Double K-shell ionization in the electron capture decays of ^{65}Zn

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The probability per K capture for double K-shell ionization in the electron capture decay branches of ^{65}Zn has been studied by recording coincidences between Cu K x rays and Cu $K\alpha$ x rays produced when the two K shell vacancies are filled. A combined probability for the two decay branches was found to be $(2.2\pm0.2)\times10^{-4}$, a value which is approximately a factor of 1.5 greater than recent theoretical predictions by Suzuki and Law. The energy shift of the hypersatellite Cu $K\alpha^H$ x ray was found to be (321 ± 17) eV.

RADIOACTIVITY Measured Cu K x-ray—$K\alpha$ x-ray coincidences in electron capture decays of ^{65}Zn; deduced combined double K-shell ionization probability and hypersatellite $K\alpha^H$ x-ray shift.

I. INTRODUCTION

In nuclear decay by electron capture (EC), double ionization of the atomic K shell may occur. Such an event happens when one K electron is captured and the other is excited to an unoccupied bound state (shakeup, SU) or completely ejected from the atom (shakeoff, SO) owing to the nearly compensating effects of the sudden reduction in nuclear charge and the sudden disappearance of the electron-electron Coulomb interaction. Filling of the two vacancies gives rise to two almost simultaneous radiations, being either two K x rays, two Auger electrons or both a K x ray and an Auger electron. Reviews of this effect have been presented by several authors.[1–3]

The first theoretical attempt to determine the probability per K capture for double K-shell ionization, P_{KK}, was done by Primakoff and Porter[4] using nonrelativistic hydrogenic wave functions with fixed effective charge screening parameters for the two K-shell electrons in the parent atom. A second study, by Intemann and Pollock[5] and Intemann,[6] used relativistic Coulomb wave functions to calculate the shakeoff probability, P_{KK}^{SO}, but not the shakeup probability, P_{KK}^{SU}

$$(P_{KK}=P_{KK}^{SO}+P_{KK}^{SU}) .$$

These initial studies have been extended in various fashions by several authors.[7,8] Mukoyama, Isozumi, Kitahara, and Shimizu (MIKS) (Ref. 9) used Dirac wave functions which were shielded according to effective charges determined from self-consistent-field calculations to determine P_{KK}^{SO} as well as P_{KK}^{SU}. The most recent theory, by Suzuki and Law (SL),[10] uses Dirac-Fock-Slater wave functions and self-consistent-field calculations in a more sophisticated description of the initial and final electron states. In their calculations SL found $P_{KK}^{SU}\ll P_{KK}^{SO}$, so that $P_{KK}\simeq P_{KK}^{SO}$.

Comparison between theoretical predictions and experimental values of P_{KK} shows that no one theory gives good overall agreement (See Tables I and II of Ref. 10). The theoretical predictions of MIKS and of Intemann have shown good agreement for several isotopes, but generally give values which range up to several times smaller than experimentally measured values. The recent theory of SL, while giving better results at low Z than the MIKS and Intemann theories, gives theoretical values which are approximately a factor of 2 larger than experimental values at high Z. To establish a firmer basis for comparison to theory, more experimental values, as well as better measurements in earlier cases, are needed. This study of double K-shell ionization in the EC decays of ^{65}Zn is an effort to help establish this experimental base.

The experimental determination of P_{KK} is generally accomplished by coincidence measurements between the K x rays emitted when the double vacancy is filled. These x rays, a K hypersatellite x ray (K^H; $1s^{-2}\rightarrow1s^{-1}2p^{-1}$), and a secondary K satellite x ray (K^S; $1s^{-1}2p^{-1}\rightarrow2p^{-2}$) are shifted to higher energy with respect to the normal K x rays, with the K^S

