EQUILIBRIUM CHARGE STATE DISTRIBUTIONS OF LOW-Z IONS INCIDENT ON THIN SELF-SUPPORTING FOILS

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Abstract

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Equilibrium charge fractions have been measured for 3 - 7 MeV lithium, boron, and carbon ions passing through thin carbon foils. The data are compared to the predictions of several semi-empirical models of charge equilibrium in the ≤ 1MeV/u regime. The current work underscores the general problem of extrapolating models developed for high-Z projectiles to ions of low-Z.

The charge fractions for lithium, boron and carbon ions passing through thin foils of carbon, aluminum, copper, silver, and gold are compared with the mean charge of the projectile, the functional form of the charge distribution, and the distribution width. These are parameters used to examine the effects of the electronic structure on charge exchange for various target-projectile combinations. Projectile shell structure is found to have a large influence on the widths of the charge state distribution and the data for these low-Z ions can be used to establish a baseline for more complicated electron systems encountered with ions of higher Z. Experimental techniques and comments on the nature of the equilibrium charge states of low-Z ions are presented.
Dedication

To those who believed...especially my wife.
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CHAPTER 1

INTRODUCTION

1.1 An Overview of Charge Exchange

The charge of a fast ion moving through matter oscillates as a result of two fundamental processes that make up charge exchange: electron capture and electron loss. In electron capture the charge state \(q\) of an ion with an atomic number \(Z\) decreases by one when accepting an electron from the target medium,

\[
Z^q + e^- \rightarrow Z^{q-1}
\]  

with \(q \geq 0\), for example Li\(^{2+}\) to Li\(^{1+}\). Conversely, electron loss is the process by which an electron is stripped off and the ion charge state is increased by one,

\[
Z^q \rightarrow Z^{q+1} + e^-
\]  

with \(q \geq -1\), for example Li\(^{2+}\) to Li\(^{3+}\). Figure 1.1 schematically represents the electron loss and capture process during charge-changing collisions between a lithium ion and a target. After a sufficient number of collisions, an equilibrium charge state distribution is established, where electron capture and loss processes become balanced and depends only on the ion species, the velocity of the ion, the target and its phase (solid or gas).
1.1 Figure 1.1. This is a schematic representation of the fundamental processes of charge exchange. a) This is Li$^{1+}$ ion before charge exchanging has occurred. b) The ion loses an electron through successive collisions within the target till the electron is stripped to become Li$^{2+}$. c) The ion can then also capture a bound target electron to become Li$^{1+}$. Note the black circles are electrons and the open circles represent electron vacancies.

1.2 Brief History of Charge Exchange

From the first decade of the twentieth century, physicists realized that an ion beam could become partially neutralized in collisions with a gas that it was passing through. Henderson in 1922 made measurements of charge-changing collisions of alpha particles from radioactive sources[27]. In 1924, Rutherford measured equilibrium fractions of helium ions exiting through foils[40]. Much of the early experimental work, post World War II, involved hydrogen and helium beams because they were readily available. Those experiments were performed using gas targets since at the time manufacturing thin solid targets was difficult. In the 1940s and 50s, groups at the University of Chicago worked in the 50 to 450 keV region, while groups at Oak Ridge National Laboratory worked up to 1 MeV using a Van de Graaff generator. The study of charge exchange research led the way to
the development of heavy ion accelerators like the tandem Van de Graaff (1960s). This device typically uses a carbon foil to strip away electrons to produce a second acceleration that allows for the production of higher energy ion beams (1 to 200 MeV)[5].

1.3 Scientific Motivations

Fast ions lose energy by Coulombic interactions with the electrons of a medium so knowledge of the charge state of the ion is essential to describing a number of fundamental properties including the stopping power of a medium and the range of the ions. Stopping power, for example, at lower energies is problematic for describing ions with different initial charge states, for example $dE/dx(^{12}C^{3+}) \neq dE/dx(^{12}C^{6+})$, see Fig 1.2.

While systematic studies in charge exchange are rarely performed there are dozens of papers published annually presenting new charge state distribution data. These results are very limited in scope, focusing only on a particular energy, ion and target that the experimenter needs to extract information. Generally, experimenters feel that the current charge exchange models provide dubious results, which necessitates a preliminary charge exchange experiment. Below is a description of several of these experiments and what they provide to the physics community in general:

1. There have been astrophysics-related experiments where helium ions collide with gas jet targets to mimic different astrophysical phenomena. In this case the ion-matter interaction imitates the solar wind colliding with cometary tails, a water gas jet target, to measure velocity distributions [16]. These charge-changing collisions emit x-ray information that can then be used to
Figure 1.2. The fluctuating stopping power in matter is due to repeated electron capture and loss, where incident charge states above or below merge at equilibrium. There is additional energy loss at each charge-changing collision[68].

compare with observational spectra lines. This data can be used to improve existing models and can lend some insight to the composition of comets and stellar wind velocity profiles.

2. Accelerator Mass Spectrometry (AMS) uses the gas-filled magnet technique to take advantage of charge-changing collisions in dilute gases to separate isobars of interest[48]. When ions have charge exchanged with a target and enter a magnetic field, a distribution of charge states can be observed. If this same magnetic region is now filled with a gas the charge state distribution will merge around the mean charge state for the given beam of ions. Multiple ion species will merge around their respective mean charge state as determined by the atomic number of the ion species. Typically, this method is used to separate isobars, for example $^{39}$K and $^{39}$Ar, see Fig 1.3.
Figure 1.3. (a) An ion beam charge exchanges with a foil and is magnetically separated into distinct charge states \((q, q+1, q+2, q+3)\).

(b) The same magnetic region is now filled with a gas and the distribution of charge states merges around the mean charge state \((\bar{q})\).

(c) A gas-filled magnet separating the isobars of argon and potassium\(^{[48]}\).
3. Radiation effects and dosimetry are two areas in health physics that are ultimately dependent on the charge state of the incident ion. The Bethe formula, which is used to calculate stopping power, is limited to high energies where the projectile ion is bare and does not carry atomic electrons with it. At lower energies, the effective charge must be used in the stopping power determination. Therefore, knowledge on the influence of charge exchange, electron loss and capture, on energy loss is important. The position of the stopping power maximum (Bragg peak) depends on the charge state and how the stopping power fluctuates along the ion trajectory, see Fig 1.2. As the charge state approaches equilibrium, where electron loss and capture collisions balance, the stopping power also stabilizes. With every charge-changing collision there is some energy loss that contributes to stopping power[68].

4. There are experimental nuclear reactions that are not easily achievable as a direct reaction, such as a beam of alpha particles on a short-lived heavy nucleus target. This scenario is where inverse kinematic reactions are more favorable by using a gas target of helium with the former target now the ion beam. Experiments by Liu et al.[37] provide charge exchange information needed for inverse kinematic nuclear reactions that are performed with a recoil mass separator, which has a single charge state acceptance. Data published by Liu et al.[37] emphasizes that few charge exchange studies focus on low energy, low-Z ions traveling through gas.

5. Ion sources use charge exchange to produce ions for the initial boost before being injected into an accelerator, see the negative ion source SNICS in chapter 3. An Electron Cyclotron Resonance Ion Source (ECRIS) is able to
produce singly-charged ions with high intensities (mA’s) and for multiply-charged ions the ECRIS can confine the ions long enough for the multiple charge-changing collisions to produce multiple ionizations and higher initial charge states for high-Z ion beams[23]. ECRIS is better known for the latter.

6. There are instances in accelerators where regions of poor vacuum or residual gas can cause unwanted charge-changing collisions [46]. This spurious charge exchange can potentially influence the outcome of experiments, especially those with sensitive detection methods such as AMS. The simple way to overcome such problems is to introduce additional pumping near the charge exchanging source [78]. There is an instance where an intentional volume of residual gas is desirable, for example, the gas canal in a tandem accelerator, where it functions as an electron stripping medium. Gas stripping is more stable than using carbon foils as those are affected by radiation damage and break. Gas stripping typically only populates lower charge states, but additional pumping at the terminal would allow for an increase in gas density thereby allowing for larger charge states to be populated, see Fig 1.4 [31].
Significant gaps exist in the data for the charge state distribution of low energy ions in solid materials, which makes it difficult to determine the trustworthiness of stopping power and range compilations. Of the many compilations and reviews that have been written, the focus has been on carbon targets. There has been no equivalent systematic study with other target species. Many of the semi-empirical models cannot reproduce the low-Z projectile results based on experiment because the formula were built from relatively high-Z projectiles \((Z > 6)\). The models cannot extrapolate well to lower energies because the charge fractions are now dominated by fully stripped and proton-like ions.

A target dependence has been noted in some papers and efforts have been made to develop expressions to incorporate target species other than carbon \([10, 51, 52, 57]\). For those not familiar with the described phenomena, extensive reviews including those by Allison\([5]\), Betz\([10]\), Wittkower\([79, 80]\), and Shima\([66]\) provide some of the theoretical background and experimental techniques as well as collected data on charge state distributions.

1.4 Outline of the Following Chapters

The work presented in the following chapters is data collected from low energy, low-Z ions through some common target materials to study the behavior of the charge state distribution, the mean charge and the projectile shell structure. Low-Z ions in this energy regime have not received much attention and can provide practical information relating mean charge and charge fractions for experimenters. These types of charge exchange studies have been performed for some high-Z ions through carbon at varying energies, but not for low-Z ions. This work will also look for evidence of target dependence at lower energies, where low-Z ions can be
used to establish a baseline for more complicated electron systems.

The remaining chapters are organized to reflect the progression from theory to experiment to results. Chapter 2 will present some of the theoretical aspects of the charge-changing process as well as several models that have and still influence experimental decisions. Chapter 3 will present the experimental facilities used to perform the present work and the preparation of solid targets. Chapter 4 will outline the charge exchange experimental procedures and discuss the results from the present work. Finally, chapter 5 will provide concluding remarks.
2.1 Introduction

In this chapter, the theoretical frameworks that can best describe charge exchange in the most fundamental terms are considered. In the first section, some of the basic mechanisms of charge exchange are listed. Next, a mathematical description that relates to charge-changing cross-sections will be derived and discussed. Then how ions are influenced by the charge exchanging medium is considered, \textit{i.e.} the density effect. Basic parameters such as the mean charge state, the distribution width and the distribution skewness (the symmetry) from charge state fractions are then defined and calculated. Finally, the most common semi-empirical models for determining equilibrium mean charge states and their limitations are described.

2.2 Mechanisms of Charge Exchange

The most basic processes in atomic charge exchange are electron capture and loss. When an ion with a charge $q$ collides with a target atom several processes may occur that change the charge into $q'$. Figure 2.1 by H. D. Betz [12, 15, 40] pictorially shows a few of the single and multiple electron processes in ion-atom collisions:
Figure 2.1. These are the processes that H.D. Betz considered the fundamental mechanisms of charge exchange[12].

1. Nonradiative (Coulomb) Electron Capture - The ion captures one or more bound electrons from a target atom into its ground or excited states. Free electrons are restricted based on momentum and energy considerations. If an electron moves from a target atom to a fast moving projectile, the electron must have components of high momenta that can match the high momenta of the projectile[22].

2. Radiative Electron Capture - The ion captures a free electron or a bound target electron into its ground or excited states accompanied by the simultaneous emission of a photon, see Fig 2.2. This competing electron capture process is not restricted by momentum and energy considerations, where the emission of a photon acts a third body carrying away energy and momentum
Figure 2.2. A schematic for radiative electron capture of a bound target electron in a collision with a moving ion, where a) shows the ion moving near a bound electron and b) where the electron capture and simultaneous photon emission occur. This mechanism of electron capture can also happen with a free electron[12].

relaxing the momentum matching required for electron capture[22].

3. **Excitation to the Continuum** - The ion loses one or more electrons, leaving the ion in either its ground or an excited state. Singly excited ions usually return quickly to the ground state; however metastable states that have long lifetimes can be produced. There is a definite possibility that ions are still excited when a subsequent collision takes place. An ion left in an excited state may undergo an Auger process, causing ionization after the actual collision has occurred. The Auger processes are the transition of an electron from a higher energy level to fill an inner shell vacancy, where the excess energy can either emit a photon or cause the emission of an electron, see Fig 2.3.

Despite the simple schematic representation of Fig 2.1, the charge exchanging processes are much more complicated. In fact for fast collision encounters, where the ion velocity is larger than the orbital velocity of the transferred electron, these processes can occur simultaneously. In addition, there are several other processes
Figure 2.3. A schematic for the production and destruction of an inner-shell vacancy moving through a solid target [12].

that potentially could occur. These processes are outside the scope of the present work and will only be mentioned and not discussed: the capture of two or more electrons, transfer excitation, transfer ionization, multiple outer-shell ionization and inner-shell ionization. References to these other processes can be found in McDaniel [40], Stolterfoht [70], Shelvelko [55].

2.3 Charge Exchange Cross-Sections

The charge exchange cross-section is described in the following manner. If there are \( N_i \) particles in the initial charge state incident on a target of number density \( n \) and linear thickness \( l \) then the areal density (thickness) is \( x = nl \) (atoms \( cm^{-2} \)). The number of particles exiting from this target in a given final charge state can be expressed as

\[
N_f = N_i x \sigma_{if},
\]  

(2.1)
where $\sigma_{if}$ is the cross-section for the change in charge from “i” the initial charge state to “f” the final charge state. Here the target is assumed to be “thin” meaning that $x\sigma \ll 1$, where $\sigma$ is any cross-section that can have an effect on the charge state. This assumption implies that a “thin” target has a small probability of interaction (cross-section), which allows for single ion-atom collisions to be the most prominent case[40]. The single ion-atom collision condition says that the ion loses or captures a single electron per collision ($q \pm n$), where $q$ is the charge state and $n$ is an integer. The positive and negative signs characterize the electron loss and capture by the ions, respectively.

Cross-sections are generally small at low energies since the electron has time to adjust adiabatically to the changing potential as the collision proceeds and the probability of a transition is small. The probability of charge exchange increases with increasing energy, but the interaction time is reduced as the velocity of the projectile is increased. This reduced interaction time eventually results in a lower probability at high energies, see Fig 2.4.

2.4 The Two-Component System

If a projectile ion makes multiple collisions while traversing a target, it may change its charge state several times. The simplest projectile to study is hydrogen and it can exist as a fully stripped ion, $H^+$, a neutral atom, $H^0$, or as a negative ion, $H^-$. Collisions of hydrogen atoms or ions with a target can lead to the product of either its original charge state or any of the other two. Therefore, there are six possible charge-changing cross-sections designated by $\sigma_{if}$, where “i” is the initial charge state and “f” is the final charge state. Figure 2.4 is an example of $H^+\text{+He}$ collision system and the related energy dependence of those cross-sections.
Above a couple hundred keV, the H$^-$ charge state becomes negligible, so that the initial charge state of the projectile is 0 or 1+. Under those conditions hydrogen becomes a two-component system with only two cross-sections, where $\sigma_{10}$ and $\sigma_{01}$ are needed to describe the charge-changing transitions.

The equations that are derived in a two-component system can be written to describe the gain and loss of neutral fractions $F_0$ and the fully stripped (1+) fractions $F_1$ after traversing through a given thickness. First, one can look at the neutral beam component, $F_0$, after it travels a distance $dx$ through the target, which will equate to the gain of neutral particles ($F_1\sigma_{10}dx$) minus the loss of neutral particles ($F_0\sigma_{01}dx$),

$$dF_0 = F_1\sigma_{10}dx - F_0\sigma_{01}dx. \quad (2.2)$$
In Eqn 2.2, \( dF_0 \) represents the number of neutral atoms after the charge exchange collision. Applying the condition \( F_0 + F_1 = 1 \), which is the summation of all measured charge states, Eqn 2.2 then becomes

\[
dF_0 = (1 - F_0)\sigma_{10}dx - F_0\sigma_{01}dx,
\]

which can be simplified to

\[
\frac{dF_0}{\sigma_{10} - F_0(\sigma_{10} + \sigma_{01})} = dx.
\]

Then integrate

\[
\int_{F_0 \to 0}^{F_0} \frac{dF_0}{\sigma_{10} - F_0(\sigma_{01} + \sigma_{10})} = \int_0^x dx,
\]

where \( F_0 \to 0 \) is the charge fraction that remains neutral after entering the target and \( F_0 \) is the neutral component of the beam upon exiting the target. The right hand side describes the thickness of the target.

