical bars on this curve indicate the range of these three sets of values. We do not expect good absolute comparison for these data, due to the uncertainty in the thermocouple calibration. The experimental points are normalized to an oven temperature of 200°C (this correction is at most 5%). The resulting slope is more steep than that calculated; one explanation for such behavior would be deviations from non-effusive flow. Since non-effusive flow would be more directed in space and more pronounced at higher temperatures, there would be a stronger temperature dependence than calculated.

3. Calibration of Scanner Motion

The sapphire rod on which the scanning slit and tungsten wire are mounted is moved by a lever arm attached to a micrometer head outside the vacuum system. It is not convenient to measure the mechanical advantage of the lever; consequently, we calibrate the scanner motion directly. A telescope is visually aligned with the
Figure 20. Comparison of the Cs\(^+\) current expected and that actually measured as a function of the temperature at the Cs reservoir.
beam axis and focused on the slit portion of the scanner. The telescope is mounted on a movable frame whose translation is measured with a vernier scale to within .005 cm. One sets the telescope cross hairs on one edge of the slit (which must be brightly illuminated) and records the telescope position as a function of micrometer settings. Figure 21 demonstrates two important features of this calibration: one, that the motion is linear with micrometer values; and two, that the deviations of individual points from the line (due to sticking and slipping) are small relative to the slit width and wire diameter.

E. Charged Particle Detection Efficiencies

The over-all detection efficiency is defined as the probability of a scaler registering a count when an appropriate event occurs. We define $\epsilon_1$ as the efficiency for the $O^-$ channel and $\epsilon_2$ as the efficiency in the $\text{Cs}^+$ channel. The over-all efficiencies are products of individual efficiencies for several processes, some of which can be determined directly and others only estimated.

1. $O^-$ Channel Efficiency

The over-all efficiency for registering a count in the $O^-$ channel scaler is

$$
\epsilon_1 = \prod_{j=1}^{4} p_j
$$

(34)

where $p_1$ = transmission of $O^-$ from interaction region to $O^-$ multiplier;
Figure 21. Calibration of the scanning slit and hot wire motion.
\[ p_2 \equiv \text{probability of producing and accelerating at least one electron on the first dynode (detection efficiency)} \]
\[ p_3 \equiv \text{probability of the amplifier output pulse exceeding the discriminator level (counting efficiency)} \]
\[ p_4 \equiv \text{probability of the pulse arriving within the gate}. \]

The transmission is determined by using the multiplier as a Faraday collector for the full \( 0^- \) beam; indeed, the focusing is empirically adjusted to optimize the transmission to the \( 0^- \) multiplier. We have found, also empirically, combinations of stray magnetic fields which can enhance the apparent transmission through the analyzer and acceleration sections and which serve to inhibit the counting of electrons presumably produced by heavy particle impact on surfaces in the analyzer region. For the lower energy beams, a positive potential on the cylinder lens downstream from the interaction region aided in increasing the transmission figure. The number, \( p_1 \), is not the same as the transmission for \( 0^- \) ions produced in the interaction region for the following reasons:

a. The laser detaches in a line of vertical dimension smaller than the ion beam; consequently the vertical loss will be less than for the whole \( 0^- \) beam.

b. The \( 0^- \) ions produce secondary electrons some of which are lost in the current collection mode, resulting in an underestimate for the current there. On the other hand the shielded Faraday cup, near the interaction region is designed to minimize secondary electron losses, which in fact it does.
c. The acceleration region after the 90° analyzer will provide some refocusing.

d. Momentum transfer in the collisions may deflect the product O ions so much that they are not detected.

The detection efficiency can be estimated from the literature.\textsuperscript{27,28} To insure the highest possible value, a guard structure and tube is carried more negative than the first dynode, acting as a secondary electron suppressor (in the direction away from the multiplier) for the first three or four dynodes. We assume the value 0.9 for $p_2$. This value is qualitatively checked by measuring the counting rate as a function of accelerations of the impact ions beyond 5 - 6 kV, so a relatively high detection efficiency is experimentally justified.

The counting efficiency, $p_3$, is defined as the number of pulses from the discriminator per electron event on the first dynode. Some information on this efficiency can be obtained by measuring the counting rate vs multiplier voltage (i.e., gain) and looking for a plateau. A more elegant and rapid way of testing the counting efficiency is to measure the pulse height distribution obtained from ion impact on the first dynode. If the pulse height distribution has a well defined peak similar to that shown in Figure 22a, one assumes that a negligible number of counts are too small to be counted; however, the converse is not necessarily true. Although the pulse height distribution is theoretically a compound Poisson function,\textsuperscript{27} experience in this and other laboratories\textsuperscript{27} indicates that such is not always the case. Figure 22b shows a pulse height
Figure 22. Pulse height distributions from $O^-$ multiplier.
distribution for which one would be unable to estimate a high counting efficiency, but for which a counting efficiency of some 90% can be deduced from experiments. Because of the difficulty of making an a priori determination of the counting efficiency from such a distribution, multiplier voltage drops and ion acceleration parameters are chosen to optimize the pulse height distribution in the sense of Figure 22a.

The large signal-to-noise ratio obtainable due to the gated signals makes the experiment possible. Because the signal and noise can be time correlated and because the signal arrival time distribution is not flat but more or less Gaussian, there can be a signal-to-noise advantage in narrowing the gate, eliminating signal in the wings of the distribution. If the gate is too narrow, the gating efficiency, $p_4$, is less than unity. In practice the gating efficiency is usually unity, with a lower limit of 0.85 in the most severe case.

2. Cs$^+$ Channel Efficiency

The same general considerations apply in the Cs$^+$ channel with the exception that it is not feasible to measure the Cs$^+$ transmission independently. The transmission is studied indirectly. Using a reduced O$^-$ beam to ionize cesium in the same region as in the O atom experiment, large Cs$^+$ signal counting rates can be obtained with excellent signal-to-noise (refer to Chapter VIII). The counting rates were optimized as functions of the following parameters:
a. Repeller potential  
b. Lens 1 potential  
c. Lens 2 potential  

Figures 23-25 show plateaus for each of these parameters. It is assumed that the existence of a flat dependence of counting rate on these parameters implies essentially complete transmission. In addition, the optical transmission of the wire screen at the interaction region has been measured to be 0.8; however, the transmission for charged particles would be expected to be larger due to field penetration between the mesh wires.

As detailed in Chapter IV, the over-all efficiency, \( \varepsilon_2 \), can be determined from the data obtained from the electron transfer experiment. These determinations regularly indicated a smaller efficiency than had been estimated. Since the interaction volume size is nearly the same as the \( \text{Cs}^+ \) exit aperture, the most likely place for transmission losses appeared to be in the interaction region. Previous field plots indicated that the \( \text{Cs}^+ \) lens 1 field penetration with no screen would provide good focusing. The wire mesh screen at the exit aperture was removed, and the lens 1 voltage is now pulsed on only after any \( \text{O}^- \) ion formed in the interaction region has left. The repeller is maintained at ground potential in this mode of extraction. The result of this change was in fact to achieve a 30% increase in the \( \text{Cs}^+ \) channel efficiency, from approximately 0.6 to 0.82.
Figure 23. Demonstration of the plateau in Cs\(^+\) count rate as a function of the potential on the repeller plates.
Figure 24. Demonstration of the flat dependence of the Cs$^+$ count rate on the Cs$^+$ lens 1 potential.
Figure 25. Demonstration of the flat dependence of the Cs⁺ count rate on the Cs⁺ lens 2 potential.
F. Arrival Time Spectrum of Cs\(^+\) Ions

When the noise in both channels is low enough, the signal-to-noise ratio in the coincidence channel is of the order of 30 or more. It is then possible to reduce the gate widths on one channel to the minimum 1 µsec (nominal) and still get enough signal in the coincidence channel to scan the arrival time spectrum of products in the Cs\(^+\) channel. Figure 26 gives the result of such a scan compared with calculated values and the previously used gate settings. (The same technique is applicable in the 0\(^-\) channel, but it is merely a verification of the value computed from multiplier 0 atom signal arrival time.) It is noteworthy that the absence of signal for all shorter and longer arrival times puts an upper limit on the amount of noise that can be attributed to 0 atom ionization of particles other than cesium. In particular, Cs\(_2\)^+ would have an arrival time of approximately 36 µsec. Other masses which can be excluded on the basis of these data are from 10 to 110 amu and from 160 to 280 amu. With some 27 coincidence counts arriving in the gate used, the upper limit for spurious positive ions is less than 4%. Other runs have verified these results.

The other information obtainable from these data is an upper limit to the momentum transfer in the cesium ionizing collisions. Because the spectral width is so narrow (<4 µsec), the momentum transfer is minimal. Making time of flight calculations for different points of origin of the Cs\(^+\) ions and including the velocity spread associated with the collimation parameters, one expects an arrival time spread of approximately 2.2 µsec. The remainder is
Figure 26. Arrival time spectrum for Cs produced in electron transport from the Cs+ channel alone, which includes all sources of positive ions.
attributed to momentum transfer in the collisions, putting an upper limit to the 0° deflection at 8 mrad. In these calculations, the width at 10% of the peak height is the one quoted.

G. Miscellaneous Monitors

1. Reservoir Temperature

The temperature controller provides both a front panel meter indication and an electrical output proportional to the deviation of the sensor temperature from the set point. Until it proved unnecessary, chart recordings were made of the voltage output as a function of time. Currently, the short term temperature changes are less than 0.1°C, so that the recorded monitor is not used. There can, however, be a long term drift in the reservoir temperature if the power input is not set at just the equilibrium value. This drift is taken into account through periodic recording of the thermocouple sensed temperature and especially through repeated measurements of the cesium beam flux.

2. Oven Temperature

In these experiments the oven temperature ranged from 170° to 240°C; but since $\frac{\nu}{c} \propto \sqrt{T(\circ K)}$, the effect of this parameter on the cross sections is minimal. It was a practice to observe the oven temperature periodically for this reason: the beam flag occasionally sticks to the oven, presumably because a layer of cesium on the underside of the beam flag can braze to the top of the oven. Such a bond reduces the oven temperature markedly, sometimes cooling it
below the reservoir temperature. Since this condition leads to condensation in the oven, the beam density would change dramatically.

Usually a much smaller temperature change is observed, but frequently a more serious difficulty accompanies sticking of the beam flag. If the flag is responsible for a drop of liquid cesium on the outside of the oven, then an intense cesium source occurs there, a large portion of which is observed to be Cs⁺.

3. Cesium Channel Continuous Count Rate

The signal from the Cs⁺ multiplier is gated according to time of flight considerations, but the gate follows the output of the integral discriminator. In parallel with the gate, a frequency meter monitors the over-all count rate in the Cs⁺ channel. This count rate registers immediately any anomalous change in the cesium beam as described above.

