Double K-Shell Ionization in the Decay of ^{131}Cs and ^{165}Er

H. J. Nagy, G. Schupp, and R. R. Hurst
Department of Physics, University of Missouri – Columbia, Columbia, Missouri 65201
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Double ionization of the K shell has been studied in the electron capture decays of ^{131}Cs and ^{165}Er by recording coincidences between K x rays emitted when the double vacancies are filled. A Si(Li) x-ray detector was used in conjunction with a NaI(Tl) detector for these measurements. The results obtained for P_{KK}, the probability per K capture that a double K-shell vacancy is formed, were $(1.33\pm 0.33)\times 10^{-5}$ for ^{131}Cs and $(6.7\pm 3.9)\times 10^{-6}$ for ^{165}Er. Both of these values are a factor of 2 smaller than previous measurements and, within the experimental uncertainties, are in agreement with predictions based on the relativistic theory of Intemann.

I. INTRODUCTION

In nuclear decay by electron capture, 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 or completely ejected from the atom due to both the sudden change 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.

The first theoretical study of K electron excitation and ejection accompanying K capture was made in 1954 by Primakoff and Porter.[1] Using nonrelativistic variational wave functions and the sudden-perturbation approximation, they derived expressions for the probability per K capture that a double vacancy is formed, P_{KK}; the probability that the excited electron is ejected from the atom, P_{KKe}; and the momentum spectrum of the ejected electrons. A second study, by Intemann and Pollock[2] and by Intemann,[3,4] used relativistic Coulomb wave functions associated with the "symmetric Hamiltonian" of Biedenharn and Swamy[5] for the initial and final electron states and the scattering matrix formalism to calculate P_{KKe} and the momentum spectrum of the ejected electrons. This latter study, however, did not actually include the probability for excitation to unoccupied bound states. Although the results of both studies gave momentum spectra similar in shape, they differed by approximately a factor of 2 in the probability for K electron ejection, with the Primakoff-Porter values being larger.

A third calculation of an expression for P_{KK} and the momentum spectrum of the ejected electrons has been made by Stephas[6] using the atomic matrix element of Stephas and Crasemann.[7] This matrix element was calculated with exact relativistic hydrogenic wave functions. However, we found that the corrected expression [Eq. (5) of Stephas and Craseman[8]] diverges at small momenta for electrons ejected in β decay. While this expression formally converges for electron-capture cases, it does so quite slowly and is therefore suspect. We obtained $P_{KK}\approx 1.5\times 10^{-3}$ for ^{131}Cs with more than 90% of this value being contributed by electrons ejected with momenta p/m_0c less than 10^{-4} (0.0026 eV).

Among the most prominent experimental work in double K-shell ionization in electron capture has been that of Lark and Perlman[9] in the decay of ^{131}Cs. Using two NaI(Tl) detectors in coincidence to detect the K x rays emitted as a result of the filling of the double vacancy, they obtained a value for P_{KK} which was almost a factor of 2 less than that predicted by the Primakoff-Porter theory. In a similar study of ^{165}Er, Ryde, Persson, and Oelsner-Ryde[10] also found P_{KK} to be almost a factor of 2 less than that predicted by Primakoff-Porter theory. The momentum spectrum of ejected electrons in the decay of ^{55}Fe was studied by Pengra and Crasemann[11] using both proportional counters and semiconductor detectors. Their measured spectrum deviated from theory[1] by being less intense in the low-energy region.

In this work the probabilities for double K-shell ionization in the electron-capture decays of ^{131}Cs and ^{165}Er were reexamined by detection of the coincident K x rays emitted. To this end, a Si(Li) x-ray detector was used in conjunction with a NaI(Tl) detector. The advantage of using the Si(Li) detector in place of a second NaI(Tl) detector was its ability to partially resolve the $K\alpha$ x rays of neighboring elements. This capability was important, since very small amounts of contaminants present in a source could give rise to true coincidences in the x-ray region, which a NaI(Tl) detector would not be able to resolve.

