INFORMATION TO USERS

This was produced from a copy of a document sent to us for microfilming. While the most advanced technological means to photograph and reproduce this document have been used, the quality is heavily dependent upon the quality of the material submitted.

The following explanation of techniques is provided to help you understand markings or notations which may appear on this reproduction.

1. The sign or "target" for pages apparently lacking from the document photographed is "Missing Page(s)". If it was possible to obtain the missing page(s) or section, they are spliced into the film along with adjacent pages. This may have necessitated cutting through an image and duplicating adjacent pages to assure you of complete continuity.

2. When an image on the film is obliterated with a round black mark it is an indication that the film inspector noticed either blurred copy because of movement during exposure, or duplicate copy. Unless we meant to delete copyrighted materials that should not have been filmed, you will find a good image of the page in the adjacent frame. If copyrighted materials were deleted you will find a target note listing the pages in the adjacent frame.

3. When a map, drawing or chart, etc., is part of the material being photographed the photographer has followed a definite method in "sectioning" the material. It is customary to begin filming at the upper left hand corner of a large sheet and to continue from left to right in equal sections with small overlaps. If necessary, sectioning is continued again—beginning below the first row and continuing on until complete.

4. For any illustrations that cannot be reproduced satisfactorily by xerography, photographic prints can be purchased at additional cost and tipped into your xerographic copy. Requests can be made to our Dissertations Customer Services Department.

5. Some pages in any document may have indistinct print. In all cases we have filmed the best available copy.
Harrison, Allen Ray

CONSTRUCTION AND CHARACTERIZATION OF A SPIN POLARIZED HELIUM ION BEAM FOR SURFACE ELECTRONIC STRUCTURE STUDIES

Rice University

Ph.D. 1982

University Microfilms International

300 N. Zeeb Road, Ann Arbor, MI 48106
PLEASE NOTE:

In all cases this material has been filmed in the best possible way from the available copy. Problems encountered with this document have been identified here with a check mark √.

1. Glossy photographs or pages
2. Colored illustrations, paper or print
3. Photographs with dark background
4. Illustrations are poor copy
5. Pages with black marks, not original copy
6. Print shows through as there is text on both sides of page
7. Indistinct, broken or small print on several pages √
8. Print exceeds margin requirements
9. Tightly bound copy with print lost in spine
10. Computer printout pages with indistinct print
11. Page(s) _________ lacking when material received, and not available from school or author.
12. Page(s) _________ seem to be missing in numbering only as text follows.
13. Two pages numbered _________. Text follows.
14. Curling and wrinkled pages
15. Other
RICE UNIVERSITY

CONSTRUCTION AND CHARACTERIZATION OF
A SPIN POLARIZED HELIUM ION BEAM
FOR SURFACE ELECTRONIC STRUCTURE STUDIES

by

ALLEN RAY HARRISON

A THESIS SUBMITTED
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE

DOCTOR OF PHILOSOPHY

APPROVED, THESIS COMMITTEE

G. K. Walters, Professor of Physics, and Space Physics and Astronomy
Chairman

F. B. Dunning, Professor of Space Physics and Astronomy, and Physics

P. Brooks, Professor of Chemistry

HOUSTON, TEXAS
APRIL, 1982
ABSTRACT

CONSTRUCTION AND CHARACTERIZATION OF
A SPIN POLARIZED HELIUM ION BEAM
FOR SURFACE ELECTRONIC STRUCTURE STUDIES

by

Allen Harrison

Ion neutralization and metastable de-excitation spectroscopy, INS and MDS, allow detailed analysis of the surface electronic configuration of metals. The orthodox application of these spectroscopies may be enhanced by electronic spin polarization of the probe beams. For this reason, a spin polarized helium ion beam has been constructed.

The electronic spin of helium metastables created within an rf discharge may be spatially aligned by optically pumping the atoms. Subsequent collisions between metastables produce helium ions which retain the orientation of the electronic spin. Extracted ion polarization, although not directly measurable, may be estimated from extracted electron polarization, metastable polarization, pumping radiation absorption and current
modulation measurements. Ions extracted from the optically pumped discharge exhibit an estimated polarization of about ten per cent at a beam current of a few tenths of a microampere.

Extraction of helium ions from the discharge requires that the ions have a high kinetic energy. However, to avoid undesirable kinetic electron ejection from the target surface, the ions must be decelerated. Examination of various deceleration configurations, in particular exponential and linear deceleration fields, and experimental observation indicate that a linear decelerating field produces the best low energy beam to the target surface.

Auger analysis of the target surface allows determination of the degree of surface contamination. Although various analog systems for Auger analysis are available, a digital, computer compatible system has been constructed.

The technique of ion neutralization spectroscopy and metastable de-excitation spectroscopy allows probing of the surface electronic structure of a metal target. With spin polarization of the probe helium ion beam, the apparatus provides a means to ascertain the existence and properties of surface magnetization. The apparatus not
only may perform the highly surface selective ion neutralization and metastable de-excitation spectroscopies upon a common target surface but also provides the additional diagnostic ability achieved with spin polarization of the probe beam.
ACKNOWLEDGEMENTS

I would like to express my deepest appreciation and thanks to all those who have in some way contributed to the production of this thesis.

I wish to thank Dr. G. K. Walters for his special understanding and guidance during both the conceptual and experimental phases leading to this thesis. To Dr. F. B. Dunning, for his ideas and recommendations, I also express my appreciation.

A special thanks go to my mother and father without whose never-ending encouragement and support this thesis would have been impossible. God bless you.

Finally, a special thanks must go to my wife, Janet, for her patience, support, love and encouragement. It is to her and my parents that this thesis is affectionately dedicated.
# TABLE OF CONTENTS

I. Introduction 1

II. Production and Characterization of Spin Polarized Helium Ions 5
   A. Rf Discharge Production 5
   B. The Optical Pumping Process 7
   C. Quantitative Analysis of the Optical Pumping Process 11
   D. Ion Production Mechanisms 17
   E. Apparatus and Experimental Observations 21
      1. The High Intensity Optical Pumping Lamp 21
      2. Discharge Cleanliness 26
   F. Characterization of the Optical Pumping Process Within the Bulb 29
      1. Mott Analysis of Discharge Electrons 30
      2. Metastable Polarization Analysis By Measurement of Resonance Line Absorption Intensity 31
      3. The Optical Signal 37
      4. Current Modulation 47
      5. Estimate of Ion Polarization 78
III. Ion Extraction, Transport and Deceleration  81
   A. Extraction Canal and Gap Lens  81
   B. Differential Pumping and Transport Optics  85
       1. Differential Pumping  85
       2. Electrostatic Optics  90
   C. Ion Beam Deceleration  93
       1. Exponential Deceleration  93
       2. Linear Deceleration  102
       3. The Deceleration Column  106

IV. Surface Characterization—The Auger Analysis System  114
   A. Auger Analysis Instrumentation  115
   B. Auger Analysis Control System  119
       1. Analog Auger Analysis  120
       2. The Digital Control System  121

V. Ion Neutralization and Metastable De-excitation Spectroscopies  130
   A. Interactions Between The Metal and Incident Ions or Excited Neutral Atoms  131
   B. Electron Polarization Effects in Ion Neutralization and Metastable De-excitation Spectroscopy  141
VI. Proposed Experiments 145
   A. Apparatus Configuration 145
   B. Initial Experiments 149

VII. Conclusion 155

Appendix: Optical Pumping Model
Assuming Two Absorption Lines 157

References 162


**TABLES AND FIGURES**

<table>
<thead>
<tr>
<th>TABLE</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Einstein A and B Coefficients</td>
<td>15</td>
</tr>
<tr>
<td>2</td>
<td>Metastable-Metastable Collision Products</td>
<td>19</td>
</tr>
</tbody>
</table>

**FIGURE 1**

<table>
<thead>
<tr>
<th>Number</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Helium Term Diagram</td>
<td>8</td>
</tr>
<tr>
<td>2</td>
<td>Optical Pumping Lamp</td>
<td>23</td>
</tr>
<tr>
<td>3</td>
<td>Color-center Laser Polarization Measurement Configuration</td>
<td>32</td>
</tr>
<tr>
<td>4</td>
<td>Laser Measurements of Metastable Polarization</td>
<td>35</td>
</tr>
<tr>
<td>5</td>
<td>Optical Pumping Configuration</td>
<td>41</td>
</tr>
<tr>
<td>6</td>
<td>Observed Optical Signal</td>
<td>45</td>
</tr>
<tr>
<td>7</td>
<td>Measured Optical Signals and Ion Currents</td>
<td>48</td>
</tr>
<tr>
<td>8</td>
<td>Configuration for Current Modulation Measurement</td>
<td>66</td>
</tr>
<tr>
<td>9</td>
<td>Measured Current Modulation</td>
<td>69</td>
</tr>
<tr>
<td>10</td>
<td>Discharge Bulb and Mounting Plate</td>
<td>82</td>
</tr>
<tr>
<td>11</td>
<td>Beam Transport and Differential Pumping Configuration</td>
<td>86</td>
</tr>
<tr>
<td>12</td>
<td>Exponential Deceleration Trajectories (theory)</td>
<td>99</td>
</tr>
<tr>
<td>13</td>
<td>Exponential Deceleration Trajectories (calculated)</td>
<td>103</td>
</tr>
<tr>
<td>14</td>
<td>Linear Deceleration Trajectories</td>
<td>107</td>
</tr>
<tr>
<td>Page</td>
<td>Section</td>
<td>Page</td>
</tr>
<tr>
<td>------</td>
<td>----------------------------------------------</td>
<td>------</td>
</tr>
<tr>
<td>15</td>
<td>Deceleration Column</td>
<td>111</td>
</tr>
<tr>
<td>16</td>
<td>Auger Analysis System</td>
<td>116</td>
</tr>
<tr>
<td>17</td>
<td>Digital Analysis System</td>
<td>122</td>
</tr>
<tr>
<td>18</td>
<td>Nickel Auger Electron Curve</td>
<td>127</td>
</tr>
<tr>
<td>19</td>
<td>INS and MDS Triangle Diagram</td>
<td>132</td>
</tr>
<tr>
<td>20</td>
<td>Resonance Neutralization and Resonance Ionization</td>
<td>136</td>
</tr>
<tr>
<td>21</td>
<td>Auger De-excitation</td>
<td>137</td>
</tr>
<tr>
<td>22</td>
<td>Auger Neutralization</td>
<td>138</td>
</tr>
<tr>
<td>23</td>
<td>Ion and Metastable Beam Aparatus Schematic</td>
<td>146</td>
</tr>
<tr>
<td>24</td>
<td>Nickel Density of States</td>
<td>151</td>
</tr>
</tbody>
</table>
INTRODUCTION

Various techniques, such as low energy electron diffraction (LEED) and photoemission are used to probe the properties of solid surfaces. The study of solid surfaces is an area of particularly current research interest with unusual scientific and technological importance. The chemistry of the solid-gas and solid-liquid interface depends, through bond formation, upon surface electronic structure. The surface electronic wave functions extend from the solid surface and thus determine the chemistry of the surface. Although techniques such as LEED and photoemission yield important information concerning surface interactions, they probe not only the surface but the first few atomic layers of the solid. However, ion and excited atom probes are much more surface specific because their neutralization and de-excitation at surfaces are accompanied by electron ejection from the target surface. Energy analysis of these ejected electrons forms the basis of ion neutralization and metastable de-excitation spectroscopies, INS and MDS.

The uniqueness of the apparatus described within this thesis lies in the spin polarization of the probe helium
ion and helium metastable beams. The electronically polarized helium ion beam and the accompanying polarized helium $^2\text{H}_\text{e}$ metastable atom beam provide not only the unique ability to probe a common target surface with both ions and metastables, but also introduces the new ability to perform spin dependent INS and MDS.

This presentation begins by discussing the technique for production of electronically polarized $^2\text{H}_\text{e}$ metastable atoms within a low pressure rf helium discharge. A discussion of the metastable-metastable collision mechanism, which is exploited to produce the polarized ions, follows. The polarized ion production process involves optical orientation of the electronic spins on neutral helium $^2\text{H}_\text{e}$ metastable atoms created within an electrical discharge and extraction of spin polarized ions produced from subsequent metastable-metastable reactions.

The next section presents the technique for extracting the polarized ions from the discharge. Ion polarization detection methods and their associated limitations are also discussed. Estimated ion polarizations of about ten percent, substantiated by several polarization indicators, were achieved.

Cleanliness requirements of the target surface require delivery of the ion beam to the target located
within an ultra-high vacuum environment. The differential pumping and ion transport optics requirements are substantial and are the subject of Chapter III. Because ion transport requires high ion kinetic energies, ion deceleration before delivery to the target, to avoid kinetic electron ejection, is required. Trajectory calculations for decelerating ions in exponential and linear retarding field configurations, to permit selection of the retarding field, are also presented.

Chapter IV discusses the general properties of ion neutralization spectroscopy and metastable de-excitation spectroscopy. A discussion of the new capabilities introduced by spin polarizing the ion and metastable beams concludes the chapter.

Determination of crystal cleanliness is accomplished by Auger electron spectroscopy and a digital analysis system, built for this apparatus, will be described. The digital system employs a digital phase locked detection technique which, although not interfaced at present, allows simple computer interconnection.

The last section discusses an experiment for examining the surface magnetic properties of nickel where the ferromagnetic itinerant electron model predicts a spin-split band for the bulk electronic states. The proposed experiment would perform spin-dependent INS, with
the polarized ion and metastable beams, to discern
differences in the surface and electronic structures for
magnetic materials. This experiment emphasizes the forte
of this apparatus: the unique ability to not only perform
the highly surface selective ion neutralization and
metastable de-excitation spectroscopies upon a common
target surface but also to use the additional diagnostic
capabilities provided by spin polarization of the probe
beams.
II
PRODUCTION AND CHARACTERIZATION OF
SPIN POLARIZED HELIUM IONS

The ion neutralization spectroscopy for probing the magnetic properties of the nickel surface requires an electronically polarized helium ion beam. The following sections present the technique for obtaining polarized ions by optically pumping helium 2(3)S metastable atoms excited within a radio frequency discharge and producing polarized ions through subsequent metastable-metastable collisions.