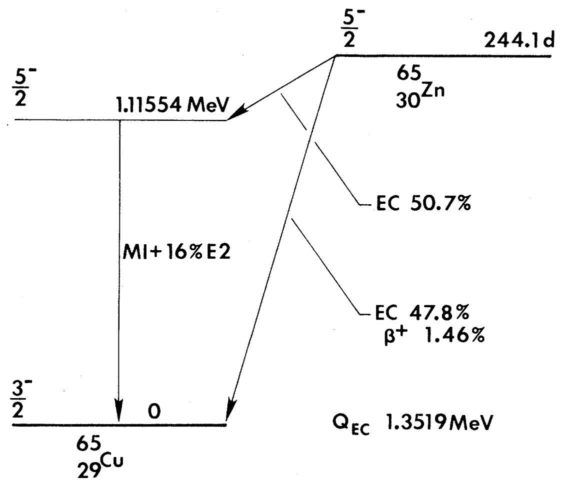

FIG. 1. Decay scheme of ^{65}Zn.

x ray shifted only about 10% of the shift of the K^H x ray. Coincidences between these x rays are generally difficult to measure, owing to the small magnitude of P_{KK} ($10^{-4}-10^{-5}$ per EC decay) and to competing events in most decays (such as K x rays produced in EC in coincidence with K x rays produced in internal conversion or in coincidence with low energy Compton photons from gamma decay). For these reasons, most experimental studies of P_{KK} have been performed on isotopes where the EC decay leads directly to the ground state and no gamma transitions are present. However, due to the development of high resolution Si(Li) x-ray detectors (as used in this experiment) and intrinsic Ge detectors, the K^H x rays can now be resolved from the normal K x rays, and problems introduced by competing events are greatly reduced.

In this experiment, the double K-shell ionization of ^{65}Zn was studied by coincidence measurements between the Cu $K\alpha^H$ x rays and Cu K^S x rays emitted when the double vacancy was filled. Figure 1 shows the decay scheme of ^{65}Zn.[11] Double K-shell vacancies are produced in both the EC to the 1.116-MeV excited level and to the ground state of ^{65}Cu. Competing events as discussed above would be present owing to the 1.116-MeV transition. While the coincidence measurement as performed in this experiment does not yield the probabilities for double K-shell ionization in the EC decays to the excited state, P_{KK} (EX), and to the ground state, P_{KK} (g.s.), separately, it does yield a combination of these probabilities which can be compared to the same theoretical combination. ^{65}Zn was chosen for this measurement owing to the very strong EC branches (98.5%), the long half-life of the isotope, and the simplicity with which one can determine the product of absorption and detector efficiency factors by coincidence measurements between the 1.116-MeV gamma rays and Cu K x rays produced in the EC decay to the excited level.

II. EXPERIMENTAL PROCEDURES

A. Electronic circuitry

In this work, detection of the coincidences between the Cu $K\alpha^H$ and K^S x rays ($K\alpha^H K^S$ coincidences) was accomplished by use of the electronic circuit shown schematically in Fig. 2. While both types of x rays were detected in both detectors, data analysis was performed on Cu $K\alpha^H$ x rays in the Si(Li) detector. Hence, as far as data analysis was concerned, Cu K^S x rays were detected in the NaI(Tl) x-ray detector. The 12.7 cm×15.2 cm NaI(Tl) detector shown in the schematic diagram was used in auxiliary experiments to determine the product of the absorption and efficiency factor for Cu $K\alpha$ x rays in the Si(Li) detector by coincidence measurements between the 1.116-MeV gamma rays and the Cu $K\alpha$ x rays produced in EC decay to that excited level. For the $K\alpha^H K^S$ coincidence measurements, pulses from the NaI(Tl) x-ray detector, windowed on the Cu K x ray, were used for the start pulses to the time-to-amplitude converter (TAC), whereas for the auxiliary coincidence measurements pulses from the 12.7 cm×15.2 cm NaI(Tl) detector, windowed on the 1.116-MeV photopeak, were used for the start pulses.

