Study of Single-Electron Capture from He, Ne and Ar by Low-Energy Ne^Q+ Recoil Ions Using Translational Energy Spectroscopy

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STUDY OF SINGLE-ELECTRON CAPTURE FROM He, Ne AND Ar
BY LOW-ENERGY Neq+ RECOIL IONS USING TRANSLATIONAL
ENERGY SPECTROSCOPY

by

Renad Qubaj-Said

A Thesis
Submitted to the
Faculty of The Graduate College
in partial fulfillment of the
requirements for the
Degree of Master of Arts
Department of Physics

Western Michigan University
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June 1993
Single-electron capture by low-energy multiply charged Ne\(^q^+\) ions (\(q = 3-6\), where \(q\) is the projectile charge state) from He, Ne, and Ar has been studied by means of translational energy spectroscopy at laboratory collision energies between 100 and 600 eV, and at scattering angles between 0° and 6°. The dominant reaction channels are due to capture into excited states of the projectile products and show a dependence on scattering angle. A reasonable description of dominant final states is obtained in terms of the reaction windows, which are calculated using a multichannel Landau-Zener model. The measured differential cross sections show that the projectile products are distributed with maximum intensity near a scattering angle \(\theta_c\) which corresponds to capture at an impact parameter equal to the crossing radius.
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Renad Qubaj-Said
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Study of single-electron capture from He, Ne and Ar by low-energy Ne\textsuperscript{aq}+ recoil ions using translational energy spectroscopy

Qubaj-Said, Renad, M.A.

Western Michigan University, 1993
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CHAPTER I

INTRODUCTION

In an ion-atom collision, the atomic interactions taking place can be categorized into three processes: excitation, ionization, and electron capture (frequently called charge transfer). The focus of this study is on single-electron capture, which is defined as a process where one electron is transferred from the target atom to the projectile ion.

In the last decade, electron capture by multiply charged ions has become an active field of study. This was mainly due to the importance of this process in areas such as energy-loss mechanisms of high temperature and astrophysical plasmas (Peguignot, 1980). Electron capture is also considered to be useful for obtaining population inversion of exited atoms and ions used in short-wavelength lasers (Louisell, Scully, & McKnight, 1975). Its importance is also known in the development of controlled thermonuclear fusion devices (Meade, 1974), in gas-filled radiation detectors, and in both heavy-ion and electron-beam ion sources (EBIS) (Arianer & Geller, 1981). Electron capture has also been experimentally useful for obtaining certain neutral beams with known distributions of states which can then be used to study other phenomena (Cooks, 1978).

For low-velocity collisions where the relative velocity of the colliding particles is smaller than the velocity of the bound target
Electrons, electron capture can be described in terms of the molecular orbitals of the quasimolecule formed by the collisional partners. Electron capture is, therefore, a transition between electronic states of the transitory molecule formed temporarily during the collision.

Several previous experimental measurements on electron capture by multiply-charged neon ions from different target atoms have been reported. Justiniano, Cocke, Gray, Dubois, Can et al. (1984) have reported experimental cross sections for electron capture and transfer ionization for collisions of Ne$q^+$, Ar$q^+$, Kr$q^+$, and Xe$q^+$ ions with He for projectile charge states (q) between 2 and 13 and for projectile energies between 100 and 250 keV. They found the cross sections for single-electron capture to be an order of magnitude greater than those for double-electron capture.

Schmeissner, Cocke, Mann and Meyerhof (1984) measured single-electron capture and transfer ionization processes in collisions of slow Ne$q^+$ ions with He for q=3-8. The dominant reactions were found to have positive energy defects between (see Sec. V) 3.6 and 31 eV.

Tunnell, Cocke, Giese, Kamber, Varghese et al. (1987) measured the angular distributions for single-electron capture by Ne ions from He at laboratory incident energies between 172 and 1200 eV for q=3-6. Giese, Cocke, Waggoner, Tunnel and Varghese (1986) used translational energy-gain spectroscopy to determine the final state populations in the single-electron capture process by Ar$q^+$ (q=4-8) and Ne$q^+$ (q=4-7) ions from Ar, D$_2$, and D at collision energies of 545 keV. They found...
that the final state populations are very sensitive to the energy-level structure of the collision system, and the results were interpreted in terms of a "reaction window." (see section on reaction window in Chapter II). Other studies, such as the one conducted by Tan and Lin (1988), applied a quantal two-channel calculation to charge-transfer differential cross sections in Ne\(^{4+}\) - He collisions at laboratory impact energies from 220 to 500 eV. Lebius, Kosloski, and Huber (1989) studied the final state population in terms of the reaction window and the multichannel Landau-Zener model for collisions of Ne\(^{3+}\) with Ne and He.

Hansen & Andersson (1989) studied single electron capture for 2 keV Ne\(^{6+}\) ions from He using the atomic expansion close-coupling approach. They found that the possibility of collision-induced core excitation is of little importance in a time-dependent model treatment in contrast to multichannel Landau-Zener calculations. Andersson, Pedersen, Barany, Bangsgaard and Hvelplund (1989) have measured translational energy gain spectra for single electron capture in Ne\(^{6+}\)-He collisions at energies of 200 - 2000 eV. They calculated theoretical energy gain spectra, which can be directly compared with the experimental data, using a multichannel Landau-Zener model based on curved classical trajectories for the nuclear motion. It was found that the model was valuable in the identification of reaction channels, especially at low projectile energies where the effects of angular scattering introduce additional structure in the spectra.

Against this background, the following goals were formulated for
this work. First, state-selective single-electron capture reactions in low-energy \( \text{Ne}^+ (q=3-6) - \text{He}, \text{Ne}, \text{and Ar} \) collisions were investigated experimentally. The experiment was performed by using the translational energy-gain spectroscopy technique. The measured energy gain allows for a determination of the excited states which are populated in the projectile ion. Secondly, angular distributions of the projectile products following single-electron capture were measured.

Chapter II of this thesis presents the theoretical considerations for single-electron capture used to explain the observed collision processes. Chapter III describes the experimental apparatus, data acquisition, and analytical methods used to measure the translational energy-gain spectra. Chapter IV presents the results of this experiment, and the results are compared with those of other studies. The conclusions drawn from these results are given in the last chapter.
CHAPTER II

THEORETICAL CONSIDERATIONS

There are three approaches to the study of the electron capture process when the collision velocity is small compared with the velocity of the captured electron. These are: (1) numerical solution of the coupled-channel equations, (2) reduction of the problem to a multi-channel model which allows analytical solution, and (3) construction of decay models for the electron capture process (Kamber & Cocke, 1991), which can be derived from the quasi-stationary state in the field of multiply-charged ion. In this study, the focus will be on those aspects which are directly related to the prediction of the final state populations of the captured electrons. Within this chapter, the following will be discussed: (1) kinematics, (2) differential and total cross sections, (3) the classical over-barrier model, which deals with the ability of the target electron to overcome the barrier potential between the collision partners, (4) the Landau-Zener model which is used to interpret the final state populations by calculating the location of the quasimolecular crossing radii where the probability of single-electron capture is large, and (5) reaction windows.

Kinematics

In this experiment, we are concerned with events in which the
basic ingredients are atomic systems, i.e., atoms and ions. Here, electron capture may take place through a variety of reaction channels. The following equation indicates the general capture reaction:

$$A^{q} + E - A^{q-1} + B^* + \Delta E$$ (2.1)

where $q$ is the initial charge state of the projectile and $\Delta E$ represents the energy defect of the reaction channel involved. $\Delta E$ is calculated according to the following formula:

$$\Delta E = I_p(A^{q-1}) - I_p(B^*) - E_j$$ (2.2)

where $I_p(A^{q-1})$ is the ionization potential of the product ion $A^{q-1}$, $I_p(B^*)$ the ionization potential of the target (assuming the target to be in its ground state and the captured electron to be the most loosely bound) and $E_j$ the excitation energy of the $j$th level of projectile product ion $A^{q-1}$.

Using classical two-body dynamics, the translational energy $E_{\text{inelastic}}$ of an ion undergoing inelastic scattering differs from the energy $E_0$ of the incident projectile ion by:

$$E_{\text{inelastic}} - E_0 = \Delta E - \Delta K$$ (2.3)

and

$$Q = \Delta E - \Delta K$$ (2.4)

where $\Delta K$ is the translational energy given to the target. $Q$ is the total change in internal energy that takes place during an inelastic
collision. The energy $Q$ can be either positive or negative depending on the binding energy of the captured electron. The former collision is called an exothermic reaction and the latter an endothermic one.

