CHARGE STATE DISTRIBUTIONS

IN MOLECULAR DISSOCIATION

DISSERTATION

Presented to the Graduate Committee
of the Department of Physics
University of North Texas

For the Degree of

DOCTOR OF PHILOSOPHY

By

Steven N. Renfrow, B.S., M.S.

Denton, TX

December, 1998
Experiments were conducted to measure the charge state distributions resulting from molecular breakup and to compare them to that obtained for atomic ions at the same velocity. Elements (N, Mg, Mn, and Zn) chosen for study are those of interest to Trace Element Accelerator Mass Spectrometry (TEAMS) because they do not readily form negative ions which are necessary for injection into the tandem accelerator. A final energy of 1.4 MeV to 1.7 MeV was used in this experiment. AlO was chosen as an additional molecule to be studied because it was known to be more tightly bound than the SiX molecules. The SiX⁺ or AlO⁺ molecules were selected after acceleration and passed into a 30° beam line for breakup and analysis of the charge states of the fragments. The SiX⁺ or AlO⁺ molecule ions were passed through an N₂ gas cell to simulate the conditions in the terminal of the accelerator during molecular breakup. The molecular fragment ions and the atomic ions at the same velocity reached equilibrium charge state distributions in the N₂ gas with an approximate target thickness of 0.43 μg/cm². Atomic ions of the fragments were produced in the terminal by electron stripping, e.g. Si⁺, N⁺, Mg⁺, Mn⁺, Zn⁺, Al⁺, and O⁺. The terminal voltage was chosen so that the singly charged ions were accelerated out of the accelerator at the respective fragment ion velocity produced in the molecular breakup. Calculations of charge state distributions resulting from molecular ion beam breakup by Cooney exist for the di-atomic molecule N₂. The measured charge state fractions for SiX molecular fragments were found essentially the same as that measured for the atomic ions at the same velocity. Both charge state distributions peak at
the same charge state and have approximately the same magnitude. Equilibrium fraction ratios with Cooney’s fitting parameters are in agreement with the Si data and the general trend of the other fragments is represented. The present work provides charge state fractions that may be used to generate TEAMS relative sensitivity factors for impurities in semiconductor materials.
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CHAPTER I

INTRODUCTION

The study of ionic charge states dates back to the first of this century, and even after 90 years it continues to be an important topic. Investigations of stopping powers of ions and molecules is very important to the implantation industry as well as others. Scientists working on fusion research must have accurate data on charge changing cross sections. These data are necessary in the design of fusion chambers to prevent performance degradation of the plasma. Accelerator designers as well as users need charge state data to optimize not only ion beam production but also the acceleration and transport of the beams. Other research areas where these data are important include: beam-foil spectroscopy, hyperfine interactions, and atomic physics experiments involving few-electron systems. Atmospheric effects following above ground nuclear blasts also are strongly dependent on charge state and charge changing cross sections. One of the most sensitive analytic techniques that is available today is Accelerator Mass Spectrometry (AMS). At the University of North Texas a trace-element AMS (TEAMS) system has been developed as a complement to secondary ion mass spectrometry (SIMS). Accurate charge state data for both ions and molecules is essential for the high sensitivities that are possible with these techniques.
The first studies of charge states that involved the neutralization of ions were conducted in 1911 by Wien. The discovery of fission prompted further research. These investigations included the need for an understanding of fast fission fragments and their stopping powers derived from charge states. Many studies have added to the overall understanding of charge state distributions. In 1940, Lamb calculated theoretical estimates for the average charge states from energetic considerations. Using the assumptions that electron velocities in the ion and atom are given by a simplified Thomas-Fermi model, Bohr derived estimates of the capture and loss cross sections of charged particles in matter in 1940. Bohr also simplified the calculation further by assuming that the ion returns to its ground state between collisions. Knipp and Teller in 1941 showed the average charge state depends primarily on the ratio of electronic to ionic velocities. In 1954, Bohr and Lindhard took into account the residual excitation of the ion, to determine the charge change mechanism. The field progressed significantly with the availability of particle accelerators.

In the late 1950's Barnett and his colleagues at Oak Ridge National Laboratory and Fogel's group in the U.S.S.R. were involved in early experiments measuring charge changing collisions. In the 1960's, heavier ions started to be studied by Betz et al. in Heidelberg and by Nikolaev in Moscow who also published a critical review of cross section data. In the early 1970's, Datz et al., Wittkower, and Ryding published heavy ion equilibrium fractions. Betz and co-workers also published a comprehensive review of experimental and theoretical calculations in 1972, 1973, and 1980. Hofmann et al. from Zurich has published a rather extensive set of measurements on C, Si, and Cl.
As was mentioned above these data are of direct interest to the AMS community for increased transmission and hence better sensitivity. Table I-1 is a list of review articles and data collections on ionic charge states with a summary of the contained data from Betz’s chapter on Heavy Ion Charge States.

**Table I-1: Review articles and data collections on ionic charge states.**

<table>
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<th>Authors</th>
<th>Year</th>
<th>Summary</th>
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<tr>
<td>Bohr</td>
<td>1948</td>
<td>Theory of atomic collisions; excitation, ionization, $\sigma_c$ and $\sigma_f$ for light and heavy ions.</td>
</tr>
<tr>
<td>Allison</td>
<td>1958</td>
<td>Experimental results on $\sigma_{ij}$ and $F(q)$ for H and He above 0.2 keV; relations $\sigma_{ij} \leftrightarrow F(q)$; experimental methods</td>
</tr>
<tr>
<td>Allison and Garcia-Munoz</td>
<td>1962</td>
<td>Experimental results on $\sigma_{ij}$ and $F(q)$ for H, He, Li, and some heavy ions</td>
</tr>
<tr>
<td>Nikolaev</td>
<td>1965</td>
<td>Review of experimental and theoretical results on $\sigma_{ij}$, $F(q)$, and $\bar{q}$ for fast light ions; experimental methods</td>
</tr>
<tr>
<td>McDowell and Coleman</td>
<td>1970</td>
<td>Theory of ion-atom collisions; excitation, ionization, $\sigma_c$ in various approximations</td>
</tr>
<tr>
<td>Lo and Fite</td>
<td>1970</td>
<td>Graphical compilation of experimental results on $\sigma_{ij}$ in N$_2$, O, O$_2$, Ne, Ar; 4 $\leq Z \leq$ 92; $v \leq v_0$; $i = 0^+$, 1+, 2+</td>
</tr>
<tr>
<td>Datz et al.</td>
<td>1971</td>
<td>Experimental results on $Y_q(x)$ and $F(q)$ for 15-140 MeV bromine and 15-162 MeV iodine in gases and solids</td>
</tr>
<tr>
<td>Mapleton</td>
<td>1972</td>
<td>Theory of charge exchange; various approximations; comparison with experimental cross section data</td>
</tr>
<tr>
<td>Betz</td>
<td>1972</td>
<td>Review of experimental methods, $\sigma_{ij}$, $F(q)$, $\bar{q}$, d; $Z &gt; 16$; relations $\sigma_{ij} \leftrightarrow F(q)$; empirical methods, density effects</td>
</tr>
<tr>
<td>Tawara and Russek</td>
<td>1973</td>
<td>Review of $\sigma_{ij}$ for H; experimental results; theory; experimental techniques</td>
</tr>
<tr>
<td>Wittkower and Betz</td>
<td>1973</td>
<td>Tabular compilation of 2525 experimental $F(q)$, $\bar{q}$, d for 26 ions $Z &gt; 2$, in 50 gases and solids</td>
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<tr>
<td>Dehmel et al.</td>
<td>1973</td>
<td>Bibliography of experimental results on $\sigma_f$ (1950-1970);</td>
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<td>Bayfield</td>
<td>1975</td>
<td>Survey of $\sigma_c$ in atomic collisions; theory and experiment</td>
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<tr>
<td>Basu et al.</td>
<td>1978</td>
<td>Theoretical review of $\sigma_c$ in atomic collisions</td>
</tr>
<tr>
<td>Belkic et al.</td>
<td>1979</td>
<td>Theoretical review of $\sigma_c$ by bare nuclei in H and heavier targets; various approximations</td>
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Studies of molecular beams and the subsequent molecular dissociation have been ongoing since the mid 1970's. When molecular projectiles are incident at MeV energies upon a foil or gas target, most of the binding electrons of the projectile molecule are stripped off within the first few μg/cm² or monolayers of the target. The resulting charged nuclei rapidly separate due to their mutual Coulomb repulsion, converting their initial electrostatic potential energy into kinetic energy of the relative motion. This dissociation process has been termed a Coulomb explosion.\textsuperscript{37,38,39} Because of this fact,
fast molecular-ion beams provide a unique source of energetic projectile nuclei that are correlated both spatially and temporally. The recognition of this feature has prompted several studies of various aspects of the interactions of these ions with matter. In particular, the foil-induced dissociation of fast molecular ions has been used by several groups to provide new information about molecular-ion structures, the charge states of fast ions inside and outside solids, the interactions of such ions with the solid, as well as other atomic collision phenomena. More recent experiments with molecular beams include measuring lifetimes of metastable highly charged molecular ions and stopping powers of molecules in solids.

