Multiple ionization of argon in coincidence with projectile ions in 60–120 MeV Si\(^{q^+}\)–Ar collisions


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Abstract. A projectile ion–recoil ion coincidence technique has been employed to study the multiple ionization and the charge transfer processes in collisions of 60–120 MeV Si\(^{q^+}\) (q = 4–14) ions with neutral argon atoms. The relative contribution of different ionization channels, namely; direct ionization, electron capture and electron loss leading to the production of slow moving multiply charged argon recoil ions have been investigated. The data reported on the present collision system result from a direct measurement in the considered impact energy for the first time. The total ionization cross-sections for the recoil ions are shown to scale as \(E_p^*\), where \(E_p\) is the energy in MeV of the projectile and \(q\) its charge state. The recoil fractions for the cases of total- and direct ionizations are found to decrease with increasing recoil charge state \(j\). The total ionization fractions of the recoils are seen to depend on \(q\) and to show the presence of a ‘shell-effect’ of the target. Further, the fractions are found to vary as \(1/j^2\) up to \(j = 8^+\). The average recoil charge state \langle j \rangle\) increases slowly with \(q\) and with the number of lost or captured electrons from or into the projectile respectively. The projectile charge changing cross-sections \(\sigma_{q\rightarrow q'}\) are found to decrease with increasing \(q\) for loss ionization and to increase with \(q\) for direct- and capture ionization processes respectively. The physics behind various scaling rules that are found to follow our data for different ionization processes is reviewed and discussed.

Keywords. Multiple ionization; recoil ions; charge state fractions; scaling rules.

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1. Introduction

Studies related to recoil ion production by energetic heavy projectiles have been the subject of extensive investigations since the pioneering work of Cocke [1] in 1979. In his measurements, the recoil ions were studied without any correlation with the charge states of the scattered projectile. A detailed knowledge about collisional processes like, electron capture, loss or ionization is of basic importance for an understanding of high temperature
plasmas since many plasma properties are significantly affected by the presence of highly charged ions. Additionally, applications of the cross-sections to astrophysical problems are well known. Over the years, a number of groups [2–6] have focused on studies of the recoil ion distributions in correlation with the scattered projectile of a given charge state. The projectile ion–recoil ion coincidence technique has been used to see the variation of the charge state distribution of the recoil ions and their partial ionization cross-sections for different ionization channels, like; direct ionization, electron capture and electron loss. The classification of such collision processes has been done on the basis of the charge change undergone by the projectile during the interaction. An exhaustive review of the work performed by the above groups can be found in the article of Cocke and Olson [7].

In the present work, we report on the measurements of the Ar\(^{2+}\) recoil ions produced in the collision of 2.14–4.20 MeV/amu partially/fully stripped Si\(^{q+}\) projectiles (q = 4–14) with neutral argon atoms. The major objective of this work was two-fold: firstly, to develop a working experimental set up called SCORPION (System for COincidences between Recoil and Projectile IONs) for carrying out the accelerator-based atomic physics experiments using the 15 UD Pelletron accelerator facility at Nuclear Science Centre (NSC), New Delhi, and secondly, to provide the experimental data on a collision system not studied before. These data are expected to throw further light on the mechanism of producing multiple ionization in the target atom through various ionization processes in energetic heavy ion-atom collisions and to provide a support for validity of various scaling rules that are established before.

2. Experiment

The present experiments were performed using the SCORPION set up at the 15 UD Pelletron accelerator facility of NSC, New Delhi [8]. A schematic diagram of the basic components of SCORPION set up is shown in figure 1. The details of each part, its design,

![Figure 1. Schematic diagram of the experimental set up.](image-url)
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fabrication and optimization etc. have been described elsewhere [9,10]. A pulsed beam of Si$^{4+}$, Si$^{5+}$ and Si$^{6+}$ ions at energies of 60, 85 and 120 MeV respectively was delivered by the accelerator. The repetition rate of the beam pulses was 1 microsec. Higher charge states of Si ions were selected by the switching magnet after the beam was passed through a post acceleration stripper foil. The number of incident ions was reduced to a desired value using a four jaw slit assembly placed at the entrance of the SCORPION set up. This was necessary because of the limited count-rate handling capacity of the parallel plate avalanche counter (PPAC). The reduced beam was passed through a time of flight (TOF) spectrometer having a tantalum aperture of 3 mm at its entrance, and interacted with a beam of neutral argon atoms effusing from a hypodermic needle in a crossed beam geometry. Collisionally induced argon recoils of various charge states were extracted from the interaction region by applying an electric field between the two parallel plates of the TOF set up. The two plates were separated from each other by 20 mm and were held at a potential of ± 500 V. After passing through a 5 mm hole in the negative plate and subsequently through a drift tube of length 30 mm, the recoils were finally registered in a channeltron detector. The single collision condition was established by studying the reaction rates of argon as a function of the gas pressure. The pressure was measured at the inlet port and just below the time of flight spectrometer box by using a Pirani and a Penning gauge respectively. The Pirani gauge showed a pressure of the order of $10^{-2}$ mbar and the Penning gauge pressure was of the order of $10^{-6}$ mbar.