II. EXPERIMENTAL PROCEDURES

A. ^{131}Cs Source

The ^{131}Cs used in this work was produced by neutron irradiation of ^{130}Ba, yielding ^{131}Ba which decays by electron capture to ^{131}Cs with a half-life of 11.7 days. A 0.3-mg sample of $Ba(NO_3)_2$, enriched 48.8% with ^{130}Ba, was irradiated in the University of Missouri research reactor for 95 h at a flux of 1.8×10^{14} neutrons/cm^2 sec. The sample was allowed to decay for a period of 13 days, at which time the ^{131}Cs was separated by a $BaSO_4$ precipitation. The $BaSO_4$ initially precipitated was placed in a Cs carrier solution for a period of 6 days, allowing the solution to pick up the active ^{131}Cs as the ^{131}Ba decayed, but leaving the active Ba in precipitate form. The solution and precipitate were then separated and a small amount of ^{131}Ba activity carried along in the solution was again precipitated. This procedure gave a ^{131}Cs solution which was nearly free of ^{131}Ba and from which a 22-nCi source was taken. The source was purposely made weak in order to make the ratio of accidental to true coincidences as small as practical.

B. ^{165}Er Sources

Because of the 10.3-h half-life of ^{165}Er, a single source could not be used for the coincidence measurements. Instead, eleven 0.1-mg samples of Er_2O_3, enriched 62.7% in ^{164}Er, were irradiated at a flux of 10^{14} neutrons/cm^2 sec for 20 h each over a period of 6 weeks. 14 h after each irradiation the rare-earth elements were separated in a Dowex 50 cation-exchange column with α-hydroxy isobutyric acid as the eluant.[12] Nevertheless, various rare-earth contaminants were present in the sources and their contributions are discussed in conjunction with the results. Each irradiation allowed 3 days of coincidence counting. Initial source strengths varied from 15 to 23 nCi.

C. Source Mounting

Each source was prepared by evaporating a drop of active solution on a 6-mg/cm^2 Mylar backing which was mounted on a 1.6-mm-thick brass plate having a 3.2-mm minimum diameter, 6.4-mm maximum diameter tapered hole. The brass plate was then placed between the detectors which were at an angle of 180° and in contact with the plate to give maximum coincidence efficiency.

D. Detectors and Coincidence System

An 8-mm diameter by 3-mm-thick ORTEC Si(Li) x-ray detector with a 0.25-mm-thick Be window was used in this work. Its full width at half maximum was approximately 600 eV for photons in the 20- to 50-keV region. Detection efficiencies were determined using sources whose decay rates were known to within 5%. Pulses from this detector and from a Harshaw 5.1×5.1-cm NaI(Tl) detector were analyzed in a fast-slow coincidence system. Fast-coincidence resolving times were 41.3 ± 0.7 nsec for the ^{131}Cs measurement and 61.8 ± 0.8 nsec for the ^{165}Er measurement. Using these values, the accidental coincidences per channel were calculated and subtracted to give the true coincidence spectra. The ratio of true to accidental coincidences was near unity for both measurements. The fast-circuit-coincidence efficiency was determined using the slow-coincidence circuit in the following manner. With a variable delay set at the maximum of a prompt curve, a coincidence measurement was made utilizing the fast-slow system and a ^{152}Eu source which gave a true fast coincidence rate, R_F. For this measurement, the energy gates on the ^{152}Eu spectrum spanned exactly the same photon energy regions as used in the corresponding ^{131}Cs or ^{165}Er coincidence run. Without changing either source or geometry, a slow-coincidence measurement was performed in a similar way and the true slow-coincidence rate, R_S, determined. Since the resolving time for the slow-coincidence circuit was 1 μsec, the circuit efficiency for it was taken to be unity. The fast-coincidence-circuit efficiency was then simply given by R_F/R_S.

E. Evaluation of P_{KK}

All data analyses were performed on the $K\alpha$ x-ray peaks of the coincidence spectra recorded with the Si(Li) detector, since the statistics on the $K\beta$ peaks were extremely poor. The probability for double K-shell ionization, P_{KK}, can be calculated from the coincidence count rate using

$$R_{K\alpha,K}=R_KP_{KK}P\epsilon_1\epsilon_2\epsilon_C\,, \qquad (1)$$

where

$R_{K\alpha,K}$ = coincidence rate for a $K\alpha$ x ray in the Si(Li) detector and a K x ray in the NaI(Tl) detector,

R_K = K-electron-capture rate of the source,

ϵ_1 = $K\alpha$ photopeak efficiency of Si(Li) detector, includes solid angle,

ϵ_2 = efficiency of NaI(Tl) detector, includes solid angle,

ϵ_C = coincidence circuit efficiency.