The two kinds of targets studied are gaseous and solid. On the average, the gaseous target gives a lower average charge state than a solid target. In addition, molecules give a lower average charge state compared to ions at the same velocity. It has been shown when dilute gas targets are used, large impact parameter collision processes dominate the excitation process and a much lower probability for multi-electron ionization results. Instead, electrons are promoted to dissociative states that can produce charged as well as neutral fragments. In foil targets, the molecule is highly ionized exiting the target, which leads to fewer neutrals after the Coulomb explosion. Cooney et al. published a semi-empirical model that related charge state distributions of molecular ions to the equilibrium distribution for monatomic ion beams in foils. Ben-Hamu, et al. published a simple model for molecular charge states in their paper on the energy loss of clusters in solids. They presented the united atomic model which assumes the molecule has the same charge state distribution as the monatomic ion of the same
mass, with the condition that the inter-nuclear distances are smaller than electron screening length in the target.

In general, the probability of finding an ion in a final charge state $q'$ after a collision with a target depends on five variables. These five variables include the atomic number of the incident ion $Z_i$, the velocity $v_i$, the initial charge state of the ion or molecule $q$, the target atomic number $Z_2$, and the density of the target $\rho$. Particle accelerators allow the study of collisions with the controllable parameters of $Z_i$, $v_i$, and $q$. The use of a gas cell allows the control of $Z_2$ and $\rho$.

The purpose of the present investigation is to measure the charge state distributions after molecular dissociation for a variety of atoms that do not form negative ions. This research is of direct interest in quantifying charge-state fractionization for use in Trace Element Accelerator Mass Spectrometry (TEAMS) and for purely fundamental investigation. The studies in the research will be used to accurately measure the impurity levels of atoms that do not form negative ions and atoms that form more abundant negative molecules. The reason for this is that negative ions or molecules are needed for injection into the tandem accelerator. Furthermore, the data on molecular dissociation can be used to improve Secondary Ion Mass Spectrometry's (SIMS) Relative Sensitivity Factors (RSF), and hence molecular dissociation data is needed. Similarly, the ion implantation industry needs data on the charge state distributions of BF and $B_2F$ for better understanding of the energy spread of the implantation beam.

For the studies in this thesis, charge state measurement of O, Al, and Si, were compared to previously measured values and theory to confirm the validity of the data.
taking and data processing procedures. Then, charge state measurements of molecular
dissociation of AlO, SiN, SiMg, SiMn, SiCa, and SiZn were performed and compared to
models for molecular dissociation charge states. The source of negative ions by Cs
sputtering (SNICS) source was used to produce the desired ions and molecules, which
were then stripped to the q = 1+ charge state at the tandem's terminal gas stripper. Next,
the High Voltage Engineering Corp. (HVEC) magnet deflected the desired molecular ion
beam into the 30° atomic beam line which has the nitrogen gas, electron stripping cell.
This allowed analysis of the different charge states with a 30° electrostatic analyzer ESA
into particle detectors.

In the dissertation, chapter II will cover theoretical models and semi-empirical
models. Chapter III will detail a description of the experimental setup and equipment.
Chapter IV will discuss the measurements made and data will be presented. Chapter V
will discuss the results of the data analysis. Chapter VI will discuss the conclusions
drawn from the data.
Atomic beams are most likely ionized in dilute gases by a succession of single electron removals, although multiple electron loss becomes more likely for higher Z_i. The case of molecular beams is more complex. For molecular ions, four basic processes can occur when passing through a target: electron loss, electron capture, electron excitation, or dissociation.

Since molecular ions are weakly bonded, with only a few eV, they dissociate when colliding with targets thicker than a few monolayers. Y.D. Kim designed a differentially pumped gas cell for molecular lifetime studies. The gas cell was used in this study to strip electrons, producing higher charge molecules and molecular dissociation.

An ion can have four types of charge changing during a collision. (1) Coulomb capture occurs when the ion captures one or more electrons from a target atom into the ground or excited states. (2) Radiative capture occurs when the ion captures an electron into a ground or excited state accompanied by the emission of a photon. (3) Excitation to the continuum happens when the ion losses one or more electrons, leaving the ion in either its ground or excited states. (4) Excitation to meta-stable bound states that later decays by the emission of an electron is the fourth type.
Charge state calculations fall into two categories, non-equilibrium and equilibrium. Non-equilibrium charge states involve a set of linear coupled differential equations.

\[
\frac{dY_q(x)}{dx} = \sum_{q' \neq q} [\sigma_r(q', q) Y_{q'}(x) - \sigma_c(q, q') Y_q(x)]
\]

Where \( q \), the initial charge state, and \( q' \), the final charge state, extends over the range of possible charge states. Also \( \sigma_c(q, q') \) is the cross section for single electron capture, \( \sigma_r(q', q) \) is the cross section for single electron loss, \( x \) is the target thickness and \( Y_q(x) \) is the non-equilibrium charge state fraction.

The charge state of a fast ion interacting with matter fluctuates as a result of electron loss and capture in collisions with the stationary atoms of the target. Charge state equilibrium is reached when a thicker target does not change the final charge state fractions, \( F(q) \) because the production of \( e^- \) vacancies and \( e^- \) filling rates are equal. They are independent of the initial distribution and reflect the ratios of the cross sections. This process can be considered in a statistical manner and in many cases the distribution of charge states is close to a gaussian shape. If one assumes single electron capture or loss, then the ratio becomes,

\[
\frac{F(q)}{F(q + 1)} = \frac{\sigma_c(q + 1)}{\sigma_r(q)}.
\]

And, if the cross sections are known \( \sigma(q, q') \), \( F(q) \) can be calculated without integration.

The cross sections can be measured using a linear growth method. Ions from a single
charge state \( q_i \) are selected and pass through a gas target of sufficiently small thickness so that single collisions prevail. Variation of the target thickness then results in an approximately linear growth of neighboring charge fractions, \( Y_{q'}(x) = \sigma(q_i, q)x \).

Other experimental methods have also been used to measure charge changing cross sections: beam-foil emissions, ion-induced x-rays, and ion-induced Auger electrons.

Electron capture theory for a simple system involves using the Oppenheimer, Brinkman and Kramer's theory as approximated by Nikolaev\(^{15}\) (OBKN) where \( n_i \) and \( n_f \) are the quantum number for the shells, \( v_i \) is the projectile velocity, \( v_{if} \) and \( v_{2i} \) are the orbital velocities of the electron on the projectile and target atom, respectively.

\[
\sigma_{OBKN}^{q} = \left( \frac{2\pi}{5} \right)^{2} \left( \frac{n_i n_f}{v_i} \right)^{2} \left( \frac{v_{if}}{v_{2i}} \right)^{5} \xi_{q}^{10} (\theta) \frac{\phi_{q} (1 - \theta) \xi_{q}^{2} (\theta)}{[1 + (1 - \theta) \xi_{q}^{2} (\theta)]^{3}} \tag{II-3}
\]

The function \( \xi_{q} (\theta) \) is a dimensionless velocity parameter given by the equation

\[
\xi_{q} (\theta) = \frac{v_{2i}}{\sqrt{v_{if}^2 + q_{q}^2 (\theta)}} \tag{II-4}
\]

The quantity \( q_{q} (\theta_i) \) approximates the minimum momentum transferred by the capture process, given as
\[
q_i (\theta_i) = \frac{1}{2} \left[ v_i + \frac{v_i^2 \theta_i - v_i^2_{i_f}}{v_i} \right].
\]

\(\theta_i\) is the reduced binding energy with \(R_y\) is the Rydberg energy and \(\hbar \omega_{2i}\) the binding energy for an electron in state \(i\)

\[
\theta_i = \frac{n_{2i}^2 \hbar \omega_{2i}}{Z_{2i}^2 R_y}.
\]

In addition, the function \(\phi_4(t)\) can be approximated for \(t < 3\) by the equation

\[
\phi_4(t) = \frac{1}{1 + 0.3t}.
\]

However, this theory ignores the target-ion interaction after capture, so that its predictions are in general too large. Lapicki and McDaniel\(^{52}\) published the ECPSSR theory, whose predictions are much closer to the data. The ECPSSR theory with direct ionization was implemented by incorporating certain modifications to the appropriate parameters of the first Born theory. Basbas, Brandt, and Laubert,\(^{53}\) Brandt and Lapicki,\(^{54}\) have included the perturbed stationary states (PSS) in considering the polarization and binding energy effects and relativistic (R) wave functions for the target electron while the projectile is passing near the atomic electron. These calculations also accounted for the energy loss (E) and the Coulomb deflection (C) of the projectile as it traverses the target.
The increase in the binding energy of the target inner-shell electrons due to the penetration of the projectile inside the inner-shell during the collision reduces the probability for ionization. The polarization of the inner-shell electron wave function due to the projectile, which keeps the projectile and target electron in contact longer, increases the ionization probability. The deflection of the projectile by the Coulomb field of the target atom increases the distance from the projectile to the target electron and therefore reduces the ionization probability. The energy loss of the projectile reduces the ionization probability if the $v_1 \ll v_{25}$. The relativistic effect of the target electron increases the mass of the electron and reduces the ionization probability.

For complex systems, several methods are used: screening parameters, Eikonal approximation, *Ab initio* calculations, and Monte Carlo simulations.

Bohr and Lindhard\(^\text{11}\) proposed electron loss cross sections caused by excitation by light particles and projectile ionization are calculated from,

\[
\sigma_i = \pi a_o^2 \frac{Z_i^{2/3} Z_T^{4/3}}{q^2} \left( \frac{v}{v_o} \right)^2
\]

where $Z$ is the projectile atomic number, $Z_T$ is the target atomic number, $a_o$ and $v_o$ is the Bohr radius and velocity, $q$ is the in projectile charge, and $v$ is the projectile’s velocity.