A. RF DISCHARGE PRODUCTION

In this particular apparatus, production of the 2(3)S helium metastables involves rf discharge induced electron impact excitation of helium atoms within a glass bulb containing a pressure of 0.1 torr of helium. An rf discharge may be created within such a bulb by placing the bulb between a pair of metal plates across which an rf potential is applied or by placing the bulb within a coil across which an rf potential is applied. In the first case, an alternating electric field excites the bulb and
in the second case an alternating magnetic field excites the bulb. Electric field excitation has been reported to produce discharges to lower pressures, down to 10**-5 torr [1], and therefore in order to achieve low discharge levels at low pressures with a minimum of rf power, the electric field excitation method was selected for this apparatus.

Application of an rf electric field to the helium filled bulb forces any free electrons to oscillate with a velocity ninety degrees out of phase with the applied field and therefore no net power from the rf source will be absorbed. However, in the event of an elastic collision between an electron and an atom, a velocity component perpendicular to the applied rf field may result and produce a change in momentum with consequent power absorption from the rf source. Each such quasi-elastic collision results in energy transfer to the electron of only about 0.001 eV [2]. Because the first excited state of helium lies 19.8 eV above the ground state, many such electron-atom collisions occur before the electron gains sufficient energy to electronically excite a ground state helium atom. Upon reaching the 19.8 eV threshold energy, the de-excitation cross section for the excited electron increases abruptly and excited helium 2(3)S atoms are produced.
In a weak discharge of 0.1 torr helium, the steady state metastable density typically lies between $10^{10}$ and $10^{11}$ per cubic cm and the metastable lifetime, governed by thermal diffusion to the cell walls, is a few tenths of a millisecond. The mean free path, inversely proportional to pressure, for helium at 0.1 torr is about 0.15 cm with a helium-helium collision rate of about $8.4 \times 10^{10}$ collisions per second [3].

B. THE OPTICAL PUMPING PROCESS


The pumping process (see Figure 1) produces an unequal population distribution among the 2(3)S magnetic sublevels of helium metastables created within the rf discharge. Without optical pumping, thermalizing processes at 300 K would produce equal sublevel populations. The 2(3)S metastable helium atoms are doubly
FIGURE 1

Partial term diagram for helium showing 2(1)S and 2(3)S metastable states and the transitions involved, D0, D1, D2 and D3, in the optical pumping process. Relative energies not to scale. Zeeman energy splitting for the triplet levels shown to the right with Mj indicated.
forbidden to radiatively decay to the singlet ground state by $\Delta s = 0$ and $S \rightarrow S$ selection rules.

For helium, the optical pumping process proceeds as follows. Resonance radiation produced by a high intensity rf helium discharge lamp is directed coaxially along the axis defined by a weak, superimposed magnetic field. This infrared radiation, at a wavelength of about 1.08 microns, passes successively through an infrared pass filter and linear polarizer (Polaroid type HR) and a quarter wave plate and thus becomes circularly polarized. The circularly polarized light excites transitions from the $2(3)S$ metastable state to the $2(3)P$ states where selection rules require the magnetic quantum number $M_j$ to increase by one for right handed circularly polarized light (RHCPL), corresponding to sigma+ transitions, and to decrease by one for left HCPH, corresponding to sigma- transitions. Subsequent spontaneous reradiation from the $2(3)P$ state back to the $2(3)S$ metastable state, occurring in about $10^{-7}$ seconds, proceeds by electric dipole radiation where $\Delta M_j = 0, +$ or $- 1$. For example, the combination of a $2(3)S$ level with $M_j = 0$ and RHCPL produces a $2(3)P$ level with $M_j = 1$. Subsequent emission from the $2(3)P$ level leading to the $2(3)S$ level produces a metastable with $M_j = 0$ or $+1$. For this particular example, a fraction of the original $2(3)S$ $M_j = 0$ atoms
become $2(3)S$ Mj = 1 atoms. For either type of pumping photon the equilibrium populations of the metastable sublevels depends upon competition between the pumping and thermalization processes which tend to equilibrate the population of the metastable magnetic sublevels.

In a weak discharge of 0.1 torr helium, the steady state metastable density lies between $10^{14}+10$ and $10^{15}+11$ metastables per cubic cm and the metastable lifetime, limited by thermal diffusion to the cell walls, becomes a few tenths of a millisecond [7].

C. QUANTITATIVE ANALYSIS OF THE OPTICAL PUMPING PROCESS

The technique of optical pumping involves changing the relative populations of the metastable $2(3)S$ helium magnetic sublevels. In the absence of the 1.08 micron pumping radiation, the normal Boltzmann distribution populates equally the three $2(3)S$ magnetic sublevels. With the introduction of the circularly polarized pumping light, the relative population among the sublevels changes. A quantitative formulation of this process follows [7].

Allow the magnetic $2(3)S$ sublevels to have populations described by $N_i$ where $i = \pm, 0$ and + corresponding to Mj values of -1, 0 and +1. The pumping
process begins with a $2(3)S$ metastable atom in the $i$-th $M_j$ sublevel absorbing a pumping photon and being excited to the $k$-th $2(3)P$ magnetic sublevel. The $k$-th sublevel is one of the nine sublevels associated with the three $2(3)P$ states: $2(3)P_0$, $2(3)P_1$ and $2(3)P_2$. This process proceeds at a rate proportional to the product of the intensity of the pumping radiation causing the transition, $F$, the absorption probability for a transition from the $i$-th $2(3)S$ sublevel to the $k$-th $2(3)P$ sublevel, $B_{ik}$, and the initial population of the $i$-th $2(3)S$ sublevel, $N_i$. The loss of population from the $i$-th $2(3)S$ sublevel becomes

$$-\sum_{k=1}^{q} F N_i B_{ik}$$

To complete the pumping cycle, those atoms pumped to the $2(3)P$ states return to the $2(3)S$ state by spontaneous emission. The repopulation rate of the $i$-th $2(3)S$ sublevel involves a two-step process where an atom in the $j$-th $2(3)S$ sublevel excites to the $k$-th $2(3)P$ sublevel and then spontaneously radiates to the $i$-th $2(3)S$ sublevel. Defining $A_{ki}$ to be the Einstein coefficient for $2(3)P (k) \rightarrow 2(3)S (i)$, then the rate for this process may be expressed as

$$+\sum_{j=1}^{3} \sum_{k=1}^{q} F N_j B_{jk} A_{ki}$$
In competition with the optical pumping process, thermalization tends to restore the normal Boltzmann distribution. Using \( N \) to represent the total population of the \( 2S \) state and \( T \) to represent the characteristic lifetime of the thermalization process, the thermalization rate may be written as

\[
+ [ (N/3) - N_i ] / T
\]

For weak magnetic where sublevel energy splitting is small, such as employed in this experiment, the Boltzmann distribution produces essentially equal populations among the \( 2S \) sublevels. Thus the \( i \)-th \( 2S \) sublevel obeys the following population rate equation

\[
d (N_i) / dt = - \sum F_{Ni} B_{ik} + \sum_{j=1}^{3} \sum_{k=1}^{q} F_{Nj} B_{jk} A_{ki} + [ (N/3) - N_i ] / T
\]

The pumping light from the high intensity helium rf discharge lamp contains three emission lines, D0, D1 and D2, indicated in Figure 1. The intensities, positions and shapes of these lines have been examined \([8, 9]\) and the D1 and D2 transitions found to be unresolvable due to Doppler broadening within the high intensity pumping lamp discharge. Therefore, the unresolved D1 and D2 lines may referred to as one line, D3. The intensities of the lines
D0, D1 and D2 as seen by absorbing 2(3)S atoms may be represented by a relative intensity ratio of K:Li:1.
Previous work [7] suggests relative intensities roughly in the ratio 0.4:1.3:1. However this ratio can only be regarded as semiquantitative at best.

In helium with n = 2, L-S coupling accurately represents the atomic states and allows the electric dipole transition probabilities to be obtained from the dipole matrix elements given by Condon and Shortley [9]. The transition probabilities Bjk and Aik have been tabulated using these matrix elements [10] and are presented in Table 1 for RHCPL, sigma+ transitions, and the selection rule \( \Delta M_J = \pm 1 \).

The population of the i-th 2(3)S sublevel may be now calculated using the transition probabilities of Table 1 and, assuming a steady state, \( \frac{d(N_i)}{dt} \) set to zero. Allowing the three magnetic sublevels of the 2(3)S state corresponding to \( M_J = \pm 1, 0 \) and \(-1\) to have populations of \( N^+, N^0 \) and \( N^- \) respectively, the steady state population densities for RHCPL are

\[
N^+ = \left( \frac{2N}{q} \right) \left[ 1 + \left( \frac{T F}{2} \right) \left( 8 + 12L + 4K \right) \right] \\
+ \left( \frac{3}{4} \right) \left( \frac{T F}{2} \right)^2 \left( 1 + L \right) \left( 5 + 9L + 8K \right)
\]

\[
N^0 = \left( \frac{2N}{q} \right) \left[ 1 + \left( \frac{T F}{2} \right) \left( 3 + 3L + 4K \right) \right]
\]

\[
N^- = \left( \frac{2N}{q} \right) \left[ 1 + \left( \frac{T F}{2} \right) \left( 3 + 3L \right) \right]
\]
**TABLE 1**

Einstein A and B coefficients for sigma+ transitions between the 2(3)S magnetic sublevels and the 2(3)P magnetic sublevels. Factors K and L refer to the pumping light intensity.
<table>
<thead>
<tr>
<th>$m_s(i)$</th>
<th>$2^3p_2$</th>
<th>$2^3p_1$</th>
<th>$2^3p_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>+2</td>
<td>+1</td>
<td>0</td>
<td>-1</td>
</tr>
<tr>
<td>+1</td>
<td>0</td>
<td>-1</td>
<td>0</td>
</tr>
<tr>
<td>0</td>
<td>-2</td>
<td>0</td>
<td>-1</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$m_k(i)$</th>
<th>$B_{j,k}$</th>
<th>$A_{j,k}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>+1</td>
<td>6 0 0 0 0</td>
<td>$B_{j,k}$</td>
</tr>
<tr>
<td>0</td>
<td>0 3 0 0 0</td>
<td>$A_{j,k}$</td>
</tr>
<tr>
<td>-1</td>
<td>0 0 1 0 0</td>
<td></td>
</tr>
</tbody>
</table>

$B_{j,k}$ and $A_{j,k}$ are likely to be coefficients or parameters in a physical or mathematical context, such as in quantum mechanics or spectroscopy.
where

\[ q = \left(\frac{3}{2}\right) (T \, F)^2 (1 + L) (5 + 9 \, L + 8 \, K) + T \, F (14 + 18 \, L + 8 \, K) + 6 \]

Note that for large relaxation times, the population ratio becomes N : O : 0 and therefore, without relaxation processes, the metastables would become completely polarized.

D. ION PRODUCTION MECHANISMS

The principal ionization reactions sustaining the discharge are direct helium ionization, cumulative ionization and metastable-metastable de-excitation. For direct helium ionization,

\[ \text{He}^+ + e^- \ (\ > 24.5 \text{ eV}) \longrightarrow \text{He}^+ + 2 \, e^- \]

the energy of the incident electron must be greater than the ionization energy of helium, 24.5 eV. For cumulative ionization,

\[ \text{He}^* + e^- \longrightarrow \text{He}^+ + 2 \, e^- \]

a previously excited, but not ionized, helium atom collides with an electron energetic enough to ionize the excited helium atom. Because metastable 2(3)S and 2(1)S atoms constitute the majority of excited atoms present in the discharge, cumulative ionization therefore becomes primarily metastable ionization.
\[ \text{He}^m + e^- \rightarrow \text{He}^+ + 2 \text{e}^- \]

Metastable-metastable ion production

\[ \text{He}^m + \text{He}^m \rightarrow \text{He} \, [1(1S)] + \text{He}^+ + \text{e}^- \]

involves a collision between two metastable atoms which produces a ground state helium atom, an ion and an electron through a chemionization reaction.

The rates for these processes have been estimated [7] and indicate that the majority of electrons and ions originate from metastable-metastable collisions.

