**L- and M-shell x-ray production cross sections of Nd, Gd, Ho, Yb, Au, and Pb by 25-MeV carbon and 32-MeV oxygen ions**

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L- and M-shell x-ray production cross sections have been measured for thin solid targets of neodymium, gadolinium, holmium, ytterbium, gold, and lead by 25-MeV \(^{12}\)C\(^{++}\) \((q=4,5,6)\) and by 32-MeV \(^{16}\)O\(^{++}\) \((q=5,7,8)\). The cross sections were determined from measurements made with thin targets (less than 2.25 \(\mu g/cm^2\)). For projectiles with one or two \(K\)-shell vacancies, the target x-ray production cross sections were found to be enhanced over those for projectiles without a \(K\)-shell vacancy. The sum of direct ionization to the continuum (DI) plus electron capture (EC) to the \(L,M,N,\ldots\) shells and EC to the \(K\) shell of the projectile have been extracted from the data. The results are compared to the predictions of first Born theories, i.e., plane-wave Born approximation for DI and Oppenheimer-Brinkman-Kramers formula of Nikolaev for EC, and to the ECPSSR that accounts for energy loss and Coulomb deflection of the projectile as well as for relativistic and perturbed stationary states of inner-shell electrons.

**I. INTRODUCTION**

Interest in inner-shell ionization in ion-atom collisions has resulted in major advances both theoretically and experimentally.\(^{1}\) This information is important in the development of tokamaks,\(^{2}\) plasma physics,\(^{3}\) ion implantation,\(^{4}\) and in particle-induced x-ray emission (PIXE).\(^{5,6}\) Initially, the majority of the experimental work was done using low-atomic-number ions, primarily protons, to ionize \(K\)-shell electrons from target atoms.\(^{7}\) With the development of high-resolution Si(Li) detectors and the increased availability of heavy-ion beams, ionization measurements have been extended to the \(L\) shell\(^{8,9}\) and \(M\) shell.\(^{10}\)

At the time when advances were being made in the experimental studies, several theories were proposed to describe the dynamics of an ion-atom collision. The plane-wave Born approximation (PWBA)\(^{11,12}\) the binary-encounter approximation (BEA),\(^{13}\) and the semiclassical approximation (SCA) (Ref. 14) were successful in describing direct ionization (DI) of the target \(K\) shell by protons. Later the PWBA was modified\(^{15,16}\) for low relative velocity effects of enhanced target electron binding and Coulomb deflection which increase with \(Z_1/Z_2\) where \(Z_1\) and \(Z_2\) are the atomic numbers of the projectile and target atoms, respectively.

In addition to DI, electron capture (EC) to vacancies in the projectile can play an important part in ionization.\(^{17}\) The first Born calculations use the PWBA to describe DI and the Oppenheimer-Brinkman-Kramers (OBK) theory\(^{18}\) as modified by Nikolaev\(^{19}\) (OBKN) to describe electron capture. Using the perturbed-stationary-states (PSS) approach that was used in DI calculations\(^{20,21}\) for \(K\)-shell ionization, Lapicki and Losonsky\(^{22}\) have developed another approach to EC, also called ECPSSR, where energy loss (E), Coulomb deflection (C), and refinements for target relativistic effects (R) have been taken into account.\(^{23}\) It has been improved and extended to the \(L\) and \(M\) shells by Lapicki and others.\(^{24–28}\)

In the present paper we report the simultaneous measurement of \(L\)- and \(M\)-shell x-ray production for projectiles of 25-MeV \(^{12}\)C\(^{++}\) \((q=4,5,6)\) and 32-MeV \(^{16}\)O\(^{++}\) \((q=5,7,8)\) incident upon thin (less than 2.25 \(\mu g/cm^2\)) targets of \(^{64}\)Nd, \(^{66}\)Gd, \(^{67}\)Ho, \(^{70}\)Yb, \(^{76}\)Au, and \(^{82}\)Pb. The purpose of this work was to make independent determinations of the \(L\)- and \(M\)-shell x-ray production cross sections and then compare them to predictions of the first Born and ECPSSR theories. Preliminary \(L\)-shell x-ray production cross sections have been previously reported.\(^{9}\) These earlier data have been improved by using more refined data analysis techniques as discussed in Sec. II B.
II. EXPERIMENTAL DETAILS

A. Target preparation

The target foils used in this experiment were made at North Texas State University (NTSU) by vacuum evaporation of the various elements onto (5–20 μg/cm²) carbon backings. Each set of target foils ranged from approximately 0.5 to 100 μg/cm². Targets with thicknesses less than 2.25 μg/cm² were used for cross-section determinations while the thicker targets were used to study target thickness dependence and to obtain initial fit parameters during data analysis (cf. Sec. II B).