H. Tests for Validity of Data

A necessary but not sufficient test of the data's validity is the demonstration of linearity of the net signal with the 0 atom and Cs atom fluxes. Ideally one would like to determine each relationship with all other parameters fixed; however, the length of time required to change cesium densities makes it unfeasible to require the 0 atom flux to be held constant. Accordingly, we first established linearity of the signal with 0 atom flux, then normalized the cesium data to a certain 0 atom flux.
Figure 27 shows a number of determinations of signal as a function of O atom flux, with the cesium density held constant but different for each group. Representative error bars include only random errors due to counting statistics. In the case of Group D, the cesium flux was fluctuating in time, but no allowance is included for that fact.

In Figure 28, signal normalized to a specific O atom flux is plotted as a function of the cesium column density. In this case, all the absolute cross section data for the nitrogen laser produced O atoms at 1 keV are included. In general these data are from different groups; consequently, the relative uncertainties in these data are nearly as large as the uncertainty in the relative cross sections. The filled squares, however, are points from a single group, for which the relative uncertainty is smaller.

A more complete test of the data's validity would include checks that the deduced cross sections are independent of all other parameters, as predicted by the theory of the experiment. Such parameters are certain potentials, background pressure, laser repetition rate, etc. In fact, most of the potentials inside the vacuum apparatus affect the efficiency in one channel or the other so that the proper test would be to deliberately reduce the efficiencies in a controlled way and check that the resulting cross section is unchanged. Although the complexity and duration of these experiments prohibited most such tests, there are some data to support the assumption that in fact the cross sections are independent of these parameters. The cross section at 1 keV O atom energy
Figure 27. Demonstration of the linearity of the deduced event rate with measured O atom flux. Representative error bars include only statistical uncertainties.
Figure 28. Demonstration of the linearity of the deduced event rate with the measured column density of Cs atoms. Representative error bars include only the uncertainty in the O atom number.
using the nitrogen laser has been measured over the range of $\text{Cs}^+$ channel efficiencies (from .5 to .8) and with the background pressure from $7 \times 10^{-8}$ to $6 \times 10^{-7}$ Torr. There is no correlation between the measured values and those parameters. The restriction is that the pressure, cesium density, O atom flux, and $\text{Cs}^+$ efficiency all take on a range of values over those nine measurements, so that there might exist some pathological systematic error whose functional dependence exactly cancels in the tests that were made. But since the O atom flux and Cs atom flux functional dependences have been checked under controlled conditions, the probability that there is such a systematic error is extremely small.
CHAPTER VI

DISCUSSION OF ERRORS

This discussion of errors includes both systematic and random errors in measuring the signal rates and neutral atom fluxes. In each case, the final uncertainty quoted represents one standard deviation; however, in many instances systematic errors can only be estimated, and in these cases we have treated a maximum error estimate as just one standard deviation.

A. Errors in Signal Determination

1. Counting

If experimental conditions remain fixed, one can estimate the statistical error in the signal and noise counts using random counting statistics for which the estimated standard deviation, \( \delta(n) = \sqrt{n} \) (shot noise). Here \( n \) represents the total number signal or noise counts over the number of runs for which conditions remain approximately uniform. Generally \( n \) for signal + noise is of order \( 10^3 \). The number of noise counts measured in a background run may be as large as \( n/2 \), although usually the background count rate in the noisier channel (0\(^{-}\)) is less than \( n/3 \). The counting error for signal in such a case will be larger than \( \sqrt{n} \), for

\[
(n \pm \sqrt{n}) - \left( \frac{n}{3} \pm \sqrt{\frac{n}{3}} \right) = \frac{2}{3} n \pm (\sqrt{n} + \sqrt{n}/3)
\]  

(35)
and the \( \text{rms} \) error is \( \sqrt{\frac{4}{3}} n \). The fractional uncertainty in the net signal using the \( \text{rms} \) error is \( \sqrt{\frac{3}{n}} \), which for \( n = 1000 \) is 5\( \frac{1}{2} \)%.

This value is typical for the net counts in the \( 0^- \) channel, with lower values characteristic of the counting error for the other two scalers because the signal-to-noise ratio is higher.

2. Efficiencies

The most important systematic error (probably with random components) is in the determination of the efficiency \( \epsilon_1 \). The sources of error in determining \( \epsilon_1 \) are the following:

a. loss of secondaries at multiplier when measuring transmission

b. difference in transmission due to acceleration focusing

c. difference in transmission for atoms vs ions due to narrowness vertically of atom beam

d. loss of ions due to momentum transfer

e. detection efficiency estimate

f. counting efficiency estimate.

The first three errors in the transmission efficiency make the actual value larger than that measured. Since the measured transmissions are in the range 0.8 to 0.9, the maximum uncertainty due to these three sources is about 25%. An upper limit to the loss in transmission efficiency due to momentum transfer is obtained from the Cs\(^+\) arrival time data. These data indicate at most an 8 m rad deflection for 90% of the product \( 0^- \) ions. The maximum loss in
transmission is estimated to be 6%, but the loss is considered negligible when lens 3 is in use. The following test is performed to check the secondary electron loss: An electrometer measuring O⁻ current to the relative 0 atom detector multiplier is biased at a positive potential with respect to ground. The difference in current for zero bias and +22 V bias is 13% of the total current detected; a plateau begins at about +9 V bias. The same test cannot be performed with the upper multiplier, but that one is better shielded and magnets are placed to optimize the current there so the value obtained for the transmission is used uncorrected. The current to the upper multiplier is typically 90% of that to the back multiplier when some secondary electron suppression is incorporated at the back multiplier, putting an upper limit of 10% on the improvement in transmission due to acceleration focusing after the 90° analyzer. If the beam is approximately cylindrically symmetric, the maximum increase in transmission due to the vertical narrowing would be to move halfway between the measured value and 100%. There is an overlap of this effect with the acceleration focusing one. The sum of all the transmission uncertainties is estimated at a maximum of 10% for the lower transmission figures sliding down to 5% for the higher transmissions. The uncertainty in the detection efficiency is estimated at 5%; whereas, the uncertainty in counting efficiency varies from ±10% to ±3%, depending upon the shape of the pulse height distribution.

The Cs⁺ channel efficiency is obtained from the data itself by applying the criterion that the signal rates deduced from the O⁻
channel and from the coincidence channel must be equal (refer to
Chapter IV). Let \( N \) be the total number of pulses for which \( \varepsilon_2 \)
may be regarded to be constant. We take \( C = N c, A_1 = N a_1, A_0 = N a_0 \),
so that approximately,

\[
\varepsilon_2 \approx \frac{C}{A_1 - A_0}
\]

(36)

and the constant of proportionality is only dependent on \( \varepsilon_2 \) in
higher order. It has been stated above that the counting error in
determining the denominator of the expression is about \( \sqrt{\frac{4}{3}} A_1 \), for
typical operating conditions. But in determining this ratio, some
of the uncertainties can be disregarded. We consider that the actual
number of events occurring, \( S \), is statistically uncertain by an
amount \( \sqrt{S} \). But this uncertainty is reflected in all the channels
detecting these events; in particular, both \( C \) and \( (A_1 - A_0) \)
fluctuate together according as \( 1/\sqrt{S} \) times their value. Thus, the
statistical uncertainty in determining \( \varepsilon_2 \) should be due to the
noise fluctuations, \( \sqrt{A_0} \), alone. This error is contained in both
terms \( A_1 \) and \( A_0 \), so the rms uncertainty is \( \sqrt{2} A_0 \). The frac-
tional uncertainty in \( \varepsilon_2 \),

\[
\frac{\Delta \varepsilon_2}{\varepsilon_2} = \frac{\sqrt{2} A_0}{A_1 - A_0}
\]

(37)

ranges from 9% to 2% over the several groups, where all data
from a group are considered together. This error assessment is based
on the assumption that \( \varepsilon_2 \) is constant for all data in a group.
This is not always the case; in particular, some changes can be
attributed to the difference in focusing of the \( 0^-/0 \) atom beam.
As a result of such changes and uncertainties, the smallest error in \( \epsilon_2 \) is estimated to be approximately \( 3\% \) and the largest as much as \( 10\% \), with most values uncertain by about \( 5\% \).

B. Errors in O Atom Measurements

The error in calibration of the absolute O atom detector is small, at most \( 5\% \). The calibration is energy independent within the random errors in these measurements, which is the theoretical result. The random errors of the absolute O atom detector are in some cases the largest single uncertainties in these experiments. The very best values for mean O atom fluxes are about \( 7\% \) uncertain, with other determinations as poor as \( 35\% \). Use of the relative atom detector allows inclusion of data from widely differing O atom fluxes. It has the disadvantage that we must take into account the possibility of different electron yields on the multiplier's first dynode for the two atom states.

C. Errors in Cs Atom Measurements

With only a few exceptions, the uncertainty in measuring the cesium flux is due to systematic errors. The results obtained make use of the assumption that every Cs atom which hits the hot wire is ionized and then collected at the repeller plate, where it is assumed that the detected current is entirely due to \( \text{Cs}^+ \) arrival. The zero for this measurement is obtained by closing the beam flag and cancelling out any leakage current; consequently, there is no way to discriminate against ions formed from the \( \text{Cs}_2 \) dimer or
against Cs$^+$ in the beam itself. We can, however, put upper limits on the dimer/monomer ratio (refer to Chapter IV); and we can independently check for the Cs$^+$ contribution. Both of these species contribute a negligible quantity to the cesium flux measurement.

1. Ionization Efficiency

Datz and Taylor$^{35}$ have studied the question of ionization efficiency of the alkali metals in detail, considering both experimental and theoretical results. They conclude that there is no reason to doubt the predicted result of 100% ionization for cesium (and rubidium); furthermore, they advance theoretical reasons to exclude possibility of reflection prior to thermal equilibration. The case of cesium is especially satisfactory because the ionization probability is flat with temperature over a wide range, above some minimum value (about 1200°K) necessary to remove surface contaminants.$^{35}$

2. Collection Efficiency

Because the collector in our version of the Langmuir-Taylor detector is not optimum in its geometry, a large bias voltage is necessary to obtain complete collection of the Cs$^+$ formed at the hot tungsten wire. Based on the current vs bias voltage plot shown in Chapter V, the Cs$^+$ collection efficiency is estimated at 100%, +0, -5%.

The other consideration with regard to the collector is the possibility of secondary electron emission at the collector surface.
Due to the bias potential there, any electron emitted will certainly be lost, leading to a spurious component in the detected current. Although direct measurements for this particular case are not available, there is experimental evidence to believe the secondary emission coefficient, \( \gamma \), is very small. Waters\(^{36} \) measured \( \gamma \) for 300 eV Cs\(^+\) on clean tungsten to be about \( 10^{-4} \), and this result was approximately confirmed by Petrov.\(^{37} \) Although one would expect from McDaniel's review\(^{38} \) to find \( \gamma \) smaller for surfaces with adsorbed gases (\( \gamma \) is down by a factor of 3 for X\(_e\) on W), Waters found the opposite to be true for Li\(^+\) on W with N\(_2\) or O\(_2\) adsorbed. Even this value is less than .01 at 300 eV, and \( \gamma \) for Li\(^+\) on clean tungsten was larger than for Cs\(^+\). We conclude, therefore, that secondary emission by Cs\(^+\) impact at a maximum energy of 270 eV is a negligible effect in this experiment.