Again, ECPSSR theory has made improvements on ionization predictions.

Atomic ion charge state models have been refined over the years. Early programs like STRIP, which uses semi-empirical fits to predict charge states, have been improved.
The program ETACHA uses theoretical cross sections for electron capture, ionization, and excitation, and scaled radiative and Auger decay rates. In addition, work has been done with Monte Carlo computer codes and data tables of known electron capture and loss cross sections to predict final charge state distributions.

However, this kind of data has not been generally available for molecular ions. P.J. Cooney, et al.\textsuperscript{49} presented a model for predicting molecular charge state distributions. In these studies the Argonne National Laboratory’s 4.5 Dynamitron was used to produce 2.1 MeV N\textsuperscript{+} and 4.2 MeV N\textsubscript{2}\textsuperscript{+} and were measured for energy, angular, and charge state distributions in a range of carbon foils of thickness 75 to 780 Å.

Two observation of the data where made:

1. For the case of molecular ion impact, there is a total shift of the charge state distributions toward lower charges when compared to those distributions measured for atomic ion impact at the same velocity.

2. Although for atomic ion impact, the yields of the dominant charges states are equilibrated in the thinnest target, the case of molecular ion impact shows apparent non-equilibrium even in the thickest targets.

The first observation is not from the redistribution of valence electrons of the exiting fragments because the charge is conserved which would not produce a shift but a narrowing of the peak. The shift is caused by an enhanced capture of target electrons. Although, inter-atom exchange effects do play a role, the yield of highly charged ions are far from the equilibrium mean. The second result implies that the cross section for
capture and loss of target electrons is dependent on the internuclear separation of the
dissociation fragments.

The effective capture and loss cross sections were extracted from the data. By
assuming single electron charge changing this simplified the ratio of capture and loss
cross sections to equation II-2. In order to reduce the number of free parameters required
to describe the equilibrium charge state distributions, two more simplifying assumptions
were made for the cross sections:

1. The capture cross sections $\sigma_c$ are assumed to be proportional to some power (p) of
the charge state $q$.

   $$\sigma_c(q) = C \sigma_0 q^p$$  \hspace{1cm} \text{(II-9)}

2. The loss cross sections $\sigma_l$ are assumed to be proportional to the number of L-shell
electrons available for each charge state.

   $$\sigma_l(q) = \sigma_0 (5 - q)$$  \hspace{1cm} \text{(II-10)}

Where $C$ and $\sigma_0$ are constants of proportionality. Using these assumptions,
Cooney et al. were able to extract the power $p$ and constant $C$ by best fitting the data.
The values of the $p=2.8$ and $C=0.05$ were determined and are within the range of
predicted values for atomic ions determined from the OBKN formula and scaled
ionization data.
For impact of molecular ions, a target thickness parameter must be introduced, to account for the increasing equilibrium charge state for thicker targets. For computing the capture cross section, they assumed an effective charge \( q^{\text{eff}} \) given by:

\[
q^{\text{eff}} = q + x\langle q \rangle.
\]

where, \( \langle q \rangle \) is the measured mean exit charge and \( x \) is a target thickness perturbation parameter to be determined. Substituting \( q^{\text{eff}} \) for \( q \) in equation II-2 and using the constants \( p \) and \( C \) from the fit atomic data, \( x \) is determined for each target thickness. The authors then ascribe a physical interpretation to the \( x \) parameter by noting for internuclear separation \( R_{ex} \) large compared to the L-shell radius \( r_L \) and write the two-center potential acting on an electron at \( r_L \) as:

\[
\frac{q}{r_L} + \frac{\langle q \rangle}{R_{ex}} = \frac{\left[ q + \frac{r_L}{R_{ex}} \langle q \rangle \right]}{r_L}.
\]

Thus, \( x \) is identified as the ratio \( r_L / R_{ex} \) and shows good agreement with both the trend and magnitude of the fitted data for \( x \). Figure II-1 taken from McDaniel et al.\textsuperscript{55} shows that target thickness parameters are also important for ionization cross section measurements with ions. Note that in the single collision realm (\(< 4\mu\text{g/cm}^2\)) in the figure,
the Electron Capture is dominant for high charge states of Si\textsuperscript{n+} bombarding thin Titanium targets.

The above theoretical model will be used in analysis of the data from this project, relating the charge state distributions measured for molecular ion impact to the equilibrium distribution measured for the case of a atomic ion beam impact. Data will be taken for the molecules and the atomic components at the same impact energy.
Figure II-1: Effective titanium cross sections for target K-shell X-ray production by \( Si^{q+} \) ions as a function of titanium target thickness.
CHAPTER III

EXPERIMENTAL APPARATUS AND TECHNIQUES

In the Ion Beam Modification and Analysis Laboratory (IBMAL) at the University of North Texas, there is a variety of techniques and equipment available to perform fundamental atomic physics studies and material analysis. A full description of the laboratory is in Appendix A.

The 3 MV Tandem Accelerator

IBMAL has a National Electrostatics Corporation\textsuperscript{56} (NEC) 9SDH-2 3 MV Tandem Pelletron which has the ability to accelerate a wide assortment of atomic or molecular ions. The tandem has three sources of negative ions, as required for injection. The source of negative ions by Cs sputtering (SNICS)\textsuperscript{51} is the most heavily used source because of the large currents and variety of ions it can produce, it is being used in the present experiment. This is a high intensity source which includes a spherical tantalum ionizer and a spray system for Cs vapor. The SNICS uses accelerated positive cesium ions striking a cold cathode to produce a negative ion beam of the cathode material. The Alphatross is a RF ion source used mainly for H\textsuperscript{+} and He\textsuperscript{+} ions. The CHIMERA\textsuperscript{57} is the ultra-clean Cs sputter source for the TEAMS system. For the SNICS and CHIMERA sources, the sample to be measured is sputtered and the negative ions or molecules are extracted and injected into the tandem. From these sources, the majority of the material
ejected from the sputtered sample is neutral; of the ions produced, small portions (~1-2%) are of singly charged ions and clusters.

Once the negatively charged ion beam is produced at an injection voltage ($V_i$) of 15-70 kV, it is bent into the pre-analysis beam line by a source selector magnet. In addition, the CHIMERA source has a 45° electrostatic analyzer (ESA) to ensure no ions sputtered from beam line components are injected. The negatively charged ion beam is momentum/charge analyzed before injection into the tandem by a double focusing 90° magnet. The ion beam is then accelerated to the positive terminal, which is at a voltage ($V_T$) from 0.15 MV to 3 MV. At the terminal, the ions reach an energy of $eV_T + eV_f$.

There the ion beam interacts with a nitrogen gas target in a 60 cm gas cell or a carbon foil target. The ions lose electrons in the collision process and depending on the energy and type of ion, several charge states are produced $q = 0, +1, +2...$ which are accelerated out of the tandem. Therefore, the exit energy of the ions depends on their charge state ($q$):

$$E = e(V_f + V_T) + qeV_T \quad III-1$$

If a molecule is injected into the tandem, it has the likelihood of breaking apart during the terminal stripping. The fragments may be broken into a number of different charge states. This introduces another factor into the final energy equation: the ratio of the mass of interest ($m$) to the molecular mass ($m_0$) injected.

$$E = \left( \frac{m}{m_0} \right) e(V_f + V_T) + qeV_T \quad III-2$$
Computer Control of Accelerator, Ion Sources, Magnets, ESA, and Data Acquisition

Computer control of the tandem accelerator and sub-systems is required for the data taking procedures. Automated magnetic and electrostatic scanning of the beam into Faraday cups by the Tandem Accelerator Control System (TASC) computer maximizes the beam current. This scanning occurs with the 30° and 90° injector magnets, the HVEC magnet and the ESA. shows the computer schematic used in this experiment.

Measurements for this experiment will be made in two ways. For molecular beams with high currents (>0.1 nA) into the 30° beam line, the Faraday cup past the 3° ESA was used. For lower current measurements, the surface-barrier particle detector was used and the data sent to the Texas Instruments Data Acquisition System (TIDAS). The system will rely on calibration of the 90° injector magnet, HVEC magnet and 3° ESA. The electronics of the system consist of two computers, controllable power supplies, GPIB meter, amplifier, and a CAMAC crate with A/D converter and multi-channel analyzer.
Figure III-1: Tandem Accelerator Control and Data Acquisition Electronics Block Diagram.
Experimental Setups used by Other Researchers for Molecular Measurements

Early molecular work at Argonne National Laboratory involved measurements of particle distribution areas to determine the separation of the diatomic molecular ions after Coulomb exploding through collisions with dilute gases and foils. The apparatus used at Argonne for this type of measurement used a magnetically analyzed molecular-ion beam from a 4 MV Dynamitron accelerator which were collimated to a maximum angular divergence of 0.09 mrad at the target position. A set of pre-deflectors plates permitted electrostatic deflection of the beam incident on the target. Similarly, a set of post-deflectors was used to deflect charged particles emerging from the target. Charge particles entering the electrostatic analyzer were energy-analyzed with a relative resolution of \(3 \times 10^{-4}\). The angular acceptance of the analyzer was 0.11 mrad.

