Recent Experimental Results on Orbital Electron Capture

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A summary is presented of experimental techniques and experimental results on electron-capture ratios, comparative half-lives, and transition energies. The effect of electron capture from the M shell and higher shells is taken into account. Transition energies are computed from the observed capture ratios for about twenty cases. A summary of mean L-fluorescence yields following nuclear excitation is brought up to date, and the origins of uncertainties in the interpretation of experimental results are discussed.

Six experiments are precise enough to be compared critically with the theory of Brysk and Rose, and the observed L/K capture ratios are found to be some ten percent greater than predicted. The discrepancies seem to be more or less independent of atomic number for $18 \notin Z \notin 53$; it is not at all clear that the effect of correlations between the coordinates of the K electrons, suggested by Odiot and Daudel, is sufficient to account for the discrepancy.

ONSIDERABLE experimental data have appeared since the publication in 1955 of a review¹ which compared experimental values of L/K capture ratios with the theoretical results of Brysk and Rose,² and which summarized our knowledge of transition energies involved in electron-capture decay and of mean L-fluorescence yields. The purpose of the present paper, therefore, is to present new data along the lines established previously.

I. EXPERIMENTAL TECHNIOUES

Essentially three fundamental techniques have been applied in the determination of x-ray intensity ratios and orbital electron-capture ratios.

A. External Source Spectrometry

The radioactivity is placed outside the sensitive volume, and the relative intensities of K and L x-rays are measured. Corrections must be applied for source self-absorption, self-scattering, and in some cases selfexcitation of fluorescent x-rays (F5),‡ differential air and window absorption, and K- and L-fluorescence yields. One must also consider that a K-shell vacancy

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Uppsala, Uppsala, Sweden. ¹ B. L. Robinson and R. W. Fink, Revs. Modern Phys. 27, 424 (1955). Errata: Table I: E_{BC} of Mn^{54} should be 0.540 ± 0.010 Mev (1955). Errata: Table 1: E_{EC} of Mn⁴⁴ should be 0.540 ± 0.010 Mev (p,n), cc. This makes no change in the capture ratios. Also a new measurement has been reported for Mn⁵⁴; $E_{EC}=0.528\pm0.010$ IB(J4). Table II(b); K⁴⁰ shoul dread (4- to 2+). Reference M4: change "1168" to "1186". ² H. Brysk and M. E. Rose, Oak Ridge National Laboratory Rept. ORNL-1830 (1955) (unpublished); Revs. Modern Phys. 30, 11400 (1059).

1169 (1958).

‡ References in the text are in the form of numbered footnotes or initial-number combinations, e.g. (A2). The latter are also the references for the Tables and for Fig. 5, and are listed with the Tables.

may be filled by an L electron either by radiative transition $(K_{\alpha} = K - L_{II}, L_{III})$ or by Auger transition (K-LL, or K-LX). The x-ray intensity ratio I_L/I_K is related to the capture ratio P_L/P_K by the expression

$$I_L/I_K = [(P_L/P_K) + n_{KL})]\bar{\omega}_L/\omega_K, \qquad (1)$$

where $\omega_K = K$ -fluorescence yield; $\bar{\omega}_L = \text{mean } L$ -fluorescence yield; and n_{KL} = number of L-shell vacancies produced in the filling of a K-shell vacancy,

$$n_{KL} = k\omega_K + a_K \{ [2(K - LL) + (K - LX)] / [\Sigma \text{ Augers}] \}; \quad (2)$$

 $k = I \kappa_{\alpha} / I_{\kappa}$ = intensity ratio of K_{α} x-rays to total K x-rays; $a_K = K$ Auger yield = $(1 - \omega_K)$; and (K - LX)= partial Auger yields; X denotes M-, N-, etc. shell electrons. (K-LX) is the probability that a K-shell vacancy is filled by an L-shell electron with the excess energy carried off by an X-shell electron.

The value of n_{KL} was computed and displayed as a function of Z (Fig. 1, reference 1); the data available for this calculation have now been re-examined, and estimates have been made of the reliability of the results^{3,4} (see Sec. IE and Fig. 1). The fraction of Kx-rays in the K_{α} group, $(I_{\kappa_{\alpha}}/I_{\kappa})$, has been computed from the data in Compton and Allison⁵ and other tabulations,^{6,7} in the case of gallium a recent determination has been made by Drever and Moljk (D6), and Lazar (L15) has communicated recent measurements for some

³ L. Slack and K. Way, *Radiations from Radioactive Atoms in Frequent Use* (U. S. Atomic Energy Commission, Washington, D. C., 1959), pp. 67-69.

⁴ Wapstra, Nijgh, and van Lieshout, Nuclear Spectroscopy Tables (Interscience Publishers, Inc., New York, 1959), pp. 82-87. It may be noted that the f_{KL_i} estimated in these Tables, pp. 86-87,

It may be noted that the *j*KL_i estimated in these 1 ables, pp. 80–87, is connected with our n_{KL} by the relation $n_{KL} = \sum_i f_{KL_i}$. ⁵ A. H. Compton and S. K. Allison, *X*-Rays in Theory and *Experiment* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1935), second edition, p. 637ff. ⁶ Reference 4, pp. 80–81. ⁷ A. E. Sandström, Handbuch der Physik (Springer-Verlag, Berlin-Göttingen-Heidelburg, 1957), Vol. XXX, p. 236.



FIG. 1. n_{KL} is the number of L-shell vacancies produced in the filling of a K-shell vacancy.

elements between terbium and lead (see Sec. V). *K*-fluorescence yields are given by Broyles, Thomas, and Haynes,⁸ Gray (G6), Laberrigue-Frolow and Radvanyi,⁹ Roos,¹⁰ and others. A recent summary of mean *L*-fluorescence yields has been given by Fink (F5) which is brought up to date in the present review. Trends in partial *L*-fluorescence yields and in Coster-Krönig yields are shown by Wapstra, Nijgh, and Van Lieshout.⁴

In some cases, all electron-capture transitions proceed via one excited state of the daughter. Then a measurement of the relative intensities or specific activities of the K x-ray and gamma ray will yield directly the fraction of captures from the K shell, P_K (H4, T3).

B. Internal Source Spectrometry

The radioactive material is dispersed throughout the sensitive volume of the detector. An advantage of this method is that a knowledge of the *L*-fluorescence yield is not required because the *L* radiations are (usually) totally absorbed in the counter.

The intensity ratio N_L/N_K of the L and K peaks may be measured directly in a gas proportional counter. The K peak arises from K Auger electrons and from K x-rays that are absorbed in the filling gas. The L peak comprises L Auger electrons and L x-rays following L capture as well as L radiation emitted after K capture, simultaneously with K_{α} x-rays that escape undetected from the counter. Neglecting L x-ray escape which is usually very small, the experimental capture ratio is given by (D6)

$$P_L/P_K = (N_L/N_K)(1 - P\omega_K) - kP\omega_K, \qquad (3)$$

where P is the probability that a K x-ray can escape from the counter without being detected, and $k = I \kappa_{\alpha}/I_K$ is the intensity ratio of K_{α} x-rays to total K x-rays, as in the foregoing.

In order to measure P_L/P_K experimentally, one chooses the dimensions of the counter and the type of filling gas so that the K x-ray escape probability P approaches either unity or zero, the former being experimentally simpler. However, if one chooses $P \rightarrow 1$, and uses a small low-pressure counter with a gas of low Z, such as methane or propane (L11, L12), the result depends critically upon ω_K . For example, for Ge⁷¹ Langevin¹¹ obtained $P_L/P_K=0.30$ using $\omega_K=0.45$; but if ω_K is taken to be about 0.50, this experiment gives $P_L/P_K=0.19$.

