$K\alpha_2/K\alpha_1$ Transition Probabilities in Elements with $Z \leq 50$

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The $K\alpha_2/K\alpha_1$ transition probabilities have been measured for 22 elements ranging in atomic number from $_{21}$Sc to $_{50}$Sn, with an estimated error of about 2%. This has been accomplished by bombarding the elements with an electron beam of constant energy, and studying the x-ray emission spectrum. The measured ratios rise gently above the classical limit as the atomic number increases, in contrast to the dip below this limit previously reported for $30 \leq Z \leq 40$. For $Z \geq 30$, the experimental results are in excellent agreement with recent calculations based on the relativistic Hartree-Slater potential. For $Z \leq 30$, although experimental results agree with theoretical calculations to within experimental error, they tend to be lower.

INTRODUCTION

Systematic measurements of the probability of the electronic transition $2P_{1/2} \rightarrow 1S_{1/2}$ relative to that of the $2P_{3/2} \rightarrow 1S_{1/2}$ have been previously carried out by Meyers,[1] by Williams,[2] by Beckman,[3] and recently by Nelson and Saunders,[4] and by Ebert and Slivinsky.[5] The averaged values of the earlier measurements were tabulated by Wapstra *et al.*,[6] and the recent measurements were performed for elements with $Z \geq 52$. The discrepancy between the recently observed values and Wapstra's table far exceeds reported experimental errors. This fact and the development of new theories lead to the present experimental measurements.

Relativistic calculations of the radiative transition probabilities were carried out earlier by Massey and Burhop,[7] by Laskar,[8] by Payne and Levinger,[9] by Asaad,[10] by Taylor and Payne,[11] and later by Babushkin,[12] and by Scofield.[13] All these calculations except Asaad's are based on a Coulomb potential. Massey and Burhop, and Laskar introduced an effective nuclear charge to account for the screening effect of the atomic electrons. Babushkin carried out his calculations with and without allowance for screening; he took into account the electron screening effect as prescribed by Slater[14] and by Burns.[15] Asaad's calculations are based on a potential derived from a self-consistent field, while Scofield assumed that the atomic electrons are under the influence of a relativistic Hartree-Slater potential.

The most recent K-transition probability measurements, which are probably the most reliable, seem to favor calculations based on a relativistic Hartree-Slater potential or a Coulomb potential screened by Burns's rule.[16,17] Scofield's relativistic calculations are in excellent agreement with measured values when transitions resulting in a close doublet are concerned, and differ from measured values by as much as 20% for large energy difference between studied transitions.[5,18,19]

EXPERIMENTAL

Most of the target elements used in this work were in the form of metal foils 2.5×3.75 cm, varying in thickness from 0.028 cm for $_{21}$Sc to 0.32 cm for the less expensive metals. Backing was provided for thin foils. Elements of low melting points $_{49}$In and $_{50}$Sn were melted in a brass container then introduced in the vacuum chamber. Pieces of $_{24}$Cr, $_{25}$Mn, and $_{38}$Sr were lead cemented into properly cut pieces of $_{29}$Cu prior to their bombardment. In the case of $_{38}$Sr, this was necessary to avoid the reaction of the sample with the anode-cooling water. The $_{32}$Ge sample was prepared by evaporating a thick layer of the element on a smooth piece of $_{4}$Be.

The electron source consists of a $_{74}$W filament in a stainless-steel focusing cup. The filament is heated by an insulated transformer. The electron energy is provided by a power supply connected in series with a voltage-line regulator and a ripple suppressor, and delivers a steady voltage with less than 0.3% ripple at a full load of 120 keV and 30 mA.

A modified high-angle goniometer was used to scan over the region of interest, and for each sample the line intensity was also measured in steps of 0.005 deg in the Bragg angle 2θ. In all, six runs for each element were recorded and averaged.

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 instrumental factors. It should be pointed out that the natural width of the $K\alpha_1$ and $K\alpha_2$ lines are very nearly the same for the same element,[20] and therefore

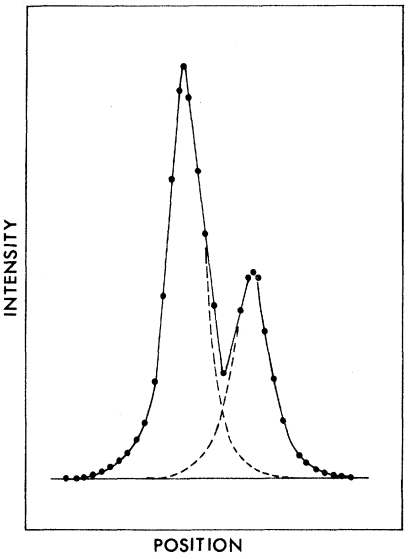

FIG. 1. $K\alpha_1$ and $K\alpha_2$ x-ray lines of Mn. The solid curve is a smooth fit to the experimental points, and the dashed portions account for contributions from the overextended Lorentzian tails.

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 $_{36}$Kr-filled counter set at the diffraction peak was considered proportional to the probability of the transition causing the peak.

To reduce the effect of self-absorption, which is complicated by photoionization,[21] the energy of the exciting electrons was kept as close to the ionization energy of the sample as practically possible, and the x-ray take-off angle was chosen to be nearly zero. It was found that the combined effect of differential self-absorption, absorption in the $_4$Be window and in the air path, and the reflectivity of the calcite crystal and the efficiency of the counter amounts, in the most severe case, $_{50}$Sn, to be less than 0.2% of the measured ratios. This is an order of magnitude smaller than experimental error, and only twice as large as the reported error in Scofield's calculations.