Distributions in angle and energy were made for particles emerging from the target by varying the voltages on the pre- and post-deflectors in conjunction with that on the analyzer. Neutrals were energy-analyzed by first stripping them in 100 Å carbon foil located just ahead of the analyzer. A gas target could also be used in lieu of foils. For coincidence measurements, there were two computer controlled movable detectors locate 16 feet upstream from the electrostatic analyzer. From these data, molecular bond lengths and orientations were calculated.

The Weizmann Institute in Israel currently uses a tandem to accelerate the molecular beam; then a Nd-YAG laser neutralizes the molecular ions by a photo-detachment process at the terminal. The charged beam is deflected away and the neutral
beam interacts with the target. The collision products are measured by time of flight for energy and a phosphor screen for spatial information.

Both single ended and tandem accelerators may be used to study molecules. All beam lines use a collimated beam, steerers, target, and magnetic or electrostatic analysis into a detector system.
Figure III-2: Experiment beam line at the IBMAL. Particle Detector (PD) and Annular Particle Detector (APD).
Accelerator Experiment Setup

In this experiment, shows the beam line setup used for these measurements. The molecule of interest was produced in the SNICS source from in-house produced cathodes. Table III-1 list the materials used for each cathode and current injected into the 30° atomic line as measured at cup #1. The materials were ground into a fine power and pressed into copper cathodes. Each cathode lasted for the entire run of 1 to 3 days.

Table III-1: Cathodes used in SNICS source.

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Cathode</th>
<th>30° Cup #1 Current</th>
</tr>
</thead>
<tbody>
<tr>
<td>AlO</td>
<td>Al₂O₃</td>
<td>20 nA</td>
</tr>
<tr>
<td>SiN</td>
<td>Si₃N₄+Ag</td>
<td>&lt;5 pA</td>
</tr>
<tr>
<td>SiZn</td>
<td>Si+Zn+Ag</td>
<td>&lt;5 pA</td>
</tr>
<tr>
<td>SiCa</td>
<td>Si₉Ca₂</td>
<td>10 pA</td>
</tr>
<tr>
<td>SiMg</td>
<td>Si+Mg+Ag</td>
<td>20 pA</td>
</tr>
<tr>
<td>SiMn</td>
<td>Si₂₆Mn₁₅</td>
<td>50 pA</td>
</tr>
</tbody>
</table>

The ion beam was \( mv/q \) analyzed with the 30° and 90° magnets and the desired mass was injected into the accelerator. The beam passed through a set of electrostatic steerers and an einzel lens, which aligns and focuses the beam on to the terminal electron stripper canal.

The ion beam was accelerated to the terminal and stripped to the \( q = +1 \) state in the 60 cm nitrogen gas cell, which had a minimum of nitrogen gas as to allow as many molecules to survive as possible. Negative atomic ions injected into the accelerator normally lose one or more electrons due to collisions with the molecules of the \( \text{N}_2 \) gas run through the stripper channel. The \( \text{N}_2 \) pressure within the channel is of significant
importance for the intensity of the accelerated ions. In case of molecular ions it was found that the N\textsubscript{2} pressure has to be roughly one order of magnitude smaller than for atomic ions to get any significant beam intensity. This is quite obvious because a large N\textsubscript{2} density increases the number of break up processes and internal excitations of the molecule which in turn can lead to disintegration processes. Moreover, a large N\textsubscript{2} density increases the number of events where several atomic constituents of a molecule become positively charged with the result that a Coulomb explosion takes place, decreasing the molecular beam intensity. The low N\textsubscript{2} density necessary for a molecular beam represents a N\textsubscript{2} thickness that is much smaller than the thickness necessary to establish a charge state equilibrium for the molecular constituents. A consequence is that events where more than two electrons are stripped in a molecule-N\textsubscript{2} collision are rather scarce. This means that neutral and singly charged positive molecular ions are generated in the stripper channel in an overwhelming majority. The maximum cluster energy is thus limited to 2eV\textsubscript{T} + the injection energy (eV\textsubscript{i}), where V\textsubscript{T} is the terminal voltage. Only in the case of AlO were a measurable amount of doubly charged molecular ions formed with the maximum energy of 3eV\textsubscript{T}+eV\textsubscript{i}. Optimum molecular beam intensities were obtained when the N\textsubscript{2} gas inlet to the stripper channel was almost closed so that a pressure of 6.2 x 10\textsuperscript{-7} torr was measured at the high energy end of the accelerator. It is estimated that the actual pressure within the terminal is only insignificantly higher in this case. This pressure has to be compared with the base pressure of several times 10\textsuperscript{-7} torr existing in the accelerator tube and the beam lines. It should be noted that the stripping processes for the molecular ions occur predominantly within the terminal. This
conclusion can be drawn from the fact that a magnetic scan of the beam produced no satellite peaks in front of the molecular peak.

After exiting the accelerator, the molecular beam was focused with the electrostatic quadrupole. The $q=+1$ molecular beam was magnetically $mv/q$ analyzed into the $30^\circ$ atomic beam line by the HVEC magnet to separate it from any atomic beams. The molecular beam was then passed through a second gas cell (Figure III-3) containing nitrogen, which is pressure controlled by feedback from a capacitance manometer. The beam then passes through a normalization detector as shown in Figure III-4, which consists of a 9 mm aperture followed by a 80 lines/inch gold mesh which has 85% transmission. A small portion of the beam is forward scattered off the gold into an annular particle detector. The rest of the beam pass through the detector’s 13 mm hole. Each set of data was taken for the same number of normalization counts. The resultant molecular fragments were analyzed with the $3^\circ$ ESA for their energy/charge. The resulting elemental fragment ions were measured in either a Faraday cup approximately 2 m from the ESA or a particle detector. The $3^\circ$ ESA must be scanned under computer control so the data can be acquired by the TIDAS system. At that point, the data was processed and the peaks integrated yields measured using PeakFit.
Figure III-3: Atomic beam line $N_2$ gas cell setup.

**Figure Description:**
- **GB Pressure Meter**
- **MKS Pressure Controller**
- **Capacitance Manometer**
- **$N_2$**
- **Valve**
- **Entrance and Exit diameter = 8 mm**
- **14 cm**
- **Effective length = 14.4 cm**

**Technical Specifications:**
- Entrance and Exit diameter: 8 mm
- Effective length: 14.4 cm
Figure III-4: Normalization detector and electronic setup.
A particle detector is also located 0.5 m from the exit of the 3° ESA; it was used to measure neutrals by setting the ESA to 30KV which deflects all charged particles into the ESA plate. Another particle detector is 3 m from the exit of the 3° ESA to measure the charged fragments as the ESA is scanned. Any negatively charged breakups can also be detected by reversing the polarity of the ESA and scanning into the final particle detector. Improvements were made to the tandem accelerator’s terminal gas stripper: a MKS 622 Baratron capacitance manometer, with a measurement range of 0.01 to 100 torr has been installed to monitor the stripper gas pressure. Custom electronics were installed to bring the pressure signal down from the terminal voltage and to shield it from sparks. The Baratron has a fiber optic relay to ground potential for a direct measurement of the pressure in the terminal gas cell. With this information, reproducible gas pressures can be set for stripping ions. Measurements can be made of the exiting charge states and maximized for highest sensitivity for the present measurements and subsequently for the TEAMS system.
CHAPTER IV

MEASUREMENTS

The measurement of charge states seems simple at first, but it soon becomes evident that many physical factors must be taken into account and many hardware hurdles overcome. Initial charge state measurements were attempted with an RBS-like system at the exit of the accelerator as used by the Lund group for carbon Charge State Distributions (CSD). However, the spectra were cluttered with ions in the different charge states and therefore different energies, and peaks from ions that were presstripped in the accelerating column before the terminal stripping canal. Therefore, a post-acceleration spectrum of molecules and the resulting fragments at many different energies and charge states would be very complicated and difficult to interpret. We therefore aborted the original idea of measuring molecules and breakup fragments directly from the tandem.

In subsequent trials, we continued to use the terminal stripping canal but used the TEAMS beams line with the HVEC magnet and large ESA for the measurements. The CSD of fluorine were measured as a function of N₂ pressure in the canal, but the pressure could only be determined from the post accelerator ion gauge. The resulting gas pressure dependence data was very erratic and not reproducible, so it was then decide to install a capacitance manometer at the terminal of the accelerator. Since equilibrium CSD data
were also desired, measurements could continue until the installation of the capacitance manometer was completed.

Subsequent measurements of AlO, SiMn, and SiMg were made but the charge state \( q = 1^+ \) could not be investigated with a terminal voltage above \( V_T = 1.0 \text{ MV} \). Because the radius of curvature was smaller for the 40° bend than for other beam lines, the molecular ions could not be bent into the TEAMS beam line, the required magnetic field exceeded the capability of the HVEC magnet. With the TEAMS system normally running at \( V_T = 1.7 \text{ MV} \), any data from this study would not cover the energy range of interest. For example, at \( V_T = 1 \text{ MV} \) and with the injection of the molecule SiMn, the HVEC magnet current must be set to 114% of maximum current to bend the Mn\(^{1+}\) fragment into the TEAMS 40° beam line. Another problem was the requirement of extremely good calibrations of the HVEC magnet and large 45° ESA over the wide range of rigidity values. With magnetic (\( \chi_m \)) and electrostatic (\( \chi_e \)) rigidity defined as:

\[
\chi_m = \frac{\sqrt{2Em}}{qe}, \quad IV-1 \\
\chi_e = \frac{2E}{qe} \quad IV-2
\]

Where \( E \) is the kinetic energy, \( m \) is mass, \( q \) is charge state, and \( e \) is the electron charge of \( 1.6 \times 10^{-19} \text{C} \). Data taken at a lower \( V_T \), with the system running in automatic mode as compared with data from a complete scan revealed systematic problems that even persisted after recalibration. In addition, without scanning both the HVEC Magnet and
ESA, the ionization chamber can not be used. Moreover, the problem of measuring neutral fragments could not be performed with the TEAMS system.

