Chapter 5

Measurements of x-ray cross section

The study of ion-atom collisions plays an essential role in developing new models for ion-atom interactions and in providing important atomic data and cross sections for other areas of science, such as atomic physics, astrophysics, plasma physics, elemental analysis, medical research, environmental protection, and industrial processing [1-6]. The theoretical description of inner-shell vacancy production in fast heavy ion-atom collisions is an extremely challenging problem due to its many-body nature and because multiple mechanisms contribute to the process. Development of a detailed understanding of inner-shell vacancy production in ion-atom collisions requires knowledge of the systematic behavior of cross sections for electron excitation, ionisation, and capture, along with the rate constants for radiative, nonradiative and collisional decay of the various excited states produced in both collision partners. So far, rigorous treatments that account only for the interactions between a single active target electron and a bare projectile nucleus have been attempted. While methods based upon the Born approximation, such as the semiclassical approximation (SCA) [7-12] and the perturbed stationary
state approximation (PSS), which incorporates the increased binding and polarisation effects of the projectile, along with corrections for projectile energy loss (E), Coulomb deflection (C) effects in the projectile trajectory, and relativistic (R) effects (ECPSSR theory) [13-18], have been most successful at predicting ionisation cross sections for projectiles of low atomic number. But in case of heavy ions with higher energies, the experimental data tend to deviate considerably from the prediction of these theories. Therefore, a detailed experimental examination of the dependence of inner-shell vacancy production cross sections on projectile energy and atomic number, and on the target atomic number, is desirable to stimulate further theoretical progress.

During ion-atom collision, ionisation of an inner shell electron of the target atom is produced by two processes; direct ionisation (DI) and electron capture (EC). The DI process is essentially the direct transfer of momentum to the bound electrons of the target by the incident projectile ion due to the Coulomb interaction whereas EC process is due to the capture of an bound state electron of the target atom to an unoccupied state of the projectile ion. The DI process is independent of projectile charge state and is the principal mechanism of ionisation for \( Z_1 \ll Z_2 \) and \( v_1 \gg v_{2s} \) where \( Z_1 \) and \( Z_2 \) refer to projectile and target atomic numbers while \( v_1 \) and \( v_{2s} \) (\( s = K, L, M... \)) are the velocities of the projectile and target inner shell electrons respectively. The EC contribution to the target inner shell ionisation is dependent upon projectile charge state and becomes significant for \( Z_1 \leq Z_2 \) and \( v_1 \leq v_{2s} \). The violent nature of the heavy ion collision creates multiple vacancies in the target atom. Aside from the increase in the electron binding energy that shifts the emitted x-rays towards higher energy, removal of the additional electrons in multiple ionisation affects the x-ray process by reducing the pool of outer shell electrons. The ionisation cross section also depends upon target thickness. As, for thick solid targets, vacancies in the projectile \( K \)-shell could be
created, which enhance the EC process resulting in an increase of ionisation cross section. The $L$-shell ionisation as well as x-ray production cross sections using heavy ion beams indicates large deviations from theoretical estimates. The effect of multiple ionisation on the radiative rates from outer $N$ and $O$ shell is yet to be studied extensively. Also, the Coster-Kronig transition probabilities of different sub-shells as well as the radiative widths of different lines are expected to change significantly. Therefore, there may be a significant change in the relative intensities of different $L$ x-ray lines.

Multiple ionisation of the target atom, induced by heavy projectiles, reduces the pool of outer shell electrons and consequently reduces the probability of Auger process. The Auger process involves two electrons whereas in radiative process only one electron is involved, the amplification of the later leading to an enhanced fluorescence yield has been observed. Apart from this, due to multiple ionisation, there is an increase in the binding energies of the electrons which leads to the shifting of emitted x-ray lines towards higher energy region. Furthermore this increase in binding energy reduces the energies of the Coster-Kronig (CK) electrons leading to the closure of the CK transitions which are being open for the single vacancy configuration.

Studies conducted for the $L$ shell ionisation by heavy ions show a large deviation from theoretical predictions. The enhancement in $L1$ sub-shell ionisation reported by Braziewicz et al [19] has been attributed to multiple ionisation of outer shells and thus it leads to enhancement in fluorescence yields. $L$ shell ionisation studies by other groups also show similar enhancement in cross sections as compared to that of the theory [20-22]. The role of multiple ionisation effects in Au sub-shell ionisation by oxygen ions [23] shows enhancements in $L$ sub-shell fluorescence yields and a decrease in Coster-Kronig transition probabilities.

The hyperfine interaction in He-like ions induces a small admixture of 1s2p
$^{13}\text{P}_1$ state to the $1s2p\ 3\text{P}_0$ state. Therefore it has been observed that, the forbidden E1 transitions from the $1s2p\ 3\text{P}_0$ state to $1s2\ 1\text{S}_0$ ground state are allowed. In light of this finding it is expected that, in case of single vacancy condition the effect of hyperfine interaction can increase the radiative transition probability.