The translational energy $\Delta K$ given to the target can be represented through the following equation (Cooks, 1978):

$$\Delta K = \frac{m_p}{M+m_p} (1-\cos \theta) \left( \frac{2ME_0 - \Delta E}{M+m_p} \right) + \frac{m_p (\Delta E)^2}{4ME_0} \cos \theta \quad (2.5)$$

where $m_p$ and $M$ are, respectively, the projectile and target masses, $E_0$ is the initial laboratory translational energy of the projectile, and $\theta$ is the scattering angle. For zero scattering angle, the translational energy reduces to

$$\Delta K = \frac{m_p (\Delta E)^2}{4ME_0} \quad (2.6)$$

Differential and Total Cross Sections

Consider a parallel beam of monoenergetic projectile particles moving in the direction of the z-axis with $N_p$ particles per second crossing a unit area perpendicular to the beam (see Figure 1). The differential cross section is defined as the probability per unit solid angle that an incident particle is scattered into the solid angle $d\Omega = \sin \theta d\theta d\phi$ at the scattering center, and can be expressed in the form $N_p I(\theta, \phi) d\Omega$, where $I(\theta, \phi)$ has the dimensions of area. The total cross section is described as the total number of particles scattered per second from an incident beam of unit flux density, and
is given by:

$$\sigma = \int_0^\pi \int_0^{2\pi} I(\theta, \phi) \sin \theta d\theta d\phi$$  \hspace{1cm} (2.7)

Since scattering is almost always isotropic in $\phi$,

$$\sigma = 2\pi \int_0^\pi I(\theta) \sin \theta d\theta$$  \hspace{1cm} (2.8)

Since the magnitude of differential cross section results directly

![Figure 1. The Basic Geometry of Scattering.](image)

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from the interactions between the colliding particles, the nature of atomic forces may be determined by examining the differential cross section.

**Classical Over-Barrier Model**

The major assumption of this model is that capture will occur if the target electron can overcome the potential barrier between the projectile and the nuclear charge of the target (Ryufuku, Sasaki & Watanabe, 1980). The classical over-barrier model (cbm) is widely used to explain the main aspects of the electron capture process. Its widespread use is largely due to the fact that resonant electron capture populates preferentially states with large quantum numbers $n$ and $l$, yielding large cross sections for capture into the principal shell $n$ (Kamber & Cocke, 1991).

The potential barrier in which the electron is captured is given by

$$V(x) = \frac{-Z}{R} \frac{1}{R-x}$$  \hspace{1cm} (2.9)

where $Z$ is the charge of the projectile, $R$ is the internuclear distance in atomic units and $X$ is the distance measured along the internuclear axis between the electron and the projectile in atomic units (see Figure 2). From Equation (2.9), the maximum of the potential can be found by differentiation with respect to $X$:

$$V_{\text{max}} = \frac{-(\sqrt{Z+1})^2}{R}$$  \hspace{1cm} (2.10)

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Figure 2. Schematic Representation of Classical Model for Capture Reaction.

which gives the height of the barrier potential.

Electron transfer takes place under two conditions. First, there must be a degeneracy between initial and final states (a curve crossing). This condition is represented by the following equation:

$$-I_e - \frac{Z}{R} - \frac{Z^2}{2n^2} - \frac{1}{R}$$

where the left hand side denotes the ionization potential of the target electron (in atomic units) followed by the Coulomb potential of the projectile. The right hand side denotes the energy of the electron when bound to a level of quantum number $n$ of the projectile.
perturbed by the Coulomb energy due to ionization of the target. Second, the energy of the target electron must exceed the barrier maximum. This condition is represented by the following equation

$$-I_e\frac{Z}{R} > \frac{-(\sqrt{Z+1})^2}{R}$$ (2.12)

In this case the energy of the bound electron in the Coulomb potential of the projectile ion must be equal to or greater than the maximum barrier height $V_{\text{max}}$.

According to the model, $n$ will satisfy the two conditions mentioned above if:

$$n \leq \left( \frac{Z^2}{2I_e} \right)^{\frac{1}{3}} \left( \frac{2/(Z+1)}{Z + 2\sqrt{Z}} \right)$$ (2.13)

Equation (2.13) will not, in general, yield an integer value for $n$. Therefore, one defines $n_{\text{cbo}}$ as the largest integer smaller than that given by the right-hand side of Equation 2.13. This gives the hydrogen-like level in the projectile into which the electron is captured. The critical radius, $R_{\text{cbo}}$, is given in terms of $n_{\text{cbo}}$ by (Ryufuku, Sasaki, & Watanabe, 1980):

$$R_{\text{cbo}} = \frac{2(Z-1)}{2^2 I_e - 2I_e}$$ (2.14)

From Equation (2.14), we can see that the crossing distance $R_{\text{cbo}}$ becomes gradually smaller with decreasing $n$. From this, we can
identify $n_{\text{em}}$ as the principal quantum number of the final state most probably populated by the transferred electron (Ryufuku, Sasaki, & Watanabe, 1980).

**Landau-Zener Model**

This model is based on the theoretical work done by both Landau (1932) and Zener (1932). Their work was based on the classical picture of the nuclear motion within the impact parameter formulation, and dealt with the transition probability at a given molecular curve crossing. The multichannel problem of electron capture into excited states of the projectile can be simplified by considering the coupling between the several available states as coupling between successive sets of two states. Then, the electron capture process can be treated by the multichannel Landau-Zener (MCLZ) theory. The transfer of an electron from atom B to ion A of initial charge $q$ is represented by:

$$A^{q}+B \rightarrow A^{q-1}+B^{+}$$  \hspace{1cm} (2.15)

In the absence of a collision interaction, the left hand and the right hand members of this equation can be imagined to form two quasimolecules. Figure 3 shows the associated diabatic potential energy curves (solid lines) in the absence of a collision interaction between the two systems, where the curves cross at an internuclear distance $R=R_x$. In a collision interaction, these diabatic curves are modified to give the adiabatic behavior indicated qualitatively by
the dashed curves in Figure 3.

Note: The Value of the Matrix Element $H_{22}$ is One-Half the Adiabatic Splitting at the Diabatic Crossing Radius $R_x$ as Indicated.

Figure 3. A Systematic Plot of the Potential Energy $V(R)$ Curves for the Incoming and Outgoing Channels Near a Curve Crossing Versus Internuclear Separation $R$; (Broken Curve), Adiabatic Basis; (Solid Curve), Diabatic Basis.

According to the Landau-Zener model discussed in (Janev & Winter, 1985), the single-crossing transition probability at each potential energy crossing $R_x$ is given by the following formula:

$$ P_a = \exp\left(-\frac{2\pi H_{22}^2}{\nu_r \Delta P}\right) $$ (2.16)
where $H_{12}$ is the coupling matrix element and is equal to one half of
the adiabatic splitting which is the minimum energy difference be­
tween non-crossing curves as indicated in Figure 3, $\Delta F$ is the differ­
ence in slopes of the diabatic potential energy curves at $R_*$ (i.e.,
where the potential curves of the initial and final channels inter­
sect), and $v_r$ is the radial velocity of the projectile at the cross­
ing distance $R_*$ which is given by:

$$v_r = v_0 \left(1 - \left(\frac{b}{R_*}\right)^2\right)^{\frac{1}{2}}$$ \hspace{1cm} (2.17)

In Equation 2.17, $v_0$ is the relative velocity of the colliding parti­
cles and $b$ is the impact parameter.

$H_{12}$ is the most important parameter in the Landau-Zener model.

Olson and Salop (1976) developed a formulation for $H_{12}$ for one­
electron capture by multi-charged ions, and the coupling matrix ele­
ment was determined for a large number of bare projectiles on atomic
hydrogen. These systems can be treated in the one-electron diabatic
molecule model which allows analysis of the pseudo-crossing (i.e., a
non curve crossing, sometimes called an avoided curve crossing) ob­served in the exact adiabatic potential (i.e., where the potential
curves of the initial and final channels do not intersect) curves for
each system. Empirically, Olson and Salop (1976) obtained a func­tional expression for the coupling matrix elements, given by

$$H_{12}^{os} = 9.13q \frac{e^2}{\hbar} \exp(-1.324aR_xq^{\frac{1}{3}})$$ \hspace{1cm} (2.18)
\[ \alpha = 2I_t^{-1} \]  

where \( \alpha \) is a coefficient in the formula, \( I_t \) is the ionization potential of the target atom in atomic units. Kimura, Iwai, Kobayashi, Matsumoto, Ohtani et al., (1984) reduced \( H_{12} \) by 40% in order to achieve agreement with their measurements of total single-capture cross sections in He targets.

The total probability for electron capture at a given impact parameter is given by:

\[ P = 2p(1-p) \]  

where \( p \) is the probability that in traversing the crossing point the system remains on the same potential curve, and \( (1-p) \) is the probability that a jump from one curve to another occurs. Since the system traverses the crossing point \( R_\ast \) twice, once each on the incoming and outgoing trajectories, the factor of two appears.