Consequently, it was decided to use the “30° atomic beam line” with the gas cell after the HVEC magnet, in order to strip electrons and breakup the molecular ions. This allowed the terminal voltage to be set at half the value and the HVEC magnetic field, low enough to bend charge state 1+ into the atomic line. Moreover, the correct CSD range could be measured that would be most useful with the TEAMS system. For that reason, AlO was used for preliminary measurements, since 20 nA of beam could be brought into the atomic line.

First, a crude calibration for the “atomic line” was determined using an oxygen beam. Then an injected AlO beam was scanned into the Faraday cup just after the gas cell (Figure IV-1) and a set of constants determined using a least squares fit routine for a quadratic equation. A particle with charge state $q$ and mass $m$ moving in a magnetic field is deflected whenever the magnetic field $B$ has a component perpendicular to the velocity $v$ of the particle. Since the magnetic field and the ion velocity are perpendicular, the force $F = qevB$ is perpendicular to the plane of $B$ and $v$ and the charged particle moves in a circle. For the circular orbit of radius $r_m$, Newton’s second law can be written as

$$ F = \frac{mv^2}{r_m} = qevB. \quad IV-3 $$

and from this it follows that
\[ \chi_m = Br = \frac{mv}{qe} = \frac{p}{qe} = \frac{\sqrt{2mE}}{q}, \]

where \( \chi_m \) denotes the magnetic rigidity of the particle and \( E \) is the kinetic energy. For particles of the same charge state \( q \), the orbital radius in a homogeneous field is proportional to the momentum \( p \). The magnetic field \( B \) is proportional to the current supplied to the current loops around the iron pole faces.

\[ B \propto \frac{\mu I}{4\pi} \]

Therefore the magnetic rigidity is proportional to the current for a constant permeability of the iron core. Fitting the current vs. \( \chi_m \) the constants were determined. The equation was used in a Excel worksheet to find the percent current value for the HVEC required for injecting the desired molecule into the atomic line as a function of mass, energy, and charge.

Table IV-1 lists the terminal voltage required to measure the ions and fragment at the same velocity. The data for SiMn and SiZn had to be at a lower terminal voltage in order to bend the \( q = 1+ \) molecule into the atomic beamline.
Table IV-1: Molecules measured and terminal voltage ($V_T$) required for fragment comparisons. $M_1$ is the first element in the molecule, $M_2$ is the second element.

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Mass 1</th>
<th>Mass 2</th>
<th>Molecular Mass</th>
<th>Final Energy</th>
<th>$V_T$ for Molecule</th>
<th>$V_T$ for M1</th>
<th>$V_T$ for M2</th>
</tr>
</thead>
<tbody>
<tr>
<td>AIO</td>
<td>26.98</td>
<td>15.99</td>
<td>42.98</td>
<td>1.7120</td>
<td>0.8230</td>
<td>0.0398</td>
<td>0.504</td>
</tr>
<tr>
<td>SiN</td>
<td>27.98</td>
<td>14.00</td>
<td>42.09</td>
<td>1.7120</td>
<td>0.8230</td>
<td>0.0407</td>
<td>0.657</td>
</tr>
<tr>
<td>SiMg</td>
<td>27.98</td>
<td>23.99</td>
<td>52.39</td>
<td>1.7120</td>
<td>0.8230</td>
<td>0.0327</td>
<td>0.573</td>
</tr>
<tr>
<td>SiCa</td>
<td>27.98</td>
<td>39.96</td>
<td>68.16</td>
<td>1.7120</td>
<td>0.8230</td>
<td>0.0251</td>
<td>0.479</td>
</tr>
<tr>
<td>SiMn</td>
<td>27.98</td>
<td>54.94</td>
<td>83.02</td>
<td>1.4700</td>
<td>0.7020</td>
<td>0.0177</td>
<td>0.354</td>
</tr>
<tr>
<td>SiZn</td>
<td>27.98</td>
<td>63.93</td>
<td>93.48</td>
<td>1.3200</td>
<td>0.6270</td>
<td>0.0141</td>
<td>0.289</td>
</tr>
</tbody>
</table>
Figure IV-1: AlO⁺ molecule passed into terminal stripper at $V_f=0.7$ MV and scanned after acceleration with the HVEC.
The 3° deflection ESA in the "atomic beam line" was calibrated in the same manner as the HVEC magnet. The electrostatic rigidity is determined from Newton's law

\[ F = \frac{mv^2}{r_e} = qeE, \]

where \( r_e \) is the radius of curvature in the electric field \( E \).

\[ \chi_E = Er = \frac{2E}{qe}. \]

The data were fit with a linear equation. The raw data were taken with the TIDAS system; relevant data stripped out and plotted as a function of cup current vs. ESA voltage (Figure IV-2). Next the peaks were identified and peak areas measured with PeakFit. Because the data is converted to counts by the ADC, based on charge collected, the data must be corrected for charge states to get particle counts. The areas of the peaks must be divided by the charge state of the identified peak. This gives the number of particles for each charge state. Each of the sets of numbers is converted to a relative percentage of total and plotted. However, as stated before, the charge state fractions do not include neutral or negative ions fractions.
Figure IV-2: Raw data from 1 MeV AlO molecule after going through the gas cell and analyzed with the 3° electrostatic analyzer (ESA).
The equilibrium thickness of the N\textsubscript{2} gas was determined for O. This was also a test of the gas cell since it was designed for low pressures. The thickness \(x\) of the gas target in \(\mu g/cm^2\) is determined by the following equation.

\[ x = \frac{mlP}{RT} \]  

Where \(m\) is the target mass (amu), \(l\) is the effective gas cell length (cm), \(P\) is the gas pressure (torr \(\times 133\)), \(R = 8.31 \text{ J/mol} \text{K}\), and \(T\) is temperature (K). From Kim’s thesis\textsuperscript{45}, the gas cell effective length is 14.4 cm, which gives an N\textsubscript{2} thickness of 1.12 \(\mu g/cm^2\) for a \(P = 50 \times 10^{-3}\) torr and \(T = 288\) K. From Figure IV-3 we see that equilibrium charge state fractions were reached at 35-50 mtorr for 3 MeV O ions. This gives an average equilibrium charge state of \(\overline{q} = 3.23\) when compared to published data\textsuperscript{19} of \(\overline{q} = 4.2\) for carbon foils and \(\overline{q} = 3.3\) from a theoretical graph for dilute gas. The average agrees well even without data for \(q = -1\) and 0 states because published data shows for oxygen ions > 1 MeV no resulting charge state \(q < 0\) has been reported.
Figure IV-3: 3 MeV Oxygen charge state distributions as a function of Nitrogen gas pressure. Lines are to guide the eye.
Oxygen data were then taken for equilibrium charge state distributions over a range of energies from 1.0 to 4.2 MeV (Figure IV-4). The gas cell was kept at a constant 40 mtorr pressure to ensure equilibrium conditions were maintained. These data compares well to the semi-empirical formula used in STRIP. The crossing points of the measured charge states are high, and may be from poor calculations of the incident energy.

Figure IV-5 shows the measured gas cell pressure needed to achieve equilibrium charge state fraction for AlO. From Figure IV-5 can be seen that equilibrium is quickly reached between 10 and 20 mtorr. At 20 mtorr, the nitrogen gas target thickness is 0.43 μg/cm². In addition, data for SiZn were taken to confirm equilibrium CSD for the heaviest molecule. Figure IV-6 shows that the SiZn fragments reach equilibrium CSD at approximately 20 mtorr of N₂. With this information, an N₂ gas pressure of 30 mtorr was used for all other CSD measurements. Which corresponds to a gas target thickness of is 0.65 μg/cm².
Figure IV-4: Oxygen equilibrium charge state distributions as a function of energy, dashed lines are from the STRIP program.
Figure IV-5: 1 MeV AlO and fragment charge state distributions as a function of Nitrogen gas pressure. Lines are to guide the eye.
Figure IV-6: Charge state distributions as a function of $N_2$ gas cell pressure for the fragments of 1.32 MeV SiZn.
Figure IV-7 through Figure IV-10 shows the charge state fractions of AlO, SiN, SiMg, SiMn and SiZn. The solid lines are data of the fragments and the dotted lines are of the atomic ion at the same velocity as the fragment. The dashed lines are a semi-empirical fit published by R.O. Sayer\textsuperscript{61} for CSD of high energy ions in dilute gases. The plots were generated with an Excel spreadsheet written by B. Doyle for the ion at the fragment energies. The fitting parameters where originally for higher energy ions (0.04 to 10 MeV/amu), therefore the slight disagreement between the present data and Sayer is expected. But a good agreement is seen for the lower masses, N and O.

As stated earlier in this paper SiCa was to be measured, but data for SiCa could not be used because of interference from other undetermined molecules which completely concealed any SiCa signal. Also for the SiMg data a large beam of SiC\textsubscript{2} was present which adds uncertainty to the Si fragment data.

