M X-ray production in the selected light rare-earth elements by 0.75-1.75 MeV helium ions

F. Naab, J. L. Duggan, O. W. Holland and F. D. McDaniel

Ion Beam Modification and Analysis Laboratory, Department of Physics, P.O. Box 311427, University of North Texas, Denton, TX 76203-1427, USA

G. Lapicki

Department of Physics, East Carolina University, Greenville, NC 27858, USA

ABSTRACT

M-shell X-ray production cross sections for the light rare-earth elements of \( \gamma \)Er, \( \gamma \)Yb, and \( \gamma \)Lu have been measured for incident \( {}^4\)He\(^+ \) ions in the energy range from 0.75 to 1.75 MeV. The measured X-ray production cross sections are compared to the predictions of the First Born approximation, the ECPSSR theory, and the eECusPSSR theory in which the ECPPSR is modified for the exact (e) evaluation of the energy loss effect and a combined united-and-separated (us) atom treatment of the PSS effect.

Keywords: M-shell X-ray production cross section; ECPSSR theory; helium ion; rare-earth elements.

1. INTRODUCTION

Recently four groups compared measured M sub-shell X-ray production cross sections with different theories \([1,2,3,4]\). Several software programs for Particle-Induced X-ray Emission (PIXE) analyses rely on theoretical values to determine the concentration of elements in a particular sample \([5]\). Measurements to check the agreement of theory and experimental values is very important not only for the total M X-ray production, but for each group of transitions (see Figure 1). They should facilitate better PIXE discrimination in the composition of a sample where there are overlaps with X-ray emissions from other elements. In the present work, we compare our data with the First Born \([6,7]\), ECPSSR \([8,9]\) and eECusPSSR theories for M-shell peaks and total X-ray production cross sections in Er, Yb, and Lu bombarded by 0.75-1.75 MeV \( {}^4\)He\(^+ \) ions. In the eECusPSSR theory, calculations are made with the exact (e) limits for the momentum transfers instead of a prefactor of the ECPSSR theory that accounts for the energy loss effect (E), and by the replacement of the separated atom treatment of the PSS effect with a united and separated (us) atom approach as proposed by Lapicki \([10]\).

2. EXPERIMENTAL SETUP

The \( {}^4\)He\(^+ \) ion beam was produced with the 2.5 MV Van de Graaff accelerator at the University of North Texas. The X-rays were detected using an ORTEC HPGe detector, which was placed at 145° to the beam direction. The resolution of the detector was 143 eV at 5.9 keV. A surface barrier detector with a resolution of \( \sim 21 \) keV was placed on the other side of the beam direction, also at an angle of 150°. The X-ray and the Rutherford scattered helium ions were measured simultaneously allowing the X-ray production cross section to be determined independent of target thickness and integrated charge.

Using the standard technique of Lenard and Phillips \([11]\), the efficiency of the X-ray detector was measured for the K\( \alpha \) transitions of F, Na, Mg, Al, Si, P, S, Cl, K, Ca, Sc and Ti using \( {}^4\)He\(^+ \) ions at 1.5 MeV. A polynomial fit to natural logarithm of the experimental values was done using the Ispline and interp functions of Mathcad 8.0.

* Correspondence to: Fabian Naab, E-mail: fun001@unt.edu. Phone: (1) 940-565-3252.
The targets used for the efficiency measurement were all less than 10 \( \mu g/cm^2 \) thick. As a result, beam energy loss corrections and X-ray absorption were unnecessary. The Er, Yb and Lu targets ranged from 40 to 100 \( \mu g/cm^2 \) in thickness, and thus corrections for beam energy loss and X-ray absorption were included in the analysis.