The scattered projectiles, which might or might not have changed their charge states during the collision then entered a parallel plate electrostatic charge analyzer [11]. The applied electric field strength of the order of 1 kV/mm across the parallel plates separated the various charge states of the scattered projectiles which were detected by a 1-dimensional position sensitive PPAC. The total flight path of the scattered projectiles from the exit point of the charge analyzer to the PPAC was about 1600 mm.

The various signals from the detectors were electronically processed and the outputs were fed into a multiparameter ADC module AD811. The strobe to AD811 was given from the channeltron. The data were collected in the list mode. The time of flight spectra of the recoils were obtained by giving a START signal to the time to amplitude converter (TAC) from the channeltron and a STOP signal from the anode of the PPAC detector through a suitable delay. The position spectrum of the various charge states of the scattered projectiles was obtained by dividing the signal from one end of the cathode of the PPAC with the sum signal from the two ends. A typical example of the argon recoil spectra corresponding to all ionization channels and that of a position spectrum of the scattered silicon projectiles produced in 60 MeV Si$^{12+}$–Ar collisions are shown in figure 2a and 2b respectively. The spectrum shown in figure 2a contains the visible peaks of $\text{H}_2\text{O}^+$, $\text{N}_2^+$, $\text{O}_2^+$, $\text{H}^+$ and $\text{H}_2^+$ as impurity recoils. The impurity recoils have times of flight quite different from those of argon recoils and they were found not to interfere with the argon recoil peaks under consideration. Further, the yield of the impurity recoils normalized to the number of projectiles was found to be constant with gas pressure. This suggested that the argon gas used for the studies was free from contaminant gases and that the residual gas was present in the interaction region as a constant background. However, the spectrum for the scattered projectiles as shown in figure 2b, has also, in addition to the main peak of the incident projectile charge state, peaks corresponding to the 1-electron capture (Si$^{11+}$) and 2-electron capture (Si$^{10+}$) events.
Figure 2a. Time of flight spectrum of argon recoils in coincidence with the integrated charge states of the projectiles for 60 MeV Si$^{12+}$–Ar collisions.

Figure 2b. Position spectrum of the scattered projectiles obtained in 60 MeV Si$^{12+}$–Ar collisions.
The data were stored on the magnetic tapes and were sorted after the completion of the experiment using the NSCSORT software. In order to obtain the recoil ion charge distribution corresponding to a particular ionization process, the suitable conditions were generated by considering the start and the end channels of the chosen scattered projectile peak. The conditions were then applied to the total recoil spectrum to generate the desired spectra. Figure 3a–c show the argon recoil ion distributions in 60 MeV Si$^{12+}$–Ar collisions for the direct ionization (Si$^{12+}$→Si$^{12+}$), 1-electron capture (Si$^{12+}$→Si$^{11+}$) and for 2-electron capture (Si$^{12+}$→Si$^{10+}$) processes respectively. As observed from the distributions,
the yield of the recoil ions peaks at \( j = 1^+ \) drops with increasing \( j \) values for the direct ionization process. However, the distributions for electron capture events are found to be ‘bell-shaped’ with the peak shifting to some intermediate charge states in contrast to the distributions for direct ionization. The recoil ion peaks tend to show a tailing effect which is more prominent for the lower charge states. This effect can be interpreted to be due to a charge loss suffered by the recoil ions due to interaction with residual gas during flight path prior to their final acceleration into the channeltron cone. In a coincidence measurement, such as the present one, there is always a possibility to observe contribution of coincidence events from those projectiles which have changed their charge states in collisions occurring outside the effective interaction region besides those which have changed their charge states actually due to electron capture or electron loss processes during their collisions with target atoms. For example, the incident beam of Si\(^{12+}\) ions may have a small component of Si\(^{11+}\) ions produced by interactions with impurity atoms prior to actual collision with the target atoms. Si\(^{11+}\) ions produced due to \( 1e^- \) capture from the argon atoms and those due to the impurity atoms in the beam will be incident at the same position of the position sensitive PPAC. Thus, the corresponding recoil ion spectrum will have recoil ions produced due to \( 1e^- \) capture as well as due to direct ionization by the beam impurity ions. In order to obtain the true yield of the recoil ions exclusively for the electron capture and for the loss processes, it is necessary to remove the unwanted contribution from the beam impurity ions. In the present case, this was done by normalizing the electron capture spectra with the direct ionization spectrum for Si\(^{12+}\) ions incident on argon following the procedure of Gray et al [2]. It was noted that for the recoil ions of lower charge states (for example, for Ar\(^{2+}\) to Ar\(^{12+}\)), the corrections in the recoil ion yields after normalization ranged from 30% to almost 100%. As a matter of fact, the maximum correction was applicable to the yield of Ar\(^{1+}\) ion.