Statistical error analysis of the experimental data was preformed. The standard deviation was calculated for each set of data runs taken for the different molecules. If a large enough sample was not available, Poisson statistics were used from the individual spectra. Sources of error for this experiment include terminal potential fluctuations of 1%; true gas cell pressure of 1%; overlapping peak area calculation of 2%; and incomplete particle counting leading to incorrect area calculations of 2%. 
Figure IV-7: Charge state distribution of Al and O fragment ions from AlO⁺ incident on N₂ at 1.71 MeV. Atomic Al and O ions at same energy are shown for comparison. Semi-empirical fits by Sayer are shown for ions in dilute gases.
Figure IV-8: Charge state distribution of Si and N fragment ions from SiN+ incident on N₂ at 1.71 MeV. Atomic Si and N ions at same energy are shown for comparison. Semi-empirical fits by Sayer⁶¹ are shown for ions in dilute gases.
Figure IV-9: Charge state distribution of Si and Mg fragment ions from SiMg+ incident on N₂ at 1.71 MeV. Atomic Si and Mg ions at same energy are shown for comparison. Semi-empirical fits by Sayer are shown for ions in dilute gases.
Figure IV-10: Charge state distribution of Si and Mn fragment ions from SiMn$^+$ incident on $N_2$ at 1.47 MeV. Atomic Si and Mn ions at same energy are shown for comparison. Semi-empirical fits by Sayer$^6$ are shown for ions in dilute gases.
Figure IV-11: Charge state distribution of Si and Zn fragment ions from SiZn\(^+\) incident on N\(_2\) at 1.32 MeV. Atomic Si and Zn ions at same energy are shown for comparison. Semi-empirical fits by Sayer\(^6\) are shown for ions in dilute gases.
When the data were analyzed, the sum of the peaks for the two fragments of the molecule was not equal. One might assume when a molecule broke apart the resulting fragments would be detected in a 1:1 ratio. In general, all the molecular data shows a higher count rate for the heavier fragment. This can only be quantified for the SiZn data because during the measurement both the Si and the Zn fragments were measure in one scan. With the other SiX fragments, two scans were performed gating on the energy peaks of interest to separate the overlapping charge state peaks.

This procedure gives rise to a built-in error of the total counts, therefore an exact comparison can not be made. The first sets of data, with the detector 285 cm from the ESA, give a ratio of sum of Si counts / sum of Zn count = 0.25. In an attempt to count all fragments, the detector used to measure neutrals 36 cm from the exit of the electrostatic analyzer (ESA) was used. A small improvement was found for the charge state scan, the ratio was sum of Si counts / sum of Zn counts = 0.31. Then, the gas cell was moved closer to the ESA (from 200 cm to 92 cm away from the exit of the ESA) because beam was being lost on the 10 mm aperture of the normalization detector. The charge state scan data again improved to a ratio of sum of Si counts / sum of Zn counts = 0.50. The final adjustment attempted was to move the ESA plates away from the beam direction. In Figure IV-12, the beam comes into the analyzer very close to the positive electrode. To prevent part of the beam from hitting the right electrode, it was moved 1 cm away from the beam to position it in the center of the plates. With a subsequent scan of the beam into the neutral particle detector the Si$^{1+}$ and Zn$^{2+}$ peaks where not resolvable and therefore the ratio could not be determined.
Figure IV-12: 3° electrostatic analyzer entrance view.
The neutral beam width was measured with the gas cell in the condensed configuration, 32 cm from the entrance of the ESA. Figure IV-13 is a graph of the beam profile measured with the neutral particle detector 36 cm from the exit of the ESA which has a 3.75 mm horizontal x 10 mm vertical slit aperture on the detector. The neutral beam width at 128 cm from the gas cell is 9.0 mm FWHM for the Si fragment and 6.0 mm FWHM for the Zn fragments. This difference in widths is most likely from the scattering in the gas cell because the neutral fragment does not receive a off axis velocity component from the Coulomb explosion and possibly a small contribution from fragments that did Coulomb explode but then electron captured to the neutral charge state.
Figure IV-13: Beam width of the neutral fragments from the breakup of 1.32 MeV SiZn on 30 mtorr of N$_2$. 

Si FWHM = 9mm

Zn FWHM = 6mm
CHAPTER V

RESULTS

Differences were found in the transmission of the breakup fragment ions. In general, all molecules exhibited the same trend of a lower relative count rate in the particle detector for the lighter fragment. With the particle detector 285 cm from the exit of the ESA, the Si count/Zn count ratio was 0.25. When the particle detector was moved to within 36 cm from the exit of the ESA, the Si count/Zn count ratio was 0.32. When the gas cell was moved from 200 cm from the entrance of the ESA to 32 cm, the Si count/Zn count ratio was 0.5. However, in each of the different hardware configurations the charge state fractions remain the same.

An external magnet was used to deflect the ion beam in the gas cell. The count rate of the lighter fragment was found to increase while the heavier fragment count rate decreased. It is believed that the lighter fragment scattering cone was larger than the scattering cone of the heavier fragment. It was found in a later measurement that the width of the neutral Si beam was 50% wider FWHM (9 mm) than the width of the neutral Zn beam (6 mm). This is consistent with the loss of more of the lighter fragment ions compared to the heavier fragment ions and the skewing of the Si count/Zn count ratio away from what you might expect from a molecular breakup, 1:1. It is believed that the beam spread is due to collisions in the gas cell and to Coulomb repulsion of the two
fragments after breakup if they are both charged. From conservation of momentum arguments, the smaller mass fragment will necessarily have the larger velocity.

Figure V-1 shows graphs of the transmission of Si and Zn ion fragments versus the N$_2$ gas cell pressure in mtorr. From 5 mtorr to 70 mtorr the transmission of the fragments decreased by 90% in the end particle detector located 285 cm from the exit of the gas cell. However, the charge state fractions remained constant at each pressure above equilibrium.

As seen in Figures IV-8 to IV-11, the measured charge state fractions for SiX molecular fragments were essentially the same as that measured for the atomic ions at the same velocity. Both charge state distributions peak at the same charge state and have approximately the same magnitude. Although, the semi-empirical calculations of Sayer REF are for atomic ions, the calculations appear to underpredict the measured charge state distributions peak for both the atomic ions and the molecular fragment ions. In some cases the Sayer calculations predict a higher charge state distribution which may be due to the fact that the calculations were for higher energy ions (0.04 to 10 MeV/amu).

In Figure IV-7, the measured charge state fractions for AlO molecular fragments and atomic ions at the same velocity are not in as good agreement partially do to the scatter in the data. This is especially true for the aluminum data. Again, the Sayer calculations predict a lower charge state distribution peak.

Table V-2 shows the average charge states for atomic ions and molecular fragment ions at the same velocity. The charge states were produced by passing the atomic ions and the molecular ions through a 30 mtorr N$_2$ gas cell. The molecular ions
broke apart into fragment ions due to soft collisions in the gas cell. The fragment ions and the atomic ions, at the same velocity, reached equilibrium charge state distributions in the 30 mtorr N₂ gas cell. The average charge state was determined as the sum of multiplying each charge state by the charge state fraction for that charge state. The difference between the average charge states for the atomic ions and the fragment ions is negligible for the four SiX molecular breakups studied. However, for the AlO molecular breakup, the difference is 0.237 of a charge state for Al and 0.60 of a charge state for O. This difference may be due to the more tightly bound AlO molecule compared to the SiX molecules. In fact, the AlO molecule has been observed in a 3+ charge state for many microseconds. Appendix B lists the data average of the charge state fraction for each charge state for all molecules measured.

Figure V-2 shows the equilibrium charge state fraction ratios $F(q)/F(q-1)$ plotted as a function of charge state $q$ for the Al and O fragment ions from the AlO molecular breakup, for the atomic Al and O ions at the same velocity, and for the semi-empirical calculations of Sayer. All of the equilibrium charge state fraction ratio curves follow the same trend and decrease with increasing charge state $q$ including the semi-empirical calculations of Sayer.

Figure V-3 to Figure V-6 show the equilibrium charge state fraction ratios $F(q)/F(q-1)$ plotted as a function of charge state $q$ for the fragment ions, atomic ions at the same velocity, and the semi-empirical calculations of Sayer for SiN, SiMg, SiMn, and SiZn molecular breakup, respectively. The same trend is observed with the equilibrium charge state fraction ratios decreasing with increasing charge state $q$. 
Figure V-7 shows the equilibrium charge state fraction ratios \( F(q)/F(q-1) \) for the four SiX molecular breakups plotted as a function of charge state \( q \). Figure V-7a is for only Si fragment ions and atomic ions at the same velocity. Figure V-7b is for all non-Si fragment ions and atomic ions at the same velocity. Also shown in Figure V-7 are the model calculations of Cooney\textsuperscript{49} for comparison. The model calculations give the following parameters

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<td>Higher Energy Si</td>
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<td>.5</td>
<td>4.1</td>
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<tr>
<td>0.4 MeV/amu</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Lower Energy Si</td>
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<td>4.1</td>
</tr>
<tr>
<td>0.15 MeV/amu</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>All Other</td>
<td>12</td>
<td>.7</td>
<td>4.1</td>
</tr>
</tbody>
</table>

*Table V-1: Cooney fitting parameters.*
Figure V-1 Normalized Fractions of the sum of the peak for 1.32 MeV SiZn fragments as a function of pressure.
Table V-2: Average charge states for ions and molecular fragments at same velocity incident on 30 mtorr N₂ gas cell.