Due to splitting of 2p levels the number of possible transitions from 2p$_{3/2}$ level increase in comparison to that of 2p$_{1/2}$ level. For example, in an isotope of Gd with non-zero nuclear spin (say $^{157}\text{Gd}$, I = 3/2), there will be a change in total angular momentum due to hyperfine interaction. This will lead in splitting of the 2p$_{3/2}$ level will split in to four levels with J values from 0 to 3 and the 2p$_{1/2}$ level in to two with J values 1 and 2 as shown in Figure 5.1. As is evident from the figure the original single E1 transition from 2p$_{3/2}$ to 1s$_{1/2}$ ($K\alpha_1$) is now replaced with a total of 8 transitions (6 E1 and 2 M2) and for 2p$_{1/2}$ there are four E1 transitions. Therefore it is expected that the intensity ratio of $K\alpha_1$ to $K\alpha_2$ may increase for elements with a non-zero nuclear spin.

The objective of the present work is to measure the $K$ and $L$ shell x-ray production cross sections for rare earth elements induced by Li ions and observe
the effect of multiple ionisation in radiative process. Furthermore, use of enriched isotopic targets to compare the intensity ratios of different $K$ x-ray lines to find out any signature of the effect of hyperfine splitting. The ion beam energy was kept above 3 MeV/u so that the effect of target sub-shell coupling resulting in rearrangement of the initial vacancies is avoided.

5.1 Experimental Details

The experiment has been carried out using the 15UD pelletron accelerator at Inter University Accelerator Centre (IUAC), New Delhi. Details of the experimental set up are discussed in chapter 3. Ion beam of $^7$Li$^{3+}$ of energy 28, 30, 32, 33.5, 35, 42 and 45.5 MeV have been used for the measurements. Both the Si(Li) and HPGe detectors were used to record the x-ray spectra of the targets. The target thicknesses were 6 $\mu$g/cm$^2$ Ti on carbon backing, 50 $\mu$g/cm$^2$ Ni on carbon backing, 91.7 $\mu$g/cm$^2$ Ba on carbon backing, 25.3 $\mu$g/cm$^2$ $^{157}$Gd on aluminium backing, 97.8 $\mu$g/cm$^2$ $^{160}$Gd on aluminium backing and 178 $\mu$g/cm$^2$ Au on carbon backing. Except the Ba target (in the form of BaCl$_2$), all other targets were elemental. The targets were mounted on the target ladder and placed at an angle of 45° with respect to the incident beam.

The Si(Li) detector gain was set for 45 keV x-rays at the maximum and the HPGe detector was set for 100 keV x-rays at the maximum. Therefore, while the HPGe detector recorded both the $K$ and $L$ x-rays from every target, the Si(Li) detector recorded only the low energy x-rays i.e. $K$ x-rays of Ti, Ni & Ba and $L$ x-rays of Ba, Gd & Au. The beam current was kept between 1 - 5 pnA according to the target in position, so that the count rate in both the x-ray detectors remains satisfactory with a dead time of $< 1\%$. As the solid angle subtended by the SSB detectors was very small, no such situation raised in case of their case. Typical $K$
and $L$ x-ray spectra from the targets are shown in Figure 5.2 - 5.5.

![Graph of x-ray spectra](image)

Figure 5.2: The $K$ x-ray spectra from Ni target in collision with 28 MeV $^7\text{Li}^{3+}$ ions

Efficiencies of the x-ray detectors were determined before the experiment as described in section 3.7. The measurement for the HPGe detector was carried out at the experimental setup by placing the radioactive source at the target position. During the experiment, except for the distance between the source and detector, everything else remained unchanged. The distance between the target and the HPGe detector was increased to 55 mm (instead of 43 mm during the efficiency measurement) in order to reduce the dead time of the detector and data acquisition system which was considerably high due to the high count rate. Therefore, after the experiment, the efficiency of the detector was again determined with $^{57}\text{Co}$ source and the previous data was normalised to the new values. The new efficiency
5.1. Experimental Details

Figure 5.3: The (a) $K$ x-ray and (b) $L$ x-ray spectra from Ba target in collision with 35 MeV $^7$Li$^{3+}$ ions.
Figure 5.4: The (a) $K$ x-ray and (b) $L$ x-ray spectra from Gd targets in collision with 45.5 MeV $^7\text{Li}^{3+}$ ions.
Figure 5.5: The (a) $K$ x-ray and (b) $L$ x-ray spectra from Au target in collision with 33.5 MeV $^7\text{Li}^{3+}$ ions.
5.1. Experimental Details

curve is shown in Figure 5.6.