After integrating, simplifying the result will yield

\[
F_0 = e^{-(\sigma_{10} + \sigma_{01})x} \left( F_0 \to 0 - \frac{\sigma_{10}}{\sigma_{10} + \sigma_{01}} \right) + \frac{\sigma_{10}}{\sigma_{10} + \sigma_{01}}
\]

and applying the condition \( F_0 + F_1 = 1 \) gives the general solution

\[
F_1 = e^{-(\sigma_{10} + \sigma_{01})x} \left( \frac{\sigma_{10}}{\sigma_{10} + \sigma_{01}} - F_0 \to 0 \right) + \frac{\sigma_{01}}{\sigma_{10} + \sigma_{01}}.
\]

Likewise, to find the general solution for the other charge state fraction one starts with this initial expression.
\[ dF_1 = F_0 \sigma_{01} dx - F_1 \sigma_{10} dx, \quad (2.8) \]

which describes the difference between the gain of the 1+ charge state from the neutral charge fraction and the loss of the 1+ from fully stripped charge fraction. Repeating the mathematical steps from the previous solution will yield

\[ F_0 = e^{-x(\sigma_{01} + \sigma_{10})} \left( \frac{\sigma_{01}}{\sigma_{01} + \sigma_{10}} - F_{1 \rightarrow 0} \right) + \frac{\sigma_{10}}{\sigma_{01} + \sigma_{10}}, \quad (2.9) \]

where \( F_{1 \rightarrow 0} \) is the charge fractions that change from 1+ to 0. An equilibrium condition can be applied to the general solution of \( F_0 \) and \( F_1 \). This equilibration condition is achieved when the charge exchanging medium has a thickness \( x \) great enough that \( x(\sigma_{01} + \sigma_{10}) \gg 1 \) to drop the exponential terms from Eqns 2.7 and 2.9 and the charge fractions remain constant. The consequences of the equilibrium condition yields

\[ F_1 \rightarrow F_{1\infty} = \frac{\sigma_{01}}{\sigma_{01} + \sigma_{10}} \quad (2.10) \]

and

\[ F_0 \rightarrow F_{0\infty} = \frac{\sigma_{10}}{\sigma_{01} + \sigma_{10}}, \quad (2.11) \]

where \( F_{1\infty} \) and \( F_{0\infty} \) are the equilibrated charge fractions for 1+ and 0, respectively. These two terms together gives

\[ \frac{F_{0\infty}}{F_{1\infty}} = \frac{\sigma_{10}}{\sigma_{01}}, \quad (2.12) \]

showing that the equilibrated charge state fraction is determined solely by the
cross-sections involved.

2.5 Heavy Ion Projectiles

Beyond the two-component system most cases will deal with much heavier ion projectiles \((A > 4)\) where the mathematics becomes complex enough to make an analytical solution difficult.

The general differential equation for an \(n\)-component system is

\[
\frac{dF_i}{dx} = \sum_{j=1, j \neq i}^{n} (\sigma_{ji} F_j - \sigma_{ij} F_i),
\] (2.13)

where \(\sum_{i=1}^{n} F_i = 1\) is the summation of all charge state fractions and at equilibrium \((x \to \infty)\) Eqn 2.13 becomes

\[
\sum_{j=1, j \neq i}^{n} (\sigma_{ji} F_j - \sigma_{ij} F_i) = 0.
\] (2.14)

Now replace \(i \to q\), where \(q\) is the charge state before collision and \(j \to q'\), where \(q'\) is the charge state after collision to get

\[
\sum_{q'} \sigma_{q' \to q} F_{q'} = \sum_{q} \sigma_{q \to q'} F_q.
\] (2.15)

Another way to interpret the expression above is that equilibrium conditions are attained when electron loss and capture into any charge state component are balanced. If it is assumed that single electrons are captured or lost, \(|q' - q| = 1\), then Eqn 2.15 can be applied. All the corresponding charge fractions can then be directly calculated when a complete set of cross-sections \(\sigma(q \to q')\) are known. Note that Eqn 2.15 reduces to Eqn 2.12 from the two-component system.
There are some limitations to the application of these formulae: (1) all ions must be in the ground state prior to a charge-changing collision otherwise the cross-sections are also a function of residual ion excitation and dependent upon target thickness and (2) experimental equilibrium distribution are well defined only as long as the target thickness for which the average energy loss of the ion can be neglected [12].

Thus far all descriptions, quantitative and qualitative, have focused on the electrons of the projectile ion interacting with a medium. Nothing has been stated on the nature of the medium. The next section will describe the influence of the target density of the medium, a phenomenon frequently referred to as the density effect.

2.6 Density Effect

Early charge state experiments with fission fragments[34, 35] show a large difference between mean charge states when going through a gaseous target as compared to a solid target. Figure 2.5 displays an example of this phenomenon with bromine ions through oxygen gas and carbon foils. The results clearly show that the charge state distribution is shifted to larger charge states having gone through the carbon target[10, 12, 15]. The higher mean charge state attributed to a solid target was first explained by Bohr and Lindhard (1954). They explained this effect to the influence of excited projectile states that are formed in the ion-atom collisions. Due to the density of the solid, the excited states do not decay fast enough to the ground state before the subsequent collision. Thus, the electron loss cross-section becomes enhanced. Another explanation of this density effect is given by Betz and Grodzins (1970), who state that equilibrium is achieved
Figure 2.5. This plot displays the charge fraction as a function of charge state, where an example of the density effect is observed with bromine ions going through oxygen gas and a carbon foil[11].

post-foil due to Auger emissions[13]. Figure 2.6 is a pictorial representation of the competing models. The Betz-Grodzins model was based on stopping power measurements that indicated the effective charges inside solid and gaseous targets were nearly identical. In general, the Bohr-Lindhard model is the more accepted of the two since later experiments cast doubt about the influence that post-foil Auger emissions contribute to the mean charge ($\bar{q}$)[65].
Figure 2.6. An illustration of the BL (Bohr-Lindhard) and BG (Betz-Grodzins) models as ions penetrate a gaseous and solid target[12].

2.7 Equilibrium Charge State Distributions

Figure 2.7 schematically displays how the charge of fast ions moving through matter fluctuates as a result of electron loss and capture in collisions with the target atom. After an adequate number of collisions, an equilibrium distribution of charge states is established that depends only on the ion species, the velocity of the ion and the nature of the target. Figure 2.8 shows the charge state fractions of carbon ions as a function of target thickness. Notice the behavior at (a) and (b) where the distribution and the mean charge ($\bar{q}$) are different. Starting at (c) the charge state distribution and mean charge ($\bar{q}$) does not change anymore since the target has reached a critical thickness and the charge states have attained equilibrium[26]. Equilibrium charge state distributions are independent of the charge state incident on the target. At equilibrium, the incident charge state does not affect the charge state fractions that are being measured. If the target thickness, $x$, is further increased the measured charge fraction will remain the same provided the total energy loss is negligible.
Figure 2.7. A simple schematic showing a negative ion from an ion source going through a carbon stripper and approaching charge state equilibrium. The adjacent diagram shows the relation between exit charge states and a charge state distribution, $\bar{q}$ [26].

There are three useful parameters for equilibrium charge state distributions: the mean charge state, the distribution width, and the skewness of the distribution. The first parameter is the mean charge state ($\bar{q}$), which is derived from the measured charge state fractions

$$\bar{q} = \sum_q q F_q,$$  \hspace{1cm} (2.16)

where $q$ is the charge state and $F_q$ is the charge state fraction. The distribution width, $d$, can be calculated from

$$d = \left[ \sum_q (q - \bar{q})^2 F_q \right]^{1/2},$$  \hspace{1cm} (2.17)

where $\bar{q}$ is the mean charge state, $q$ is the charge state and $F_q$ is the charge state fraction.
Figure 2.8. Charge fractions as a function of target thickness for carbon through nitrogen gas. The plot shows the fractions of the measured charge states as a function of target thickness. The charge state behavior at various target thicknesses is shown at (a)-(c). At (c) the target has attained a critical thickness and equilibrium is attained[26]. Note that the green bar represents the mean charge of a given charge state distribution.
state fraction. The last parameter describes the skewness (symmetry), $s$, of the distribution

$$s = \sum_q (q - \bar{q})^3 (F_q / d^3),$$

where $\bar{q}$ is the mean charge state, $q$ is the charge state, $d$ is the distribution width and $F_q$ is the charge state fraction.

The charge state distribution for many cases looks like a Gaussian distribution. The charge state fractions can then be written in the Gaussian functional form as

$$F_q = (1/d\sqrt{2\pi}) exp[-(q - \bar{q})^2 / 2d^2],$$

where $q$ is the charge state, $\bar{q}$ is the mean charge state and $d$ is the distribution width. The skewness for a Gaussian distribution is ideally equal to zero and the full width at half maximum can be defined as

$$FWHM = 2d(2ln2)^{1/2} \approx 2.35d.$$
mean charge state can be calculated by building semi-empirical models, which are explained below.

2.8 Semi-empirical Relationships for Equilibrium Mean Charge States

Charge state distributions of ions penetrating through matter can be calculated from electron capture and electron loss cross-sections. Such cross-sections are difficult to obtain so semi-empirical techniques have been utilized as a substitute with varying degrees of success. It should be noted that these semi-empirical expressions present little to no physics. They are equations that are parameterized to a given data pool. The last stringent effort based on physics came from Bohr in 1948[19]. Below are the most frequently cited semi-empirical models, mostly in chronological order, where the model is briefly introduced and limitations presented.
2.8.1 Thomas-Fermi Model

This model treats electrons in an atom as a statistical assembly of electrons obeying Fermi-Dirac statistics, where the exclusion principle has been taken into account[81]. The Thomas-Fermi model of the atom ignores shell structure but provides useful estimates for the dependence of an atom as a function of Z, the atomic number of the ion[1]. Thomas-Fermi velocity can be defined as \( v_{TF} = v_B Z^{2/3} \), where \( v_B = 2.188 \times 10^6 \) m/sec is the Bohr velocity\(^1\) and the mean charge state \( \bar{q} \) can be written as

\[
\bar{q} = Z[1 - \exp(-v/v_{TF})], \tag{2.21}
\]

where \( Z \) is the atomic number of the ion, \( v \) is the ion velocity and \( v_{TF} \) is the Thomas-Fermi velocity. The Thomas-Fermi velocity is the limit where the projectile is expected to be stripped of the majority of their electrons at energies exceeding that limit. This formalism has become the basis for most, if not all, semi-empirical expressions.

2.8.2 Bohr Model

Bohr described a criterion for the mean charge state of a fast heavy ion passing through a gas. The model calls for an electron with orbital velocities smaller than the velocity of the ion to be lost in collisions, while those electrons with larger velocities are able to adjust adiabatically and are therefore retained by the ion. In electron capture, the electron must change its momentum when picked up by the projectile. Electron capture is less likely as the projectile velocity increases

\(^1\)The Bohr velocity is derived from angular momentum and Coulomb force considerations to get \( v_B = \frac{e^2}{4\pi\epsilon_0 h} = 2.188 \times 10^6 \) m/sec, where \( e \) is the charge of the electron, \( \epsilon_0 \) is the permittivity of free space and \( h \) is the reduced Plank constant.
Bohr derived a formula for the mean charge in terms of $Z$ of the ion:

$$\bar{q} = Z\left[\frac{v}{v_B Z^{2/3}}\right], \quad (2.22)$$

where $v$ is the ion velocity, $v_B$ is the Bohr velocity. Its limitation, $v_B < v < v_B Z^{2/3}$, is based on ion velocity where the model fails with slow ions, below the Bohr velocity, and very fast ions above the Thomas-Fermi velocity[19].

2.8.3 Betz Model

H.D. Betz derived an expression to estimate the equilibrium mean charge state from experimental data using stripped sulfur, arsenic, iodine and uranium ions in air and in formvar foils at an energy range from 5 to 80 MeV to derive

$$\bar{q} = Z\left[1 - C\exp\left(-\frac{v}{v_B Z^\gamma}\right)\right]. \quad (2.23)$$

The terms within the expression include ($Z$) the atomic number of the ion, ($v$) the ion velocity, ($v_B$) the Bohr velocity and fitting parameters $C$ and $\gamma$ are determined empirically for each ion and target pair. Note that previously $\gamma$ was define by Thomas-Fermi as $2/3$. Liu et al. noted that Betz also made $v_B$ a free parameter (later renamed $v'$) to achieve a better fit, which became the new standard velocity term[36]. They developed an alternate form of the Betz equation to fit hydrogen and helium gas targets that can be found in the previous citation.
2.8.4 Nikolaev-Dmitriev Model

Nikolaev and Dmitriev in their paper “On the Equilibrium Charge Distribution in Heavy Element Ion Beams”[47] built what they described as a unified approximate semi-empirical formula for the mean charge of ions ($Z \geq 20$) through solids. Their formula for the mean charge state has the functional form

$$\bar{q} = Z[1 + (v/Z^\alpha v')^{-1/k}]^{-k}, \quad (2.24)$$

where $Z$ is the atomic number of the ion, $v$ is the ion velocity, $\alpha=0.45$, $k=0.6$ and $v'=3.6 \times 10^8$ cm sec$^{-1}$, the free parameter from the previous section. They determined that the relative ionization $\bar{q}/Z$ should be proportional to $Z^\alpha$ and ions with $Z \geq 20$ passing through solids that $\alpha$ is nearly independent of the relative ionization. Nikolaev and Dmitriev treated $v/Z^\alpha$ as a universal function for all $Z$, where the relative ionization is of the form presented in Eqn 2.24.

The Nikolaev-Dmitriev expression can be re-written to look like

$$\bar{q} = Z[(1 + X^{-5/3})^{-3/5}], \quad (2.25)$$

where $X = 3.86Z^{-0.45}\sqrt{E(\text{MeV})/m(\text{u})}$ is the reduced velocity. The benefit of writing the equation in terms of $X$ is that it will have the same dependence and similar structure to later empirical models. The limitations of this model are based on the data pool used by Nikolaev-Dmitriev and include $0.3 < \frac{\bar{q}}{Z} < 0.7$ and $E > 0.5\text{MeV/amu}$ [20, 30, 47]. This model is used in the Nuclear Science Laboratory program called ZDIST.
2.8.5 To-Drouin Model

The To-Drouin model was specifically developed for ions from boron to argon through carbon targets, which partially encompasses the present work. Their model gives the mean charge as

\[ \bar{q} = Z[1 - \exp(-X)], \quad (2.26) \]

where \( X = 3.86Z^{-0.45} \sqrt{E(\text{MeV})/m(u)} \) is the reduced velocity with the limitations \( 0.2 \leq X \leq 1.6 \) and \( 5 \leq Z \leq 18 \) [75, 76]. The limitations are based on the available data pool used by To-Drouin.

2.8.6 Shima-Ishihara-Mikumo Model

Previous empirical models from Bet [10] and Nikolaev and Dmitriev[47] did not fit new charge state data taken at higher energies, so the Shima-Ishihara-Mikumo[57] model was built.

The first thing that Shima-Ishihara-Mikumo noted was that all of the carbon target data seemed to follow the same track. They used that trend to build the following formula

\[ \bar{q}(Z_2 = 6) = Z[1 - \exp(-1.25X + 0.32X^2 - 0.11X^3)], \quad (2.27) \]

where \( X = 3.86Z^{-0.45} \sqrt{E(\text{MeV})/m(u)} \) is the reduced velocity and \( Z_2 \) is the atomic number of the target[57]. Next using the non-carbon target data, Shima-Ishihara-Mikumo approximated a linear combination of \( (Z_2 - 6)\sqrt{X} \) with a correction term of a \((1 + g(Z_2))\), where \( g(Z_2) \) is a function of target dependence, to be multiplied to the original expression to get
\[ \bar{q} = \bar{q}(Z_2 = 6) \cdot [1 + g(Z_2)], \quad (2.28) \]

where

\[ [1 + g(Z_2)] = [1 - 0.0019(Z_2 - 6)\sqrt{X} + 10^{-5}(Z_2 - 6)^2 X]. \quad (2.29) \]

The authors cite the limitations of their target dependent model are \( Z \geq 8, \)
\( 4 \leq Z_2 \leq 79 \) and \( E < 6\,MeV/u. \) The limitations are based on the available data pool used by Shima-Ishihara-Mikumo.