In metastable-metastable reactions, conservation of spin angular momentum, which has been verified experimentally [7, 11], predicts the electronic spins of the product ions and electrons. Table 2 lists the possible reactions between metastables of various magnetic quantum numbers, $M_j$. For each combination of $M_j$ values of the colliding metastables, the table indicates the electronic spin of the product ion and electron. The table further indicates the relative number of product ions and electrons having spins "up" and spins "down" and the polarization, defined as the difference between the number of electrons with spin up and the number with spin down, divided by the total number of electrons produced. In a steady state, the numbers of spin up and spin down electrons are time independent, $\frac{d\langle nu \rangle}{dt} = \frac{d\langle nd \rangle}{dt} = \ldots$
TABLE 2

Table 2 is the metastable-metastable collision products table. Columns one and two, from the left, indicate the Mj value and population of the two colliding metastables. The third column gives Ms of the product ion and column six gives Ms of the product electron. Note that two combinations of colliding metastables do not produce ions or electrons due to spin conservation requirements. Column four and five, for the product ion, and column seven and eight, for the product electron, give the number of ions or electrons produced with Ms = -1/2 and +1/2. Polarization is calculated from the total number of product electrons or ions from columns four and five or columns seven and eight.
\[ \text{He}^m_1 + \text{He}^m_2 \rightarrow \text{He} \left(1^2\text{S}_0\right) + \text{He}^+ + e^- \]

<table>
<thead>
<tr>
<th>((N_{j,1}', N_{j,1}))</th>
<th>((N_{j,2}', N_{j,2}))</th>
<th>(N_{s, \text{He}^+} n_{1/2}^+ n_{-1/2}^+)</th>
<th>(N_{s,-} n_{1/2}^- n_{-1/2}^-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>((0, N_s))</td>
<td>((0, N_s))</td>
<td>(3/2) (1/2 ) (N_s^2) (1/2 ) (N_s^2)</td>
<td>(3/2) (1/2 ) (N_s^2) (1/2 ) (N_s^2)</td>
</tr>
<tr>
<td>((+1, N_s))</td>
<td>((0, N_s))</td>
<td>(+1/2) (N_{sN}^+) (0)</td>
<td>(+1/2) (N_{sN}^+) (0)</td>
</tr>
<tr>
<td>((0, N_s))</td>
<td>((0, N_s))</td>
<td>(+1/2) (N_{sN}^+) (1/2 ) (N_s^2)</td>
<td>(+1/2) (N_{sN}^+) (1/2 ) (N_s^2)</td>
</tr>
<tr>
<td>((-1, N_s))</td>
<td>((0, N_s))</td>
<td>(-1/2) (0) (N_s^-)</td>
<td>(-1/2) (0) (N_s^-)</td>
</tr>
<tr>
<td>(+1, N_s)</td>
<td>((0, N_s))</td>
<td>(+1/2) (N_{sN}^+) (0)</td>
<td>(+1/2) (N_{sN}^+) (0)</td>
</tr>
<tr>
<td>((0, N_s))</td>
<td>((-1, N_s))</td>
<td>(+1/2) (N_{sN}^+) (1/2 ) (N_s^2)</td>
<td>(+1/2) (N_{sN}^+) (1/2 ) (N_s^2)</td>
</tr>
<tr>
<td>((0, N_s))</td>
<td>((0, N_s))</td>
<td>(+1/2) (N_{sN}^+) (0)</td>
<td>(+1/2) (N_{sN}^+) (0)</td>
</tr>
<tr>
<td>((-1, N_s))</td>
<td>((0, N_s))</td>
<td>(+1/2) (N_{sN}^+) (1/2 ) (N_s^2)</td>
<td>(+1/2) (N_{sN}^+) (1/2 ) (N_s^2)</td>
</tr>
<tr>
<td>((0, N_s))</td>
<td>((-1, N_s))</td>
<td>(-1/2) (0) (N_s^-)</td>
<td>(-1/2) (0) (N_s^-)</td>
</tr>
<tr>
<td>(N_s) = singlet population</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(N_{s, \text{He}^+}) = triplet population for (N_{j,1} = +1, 0, -1) respectively</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(N_{s,-}) = spin of electron on (\text{He}^+)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(n_{1/2}^+, n_{-1/2}^+) = number of (\text{He}^+) with spin (+1/2) and (-1/2) respectively</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(n_{1/2}^-) = spin of product electron</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(n_{-1/2}^-, n_{-1/2}^-) = number of electrons with spin (+1/2) and (-1/2) respectively</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\[
\frac{dn_{1/2}}{dt} = \frac{1}{2} N_s^2 + N_{sN}^+ + 2 N_{sN}^- + 2 N_{sN}^+ + N_{sN}^- + 1/2 N_0^2
\]

\[
\frac{dn_{-1/2}}{dt} = \frac{1}{2} N_s^2 + N_{sN}^+ + 2 N_{sN}^- + 2 N_{sN}^+ + N_{sN}^- + 1/2 N_0^2
\]

\[
p = \frac{n_{1/2} - n_{-1/2}}{n_{1/2} + n_{-1/2}} = \frac{2 (N_{s, \text{He}^+} - N_{s,-})}{N_s + N_{sN}^+ + N_{sN}^- - N_0^2 - N_s^2 - N_0^2}
\]
0, and the polarization becomes
\[ P = \frac{\nu_0 - nd}{\nu_0 + nd} \]
\[ = \frac{2(\nu_0 + \nu_s)(\nu_s^+ - \nu_s^-)}{\nu_s^2 + \nu_s^0 + (\nu_0 + \nu_s^-)^2 - \nu_s^+^2 - \nu_s^-^2} \]

A polarization of about 8 to 10% was measured [7] for electrons extracted from a configuration similar to that described in this thesis. Due to the symmetry between ion and electron production as seen from Table 2, the ion polarization of the present configuration, which has not been directly measured, should be comparable.

E. APPARATUS AND EXPERIMENTAL OBSERVATIONS

1. THE HIGH INTENSITY OPTICAL PUMPING LAMP

The high intensity optical pumping lamp provides the resonance radiation for orientation of the helium metastables in the low intensity discharge bulb. Intense helium discharge lamps have been discussed in the literature [5, 12] and the excitation circuitry chosen for this particular lamp was reported by Salomaa [13]. However, these lamps employ a sealed discharge tube with which past experience in this laboratory has indicated three major drawbacks. A sealed tube does not allow selection of helium pressure, nor purge of contaminants
within the tube, nor ease of replacement of a damaged tube. Initially the effect of helium pressure upon the intensity of 1.08 micron resonance radiation was unknown and therefore the ability to vary operating pressure was desired. Contamination of the tube by atoms other than helium was known to substantially degrade light output and construction of new sealed tubes required a lengthy and tedious vacuum cleaning procedure. To circumvent these problems, a flowing, variable pressure discharge lamp was constructed.

The lamp is excited by a 150 megacycle oscillator with circuitry as employed by Salomaa but of simpler and more compact construction than those previously employed in this laboratory. The entire lamp, see Figure 2, discharge cell and oscillator, was placed within a copper enclosure to shield the laboratory from the rf fields created by the 500 watt oscillator. The discharge cell, made of quartz to withstand the high temperatures associated with the intense discharge and to efficiently pass the radiation, consists of a quartz disc of 1.4 inch diameter, 0.5 inch thickness and 1 mm walls. Extending from the disc plane, two 6 mm quartz tubes provide the helium gas entrance and exit canals which conveniently mate with o-ring fittings. The design allows pressure selection through adjustment of the helium flow with a
FIGURE 2

Diagram of the pumping lamp indicating oscillator circuitry, discharge cell, helium flow, parabolic mirror, linear polarizer, quarter wave plate and copper shielding enclosure.
needle valve between the entrance canal and helium supply regulator. Furthermore, helium flow continuously flushes away any contaminants and ensures discharge cleanliness. With time all quartz tubes, both sealed and flowing, exhibit crazing of the cell surface. The sealed cell becomes useless with such crazing and must be discarded, while the inexpensive flowing cell is quickly and easily replaced.

The high intensity optical pumping lamp employed with this experiment exhibits the following characteristics. Maximum 1.08 micron intensity is observed to be moderately pressure dependent and optimum in the vicinity of 1 torr despite there being less total power coupled to the discharge at this optimum pressure for maximum intensity than at other pressures where more total power may be absorbed.

The 1.08 micron radiation was monitored with a 1.08 micron pass filter, which passes 72% of the incident 1.08 micron radiation, and a germanium photodiode [14] of one millimeter detector diameter and sensitivity at 1.08 microns of about 0.5 A/W. At about six inches from the lamp with the circular polarizer in place, a 1.08 micron radiation intensity of up to 5 milliwatts per square centimeter or $2.8 \times 10^{16}$ photons per second per square centimeter was achieved. The total power incident upon
the optically pumped sample bulb is estimated to lie between 40 and 60 milliwatts.

2. DISCHARGE CLEANLINESS

Atoms other than helium atoms within the discharge bulb may undergo chemiionization reactions with the helium metastables.

\[
\text{He}(m) + X \rightarrow \text{He}^{(1S)} + X^{+} + e^{-}
\]

Such reactions produce ions other than helium ions which reduce the helium ion polarization by reducing the number of metastable-metastable collisions. Therefore, discharge cleanliness is imperative.

Various indicators may be used to confirm discharge cleanliness. The most general technique involves observation of light from the discharge bulb with a hand spectroscope. With a clean discharge, the spectroscope slits open wide and the room lights off, discharge observation with the spectroscope shows only the bright helium lines. Experience has shown that any other light, usually bands in the green or haze in the red, indicates unacceptable levels of contamination. Under such contaminated conditions, observations of the optical signal or current modulation, which are strong indicators of degree of optical pumping to be presented in the
following two sections, indicate serious degradation of the metastable polarization.

Various techniques for obtaining contaminant free discharges have been suggested. The following cleaning procedure has yielded reproducible results. First, any new bulb or one whose surface may have been contaminated should be first cleaned with acetone and dried with a methyl alcohol rinse. After being placed on the vacuum system and then evacuated, the bulb should be heated to release remaining contaminants from the bulb walls. The system constructed for this experiment employs zeolite molecular sieve traps on the gas inlet lines which, after being exposed to the atmosphere, saturate with water. Therefore, as the bulb is baked, so are the inlet lines in order to purge the gas system as completely as possible. Because viton o-rings seal the bulb, the bulb temperature is monitored with a mercury thermometer and held below 150° C. After the usual overnight bake, the gas handling system is cooled while the bulb remains hot to minimize contaminant sticking to the bulb. Next the bulb is allowed to cool to room temperature.

Observations at this point always indicate a contaminated discharge. The next step involves lighting a very intense discharge within the bulb. To this end, an oscillator similar to that used with the high intensity
optical pumping lamp is employed to produce a discharge within the bulb, with helium pressure between 200 to 600 microns and input power of about 100 watts, whose visible radiation is uncomfortable to the eye. This discharge will usually produce a clean bulb between one and three hours after lighting.

Although the above recipe produces a clean discharge, care must be taken with other elements of the system to avoid re-contamination. The gas handling lines are not only bakable but also contain molecular sieve traps which, when immersed in liquid nitrogen, collect contaminants in the incoming helium and also contaminants remaining in the lines themselves. Another important element to ensure maximum cleanliness involves flowing helium within the discharge bulb, just as discussed earlier with respect to the pumping lamp, to flush out contaminants released from the bulb. The cleaning process described above relates to this particular flowing helium configuration.

In general, once a bulb has been cleaned, cleanliness will be retained until vacuum has been breeched. If contamination returns, such as has been observed after periods during which no discharge has been lit, and the bulb has remained under decent vacuum, less than 10**-5 torr, cleanliness may be re-attained by carefully flame heating the bulb. A continuous low intensity discharge
during periods of disuse usually avoids this potential problem.

F. CHARACTERIZATION OF THE OPTICAL PUMPING PROCESS WITHIN THE BULB

Production of a contamination free helium discharge and optical pumping with the high intensity lamp should produce the desired metastable sublevel population inversion. Four techniques are available to determine whether optical pumping is indeed occurring. First, Mott scattering of electrons from the discharge allows determination of their spin polarization which is expected to be the same as that of the ions. For this particular apparatus, a Mott scattering system is unavailable.

Second, exact determination of the metastable sublevel populations by optical absorption measurements permits calculation of the metastable polarization. Such an experiment employing an infrared color-center laser has been completed on this apparatus and will be mentioned briefly below. However, such measurements do not provide a direct measure of the ion polarization. Third, measurement of the optical signal indicates the presence of optical pumping and provides a method by which pumping conditions may be monitored. This measurement, although
not providing a quantitative measure of polarization, does provide a useful diagnostic tool by measuring pumping light absorption during pumped and unpumped discharge conditions. Fourth, analysis of ion production predicts that the total extracted ion current depends upon the triplet sublevel populations and this has been measured in experiments described below.

1. MOTT ANALYSIS OF DISCHARGE ELECTRONS

As discussed in the section on ion production mechanisms, those ions and electrons produced by metastable-metastable collisions should have the same polarization. However, those electrons produced by other processes, from the cell walls or metastable-contaminant collisions, tend to degrade the extracted electron polarization. Polarization analysis of electrons from the discharge therefore will place a lower limit upon the metastable and ion polarization.

The technique of Mott scattering, which is based on electron scattering asymmetry due to spin-orbit coupling of high energy electrons with heavy nuclei [15], allows direct polarization measurement of those electrons extracted from the discharge. Although this particular apparatus does not at present have provisions for Mott
analysis, electron polarizations of 10% have been measured with a similar discharge source [7]. Although the expertise is available in this laboratory for Mott analysis [16], other more convenient and expeditious measurements have been made and will now be presented.

2. METASTABLE POLARIZATION ANALYSIS BY MEASUREMENT OF RESONANCE LINE ABSORPTION INTENSITY

Because of Doppler line broadening within the high intensity pumping lamp, observation of sample absorption of the individual pumping lines is nearly impossible because of light intensity considerations. Recently, however, a color-center laser tunable over the 1.08 micron region has been constructed in this laboratory [17] which, with the narrow laser line width, permits examination of the D0, 2(3)S \rightarrow 2(3)P0, transition separately from D3, 2(3)S \rightarrow 2(3)P1,2 transition. The experimental configuration is illustrated in Figure 3.

To probe the D0 transition, circularly polarized light from the laser, tuned to the D0 transition and attenuated so as not to alter the triplet metastable sublevels, passes through the bulb co-axially with the pumping light and magnetic field. For RHCP laser light, transitions from \( M_j = -1, \ 2(3)S \) to \( M_j = 0, \ 2(3)P \)
FIGURE 3

Experimental configuration for color-center laser measurement of triplet metastable polarization. The figure shows the high intensity helium discharge lamp with associated parabolic mirror, linear polarizer and rotating quarter-wave plate. The weak 1.08 micron probe radiation from the color-center laser passes through the discharge and is reflected to an IR detector and amplifier. The DC signal measures the percent absorption of 1.08 radiation and indicates the discharge intensity. The AC component, oscillating with phase and frequency determined by the rotating quarter-wave plate and applied to the lock-in amplifier, indicates the change in absorption during the pumped and unpumped discharge conditions.
occur. Because of the symmetry of the \( 2(3S) \) sublevels, the difference in populations of the \( M_j = +1 \) and \( M_j = -1 \) sublevels may be determined by keeping the sense of laser polarization constant and rotating the quarter wave plate in front of the pumping lamp. With this arrangement, one effectively measures first the population of the depleted sublevel and then the population of the accreted sublevel. The metastable polarization is

\[
P_{\text{2(3S)}} = \frac{(N^+ - N^-)}{N_t}
\]

where \( N_t = N^+ + N^0 + N^- \), the total number of metastables which remains constant for either pumped or unpumped discharges. Since absorption is proportional to population, the polarization may be determined by measuring the intensity component of transmitted laser light oscillating in synchronization with the rotating pumping lamp quarter wave plate which modulates the \( M_j = -1 \) sublevel between depleted and accreted limits. Therefore determination of metastable polarization involves measurement of absorption modulation intensity, which provides the extent of sublevel population inversion for the numerator in the above expression, and unpumped discharge absorption, which provides the total number of metastables for the denominator. Figure 4 presents metastable polarizations based upon such measurements for several helium pressures as a function of discharge
FIGURE 4

Measured helium triplet metastable polarization as a function of discharge intensity, indicated by percent absorption of 1.08 micron pumping radiation, for selected helium pressures. Data were calculated from color-center laser absorption measurements.
intensity indicated by per cent absorption of the 1.08 micron pumping light [18].

