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$K\beta_3/K\beta_1$ Transition Probabilities in Elements of Medium and High Atomic Number*

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The $K\beta_3/K\beta_1$ transition probabilities have been measured for the following elements: $_{49}$In, $_{50}$Sn, $_{52}$Te, $_{58}$Ce, $_{65}$Tb, $_{73}$Ta, $_{75}$Re, $_{79}$Au, $_{82}$Pb, $_{90}$Th, $_{92}$U, $_{93}$Np, $_{94}$Pu, and $_{95}$Am. This has been accomplished using a spectrometer with a bent quartz crystal in conjunction with a Ge(Li) detector. The measured transition probabilities for elements of medium atomic numbers are in good agreement with calculations based on the relativistic Hartree-Slater potential, while in the region of high atomic number the measured values seem to favor theoretical values obtained assuming a Coulomb potential with the electron screening effect prescribed by Burns.

INTRODUCTION

Systematic measurements of the probability of the electronic transition $3P_{1/2} \rightarrow 1S_{1/2}$ relative to that of the $3P_{3/2} \rightarrow 1S_{1/2}$ for elements of high atomic numbers were made by Beckman,[1] but his results are not conclusive as to the dependence of this ratio on the atomic number.

Relativistic calculations of the radiative transition probabilities were carried out earlier by Massey and Burhop,[2] Laskar,[3] Payne and Levinger,[4] Asaad,[5] Taylor and Payne,[6] and later by Babushkin[7] and Scofield.[8] All these calculations except those of Asaad and Scofield are based on a Coulomb potential. Massey and Burhop and Laskar introduced an effective nuclear charge to account for the screening effect on the atomic electrons. Babushkin carried out his calculations with and without allowance for screening; he took into account the screening effect of the electrons, as prescribed by Slater[9] and Burns.[10] Asaad's calculations are based on a potential obtained from a self-consistent field calculation, while Scofield assumed that the atomic electrons are under the influence of a relativistic Hartree-Slater potential.

Experimental measurements of the $K\beta_1'/K\alpha_1$ transition probabilities[11] seem to favor Babushkin's calculations with the electron screening effect taken into account by the Burns rule; on the other hand the most recent experimental values[12] of $K\alpha_2/K\alpha_1$ are in agreement with Scofield's calculations. The two theories exhibit the same dependence of $K\alpha_2/K\alpha_1$ and almost the same dependence of $K\beta_1'/K\alpha_1$ on atomic number, with slight difference in the magnitude of these ratios. In the case of $K\beta_3/K\beta_1$ radiative transition probability, Babushkin's theory predicts a gradual increase with increasing atomic number while Scofield's cacula-tion results in a ratio that decreases as the atomic number increases above $Z = 65$. As these two refined calculations are at variance in the prediction of the dependence of $K\beta_3/K\beta_1$ on the atomic number, systematic measurements of this ratio should offer a good testing ground of the validity of these calculations.

EXPERIMENTAL

Most of the target elements used in this work were in the form of metal foils 2.5×7.6 cm and varying in thickness from 0.02 to 0.05 cm. The Am, Pu, and Np targets were sealed in Al capsules to reduce the hazard of their radioactivity. To excite their spectra, the elements were exposed to a ^{182}Ta radioactive source which was made up of a Ta foil sealed in an Al casing and rendered radioactive by exposure to neutrons from the Lawrence Radiation Laboratory (LRL) pool-type reactor for several months. The activity of the fresh source is about 50 Ci.

The present work requires an instrument of high resolution, as the wavelengths of the two lines $K\beta_1$ and $K\beta_3$ of the lowest-Z element under investigation differ by only 5×10^{-4} Å. Such high resolution was provided by a bent crystal spectrometer constructed in the Cauchois geometry.[13] The quartz crystal is 2 mm thick, bent to a radius of 2 m, and is exposed to the incident beam of 12-mm effective width. The rays are diffracted by the (310) planes and are made to converge at the 0.176-mm-wide detector slit. The relative intensity is then measured using a 2-cm^3 thin-window Ge(Li) detector, designed and built at the LRL.