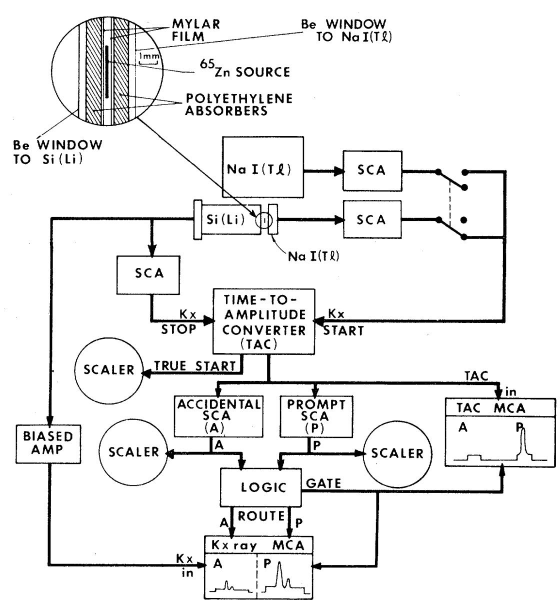

FIG. 2. Schematic diagram of detectors and circuits. Preamplifiers, amplifiers, and delay circuits have been omitted for simplicity.

The overall circuit simultaneously recorded the accidental and true-plus-accidental coincidences by use of both the "accidental" and "prompt" single channel analyzers (A SCA and P SCA) on the output from the TAC. These single channel analyzers routed P and A pulses to different halves of the multichannel analyzer (MCA) memory recording the Si(Li) K x-ray spectra. P and A window widths corresponded to about 300 nsec. Since subtraction of accidental coincidences requires that the relative window widths be accurately known, periodic checks on both the P SCA and A SCA window widths were made during the course of each run. To do this, the accidental rate was temporarily increased by lowering the discriminator on the Si(Li) pulses leading into the TAC stop into the noise. Necessary adjustments would then be made on the window widths to equalize them. Typically, such adjustments would change a window width by less than 3%.

The P and A scalers shown on the schematic diagram were used as part of an overall check of the system. At the end of each run, the P scaler reading was compared to the total number of counts in the P window of the TAC spectrum and to the total number of counts in the P Si(Li) x-ray spectrum. Similar comparisons were made between the A scaler and the A spectra. Typically, a set of such readings agreed to better than 3%.

B. Detectors

This investigation used an Ortec 7000 series Si(Li) x-ray detector with a 4 mm diameter, a 4.2 mm sensitive depth, an 0.008 mm Be window, a 10 mm window to detector distance, and a resolution width (FWHM) of 160 eV at 5.9 keV. The NaI(Tl) x-ray detector had a 5.1 cm diameter, a thickness of 3 mm, and a 0.13 mm Be entrance window.

C. ^{65}Zn source

A 0.98 μCi ^{65}Zn source prepared from ^{65}Zn activity produced by neutron irradiation of 10 mg of ^{64}Zn for 24.25 h at 4.6×10^{14} n/cm^{2}sec was used in this measurement. The source had decayed for approximately four half-lives. The source was prepared by evaporating several drops of active solution onto a 0.07 mm thick Mylar backing. After drying, the source was covered with a second piece of Mylar and sandwiched between two 1 mm thick polyethylene absorbers. This combination was then placed in contact with the Be window on the Si(Li) detector to provide maximum efficiency for this detector. During the course of the measurement three separate runs, whose durations were 188.7, 88.2, and 183.1 h, were made using this source. A Ge(Li) spectrum of the source the only contaminant to be ^{60}Co, with an activity level of 10^{-3} times that of the ^{65}Zn.

III. DATA ANALYSIS

The total number of $K\alpha^H K^S$ coincidences recorded during a run, $N(K\alpha^H K^S)$, is given by

$$N(K\alpha^H K^S)=N_0[f_{\rm EX}(K/T)_{\rm EX}P_{KK}({\rm EX})+f_{\rm g.s.}(K/T)_{\rm g.s.}P_{KK}({\rm g.s.})]\omega_K^S(aE)_{\rm NaI}^S\omega_K^H(K_\alpha/K_T)^H(aE)_{\rm Si(Li)}^H E_T\ , \quad (1)$$