The Landau-Zener formula can be generalized to a multi-channel model (i.e., many possible exit channels). Considering a collision system with \( n \) reaction channels and with no interference between the different paths leading to a particular final state as shown in Figure 4, the probability \( p_n \) \((n=1,2, \ldots, N)\) for capture into the \( n \)th final state is given by:

\[ p_n = p_1p_2 \cdots p_n(1-p_n)(1+(p_{n+1}p_{n+2} \cdots p_N)^2) \]

\[ + (p_n+1p_{n+2} \cdots p_N)^2(1-p_n)^2+(1-p_{n+1})^2 \]

\[ + \cdots + p_{n+1}^2(1-p_{n+2})^2+(1-p_{n+1})^2 \]
Each partial cross-section is found by integrating over the impact parameter, and the total cross section is then obtained by summing as follows:

$$\sigma_i = 2\pi \int_{b_1}^{b_2} dp_x$$  \hspace{1cm} (2.22)

and

$$\sigma = \sum \sigma_i$$  \hspace{1cm} (2.23)

The predictive ability of the multichannel Landau-Zener model can be enhanced by calculating the coupling matrix element $t_{12}$ for individual reaction channels of a specific collision system. For the case of electron capture by partially-stripped projectiles, for which
the subshell degeneracy is removed, we use here the Taulbjerg (1986) expression for $H_{12}$:

$$H_{12} = f_{nl}^* H_{12}$$

(2.24)

and

$$f_{nl} = -\frac{1}{2} \frac{n+1}{n+2} \frac{(2l+1)}{(n+l+1)(n+l+2)} \frac{\Gamma(n)}{\Gamma(n+1)}$$

(2.25)

where $n$ and $l$ characterize the final orbital quantum numbers of the captured electron, and $\Gamma$ is the gamma function or the generalized factorial function.

It should be noted that the Landau-Zener model has a number of limitations. Its validity is restricted to the adiabatic energy region. Furthermore, the multichannel Landau-Zener model neglects electron transitions caused by rotation of the internuclear axis. This may result in an inaccuracy of the cross sections in the low-energy region by a factor of two or more. Janev (1983) has proposed a modified Landau-Zener model including rotational mixing for the total transition probability for the collision of fully-stripped ions with hydrogen atoms.

**Reaction Window**

Electron capture in slow collisions of highly charged ions with atoms has been discussed in terms of the so-called reaction window (see Figure 5). This concept suggests that, in collisions of highly charged ions with atoms or molecules, transitions in slow atomic
collisions are strongly favored because the interaction in the entrance channel is controlled by the relatively small polarization attraction at large internuclear separations, while the interaction in the exit channels is controlled by the ion-ion Coulomb repulsion. The significant avoided curve crossings (i.e., non curve crossings) of the adiabatic potential energy curves related to the ion-atom channel and various ion-ion channels are those which take place at moderate internuclear separations (R = 3-10 a.u.), depending on the initial projectile charge state. Kimura et al. (1984) and Taulbjerg (1986) refer to this intermediate range of separation as the reaction window for the electron capture process. According to Taulberg (1986), the location of the reaction window depends modestly on the relative collision velocity, v. The adiabacity at inner crossings increases when v is reduced while the interaction time becomes larger. The reaction window, therefore, shifts towards larger internuclear separations R if v is reduced and vice versa. A reaction window is shown in Figure 5 as a function of the internuclear separation; also shown is the velocity dependence mentioned above.

In most cases, the location of the reaction window is calculated as a function of the internuclear separation R. This location is then compared to the crossing radii of specific final states. It is expected that those crossing radii falling within the reaction window will have the largest cross section. The crossing radii can be calculated from the simple relation value, $Q = (q-1)/R_x$, where q is the projectile charge (Kamber, Cocke, Giese, Pederson & Waggoner, 1987a).
Figure 5. Reaction Window for Various Collision Velocities ($v$); $\sigma$ is the Landau-Zener Cross Section; $v_1$ (Dotted Curve) = 30 a.u.; $v_2$ (Broken Curve) = 0.05 a.u.; $v_3$ (Solid Curve) = 0.005 a.u.
CHAPTER III

EXPERIMENT

This chapter describes the experimental aspects of this study. The experiment, described below, was performed at WMU using the 6 MV EN tandem Van de Graaff accelerator. The general layout of this facility is shown in Figure 6. The low-energy highly-charged Ne projectile ions used in this experiment were produced in a recoil ion source by using a fast beam of 25 MeV F^+.

Singly-charged negative ions are produced in the SNICS ion source, extracted, accelerated and then passed through a 20° inflection magnet which allows selection of the desired ion species; this beam is then deflected into the accelerator beam line. Negative ions are accelerated toward the positively-charged high voltage terminal in the center of the accelerator where they pass through a gas stripper in which electrons are removed from the ions so that the ions become positively charged. The beam emerging from the accelerator consists of several charge states of different beam energies. For the purpose of this experiment, the desired charge state of the fluorine ions (F^-) was selected by the 90° analyzing magnet which focused the ions onto a set of image slits. A second magnet (switching magnet) then deflected the F^- ions into the experimental region. The switching magnet is used to select among several beam lines set up for different experiments in the target room. The selected beam
Figure 6. Schematic Diagram of the Western Michigan University Tandem Van de Graaff Accelerator Laboratory.
of 25 MeV F^{4+} was poststripped to a median charge state of 7+ by using a carbon foil and then collimated with a set of four jaw slits as shown in Figure 7. After collimation, the beam entered the recoil-ion source which was designed and built at WMU. This source consists of a collision chamber, pusher, and two lenses (L1 and L2) (see Figure 7). The fast F^{4+} beam passed through the collision chamber which has entrance and exit apertures of 2.5 and 3.3 mm diameter, respectively.

The fast beam was collected by a Faraday cup after it passed through the interaction region in the recoil-ion source. Target ions formed in the collision chamber were extracted perpendicular to the fast beam with an acceleration voltage \( V_{acc} = V1 \) through a 2.5 mm diameter aperture in lens L1, under the action of an electric field from the lens L2 which penetrates into the collision chamber through the aperture in L1. The pusher was kept at the same potential as the collision chamber. The chamber was maintained at pressures on the order of \( 2 \times 10^{-6} \) Torr. The voltages \( V1 \) (the voltage on the pusher) and \( V2 \) (the voltage on the L2) were set so as to optimize the resolution and transmission of the detected ions. Typical voltages for \( V1 \) and \( V2 \) of 99 and 100 volts were used, in which the \( V_{acc} \) is equal to \( V1 \), yielding a recoil-ion energy given by:

\[
E = qeV1
\]  

(3.1)

where \( q \) is the charge state and \( e \) is the electron charge.

An Einzel lens was used to focus the ion beam extracted from the
recoil source into a 180° double focusing magnet. Here, the various charge states of the recoiling projectile ions were separated due to their different mass-to-charge ratios by passing them through the double focusing magnet. Following that the ion beam was again focused by two pairs of deflectors D1 and D2 and directed into a gas cell 3 mm long. The entrance and the exit apertures of the cell were 1 and 2 mm, respectively. Ions scattered through a nominal angle θ
into a solid angle ($\Delta\Omega$) of about $3 \times 10^3$ sr were energy analyzed by a $90^\circ$ double focusing electrostatic analyzer (ESA). The scattering angle $\theta$ is selected by means of an aperture Al (1 mm diameter) in front of the ESA. The analyzed ions were then detected by a one-dimensional position-sensitive microchannel plate detector (MCP), which is located at the focal plane of the ESA. The detector is constructed from two 3.5 cm diameter microchannel plates (MCP) and a resistive anode.

In this experiment, the detector operates in the following manner. When an ion strikes the front channel plate the resulting electron cascade from the two microchannel plates is collected by the anode which resides in the PSA. The fraction of charge collected on electrodes located at each side of the anode is proportional to the distance from the electrode where the electrons hit the anode. The relative position of an ion striking the MCP is then determined by the ratio between the resulting voltage pulse at one electrode and the sum of the voltage pulses of the two electrodes, that is

$$X = \frac{V_u}{V_u + V_l}$$

(3.2)

where $X$ is the relative position along the active dimension, $V_u$ is the voltage of the upper electrode, and $V_l$ is the voltage of the lower electrode (see Figure 8).

The direct beam was positioned at different locations so as to calibrate the detector, while varying the applied voltage to the ESA.
Spectra were recorded at each voltage in order to generate a calibration curve.

The present data were collected using the electronics set up shown schematically in Figure 8. Signals from each electrode of the position sensitive anode (PSA) were first identically amplified by using pre-amplifiers (Ortec model 109A) and spectroscopy amplifiers (Ortec model 451). The sum was determined by using a dual sum/invert amplifier (DSI, Tennelec model TC253). The signal then passed to the position sensitive detector (PSD) analyzer (Ortec model 466), in which the division stated in Equation (3.2) was performed. These
analyzed signals were then recorded by an analog-to-digital converter (ADC). The pulse rate of the output signal (energy output) was measured using a ratemeter (RM) (Ortec model 744).

A STARBURST interface module was used to transfer data to a MicroVAXII computer system enabling conversion from position distribution information to energy distribution information. In order to minimize distortion due to small changes in linearity near the edges of the MCP, only signals coming from the central portion of the microchannel plates were used.
CHAPTER IV

DATA ANALYSIS

In this study, energy-gain spectra for single-electron capture by Ne\(^+\) on He, Ne, and Ar are taken at selected projectile scattering angles. The position distribution of the scattered ions following single-electron capture was measured, with the voltage applied to the ESA given by

\[ V = \frac{qV_0}{q'} \]  \hspace{1cm} (4.1)

where \(V_0\) is the ESA voltage for which the direct beam is passed, and \(q\) and \(q'\) are the charge states of the projectile before and after the collision, respectively.