![Figure 5.6: The total efficiency of the HPGe detector for the target - detector geometry during the experiment.](image)

The Si(Li) detector was positioned at a distance of 155 mm from the target and equipped with a 6 mm diameter collimator. During the experiment, the dead time of the detection system increased with increase in the incident beam energies. To avoid this, the distance between the target and the Si(Li) detector was increased to 290 mm for the beam energies $> 30$ MeV. Except for the $^{152}$Eu source, the activities of other radioactive sources were quite low. Therefore it was not possible to get enough statistics from the Si(Li) detector by placing the sources at the target position, within a reasonable time period, in order to determine the efficiency points. Therefore, the efficiency data was taken with the $^{152}$Eu source at the target position and the previous data were normalised with the new efficiency
5.1. Experimental Details

points. This procedure was followed for both the distances of 155 mm and 290 mm as shown in Figure 5.7. Fortunately, the new efficiency curves matched well with the theoretical calculations for the new source - detector geometry similar to the previous measurements reported in section 3.7.

![Efficiency Curves](image)

Figure 5.7: The total efficiency of the Si(Li) detector for the target - detector geometry during the experiment.

Although the efficiency points for both the detectors follow a certain trend, the fitting of those points to the standard model failed for the HPGe detector as discussed in section 3.7. The modified model for the detector also showed considerable deviation from the measurements by up to 30% in the low energy region and around the Ge K absorption edge region. Though it is possible to fit the HPGe data with any arbitrary function, its difficult to precisely predict the behaviour around the Ge K edge leading to very high uncertainties in the region
of 10 - 14 keV. But the modelled data matched well for the higher energy region beyond 30 keV. Whereas, the model for the Si(Li) detector matched quite well with the measured data within 3% uncertainty. But the efficiency of this detector drastically decreases in the higher energy region of > 20 keV. Considering the above facts, the low energy x-ray spectra i.e. the K x-rays of Ti & Ni and L x-rays of Ba, Gd & Au, obtained from the Si(Li) detector and the x-ray spectra in the higher energy region i.e. the K x-rays of Ba, Gd & Au were used for the extraction of the x-ray production cross section values.

The x-ray production cross section was evaluated from the measure yield of a particular line using the following relation:

\[
\sigma^x = \frac{Y_x \sigma_R(\phi)d\Omega_p}{\epsilon N_p A_s} \tag{5.1}
\]

Where, \( \sigma^x \) is the x-ray production cross section for the x-ray peak of interest; \( \sigma_R(\phi) \) is the Rutherford scattering cross section at an angle 15.5° (for the present measurement), \( d\Omega_p \) is the solid angle subtended by the particle detector at the target and \( Y_x \) is the peak integral of the x-ray line. With the peaks due to the elastic scattering from the target and its backing material well separated, \( N_p \) is the number of the projectiles that scatter elastically from the target element. \( A_s \) is the correction factor due to slowing down of the projectiles in the target of finite thickness, which includes corrections due to self absorption of the x rays in the target, \( \epsilon \) is the total efficiency of the x-ray detector including the fractional solid angle. No corrections were made for the projectile energy loss as; in the present measurements it was estimated to be less than 0.3% in all the cases. The correction for self-absorption of the x-rays in the target which, for uniform x-ray
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production through out is given by,

\[ A_s = \frac{1}{\mu d} \left(1 - e^{-\mu d}\right) \]  \hspace{1cm} (5.2)

Here \( \mu \) is the mass absorption coefficient taken from the XCOM [24] data base and \( d \) is the target thickness in g/cm\(^2\). For the present measurements, this largest self-absorption correction factor was for the \( Ll \) x-ray line of Ba and amounted to be 2.8\%. For the instances when the projectile energy is higher than the Coulomb barrier of the target projectile combination, total integrated charge from the Faraday cup was used instead of the elastic scattering data to obtain the cross section. In order to check the reliability of the current integration, the Au \( K \) x-ray production cross section at 28 MeV beam energy was evaluated by both the methods and the values agreed within 2\%. In addition to this, the x-ray production cross sections extracted from the data recorded by both the detectors were also compared. The \( K\alpha \) x-ray production cross section of Ni and Ba were extracted from the spectra recorded by both the Si(Li) and HPGe detector and the ratio between them is compared at different beam energies as shown in Figure 5.8.

It can be seen that both the cross sections are quite comparable to each other with their ratios being close to unity. The Ni data for the HPGe detector is generally less than that of the Si(Li) data. This may be due to the overestimation of the HPGe detector efficiency points for the Ni \( K \) x-ray energies of 7.48 keV and 8.26 keV, which were extracted by extrapolation of the experimental data. In this region, there was no experimental point and the disagreement between the model prediction and the experimental data is very high. On the other hand, the Ba data has also shown good agreement between both the detectors except that the Si(Li) data is little less than the HPGe data. The Ba \( K \) x-ray spectra in the Si(Li) detector suffers from very poor statistics due to the low total efficiency.
5.2 Analysis and results

5.2.1 $K$ shell ionisation

As shown in Figures 5.2-5.5, the $K\alpha$ and $K\beta$ lines are well separated for all the targets. In case of Gd and Au the $K\alpha_1$ and $K\alpha_2$ lines are also resolved. The
5.2. Analysis and results

intensities of the $K\alpha$ and the $K\beta$ group of lines were obtained separately using a
standard peak-fitting program. Using Equation (5.1) the x-ray production cross
section for both the $K\alpha$ and $K\beta$ group of x-rays were extracted. The measured
$K$ shell x-ray production cross sections for different elements are tabulated in
Table 5.1. The overall experimental errors in the measured cross sections were

Table 5.1: $K$ shell x-ray production cross sections (in barns) for Ti, Ni, Ba, $^{157}$Gd,
$^{160}$Gd and Au induced by Li$^{3+}$ ions.