3. Results and discussion

3.1 Energy and charge state scaling laws

The first attempt to obtain a scaling law for the net ionization cross-sections \( \sigma_{tot} \) for recoil ions was made by Schlachter et al [12]. They have plotted \( \sigma_{tot} \) divided by the projectile’s charge state \( q(\sigma_{tot}/q) \) against the energy per nucleon per charge state of the projectile \( (E_p/amu)/q \). For the given target species, the plotted values were found to reduce to a single curve with the experimental results tending to follow a \( q^{3/2}/E_p^{1/2} \) scaling law.

Further emphasis on the scaling laws was made by Be et al [13]. These workers found the net ionization cross-section \( \sigma_{tot} \) of He, Ne and Ar atoms using partially and fully stripped projectiles of He, C, O, Ne and Ar at a fixed energy of 1.05 MeV/amu. For the case of fully stripped projectiles, the cross-sections were found to scale as \( q^{1.7} \) for neon and argon targets. For partially stripped projectiles in low charge states, some deviation of scaling law for targets having low cross-sections was observed. In particular, data for the partially ionized ions with \( q = 2^+ \), were found to significantly deviate from those for He\(^{2+}\) ions. However, the deviation among the ions having different number of screening electrons (C\(^{2+}\), O\(^{2+}\) and Ne\(^{2+}\)) was found to be small. To account for these screening electrons, the authors found out the scaling laws for the variation of the cross-sections as a function of the effective charge of the projectile. For the case of argon targets bombarded...
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by projectiles ranging from $\text{H}^+$ to $\text{Ne}^{10+}$, the scaling parameter was found to be $q^{1.7}$. A more detailed discussion on scaling laws is in order.

To establish the scaling law as a function of the energy and charge state of the incoming ion, total ionization cross-sections for argon recoils divided by the projectile’s charge state ($\sigma_{\text{tot}}/q$) have been plotted as a function of the energy of the projectile divided by its charge state ($E_p/q$). Figure 4a shows the data points corresponding to the measurements performed by various workers [12,13] over a wide range of energy and charge states of the incoming projectiles, like He, C, O, Ne, Fe, Nb and Pb. The straight line passing through the experimental points shows the cross-sections scaled as $q^{1.7}/E_p^{0.5}$ with the net ionization cross-sections for 1.05 MeV $\text{He}^{2+}$ ions incident on argon as the reference. For the range of $E_p/q$ between 0.1 and 0.6 (present range over which the measurements have been performed), the agreement between the measured values and the scaled values is seen to be within 25%.

In the present studies, it was difficult to measure the number of target atoms in the interaction region. Hence, the scaling law mentioned above was used to find out the total ionization cross-sections for 85 MeV $\text{Si}^{14+}$ ions bombarding the argon atoms. For a normalization purpose, the net ionization cross-section for 1.05 MeV/amu $\text{Ne}^{10+}$ on argon were taken from the work of Be et al [13]. The desired cross-sections were obtained by using the following relationship,

$$\sigma_{\text{tot}} = \sigma_{\text{tot}}(3.04 \text{MeV/amu } \text{Si}^{14+}) \times \left( \frac{1.05}{10} \right)^{1.7} \times \left( \frac{3.04}{0.05} \right)^{0.5}.$$

The approximate number of target atoms $N_a$ in the interaction region was then obtained using the equation,

$$N_a = \left( \sigma_{\text{tot}} \right)_{3.04 \text{MeV/amu } \text{Si}^{14+}} \times N_p N_a,$$

where $N_p$ is the number of projectiles. The value of the argon atoms in the interaction zone was found to be $10^{12}$ atoms/cc.

Since the flow of the target gas was kept constant throughout the experiment, $N_a$ was taken to be the same for all measurements. Using this number, the net ionization cross-sections for the argon recoils were estimated for the other charge states and energies of the silicon ions. Figure 4b shows the variation of $\sigma_{\text{tot}}/q$ with $E_p/q$ for the present measurements. The straight line in the plot shows the cross-sections predicted by the scaling rule ($q^{1.7}/E_p^{0.5}$) using the cross-section for 85 MeV $\text{Si}^{14+}$ as the reference. All the cross-sections are seen to follow the above scaling rule within the experimental uncertainty of about 15%.

3.1.1 Total ionization cross-section: Total recoil ion production cross-sections $\sigma_{\text{tot}}$ for collisions of projectiles having energy from 0.10 to 4.75 MeV/amu in various charge states with $\text{H}_2$ and different rare gas targets have been studied experimentally as well as theoretically by Cocke [1] and by Schlachter et al [12,14]. Theoretical calculations employing the classical trajectory Monte Carlo (CTMC) method were used to calculate the ionization cross-sections for multi-electron targets by use of the independent electron model [15,16]. Cocke [1] measured $\sigma_{\text{tot}}$ for $\text{Cl}^{1+}$ projectiles ($q = 6$–14) at energies ranging from 0.8 MeV/amu to 1.2 MeV/amu on He, Ne and Ar targets. He found $\sigma_{\text{tot}}$ to decrease slowly.
with projectile impact energy; the similar trend is also found in our present work (see, figure 4b). Cross-sections for singly charged recoil ions were found to increase very slowly with initial projectile charge $q$, closer to linearly with $q$ than quadratically. For higher recoil ion charge states, the $q$-dependence was found to be much steeper. He further pointed out that the CTMC calculations could more appropriately describe the lower ionization states of the target while the energy deposition model [17] accounted for most of the features of the higher recoil ion charge states.