The energy gain \(Q\) corresponding to \(V_{\text{acc}}\) (see Chapter III) is given by (Kamber, Cocke, Giese, Pederson, Waggoner et al., 1987b):

\[ Q = (q'V - 1)qV_{\text{acc}}. \]  \hspace{1cm} (4.2)

In terms of the position of the projectile peak, \(Q\) is given by (Kamber et al., 1987b):

\[ Q(x) = \frac{S(x) - S(0)}{FV_0 + S(x) - S(0)} qV_{\text{acc}} \]  \hspace{1cm} (4.3)
where $S(0)$ and $S(x)$ are the positions of the main peaks of the uncharge-changed projectiles and the charge-changed product ions, respectively, and $F$ is the slope of the position of the projectile peak as a function of ESA voltage (see Figure 9).

![Figure 9. A Typical Calibration Showing Channel Number Versus ESA Voltage.](image)

The energy resolution of the ESA for a 100 eV Ne$^{4+}$ projectile beam was found to be 1.52 eV as determined from the full-width half-maximum (FWHM) of the projectile peak as a function of analyzer voltage as shown in Figure 10 ($\Delta E/E = \Delta V/V$). Scanning the 180° double focussing magnet (see Figure 7) showed there to be Ne$^{3+}$ recoil ions.
with charge states ranging from 1+ to 6+ (see Figure 11). Using the charge-state values indicated in Figure 11, the square root of the mass-to-charge ratio of the recoil ion peaks was plotted as a function of the magnet current as shown in Figure 12 to verify the assigned charge states. The linearity of this plot indicates the correct assignment of charge states has been made. A small background contribution was subtracted from the energy-gain spectra based on the total amount of fast beam. These background contributions were determined from measurements with no gas in the target cell (see Figure 13).

The differential cross section \(\frac{d\sigma}{d\Omega}\), which is directly proportional to \(\sigma\), is obtained by multiplying the total number of scattered ions at an angle \(\theta\) by \(2\pi\sin\theta\). Absolute cross section scales were assigned to the present data by normalizing to the total cross section measurements of Tunnell et al. (1987). This procedure is approximate only, since the angular range of our data is smaller than the range of the angular distribution measured by Tunnell et al. (1987). The fraction of the total cross section occurring outside the angular range of our measurements was estimated by extrapolation. The error bars that are shown in the graphical presentations of the differential cross sections (Figures 15, 20, 22, and 26) represent the statistical uncertainties only. Errors in the measured Q-values are caused by the least squares fit to the calibration curve (± 2%) and the stability of the voltage applied to the ESA (± 1%). In this respect, errors in the energy scale, as calibrated against the
Figure 10. Full-Width Half-Maximum (FWHM) of 100 eV Ne$^{4+}$ Recoil Ions as a Function of the Analyzer Voltage.

Figure 11. Ne$^{8+}$ Recoil-Ion Charge-State Intensities for 25 MeV F$^{4+}$ Incident on Ne Gas as a Function of the Magnet Current.

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scattered ions, were always less than ±1 eV.

The final state populations (outgoing reaction channels) for Ne$^{q+}$1 were determined by comparing the spectra with theoretical energy-gain values. The possible outgoing channels following electron capture are listed in Tables 1-6. The energy levels used in preparing the tables are taken from Baskin and Stoner (1978).
Note: Spectrum (a) = Single-Electron Capture by 250 eV Ne$^{5+}$ Ions from He and Background Gas. Spectrum (b) = Single-Electron Capture by 250 eV Ne$^{5+}$ Ions from Background Gas. Spectrum (c) = Spectrum (a) - Spectrum (b).

Figure 13. Typical Subtraction Procedure for the Energy-Gain Spectra. Channel Number is Proportional to the Q-Value.
Table 1