For elements with atomic number equal to or smaller than 32, the two characteristic lines, although well resolved, were not far enough apart for the peak of one to be completely free of contribution from the long tail of the other (Fig. 1). Such contributions are disproportionate and, if not corrected for, result in a large value for $K\alpha_2/K\alpha_1$. This is overcomplicated by the asymmetric shape of the lines of these elements.[22] However, the characteristic lines were unfolded, and contributions from their overextended Lorentzian tails were measured and their effect eliminated.

Several different sources of errors were considered. These include: (i) counting statistics, (ii) systematic errors, (iii) voltage ripple, (iv) tungsten contamination of anode from heated filament, and (v) spectrum unfolding. To reduce statistical errors, the spectrum of each element was measured six different times, and the average value of the six measurements is reported. Systematic errors were also reduced by stepping the detector for a complete run in the increasing direction of the Bragg angle and for the following run in the opposite direction. An error analysis was carried out for all the measured ratios and the results which amount to about 2% are indicated by error bars in Fig. 2.

RESULTS AND DISCUSSIONS

In the literature, the two terms "transition probability" and "intensity ratio" have been used interchangeably. In this article, the following distinction is made. The transition probability is proportional to the number of emitted photons,

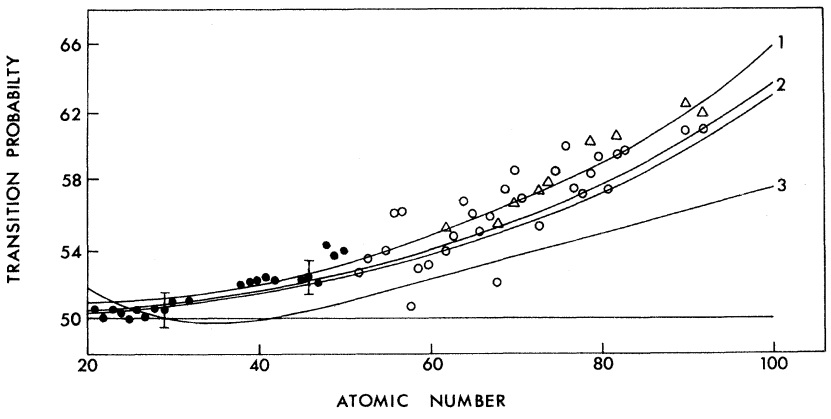

FIG. 2. $K\alpha_2/K\alpha_1$ transition probability as a function of the atomic number. The points are experimental; ○ from Ref. 4., △ from Ref. 5, and ● from the present work. Curve 1 is Scofield's; curves 2 are Babushkin's and were obtained assuming a Coulomb potential with the electron screening effect prescribed by Slater (top) and by Burns; curve 3 is from Wapstra's table. The straight line gives the classical value.

TABLE I. $K\alpha_2/K\alpha_1$ transition probability ratio.

Elements	Measured values	Most probable values	Ref. 13 values
$_{21}$Sc	0.507	0.505	0.509
$_{22}$Ti	0.502	0.505	0.509
$_{23}$V	0.507	0.505	0.509
$_{24}$Cr	0.505	0.506	0.510
$_{25}$Mn	0.500	0.506	0.510
$_{26}$Fe	0.506	0.506	0.511
$_{27}$Co	0.502	0.507	0.511
$_{28}$Ni	0.507	0.507	0.512
$_{29}$Cu	0.507	0.508	0.512
$_{30}$Zn	0.513	0.509	0.513
$_{32}$Ge	0.512	0.511	0.516
$_{38}$Sr	0.521	0.518	0.521
$_{39}$Y	0.523	0.519	0.523
$_{40}$Zr	0.524	0.520	0.524
$_{41}$Nb	0.526	0.521	0.525
$_{42}$Mo	0.525	0.523	0.526
$_{45}$Rh	0.525	0.527	0.528
$_{46}$Pd	0.526	0.528	0.529
$_{47}$Ag	0.522	0.530	0.531
$_{48}$Cd	0.544	0.531	0.532
$_{49}$In	0.539	0.532	0.533
$_{50}$Sn	0.541	0.533	0.534

while the intensity ratio is proportional to the emitted energy. Thus, for a transition $a \to b$, we have

$$I_{ab} = P_{ab} \times \hbar\omega_{ab} \quad . \tag{1}$$

The observed transition probabilities are listed in Table I. Also listed are the most probable values of the transition probabilities which were obtained from a least-squares computer to fit the experimental points. These are compared with Scofield's theoretical calculations.

The measured transition probabilities are plotted as a function of the atomic number in Fig. 2. Also plotted are the experimental values of Refs. 4 and 5 and the smooth curve of Wapstra *et al.*[6] For comparison, we have included the theoretical results of Scofield and those of Babushkin as well as the classical values. The maximum difference between the result of this work and Wapstra's table is about 4%. This could be of importance in this region as the value given in the table shows that the transition probability $K\alpha_2/K\alpha_1$ sinks below its classical limits for $30 \leq Z \leq 40$, and exhibits an increase in value as the atomic number decreases below this region. The present results show a gentle increase in the $K\alpha_2/K\alpha_1$ ratio with increasing atomic number for all the elements studied. Although the experimental results fall within experimental error from the theroretical curves shown, it is apparent that experimental values are larger than Babushkin's results for $Z \geq 38$ and are smaller than Scofield's results for $Z \leq 29$. Both Scofield and Babushkin applied relativistic calculations and included retardation effect, and both neglected the finite size of the nucleus which, if included, would have lowered their results of $K\alpha_2/K\alpha_1$ ratio of $_{92}$U by about 0.4% only. Therefore, the main difference in their calculations lies in their choice of different potentials.

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