The alternative method, in which $P \rightarrow 0$, gives results independent of ω_K as shown by Eq. (3), and all radiations resulting from a K-capture or an L-capture event are integrated by the counter into the corresponding K or L peak, giving P_L/P_K directly, with an extremely small correction for K escape. The $P \rightarrow 0$ technique has been pioneered by Drever, Molik, and Curran (D6, D7) who used an anticoincidence arrangement of two concentric proportional counters without an intervening wall ("wall-less" counter). An inner circle of wires acts as cathode for the central cylindrical proportional counter, while alternate anode and cathode wires in the outer annular region turn the surrounding layer of gas into a separate annular counter. With six atmospheres of argon-methane mixture containing radioactive Ge⁷¹H₄ the probability that a 10.2-kev K x-ray escapes from the central counter and is not detected in the annular counter is less than 10⁻⁴. The counting rate in the annular counter is some 15 times that in the central counter, which is 4.5 cm in diameter and has a sensitive length of 76 cm.

In another approach, scintillation crystals (M4) have been grown containing radioactive Cd¹⁰⁹ and I¹²⁵ (M5). The contribution of $L+M+\cdots$ capture was deduced from the difference between the number of K x-rays and the total number of gamma-ray transitions, under the assumption that one such transition, either photon or conversion electron, accompanies each decay. More recently, I¹²⁶ has been produced by the reaction I¹²⁷(γ ,n)I¹²⁶ in a crystal of NaI(Tl) introduced into the x-ray beam of a synchrotron (S12). In this case the K and L x-radiations were observed when the crystal was used as a scintillation spectrometer.

C. Coincidence Spectrometry

If electron-capture proceeds to an excited state of the daughter nucleus which in turn decays promptly by gamma-ray emission, then the branching probability for K capture P_K can be determined by a coincidence experiment. Let R_{γ} and $R\kappa_{\gamma}$ be the counting rates for gamma rays and x-ray—gamma-ray coincidences, re-

¹¹ M. Langevin, Compt. rend. 239, 1625 (1954).

⁸ Broyles, Thomas, and Haynes, Phys. Rev. 89, 715 (1953). ⁹ J. Laberrigue-Frolow and P. Radvanyi, J. phys. et radium 17, 944 (1956).

¹⁰ C. E. Roos, Phys. Rev. 105, 931 (1957).

(4)

spectively;

$$R_{\gamma} = D\epsilon_{\gamma}S_{\gamma}, \quad R\kappa_{\gamma} = D\epsilon_{\gamma}S_{\gamma}P_{K}\omega_{K}\epsilon_{K}S_{K},$$

and

$$R_{K\gamma}/R_{\gamma}=P_{K}\omega_{K}\epsilon_{K}S_{K},$$

where D is the disintegration rate, S and ϵ are solid angles and intrinsic detection efficiencies of the counters, and $\omega_{\mathcal{K}}$ is the K-fluorescence yield. The presence of internal conversion and other branches may complicate the analysis, but the result remains essentially unchanged (K4, B25). This technique is particularly useful for relatively complicated decay schemes, provided that there is a high-energy gamma-ray transition directly to the ground state or to a low-lying state. The most highly excited state will be populated by the electron-capture transition of the lowest energy, and this in turn is the most favorable circumstance for inferring the capture energy from the capture ratio. "Crystal summing" experiments as well as ordinary coincidence experiments can be analyzed in this way (G7, G9).

D. Capture from Higher Shells

One must distinguish between the experiments in which the L/K capture ratio is determined by observation of the L and K radiations, and those in which the ratio of K capture to total capture is measured, as in the coincidence experiments mentioned in (IC). In the latter cases one may readily deduce the ratio $(P_L + P_M + \cdots)/P_K \equiv P_{LM} \dots / P_K$, and this value is frequently reported. Distinction will be made between these ratios in both the experimental and theoretical results reported in the following.

E. Uncertainties in the Interpretation of **Experimental Results**

The interpretation of the experiments on electroncapture requires knowledge of several auxiliary quantities from the domain of x-ray physics, and uncertainties in the latter are propagated into the results of the nuclear physics experiments.

In many experiments in which K x-rays are counted, a knowledge of the K-fluorescence yield is required. The best measurements to date are those of Roos,¹⁰ which delineate the steeply rising portion of the ω_K vs Z curve for $26 \leq Z \leq 50$, and which are stated to some two or three percent. It would be of greatest value to know the K-fluorescence yields with a precision of one percent or better. Exact measurements and good agreement between the results of different experiments are especially lacking for 10 $\gtrsim Z \approx 25$, a region which contains some interesting cases, notably K^{40} .

In experiments in which both K and L x-rays are counted, one must take into account the L-shell vacancies produced in the filling of K-shell vacancies, a quantity called n_{KL} above. Re-examination of the data used in calculating n_{KL} leads to the assignment of \cdot uncertainties as shown in Fig. 1. For Z > 30, the chief source of uncertainty lies in the K-fluorescence yield; if ω_K were known to one percent, then the uncertainty in n_{KL} would be halved. If in addition the ratio of the intensities of the K_{α} and K_{β} x-ray groups (which radiatively transfer the K-shell vacancies to the L shell and to higher shells, respectively) were known to one percent, then the uncertainty in n_{KL} would be halved again for Z > 50; for 30 > Z > 50, improved Auger yield measurements are also required. For Z > 30, we need more precise knowledge of the total Auger yield (or, equivalently, the fluorescence yield, since $\omega_K + a_K \equiv 1$), and of the relative intensities of the K-LL, K-LX, and K - XY groups of Auger electrons.

II. CALCULATED CAPTURE RATIOS

Table I is essentially a supplement to Table I of reference 1. It is slightly different in that (a) L_{I} and L_{II} capture are taken together, (b) capture from higher shells is computed, and (c) $\log f_n t$ is given for *n*th forbidden transitions with maximum spin change (the so-called unique transitions). The only nuclides repeated in Table I are Np²³⁵ and K⁴⁰, which are discussed in Sec. IV.

A. Capture from Higher Shells

The contribution of electron-capture from shells of principal quantum number greater than two is not negligible, and may be quite important in transitions of relatively low energy and in forbidden transitions.

As a zeroth approximation,² one might examine the nonrelativistic wave functions for a hydrogenlike atom.¹² The s-electron wave functions are proportional to $(Z/n)^{\frac{3}{2}}$, and thus the transition probabilities are proportional to n^{-3} . (This gives 0.125 for the 2s/1s ratio, a value which may be compared with the results of Brysk and Rose, which run from 0.08 for Z = 15 to 0.16 for Z=86.) For p electrons the capture from shells with n=2, 3, 4, and 5 is found to be in the proportion 1:0.352:0.156:0.082

As a somewhat refined approximation, Wapstra and Van der Eijk (W4) have used Slater screening constants in hydrogenlike wave functions. Alternatively, Ketelle, Thomas, and Brosi (K4) determined relative densities of M-, N-, etc., shell electrons from the results of the self-consistent field (SCF) calculations of Hartree and others.¹³ Following the latter, we have calculated the total contribution to s-electron capture and p-electron

¹² See for instance L. Pauling and E. B. Wilson, Jr., Introduction to Quantum Mechanics (McGraw-Hill Book Company, Inc., New York, 1935), pp. 135–139.
¹⁸ D. R. Hartree, Proc. Roy. Soc. (London) A141, 282 (1933), (Cl⁻, Cu⁺); D. R. Hartree, Phys. Rev. 46, 737 (1934), (Hg⁺); D. R. Hartree, Proc. Roy. Soc. (London) A143, 500 (1934), (Cs⁺, K⁺); D. R. Hartree and W. Hartree, Proc. Roy. Soc. (London) A143, 500 (1934), (Cs⁺, K⁺); D. R. Hartree and W. Hartree, Proc. Roy. Soc. (London) A149, 210 (1935), (Ca, Hg); Douglas, Hartree, and Runciman, Proc. Cambridge Phil. Soc. 51, 486 (1955), (Au⁺, Tl⁺); W. G. Henry, Proc. Phys. Soc. (London) A67, 789 (1954), (Au⁺); E. C. Ridley, Proc. Cambridge Phil. Soc. 51, 702 (1955), (Mo⁺). (Mo+́).

