K-Electron Excitation Accompanying K Capture in Cs^{131}†

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The probability of production of an atom with a completely vacant K shell by excitation of the second K electron during the K-electron capture process has been determined to be $(2.5\pm0.2)\times10^{-5}$ per K-capture event in Cs^{131}. A scintillation coincidence spectrometer with a coincidence resolution time of ten millimicroseconds for 30-kev x rays was used for the measurement. The half-life of Cs^{131} has been redetermined to be 9.69 ± 0.05 days. Previous measurements of electron excitation phenomena accompanying K capture are summarized and the results are compared with the theory of Primakoff and Porter.

INTRODUCTION

THE electronic structure of an atom must change to adapt to the modified potential field when the nuclear charge changes during radioactive decay. Although usually the electronic rearrangement is minor and only small amounts of energy are involved,[1] there is a finite probability of more drastic interaction leading to highly excited states of the final atom.[2] An especially interesting case is that of K-electron excitation during K capture, which produces an atom with a completely vacant K shell. The probability of occurrence of this phenomenon was calculated by Primakoff and Porter[3] in 1953. Since then seven observations of the various manifestations of this process have been reported.[4–10] The results of the more accurate measurements, those on Ar^{37}, Ge^{71}, and Cs^{131}, seem to indicate that the theory is valid to within fifty percent or less. This apparent agreement may be misleading, however, for in some of the determinations only part of the effects were observed and the theory itself was used to estimate the total electron excitation probability.

The theoretical calculations are independent of the poorly known nuclear transition probabilities and depend only on electronic wave functions. The results are especially sensitive to the effects of electron-electron correlation in the initial state of the two K electrons. Thus more accurate measurements may lead to a better knowledge of this aspect of atomic structure.

EXPERIMENTAL CONSIDERATIONS

The two vacancies in the K shell are filled primarily by the emission of two K x rays for elements with atomic number greater than about forty. This makes it possible to measure this phenomenon by x ray, x-ray coincidence counting with scintillation crystals as detectors. The potential efficiency of this technique is high, being limited only by the competition from Auger transitions and by source-detector geometry. The sensitivity is limited by the background and the finite resolving time of the coincidence circuit and by the problems of source preparation.

X-ray coincidence counting can be used to detect the process of interest only for a nuclide which decays in such a simple manner that there is no other possible source of coincident radiations. This excludes from consideration nuclides which decay partly by positron emission or which decay by K capture to short-lived excited states. Even for a long-lived excited state, internal conversion can lead to spurious coincidences between the x rays and the conversion electrons or bremsstrahlung from the electrons. Negative beta decay can also be a source of obscuring coincident gamma radiation; the probability of a single beta particle producing one internal bremsstrahlung quantum as it is emitted and producing an external bremsstrahlung quantum as it interacts with surrounding matter is comparable to the probability of the electron excitation process in K capture. The nuclide to be studied must be one which can be produced with high specific activity and free from radioactive contaminants to a very high degree. Careful consideration indicates that only Cs^{131} is suitable with the techniques now available. Cs^{131} decays only by electron capture to the ground state of stable Xe^{131} with a half-life of ten days.[11,12] The fluorescence yield of xenon is 0.87 and its K x-ray energy is 30 kev.[13]

† This research was performed under the auspices of the U. S. Atomic Energy Commission. It is described in greater detail in the Ph.D. thesis of N. Lark, Cornell University, 1960 (unpublished).

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[3] H. Primakoff and F. T. Porter, Phys. Rev. **89**, 930 (1953).
[4] F. T. Porter and H. P. Hotz, Phys. Rev. **89**, 938 (1953).
[5] G. Charpak, Compt. rend. **237**, 243 (1953).
[6] J. A. Miskel and M. L. Perlman, Phys. Rev. **94**, 1683 (1954).
[7] E. A. Soltysik, Ph.D. thesis, Indiana University, 1955 (unpublished).
[8] M. Langevin, Compt. rend. **245**, 664 (1957); J. phys. radium **19**, 34 (1958).
[9] R. W. Kiser and W. H. Johnston, J. Am. Chem. Soc. **81**, 1810 (1959); R. W. Kiser, Ph.D. thesis, Purdue University, 1958 (unpublished).
[10] H. Daniel, G. Schupp, and E. N. Jensen, Phys. Rev. **117**, 823 (1960).
[11] Nuclear Data Sheets (National Academy of Sciences—National Research Council, Washington, D. C.).
[12] D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958).
[13] A. H. Wapstra, G. J. Nijgh, and R. Van Lieshout, *Nuclear Spectroscopy Tables* (Interscience Publishers, Inc., New York, 1959).