3. THE OPTICAL SIGNAL

The optical signal is defined as the change in pumping light absorbed by the sample cell for the pumped versus unpumped conditions. Allowing $I_u(n)$ and $I_p(n)$ to represent the absorbed light intensity in the unpumped and pumped conditions respectively, with $(n)$ designating the contribution of the absorption from the D$n$ component of the pumping radiation, the optical signal is

$$
\Delta I = \sum_{n=1}^{q} I(n) - I_p(n)
$$

Because $\Delta I$ depends upon absolute light intensity and geometrical factors, a more convenient and meaningful measure for the degree of $2(3)6$ sublevel inversion is the optical signal normalized to $I_a$, the total light absorbed by the unpumped metastable population in the cell.

$$
\Delta I / I_a = \left[ \sum_{n=1}^{q} I_u(n) - I_p(n) \right] / \left[ \sum_{n=1}^{q} I_a(n) \right]
$$

$$
= \sum_{n=1}^{q} \left[ I_u(j) - I_p(j) \right] / \left[ IO - It \right]
$$

where $IO$ is the total pumping light intensity transmitted with discharge off and $It$ is the transmitted light
intensity with discharge on.

In terms of the absorption probabilities, $K_{11}$ for D0, D1 and D2 components of the pumping radiation, and the sublevel populations, the individual absorption components for unpumped and pumped discharges may be written as,
using Table 1,

$$I_{u}(0) = (2 \, K) \, (1/3) \, N$$

$$I_{u}(1) = (3 \, L) \, (N/3) + (3 \, L) \, (N/3)$$
$$\quad = (6 \, L) \, (N/3)$$

$$I_{u}(2) = 6 \, (N/3) + 3 \, (N/3) + (N/3)$$
$$\quad = (10/3) \, N$$

$$I_{p}(0) = (2 \, K) \, N^-$$

$$I_{p}(1) = (3 \, L) \, (N^0 + N^-)$$

$$I_{p}(2) = 6 \, N + 3 \, N^0 + N^-$$

where the numbers $n = 0, 1, 2$ in parentheses on the left indicate the pumping line $2(3)S1 \rightarrow 2(3)Pn$. Note also that the intensity ratio of $K_{11}$ has been assumed to remain constant throughout the cell. Substitution of the previously obtained values for $N_i = N^0, N^+$ and $N^-$ yields

$$K \, (5 + 9L + 8K) \, (2T \, F + 3(T \, F)^2 \, (1 + L)$$

$$\Delta I / Ia = \frac{(10 + 9L + 2K) \, [6 + T \, F \, (14 + 18L + 8K)$$
$$\quad + (3/2) \, (T \, F)^2 \, (1 + L) \, (5 + 9L + 8K)]}{(10 + 9L + 2K) \, [6 + T \, F \, (14 + 18L + 8K)$$
$$\quad + (3/2) \, (T \, F)^2 \, (1 + L) \, (5 + 9L + 8K)]}$$

Furthermore because of Doppler broadening within the high intensity pumping lamp and the small separation of the D1 and D2 emissions, these two lines are unseparable
and together are designated the D3 pumping line [5]. The rate of absorption for the D0 and D3 lines depends upon the metastable density, the thickness of the absorber and absorption probability; in particular the absorption rate of the D0 line differs from that of the D3 line. For the sample cell used in this experiment, overall absorptions as high as 60% have been observed. Because of the difference in absorption rates of the D0 and D3 components, the local intensity ratio between the two pumping lines depends upon their position within the discharge and the metastable density [6]. As a result, the local degree of pumping due to each line depends upon the depth within the bulb. The previous discussion assumed that the intensity ratio remained constant, as might be expected in the limit of a thin absorber, but for the higher absorption rates encountered with this apparatus the analysis becomes more complex. At high absorption rates, the intensity ratio D0:D3 of the light exiting the bulb may be quite different from the initial intensity ratio. Furthermore, because the pumping light line widths are appreciably wider than the absorption lines of the bulb discharge, the light exiting the bulb may exhibit intensity holes corresponding to the narrower discharge absorption lines.
Because of the complications noted above, the optical signal does not yield a unique value for the metastable polarization. In general, for a weak absorber, the greater the signal, the greater the metastable polarization but, as will be discussed in the following paragraphs, geometrical and metastable density considerations preclude quantitative polarization measurements. The Appendix presents a simple model illustrating the non-unique nature of the optical signal for polarization measurement.

Observation of the optical signal may be attempted by several methods, which although not entirely successful, present useful information concerning the discharge within the bulb.

The basic experimental configuration, Figure 5, includes the discharge bulb, Helmholtz coil pairs for superimposing a constant magnetic field along the optical pumping axis, a circularly polarized pumping lamp, an infrared detector and a Larmor coil with axis perpendicular to the pumping axis. The Larmor coil, when excited at the frequency corresponding to the Zeeman energy separation of the \(2(3)S\) sublevels, stimulates magnetic dipole transitions between the triplet metastable magnetic sublevels. With sufficient intensity, the Larmor radiation short circuits the pumping process by forcing
FIGURE 5

Optical pumping experimental configuration. The figure illustrates the high intensity helium pumping lamp with associated parabolic mirror, linear polarizer and quarter wave plate. An IR detector and amplifier combination provided both AC and DC signals for oscilloscope display of the optical signal and for measurement of percent absorption, respectively. A reference signal from the Helmholtz coil oscillator provides the X-axis trace for the oscilloscope. The Larmor coil, with center axis perpendicular to the pumping axis, forces mixing of the metastable sublevels when the applied oscillating magnetic field corresponds to the Larmor frequency.
equal sublevel populations, $N^+ = N^0 = N^-$. The Zeeman splitting, $\Delta E = g \cdot H \cdot \mu$ where $g = 2$, $H$ is the strength of the magnetic field and $\mu$ is the Bohr magneton, $\mu = 9.27 \times 10^{-27}$ erg/gauss, corresponds to a Larmor frequency,

$$f = \left( 2.8 \times 10^6 \text{ Hz/gauss} \right) \times H$$

Because the optical signal requires measurement of the light absorption between pumped and unpumped discharge conditions, the following experimental arrangements could provide the desired information.

First, by exciting the Larmor coil at the Zeeman frequency and then applying a square wave voltage to an additional pair of Helmholtz coils with longitudinal axis along the pumping direction, one expects the transmitted light intensity to modulate in step with the coil modulation. However, coil inductance precludes magnetic field modulation at a convenient frequency. The observed optical signal resembled that observed with a sinusoidal magnetic field oscillation which will be discussed presently.

Second, by setting the magnetic field to a constant level and modulating the Larmor coil on and off, one expects the transmitted light intensity to follow the Larmor coil modulation. Observations indicated transmitted light intensity modulation at low discharge intensity even with the DC magnetic field shifted such
that the applied Larmor coil frequency no longer corresponded to the sublevel spacing. The signal observed at low discharge levels therefore could be due to discharge intensity modulation through interaction with the Larmor coil oscillation.

In the third method, similar to the first and illustrated in Figure 5, the Larmor oscillator is set to the Zeeman frequency and the magnetic field oscillated sinusoidally by means of the additional pair of Helmholtz coils. In this case one observes dips in transmitted light intensity corresponding in phase and breadth to that period during which the field at the bulb corresponds to the applied Larmor coil frequency. Figure 6 illustrates the observed signal as monitored on an oscilloscope. However, the Larmor frequency necessary to produce the optical signal was observed to vary with discharge intensity. Calculation of the magnetic field [19] across the diameter of the bulb indicates a field variation of about 1.5%. At the nominal 5 gauss field used in this experiment, the calculated 0.08 gauss variation across the bulb corresponds to 0.2 MHz spread in the Larmor frequency across the bulb. The Larmor frequency drift with discharge intensity was observed to be about 0.2 MHz and thus leads to the conclusion that the optical signal being observed originates predominately from different areas within the
FIGURE 6

Optical signal as observed with an oscilloscope.
LIGHT INTENSITY
AT THE DETECTOR

MAGNETIC FIELD

ΔI

I_u

DETECTOR OUTPUT LEVEL
WITH NO LIGHT ON DETECTOR
bulb for different discharge intensities. This is a result of the D0:D3 ratio of the pumping light being a function of position within the discharge. Therefore, although the optical signal indicates that optical pumping is occurring and is reproducible for the geometry of this particular apparatus, the optical signal does not deliver a quantitative value of the degree of metastable polarization.

Figures 7a through 7k display the observed optical signals as a function of discharge intensity, as indicated by per cent absorption of the 1.08 micron pumping radiation, for various bulb pressures. Bulb pressure measurements were made with a Hastings thermopile pressure gauge tube, DV-6, with corrected helium pressure indicated and the actual gauge reading in parentheses. Because of the importance of geometrical factors, the figures present data which, although very reproducible for this particular apparatus, are not readily transferrable to other experimental geometries.

4. CURRENT MODULATION

Assuming that all ions extracted from the discharge bulb originate from metastable-metastable collisions, as discussed in the earlier section on ion production
FIGURES 7a - 7k

Measured optical signals for various pressures as a function of discharge intensity indicated by percent absorption of 1.08 micron pumping radiation. Indicated ion currents, measured immediately after the gap lens, provide a relative measure between various discharge intensities and helium pressures.
230(200) x 10^{-3} \text{ torr}

OPTICAL SIGNAL $\Delta I/I$ (%) vs.

$\%$ ABSORPTION vs.

CURRENT ($\mu$A)
(80) × 10^{-3} \text{ torr}
mechanisms, Table 2 presents the available production paths with respect to metastable sublevel populations. From this table, the ion current as a function of metastable populations becomes

\[ I = (N_s)^2 + (N_0)^2 + 2(N_0 + N^+ + N^-) N_s + 2(N^+ * N^-) + 2(N^+ * N_0) + 2(N_0 * N^-) \]

Where \( n_i = \frac{(N_i)}{(N_u)} \) and \( N_u \) is 1/3 of the total triplet population, \( n_i \), of the unpumped discharge. For the singlet population, \( n_s = \frac{(N_s)}{(N_u)} \) and is identical for the pumped and unpumped discharges. The unpumped current is

\[ (I')u = (n_s)^2 + 6 ns nu + 7 (nu)^2 \]

The difference in extracted ion current for pumped and unpumped discharges is then

\[ \Delta I' = (I')p - (I')u \]

\[ = -7(nu)^2 - 6 ns nu + (n_0 p)^2 + 2 n_0 p (n_0 p + n^+ + n^-) + 2 (n^+ n^- + n_0 p n^+ + n_0 p n^-) \]

where the prime on \( I' \) indicates that \( I \) has been divided for convenience by \( N_u \); \( p \) and \( u \) indicate pumped and unpumped values of \( n^+ \), \( n_0 \), and \( n^- \). The current modulation normalized to the ion current from the unpumped discharge is then

\[ \frac{-7(nu)^2 - 6 ns nu + (n_0 p)^2 + 2 n_0 p (n_0 p + n^+ + n^-) + 2 (n^+ n^- + n_0 p n^+ + n_0 p n^-)}{I} = \frac{(n_s)^2 + 6 ns nu + 7 (nu)^2}{(ns)^2 + 6 ns nu + 7 (nu)^2} \]

The ion polarization, from Table 2, is (all values
with reference to the pumped discharge)

\[ P(\text{ions}) = \frac{2(\text{ns} + \text{n0})(\text{n}^+ - \text{n}^-)}{(\text{ns} + \text{n}^+ + \text{n0} + \text{n}^-)^2 - (\text{n}^+)^2 - (\text{n}^-)^2} \]

The metastable polarization is given simply by (again all values refer to the pumped discharge condition)

\[ P(\text{triplet}) = \frac{[\text{n}^+ - \text{n}^-]}{[\text{n}^+ + \text{n0} + \text{n}^-]} \]

Note that the ion polarization does not equal the metastable polarization because the singlet population dilutes the polarization. Furthermore, even with zero singlet population, the ion polarization does not reduce to the metastable polarization. For example, note that for no = 0 and ns = 0, the ion polarization becomes zero regardless of metastable polarization. Therefore inference of the ion polarization requires knowledge of the singlet populations.

The experimental configuration, Figure 8, includes a Faraday cup to collect ions extracted from the discharge cell, a rotating quarter-wave plate and a lock-in amplifier. By rotating the quarter-wave plate, the pumping light oscillates between linear and circular polarization and consequently the discharge oscillates between the pumped and unpumped conditions. Therefore, using the rotating quarter-wave plate to generate the reference signal for the lock-in amplifier and measuring the current modulation at that frequency, the amount of
FIGURE 8

Experimental arrangement for measuring current modulation. The rotating quarter-wave plate following the linear polarizer provides pumping radiation which oscillates between linear and circular polarization and thus produces unpumped and pumped discharges. Ions extracted from the discharge enter the Faraday cup and an electrometer provides a voltage proportional to the current. The lock-in amplifier detects the amplitude of current oscillation with frequency and phase determined by the rotating quarter-wave plate. The IR detector and amplifier measure 1.08 micron radiation transmission for percent absorption measurements which characterize discharge intensity.
current modulation may be measured. For these measurements, current modulation was at 20 Hz which was much longer than that time required for metastable polarization relaxation (see section II B).