3. Mean Velocity

Since \( \sqrt{\frac{v}{c}} \propto \sqrt{T} \), errors in the oven temperature are of little consequence. The thermocouple and pyrometer were not calibrated absolutely, so the temperature error is taken as \( \pm 10^\circ \text{C} \). This uncertainty leads to at most a 1% error in the mean velocity.

There might be some question whether the velocity distribution for an ideal effusive beam is appropriate, since the cesium flux vs temperature relation is not quite ideal. The deviation from a Maxwell-Boltzmann distribution has been shown by Herschbach\(^{39} \) and co-workers to be pressure dependent; and the quantitative dependence is also a function of the slit width and channel length. The Laval
nozzle used in Herschbach's experiment was in fact remarkably similar in its dimensions to the channel used in this work. They found that as the source pressure increases, the velocity distribution narrows and shifts to higher velocities; but that this effect is negligible in a potassium beam with a source pressure about 0.02 Torr. At 0.2 Torr, the most probable velocity is higher by about 10%. The highest source pressure in our experiments is about 0.015 Torr; therefore, the upper limit to this correction is considered to be less than 1%.

4. Scanning Errors

There are four possible errors to be considered under the heading of scanning errors: calibration, slipping, wire-slit offset, and divergence of the cesium beam. The values for the slit scanner calibration have proved exceedingly repeatible; consequently the maximum error associated with the calibrations obtained is about 2%. Figure 21 indicates the slipping must be smaller than a slit width and no larger than a wire diameter. From these considerations and from repeatability of cesium beam width measurements, any error due to slipping of the scanning arm is estimated to be smaller than 3%. The offset of the hot wire from the center of slit is measured using the telescope as for the scanner calibrations. Although this value is accurate only to within 10% or so, the effect of that uncertainty in calculating the form factor is only 3% for the worst case of a rather broad 0 atom beam.
Finally there is a possible systematic error which arises because the cesium beam diverges through the interaction volume (the angle is approximately 9°). The hot wire measures the cesium beam at the nominal center of the 0 atom beam. If the wire is not at the actual center of the 0 atom beam, or if the 0 atom beam has a vertical distribution other than flat, then there will be an error introduced by the divergence of the cesium beam. The maximum error due to this effect is approximately 3 %.

D. Error in 1D Cross Sections

The cross section for Cs + 0(1D) → Cs⁺ + 0⁻ is dependent on the fraction of 0(1D) in the nitrogen laser detached atom beam. This fraction is determined from the measured photodetachment cross section by extrapolation, and the uncertainty is greater than 10 %. The uncertainty in the 1D cross section is even larger. Since

$$\sigma_N = f\sigma_1 + (1 - f)\sigma_3$$  \hspace{1cm} (38)

where $\sigma_3$ is the cross section for 0(3P) + Cs → 0⁻ + Cs⁺

$\sigma_1$ is the cross section for 0(1D) + Cs → 0⁻ + Cs⁺

$\sigma_N$ is $N_2$ laser cross section

$\sigma_D \equiv \sigma_3$ is Dye laser cross section

f is 1D fraction in atom beam

then

$$\frac{\sigma_1}{\sigma_3} = \frac{1}{f}\left[\frac{\sigma_N}{\sigma_D} + f - 1\right]$$  \hspace{1cm} (39)

and

$$\Delta \left(\frac{\sigma_1}{\sigma_3}\right) = \left(1 - \frac{\sigma_N}{\sigma_D}\right) \frac{\Delta f}{f^2} + \frac{1}{f} \Delta \left(\frac{\sigma_N}{\sigma_D}\right)$$.  \hspace{1cm} (40)
The fractional uncertainty in \( \frac{\sigma_1}{\sigma_3} \) does not take a simple form, but as \( \sigma_1/\sigma_3 \) approaches zero, the fractional uncertainty becomes very large.

E. Summary

The limit of systematic error in the measurements for a given laser is taken to be the linear sum of the errors discussed above. Other than the error in \( \epsilon_1 \), the limit of errors is \( +12, -17 \% \); the limit of errors in \( \epsilon_1 \) ranges from \( \pm 25 \% \) to \( \pm 13 \% \). The total standard (rms sum) systematic error assuming each source of error has a Gaussian distribution is \( \pm 17 \% \) in the worst cases. The random errors vary from subgroup to subgroup; these uncertainties are due to statistical fluctuations in the count rates and in the output of the absolute \( 0^+ \) atom detector. In the next chapter, we shall be concerned with errors in the determination of the relative cross section as a function of energy. Although each measurement is an absolute one, the systematic errors in principle are independent of energy and group. One exception is the assessment of the \( 0^- \) channel efficiency \( \epsilon_1 \). The error in the transmission efficiency is energy (and efficiency) dependent, and the estimate for counting efficiency is a new one for each group. The random contributions to the uncertainty in \( \epsilon_1 \) are estimated to be from 5 to 10 \%, depending on the transmission and the counting efficiency estimate. The total relative error is taken to be the \( \text{rms sum} \) of these random contributions; i.e., the fractional relative error,
\[
\frac{\Delta \sigma}{\sigma}_{\text{rel}} = \left[ \left( \frac{\Delta F_0}{F_0} \right)^2 + \left( \frac{\Delta C}{C} \right)^2 + \left( \frac{\Delta \varepsilon_2}{\varepsilon_2} \right)^2 + \left( \frac{\Delta \varepsilon_1}{\varepsilon_1}_{\text{rel}} \right)^2 \right]^{1/2} \tag{41}
\]

where \( \sigma \) is the electron transfer cross section

\( C \) is the total number of coincidence counts for a subgroup

\[
\left[ \left( \frac{\Delta C}{C} \right)^2 = \frac{1}{C} \right]
\]

\( \frac{\Delta \varepsilon_1}{\varepsilon_1}_{\text{rel}} \) is the random contribution to the uncertainty in \( \varepsilon_1 \).
CHAPTER VII

RESULTS

Although the principal physical results of these experiments are cross sections for electron transfer in oxygen-cesium collisions, there is some additional information obtained. The data at 1 keV allows a small adjustment in the $0^1D$ fraction in the nitrogen laser detached beam. These results are presented first in order to make use of this information in the subsequent cross section reduction. The relative O atom detector data gives unique information on the relative secondary emission coefficients for the $^1D$ and $^3P$ states of the O atoms on a Be-Cu surface.

A. $^1D$ Fraction with $N_2$ Laser

The fraction of neutral atoms in the $^1D$ state resulting from photodetachment at 337.1 nm was estimated from photodetachment data at 0.37 with an uncertainty of $\pm 0.05$. It has been shown that the error in deducing cross sections for $0^1D$ atoms is sensitive to the uncertainty in the $^1D$ fraction in the beam. Some of the data, however, allows a refinement in the determination of this fraction and gives the results some independence from photodetachment data.

Three measurements have been made of the $N_2$ laser/dye laser ratio of cross sections for electron transfer at 1 keV. Cross
sections labeled by the laser used are simply the numbers calculated from data taken with the laser indicated.

Table IV

<table>
<thead>
<tr>
<th>Group</th>
<th>(\sigma(N_2 \text{ laser})/\sigma(\text{Dye laser}))</th>
<th>Standard Error</th>
<th>Implied Ratio for (f = .37)</th>
<th>Implied Ratio for (f = .42)</th>
</tr>
</thead>
<tbody>
<tr>
<td>F</td>
<td>.70</td>
<td>.10</td>
<td>+.19</td>
<td>+.29</td>
</tr>
<tr>
<td>H</td>
<td>.43</td>
<td>.07</td>
<td>-.54</td>
<td>-.36</td>
</tr>
<tr>
<td>I</td>
<td>.54</td>
<td>.08</td>
<td>-.24</td>
<td>-.04</td>
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<tr>
<td>mean ratio</td>
<td>.56</td>
<td>.15</td>
<td>-.19</td>
<td>-.05</td>
</tr>
<tr>
<td>ratio of mean cross sections</td>
<td>.60</td>
<td>.07</td>
<td>-.08</td>
<td>+.05</td>
</tr>
</tbody>
</table>

As the table shows, our data are not consistent with a \(^1D\) fraction of 0.37. Although these data cannot establish an upper limit for the \(^1D\) fraction, neither experimental nor theoretical considerations permit a value much in excess of 0.5. In the following discussion, the value for \(f\) is taken to be \(0.42 \pm 0.05\). Although this value is not very different from the extrapolated one, it allows a more realistic assessment of the \(^1D\) cross sections.