<table>
<thead>
<tr>
<th>E (MeV)</th>
<th>Ti</th>
<th>Ni</th>
<th>Ba</th>
<th>$^{157}$Gd</th>
<th>$^{160}$Gd</th>
<th>Au</th>
</tr>
</thead>
<tbody>
<tr>
<td>28</td>
<td>7233</td>
<td>1944</td>
<td>6.83</td>
<td></td>
<td></td>
<td>0.424</td>
</tr>
<tr>
<td>30</td>
<td>2167</td>
<td>2.01</td>
<td>1.84</td>
<td>0.437</td>
<td></td>
<td></td>
</tr>
<tr>
<td>32</td>
<td>7601</td>
<td>2701</td>
<td>12.84</td>
<td></td>
<td>0.521</td>
<td></td>
</tr>
<tr>
<td>33.5</td>
<td></td>
<td></td>
<td></td>
<td>0.672</td>
<td></td>
<td></td>
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<td>35</td>
<td>8240</td>
<td>3495</td>
<td>16.44</td>
<td>3.14</td>
<td>3.14</td>
<td>0.697</td>
</tr>
<tr>
<td>42</td>
<td>4688</td>
<td>23.28</td>
<td>7.05</td>
<td>6.67</td>
<td></td>
<td></td>
</tr>
<tr>
<td>45.5</td>
<td>4988</td>
<td>9.17</td>
<td>8.83</td>
<td>1.490</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

estimated to be about 7 - 12% arising from the uncertainties in the determination
of the absolute efficiency of the detector, total charge collected in the Faraday cup
and the target thickness. The ionisation cross sections ($\sigma_k^I$) were obtained using
the following relation;

$$\sigma_k^I = \frac{\sigma_{kj}^x}{\omega_k F_{kj}} \quad (j = \alpha, \beta)$$  \hspace{1cm} (5.3)

Here, $\sigma_{kj}^x$ is production cross section, $\omega_k$ is the $K$-shell fluorescence yield, and
$F_{kj}$ is the fractional emission rate for the $Kj$ group of x-rays. Single vacancy
fluorescence yield tabulated by Krause [25] were used for the $\omega_k$ values whereas,
the Dirac-Fock values for the $K$ x-ray emission rates computed by Scofield [26]
and further revised by Campbell et. al. [27] were used for the value of $F_{kj}$. Thus obtained ionisation cross sections for different elements were compared with the predictions from first Born approximation (FBA) and ECUSAR [28] theories in Figures 5.9 - 5.14.

![Graph](image)

Figure 5.9: $K$ shell ionisation of Ti by Li ions. The solid line represents the predictions by ECPSSR theory and the dotted line is for the FBA predictions.

In these figures, the $K$-shell ionisation cross sections for all the targets have been plotted as a function of projectile energy. It is observed that the cross sections were well reproduced by the ECUSAR theory in general than the FBA. For the low $Z$ elements like Ti and Ni, as shown in Figures 5.8 and 5.9, both the FBA and ECUSAR predictions grossly underestimate the measurement. The deviations are more pronounced in case of Ni, where the experimental data is under predicted by the ECUSAR by about 20%. In the present experimental scenario, the $v_1/v_2$ for Ti and Ni ranges from 0.51 to 0.74. So in this region of velocities and asymmetric
(Z_1/Z_2 < 1) collision conditions only the direct ionisation phenomena is expected to be prevalent in the K shell ionisation process. Therefore, the experimental data should match well with the theoretical predictions. In the case of Ba, Gd and Au targets, the v_1/v_2s value ranges from 0.16 to 0.29 and Z_1/Z_2 \ll 1 which indicates the direct ionisation process to be most prominent. In case of Au the ECUSAR predictions provide excellent match to the experimental data for all the energies (Figure 5.14) whereas both the theories overestimate the data for Ba as well as for both the isotopes of Gd in the lower energy regime (Figures 5.10 -12). In the higher energy regime, the ECUSAR predictions are closer to the measurements for Ba and Gd.