<table>
<thead>
<tr>
<th></th>
<th>Al</th>
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<tr>
<td>Ion</td>
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<td>1.92</td>
</tr>
<tr>
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<td>1.24</td>
</tr>
<tr>
<td>Difference</td>
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</tr>
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<table>
<thead>
<tr>
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<th>Si</th>
<th>N</th>
</tr>
</thead>
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<tr>
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<td>1.18</td>
</tr>
<tr>
<td>Fragment</td>
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<td>1.16</td>
</tr>
<tr>
<td>Difference</td>
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<td>0.02</td>
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</tbody>
</table>

<table>
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<th>Si</th>
<th>Mg</th>
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</thead>
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<td>Ion</td>
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<td>1.49</td>
</tr>
<tr>
<td>Fragment</td>
<td>1.56</td>
<td>1.61</td>
</tr>
<tr>
<td>Difference</td>
<td>-0.04</td>
<td>-0.12</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>Si</th>
<th>Mn</th>
</tr>
</thead>
<tbody>
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<td>Ion</td>
<td>1.27</td>
<td>1.30</td>
</tr>
<tr>
<td>Fragment</td>
<td>1.19</td>
<td>1.30</td>
</tr>
<tr>
<td>Difference</td>
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<td>0.00</td>
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<table>
<thead>
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<th>Zn</th>
</tr>
</thead>
<tbody>
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<td>1.42</td>
</tr>
<tr>
<td>Fragment</td>
<td>1.24</td>
<td>1.48</td>
</tr>
<tr>
<td>Difference</td>
<td>0.02</td>
<td>-0.06</td>
</tr>
</tbody>
</table>
Figure V-2: Equilibrium fraction ratio of 1.72 MeV AlO fragments on N₂.
Figure V-3: Equilibrium fraction ratio of 1.72 MeV SiN fragments on \( \text{N}_2 \).
Figure V-4: Equilibrium fraction ratio of 1.72 MeV SiMg fragments on N$_2$. 
Figure V-5: Equilibrium fraction ratio of 1.47 MeV SiMn fragments on N₂.
Figure V-6: Equilibrium fraction ratio of 1.32 MeV SiZn fragments on N₂.
Figure V-7: Equilibrium fraction ratio for all Si beams and all other beams.
CHAPTER VI

SUMMARY AND CONCLUSIONS

Experiments were conducted to measure the charge state distributions resulting from molecular breakup and to compare them to that obtained for atomic ions at the same velocity. Elements (N, Mg, Mn, and Zn) chosen for study are those of interest to Trace Element Accelerator Mass Spectrometry (TEAMS) because they do not readily form negative ions which are necessary for injection into the tandem accelerator. For these elements to be studied by TEAMS, they have to be transported to the terminal of the tandem accelerator as a molecule and broken apart through soft collisions in the N\textsubscript{2} gas cell in the terminal. AlO\textsuperscript{3+} was chosen as an additional molecule to be studied because it was known to be more tightly bound than the SiX molecules and also it exist in a 3+ charge state for many microseconds.$^{45}$

In order to better control experimental parameters, like N\textsubscript{2} gas pressure and target thickness, the measurements were conducted after production of the SiX\textsuperscript{+} or AlO\textsuperscript{+} molecules in the ion source and acceleration of the molecular ions to the terminal where they were stripped of electrons. Only the SiX\textsuperscript{+} or AlO\textsuperscript{+} molecules were selected after acceleration and passed into a 30° beam line for breakup and analysis of the charge states of the fragments. The SiX\textsuperscript{+} or AlO\textsuperscript{+} molecule ions were passed through an N\textsubscript{2} gas cell to simulate the conditions in the terminal of the accelerator during molecular breakup. The
molecular fragment ions and the atomic ions at the same velocity were found to reach
equilibrium charge state distributions in the N$_2$ gas at a pressure of approximately 20
mtrr corresponding to an N$_2$ target thickness of 0.43 $\mu$g/cm$^2$.

In order to make realistic comparisons to atomic charge state distributions, atomic
ions of the fragments were produced in the terminal by electron stripping, e.g. Si$^+$, N$^+$,
Mg$^+$, Mn$^+$, Zn$^+$, Al$^+$, and O$^+$. The terminal voltage was chosen so that the singly charged
ions were accelerated out of the accelerator at the respective fragment ion velocity
produced in the molecular breakup. Since the two fragments have the same velocity after
breakup, the kinetic energy of the molecule is shared between the two fragments
according to their masses.

No measurements of the charge state distributions resulting from molecular ion
beam breakup were found in the literature except for the work with di-atomic molecules,
like O$_2$ and N$_2$ breakup of the Argonne National laboratory group. No calculations of
charge state distributions resulting from molecular ion beam breakup were found to exist
except for the model of Cooney et al.\textsuperscript{49} Much work has been done on ionic charge states
as listed in Table I-1, and several individuals such as Betz, Datz and Sayer have given
much time and effort to the theoretical understanding and semi-empirical fits of charge
state distributions.

The measured charge state fractions for SiX molecular fragments were found to
be essentially the same as that measured for the atomic ions at the same velocity. Both
charge state distributions peak at the same charge state and have approximately the same
magnitude. The semi-empirical calculations of Sayer\textsuperscript{61} predict lower charge state
distribution peaks than both the atomic ions and the molecular fragment ions. The Sayer calculations predict a higher charge state distribution for some ions which may be due to the fact that the calculations were for higher energy ions. The measured charge state fractions for AlO molecular fragments and atomic ions at the same velocity are not in as good agreement, especially for the aluminum data. Sayer’s calculations again predict a lower charge state distribution peak.

The average charge state for each ion was determined from the charge state fractions by multiplying each charge state by the charge state fraction for that charge state. The average charge state difference for the atomic ions and the fragment ions was found to be negligible for the four SiX molecular breakups. For the AlO molecular breakup, the difference was 0.237 of a charge state for Al and 0.60 of a charge state for O. This difference may be due to the more tightly bound AlO molecule compared to the SiX molecules.

Figure V-7 shows the equilibrium fraction ratios with Cooney’s fitting parameters. The agreement with the Si data is very good and the general trend of the other fragments is represented.

At present, TEAMS requires standards to analyze unknown impurity concentrations. The users of SIMS have developed Relative Sensitivity Factors (RSFs) for almost all-possible contaminants in Si, GaAs, and other semiconductor materials, for both ions and molecules. A TEAMS-RSF would use the SIMS-RSF\textsuperscript{50} multiplied by a charge state factor and a gas cell attenuation factor as given in Equation VI-1.
\[ \rho_i = \frac{I_i e^{n \sigma x} A_m CS_m RSF}{I_m e^{n \sigma x} A_i CS_i} \]  

where \( \rho_i \) is the impurity atom density (atoms/cm\(^3\)); subscript \( m \) is for the matrix element; subscript \( i \) is for the impurity element; \( I \) is secondary ion intensity (counts/s); \( A \) is the isotope abundance fraction; \( CS \) is the charge state fraction; \( n \) is the gas number density (cm\(^{-3}\)); \( \sigma \) is the cross section for collision (cm\(^2\)); \( x \) is the path length in the gas (cm); \( RSF \) is the relative sensitivity factor (atoms/cm\(^3\)).

Hence, the present work provides charge state fractions that may be used to generate TEAMS-RSF's for impurities in semiconductor materials.
APPENDIX A

ION BEAM MODIFICATION AND ANALYSIS LABORATORY
In the ion beam modification and analysis laboratory (IBMAL) at UNT, there is a variety of techniques and equipment available for accelerator-based atomic and molecular physics and material analysis. The laboratory contains approximately $6,000,000 in equipment and has an annual budget of $300,000 for students, post-doctoral researchers, maintenance, and operation. The laboratory has five accelerators, Figure A-1 shows the main laboratory with 3 of the accelerators. The largest is a National Electrostatics Corporation (NEC) 9SDH-2 3 MV Tandem Pelletron to accelerate a wide assortment of atomic or molecular ions. Also, two 2.5 MV Van de Graaff accelerators are in the IBMAL, one is for teaching accelerator techniques and for visiting faculty and students, the other is being setup for routine resonance nuclear reaction analysis (RNRA) of amorphous silicon dioxide. A 200 kV Cockcroft-Walton accelerator is used for sputter initiated resonance ionization spectroscopy (SIRIS) studies. A 200 kV Eaton NV200-4 implanter is operational but has no beam line attached at present.

At IBMAL several areas of study are ongoing: material characterization using Rutherford backscattering spectroscopy (RBS), particle induced x-ray emission (PIXE), elastic recoil detection analysis (ERDA), and resonance nuclear reaction analysis (RNRA); trace element accelerator mass spectrometry (TEAMS) for sub-part per billion contamination measurements of semiconductor samples; X-ray cross sections and electron capture in inner-shell ionization; sputtered particle depth of origin and yield; electron yields from ion bombarded targets; single event upset (SEU); ion beam induced charge collection (IBICC); nuclear microprobe studies; and molecular lifetime and charge state studies. Presently, four professors, two post-docs and nine graduate students are
active in these areas. The laboratory also has access to two electrical technicians and two machinists, and one administrative assistant. Some of the other significant equipment available to facilitate research at IBMAL: depth profilometer for measuring sputtered crater depths; clean room for mounting TEAMS samples; evaporator for making foil targets; 25 computers; 100's of NIM modules; tube furnace; and a student machine shop.