B. Cross Sections

In Table V the cross sections for both lasers are tabulated as functions of both 0 atom energy and subgroup identification. The group labels, Roman letters, identify a single opening of the vacuum system; and the subgroup labels, Arabic numerals, identify sets of
| O Atom Energy (eV) | Laser | Sub-Group | ε<sub>1</sub> | ε<sub>2</sub> | σ<sub>1</sub> (10<sup>-16</sup> cm<sup>2</sup>) | σ<sub>2</sub> (10<sup>-16</sup> cm<sup>2</sup>) | Δσ<sup>b</sup> | S<sub>3</sub>/S<sub>2</sub> | S<sub>0</sub>/S<sub>2</sub> | σ<sup>′</sup> (10<sup>-16</sup> cm<sup>2</sup>) | Remarks |
|-------------------|-------|-----------|-------------|------------|---------------------------------|---------------------------------|----------|----------------|----------------|-----------------|-----------|------------|
| 200               | Dye   | K2        | .72         | .82        | 9.5                                            | 1.9                                            | 1.03     | .95            | .3             |                 |           |            |
|                   |       | K1        | .75         | .82        | 9.8                                            | 1.3                                            | 1.37     | 1.04           | 3.6            |                 |           |            |
|                   |       | L5        | .6          | .7         | 8.9                                            | 1.9                                            | .99      | 1.03           | <0             |                 |           |            |
|                   |       | L6        | .59         | .82        | 7.3                                            | 0.9                                            | .96      | .88            | <0             |                 |           |            |
|                   |       | L7        | .59         | .82        | 7.0                                            | 0.8                                            | 1.07     | 1.06           | .5             |                 |           |            |
|                   |       | L8        | .59         | .82        | 7.1                                            | 0.8                                            | 1.06     | 1.04           | .4             |                 |           |            |
| 300               | Dye   | I3        | .68         | .82        | 15.2                                           | 2.3                                            | 1.09     | 1.06           | 1.4            |                 |           |            |
|                   |       | I1        | .59         | .82        | 18.8                                           | 7.1                                            | 1.16     | 1.04           | 3.0            |                 |           |            |
|                   |       | I2        | .67         | .82        | 22.9                                           | 8.5                                            | 1.11     | 1.04           | 2.5            |                 |           |            |
|                   |       | L1        | .7          | .82        | 13.3                                           | 1.5                                            | 1.12     | 1.07           | 1.6            |                 |           |            |
|                   |       | L2        | .7          | .5         | 10.9                                           | 2.0                                            | 1.02     | .97            | 0.2            | ε<sub>2</sub> unexplained |           |            |
|                   |       | M         | .65         | .8         | 19.6                                           | 2.5                                            | 1.00     | 1.00           | 0              | note a |           |            |
| 500               | Dye   | G3        | .68         | .57        | 9.5                                            | 2.1                                            | 1.15     | .91            | 1.4            |                 |           |            |
|                   |       | G1        | .67         | .57        | 9.6                                            | 1.4                                            | 1.56     | .90            | 5.4            |                 |           |            |
|                   |       | G2        | .68         | .57        | 11.3                                           | 1.7                                            | 1.10     | .99            | 1.1            |                 |           |            |
|                   |       | L3        | .58         | .9         | 8.7                                            | 0.9                                            | 1.16     | .99            | 1.4            |                 |           |            |
|                   |       | L4        | .59         | .9         | 9.9                                            | 1.2                                            | 1.02     | 1.02           | 0.2            |                 |           |            |
| 1000              | Dye   | F3        | .81         | .62        | 13.0                                           | 2.0                                            | 1.25     | .89            | 3.2            |                 |           |            |
|                   |       | H5        | .71         | .62        | 16.7                                           | 4.5                                            | 1.15     | 1.16           | 2.5            |                 |           |            |
|                   |       | I4        | .66         | .82        | 13.4                                           | 1.3                                            | 1.00     | .90            | 0              |                 |           |            |
|                   |       | E1        | .6          | .46        | 9.6                                            | 2.0                                            | 1.29     | 1.12           | 2.8            |                 |           |            |
|                   |       | E2        | .6          | .46        | 8.7                                            | 1.8                                            | 1.12     | 1.02           | 1.0            |                 |           |            |

---

*Note: *Excessive signal and noise are indicated in the Remarks column.
Table V (continued)

<table>
<thead>
<tr>
<th>O Atom Energy (eV)</th>
<th>Laser</th>
<th>Sub-Group</th>
<th>$\varepsilon_1$</th>
<th>$\varepsilon_2$</th>
<th>$\sigma$ (10$^{-16}$ cm$^2$)</th>
<th>$\Delta_0$</th>
<th>$\sigma_{3/2}$ (10$^{-16}$ cm$^2$)</th>
<th>$\sigma_{0/2}$ (10$^{-16}$ cm$^2$)</th>
<th>Remarks</th>
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<td>1000</td>
<td>N$_2$</td>
<td>F1</td>
<td>.87</td>
<td>.62</td>
<td>9.3</td>
<td>1.7</td>
<td>1.18</td>
<td>.93</td>
<td>1.7</td>
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<tr>
<td></td>
<td></td>
<td>F2</td>
<td>.87</td>
<td>.62</td>
<td>8.6</td>
<td>1.5</td>
<td>1.17</td>
<td>1.10</td>
<td>1.5</td>
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<td>1.7</td>
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<td>.89</td>
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<td></td>
<td></td>
<td>G4</td>
<td>.73</td>
<td>.50</td>
<td>10.1</td>
<td>4.1</td>
<td>1.38</td>
<td>1.01</td>
<td>3.9</td>
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<td></td>
<td></td>
<td>H4</td>
<td>.73</td>
<td>.62</td>
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<td>.82</td>
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<td>1.13</td>
<td>.99</td>
<td>0.8</td>
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<td></td>
<td></td>
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<td>.82</td>
<td>7.7</td>
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<td>1.16</td>
<td>.95</td>
<td>1.2</td>
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<td>Dye</td>
<td>H2</td>
<td>.75</td>
<td>.62</td>
<td>11.3</td>
<td>1.4</td>
<td>1.01</td>
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<td>.82</td>
<td>10.8</td>
<td>2.2</td>
<td>1.00</td>
<td>.85</td>
<td>0</td>
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<td>J3</td>
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<td>.82</td>
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<td>1.08</td>
<td>1.01</td>
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<td></td>
<td></td>
<td>J4</td>
<td>.71</td>
<td>.82</td>
<td>10.4</td>
<td>2.1</td>
<td>1.10</td>
<td>1.05</td>
<td>1.0</td>
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<tr>
<td></td>
<td></td>
<td>H1</td>
<td>.4</td>
<td>.62</td>
<td>9.4</td>
<td>2.2</td>
<td>1.01</td>
<td>1.00</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>N$_2$</td>
<td>H3</td>
<td>.74</td>
<td>.62</td>
<td>10.6</td>
<td>1.9</td>
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<td>.82</td>
<td>8.7</td>
<td>1.7</td>
<td>1.12</td>
<td>1.01</td>
<td>1.0</td>
</tr>
</tbody>
</table>

a The form factor and cesium density were not measured due to apparatus failure, but estimated from a new measurement of the $O^-$ ionization of cesium rate; other estimates for the cesium density give lower cross sections.

b The absolute error in these cross sections is the (rms) sum of the relative cross tabulated and the systematic error: ± 17%.
data runs taken under similar experimental conditions. All data is presented for which absolute cross sections could be computed. Relative errors in the electron transfer cross section (Δσ) are included in the tabulation, where relative error is taken to be the standard error of each measurement relative to those from other groups (refer to Chapter VI); whereas, the absolute error includes those errors and uncertainties which apply equally to the entire set of data. The relative errors arise mostly from statistical and 0 atom uncertainties. The electron transfer cross section, σ, is taken from the coincidence channel data. The ratio $S_0 / S_2$ is the ratio of the event rate deduced from the $0^-$ channel to that deduced from the coincidence channel. Since both channels detect only electron transfer events, this ratio should be unity. The cross section for $0 + Cs \rightarrow 0 + Cs^+ + e^-$ is deduced by comparing the event rate in the $Cs^+$ channel ($S_3$) with that in the coincidence channel ($S_2$). $S_3$ is the sum $S_1 + S_2$ where $S_1$ is the rate for cesium ionization without electron attachment to 0 (as before).

Ratios of cross sections obtained from nitrogen and dye laser runs are computed from data belonging to a single group, to minimize relative errors. These ratios are tabulated below as a function of energy:

### Table VI

<table>
<thead>
<tr>
<th>Energy (eV)</th>
<th>$\sigma(N_2)/\sigma(Dye)$</th>
<th>Standard Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>200</td>
<td>1.02</td>
<td>.23</td>
</tr>
<tr>
<td>300</td>
<td>1.37</td>
<td>.43</td>
</tr>
<tr>
<td>500</td>
<td>1.16</td>
<td>.15</td>
</tr>
<tr>
<td>1000</td>
<td>.56 (mean)</td>
<td>.15</td>
</tr>
<tr>
<td>1800</td>
<td>.88 (mean)</td>
<td>.23</td>
</tr>
</tbody>
</table>
The absolute cross sections for the nitrogen and dye lasers are plotted vs energy in Figure 29. Data from the same group are averaged for presentation in this figure; and the error bars represent the relative error only. The dye laser cross section is of course the cross section for \( O(3P) \) atoms only. Figure 30 shows the mean \( O(3P) \) and \( O(1D) \) cross sections as a function of \( O \) atom velocity. The \( 1D \) cross section plotted here assumes the fraction 0.42 obtained earlier and the cross section ratios from Table VI. The error bars in this figure include the relative errors, and for the \( (1D) \) points, the uncertainty in \( 1D \) fraction. In Figure 31, these data are shown again on a logarithmic scale together with theoretical calculation of van den Bos.\(^7\) In this case we have indicated the total standard error (rms uncertainty) in the absolute cross sections.

As indicated in Table V, our data indicates, with a large degree of uncertainty, a non-zero cross section for the reaction

\[
O + Cs \rightarrow O + Cs^+ + e^-;
\]

(42)

however, the uncertainties are sufficiently large to make the difference between \( 1D \) and \( 3P \) contributions undetectable. These results are also given in Figure 31 with estimated errors. Because of their small magnitude, these cross sections are extremely sensitive to the absolute detection efficiency in the \( O^- \) channel, as well as to statistical uncertainties.
Figure 29. Absolute cross sections for electron transfer events as a function of O atom energy. The error bars represent one standard deviation on the relative error.
Figure 30. Absolute cross sections for electron transfer events as a function of O atom velocity. Relative errors are indicated.
Figure 31. Comparison of theoretical and experimental results for the electron transfer cross sections, and the experimental results of the cesium ionization reaction, $O + Cs \rightarrow O + Cs^+ + e^-$. The total errors in the absolute cross sections are indicated.
C. Discussion

The discrepancy between the Landau-Zener-Stueckelberg approximation calculation and these results is a factor of about 4. Since the total standard error (systematic + random) is about 30%, the discrepancy is much greater than 3 standard deviations. One explanation for this difference may be the effect of competing outgoing channels which were not considered in the calculation.

The other features of the data are less well established; but, there is a strong suggestion that the cross section functional dependence is considerably sharper than that indicated by the calculated result. The cross sections for $^1D$ atoms and $^3P$ atoms at 1 keV are definitely different. At other energies, the $^1D$ cross section is much more uncertain. The nitrogen laser/dye laser cross section ratio at 300 eV has been taken to be that indicated by the Group I measurements of both cross sections. These data are supported by a subsequent measurement of the relative O atom detector response to the dye and nitrogen laser O atom fluxes, but this recalibration was conducted under difficult conditions and may not be reliable. The results shown in Figure 30 also serve to establish bounds for the $^1D$ cross section, which from a crude picture might be expected to be much larger than the cross section for ground state atoms.

It is noteworthy that the cross sections for ionization of cesium without electron attachment to the O atom are almost negligible compared with the electron transfer cross section; indeed, the uncertainties in the $^1D$ channel efficiency $\varepsilon_1$, are great enough
that the total cross section for ionization of cesium by O atom impact may be entirely due to electron transfer.

D. Secondary Electron Yield on Be–Cu Surfaces

The calibrations of the relative O atom detector (multiplier) yield unique information on the relative secondary electron yield, \( \gamma \), of beryllium–copper surfaces as a function of the state of the O atom for each of the energies used. There is also some data available on the yield as a function of energy, but it is very sketchy, as one cannot rely on the multiplier efficiency and gain to remain constant from one opening to the next. Qualitatively, the secondary electron emission increases with increasing O atom energy, as expected. Table VII shows the ratio of electron yields for \( ^1D \) and \( ^3P \) oxygen atoms on the Be–Cu surface of an EMI 9603 B venetian blind multiplier as a function of atom energy. In Figure 32, the quantity \( [\gamma(N_2)/\gamma(Dye) - 1] \) is plotted as a function of O atom velocity. This quantity is chosen since it is more closely related to the raw data than the ratio \( \gamma(^1D)/\gamma(^3P) \). The values shown here and in Table VII are weighted means, where the weighting factors are inversely proportional to the random errors in the absolute O atom number.