where N_0 is the total number of decays during a run, $f_{\rm EX}=0.507$ and $f_{\rm g.s.}=0.478$ are the fraction of decays which go by EC to the excited level or to the ground state,[11] $(K/T)_{\rm EX}$ and $(K/T)_{\rm g.s.}$ are the K-shell-to-total capture ratios for the EC decays to the excited level and to the ground state, $P_{KK}({\rm EX})$ and $P_{KK}({\rm g.s.})$ are the probabilities for double K-shell ionization in the two EC decays, ω_K^S and ω_K^H are the K-shell fluorescence yields for the Cu K^S and K^H x rays, $(aE)_{\rm NaI}^S$ and $(aE)_{\rm Si(Li)}^H$ are the products of absorption factors (a) and total detection efficiencies (E) for the Cu K^S x rays in the NaI(Tl) detector and for the Cu $K\alpha^H$ x rays in the Si(Li) detector, $(K_\alpha/K_T)^H$ is the $K\alpha^H$ fraction of all the K^H x rays, and E_T is the TAC coincidence efficiency. Taking

$$(K/T)_{\rm Ex}=(K/T)_{\rm g.s.}=(K/T)=0.90$$

(Ref. 11) and defining P_{KK} such that

$$P_{KK}=f_{\rm EX}P_{KK}({\rm EX})+f_{\rm g.s.}P_{KK}({\rm g.s.})\ , \quad (2)$$

Eq. (1) becomes

$$N(K\alpha^H K^S)=N_0(K/T)P_{KK}\omega_K^S(aE)_{\rm NaI}^S \times\omega_K^H(K\alpha/K_T)^H(aE)_{\rm Si(Li)}^H E_T\ . \quad (1a)$$

The total number of Cu K x rays detected in the NaI(Tl) x-ray detector during a run, $N(K)$, is given by

$$N(K)=N_0(K/T)[f_{\rm EX}+f_{\rm g.s.}]\omega_K(aE)_{\rm NaI}\ , \quad (3)$$

where ω_K is the K-shell fluorescence yield for normal Cu K x rays and $(aE)_{\rm NaI}$ is the product of the absorption factor and total detection efficiency for normal K x rays in the NaI(Tl) x-ray detector. The very low intensity contribution to the K x rays by internal conversion of the 1.116-MeV transition has been neglected in Eq. (3), as well as the contributions of K^H and K^S. Assuming the $\omega_K^S=\omega_K^H=\omega_K$, where $\omega_K=0.445$,[11] and that $(aE)_{\rm NaI}^S=(aE)_{\rm NaI}$, the ratio of Eq. (1a) to Eq. (3) gives

$$P_{KK}=\left[\frac{N(K\alpha^H K^S)}{N(K)}\right]\left[\frac{f_{\rm EX}+f_{\rm g.s.}}{E_T}\right]\times\left[\frac{1}{(aE)_{\rm Si(Li)}^H}\right]\left[\frac{1}{\omega_k(K\alpha/K_T)^H}\right]. \quad (4)$$

The $(aE)_{\rm Si(Li)}^H$ term of Eq. (4) was taken as equal to the $(aE)_{\rm Si(Li)}$ term for normal $K\alpha$ x rays and was determined by both singles spectra measurements of the $K\alpha$ x-ray count rate using the known decay rate of the ^{65}Zn source and by auxiliary coincidence measurements between $K\alpha$ x rays produced in the EC decay to the 1.116-MeV excited level and the 1.116-MeV gamma rays. For each of the $K\alpha^H K^S$ runs, a singles measurement and an auxiliary coincidence measurement were taken at both the start and the end of the run. The average of the four values of $(aE)_{\rm Si(Li)}$ acquired was then used in Eq. (4). The four values acquired for any one run were consistent to less than 3%.