2.8.7 Itoh Model

Itoh et al. measured the charge state distributions of lithium ions passing through carbon targets with an energy range 0.8 to 6 MeV. They noted a lack of information for this energy range and in general for low-Z ions [30]. Their results match with the next data set collected from Stocker and Berkowitz [69] at higher energies. Itoh built an expression similar to that by Shima-Ishihara-Mikumo specifically for lithium ions through carbon targets,

\[ \bar{q} = Z[1 - \exp(0.706 - 1.98X + 0.0883X^2)], \quad (2.30) \]

where \( X = 3.86Z^{-0.45}\sqrt{E(MeV)/m(u)} \) is the reduced velocity with the limitation of \( 0.8 < X < 5. \) The limitations are based on the available data pool used by Itoh.
2.8.8 Ziegler-Biersack-Littmark Model

The Ziegler, Biersack and Littmark model is used in the well known SRIM (Stopping and Range of Ions in Matter) code. The expression for helium ions is given as

\[ \gamma^2 = 1 - \exp \left[ - \sum_{i=0}^{5} a_i \ln(E)^i \right], \quad (2.31) \]

where \( \gamma \) is defined as the fractional effective charge of the ion, \( a_i \) are fitting constants that were determined to be \( a_0 = 0.2865 \), \( a_1 = 0.1266 \), \( a_2 = -0.001429 \), \( a_3 = 0.02402 \), \( a_4 = -0.01135 \) and \( a_5 = 0.00175 \) and \( E \) is in units of keV/amu. This equation can be written into a form similar in notation to the other semi-empirical expressions used in this work:

\[ \tilde{q} \approx Z \left[ 1 - \exp \left[ - \sum_{i=0}^{5} a_i \ln(E)^i \right] \right]^{1/2}, \quad (2.32) \]

where \( Z \) is the atomic number of the ion. The fractional effective charge \( \gamma \) is approximately equal to the relative ionization \( (\tilde{q}/Z) \) based on the definition of effective charge, \( q_{eff} = Z \gamma \). Some assumptions that are made for the derivation of Eqn 2.32 include that the effective ion charge is independent of target material and that the effective ion charge of H is always unity.

For ions of \( Z \) greater than 2 the Ziegler, Biersack and Littmark formula can be written as:

\[ \tilde{q} = Z \left[ 1 - \exp \left[ 0.803y_r^{0.3} - 1.3167y_r^{0.6} - 0.381557y_r - 0.008983y_r^3 \right] \right], \quad (2.33) \]

where \( y_r \) is the reduced velocity as given by \( y_r = \frac{v_r}{v_e Z^{2/3}} \) and \( v_r \) is the relative
velocity as given by

$$v_r = v \left( 1 + \frac{v_F^2}{5v^2} \right)$$

(2.34)

for \( v > v_F \) and

$$v_r = \frac{3v_F}{4} \left( 1 + \frac{2v^2}{3v_F^2} - \frac{v^4}{15v_F^4} \right)$$

(2.35)

for \( v \leq v_F \) [82] where \( v \) is ion velocity and \( v_F \) is the Fermi velocity of the medium.

The Fermi velocity is the electron velocity of the highest occupied energy level for conduction electrons in the solid.

2.8.9 Schiwietz Model

A relatively new formula created by Schiwietz et al. [53] is a highly parameterized least-square fit built from an array of over 800 data points that span a wide variety of ions and targets, see Fig 2.10. The expression for the relative ionization is given as

$$\bar{q} = Z \left[ \frac{8.29x + x^4}{0.06/x + 4 + 7.4x + x^4} \right],$$

(2.36)

where \( Z \) is the atomic number of the ion

$$x = c_1(\bar{v}/c_2/1.54)^{1+1.83/Z},$$

(2.37)

is a reformulated reduced velocity and the power term is used to adjust the steepness of the charge state curves as a function of \( x \) with the following correction terms:
Figure 2.10. Displays the relative ionization ($\bar{q}/Z$) as a function of the velocity scaling variable $x$. The solid curve is the fit for the Schiwietz model and the elements noted on the plot are some of the ions used[53].

\[c_1 = 1 - 0.26 e^{\left[-\frac{Z_2}{11}\right]} e^{\left[-\frac{(Z_2 - Z)^2}{9}\right]}\]  \hspace{1cm} (2.38)

and

\[c_2 = 1 + 0.030 ln(Z_2),\]  \hspace{1cm} (2.39)

where $Z_2$ is the atomic number of the target. The first term accounts for resonant electron capture, which reduces the mean charge state or similarly $x$ for
symmetrical ion-target combinations, while the second correction term allows for a target dependent deformation at high velocities. The final component in the reformulated reduced velocity is the scaled projectile velocity

\[ \tilde{v} = Z^{-0.543} v / v_B. \]  

(2.40)

The limitations noted for this model are that the ratio of the projectile velocity to the Bohr velocity for protons and helium must be greater than 2 and for all other ions the ratio must be greater than 0.4.

Of the models presented, there are three models (To-Drouin, Ziegler-Biersack-Littmark, Schiwietz) whose limitations fit the energy range for low-Z ions in the present work. The To-Drouin model works for boron and carbon, but has not been tested for low-Z ions less than boron. The Ziegler-Biersack-Littmark and Schiwietz models are unique in that they provide a target dependence. The Schiwietz model has only been compared with high energy and high-Z charge state data, so testing that model for low energy, low-Z ions will be a first.

2.9 Conclusion

This chapter has reviewed the fundamental empirical frameworks for charge exchange necessary to apply to the study of low energy, low-Z ions. The following chapter describes the experimental facilities required to gather the needed data for low-Z ion information i.e. the ion source, accelerator and preparation of charge exchanging solid targets.
CHAPTER 3

EXPERIMENTAL FACILITIES

3.1 Introduction

In this chapter, the experimental facilities of the Nuclear Science Laboratory at the University of Notre Dame that are necessary to measure and study the behavior of charge state distributions for low-Z ions (Li, B and C) will be described. For the present work the ion source, the FN tandem accelerator, the Browne-Buechner Spectrograph and associated detectors that are needed for charge exchange experiments will be explained in more detail in the following sections. Figure 3.1 displays the current layout of the Nuclear Science Laboratory. All experiments using the FN tandem start at the ion source area and the post accelerated ions are then sent to various experimental areas. There are two smaller accelerators (JN and KN) noted in the figure, which run at lower voltages.

3.1.1 Ion Source - SNICS

The operation of the SNICS (Source of Negative Ions by Cesium Sputtering) sputter ion source starts with a reservoir of cesium, which is heated so that a cesium vapor is formed, see Fig 3.2. This vapor rises from the reservoir to an enclosed region between the cooled cathode and the heated ionizer. Some of the cesium condenses onto the cool surface of the cathode, while some ionizes
with contact on the hot surface of the ionizer. The positively charged cesium ions leaving the ionizer are accelerated toward and focused onto the cathode, sputtering material from the cathode at impact. Some of this sputtered material gains an electron in passing through the cesium coated layer and forms negatively charged ions. Since the entire source is operated at approximately 80 kV below electrical ground, this negative beam is accelerated out of the source and is available for injection into the FN tandem accelerator.

A wide variety of ion beams can be produced with this source, depending upon the nature of the cathode material used. The “Negative Ion Cookbook” by Roy Middleton is a good source of information on cathode material and beam
Figure 3.2. A basic diagram of ion production inside a SNICS ion source [3].

production [41]. The cathode is a cylindrical piece of aluminum or copper, which has a small cavity drilled into the cylinder’s axis. Typically, a powder containing the isotope of interest is pounded or pressed into the cavity. Cathode material lifetime varies with isotope, some lasting for a week or more, while others for only hours.

Most accelerator laboratories typically have a single-cathode ion source, but in 2008 the Nuclear Science Laboratory received funding for a Multi-Cathode SNICS as seen in Fig 3.3. An advantage of a Multi-Cathode ion source is the relatively short time it takes to switch between cathodes, otherwise the process can take several hours. The Multi-Cathode SNICS has proven invaluable for Accelerator Mass Spectrometry experiments as AMS usually relies on frequent cathode changes. Frequent cathode changes on a single-cathode SNICS could
potentially introduce instabilities and changes in ion transport transmission, which is a serious problem for sensitive AMS experiments.

The operation of the Multi-Cathode SNICS, in principle, functions the same as the single-cathode SNICS. The obvious difference is the ability to hold 20 cathodes that can be changed by a piston that rotates the cathode wheel.

3.1.2 FN Tandem Accelerator

The model FN tandem Van de Graaff accelerator, see Fig 3.4, has been in service since 1968[26]. A centralized metal electrode, referred to as the terminal shell, is charged to a positive potential. The negatively charged ion beam produced by the SNICS ion source is transported, in vacuum, toward the terminal. The first
stage of acceleration corresponds to the negative ion moving toward the positively charged terminal shell. As the beam enters the area within the terminal shell, it passes through a thin charge exchanging carbon foil ($\sim 3 \, \mu g/cm^2$) that strips electrons from the ions, leaving the ions positively charged. The second stage of acceleration is where the positively charged ion beam accelerates away from the positive potential at the terminal. The development of a heavy ion accelerator like the FN tandem is a perfect example of charge exchange in action. The development of tandem accelerators has proven invaluable to the physics community.

After exiting the accelerator the ions are selectively bent around a dipole “analyzing” magnet where the final energy of the beam can be written as:

$$E(MeV) = V_T(q + 1),$$  \hspace{1cm} (3.1)
where $V_T$ is the terminal voltage in MV and $q$ is the positive charge state of the beam exiting the stripping region.

### 3.2 Beam Transport and Scattering Chamber

The accelerated beam, as mentioned in the previous section, is sent directly into a 90° dipole “analyzing” magnet where the beam direction is changed and the components are analyzed. The path of the ion beam in a magnetic field is based on the radius of curvature and its momentum,

$$\vec{B} \rho = \frac{\vec{P}}{Q},$$

(3.2)

where $\vec{B}$ is the magnetic field in Tesla, $\rho$ is the radius of curvature of the magnet in meters, $\vec{P} = \sqrt{2mE}$ is the momentum defined by mass in terms of atomic mass, energy in MeV and $Q = qe$, where $q$ is the charge state of the ion and $e$ is the charge of the electron.

Beyond the accelerator all other beamline components are used for the purpose of beam transport and tuning. These elements are used for focusing, steering, and monitoring beam. Figure 3.5 shows the experimental layout for all charge exchange experiments. The start of the experiment is at the Multi-Cathode SNICS ion source. Then the path of the ion beam continues through the accelerator and is bent around a dipole magnet to get directed into the AMS beamline. Finally, the ion beam will arrive at the scattering chamber, which houses all of the charge exchanging targets. The ions charge state fractions are separated in the spectrograph to be measured by a Faraday cup.
Figure 3.5. This schematic displays the experimental layout at the Nuclear Science Laboratory that is used for a charge exchange experiment, from the ion source to the AMS beamline with the spectrograph and other important elements labeled.
3.2.1 Scattering Chamber

The scattering chamber sits directly in front of the spectrograph, see Fig 3.6. The scattering chamber was part of the original spectrograph infrastructure and only slightly modified by the AMS group. The interior of the scattering chamber has a target ladder that can hold six targets or five targets and a small Faraday cup. There are two plates that can rotate about the target ladder and are used to hold silicon detectors or a camera, see Fig 3.7.

The exterior of the scattering chamber has several ports that are used for pressure gauges, slow pump valve, vent valve, a plastic viewer to look inside, and BNC (Bayonet Neill-Concelman) electrical connections.
3.3 Browne-Buechner Spectrograph

The Browne-Buechner spectrograph, see Fig 3.8, was first installed in the early 1970s for highly accurate energy measurements from nuclear reactions [49]. Its design is built around a $90^\circ$ single dipole magnet with a radius of curvature of 100cm at $90^\circ$. The magnet gap is in the vertical plane with a detection system originally designed to use photographic plates at the focal surface. The complete system can rotate about the stationary scattering chamber with a radius of about 2.5m up to an angle of $150^\circ$. Table 3.1 summarizes the design specifications of the Browne-Buechner Spectrograph. Despite good resolution and high accuracy, the spectrograph was abandoned more than ten years ago, as work in the laboratory moved into different fields and techniques.

When renovation began in 2003 with the formation of the Accelerator Mass
Figure 3.8. The Notre Dame Browne-Buechner spectrograph.

Spectrometry group at the Nuclear Science Laboratory, neglect and cannibalism had left the support systems in a very poor condition. The renovation required an extensive overhaul of the electrical, cooling and vacuum systems. The electrical systems had excess electrical cables that were not connected and some exposed live wires. The entire system needed to be mapped and stripped bare to make room for other upgrades. The spectrograph magnet cooling systems were corroded, leaking and needed to be flushed with a calcium-lime remover. The vacuum systems were old diffusion and belt-driven pumps that barely functioned and were replaced by turbo-molecular and rotary-type pumps. The spectrograph over time has been optimized for AMS experiments such as the removal of the focal plane.
TABLE 3.1

DESIGN SPECIFICATIONS FOR THE BROWNE-BUECHNER
SPECTROGRAPH

<table>
<thead>
<tr>
<th>Specification</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Orbit radius</td>
<td>100 cm</td>
</tr>
<tr>
<td>Deflection</td>
<td>90°</td>
</tr>
<tr>
<td>Max field strength</td>
<td>1.2 Tesla</td>
</tr>
<tr>
<td>Pole gap</td>
<td>50.8 mm</td>
</tr>
<tr>
<td>Total weight</td>
<td>26,580 kg</td>
</tr>
</tbody>
</table>

and photoplate detection system for the gas-filled AMS detectors and converting valve controls and pressure readouts to LabView.

3.4 Detectors

There are two detectors types that are needed for measuring charge state fractions. The first is the silicon “monitor” detector, which “monitors” the ion beam intensity before the charge state fractions are measured. The second detector is a Faraday cup, which measures the current of each charge state as they are magnetically separated in the spectrograph. These two detector types must work in tandem for the present charge exchange experiments to be successful. Below are some more details about each detector.
3.4.1 Silicon “Monitor” Detector

The sole purpose of the monitor detector is to measure the ions scattered from a target foil prior to entering the spectrograph. This detector will “monitor” changes in beam intensity, thus the scattered beam is used as a normalization tool. This capability becomes critical when the experimental setup does not permit the entire charge state distribution to be measured at once. Rather each charge state has to be measured individually and during that period of time the beam intensity will not remain constant.

The general physics and characteristics of semiconductor detectors is given in Appendix A. There are many varieties of silicon detectors and no specific type is preferred for the role of beam monitor, though for charge exchange studies a monitor detector does not need to have high resolution. The standard precautions are true for nearly all types, which include sensitivity to light when biased. Another precaution is to limit the count rate to around 500 cps, since radiation damage will affect the lifetime of the detector.

3.4.2 Faraday Cup

The most basic type of detector for collecting direct current of charged particles is a Faraday cup. The measurement of particle current is the most important diagnostic tool necessary for optimizing ion source output, transmission and beam transport. A well-designed Faraday cup does not allow secondary electrons from the body of the cup to escape, otherwise the loss of those electrons appear to an ammeter as an extra ion. To prevent the escape of secondary electrons, the general rule is that the length of the cup should be at least five times the size of the diameter [43]. The bottom of the cup is built at an angle, see Fig 3.9. This
angle will minimize the probability of electrons being ejected (backscattered) out of the deep well of the cup. A final piece added to the cup is the suppression piece where a $-300\, \text{V}$ potential will provide a final obstacle to secondary electron emission. The potential is created between the suppression piece and the body of the Faraday cup. The following reference by Turner et al. [77] illustrates the importance of suppressing secondary electrons.

3.5 Preparation of Thin Nuclear Targets

3.5.1 Introduction

Thin self-supporting targets are needed for many applications in low-energy nuclear physics experiments. There are several sources that review a number of techniques used in the preparation of nuclear targets, which can be a useful guide[28, 38, 39, 44]. Vacuum evaporation is the most commonly used technique.
In the next sections, the high-vacuum evaporation setup is described and a method for \textit{in-situ} film thickness monitoring with a quartz crystal is explained. The stages in the fabrication and mounting of self-supporting foils are described with an emphasis given to the preparation of substrates and release agents related to different material.

