Precise Energies of K-Röntgen-Lines of Tm, Th, U and Pu

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The energies of the Kα₁- and Kα₂-Röntgen lines of Tm, Th, U and Pu have been measured with high accuracy relative to the 411.794 keV γ-line from a gold source. In addition, the energies of a few Kβ lines have been determined.

1. Introduction

Accurate energies of K-Röntgen-lines of heavy elements are of interest for a comparison with a theory which allows precise calculations of atomic electron binding energies. They are also needed for the calibration of Röntgen- and Ge(Li)-spectrometers.

In practice the K-Röntgen-line energies are calculated from the atomic level energies listed in standard tables such as those given by Hagström, Nordling and Siegbahn or by Lederer, Hollander and Perlman or they are calculated from the energies recommended by Bearden and Burr. Inspection of such tables shows that differences up to 5 × 10⁻⁴ in relative energy (or 58 eV at 121.8 keV) still exist for the energy of the Is state in Pu.

Therefore, the Kα₁- and Kα₂-line energies of a few heavy elements have been determined with very high accuracy with our interferometric curved crystal diffractometer.

2. Experimental Method

The measurements were performed with the aid of the curved crystal spectrometer which has been described elsewhere. For the Tm measurement a very thin source was fabricated out of 9 mg of Yb₂O₃ enriched in¹⁸⁸Yb to about 20%. The U-measurement was carried out with the use of a similar probe made out of 25 mg of metallic thorium powder. For the measurement of the Pu-lines a metallic uranium foil with a weight of 91 mg and depleted with respect to ²³⁵U was used as source. All of these sources were activated in the reactor Merlin of the Kernforschungsanlage Jülich. The thorium lines were measured twice: a) during the U-measurement (fluorescence due to the ²³³Pa decay) and b) during the irradiation of a small sample of 1.5 mg of thorium metal powder located between two gold sources of a total activity of about 50 Ci. In this latter case, a ¹⁶⁹Yb source was used as standard. The U-fluorescence measurement was performed in the same manner but with a gold source as standard because of the proximity of the 93 keV line of ¹⁶⁹Tm. During the other measurements either a gold source or a ¹⁶⁹Yb source provided the reference line. All measurements were performed in the manner outlined in detail previously. The resolution ΔE (FWHM) at the diffractometer was ΔE ≈ 50 eV during the Tm-Kα-measurement which could only be performed in the first order of reflection (see Fig. 1) because of the limited angular range of the instrument. It was ΔE ≈ 33 eV for the Tm Kβ₁ line and ΔE ≈ 63 eV during the Kα-measurement of Pu (ΔE ~ E²).

3. Results and Discussion

The results of the present experiment are listed in Table 1. The relative energies are based on the 411 keV gold standard. For an estimate of the influence of the particular choice of the standard ener-

Fig. 1. Spectrum of the K-Röntgen lines emitted from a ¹⁸⁸Yb source and measured in the first order of reflection.
Table 1. Energies $E_{\text{relative}}$ and absolute energy errors $dE$ of K-Röntgen lines of Tm, Th, U and Pu excited during the decay of 169Yb, fluorescence, the decay of 233Pa and of 239Np respectively and measured relative to the 411.794 keV γ-ray standard from the decay of 198Au. The error of the 411.794 keV line, which is given as ±7 eV has therefore not been included in the results listed in column 4 of the table. For comparison with previous data, level-energy differences are tabulated in the columns 5, 6 and 7.

<table>
<thead>
<tr>
<th>Element</th>
<th>Radioactive Source</th>
<th>K-Röntgen Line</th>
<th>$E_{\text{relative}} \pm dE$/eV</th>
<th>Hagström et al.</th>
<th>Lederer et al.</th>
<th>Bearden and Burr</th>
<th>$E$/eV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tm</td>
<td>169Yb $\rightarrow$ 169Tm</td>
<td>K$\alpha_1$</td>
<td>50,740.52 ± 0.09</td>
<td>50,742</td>
<td>50,742</td>
<td>50,741.6</td>
<td>0.6</td>
</tr>
<tr>
<td></td>
<td>20% 168YbO$_3$</td>
<td>K$\beta_1$</td>
<td>57,507.68 ± 0.15</td>
<td>57,502</td>
<td>57,505</td>
<td>57,505.1</td>
<td>1.2</td>
</tr>
<tr>
<td>Th</td>
<td>233Pa in</td>
<td>K$\alpha_1$</td>
<td>93,346.0 ± 0.6</td>
<td>93,349</td>
<td>93,351</td>
<td>93,350.6</td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>Th metal fluorescence</td>
<td>K$\alpha_1$</td>
<td>89,955.5 ± 0.7</td>
<td>89,957</td>
<td>89,958</td>
<td>89,957.7</td>
<td>1.0</td>
</tr>
<tr>
<td>Pu</td>
<td>239Np $\rightarrow$ 239Pu</td>
<td>K$\alpha_1$</td>
<td>98,431.9 ± 0.6</td>
<td>98,436</td>
<td>98,438</td>
<td>98,439.8</td>
<td>1.6</td>
</tr>
<tr>
<td>W</td>
<td>fluorescence</td>
<td>K$\alpha_1$</td>
<td>94,651.0 ± 0.7</td>
<td>94,656</td>
<td>94,658</td>
<td>94,658.5</td>
<td>1.6</td>
</tr>
</tbody>
</table>

Notes:

1. The energy of the K$\alpha_1$ line of tungsten is systematically larger by about 1 eV. The energy of our K$\beta_1$ line of Tm is, however, significantly larger than the other values.