Schlachter et al [12,14], as mentioned above, have shown that for a given rare gas target, the cross-sections $\sigma_{\text{tot}}$ reduce to a common curve when plotted as cross-section divided by charge state versus energy per nucleon divided by charge state (see, figure 4a). This feature is found strictly true for the ionization cross-sections of targets by fully stripped projectiles while the cross-sections by partially ionized projectiles result in different curves indicating the importance of the screening electrons. Hence, it is necessary to consider the cross-sections as a function of the effective charge $q_{\text{eff}}$ of such partially ionized projectiles instead of the projectile charge itself, by taking into account the screening of the nucleus by

![Figure 4a](image)

**Figure 4a.** Scaling law for total ionization measurements. Solid line shows the $q^{1.1}/E_p^{0.2}$ dependence of the cross-sections on the projectile charge state $q$ and the energy $E_p$. Experimental data taken from [12,13].
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Figure 4b. Variation of \( \sigma_{\text{tot}}/q \) vs \( E_p/q \) for the present data.

electrons. Further, this is of interest that the reduced plot of figure 4a is inapplicable to the cross-sections \( \sigma_j \) calculated for the specific charge state \( j \) of the target for ionization; such a behavior is expected since the Bohr–Lindhard arguments for the lower reduced energy collisions apply only to the total charge removed from a target atom. The above feature is indeed supported by our data (see, figure 4b) for the considered energy and charge states of the silicon projectiles.

3.1.2 Individual ionization cross-sections: Measurements of recoil ion charge state fractions \( F_j \) created by passage of a fast projectile through a gas jet have provided the cross-sections \( \sigma_j \) for producing a recoil ion in charge state \( j \). Cross-sections \( \sigma_j \) for producing \( j \)-times ionized recoil ions in He, Ne, Ar, Kr and Xe targets have been studied for 1.4 MeV/amu \( \text{U}^{4+} \) ions and were compared with the CTMC calculations for 1–5 MeV/amu \( \text{U}^{4+} \) projectiles by Schlachter et al [14]. The agreement between experiment and theory was found to be only qualitative. The discrepancies in the theoretical description were pointed out due to two reasons: (i) the average binding energy for all electrons within a given shell was taken as input parameter in the independent electron model and (ii) the
electrons are removed sequentially if the collision is not sufficiently sudden, with the result that the last few electrons removed from a shell have binding energies much larger than the average value of the model. Thus, the calculations would generally overestimate the cross-section for production of higher charge states. Further, the cross-sections $\sigma_j$ for low $j$-charge states were found to underestimate the CTMC calculations which reflects the classical description of the radial electron distribution in the target atom. Since tunneling is not allowed classically, the electron distribution is not accurately portrayed at large electron-nucleus separations. Generally, the low charge state recoil-ions are generated by collisions at large impact parameters (soft-collisions). Comparison of theory with experimental data show that the collisions are of longer range than calculated in ref. [14].

3.1.3 $F_j$ and $1/j^2$ scaling: Theoretical as well as experimental $F_j$-values for 1.4 MeV/amu U$^{14+}$, Ar, Kr and Xe have been found to lie on a single curve by Schlachter et al [14] except for $F_9$ and $F_{10}$ in Ne. The CTMC calculations show a pronounced effect due to the ‘shell-structure’ of the target atom while there is no such indication in their experimental data. This kind of feature indicates that Auger processes must be contributing significantly to the production of highly charged recoil ions as it is seen in their experimental data. The production of highly charged recoil ions through the Auger process is, however, not included in the CTMC calculations. Contrary to this, in the present results for collisions of 2.1–3.0 MeV/amu Si$^{14+}$ ions with argon atoms, the total ionization fractions $F_j$ show a clear indication of a ‘shell-structure’ of the target atom at $j = 8+$ (see, eg., figures 5a and 5b). In these figures, the solid lines represents a $1/j^2$ scaling behavior when the data

Figures 5a.
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Figures 5b,c.