3.5.2 High Vacuum Deposition

3.5.2.1 Resistive Heating

The simplest type of vacuum deposition and the only one available in the Nuclear Science Laboratory is electrical resistance heating. This technique is where the passage of electric current through a conductor (copper clamps) into refractory metal (W, Ta, Mo) “boats” releases heat to melt and evaporate the target material. The resistive heating process just described can be seen in Fig 3.10.

3.5.2.2 Evaporator Setup and Cleaning

The evaporator at the Nuclear Science Laboratory, see Fig 3.11, has a basic setup with a large bell jar that can be pumped out and isolated from the vacuum system by a butterfly valve. The system can reach pressures of $10^{-6}$ torr with a diffusion and mechanical pump. Inside the bell jar are water-cooled copper clamps that supply electric current to the evaporation source, \textit{i.e.} a filament or boat. The copper clamps also hold the boat with evaporant in place. A substrate holder with three pyrex (microscope) slides rest above the clamped boat. Once evaporation is complete, the bell jar is isolated via the butterfly valve and slowly vented to atmosphere. Then the material coated substrates are carefully removed and stored for the next stage of target fabrication.
Cleaning the evaporator should be done before using a different evaporant material. The support rods and all other pieces are cleaned thoroughly in order to prevent contamination for subsequent evaporations. The method that seems to be most effective is to wash everything in a 25−50°C HNO₃ solution (or any other weak acid)[39]. Then rinse thoroughly in deionized water and dry completely with acetone. Sometimes a thorough scrub down with a powdered detergent is sufficient and used for cleaning the bell jar’s inner surface.

3.5.3 Quartz Crystal Monitor

A quartz crystal monitor can measure the thickness of a thin film by taking advantage of the piezoelectric effect in quartz crystals. When an electric potential is applied across the crystal, it induces a mechanical shear strain. The strain reverses when the polarity of the electric potential is reversed. Thus rapid oscil-
Figure 3.11. Diagram of the typical evaporator setup[44].
lations in the polarity leads to vibrational motion of the quartz crystal [7]. With
the proper conditions a standing wave between the crystal faces is induced and
a resonant frequency is achieved. This resonant frequency is proportional to the
thickness of the quartz crystal and the change in this frequency by adding addi-
tional material is then the thickness of the additional material. The thickness of
the thin film can be calculated using

\[ T = \frac{P_{\text{quartz}} N_{\text{quartz}} (f_{\text{quartz}} - f)}{f_{\text{quartz}}^2} = \frac{P_{\text{quartz}} N_{\text{quartz}} (\Delta f)}{f_{\text{quartz}}^2}, \]  

(3.3)

where \( P_{\text{quartz}} \) is the density of quartz (2.648g/cm\(^3\)), \( N_{\text{quartz}} \) is the frequency con-
stant and for AT-Cut crystals is 1.668 \times 10^5 Hz cm, \( f_{\text{quartz}} \) is the resonant frequency
of the quartz crystal and \( \Delta f \) is the frequency shift when material is deposited on
the crystal. The thickness of the deposited material \( T \) is in terms of an areal
density (mass/area). Equation 3.3 is valid only when the resonant frequency is
much larger than the frequency shift (\( f_{\text{quartz}} \gg \Delta f \)), which requires a thin film[9].

Multiplying out the constants related to quartz and using a crystal with a
resonant frequency of 6MHz, Eqn 3.3 can be written in a more practical form

\[ T(\mu g cm^{-2}) = \frac{\Delta f}{81.5}, \]  

(3.4)

where \( \Delta f \) is the difference in the initial and final frequency. Equation 3.4 is only
valid for a 6MHz crystal, otherwise it has to be recalculated.

The damping of the crystal varies with changes in its surrounding environ-
mental setup; this damping is indicated by a small frequency shift during the
evacuation of the evaporation unit. Mechanical vibrations may cause frequency
shifts if the crystal is not securely mounted. The most serious factor influencing
frequency shift, and the most difficult to eliminate is variation in crystal temper-
ature. Crystals are selected with a low frequency-temperature coefficient and to attach in a water-cooled mount, see Fig 3.12[45].

Quartz crystal monitors are possibly the most commonly used thin-film monitoring devices. Quartz crystals can be used *in-situ* to determine the areal density of thin films. The thicknesses of the films need to be confirmed by an independent method post evaporation, which will be mentioned later.

3.6 Targetry

3.6.1 Target Backings

Frequently a target cannot be made self-supporting, especially thin targets (<100µg cm$^{-2}$), and require some additional support, either permanent or temporary. Some thin films are coated with Collodion, post evaporation, as a support that can be removed afterwards[71, 72]. With Collodion an intense heat lamp or methanol can be used to remove it.
3.6.2 Release Agents

Release agents are used to separate the thin film from the substrate. The choice of release agent should provide the least amount of mechanical stress to the thin film. There are two basic water soluble types of release agents: salts and detergents. Their solubility becomes important when making the thin film into targets, which will be explained in a later section.

3.6.2.1 Salts

Barium chloride, BaCl$_2$ is the most commonly used salt as a release agent, though any salt can be used since it is water soluble. The advantage with BaCl$_2$ is that it layers on the substrate very evenly. It is necessary to heat or bake the salt to remove moisture before loading it into a boat so that all the absorbed water vapor is driven out. The melting point of BaCl$_2$ is greater than 900$^\circ$C, which makes it ideal for targets that must be deposited on heated backings since it will not re-evaporate. Enough salt must be evaporated onto the substrate to give a coating of about 10 – 20 $\mu g cm^{-2}$. This amount is usually enough to allow the film to be easily stripped. The salt can be evaporated from molybdenum or tantalum boat, where this boat can be reused many times.

3.6.2.2 Detergents

The most commonly used detergent is Teepol 610, which can be purchased from Sigma Aldrich. This application involves putting a drop of the detergent on the center of the slide. The drop is then smeared over the entire substrate as evenly as possible. The substrate is then polished until no trace of the detergent is left. At this point there is a monatomic layer of the detergent remaining on the
substrate, which is more than sufficient to allow a film to release. Teepol has a low melting point (about 100 – 200°C) and any heat reaching the substrate will make it evaporate. For this reason, Teepol should only be used for low temperature evaporations or those in which the radiant heating from the source is low. Another way to minimize heating restrictions is to increase the distance between the radiating boat and the Teepol coated substrate.

3.7 Floating and Mounting of the Foils

The method for floating a completed film from the substrate starts with scraping a line all around the edges of the film removing any excess material that may interfere, see Fig 3.13 and 3.14. The film is cut into appropriately sized pieces for mounting by etching out the desired area with a razorblade, which will allow water to interact with the release agent. The substrate should then be clamped on a stand above a large beaker of distilled or deionized water making an angle with the surface of the water, approximately 45° for the best results. The beaker of water should be on top of some sort of laboratory jack to raise or lower evenly. The water is then slowly raised until the surface makes contact with the cut edge of the film or vice versa. As the film starts to lift from the substrate, the water is slowly raised to continue the process. When the water level reaches the end of one of the foils it will separate completely from the substrate and float on top of the water. If the film does not completely release from the substrate, carefully use a needle to gently free it.

The target holders used for mounting a foil are made of 0.03in thick aluminum and are 0.846in wide by 0.68in high. There is a 0.50in diameter hole in the center that is smooth and deburred on all sides. The target holder is immersed
in the water perpendicular to the surface and is positioned near the floating foil. When the foil is positioned properly, the target holder is then gently and evenly withdrawn from the water, making sure that it is approximately perpendicular to the water’s surface. In some cases the surface tension of the water can break the film during the lifting operation. Once the mounted foil has been removed from the water it must be stored away from breezes or changes in pressure and given time to dry. These foils are typically stored in plastic cases where the mounted foils are pressed into modeling clay. The case is carefully closed and then stored in a cabinet.

Much of what has been described can be found in John O. Stoner Jr.’s “Methods for Floating and mounting standard carbon foils” from www.ACFMetals.com website.

3.8 Target Materials

These target materials represent some of the more commonly used targets for charge exchange. Carbon is the most commonly used and provides a baseline to verify the reproducibility of previously published data. Aluminum has existing
Figure 3.14. a) Under the bell jar in a evaporator. b) Three thin films of aluminum prepared. c) A thin film etched in preparation for floating targets. d-e) Shows the release of a section of aluminum film and being caught by a target holder. f) The newly made target is removed and stored in a plastic container. The pictures were taken by M. Carilli making aluminum targets.
low-Z ion data, but at higher and lower energies than the present work thus allowing for the opportunity to include new data for a given ion-target system. Copper, silver, and gold all belong to the same family on the periodic table and all of the data collected will be new and perhaps will give some insight for charge exchange processes.

This discussion will cover the various target materials used for producing self-supporting foils for the present charge exchange experiments. Each of the materials mentioned have their own unique difficulties that must be overcome in order to make them useful as a thin target. All evaporations were done with a separation of 15cm between the boat and the substrate, which produces a more uniform film and minimizes heating effects. All target making attempts started with suggestions from a paper by Maxman[39].

3.8.1 Carbon

The carbon foils were the only material that was purchased as a pre-made film from the Arizona Carbon Foil company (ACF). The films were cut and scraped to match the shape of the target holders and then floated in the manner previously described.

3.8.2 Aluminum

Aluminum is evaporated in a tungsten boat, which has the advantage that aluminum wets on tungsten very easily, but has the disadvantage that it also easily alloys with that boat. Therefore the number of evaporations with a given boat is very limited. Another problem with these kinds of targets is that they oxidize, which makes them susceptible to hydrocarbons from the evaporator’s
pumping systems. A clear sign of this is a yellowing effect. Teepol is the release
gent that works best for aluminum.

3.8.3 Copper

Copper is by far the easiest material to evaporate and works best with a
tantalum boat, which can be used for about a dozen times. When the boat
is nearly depleted of copper there is a layer of the remaining impurities. The
substrate can be coated either with Teepol or BaCl₂. The copper removes easily,
but fractures and is incredibly fragile making it one of the more difficult targets.

3.8.4 Silver

A molybdenum boat works best when evaporating silver, where little material
is needed for multiple evaporations. The substrate can be coated with Teepol
or BaCl₂ can be evaporated and used as a release agent. The subsequent film
removes very easily. The foils can be mounted onto target frames with very little
effort. Out of all of the materials used, silver was the easiest to evaporate and
float.

3.8.5 Gold

With gold, either a tungsten or tantalum boat can be used. Since gold melts
easily, care should be taken that the temperature is not too high since gold tends
to spit at high evaporation rates. The substrate should be coated with Teepol and
the subsequent film is fairly easy to float. For very thin foils, \( \leq 60 \mu gcm^{-2} \), the
film “pops” when removed from the water due to surface tension. To overcome
this dilemma the gold covered slides are coated with a layer of collodion prior to
floating. The process of making collodion and using it as a strengthening agent is given in a paper by John O. Stoner Jr., which was closely followed [72]. The same laboratory jack used to lower thin films into water to float foils is used in the coating of the thin films. The laboratory jack provides a smooth and steady insertion of the thin film into the collodion mixture giving a uniform collodion coating.

3.9 Measuring Foil Thicknesses

As mentioned earlier, quartz crystal monitors can provide an approximate thickness because of the many assumptions and conditions of the evaporator environment. Other methods must be used to confirm or give an upper limit to the target thickness.

3.9.1 Weighing

The general idea is to measure the weight of the substrate plus release agent before evaporation has taken place and then weigh it again with the evaporated material. The difference in weight, assuming a uniform distribution of evaporant, divided by the area of the substrate will give the approximate thickness. The problem lies in the sensitivity of the balance used to measure the slight difference from before and after the evaporation process. The weighing method works best with foils that are in the $mg/cm^2$ range where the present work are near 100 $\mu g/cm^2$. The more effective method is the energy loss approach.
3.9.2 Energy Loss

The method that is used most often for determining the thickness of thin foils is the measurement of energy loss of charged particles as they traverse the target. The target to be measured is placed between an alpha source, like $^{241}$Am, and a silicon detector, where there will be a noticeable shift in energy: $\Delta E = E_\alpha - E$, where $E$ is the energy on exiting the foil. The energy of the alpha, $E_\alpha$ from $^{241}$Am is 5486 keV. Once the energy loss for the target has been measured the target thickness can be calculated by dividing the loss measured by the rate of loss per unit thickness (or stopping power). The stopping power $dE/dx$ can be found in tables or more conveniently can be displayed by the program called SRIM, the Stopping and Range In Matter program. The expression below summarizes what has been described above and works only for thin foils. Thin foils allow for a linear approximation since there is a minimum energy loss.

$$\left(\frac{dE}{dx}\right)_{SRIM} \approx \left(\frac{\Delta E}{\Delta x}\right) \rightarrow thickness \approx \Delta E / \left(\frac{dE}{dx}\right)_{SRIM} \quad (3.5)$$

The following chapter describes the experimental setup and procedures for performing a charge exchange experiment. The results of these experiments will be discussed and compared with the relevant empirical models and what various parameters like mean charge and the distribution width can reveal about the projectile shell structure.
CHAPTER 4

RESULTS AND DISCUSSIONS

4.1 Introduction

This chapter will highlight the experimental setup and procedures used for data collection for charge exchange experiments. The purpose for these charge exchange experiments will be to present new data in the energy regime (3 - 7MeV) not currently explored for low-Z ions and with target material other than carbon. Since there is a large carbon target data pool, the reliability of the experimental setup can be verified by measuring lithium, boron and carbon charge state fractions through carbon foils. Then using the carbon data pool for low-Z ions (He, Li, Be, B and C), a comparison between relevant empirical models are followed by exploring the behavior of global and local models. Targets other than carbon will provide a look at target dependence in this energy regime especially when compared as a function of mean charge. Finally, the study of low-Z, low energy ions through various materials will highlight projectile electron shell effects and how they affect the functional form of the charge state distribution as indicated by the distribution width.
TABLE 4.1

RESULTS: AMS DETECTOR EXPERIMENT

<table>
<thead>
<tr>
<th>Incident Ion</th>
<th>Incident Energy (MeV)</th>
<th>Exit Energy (MeV)</th>
<th>Foil Thickness (µg/cm²)</th>
<th>Charge State Fractions</th>
</tr>
</thead>
<tbody>
<tr>
<td>⁷Li²⁺</td>
<td>12</td>
<td>11.961 (Au) 85</td>
<td>2.94</td>
<td>94.2</td>
</tr>
<tr>
<td>⁷Li²⁺</td>
<td>15</td>
<td>14.965 (Au) 85</td>
<td>2.96</td>
<td>96.2</td>
</tr>
</tbody>
</table>

4.2 Experimental Tests

The first experiments to measure charge state fractions used the AMS gas-filled position sensitive detector coupled with a silicon detector to monitor beam fluctuations. The experiment elucidated technical difficulties with the AMS detector count rate acceptance. This detection method was abandoned because the low beam intensity needed for the AMS detector (less than femtoamps of ion beam current) made the scattered count rate too low to effectively monitor beam fluctuations, i.e. monitor count rate of 0.64 counts per second while the AMS detector had a count rate of 2500 counts per second. The results for lithium ions traversing a gold foil are summarized in Table 4.1. In addition, the extra charge exchanging material that the ions have to travel through, a mylar window and the isobutane fill-gas, would have skewed charge state results.

Based on the AMS detector experience, it was decided that the coupled detector scheme should use two similar types of detectors. The detector scheme would
use simple photocells to monitor ion beam fluctuations and collect charge state data. The development of the photocell detector for charge exchange experiments is given in Appendix A. Ultimately, that detector setup was abandoned because of inconsistent production and photocell quality by the manufacturer.

Eventually it was decided to use a standard detection method consisting of a Faraday cup coupled with a silicon detector as the beam monitor. The Faraday cup was not built originally to suppress electrons and so was tested with a lithium beam traversing carbon and aluminum targets. The original goal was that the geometry of the Faraday cup, the length of the cup and the angled back end, would suffice in preventing the escape of secondary electrons. Despite the successful tests, it was determined to rebuild the Faraday cup so that it has suppression capabilities to compensate for beam components that will strike the front edge or sides of the Faraday cup as a precaution.

The suppression voltage was experimentally determined, for an example see Fig 4.1. These experiments found that a $-300 \text{ V}$ is sufficient to suppress secondary electrons [29]. With a functioning Faraday cup in place, the experimental setup is complete and will be explained in the following section.