Figures 9a through 9h present measurements of normalized current modulation as a function of discharge intensity for various pressures. The data indicate that for all pressures, current modulation decreases with an increase in discharge intensity indicating a depolarization of both ions and metastables. Note that the behavior exhibited by the current modulation follows, as expected, the metastable polarization as measured with the color-center laser. Metastable de-polarization at high discharge levels probably reflects the increase in metastable - metastable collision rate, hence decreased metastable lifetime, due to greater metastable density. With removal of the pumping light or magnetic field, or application of the Larmor mixing field, the current modulation was observed to vanish thus assuring that the observed modulation originates from the optically pumped metastables. Indeed the 2(3)S magnetic resonance can be observed by monitoring the current modulation as the Larmor frequency is applied.
FIGURES 9a - 9h

Measured extracted current modulation for various pressures as a function of discharge intensity indicated by percent absorption of 1.08 micron pumping radiation.
5. ESTIMATE OF ION POLARIZATION

Measurements of metastable polarization, optical pumping signals and ion modulation indicate, but do not quantitatively measure, ion polarization. The derived expressions for polarization require knowledge of singlet and n0 triplet sublevel populations if they are to yield polarization values. Best estimates of these populations set the singlet population at a constant value of n = 1/3 and sets the n0 triplet sublevel at the unpumped value of 1/3 where n = (Ni) / Nu as defined earlier. For n0 = 1/3, then n+ and n− are forced to be 1/3 + X and 1/3 − X where X indicates the extent of asymmetry from the normal, unpumped triplet sublevel population introduced by optical pumping. With this estimate, the expressions for polarization and current modulation reduce to

$$\Delta I / I = (-9/7) \cdot (X)^2$$

$$P \text{ (ions)} = (12 \cdot X) / [7 - 9 \cdot (X)^2]$$

Setting ΔI/I at the measured value of 0.005, the ion polarization and metastable polarization estimates become 11% and 12% respectively with, as anticipated, the ion polarization less than the metastable polarization. The estimate for metastable polarization approximates those values found by the color-center laser absorption experiment and the estimate of ion polarization also
approximates the electronic polarization observed from a similar discharge source [7].

The calculation of ion polarization becomes feasible with knowledge of the singlet and NO populations. Without such information, only estimates of ion polarization may be made. However, the suitability of setting \( n0 = 1/3 \) may be explored through the following experiment. With circularly polarized light from the color-center laser probe, \( (n^+ - n^-) \) has been determined and was used to determine the metastable polarization. With linear laser light, the selection rules are such that \( (n^+ + n^-) \) may be determined. With the same modulation arrangement as before, the absorption modulation of the linearly polarized laser probe beam indicates the degree of population asymmetry between the \( n^+ \) and \( n^- \) sublevels. With no or little modulation detected, the \( n^+ = 1/3 + X \) and \( n^- = 1/3 + X \) approximation is confirmed. Furthermore, along with \( n^+ + n^- + n0 = 1 \), exact determination of \( n^+ \), \( n0 \) and \( n^- \) becomes possible with this additional measurement. However, the unsolved singlet population remains and prevents calculation of ion polarization. This population can be determined by measuring absorption of \( 2(1)S \rightarrow 2(1)P \) radiation at 2.06 microns. While this measurement was not undertaken in the present study, earlier work on similar helium discharges suggests that the
singlet-to-triplet population ratio is indeed in the vicinity of the value of 1/3 assumed above.

Another method for determination of ion polarization involves production of metastable helium atoms by charge exchange between the helium ions and alkali metal atoms [44]. Charge exchange between He+ and Na has been shown to produce a beam of triplet helium metastable atoms. Subsequent Stern-Gerlach analysis of the metastables, as done with the helium metastable beam source accompanying this ion source [40], would then allow calculation of ion polarization. However, this experiment requires a crossed beams configuration and an additional Stern-Gerlach apparatus and was therefore not attempted with this source.
III

ION EXTRACTION, TRANSPORT AND DECELERATION

Ions created within the discharge bulb must be extracted and formed into a beam for transport to the target surface. Because the pressure within the bulb is about nine orders of magnitude greater than the pressure in the target chamber, transport of the ion beam requires moving the beam through a series of differential pumping regions. Furthermore, the desire for a low energy beam incident upon the target together with the necessity of high energy beam transport dictates the need for deceleration optics in the final target chamber. The following sections discuss beam extraction, differential pumping, transport optics and deceleration.

A. EXTRACTION CANAL AND GAP LENS

The ion source, diagrammed in Figure 10, consists of a three inch diameter pyrex glass bulb with an evacuation line for cleaning, a tungsten anode for setting the discharge plasma potential and mounting tube for mating to the extraction optics. The evacuation line for cleaning consists of a 3/8 inch pyrex tube which, through a
FIGURE 10

Discharge bulb, mounting plate and extraction optics.
shut-off valve, leads to the first pumping chamber. During the cleaning process, described earlier, and during normal operation, the open valve allows continuous helium flow through the bulb to prevent contaminant accumulation.

The discharge bulb is mounted upon a Macor (machinable Corning glass) plate with a Maycor tube extending into the discharge region [20, 21, 22, 23, 24, 25, 44]. The Macor plate fits in turn onto an aluminum base plate which is o-ring vacuum sealed to the bulb. The aluminum extraction canal extends from the aluminum base into the Macor tube. In operation, a potential of between 1000–2000 volts across the tungsten anode and aluminum base plate forms an electric field whose polarity determines whether electrons or ions are extracted. The Macor plate shields the discharge from the aluminum base plate. The Macor tube surrounding the metal extraction canal charges so as to form a lens which focuses the ion beam through the extraction canal [23, 25]. With the discharge lit and potentials applied, a dark dischargeless region may be observed forming an umbrella over the Macor tube and extraction canal. Ions entering the extraction canal have kinetic energy equal to the potential across the bulb and potential energy, with respect to ground, equal to the anode potential.
After passing through the canal, ions encounter a gap lens [23, 25] which produces a tight parallel beam of about 2 mm in diameter. This parallel beam next enters the transport optics necessary to pass the beam through the differential pumping stages and into the ultra-high vacuum target chamber.

B. DIFFERENTIAL PUMPING AND TRANSPORT OPTICS

1. DIFFERENTIAL PUMPING

As illustrated in Figure 11, three regions of differential pumping exist in the apparatus: first, between the discharge bulb and the first chamber; second, between the first chamber and the intermediate chamber; and third, between the intermediate chamber and target chamber. Typical pressures are 0.1 torr of helium in the discharge bulb, $5 \times 10^{-7}$ torr in the first chamber, $1 \times 10^{-9}$ torr in the intermediate chamber and low $10^{-10}$ torr in the target chamber.

A six inch diffusion pump with water cooled baffle pumps the first chamber at 900 and 1125 liters/sec for air and helium respectively. With typical operating pressures of about 0.1 torr of helium in the discharge bulb and the evacuation line open to the first chamber, the first
FIGURE 11

Beam transport optics and differential pumping diagram.
chamber pressure rises less than 20% above the base pressure. At bulb pressures greater than 1 torr, chamber pressures in excess of $10^{-5}$ torr have been observed with a corresponding increase in backing line pressure.

The intermediate chamber is pumped through a bellows by another six inch diffusion pump with refrigerated helium cold trap. A 2 mm diameter aperture separates the first and intermediate chambers. The conductance of a circular aperture under conditions of molecular flow at 20°C may be calculated from [26]

$$C_{\text{air}} = 9.16 \times D^2 \text{ liters/sec}$$

$$C_{\text{He}} = 24.5 \times D^2 \text{ liters/sec}$$

for diameter $D$(cm). The conductance for this particular aperture is then 0.37 and 1.0 liters/sec for air and helium respectively. The throughput, $Q$, between chamber one with pressure $P_1$ and chamber two with pressure $P_2$ is given by

$$Q = C (P_1 - P_2)$$

With conservative pressures in the first chamber of $P(\text{air})$ and $P(\text{He})$ of $10^{-6}$ torr and $5 \times 10^{-6}$ torr respectively and intermediate chamber pressure assumed much lower than this, the throughput becomes less than $4 \times 10^{-7}$ and $5 \times 10^{-6}$ torr-liters/sec of air and helium respectively.

The conductance of the bellows connecting the intermediate chamber to the diffusion pump may be
approximated by the conductance of a long tube,

\[ C_{\text{air}} = 12.1 \, (D)^3 / L \]

\[ C_{\text{He}} = 32.6 \, (D)^3 / L \]

for diameter \( D \text{(cm)} \), length \( L \text{(cm)} \) and conductance \( C \text{(liters/sec)} \). The bellows, about 2.25 inches in diameter and 10 inches long, then has a conductance of about 90 and 240 liters/sec for air and helium respectively. Using these values for the pumping speed in the intermediate chamber, predicted pressures of \( 4 \times 10^{-9} \) and \( 2 \times 10^{-8} \) torr of air and helium are calculated and agree closely with measured pressures.

The aperture between intermediate and target chamber, which also has a 2 mm diameter, is characterized by a conductance of 0.37 and 1.0 liters/sec for air and helium respectively. With intermediate chamber pressure of \( 4 \times 10^{-9} \) and \( 2 \times 10^{-8} \) torr, the throughput becomes \( 1.5 \times 10^{-9} \) and \( 2 \times 10^{-8} \) torr-liters/sec of air and helium respectively. The ion pump for the target chamber has a speed of 500 and 150 liters/sec for air and helium. The calculated target chamber pressure is then \( 3 \times 10^{-12} \) and \( 1.3 \times 10^{-10} \) of air and helium.

These calculations begin with conservative first chamber pressures and conclude with target chamber throughputs which indicate excellent differential pumping. Furthermore, actual pressure measurements, which
show no increase in target chamber pressure upon opening the gate valve between first and intermediate chamber, substantiate this conclusion. With this information and the evidence to be presented in the following section concerning the loss of beam current at the first aperture, future consideration should be given to increasing the diameter of the apertures to increase the current delivered to the target. All apertures may be easily, although tediously, replaced as the situation may demand.

2. ELECTROSTATIC OPTICS

Figure 11 also depicts the electrostatic optics configuration for transport of the ion beam to the target chamber. Ion optics, which behave identically to electron optics except that the signs of applied voltages are reversed, may be designed using various electrostatic optics tables [27, 28]. The following section describes the operation of the transport optics.

After passing through the gap lens, the ions form a parallel beam with diameter of about 2 mm which enters the first einzel lens. The einzel lens system in the first chamber focuses the beam through the aperture between the first and intermediate chambers. Before and after the lens system, beam steering is accomplished by two
orthogonal pairs of plates parallel to the beam axis with voltages applied such that the two plates opposite each other are $+\Delta$ and $-\Delta$ from the beam potential. By selection of the steering voltages, the beam may be moved up-and-down and left-and-right and hence be steered through the einzel lens and apertures.

Inside the intermediate chamber, the second einzel lens takes the diverging beam which was focused through the aperture and reproduces a parallel beam. This parallel beam subsequently enters the second einzel lens which focuses the beam through the aperture between the intermediate chamber and the target chamber. Within the intermediate chamber, steering plates allow fine adjustment of beam entrance into the einzel lenses and through the aperture.

Upon entering the target chamber, the beam passes through the last einzel lens which again reforms a parallel beam before entrance into the deceleration column. The deceleration column, which will be discussed in detail in the following section, reduces the ion energy before incidence upon the target surface. Again steering plates are available for fine adjustment.

The voltages required for delivery of the ion beam to the target are given in Figure 11. Because of close alignment of the differential pumping chambers and optics,
steering of the beam is quite simple. In operation, only those plates in the first chamber, which take the beam from the gap lens, launch it into the first einzel lens and then steer the beam through the first aperture, required application of the \( A \) voltage in order to place the beam on target. However, all steering plates were able to move the beam sufficiently off axis to stop the beam from passing through the chambers.

For 200 to 500 volt ions, beam transmission through the entire system, including deceleration column and into a Faraday cup located in the target chamber, is estimated to lie between 25 and 50%. The uncertainty in transmission is due to difficulties in estimation of the initial beam current in the first chamber without the use of a Faraday cup. With the current configuration, placement of a Faraday cup in the first chamber requires breach of vacuum. However, because the transport optics appear to work satisfactorily and first chamber beam currents may be obtained within factors of two, their measurement was not deemed essential. The major transmission loss results from beam divergence at the aperture between first chamber and target chamber caused by an electrostatically unshielded region just upstream of the aperture where a gate valve is installed. The convenience of the valve outweighs the loss of current
because the valve allows the ion source to be isolated from the rest of the vacuum system. Transmission losses at the other aperture and in the deceleration column account for about 20% of the total transmission loss.

C. ION BEAM DECELERATION

Transport of the ion beam through the various differential pumping stage apertures requires higher beam energies than those desired at the target. Currents on the target crystal are measured with an electrometer. Experimental considerations dictate that the electrometer be grounded rather than placed at high potential. The tungsten anode on the ion source sets the discharge potential relative to the grounded target crystal and thus the incident ion energy. However, extraction from the discharge and transport through the differential pumping apertures requires much higher beam energies (see Figure 11), and as a result deceleration of the ions prior to incidence upon the target becomes necessary.

1. EXPONENTIAL DECELERATION

Beam deceleration, in general, is accompanied by beam divergence. Therefore various deceleration configurations
have been considered to minimize beam divergence and place as many ions upon the target as possible. One particular deceleration arrangement employs a series of apertures with equidistant separation. The voltage upon each aperture is such that the deceleration voltages exponentially decrease with distance from the first element to the last. Thus for voltage \( V \) on the first aperture, the voltage on any subsequent aperture distance \( x \) from the first will be given by \( V \exp(-Ax) \) where \( A \) depends upon the length of the aperture column.

Under the paraxial approximation [29] which requires that both the radial distance from the cylindrical axis and \( dr/dz \) be small with respect to lens dimensions, the exponential deceleration column has been predicted to focus the beam at various distances from the column entrance [30]. However, detailed analysis of beam trajectories shows this assertion to be incorrect. The following analysis begins with Laplace's equation and ends with plots of the particle trajectories for an exponential deceleration column.