<table>
<thead>
<tr>
<th>0 Atom Energy (eV)</th>
<th>( \frac{\gamma(N_2 \text{ Laser})}{\gamma(Dye \text{ Laser})} )</th>
<th>Standard Error</th>
<th>( \frac{\gamma(^1D)}{\gamma(^3P)} )</th>
<th>Standard Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>200</td>
<td>3.1</td>
<td>.6</td>
<td>6.0</td>
<td>.8</td>
</tr>
<tr>
<td>300</td>
<td>1.9</td>
<td>.4</td>
<td>3.1</td>
<td>.7</td>
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<td>500</td>
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<tr>
<td>1000</td>
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<td>.17</td>
<td>1.1</td>
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<tr>
<td>1800</td>
<td>1.00</td>
<td>.23</td>
<td>1.0</td>
<td>.6</td>
</tr>
</tbody>
</table>
\[
\frac{\gamma N_2}{\gamma \text{DYE}} - 1 = f \left( \frac{\gamma 1 - \gamma 3}{\gamma 3} \right)
\]

Figure 32. Velocity dependence of the secondary electron yield enhancement obtained with nitrogen laser detached O atoms over that obtained with dye laser detached O atoms. The surface is the first Be-Cu dynode of a 15 stage venetian blind multiplier; it is not clean.
CHAPTER VIII

IONIZATION OF CESIUM IN COLLISIONS WITH NEGATIVE IONS

Earlier we mentioned the use of a small $^0_-$ beam to provide a continuous source of $\text{Cs}^+$ ions. It represents only a small additional effort to measure the absolute cross section for the process

$$\text{Cs} + X^- \rightarrow \text{Cs}^+ + \text{products} \quad (43)$$

where $X^-$ is usually $^{18}_0^-$, but in general any atomic or molecular negative ion. The total cross section for cesium ionization by $^0_-$ impact has been measured as a function of energy from 300 eV to 1.8 keV. In addition we measured the cesium ionization cross section for various impurity negative ions from $^1_1H^-$ to $^{35}_{17}Cl^-$ at an ion energy of 1 keV.

A. Description of Experiment

In general the experimental arrangement is the same as the one described for 0 atom collisions with cesium, the principal exception being that the $^0_-$ beam continues straight into the interaction region. It is measured periodically at the shielded Faraday cup by deflecting it up in the usual way. It is necessary to use an ion beam of the order of 100 pA in order to avoid count rate saturation in the electronics when interacting with a cesium beam of the same magnitude used in the 0 atom collision experiments. The signal-to-
noise ratio for this experiment is so great that neither pulsed nor ac modulation techniques are necessary. Typically the signal-to-noise ratio is of the order of 20. The background counting rates are obtained by summing the rates for the two cases of one beam on and one off, and subtracting the rate obtained with both beams off.

The cross section as a function of measured parameters is given by

\[ \sigma = 0.944 \times 10^{-18} \frac{S\sqrt{T}}{I_o F(1) \epsilon_2} \]  

where \( S \) is the number of signal counts per second

\( I_o \) is the negative ion current in pA

\( F(1) = \int I_c dz \); the numerical factor relating this quantity to \( F_c \) is contained in the coefficient and the other parameters as previously defined. We have made the same relative velocity approximation as before. The cesium profile and form factor are measured in the same way, except that the relative 0 atom detector multiplier is operated as a Faraday cup for the \( 0^- \) beam. The value for \( \epsilon_2 \) is obtained from the 0 atom-cesium experiment.

B. Theoretical Considerations

To our knowledge, there are no calculations relating to this particular class of problems. Figure 33 gives the simplified potential curve diagram analogous to Figure 5. It should be remarked that the processes leading to \( 0^- + Cs^+ \) via the \( 0^- - Cs^+ \) curve involve ionization of the \( Cs \) atom followed by recombination with that electron, which is probably unlikely. In other respects the potential map is similar to that for \( 0^+ + Cs \). On these grounds
Figure 33. Non-adiabatic Coulomb potentials for the $O^-$ – Cs systems, showing some of the excited state levels.
alone, it seems plausible to expect a cross section of the same order as that for the electron transfer process in the neutrals.

C. Discussion of Results

1. Consistency Checks

Figures 34 and 35 show the linearity of event rate with $0^-$ current and with cesium column density. The $0^-$ linearity data is taken at a fixed cesium beam density by attenuating the $0^-$ beam at the mass spectrometer slit. There is no correction for possible changes in form factor, which were not investigated. The data in Figure 35 are normalized to 50 pA $0^-$ current, and the currents actually used were not different by more than a factor of 2.

2. Results

Figure 36 gives the measured absolute cross section for ionization of cesium by $0^-$ ions as a function of energy. These cross sections are smaller by a factor of 2 to 3 than the total cross section for cesium ionization by O atom impact. In Figure 37, both cross sections are shown. In this figure the bars show the errors in absolute values. We take these data as evidence that the ionization of cesium atoms is sensitive to the details of the electronic interaction between the colliding species.

The data shown in Table VIII are the cesium ionization cross sections for collision with the various negative ions present when operating the source normally for $0^-$ at an energy of 1 keV. The beam current was attenuated at two levels for these data and the form
Figure 34. Demonstration of the linearity of Cs\(^+\) signal rate with \(O^-\) current.
Figure 35. Demonstration of the linearity of Cs$^+$ signal rate with cesium column density.
Figure 36. Total cross section for ionization of cesium by $O^-$ impact.
Figure 37. Total cross sections for ionization of cesium by O atom and by O⁺ ion impact. Total errors are indicated.
Table VIII
Cesium Ionization Cross Sections at 1 keV

<table>
<thead>
<tr>
<th>Mass No.</th>
<th>Ion</th>
<th>$\sigma\left(10^{-16} \text{ cm}^2\right)$</th>
<th>Standard Error $\left(10^{-16} \text{ cm}^2\right)$</th>
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<td>1</td>
<td>H$^-$</td>
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<td>12</td>
<td>C$^-$</td>
<td>2.21</td>
<td>.15</td>
</tr>
<tr>
<td>13</td>
<td>$^{13}$C$^-$</td>
<td>1.68</td>
<td>.84</td>
</tr>
<tr>
<td>17</td>
<td>OH$^-$</td>
<td>4.30</td>
<td>.34</td>
</tr>
<tr>
<td>18</td>
<td>$^{18}$O$^-$</td>
<td>3.70</td>
<td>.26</td>
</tr>
<tr>
<td>19</td>
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<td>2.59</td>
<td>.18</td>
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</table>

factor measured for each attenuation. The species $^{16}$O$^-$ was not attempted in this series, since the cross section had previously been established to be the same as for $^{18}$O$^-$ within the limits of error. These experiments were plagued by rather high background counting levels due to corona induced by the Cs$^+$ lens 2 voltage. The errors listed are the random errors. The systematic errors are considered later.
3. Relative Cs⁺ Channel Efficiency ($\varepsilon_2$)

With the signal rate and the signal-to-noise ratio both high, one would expect minimal random errors. It should be possible to exploit this circumstance to establish an excellent relative scale of $\varepsilon_2$'s for differing sets of experimental conditions. (Even if all data were taken under identical conditions, there can still be a decrease in multiplier sensitivity and gain each time the vacuum system is opened.) Figure 38 is a plot of the absolute quantity that is measured in these experiments, $\varepsilon_2^\sigma$, as a function of the $\varepsilon_2$ obtained from the nearest O atom-Cs data run. These $\varepsilon_2^\sigma$'s are for 1 keV $0^-$ ions (in most cases $18^0$). The variation in $\varepsilon_2$ is understood in terms of specific modifications in either apparatus or procedure designed to enhance the Cs⁺ channel efficiency. The uncertainty in $\varepsilon_2$ is due to the random error associated with the signal to noise ratio in the $0^-$ channel (refer to Chapter IV for details on the calculation of $\varepsilon_2$ from data).

One would hope to find a least squares fit to a line passing through the origin, which one could use as a more accurate calibration for $\varepsilon_2$. It is apparent that the scatter around such a line is greater than expected; in addition, some of the scattered data plotted at the same $\varepsilon_2$ was taken under nominally the same conditions. We conclude from these observations that, although the $0^-$ - Cs experiment provides a qualitative corroboration of the $\varepsilon_2$'s obtained from the atom-atom experiment, there are (random) fluctuations in this efficiency which are uncorrelated with the nominal parameters. Causes for these fluctuations include $0^-$ beam
Figure 38. Demonstration of the linearity of the measured quantity $\epsilon_2^0$ with the value for $\epsilon_2$ obtained from the O atom - Cs experiment.
displacements, non-uniformities in the cesium beam, and fluctuations in cesium beam column density. This last factor is attributed to a partial block of the oven or collimating slit by cesium metal moved by the beam flag. This effect has been observed in these experiments, and data known to be unreliable has been eliminated. The fluctuations due to these factors in apparent \( \varepsilon_2^\sigma \)'s are of the same order as the random errors associated with \( \varepsilon_2 \)'s obtained from the 0–Cs data, so it is not in fact possible to reduce their uncertainties.

4. Errors

The limit of systematic errors in measuring \( \varepsilon_2^\sigma \) is estimated to be +16, -21 \%. The random errors in that measurement are generally negligible. The error in \( \varepsilon_2 \) is again estimated to be 3 to 5 \%. The standard error (rms) in the total absolute ionization cross section is then just 9 \%. 
CHAPTER IX

CONCLUSIONS AND SUGGESTIONS FOR FURTHER EXPERIMENTS

These experiments are among the first to provide information on atomic collision cross sections as a function of the electronic states of the participating atoms. With appropriate modifications in the original design, it has been a straightforward procedure to measure reliable cross sections, even at low signal rates. With additional modifications, it will be feasible to collect data at faster rates, thus overcoming the limitations imposed by the finite source life of both the negative ion and atomic alkali beams. These modifications are lasers capable of faster repetition rates and some automatic data recording system. The repetition rate limitation is not a fundamental one, even with the dye laser, because of the feasibility of pumping dyes with the $N_2$ laser. Although not all of the suggested experiments depend on these improvements, they become more attractive if they can be done more quickly.