The factor P is the sum of probabilities that, given a double vacancy, a $K\alpha$ and a K x ray will be emitted; i.e., two $K\alpha_1$ x rays, two $K\alpha_2$ x rays, a $K\alpha_1$ and a $K\beta_2'$ x ray, etc. In cases where each x ray is a $K\alpha$ x ray, the probability must be multiplied by a factor of 2, since either x ray can be detected in either detector. Also incorporated into P is a modification of the usual K x-ray relative intensities[13] to account for the specific vacancy left by the first transition. The fluorescence yield, ω_K, was taken to be the same for both K-shell vacancies.

Using the NaI(Tl) K x-ray count rate,

$$R_{\mathrm{NaI}} = R_K \omega_K \epsilon_2 , \qquad (2)$$

and multiplying by the total time of the measurement in Eq. (1) gives

$$N_{K\alpha,K} = N_{\mathrm{NaI}} P_{KK} P \epsilon_1 \epsilon_C / \omega_K , \qquad (3)$$

where $N_{K\alpha,K}$ and N_{NaI} refer to the total number of $K\alpha, K$ coincidences and the total number of NaI(Tl) K x-ray gating pulses, respectively. N_{NaI} was determined by subtracting a small (<5%) continuum from the NaI(Tl) singles spectrum to obtain a multiplicative factor which was applied to the scaled output of the NaI(Tl) single-channel analyzer. The energy width of this single-channel gate was about 8 keV in both measurements.

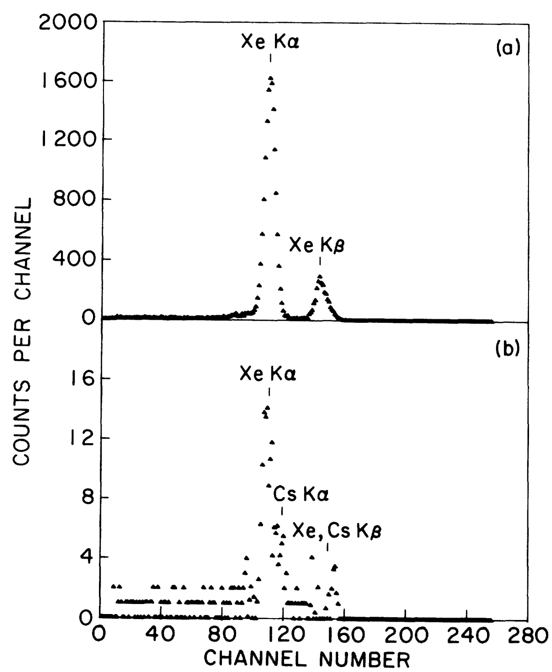

FIG. 1. (a) Si(Li) singles spectrum of ^{131}Cs from 20 to 40 keV. (b) True coincidence spectrum obtained in the study of ^{131}Cs after subtracting accidentals.

III. RESULTS

Figure 1(a) shows the Si(Li) singles spectrum for the decay of ^{131}Cs in the 20- to 40-keV region. This region was set with a single-channel analyzer and offset from zero by use of a biased amplifier. The x rays produced in the decay are characteristic of Xe. Figure 1(b) shows the resultant true coincidence spectrum after 17 days of counting. Some Cs $K\alpha$ x rays from the decay of ^{131}Ba can be seen on the upper side of the Xe $K\alpha$ peak. The low-intensity continuum below the peaks was due to internal bremsstrahlung following the electron capture of ^{131}Cs and to interactions involving the ^{131}Ba contaminant.

Figure 2 shows the Xe $K\alpha$ x-ray region of Fig. 1(b) on an expanded scale. The continuum in this region was determined by a linear least-squares fit to three regions of the coincidence spectrum, channels 9 to 25, 49 to 65, and 130 to 141. The smooth curve, designated as Xe $K\alpha'$, was synthesized by adding one Xe $K\alpha$ singles distribution in the usual position to a second singles distribution of equal intensity shifted upward by 0.43 keV.