The deviations of the experimental data from theoretical predictions for Ti and Ni might be due to the simultaneous L shell ionisation along with the K
Figure 5.11: Same as that of Figure 5.9 except for Ba target.

shell. As the $K$ shell fluorescence yield for these two elements are quite small, a small fraction of the simultaneous $L$ shell ionisation has considerable effect on it. The measurements of Li et. al. [29, 30] suggest considerable enhancements in the fluorescence yields of multiply ionised atoms. The best way to observe the effect of multiple ionisation of target inner shells is by measuring energy shift of the $K\beta$ line and the variation of the width of its peak. In the experimental conditions of the present measurements, it was not possible to measure neither the energy shift of the change in the width of the $K\beta$ line unambiguously. So the only way remained was to measure the intensity ratios of the $K\beta$ line to that of the $K\alpha$ line for a qualitative estimation of multiple ionisation. The measured $K\beta/K\alpha$ intensity ratios of different elements are tabulated in Table 5.2.

The $K\beta/K\alpha$ ratios for Ni and Ba show considerable enhancements from the
theoretical values whereas in case of both the isotopes of Gd they are less than the theoretical values. But in case of Au the measured values match excellently with the theoretical values. The same trend is observed in case of their $K$ shell ionisation cross sections where the Au data matches well with the theoretical values whereas the Ni data is consistently higher than the theoretical predictions. This deviation can be attributed to the simultaneous ionisation of Ni $L$ shell along with the $K$ shell which in turn affects the level widths for both the Auger and radiative processes. The former process, being a two electron process, is affected more than the later one and hence the fluorescence yield gets enhanced. In their measurements with 2.35 MeV/amu carbon projectiles, Li et.al. [30] observed an increase in $K\beta/K\alpha$ intensity ratio by 24% and 18% for Ti and Ni respectively. In the present experiment the observed deviation of the Ni ionisation cross section can
be attributed to the enhancement of fluorescence yield due to multiple ionisation.

5.2.2 \( L \) shell ionisation

From Figures 5.2-5.5, it can be observed that the dominant components of the \( L \) x-rays of all three elements are well resolved. The separation between the components increases as the target \( Z \) increases. The peak areas in the x-ray spectra were estimated using a multi-Gaussian least-squares-fitting program with the possibility of choosing variable widths of the lines and background subtraction. From the measured x-ray yields, the x-ray production cross sections were estimated using the relation given in Equation (5.1). The measured x-ray production cross sections for the most commonly resolved \( L\alpha \), \( L\beta \), and \( L\gamma \) peaks plus the \( L\gamma \) subpeaks are listed in Table 5.3.
5.2. Analysis and results

Figure 5.14: Same as that of Figure 5.9 except for Au target.

The x-ray production cross sections \( \sigma^x_p, \ p = \alpha, \beta, \gamma, \ldots \) for the most commonly resolved \( L\alpha, \ L\beta, \) and \( L\gamma \) series peaks are then related to the three subshell ionisation cross sections \( \sigma^I_i \ (i = 1,2,3) \) and the various atomic parameters in the following ways:

\[
\sigma^x_\alpha = [\sigma^I_1 (f_{12} f_{23} + f_{13}) + \sigma^I_2 f_{23} + \sigma^I_3] \omega_3 S_{\alpha,3}, \tag{5.4}
\]

\[
\sigma^x_\beta = \sigma^I_1 [\omega_1 S_{\beta,1} + f_{12} \omega_2 S_{\beta,2} + (f_{12} f_{23} + f_{13}) \omega_3 S_{\beta,3}] + \sigma^I_2 \omega_3 S_{\beta,3} \tag{5.5}
\]

\[
\sigma^x_\gamma = \sigma^I_1 [\omega_1 S_{\gamma,1} + f_{12} \omega_2 S_{\gamma,2}] + \sigma^I_2 \omega_2 S_{\gamma,2} \tag{5.6}
\]

\[
\sigma^x_{\gamma,1} = \sigma^I_1 f_{12} \omega_2 S_{\gamma,1,2} + \sigma^I_2 \omega_2 S_{\gamma,1,2} \tag{5.7}
\]
Table 5.2: Measured $K\beta/K\alpha$ intensity ratios for different elements induced by $^7$Li$^{3+}$ ions. Values given just below the element (second row from top) are the theoretical values.

<table>
<thead>
<tr>
<th>E (MeV)</th>
<th>Ti</th>
<th>Ni</th>
<th>Ba</th>
<th>157Gd</th>
<th>160Gd</th>
<th>Au</th>
</tr>
</thead>
<tbody>
<tr>
<td>28</td>
<td>0.143</td>
<td>0.15</td>
<td>0.276</td>
<td></td>
<td></td>
<td>0.271</td>
</tr>
<tr>
<td>30</td>
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<td>0.194</td>
<td>0.23</td>
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<tr>
<td>32</td>
<td>0.138</td>
<td>0.144</td>
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</tr>
<tr>
<td>33.5</td>
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<td></td>
<td></td>
<td>0.27</td>
</tr>
<tr>
<td>35</td>
<td>0.129</td>
<td>0.147</td>
<td>0.261</td>
<td>0.21</td>
<td>0.241</td>
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</tr>
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<td></td>
<td>0.273</td>
</tr>
<tr>
<td>45.5</td>
<td>0.151</td>
<td></td>
<td>0.227</td>
<td>0.229</td>
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<td>0.271</td>
</tr>
</tbody>
</table>