SOURCE PREPARATION AND PURITY

To obtain Cs^{131}, $BaCO_3$ was irradiated in the Brookhaven reactor. The irradiated barium was purified by repeated precipitation as $BaCl_2$, with special attention paid to removal of cesium. After about two weeks the Cs^{131} which had been produced from the decay of 12-day Ba^{131} was separated from the barium by a similar precipitation procedure. The cesium was further purified by a procedure described by Hyde,[14] which utilizes coprecipitation of cesium silicotungstate with silicotungstic acid and an ion exchange separation using Dowex 50. Two separate preparations of Cs^{131} were made. Both were practically carrier-free. Initially, sources were mounted between two layers of 0.00025-inch Mylar film stretched across a $1\frac{1}{2}$-inch diameter hole in an aluminum plate. Later, sources were mounted as shown in Fig. 1.

Since almost any radioactive contaminant would be a source of coincident radiation, it was essential to establish clearly the radiochemical purity of the Cs^{131}. X-ray and gamma-ray spectra of both preparations were measured both shortly after purification and about ten half-lives later. Careful analysis of these spectra indicated no radiation was present except the xenon K x ray and internal bremsstrahlung associated with the Cs^{131} electron-capture decay. The upper limit measured for radioactive impurities was much too small to account for the observed x-ray coincidence effect. By use of several samples with different amounts of cesium, it was possible to follow the decay of the Cs^{131} activity for very long periods, fourteen half-lives for the first preparation and twenty-four half-lives for the second. The decay curves showed no deviation from straight lines. The average value for the half-life obtained from six individual determinations is 9.69 ± 0.05 days. This value is in accord with previously reported values and has a smaller uncertainty.

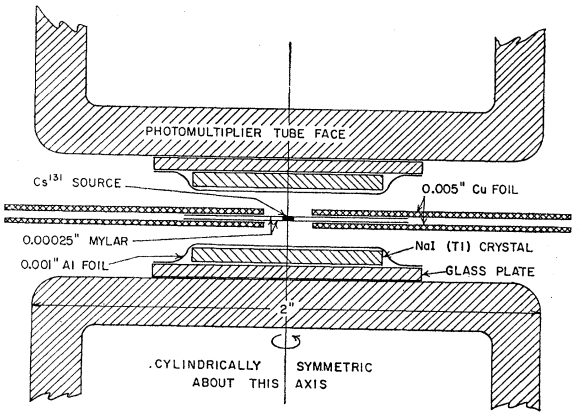

FIG. 1. Details of the source mount and detector arrangement. To maintain clarity, the aluminum foil cover of the photomultiplier tube and the aluminum absorbers between the source and the detectors have not been shown.

[14] E. K. Hyde, J. Am. Chem. Soc. 74, 4181 (1952)

For calibration purposes to be explained later, I^{125} was used as a convenient source of effectively coincident x rays. It decays primarily by K-electron capture followed by a 35-kev transition which, to a large extent, is internally converted in the K shell[11,12]; thus a considerable fraction of the decay events leads to emission of two K x rays. The I^{125} was obtained by neutron irradiation of gaseous xenon to which a small amount of elemental iodine had been added as a carrier. Eighteen-hour Xe^{125} is formed by neutron capture in Xe^{124} and decays to 60-day I^{125}. After irradiation the iodine was dissolved in dilute ammonia. The solution was placed in contact with a thin copper foil and acidified with sulfuric acid. The iodine then reacted with the copper to produce an adherent deposit of cuprous iodide.[15]

THE COINCIDENCE SCINTILLATION SPECTROMETER

The coincidence spectrometer used for these measurements is shown schematically in Fig. 2. The detectors are NaI(Tl) crystals, 3 cm^2 in area and 2 mm thick, coupled to RCA-6342 photomultiplier tubes. Descriptions of similar fast-slow coincidence circuits and discussion of the pulse-clipping technique are available in the literature.[16–18] The resolving time and the coinci-