A. Collision Processes

It would be of considerable interest to extend these studies to lower energies. The present limitations in energy range are imposed by the apparatus, but they are not insuperable with appropriate design changes. There are, however, a number of experiments which can be done with little or no change in the vacuum apparatus. Other
alkali metals could be investigated, although there begin to be
detection problems for those metals lighter than rubidium. Of more
interest might be the study of electron transfer and cesium ion-
ization processes for a variety of fast atoms. In particular sulfur,
in the outer shell, is isoelectronic with oxygen; and the $^1D$ state
is also obtainable by photodetachment with the $N_2$ laser. It is
also possible to obtain large currents of $H^-$, which has a photo-
detachment cross section maximum at 850 nm.\(^{40}\) It is not difficult
to prepare large currents of the halogen negative ions, all of which
have electron affinities smaller than the $N_2$ laser photon energy.
No state selection is possible for these atoms but a system such as
Cs + F might be more tractable theoretically.

Another experiment which suggests itself is to look for cesium
resonance radiation from the oxygen-cesium collision. The potential
diagram suggests that excited cesium atoms may be produced in large
quantities. Because the detection efficiency would be much lower
for photons, it might be useful to try first to see $\text{Cs} + 0^- \rightarrow$
Cs ($^2P$) + 0 + e\(^-\), where a larger count rate can be obtained.

B. Photodetachment Studies

The advent of the tunable dye laser makes possible a thorough
investigation of the structure of negative ions and of electron
affinities for a large number of species. This organic dye laser
has been tuned using a blazed diffraction grating at one end of the
optical cavity. Line widths (FWHM) of about .1 nm are regularly
obtained with the grating; the output power is wavelength dependent,
but much larger than the ratio of line width to the broadband line-
width.

Since the photodetachment cross section just above threshold
is proportional to $E^{1/2}$ for detachment of $p$ electrons,\textsuperscript{41} the slope
of a $\sigma$ vs $E$ plot is infinite at threshold, giving a clear in-
dication when additional states participate. We have obtained pre-
liminary data on the relative photodetachment cross section of $S^-$
near threshold, as shown in Figure 39. The $S^-$ state is expected
to be a $^2P$ state, analogous to the ground state of $Cl$. We measure
the threshold and electron affinity photon energies, as well as
other thresholds corresponding to detachment into different final
states of the atom. The difference between the threshold and elec-
tron affinity is the fine structure splitting of the $S^-$ state,
$482 \pm 5$ cm\textsuperscript{-1}, the best such measurement obtained for any negative
ion. With the prospect of several dyes extending the wavelength
range of tunable lasers, many other atomic and molecular ions lend
themselves to such measurements. In particular we suggest the study
of $O^-$ both at threshold and at the $^1D$ threshold, as well as
continued study of the $S^-$ problem, which is not yet fully resolved.
It would also be of interest to study negative ion states of dif-
ferent multiplicities. Although more difficult, it is also appro-
priate to study threshold behavior in those negative ions which have
$s$ and $d$ electrons in the outer shell. These systems should both
exhibit $E^{3/2}$ threshold laws, and it would be of interest to compare
the two cases.
Figure 39. Relative cross section for photodetachment of $S^{-}$ as a function of photon energy. Not all of the measured values are shown due to the scale of the figure.
It should be pointed out that the high signal-to-noise ratio in these experiments makes possible their execution with much smaller ion beams than ever before. Especially if the detailed structure of the ion is not required, photodetachment threshold experiments can be conducted with very small ion currents, such as those characteristic of impurity ions in the source. Suppose a laser is bright enough to detach only 1 ion in 1000 over a path length of 1 cm in the beam. Then to get 1 count in 10 laser flashes, we require about $10^2$ ions per cm. If the ion velocity is $6 \times 10^6$ cm/sec, one needs only 100 pA. With faster repetition rates, one could afford to count more slowly, dropping the minimum ion current to perhaps 10 pA. It appears to be almost trivial to obtain ion beams of the order of 100 pA by admitting small quantities of impurities to the source. The following table is a partial list of ions whose electron affinities could probably be determined with a tunable dye laser operating between 700 and 350 nm:
### Table IX

Candidates for Electron Affinity Measurements

<table>
<thead>
<tr>
<th>Ion</th>
<th>Approximate Electron Affinity, 42,43 A(eV)</th>
<th>Excited Atomic States</th>
<th>Probable Orbital</th>
</tr>
</thead>
<tbody>
<tr>
<td>Li⁻</td>
<td>.7</td>
<td>²p</td>
<td>s</td>
</tr>
<tr>
<td>C⁻</td>
<td>1.25</td>
<td>¹d</td>
<td>p</td>
</tr>
<tr>
<td>F⁻</td>
<td>3.45</td>
<td></td>
<td>p</td>
</tr>
<tr>
<td>Na⁻</td>
<td>.5</td>
<td>²p</td>
<td>s</td>
</tr>
<tr>
<td>Si⁻</td>
<td>1.4</td>
<td>¹d</td>
<td>p</td>
</tr>
<tr>
<td></td>
<td></td>
<td>¹s</td>
<td></td>
</tr>
<tr>
<td>P⁻</td>
<td>.9</td>
<td>²d</td>
<td>p</td>
</tr>
<tr>
<td></td>
<td></td>
<td>²p</td>
<td></td>
</tr>
<tr>
<td>Cr⁻</td>
<td></td>
<td>⁵s</td>
<td>.94</td>
</tr>
<tr>
<td></td>
<td></td>
<td>⁵d</td>
<td>.96–1.03</td>
</tr>
<tr>
<td>Cu⁻</td>
<td></td>
<td>²d</td>
<td>1.39–1.64</td>
</tr>
<tr>
<td>Br⁻</td>
<td>3.36</td>
<td></td>
<td>p</td>
</tr>
<tr>
<td>I⁻</td>
<td>3.06</td>
<td></td>
<td>p</td>
</tr>
<tr>
<td>SH⁻</td>
<td>2.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>C₂⁻</td>
<td>3.1–4.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>SCN⁻</td>
<td>2.2</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
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APPENDIX A

NITROGEN LASER

1. Design Details

Figure 40 is a complete circuit diagram for the switching system of the nitrogen laser used in these experiments. A few points are worth noting. Although the instantaneous current and power ratings for the EGG model 1802 hydrogen thyratron are exceeded in this application, these ratings are pulse length dependent and were established assuming longer "on" times. The same thyratron was used for a number of shots approaching $10^8$ before failure. In that time the optimum reservoir current was increased by a most 5%. The failure mode was an inability to hold off an anode potential $\geq 5$ kV. By comparison we have had only limited success in using the EGG pressurized spark gaps. The spark gaps have a repetition rate limitation of at most 20, have greater jitter, are more noisy electrically, and require high voltage trigger pulses, in themselves difficult to generate in an optimum way. In addition, the spark gaps have a more limited life, although our experience is inconsistent in the actual values.

One other point regarding the hydrogen thyratrons is of considerable practical importance. When the tube fires, the grid potential rises far off ground (at least to $\pm 5$ kV). This fact makes necessary the high voltage rectifier shown in Figure 40 as well as attention to insulation of the grid and the lead to it.
Figure 40. Switching circuit used for the pulsed nitrogen laser.
The discharge cables from the capacitor to laser channel are twenty-nine 180 cm segments of YK - 217 coaxial cable, manufactured by the Belden Co. The parameters of this cable are given in Table X below:

Table X

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capacitance</td>
<td>~360 pF/m</td>
</tr>
<tr>
<td>Inductance</td>
<td>~95 nH/m</td>
</tr>
<tr>
<td>Characteristic Impedance</td>
<td>~17 $\Omega$</td>
</tr>
<tr>
<td>Breakdown voltage</td>
<td>50 kV (1 min. test; actual value not determined.)</td>
</tr>
</tbody>
</table>

The reasons for selecting this cable become clear in the next section; and the paper of M. Geller et al.\textsuperscript{24} discusses a theoretical model explaining the observed increase in laser performance with cable parameters. In the equivalent transmission circuit for the electrical discharge, the simplest lumped parameter model would predict that the rise time of the output voltage pulse would be proportional to the sum of the lumped inductances. We have found, however, that the performance improves linearly with the number of coaxial cables forming the transmission line even when the inductance of the cables is negligible with respect to that of the switch and capacitor (see Fig. 44).

Two basic types of discharge channels have been studied; however, the comparison between the two is still not firmly established. The
"narrow" channel geometry is basically that due to Avco Everett researchers$^{22,23}$ and has been studied in this laboratory with both plexiglass (lucite) and glass as the side wall material (see Fig. 41). In addition, we have studied minimum inductance configurations, and a number of the laser parameter tests were conducted using a discharge channel of this design. The discharge is confined to a volume very near one or both side walls of the channel; in the version shown in Fig. 41, it will be only on the lower plate, presumably because that is the lowest inductance path. This feature has both advantages and disadvantages. The advantage is that the volume of excited molecules is well defined; a fact which is at least in part offset by the possibility that deviations from flatness in the

Figure 41. Example of the narrow channel (flat) N$_2$ laser (cross section view).
wall surface may prohibit a substantial fraction of the excited molecules from participating in stimulated emission.

The other type of discharge channel, the "round" tube, is adapted from a design by EGG Nuclear (Santa Barbara). In this configuration, the walls are far removed from the electrical discharge (see Fig. 42). A larger voltage across this gap is required to form a discharge in the tube than in the narrow channel design; and it is clear that the output power per pulse is smaller if a glass plate is inserted down the tube. It might be that the rise time of the current pulse through the nitrogen is smaller when the breakdown voltage is higher. This type of laser is the one used in the experiments reported here, largely due to the more straightforward assembly associated with the cylindrical symmetry. We have

Figure 42. Example of the round channel \( N_2 \) laser (cross section view).
introduced modular design into the current versions of these tubes. The output window, mirror, and gas ports are constructed in modules which seal on both ends of a discharge module containing the electrodes. We have studied a variety of end configurations including a Brewster window, with minimum effort because of this modular construction. In addition, there are two discharge modules available, one 74 cm long, and one 147 cm long. No clear difference in performance can be found between the two different length lasers.

2. Performance Data

In this section we discuss the ultra-violet output of the N₂ laser, with the principal emphasis on the functional dependence of output power with various parameters. In most cases these data are presented without an attempt at interpretation. The output light pulse has been measured as a function of time with an ITT planar photodiode, type 4000 with S5 spectral response, and a Tektronix oscilloscope, model 519. This combination has a rise time of about ½ nsec. The temporal shape of the light pulse is approximately a skewed Gaussian with a duration of about 10 nsec at the 20% points. The spectral distribution from one of the lasers is shown in Fig. 44.