Usually target contamination is dominated by low-Z elements which have x rays at the same energies as the soft M-shell x rays. Steps taken during target preparation to reduce the presence of these contaminants are described elsewhere. They consisted primarily of repeated bathing of the carbon foils in weak solutions of warm (40–60°C) acetone. The foils, after mounting on target frames, were analyzed to ensure that they were relatively contaminant-free. The 2.5-MV Van de Graaff accelerator at NTSU was used to provide protons for target analysis by proton-induced x-ray emission.

While target contamination was being monitored, Rutherford ion backscattering was used to determine the target thicknesses. The scattered-particle yields were measured with a Si surface barrier detector placed at 150° relative to the incident ion beam direction. These yields, the theoretical Rutherford scattering cross sections, and the number of incident protons were used to determine the target thicknesses. The resulting thicknesses have a total uncertainty of ±7% due to uncertainties in source calibration (3%), solid angle (2%), ion energy (3%), and counting statistics (5%).

B. X-ray cross-section measurements

The ions used in this experiment were obtained from the 6.5-MV EN Tandem Van de Graaff accelerator at the Oak Ridge National Laboratory. Primary beams of C⁺ and O⁺ were produced at energies of 25 and 32 MeV, respectively. Each beam was then electromagnetically analyzed for the desired charge state before being focused and directed into the target chamber. Prior to entry into the target chamber, the beam was collimated to 6.4-mm diameter. A pair of carbon collimators, 12.5- and 5-cm in front of the target holder, further narrowed the beam to 3.2-mm diameter. The central target holder was oriented at 45° relative to the incident beam direction. An Ortec Si(Li) detector was positioned at 90° relative to the incident beam direction and in such a manner as to view the incident side of the target.

To obtain the best available values for the detector efficiency, a technique by Lennard and Phillips was employed which suggested using theoretical x-ray cross sections in a "reverse" calculation. Bombardment of several thin targets of low-Z elements (Al, Si, P, Cl, and K) provided K-shell x rays. The Rutherford scattered-particle yields of the projectile were collected simultaneously and were used to normalize the x-ray yields. This information, along with the theoretical x-ray production cross sections from the ECPSSR and the fluorescence yields and transition rates of McGuire were used to determine the Si(Li) detector efficiency at points below 3.3 keV. For energies above 3.3 keV, calibrated sources of Fe, Co, and Am provided x rays of known intensities from which the efficiencies were determined. The theoretical efficiency curve of the detector above 3.3 keV was obtained by calculating the x-ray attenuation in the Be entrance window, the Si dead layer, and the Au contact layer. This curve was then normalized to the data points for the radioactive sources, and the efficiencies corresponding to L- and M-shell x-ray energies were determined.

While x rays were being counted by the Si(Li) detector, Rutherford scattered particles were simultaneously being collected by a solid-state charged-particle detector positioned at either 45° or 135° relative to the incident beam direction. These results were used to find the product of the number of ions incident upon each target and the target thickness which was used to normalize the x-ray yields.

Each set of spectra, containing L- and M-shell x rays and Rutherford yields, was processed with a common configuration of signal processing equipment and then stored on the ORNL computer facilities for later recall and analysis. This equipment consisted of a PDP 11/45 computer and a Hewlett Packard CRT with light-pen capabilities. Throughout the experiment the x-ray yields were monitored for consistency. Each spectrum was displayed on the CRT and a background was drawn in by hand. The resulting yields along with initial estimates of the detector efficiency were used to monitor the experimental results as they were generated. This control was especially useful because of the long counting times required to obtain acceptable yields from the thinnest targets. After completing the experiment, an Ortec 7050 Data Acquisition and Analysis System with peak-fitting capabilities was used to analyze the relatively clean L-shell spectra. A least-squares-fitting routine (FACEFIT) (Ref. 33) gave complete analysis of each M-shell spectrum. This program was capable of fitting multiple Gaussian peaks on a linear background. A typical M-shell x-ray spectrum and the results of a fit are illustrated in Fig. 1. After initially reporting the L-shell results obtained with the Ortec system, the data were analyzed using the FACELIFT procedure. The previously extracted data obtained by the light-pen fit differed very little from that obtained by the FACELIFT procedure.