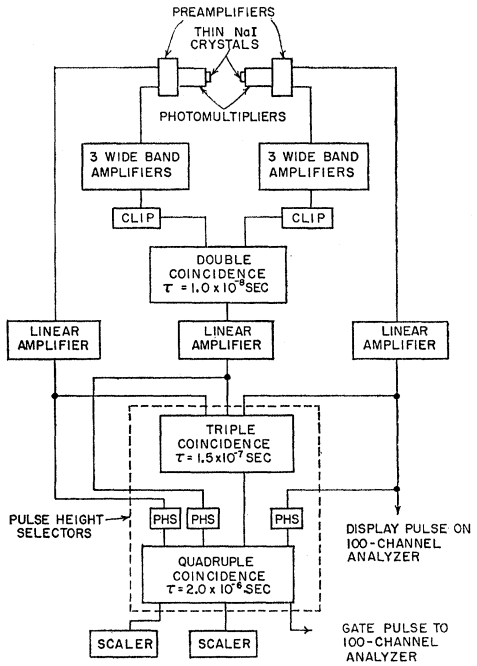

FIG. 2. Schematic diagram of the fast-slow coincidence spectrometer.

[15] G. Friedlander and W. C. Orr, Phys. Rev. 84, 484 (1951).
[16] F. K. McGowan, Phys. Rev. 93, 163 (1954).
[17] P. R. Bell, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. V.
[18] R. E. Bell, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. XVIII.

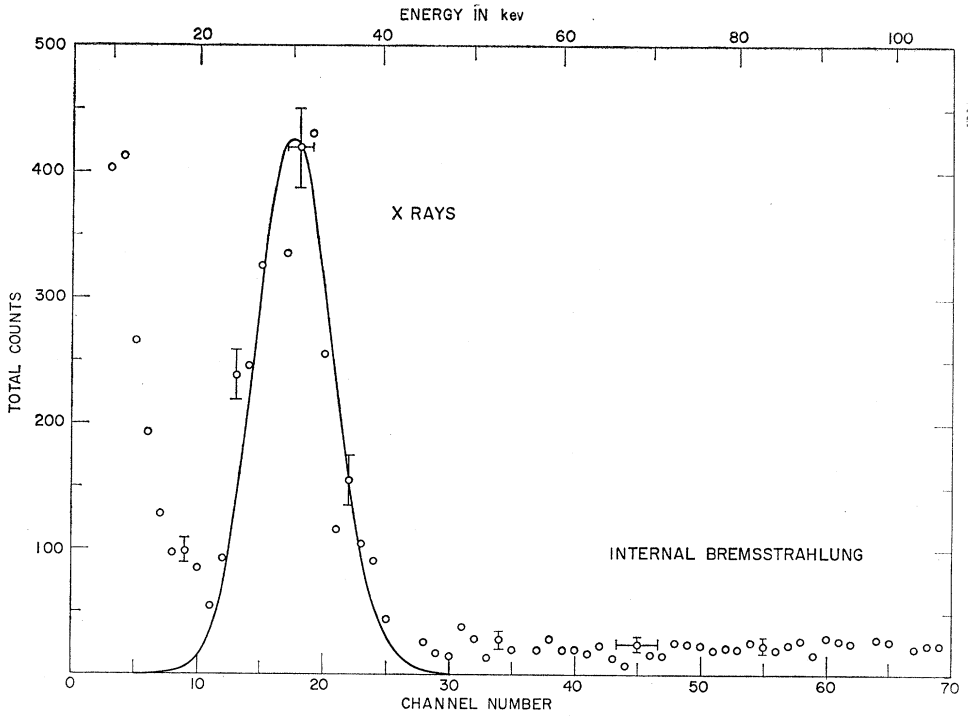

FIG. 3. Pulse-height spectrum of radiation in coincidence with K x rays from the decay of Cs^{131}. Natural background and accidental coincidences have been subtracted. The smooth curve shows the shape of a single x-ray spectrum for comparison. The uncertainty in the number of counts in each channel is based on the statistical uncertainty due to the small number of gross counts in that channel. The uncertainty in the abscissa is the uncertainty in the energy calibration due to the drifting of the pulse amplification.

dence efficiency of the over-all circuit are directly related through the behavior of the fast double coincidence unit. The essential component of this unit, a 6BN6 coincidence gate tube, gives a small output pulse whenever either of its control grids receives an input pulse and gives a much larger output pulse when both grids receive pulses simultaneously. Thus it is necessary to use pulse-height discrimination on the output. The optimum discriminator setting gives a coincidence counting efficiency for the circuit of about 90% and a resolving time of ten millimicroseconds for 30-kev x rays.