$$\sigma_{\gamma_{5}} = \sigma_{1}^{I}f_{12}\omega_{2}S_{\gamma_{5},2} + \sigma_{2}^{I}\omega_{2}S_{\gamma_{5},2}$$  \(\text{(5.8)}\)

$$\sigma_{\gamma_{2+3}} = \sigma_{1}^{I}\omega_{1}S_{\gamma_{2+3},1}$$  \(\text{(5.9)}\)

$$\sigma_{\gamma_{4+4'}} = \sigma_{1}^{I}\omega_{1}S_{\gamma_{4+4'},1}$$  \(\text{(5.10)}\)

Where $S_{p,i}$ is the fraction of the radiative transition to the $i$th subshell associated with the $Lp$ peak (e.g., $S_{\alpha,3} = (\Gamma_{\alpha1} + \Gamma_{\alpha2})/\Gamma_{3}$, where $\Gamma$’s are the radiative widths), $\omega_i$ is the fluorescence yield of the $i$th subshell and $f_{ij}$ are the Coster-Kronig transition probabilities. Equations (5.4) - (5.10) link the experimentally measured line intensities to the three unknown subshell ionisation cross sections $\sigma_{i}^{I}$ ($I = 1, 2, 3$).

Several combinations of these seven equations can be used to solve for the experimentally measured ionisation cross sections. Therefore, one has a choice of a particular set of peaks, which may be used to deduce the ionisation cross-sections.
5.2. Analysis and results

Table 5.3: Measured $L$ x-ray production cross sections (in barns) for different elements induced by $^7$Li$^{3+}$ ions

<table>
<thead>
<tr>
<th>E (MeV)</th>
<th>$L\alpha$</th>
<th>$L\beta$</th>
<th>$L\gamma_1$</th>
<th>$L\gamma_2+3+4,4'$ or $L\gamma_4,4'$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$L\gamma_{2+3+6}$</td>
</tr>
<tr>
<td>Ba</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>28</td>
<td>3147.3</td>
<td>2687.5</td>
<td>240.8</td>
<td>142.4</td>
</tr>
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<td>3223.5</td>
<td>2877.4</td>
<td>324.8</td>
<td>160.8</td>
</tr>
<tr>
<td>35</td>
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<td>2994.7</td>
<td>325.7</td>
<td>172.7</td>
</tr>
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<td>3467.2</td>
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<td>333.7</td>
<td>199.3</td>
</tr>
<tr>
<td>$^{157}$Gd</td>
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<td>3411.7</td>
<td>2263.3</td>
<td>165.5</td>
</tr>
<tr>
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sections. Different sets of equations can give rise to a large spread in the final results and may account for some of the discrepancies, which often amount to several times the quoted errors [31]. However, four most commonly used methods
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are discussed below.

Method 1 or the $\alpha\beta\gamma_{\text{total}}$ method. Most of the $L$ x-ray spectrum is split into three groups, namely $L\alpha$, $L\beta$, and $L\gamma$ regions. Equations (5.4) - (5.6) are used to solve for ionisation cross sections. Since all the major $L$ lines are used in this technique, statistical counting errors and background subtraction problems are minimal here. One main disadvantage of this method is the usage of the maximum number of atomic parameters, which are known for single vacancy atoms only. For heavy-ion-induced collision, multiple ionisation causes additional complications since the atomic parameters are changed, which ultimately affects the cross-section values. Furthermore, it has been shown [31-33] that particle-induced x rays originating from the states with $j = 3/2$ are anisotropic. Therefore, using this method to extract ionisation cross sections is not the most desirable. In the present work, only the $L\alpha$ and $L\gamma$ lines will be used for the extraction of subshell ionisation cross sections. $L\gamma$ lines originate from the states with $j = 1/2$, and so their cross sections are totally isotropic. $L\alpha$ x rays are practically isotropic because its $L\alpha_1$ and $L\alpha_2$ components have opposite anisotropy.

Method 2 or the $\alpha\gamma_{1,4,4'}$ method. In this approach, the intensity of the $L\gamma_1$, $L\gamma_{4,4'}$, and $L\alpha$ peaks is needed and Equations (5.4), (5.7), and (5.10) are used. Compared to method 1, here the number of atomic parameters used is much lower, but the main disadvantage is that the total counts under the $L\gamma_{4,4'}$ peak are much smaller than those in the $L\alpha$ or $L\gamma_1$ peaks. Thus the method suffers from poor statistics. Moreover, the peak is situated at the falling portion of the $L\gamma_{2,3,6}$ peak. Inaccurate background subtraction for such a small peak introduces a large error in the determination of $\gamma_{4,4'}$ and hence $\sigma_I$. This situation is more aggravated because of larger widths as a result of multiple ionisation. However, this is the most favoured method provided good statistics under the $L\gamma_{4,4'}$ line is achieved, and in fact it has been used by several experimentalists.
Method 3 or the $\alpha \gamma_1 \gamma_{total}$ method: In this approach, the intensities of the $L\gamma_1$, the total $L\gamma_{total}$, and the $L\alpha$ peaks are needed for which Equations 5.4, 5.6, and 5.7 are used. Compared to method 1, the atomic parameters used here are much less in number. Since $L\gamma_1$ is the strongest line among the $L\gamma$ structure, there is also no problem of Gaussian peak fitting or background subtraction. But compared to method 2, the number of atomic parameters used here is higher.