The fitting procedure for the M-shell targets consisted of using spectra of thicker targets (ρt = 2.5 to 100 μg/cm²) in the initial phase to determine line shapes and peak locations. As might be expected these thicker targets have a much higher ratio of desirable target x rays versus undesirable contaminant x rays. With thick target ratios as initial values, the fitting process for the thinner targets was expedited. The thick target yields have an estimated uncertainty of ±10%. Due to the buildup of silicon contamination (from diffusion pump oil), the uncertainty in the thinnest target yields was estimated to be ±20%. Combining the efficiency (±6%),
fluorescence yield (±5%), target thickness (±7%), and x-ray yield uncertainties (±10 or ±20%), the overall uncertainty in the x-ray cross sections is ±15% for ρt > 2.5 μg/cm² and ±23% for ρt < 2.5 μg/cm². To the extent that these thin targets represent single-collision conditions, the cross sections are a good approximation of the cross sections for vanishingly thin targets.

Because the L-shell spectra were much less contaminated with low-atomic-number elements, the uncertainties in the x-ray yields were only ±2% for all targets. The uncertainty in the efficiency of the Si(Li) detector is also better known (±3%) in the L-shell energy region. Combining these values with those for fluorescence yield and target thickness, the overall uncertainty in the L-shell x-ray cross sections is ±10% for all measurements.

III. RESULTS

Effective M-shell x-ray production cross sections as a function of target thickness are plotted in Fig. 2 for different charge state ions.14 Similar results were obtained for the L-shell spectra. The L-shell data for carbon and oxygen ions for the thinnest targets are presented in Tables I and II, respectively. The M-shell data for carbon and oxygen ions for the thinnest targets are presented in Tables III and IV, respectively. The thickness dependence of the yield must be carefully interpreted because the electron configuration of the projectile inside a solid target is not well understood.15 Early studies by Hopkins16 and Groeneveld et al.17 indicated that target x-ray yields, from ions which first passed through the carbon backing of the target foil and had more K-shell vacancies, were considerably enhanced over the target x-ray yields when the beam was incident on the target side of the foil.

This dependence of x-ray yields on projectile charge state and target thickness have also been investigated by Gray et al.18 and McDaniel et al.19 They measured target K-shell x-ray yields for projectiles with zero, one, and two K-shell vacancies as a function of target thickness. By varying the number of projectile K-shell vacancies and target thicknesses, they controlled the amount of K-shell to K-shell EC from the target to the projectile. Both groups found that x-ray yields were strongly influenced by the presence of projectile inner-shell vacancies and that the projectile charge state would equilibrate at large target thicknesses. McDaniel et al.19 compared their results favorably with the ECPSSR. McDaniel et al.26 later extended this investigation to the production of L-shell x rays and found similarly good agreement. McDaniel et al.26,39 used the thinnest possible targets (approximately 1 μg/cm²), which approximated single-collision conditions, to extract DI and EC cross sections.

The variation of M-shell x-ray production cross sections with projectile inner-shell vacancies has recently been measured10,32,40 and compared to the first Born and ECPSSR theories. Again it was found that the ECPSSR was in good agreement with the data.

In Figs. 2–6 the filled circles represent target cross sections σ(0) for projectiles with no K-shell vacancies, while the half-filled circles and open circles show target cross sections for projectiles with one σ(1) and two σ(2) K-shell vacancies, respectively. Whereas the σ(0) results in Fig. 2 are constant within statistical limits for all but the thinnest targets, the σ(1) and σ(2) results rise
TABLE I. L-shell x-ray production cross sections of several thin targets for incident projectiles of 25 MeV $^{1}C^{+}$ ($q$ = +4, 5, 6) (2.08 MeV/amu). From top to bottom, the first three cross sections represent DI + EC for zero, one, and two K-shell vacancies in the projectile. The last two groups are the inferred EC cross sections for one and two K-shell vacancies. Cross sections are in kilobarns. Uncertainties are ±10% and ±14% for the measured and inferred results, respectively.