Single-Electron Capture Reaction Channels for Ne\(^{3+}\) – He Collisions

<table>
<thead>
<tr>
<th>Reactant and initial states</th>
<th>Product and final states</th>
<th>(\Delta E) (eV)</th>
<th>Designation of reaction process</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ne(^{3+}) ((2s^2, 2p^3, 1S)) + He((1s^2, 1S)) (\rightarrow) Ne(^{3+}) ((2s, 2p^1, 3P)) + He(^+) ((1s, 2S))</td>
<td>(\rightarrow) Ne(^{3+}) ((2s, 2p^3, 1P)) + He(^+)</td>
<td>13.55</td>
<td>I(\Xi)X</td>
</tr>
<tr>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3S \ 3S)) + He(^+)</td>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3S \ 3S)) + He(^+)</td>
<td>-0.7</td>
<td>I(\eta)X</td>
</tr>
<tr>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3p \ 3P)) + He(^+)</td>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3p \ 3P)) + He(^+)</td>
<td>-4.84</td>
<td>I(\Omega)X</td>
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<tr>
<td>Ne(^{3+}) ((2s^2, 2p^3, 1D)) + He((1s^2, 1S)) (\rightarrow) Ne(^{3+}) ((2s, 2p^3, 3P)) + He(^+)</td>
<td>(\rightarrow) Ne(^{3+}) ((2s, 2p^3, 3P)) + He(^+)</td>
<td>+18.63</td>
<td>II(\Xi)X</td>
</tr>
<tr>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3S \ 3S)) + He(^+)</td>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3S \ 3S)) + He(^+)</td>
<td>+5.01</td>
<td>II(\iota)X</td>
</tr>
<tr>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3p \ 3P)) + He(^+)</td>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3p \ 3P)) + He(^+)</td>
<td>+4.38</td>
<td>II(\eta)X</td>
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<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3p \ 3P)) + He(^+)</td>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3p \ 3P)) + He(^+)</td>
<td>+0.24</td>
<td>II(\Omega)X</td>
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<td>Ne(^{3+}) ((2s^2, 2p^3, 3P)) + He((1s^2, 1S)) (\rightarrow) Ne(^{3+}) ((2s, 2p^3, 1P)) + He(^+)</td>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3S \ 3S)) + He(^+)</td>
<td>+10.69</td>
<td>III(\Xi)X</td>
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<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3S \ 3S)) + He(^+)</td>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3S \ 3S)) + He(^+)</td>
<td>+7.63</td>
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<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3p \ 3P)) + He(^+)</td>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3p \ 3P)) + He(^+)</td>
<td>+7.00</td>
<td>III(\eta)X</td>
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<tr>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3p \ 3P)) + He(^+)</td>
<td>(\rightarrow) Ne(^{3+}) ((2s^2, 2p^3, ('S), 3p \ 3P)) + He(^+)</td>
<td>+2.86</td>
<td>III(\Omega)X</td>
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<table>
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<tr>
<th>Reactant and initial states</th>
<th>Product and final states</th>
<th>ΔE(eV)</th>
<th>Designation of reaction process</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ne$^3^*$ ($2s^2 2p^3 4,^1S) + Ne(2p^6 1,^S)\rightarrow$</td>
<td>Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$ ($2p^5 2,^P)\rightarrow$</td>
<td>+16.57</td>
<td>I6X</td>
</tr>
<tr>
<td>Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
<td>+6.01</td>
<td>I$\varepsilon$X</td>
<td></td>
</tr>
<tr>
<td>$→$ Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
<td>+2.95</td>
<td>I</td>
<td>X</td>
</tr>
<tr>
<td>$→$ Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
<td>+2.3</td>
<td>$\eta$X</td>
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<tr>
<td>$→$ Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
<td>−1.82</td>
<td>I0X</td>
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<td>$→$ Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
<td>+14.99</td>
<td>I</td>
<td>A</td>
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<tr>
<td>$→$ Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
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<td>I</td>
<td>C</td>
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<td>$→$ Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
<td>+11.39</td>
<td>I</td>
<td>D</td>
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<tr>
<td>$→$ Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
<td>+11.79</td>
<td>I</td>
<td>B</td>
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<tr>
<td>$→$ Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
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<td>I</td>
<td>G</td>
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<td>Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
<td>+21.65</td>
<td>II</td>
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<td>Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
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<td>II</td>
<td>$\varepsilon$X</td>
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<td>$→$ Ne$^2^+$ ($2s^2 2p^5 3,^P) + Ne^+$</td>
<td>+8.03</td>
<td>II</td>
<td>JX</td>
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<td>Reactant and initial states</td>
<td>Product and final states</td>
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<td>Designation of reaction process</td>
</tr>
<tr>
<td>----------------------------</td>
<td>--------------------------</td>
<td>----------------</td>
<td>-------------------------------</td>
</tr>
<tr>
<td>→ Ne$^{3+}$ ($2s^2 2p^3 \ (3S)$ $3s \ 3S$) + Ne$^*$</td>
<td>Ne$^{3+}$ ($2s^2 2p^3 \ (3P)$ $3s \ 3P$) + Ne$^*$</td>
<td>+7.38</td>
<td>II$\eta$X</td>
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<tr>
<td>→ Ne$^{3+}$ ($2s^2 2p^3 \ (3P)$ $3s \ 3S$) + Ne$^*$</td>
<td>Ne$^{3+}$ ($2s^2 2p^3 \ (3P)$ $3s \ 3P$) + Ne$^*$</td>
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<td>→ Ne$^{3+}$ ($2s^2 2p^4 \ (3P)$) + Ne$^+$ ($2s^2 2p^6 \ (3S)$)</td>
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<td>+20.07</td>
<td>II$\alpha$A</td>
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<td>Ne$^{3+}$ ($2s^2 2p^4 \ (3P)$) + Ne$^+$ ($2s^2 2p^6 \ (3S)$)</td>
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<td>→ Ne$^{3+}$ ($2s^2 2p^4 \ (3P)$) + Ne$^+$ ($2s^2 2p^4 \ (3P)$ $3s \ 3P$)</td>
<td>Ne$^{3+}$ ($2s^2 2p^4 \ (3P)$) + Ne$^+$ ($2s^2 2p^4 \ (3P)$ $3s \ 3P$)</td>
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<td>II$\alpha$D</td>
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<td>→ Ne$^{3+}$ ($2s^2 2p^4 \ (3P)$) + Ne$^+$ ($2s^2 2p^4 \ (3P)$ $3s \ 3P$)</td>
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<td>→ Ne$^{3+}$ ($2s^2 2p^4 \ (3P)$) + Ne$^+$ ($2s^2 2p^4 \ (3P)$ $3s \ 3P$)</td>
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<td>→ Ne$^{3+}$ ($2s^2 2p^4 \ (3P)$) + Ne$^+$ ($2s^2 2p^4 \ (3P)$ $3s \ 3P$)</td>
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<td>+9.56</td>
<td>II$\gamma$D</td>
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*Note: The reactions involving Ne$^{3+}$ and Ne$^*$ are simplified representations for the purpose of this table.*
<table>
<thead>
<tr>
<th>Reactant and initial states</th>
<th>Product and final states</th>
<th>$\Delta E$(eV)</th>
<th>Designation of reaction process</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\rightarrow$ Ne$^3^+$ (2s$^2$ 2p$^4$ 'D) + Ne$^+$ (2s$^2$ 2p$^4$ (3P) 3p $^4$P)</td>
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<td>+15.89</td>
<td>III$\beta$D</td>
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<td>$\rightarrow$ Ne$^2^+$ (2s$^2$ 2p$^4$ 'S) + Ne$^+$ (2s 2p$^6$ 'S)</td>
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<td>+15.78</td>
<td>III$\gamma$A</td>
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<td>$\rightarrow$ Ne$^3^+$ (2s$^2$ 2p$^4$ 'S) + Ne$^+$ (2s$^2$ 2p$^4$ (3P) 3p $^4$P)</td>
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<td>+12.18</td>
<td>III$\gamma$D</td>
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<td>$\rightarrow$ Ne$^3^+$ (2s$^2$ 2p$^4$ 'P) + Ne$^2^+$ (2s$^2$ 2p$^4$ 'P) + e</td>
<td></td>
<td>+8.52</td>
<td>III$\alpha$Y</td>
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<table>
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<th>$\Delta E$ (eV)</th>
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<td>$\text{Ne}^{3+}$ $(2s^2 , 2p^3 , {}^4S)$ + $\text{Ar}$ $(3p^6 , {}^1S)$</td>
<td>$\text{Ne}^{2+}$ $(2s , 2p^2 , {}^3P)$ + $\text{Ar}^+$ $(3p^3 , {}^3P)$</td>
<td>+22.37</td>
<td>$\text{IX}$</td>
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<td>$\rightarrow$</td>
<td>$\text{Ne}^{2+}$ $(2s , 2p^3 , {}^1P)$ + $\text{Ar}^+$</td>
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<td>$\rightarrow$</td>
<td>$\text{Ne}^{2+}$ $(2s^2 , 2p^3 , {}^4S , 3p , {}^5P)$ + $\text{Ar}^+$</td>
<td>+3.98</td>
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<td>$\text{Ne}^{2+}$ $(2s^2 , 2p^3 , {}^1P , 3s , {}^3P)$ + $\text{Ar}^+$</td>
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<td>$\rightarrow$</td>
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<td>+3.47</td>
<td>$\text{I}_{\epsilon}X$</td>
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<td>$\rightarrow$</td>
<td>$\text{Ne}^{2+}$ $(2s^2 , 2p^3 , {}^1P , 3s , {}^3S)$ + $\text{Ar}^+$ $(3s , 3p^6 , {}^2S)$</td>
<td>+8.89</td>
<td>$\text{I}_{\delta}A$</td>
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<tr>
<td>$\rightarrow$</td>
<td>$\text{Ne}^{2+}$ $(2s^2 , 2p^3 , {}^1P , 3d , {}^3D)$ + $\text{Ar}^+$ $(4s , {}^4P)$</td>
<td>+5.97</td>
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<td>+5.73</td>
<td>$\text{I}_{\delta}C$</td>
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<tr>
<td>$\rightarrow$</td>
<td>$\text{Ne}^{2+}$ $(2s^2 , 2p^3 , {}^1P , 3d , {}^4F)$ + $\text{Ar}^+$ $(3d , {}^4P)$ + e</td>
<td>+5.23</td>
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<td>$\text{I}_{\delta}E$</td>
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<td>+16.75</td>
<td>$\text{I}_{\beta}Y$</td>
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Table 3—Continued

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<tr>
<td>$\rightarrow$ Ne$^{2+}$ (2s 2p$^3$ 3P) + Ar$^+$ (4s $^2$P)</td>
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<td>III6C</td>
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<tr>
<td>$\rightarrow$ Ne$^{2+}$ (2s 2p$^3$ 3P) + Ar$^+$ (4s $^2$P)</td>
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<td>( \text{Ne}^3+ (2p^2 3p 4\text{S}) )</td>
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<th>Ar</th>
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<td>( \text{Ne}^{++}(2s^2 , 2p , {}^3P) \text{ Ne} )</td>
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<td>13.78</td>
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CHAPTER V

RESULTS AND DISCUSSION

Graphical presentations of energy-gain spectra and total differential cross sections for single-electron capture processes are shown in Figures 14-18 and 20-29. The observed reaction channels for Ne$^{3+}$ are shown in Figures 14, 16, and 17, respectively, and are labeled according to Hasted's notation as previously used by Kamber (1988). The observed reaction channels are assigned the designations I, II, and III, representing the ground, first and second metastable states of the incident Ne$^\ast$ ion beam, respectively; $\alpha, \beta, \gamma, \ldots$ represent the ground and successively higher excited states of the projectile product Ne$^\ast$; and X, A, B, $\ldots$ represent the ground and successive excited states of the target product; Y represents the second ionization state of the target.

The multichannel Landau-Zener model was used to interpret the final state populations by calculating the location of the reaction window in terms of the Q-value of the final states. The reaction windows were calculated based on the expression for $H_{12}$ with Taulbjerg (1986) factors $f_{n} = 1$ (i.e., $H_{12}$ contains the factor 9.13) (see Equations 2.18, 2.24, and 2.25) and the $f_{m}$ value obtained with the quantum numbers $n$ and $l$ for the captured electron (see Equation 2.25). For fully-stripped ions $H_{12} = f_{n} H_{12}^{\ast}$ and for partially-stripped ions $H_{12} = H_{12}^{\ast}$. 42
Before delving into the results of the present study, a number of comments need to be made. First, for each charge state, at least three collision energies were studied. However, because of limited space, one or two collision energies were chosen and discussed for each charge state. Second, for each system, single-electron capture was studied using a large number of scattering angles between 0°-6°. The angles presented here were chosen based on high statistics.