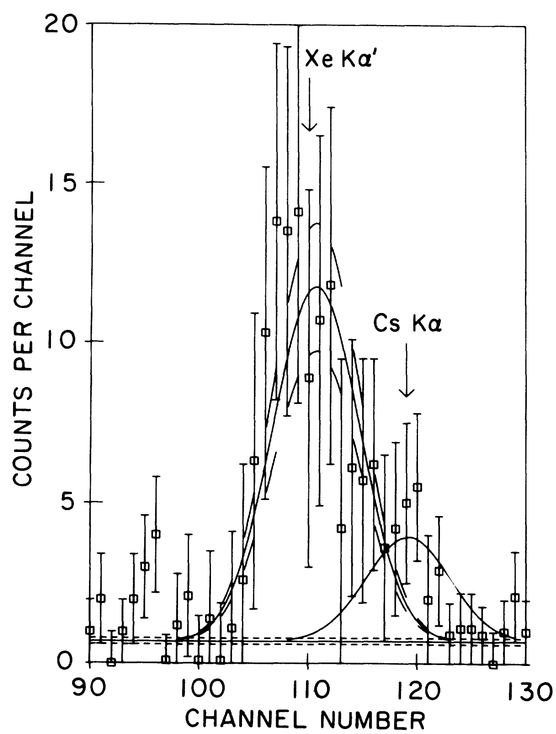

FIG. 2. Xe $K\alpha$ x-ray region of the true coincidence spectrum shown in Fig. 1(b). The smooth curve designated Xe $K\alpha'$ is a synthesized distribution described in the text, whose area has been normalized to the net counts obtained by summing channels 101 through 120. Dashed curves indicate standard deviations.

This shifted distribution is caused by an L electron making a transition into a completely vacant K shell. We used a Slater screening constant, $\Delta Z = 0.3$, to estimate the increase in energy for the shifted $K\alpha$ x ray. Observation of a "hypersatellite" of this type has recently been reported by Briand *et al.*[14] in studies on ^{71}Ga. Although the distribution occurring in coincidence with the hypersatellite is really a (relatively close-lying) KL satellite, it was taken at the position of the usual $K\alpha$ distribution in this evaluation. This synthesized distribution was used in the data analysis only to the extent of determining the channels to be summed to obtain $N_{K\alpha,K}$ and was subsequently normalized to give the appropriate area. The channels summed were 101 through 120 and contributions of 14 ± 2 and 20 ± 4 counts due to the continuum and the Cs $K\alpha$ x rays, respectively, were subtracted. The contribution of the Cs $K\alpha$ x rays was determined by a two Gaussian fit to the Xe, Cs $K\alpha$ x-ray region using the program of Putnam *et al.*[15] Parameters obtained for the Xe $K\alpha$ peak in this two Gaussian fit (not shown) indicated a broadening of the peak with respect to those found for the singles spectrum $K\alpha$ peak shown in Fig. 1(a). The experimental uncertainties, however, prevented a meaningful analysis of the hypersatellite.

Figure 3(a) shows the singles spectrum for the decay of ^{165}Er in the 35- to 60-keV region. The x rays are characteristic of Ho. Figure 3(b) shows the resultant true coincidence spectrum after 30 days of counting. The Sm and Eu x rays are from ^{152m_1}Eu and ^{153}Sm contaminants, respectively, while the Tm x rays are from the decay of ^{171}Er. Very few Ho $K\alpha$ x rays can be seen. The continuum in this case was caused principally by the Compton distributions of photons in coincidence with the Sm, Eu, and Tm x rays.

The $K\beta$ x rays of both Sm and Eu have approximately the same energy as the Ho $K\alpha$ x rays and a correction was necessary for them. To make

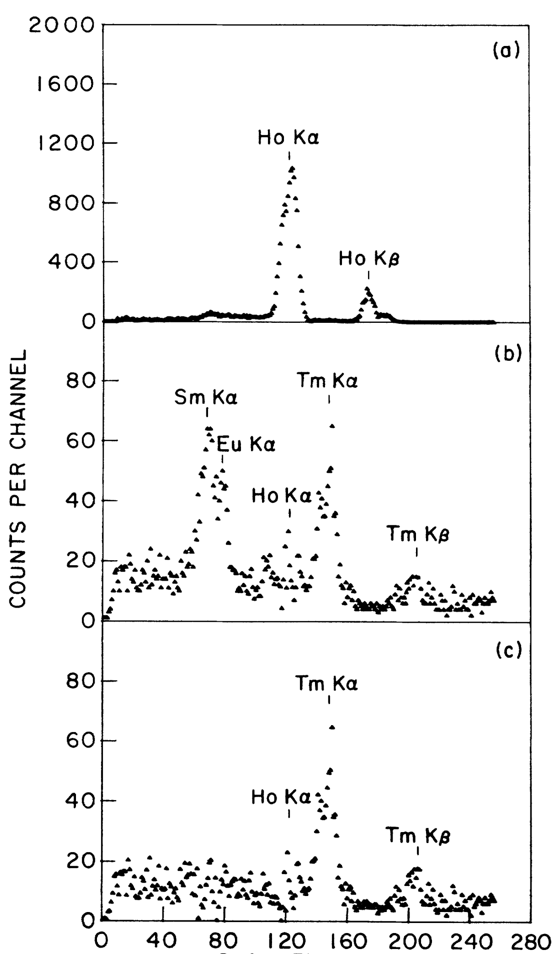

FIG. 3. (a) Si(Li) singles spectrum of ^{165}Er from 35 to 60 keV. (b) True coincidence spectrum obtained in the study of ^{165}Er after subtracting accidentals. (c) Coincidence spectrum after subtracting out contributions from the Sm and Eu contaminants.