<table>
<thead>
<tr>
<th>Targets</th>
<th>$\omega_{\text{Nd}}$</th>
<th>$\omega_{\text{Gd}}$</th>
<th>$\omega_{\text{Ho}}$</th>
<th>$\gamma_{\text{Yb}}$</th>
<th>$\gamma_{\text{Au}}$</th>
<th>$\gamma_{\text{Pb}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Z_{1}/Z_{2}$</td>
<td>0.132</td>
<td>0.125</td>
<td>0.119</td>
<td>0.114</td>
<td>0.101</td>
<td>0.098</td>
</tr>
<tr>
<td>$\rho \times 10^{4}$ (µg/cm$^{2}$)</td>
<td>0.48</td>
<td>0.45</td>
<td>0.43</td>
<td>0.41</td>
<td>0.36</td>
<td>0.35</td>
</tr>
</tbody>
</table>

significant in all targets as their thicknesses decrease. The rise in $\sigma^{(0)}$ for the thinnest targets is attributed to the increasing contribution of contaminate x rays from low-atomic-number elements. Assuming a constant amount of contaminant, the associated error will increase as the target of interest gets thinner. The disproportionately large enhancement of $\sigma^{(1)}$ and $\sigma^{(2)}$ reflects the contribution of EC cross sections when the projectiles carry K vacancies into the collision and have no chance to equilibrate in the thinnest targets.

The x-ray cross sections for all targets were determined from the results for the thinnest possible targets where approximately single-collision conditions exist. The agreement between the ECPSSR theory and data for the L shell is very good (Fig. 3) as compared with the M-shell discrepancies (Fig. 4). The M-shell data are subject to larger uncertainties than the L-shell data because of the greater complexity of their spectra, the contribu-

TABLE II. L-shell x-ray production cross sections of several thin targets for incident projectiles of 32 MeV $^{3}O^{+}$ ($q$ = +5, 7, 8) (2.00 MeV/amu). From top to bottom, the first three cross sections represent DI + EC for zero, one, and two K-shell vacancies in the projectile. The last two groups are the inferred EC cross sections for one and two K-shell vacancies. Cross sections are in kilobarns. Uncertainties are ±10% and ±14% for the measured and inferred results, respectively.

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<th>Targets</th>
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</tr>
<tr>
<td>$\rho \times 10^{4}$ (µg/cm$^{2}$)</td>
<td>1.4</td>
<td>0.8</td>
<td>0.48</td>
<td>1.35</td>
<td>0.55</td>
<td>1.24</td>
</tr>
</tbody>
</table>

$\sigma^{(0)}$ | First Born | 16.8 | 10.9 | 8.06 | 5.94 | 2.47 | 1.85 |
    | ECPSSR | 6.42 | 4.11 | 3.03 | 2.25 | 0.99 | 0.75 |
    | Expt. | 7.25 | 4.82 | 3.49 | 2.83 | 1.06 | 0.71 |

$\sigma^{(1)}$ | First Born | 32.9 | 19.3 | 13.2 | 9.04 | 3.15 | 2.25 |
    | ECPSSR | 9.10 | 5.39 | 3.78 | 2.69 | 1.08 | 0.81 |
    | Expt. | 8.56 | 5.90 | 3.57 | 2.99 | 1.12 | 0.73 |

$\sigma^{(2)}$ | First Born | 49.0 | 27.6 | 18.3 | 12.1 | 3.83 | 2.65 |
    | ECPSSR | 11.8 | 6.67 | 4.53 | 3.13 | 1.17 | 0.86 |
    | Expt. | 10.45 | 5.99 | 3.99 | 3.21 | 1.16 | 0.85 |

$\sigma^{(3)}$ | First Born | 16.1 | 8.40 | 5.14 | 3.10 | 0.68 | 0.40 |
    | ECPSSR | 2.68 | 1.28 | 0.75 | 0.44 | 0.09 | 0.06 |
    | Expt. | 1.31 | 1.08 | 0.08 | 0.16 | 0.06 | 0.02 |

$\sigma^{(4)}$ | First Born | 32.2 | 16.7 | 10.2 | 6.16 | 1.36 | 0.80 |
    | ECPSSR | 5.38 | 2.56 | 1.50 | 0.88 | 0.18 | 0.11 |
    | Expt. | 3.20 | 1.17 | 0.50 | 0.38 | 0.10 | 0.13 |
TABLE III. M-shell x-ray production cross sections of several thin targets for incident projectiles of 25 MeV $^{12}$C$^+$ ($q = +4,5,6$) (2.08 MeV/amu). From top to bottom, the first three cross sections represent DI + EC for zero, one, and two K-shell vacancies in the projectile. The last two groups are the inferred EC cross sections for one and two K-shell vacancies. Cross sections are in kilobarns. Uncertainties are $\pm 23\%$ and $\pm 33\%$ for the measured and inferred results, respectively.