Figure 5. Argon recoil ion fractions distribution as a function of recoil charge state $j$ for (a) total ionization in 60 MeV $Si^{2+}–Ar$ collisions; (b) direct ionization in 60 MeV $Si^{1+}–Ar$ collisions; (c) 1-electron capture for different incident energies of $Si^{12+}$ ions; (d) 1–4 electron loss case for 60 MeV $Si^{10+}–Ar$ collisions. Solid lines in figures 5a and 5b show the $1/j^2$ dependence of the fractions. Lines joining the points in figures 5c and 5d are a guide to eyes.

for $F_j$ are plotted versus recoil ion charge states $j$. Deviations of $F_j$ values at high $j$ from $1/j^2$ scaling behavior is discussed below. The above scaling behavior can be understood by noting the prediction of CTMC calculations according to which the recoil ion energy $E_t$ in eV for simple Coulomb scattering is given by [18],

$$E_t = \left( C_r q^2 j^2 \right) / (M_t E_p b^2),$$  

(3)

where, $q$ and $j$ are charge states of the projectile and that of the recoil ions respectively. $M_t$ is the mass of the target in amu, $E_p$ is the energy of the projectile in MeV/amu and $b$ is the impact parameter in units of $a_0$ (Bohr radius). The coefficient $C_r$ equals $4 \times 10^{-4}$ if one assumes the projectile-target nucleus interaction to be zero until the distance of closest approach where the electrons are ejected. For a given $M_t, E_p$ and $b, E_t$ follows a $j^2$ behaviour suggesting that the intermediate charge states of the recoil ions (up to $j = 8+$) are born out of the interactions taking place approximately within the same range of impact parameters.

Further, our results reported earlier [19] have shown that there exists a linear relationship between recoil fractions and the corresponding values of $1/E_t$ for almost all the charge
states considered in the investigation. This result has pointed to the fact that the recoil fraction must show a $1/j^2$ behaviour which is indeed observed in the present investigation and is shown in figures 5a, 5b. This dependence can be expected from the binding energy consideration for various shells of the target atom.

Outer shell electrons (low recoil charge states) are produced mainly due to the soft collisions as mentioned above and their number increases following the cascade process via Auger transitions to further inner shell vacancies. The $1/j^2$-scaling behavior is indeed verified by our data for ‘total’ as well as for ‘direct’ fractional ionization in collisions of 60–85 MeV Si$^{q+}$ ions ($q = 4–14$) with argon target (see, figures 5a and 5b). However, deviations between experiment and $1/j^2$ scaling behavior at a given impact energy of the projectile increase more and more as the charge state of incoming projectile reduces from a high value to a low value. These deviations are understood by noting the fact that Coulomb ionization cross-sections are proportional to square of the charge state of the incoming projectile; projectiles of high charge states produce highly charged states of the recoils more efficiently thus bringing a better and better agreement between experiment and theory. This trend indirectly improves the match between $F_j$ and $1/j^2$ relationship as one moves from lower to higher charge states of the projectile at a considered impact energy.

It may be pointed out here that the present results for multiple ionization of the argon atoms in 2.1–4.2 MeV/amu Si$^{q+}$ Ar collisions ($q = 4–14$), clearly show a ‘shell structure’ at the recoil ion charge state $j = 8+$ and are found to follow a $1/j^2$-scaling behavior for ‘total’ as well as for ‘direct’ recoil charge state fractions.

Deviations of recoil charge state fractions $F_j$ from the $1/j^2$-scaling behavior at $j$-values higher than $j = 8$ can be explained by noting the fact that the ionization of 8-electrons of the outer most shell (M-shell) of argon atom takes place generally in large impact parameter encounters (see discussion above); their ionization transition probabilities are calculated using one electron formalism and extending it to represent a multi-electron target by use of the independent electron model. However, the electrons of next neighboring inner shell (i.e., L-shell) of argon atom are known to have many fold larger binding energies than those of M-shell electrons. This suggests that the ionization probability of an L-shell electron reduces to a considerably smaller value. As a matter of fact, the $F_j$ values for $j > 8$ are seen to fall down more steeply than the $1/j^2$ prediction.

3.2 The charge changing cross-sections

The charge changing cross-sections for a particular ionization channel have been obtained using the expression

$$\sigma_{q'q} = (N_p)^{q'} / N_p N_T,$$

(4)

where $q$ and $q'$ represent the charge states of the projectile before and after the collision respectively, $N_T$ is the number of target atoms per unit volume. For the direct ionization process, $q = q'$; whereas for electron loss or electron capture process, $q'$ differs from $q$ depending on the number of electrons lost or captured by the incident ion during the interaction respectively. The projectile charge changing cross-sections for 85 MeV Si$^{q+}$ ions interacting with argon, plotted as a function of the incident projectile charge state are shown in figure 6. The cross-sections corresponding to direct ionization and to electron capture processes are found to increase with the projectile charge state. For the electron
loss channel, the cross-sections decrease with the increasing projectile charge state or with the increasing number of electrons lost by the projectile during the collision. The ratio of cross-sections for 2-\(e^-\) and for 1-\(e^-\) capture is found to be 0.24. This value is comparable to those quoted by earlier investigators [20,21].

