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Lawrence Radiation Laboratory
UNIVERSITY OF CALIFORNIA
LIVERMORE

MEASUREMENT OF RELATIVE K-RADIATIVE DECAY RATES

V. W. Slivinsky

P. J. Ebert

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MEASUREMENT OF RELATIVE K-RADIATIVE DECAY RATES*

V. W. Slivinsky and P. J. Ebert

Lawrence Livermore Laboratory, University of California
Livermore, California 94550

ABSTRACT

Relative K-radiative decay rates for elements between $Z = 18$ and 92 were measured. In the range $18 \leq Z \leq 39$, pure samples of thickness between $40\text{-}90 \mu\text{g}/\text{cm}^2$ were evaporated on $0.5 \text{ mg}/\text{cm}^2$ thick Be substrates. A strongly collimated x-ray beam from a Mo target x-ray tube operated at 30 kV was filtered with $50 \text{ mg}/\text{cm}^2$ Mo and $140 \text{ mg}/\text{cm}^2$ Al. The transmitted x-rays (mainly Mo fluorescence) excited the samples which were mounted in a vacuum. A Be window Si(Li) spectrometer with known efficiency was used to count fluorescent x-rays. Spectrometer resolution was 185 eV at 6.4 keV, and for the lower Z elements the fluorescent peaks had to be graphically separated. The data were corrected for scattered background, detector efficiency, Si escape peaks, Be x-ray transmission, and incomplete charge collection in the spectrometer (tailing), all of which were measured. These corrections were generally $<2\%$, except for $18 \leq Z \leq 22$, where 3-6% corrections were applied. The total error was estimated to be 1% for all elements with $Z \geq 22$; the largest error was 6% at $Z = 18$.

In the range $29 \leq Z \leq 92$, pure metal oxide samples $<1 \text{ mg}/\text{cm}^2$ were supported by 2.5×10^{-4} cm thick mylar films. Appropriately filtered x-rays from a W-target x-ray tube excited the samples in air. Sample

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thickness was determined by comparing count rates with those from foils of known thickness. A Ge(Li) spectrometer with known efficiency having a resolution of 550 eV at 60 keV was used to count the fluorescent x-rays. For $Z \geq 62$, it was possible to resolve the K_{α_1} , K_{α_2} , K_{β_1} and K_{β_3} x-ray peaks. Some of the x-ray peaks were graphically separated, and the data were corrected for the effects mentioned above. For the larger peaks the corrections generally were <10%. Estimated experimental errors in peak areas were 1.5-6%.

The K_{β}/K_{α} ratios for all elements are consistently higher than those predicted by Scofield, but show qualitative agreement in that the shapes of the K_{β}/K_{α} vs. Z curves are similar. Both the calculation and the experiment show K_{β}/K_{α} to be relatively constant for $21 \leq Z \leq 30$, where the 3d levels are being filled. For $Z \geq 62$, the $K_{\alpha_2}/K_{\alpha_1}$ ratios are in excellent agreement with predictions while the ratios K_{β_1}/K_{α_1} and K_{β_2}/K_{α_1} are consistently higher than predicted.

I. INTRODUCTION

As late as 1969, values used for the relative K radiative decay rates were from the tabulation of Wapstra et al.,¹ which was based on measurements by Williams² for $Z < 50$ and by Beckman³ for $Z > 70$. Beckman and Williams used electrons to produce K-shell vacancies and both used diffractometers to obtain the resolution required to measure the line intensities. Since 1969, much experimental and theoretical work on this subject has been reported. This has been summarized by Nelson et al.⁴ and more recently by Rao et al.⁵

This report summarizes the work which we have done in Livermore since 1969 in the measurement of the relative K-radiative decay rates.^{6,7,8,9} All of these measurements have utilized semiconductor x-ray spectrometers to obtain high resolution, and in all but a single measurement, K-shell vacancies were produced by x-rays. We compare these measurements with the theoretical results of Scofield.¹⁰