2. Since a comparison of line-energies based on different standards is always difficult, we choose to compare line-energy differences. In this case the differences of the standard energies have relatively little or negligible influence on the numbers to be compared. One can immediately realize that our figure agrees well with the result of Bearden and Burr and fairly well with the identical results of Hagström et al. and Lederer et al. regarding $E(L_{\text{III}}) - E(L_{\text{III}})$.

3. The K$\beta_1$ line of Tm we obtain $E(K_{\beta_1}) - E(K_{\alpha_2}) = 6,767.16 \pm 0.17$ eV which does not agree with the differences of 6.760, 6.763 and 6.763 ± 1.4 eV of the tabulated level energies. In order to understand this discrepancy we have studied the energy systematics of the $E(L_{\text{III}}) - E(M_{\text{III}})$ distance which should equal the difference $E(K_{\beta_1}) - E(K_{\alpha_2})$ in the absence of line asymmetries. It must be emphasized that our K-Röntgen energies listed in Table 1 are those of the quanta emitted by the sources characterized in the second column of that table. For other sources shifts may arise which may exceed our quoted errors. It is obvious that this energy distance should increase monotonously with Z. Testing the tabulated data in this respect should, therefore, provide insight into the magnitude of the statistical uncertainties and reveal possible errors of individual distances.
Table 2. Comparison between the K-Röntgen line-energy differences as obtained in the present work and the tabulated level energy differences. The units are eV.

<table>
<thead>
<tr>
<th>Element</th>
<th>Levels</th>
<th>Bearden and Burr</th>
<th>Lederer et al.</th>
<th>Hagström et al.</th>
<th>this work</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tm</td>
<td>$E(L_{II}) - E(L_{III})$</td>
<td>$1.031.1 \pm 0.8$</td>
<td>1.030</td>
<td>1.030</td>
<td>$1.031.21 \pm 0.15$</td>
</tr>
<tr>
<td>Th</td>
<td>$E(L_{II}) - E(L_{III})$</td>
<td>$3.392.9 \pm 0.5$</td>
<td>3.393</td>
<td>3.392</td>
<td>3.390.7 $\pm 1.7$</td>
</tr>
<tr>
<td></td>
<td>$E(L_{III}) - E(M_{II})$</td>
<td>$14.862.6 \pm 0.6$</td>
<td>14.862</td>
<td>14.861</td>
<td>14.862.5 $\pm 1.7$</td>
</tr>
<tr>
<td>U</td>
<td>$E(L_{II}) - E(L_{III})$</td>
<td>$3.781.3 \pm 0.4$</td>
<td>3.780</td>
<td>3.780</td>
<td>3.781.0 $\pm 0.8$</td>
</tr>
<tr>
<td>Pu</td>
<td>$E(L_{II}) - E(L_{III})$</td>
<td>$4.209.4 \pm 0.9$</td>
<td>4.209</td>
<td>4.210</td>
<td>4.210.8 $\pm 1.4$</td>
</tr>
</tbody>
</table>

In the region $60 \leq Z \leq 78$ the distance between the $L_{III}$ and $M_{III}$ levels can be described very well with the aid of the empirical relation:

$$E(L_{III}) - E(M_{III}) \approx \bar{E} = 6767 \text{ eV} + (Z - 69) \times 222.5 \text{ eV} + (Z - 69)^2 \times 1.8 \text{ eV}.$$

In the lower part of Fig. 2 we have plotted the differences $[E(L_{III}) - E(M_{III})] - \bar{E}$. The figure clearly shows that the tabulated $2-4$ level distance $E(L_{III}) - E(M_{III})$ is not consistent with the expectation. The result of the present experiment is, however, in full agreement with the systematics. The discrepancy is due to the large errors and inconsistency of the tabulated $M_{III}$ level energies $2-4$ for $Z = 69$. This is demonstrated in the upper part of Fig. 2 where again a smooth function is expected [with $\bar{E} = 70 \text{ eV} + (Z - 69) \times 2 \text{ eV}$].

Our energies for the thorium lines are systematically somewhat smaller than the tabulated level distances. This is also true for the differences of $E(K\alpha_1) - E(K\alpha_2)$ and of $E(L_{II}) - E(L_{III})$. In case of the distances $E(K\beta_3) - E(K\alpha_2)$ and $E(L_{II}) - E(M_{II})$ very good agreement is observed with the tabulated data (see Table 2).

The $K\alpha_1$ and $K\alpha_2$ Röntgen-line energies of U of $E_{\text{relative}} = 98,431.6 \pm 0.5$ eV and 94,650.6 $\pm 0.6$ eV are considerably smaller than the tabulated level-energy differences. Inspection of Table 2 shows very good agreement between the $L_{II} - L_{III}$ level energy differences and our difference $E(K\alpha_1) - E(K\alpha_2)$. This indicates that the tabulated K-shell energies are to large by 5—8 eV.

In case of plutonium, the tabulated $2,3$ K-shell energies differ by as much as 58 eV. The uncertainty of the result by Bearden and Burr is 44 eV. Our K-line energies are larger than the energies of Hagström et al. $5$ and smaller than those of Lederer et al. $3$. They agree within the 44 eV error with the energies of Bearden and Burr. In view of our much higher accuracy we have again compared the line-energy differences with the tabulated $2-4$ level-energy distances. Table 2 shows agreement for the $L_{II} - L_{III}$ spacing.

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