For normal Cu x rays $(K\alpha/K_T)=0.88$.[11] Åberg *et al.*[12] pointed out, however, that the ratio of $K\alpha_1^H$ x rays to $K\alpha_2^H$ x rays decreases considerably in the case of low Z. For normal Cu x rays, this ratio is 1.96,[11] whereas this ratio is about 0.3 (Ref. 12) for hypersatellite x rays. This value of 0.3 has been verified by Briand *et al.*[13] in studies where the hypersatellites were produced by electron bombardment of Cu targets. Assuming only the $K\alpha_1^H$ transition is suppressed, then $(K\alpha/K_T)^H=0.77$. Isozumi *et al.*[14] in their study of double K-shell ionization in the EC decay of ^{131}Cs found the $(K\alpha/K_T)^H$ ratio for Xe K^H x rays to be very near that of normal K x rays. The suppression of the $K\alpha_1^H$ transition is not as large at higher Z,[12] however. For this study a value of

$$(K\alpha/K_T)^H=0.83\pm0.05$$

was used in Eq. (4).

The $N(K\alpha^H K^S)$ term of Eq. (4) was determined from least-squares computer fits to the $K\alpha$-$K\alpha^H$ region on the true coincidence spectra recorded with the Si(Li) detector. For a given run the true coincidence spectrum was acquired by the subtraction of the computer fit accidental spectrum from the actual prompt spectrum. The accidental spectrum had been fit in the $K\alpha$ region with the sum of a modified Gaussian distribution and a linear continuum. The modified Gaussian distribution used had the form suggested by Jorch and Campbell.[15] The $K\alpha$-$K\alpha^H$ region of the true coincidence spectrum was then fit with the sum of two modified Gaussian distributions, one representing the $K\alpha$ peak and the other representing the $K\alpha^H$ peak, and a linear continuum. Since it was felt that the overall shape of the x ray peaks was determined by detector characteristics rather than line characteristics, the shape parameters were taken as the same for both the $K\alpha$ and $K\alpha^H$ peaks in the computer fit to the true spectrum. A measure of agreement between the experimental data and the computer fit in the $K\alpha$-$K\alpha^H$ region was taken as

$$\chi^2=\frac{1}{n}\sum_{i=1}^{n}\left[\frac{N_i-F_i}{\Delta N_i}\right]^2, \quad (5)$$

where n is the number of channels involved in the fit (typically 90 channels), N_i is the number of counts in channel i, F_i is the number of counts in channel i as given by the computer fit, and ΔN_i is the statistical error associated with N_i. The agreement between data and fit was satisfactory, with an average χ^2 value of 1.83 for the three runs.

$N(K)$ was determined on the TAC true starts scaler with a 6% reduction due principally to the Compton continuum from the 1.116-MeV gamma ray in the Cu K x-ray region. E_T was determined from TAC spectra recorded simultaneously during the course of each run and was equal to 0.95 ± 0.01 for all three runs.

IV. RESULTS

Figure 3(a) shows the $K\alpha$-$K\alpha^H$ region of the true coincidence Si(Li) spectrum for the 188.7 h run. The solid curve indicates the overall computer fit to the region, whereas the dashed curves indicate the components of the computer fit. Figure 3(b) shows the $K\alpha$-$K\alpha^H$ region of the accidental spectrum, normalized to the height of the true spectrum, for com-

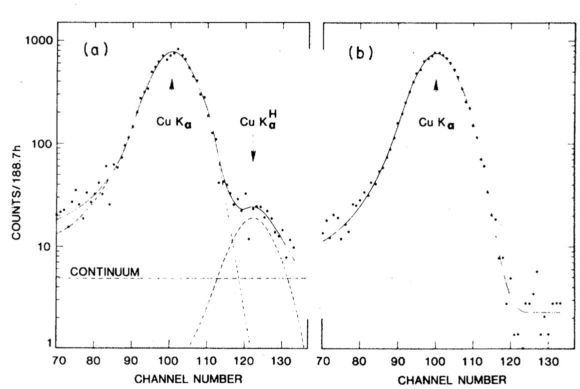

FIG. 3. (a) True coincidence spectrum in the $K\alpha$-$K\alpha^H$ region. The solid curve gives the overall computer fit to the data, whereas the dashed curves give the components of this fit, the $K\alpha$ x-ray peak, the $K\alpha^H$ x-ray peak, and the linear continumm. (b) Normalized accidental spectrum in the $K\alpha$-$K\alpha^H$ region.