The SNICS Source

The Source of Negative Ions by Cesium Sputtering (SNICS) is an ion source specifically developed for tandem accelerator systems. It operates on the phenomenon of production of negative ions of sputtered particles by low energy cesium ions. Because nearly all atoms can form negative ions (with varying degrees of success), the SNICS is applicable when any ion beam is required. The cesium positive ions are produced through thermal ionization by a high temperature ionizer. Cesium vapor is supplied to the ionizer by heating a reservoir of solid cesium and allowing the vapor to travel to the ionizing region via a transfer tube. The cesium ions are attracted to and focused on a target cathode which is biased at a negative voltage (~5 to -10 kV). The negative ions are also accelerated away from the cathode due to this bias.

The negative ion beam is then shaped, focused, and accelerated further by an extractor voltage. The negative ions then receive the final acceleration from the source bias applied to the entire source cage.
Figure A-1: IBMAL Main Room Layout
Initial mass selection is done with the 30° magnet. More precise mass resolution is obtained by going into a 90° magnet, which steers the beam into the accelerator. Various electrostatic optical elements focus, steer and deflect the ion beam for maximum transmission of the beam through the accelerator.

The Tandem Accelerator

The Tandem Accelerator (Tandem) is a single-terminal accelerator specifically designed to accept negative ions from sources and produce multiply charged positive ions. This is done by using a positive terminal voltage, which attracts the negative ions produced in any of the SNICS, Alphatross, or CHIMERA ion sources. At the terminal voltage is a gas stripping cell, which uses nitrogen gas to "strip" the negative ions to a positive charge state. That is, the collisions between the nitrogen gas atoms and incoming negative ions causes electrons to be knocked off the negative ions and produces any of several positive charge states of the incoming ion. The positive ions are then accelerated away from the terminal under the influence of the positive terminal voltage. The beam energy of the positive ions is given by the equation.

\[ E = e [ V_T + V_T (q + l)] \]

Where \( e \) is the elementary charge, \( V_T \) is the energy of the negative ions out of the respective ion source, \( V_T \) is the terminal voltage, and \( q \) is the final, positive charge state of the ion as it is accelerated away from the terminal.
The terminal voltage is applied to the terminal by depositing charge on two charging chains inside the tandem tank which run parallel to the beamline. The terminal voltage is attained by "inductive picking off" the charge from the chains at the terminal. A power supply inside the tandem deposits variable amounts of charge onto the chains depending on the terminal voltage needed. The terminal voltage is measured by means of a Generating VoltMeter (GVM), basically an AC capacitive divider between the terminal and ground whose output is proportional to the terminal voltage.

A feedback loop, characterized by the control gain and CPO gain, reads this voltage continuously and adds corrections to hold the preset voltage at a stable setting. The feedback loop is coordinated through three, needle-point corona probes whose position with respect to the terminal is variable. These probes provide a leakage current to ground, which is converted, to a bias voltage.

The high voltage on the terminal is distributed along the accelerating tube, in each direction, by means of a series of resistors. Every section of these columns is equal in resistance value, meaning that each section of the column has the same voltage drop across them, which provides a steady and uniform electrical field and therefore acceleration to the ion beam. The high voltage is insulated from the accelerator tank by filling the space between the columns/terminal and the inside of the tank with an insulating gas. Many insulating gases can be used for this purpose depending on the voltage. In the case of the tandem, Sulfur Hexafluoride (SF$_6$) is used. The tank is pumped out to a rough vacuum before being pressurized to ~75-80 psi of SF$_6$. The SF$_6$ is continuously circulated through filters (to remove contaminants) and dryers (to remove moisture) while it is in use.
Excessive moisture/contaminants in the insulating gas will cause instability in the high voltage and poor control of the ion beam.

The nitrogen stripping gas is controlled from the control room by the stripper controller panel. When admitted, the nitrogen gas is confined to the gas cell by the turbo pump on the terminal. This pump recirculates the nitrogen in the cell in a closed loop.

**Existing Tandem Accelerator Beam Lines and Their Functions**

After acceleration the ion beam is bent with the HVEC magnet and passed into one of seven beam lines. The 55° left beam line is used for experiments measuring electron yields from ion bombarded targets, which is important for the semiconductor industry for studying of surface potentials during the implantation process. The 40° left beam line is the TEAMS analysis line, where the ions are magnetically analyzed at 40° for \( mv/q \) \((M/\Delta M \sim 300)\) and electrostatically analyzed at 45° for \( E/q \) \((E/\Delta E \sim 250)\). This gives determination of \( m/q \), to eliminate atomic interference from molecular breakup fragments. Then, the total energy is measured in an ionization chamber. The 30° left is the atomic line, it has been designed for charge state studies, both distributions and x-ray yield vs. charge state. It has also been used to study samples with PIXE and RBS. The 30° left is the beam line used in this experiment, further details will be presented later. The 15° left is an aperatured microbeam used by Texas Instruments for SEU studies of memory chips. The 15° right has a chamber for RBS, ERDA and NRA. The 30° right is not used at present and the 40° right is the focused nuclear microprobe. The microprobe can focus an ion beam to a diameter of 1 \( \mu \)m and map a sample using RBS, NRA, PIXE,
or IBIC. SEU studies can also be preformed. Both before and after the accelerator are electrostatic lensing and steering elements, Faraday cups, beam attenuators and beam profile monitors.

Electronic signals from x-ray and particle detectors are processed with NIM electronics and data is acquired with multi-channel analyzer (MCA) cards in computers. This data is processed on several PC computer systems.
APPENDIX B

TABLES OF EQUILIBRIUM CHARGE STATE FRACTIONS
Table B-1: Equilibrium charge state distributions for 1.71 MeV AlO$^+$ on 0.65 μg/cm$^2$ of $N_2$ gas. Fragment and ions at equal MeV/amu.

<table>
<thead>
<tr>
<th>Q</th>
<th>Al Fragment</th>
<th>Al Ion</th>
<th>O Fragment</th>
<th>O Ion</th>
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</thead>
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<tr>
<td>0</td>
<td>0.0173</td>
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<td>0.1286</td>
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<td>0.4447</td>
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<tr>
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Table B-2: Equilibrium charge state distributions for 1.71 MeV SiN$^+$ on 0.65 μg/cm$^2$ of $N_2$ gas. Fragment and ions at equal MeV/amu.

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<thead>
<tr>
<th>Q</th>
<th>Si Fragment</th>
<th>Si Ion</th>
<th>N Fragment</th>
<th>N Ion</th>
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<td>0.0137</td>
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<td>0.2264</td>
<td>0.2326</td>
</tr>
<tr>
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<td>4</td>
<td>0.0060</td>
<td>0.0067</td>
<td>0.0003</td>
<td>0.0004</td>
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</table>

Table B-3: Equilibrium charge state distributions for 1.71 MeV SiMg$^+$ on 0.65 μg/cm$^2$ of $N_2$ gas. Fragment and ions at equal MeV/amu.

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<thead>
<tr>
<th>Q</th>
<th>Si Fragment</th>
<th>Si Ion</th>
<th>Mg Fragment</th>
<th>Mg Ion</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.0700</td>
<td>0.0322</td>
<td>0.0080</td>
<td>0.0145</td>
</tr>
<tr>
<td>1</td>
<td>0.5205</td>
<td>0.5072</td>
<td>0.4321</td>
<td>0.5085</td>
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<tr>
<td>2</td>
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<td>0.3846</td>
<td>0.5306</td>
<td>0.4481</td>
</tr>
<tr>
<td>3</td>
<td>0.0682</td>
<td>0.0684</td>
<td>0.0282</td>
<td>0.0275</td>
</tr>
<tr>
<td>4</td>
<td>0.0045</td>
<td>0.0077</td>
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<tr>
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<td></td>
<td></td>
<td></td>
<td>0.0008</td>
</tr>
</tbody>
</table>
Table B-4: Equilibrium charge state distributions for 1.47 MeV SiMn on 0.65 µg/cm² of N₂ gas. Fragment and ions at equal MeV/amu.

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<tr>
<th>Q</th>
<th>Si Fragment</th>
<th>Si Ion</th>
<th>Mn Fragment</th>
<th>Mn Ion</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.0844</td>
<td>0.0642</td>
<td>0.0636</td>
<td>0.0635</td>
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<td>0.6369</td>
<td>0.6458</td>
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<td>0.2447</td>
<td>0.2607</td>
</tr>
<tr>
<td>3</td>
<td>0.0186</td>
<td>0.0221</td>
<td>0.0368</td>
<td>0.0359</td>
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<tr>
<td>4</td>
<td>0.0006</td>
<td>0.0003</td>
<td>0.0106</td>
<td>0.0056</td>
</tr>
<tr>
<td>5</td>
<td></td>
<td></td>
<td>0.0017</td>
<td>0.0013</td>
</tr>
</tbody>
</table>

Table B-5: Equilibrium charge state distributions for 1.32 MeV SiZn⁺ on 0.65 µg/cm² of N₂ gas. Fragment and ions at equal MeV/amu.

<table>
<thead>
<tr>
<th>Q</th>
<th>Si Fragment</th>
<th>Si Ion</th>
<th>Zn Fragment</th>
<th>Zn Ion</th>
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REFERENCES


15. V.S. Nikolaev, Sov. Phys, Usp. 8, 269 (1965).


National Electrostatic Corporation, 7540 Graber Road, PO Box 620310, Middleton, Wisconsin, 53562-0310.


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