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Measurements of Relative K Radiative Decay Rates*

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Relative radiative decay rates were measured for K-shell vacancies for elements between $Z=62$ and 92 with a high-resolution Ge(Li) spectrometer. The ratios α_2/α_1, β_1'/α_1, and β_2'/α_1 (Siegbahn notation) were determined, with α_2/α_1 significantly higher (4–14%) than those reported by Beckman but in excellent agreement with recent Hartree-Slater calculations of Scofield. The ratios β_1'/α_1 and β_2'/α_1 do not agree with either Beckman's experiment or Scofield's calculations.

I. INTRODUCTION

Relative radiative decay rates of K-shell vacancies are of both fundamental and practical importance in atomic physics and nuclear spectroscopy. They are used in the determination of internal conversion and electron-capture coefficients in experiments which count fluorescent photons from inner-shell vacancies produced by these phenomena. They are necessary for the determination of L-shell fluorescence yields and for calculating the average x-ray energy emitted from x-ray sources used to determine detector energy response. Experimental determination of the radiative decay rates of K-shell vacancies are also needed to test the various theoretical models used to calculate these transitions.

Relative radiative decay rates are determined experimentally by measuring the relative intensities of characteristic x rays. In the case of the K shell, the relative intensities of the x-ray lines are independent of the method of production of shell vacancies. Previous experimental results[1,2] in the ranges of $24 \leq Z \leq 52$ and $73 \leq Z \leq 92$ have been tabulated by Wapstra, Nijgh, and van Lieshout.[3] The gap in the data for $53 \leq Z \leq 72$ led to this investigation, and (subsequent) differences between our results and the Wapstra extrapolation in this region led to an expanded effort to remeasure previously published values, some of which were measured as far back as 1933.

II. EXPERIMENT

A. Apparatus

A schematic diagram of the experimental apparatus is shown in Fig. 1. K-shell vacancies are produced by exposing pure samples to the bremsstrahlung produced by a 150-kV x-ray machine. Samples of metal or metal oxide were mounted on 2.5×10^{-4}-cm-thick Mylar film. Each sample was less than 1 mg/cm^2 thick as determined by comparing count rates from the samples with those from metal foils having a known thickness.

Fluorescent x rays were detected by a highly collimated Ge(Li) spectrometer placed perpendicular to the bremsstrahlung beam. In this experiment, two spectrometers were used interchangeably; one having a resolution of 640 eV full width at half-maximum (FWHM) at 60 keV and one having a resolution of 550 eV at 60 keV. The photopeak efficiency of each spectrometer was measured and reported elsewhere.[4] The spectrometer was placed 25 cm from the sample. Amplified pulses were fed into a 1024-channel analyzer. An example of the fluorescent x-ray spectrum for

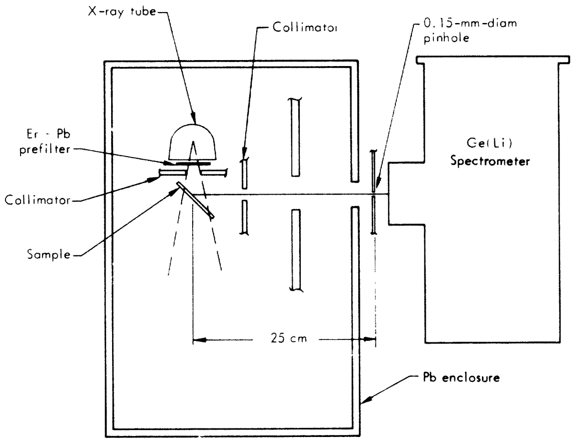

FIG. 1. Experimental geometry.

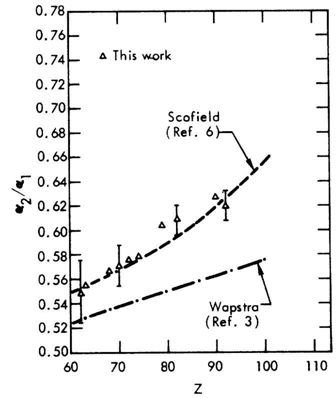

FIG. 3. α_2/α_1 x-ray radiative-decay-rate ratios Shown are results of this experiment, previous measurements (Ref. 3), and recent calculations (Ref. 6).

tungsten is shown in Fig. 2. In these spectra, the fluorescent x-ray peaks are superimposed on a background of scattered bremsstrahlung. For $68 \leq Z \leq 72$, the average background count was complicated, as a result of scattering of W fluorescence from the x-ray tube. To minimize this structure in an otherwise smooth background, the tube x rays were filtered with Er and Pb foils. In this way, most of the tungsten K x rays and virtually all of the tungsten L x rays were absorbed. When scattered tungsten K x rays were not close to the photopeaks of interest, an Fe + Al prefilter was used to absorb the tungsten L x rays from the tube, and thereby, reduce the rate of unwanted counts. The spectrometer count rate could be controlled by adjusting the x-ray tube current. In this experiment, it was always less than 500 counts per second for optimum spectrometer resolution.

