M-Shell X-Ray Transition Rates for Heavy Rare Earth Elements

Sera K., Ishii K., Orihara H., Morita S.

CYRIC annual report

Volume

Page range

Year

URL

http://hdl.handle.net/10097/49326

<table>
<thead>
<tr>
<th>著者</th>
<th>Sera K., Ishii K., Orihara H., Morita S.</th>
</tr>
</thead>
<tbody>
<tr>
<td>区部</td>
<td>CYRIC annual report</td>
</tr>
<tr>
<td>年度</td>
<td>1986</td>
</tr>
<tr>
<td>巻</td>
<td>52-59</td>
</tr>
<tr>
<td>URL</td>
<td><a href="http://hdl.handle.net/10097/49326">http://hdl.handle.net/10097/49326</a></td>
</tr>
</tbody>
</table>
M-Shell X-Ray Transition Rates for Heavy Rare Earth Elements

Sera K., Ishii K., Orihara H. and Morita S.*
Cyclotron and Radioisotope Center, Tohoku University
Research Center of Ion Beam Technology, Hosei University*

Introduction

X-ray emission rates, Auger transition rates and Coster-Kronig transition rates for K- and L-shell vacancies have been well studied both theoretically and experimentally. For M-shell, however, number of studies have been limited owing mainly to the complexity of M X-ray spectrum.

Recently, we and Inagaki pointed out that the intensity of Dy-M\(_{\alpha,\beta}\) line produced by the M-electron capture of \(^{163}\)Ho show a significant disagreement with the theoretical prediction. The theoretical intensity for M\(_{\alpha,\beta}\) line is 1.8 times larger than the experimental one, where we normalized both intensities at M\(_{\gamma}\) line. The intensities of these M X-rays can be used to determine the mass of the electron neutrino (m\(_{\nu_e}\)). Therefore, the X-ray transition rates play an important role for the determination of m\(_{\nu_e}\).

On the other hand, it has been found from recent studies of XPS that the empty 4f states of heavy rare earth elements hybridize with the valence-band states. This effect is expected to affect the X-ray transition rates of M\(_{\alpha,\beta}\) line (4f-3d).

In the present work, we have systematically measured the M X-rays of heavy lanthanides produced by proton impact to investigate the discrepancy between theory and experiment.

Experiment and Results

We have measured M and L X-rays of heavy rare earth elements (Z=66-71) by proton 3 MeV impact with an ORTEC Si(Li) detector. Very thin targets (<2 µg/cm\(^2\)) were prepared to avoid self absorption due to the resonance excitation (3d-4f).

After correcting for detection efficiency, we obtained the partial M X-ray cross sections by referring to the counts of L\(_{\beta_1}\) peak,

\[ \sigma_{M_a}^X = \frac{(Y_{M_a}/(\text{eff})_{M_a})}{(Y_{L_{\beta_1}}/(\text{eff})_{L_{\beta_1}})} \times \sigma_{L_{\beta_1}}^{X-\text{theory}} \]

(1)

where \(\text{eff}\) is the detection efficiency, \(\sigma_{L_{\beta_1}}^{X-\text{theory}}\) the theoretical X-ray production cross section for L\(_{\beta_1}\) line, \(Y_{M_a}\) and \(Y_{L_{\beta_1}}\) the peak yields of M\(_a\) and L\(_{\beta_1}\) lines, \(\sigma_{M_a}^X\) is the experimental X-ray production cross section for M\(_a\)}
subline. In equation (1), errors from target thickness, solid angle, and the number of incident particles are cancelled out.

On the other hand, the theoretical X-ray production cross sections for M-shell are given by the following formulas (Eq. 2–7),

\[
\sigma_{\text{X-theory}}^{M_a} = \omega_{M_{1a}} \cdot \frac{\Gamma_{M_{1a}}^{X}}{\sum_{a} \Gamma_{M_{1a}}^{X}} \cdot \sigma_{M_{1}}^{h}
\]

\[
\sigma_{M_{1}}^{h} = \sigma_{M_{1}}^{I}
\]

\[
\sigma_{M_{2}}^{h} = \sigma_{M_{2}}^{I} + S_{12} \sigma_{M_{1}}^{I}
\]

\[
\sigma_{M_{3}}^{h} = \sigma_{M_{3}}^{I} + S_{23} \sigma_{M_{2}}^{I} + (S_{31} + S_{12} \cdot S_{23}) \sigma_{M_{1}}^{I}
\]