For a given element, the positions of the diffraction peaks were located from a plot of the intensity versus position by automatically stepping the detector slit over the proper region. These steps were 0.01 mm each, and the counting period varied depending on the intensity of the lines under investigation. A typical spectrum is shown in Fig. 1. With the precise positions of the $K\beta_1$ and $K\beta_3$ diffraction maxima established, the detector slit was set at one of the diffraction maxima for a given period and then on the other. For each element, this setting was alternated several times between the two diffraction peak positions. This eliminated the effect of the minute reduction in intensity caused by the decay of the fluorescing ^{182}Ta source. The period of measurement for each line varied from 4 to 10 h, depending on its intensity. Although the ratio of the areas under the curves obtained by stepping over the diffraction peaks is of more physical significance, the ratio of the peak intensities was considered more reliable in determining the transition probabilities as it is less susceptible to

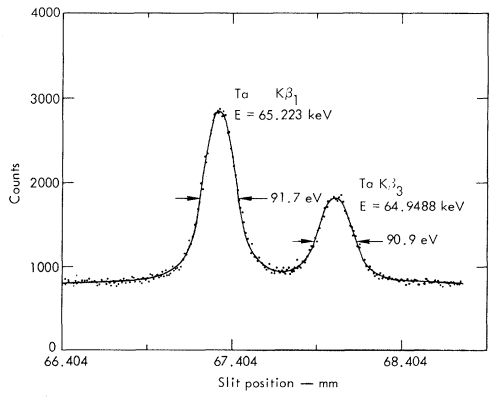

FIG. 1. $K\beta_1$ and $K\beta_3$ x-ray lines of Ta. The points are experimental and were obtained by stepping the detector slit in 10^{-2}-mm steps. The smooth curve is a computer fit.

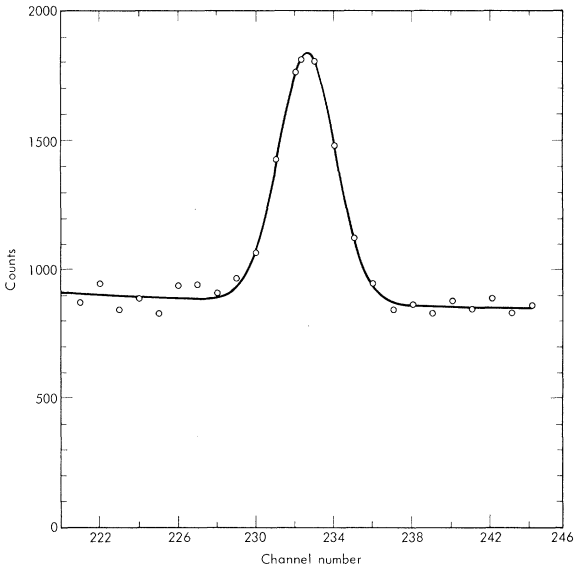

FIG. 2. Computer fit to the experimental points of $UK\beta_1$ pulse-height full-energy peak.

instrumental factors. It should be pointed out that the natural widths of the $K\beta_1$ and $K\beta_3$ lines are very nearly the same for the same element, and therefore the ratio of the peak intensities should be very nearly the same as the ratio of the physical areas under the curves. Thus the number of counts recorded with the Ge(Li) detector set on the diffraction peak was considered proportional to the probability of the electronic transition causing the peak.

With the detector and slit on the proper peak position, the x-ray pulse-height spectrum was recorded on a multichannel analyzer. The area under the pulse-height full-energy peak represented the relative transition probability, and was obtained from a nonlinear least-squares fit of the sum of a Gaussian and a quadratic background to the experimental points. Figure 2 is a computer fit to the $K\beta_1$ pulse-height peak of $_{92}$U.

The raw data were corrected for absorption, reflectivity of the quartz crystal, and the varying efficiency of the Ge(Li) detector with photon energy. For elements with atomic numbers equal to or smaller than 65, compensations were made for the tails characteristic of the Lorentzian shape of the x-ray lines.