B. Experimental Results

To determine the intensity of each line, the counts in each photopeak were corrected for relative detector efficiency after the background was subtracted. Air and sample self-absorption was small and accounted for in the analysis. Partially resolved peaks were unfolded both graphically and by computer. Preliminary results for the α/β ratios in the range $29 \leq Z \leq 92$ were published,[5] and further data analysis does not warrant changing that tabulation. The α_2/α_1 data are plotted in Fig. 3, while the β_2'/α_1 and β_1'/α_1 results are shown in Fig. 4.

C. Experimental Error

An error analysis was carried out for the measured ratios and these results are indicated by the error bars in Figs. 3 and 4. Sources of errors included: (i) counting statistics (< 3%); (ii) relative efficiency of the Ge(Li) spectrometer (0.5%); (iii) spectrum contamination by scattered tungsten

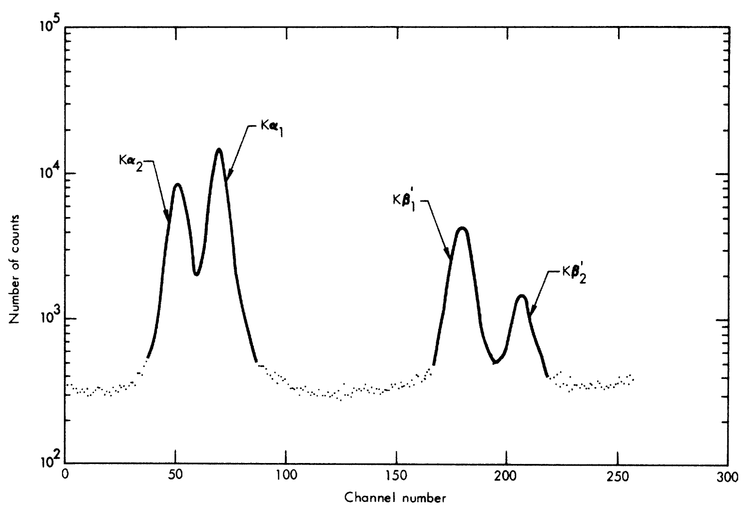

FIG. 2. W-fluorescent spectrum as measured with Ge(Li) spectrometer having 550-eV resolution at 60 keV.

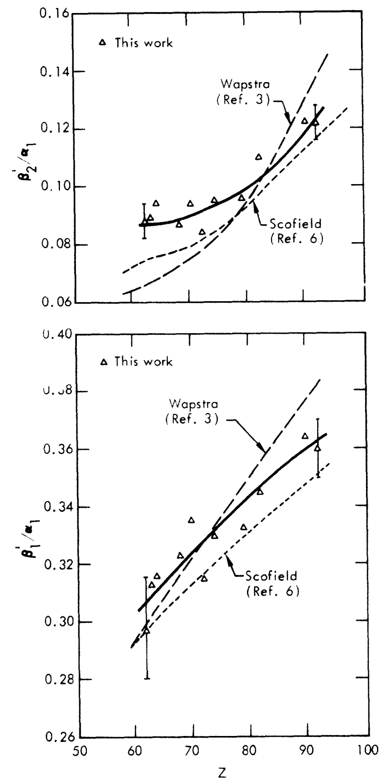

FIG. 4. β_2'/α_1 and β_1'/α_1 x-ray radiative-decay-rate ratios. Shown are the results of this experiment, previous measurements (Ref. 3), and recent calculations (Ref. 6).

K x rays (< 2%); (iv) spectrum unfolding (2–5%). Generally, the errors become larger with decreasing *Z*.

III. DISCUSSION

Recently, radiative-decay-rate calculations were made by Scofield,[6] who considered the atomic electrons to be in single-particle states in a central potential given by the relativistic Hartree-Slater theory. Using Scofield's α and β components, the measured ratios were calculated for comparison. The comparison for the α/β ratios is shown in Ref. 5.

The results of Scofield for the α_2/α_1 ratios are shown in Fig. 3 as a smooth line which fits the data very well. The Wapstra tabulation, also shown in Fig. 3, is consistently low. Saunders and Nelson, of the Lawrence Radiation Laboratory, Livermore, California, have independently measured α_2/α_1 with a crystal spectrometer in the region $51 \leq Z \leq 95$. Their unpublished data are in excellent agreement with our results.[7]

Scofield's β_1'/α_1 and β_2'/α_1 ratios (Fig. 4) are somewhat lower than experiment. Since the α/β ratios[5] for $29 \leq Z \leq 62$ were found to be consistently lower than Scofield's calculated ratios, we can speculate that the β_1'/α_1 and β_2'/α_1 ratios should be higher than the calculated ratios in that region.

The data presented here represent an improvement in accuracy over previously measured values for the high-*Z* elements because of the simplicity of the experiment. Beckman made sizable corrections to his data because of target self-absorption (11–21%) and crystal reflection efficiency (29%). Inaccuracies in these corrections, especially in calculating the spatial distribution of *K*-shell vacancies produced by 500-keV electrons bombarding thick targets, could account for the difference between the two measurements. Target self-absorption corrections for this experiment were always less than 0.5%.

The accuracy of some of the most commonly used *K* radiative decay rates has been improved with this experiment, and significant differences were found when compared with previous measurements. It is clear from our results that further high-resolution experiments are needed to accurately measure decay rates for the lower-*Z* elements.

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