Absolute power measurements are made with this detector and with an Eppley thermopile having a response time of about 2.4 sec. At repetition rates above 2 or 3 per second, the thermopile makes a relatively good measurement of the average power delivered. The comparison between the two detectors is within a factor of 2 of
Figure 43. Output spectrum from a small $N_2$ laser of the round channel type.
Figure 44. $N_2$ laser output power as a function of measured capacitance in bank.
Figure 45. \( \text{N}_2 \) laser output power as a function of number and type of coaxial cables.
Figure 46. $N_2$ laser output power as a function of applied voltage.
Figure 47. $N_2$ laser output power as a function of $N_2$ pressure.
agreement. Generally the thermopile does not detect the entire 
laser beam, but only a segment of a line focused beam. This geo-
metrical arrangement permits a reliable estimate to be made of the 
fraction of the beam being measured. Typical outputs are in the 
range from 2 to 4 mJ per pulse in the final configurations.

3. Infra-Red Output

The existence of laser action in the first positive system 
(B \( ^3\Pi \rightarrow A \ ^3\Sigma^+ \)) of the nitrogen molecule has been reported previ-
ously;\(^{45,46}\) however, the power obtainable from the version developed in 
this laboratory is much greater than previously reported. It is not 
yet possible to make a complete report on this laser transition, so 
these remarks should be regarded only as preliminary information.

The usual 2\(^{nd}\) positive system output at 337.1 nm is denoted UV; the first positive system output in the infra-red is denoted IR. 
The IR output is observed in the round lasers whenever two mirrors 
are aligned with the electrical discharge of the laser and parallel 
with each other. Even the reflections from the output window are 
sufficient to produce gain in some of the IR transitions. The 
data reported here are obtained from cavities with losses of 
approximately 5%.

Absolute power levels in the infra-red have been measured only 
for the sum of all IR lines. As much as 30 mJ per pulse inside 
the cavity has been observed in these lines; this figure is inde-
pendent of the type of coaxial cables used, whereas the UV power 
increased by almost a factor of 3 in changing from RG-8 to YK-217.
Figure 48 gives the temporal characteristics of both the UV and IR lines, measured with an S-1 photodiode and a Tektronix 547 oscilloscope, which has a risetime of 7 nsec. The first spike is clearly identified with the UV lines, since it disappears when a UV filter is inserted in the optical path, while the second feature is unchanged.

---

Figure 48. Temporal differences in UV and IR outputs from the N₂ laser.
A limited amount of spectroscopy has been performed in this system, using a Jarrell-Ash ½ Meter monochromater. The relative intensities of the IR lines are voltage and pressure dependent, but the greatest total IR power occurs at very nearly the optimum place (in voltage - pressure space) for the UV laser output. Not all of the lines are even present under different sets of operating conditions. Table XI lists the most prominent wavelengths observed when the IR power is near maximum. It is noted that the strongest lines observed in the IR are of the same electronic and vibrational state as the terminal state for the 337.1 nm UV transitions.

Table XI

Nitrogen Laser Infra-Red Output

<table>
<thead>
<tr>
<th>( \lambda \text{ (approx)} ) (nm)</th>
<th>( \Delta \lambda \text{ (FWHM)} ) (nm)</th>
<th>( \text{N}_2 ) \text{ Band}</th>
<th>Relative Intensity</th>
</tr>
</thead>
<tbody>
<tr>
<td>770</td>
<td>20</td>
<td>2-0</td>
<td>1</td>
</tr>
<tr>
<td>870</td>
<td>6</td>
<td>2-1</td>
<td>9</td>
</tr>
<tr>
<td>880</td>
<td>6</td>
<td>1-0</td>
<td>9</td>
</tr>
<tr>
<td>1050</td>
<td>6</td>
<td>0-0</td>
<td>10</td>
</tr>
<tr>
<td>1230</td>
<td>20</td>
<td>0-1</td>
<td>3</td>
</tr>
</tbody>
</table>

\[ ^a \text{ The band assignments are based on identifications given in Reference 46. } \]
APPENDIX B

ION SOURCE

This section is meant to be a brief review of operational characteristics of the hot cathode ion source used in these experiments. The working gas was carbon monoxide (and carbonyl sulfide for the $S^-$ photodetachment); the figures for source parameters may be somewhat dependent on the gas used, but were the same for CO and COS.

The voltage across the arc (cathode to anode) can range from 150 to 450 V, but the lower voltages are preferred. At about 200 V across the arc, the beam is most stable, and the beam current can remain constant for as long as 10 hours. This stability has never been observed at the higher operating voltages; however, the current available is generally much larger at 400 - 450 V. In excess of 200 nA has been observed at 450 V; whereas, at 200 V the largest focused ion beam is about 80 nA. In addition to stability, the lifetime of the source is much greater when operated at 200 V instead of 400 - 450 V.

The pressure in the source is not measured, but the pressure at the diffusion pump is monitored by an ion gauge. Maximum beam currents are obtained when the ion gauge pressure is between 10 and $30 \times 10^{-6}$ Torr. The pressure dependence is a function of other source parameters and is not well defined. Current peaks have been
found throughout this range without an obvious correlation with other parameters; occasionally, the distribution has more than one peak.

The third and final adjustable parameter in the ion source is the filament emission current. This current is supplied by the arc voltage power supply but is dependent on the filament temperature. A value of 50 mA is typical for optimum beam currents, and the ion beam current is very sensitive to the actual value when the other parameters are fixed. Consequently, the filament heating current is regulated to provide a constant emission current. Figure 49 gives the circuit diagram for this device.

The length of the filament appears to have some influence on the properties of the ion beam. The optimum length from the tip of the tungsten wire to the top of the jaws holding the wire is $2\frac{1}{2} \pm \frac{1}{2}$ mm. The position of the disc nearest the anode has been found to be variable by a small amount, explaining large changes in the focusing parameters immediately after the ion source. Some effort is now made to center that plate over the anode hole.

The ion source has a finite life, and the limiting element is ultimately the hot tungsten filament. This filament draws 20 A at first, and can operate with as little as 7 A. There are, however, other limitations which are less well understood. Deposits of tungsten and especially carbon on the discs and ceramic spacers are deleterious to the source. If a carbon whisker shorts one of the discs to the filament (at cathode potential) the discharge will not start. A carbon deposit on the ceramic spacers is also a condition
Figure 49. Circuit diagram for the filament current regulator. Q1, 5, 6 and 9 are 2N 404; Q3, 8, 10, 11, and 14 are 2N 1039; Q2 and Q7 are 2N 1308; Q12, 13, 15 and 16 are 2N 2156; and Q4 and Q17 are 2N 2801.
which renders it impossible to start a discharge, presumably because
the discs become electrically connected by other than the plasma.
These carbon (and tungsten) deposits are also arc voltage dependent:
they are much less severe at the lowest voltages used.

The least well understood phenomenon associated with the ion
source is that there are clearly different "modes" of the discharge,
for which the extracted current can differ by as much as an order of
magnitude. These modes are definitely associated with the hot plasma
itself, for frequently the visual appearance of the discharge changes
from mode to mode. The existence of more than one mode under given
source conditions gives rise to mode-switching, a phenomenon which
takes one of two forms. In one case, an apparently stable beam
suddenly changes in current by from 5 to 95% and remains at
the new value indefinitely. These changes have been both increases
and decreases, although the larger changes are usually decreases.
The other case is the change from a relatively stable mode to an
unstable one. The unstable mode immediately changes back to the
stable one. These mode jumps appear as spikes, usually downgoing,
in a trace of current vs time. The "rise time" of the spikes is
limited by the electrometer, and no further information on their
duration is available.
APPENDIX C

TIME OF FLIGHT

This appendix gives details of the O atom/O⁻ ion and Cs⁺ ion time of flight calculations. The actual results of the Cs⁺ time of flight calculations are not of as much interest as they were before the Cs⁺ arrival times were measured directly.

Since the O atoms/O⁻ ions travel at constant velocity over all but a small fraction of their path, the calculations are straightforward. The calculation of the time of flight of O⁻ ions to the upper multiplier does take into account the post-acceleration.

In the case of Cs⁺ ions, the trajectories and potentials must be defined in order to make time of flight calculation. Figures 50 and 51 are reproductions of the two-dimensional field map made of the y-z plane in the interaction region, with and without the exit screen respectively. Similar plots were made in the x-z plane, but they do not differ in their qualitative results. Trajectories and time of flight calculations have been made only for the case illustrated in Figure 50, with the screen in place. The arrival time spectra without the screen is broader than it is with it, presumably because there is a greater range of trajectories. Note that the potentials are at most a few volts; these values are scaled up to the actual potentials in the time of flight calculation. This calculation is performed on the time-sharing computer using the
(crude) potential assumption shown in Figure 52. The program is
given in Table XIII.

<table>
<thead>
<tr>
<th>Energy (eV)</th>
<th>Velocity ($10^6$ cm/sec)</th>
<th>Time of Flight (μsec)</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>to Faraday Cup</td>
<td>to Interaction Region</td>
</tr>
<tr>
<td>100</td>
<td>3.48</td>
<td>5.9</td>
<td>8.2</td>
</tr>
<tr>
<td>200</td>
<td>4.92</td>
<td>4.2</td>
<td>5.80</td>
</tr>
<tr>
<td>300</td>
<td>6.02</td>
<td>3.4</td>
<td>4.73</td>
</tr>
<tr>
<td>500</td>
<td>7.77</td>
<td>2.6</td>
<td>3.67</td>
</tr>
<tr>
<td>800</td>
<td>9.83</td>
<td>2.1</td>
<td>2.90</td>
</tr>
<tr>
<td>1000</td>
<td>10.99</td>
<td>1.9</td>
<td>2.59</td>
</tr>
<tr>
<td>1500</td>
<td>13.46</td>
<td>1.5</td>
<td>2.12</td>
</tr>
<tr>
<td>1800</td>
<td>14.75</td>
<td>1.4</td>
<td>1.93</td>
</tr>
<tr>
<td>2000</td>
<td>15.55</td>
<td>1.3</td>
<td>1.83</td>
</tr>
<tr>
<td>2500</td>
<td>17.38</td>
<td>1.2</td>
<td>1.64</td>
</tr>
</tbody>
</table>

Table XII
O Atom/O$^-$ Ion Times of Flight
Figure 50. Potential plot obtained for the interaction region y-z plane with a modeled screen.
Figure 51. Potential plot obtained for the interaction region y-z plane without screen.
Figure 52. Assumed Cs$^+$ acceleration potentials.
Table XIII

Cs+ Time of Flight Program

/CESIUM/
1.0 SET E = 1.602E-11
1.1 SET M = 2.076E-24
2.0 DEMAND Z
2.1 DEMAND V(I), S(I) FOR I = 1 TO Z
2.15 DEMAND U(R)
2.2 TO STEP 2.5
2.3 DEMAND X, Y
2.4 DEMAND V(J) FOR J = X TO Y
2.5 DO PART 3 FOR I = 1 TO Z
2.6 TO STEP 4.1
3.2 SET A(I) = E*(V(I) - V(I-1))/(E*(S(I) - S(I-1)))
3.3 SET T(I) = 1/A(I)*(-U(I-1) + SQRT(U(I-1)*2 + 2*A(I)*(S(I) - S(I-1)))) UNLESS V(I) = V(I-1)
3.4 SET T(I) = (S(I) - S(I-1))/U(I-1) IF V(I) = V(I-1)
3.5 SET U(I) = U(I-1) + A(I)*T(I)
4.1 SET T(0) = -2*U(0)*M*(S(1) - S(0))/(E*(V(1) - V(0))) IF U(0) < 0
4.2 SET T(0) = 0 UNLESS U(0) < 0
4.3 SET Q = SUM(I = 1 TO Z : T(I))
4.4 TYPE Q
5.0 PAUSE
5.2 TO STEP 2.3
APPENDIX D

ABSOLUTE O ATOM DETECTOR DETAILS

Figure 11 gives a schematic diagram of the circuitry of the absolute O atom detector. The preamp is an Ortec 109A charge sensitive preamplifier. The differentiator and filter is a passive RLC network. The amplifier is a Hewlett-Packard 450A operated with 40 dB gain with an emitter follower stage added to provide impedance matching to the EG&G linear gate. The Gaussian filter and amplifier stages are shown in Figure 53, and the slow gate and integrator in Figure 54. The trigger signals are derived from a master synchronization pulse using integrated circuit one-shots. The difference amplifier is an integrated circuit operational amplifier, μA 709C.