The projectile charge changing cross-sections obtained for 1-\(e^-\) capture in the present measurements have been compared with the scaling rule of Schlachter et al [22]. According to their scaling rule, the charge changing cross-sections for all atomic and molecular targets reduce to a single curve (figure 7) when the reduced cross-section \(\tilde{\sigma}\) was plotted as a function of the reduced energy \(\tilde{E}\), where \(\tilde{\sigma}\) and \(\tilde{E}\) are given by the relations:

\[
\tilde{\sigma} = \sigma \left( \frac{Z^{1.8}}{q^{0.5}} \right) \quad \text{and} \quad \tilde{E} = \left( \frac{E_p}{Z^{1.5} q^{0.7}} \right).
\]  

(5)

In the above expression \(\sigma\) is the electron capture cross-section experimentally obtained for a given combination of the target atom of atomic number \(Z\), the projectile ion of charge state \(q\) and the impact energy \(E_p\) (keV/amu). The scaling rule was quoted to be valid within a factor of 2 by the authors. The present experimental data are shown by filled triangles on the reduced plot which are found to be in good agreement with the scaling law within the region of its validity (see figure 7).

Figure 6. Beam charge changing cross-sections for the various ionization channels in collisions of 85 MeV Si\(^{2+}\) ions with argon as a function of the projectile charge state.
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Figure 7. Plot for reduced cross-sections as a function of the reduced energy showing the scaling rule for $1-e^-$ capture cross-sections. Present data are shown on the plot with filled triangles. Additional data are taken from [12].

Table 1. Comparison of the cross-sections obtained for the 1-electron loss case with those obtained using the empirical scaling rule of Alton et al [23].

<table>
<thead>
<tr>
<th>Beam $E_{\text{p}}/q$</th>
<th>Scaled $\sigma$ (cm$^2$) (Alton et al)</th>
<th>Experiment $\sigma$ (cm$^2$) (present values)</th>
</tr>
</thead>
<tbody>
<tr>
<td>60 MeV (4+)</td>
<td>$2 \times 10^{-16}$</td>
<td>$1.4 \times 10^{-16}$</td>
</tr>
<tr>
<td>60 MeV (10+)</td>
<td>$6.3 \times 10^{-17}$</td>
<td>$5.3 \times 10^{-17}$</td>
</tr>
<tr>
<td>85 MeV (6+)</td>
<td>$9.01 \times 10^{-17}$</td>
<td>$7.0 \times 10^{-17}$</td>
</tr>
<tr>
<td>85 MeV (10+)</td>
<td>$4.7 \times 10^{-17}$</td>
<td>$4.3 \times 10^{-17}$</td>
</tr>
</tbody>
</table>

An empirical scaling rule for the $1-e^-$ loss cross-sections was given by Alton et al [23]. The authors had modified the Bohr formula by taking into account the shell from which the electron is lost and that of the ionization potential of the considered electron. The expression for $1-e^-$ loss cross-section is given by

$$\sigma = N_j \frac{13.6}{I_j} \left[ \frac{V_o}{V_i} \right]^2 \left[ Z_1^{1/3} + Z_2^{1/3} \right] \pi a_0^2,$$  

where, $\sigma$ is the electron loss cross-section, $Z_1$ and $Z_2$ are the atomic numbers of the projectile and the target respectively, $N_j$ is the $j$th shell from which the electron is lost, $I_j$ is the ionization potential of the $j$th electron being lost, $V_o$ is the orbital velocity of the electron, $V_i$ is the velocity of incident projectile and $a_0$ is the Bohr radius. The loss cross-sections calculated using the above expression are listed in table 1 along with the experimental values. The values for the ionization potential were taken from the work of Carlson et al [24]. The agreement between the two values is found to be within 20%.
3.3 Recoil ion fraction and average charge state

The fraction $F_j$ of the recoil ion in the $j$th charge state is obtained by normalizing the area $A_j$ of the peak under this charge state by the total area of the peaks under all the recoil charge states, i.e.,

$$F_j = \frac{A_j}{\sum_{j=1}^{n} A_j}. \quad (7)$$

Further, the average recoil ion’s charge state $\langle j \rangle$ is obtained by using the expression,