TABLE I. Data necessary for the calculation of P_{KK} from Eq. (4).

Run	Time (h)	$N(K\alpha^H K^S)$	$N(K)$ $(\times 10^{-6})$	$(aE)_{\mathrm{Si(Li)}}$ $(\times 10^3)$	P_{KK} $(\times 10^4)$	ΔE (eV)
I	188.7	278±17	706	5.26±0.15	2.1±0.2	304±16
II	88.2	128±11	304	5.19±0.15	2.3±0.3	321±17
III	183.1	270±16	654	5.16±0.15	2.2±0.2	337±18
Average					2.2±0.2	321±17

parison. The large number of counts in the $K\alpha$ x-ray peak came from several sources, namely: (a) $K\alpha^S$ x rays in coincidence with $K\alpha^H$ x rays in the NaI(Tl) x-ray detector; (b) $K\alpha$ x rays in coincidence with K x rays in the NaI(Tl) x-ray detector produced by coincidences between EC K x rays and K x rays from the K internal conversion of the 1.116-MeV transition; (c) $K\alpha$ x rays following K EC in coincidence with internal bremsstrahlung photons produced in the EC decays and detected in the NaI(Tl) x-ray detector; and (d) $K\alpha$ x rays following K EC in coincidence with the low-energy Compton distribution of the 1.116-MeV gamma rays in the NaI(Tl) x-ray detector.

Table I shows the data necessary to calculate P_{KK} from Eq. (4) for each run as well as the results of these calculations. The uncertainties on the individual P_{KK} values are statistical uncertainties based upon the uncertainties associated with $N(K\alpha^H K^S)$, $(aE)_{\mathrm{Si(Li)}}$, and $(K\alpha/K_T)^H$. The average value of P_{KK}, $(2.2\pm0.2)\times 10^{-4}$, is the unweighted average of the individual P_{KK} values. The uncertainty on the average value of P_{KK} is the average value of the uncertainties on the individual P_{KK} values and has not been statistically reduced because of systematic errors which may have existed in detection efficiencies, computer fitting procedures, and the assigned value of $(K\alpha/K_T)^H$.

Two other processes which could give rise to simultaneous, rather than successive, K vacancies and thus to $K\alpha^H$ x rays in the coincidence spectra, and thereby affect the P_{KK} value, were considered by the authors:

(1) K-shell internal conversion of the 1.116 MeV transition before the K-shell vacancy produced by K EC to the 1.116 MeV excited level is filled. The K vacancy lifetime in Cu, however, is about 5×10^{-16} sec (Ref. 16), while the lifetime of the 1.116 MeV excited state is 2.6×10^{-13} sec.[11] Hence any contribution due to this effect is completely negligible.

(2) Double K-shell ionization from K-shell shaking accompanying the K-shell internal conversion of the 1.116 MeV transition. Since the probability per K internal conversion for K-shell shaking is typically on the order of 10^{-4} and the K-shell internal conversion coefficient for the 1.116 MeV transition is also on the order of 10^{-4}, the probability for double K-shell ionization produced by this process will be on the order of 10^{-8} per ^{65}Zn decay to this level. Hence the probability for this effect would be negligible, only about 10^{-4} times the measured P_{KK} value.