Method 4 or the $\alpha \gamma_1 \gamma_{2+3}$ method. The intensities of $L\gamma_1$, $L\gamma_{2+3}$, and $L\alpha$ peaks are needed and Equations (5.4), (5.7) and (5.9) are used in this method. In case of the $L$ x-ray spectra of most elements detected using an energy dispersive detection system, the $L\gamma_6$ line and the $L\gamma_{2+3}$ line is not resolvable and form a composite peak called $L\gamma_{2+3+6}$. Now, the $L\gamma_{2+3}$ line originates due to a vacancy in the $L1$ subshell, while $L\gamma_6$ is from the $L2$ subshell. So to obtain the sub-shell ionisation cross sections one has to subtract the contribution from the $L\gamma_6$ line. In order to achieve this, generally, Datz’s prescription [34] to estimate the contribution of the $L\gamma_6$ intensity in the $L\gamma_{2+3+6}$ is followed. According to this technique, a straight-line fit to the intensity ratio $y \equiv (I_{\gamma_{2+3+6}}/I_{\gamma_1})$ plotted against $x \equiv (I_{\gamma_{4,4'}}/I_{\gamma_1})$ yields the ratio of the $L\gamma_6$ intensity relative to the $L\gamma_1$ intensity. It is the intercept of that line with the $y$ axis. But in this method if the intensity of the $L\gamma_{4,4'}$ is low and there are few number of data points then the error bar in the x-axis will lead to some unrealistic value of the $I_{\gamma_6}/I_{\gamma_1}$ ratio.

In the present measurement the $L\gamma_{4,4'}$ peak of Au is well resolved by the Si(Li) detector and the intensity of the $L\gamma_{4,4'}$ peak is more than 10000 (amounting to < 1% statistical uncertainty) for every projectile energies. In case of the Gd $L$ x-ray spectra the $L\gamma_{4,4'}$ intensity is very low and not so well separated from the $L\gamma_{2+3+6}$ peak. Also the $L\gamma_1$ and $L\gamma_5$ lines, being closely spaced, form a single peak. Though there is no $L\gamma_6$ line in case of Ba $L$ x-ray spectra because of the absence of the $OIV$ electron in its shell, the $L\gamma_{4,4'}$ line merges with the $L\gamma_{2+3}$ peak and the
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$L\gamma_1$ and $L\gamma_5$ form a single peak as in case of Gd. Hence, for the extraction of $\sigma_i^I$ ($i=1,2,3$) ionisation cross sections for all the elements, the $\alpha\gamma_1\gamma_{total}$ method was employed with a little modification as the $L\gamma_1$ and $L\gamma_5$ components of two elements merge together in the spectra. Therefore instead of using Equation (5.7) alone, Equations (5.7) and (5.8) are combined together and used along with Equations (5.6) and (5.4) to extract the $\sigma_i^I$ ($i=1,2,3$) value. Now onwards this method is referred as the $\alpha\gamma_1+\gamma_{total}$ method. Furthermore, in order to check the agreement between various methods the $L$ subshell ionisation cross sections for Au and Ba were derived using two methods namely $\alpha\gamma_1\gamma_{4,4'}$ and $\alpha\gamma_1\gamma_{total,1}$ and $\alpha\gamma_1\gamma_{2+3+4,4'}$ and $\alpha\gamma_1\gamma_{total}$ respectively. As the $L\gamma_{2+3}$ and $L\gamma_{4,4'}$ lines form a single peak in the $L$ x-ray spectra of Ba, Equations (5.9) and (5.10) were merged together and used along with Equations (5.6) and (5.4) to extract the ionisation cross sections. The subshell ionisation cross sections obtained by both the methods are inside each others error bars for both the elements as the maximum relative deviation among them is 5%. In all the methods, as the intensity of $L\gamma_1$ or $L\gamma_{1+5}$ is common, there is not much difference in the extracted $L1$ and $L2$ ionisation cross sections. The change in $L3$ cross section is very small as it is comparatively higher than the other two. This suggests that the extracted ionisation cross sections are invariably identical and regardless of the method or the number of parameters used. The only thing it is sensitive to is the recorded statistics of the line used in extraction. If the statistics under the line of interest is high then, the uncertainties in the extracted cross section values are minimal.