Corrections were made for self-absorption in the irradiated foil. Here the relative intensity[14] of the ^{182}Ta γ rays, as well as the excitation cross section of the K electrons of the irradiated element, was taken into consideration to determine the distribution of the formation of the K x rays in the target. The β^- particles emitted by

the radioactive ^{182}Ta source are mostly absorbed in the Al casing and contribute very little to the excitation of the target atoms. Corrections for absorption in the air path and in the quartz crystal were also made. The values of mass-absorption coefficients used were those tabulated by Storm *et al.*[15]

The reflectivity of the quartz crystal as a function of the energy has been investigated by many. Edwards[16] found that the reflectivity of bent quartz crystal varies as $E^{-1.987\pm0.022}$ over the energy range 60 keV $\geq E \geq$ 400 keV. Line *et al.*[17] reported an E^{-2} dependence over the energy range 25 keV $\geq E \geq$ 1332 keV. For the present work, an E^{-2} dependence was assumed.

The efficiency of the Ge(Li) detector as a function of the photon energy was determined using calibrated ^{57}Co, ^{109}Cd, and ^{241}Am radioactive sources. The combined net effect of the above corrections is shown in Fig. 3 and amounts in the most severe case, $_{95}$Am, to little less than 4% of the measured ratios.

The bent crystal spectrometer is capable of resolving the $K\beta_1$ and $K\beta_3$ lines of the lowest-Z element studied. But, for elements with atomic number $Z \leq 65$, the two characteristic lines were not far enough apart for the peak of one to be completely free of contribution from the long tail of the other. Such contributions are disproportionate and, if not corrected for, result in too large a $K\beta_3/K\beta_1$ ratio. The shape of these lines was assumed symmetrical with respect to a straight line drawn through the intensity maximum[18] which for a transition $a \rightarrow b$, occurs at $\omega = \omega_{ab}$. The characteristic lines were then unfolded, and contributions from their over extended tails were measured and their effect eliminated.

RESULTS AND DISCUSSION

In the literature, the two terms "transition probability" and "intensity ratio" are used interchangeably. Here, transition probability stands for the number of photons, whereas intensity designates energy flux. Thus, for a transition i,

$$I_i = T_i \cdot \hbar\omega_i \quad . \tag{1}$$

The observed transition probabilities as well as the intensity ratios are listed in Table I. Also listed are the most probable values of the transition probabilities which were obtained from a least-squares computer fit to the experimental points. The over-all error is estimated to be about 2% for elements with $Z \leq 80$ and is probably as high as 3% for elements with $Z \leq 90$. The larger error at high Z is partially due to the relatively low observed intensities of the characteristic lines of these elements. The energies of the $K\beta_1$ and $K\beta_3$ transitions for the elements $_{93}$Np, $_{94}$Pu, and $_{95}$Am had not been published before and had to be measured to calculate the $K\beta_3/K\beta_1$ intensity ratios for these elements (these energies will be published elsewhere).

The measured transition probabilities are plotted as a function of the atomic number in Fig. 4. Also plotted are the experimental values obtained by Beckman and the theoretical calculations of Babushkin and Scofield.

Our experimental approach is similar to that of Beckman, but two points of variance should be noted: (i) Beckman used high-energy electrons to irradiate his targets, whereas we used γ rays. Corrections for self-absorption when the excitation is caused by light charged-particles are very complicated because of photoionization[19] but become simple when the ionizing particles are γ rays. (ii) The ^{182}Ta radioactive source with its long half-life provides much better stability than the Van de Graaff generator used to furnish the high-energy electrons in Beckman's work. This could be Beckman's main difficulty as the intensity of characteristic lines is very sensitive to the applied voltage.

The radiative transition probability for an electric dipole to go from a state a or a state b to a state c, emitting a photon of frequency ω_{ac} or ω_{bc} is

$$\Gamma_{ac}/\Gamma_{bc} = (\omega_{ac}/\omega_{bc})^2 f_{ac}/f_{bc} \quad . \tag{2}$$

Here, f is the sum of the oscillator strengths; it explicitly contains the radial matrix elements of these transitions. Thus, the calculations of transition probabilities are a matter of evaluating the oscillator strengths. This involves the selection of the right potential, the inclusion of relativistic and retardation effects, and the effect of the finite size of the nucleus.