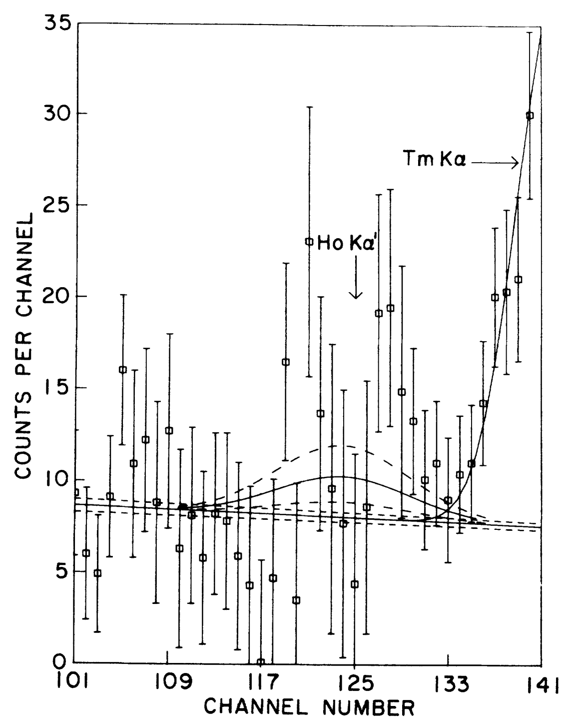

FIG. 4. Ho $K\alpha$ x-ray region of the coincidence spectrum shown in Fig. 3(c). The smooth curve designated Ho $K\alpha'$ is a synthesized distribution, whose area has been normalized to the net counts obtained by summing channels 112 through 134. Dashed curves indicate standard deviations.

TABLE I. Data used for the determination of P_{KK}. Symbols defined in text.

	^{131}Cs	^{165}Er
$N_{K\alpha,K}$	102 ± 20	38 ± 22
$N_{\rm NaI}$	$(2.17 \pm 0.02) \times 10^8$	$(3.22 \pm 0.02) \times 10^8$
ϵ_1	$(5.06 \pm 0.61) \times 10^{-2}$	$(1.84 \pm 0.20) \times 10^{-2}$
P	$1.61\ \omega_K^2$ [a]	$1.58\ \omega_K^2$ [a]
ϵ_C	0.48 ± 0.04	0.63 ± 0.05

[a] Values used for ω_K were taken from V. O. Kostroun, M. H. Chen, and B. Crasemann, Phys. Rev. A 3, 533 (1971).

this correction, a ^{152}Eu source was used to generate a Sm K x-ray distribution. This distribution, adjusted for intensity, was then subtracted from the coincidence spectrum. This same distribution was then shifted to a slightly higher energy to represent a Eu x-ray distribution. This shifted distribution, appropriately adjusted for intensity and for $K\alpha$, $K\beta$ energy differences, was also subtracted from the coincidence spectrum. Figure 3(c) shows the coincidence spectrum after these corrections had been made.

Figure 4 shows the Ho $K\alpha$ x-ray region of Fig. 3 on an expanded scale. The smooth curve in Fig. 4 shows the shape of the Ho $K\alpha'$ x-ray distribution expected in coincidence. This curve was synthesized in a manner similar to that described for the ^{131}Cs data. The continuum through the region was determined by a linear least-squares fit to four regions of the true coincidence spectrum shown in Fig. 3(c) and identified by channels 20 to 51, 90 to 101, 170 to 181, and 220 to 251. The 170 to 181 region was included even though a few Ho $K\beta$ pulses may be present in it. To determine $N_{K\alpha,K}$, the counts in channels 112 through 134 were summed and the continuum in these channels amounting to 176 ± 7 counts was subtracted. A contribution from the Tm $K\alpha$ x-ray tail amounting to 3 ± 1 counts was also subtracted. Table I shows the data used for the evaluation of P_{KK} from Eq. (3) for both the ^{131}Cs and ^{165}Er measurements. The poorer statistics for the Er measurement compared with those for Cs were due to a lower x-ray detection efficiency and a lower probability for P_{KK} as well as the contaminant subtractions. Values for P_{KK} obtained in this work are given in Table II, which summarizes experimental results and theoretical predictions for P_{KK} and P_{KKe} in ^{131}Cs and ^{165}Er.