Laplace's equation in cylindrical coordinates, with the appropriate gauge and axial symmetry becomes

\[
\frac{\partial^2 \phi}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial \phi}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2 \phi}{\partial \phi^2} + \frac{\partial^2 \phi}{\partial z^2} = 0
\]

\[
\nabla^2 \phi = 0
\]

\[
\frac{\partial^2 \phi}{\partial \phi^2} = 0
\]
\[
\frac{\partial^2 \phi}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial \phi}{\partial \rho} + \frac{\partial^2 \phi}{\partial z^2} = 0
\]

Assuming a product type solution, the equation may be separated as follows:

\[
\phi = R(\rho) Z(\zeta)
\]

\[
Z(\zeta) \frac{\partial^2}{\partial \zeta^2} R(\rho) + Z(\zeta) \rho \frac{\partial^2}{\partial \rho^2} R(\rho) + R(\rho) \frac{\partial^2}{\partial z^2} Z(\zeta) = 0
\]

\[
\frac{\partial^2}{\partial z^2} Z(\zeta) = k^2 \quad \frac{\partial^2}{\partial \rho^2} R(\rho) + \frac{1}{\rho} \frac{\partial^2}{\partial \rho^2} R(\rho) = -k^2
\]

Expand \( \phi \) in a series dependent upon the axial potential

\[
\phi(\zeta, \rho) = b_0(\zeta) + b_2(\zeta) \rho^2 + b_4(\zeta) \rho^4 + \ldots
\]

\[
\frac{\partial^2 \phi}{\partial \zeta^2} = b'_0(\zeta) + b'_2(\zeta) \rho^2 + b'_4(\zeta) \rho^4 + \ldots, \quad \phi'' = \frac{\partial^2 \phi}{\partial \zeta^2}
\]

\[
\frac{\partial \phi}{\partial \rho} = 2 b_2(\zeta) \rho + 4 b_4(\zeta) \rho^3 + \ldots
\]

\[
\frac{\partial^2 \phi}{\partial \rho^2} = 2 b_2(\zeta) + 12 b_4(\zeta) \rho^2 + \ldots
\]

and note

\[
b_0(\zeta) = \phi(\zeta)\bigg|_{\rho=0} \equiv \phi_0(\zeta)
\]

Equating coefficients allows the following
simplification:

\[ b_n(z) = \frac{(-1)^n}{2^24^2\cdots(2n)^2} \phi_n^2(z) \]

\[ \phi(z, \rho) = \phi_0(z) - \frac{\rho^4}{4} \phi_4^2(z) + \frac{\rho^4}{4^3} \phi_6^2(z) + \cdots \]

\[ \phi_0^2(z) = \frac{\alpha}{\beta^2} \phi_0(z) \]

For a force \( F = +q \mathbf{E} \) on the charged particle,

\[ F_\rho = -q \frac{\partial \phi}{\partial \rho} = 0 \Rightarrow F_\rho = 0 \]

\[ F_\rho = -q \frac{\partial \phi}{\partial \rho} = m \dot{\rho} \]

\[ F_\rho = -q \frac{\partial \phi}{\partial \rho} = m (\ddot{\rho} + \rho \dot{\rho}^2) \]

Equating kinetic energy and field potential, measured relative to that potential at which the particles have zero kinetic energy, allows the following simplification for the axial velocity:

\[ -q \phi = \frac{1}{2} m (\dot{z}^2 + \dot{\rho}^2) \]

\[ \dot{z}^2 = -\frac{q}{m} 2 \phi - \dot{\rho}^2 \]

\[ = -\frac{q}{m} 2 \phi \left[ 1 + (\rho^2)^2 \right]^{-1} \]

Appropriate manipulation yields the expressions for the axial and radial velocities:
\[ \ddot{\rho} = \frac{d}{dt} \dot{\rho} = \frac{d}{dt} \left( \dot{\rho} - \frac{2}{3} \dot{\rho} \right) = \dot{3} \rho + \ddot{\rho} \frac{m \ddot{z}}{g} = -\frac{2}{3} \frac{\dot{\rho}}{3} \]

\[ F_\rho = m \ddot{\rho} = -g \frac{\ddot{\rho}}{3} \rho = m \left( 3 \rho + \ddot{\rho} \frac{\rho}{m} \right) \]

\[ \frac{2}{m} \frac{\ddot{\rho}}{\rho} = \frac{2}{m} \rho \frac{\ddot{\rho}}{\rho} + \frac{1}{m} \frac{\ddot{\rho}}{\rho} \left[ 1 + \left( \frac{\rho}{m} \right)^2 \right]^{-1} \]

\[ D = \left[ 1 + \left( \frac{\rho}{m} \right)^2 \right] \left\{ \rho \frac{\ddot{\rho}}{\rho} - \frac{\ddot{\rho}}{\rho} \right\} + 2 \frac{\ddot{\rho}}{\rho} \rho \]

\[ D = \rho \frac{\ddot{\rho}}{\rho} + \frac{1}{2} \frac{\ddot{\rho}}{\rho} \left\{ \rho \frac{\ddot{\rho}}{\rho} - \frac{\ddot{\rho}}{\rho} \right\} \]

The general equation of motion as a function of potential may now be written as

\[ \frac{d^2 \rho}{d z^2} = \frac{1 + \left( \frac{\rho}{m} \right)^2}{2 \rho} \left\{ \frac{\ddot{\rho}}{\rho} - \frac{\ddot{\rho}}{\rho} \frac{\ddot{\rho}}{\rho} \right\} \]

Because the potential has been expanded as a series dependent upon the potential at the axis, various approximations may be made:

\[ \frac{3 \ddot{\rho}}{2 \rho} \sim \frac{3 \ddot{\rho}}{2} \frac{d^2}{d z^2} \phi(z) \]

\[ \frac{\ddot{\rho}}{\rho} \sim \frac{3 \ddot{\rho}}{2} \frac{d^2}{d z^2} \phi(z) \]

Substitution into the equation of motion yields
\[
\frac{d^2\rho}{dz^2} = \frac{1+(\frac{d\phi}{dz})^2}{2\Phi} \left\{ \frac{1}{2} \frac{d^2\Phi}{dz^2} - \frac{d\rho}{dz} \frac{d\Phi}{dz} \right\}
\]

Assuming the particles remain very close to the axis,

\[
\left(\frac{d\Phi}{dz}\right)^2 << 1 \quad \Rightarrow \quad \Phi \approx \Phi_0
\]

The equation of motion for the particles becomes

\[
\frac{d^2\rho}{dz^2} + \frac{\rho}{4\Phi} \frac{d^2\Phi}{dz^2} + \frac{1}{2\Phi} \frac{d\Phi}{dz} \frac{d\rho}{dz} = 0
\]

For an exponential field along the axis,

\[
\Phi_0 = C \exp\left(-\alpha z\right)
\]

the equation of motion, for particles remaining very close to the axis, is

\[
\frac{d^2\rho}{dz^2} + \frac{\rho}{4} \alpha^2 - \frac{\alpha}{2} \frac{d\rho}{dz} = 0
\]

with solution

\[
\rho = C \exp\left(\frac{\alpha z}{4}\right) \sin\left(\frac{\sqrt{3} \alpha z}{4} + \delta\right)
\]

\[
\rho_0 = C \sin \delta
\]

where \( \delta \) is the initial launch angle measured from the z axis and \( r_0 \) is the initial radial distance from the z axis. Figure 12 plots \( r \) as a function of \( z \). These equations do predict beam focusing but the following analysis indicates that the approximations are too
FIGURE 12

Trajectories for charged particles in the non-physical radially independent exponential deceleration potential.
\[
\phi = U_0 + U \exp(-\alpha z)
\]

\[
U_0 = 0
\]

\[
\alpha = \frac{4\pi}{\sqrt{3} z_f}
\]
restrictive for real beams.

Assuming the axial potential to be given, the potential at any distance \( r \) from the axis may be calculated from the series expansion of \( \Phi \) given above. Furthermore, the acceleration due to the potential may be evaluated and the incremental motion of the particle as a function of time calculated:

\[
\begin{align*}
\ddot{z}_0 &= \frac{e}{m} \frac{\partial}{\partial z} \Phi |_{z_0} \\
\dot{z}_0 &= \dot{z}_0 + \ddot{z}_0 \Delta t \\
\ddot{z}_1 &= \ddot{z}_0 \Delta t \\
\dot{z}_1 &= \dot{z}_0 + \ddot{z}_0 \Delta t \\
\Delta s &= \frac{\dot{z}_0}{\ddot{z}_0} \\
\end{align*}
\]

The initial particle trajectory is defined by the initial launch angle and velocity. Iteration allows the path of the particle to be evaluated in increments of time \( \Delta t \).

For the exponential axial potential,

\[
\Phi(z, \rho) = U_0 + U \exp(-\alpha z)
\]

the potential throughout space may be evaluated from the series,

\[
\Phi(z, \rho) = U_0 + U \exp(-\alpha z) + \sum_{m=1}^{\infty} \frac{(-1)^m (\rho \alpha)^{2m}}{2^m (m!)^2} U \exp(-\alpha z)
\]

from which the required derivatives may be found.
Trajectories for particles initially off-axis and with various initial velocity vectors are plotted in Figures 13a and 13b. Note that no focusing occurs. Each point represents ten time increments, thus ten iterations between points, and the particle's speed along the trajectory may be inferred from the separation of the points. The complex behavior of the trajectories depends upon the velocity component parallel to the electric field. For an exponential axial field, the off-axis field is divergent. The complex behavior of these trajectories compared to those originally predicted lies in the unphysical assumption that the field everywhere is parallel to the axis. These calculations show that this assumption is valid only for perfectly on-axis initial particle trajectories. Any particle entering off-axis immediately encounters a defocusing field. Thus, another field configuration was sought.

2. LINEAR DECELERATION

Substitution of any axial potential into the equation of motion derived above allows calculation of particle trajectories. This section examines the characteristics of a linear deceleration potential.
FIGURES 13a and 13b

Trajectories for charged particles in an exponential deceleration potential.
\[ \phi \big|_{\rho_0} = U_0 + U \exp(-\alpha z) \]

\[ U_0 = 0.01 U \]

\[ \alpha = \frac{4\pi}{\sqrt{3}} z_f \]
The linear potential may be expressed as
\[ \phi = U_i \left[ 1 - \left( \frac{z}{z_f} \right) \right] + U_0 \]
where \( z \) indicates the axial distance, \( z_f \) the point at which the potential becomes \( U_0 \), \( U_i \) the initial potential and \( U_0 \) a constant offset potential.

Figures 14a through 14c plot these trajectories, using iteration, for various initial conditions and field configurations. Note that because the potential remains constant for any \( z \) regardless of radial distance, the initial radial offset appears as a constant radial shift of the trajectories. Furthermore, particles with non-zero launch angle never reach the potential plane equal to the particle's initial energy because the axial component of the energy determines the maximum potential the particle may reach. The velocity component perpendicular to the axis, unaffected by the potential, remains constant and gives the particle a constant speed perpendicular to the axis.

3. THE DECELERATING COLUMN

A series of fifteen thin stainless steel plates with quarter-inch separation forms the deceleration column (see Figure 15). Each plate mounts into an outer ring which mates, with sapphire ball insulators, to other aperture
FIGURES 14a, 14b and 14c

Trajectories for charged particles in a linear deceleration potential.
\[ U_0 = 0 \]
\[ \phi = U \left( 1 - \frac{Z}{Z_f} \right) + U_0 \]
\[ U_0 = 0.05U \]
\[ \phi = U \left( 1 - \frac{Z}{Z_f} \right) + U_0 \]
\[ U_0 = 0.1U \]
\[ \phi = U \left(1 - \frac{Z}{Z_f}\right) + U_0 \]
FIGURE 15

Deceleration column assembly diagram.
rings on both sides, thus forming a sandwich of apertures. The entire column slides into a stainless steel shielding tube. Holes in the side of the outer tube allow electrical connection to the individual plates. A twenty pin feedthru provides the outside deceleration voltage connection.

Column potentials were initially set to form a linear deceleration potential. However ions incident upon the first aperture produced quantities of electrons which, because the column decelerates ions, accelerated through the column. Setting the second aperture at a potential lower than the first aperture potential eliminated this problem. No electron current, as measured with a Faraday cup biased to repel all incident ions, was then observed to exit the column, thus indicating that few ions hit the inner apertures and/or that the plates act as a baffle to those electrons originating within the column. Transmission through the column was measured to be in excess of eighty percent of the ion current measured without deceleration.

Having achieved low energy ion delivery to the target chamber, target surface characterization becomes important. The following chapter discusses surface characterization using Auger ejection electron energy analysis.
IV
SURFACE CHARACTERIZATION —
THE AUGER ANALYSIS SYSTEM

To perform meaningful surface physics experiments, the conditions at the surface, specifically surface contamination, must be known. In ion neutralization studies of a nickel surface for example, the incoming ions may interact with the everpresent adsorbed carbon atoms rather than the desired nickel atoms. Furthermore, contaminants present in the bulk of any target crystal can segregate to the surface and interact with the ions. Contaminants alter the surface electronic configuration which this apparatus is designed to probe and thus both crystal cleaning and diagnostic capabilities are necessary.

Auger electron spectroscopy allows elemental analysis of the surface by examining the energies of electrons ejected from the target due to Auger processes following inner-shell ionization by an incident electron beam. After inner-shell ionization, an electron from an outer shell fills the inner shell vacancy within $10^{\pm 17}$ to $10^{\pm 12}$ seconds. The energy lost in this process produces either X-radiation or may be given to an outer-shell
electron which then may be ejected from the crystal surface. The energy of an ejected electron is equal to the energy released in the inner-shell decay process and hence is characteristic of the atom initially excited. Therefore, the energy spectrum of those electrons, emitted from a crystal surface following excitation by an incident electron beam will reflect the elemental composition of those atoms near the surface.