$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Z A (Class	Half-life	E_{BC} (Mev) and method	References ^a	Initial state	Final state (Mev)	% EC	Log fut	$\operatorname{Log} f_n t$	$Th (L_1+L_{r1})/R$	eoretical capture ra Lun/K I/K	atios 7 M / V	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	19 K 40	В	$1.3 \times 10^9 \mathrm{y}$	$ \begin{cases} 0.043 \pm 0.008 \ MS, cc \\ 0.060 \pm 0.008 \ (p,n)cc \end{cases} $	$\left. \begin{array}{c} G13\\ H15 \end{array} \right\}$	4-	2+ 1.46	11%					TT /	See discussion
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	27 Co 57	в	270 d	$ \begin{cases} 0.434 \pm 0.030 \ IB \\ 0.755 \pm 0.030 \ (p,n)cc \end{cases} $	$\left. {{ m J4}\over { m E2}} \right\}$	7/2-	5/2 - 0.136	$\sim 100\%$	6.0 6.4		0.093		0.107	01 200, 14
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	33 As 74	A	18 d	$2.55\pm0.02\ eta^+$ $1.94\pm0.02\ eta^+$	J6	2-	0+ ground 2+ 0.596	1 2% 38%	8.4 6.8	8.7	0.095	0.001 0.096	0.113	
	37 Rb 84	A	33 d	$2.65\pm0.01 \ eta^+$ $1.77\pm0.02 \ eta^+, cc$ $0.75\pm0.02 \ cc$	W6	2-	$\begin{array}{c} 0+ \\ 2+ \\ 2+ \\ 0.88 \\ 2+ \\ 1.90 \end{array}$	1 9% 66% 1.4%	8.1 7.0 7.9	8.6	0.101 0.102 0.103	0.001 0.102	0.111	
	38 Sr 85	в	64 d	~ 0.6 (est)	W7	9/2+	9/2+ 0.513	100%	6.2		0.108		0.130	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	43 Tc 96	в	4.2 d	~0.6 (est)	W8		2.42, 2	.73	~ 5.3		0.115		0.130	
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	51 Sb 119	A	38 hr	$0.555 \pm 0.020 \ IB$	02	5/2+	3/2 + 0.024	${\sim}100\%$	5.0		0.126		0.155	
	53_I 126	V I	13 d	$2.13\pm0.02 \beta^+$ $1.48\pm0.03 cc$ $0.73\pm0.04 cc$	K9	2-	$\begin{array}{c} 0+ & {\rm groun}\\ 2+ & 0.65\\ 2+ & 1.40 \end{array}$	d 24% 27% 4%	7.8 7.5 7.7	8.0	0.125 0.123 0.128	0.004 0.129	0.158 0.151 0.157	
$ \begin{array}{llllllllllllllllllllllllllllllllllll$	62 Sm 145	A	340 d	0.585 ± 0.015 IB 0.160 ± 0.015 cc	B25	7/2-	7/2 + 0.061 3/2 + 0.485	92% 0.0027%	7.8 11.3	8.8	$ \{ \begin{matrix} 0.151 \\ 0.41 \\ 0.23 \end{matrix} \}$	0.34 0.75	0.189 0.99 0.29	Forbidden Allowed
84 Po 209 B 103 y 0.99±0.03 cc F6, S14 1/2 - 7/2 - 0.91 $\sim 5\%$ 13.2 12.1 $\begin{pmatrix} 0.274 \\ 0.200 \\ 0.26 \\ 0.26 \\ 0.26 \\ 0.26 \\ 0.31 \\ $	83 Bi 208g	C	$3 \times 10^4 \mathrm{y}$	0.31 ± 0.08 (est)	F6, S14	4,5+	3- 2.165	100%	12.4	(10.4)	$\left\{ \begin{matrix} 0.545 \\ 0.302 \end{matrix} ight\}$	0.163 0.71	0.92 0.39	Forbidden Allowed
85 At 211 B 7.2 hr $0.75 < E_{E_{C}} < 0.82 \ cc$ F6, S14 9/2 ground 59% 6.0 0.215±0.002 0.31 93 Np 235 A 410 d 0.13±0.02 cc F6, S14 1/2+ 1/2+ ground ~100% ~7 11.3 ⁺ no limit 16.3 ⁺ no limit 16.3 ⁺ -12.5 94 Pu 237 A 46 d 0.21±0.02 cc F6, S14 7/2- 5/2+ ground 60% 6.3 0.86 ⁺ 0.30 1.19 ⁺ 0.38 0.18±0.02 cc 7/2+ 0.033 22% 6.6 1.4 ^{+1.0} 1.4 ^{+1.0} 1.90 ^{+1.4} 0.15±0.02 cc 5/2 0.060 15% 6.4 3.0 ^{+2.0.4} 1.4 ^{+1.0} 1.90 ^{+1.4} 0.15±0.02 cc F6, S14 7/2- 6.6 1.4 ^{+1.0} 1.4 ^{+1.0} 1.90 ^{+1.4} 0.15±0.02 cc 7.2 0.060 15% 6.4 3.0 ^{+2.0.4} 0.42 ^{+2.13} 97 Bk 245 A 4.98 d 0.59±0.02 cc F6, S14 0.65 4.70 0.32±0.01 0.42±0.01 1.5 ^{-0.15} 0.42 ^{+0.44} 0.51 ^{+0.44} 0.55 ^{+0.36} 1.5 ^{+0.36} 0.53 4.2 ^{+0.44} 0.51 ^{+0.44} 0.55 ^{+0.44} 0.50 ^{+0.44} 0.51 ^{+0.44} 0.51 ^{+0.44} 0.55	84 Po 209	В	103 y	0.99±0.03 66	F6, S14	1/2 -	7/2 - 0.91	~5%	13.2	12.1	$\{0.274\ 0.200\$	0.024 0.298	0.39	Forbidden
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	85 At 211	в	7.2 hr	$0.75 < E_{BC} < 0.82 \ cc$	F6, S14	9/2	groune	1 59%	6.0		0.215 ± 0.002		0.31	no Motter
94 Pu 237 A 46 d $0.21\pm0.02 \ cc$ F6, S14 7/2- 5/2 + ground 60% 6.3 $0.86+0.30$ 1.19+0.38 1.19 -0.21 0.18\pm0.02 \ cc 7/2+ 0.033 22% 6.6 $1.4+1.0$ 1.90+1.4 1.90+1.4 0.15\pm0.02 \ cc 5/2 0.060 15% 6.4 $3.0+2.0$ 1.90+1.4 1.90+1.4 0.52 2.2 0.060 15% 6.4 $3.0+2.0$ 1.90+1.4 1.0 -0.6 1.8 245 A 4.98 d $0.59\pm0.02 \ cc$ F6, S14 0.25 4% 7.0 $0.32\pm0.01 \ 0.42\pm0.01 \ 0.42\pm0.01 \ 1.52+0.35 \ 1.52+$	93 Np 235	A I	410 d	0.13±0.02 cc	F6, S14	1/2+	1/2+ ground	i ~100%	~		11.3 ± 10 limit -8.5		$16.3 \pm $ no limit 16.3 ± 12.5	
$\begin{array}{ccccccc} 0.18\pm0.02\ \mathrm{cc} & 7/2+0.033 & 22\% & 6.6 & 1.4+1.0 & 1.90+1.4 \\ 0.15\pm0.02\ \mathrm{cc} & 5/2 & 0.060 & 15\% & 6.4 & 1.4+1.0 & 1.90+1.4 \\ 0.15\pm0.02\ \mathrm{cc} & 5/2 & 0.060 & 15\% & 6.4 & 3.0+2.0 & 4.2+2.3 \\ 97\ \mathrm{Bk}\ 245\ \mathrm{A}\ 4.98\ \mathrm{d}\ 0.21\pm0.02\ \mathrm{cc}\ \mathrm{F6}, \mathrm{S14} & 0.23 & 4\% & 7.0 & 0.32\pm0.01 & 0.42\pm0.01 \\ 0.23 & 6\% & 7.0 & 0.32\pm0.01 & 0.42\pm0.01 & 0.44\pm0.01 & 0.44\pm0$	94 Pu 237	V	46 d	0.21±0.02 cc	F6, S14	7/2-	5/2+ ground	ф 09 р	6.3		0.86+0.30		1 10+0.38	
$\begin{array}{ccccccc} 0.15\pm0.02\ cc & 5/2 & 0.060 & 15\% & 6.4 & \frac{1.7}{3.0}+2.0 & 0.6 \\ 97\ \mathrm{Bk}\ 245 & \mathrm{A} & 4.98\ \mathrm{d} & 0.59\pm0.02\ cc & \mathrm{F6},\mathrm{S14} & 0.25 & 4\% & 7.0 & 0.32\pm0.01 \\ 0.21\pm0.02\ cc & \mathrm{F6},\mathrm{S14} & 0.63 & 6\% & 7.0 & 0.32\pm0.01 \\ 0.25 & 6\% & 7.0 & 1.15\pm0.35 & 1.52\pm0.01 \\ 1.15\pm0.35 & 1.52\pm0.01 \\ 0.25 & 0.63 & 6\% & 7.0 & 0.15\pm0.02 \\ 0.25 & 0.63 & 6\% & 7.0 & 0.15\pm0.01 \\ 0.25 & 0.63 & 0.63 & 0.63 & 0.63 & 0.63 \\ 0.25 & 0.63 & 0.63 & 0.63 & 0.63 & 0.63 \\ 0.25 & 0.63 & 0.63 & 0.63 & 0.63 & 0.63 \\ 0.25 & 0.63 & 0.63 & 0.63 & 0.63 & 0.63 \\ 0.25 & 0.63 & 0.63 & 0.63 & 0.63 & 0.63 \\ 0.25 & 0.63 & 0.63 & 0.63 & 0.63 & 0.63 \\ 0.25 & 0.63 & 0.63 & 0.63 & 0.63 & 0.63 \\ 0.25 & 0.25 & 0.64 & 0.64 \\ 0.25 & 0.64 & 0.64 & 0.64 \\ 0.25 & 0.64 & 0.64 & 0.64 \\ 0.25 & 0.64 & 0.64 & 0.64 & 0.64 & 0.64 \\ 0.25 & 0.64 & 0.64 & 0.64 & 0.64 & 0.64 \\ 0.25 & 0.64 & 0.64 & 0.64 & 0.64 & 0.64 \\ 0.25 & 0.64 & 0.64 & 0.64 & 0.64 & 0.64 \\ 0.25 & 0.64 & 0.64 & 0.64 & 0.64 & 0.64 & 0.64 \\ 0.25 & 0.64 & 0.64 & 0.64 & 0.64 & 0.64 & 0.64 & 0.64 \\ 0.25 & 0.64 & 0.$				0.18±0.02 cc			7/2 + 0.033	22%	6.6		$^{-0.15}_{14}$ + 1.0		$\frac{1.19}{100} - 0.21$	
97 Bk 245 A 4.98 d 0.59 ± 0.02 c F6, S14 0.25 4% 7.0 0.32 ± 0.01 0.42 ± 0.01 0.42 ± 0.01 0.42 ± 0.01 0.21 ± 0.02 c 1.52 ± 0.02 0.63 6% 7.0 1.15 ± 0.35 1.52 ± 0.46 1.52 ± 0.46				0.15±0.02 cc			5/2 0.060	15%	6.4		$3.0^{+20}_{-1.2}$		4.2 + 23 4.2 + 23 -1.8	
	97 Bk 245	A	4.98 d	0.59±0.02 cc 0.21±0.02 cc	F6, S14		0.25 0.63	4% 6%	7.0 7.0		$\begin{array}{c} 0.32 \pm 0.01 \\ 1.15 + 0.35 \\ 1.15 - 0.26 \end{array}$		$\begin{array}{c} 0.42 \pm 0.01 \\ 1.52 \pm 0.46 \\ 1.52 - 0.35 \end{array}$	