\[
\sigma_{M_{4}}^{h} = \sigma_{M_{4}}^{I} + S_{34} \sigma_{M_{3}}^{I} + (S_{24} + S_{23} \cdot S_{34}) \sigma_{M_{2}}^{I}
\]

\[
+ (S_{14} + S_{12} \cdot S_{24} + S_{13} \cdot S_{34} + S_{12} \cdot S_{23} \cdot S_{34}) \sigma_{M_{1}}^{I}
\]

\[
\sigma_{M_{5}}^{h} = \sigma_{M_{5}}^{I} + S_{45} \sigma_{M_{4}}^{I} + (S_{35} + S_{34} \cdot S_{45}) \sigma_{M_{3}}^{I}
\]

\[
+ (S_{25} + S_{24} \cdot S_{45} + S_{23} \cdot S_{35} + S_{23} \cdot S_{34} \cdot S_{45}) \sigma_{M_{2}}^{I}
\]

\[
+ (S_{15} + S_{12} \cdot S_{25} + S_{13} \cdot S_{35} + S_{14} \cdot S_{45} + S_{12} \cdot S_{23} \cdot S_{35}
\]

\[
+ S_{12} \cdot S_{24} \cdot S_{45} + S_{13} \cdot S_{34} \cdot S_{45} + S_{12} \cdot S_{23} \cdot S_{34} \cdot S_{45}) \sigma_{M_{1}}^{I}
\]

(7)

here \(\omega_{M_{1}}\) and \(S_{ij}\) are the fluorescence and Coster-Kronig yields for \(M_{1}\) subshell \(^{1}\), \(\Gamma_{M_{1a}}^{X}\) the theoretical radiative transition rates for \(M_{a}\) subline \(^{8}\), \(\sigma_{M_{1}}^{I}\) the theoretical ionization cross sections calculated by PWBA \(^{9}\), \(\sigma_{M_{1}}^{h}\) means the total probability for \(M_{1}\) subshell vacancy prior to the emission of M X-rays. Since it is verified that the prediction of PWBA for ionization cross section agree with the experimental value within 10% in this energy region, it is expected that the values of transition rate are mainly reflected in the comparison of X-ray production cross sections between experiment and theory.

In Fig. 1 are shown the raw X-ray spectrum for Ho metal and background spectrum from 10 μm mylar backing together with the net spectrum after background subtraction. Figure 2 shows the results of the peak separation of M X-rays for each elements, and Fig. 3 shows the example of the fitting of \(L_{B1}\) line used for the intensity normalization.

Figure 4a–4d show the results for each M X-ray subline. As shown in
these figures, the theoretical values for $M_\gamma$ line are considerably smaller than the experimental values, while those of $M_{\alpha,\beta}$ line agree with the experimental values. In the case of $M_{\gamma}$ line, theoretical values always underestimate the experimental ones. $M_{2-N_4}$, $M_{1-N_3}$ and $M_{1-N_2}$ lines are in good agreement within experimental errors. These results are shown in Table 1. Errors are mainly come from fitting and the detection efficiency.

Discussion

In Fig. 5a, 5b are shown these results for each element. It is found in this figure that the good agreement between theory and experiment are achieved for $M_{\alpha,\beta}$ line. For $M_\zeta$ and $M_{\gamma}$ lines, however, there are distinct disagreements. Particularly for Dy, which is the daughter atom of $^{163}$Ho, the experimental value of $M_{\gamma}$ is nearly 1.8 times larger than the theoretical one. As mentioned above, the experimental intensity for $M_{\alpha,\beta}$ line is about 1.8 times larger than the theoretical one in $^{163}$Ho spectrum. It is found that this discrepancy are due to the intensity normalization at $M_{\gamma}$ line, while the relative intensities of $^{163}$Ho spectrum are consistent with the present results.

After all, despite of the transition from 4f, there is no clear discrepancy between experiment and theory for $M_{\alpha,\beta}$ line. On the other hand, the reason for disagreement in the case of $M_\zeta$ and $M_{\gamma}$ lines is not explainable at the present time.