Both Babushkin and Scofield neglected the ef-

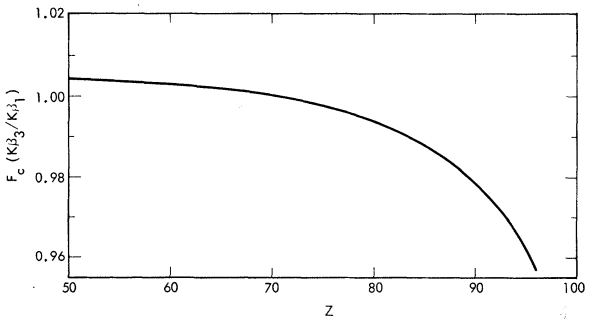

FIG. 3. Calculated correction factor applied to $K\beta_3/K\beta_1$ as a function of the atomic numbers. This includes absorption, reflectivity of the crystal, and the efficiency of the detector.

TABLE I. $K\beta_3/K\beta_1$ transition probabilities and intensity ratios.

Elements	$K\beta_3/K\beta_1$ transition probabilities measured values	$K\beta_3/K\beta_1$ transition probabilities most probable values	$K\beta_3/K\beta_1$ intensity ratios most probable values
$_{49}$In	0.517	0.515	0.514
$_{50}$Sn	0.515	0.515	0.514
$_{52}$Te	0.513	0.516	0.515
$_{58}$Ce	0.520	0.520	0.519
$_{65}$Tb	0.525	0.524	0.522
$_{73}$Ta	0.527	0.529	0.527
$_{75}$Re	0.532	0.531	0.529
$_{79}$Au	0.534	0.534	0.531
$_{82}$Pb	0.539	0.537	0.534
$_{90}$Th	0.560	0.544	0.540
$_{92}$U	0.537	0.547	0.543
$_{93}$Np	0.554	0.548	0.544
$_{94}$Pu	0.530	0.550	0.545
$_{95}$Am	0.561	0.551	0.546

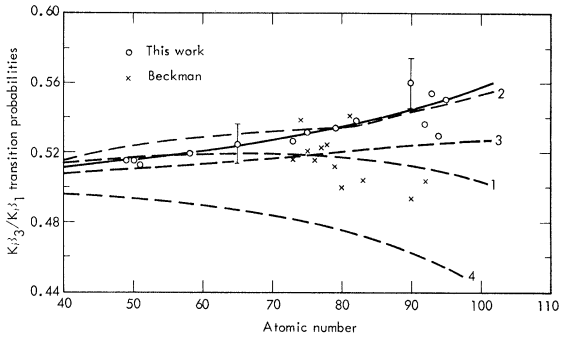

FIG. 4. $K\beta_3/K\beta_1$ transition probability as a function of the atomic number. The solid line represents the most probable experimental values from this work; curve (1) from Scofield's calculation based on a relativistic Hartree-Slater potential; curve (2) from Babushkin's calculation based on a Coulomb potential screened by Burns's prescription; curve (3) from Babushkin's calculation based on a Coulomb potential screened by Slater's prescription; and curve (4) from Babushkin's calculation based on a Coulomb potential without allowance for screening.

fect of the finite size of the nucleus which, if included, would have lowered their results of the $K\beta_3/K\beta_1$ ratio of $_{92}$U by about 0.35%. They applied relativistic calculations and included retardation effect. Therefore, the main difference in their calculations lies in their choice of the potential.

As Fig. 4 indicates, the experimental results are in agreement with calculations based on the relativistic Hartree-Slater potential in the region $49 \leq Z \leq 60$ but begin gradually to rise above the calculated values attaining a maximum discrepancy of about 8% at $_{95}$Am. For elements of atomic numbers $Z \geq 80$, the measured transition probabilities begin to agree with calculations based on a Coulomb potential with the electron screening effect prescribed by Burns. Calculations based on this type of potential seem to give too large a value of the $K\beta_3/K\beta_1$ for elements of medium and low atomic numbers.

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