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See list at end of article.

capture from shells of $n \ge 2$. In Fig. 2 the results are given as the ratio of $(M+N+\cdots)/L$, neglecting the dependence on neutrino energy.

In the process of obtaining these results we had the opportunity to compare the density ratios of 2s and 1s electrons calculated by Brysk and Rose² with those obtained from the work of Hartree and his collaborators, whose nonrelativistic SCF results do not even approach the hydrogenlike value of 0.125, but range from 0.08 for Z=20 to 0.11 for Z=80. However, the value obtained from Mayers' relativistic SCF calculations for mercury¹⁴ is 0.1485, in good agreement with the value of 0.150 which one reads from the graphical presentation of Brysk and Rose. On the other hand, there is good agreement between the relativistic and nonrelativistic SCF results for mercury for the ratios 3s/2s, 4s/2s, 3p/2p, and 4p/2p; and one is therefore led to have reasonable confidence in the validity of the results given in Fig. 2.

B. Comparative Half-Life

As before, the logarithms of the comparative halflives $(\log ft)$ have been obtained from the nomograms of Moszkowski¹⁵ or by the method given by Major and Biedenharn¹⁶ in the case of low-energy transitions.

Davidson¹⁷ has calculated and correlated $\log f_n t$ values for the *n*th forbidden unique beta decays. By analogy we have calculated $\log f_n t$ values for electroncapture transitions from the relations given by Major and Biedenharn¹⁶ and by Brysk and Rose²:

 $ft = f_0 t = (\pi/2) g_K^2 q_K^2 [1 + (P_{LM}.../P_K)]t,$

 $f_1 t = (\pi/24) g_K^2 q_K^4 [1 + (P_{LM}.../P_K)] t$

and

$$f_2 t = (\pi/2160) g_K^2 q_K^6 [1 + (P_{LM}.../P_K)] t. \quad (5)$$

For completeness we give here the $\log f_1 t$ values for the first forbidden unique transitions listed in reference 1: Ca⁴¹, $\log f_1 t = 9.4$; Tl²⁰⁴, $\log f_1 t = 8.4$. These two transitions, the ground state transitions of As⁷⁴, Rb⁸⁴, and I¹²⁶, and the weak inner transition in Sm¹⁴⁵ all have $\log f_1 t$ values that are within the range of the $\log f_1 t$ values for this class of beta emitters tabulated by Feenberg,¹⁸ although Ca⁴¹ is at the upper limit.

The $\log f_2 t$ value for the second forbidden unique transition of Po²⁰⁹ is also near the values calculated by Davidson¹⁷ for Be¹⁰ and Na²². On the basis of comparative half-life, one is tempted to classify the electroncapture of Bi^{208g} as second forbidden ($\Delta I = 2$, no), but neither the half-life nor the transition energy is well



FIG. 2. Ratio of electron densities for $M+N+\cdots$ shells and L shell at the nuclear radius. Self-consistent field results for s electrons and p electrons.

enough known to permit any degree of assurance in this assignment.

