

# Atomic-K-X-Ray Intensity Ratios

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*Dedicated to Prof. Dr. H. Maier-Leibnitz on his 60th birthday*

Experimental K-X-ray intensity ratios  $K\alpha_2/K\alpha_1$ ,  $K\alpha_3/K\alpha_1$ ,  $K\beta_1/K\alpha_1$ ,  $K\beta_2/K\beta_1$ ,  $K\beta_3/K\beta_1$ ,  $K\beta_4/K\beta_1$  and  $K\beta_5/K\beta_1$  have been determined for several elements with  $63 \leq Z \leq 92$ . These ratios agree very well with the results of a fully relativistic calculation of transition probabilities by Rosner and Bhalla. The theoretical values are tested with an accuracy of better than one per cent for the ratios  $K\alpha_2/K\alpha_1$ ,  $K\beta_2/K\beta_1$  and  $K\beta_3/K\beta_1$ .

## Introduction

Recently, ROSNER and BHALLA<sup>1</sup> have performed a relativistic calculation of atomic X-ray transition rates. Including the effect of the retardation of the radiation field, they used the self-consistent relativistic Hartree-Fock method with the Slater approximation for the exchange term and computed EL and ML multipole transition rates for all transitions to the K-, L- and M-shells for the elements with  $21 \leq Z \leq 93$ .

A detailed comparison of experimental data with the predicted transition rates provides a critical test of the theoretical results and shows to which extent the theory<sup>1</sup> gives a proper description of the atomic electron structure.

Extensive K-X-ray intensity measurements have been carried out by BECKMAN<sup>2</sup> in the region of heavier elements ( $Z \geq 73$ ) already in 1955. BECKMAN has also observed the forbidden (M1)  $L_1 \rightarrow K$  transition as a very weak bump on the low energy side of the  $K\alpha_2$  line of tungsten and gold. However, a systematic determination of the intensity of the  $L_1 \rightarrow K$  line of the heavier elements has not been made.

For this reason and in order to obtain an independent set of data, we have measured K-X-ray spectra of several elements with  $Z \geq 63$ .

## Experimental Method

The K-X-ray spectra were recorded together with gamma lines from the capture of slow neutrons in natural elements or enriched isotopes. The purpose of these measurements, which were performed in the period from 1963 to 1970, has always been the investigation of the low energy ( $n, \gamma$ )-spectrum for the study of nuclear structure<sup>3</sup>. The K-X-ray data, to be discussed in this paper, were only a by-product.

The spectra were measured with the curved crystal diffractometer located at one of the tangential channels of the reactor DR-3 at Risø. The instrument has been described elsewhere<sup>4</sup>. The gamma- and X-ray sources were 20–30 mm high, 0.02–0.15 mm wide and 2–4 mm deep along a direction in parallel with the beam of radiation impinging on the spectrometer crystal.

The narrow source widths permitted the recording of X-ray spectra with an instrumental resolution of

$$600 < E/\Delta E < 7000,$$

where  $E$  is the energy and  $\Delta E$  the full energy width at half maximum. The observed widths of the reflections of the X-ray lines were considerably larger than the instrumental widths in most cases because of the large natural X-ray line widths<sup>5</sup>. The source

<sup>1</sup> H. R. ROSNER and C. P. BHALLA, Z. Physik **231**, 347 [1970].

<sup>2</sup> O. BECKMAN, Ark. Fysik **9**, 495 [1955].

<sup>3</sup> Most of the ( $n, \gamma$ ) data and their interpretation have been published already (see e. g. Nuclear Data **3**, 367 [1967], **5**, 1 [1968] and **5**, 243 [1969]).

<sup>4</sup> U. GRUBER, B. P. MAIER, and O. W. B. SCHULT, Kerntechnik **5**, 17 [1963], B. P. MAIER, U. GRUBER, and O. W. B. SCHULT, Kerntechnik **5**, 19 [1963], O. W. B. SCHULT, U. GRUBER, and B. P. MAIER, Kerntechnik **7**, 101 [1965].

<sup>5</sup> G. C. NELSON, W. JOHN, and B. G. SAUNDERS, Phys. Rev. **187**, 1 [1969], G. C. NELSON, W. JOHN, B. G. SAUNDERS, UCRL-71856.



depth was chosen such as to yield a high sensitivity for  $(n, \gamma)$ -rays with energies around 500 keV which implies that X-ray quanta suffer strong absorption within the source. Because of the small source width it is experimentally difficult to produce a geometrically very well defined and homogeneous source. Therefore, the effect of the X-ray absorption within the source cannot be calculated with high accuracy. Consequently, precise absolute X-ray intensities cannot be obtained from these data. However, the uncertainty of the relative X-ray absorption is rather small in the narrow energy region covered by the K-X-ray lines, where also the relative detection efficiency of the crystal spectrometer is well known. For these reasons, the intensity ratios of close lying lines can be determined quite accurately.