The slower components of the circuit are practically loss-free; the fast double coincidence circuit is the only component for which the electronic efficiency must be determined. This was measured quite simply by the use of I^{125} as a source of effectively coincident x rays. The coincidence counting rate of the over-all circuit was measured with and without the requirement of an output pulse from the fast double coincidence circuit. The ratio of these two counting rates gave directly the efficiency of the fast circuit, since the single counting rates were low enough that the accidental coincidence counting rate was negligible even with the long coincidence resolution time of the slow circuit.

ANALYSIS OF THE DATA

The quantity determined in this investigation, the probability per K capture of excitation of the other K electron, is equal to the number of decays per unit time which produce two K-shell vacancies, D_{KK}, divided by the K-capture decay rate, D_K. This probability, designated $P_{(2)}$, may be expressed in terms of directly measured quantitities only, as shown by the following considerations.

The counting rate of true x–x coincidence is given by

$$R_{AB}=2D_{KK}\omega^2\eta_A\eta_B\eta_C, \tag{1}$$

where ω is the K fluorescence yield,[19] η_C is the fast coincidence efficiency previously discussed, and η_A and η_B are the over-all efficiencies of the two detectors, given by

$$\eta_A=R_A/D_K\omega \quad \text{and} \quad \eta_B=R_B/D_K\omega. \tag{2}$$

Here R_A and R_B are the single x-ray counting rates with pulse amplitude requirements imposed. Rearrangement of (1), with substitutions, (2), made for the detector efficiencies, gives

$$P_{(2)}=D_{KK}/D_K=R_{AB}D_K/2R_AR_B\eta_C. \tag{3}$$

For each of the sources used for coincidence measurements, D_K was determined periodically by counting K x rays with the source at several accurately measured distances from the crystals, the areas of which had also been measured. Corrections for absorption and scattering in the foils covering the crystals and in the air were very small, and over-all uncertainties were no larger than two or three percent.

The determination of R_{AB} from the observed coincidence counting rate was by far the most difficult step

[19] The use of the normal fluorescence yield for xenon for this case has been justified in detail in the Appendix of the thesis of N. Lark. The fluorescence yields for the appropriate configurations of the excited xenon atom were estimated to be only about one percent different from the normal fluorescence yield.

in the measurement. In order to facilitate analysis of the gross coincidence data, the measurements were made with the pulse-height discriminator on one detector set to pass only the x-ray peak and the discriminator on the second detector set to pass a much wider energy band. Whenever all the coincidence requirements were met, the pulse from the second detector was registered in a 100-channel analyzer. With these same conditions, the natural background of the system was measured by counting without a source; and the accidental coincidence background was measured by separating the two detectors and counting with two sources, one at each detector. A typical spectrum, from which the natural and accidental backgrounds have been subtracted, is shown in Fig. 3. Although the x-ray peak is clearly evident, it is apparent that there are other types of coincident events.

The continuum at energies above the x-ray peak has a shape consistent with that expected for internal bremsstrahlung,[20,21] if a correction is applied for the decreasing efficiency of the thin crystals at the higher energies. The relative magnitude of this continuum is also in agreement with theory, within the rather large limit of uncertainty due to the small number of counts observed. Since the correction was small, the simple shape of the internal bremsstrahlung originally calculated by Morrison and Schiff[20] was used to extrapolate under the x-ray peak. The spectrum of internal bremsstrahlung in coincidence with K x rays has already been measured[22,23] for Cs^{131}, so this effect was not investigated further.