4.3 Current Experimental Setup, Procedure, and Data

A variety of experimental procedures for measuring the charge fraction of ions passing through thin foils have been employed and the current work has adapted a technique similar to that used by Ishihara et al. [29]. The incident ions pass down the AMS beamline and through a foil in the scattering chamber. The scattering chamber has a moveable target ladder that holds three foil targets, a Faraday cup and an empty frame to allow passage of the bare beam, see Fig 4.2. Some
of the ions are Rutherford scattered into a silicon (monitor) detector located in the scattering chamber, which acts as a normalization tool. The un-scattered beam is sent directly into a Browne-Buechner Spectrograph where the charge state fractions are separated magnetically and measured by an electron suppressed Faraday cup. The spectrograph Faraday cup is mounted on a set of rails referred to as the “zero degree rails” as shown in Fig 4.2. All of the ions are magnetically selected by the dipole “Analyzing” magnet to give ions of well defined energy initially in the 2+ charge state. Passage of the ions through a target foil results in a distribution of charge states (Li: 1+ to 3+, B: 1+ to 5+, C: 1+ to 6+). The spectrograph magnet is carefully scanned to bring each of the charge states onto the Faraday cup and the identity of each charge state is then verified using the following scaling procedure:

Figure 4.1. An example of testing the Faraday cup suppression voltage. The red line is there to guide the eye.
Figure 4.2. Schematic of the experimental setup specific for measuring charge state fractions. For a full overview of the entire AMS beamline see references [49, 50]. The (*) indicates the target ladder and its general makeup, the silicon monitor detector in relation to the foils and the Faraday cup (FC) mount on the zero degree rails.
where $B$ is the magnetic field of the spectrograph, $I_{\text{magnet}}$ is the current supplied to the spectrograph magnet and $q$ is the charge state. With the charge state distribution mapped, the next step is to bring the first charge state back onto the Faraday cup and maximize the current by adjusting the magnetic field. Then several short scans are taken while measuring the integrated current on the spectrograph Faraday cup and the scattered beam on the monitor detector to check reproducibility. Once the first charge state has been measured then the next charge state is moved onto the Faraday cup and the procedure is repeated. When a charge state distribution for a given energy has been measured, the charge fractions can be calculated using $F_q = N_q / \sum N_q$, where $F_q$ is the charge fraction and $N_q$ is $N_q = I_q / (qeW)$ with $I_q$ being the current read from the Faraday cup, $q$ is the charge state, $e$ is $1.6 \times 10^{-19}$ C and $W$ is the normalization counts from the monitor. Following the determination of the charge fractions, the mean charge can be determined using $\bar{q} = \sum qF_q$ and the distribution width, $d = \sqrt{\sum (q - \bar{q})^2 F_q}$, can also be calculated. This procedure is repeated for multiple ions and energies.

Tables 4.2, 4.3, and 4.4 are the data collected for the current work\textsuperscript{1}. Each table represents a different ion (Li, B, C) and each charge exchanged with the same target material: carbon ($Z_2 = 6$), aluminum ($Z_2 = 13$), copper ($Z_2 = 29$), silver ($Z_2 = 47$), and gold ($Z_2 = 79$). The charge fractions, $F_q$, were directly measured at the given incident energies and the mean charge ($\bar{q}$), the distribution width ($d$), and the skewness ($s$) were calculated using the equations from the previous paragraph. Effective charge ($q_{\text{eff}}$) is explained in section 4.5.2. Section 4.4 looks

\textsuperscript{1}Estimated errors as based on statistics of counts and reproducibility of the repeated measurements are on the order of a few percent or less.
at low-Z ions emerging from carbon targets to demonstrate the validity of the experimental setup. Then a comparison of relevant semi-empirical models to check which model works best for low-Z ions at this energy regime. Finally, the use of local models can give information as to why mean charge models partly fail by examining two common regression fits (linear and polynomial). And section 4.5 will focus on two aspects of charge exchange for low-Z ions that have never been expressed before: 1) the target dependence as a function of the mean charge state and 2) the projectile electron structure, both extending the knowledge of the charge exchange process in solid targets.

4.4 Equilibrium Mean Charge States for Low-Z Ions at $\leq$ 1 MeV/u in Carbon

4.4.1 Introduction

Fast ions traversing a medium will undergo a series of electron capture and loss collisions. If the medium is thick enough then an ample number of collisions will occur to establish an equilibrium charge state distribution. Various experimental studies have been performed using a variety of ion and target combinations covering a wide range of energies. Several reviews and tables have accumulated the data for equilibrium charge state distributions for Be, B, and C ions using carbon foils [5, 10, 19, 62, 66, 79]. Lithium ion equilibrium charge state distributions have been reported by Itoh [30] in the energy range of 1 - 6 MeV and by Stocker and Berkowitz [69] in the energy range of 5.8 - 16.4 MeV. Equilibrium charge state distributions for He ions have been reported by Armstrong [6]. Thin carbon foils of well known thicknesses are relatively simple to make and easy to use. Further-
<table>
<thead>
<tr>
<th>$^7\text{Li}^{2+}$ Incident energy (MeV)</th>
<th>$\eta$</th>
<th>$d$</th>
<th>$s$</th>
<th>$F_2$</th>
<th>$F_3$</th>
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<tbody>
<tr>
<td>$Z_2 = 6$</td>
<td>3</td>
<td>2.65</td>
<td>0.48</td>
<td>-0.63</td>
<td>35.26</td>
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<tr>
<td></td>
<td>4</td>
<td>2.70</td>
<td>0.46</td>
<td>0.82</td>
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<td></td>
<td>5</td>
<td>2.83</td>
<td>0.40</td>
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<tr>
<td></td>
<td>6</td>
<td>2.83</td>
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<td>-1.75</td>
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<td>0.48</td>
<td>-0.63</td>
<td>35.49</td>
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<td>0.36</td>
<td>-1.65</td>
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<td></td>
<td>6</td>
<td>2.81</td>
<td>0.41</td>
<td>-1.53</td>
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<td></td>
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<td>$Z_2 = 29$</td>
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**BORON ION EXPERIMENTAL RESULTS**

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TABLE 4.4

CARBON ION EXPERIMENTAL RESULTS

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more, equilibrium charge state distributions in carbon foils are important because of its common use as a standard for comparison between experiments and with established models.

Energy loss through the foil should be negligible so as to not influence the measurements of the charge fractions. The SRIM2008 program by Ziegler [82] suggests that lithium of 3 - 7 MeV will have an energy loss range of approximately 43 - 60 keV for a 20μg cm\(^{-2}\) thick carbon foil.\(^2\) Boron and carbon ions at 3 - 6 MeV will lose approximately 110 - 118 keV and 143 - 146 keV, respectively, in a carbon foil of the same thickness. These expected energy losses are too small to significantly change the incident ion energy and thereby affect the charge state distribution.\(^3\) Estimated errors as based on statistics of counts and reproducibility of the repeated measurements are on the order of a few percent or less.

In this work, equilibrium charge state distribution measurements have been performed for lithium, boron and carbon ions emerging from a carbon foil in the energy range of 3 - 7 MeV. A comparison with experimental literature values has been made to establish the accuracy of the techniques used for these studies. The combined experimental data have also been compared to the predictions of a variety of semi-empirical formalisms to show their suitability and range of application. Comments and suggestions on the nature of the equilibrium charge state distributions of low-Z ions are presented in section 4.4.4.

4.4.2 Comparison with Literature Data

Since the present configuration has never been used to measure charge state distributions a direct comparison with literature values in carbon foils was first

\(^2\)SRIM2008 can be downloaded at www.SRIM.com

\(^3\)\(^{11}\)B ions were used and then scaled to \(^{11}\)B to compare with established data when necessary.
performed. Figure 4.3 shows the present results for lithium ions with the data for lithium ions by Itoh [30]. Also shown in Fig 4.3 are the present results for boron and carbon ions with carbon ion data by Shima [62]. The agreement between the sets of data for lithium, boron and carbon ions is excellent, which indicates that the present technique is trustworthy. The differences between the different sets of measurements are estimated to be less than one percent. A model developed by Itoh for the prediction of the mean charge state for lithium ions gives excellent agreement to the data as shown in Fig 4.3. A slightly different model for higher Z ions was developed by To-Drouin for boron and carbon, but was tested with ions up to argon [76]. The predictions of the latter model are seen in Fig 4.3 to agree well with the boron and carbon ion data. Table 4.5 summarizes the experimental mean charge states for each ion species at the incident ion energy examined by this group. In all cases, the mean charge state increases monotonically with increasing ion energy. Ions of lower energy could not be examined because of instabilities in the beam transport using the FN tandem accelerator.

4.4.3 Comparison with Established Semi-empirical Models

A variety of semi-empirical models have been developed to predict the experimental mean charge state. These models were usually constructed from data for a limited number of ions and targets and were optimized over a finite energy range. These three models are important because they were designed to fit the low energy range covered in this work. In all the models, the relative ionization ($\bar{\eta}/Z$) is given. This quantity is defined as the mean charge state of the ion divided by its Z.

The To-Drouin model was specifically developed for application to the range
TABLE 4.5
LITHIUM, BORON, AND CARBON ION RESULTS

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<td>0.63</td>
</tr>
<tr>
<td>6</td>
<td>2.83</td>
<td>0.38</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>2.89</td>
<td>0.32</td>
<td></td>
<td></td>
<td></td>
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</tr>
</tbody>
</table>

of ions from boron to argon and the relative ionization is represented as:

$$\bar{q} = Z[1 - \exp(-X)],$$

(4.2)

with the limitations $0.2 \leq X \leq 1.6$ and $5 \leq Z \leq 18$ [75, 76].

The formula created by Schiwietz et al. [53] is a highly parameterized least-square fit built from an array of over 800 data points that span a wide variety of ions and targets. The expression for the relative ionization is given as

$$\bar{q} = Z\left[\frac{8.29x + x^4}{0.06/x + 4 + 7.4x + x^4}\right],$$

(4.3)

where x is a reformulated reduced velocity.

The last expression to be examined here is the Ziegler, Biersack and Littmark
Figure 4.3. Variation of the relative ionization as a function of the incident energy for lithium ions from Itoh [30] (○) and current work (●) compared to the Itoh model (solid line) and similarly for boron and carbon ions from ref [62] (▽, †, respectively) and the current work (▼, ‡, respectively) using the To-Drouin model (solid line).

model that is used in the well known SRIM/TRIM codes. The expression for He ions is given as

$$\bar{q} \approx Z \left[ 1 - \exp \left( - \sum_{i=0}^{5} a_i \ln(E)^i \right) \right]^{1/2}$$

(4.4)

where $a_i$ are fitting constants that were determined to be $a_0 = 0.2865$, $a_1 = 0.1266$, $a_2 = -0.001429$, $a_3 = 0.02402$, $a_4 = -0.01135$ and $a_5 = 0.00175$ and $E$ is in units of keV/amu. For ions of $Z$ greater than 2 the Ziegler, Biersack and Littmark formula can be written as:
\[ \bar{q} = Z[1 - exp(0.803 y_r^{0.3} - 1.3167 y_r^{0.6} - 0.381557 y_r - 0.008983 y_r^3)], \quad (4.5) \]

where \( y_r \) is the reduced velocity. The details of each of these models are described in detail in Chapter 2.

Experimental charge state distributions for Li, B, and C ions are presented with the three model predictions in Fig 4.4, where the relative ionization is given as a function of incident energy per amu. The figures readily show how well the data, current and older, compare with the To-Drouin, Schiwietz and Ziegler-Biersack-Littmark models, respectively. The comparison is quantified by examining the percent deviation between the experimental values and the calculated ones.

The To-Drouin model works best with the higher Z ions such as B and C. Huge deviations are observed between this model and data with the lighter Z ions. For example, the worst case is with He ions, where the deviation is as large as 17%. The deviation decreases to about 12% with Li ions and to about 5.1% for Be ions. Even with B and C ions, the deviation of 8% is the greatest at the lowest energy and less than 3% at higher energies. The To-Drouin model clearly has large errors with ions of low-Z and it is not very accurate with any light ion at low energy.

The comparison of the Schiwietz model with the experimental data, Fig 4.4, shows a reasonably good fit with the data for every ion especially those with Z below that of B ions. The maximum deviations between model predictions and the data are < 3.1% with He ions, < 4.5% with Li ions and < 1.8% with Be ions. As observed with the To-Drouin model, the largest deviations with B and C ions are at the lowest energies with 3 and 6% deviation, respectively. The higher energy B and C ions have deviations of less than 3% and 2%, respectively.
Figure 4.4. Variation of the relative ionization as a function of initial ion energy for the (a) To-Drouin model, (b) Schwietz model and (c) Ziegler-Biersack-Littmark model (solid lines), He ions (□) [6], Li ions (○) [30], Be ions (△) [62], B ions (▽) [62] and C ions (♢) [62]. Closed data points are from this work and open ones are from the literature.
The Ziegler-Biersack-Littmark model predictions for all of the ions other than He are within 9% of the data. The general trend of the model is to give larger deviations at lower energies and better agreement with the data as the energy increases. Except at the lowest energies, the Schiwietz model reproduces the data very well and it seems to be the best of the three models in this regime. With increasing Z, the To-Drouin model starts to give values similar to that with the Schiwietz model, but the upper bound on the To-Drouin model is argon ions whereas the Schiwietz model has been tested with ions of much larger Z. In general, all the models seem to improve with increasing energy. None of models examined here specifically addresses the shell effects that have been reported with Z and $Z_2$ oscillations [63]. These shell effects originate with the electronic structure of the projectile ion and include the ability of the target atom to be an electron sink or reservoir. An example of this shell effect is as follows, when bromine ions are stripped in solids near 140 MeV the mean charge reaches 25[79]. Further ionization requires removal of L-electrons from the ion. However, those electrons are more tightly bound and the probability of removing them becomes small in comparison to lower energy M-electrons. This kind of behavior leads to oscillatory structure instead of smooth trends in the relative ionization as a function of energy. The observation of this type of data is an indicator of some of the underlying physics in charge exchange.

There seems to be a systematic deviation between model predictions and the data that are more prominent at lowers energies. In the higher energy regime, the ions are fully-stripped or are proton-like projectiles where shell effects have become negligible and the relative ionization ($\bar{q}/Z$) becomes Z-independent. A compensation Z (ion)-dependent term to allow for shell effects in the low energy
region has been proposed in addition to the standard X value presented in the semi-empirical models for the mean charge state or relative ionization [30].

The parameterizations of some of the models for the prediction of the mean charge state have suffered from the lack of systematic data. The use of only a few data points can lead to oversimplification and erroneous extrapolation to ion of other Z or different energy regimes. On the other hand, there has been a general trend to look at massive pools of data to build a master expression that covers everything. This approach often misses potential underlying features. Figure 4.5a shows the relative ionization of carbon ions emerging from a carbon foil over the energy range of 0 - 5 MeV/u. The figure contains three sets of data that give what looks like a relatively smooth variation in charge as a function of energy. However, examination of the data over a smaller energy range such as 0 - 0.7 MeV/u as in Fig 4.5b gives a different view of the data. The solid line drawn in Fig 4.5 is there to guide the eye and shows a distinctive step behavior. All three sets of data that are shown have the same response, which strongly suggests that this effect is real and reproducible.

The approach to the fitting of data in Fig 4.5a is commonly used to achieve a global fit of the data. Such a global model is built with the intention of covering of wide array of ions, target materials, and energy, but at the loss of accuracy in some energy regimes. The question now arises as to how a local model compares to the best of the global models. A local model would give an expression that covers a smaller range of energies and a specific ion-target combination. There are several regression fits that can be used to build such a local model. The following section explores several approaches and present arguments on the physicality of such choices.
Figure 4.5. Relative ionization of carbon ions through carbon foils for an energy range from 0 - 5 MeV/u (a) and 0 - 0.7 MeV/u (b). The line through the data points is to guide the eye. Open squares are from ref [79]. Closed squares are current data. Open squares with a plus are from ref [62].
4.4.4 Development of Local Models

Most semi-empirical relative ionization models (global and local) are built based on the work of Betz[14] who in turn used a suggestion from Heckman et al. [25] to focus on $ln(1 - \bar{q}/Z)$, which represents the mean relative number of electrons carried by the ions, as a function of a reduced velocity. Following a simple rearrangement, the relative ionization can take the form of $\bar{q} = Z[1 - T exp(-\frac{\nu}{v_B Z})]$, where $T$ and $\gamma$ are determined empirically and $\nu$ is the ion velocity and $v_B$ is the Bohr velocity. The quantity inside the parentheses is readily identified as the reduced velocity.