### Results

Ratios of K-X-ray line intensities have been determined for Eu, Gd, Yb, Lu, Au, Hg, Th and U. The results are shown in the figure, which also includes the data obtained by BECKMAN<sup>2</sup>. The nomenclature

is as follows:  $K\alpha_3 = (L_1 \rightarrow K)$ ,  $K\alpha_2 = (L_2 \rightarrow K)$ ,  $K\alpha_1 = (L_3 \rightarrow K)$ ,  $K\beta_3 = (M_2 \rightarrow K)$ ,  $K\beta_1 = (M_3 \rightarrow K)$ ,  $K\beta_5 = (M_4 \rightarrow K) + (M_5 \rightarrow K)$ ,  $K\beta_2 = (N_2 \rightarrow K) + (N_3 \rightarrow K)$ ,  $K\beta_4 = (N_4 \rightarrow K) + (N_5 \rightarrow K)$ . The intensity  $I$  of a particular X-ray line is set equal to the number of quanta per sec. of this line, so that  $I$  is proportional to its transition rate. The ratio  $I(K\alpha_3)/I(K\alpha_1)$  of Yb could not be given because of a strong  $\gamma$ -line superimposed on the  $K\alpha_3$ -transition. Instead the corresponding ratio of Tm has been plotted in the figure.

### Discussion

An inspection of the figure shows that the predicted ratio<sup>6</sup>  $I(K\alpha_2)/(K\alpha_1)$  is in extremely good agreement with the experimental result derived from the Risø data. Assuming that the slope of the theoretical ratio is correct, we obtain a difference between the experimental and predicted magnitudes of only  $\Delta(\alpha_2/\alpha_1) \approx 0.7\%$ , which follows from the scattering of the experimental points around the theoretical curve. The centre of gravity of the experi-

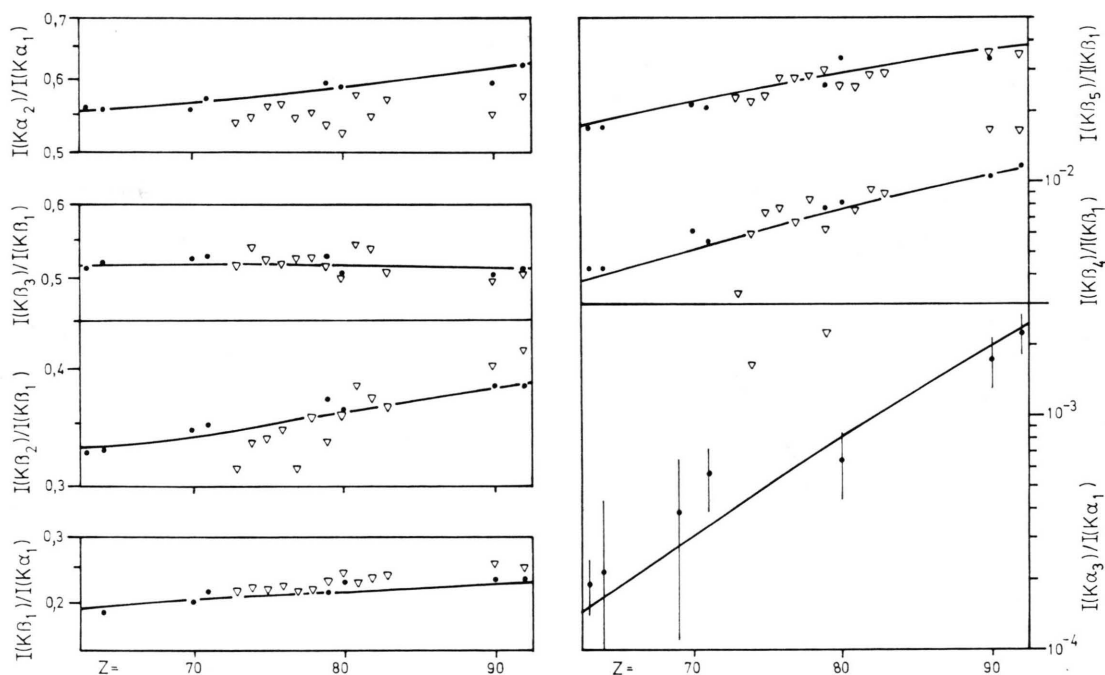


Fig. 1. Comparison between experimental and theoretical<sup>1</sup> K-X-ray intensity ratios. The solid curves give the predicted ratios. The experimental data by BECKMAN<sup>2</sup> are characterized by triangles, the Risø data by points.