References

2) Inagaki Y., Same Proceedings as Ref. 1) but p. 15.
<table>
<thead>
<tr>
<th>Elements</th>
<th>( L_{61} )</th>
<th>( M_x )</th>
<th>( M_y )</th>
<th>( M_{61} )</th>
<th>( M_{x-y} )</th>
<th>( M_{x-N} )</th>
<th>( M_{y-N} )</th>
<th>( M_{x-N} )</th>
<th>( M_{y-N} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( ^{159}Yb )</td>
<td>36.5</td>
<td>(36.5)</td>
<td>504.0</td>
<td>72.5</td>
<td>2894.1</td>
<td>2917.4</td>
<td>276.7</td>
<td>36.7</td>
<td></td>
</tr>
<tr>
<td>( ^{161}Yb )</td>
<td>33.3</td>
<td>(33.3)</td>
<td>386.7</td>
<td>40.0</td>
<td>2922.0</td>
<td>220.8</td>
<td>34.9</td>
<td>27.2</td>
<td></td>
</tr>
<tr>
<td>( ^{169}Dy )</td>
<td>46.0</td>
<td>(46.0)</td>
<td>2592.3</td>
<td>1937.5</td>
<td>243.0</td>
<td>98.4</td>
<td>7.7</td>
<td>44.6</td>
<td></td>
</tr>
<tr>
<td>( ^{169}Ln )</td>
<td>46.0</td>
<td>(46.0)</td>
<td>2592.3</td>
<td>1937.5</td>
<td>243.0</td>
<td>98.4</td>
<td>7.7</td>
<td>44.6</td>
<td></td>
</tr>
<tr>
<td>( ^{171}Er )</td>
<td>43.2</td>
<td>(43.2)</td>
<td>700.2</td>
<td>87.5</td>
<td>266.4</td>
<td>114.4</td>
<td>22.0</td>
<td>33.1</td>
<td></td>
</tr>
<tr>
<td>( ^{173}Er )</td>
<td>43.2</td>
<td>(43.2)</td>
<td>700.2</td>
<td>87.5</td>
<td>266.4</td>
<td>114.4</td>
<td>22.0</td>
<td>33.1</td>
<td></td>
</tr>
<tr>
<td>( ^{175}Ho )</td>
<td>59.1</td>
<td>132.1</td>
<td>529.4</td>
<td>54.0</td>
<td>2399.7</td>
<td>171.4</td>
<td>38.9</td>
<td>28.3</td>
<td></td>
</tr>
<tr>
<td>( ^{177}Ho )</td>
<td>59.1</td>
<td>132.1</td>
<td>529.4</td>
<td>54.0</td>
<td>2399.7</td>
<td>171.4</td>
<td>38.9</td>
<td>28.3</td>
<td></td>
</tr>
<tr>
<td>( ^{179}Dy )</td>
<td>61.9</td>
<td>171.2</td>
<td>2050.3</td>
<td>463.6</td>
<td>112.3</td>
<td>22.4</td>
<td>47.3</td>
<td>10.0</td>
<td></td>
</tr>
<tr>
<td>( ^{181}Dy )</td>
<td>61.9</td>
<td>171.2</td>
<td>2050.3</td>
<td>463.6</td>
<td>112.3</td>
<td>22.4</td>
<td>47.3</td>
<td>10.0</td>
<td></td>
</tr>
<tr>
<td>( ^{183}Er )</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td></td>
</tr>
<tr>
<td>( ^{185}Er )</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td></td>
</tr>
<tr>
<td>( ^{187}Er )</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td></td>
</tr>
<tr>
<td>( ^{189}Er )</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td></td>
</tr>
<tr>
<td>( ^{191}Er )</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td></td>
</tr>
<tr>
<td>( ^{193}Er )</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td>55.2</td>
<td>_+4.2</td>
<td>_46.7</td>
<td>_+10.0</td>
<td></td>
</tr>
</tbody>
</table>

*Table 1. Partial M X-ray Production Cross Sections (in barn).*
Fig. 1. M and L X-ray spectra for Ho. Besides the raw spectrum, background spectrum from 10 \( \mu \)m mylar backing and the net spectrum after background subtraction are also shown.
Fig. 2(a-f). Spectra of M X rays for Dy-Lu. Smooth curves show peak separation by a least-squares-fitting assuming Gaussian shapes and 5-th order polynomial background.
Fig. 3. Same as Fig. 2 but for Er L X-rays.

Fig. 4(a-d). Partial M X-ray production cross sections for 4 sublines (a); M,; b; M,; c; M,; d; (M, - N,)+(M, - N,)+(M, - N,). The lines labeled "theory" are obtained using Eq. (2). For present data errors are shown with bars.
Fig. 5(a,b). Same as Fig. 4 but for each element (a; Dy, Ho, Er, b; Tm, Yb, Lu).