The general trend of the experimental data is that the mean charge state increases uniformly (monotonically) with energy, which suggests a linear function as a valid choice. The appearance of the regression fit will take the form of $AX + B$ where $A$ is the slope and $B$ is the y-intercept in the linear relationship. The mean charge will take the form of $\bar{q} = Z[1 - exp(AX + B)]$. A second regression fit that will be examined here is a third-order polynomial fit in which the mean charge will take the form of $\bar{q} = Z[1 - exp(CX^3 + DX^2 + EX)]$ where $C$, $D$, $E$ are fitting constants and the y-intercept is set to zero to mimic the look of the model developed by Shima-Ishihara-Mikumo [62, 63].

Due to the apparent linear nature of the relative ionization data and models in the literature, a linear regression fit was deemed suitable as a first attempt to develop a good local model. The values for $ln(1 - \bar{q}/Z)$ as a function of the reduced velocity, $X$, are shown in Fig 4.6 for the five ion species through a carbon target. The resulting coefficients are displayed in Tables 4.6 and 4.7 with the corresponding $R^2$ values, which describe statistically how well the regression fits to the data. A $R^2$ value of 1 is considered a perfect fit.
Figure 4.6. Fit of the parameter $\ln(1 - \bar{q}/Z)$ as a function of the (a) linear and (b) polynomial coordinate $X$ (solid lines) with He ions (□) [6], Li ions (○) [30], Be ions (△) [62], (▽) [62] and C ions (◇) [62]. Closed symbols are this work and open symbols are from the literature.
TABLE 4.6
THE COEFFICIENTS AND $R^2$ VALUES RELATED TO LINEAR FITS

<table>
<thead>
<tr>
<th></th>
<th>A</th>
<th>B</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>-2.2469</td>
<td>0.9731</td>
<td>0.9914</td>
</tr>
<tr>
<td>Li</td>
<td>-1.7038</td>
<td>0.5112</td>
<td>0.9956</td>
</tr>
<tr>
<td>Be</td>
<td>-1.2787</td>
<td>0.2195</td>
<td>0.9992</td>
</tr>
<tr>
<td>B</td>
<td>-1.1764</td>
<td>0.1951</td>
<td>0.9937</td>
</tr>
<tr>
<td>C</td>
<td>-0.9866</td>
<td>0.0195</td>
<td>0.9929</td>
</tr>
</tbody>
</table>

The fits for He and Li ions in Fig 4.6a agree well with the lines either going through the data points or just grazing them, while Be and higher Z ions seem to be well represented with linear functions. With the development of a functional fit, a balance of attaining the highest $R^2$ value must be tempered with a little bit of physics. For example, as $X \to 0$ so does $\bar{q}/Z$ and the expression must have a y-intercept of zero. The fits to the data of Fig 4.6 clearly show that the best regressions have non-zero intercepts, which leads to unrealistic physics as $X \to 0$. Forcing a zero y-intercept on the linear fits creates an equally poor situation where the fit does not conform to the data. The results suggest that despite a linear trend in the data higher order terms are needed.

Third-order polynomial fits of the data are given in Fig 4.6b. The rational for this type of functional dependence is based on the model of Shima-Ishihara-Mikumo, but the energy ranges of that model are not applicable to the current
TABLE 4.7

THE COEFFICIENTS AND $R^2$ VALUES RELATED TO THIRD ORDER POLYNOMIAL FITS

<table>
<thead>
<tr>
<th></th>
<th>C</th>
<th>D</th>
<th>E</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>0.4825</td>
<td>-2.0204</td>
<td>0.3054</td>
<td>0.9924</td>
</tr>
<tr>
<td>Li</td>
<td>0.3697</td>
<td>-1.4065</td>
<td>-0.1225</td>
<td>0.999</td>
</tr>
<tr>
<td>Be</td>
<td>0.1242</td>
<td>-0.4737</td>
<td>-0.7054</td>
<td>0.9995</td>
</tr>
<tr>
<td>B</td>
<td>0.3373</td>
<td>-0.9413</td>
<td>-0.3793</td>
<td>0.995</td>
</tr>
<tr>
<td>C</td>
<td>0.4214</td>
<td>-0.858</td>
<td>-0.5474</td>
<td>0.9982</td>
</tr>
</tbody>
</table>

work [62, 66]. The $R^2$ values for the polynomial fits are slightly better than the linear fits. Statistically, both regression fits are close enough to be interchangeable, but the third-order polynomial fit has no y-intercept to give more realistic physics.

Does a better fit always imply the correct physics? The argument could be made that until enough information is collected the best fit would be the more correct approach. It should be noted that the Itoh model has a non-zero y-intercept, but as stated in reference [30] one of its limitations is the very low energy regime. The physics of charge exchange in the very low energy regime appears to be different than in the swift velocity regime. Low energies lead to a more quasi-molecular state being prevalent and energy loss through the medium is dominated by nuclear interactions. Perhaps the non-zero y-intercept term mimics some of the underlying physics, but the problem persists as to the nature of that physics and how to quantify it correctly in future mathematical fits.

The expectation is that a local model should provide a better fit than a global
model because the regression fits are not weighed down by a large pool of data in various energy regimes. A more accurate model with respect to the variation in incident ions can be obtained when the target medium remains the same. As noted earlier, this approach clearly provides the advantage of observing potential underlying structure.

4.4.5 Conclusion

Experimental observations for charge fraction measurements of Li, B, and C ions on carbon targets are found to be in excellent agreement with established data for the 3 - 7 MeV energy range. Comparison of the data with models that were developed for the energy range examined here shows that these models are not very accurate at very low energies (< 3 MeV). Of the three models examined, the Schiwietz model clearly shows the best response for lower energy low-Z ions. The Schiwietz model follows one of the suggestions by Itoh et al.[30] and includes a Z-dependence. None of the models presented here for determination of mean charge take into account atomic phenomena like shell structure and oscillation effects that have been reported by Shima[64]. The nature of the Z-dependence in this energy range should continue to be explored with these same projectiles, but to include a systematic study of energy ranges as well as other target material. The following section will include target material other than carbon and focus on the charge state distribution, the distribution width and charge fractions as parameters to evaluate charge exchange processes.
4.5 Equilibrium Charge State Distributions of Low-Z ions Through Various Foils

4.5.1 Introduction

Various experimental studies have been performed using a variety of ion and target combinations covering a wide range of energies and assembled in several compilations of charge state data [5, 6, 10, 24, 30, 62, 66, 69, 79]. However, these studies principally use carbon targets. Only a few papers highlight the dependence of charge states on target media, and these studies are typically for large energies and high-Z projectiles. An understanding of target dependence for the charge transfer, exchange and equilibration of low-Z ions will greatly advance the fundamental science of this field and aid in many practical applications involving the transport of ions.

The following sections will examine the effects of target materials on ion transport by describing: the dependence of the equilibrium mean charge state ($\bar{q}$) and the effective charge ($q_{eff}$) on the target atomic number ($Z_2$); the behavior of individual charge state fractions as a function of charge state and energy; and the influence of charge state distributions on a variety of parameters.

4.5.2 Equilibrium Mean Charge State and Effective Charge

The mean charge state typically displays a monotonic behavior as a function of ion energy for specific ion-atom systems and can be plotted with unitless parameters, such as a reduced velocity, X. Relative ionization, $\bar{q}/Z$, is often used to combine data from multiple systems to build universal models for predicting the mean charge state. Despite these efforts, the predictive capabilities of these models are limited by the underlying physics and the associated data pool used for parameterization. Of the many limitations to charge state models, one is a
lack of target dependence information. The most direct method to probe the target dependence in ion-atom collisions is to examine the mean charge state as a function of the target’s atomic number.

Figure 4.7 shows a typical variation of the mean charge state for a 4 MeV carbon ion through different solid targets (C, Al, Cu, Ag and Au). The mean charge state shows a significant variation as a function of the target atomic number. Model predictions are also shown in this figure for comparison. The model developed by To-Drouin was designed for carbon targets, which it can handle well [76]. However, this model has no target dependence with the expected result as
shown in Fig 4.7. Three other models examined here incorporate some form of target dependence with varying degrees of success. The Shima-Ishihara-Mikumo model undershoots the data severely with increasing target Z [57]. This model was developed using high-Z projectiles where the ion-atom interaction physics is not the same as with low-Z projectiles. The Ziegler-Biersack-Littmark model accounts for the target media using condensed matter considerations, but the result does not display the same target dependence as the experimental data [82]. Finally, a model developed by Schiwietz et al. utilized a wide ranged data pool to develop a formalism that includes shell-corrections and deformation affects at different energies [53]. This model gives the best agreement with the data than any other model presently available, yet its predictions are still not acceptable. A better understanding of charge exchange processes for low-Z projectiles could provide some insight to help correct future models or provide corrections to some of the current models.

Mean charge states are shown in Fig 4.8 for (a) boron and (b) carbon ions of 3 - 6 MeV energy through a variety of target materials (C, Al, Cu, Ag, Au). Both ions exhibit an increase in their mean charge state with increasing ion energy in all targets. Boron ions, Fig 4.8a, have a relatively greater mean charge state in silver targets than in the other targets. Carbon ions, Fig 4.8b, behave differently by having copper or aluminum produce the largest mean charge state at the higher energies and copper or silver at the lower energies. The difference in behavior for the two ions with the same target is interesting, which suggests that charge exchange can be influenced by the projectile as well as the target. Mean charge states with lithium ions do not fluctuate as much as with boron and carbon ions, which is probably due to the lithium ions being nearly stripped at these energies.
Figure 4.8. Equilibrium mean charge states as a function of $Z_2$ for: (a) boron ions of incident energy (■) 3.3 MeV, (○) 4.4 MeV, (▲) 5.5 MeV, (▼) 6.05 MeV and (◇) 6.6 MeV, and (b) carbon ions (■) 3 MeV, (○) 4 MeV, (▲) 5 MeV, (▼) 5.5 MeV and (◇) 6 MeV.
The electrons remaining on the projectile are influencing the mean charge state.

An alternate way to display the target dependence is to look at the mean charge as a function of incident energy for each target. This presentation allows for an easier comparison in terms of energy and the evolution of the mean charge. Figure 4.9 shows boron ions from various target species. The insert focuses on the region of interest for the current work. As mentioned in Fig 4.8 at higher energy, silver has the largest mean charge followed by copper, carbon, aluminum, and then gold. At lower energies something interesting happens, all target species present some fluctuations to the projectile mean charge. This fluctuation then can cause a shift in the mean charge that could be extreme. For example, at 3.5 MeV carbon decreases low enough that the gold target data have a higher mean charge. Similar behavior can be seen in Fig 4.10 with carbon ions emerging from the same target materials as the boron ions. The largest mean charge at high energy is now aluminum and copper and gold remains the lowest mean charge. At lower energies the mean charge order shifts with copper and silver as the largest mean charge and aluminum, carbon, and gold having the lowest mean charge. The typical behavior cited by Shima et al.[60] has gold with the lowest mean charge state and the lower the target atomic number ($Z_2$), the larger the mean charge. This behavior is not present for low-Z ions at this energy regime. It is unclear if this is unique for the energy regime covered by the present work or if the projectile shell structure shifts at high energies to fit the Shima description[60].

In addition to the target having an effect on the mean charge state, $\bar{q}$, the target will also influence the effective charge $q_{eff}$, which is a quantity based on stopping power of the medium[56, 67, 68]. The stopping power $S$ in the Bethe-Bloch energy regime is proportional to the square of the effective charge of the
Figure 4.9. Equilibrium mean charge states as a function of incident energy for various targets, where (*) is a closeup of the current target data: C(□, ■), Al(○, ●), Cu(▲), Ag(▼) and Au(♦). The open symbols belong to previous published data [80] and the closed symbols from the current work.
Figure 4.10. Equilibrium mean charge states as a function of incident energy for various targets, where (*) is a closeup of the current target data: C(□, ■), Al(○, ●), Cu(▲), Ag(▼) and Au(♦). The open symbols belong to previous published data [80] and the closed symbols from the current work.
ion, $S = (q_{eff})^2 \Theta$, where $\Theta$ is a function of ion energy, mean ionization potential and correction terms. Stopping power compilations sometimes assume that $\Theta$ is approximately the same for light ions, typically a proton, as for heavy ions of the same velocity in a given medium. The quantity $q_{eff}$ for an ion to induce a stopping power $S$ in a given medium can be defined as

$$q_{eff} = (q_{eff})_{proton} \left( \frac{S}{S_{proton}} \right)^{1/2},$$

(4.6)

where the effective charge of the proton is assumed to be unity. The effective charge is a quantity that describes the degree to which the charge states inside the foil are excited, whereas $q$ is the ground state of the mean charge state on leaving the foil. Ions passing within a foil will undergo sufficiently rapid collisions that the electrons are not in an equilibrium state and only after the ion leaves the foil will its charge state relax to the mean charge state.

The values of the effective charge ($q_{eff}$) for boron and carbon ions are shown in Tables 4.3 and 4.4, respectively. The effective charge was calculated from Eqn 4.6 where the stopping power of the heavy ion and the proton at the same velocity is determined by the SRIM2008 program developed by J.F. Ziegler et al. The values of the effective charge are always larger than the corresponding ones for the mean charge state, which means that ions have slightly fewer electrons in a material than the average number of electrons after exiting that material. Shima et al. [60] note that highly charged ions have good agreement between effective charge and the mean charge state since the Auger electron emission process that occurs post foil contributes little and can be neglected. This assumption has been shown to be valid for high energy (nearly fully stripped) boron and carbon ions in carbon [66]. Agreement between effective charge and the mean charge state is good at the
lower energies where the carbon ion becomes neutral and at the highest energies (59.9 MeV: \( q_{\text{eff}} = 5.94 \) and \( \overline{q} = 5.96 \)) [66] corresponding to a fully stripped carbon ion, see Fig 4.11. Similar behavior is observed for boron ions.

4.5.3 Charge State Distributions

In addition to developing semi-empirical models for equilibrium mean charge states, there is also a need to calculate the equilibrium charge fractions. Equilibrium charge state fractions give information on the relative rate of change of charge states that can be used to investigate the underlying physical processes responsible for charge equilibrium. Charge state distributions for ions of \( Z \) greater
than beryllium are often approximated by a Gaussian (symmetric) distribution. The expression for a Gaussian distribution can be described in terms related to charge states as the following: 

\[ F_q = \frac{1}{d\sqrt{2\pi}} \exp\left[-\frac{(q - \bar{q})^2}{2d^2}\right] \]

where \( F_q \) is the charge state fraction, \( d \) is the charge state distribution width, \( q \) is the charge state and \( \bar{q} \) is the mean charge state (Eqn 2.19) [62]. The physical description is that when the outermost electrons of the ion are distributed over a shell \( n \), where \( n \) is the principal quantum number, then the distribution can be approximated as a Gaussian [66]. There are instances when the charge state distributions are not symmetric. Asymmetric charge state distributions occur when the outermost electrons are spread between two adjacent \( n \) shells as has been shown by Shima et al. [58] to be approximated by two Gaussians. However, this phenomenon has only been demonstrated with high-Z ions where this shell effect seems to be more enhanced [8, 33, 42, 58]. Shell effects have been observed in both equilibrium and non-equilibrium charge states with closed-shell electron structures. In addition, there are special cases where asymmetric behavior has been observed with low and high velocity ions. According to Baudinet-Robinet, charge state distribution observed behind carbon targets can be approximated by \( \chi^2 \), Gaussian, and reduced \( \chi^2 \) distributions for low-, intermediate-, and high-charge ions, respectively [42, 62].

Skewness can be used to check the symmetry of the charge state distribution using the definition of 

\[ s = \sum_q (q - \bar{q})^3 F_q / d^3 \]

where \( q \) is the charge state, \( \bar{q} \) is the mean charge state, \( F_q \) is the charge state fraction and \( d \) is the distribution width.