As noted by Cohen [35], the disagreement of the ionisation data with the predictions of different theoretical models might depend to some extent on the use of different atomic parameters. But Lapicki et. al. [36] observed that combination of different parameters resulted in a deviation of less than 10%. Therefore, in the present work the most recent atomic parameters were used in extraction of
the $Li$-subshell ($i = 1, 2, 3$) ionisation cross sections. The fluorescence yields and Coster-Kronig (CK) transition rates the values recommended by Campbell [37] and the branching ratios from the table of Campbell and Wang [38] have been used. Recently, Campbell [39] has further revised the fluorescence yields and CK transitions for the $L_1$ subshell for the elements with $Z \geq 64$. The new recommended values differ up to 10% from the previous values. The $L_1$ subshell ionisation cross sections are very sensitive to the atomic parameters and it also affect the extraction of $L_3$ subshell ionisation cross section. Therefore, the $Li$-subshell ionisation for both the Gd isotopes and Au were extracted using both set of parameters and has been observed that the $L_1$ subshell ionisation cross sections are enhanced by up to 10% and the $L_3$ ionisation cross section is decreased by up to 3%. The change in the $L_2$ ionisation cross section was less than 1%. Finally, the new set of parameters has been used for the extraction of $Li$-subshell ionisation at all the energies. In order to maintain uniformity across different targets, the extracted $Li$ ionisation cross sections using the $\alpha \gamma_1 \gamma_{total}$ total method are used in further discussion.

Comparison of the measured $L_1$, $L_2$, $L_3$ and total $L$ shell ionisation cross sections along with the predictions of FBA and ECUSAR are plotted as a function of projectile energy in Figures 5.15-5.17.

In case of Ba and both the Gd isotopes the data compares well with the predictions of the ECUSAR theory whereas the predictions of FBA overestimate the data in general. But in case of Au the theoretical predictions largely underestimate the data at all the energies. The differences between the data and the ECUSAR predictions are highest for the $L_1$ subshell cross section. In this case the data is about a factor of 2 higher than the theoretical predictions. Where as this difference reduces in case of $L_2$ and $L_3$ subshell ionisation cross sections. This discrepancy may be attributed to the effect of multiple ionisation of the Au $M$ and
Figure 5.15: The $L_1$, $L_2$, $L_3$ and total $L$ shell ionisation cross sections for Ba induced by Li ion at different energies. The solid line represents the ECUSAR predictions and the dotted line represents the FBA predictions.
Figure 5.16: Same as Figure 5.15 except for $^{157}\text{Gd}$ target.
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Figure 5.17: Same as Figure 5.14 except for $^{160}\text{Gd}$ target.
Figure 5.18: Same as Figure 5.14 except for Au target.
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Table 5.4: The ratios of the intensity of $K\alpha_2$ and $K\beta_{1,3}$ to that of the $K\alpha_1$ x-rays for both the isotopes of Gd induced by Li$^{3+}$ ions at different energies. The figures just below the column heading represent the theoretical values.

<table>
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<th>E (MeV)</th>
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<th>$^{160}$Gd</th>
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<td>$K\alpha_2/K\alpha_1$</td>
<td>$K\beta_{1,3}/K\alpha_1$</td>
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<td>0.505</td>
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<td>0.363</td>
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</table>

$N$ shells along with the $L$ shell which is being responsible for the enhancement of the $L$ shell florescence yields.

5.2.3 Isotopic effects on $K$ shell x-ray production

In the present measurement two isotopes of Gd were used for the study of inner shell ionisation in order to access the effect of hyperfine splitting in case of the inner shell x-ray emission by ion atom collisions. As discussed above, the $K$ and $L$ shell ionisation of both the isotopes follow a similar trend. It was not possible to observe the effects of hyperfine splitting from the ionisation cross sections as the uncertainty in the measurements were too high. In case of Gd, both the components of $K\alpha$ x-rays are well separated in spectra recorded by the HPGe detector (Figure 5.4). Hence, the ratio between the intensities of different components of $K$ shell x-rays can be measured which provide some evidence of the phenomena. The intensity ratios of $K\alpha_2$ and $K\beta_{1,3}$ to that of the $K\alpha_1$ for both the isotopes at different beam energies have been measured and is tabulated in Table 5.4.
It is observed that the $K\alpha_2/K\alpha_1$ values for $^{157}\text{Gd}$ are lower than that of the $^{159}\text{Gd}$ in general. The $K\beta_{1,3}/K\alpha_1$ ratio follows the same trend as the previous one. As the experimental conditions were identical in both the cases such deviation is not expected and this implies that, the enhancement of the $K\alpha_1$ intensity for $^{157}\text{Gd}$ in comparison to that of the $^{157}\text{Gd}$ may be due to the effect of hyperfine splitting which favours the emission of $K\alpha_1$ x-rays to that of the $K\alpha_2$ x-rays. But in this measurement the number of data points were quite few in order to confirm such effects. Further measurements with different targets and different beams and different energy regime is required to study this phenomena in detail.
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