3. DATA ANALYSIS

A typical M-shell X-ray spectrum for Lu induced by 1.75 MeV \(^4\)He ions is shown in Fig.1. The spectra were analyzed using the AXIL code [12]. Five transition groups were considered: peak 1 (M4-N2, M5-N3, M4-N3), peak 2 (M3-N1, M5-O3, M5-N6, M4-O2, M4-O3, M4-N6), peak 3 (M2-N1, M3-N4, M3-N5), peak 4 (M3-O1, M3-O4, M3-O5, M2-N4, M1-N2, M1-N3, M2-O1, M2-O4), and peak 5 (M1-O2, M1-O3). Due to the weak intensity of M2-O1 and M2-O4 we merge them into peak 4. We assume that the characteristic M-shell X-ray emission is isotropic. The efficiency for the energy centroid of each peak was used to calculate the corresponding cross section.

![FIGURE 1. M X-ray spectrum for Lu induced by \(^4\)He at 1.75 MeV. The transitions shown are the ones used to fit each spectrum for Er, Yb and Lu. In the inset, the measured spectrum is compared to our fit (solid lines) for its peaks and background.](image)

### TABLE 1. M X-ray production cross sections of the five transition groups for Er, Yb and Lu as a function of \(^4\)He ion energy. The cross section uncertainty for each transition group is ~17 %, ~16 %, ~16 %, ~18 % and ~22 %, respectively. The uncertainty for the total cross sections is ~16 %.

<table>
<thead>
<tr>
<th>Energy [MeV]</th>
<th>Peak 1</th>
<th>Peak 2</th>
<th>Peak 3</th>
<th>Peak 4</th>
<th>Peak 5</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Er</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.75</td>
<td>52</td>
<td>251</td>
<td>3.0</td>
<td>2.0</td>
<td>0.063</td>
<td>308</td>
</tr>
<tr>
<td>1.00</td>
<td>75</td>
<td>432</td>
<td>6.4</td>
<td>3.3</td>
<td>0.072</td>
<td>517</td>
</tr>
<tr>
<td>1.25</td>
<td>111</td>
<td>646</td>
<td>12</td>
<td>5.9</td>
<td>0.13</td>
<td>774</td>
</tr>
<tr>
<td>1.50</td>
<td>152</td>
<td>845</td>
<td>20</td>
<td>9.6</td>
<td>0.26</td>
<td>1030</td>
</tr>
<tr>
<td>1.75</td>
<td>180</td>
<td>1060</td>
<td>30</td>
<td>15</td>
<td>0.42</td>
<td>1290</td>
</tr>
<tr>
<td>Yb</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.75</td>
<td>26</td>
<td>301</td>
<td>3.2</td>
<td>1.6</td>
<td>0.033</td>
<td>332</td>
</tr>
<tr>
<td>1.00</td>
<td>60</td>
<td>549</td>
<td>6.8</td>
<td>3.0</td>
<td>0.047</td>
<td>619</td>
</tr>
<tr>
<td>1.25</td>
<td>83</td>
<td>786</td>
<td>11</td>
<td>5.4</td>
<td>0.072</td>
<td>886</td>
</tr>
<tr>
<td>1.50</td>
<td>99</td>
<td>1050</td>
<td>19</td>
<td>8.7</td>
<td>0.11</td>
<td>1170</td>
</tr>
<tr>
<td>1.75</td>
<td>118</td>
<td>1330</td>
<td>27</td>
<td>13</td>
<td>0.14</td>
<td>1490</td>
</tr>
<tr>
<td>Lu</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.75</td>
<td>30</td>
<td>288</td>
<td>3.0</td>
<td>1.5</td>
<td>0.037</td>
<td>322</td>
</tr>
<tr>
<td>1.00</td>
<td>37</td>
<td>498</td>
<td>6.1</td>
<td>2.6</td>
<td>0.049</td>
<td>543</td>
</tr>
<tr>
<td>1.25</td>
<td>66</td>
<td>744</td>
<td>10</td>
<td>4.3</td>
<td>0.065</td>
<td>824</td>
</tr>
<tr>
<td>1.50</td>
<td>91</td>
<td>1010</td>
<td>18</td>
<td>6.8</td>
<td>0.10</td>
<td>1130</td>
</tr>
<tr>
<td>1.75</td>
<td>103</td>
<td>1300</td>
<td>26</td>
<td>11</td>
<td>0.19</td>
<td>1440</td>
</tr>
</tbody>
</table>

For the determination of the X-ray detector efficiency we used the fluorescence values from Hubbell et al. [13], except for Al where we used Krause's value for solid targets 0.036 [14], which lies between 0.033 of Hubbell et al. (see Eq. 26 and Table 7 in Ref. [13]) and 0.0387 of Schönfeld and Janben [15].
Accurate fluorescence values are important since they can shift the efficiency curve by ~10% affecting the total X-ray production cross section, or change the ratio between different peaks.