$$\langle j \rangle = \sum_{j=1}^{n} j F_j. \quad (8)$$

The variation of fractions of the recoil ions for the total ionization as well as for each of the ionization processes has been studied by plotting the fractions as a function of the recoil charge state. It is found that the recoil ion fractions decrease with the increasing recoil ion charge state for both, the total- and the direct-ionization processes (see figures 5a, 5b). Uncertainties in the relative charge state fractions are estimated to range from 1% for Ar$^{+}$ to 36% for Ar$^{10+}$. The decrease in fractions of the recoil ions for both the cases is seen to be much faster beyond $j = 8+$. This behavior is attributed to the shell-effects, which arise due to a sudden change in the binding energy in going from $M$-shell to $L$-shell of argon atom [1]. Also, for the total ionization case, the fractions and hence the ionization probabilities for producing the higher recoil charge states increase as the incident projectile charge state changes from $8^+$ to $10^+$ (see figures 5a and 5b). It may be noted here that Si$^{10+}$ has six $L$-shell vacancies while no $L$-vacancies are present in Si$^{4+}$. Thus, the increase in the ionization probability of argon atoms points to a possible transfer of the $L$-shell electrons of the target to the $L$-vacancies of the projectile [25]. The process of transfer followed by Auger emission results into the highly ionized recoils with appreciable fractions. This feature is further highlighted in the discussions related to the electron capture in following paragraphs. It is also noted that the fractions of the recoils seem to follow a $1/j^2$ dependence for the total ionization processes as shown by solid lines in the plots. This behavior is observed for the recoil ions up to $j = 8+$. However, for the direct ionization case, a considerably large disagreement between the experimental fractions and the prediction of $1/j^2$ dependence is observed for the incident ions in charge state $4^+$. The disagreement reduces for Si$^{6+}$ ions and almost vanishes for incident projectiles having charge states ranging from $10^+$ to $14^+$. The direct ionization fractions are also found to be independent of the incident energy of the projectile. The value of average recoil ion charge state for the direct ionization channel shows only a slight change from 1.3 for Si$^{4+}$ to 1.8 for Si$^{12+}$ (see figures 8a and 8b). Thus, from the studies of recoil ion distribution and that of average recoil ion charge states, it can be concluded that the lower recoil charge states, particularly singly charged recoil ions are dominantly produced by the direct ionization process. These results point to the fact that the direct ionization collisions generally occur at large impact parameters.

It may be interesting to compare the charge state distribution of recoil ions produced in slow-collisions between multiply charged ions and free atoms, that is, collisions at velocities less than 1 atomic unit ($2.2 \times 10^8$ cm s$^{-1}$) with that produced in energetic (a few
Multiple ionization of argon collisions, such as the collision of present case. At low collision velocities, the cross-sections are sensitive to the details of the potential energy of the quasi-molecular system; many properties can be understood on the basis of energy difference between the initial and final states of the system. In these collisions, the internal potential energy often dominates effects due to kinetic energy of the projectile, especially for ionization; direct or impact ionization becomes increasingly unlikely as the collision energy decreases. The results obtained in collisions of keV-energies show the importance of ‘transfer ionization’ as a contribution to charge-transfer processes; for instance, electron capture events which are found to cause a large shift in the recoil charge state distribution towards higher charge states. An overall correlation of the recoil ion charge states is found with the maximum internal electronic excitation energy available in the collision system. In contrast, the results obtained in collision at MeV energies show the importance of electronic excitation/ionization of the target atom for the similar shifts due to the momentum transfer to the target atom by the energetic projectile.

The recoil ion fractions for 1-electron capture channel show a bell-shaped distribution with the peak at some intermediate charge state (see figure 5c). Also, for the same incoming charge state (Si$^{12+}$), the recoil ion fractions are seen to be independent of the impact energy. The peak of the distribution for recoil fraction and the value of fractions for the higher recoil charge states are seen to depend on the charge state of the incident projectile.

Figure 8a.
ion. The distribution of recoil fractions peaks at a charge state of 6+ for an incident silicon ion of charge state 9+. The peak shows a gradual shift by a unit charge of the recoils for a unit change in the projectile charge state. However, it is noted that there occurs a saturation in the peak value of the distribution at $j = 8+$. Such a behavior can be understood if one takes into account the velocities of the incoming ion and those of electrons in various shells of the argon atom. In the present study, the velocity of incident projectile varies from 2.04 cm/nsec (60 MeV Si$^{9+}$) to 2.98 cm/nsec (120 MeV Si$^{7+}$); whereas the average velocity of the argon L-shell electron is 1.97 cm/nsec. The velocity matching between the projectile and the argon L-shell electrons suggests that the projectile should penetrate into the vicinity of the L-shell of the argon atom. Hence, as compared to the direct ionization process, which is found to occur at a large impact parameter, the capture process occurs at a much smaller impact parameter. In addition to creating a binomial distribution of M-shell electrons in such violent collisions, a hole is created due to capture of an electron from the L-shell of the argon atom [6,26]. The hole is filled by the electrons falling from the higher shells and a number of Auger electrons are ejected out. This process results into a broad charge state distribution of recoils with a peak at an intermediate charge state. The
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increase in the average recoil charge state and that in the cross-sections with the increasing projectile charge state can be understood by considering two factors [21], namely:

(i) with the increasing number of $L$-shell vacancies in the incident ion, the average number of states into which the capture of $L$-shell electrons of argon atom can take place increases,

(ii) in such capture collisions, the average impact parameter (roughly the sum of the radii of the outer shell radius of $\text{Si}^{\text{III}^+}$ and the $L$-shell of argon [27]) reduces.