II. EXPERIMENT

A. Production of K-Shell Vacancies

There are several ways in which K-shell vacancies can be produced. Electrons directly excite K-shell vacancies,^{1,2,11,12} but a large bremsstrahlung background is produced along with the fluorescent lines. Uncertainty in background subtraction can introduce a large error in measurement of lines. Heavy charged particles have also been used to produce K vacancies.¹³ While there is no bremsstrahlung background produced, the slowly moving particle perturbs the outer shell electrons. This alters the line energies, and also changes the relative radiative decay rates. Furthermore, excitation of K-vacancies by heavy ions is very inefficient for high Z elements. Radioactive sources also emit K x-rays. A radioactive source to be used for this purpose must be thin to its own fluorescent x-rays or be of a known, uniform thickness so that the difference in self-absorption at the fluorescent energies can be accounted for accurately. The source should decay primarily by K capture or K conversion, so that there is small background near the fluorescent peaks from higher energy gamma rays scattering out of the detector. In the low Z region, a limited selection of easily produced radioactive isotopes has these properties. For these reasons, we used bremsstrahlung from heavily filtered x-ray tubes to produce the K vacancies. In the low Z region ($18 \leq Z \leq 39$), the tube had a Mo anode. With appropriate filtration, ($50 \text{ mg/cm}^2 \text{ Mo}$ and $140 \text{ mg/cm}^2 \text{ Al}$) almost negligible background was present near the fluorescent peaks.

For $29 \leq Z \leq 92$ the tube had a W anode, and was filtered with 100 mg/cm^2 Fe and 140 mg/cm^2 Al to remove W L x-rays. Tube voltage was adjusted between 50 and 150 kV. In the range $69 \leq Z \leq 74$, the background was complicated by scattering of W K x-rays. To minimize this structure on an otherwise smooth bremsstrahlung background, the tube x-rays were filtered with Er and Pb foils to absorb most of the W fluorescence.

B. Samples

1. Elements with $18 \leq Z \leq 39$

Samples of 16 elements were used; each was placed in the collimated x-ray beam. The samples were made by evaporating high-purity metals or compounds on 2.5×10^{-4} cm thick Be substrates. The thickness of the samples ranged from $40\text{--}90 \times 10^{-6} \text{ g/cm}^2$. This is thin enough that there was no significant correction to the data for the difference in self-absorption between the K_α and K_β lines. The sample chamber was evacuated to 10^{-3} Torr to eliminate an air-attenuation correction to the data. For $Z = 18$ (Ar) and $Z = 36$ (Kr), the chamber was filled with high-purity gas to a pressure of 25 Torr.

Since it was possible for the chemical composition of the sample to effect the results,¹⁴ we also used thick samples (2.5×10^{-3} cm) of high-purity Sc, Ti, Cu, and Zn. We compare these results with thin-sample results for the same elements. A K-capture isotope, a thin (10^{-6} g/cm^2) ^{55}Fe source with an activity of 1×10^{-6} Ci, was used in an attempt to duplicate the

method of excitation used by Hansen et al.;¹⁵ we compare that result with our other results.

2. Elements with $29 \leq Z \leq 92$

Twenty-two samples of metal or metal oxide were mounted on 2.5×10^{-4} cm thick mylar film. Each sample had an average thickness of less than 1 mg/cm^2 as determined by comparing count rates from the samples with those from metal foils having known thicknesses. However, some of the samples were nonuniform and may have been considerably thicker than the average in some places. These samples were mounted in air with a 25 cm separation between the sample and the spectrometer.

C. Spectrometers

1. Si(Li) Spectrometer

The Si(Li) spectrometer has a pulsed light feedback pre-amplifier and a low-noise amplifier. It was fabricated at the Lawrence Berkeley Laboratory. The Si wafer is approximately 3 mm thick and has a collimated front surface area of 12.3 mm^2 . The vacuum window on the system is high-purity Be whose thickness, $9.613 \times 10^{-3} \text{ g/cm}^2$, was measured by weighing, and whose purity was checked by measuring its transmission to 4.0 keV x-rays and comparing with the calculated transmission using the measured cross section.¹⁶ The Si wafer also has a thin layer of Au, whose thickness is approximately 1×10^{-6} cm on its front surface, and a Si dead layer with a thickness of about 1×10^{-5} cm.¹³ This system has a resolution of 185 eV for the 5.9 keV photons of ⁵⁵Fe.