Several effects contributed to the rise at the low-energy end of the spectrum. One was true coincidences between L x rays and K x rays from the same atom. This contribution was removed in later runs by the use of sufficient aluminum on each side of the source to absorb the L x rays. Another source of spurious coincidences was scattering of a single K x ray in one crystal with the scattered x ray being detected in the second crystal. This deposits about 3 kev in the first crystal and 27 kev in the second. This effect was markedly reduced in later runs, but not entirely eliminated, by mounting the sources in a hole in a copper baffle which partly shielded the crystals from each other, as shown in Fig. 1. A third source of the rise at the low-energy end of the spectrum was an inherent malfunction of the coincidence circuitry. One final small correction had to be made after the resolution of the observed pulse-height spectrum was complete; this was the subtraction of events in which an x ray was observed in the display crystal, but internal bremsstrahlung or the tail of the low-energy rise fell within the channel on the other detector.

[20] P. Morrison and L. I. Schiff, Phys. Rev. **58**, 24 (1940).
[21] R. J. Glauber and P. C. Martin, Phys. Rev. **104**, 158 (1956); P. C. Martin and R. J. Glauber, Phys. Rev. **109**, 1307 (1958).
[22] A. Michalowicz, Compt. rend. **242**, 108 (1956).
[23] M. H. Biavati, S. Nassiff, and C. S. Wu, Bull. Am. Phys. Soc. 4, 278 (1959).

EXPERIMENTAL RESULTS

The first series of complete measurements was made with sources and detectors placed in only approximately reproducible positions, and the source strength was varied over quite a wide range. The quantity measured as $P_{(2)}$ varied from run to run, increasing roughly linearly with increasing source strength. This functional behavior indicated the presence of an unaccounted-for source of accidental coincidences. Supplementary experiments confirmed this explanation and led to the conclusion that the origin of these accidental coincidences was the scattering effect mentioned previously. When this scattering occurred, the fast coincidence requirements could be met and the over-all resolving time was thus increased to two microseconds, the time necessary for pulse-height discrimination. If within this time an unrelated x ray fell into the detector which received the 3 kev initially, the circuit registered a spurious coincidence event: 27 kev in one detector and 33 kev in the other. In accord with this interpretation, the quantity measured as $P_{(2)}$ for each determination was plotted versus the activity of the source. A least-squares analysis was used to extrapolate to zero source strength to obtain a true measurement of $P_{(2)}$ for the entire series. The result is $P_{(2)}=(2.9\pm0.7)\times10^{-5}$. The results of the individual determinations in this series, designated by Roman letters, are given in the first half of Table I and are shown plotted versus the source

TABLE I. Summary of the individual determinations of $P_{(2)}$.

1	2	3	4	5	6	7
Run	Duration (min)	Net x–x counts	Average singles rate (10^4 counts/min)	Resolving time (2τ) (10^{-8} sec)	Coincidence efficiency (%)	$10^5P_{(2)}$
N	441	993	8.42	1.9	91	7.74
O	398	608	7.71	2.2	90	5.74
P	555	785	7.09	2.1	89	5.69
Q	3040	2861	5.63	2.0	89	4.92
R	1227	5666	15.65	2.3	91	8.92
S	1902	1119	3.46	2.5	91	4.91
T	1045	363	2.97	2.5	90	3.24
U	360	124	2.70	2.7	90	3.68
V	1075	1480	9.33	2.6	91	4.29
W	1747	3104	5.10	2.6	91	6.39
X	1906	1711	4.89	3.3	90	5.16
ϵ	408	588	4.71	2.5	89	(2.52±0.38)
ζ	441	636	4.64	2.4	88	(2.90±0.37)
η	575	733	4.13	2.5	89	(2.97±0.31)
θ	510	654	4.02	2.5	90	(2.65±0.27)
ι	935	736	3.10	2.5	88	(2.75±0.27)
κ	951	342	1.953	3.0	89	(2.50±0.60)
λ	1384	419	1.680	2.5	88	(2.44±0.25)
μ	1260	322	1.397	2.5	81	(3.14±0.28)
ν	746	205	1.195	6.3	88	(1.85±0.43)
ξ	711	173	1.078	7.6	89	(1.99±0.50)
o	2685	382	0.945	3.2	80	(2.40±0.25)
π	3370	277	0.589	3.7	80	(2.44±0.26)
ρ	3470	161	0.345	2.4	79	(2.98±0.32)