The average recoil charge state corresponding to 1-electron capture is seen to shift by two units as compared to that for 2-electron capture (see, for example, $\text{Si}^{\text{II}^+}$ in figure 8a and $\text{Si}^{\text{I}^+}$ in figure 8b). Similar shifts have also been observed by other investigators [20,28] and have been explained by considering the fact that the magnitude of ionization due to vacancy cascades which follow the production of two holes in the $L$-shell is much larger than those due to the production of one hole. However, a small increase is noted in the recoil fractions for the two processes. This observation suggests that both types of the electron capture events occur in almost the same region of impact parameter space [26].

The recoil ion fractions for the electron loss events also exhibit a ‘bell shaped’ structure. Corresponding to each additional electron lost by the projectile during the interaction, the average of the charge state of the recoil ions increases by unity (see, figure 5d). This feature, as observed by other workers also [20,28], points to the fact that with the increasing number of the lost electrons from the projectile during the interaction, an additional electron is ejected from the same shell of the argon atom as it happens in the case of 1-electron loss. The average recoil ion charge state (see figure 8a and 8b) increases with the charge state of the incident projectile. This is due to the deeper penetration of the incident projectile into the target atom. It may be noted here that for the loss ionization case, the impact parameter is roughly equal to the radius of the shell from which the electron is lost and that the radius of the $\text{Si}^{\text{I}^0^+}$ ion is roughly half the radius of the $\text{Si}^{\text{I}^+}$ ion.

3.4 Recoil ion cross-sections

The partial cross-sections $\sigma_j^{q^+\lambda}$ for the recoil ion in a charge state $j$ and produced by a projectile undergoing a charge change from $q$ to $q^\prime$ during interaction with the target atom in a given ionization channel have been obtained by multiplying the fraction of the recoil ion charge state $F_j$ with the charge changing cross-section ($\sigma_j^{q^+\lambda}$) of the projectile (see, equation (4)), i.e.,

$$\sigma_j^{q^+\lambda} = F_j \sigma_j^{q^+\lambda}.$$  \hspace{1cm} (9)

The cross-sections thus obtained are plotted as a function of the recoil ion charge state and are shown in figure 9 for various ionization channels under consideration. As observed earlier, the cross-sections are strongly correlated to the final charge state of the scattered projectile. One of the interesting features observed for $\text{Si}^{\text{II}^0^+}$ ion interacting with argon is that the recoil ion distributions for $\text{le}^-\text{capture}$ and for $\text{le}^-\text{loss}$ processes are similar for the same average recoil charge state. This observation suggests that both, the loss ionization and the capture ionization processes occur at the similar impact parameters [20].
Figure 9. Cross-sections of the argon recoil ion production in 85 MeV Si$^{10+}$–Ar collisions for direct ionization, 1-electron loss and 1-electron capture events (with $q = 10+$), as a function of recoil charge state $j$. Sum refers to the cross-sections obtained for the total ionization, wherein the total recoil spectrum is considered for all the charge states of the projectiles incident on the PPAC.

4. Conclusions

In the present work, the recoil ion charge distributions for electron capture, and electron-loss and direct ionization processes have been studied in detail by performing coincidences between argon recoils and charge selected silicon ions. The energy of the incident beam of silicon projectiles was changed from 60 MeV to 120 MeV and the incident charge state was varied from 4+ to 14+.

The total ionization cross-sections are found to scale as $q^{1.7}/E_p^{0.5}$. The cross-sections for the 1-electron capture case are found to agree with the scaling rule of Schlachter et al and those for 1-electron loss with the empirical scaling rule of Alton et al. The ratio of the cross-sections for the 2-electron capture to the 1-electron capture channel is seen to be 0.24 which is in good agreement with the values reported by earlier workers.

The distributions for the fractions of recoil ions in case of the total ionization and that of the direct ionization processes show the presence of a ‘shell-effect’. While the probability
of production of higher charge states increases with the incident projectile charge state for the total ionization case, the distributions are found to be independent of the beam parameters for the direct ionization process. Also, in the case of total ionization, recoil fractions are found to vary as $1/j^2$. This dependence of fractions on the recoil charge state shows poorer agreement for the lower charge states of the projectile in the direct ionization channel. However, for higher incident charge states, the disagreement is seen to gradually reduce. The distributions for the 1-electron capture and for the 1-electron loss channels are seen to be bell-shaped with the peak at some intermediate charge states. The peak is dependent on the incident projectile charge state. A shift of two charge states in the peak of the distribution is found on comparing the 1-electron capture and 2-electron capture recoil spectra. The variation in cross-sections for argon recoil ion and the average recoil ion charge states for the 1-electron capture and for the 1-electron loss are seen to be similar for $\text{Si}^{10+}$ ions interacting with argon.

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References


  P Richard (Academic, New York, 1975)
  **A27**, 3372 (1983)
[23] C D Alton, L B Bridwell, M Lucas, C D Oak, P D Miller, C M Jones, Q C Kessel, A A Antar
  National Laboratory (1970)
  Rev.* **A38**, 2674 (1988)