A. AUGER ANALYSIS INSTRUMENTATION

The Auger analysis system (see Figure 16) consists of three distinct parts: the electron gun which forms the beam of energetic electrons to excite the target atoms, the target surface and the ejected electron energy analysis system. The analysis system uses a combination LEED-Auger analysis system manufactured by Physical Electronics Industries, Inc., a division of Perkin-Elmer. The analysis configuration has the target centered on a group of four hemispherical grids and a hemispherical screen. An electron gun, trained on the target crystal through the screen and grids, bombards the target crystal with electrons of energy variable between 2000 and 5000 volts with respect to the grounded target. Electrons emitted from the surface toward the grids travel first
FIGURE 16

Schematic of the Auger analysis system.
through the equipotential region between the target surface and the first grid. After passing through the first grid, the electrons encounter the second and third grid which are at the same potential and provide a retarding potential through which those electrons with energy less than the retarding potential cannot pass. The fourth grid, set at ground potential, provides field symmetry, shields the screen from the retarding potential and suppresses electron ejection from the screen. The screen thus collects electrons which pass through the retarding grid potential and gives a current dependent upon the energy of the incident electrons.

The screen current versus the retarding potential gives the integral electron energy distribution curve. For any particular retarding potential, a point on the integral curve plots the number of electrons with energy greater than the potential of the retarding grids. Actual integral curves from Auger analysis, however, are extremely flat and featureless and thus difficult to analyze. The first derivative of the integral curve exhibits peaks indicating the energy at which the electrons leave the target. However, these peaks are rather small and appear upon a large continuous background.
The second derivative curve is the curve from which analyses may be made. The second derivative erases the continuous background of the first derivative and at each peak of the first derivative, the second derivative displays a distinctive positive and negative spike with a central pass through zero. Comparison with published Auger second derivative curves [31] allows identification of the Auger peaks and therefore surface contaminants.

B. AUGER ANALYSIS CONTROL SYSTEM

The Auger control system (see Figure 16) contains three major components: electron gun power and control, retarding potential voltage supply, and curve differentiation system. The electron gun circuitry consists of a filament current source provided by a commercial current supply and electrostatic lens voltage supplies derived from a commercial high voltage supply and resistive divider. The second derivative curve has, as its ordinate, the retarding grid voltage and, as its abscissa, the second derivative of the current to the screen. This curve may be produced by two methods, one analog and the other digital. The following sections present both methods with emphasis upon the digital system constructed for this apparatus.
1. ANALOG AUGER ANALYSIS

The analog system produces the second derivative of the integral curve by the following method. A sinusoidal voltage modulation with frequency \( f \) superimposed upon the retarding voltage produces a screen current modulation of frequency \( f \). The amplitude of the screen current at frequency \( f \) is directly proportional to the first derivative and the second derivative is directly proportional to the current at frequency \( 2f \).

\[ I(V) = \text{current to screen when retarding voltage is } V \]
\[ N(E) = \text{number of electrons with energy } E \text{ from the target} \]
\[ V = \text{retarding potential} \]
\[ E_0 = \text{energy of electrons incident upon target} \]

\[ I(V) = \int_{V}^{E_0} N(E) \, dE \]

\[ I(V + dV) = I(V) + dV \left[ I'(V) + (dV)^2 \frac{I''(V)}{2!} + \ldots \right] \]
\[ dV = A \sin(wt) \]

\[ I(V + dV) = I(V) + N(V) \left[ A \sin(wt) \right] \]
\[ - \frac{dN}{dV} \left[ \frac{A^2}{2} \right] \cos(2wt) + \ldots \]
2. THE DIGITAL CONTROL SYSTEM

The digital control system, Figure 17, provides the potential for the retarding grids and the necessary modulation for obtaining the second derivative curve. The following section describes the digital processing involved and will be rather superficial of necessity; the actual digital circuitry involves over 100 integrated circuits, 800 hundred feet of wire and 1200 interconnections.

Processing begins with the screen current where an operational amplifier electrometer converts the screen current into a voltage proportional to the current. A voltage to frequency converter in turn converts the electrometer voltage into a frequency proportional to the collector current.

For all curves, integral and first and second derivatives, a voltage step generator provides the potential applied to the retarding grid. The generator circuit (see Figure 17) contains a clock, step counter, digital-to-analog converter and analog amplifier. Each step of the generator, controlled by the clock, corresponds to one count in the step counter, which in turn increments the converter voltage and therefore the
FIGURE 17

Block diagram of the digital analysis system.
retarding potential. Production of the integral curve involves stepping the retarding potential in time and integrating the pulses produced during each step. By holding the potential constant for time \( T \) and counting pulses from the I/V/F converter during this time, the integral curve, to be plotted by an X-Y chart recorder, is obtained by applying the count accumulated during time \( T \) to a digital to analog conversion IC. Application of this analog output to the \( Y \) axis and the retarding potential to the \( X \) axis produces the integral curve.

Production of the derivative curve requires introduction of a voltage modulation upon the retarding potential. With the digital system, the modulation is a square wave of amplitude \( \Delta V \) and frequency \( f \). At any particular retarding potential \( V \), the derivative may be found by allowing a counter, the derivative counter, to count up the I/V/F pulses, while the square wave modulation is high, \( V + \Delta V \), and to count down the pulses while the modulation is low, \( V - \Delta V \). The measured derivative is thus the change in count rate between retarding potentials \( V + \Delta V \) and \( V - \Delta V \). A plot of derivative counter counts versus retarding potential produces the derivative curve.

The second derivative may then be found from the difference between the first derivative of two adjacent
retarding potential voltage steps. The following sequence
yields the second derivative. With the ramp at step Sn,
the first derivative counter, D1, counts up and counts
down with the applied square wave modulation as described
above. After a prescribed number of square wave
modulation cycles during ramp step Sn, the following
processing sequence begins. First, the counts in C1, the
first derivative at ramp step Sn, presets another counter,
C2. Also the counts in C1 move into a latch, L1, which
feed a digital to analog converter and provide the abscissa
for the first derivative curve. Second, C1 resets to
zero, the ramp moves to the step Sn + 1 and the processing
sequence ends. The modulation resumes and C1 finds the
first derivative for step Sn + 1. C2, which is preset to
the previous first derivative, counts up while the square
wave modulation is low and counts down while the
modulation is high. This out-of-phase counting of the
I/V/F pulses by C2 with respect to C1 causes C2 to produce
the second derivative. During the processing sequence,
the contents of C2 move into latch L2 which determines the
output of another digital to analog converter and yields
the second derivative for a chart recorder. C2 presets
again to the count in C1, then the retarding potential
increments to the next step and the cycle repeats.
Figure 18 displays an Auger second derivative curve taken with the digital system of a heavily carbon contaminated nickel crystal. Random noise, particularly room vibration, with period greater than the time spent on one retarding voltage step will be averaged to zero because the up-down counting sequence accumulates an identical number of counts-up as counts-down. With low differential count rates, long dwell times produce more total differential counts and thus more sensitivity. Sensitivity may also be increased, at the expense of energy resolution, by increasing the modulation amplitude because the modulation amplitude determines, for a constant retarding potential, the differential count rate. Energy resolution depends upon the magnitude of the retarding voltage step. With smaller steps and the same dwell time on each step, resulting in a longer time for the scan of a set energy range, derivative evaluation occurs more often which yields greater energy resolution. Typical measurements required a ten volt peak-to-peak modulation, ten volt step increments and three second dwell times.

Because the modulation and step generation have digital form, the system handles inherently digital information. Although the ejected electrons form an analog signal, early digitization allows nearly complete
FIGURE 18

Auger electron second derivative curve taken with the digital analysis system of a carbon contaminated nickel surface.
10 V p-p modulation
2.5 sec/step

100 V  SULFUR  CARBON  400 V
digital analysis. With this design future interfacing of the control system to a digital computer becomes straightforward and desirable.
ION NEUTRALIZATION AND METASTABLE DE-EXCITATION SPECTROSCOPIES

The electronically spin-polarized ion beam was developed for study of the electronic structure of metal surfaces. The following section presents a qualitative description of ion neutralization spectroscopy (INS) and the associated metastable de-excitation spectroscopy (MDS).

Oliphant and Moon [32] reported in 1930 on "the liberation of electrons from metal surfaces by positive ions" and presented an early theoretical treatment for the ejection of secondary electrons [33, 34]. In their theory they proposed that the neutralization of incident ions induced field emission of secondary electrons and they were the first to suggest a resonant capture tunneling mechanism as an alternative to ion induced field emission. Cobas and Lamb [35] presented in 1944 both theory and experimental evidence for a two step process of resonance neutralization of the incoming ion followed by Auger de-excitation of the neutralized, excited atom. Schekhter [36] proposed in 1937 an Auger neutralization process which behaved as a one step direct neutralization
process. Beginning in the early 1950's, Hagstrum began
work with slow noble gas ions incident upon various metal
surfaces. In 1954, he published a detailed article
presenting a theory of electron ejection from metals
[37]. As the secondary electron ejection processes were
studied, spectroscopy of the ejected electrons associated
with ion neutralization evolved into a method for probing
the electronic structure at the surface of a solid [38].
This spectroscopy forms the basis for the proposed method
for studying the spin dependences of ion interactions at
surfaces.

A. INTERACTIONS BETWEEN THE METAL AND INCIDENT
IONS OR EXCITED NEUTRAL ATOMS

Figure 19 illustrates the possible electronic
interactions between a metal surface and incident helium
ions or helium metastable atoms. The figure, commonly
referred to as the triangle diagram, shows four distinct
interactions involving the three states of the atom-metal
system. These three states are presented in notation
similar to that of a chemical equation. The symbols X+, X*
and e(m)—represent an ion, a metastable and an
electron in the metal, respectively and the prefixes to
FIGURE 19

INS and MDS triangle diagram.
e(m) - indicate the number of electrons present in the
metal. Note that horizontal arrows involve no change in
the total number of electrons in the atom-metal system,
while arrows with a vertical component represent processes
in which an electron, indicated by e(e) -, may be ejected
from the system. Note that only Auger processes, not the
resonance processes, produce ejected electrons. The four
interaction processes will now be discussed.

Figures 20, 21 and 22 qualitatively illustrate the
four possible transitions: resonance neutralization,
resonance ionization, Auger de-excitation and Auger
neutralization. In each figure the potential well to the
left represents the metal and the potential well to the
right represents the incoming ion or neutral atom. The
horizontal line above the two potential wells indicates
the vacuum level potential, VL. The metal, assumed to
have a smooth, structureless surface, is characterized by
ϕ, the work function, and ε₀, the bottom of the conduction
band lying between ϕ and ε₀. The incoming ion or
metastable is characterized by Ei’, the ionization
potential, and Ex’, the energy of an excited state above
the ground state. Interactions between the incoming ion
or atom and the metal alter slightly these energies from
those of an isolated ion or atom [37, 38, 39]. Primed Ei
FIGURE 20, 21 and 22

Resonance neutralization and resonance ionization, Auger neutralization, and Auger de-excitation.
and $E_x$ indicate the ionization potential and excited state energy for that separation at which the transition occurs. In the figures solid circles represent the positions of electrons before the transition and open circles represent vacancies before the transition. The electrons in the conduction band form a continuum of energy states between $\phi$ and $E_0$.

In the resonance neutralization process an electron at energy $\alpha$ below $VL$ resonantly tunnels to a corresponding level of the incoming ion $\alpha$ below $VL$. For this process to occur, an energy level of the neutral atom must have an energy state resonant with the energy of the tunneling metal electron. Therefore resonance neutralization requires

$$E_0 < E_{\text{I}' - E_{\text{x}}'} < \phi$$

Resonance ionization, the reverse process of resonant neutralization, produces ionized atoms from incoming excited neutral atoms. An electron from the neutral atom resonantly tunnels to an unoccupied level above the conduction band in the metal. Hence resonance ionization requires

$$E_{\text{I}' - E_{\text{x}}'} > 0$$

Resonance ionization and resonance neutralization are represented by the horizontal lines at the top of the
triangle diagram.

The other two processes, Auger neutralization and Auger de-excitation, each require simultaneous radiationless transitions involving two electrons. In contrast to the resonant processes, one of these electrons may be ejected from the metal. These two processes are represented by the vertical sides of the triangle diagram.

In Auger neutralization two metal electrons simultaneously undergo a transition of equal energy, with one gaining the energy lost by the other. Initially the two electrons are located at energies $\alpha$ and $\beta$ below VL. One electron at energy $\beta$ gains potential energy and moves into the continuum above VL while the second electron loses an equal amount of potential energy and neutralizes the incoming ion. The energy limits of the ejected electron are

$$E_k [e(e)-] = E_i' - \alpha - \beta$$

$$E_k [e(e)-] \text{ minimum} = E_i' - 2E_o$$

$$E_k [e(e)-] \text{ maximum} = E_i' - 2\phi$$

Furthermore, for production of any ejected external electrons

$$E_i' > 2\phi$$

Similarly the Auger de-excitation process may be
considered as a process involving two electrons, one at energy $\beta$ below VL and the second, an excited atomic electron, at energy $E_x'$ above the ground state of the neutral atom. The two equivalent processes indicated in the figure differ only as to which electron is ejected into the continuum: either an electron from the atom or one from the metal. In either channel, an ejected electron is produced with energy

$$E_k [e(e)\rightarrow] = E_x' - \beta$$

For an incident ion one competing process involves radiative neutralization; however Shekhter [36] has shown that because of the relatively long radiation lifetime, about $10^{\#\#}\times 8$ sec, compared to the time which a thermal particle spends near a surface, about $10^{\#\#}\times 12$ sec, the probability of radiative neutralization is low, about $5\times 10^{\#\#}\times 7$. Another possibility, tunneling between the ground state of an incident ion and a core level of the solid, requires close energy equality, about 10 eV, and is also negligible in comparison to Auger processes.

**B. ELECTRON POLARIZATION EFFECTS IN ION NEUTRALIZATION AND METASTABLE DE-EXCITATION SPECTROSCOPY**

Consider the electronic processes available to an
incoming electronically polarized helium ion or metastable atom. For Auger neutralization of a polarized ion to the ground state, the neutralizing electron must have spin opposite to that of the electron on the incident ion because the helium singlet ground state requires two electrons of opposite spin. The polarization of the electron ejected from the metal will depend upon correlation effects between the neutralizing electron and the ejected electron. Because little is known about the strength of such correlations, one of the proposed experiments involves a search for such a correlation by measuring the polarization of the ejected electron as a function of incoming ion polarization.