Third, other investigators have discussed angular distributions for Ne3+, Ne4+, Ne5+, and Ne6+ (Tunnell et al., 1987). Angular distributions for Ne4+ - Ne and Ne5+ - Ne were measured here in order to contribute to the present literature on single-electron capture (see also Roncin, Barat, & Laurent, 1986). However, the choice of angular distributions presented here was done to be as representative as possible given the space allowed for this study. Fourth, our angular measurements show the differential cross section $\frac{d\sigma}{d\theta}$ rather than $\frac{d\sigma}{d\Omega}$, which has to be multiplied by $2\pi\sin\theta$ in order to obtain $\frac{d\sigma}{d\theta}$. Thus, the differential cross section $\frac{d\sigma}{d\theta}$ tends to go to zero for $\theta = 0^\circ$. Fifth, in the present measurements, projectile ions lose some translational energy in an electron capture collision process. Thus, the energy-gain spectra of the scattered particles are conveniently expressed in terms of the Q-value rather than the energy defect $\Delta E$ as indicated in Tables 1-6. In the following sections, the results of individual collision systems are presented and discussed.
Ne$^{3+}$ - He Collisions

Figure 14 shows the translational energy-gain spectra obtained for single-electron capture by 150 eV Ne$^{3+}$ ions from He at different scattering angles. The possible reaction channels are listed in Table 1. At 0° scattering angle, the dominant reaction channel is \text{IIX} due to single-electron capture from the $2p^{3}\frac{3}{2}P$ metastable state of Ne$^{3+}$ into the $2s2p^5 \frac{1}{2}P$ state of Ne$^{2+}$ with crossing radius at $R_x = 5.44$ a.u., where $R_x = (q-1)27.2/Q$ (eV), (Kamber, 1988) neglecting polarization (Dalgaro & Butler, 1978). Since the significant non curve crossing must occur in the range $R = 3 - 10$ a.u. to be consistent with the reaction window theory, $R_x = 5.44$ a.u. conforms to this theory. A small peak at $Q = 13$ eV is due to capture from ground state Ne$^{3+}$ into the $2s2p^5 \frac{3}{2}P$ state of Ne$^{2+}$ via reaction channel \text{IX}. The other peak at around 6 eV is due to capture into the $3s \frac{5}{2}S$ and $3s \frac{3}{2}S$ states via reaction channels \text{III}_{\geq}X$ and \text{III}_{<}X$, respectively, with an additional contribution from the \text{IIX} channel. Both peaks can only be explained due to core-changing (excited core) single-electron capture transitions since the final Ne$^{2+}$ ($2s2p^5$) states cannot be described by single configuration states (Winter, 1991).

Lebius, Koslowski, and Huber (1989) used translational energy-gain spectroscopy to determine the relative populations of ground and metastable states in a Ne$^{3+}$ ion beam produced in an electron impact ion source. They determined the $^4S$, $^2D$ and $^2P$ state abundances to be 0.2, 0.5 and 0.3, respectively, in accordance with their statistical populations. The measurements obtained in the present study

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for reaction channel IIIεX show good agreement with those of Lebius et al. (1989) at a collision energy of 600 eV, but there is no clear evidence in their measurement of a contribution from reaction IδX. Their measurements also show smaller contributions from the processes IIεX, IIIεX and IIIηX. The present results disagree substantially, however, with those of Schmeissner et al. (1984) for which the dominant reaction channel was observed to be IδX and there is no evidence of contributions from metastable states in the Ne^3* beam. The reason for disagreement with the results of Schmeissner et al. (1984) and those of Lebius, Koslowski, and Huber (1989) is not known.

As the projectile scattering angle is increased, reaction channel IδX becomes more pronounced relative to the reaction channels IIIεX, IIIηX and IIηX. This indicates that angular distributions for capture by metastable Ne^3* ions into 2s2p^5 1P and 3s^3 3S states of Ne^2* are more strongly peaked in the forward direction than for process IδX. At a scattering angle of 3.15°, the dominant peak is observed to be shifted toward a lower Q-value due to the additional translational energy given to the target as the scattering angle increases.

Reaction windows are also shown in Figure 14. These are calculated using a multichannel Landau-Zener model (Salop and Olson, 1975), based on the Taulbjerg expression for H_{12} with Taulbjerg factors f_n = 1 and f_n(2p) = 0.707 (where n = 2 and l = 1) (see Equation 2.25) (Taulbjerg, 1986). Calculated peak values have been normalized to our observed peak values in the energy spectrum. Here, it can be
seen that the reaction window based on $f_{\text{fd}} = 1$ favors reaction channel IIeX (dotted curve), while the reaction window based on $f_{\text{fd}} = 0.707$, which is calculated according to capture into 2s2p$^5$ state (broken curve), does not describe the position of the dominant process since it involves a core-changing transition; i.e., a two-electron process.

Note: Also Shown are Reaction Windows Calculated on the Basis of a Single-Crossing Landau-Zener Theory Using Values of $H_1$ (see Equations 2.18, 2.24, and 2.25) Containing Factors 9.13 (Dotted Curve) and 6.455 (Broken Curve).

Figure 14. Translational Energy Spectra for Single-Electron Capture by 150 eV Ne$^+$ Ions from He at Different Scattering Angles.
The measured differential cross section for single-electron capture by Ne\(^{3+}\) ions from He at a collision energy of 300 eV is shown in Figure 15. Absolute cross section scales were assigned to the data by normalizing the results of this study to the total cross section measurements of Tunnell et al. (1987). The experimental data show a broad distribution with a maximum near a scattering angle of about 2.1°. The data also show that the projectile products are distributed mostly at angles larger than \(\theta_c = 1.02°\), which corresponds to capture at an impact parameter equal to the crossing radius. For small angles, \(\theta_c = Q/2E\), where \(Q\) is the exoergicity of the collision and

**Figure 15.** Total Differential Cross Section for Single-Electron Capture by 300 eV Ne\(^{3+}\) from He.

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E is the laboratory impact energy (Cocke, Kamber, Tunnel, Varghese & Waggoner, 1987). In measurements by Tunnell et al. (1987) for this collision system at 285 eV impact energy, the angular distribution rises to a maximum at 0.57° in disagreement with this experiment where the angular distribution peaks at 2.1°. The reason for this disagreement might be due to the fact that their measurements were recorded with higher angular resolution compared to ours.

Ne⁵⁺ - Ne Collisions

Figure 16 shows the translational energy-gain spectra of the product Ne²⁺ following single-electron capture in Ne⁵⁺ - Ne collisions at a collision energy of 150 eV and different projectile scattering angles. The possible exit channels are listed in Table 2. At θ = 0°, capture from the ground state of Ne⁵⁺ into the ground state of Ne²⁺ accompanied by excitation of the target product dominates the single-electron capture processes via reaction channels IαA and IαC, with additional contributions from the IIIγA and IIIβD channels. The broad peak at about 8 eV is due to capture into the 3s 3S and 3S states via reaction channels IIγX and IIγX, and with contributions from the channels IγA, IγC, and IIIaY. Again, the results of Lebius, Koslowski, and Huber (1989) show good agreement with the measurements obtained in this study. The reaction window based on W₁ favors smaller energy-gain values than observed, and accommodates part of the observed peak at Q = 8 eV. Other reaction windows are not calculated for this collision system because the
Note: Also Shown is the Reaction Window Calculated on the Basis of a Single-Crossing Landau-Zener Theory Using Values of $H_{12}$ Containing the Factor 9.13 (Dotted Curve).

Figure 16. Translational Energy Spectra for Single-Electron Capture by 150 eV Ne$^{+}$ Ions from Ne at Different Scattering Angles.

Dominant reaction channels are due to capture from metastable state.

As the scattering angle is increased, reaction channel IaA remains dominant, but the relative importance of capture into the 3s states of Ne$^{2+}$ (the peak at about 8 eV) is strongly increased at $\theta = 1.86^\circ$. This indicates that the angular distribution for capture into the 3s states is not maximum at 0° but instead peaks at a greater angle. For a scattering angle of 2.8°, the reaction channels IIIγA, IIIβD, and IδX are significantly populated and become equivalent to the height of reaction channel IaA. In general, at the
smaller angles, the larger impact parameter collisions play an important role in the electron capture process since the non-crossings at small internuclear separation cannot be reached, and make no contribution. As the angle is increased, contributions from successively smaller internuclear separation regions make their appearance (Kamber, 1988). Therefore, the peak at 8 eV disappears between $\theta = 2.8^\circ$ and $4.73^\circ$ as shown in Figure 16.

**Ne$^3+$ - Ar Collisions**

Figure 17 shows typical translational energy spectra for collisions of Ne$^3+$ with Ar at a collision energy of 150 eV and different scattering angles; the possible exit channels following single-electron capture are listed in Table 3. At 0° scattering angle, two peaks are clearly resolved. The strongest peak correlates with single-electron capture from ground state incident Ne$^3+$ $(2s^2 \ 2p^3 \ 4S)$ into $3s$ states of Ne$^{2+}$ via reaction channels $I_{\pi}X$ and $I_{\sigma}X$, with additional contributions from the reaction channels $II_{\pi}X$ and $II_{\sigma}X$. The other peak arises from capture into the $2s2p^1 \ 3P$ state of Ne$^{2+}$ by the Ne$^3+$ $(2s^2 \ 2p^3 \ 4S)$ ground state incident beam accompanied by excitation of the target product via reaction channels $II_{\delta}B$, $II_{\delta}C$, $II_{\delta}D$ and $II_{\delta}E$. The calculated reaction windows based on $f_{n} - 1$ and $f_{n} (3s) = 0.866$ ($n = 3$ and $l = 0$) are shown in the spectrum at 0°. The reaction window based on the $f_{n} = 1$ provides the best description of the observed peak at $Q = 5$ eV, but neither agrees very well.

As the projectile scattering angle is increased, single-electron
Note: Also shown are reaction windows calculated on the basis of a single-crossing Landau-Zener theory using values of $H_2$ containing factors 9.13 (dotted curve) and 7.906 (broken curve).

Figure 17. Translational energy spectra for single-electron capture by 150 eV Ne$^{3+}$ ions from Ar at different scattering angles.

capture into the 3s states remains strong, but the relative importance of the processes ISB, IS$^6$C, IS$^6$D, and IS$^6$E strongly decreases.