Also shown in Table I are the energy differences between the Cu $K\alpha^H$ and Cu $K\alpha$ x rays,

$$\Delta E = E(K\alpha^H) - E(K\alpha)$$

for each run. The $E(K\alpha)$ value of this expression is taken as the weighted average of $E(K\alpha_1)$ and $E(K\alpha_2)$, $E(K\alpha)=8041$ eV.[11] The uncertainties on the individual ΔE values are statistical uncertainties based upon the energy calibration of each spectrum and the peak positions. The average value of ΔE, $(321\pm17$ eV) is the unweighted average of the individual ΔE values; the uncertainty on the average ΔE value is the average value of the uncertainties on the individual ΔE values, unreduced as in the case of P_{KK}. This average ΔE value is about 6% larger than the $E(K\alpha_2^H) - E(K\alpha_2)$ energy difference in Cu of (303 ± 3) eV measured by Briand *et al.*[13]

V. DISCUSSION

Table II shows the theoretical predictions of SL for the K-shell electron shakeoff probabilities in the two decay branches of ^{65}Zn, P_{KK}^{SO}(g.s.) and P_{KK}^{SO}(EX), as well as the theoretical value for the combined probability, P_{KK}^{SO}. The values shown in the SCF column arise when SL use the optimized self-

TABLE II. Theoretical predictions of SL[a] for P_{KK}^{SO}(g.s.), P_{KK}^{SO}(EX), and P_{KK}^{SO} and of MIKS[b] and Intemann[b] for P_{KK}^{SO} $(\times 10^4)$.

	SL			
	SCF	LDA	MIKS	Intemann
P_{KK}^{SO}(g.s.)	1.60	1.71		
P_{KK}^{SO}(EX)	1.32	1.40		
P_{KK}^{SO}	1.43	1.53	≃0.6	≃0.6

[a]Private communication from J. Law.

[b]Z interpolated values acquired from Ar, Fe, and Ge values in Table I of Ref. 10.

consistent-field potential which emerges from the daughter wave function to describe the final continuum electron state. The values shown in the LDA column arise when SL modify the exchange part of the potential and use the local eleectron density to approximate the Fermi momentum in describing the final continuum electron state. Also shown in Table II are theoretical values of P_{KK}^{SO} for the MIKS and Intemann theories. These values were acquired from Z interpolations of their P_{KK}^{SO} values for Ar, Fe, and Ge (see Table I of Ref. 10). While two different decay branches exist in the EC decay of ^{65}Zn, both of these theories (as well as the theory of SL) predict that P_{KK}^{SO} is nearly independent of decay energy when the decay energy is much larger than the K-shell binding energy, as in both decay branches of ^{65}Zn. Hence, it would be expected that

$$P_{KK}^{SO}(\text{g.s.}) \simeq P_{KK}^{SO}(\text{EX}) \simeq P_{KK}^{SO}$$

in both the MIKS and Intemann theories for ^{65}Zn.

The P_{KK} value of $(2.2\pm0.2)\times10^{-4}$ as determined in this investigation is (1.5 ± 0.2) times larger than the SL SCF P_{KK}^{SO} value, (1.4 ± 0.2) times larger than the SL LDA P_{KK}^{SO} value, and (3.7 ± 0.3) times larger than the theoretical predictions of both MIKS and Intemann. While the measured value is larger than predicted by all three theories, it is consistent with other experimentally determined values of P_{KK} in the same Z region and is very similar to the latest P_{KK} measurment for ^{71}Ge by Briand *et al.*[17] for which the experimental value is 2.2 and 2.0 times larger than the SL SCF P_{KK}^{SO} and LDA P_{KK}^{SO} values, respectively, and 5.2 times larger than the value predicted by MIKS and Intemann.

The value of P_{KK} as determined in this investigation is the sum of the probabilities for K-shell electron shakeoff and shakeup, $P_{KK}^{SO}+P_{KK}^{SU}$. Hence, addition to the theoretical P_{KK}^{SO} values of a P_{KK}^{SU} value would bring about better agreement for this measurement. However, SL calculated the probability for K-shell electron shakeup to be only about 1% of the probability for K-shell electron shakeoff in β^- decay,[18] and they expect a similar shakeup-to-shakeoff ratio in EC decay. If their assumption is correct, a shakeup contribution could not account for the observed difference between P_{KK} and P_{KK}^{SO}.

The result of this investigation of the probability for double K-shell ionization in the EC decays of ^{65}Zn then shows a P_{KK} value which is in fair agreement with the SL theory but well above the theoretical predictions of both MIKS and Intemann.

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