IV. DISCUSSION

If, in the decay of ^{165}Er, a $\leq 0.0002\%$ electron-capture decay to a 94.7-keV level in ^{165}Ho were present,[13] it would give rise to true coincidences with a probability of $\leq 1.3 \times 10^{-6}$. This possible source of coincidences would then reduce the measured value of P_{KK} by $\leq 19\%$.

Another effect which could give rise to true coincidence $K\alpha$ x rays in the Si(Li) spectra in both the ^{131}Cs and ^{165}Er measurements is the internal bremsstrahlung associated with electron capture. However, due to the low intensity of the internal bremsstrahlung spectrum in the low-energy regions[16] and the narrow NaI(Tl) gates, this effect was estimated to contribute only 1×10^{-6} per K capture. This contribution would be well within the experimental uncertainties and no corrections were made.

The values obtained for P_{KK} (in both measurements) are 3 to 4 times lower than the values predicted by the Primakoff-Porter theory but are midway between the P_{KKe} predictions of Primakoff-Porter and Intemann. The difference between the Primakoff-Porter P_{KK} value, calculated by subtracting from 1 the probabilities per K capture for producing 1 or 0 holes in the K shell, and the P_{KKe} value, calculated directly, formally suggests that

TABLE II. Summary of experimental results and theoretical predictions for P_{KK} and P_{KKe} in ^{131}Cs and ^{165}Er.

	Experimental P_{KK}		Theoretical P_{KK} and P_{KKe}		
	This work	Previous work	P_{KK} (P-P) [a]	P_{KKe} (P-P) [a]	P_{KKe} [b]
^{131}Cs	$(1.33 \pm 0.33) \times 10^{-5}$	$(2.5 \pm 0.2) \times 10^{-5}$ [c] $(5.0 \pm 1.0) \times 10^{-5}$ [d]	4.13×10^{-5}	1.68×10^{-5}	9.82×10^{-6}
^{165}Er	$(6.7 \pm 3.9) \times 10^{-6}$	$(1.5 \pm 0.4) \times 10^{-5}$ [e]	2.7×10^{-5}	0.85×10^{-5}	4.32×10^{-6}

[a] (P-P) refers to the theory of Primakoff and Porter. The multiplicative factor of $\frac{2}{3}$ suggested in Ref. 1 has been used. See discussion of $p^{(0)}$ term below Eq. (15), Ref. 1. The P_{KKe} (P-P) values were calculated using Eq. (17a) of Ref. 1.
[b] Private communication from R. L. Intemann.
[c] Experimental result from Ref. 9.
[d] H. Daniel, G. Schupp, and E. N. Jensen, Phys. Rev. 117, 823 (1960).
[e] Experimental result from Ref. 10.

the excitation process has a large contribution, whereas Intemann has expected this contribution to be small.[4] If excitation is <10% of P_{KKe} the experimental values agree with either calculation; if ~30% the Intemann theory would be preferred.

A recently reported measurement[17] of the spectrum of electrons ejected in the ^{131}Cs decay with kinetic energy greater than 80 keV gave $P_{KKe} = (8.4 \pm 1.5) \times 10^{-7}$. Assuming the spectrum shape predicted by Primakoff-Porter (essentially identical to Intemann[4]), we find that the measured energy range corresponds to 5.9% of the total ejected electron intensity or to a total P_{KKe} value of $(1.43 \pm 0.26) \times 10^{-5}$. This value then compares favorably with our value of $(1.33 \pm 0.33) \times 10^{-5}$ for P_{KK} but is somewhat higher than the Intemann prediction.

It is worth noting that the various contaminant peaks resolved by the Si(Li) detector and subsequently excluded would have been included in a NaI(Tl) x-ray peak and would have given erroneously large values for P_{KK}. This effect may have led to the larger values obtained for P_{KK} in the past. In this respect, a higher-resolution detector and isotope-separator-prepared sources should lead to better experimental values for P_{KK} as well as information concerning the hypersatellites.

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