C. Forbidden Transitions

Previously we discussed briefly the forbidden electron-capture transition of Ni⁵⁹ ($\Delta I = 2$, no; log ft = 11.9). For forbidden transitions with $\Delta I \ge 2$, the multiplicity of matrix elements precludes making precise connection between the energy of the transition and the capture ratios. Nevertheless some conclusions can be drawn from an examination of the expressions given by Brysk and Rose² in which g and f, the large and small parts of the Dirac wave function, are mixed.

(i) There is no effect on the $L_{\rm I}/K$ ratio for two reasons. In the first place, $(f_{LI}/f_K)^2 = (g_{LI}/g_K)^2$. Secondly, the interference terms, which may be negative, are proportional to the neutrino energy q; but when q becomes large the energy dependence of the capture ratio virtually vanishes for all transitions.

(ii) The $L_{\rm III}/L_{\rm I}$ ratio also depends on the matrix elements, and precise determinations of both the capture energy and the $L_{\rm III}/L_{\rm I}$ ratio might serve to identify the matrix elements (or their ratio) in these transitions. This is equivalent to the shape fitting of beta-ray spectra of these classes of forbidden transitions. The cases in point are Ni⁵⁹, Tc^{97g}, and perhaps Bi^{208g}.

III. COMPARISON OF THEORY AND EXPERIMENT

In Table II we have compared experimentally determined capture ratios with those calculated from known transition energies (transcribed from Table I or from Table I of reference 1) for 15 cases. Table II corresponds to Table II(a) of reference 1 and includes the cases mentioned therein with the exception of Pd103; the latest experimental results on Pd¹⁰³ make the situation appear inconclusive at this time.¹⁹

¹⁴ D. F. Mayers, Proc. Roy. Soc. (London) A241, 93 (1957),

Hg⁺, relativistic).
 ¹⁶ S. A. Moszkowski, Phys. Rev. 82, 35 (1951); reproduced in part in reference (S14), pp. 597-600.
 ¹⁶ J. K. Major and L. C. Biedenharn, Revs. Modern Phys. 26, 221 (1957).

^{321 (1954).}

 ¹⁷ Jack P. Davidson, Jr., Phys. Rev. 82, 48 (1951).
 ¹⁸ Eugene Feenberg, *Shell Theory of the Nucleus* (Princeton University Press, Princeton, 1955), Chap. V.

It may be said that there appears to be gross agree-

¹⁹ Avignon, Michalowicz, and Bouchez, J. phys. et radium 16, 404 (1955).

ZA	Ex; Methodª	perimental n References	nethod and results P_L/P_K	Р _{LM} /Рк	Theoretical pr P_L/P_K	edictions PLM/I	2 <i>K</i> Remarks
18 A 37	B(P small)	{L11 K10	$\left.\begin{smallmatrix} 0.092 + 0.010 \\ 0.092 - 0.005 \\ \pm 0.008 \end{smallmatrix}\right\}$		0.082		h
19 K 40	Ab	$\mathbf{H4}$					See Sec. IV for discussion
26 Fe 55	$\mathbf{B}\left(P\rightarrow0\right)$	S13	0.108 ± 0.006		0.097		h · ·
27 Co 57	C	M12		0.20 ± 0.13		0.107	
32 Ge 71	$B(P \rightarrow 0)$	D6, D7 D9	$0.128 \substack{+0.005 \\ -0.003 \\ 0.116 \pm 0.005}$		0.106		^h This result is considered to supercede that given in reference D6
33 As 74	$\mathbf{B}(P\to 0)$	S9	0.085 + 0.020		0.095		
36 Kr 79	$\begin{array}{l} \mathbf{B}\left(P \text{ not small}\right)\\ \mathbf{B}\left(P \rightarrow 0\right) \end{array}$	L12 D9	$0.16 {\pm} 0.03 \\ 0.108 {\pm} 0.005$		0.101		i h
37 Rb 84	C ^d	W6		$0.12{\pm}0.05$		0.121	Capture to 0.89-Mev level
38 Sr 85	Cď	B26		0.14±0.05		0.130	Capture to 513-kev level; reported as $P_{\kappa} = (88 \pm 4)\%$; theoretical value is 89%
53 I 126	${\rm B}(P \to 0)^{\rm e}$	S12	$0.142 \substack{+0.005 \\ -0.018}$		0.123		b
62 Sm 145	C ^f C ^d	B25		$0.20 \pm 0.02 \\ 0.6 \ \pm 0.1$		0.189 0.99 0.29	Capture to 61-kev level Forbidden transition to 485-kev level Allowed transition to 485-kev level
85 At 211	Α	H8	0.143		$0.215 {\pm} 0.002$		See also (G6)
93 Np 235	A	$\begin{cases} H10\\ G8 \end{cases} ;$	30 ± 2 36.7 ± 4		$11.3^{+no limit}_{-8.5}$		j
94 Pu 237	Ae	H14	2.8±0.8		$^{+20}_{3.0-1.2}$		Capture to 59.6-kev level; see Table III
97 Bk 245	Cď	M13 ~	~0.33		0.32 ± 0.01		Capture to 0.25-Mev level; see Table IV for L capture to 0.63-Mev level

TABLE II. Comparison of theory and experiment.

* A, B, and C refer to external source, internal source, and concruence operations the counter in internal source spectrometry. • Specific activity for K x-rays and gamma rays. • Beta-ray spectrometry of Auger and internal conversion electrons. • K and L radiation coincidences. • K and L radiation coincident with gamma-ray. • K and L radiation coincidentes. • K and L radiation coincidences. • L A content of the content of th A, B, and C refer to external source, internal source, and coincidence spectrometry, respectively; P is the probability that a K x-ray escapes

ment between theory and experiment. The apparent discrepancies in the cases of K⁴⁰, At²¹¹, and the inner branch of Sm¹⁴⁵ can in all likelihood be attributed to insufficiently precise data; and the result of Langevin for Kr⁷⁹ (L12) depends critically upon the K-fluorescence yield. However, the precision measurements for A³⁷, Fe⁵⁵, Ge⁷¹, Kr⁷⁹, and I¹²⁶ are not in accord with the predictions of the theory of Brysk and Rose.² The six experimental results which are precise enough for critical comparison with the predictions of Brysk and Rose² are summarized in Fig. 3. The correction proposed by Odiot and Daudel²⁰ for A³⁷ is also indicated in Fig. 3.

There appears to be a small but consistent discrepancy of some ten percent between these precision experimental measurements and the theoretical results of Brysk and Rose. Brysk has informed us that the latter were not intended to be more precise than a few percent.21

A. Correlations

Odiot and Daudel²⁰ have treated the problem of electron capture for small Z, taking into account the correlations that exist between the positions of the electrons. These arise from the facts that (i) the wave functions are not the same before and after capture, since the nuclear charge has changed by one unit, (ii) the wave functions must be antisymmetric in the coordinates of the electrons (Pauli exclusion principle), and (iii) the mutual repulsion of the K electrons may not be completely negligible compared to the attraction of

²⁰ S. Odiot and R. Daudel, J. phys. et radium 17, 60 (1956).

²¹ H. Brysk (private communication 1959).

the nucleus. (The latter is in a fashion taken into account by Brysk and Rose² in their use of screened wave functions.) Using nonrelativistic wave functions, Odiot and Daudel found that these considerations operate to suppress the capture of K electrons relative to L electrons by some 25% for Z=18.