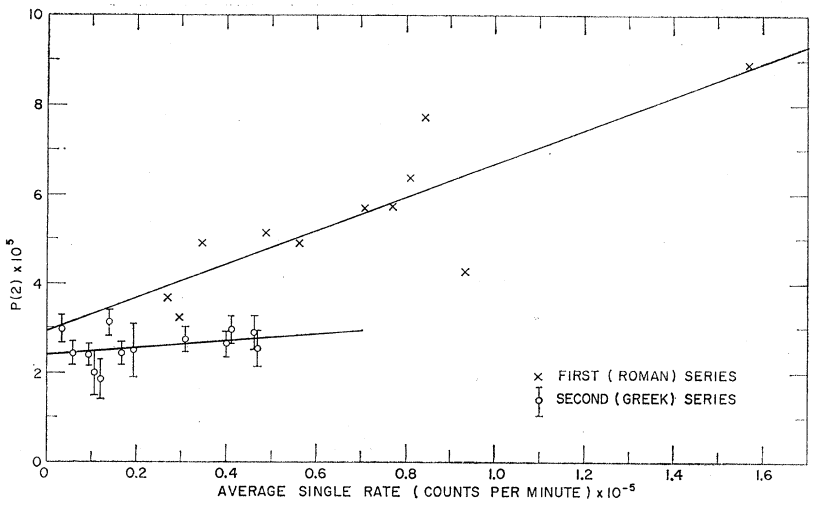

FIG. 4. Determinations of $P_{(2)}$, the probability per K capture of production of a second vacancy in the K shell. The individual measurements shown here include the effects of a spurious source of accidental coincidences which is explained in the text. The straight lines are the least-squares fits to the data, used to extrapolate to zero source strength to obtain a true determination of $P_{(2)}$.

strength in Fig. 4. Two reasons for the comparatively large deviations from run to run were the erratic drifting in the effective resolving time of the fast coincidence circuit and the variations in the relative positions of the sources and detectors.

To obtain a more reliable result, another series of determinations was made with generally weaker sources mounted as shown in Fig. 1 in order to reduce the x-ray scattering effect. For this second series the resolving time was measured both before and after each run in order to minimize effects of its drifting, and the drifting itself was reduced by additional voltage stabilization. The source and detector positions were carefully reproduced for each determination so that the results would be directly comparable. Values obtained in this series, shown designated by Greek letters, scatter relatively little from a straight line, and the slope of the line is considerably reduced from that of the earlier series, as anticipated. Extrapolation to zero source strength by least-squares analysis gives a value for $P_{(2)}$ of $(2.4\pm0.2)\times10^{-5}$. The results of the individual determinations are tabulated in Table I and shown in Fig. 4. The weighted average value of $P_{(2)}$ from the two series of runs is $(2.5\pm0.2)\times10^{-5}$.

THEORETICAL CALCULATIONS

The only published calculations concerning electron excitation phenomena accompanying K capture are those of Primakoff and Porter.[3] These calculations are essentially based on the use of the sudden perturbation approximation like the published calculations of electron excitation effects accompanying beta decay.[2,24–26] By computing the overlap between the initial and final $1s$ states for the uncaptured K electron, Primakoff and Porter obtained the probability that the electron is not excited. Subtraction of this quantity from unity gave the probability that the electron *is* excited to either bound or continuum states during the K-capture process:

$$P_{(2)}\cong\tfrac{3}{4}(1-\gamma)^2/Z^2. \quad (4)$$

Here Z is the charge on the initial nucleus and $(Z-\gamma)$ is the effective nuclear charge for the initial state of the electron. Gamma, which includes effects of both screening and electron-electron correlation, was evaluated as 0.50. This subtraction from unity depends on completeness arguments which are justifiable only for an initial two-electron atom. Primakoff and Porter estimated that for a real many-electron atom $P_{(2)}$ would be reduced by about one-third by the effects of the occupied orbitals. There is some uncertainty regarding the value of this correction, which was estimated by calculating the overlap between a final $1s$ state and all the initially occupied higher s states.

For Cs^{131} these calculations predict $P_{(2)}$ to be 6.2×10^{-5} [Eq. (4)] or 4.1×10^{-5} [Eq. (4) multiplied by two-thirds].

Calculation of the overlap between an initial $1s$ state and the continuum of final ejected states gave $P_{(ej)}$, the probability that the electron is ejected completely from the atom,[27]

$$P_{(ej)}\cong0.32(1-\gamma)^2/(Z-\gamma)^2. \quad (5)$$

This can be considered a partially independent estimate of the lower limit for the total probability of K-electron excitation accompanying K capture. Primakoff and Porter obtained also an expression for the momentum distribution of the ejected electrons.