The efficiency of the spectrometer was measured between 8 and 25 keV using a technique described elsewhere.¹⁷ This upper-energy calibration determined the energy at which the Si wafer transmits an appreciable number of incident photons. The Si(Li) detector was found to be totally absorbing¹⁸ up to 15 keV.

The spectrometer was also calibrated at the Sc K_{α} energy, 4.09 keV, by comparing it with a thin-window gas proportional counter whose efficiency was calculated. The Si(Li) spectrometer was 92% efficient at this energy. Photons absorbed in the Be window account for 7% of this loss, and the other 1% is presumably absorbed in the Au front surface and in the Si dead layer. The attenuation of 4.09 keV photons in 1×10^{-6} cm Au and 1×10^{-5} cm Si is calculated to be 3%. Since the accuracy of the calibration was $\pm 3\%$, we concluded that the layers of Au and Si were about as thick as stated by the manufacturer. Below 4.09 keV, for the $Z = 18$ and $Z = 19$ fluorescent energies, the efficiency of the spectrometer was calculated using the above layer thicknesses.

2. Ge(Li) Spectrometer

For $29 \leq Z \leq 92$ two Ge(Li) spectrometers were interchangeable; one having a resolution of 640 eV full width at half maximum at 60 keV and one having a resolution of 550 eV at 60 keV. The photopeak efficiency and escape peak-to-photopeak ratios for each spectrometer were measured and reported elsewhere.¹⁷ These results were used to correct the data.

III. SPECTRUM ANALYSIS

A. Elements with $18 \leq Z \leq 39$ -- Si(Li) Spectrometer

The Mo target x-ray tube was generally run at 30 kV and 75 mA. The bremsstrahlung beam from the tube was filtered with high-purity Al for the low-Z targets and with Mo and high-purity Al for the higher-Z elements. This filtering considerably reduced the scattered background near the fluorescent peaks. Figure 1 shows the Zn spectrum plotted on a log scale to emphasize the low background. For the lower-Z elements, tailing in the detector was more significant. It is important to stress that there is no contribution to the tail from x-rays with lower energies than the fluorescent lines because of the heavy filtering of the excitation source. This is also evident from the energy spectrum from the ^{55}Fe isotope, which decays entirely by K capture but still shows counts below the photopeaks (see Figure 2). For the K_β/K_α measurements, therefore, the background under the photopeaks was determined by the background level just above the K_β peak; in most cases, this was about 0.1% of the K_β peak height.

The tail counts are due to incomplete charge collection, and they properly belong in the photopeaks. Since the K_α peak is so much more intense than the K_β peak, we could determine the total number of tail counts per photopeak count as a function of x-ray energy (Figure 3). This curve was used to determine the number of tail counts to be added to the K_α and K_β photopeaks for the K_β/K_α measurements.

It should be noted in Figures 1 and 2 that the escape peak associated with the K_β photopeak is obscured by the tailing of the K_α peak. To determine what fraction of the K_β photons are in its escape peak,

we measured the escape peak-to-photopeak ratios for the K_{α} lines. These results are shown in Figure 4; the error bars show the uncertainty due to the statistics in counting the escape peaks. The correction to the K_{β}/K_{α} measurements for the difference in the escape peak-to-photopeak ratio for the K_{α} and K_{β} energies was less than 0.1% in the worst case and therefore was ignored.

Of particular concern was the presence of impurities in the Be substrate that might introduce small fluorescent peaks under the peaks of interest. A Be substrate was run without any sample material on it. Nanogram quantities of Fe and Mn were found in the Be, but counts from these impurities were negligible when compared with the counts from an actual target.