Data for boron and carbon ions given in Tables 4.3 and 4.4 clearly show in all instances that the skewness of the charge state distributions suggests a Gaussian distribution as a suitable choice for representing the charge state distri-
bution. On the other hand, the lithium ion data in Table 4.2 show heavily skewed values. Lithium ions are exchanging electrons mainly in its K shell at these energies, while the boron and carbon ions are exchanging electrons in both their K and L shells. For a Gaussian distribution, the skewness is ideally equal to zero and the distribution width can be related to the full width at half maximum as 

\[ 2d(2\ln(2))^{1/2} \approx 2.35d. \]  

[10, 79]

The skewness values for the distributions of boron and carbon ions through the various foils in the energy range of this study were less than 1% when using a Gaussian distribution suggesting that the maxima of the distributions are nearly equal to the mean charge states. The energy range examined in this work does not allow the peripheral charge states (2+ and 5+ for boron, 2+ and 6+ for carbon) to ever dominate, which also helps maintain a Gaussian distribution.

Because of the relatively small skewness in the data for boron and carbon ions, Gaussian distributions can be used to fit the charge exchange data to examine the evolution of the charge states as the incident ions lose energy. The evolution of the charge state distributions as functions of incident ion energy are shown in Fig 4.12a for boron in the target materials examined here. All of the targets show the same trend for the variation in the distributions with respect to the boron ion energy despite the differences in mean charge state. One might expect the distribution to simply shift to lower values of the charge state as the projectile gains electrons with decreasing velocity. Instead, Fig 4.12a shows that the distribution widens with a corresponding decrease in the maximum value as the charge state decreases from 4 to about 3.5. With further decrease in boron ion energy, the distribution then increases its maximum value and narrows as the charge state approached the value of 3. The observed similarity in the variation of the charge
state distributions with change in projectile velocity for all the targets suggests that the charge exchange process is influenced by the proximity of the mean charge state to the projectile electronic shell structure, especially if the electronic shell is closed. This phenomenon has been described as a shell effect by Moak et al.[42]. For boron and carbon ions, the only closed shell is the K shell (1s^2) electrons, which are the inner and most tightly bound electrons.

The charge state distributions for boron ions in the energy range examined here suggests that electron exchange processes involving the K-L shell boundary occur at about 3.3 MeV as indicated in Fig 4.12a by the sizeable jump in the distributions on passing through this energy. The data for boron ions in carbon targets make a suitable baseline to judge this trend since there are previously published data that extend below the energy range of the current work[62]. The distributions at 3.3 MeV for aluminum, copper, silver, and gold targets are similar to that for carbon suggesting that the influence of the K shell is observed for all materials. There is a readily observed anomaly in the charge state distribution for gold at 4.4 MeV that can also be observed to a lesser degree in the other targets. This variation reflects the loss of the last L shell electron due to its interactions with electron orbitals of the medium. Charge state distributions also show some variation with target atoms at the higher energies. Whereas the 3.3 MeV distributions are relatively narrow, the distributions at higher energies are broad. This result suggests a quality previously noted for gold targets as a weak charge exchanging medium, either as a poor electron stripper or as an abundant and rich electron reservoir for interaction with the incident ion[56].

The charge state distributions for carbon ions in the energy range examined here suggests the K-L shell boundary is at about 5 MeV as indicated in Fig 4.13a
by the narrowing of the distribution when passing through this energy. This observation is most prevalent for the carbon and aluminum targets, where the adjacent distributions (at 3, 4, 5.5 and 6 MeV) are nearly identical in behavior. For copper, silver and gold the shell effect is not as strong at 5 MeV since the mean charge state has started to shift further from $Z - \bar{q} = 2$, the K-L shell boundary condition. This shift is distinctly observed with gold targets as the K-L shell boundary condition is not met until the upper energy limit of the current work is reached. This result suggests that a gold target is a weak charge exchanging medium, either as a poor electron stripper or as an abundant and rich electron reservoir to interact with the incident ion [56].

Charge state fractions shown in Figs 4.12b and 4.13b are for boron and carbon ions, respectively, in carbon, aluminum, copper, silver and gold targets as a function of incident ion energy. Figures 4.12b and 4.13b also contain dashed lines representing the charge state fractions as calculated from an energy loss program called CasP, Convolution approximation for swift Particles, developed by Schiwietz and Grande [53]. In general, there is reasonable agreement between the data and CasP calculations. Typically the most dominant charge states have the best agreement otherwise the deviation can be interpreted as experimental error due to a larger influence that beam fluctuations have on lower populated charge states and the limitations in any model or calculation. The instances where the dashed lines abruptly end are where CasP does not calculate charge state fractions of less than 1%.

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4The CasP software is windows based and can be locate at http://www.hmi.de/people/schiwietz/casp.html with additional references and publications.
Figure 4.12: (a) Gaussian charge state distributions in carbon, aluminum, copper, silver and gold targets for: (a) boron ions at incident energies of (›) 2.5 MeV, (<) 3.0 MeV, (>) 3.5 MeV and (▽) 4.5 (■) 3.3 MeV, (●) 4.4 MeV, (▲) 5.5 MeV, (▼) 6.05 MeV and (♦) 6.6 MeV. The solid symbols are current data, open symbols from reference [3], and the solid lines are Gaussian fits for different energies. (b) Charge state fractions as a function of incident energy for boron ions. The solid data points represent current data and open data points are from reference [4]. The charge states are (■) 2+, (●) 3+, (▲) 4+, (▼) 5+ and (♦) 6+. The solid lines are from the polynomial fit and the dashed lines are from the CasP program.
Figure 4.12. See previous page
Figure 4.13: Gaussian charge state distributions in carbon, aluminum, copper, silver and gold targets for: (a) carbon ions of incident energies (■) 3 MeV, (●) 4 MeV, (▲) 5 MeV, (▼) 5.5 MeV and (♦) 6 MeV. The solid symbols are current data, open symbols from reference [79], and the solid lines are Gaussian fits for different energies. (b) Charge state fractions as a function of incident energy for carbon ions. The solid data points represent current data and open data points are from reference [4]. The charge states are (■) 2+, (●) 3+, (▲) 4+, (▼) 5+ and (♦) 6+. The solid lines are from the polynomial fit and the dashed lines are from the CasP program.
Figure 4.13. See previous page
Another approach for determining charge state fractions, as outlined in ref [73], is to fit polynomial curves to each of the charge fraction data as a function of energy. The charge fraction can then be written as \( F_q(\%) = 10^Y \), where \( Y = A + B1 \times E(\text{MeV/u}) + B2 \times E(\text{MeV/u})^2 \). Such an approach has limitations in that no insight is gained on the nature of charge exchange, the expressions can only be used within the energy range employed for constructing them and the expressions have to be based on existing experimental data. The resulting fits for boron and carbon ions are shown in Figures 4.12b and 4.13b, respectively.

The observed equilibrium charge state fractions of adjacent charge states, \( F_q \) and \( F_{q+1} \), can be plotted as functions of charge state and incident ion energy as an effective means of inspecting shell effects on the charge state distribution. When only single electron capture and loss processes are considered then the ratio of \( F_{q+1}/F_q \) can be treated as \( \sigma_{\text{loss}}/\sigma_{\text{capture}} \), where this value is the ratio of electron loss cross-sections to electron capture cross-sections [10]. Figure 4.14a displays the ratios of \( F_{q+1}/F_q \) as functions of the charge states for 5 MeV carbon ions in all of the target materials used in the current work. In Fig 4.14a each target is scaled by multiplying it by a constant to prevent data overlap: carbon by 0.01, aluminum by 0.1, copper by 1, silver by 10, and gold by 100. Figure 4.14b displays the ratios of \( F_{q+1}/F_q \) as functions of the charge states for different energy boron ions in aluminum. The results show that in general \( F_{q+1}/F_q \propto q^{-\text{constant}} \) holds true. The data of Fig 4.14a for 5 MeV carbon emphasizes the breakdown of the aforementioned power law relation and indicates that the charge state distribution deviates from the Gaussian shape. This deviation reflects the difference with the electron loss and capture processes that take place at the K-L shell boundary [29, 61]. Any additional fluctuations in slope other than that can be attributed to the
Figure 4.14. Adjacent charge state fractions as a function of charge state for: (a) 5 MeV carbon ions in (■) carbon, (●) aluminum(▲) copper, (▼) silver and (▲) gold; and (b) boron ions in aluminum at incident energies of (■) 3.3 MeV, (●) 4.4 MeV, (▲) 5.5 MeV, (▼) 6.05 MeV. Adjacent charge state fractions as a function of energy for: (c) carbon ions in carbon and (d) boron ions in aluminum. In parentheses is the charge state ratio represented.
proximity to an adjacent closed electron shell boundary, subshell interactions and sometimes a statistical anomaly related to experimental equipment. In Fig 4.14c, the ratio of the adjacent charge states as a function of energy show deviation from the power law trend \( \left( \frac{F_{q+1}}{F_q} \propto E^{\text{constant}} \right) \) at the energy where the shell effect should be observed at \( (5 \text{ MeV}) \) and likewise in Fig 4.14d the effect does not exist in that ion/atom system for the range of energies shown.

4.5.4 Charge State Distribution Width

The first serious attempt at an analysis of the width of the charge state distribution was performed by Shima et al.[59]. They developed a model from an examination of the charge state distribution width as a function of the reduced velocity. Most of the detailed data considered were chlorine, bromine and copper ions, but data for oxygen ions were also included. This work will focus on the charge distribution widths for carbon foils since a wide range of projectile velocity data already exist. General observations from carbon targets can then be used as a baseline for other target species.

No previous work has examined the width of the charge state distributions with low-Z ions in carbon targets, let alone for a variety of targets. Several empirical models have been developed for the prediction of charge state distribution widths (Baudinet-Robinet [8], Nikolaev-Dmitriev [47], and Shima-Ishihara-Mikumo [59]). Predictions from these models do not agree well with the available low-Z ion data, since the models are either based on experimental data that cover a limited energy range or they are based on experimental data obtained with high-Z ions.

The charge state distributions for carbon ions in carbon are presented in Fig 4.15 for better observation. A close examination of the charge state dis-
Figure 4.15. Charge state distributions for carbon ions in carbon as a function of the charge state where the closed symbol data come from Schmitt et al. [54] and the open symbol data comes from Wittkower[79]. The dashed line marks where the carbon ion has a mean value of two electrons.

The distribution shows that the distribution with the most narrow width (5 MeV, ▼) and smallest d value occurs when the mean number of remaining electrons is 2, \((Z - q) = 2\), which corresponds to a closed shell boundary associated with the K shell. The discontinuity due to a shell effect can be seen more clearly in Fig 4.16 where the charge distribution width is plotted as a function of the reduced velocity, where \(X = 3.86Z^{-0.45} \sqrt{E(MeV)/m(u)}\). In this figure, the K shell boundary can be located at \(X \approx 1.1\) where the width shows a sharp discontinuity. Figure 4.16 gives the first indications of structure associated with the projectile, especially with the K-L shell boundary clearly marked.
Despite there being twice as many electrons in the L as in the K shell, relatively little change in incident ion velocity is required to strip the L electrons away. A few characteristics that can be attributed to the carbon ion structure are correlated to specific velocity regions. The region pertaining to the K shell electrons that starts near \( X = 2 \) and \( Z - \bar{q} = 0.95 \), and continues to higher reduced velocities is where the mean charge state is similar to \( Z \) and the width of the charge state distribution decreases linearly with the increase in projectile energy. This region is where the outermost electrons of the projectile are distributed in the K shell and is probably
Figure 4.17. Charge distribution width, d, as a function of the mean number of remaining electrons for: (■) helium, (●) lithium, (▲) beryllium, (▼) boron and (◇) carbon ions. The closed symbols are current data and open from [6, 24, 30, 62, 66, 69, 79]. The vertical line defines the K-L boundary, while the vertical dashed lines mark reduced velocity X regions.
Figure 4.18. Distribution widths for boron ions through various target materials as a function of reduced velocity $X$ with (*) displays a close up of the current target data: Al (●), Cu (▼), Ag (▲), and Au (◆) and the open symbols C (□) and Al (○) from ref[79]. The solid line is drawn to guide the eye.
representative of all collisions in which the projectile is nearly stripped. The K-L shell boundary at $Z - q = 2$ can be thought of as the region where K and L shell electrons are mixing and provides the most distinctive feature in Fig 4.16. From this region down to where $Z - q = 5.01$ the outermost electrons of the projectile are distributed in the L shell. Beyond this region the width decreases rapidly with decreasing projectile energy as the projectile becomes neutral. Shima et al. noted a different trend for high-Z ions and suggested that a similar response would not be expected for low-Z ions[59]. On the contrary, shell effects can be observed for low-Z ions, which show that electronic effects have an important role in charge exchange processes.

Figure 4.17 displays the charge distribution width values plotted as a function of the mean number of electrons remaining on the projectile for ions with $Z \leq 6$ passing through carbon foils. The strongest influence on the charge distribution width can be seen at the $Z - q = 2$ boundary corresponding to the K-L boundary. Each of the ions exhibits a node or envelope relating to the individual electronic subshells. Helium has only K shell electrons ($1s^2$) and the current data for lithium mainly displays K shell electrons, which implies that the lone L shell electron on the lithium ($2s^1$) is easily stripped away. Beryllium is the first low-Z ion that has a second filled L subshell ($2s^2$) although these electrons do not bind as strongly as the K shell electrons. A second node/envelope is clearly visible with beryllium ions above $Z - q = 2$ corresponding to the L shell. Boron and carbon both have filled $1s^22s^2$ subshells, but have loosely bound $2p$ electrons where the oscillation in the charge distribution width is more pronounced. Overall, the presentation of the data as in Fig 4.17 gives the most sensitive profile of shell effects ever presented. As $q = Z$ at high projectile velocities almost all Z dependence is lost,
which is observed as the linear region in Fig 4.16 and the common response region of Fig 4.17.

A wide range of data exists with carbon foils, but data for other targets are scant and more dispersed. However, there is sufficient data to demonstrate the charge distribution width dependence on target material for low-Z ions. Figure 4.18 presents the values of the charge distribution width as a function of the reduced velocity for boron ion projectiles, where carbon target data are used as a baseline (the solid line). The other target material (aluminum, copper, silver and gold) are enclosed in the insert to provide a close up examination of target dependences. Figure 4.18 clearly shows that at one extreme the aluminum targets behave similarly to the carbon targets, while at the other gold shows the greatest deviation. These results are consistent with what has already been observed in the previous figures such as Fig 4.16.

4.5.5 Empirical Formula’s for Low-Z Ions through Carbon

Empirical models for charge distribution widths of low-Z ions do not currently exist. The ability to predict the distribution width, d, is important when the charge state distribution mimics a Gaussian function because it can be used to calculate charge fractions. All that is needed is a reliable model for the equilibrium mean charge then the predictive power to determine charge states fractions using Eqn 2.19 exists. The challenge in determining the distribution width comes from the projectile electronic structure, which makes each ion unique and therefore difficult to develop a universal model.

In Fig 4.19, a plot of \(d/Z\) is rescaled to take advantage of larger reduced velocities since at those velocities a linear trend is observed. The results show
that at $X \geq 2$ for He to B ions and at $X \geq 2.5$ for C ions there is a linear trend that can be described as

$$\log(d/Z^{1.2}) = -0.360X - 0.2397.$$  \hfill (4.7)

As mentioned this kind of expression has been missing for low-Z ions because atomic structure dominates at $X < 2$ with obvious fluctuations due to the projectile Z. A polynomial of order $m$ can be used to fit the varied envelopes, where $m = 1 + Z$ with Z being the ion’s atomic number. The polynomial fit can be generalized to $y = A + \sum_{i=1}^{m} B_{i}X^{i}$, where A and B are constants determined by the fit and X is the reduced velocity and is limited by $0.25 \leq X \leq 2$. Table 4.8 displays the polynomial fitting parameters for ions of helium to carbon. An example of how well this model functions can be demonstrated with a 6MeV carbon ion through carbon target, where the experimentally determined mean charge and distribution width are 4.19 and 0.63, respectively. The Schiwietz model, providing the best results for mean charge, calculates a mean charge of 4.19 and the empirical expression for the distribution gives 0.645. The calculated charge fractions deviated less than 3% from the experimental results.