<sup>6</sup> The ratios of the theoretical transitions rates may have uncertainties of 1% (C. B. BHALLA, private communication).

mental values differs even by only  $\delta(\alpha_2/\alpha_1) \approx 0.4\%$  from the theoretical result. BECKMAN's results are somewhat low.

Very good agreement between experiment and theory is also found for the ratio  $I(K\beta_3)/I(K\beta_1)$ , where we have  $\Delta(\beta_3/\beta_1) \approx 0.6\%$  and  $\delta(\beta_3/\beta_1) \approx 0.1\%$  from the Risø data, and  $\Delta_B(\beta_3/\beta_1) \approx 0.8\%$  and  $\delta_B(\beta_3/\beta_1) \approx 0.2\%$  from BECKMAN's data.

For the ratio  $I(K\beta_2)/I(K\beta_1)$  BECKMAN's points scatter more about the predicted curve

$$[\Delta_B(\beta_2/\beta_1) \approx 1.7\%, \delta_B(\beta_2/\beta_1) \approx 1.4\%]$$

than the results of the present experiments, which give  $\Delta(\beta_2/\beta_1) \approx 0.7\%$  and  $\delta(\beta_2/\beta_1) \approx 0.5\%$ , in support of the theory.

The accuracy of the Risø data is less in case of the ratio  $I(K\beta_1)/I(K\alpha_1)$  because of the larger energy spacing of the  $K\beta_1$  and  $K\alpha_1$  lines. Still good agreement is found between theory<sup>6</sup> and our data:  $\Delta(\beta_1/\alpha_1) \approx 1.4\%$  and  $\delta(\beta_1/\alpha_1) \approx 1.3\%$ . BECKMAN's results are systematically higher by  $6 \pm 1\%$ .

The  $K\beta_5$ ,  $K\beta_4$  and  $K\alpha_3$  lines are relatively weak and, furthermore, they are located in the vicinity of the quite strong  $K\beta_1$ ,  $K\beta_2$  and  $K\alpha_2$  lines. Therefore, the relative intensity accuracy is limited both by the statistics of the measurement and by the difficulty to precisely fit the tails of the strong X-ray lines, so that the experimental data have errors which are considerably larger than those of the ratios discussed above. Complete agreement is found between the Risø data and the predicted ratios<sup>6</sup>. BECKMAN's results give full support to the theory for the  $K\beta_5/K\beta_1$  ratios, where their quality compares with that of our data. The 1955 data<sup>2</sup> also

agree with the theoretical ratio  $K\beta_4/K\beta_1$ , with the exception of a few points which show larger deviations from the expected values. However, the previous results<sup>2</sup> for the ratio  $K\alpha_3/K\alpha_1$  are about a factor of three above the predicted values<sup>1</sup>. This discrepancy is probably due to the fact that BECKMAN took the full width at half maximum (FWHM) of the  $K\alpha_3$ -line twice the FWHM of the other  $K\alpha$ -lines. We find that the FWHM of the  $K\alpha_3$ -line equals the FWHM of the  $K\alpha_2$ - and  $K\alpha_1$ -lines within the experimental uncertainty.

## Conclusion

The present study has shown that the experimental K-X-ray intensity ratios agree well with the theoretical results<sup>6</sup> obtained by ROSNER and BHALLA<sup>1</sup>. The experimental data permit a test of the theory to better than 1% for the ratios  $I(K\alpha_2)/I(K\alpha_1)$ ,  $I(K\beta_3)/I(K\beta_1)$  and  $I(K\beta_2)/I(K\beta_1)$ , for which theoretical values would be desirable with computational errors less than 0.3%.

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*Note added in proof:* The ratios  $I(K\alpha_3)/I(K\alpha_1)$  of Hf and Au reported by R. K. SMITHER, M. S. FREEDMAN, and F. T. PORTER, Phys. Letters **32 A**, 405 [1970] are considerably smaller than our result. We have no explanation for this discrepancy.