The use of the sudden perturbation approximation for these calculations is easily justified in terms of the criterion given by Schiff.[28] For the nuclides of interest, the energy available in the transition is at least an

[24] E. L. Feinberg, J. Phys. (U.S.S.R.) 4, 423 (1941).
[25] A. Migdal, J. Phys. (U.S.S.R.) 4, 449 (1941).
[26] H. M. Schwartz, J. Chem. Phys. **21**, 45 (1953).
[27] This overlap integral has also been evaluated for the beta-decay problem (references 2, 24, and 25).
[28] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), 2nd ed.

order of magnitude larger than the K binding energy, so the uncertainty due to failure of the approximation is of the order of 10%. The unappraised adequacy of the wave functions is the primary source of uncertainty in the results of the calculations.

SUMMARY OF PREVIOUS EXPERIMENTAL RESULTS

The existence of eight published measurements of electron excitation effects accompanying K capture encourages an attempt to summarize these results and compare them with the theory. However, about half of the determinations contribute very little to the comparison because of the large uncertainty in the results. The summary is also made difficult by the fact that most of the experiments were restricted to energy ranges which included only part of the events in which K capture was accompanied by K-electron excitation. Thus these individual experiments can be compared with the theoretical predictions for the measured energy ranges, but they cannot be directly compared with each other. In order to summarize the results graphically in Fig. 5 it was necessary to use the theory to calculate the total of $P_{(2)}$ or $P_{(ej)}$ from the portion of these quantities which was actually determined.

Soltysik's beta-ray spectrometer measurement[7] of the ejected electrons from Be^7 samples gave a result about forty times larger than the magnitude predicted by the theory for the momentum range measured. The observed shape of the spectrum was also quite different from that predicted.

Porter and Hotz were able only to estimate an upper limit for $P_{(ej)}$ from their cloud-chamber measurement[4] of electrons ejected in the decay of Fe^{55}. For energies greater than approximately 30 kev, they concluded that less than 0.6×10^{-6} electron was ejected per disintegration. The theory predicts 3.6×10^{-6} electron per disintegration for this energy range. Accordingly, in Fig. 5 an experimental upper limit is shown as one-sixth the theoretical value of $P_{(ej)}$.

Charpak's attempt to determine $P_{(2)}$ for Fe^{55} by measuring coincident radiations with two proportional counters[5] gave the result $P_{(2)}=(3.8\pm1.7)\times10^{-4}$. This uncertainty estimate was based on statistical considerations alone. When systematic errors were included, the uncertainty became larger than the measured value itself.

Three measurements have been reported in which an electron-capturing nuclide was added to the gas filling a proportional counter and the pulse-height spectrum was analyzed in the vicinity of twice the K binding energy. Miskel and Perlman[6] were able to measure pulses corresponding to an energy range which included 73% of the theoretically predicted electron excitation and ejection effects in Ar^{37}. They observed $(3.9\pm0.7)\times10^{-4}$ excitation event per K-electron capture. Wolfsberg later calculated[29] that 18% of the

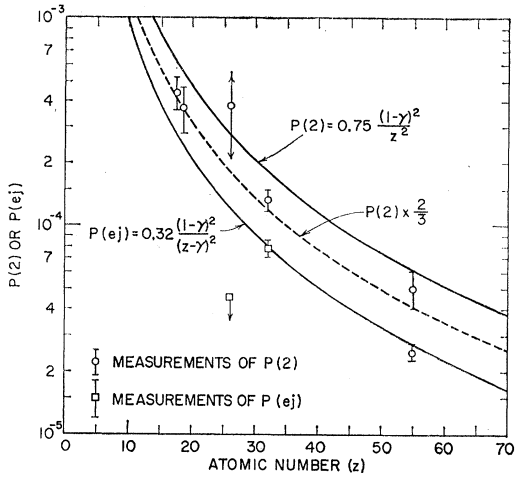

FIG. 5. A summary of the measurements which have been reported of $P_{(2)}$, the total probability per K capture of excitation of the other K electron, and $P_{(ej)}$, the probability per K capture of ejection of the second K electron from the atom. The two results for Ar^{37} have been slightly off-set horizontally for clarity. Most of the values shown were not directly determined, but were calculated from the measured results by use of the theory; the individual results can not all be directly compared with each other without recourse to theory. The uncertainty shown for each measurement is that estimated by the original authors. The solid curves are the theoretical values of $P_{(2)}$ and $P_{(ej)}$ given by Eqs. (4) and (5) of this paper [Eqs. (15) and (17) of Primakoff and Porter] with γ equal to 0.50. The dashed curve is $P_{(2)}$ multiplied by two-thirds, the correction factor estimated by Primakoff and Porter to account for effects of the occupied bound electron states.