This indicates that the angular distribution for these later processes is much more forward peaked than it is for the 3s states. The broad peaks, centered around 15 eV, are contributions due to transfer ionization (TI) resulting from double-electron capture into autoionizing states which emit electrons before detection and thus appear to have captured only a single-electron. TI processes strongly increase
with projectile scattering angle as contributions from smaller intermolecular separations make their appearance.

**Ne$^{4+}$ - He Collisions**

Figure 18 shows the translational energy-gain spectra obtained for single-electron capture by 100 eV Ne$^{4+}$ ions from He at different scattering angles. At 0° scattering angle, one peak is clearly seen.

![Figure 18](image_url)

Note: Also shown are reaction windows calculated on the basis of a single-crossing Landau-Zener theory using values of $H_{12}$ containing factors 9.13 (Dotted Curve) and 7.906 (Broken Curve).

Figure 18. Translational Energy Spectra for Single-Electron Capture by 100 eV Ne$^{4+}$ Ions from He at Different Scattering Angles.

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This peak correlates with capture into the excited state $3s \, ^4P$ of the Ne$^{3+}$ ion from ground state incident Ne$^{4+}$ ($2p^2 \, 3P$) ions (see Table 4), in agreement with previous measurements at higher energies and forward scattering angles (Schmeissner et al., 1984; Tunnell et al., 1987). There is an unresolved contribution at about 10.5 eV involving capture into the 3s $^2P$ state. The reaction windows, however, are positioned closer to the reaction channel correlated with capture into the 3s $^2P$ state.

Besides ground state $2p^2 \, 3P$, incident Ne$^{4+}$ ions in the collision system may contain the low-lying metastable states $^1D$ and $^1S$, with excitation energies of 3.75 and 7.92 eV, respectively. Assuming that the metastable fraction ($F$) in the Ne$^{4+}$ ion beam is statistically distributed, then $F(^3P) : F(^1D) : F(^1S) = 9:5:1$. Therefore, the only reaction channel associated with the presence of metastable states in the incident ion beam and compatible with capture into the 3s $^4P$ state is identified with the process (see Table 4):

$$\text{Ne}^{4+}(2p^2^1D) + \text{He} \rightarrow \text{Ne}^{3+}(2p^2^1D) \, 3s^2D) + \text{He}^+ + e^-(11.49 \text{ eV})$$

representing capture from the lowest metastable state of Ne$^{4+}$ to an excited state of Ne$^{3+}$. Furthermore, this process is a core-conserving (core remains passive) reaction.

As the scattering angle is increased, a second peak due to capture into the 3s $^2P$ state of Ne$^{3+}$ is found to open at $\theta \approx 1.5^\circ$. As the scattering angle increases further, contributions from capture into the 3s $^2P$ state increase, and its maximum shifts toward lower
exothermicity. For this collision system, the Q values for the observed channels are 11.65 and 10.4 eV respectively. Calculated kinematics and experimental values for these reaction channels are shown in Figure 19, where Q($\theta_p$) functions are shown. It is interesting to note that at the large projectile scattering angles where the translational energy of the recoil ions becomes important, the Q values decrease as would be expected from kinematic effects. However, the experimental Q values for capture into 3s $^4P$ remains constant with increasing projectile scattering angles. The reason for angular

![Figure 19. Experimental and Calculated Energy-Gain (Q) Values for Capture into 3s $^4P$ and 3s $^2P$ Versus Projectile Scattering Angle $\theta_p$.](image)

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scattering influencing the channel correlated with capture into 3s^2P and not with that due to capture into 3s^4P will be the subject of further investigation and will be discussed in detail in a forthcoming work. This is the only collision system for which we observed this effect.

The experimental differential cross section is shown in Figure 20. The experimental data show a broad distribution with a maximum near a scattering angle of 2°, which is greater than $\theta_c = 0.85°$.

![Figure 20](image.png)

**Figure 20.** Total Differential Cross Section for Single-Electron Capture by 400 eV Ne$^{+4}$ from He.

Tunnell et al. (1987) have studied the Ne$^{+4}$-He collision system.
in more detail. Their high resolution measurements show a strong forward peak close to 0° and an oscillatory structure with two discernable secondary maximum for angles greater than 0°. These oscillations were well reproduced by Tan and Lin (1988) using a quantal two-channel calculation. However, these oscillations are not seen in the present work.

**Ne⁴⁺ - Ne Collisions**

Figure 21 shows the translational energy spectra of product Ne³⁺.

![Figure 21](image)

*Note: Also Shown are Reaction Windows Calculated on the Basis of a Single-Crossing Landau-Zener Theory Using Values of H₂ Containing Factors 9.13 (Dotted Curve) and -6.464 (Broken Curve).*

**Figure 21.** Translational Energy Spectra for Single-Electron Capture by 400 eV Ne⁴⁺ Ions from Ne at Different Scattering Angles.

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for Ne$^{4+}$ - Ne collisions at an impact energy of 400 eV and different scattering angles. At 0° scattering angle, capture into $2p^2(^3P)3p$ states of Ne$^3+$ at $Q = 10.55$ and 9.49 eV are observed to be the dominant reaction channels (see Table 4). There are smaller contributions due to capture into $2p^2(^3P)3s$ states at $Q = 16.22$ and 15.01 eV. There is probably also some contribution due to core-changing single-electron capture into the $2p^2(^1D)3s$ state (shown in Figure 21 as 3s'). The reaction window based on the factor $f_n(3p) = -0.708$ ($n = 3$ and $l = 1$) accommodates most of the dominant reaction channel, but the curve for $f_n = 1$ is also in quite good agreement.

As the angle is increased, capture into the 3p state of Ne$^3+$ remains dominant, but contributions from successively smaller internuclear separations (large Q-values) make their appearance as would be expected. This indicates that the 3p state produced in the single-electron capture process is strongly forward scattered. The reaction channels are significantly populated in the collision process and are comparable to the dominant channel for scattering angles approaching 4°. This is due to capture into 3s and 3p states of Ne$^3+$, respectively, from the ground state Ne$^{4+}$ ($2p^2\,{}^3P$) and the low-lying metastable states.

Figure 22 displays the measured differential cross section for single-electron capture by Ne$^{4+}$ ions from Ne at a collision energy of 400 eV. The total differential cross section rises to a maximum at an angle of 1.4°, which is greater than $\theta_c = 0.752^\circ$, where $\theta_c = Q/2E$. The oscillations between $\theta = 0.5^\circ$ and 1° in the total differential cross section.
cross section have no physical significance.

Figure 22. Total Differential Cross Section for Single-Electron Capture by 400 eV Ne$^{4+}$ from Ne.

Ne$^{4+}$ - Ar Collisions

Typical translational energy-gain spectra showing peaks due to single-electron capture into the excited states of the projectile product are shown in Figure 23 for 200 eV Ne$^{4+}$ - Ar collisions for several scattering angles. The zero-angle spectrum shows one peak, with a shoulder at $Q = 11.5$ eV. The main peak arises from capture into the 3d state of Ne$^{3+}$ from ground state Ne$^{4+}(2p^{23}P)$ (see Table 4).
Note: Also shown are reaction windows calculated on the basis of a single-crossing Landau-Zener theory using values of $H_{12}$ containing factors 9.13 (dotted curve) and 3.725 (broken curve).

Figure 23. Translational energy spectra for single-electron capture by 200 eV Ne$^{4+}$ ions from Ar at different scattering angles.

The unresolved peak at about $Q = 12$ eV is due to core-changing single-electron capture into the $2p^2$ ($^1D$) 3p state of Ne$^{3+}$. Comparison with the results of Giese et al. (1986) at 2180 eV shows good agreement with the present measurements. This is due to the fact that electron capture occurs within a five-subshell range; and, although different subshells are populated at different energies, the same spectrum is obtained within the same shell. The reaction window based on the factor $f_{n}(3d) = 0.408$ (where $n = 3, 1-2$) is positioned...
closer to the observed peak.

Other reaction channels having higher exoergicity due to capture into 3p states open at scattering angles larger than 1.5°. As the scattering angle is increased, capture into the 3d state of Ne stops remains dominant, but the relative importance of capture into 2p²(3P)3p and 2p²(3P)3p states (see Table 4) is strongly increased.

Ne²⁺ - He Collisions

The translational energy-gain spectra for this collision system are shown in Figure 24, and the relevant reaction channels are indicated in Table 5. The observed spectra and Table 5 show that the dominant reaction channel is due to capture from ground state Ne²⁺ into the excited states 3d of Ne⁴⁺. However, there may also be a contribution from the reaction involving capture into the 3p state. For this collision system, it can be seen that the calculated reaction window based on the factor \( f_n(3d) = 0.408 \) (n = 3, and \( l = 2 \)) accommodates most of the observed features, while the reaction window based on the factor 9.13 favors smaller Q-values than observed.