<table>
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<tr>
<th>Targets</th>
<th>$^{60}$Nd</th>
<th>$^{64}$Gd</th>
<th>$^{67}$Ho</th>
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<td>0.119</td>
<td>0.114</td>
<td>0.101</td>
<td>0.098</td>
</tr>
<tr>
<td>$\sigma_{MK}^{(i)}$</td>
<td>0.48</td>
<td>0.45</td>
<td>0.43</td>
<td>0.41</td>
<td>0.36</td>
<td>0.35</td>
</tr>
<tr>
<td>$\rho t$ (µg/cm$^2$)</td>
<td>1.4</td>
<td>0.8</td>
<td>0.48</td>
<td>1.35</td>
<td>1.07</td>
<td>1.24</td>
</tr>
</tbody>
</table>

TABLE IV. M-shell x-ray production cross sections of several thin targets for incident projectiles of 32 MeV $^{16}$O$^+$ ($q = +5,7,8$) (2.00 MeV/amu). From top to bottom, the first three cross sections represent DI + EC for zero, one, and two K-shell vacancies in the projectile. The last two groups are the inferred EC cross sections for one and two K-shell vacancies. Cross sections are in kilobarns. Uncertainties are $\pm 23\%$ and $\pm 33\%$ for the measured and inferred results, respectively.

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fore a small error in the DI cross section can result in a large error in the inferred L- to K-shell EC cross section. To the extent that the cross sections \(\sigma^{(1)}\) are uncorrelated, the uncertainty of the inferred results is the square root of the sum of the squares of the individual uncertainties (i.e., \(\pm 14\%\)).

As with the L-shell data, the inferred M-shell EC cross sections (Fig. 6) are reported as the difference between the zero K-shell vacancy results \(\sigma^{(1)}_{MX}\) and the one \(\sigma^{(2)}_{MX}\) and two \(\sigma^{(3)}_{MX}\) K-shell vacancy results. Again, the uncertainty of the inferred results (\(\pm 33\%\)) is greater than the individual uncertainties (\(\pm 23\%\)). While both theories overestimate the M-shell data, the ECPSSR is closer to the data and the agreement appears to improve with increasing \(Z_1/Z_2\).

In summary the L- and M-shell x-ray production cross sections for 25 MeV \(^{12}\)C\(^{+}\) (\(q = 4, 5, 6\)) and 32 MeV \(^{16}\)O\(^{7+}\) (\(q = 5, 7, 8\)) incident upon Nd, Gd, Ho, Yb, Au, and Pb have been measured as a function of target thickness. Measurements made for the thinnest targets (0.48 to 2.25 \(\mu g/cm^2\)) were used to extract cross sections for DI plus EC to L, M, . . . shells and electron capture to the K shell. In comparing the L-shell results to the first Born and the ECPSSR theories, it was found that the PWBA plus OBK theory of Nikolaev consistently overpredicted the data by an order of magnitude or more.

![FIG. 3. L-shell x-ray production cross sections by carbon ions. The curves represent the sum of direct ionization plus electron capture from the L shell to the projectile L, M, . . . shells, \(1/2 K, L, M, . . .\) shells, and \(K, L, M, . . .\) shells for \(\sigma^{(i)}_{LX}\) \(i = 0, 1, 2\), respectively. The absolute uncertainty is \(\pm 10\%\) and is approximately the extent of the circular data points.](image1)

![FIG. 4. M-shell x-ray production cross sections by oxygen ions. The curves represent the sum of direct ionization plus electron capture from the M shell to the projectile L, M, . . . shells, \(1/2 K, L, M, . . .\) shells, and \(K, L, M, . . .\) shells for \(\sigma^{(i)}_{MX}\) \(i = 0, 1, 2\), respectively. The absolute uncertainty is \(\pm 23\%\) as shown by the representative error bars.](image2)
while the ECPSSR theory was in very good agreement with the data. For the $M$ shell the agreement between either theory and the data was not as good. Nevertheless, the ECPSSR gave better overall agreement.

**ACKNOWLEDGMENTS**

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1. Present address: Radiation Research Associates, 3815 Lisbon, Ft. Worth, TX 76107.
10/11, 181 (1985), and references therein.
33FACELIFT is a least-squares routine which fits several Gaussian peaks plus a linear background simultaneously. It was obtained from R. Watson at the Physics Department, Texas A&M University.
34Figure 2 was taken from Ref. 10.