effects observed by Miskel and Perlman were due to L-electron excitation accompanying K capture and K-electron excitation accompanying L capture. An estimate of $P_{(2)}$ from this experiment is then $(3.9\pm0.7)\times10^{-4}\times0.82/0.73=(4.4\pm0.8)\times10^{-4}$. The theoretical value of $P_{(2)}$ for argon is[30] 5.8×10^{-4}.

Kiser and Johnston reported[9] $P_{(2)}=(3.7\pm0.9)\times10^{-4}$ for Ar^{37}. They gave no details of the analysis, but this number has apparently not been corrected for the effects discussed by Wolfsberg. In the work of Kiser and Johnston, especially the thesis, we have found several numerical mistakes, misinterpretations of the calculations of Primakoff and Porter and of Wolfsberg, and misstatements regarding previous experimental work.

The data from Langevin's measurement[8] on Ge^{71} were analyzed to obtain values for both $P_{(2)}$ and $P_{(ej)}$. The experimental values, $(1.33\pm0.14)\times10^{-4}$ and $(0.78\pm0.07)\times10^{-4}$, respectively, compare favorably with the theoretical values of 1.83×10^{-4} and 0.81×10^{-4}. However, the experimental results should be reduced somewhat to account for the effects of L excitation in K capture and K excitation in L capture. Over the region where it is distinguishable, the shape of the ejected electron spectrum agrees well with the theory.

[29] M. Wolfsberg, Phys. Rev. **96**, 1712 (1954).

[30] The theoretical value of $P_{(2)}$ uncorrected for effects of occupied states is used in these comparisons because of the uncertainty regarding the magnitude of the correction.

Daniel, Schupp, and Jensen have recently reported[10] an investigation of the electron excitation effects in the decay of Cs^{131}. They found fair agreement with the theoretical $P_{(ej)}$ over a limited electron momentum range and measured $P_{(2)}$ as $(5.0\pm1.0)\times10^{-5}$, where theory predicts 6.2×10^{-5}. However, their sources were contaminated by an unidentified beta emitter.

The lack of ambiguity in the present measurement on Cs^{131} is apparent from the simplicity of Eq. (3). The probability per K capture of production of an atom with two K-shell vacancies is given by directly measured counting rates and the easily determined coincidence counting efficiency. The value of $P_{(2)}$ thus obtained is independent of any theoretical considerations. The uncertainty quoted is based on statistical analysis of many measurements, and it is consistent with careful estimates made of the uncertainties of the individual measurements. The purity of the sources was clearly demonstrated. The result, $P_{(2)}=(2.5\pm0.2)\times10^{-5}$, is lower by a factor of about two than both the previous measurement and the theoretical value.

DISCUSSION

From the results discussed here, which are summarized in Fig. 5, it can be concluded that there is better than order-of-magnitude agreement with the results of the calculations of Primakoff and Porter. However, it is essential to use caution in any more quantitative comparison. The measurements are very difficult. Traces of radioactive impurities and several other effects can cause spurious high results if they are not properly identified. The theory itself is not expected to be more accurate than roughly a factor of two. Despite this, a few further comments can be made. If the theoretical expression for $P_{(2)}$ is multiplied by two-thirds, the correction factor estimated by Primakoff and Porter to account for the effects of occupied bound states, the agreement with the experiments is generally better. The dependence of the electron excitation probabilities on the inverse square of the atomic number is roughly confirmed, but from the present measurement the probabilities would appear to be lower than predicted for heavier elements. This effect cannot be explained by relativistic effects, which would instead increase the excitation probabilities.[2] It could perhaps be explained by the increased number of occupied electronic states for the heavier elements.

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