4.5.6 Conclusion

Equilibrium charge state distributions of Li, B, and C ions through carbon, aluminum, copper, silver, and gold targets have been measured and compared with the previously published results to provide insight on the dependence of charge exchange on target materials. The charge distributions for these ions and targets at the measured energies can be modeled with a Gaussian. Shell effects
Figure 4.19. Log(d/Z^{1/2}) as a function of the reduced velocity. The open symbols are older data where (□) He, (○) Li, (△) Be, (▽) B and (♢) C are from [6, 24, 30, 62, 66, 69, 79]. The dashed line indicates a fit to the data.
<table>
<thead>
<tr>
<th>Ion</th>
<th>He</th>
<th>Li</th>
<th>Be</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>-1.45151</td>
<td>-2.26797</td>
<td>-1.00947</td>
<td>-1.16557</td>
<td>-1.3736</td>
</tr>
<tr>
<td>$B_1$</td>
<td>1.67547</td>
<td>3.08476</td>
<td>0.08991</td>
<td>1.15722</td>
<td>2.25812</td>
</tr>
<tr>
<td>$B_2$</td>
<td>-1.0876</td>
<td>-2.02953</td>
<td>0.1345</td>
<td>-2.45451</td>
<td>-5.23364</td>
</tr>
<tr>
<td>$B_3$</td>
<td>0.1842</td>
<td>0.49942</td>
<td>-0.1461</td>
<td>2.24786</td>
<td>5.38241</td>
</tr>
<tr>
<td>$B_4$</td>
<td>-</td>
<td>-0.04372</td>
<td>0.03489</td>
<td>-0.97857</td>
<td>-2.85544</td>
</tr>
<tr>
<td>$B_5$</td>
<td>-</td>
<td>-</td>
<td>-0.00266</td>
<td>0.19618</td>
<td>0.81527</td>
</tr>
<tr>
<td>$B_6$</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-0.01465</td>
<td>-0.11999</td>
</tr>
<tr>
<td>$B_7$</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.00718</td>
</tr>
</tbody>
</table>
previously observed with high-Z projectiles are also found with these low-Z ions. Exploration of these well-known effects in few electron systems can elucidate the specific influence of the K and L shell electrons of the projectile, including the subshells, on atomic substructure and ultimately the mean charge state. Many of these details are obscured in the large electron systems of high-Z projectiles through the complicated overlap of multiple shells and subshells.
Fast ions traversing a medium will undergo a series of electron capture and loss collisions. For sufficiently thick media, an ample number of collisions must occur for the ion to establish an equilibrium charge state distribution. The present work highlights and probes some of the underlying physics that are involved with charge state distributions.

Equilibrium mean charge states have been measured for 3 - 7 MeV lithium, boron, and carbon ions passing through thin carbon foils. The data have been compared to the predictions of several semi-empirical models of charge equilibrium. Of the relevant models, the Schiwietz model[53] presents the best results for low-Z ions. A compilation of experimental data for low-Z ions in the low energy regime has been used to re-parameterize a few of the charge equilibrium models.

An in-depth examination of global and local models has provided some structural insight as well as a potential source of error for semi-empirical models. The global model uses large data pools of various ions and targets to build expressions to encompass everything. By taking that approach, it develops models that present mean charge increasing smoothly with increasing energy. Charge state data confined to a smaller energy range can identify anomalies that are related to projectile shell structure. The local model perspective can be used to make,
eventually, a better and a more physics based charge state model. Another important item to be stressed is how disposable an empirical model is, as noted when comparing the linear and polynomial fits for the given low-Z ions. The current use of empirical fits is devoid of any physics and provides no scientific value to the charge exchange processes. Using that information is just one of the potential clues to developing a more meaningful model.

The large pool of carbon target data for low-Z ions have never been analyzed to look for projectile shell structure effects. Previous authors of charge state studies, like Shima, did not expect much shell structure influence with low-Z ions and therefore this structure has been generally ignored. Low-Z ions provide a clear and clean picture of electron interactions than high-Z ion systems, especially the K-L electron shell boundary see Fig 4.16. The high-Z ion systems have this structure, but it is masked due to there being more electron shell and subshells interactions.

Equilibrium charge state distributions were also measured with lithium, boron and carbon ions passing through various thin foils of carbon, aluminum, copper, silver and gold. Figure 4.7 demonstrates clearly the present state for understanding target dependence in charge exchange. Even the “best” of the empirical models, Schiwietz model, fail to predict the mean charge for the lower energy regime of the current research. This result suggests that there needs to be an extended effort in charge state experiments for various materials to gather systematic data. Only with more data can the empirical model discrepancies associated with target dependence and observation be rectified.

The current data are compared to other low-Z ions to examine the relative target dependence and shell effects for each ion species. The results show a strong
dependence of the mean charge state on target material and the charge distribution width. There are a few empirical models to include target dependence, but none of those can represent the charge state behavior in this energy regime. The typical behavior noted in the literature, i.e. the larger the target atomic number, the lower the mean charge does not seem present here. There is some indication that the mean charge as a function of target material may evolve with increasing energy to match what is noted in the literature. The use of the carbon target data pool can provide a baseline, not only for other target material, but also for more complicated electron systems.

The compilation of a large charge state data pool for low-Z, low energy ions are important. Low-Z ions are fundamental for studying projectile shell structure influences and examining the underlying behavior related to charge exchange. The long-term goal is to develop a systematic understanding of target dependence and the underlying behavior of charge exchange. In order to gather systematic target data there needs to be some development into making thin (≤ 100 µ g cm$^{-2}$) self-supporting foils of other material (tungsten, molybdenum and tantalum), which are difficult to make for this low energy experiments. For solid targets, it is trivial to attain an equilibrium charge state distribution, but there is a need to minimize energy loss to prevent any unwanted influence on the exiting charge states.

The recommendation for future charge exchange studies would be to continue to collect data at lower energies. For low-Z ions, the current experimental setup limits the energy range that can be studied. A solution would be to develop an experimental setup that uses electrostatic plates to separate the charge states and a position sensitive silicon detector to measure the entire charge state distribution. A similar setup was used by Itoh [30] and the results can be seen in Fig 5.1. The
left plot is a calculation of charge state separation as a function of distance traveled in an electric field. The right spectra comes from Itoh [30] for the same ion and energy. The obvious benefit is that the entire distribution is measured together without the need to normalize.

In addition, the types of charge exchange experiments need to go beyond the standard post-foil measurements. There needs to be a variety of experiments, beyond the scope of the current research, to try to map and categorize the charge exchange behaviors. Experiments that look into x-ray and electron emission, solid-gas effects, multiple foils at varied distances from one another, attempt non-equilibrium measurements and to test very low- and high-Z targets for comparison to literature behavior. Looking at x-ray emissions, for example, to probe the electron interactions during the collision instead of purely post-foil measurements

Figure 5.1. On the left is a plot charge state separation as a function of distance traveled. On the right is a sample spectra of counts as a function of channel number from Itoh [30].
could potentially elucidate K and L shell electron behaviors. Those efforts may provide other crucial pieces of information to developing a more physics based charge state model. The overall outcome for charge exchange studies at the Nuclear Science Laboratory were very successful and show promise for a continuing experimental program.
APPENDIX A

PHOTOCELL DETECTORS

A.1 Introduction

The basic motivation was to find an inexpensive alternative to using expensive silicon detectors for basic tasks such as monitoring ion beams, which does not require high energy resolution. One of the detriments of high quality silicon detectors is damaging the detector beyond any utility due to an accidental high dosage of radiation. There is no such worry associated with photocells, since the cost of a photocell is typically less than forty dollars in comparison to several hundred for a silicon detector. If the principle usage for a detector is to monitor fluctuations in ion beams then a low-grade, low-resolution detector makes economical sense. Other than a beam monitor, photocells can be used in a laboratory setting as an effective teaching and diagnostics tool.

A.2 Diodes

The photocell is a basic silicon photodiode that is a p-n junction, where p(positive)-type material typically has more majority carriers called “holes” and electrons with n(negative)-type material being just the opposite. The junction between the p- and n-type materials creates a neutral depleted region. When a negative bias is applied to the photocell it operates in photoconductive mode. In
this mode, the positive potential is applied to the n-type material and the negative potential to the p-type material.

The p-type section now has a negative potential forcing the “holes” in the p-type region to be pulled away from the junction and likewise with the n-type region has the positive potential forcing the electrons to cause the width of the depleted region to expand. This process effectively increases the potential barrier and increases the electrical resistance against the flow of electrons.

The depleted region is created to stand-off the reverse voltage. The width of the depleted region widens with higher voltage as does the electric field. When the electric field increases beyond a critical level, the junction breaks down and current begins to flow by avalanche breakdown. This breakdown causes drifting electrons to have secondary ionization events.

When radiation passes through the depleted region it produces electron-hole pairs along its path. The created charge moves in opposite directions and constitutes an electrical current that can be measured, see Fig A.1. The measured current is proportional to the energy deposited by the incident radiation[4, 21, 43]. The advantage is that there are very few free charge pairs and therefore with no radiation the leakage current is minimal. The brand of photodiode used was purchased from Advanced Photonix whose specifications can be seen at Fig A.2.

A.3 Experimental Setup

Before any beam tests could be performed a test chamber was assembled to determine the photocell characteristics, see Fig A.3. This test chamber consists of a pumping port, venting port, pressure gauge, and BNC’s for electrical connections. The interior houses the electrically isolated detector mounts on which
Figure A.1. This schematic shows how a photocell detects particles through interaction in the depleted region [4].

the photocells are attached. The electronics are placed in a NIM (Nuclear Instrumentation Module) crate and include a power supply, pre-amp, amplifier and an oscilloscope. The radioactive source used for all of the tests is $^{241}\text{Am}$, an alpha source.

A.4 Photocell Characteristics

Photocell characteristics were investigated in a consistent manner, where the distance between the radioactive source to the photocell, the pressure inside the test chamber and biasing procedures were the same for every photocell. Tests performed included shaping time, current-voltage behavior, counting efficiency,
count rate and repeatability. These results are summarized in Table A.1.

Generally, the first test of any detector is determining its resolution as a function of applied bias. The photocells response to applied negative bias can be seen in Fig A.4, where the optimum energy resolution is at -10V. The results were consistent whether the photocell was biased to -30V or bias was taken off. To calculate the effective resolution, E.R., the following expressions were used:

\[ E.R. = \frac{FWHM}{Centroid}, \]  \hspace{1cm} (A.1)

where FWHM is the Full Width at Half-Maximum. Resolution is more commonly given in terms of energy. For silicon type detectors an expression that can be used is
Figure A.3. a) The photocell testing chamber setup. b) A schematic of the photocell test chamber setup.
\[ FWHM(\text{keV}) = 2.35 \sqrt{E_{\alpha}(\text{keV})3.62F}, \]  
(A.2)

where \( E_{\alpha}(\text{keV}) \) is the alpha energy of the radioactive source and \( F \) is the Fano factor\[32\]. The Fano factor is

\[ F = \left( \frac{E_{\alpha}(\text{keV})}{3.62} \right) \left( \frac{E.R.}{2.35} \right)^2, \]  
(A.3)

which is a statistical noise term indicative of certain detector type materials\[32\]. The three expressions can be combined to simplify the calculation to

\[ FWHM(\text{keV}) = \sqrt{2.35E_{\alpha}(\text{keV})E.R.}. \]  
(A.4)

The optimum resolution at -10V gives an energy resolution of about 60keV.

Varying the shaping time on the amplifier affects the energy resolution of the photocell. Figure A.5 displays FWHM(keV) as a function of shaping time, where it can be seen that 2\( \mu \text{sec} \) gives the best resolution.

Leakage current is a sensitive indicator for photocells. Typically, -10V has a current draw of about 0.05 \( \mu \text{A} \) and remains constant. Figure A.6 displays current as a function of applied bias and maps the response behavior of the photocell. The displayed behavior is typical for a diode. Positive applied bias has a linear response, while a negative applied bias has a shallow slope that becomes steeper after a “knee” bend. This “knee” bend is the voltage “avalanche” breakdown mentioned earlier.

As mentioned before leakage current is a good indicator of photocell quality. There is a problem when the leakage current that is expected to remain constant does not. Does this runaway current affect measurements with a photocell. In
### TABLE A.1

PHOTOCELL RESULTS

<table>
<thead>
<tr>
<th>Objective</th>
<th>Results</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy Resolution</td>
<td>60 keV</td>
<td>$^{241}\text{Am}$ source</td>
</tr>
<tr>
<td>Shaping Time</td>
<td>2 µsec</td>
<td></td>
</tr>
<tr>
<td>Detector Efficiency</td>
<td>98</td>
<td>Compared to Si detector (100)</td>
</tr>
<tr>
<td>Repeatability</td>
<td>-</td>
<td>Centroid: 612 ± 0.66 channels</td>
</tr>
<tr>
<td></td>
<td></td>
<td>FWHM: 59 ± 6.24 keV</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Rate: 28.3 ± 0.87 Hz</td>
</tr>
</tbody>
</table>

fact it does, making none of the measurements consistent and repeatable.

A.4.1 Counting Efficiency

Since the purpose of the photocell is to be used as a beam monitor, resolution is not of interest, but counting efficiency is very important. The definition of counting efficiency is what the detector sees from the radioactive source as compared to the radioactive source output. The rate of the source, $Rate_{\text{source}}$, is calculated from the initial activity of the source and its date of birth. The source used in this work decayed at a rate of 3636 decays per second. The next step in calculating the efficiency is the solid angle, $\Omega$, of the detector, which is defined as

$$\Omega = \frac{A}{d^2}, \quad (A.5)$$

where $A$ is the area of the detector and $d$ is the distance of the source to the
Figure A.4. The resolution of a photocell as a function as the applied negative bias. The closed symbols in data points indicate that bias was increased from zero to -30V and the open symbols is where bias was taken down from -30V to zero.

detector. The expected rate of the detector can be related to the solid angle and source rate as

$$Rate_{expected} = \left( \frac{\Omega}{4\pi} \right) Rate_{source}. \quad (A.6)$$

A standard silicon detector, not a photocell, with a 3mm collimator and a 3 inch distance from the source gives an experimental rate of 0.352Hz for an efficiency of 100%. A photocell with the same collimation and distance gives an experimental rate of 0.358Hz for an efficiency of 98%.
Figure A.5. This plot displays the FWHM as a function of shaping time.

A.4.2 Irradiation Tests

The second phase of testing photocells is to irradiate them with ions. For these irradiation tests, a 21 MeV oxygen beam is used and the photocell will see either direct or scattered beam. The photocells were exposed to a count rate from 1 to 67kHz of beam. The standard spectra of a new photocell with an $^{241}\text{Am}$ source looks like Fig A.7a. An irradiated photocell clearly shows damage as indicated by the jagged spectra seen in Fig A.7b. The result of radiation damage is also quantitatively clear when returning the photocells to the test chamber and measuring their resolution. The details are summarized in Table A.2. It should be noted that these tests were a bit extreme and should be thought as a durability test. If these photocells were treated with the same precautions as a standard silicon detector then the damage should be minimal. The overall lifetime of a
Figure A.6. This plot displays the current draw as a function of the applied bias for a photocell.

A.5 Conclusion

Ultimately the photocell part of the project was abandoned due to the unreliability of the manufacturer to supply photocells that behave like the aforementioned sections have described. Previous work completed at Yale University by Thomas [74] suggests that the photocell approach could have been successful.
<table>
<thead>
<tr>
<th>Irradiation</th>
<th>Applied</th>
<th>Leakage</th>
<th>Resolution</th>
<th>Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time (min)</td>
<td>Bias (V)</td>
<td>Current (µ A)</td>
<td>(keV)</td>
<td>(%)</td>
</tr>
<tr>
<td>0 (chamber)</td>
<td>-10</td>
<td>0.05</td>
<td>60</td>
<td>98</td>
</tr>
<tr>
<td>66 (direct)</td>
<td>-10</td>
<td>0.94</td>
<td>366</td>
<td>NA</td>
</tr>
<tr>
<td>120 (direct)</td>
<td>-10</td>
<td>0.94</td>
<td>564</td>
<td>75</td>
</tr>
<tr>
<td>66 (scattered)</td>
<td>-10</td>
<td>0.9</td>
<td>187</td>
<td>80</td>
</tr>
</tbody>
</table>

Figure A.7. a) This plot displays a spectra of a new photocell and b) an irradiated photocell with counts as a function of channel number.
BIBLIOGRAPHY


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