Finding the peak areas for most elements simply amounted to subtracting the background and then adding the counts in the peaks that were well separated. For the lower-Z elements, however, the K_{α} and K_{β} peaks were not completely resolved and had to be graphically separated. This was done by constructing similarly shaped peaks whose sum, in all cases, was equal to at least 99.8% of the total number of K_{α} and K_{β} counts. Spectrometer resolution as a function of photopeak energy was accounted for in this method. For $Z \geq 21$, more than 99% of the constructed peak areas was used to determine K_{β}/K_{α} .

B. Elements with $29 \leq Z \leq 92$ -- Ge(Li) Spectrometer

Except for the lowest Z elements the K_{α} and K_{β} peaks were well-resolved, and peak areas were readily obtainable after subtraction of bremsstrahlung background. For $Z > 62$, it was also possible to resolve the K_{α_1} , K_{α_2} , K_{β_1} and K_{β_2} lines. A typical spectrum for the W fluor-escer is shown in Figure 5.

The same considerations mentioned in Section III.A. were included in the analysis of spectra taken with the Ge(Li) systems. Notable differences were that the detector tailing was less, but the escape peak-to-photopeak ratios were larger. Partially resolved peaks were graphically separated by hand and mathematically separated by a computer code which fitted the peaks to gaussian curves.

IV. EXPERIMENTAL ERROR

Sources of error in these ratio measurements were as follows:

	Si(Li)	Ge(Li)
sample thickness	<0.1%	<0.5% ^a
relative spectrometer efficiency	0.1-0.5%	0.5%
tailing	0.14%	-
backgrounds (contaminants)	0.1%	<2%
counting statistics	<0.7%	<3%
peak separation (when necessary)	0.5-3%	2-5%

^aThis assumes uniform sample thickness. For $Z < 35$, sample non-uniformity would increase this error significantly - see Section V.

Representative error is indicated in Figures 6, 7, 8 and 9, and in Table I.

V. RESULTS

A. K_{β}/K_{α}

The K_{β}/K_{α} ratios for $18 \leq Z \leq 92$ are plotted in Figure 6 and are listed in Table 1. The Si(Li) results are systematically lower than the results using the Ge(Li) detector with powdered samples for $Z < 35$. A probable cause was that the metal oxide powders had some thick areas where differential absorption was important. No correction was made to those data for differential absorption. We believe the Si(Li) results to be more reliable for the low Z elements.

In Figure 6 are plotted the K_{β}/K_{α} ratios calculated by Scofield.¹⁰ It can be seen that the experiment and theory are in qualitative agreement in that both are relatively flat in the region $21 \leq Z \leq 30$, and that a general increase in K_{β}/K_{α} with Z is apparent. Scofield's calculations show dips at $Z = 24$ (Cr) and $Z = 29$ (Cu) where the atom has an extra 3d electron instead of two 4s electrons. We studied these elements very closely, but found no evidence of such behavior.

The K_{β}/K_{α} result from the experiment using an ^{55}Fe source (Mn x-rays) was 0.1379 ± 0.0014 , while the result from a Mn sample excited by x-rays was 0.1361 ± 0.0014 . The difference in K_{β}/K_{α} for two different modes of excitation of Mn is within the experimental errors of the measurements.

The K_{β}/K_{α} results from the thick sample experiments were in excellent agreement with the K_{β}/K_{α} results from the thin samples. Even though self absorption correction factors were about 10%, the final results were within 1% of the thin target results.

All ratios are in excellent agreement with Hansen, et al. for $Z > 40$ and with McCrary, et al.¹⁹ for $18 \leq Z \leq 92$.