The effect increases rapidly as Z decreases. Because reasonably exact wave functions are available, Odiot and Daudel made complete calculations for Z=2, although they are only of theoretical and academic interest since there are no L electrons in the ground state of helium, to say nothing of the fact that helium has no electron-capturing isotopes. At any rate, the capture ratio, P_L/P_K , is found to be increased tenfold for helium, while less exact calculations indicate that the increase is about threefold for Z=4. Unfortunately 4Be⁷ seems to be quite inaccessible to experimental verification; however, some of the other low-Z electroncapturers ($_9F^{18}$, $_{11}Na^{22}$, $_{13}Al^{26}$, and $_{17}Cl^{36}$) would be worth study, although the experiments would be exceedingly difficult.

On the other hand, the effect falls off rapidly as Z increases, and although it was plainly insufficient to account for the large discrepancy previously ascribed¹¹ to $_{32}$ Ge⁷¹, it may perhaps account for the small discrepancies found in the recent precision experiments.

B. Discussion of Cases in Table II

Argon-37.—The effect of correlations has been calculated only for this case,²⁰ and it appears to increase the theoretical value of P_L/P_K from 0.080 to 0.100. The experimental values lie between these two numbers, but might be said to be in slightly better agreement with the latter than with the former.

Krypton-79.—The experiments of Langevin (L12) were performed under circumstances which make the results strongly dependent on the correction for the escape of K x-rays from the sensitive volume of the counter, and upon the value chosen for the K-fluorescence yield of bromine, as indicated in footnote (e) of Table II. Using the "wall-less" counter, Drever (D7, D9) has found a capture ratio for Kr⁷⁹ which is in gross agreement with the theory of Brysk and Rose.

Astatine-211.—The experimental results on At^{211} are reported only as the ratio of the K and L x-ray intensities: $I_K/I_L=3.1$. This appears to be the result of absolute measurements of the K and L x-ray intensities in scintillation and proportional counter spectrometers, respectively; but there is little description of the experimental work which one might base an estimate of the precision of the intensity ratio quoted.

IV. TRANSITION ENERGIES DEDUCED FROM EXPERIMENTALLY DETERMINED CAPTURE RATIOS

Despite this rather uncertain situation as regards the detailed reliability and validity of the theory of electron capture, we proceed to Table III, in which are listed



theoretical L/K capture ratio of Brysk and Rose

The open square at Z=18 represents the correlation effect computed by Odiot and Daudel (reference 20). The uncertainty in Rindicated by the length of the vertical bars does not include about 4% uncertainty in the theoretical results of Brysk and Rose.

some capture energies which were calculated from experimentally determined capture ratios. We feel some confidence in these calculations because they lie in the relatively high-Z region for which we might expect any discrepancy to be small, and because most of the transition energies are found to be quite small and therefore are not very sensitive to small changes which may have to be made in the theory of electron capture. The case of K^{40} discussed in the following is a somewhat extreme example.

Table III supercedes Table II(b) and Table IV of reference 1. The results for Cd¹⁰⁹, I¹²⁵, and W¹⁸¹ are merely repeated; there are new experimental results for Ba¹³³, Os¹⁸⁵, Au¹⁹⁵, and Tl²⁰², and twelve other radio-activities; K⁴⁰ is also included, although the experimental results for this case are somewhat contradictory as will be discussed in the following in detail.

We have also calculated $\log ft$ values for the transitions in Table III. With the exception of K⁴⁰, Tc^{97g}, La¹³⁸, and Er¹⁶⁵, the comparative half-lives seems to correspond to allowed or first forbidden transitions, that is, $|\Delta I| = 0$ or 1. The low value obtained for Er¹⁶⁵ is not understood; Tc^{97g} is a second forbidden transition $(\Delta I=2, no)$ and its comparative half-life is in the range of the beta decays listed by Feenberg¹⁸; the other two exceptions are discussed below.

Table IV lists six pure or predominantly *L*-capture transitions. Some of the transition energies are established only between *K*- and *L*-ionization energies as limits. We have calculated $\log ft$ values for the two cases in which the energy is rather well known, and estimated $\log f_1 t$ for Pb²⁰²; the latter value falls within the range of $\log f_1 t$ values for other first forbidden unique transitions.

Several cases occur in both Table I and Tables III or IV. In Table I auxiliary information is used to determine or to estimate the transition energies, and capture ratios are computed. In Tables III and IV the argument

Ą	mental method ^a	Initial state	Final state ((Mev)	% EC	perimental res L/K	$ults LM\cdots/K$	References ^h	Calculated Erc (Mev)	Half_life	Lowfor	Demon
K 40	$\mathbf{A}^{\mathbf{p}}$	4—	2+	1.46	11%		-			1 2/100	10/307	
Ic 97g	٩°	9/2+	5/2+	Ground	100% ($0.21 \pm 0.14 - 0.10$		K8	$0.185 + \frac{100}{0.055}$	1.3×10° y 2.6×10° v	12.5	a - 113
Cd 109	$\mathbf{A}^{\mathbf{p}}$	5/2+	7/2+	0.088	100% ().124±0.04	0.24 ± 0.04	W4	0.078 ± 0.010	470 d	ν.	
in 113	Cq	1/2 or 3/2+	1/2 or 3/2 +	0.650	3.5%		$2.23 \substack{+1.12 \\ -0.88}$	B24	$0.036 \substack{+0.004 \\ -0.001}$	119 d	5.0	
125	B°	5/2+	3/2+	0.0353	100% ().23±0.03		M5	$0.108 \substack{+0.020\\-0.010}$	60 d	4.7	
3a 133g	Cª	1/2+	1/2 +	0.437	~95%		$1.17 \substack{+0.26 \\ -0.20}$	G10	0.054 ± 0.003	7.5 y	5.8	
a 138	$\mathbf{A}^{\mathbf{p}}$	5+	2+	1.43	60%		1.4 ± 0.25	T3	0.185 ± 0.015	1 1 × 10 ^µ ↔	181	i.i 10. 4 4 10
Ce 139g	B°	3/2+	5/2+	0.166	100%		0.37 ± 0.02	K4	0 104 ± 0 006	140 J	1.01	17 = 10g/20 = 17
² m 145	Cť	5/2+	3/2-	0.067	26		0.85±0.03	B25	0.073 ± 0.010	18 v	C.C 7.7	
3d 153	Cq	7/2-	7/2+	0.104	$\{ \sim 100\% \ \sim 75\% \$		0.68 ± 0.02 0.34 ± 0.03	B13 G12	0.092 ± 0.002 0.149 ± 0.010	- <u>-</u>	2.6	<u>بر</u>
Jy 159	Ag			Excited		$1.0 {\pm} 0.3$		G11	0.079 + 0.010 -0.005	134 d	5.1	I
îr 165	Ag	7/2-		Excited		1.2 ± 0.4		G11	$0.082 \substack{+0.010\\-0.005}$	10 h	3.7	
la 179	Ag	7/2+	9/2+	Ground	100%	$1.4{\pm}0.4$		B15	0.094 ± 0.009	P 009	6.2	
V 181	As		7/2+	Ground	100%	1.54 ± 0.10		B5	0.092 ± 0.009	140 d	17 17	В
)s 185	Cq	1/2 -	3/2+	0.87	17%	~	1.08 ± 0.04 1.04±0.04	$\left. \begin{array}{c} \mathrm{B17} \\ \mathrm{I5} \end{array} \right\}$	0.110 ± 0.005	93 d	6.7	
Au 195	Cd	3/2+	5/2-	0.128	\sim 50%		5.5±0.9	B27	0.094 ± 0.002	192 d	7.2	
Au 196	Cq	2	2+	0.69	25%		0.41 ± 0.12	G9	0.234 ± 0.180 -0.050	5.6 d	6.1	
ri 202	Cq	2-	2+	0.44	61%		0.56 ± 0.04	H13, G7	0.197 ± 0.007	12 d	5.9	
Vp 235	As	5/2+	7/2-	Ground	<pre>>98% >-100%</pre>	30±2 36.7±4		H10 G8	$0.130 \\ 0.123\pm0.001$	410 d	7.1	
237 u 237	As	7/2-	5/2-	0.059	15%	2.8±0.8		H14	$0.153 \substack{+0.010\\-0.005}$	46 d	6.8	

TABLE III. New decay energies from capture ratios.

and the latter. dK x-ray-gamma-ray coincidences. 4π scintillation counter $(P \to 0)$. K x-ray-conversion electron coincidences. K and L x-ray intensities. ^b See list at end of article.