For resonance neutralization to an excited state of the neutral atom, the neutralizing electron may have spin up or spin down. The population distribution among the resulting excited atomic states depends upon available atomic energy levels and the spin dependent energy and density of states of the neutralizing electron from the metal. In particular, for an incident ion with electronic spin up, the possible metastable states are 2(3)S with $M_j = 0$ and $+1$ for spin-down and spin-up neutralizing electrons respectively, and 2(1)S with a spin down neutralizing electron only. However, because the 2(1)S
state lies 0.8 eV higher in energy than the 2(3)8 state, neutralization may occur only to the triplet state if the Fermi level of the metal lies between the 2(3)8 and 2(1)8 atomic energy levels. Even assuming that neutralization to both singlet and triplet states is possible, the neutralizing electrons originate at different energies and corresponding densities of states.

Following resonance neutralization, the only available de-excitation process for the excited product atom is Auger de-excitation, which may proceed by one of the two equivalent channels discussed above and indicated in Figure 22. However, because of the selection rule $\Delta S \neq 0$, the 2(3)8 metastables can de-excite only by ejection of an electron from the metal.

According to the description above, an incoming particle will undergo one of the Auger processes indicated in the triangle diagram. An incoming ion will Auger neutralize and an incoming metastable will resonance ionize and then Auger neutralize if the Fermi level of the target surface is below the energy of the first excited level of the incoming ion. Alternatively, an incoming ion will resonance neutralize and then Auger de-excite or an incoming metastable will Auger de-excite if the Fermi level of the target surface is above the energy of the
first excited level of the incoming ion. Therefore, identical total ejected electron yields and energy distributions are expected regardless of whether the incoming projectile is a metastable or an ion. However, recent experiments call into question the validity of the model, in that measured distributions do not always appear to be the same. To date, however, no experiment has been reported in which either ions or metastables could be projected onto a common target surface. With this apparatus, a 2(3)8 helium metastable beam, in addition to the ion beam described in this thesis, is available [40]. This allows direct comparison of distributions for metastable and ion metastable and ion ejected electron energy distributions.
VI

PROPOSED EXPERIMENTS

The electronically polarized helium ion beam presented in this thesis has been constructed to perform ion neutralization spectroscopy upon various target surfaces. An electronically polarized helium metastable beam source has also been recently completed which intersects the ion beam at the target chamber. With completion of these two beams complimentary information on surface electronic structure may be explored by means of INS and MDS discussed in the previous chapter. After a brief description of the apparatus, initial experiments planned on magnetized surfaces will be discussed in the following sections.

A. APPARATUS CONFIGURATION

Described briefly, the metastable beam line [40] is orthogonal to the ion beam (see Figure 23). Both 2(3)S and 2(1)S metastable helium atoms originate from an electron impact excitation source. The 2(1)S metastables are quenched by inducing transitions to the 2(1)P state, which decay to the ground state leaving only ground state
FIGURE 23

Electronically polarized ion neutralization and metastable de-excitation spectroscopy apparatus diagram.
helium atoms and triplet metastables in the beam. The triplet helium atoms, after being optically pumped, enter the target chamber where they may interact with the target surface or pass across the chamber to a Stern-Gerlach polarization analysis system. This analysis system indicates a metastable polarization of about fifty percent has been achieved.

The ion beam, the primary subject of this thesis, is perpendicular to the metastable beam and enters the target chamber opposite the LEED-Auger system. The deceleration column within the target chamber delivers low energy polarized ions to the target.

Initial experiments, to be performed with the beams, require ion bombardment cleaning of the target surface, measurement of currents on the target surface, temperature control of the target and magnetization of the target. An ion bombardment gun, electrometer connection to the crystal, cooling coils on the target holder and target magnet provide the necessary components within the ultra-high vacuum chamber.
B. INITIAL EXPERIMENTS

Ion neutralization and metastable de-excitation spectroscopies probe the "tails" of electronic wave functions extending into the vacuum from the target surface. Because the interactions occur within a few Angstroms of the target surface, MDS and INS provide extremely surface selective measurements of electronic structure.

The most interesting experiment, and the one to be explored first, involves ferromagnetic materials. The surface selectivity of the ion and metastable spectroscopies enable examination of the electronic configuration at the outermost surface layer of magnetic materials. Although much is known about bulk magnetization, little is known about surface magnetic properties. With the proposed apparatus, separate measurements of spin-up and spin-down surface electron distributions should allow determination of surface magnetic properties.

The ferromagnetic material of particular interest for the initial experiment is nickel. The bulk ferromagnetic properties of nickel may be partially described by a combination of the band and itinerant electron models. The iron group transition metals, including nickel,
display characteristics described by such a model where
the Pauli exclusion principle produces spatial and
momentum distributions for the electrons dependent upon
the relative spin orientations. Itinerant electron
exchange effects, tied to the metallic nature of the
conduction electrons and localized magnetic moments,
describe much of the magnetic character of nickel. In the
itinerant electron model, the electrons remain on a single
atom for such a short period that formation of atomic \( \hat{S} \)
does not occur and the outer 3d and 4s electrons move
freely from ion to ion while retaining some of their
atomic 3d and 4s character. Without spin dependent
effects, the density of states of these bands may be
depicted qualitatively by the upper half of Figure 24,
where 10N electrons fill the 3d-4s bands to the Fermi
level.

However, the 3d-like electrons in the itinerant
electron model have spin dependent energies as a result of
the exchange interaction [41, 42]. This interaction
splits the spin-up and spin-down bands by
\[
\Delta \varepsilon (k) = \varepsilon (k, \text{down}) - \varepsilon (k, \text{up})
\]
which, on the average, is proportional to the
magnetization. For nickel, the average energy shift,
\[
\Delta \varepsilon = 0.25 \text{ to } 0.4 \text{ eV},
\]
depends almost entirely upon \( \Delta N = [N(\text{up}) - N(\text{down})] \), the
Qualitative diagram of the spin-split density of states of nickel.
number difference between spin-up and spin-down electrons per crystal volume.

Figure 24 illustrates the exchange interaction which shifts the energy bands of the spin-up and spin-down electrons. The densities of states of nickel are such that the relative shift between spin bands allows the 3d-like spin-up band to fill with 5N electrons while the 3d-like spin-down band fills to the Fermi level with 4.4N electrons. The 4s-like band contains the remaining 0.6N electrons with 0.3N in each of the spin-up and spin-down bands.

For an incident polarized helium ion, the neutralizing metal electron must have spin opposite to that of the ion. Therefore, for spin dependent electronic state densities such as in nickel, the total ejected electron yield and distribution can be expected to depend upon incident ion polarization. The ion polarization can be selected to be spin-up or spin-down relative to the surface magnetization vector and thus will probe separately the spin-down and spin-up 3d bands respectively. The contribution of ejected electrons from the 4s band, which has the same density of states for both electronic spins, will however contribute a spin independent background.
In conclusion, the extreme surface sensitivity of ion neutralization spectroscopy allows measurement of surface magnetic properties where other spectroscopies measure an average over two or more atomic layers.
VII
CONCLUSION

This thesis described an apparatus constructed to produce electronically spin polarized helium ions for spin dependent ion neutralization spectroscopy experiments. The discussion began with a review of the optical pumping process used to produce spin polarized helium metastables in a helium discharge with subsequent polarized helium ion production by metastable-metastable collisions. Experimentally obtained evidence indicated an ion polarization of about ten percent.

Ion extraction, transport and deceleration as well as the differential pumping procedure were also discussed. In particular, calculated ion trajectories for exponential and linear deceleration fields led to the choice of a linear decelerating column.

A digital Auger analysis system for determination of surface cleanliness was also presented. This system provides a digital controller for determination of integral, first derivative and second derivative retarding potential energy curves. Because of the completely digital design, future computer interfacing is straightforward.
The process of INS and MDS permits exploration of the surface electronic structure of a metal target. The unique ability of this apparatus to perform spin dependent surface electronic studies was presented.

The final section discussed the proposed experiment on a nickel crystal to ascertain the existence and properties of surface magnetization. This particular experiment would use the spin polarization of both the helium ion beam and metastable beam to examine the orientation of surface electronic spin. Furthermore, this experiment emphasises the forté of this apparatus: the unique ability to not only perform the highly surface selective ion neutralization and metastable de-excitation spectroscopies upon a common target surface but also to use the additional diagnostic capabilities provided by spin polarization of the probe beams.
APPENDIX

OPTICAL PUMPING MODEL ASSUMING
TWO ABSORPTION LINES

The following discussion presents a model for the absorption of the D0 and D3 optical pumping lines. The model illustrates the difference in absorption rates and the inability of the optical signal to determine exact metastable polarization.

Begin by assuming that the metastable sublevel densities, N+, N0 and N- do not change across the discharge bulb and that the pumping light consists of two lines, D0 and D3, whose spectral width equals that of the discharge. This latter assumption ignores the certain greater width of the pumping light, due to the higher velocities of the helium atoms within the high intensity discharge lamp, but permits a less cumbersome presentation.

The differential absorptions for the lines with respect to position within the absorbing discharge, allowing $\sigma_{ij}$ to indicate absorption cross section, are
\[
\frac{d \, I_0(x)}{dx} = - I_0(x) \sum_{i=-1}^{+1} \sigma_{I_0} N_i \\
\frac{d \, I_3(x)}{dx} = - I_3(x) \sum_{i=-1}^{+1} \sigma_{I3} N_i
\]

where the symbols are defined in Chapter II. Taking \( \sigma \) to be the cross section common to all the transitions, the relative cross sections for each transition between sublevels are

**2(3)S to 2(3)P0 (D0)**

\[
\sigma_{-1 \, 0} = 2 \, K \sigma \quad \sigma_{0 \, 0} = 0 \sigma \quad \sigma_{+1 \, 0} = 0 \sigma
\]

**2(3)S to 2(3)P1 (D1, D2)**

\[
\sigma_{-1 \, 1} = 3 \, L \sigma \quad \sigma_{0 \, 1} = 3 \, L \sigma \quad \sigma_{+1 \, 1} = 0 \sigma \\
\sigma_{-1 \, 2} = 1 \sigma \quad \sigma_{0 \, 2} = 3 \sigma \quad \sigma_{+1 \, 2} = 6 \sigma
\]

**2(3)S to 2(3)P1 (D3)**

\[
\sigma_{-1 \, 3} = (3L+1) \sigma \quad \sigma_{0 \, 3} = (3L+1) \sigma \quad \sigma_{+1 \, 3} = 6 \sigma
\]

The total cross section for each line and the resulting differential absorption is then
\[
\sum_{i=-1}^{+1} \sigma^i 0_{Ni} = (2K N^- + O NO + O N^+) \sigma
\]
\[
= 2K N^- \sigma
\]
\[
\sum_{i=-1}^{+1} \sigma^i 3_{Ni} = [(3L + 1) N^- + (3L + 3) NO + 6 N^+] \sigma
\]

The intensity as a function of position within the absorbing discharge, allowing \(N^*(n^+) = N^+, \ N^*(n^-) = N^-\) and \(N^*(n0) = NO\), becomes

\[
\frac{d \ IO(x)}{dx} = -IO(x) \ 2K N^- \sigma
\]
\[
\frac{d \ I3(x)}{dx} = -I3(x) \ [(3L + 1) N^- + (3L + 3) NO + 6 N^+] \sigma
\]
\[
IO(x) = AO \ exp[-2K N n^- x \ \sigma]
\]
\[
I3(x) = A3 \ exp[-x N \ \sigma ((3L + 1) n^- + (3L + 3) n0 + 6 n^+)]
\]

The ratio of initial line intensities, \(AO / A3\), with \(D0: D1: D3\) as \(K=I:1\) becomes \(K / (L+1)\). Notice that pumping light intensity depends analogously upon position \(x\) and metastable density \(N\). The product \(x*N\) gives the total number of metastables per unit area to which the pumping radiation has been exposed. With respect to the amount of absorbed pumping light then, a bulb with lower metastable density and larger diameter is equivalent to one with higher metastable density and smaller diameter.

The intensity exiting the pumped bulb, with \(x0 = \sigma^* N^* x0\) where \(x0\) is the diameter of the bulb, is
\[ I_0(x_0) = A_0 \exp[-2x_0N n-] \]
\[ I_3(x_0) = A_3 \exp[-x_0 ((3L + 1)n- + (3L + 3)n0 + 6n+)] \]

Replacing \( n0 \) with \( 1 - (n+) - (n-) \) leaves

\[ I_3(x_0) = A_3 \exp[-x_0 (-2n- + (-3L + 3) + 3L + 3)] \]

For an unpumped bulb, \( n+ = n0 = n- = 1/3 \), the exiting intensity is

\[ I_0(x_0) = A_0 \exp[-2x_0K (1/3)] \]
\[ I_3(x_0) = A_3 \exp[-x_0 ((-2/3) + (-L+1) + 3L + 3)] \]
\[ = A_3 \exp[-x_0 (10/3 + 2L)] \]

The optical signal, \( \Delta I / I(\text{abs}) \), is

\[ \Delta I = \frac{[I_0(x_0) - I_0(x_0)]_{\text{pumped}} - [I_0(x_0) + I_3(x_0)]_{\text{unpumped}}}{I(\text{abs})} \]
\[ = \frac{(A_0 + A_3) - [I_0(x_0) - I_3(x_0)]}{(A_0 + A_3)} \]

In the unpumped condition the exiting intensity is known. However, under pumped conditions, the ratio of \( n+ : n0 : n- \) remains unknown and therefore for any given percent absorption and optical signal there exists a range of values for \( n+ \), \( n0 \) and \( n- \).

Further problems with optical signal analysis involve pumping line width, nonuniform incident radiation intensity, differing path lengths through the spherical bulb, nonuniform metastable density and polarization.
distribution within the bulb discharge, and magnetic field nonuniformity over the bulb preventing Larmor coil saturation of all metastables. These problems are discussed in the main body of the thesis.

In conclusion, optical pumping signal measurements, while indicating the presence of optical pumping, do not yield even under best conditions the metastable polarization.
REFERENCES


14 Rofin Optics and Electronics, Newton Upper Falls, Mass, model number 7460, Ge photodiode.


17 K. Giberson, Chu Cheng, F. Tittel, F. B. Dunning, to be published.