B. $Z \geq 62$

For $Z \geq 62$, the K_{α_1} , K_{α_2} , K_{β_1} and K_{β_2} lines could be resolved. These results are shown in Figures 7, 8 and 9, where they are compared with Scofield's calculations. The agreement for the $K_{\alpha_2}/K_{\alpha_1}$ ratio is excellent, while the K_{β_1}/K_{α_1} and K_{β_2}/K_{α_1} ratios appear systematically higher than those predicted by Scofield. The $K_{\alpha_2}/K_{\alpha_1}$ ratios are in excellent agreement with the results of Nelson and Saunders,²⁰ and all ratios agree very well with the recent work of McCrary, et al.¹⁹

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TABLE I
Final Results of K_{β}/K_{α} Measurements

Z	Element	Si(Li) K_{β}/K_{α}	Ge(Li) K_{β}/K_{α}
18	Ar	0.1032 $\pm$ 0.0067	
19	K	0.1217 $\pm$ 0.0055	
21	Sc	0.1302 $\pm$ 0.0026	
22	Ti	0.1319 $\pm$ 0.0017	
23	V	0.1339 $\pm$ 0.0011	
24	Cr	0.1344 $\pm$ 0.0011	
25	Mn	0.1361 $\pm$ 0.0011	
26	Fe	0.1366 $\pm$ 0.0011	
27	Co	0.1376 $\pm$ 0.0011	
28	Ni	0.1385 $\pm$ 0.0011	
29	Cu	0.1387 $\pm$ 0.0011	.147 $\pm$ 5%
30	Zn	0.1418 $\pm$ 0.0011	.152 $\pm$ 5%
32	Ge	0.1493 $\pm$ 0.0012	
34	Se	0.1595 $\pm$ 0.0013	.169 $\pm$ 3%
36	Kr	0.1715 $\pm$ 0.0014	
38	Sr		.181 $\pm$ 3%
39	Y	0.1642 $\pm$ 0.0015	.186 $\pm$ 3%
40	Zr		.190 $\pm$ 3%
42	Mo		.197 $\pm$ 3%
46	Pd		.210 $\pm$ 3%
52	Te		.228 $\pm$ 3%
58	Ce		.242 $\pm$ 3%
59	Pr		.243 $\pm$ 3%
62	Sm		.254 $\pm$ 3%
63	Eu		.256 $\pm$ 3%
64	Gd		.258 $\pm$ 3%
68	Er		.256 $\pm$ 3%
70	Yb		.268 $\pm$ 3%
72	Hf		.269 $\pm$ 3%
74	W		.271 $\pm$ 3%
79	Au		.279 $\pm$ 3%
82	Pb		.282 $\pm$ 3%
90	Th		.294 $\pm$ 3%
92	U		.295 $\pm$ 3%

FIGURE CAPTIONS

- Fig. 1. The x-ray spectrum from a thin Zn target excited by a bremsstrahlung from a heavily filtered Mo anode x-ray tube.
- Fig. 2. The x-ray spectrum from the ^{55}Fe source.
- Fig. 3. Total number of counts in the tail per number of counts in the photopeak.
- Fig. 4. Escape peak-to-photopeak ratio as a function of photopeak energy.
- Fig. 5. W-fluorescent spectrum as measured with Ge(Li) spectrometer having 550-eV resolution at 60 keV.
- Fig. 6. The K_{β}/K_{α} results as a function of Z. The solid curve is from Scofield's calculations (Ref. 10).
- Fig. 7. $K_{\alpha_2}/K_{\alpha_1}$ as a function of Z. Shown are results of this experiment, previous measurements (Ref. 1), and recent calculations (Ref. 10).
- Fig. 8. K_{β_1}/K_{α_1} as a function of Z. Shown are the results of this experiment, previous measurements (Ref. 1), and recent calculations (Ref. 10).
- Fig. 9. K_{β_2}/K_{α_1} as a function of Z. Shown are the results of this experiment, previous measurements (Ref. 1), and recent calculations (Ref. 10).

Slivinsky - Fig. 1

Slivinsky - Fig. 2

Slivinsky - Fig. 3

Slivinsky - Fig. 4

Slivinsky - Fig. 6

Stivinsky - Fig. 7

Slivinsky - Fig. 8

Stivinsky - Fig. 9