4. RESULTS AND DISCUSSION

In Table 1, the X-ray production cross sections for the five transition groups and the total M-shell for Er, Yb and Lu are listed as a function of the helium ion energy. As can be seen, peak 2 is the main contributor to the total cross section. The smallest peak (M1-O2, M1-O3) is not included in Figure 3 because of its low relative intensity and hence large uncertainty. In Fig. 2 and 3, experimental and theoretical cross sections are compared. The major disagreement is for the data corresponding to peak 1 for each element. In the energy region for peak 1 of each element, it is difficult to do a good fit because of the L absorption edges for Ge, which is present in the dead layer of the X-ray detector.

For conversion of the calculated ionization cross sections to X-ray production cross sections, we used the Coster-Kronig factors and the M-shell fluorescence values by interpolation from Bambynek et al. [16], and the probabilities of radiative transitions from Chen et al. [17]. The precision of all these atomic parameters is important when comparing experimental and theoretical values.

![Figure 2](image_url)

FIGURE 2. Total M X-ray production cross sections in Er, Yb, and Lu by $^4$He ions. The values for each theory correspond to $^4$He ions with charge state +1. Reported uncertainties in Ref. [19]: 10-25%.

![Figure 3](image_url)

FIGURE 3. Ratios of experimental to theoretical cross sections for the 4 most intense groups of transitions. Left plots: ECPSSR theory. Right plots: eECusPSSR theory.

5. CONCLUSION

This work reports the X-ray production cross sections for the main transition groups in Er, Yb, and Lu bombarded by $^4$He ions, and the total M-shell X-ray production cross sections. We found no data in the literature for M-shell X-ray production for the major peaks of the spectrum for these targets and for the total M-shell X-ray production for Lu bombarded by helium ions. The theories shown overestimate the data to an increasing degree with increasing energy. Recent data for a number of rare-earth elements ionized by lithium ions in the similar velocity range show a similar trend [4]. However, the results in Ref. [18] for the M-shell ionization in ytterbium by protons in the same velocity range of the projectile are particularly interesting. These data are also overestimated by the ECPSSR theory but in a manner that...
increases with the decreasing projectile energy. A good agreement is shown for total, peak 2, peak 3 and peak 4 X-ray production cross sections for Yb and Lu. Overall, with exception of the possibly erroneous erbium datum at 2 MeV, our measurements agree with previous results from a work of our group [19] for total M X-ray production cross sections for Er and Yb. We found no other data for M-shell ionization by $^4$He ions in light ($Z_2 = 58-71$) rare-earth elements since Thornton et al. [20] published their cross sections for Tb three decades ago. The lack of such data calls for more M-shell measurements in the MeV range of our He ions—which is the most common projectile energy range in PIXE applications—and on the $Z_2 = 58-71$ targets because the rare-earth elements are not necessarily rare in PIXE analyses [21,22]. A better determination of the detector efficiency taking into account absorption edges and precise K-shell fluorescence values, with an account for the effects of multiple ionization on the assumed M-shell atomic parameters, would help to avoid systematic errors and would make a comparison of the data with theories more meaningful. An account for multiple ionization would shift the curves upward resulting in less agreement between the data and the theories for the total M X-ray production; we expect that the degree of that shift is, however, smaller than the present discrepancies between the data and the eECusPSSR theory as seen in Fig. 2.

ACKNOWLEDGMENTS

Work at UNT supported in part by NSF, ONR, Texas Advanced Technology Program, and the Robert A. Welch Foundation.

REFERENCES


Proceedings of the 10th International Conference on Particle Induced X-ray Emission and its Analytical Applications, Portorož, Slovenia, June 4-8, 2004