¹ See detailed discussion in Sec. IV. ¹ Eac (recisculated by the authors of this paper. ¹ More complicated decay schemes are reported by M15 and M16; see also B18 for another report. ² See Table IV for *L* capture to an excited state. A detailed elucidation of this decay scheme is reported by K11. ^m More complicated decay schemes are reported by D8 and C5.

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is inverted, and observed capture ratios are used to determine transition energies. In the cases of Bk²⁴⁵, Pu²³⁷, and Np²³⁵, the transition energies deduced from capture ratios are in agreement with the energies deduced from other data, within the limits of uncertainty assigned. Indeed, because of the steepness of the curve of capture ratio vs transition energy near the K-capture threshold, modestly well-determined capture ratios serve to define the transition energy with precision, e.g., Np²³⁵.

Potassium-40.-The decay scheme and decay constants of naturally occurring K⁴⁰ have been the subject of an unusual amount of interest primarily because of its geochronological importance. The decay scheme suggested by Morrison²² has been confirmed and is generally accepted. The nuclear spin of K⁴⁰ has been measured²³ and is found to be 4, and odd parity is assigned according to the shell model.24 The ground states of Ca⁴⁰ and A⁴⁰ are of course 0+. The 1.328 ± 0.010 -Mev beta decay of K⁴⁰ has a unique shape which is characteristic of a third forbidden transition $(\Delta I = 4, \text{ yes})$ ²⁵ The electron capture of K⁴⁰ proceeds (almost) entirely through the first excited state of A⁴⁰ which is of course believed to be 2+. Helm²⁶ has questioned this assignment on the basis of experiments on the inelastic scattering of 187-Mev electrons from argon, but his alternative assignment of 0+ cannot apply to the state which undergoes a radiative transition to the 0+ ground state. It is generally concluded that the electron-capture is a first forbidden unique transition $(\Delta I = 2, \text{ yes}).$

Mass spectrometric and particle reaction results appear in Table I. Giese and Benson (G13) have compared the doublets A^{40} – (C₃H₄) and K^{40} – (C₃H₄) in a high-resolution mass spectrometer, and find that the mass difference corresponds to 1505 ± 3 kev. It may be mentioned in passing that their result for the K⁴⁰-Ca⁴⁰ mass difference is 1320 ± 3 kev, in good agreement with the beta-decay results. Holland and Lynch (H15) have observed neutron spectra of the reaction $A^{40}(p,n)K^{40}$, and obtain an energy difference of 1522 ± 6 kev. The origin of this discrepancy is not known.

There are a number of reports of the energy of the K^{40} gamma ray, and an average value of 1462 ± 5 kev has been adopted by Endt and Braams,²⁷ although it may be noted that they frequently refer to the "level at 1.48 ± 0.02 Mev." If one combines the gamma-ray energy adopted by Endt and Braams with the mass spectrometric and particle reaction results, one obtains "closed cycle" values of 43 ± 8 and 60 ± 8 kev, respectively, for the capture energy of K^{40} .

²² P. Morrison, Phys. Rev. 82, 209 (1951).

- ²⁴ Reference 18, p. 32, for instance. ²⁵ L. Feldman and C. S. Wu, Phys. Rev. 87, 1091 (1952). ²⁶ R. H. Helm, Phys. Rev. 104, 1466 (1956). ²⁷ P. M. Endt and C. M. Braams, Revs. Modern Phys. 29, 683 (1957).

					-	7	4		
Z A	Half-life	Initial state	Final st	ate	Observed ca L/K	the ratios M/L	Referencesª	$E_{BC}(\mathrm{kev})$	Remarks
66 Dy 159	144 d	3/2 or 1/2-	5/2 or 3/2+	350 kev	510		K11	$9 < E_{EC} < 54$	350-kev gamma ray is not in coincidence with K x-rays
78 Pt 193g	\sim 500 y	3/2 or $1/2-$	3/2 +	Ground	>1000		N1	$13 < E_{BC} < 77$	Only L x-rays were observed
82 Pb 202	3×10 ⁵ y	+0	- 2	Ground	>200	~ 0.40	H5	$15 < E_{BC} < 86$	Observed P_{M}/P_{L} implies that E_{EC} is well above lower limit; $\log f_{i}(\cong 9.3)$
82 Pb 205g	$\sim 2 \times 10^7 \mathrm{y}$	5/2-	1/2 +	Ground	>1600		W5	$15 < E_{EC} < 86$	Only L x-rays were observed; $E_{BC} \simeq 110$ kev (cal- culated with neutron binding energies, F6)
93 Np 235	410 d	1/2+	1/2+	Ground	36.7±4	0.46 ± 0.05	G8, H10	123 ± 1	Observed P_M/P_L gives $g_M^2/g_{L^2}=0.35\pm0.04$, which may be compared with 0.32 (Fig. 2); $\log/t=7.1$
97 Bk 245	4.98 d			630 kev			M13	185±15	$P_L = 0.48 \ (\pm \sim 0.05); P_R \cong 0.1;$ these observations are consistent with E_{BC} given here and $P_{M'}/P_L$ =0.4; see Table I and Fig. 2; log/t=7.0
a See list at e	nd of article.			-					

 ²⁸ Da vis, Nagle, and Zacharias, Phys. Rev. 76, 1068 (1949); J.
 R. Zacharias, Phys. Rev. 61, 270 (1942).



FIG. 4. (top) Comparative half-life of K^{40} vs capture energy for a second forbidden unique transition. (bottom) Electron capture ratio vs transition energy for K^{40} . (a) L/K ratio, Brysk and Rose (reference 2); (b) LM/K ratio (reference 2 and Sec. IIA); (c) L/Kratio including effects of correlations, Odiot and Daudel (reference 20); (d) LM/K ratio including correlations (reference 20 and Sec. IIA). (Curves b and c are omitted from the drawing but are approximately uniformly spaced between curves a and d.) [The spin of K⁴⁰ is 4 and the parity is odd.]

The discussion can best be continued in terms of Fig. 4, in which we have plotted some curves of capture ratio vs transition energy for this first forbidden unique transition. The curves a to d are the following: (a) L/K capture ratio²; (b) LM/K capture ratio (reference 2 and Sec. IIA); (c) L/K capture ratio including the effects of correlations as calculated by Odiot and Daudel²⁰ for A³⁷; and (d) LM/K capture ratio including the effects of correlations (reference 20 and Sec. IIA). (Curves b and c have been omitted from the drawing in the interest of clarity, and lie approximately uniformly spaced between curves a and d.) The closed cycle values of the electron-capture energy are indicated by the bars along the energy axis.

Heintze (H4) determined the value of P_K from the specific activity of potassium for the emission of K x-rays and gamma rays. The capture ratio, $P_{LM}/P_K = 1.35\pm0.25$, is indicated by the bar along the capture ratio axis of Fig. 4. In obtaining this value Heintze measured the K-fluorescence yield of argon in an

auxiliary experiment²⁸ and found $\omega_{K}=0.13$. The transition energy inferred from Heintze's capture ratio is 20 ± 3 kev. Although the fluorescence yields obtained by Heintze²⁸ for higher-Z noble gases are a trifle larger than those reported by others¹⁰ for neighboring elements, it would seem unlikely that the over-all error in P_{K} is large enough to lead to an appreciable change in the inferred capture energy, especially in view of the steepness of the curves of Fig. 4 in this region.