Schmeissner et al. (1984) have studied the same collision at an incident energy of 358 eV. They found the dominant reaction channels are due to the 3d states with a contribution from the 3p state, which is in agreement with the results obtained in this experiment. Tunnell et al. (1987) have studied this collision system at an energy of 2821 eV. They found that the reaction channel corresponding to capture into the 3p excited state produced a larger contribution than
did the channel corresponding to capture into the 3d state. This could be attributed to the high collision energy used, since the position of the reaction windows depends modestly on the collision energy.

Note: Also shown are reaction windows calculated on the basis of a single-crossing Landau-Zener theory using values of $H_1$ containing factors 9.13 (dotted curve) and 3.725 (broken curve).

Figure 24. Translational energy spectra for single-electron capture by 250 eV Ne$^{+}$ ions from He at different scattering angles.

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energy (Taulbjerg, 1986).

\section*{Ne$^5^+$ - Ne Collisions}

Figure 25 shows the translational energy-gain spectra of the

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure25.png}
\caption{Translational Energy Spectra for Single-Electron Capture by 250 eV Ne$^5^+$ Ions from Ne at Different Scattering Angles.}
\end{figure}

Note: Also Shown are Reaction Windows Calculated on the Basis of a Single-Crossing Landau-Zener Theory Using Values of $H_{12}$ Containing Factors 9.13 (Dotted Curve) and 3.725 (Broken Curve).

product Ne$^4^+$ ions in the Ne$^5^+$ - Ne collisions at an impact energy of 250 eV and different scattering angles. The observed collision spectrum at 0° scattering angle is dominated by single-electron capture.
from ground state Ne\textsuperscript{5+} (2p \textsuperscript{2}P) ions into the 3d \textsuperscript{1}F state of Ne\textsuperscript{4+} product ions (see Table 5). The broad peak at about Q = 10 eV can be attributed to double-electron capture processes (Roncin et al. 1986). The reaction window based on the factor 9.13 favors Q-values smaller than those observed, and is positioned closer to the peak correlated with autoionizing double capture processes.

As the scattering angle is increased, capture into 3d state of Ne\textsuperscript{4+} remains dominant, and the relative cross section for the autoionizing double-capture process goes through a maximum at an angle of \( \approx 2^\circ \) and the peak completely disappears for scattering angles larger than 4.5\(^\circ\). Roncin, Barat, and Laurent (1986) measured the differential cross sections for single- and double-electron capture by multiply-charged ions. They found that for single-electron capture, scattering occurs in the forward direction, whereas for electrons captured successively, scattering occurs at finite angles.

Figure 26 displays the measured total differential cross section for single-electron capture by 500 eV Ne\textsuperscript{5+} ions from Ne. The data show that the distribution for capture is peaked at about 1.6\(^\circ\) and is a relatively smooth function. For capture into the 3d state with \( q = 15 \) eV, the corresponding value of \( \theta_c \) is 0.85\(^\circ\).

\textbf{Ne\textsuperscript{5+} - Ar Collisions}

Typical translational energy-gain spectra of product Ne\textsuperscript{4+} for 250 eV Ne\textsuperscript{5+} - Ar collisions are shown in Figure 27. At 0\(^\circ\) scattering angle, the observed spectrum is dominated by single-electron capture into the
4s excited state of Ne$^{4+}$. The structures in the spectra at around 18 and 30 eV are contributions from transfer ionization processes (T11 and T12), (see section Ne$^{3+}$ - Ar), which are possibly due to two-electron capture into doubly-excited states which autoionize back to different excited states of Ne$^{4+}$ before detection (Giese, Cocke, Waggoner, Pedersen, Kamber et al., 1987). The calculated reaction windows $f_{nl} = 1$ and $f_{nl}(4s) = -0.866$ (where $n = 4$ and $l = 0$) do not accommodate the dominant channel and favor smaller Q-values than the observed ones.

Again, as the scattering angle is increased, capture into the 4s
state of Ne$^{4+}$ remains dominant, but the relative importance of capture into the 3d state of Ne$^{4+}$ and the contributions from transfer

![Graph showing energy spectra for Ne$^{4+}$ collisions with He and Ar at different scattering angles.]

**Note:** Also shown are Reaction Windows Calculated on the Basis of a Single-Crossing Landau-Zener Theory Using Values of $H_{12}$ Containing Factors 9.13 (Dotted Curve) and -7.907 (Broken Curve).

**Figure 27.** Translational Energy Spectra for Single-Electron Capture by 250 eV Ne$^{5+}$ Ions from Ar at Different Scattering Angles.

ionization processes are increased.

Ne$^{6+}$ - He Collisions

**Figure 28** shows the translational energy-gain spectra for single-electron capture by 600 eV Ne$^{6+}$ ions from He at different scattering angles. At 0° scattering angle, two peaks are clearly resolved. The
peak at about 23 eV correlates with core-conserving single-electron capture into a 3d state while the other peak near 15 eV arises from core-changing capture into the 2s2p (\(^{1}\)P) 3l state of Ne\(^{5+}\). The spectrum shows that the relative cross section for the core-conserving channel is comparable with that for the core-changing channel. For this collision system, the reaction window based on the factor 9.13 is nearly positioned at the center of the peak at \(Q = 14\) eV. As the scattering angle is increased, the core-conserving capture channel

Note: Also Shown are Reaction Windows Calculated on the Basis of a Single-Crossing Landau-Zener Theory Using Values of \(H_2\) Containing Factors 9.13 (Dotted Curve) and 3.725 (Broken Curve).

Figure 28. Translational Energy Spectra for Single-Electron Capture by 600 eV Ne\(^{5+}\) Ions from He at Different Scattering Angles.

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(3d) remains dominant, while the relative importance of capture into the core-changing capture channel is strongly decreased.

Andersson et al. (1989) have studied the Ne$_8^*$ - He system at collision energies of 200, 500 and 2000 eV. Based on a simple semi-classical multistate collision model, they attribute the observed translational energy-gain spectra maxima entirely to core-changing single-electron capture processes. Hansen and Andersson (1989) studied single-electron capture in Ne$_6^*$ - He collisions at an energy of 2000 eV using atomic expansion close-coupling calculations, representing the target by a one-electron wavefunction with the correct binding energy. They showed that core-conserving single-electron capture into the 3d state is the dominant reaction channel, thus confirming the interpretations of this study. Recently, Beijers, Hoekstra, Morgenstern and de Heer (1992) have studied the competition between core-conserving and core-changing single-electron processes in Ne$_6^*$ - He collisions for impact energies between 1400 and 2400 eV by measuring the spectra of the VUV radiation emitted by the decaying product ions. Their results show that the capture cross sections for the core-conserving processes involving metastable primary ions are comparable with those for the ground state primary ions, and they identified capture into the Ne$_9^*$ (2s$^2$ 3d) state as the dominant reaction channel over the entire impact energy range investigated.

Ne$_6^*$ - Ne Collisions

In 600 eV Ne$_8^*$ - Ne collisions, the zero-angle spectrum
indicates that the dominant process is due to core-changing single-electron capture into the 2s 2p ($^3P$) 3s states. The structure in the spectrum at around 28 eV is due to core-conserving single-electron capture into the 3d state. However, as the scattering angle is increased, core-conserving single-electron capture into the 3d and 3p states of Ne$^5+$ starts to contribute. There is also some contribution due to transfer ionization. The calculated reaction based on the factor 9.13 accommodates most of the dominant peak in the spectrum at 0° (see Figure 29).

Note: Also Shown are Reaction Windows Calculated on the Basis of a Single-Crossing Landau-Zener Theory Using Values of $H_1^2$ Containing Factors 9.13 (Dotted Curve) and 3.725 (Broken Curve).

Figure 29. Translational Energy Spectra for Single-Electron Capture by 600 eV Ne$^8+$ Ions from Ne at Different Scattering Angles.
CHAPTER VI

CONCLUSION

Single-electron capture by low-energy multiply charged Ne\textsuperscript{x}\textsuperscript{*} ions from He, Ne, and Ar has been studied by means of translational energy-gain spectroscopy at laboratory collision energies between 100 and 600 eV, and at scattering angles between 0° and 6°. This study shows the importance of dynamic effects in single-electron capture processes. In particular, there appears to be an impact parameter (scattering angle) dependence of the electron capture spectra as expected, i.e., as the scattering angle is increased, contributions from successively smaller internuclear separations (large Q-values) make their appearance. It is also clear that the final-state populations of the projectile products following single-electron capture are very sensitive to the energy-level structure of the collision systems.

The energy-gain spectra were interpreted qualitatively in terms of the reaction window. The reaction windows were calculated based on the Taulbjerg (1986) expression for H\textsubscript{2} to describe electron capture by partially-stripped ions. The reaction window based on the factor for the dominant channel usually provides the best description of the observed features.

The measured differential cross sections show that the projectile products are distributed near a scattering angle $\theta_c$ corresponding.
to capture at an impact parameter equal to the crossing radius as expected.

In future work, a quantitative comparison between experiment and theory should be made with a model constructed by means of an extended version of the multi-state semiclassical model.
BIBLIOGRAPHY


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