The relative atom detector differs in the commercial units used and in that no delayed comparison signal is required. The over-all gain in the electronics is smaller, since there is a substantial gain in the multiplier detector.
Figure 53. Circuit diagram of Gaussian filter amplifier.
Figure 54. Circuit diagram of gated integrator. The switches and operational amplifier for this device are Burr-Brown modules.
APPENDIX E

CALCULATION OF CROSS SECTIONS FROM DATA

1. Procedure

From Equation 17 we have that

\[ \sigma = \frac{S_v c}{F_0 F c^2} \text{ cm}^2 \quad (E1) \]

and from Equation 33 that

\[ F_c = 2.50 \times 10^{14} \text{ F}(1) \text{ atoms/sec} \quad (E2) \]

where \[ F(1) = \int I_c (\mu A) \text{ d}z \quad (E3) \]

The quantities \( S, F_0, \text{ and } F_c \) are all rates; i.e., \#/sec; however, we measure atom numbers and a number of collision events over some given number of laser pulses \( N \). Since the factor to convert atoms and events to rates is \( R/N \) in both cases (where \( R \) is the laser repetition rate), these factors cancel out, and we can substitute the measured numbers directly for the rates.

The \( 0 \) atom flux,

\[ F_0 = 80_i \text{ atoms} \quad (E4) \]

where \( i \) is the absolute \( 0 \) atom detector calibration constant, and \( 0_i \) is the output of the absolute \( 0 \) atom detector. The mean cesium atom velocity,
\[ \overline{v}_c = \frac{3}{4} \sqrt[2]{\frac{2\pi kT_0}{m_c}} \]  \quad (E5)

therefore,

\[ \overline{v}_c = 1.475 \times 10^3 \sqrt{T_0} \text{ cm/sec.} \quad (E6) \]

After combining arithmetic factors,

\[ \sigma = 5.90 \times 10^{-12} \frac{S\sqrt{T_0}}{\beta_0 F(1)f} \text{ cm}^2 \quad (E7) \]

\( T_0 \) is the measured oven temperature in °K, \( 0_1 \) is the mean of the absolute 0 atom detector measurements (over a subgroup), and the quantities \( S, F(1), \) and \( f \) are computed on the time-sharing computer.

In Chapter IV the event rate \( S \) was expressed (indirectly) in terms of the measured parameters: \( \epsilon_1, \epsilon_2, a_0, b_0, a_1, b_1, \) and \( c \) and the estimates \( \alpha, \beta. \) The program shown in Table XIV is used to compute the second order solutions to the algebraically complex set of equations. A similar procedure can be used in 3rd order; however, those refinements are significant only in a few severe cases. The number \( S \) to be substituted in Equation E7 is the value \( S_2, \) the coincidence channel event number.
Table XIV
Extraction of Collision Event Rates from Data

/SECOND/
0 N=1000
1 PRINT "ALPHA, BETA, J, DATA ="
2 INPUT A,B,J,A(O),B(I),I(I),J(I),O(I)
3 A=O(I)/N
4 A1=A(I)/N
5 C=C(I)/N
6 D=J(I)/N
7 E1=I(I)/N
8 PRINT "EFFICIENCIES E1,E2 ="
9 INPUT E1,E2
10 I=0
11 Y=2-E1
12 Z=(60-A1)/E1
13 GO TO 150
14 S=1*4
15 P2=C/(E1*E2)
16 Y=2-E2+P2
17 Y=1-A2*Y*P2
18 Z=(E2-E1)/E2+(1-A2)*P2*Y*P2
19 S=S+1*4
20 P1=Y
21 P2=Z
22 Y=(Z-E1)*C/E1
23 Z=1+(1-E2)*X2*(I2+P2)/Z-(1-E1)*X1/E1
24 Z=A2*P2+A2*E2*E2*P1/(E1*E2)
25 GO TO 150
26 P2=Y
27 P2=Z
28 S=S+1*4
29 I=I+1
30 IF I<4 THEN 31
31 PRINT "SECOND ORDER SOLUCTIONS ARE: SUBGROUP " J
32 PRINT "S2=S S1=S1/S2"
33 PRINT "SD=SD SD=SD/S2"
Table XIV (continued)

55 GO TO 8
145 PRINT X
146 STOP
150 D=SQR(Y+2-4*X*Z)
151 W1=.5*(-Y+D)/X
152 W2=.5*(-Y-D)/X
153 IF (Y+2-4*X*Z)<0 THEN 145
155 IF (W1-W2)<0 THEN 160
156 IF (W2-W1)<0 THEN 165
160 W=W1
161 GO TO 170
165 W=W2
170 IF W<0 THEN 175
171 GO TO 120
173 PRINT "TWO POSITIVE ROOTS: W1, W2 = " W1, W2
174 STOP
175 IF W2<0 THEN 178
177 W=4*?+W2
178 RETURN
The integrals $F(1)$ and $f$ are computed numerically by a trapezoidal fit to the cesium and oxygen atom profile data. Table XV gives the program for these computations, in which $f$ is given by $\frac{F(2)}{F(1)F(0)}$.

2. Sample Calculation

The calculation of the electron transfer and cesium ionization (only) cross section from raw data are presented here for Subgroup L7. In this case, data was also obtained the same day at both smaller and larger 0 atom fluxes; consequently, one good calibration number for the relative 0 atom detector can be obtained. The mean data for L7 are as follows:

\[
\begin{align*}
A_0 &= 15 \\
B_0 &= 12 \\
A_1 &= 57 \\
B_1 &= 72 \\
C &= 34
\end{align*}
\]

relative 0 atom number, $O_2 = 13.2$

where we define $O_2 = \gamma O_1$. From this day's run, we find $\gamma = 4.00 \pm .26$; therefore, $O_1 = 3.30$. The estimated $0^-$ channel efficiency is 0.59; and the trial value for $\varepsilon_2$ is 0.9. The results of the program /SECOND/ are:

\[
\begin{align*}
S_0 &= 74.7 & S_0/S_2 &= 1.16 \\
S_2 &= 64.4 & S_1/S_2 &= .07 \\
S_1 &= 4.3
\end{align*}
\]
Table XV

Form Factor and Cesium Flux Program

/Form FAC/
1.1 DEMAND IN FORM 2: G, C(I), S(I), O(I) FOR I = 1 TO N
1.2 TO STEP 2.0
1.5 OPEN "FGS" FOR INPUT AS FILE 76
1.6 READ FROM 76: N, I, S, O
1.7 READ FROM 76: I, M(I), C(I), L(I) FOR I = 1 TO N
1.8 CLOSE 76
1.9 Z = 4.97
2.0 D(I) = M(I)*Z FOR I = 1 TO N
2.1 F(P) = SUM(I = 1 TO N: (C(I) + C(I-1)) * (O(I) - O(I-1))/2)
2.2 F(I) = SUM(I = 1 TO N: (C(I) + C(I-1)) * (O(I) - O(I-1))/2)
3.0 E(J) = D(J) + S FOR J = 1 TO (N-1)
3.1 B(I) = C(I) + (E(J) - J(I)) * (C(I+1) - C(I)) / (D(I+1) - D(I)) IF E(J) = J(I) AND E(J) < J(I+1) FOR I = 2 TO (N-1) FOR J = 0 TO (N-R)
3.2 L(J) = B(J) * S(J) FOR J = 1 TO (N-1)
3.3 F(2) = SUM(J = 1 TO (N-R): (L(J) + L(J-1)) * (S(J) - D(J-1))/2)
4.0 TYPE F(1), F(2)
4.1 TYPE F(2)/(F(P)*F(I))
5.0 PAUSE
5.9 TYPE IN FORM 2:
6.0 TYPE IN FORM 3: E(J), S(J), B(J) FOR J = 1 TO (N-R)
7.0 PAUSE

FORM 2:
DIMENSION(CM) G ATOMS(V) CESIUM (MIC A)
FORM 3:
% % 
% % 
% % % % 
% % % % 
FORM 4:
# # # # #
whereas, when the value 0.82 is used for $\varepsilon_2$, we have:

\[
\begin{align*}
S_0 &= 74.7 \quad S_0/S_2 = 1.06 \\
S_2 &= 70.2 \quad S_1/S_2 = 0.07 \\
S_1 &= 5.2.
\end{align*}
\]

This value for $\varepsilon_2$ provided the best value for the 3 subgroups taken together; therefore, we used the latter set of results. Note that the 1st order solution, $S_2 = C/\varepsilon_1 \cdot \varepsilon_2$ gives $S_2 = 70.3$.

The cesium beam and form factor data is as shown in Table XVI. The slit scanner calibration is 4.97 cm/in and the wire slit offset is 0.32 cm. The program /FORM FAC/ gives the cesium beam integral, $F(1) = 3.02$, and the form factor, $f = 1.71$. The oven temperature measured during the data run was unusually low, 149°C; whereas, it was 171°C at the time of the above beam density measurements. This difference contributes a negligible uncertainty. Substituting these values into Equation E7, the electron transfer cross section,

\[
\sigma = 5.90 \times 10^{-12} \frac{70.2\sqrt{422}}{(.74 \times 10^{-6})(3.30)(3.02)(1.71)} \text{ cm}^2
\]  

(E8)

\[
\therefore \quad \sigma = 6.7 \times 10^{-16} \text{ cm}^2
\]  

(E9)

and the cross section for cesium ionization without electron attachment is only 7% of that value.
### Table XVI

Cesium and Oxygen Beam Profile Data

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<th>Mike Position (in)</th>
<th>Relative O Atom Detector (Volts)</th>
<th>Cs(^+) Current to Repeller Plate (μA)</th>
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