Figure 4 also contains a plot of $\log f_1 t$ vs transition energy for K⁴⁰. Feenberg¹⁸ finds that first forbidden unique beta transitions have 7.5 $\geq \log f_1 t \geq 9.5$, and we find a similar range for this class of electron-capture transitions (see Sec. IIB). The energy obtained from the capture ratio leads to a value of $\log f_1 t$ in this range; the other energies do not.

If the argument is now inverted so that one accepts 20 ± 3 kev as the electron-capture energy, and 1505 ± 3 kev (MS) or 1522 ± 6 kev (p,n) for the mass difference; then the separation between the ground state and the first excited state of A⁴⁰ is 1485 ± 5 kev (MS) or 1502 ± 7 kev (p,n). The former value is not badly in disaccord with most of the measurements of the K⁴⁰ gamma-ray energy.

Lanthanum-138.—The nuclear spin of La¹³⁸ has been measured²⁹ to be 5, and even parity is assigned by the shell model. La¹³⁸ undergoes a beta transition to the first excited state of Ce¹³⁸ at 0.81 Mev, and an electroncapture transition to the first excited state of Ba¹³⁸ at 1.43 Mev (T3). The gamma rays are assigned by their appearance in the spectra of Pr¹³⁸ and Cs¹³⁸, respectively; furthermore, in the elucidation of the decay scheme of La¹³⁸, it was shown that the 1.43-Mev gamma ray is coincident with the barium K x-ray. Turchinetz and Pringle (T3) also determined the specific activity of La^{138} for barium K x-rays and for gamma rays, and found the capture ratio to be $P_{LM}.../P_{K}=1.4$ ± 0.25 . We have recalculated the transition energy for a second forbidden unique transition ($\Delta I = 3$, no) including the effect of capture from higher shells, and find that the capture energy is 185 ± 15 kev. However we are unable to include the contribution of d electrons, and this transition energy may be too small. $\text{Log } f_2 t$ is 12.9, and it is comparable to other transitions of this class.¹⁷

Samarium-145.—As a final indication of the applicability of electron-capture theory, we turn to the experiments (B25) on the decay scheme of Sm¹⁴⁵. The electron capture proceeds predominantly through the 61-kev state of the daughter, and the energy of this transition has been measured by observing the inner bremsstrahlung of electron capture. There is a very weak branch to a highly excited state of the daughter, and the capture energy of this branch can be calculated from the decay scheme. By coincidence technique the capture ratio is measured for this weak low-energy transition. Comparing it with theoretical predictions for various

²⁸ J. Heintze, Z. Physik 143, 153 (1955).

²⁹ P. B. Sogo and C. D. Jeffries, Phys. Rev. 99, 613 (1955).

Z	$\omega_{L_{\mathrm{III}}}$	$\omega_{L_{\mathrm{II}}}$	ω_{LI}	$\overline{\omega}_L$	K_{β}/K_{α} Refe	rencesa	Method of excitation
65 Tb 70 Yb 72 Hf 80 Hg 81 Tl 82 Pb				0.19 0.20 0.24 0.34 0.37 0.385	0.22 0.24 0.25 0.29 0.29 0.29 0.29	L15	Electron capture
73 Ta 78 Pt 79 Au 82 Pb	0.23 ± 0.02 0.275 ± 0.03 0.32 ± 0.03 0.35 ± 0.04	0.23 ± 0.04 0.31 ± 0.04 0.27 ± 0.04 0.24 ± 0.04	0.28 ± 0.07 0.35 ± 0.08 0.36 ± 0.09 0.37 ± 0.09		· · · · · ·	R6	Monochromatic x-rays
80 Hg	$0.34{\pm}0.01$	0.42 ± 0.09		0.371 ± 0.035]	H16	Internal conversion in Au ¹⁹⁹
80 Hg				$0.34{\pm}0.04$		S10	Electron capture in Tl ²⁰⁴
81 Tl 83 Bi	0.33 ± 0.02			0.32 ± 0.02 0.40 ± 0.02		W9	Internal conversion in $Pb^{212}-Bi^{212}-Tl^{206}$ (ThB-C-C'').
					•		

TABLE V. Recent measurements of L-fluorescent yields.

^a See list at end of article.

degrees of forbiddenness, Brosi, Ketelle, Thomas, and Kerr concluded that the capture transition is forbidden, and used the fact in the further elucidation of their decay scheme.

V. SUMMARY OF DATA ON MEAN L-FLUORESCENCE YIELDS

The determination of mean L-fluorescence yields was discussed in reference 1, and a more recent summary has been given by Fink (F5). Since that time additional measurements have appeared from studies on radioactive Tb, Yb, Hf, Au, Hg, Tl, Pb, and Bi. Roos (R6) has informed us of the results of x-ray excitation experiments on Ta, Pt, Au, and Pb. Lazar (L15) has also made recent measurements of the ratio of the K_{α} and K_{β} x-ray intensities. These new results are summarized in Table V; and these values, as well as all other available nuclear data on ω_L , are shown in Fig. 5, in which the values obtained by Lay (L14) for x-ray excitation are shown for comparison. The break at $Z \approx 73$ is interpreted to mean that above this point Coster-Krönig transitions of the type $L_{\rm I} - L_{\rm III} M_{\rm IV,V}$ contribute, thus shifting $L_{\rm I}$ vacancies to the $L_{\rm III}$ subshell for which the partial fluorescence yield is greater. These transitions are energetically forbidden for $Z \gtrsim 73$, and other Coster-Krönig transitions which transfer vacancies between the L subshells occur only to a very small extent.³⁰

As pointed out previously,¹ the mean *L*-fluorescence yield is an average of the fluorescence yields of the Lsubshells, weighted in a manner which is not at all well known, and weighted differently for different modes of excitation. Ross, Cochran, Hughes, and Feather³¹ analyzed this problem in very great detail,

and extracted the partial L-fluorescence yields for bismuth from experimental results on x-ray fluorescence excitation and on nuclear excitation by internal conversion of the 47-kev gamma ray of RaD(Pb²¹⁰). This analysis is summarized briefly by Wapstra, Nijgh, and Van Lieshout.⁴ Unfortunately there is very little detailed data except in the region around Z=82, and we can only echo the call³¹ for more precise and more extensive experimental measurements in this field.



FIG. 5. Mean L-fluorescence yields vs atomic number. The solid points are the results of Lay (L14) from x-ray excitation. Open points are all derived from nuclear excitation and represent the following work (see list of references at end of article): (a) Ag^{109} , from EC in Cd¹⁰⁹ (B3), (b) Xe¹³¹, from EC in Cs¹³¹ (F3), (c) Eu¹³³, from decay of Gd¹⁶³ (B13), (d) Hf¹⁷⁹, from EC in Ta¹⁷⁹ (B15), (e) Hg²⁰⁴, from EC in Tl²⁰⁴ (S10), (f) Bi²¹⁰, from decay of Pb²¹⁰ (RaD) (F5), (g) Tl²⁰⁸, from alpha decay of Bi²¹²(ThC) (W9) (see also B22), (h) Bi²¹², from decay of Pb²¹²(ThB) (W9) (see also B22), (i) Ra²²⁶, from alpha decay of Th²³⁰(Io) (B23), (j) Hg¹⁹⁹, from decay of Au¹⁹⁹ (H16). Open triangles are the data from (L15).

³⁰ E. H. S. Burhop, *The Auger Effect* (Cambridge University Press, Cambridge, England, 1954). ³¹ Ross, Cochran, Hughes, and Feather, Proc. Phys. Soc. (London) A68, 612 (1955).

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