# Recent Experimental Results on Orbital Electron Capture

BEROL L. ROBINSON,\* Department of Physics, Western Reserve University, Cleveland, Ohio

AND

RIGHARD W. FINK,<sup>†</sup> Department of Chemistry, University of Arkansas, Fayetteville, Arkansas

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  $18\angle Z \leq 53$ ; 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.

**VONSIDERABLE** experimental data have appeared  $\overline{\phantom{a}}$  since the publication in 1955 of a review<sup>1</sup> which compared experimental values of  $L/K$  capture ratios with the theoretical results of Brysk and Rose,<sup>2</sup> and which summarized our knowledge of transition energies involved in electron-capture decay and of mean L-Auorescence yields. The purpose of the present paper, therefore, is to present new data along the lines established previously.

## I. EXPERIMENTAL TECHNIQUES

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

\* Supported in part by the National Science Foundation and the Department of Defense through the U.S. Air Force Office of Scientific Research.

\$ Supported in part by the National Science Foundation and the U. S. Atomic Energy Commission. Present address: The Gustaf Werner Institute for Nuclear Chemistry, University of Uppsala, Uppsala, Sweden.

<sup>1</sup>B. L. Robinson and R. W. Fink, Revs. Modern Phys. 27, 424<br>(1955). Errata: Table I:  $E_{BC}$  of Mn<sup>54</sup> should be 0.540 $\pm$ 0.010 Mev  $(p,n)$ , cc. This makes no change in the capture ratios. Also a new measurement has been reported for  $Mn^{54}$ ;  $E_{EC} = 0.528 \pm 0.010$ <br>*IB*(J4). Table II(b); K<sup>40</sup> shoul dread (4- to 2+). Reference M4:<br>change "1168" to "1186".<br><sup>2</sup> H. Brysk and M. E. Rose, Oak Ridge National Laboratory<br>Rept

1169 (1958).

f. 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  $(K-LL, \text{ or } K-LX)$ . The x-ray intensity ratio  $I_L/I_K$ is related to the capture ratio  $P_L/P_K$  by the expression

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

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

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

 $k = I_{K_\alpha}/I_K = \text{intensity ratio of } K_\alpha$  x-rays to total K x-rays;  $a_K = K$  Auger yield =  $(1 - \omega_K)$ ; and  $(K$ = partial Auger yields; X denotes  $M$ -, N-, etc. shell  $\begin{array}{ll} L & K \ LX \end{array}$ 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<sup>3,4</sup> (see Sec. IE and Fig. 1). The fraction of  $K$ results at  $K-LX$  is the probability that a  $K$ -bild electrons.  $(K-LX)$  is the probability that a  $K$ -shell electrons.  $(K-LX)$  is the probability that a  $K$ -shell vacancy is filled by an  $X$ -shell electron with the excess en x-rays in the  $K_{\alpha}$  group,  $(I_{K_{\alpha}}/I_K)$ , has been computed from the data in Compton and Allison' and other tabulations,<sup>6,7</sup> in the case of gallium a recent determinations,<sup>6,7</sup> in the case of gallium a recent determination has beeri made by Drever and Moljk (D6), and Lazar (L15) has communicated recent measurements for some

<sup>&</sup>lt;sup>8</sup> L. Slack and K. Way, Radiations from Radioactive Atoms in Frequent Use (U. S. Atomic Energy Commission, Washington, D. C., 1959), pp. 67–69.

<sup>&</sup>lt;sup>4</sup> Wapstra, Nijgh, and van Lieshout, *Nuclear Spectroscopy*<br> *Tables* (Interscience Publishers, Inc., New York, 1959), pp. 82–87.<br>
It may be noted that the *fxL*<sub>i</sub> estimated in these Tables, pp. 86–87,

is connected with our  $n_{KL}$  by the relation  $n_{KL} = \sum_i f_{KL_i}$ .<br>
<sup>5</sup> A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., Princeton, New<br> *Ersey, 1935*), second edition, p. 637f

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). E-fluorescence yields are given by Broyles, Thomas, and Haynes,<sup>8</sup> Gray (G6), Laberrigue-Frolow and Radvanyi,<sup>9</sup> Roos, $^{10}$  and others. A recent summary of mean  $L$ -fluorescence yields has been given by Fink (FS) 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.<sup>4</sup>

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  $K_{\alpha}$  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)

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

where  $P$  is the probability that a  $K$  x-ray can escape from the counter without being detected, and  $k=I_{K_{\alpha}}/I_{K_{\alpha}}$ is the intensity ratio of  $K_{\alpha}$  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  $\omega_K$ . For example, for Ge<sup>71</sup> Langevin<sup>11</sup> obtained  $P_L/P_K$ =0.30 using  $\omega_K$ =0.45; but if  $\omega_K$  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  $\omega_K$  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  $P_L/P_R$  directly, with an extremely small correction for K escape. The  $P \rightarrow 0$ technique has been pioneered by Drever, Moljk, 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 Ge<sup>71</sup>H<sub>4</sub> 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  $Cd^{109}$  and  $I^{125}$  (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,  $I^{126}$  has been produced by the reaction  $I^{127}(\gamma,n)$ I<sup>126</sup> 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  $R_{\gamma}$  and  $R_{K_{\gamma}}$  be the counting rates for gamma rays and x-ray—gamma-ray coincidences, re-

<sup>&</sup>lt;sup>8</sup> Broyles, Thomas, and Haynes, Phys. Rev. 89, 715 (1953).<br><sup>9</sup> J. Laberrigue-Frolow and P. Radvanyi, J. phys. et radium 17, 944 (1956).

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

<sup>»</sup> M. Langevin, Compt. rend. 239, 1625 (1954).

spectively;

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

and

$$
R\kappa_{\gamma}/R_{\gamma} = P_{K}\omega_{K}\epsilon_{K}S_{K}, \qquad (4)
$$

where D is the disintegration rate, S and  $\epsilon$  are solid angles and intrinsic detection efficiencies of the counters, and  $\omega_K$  is the K-fluorescence yield. The presence of internal conversion and other branches may complicate the analysis, but the result remains essentially unchanged (K4, 825). 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 (67, 69).

#### 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  $(P_L+P_M+\cdots)/P_K \equiv P_{LM}.../P_K$ , and this value is frequently reported. Distinction mill 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 a knowledge of the K-fluorescence yield is required. Th<br>best measurements to date are those of Roos,<sup>10</sup> whicl delineate the steeply rising portion of the  $\omega_K$  vs Z curve for 26  $\ll$  Z  $\ll$  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  $ZZ$   $Z$  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. uncertainties as shown in Fig. 1. For  $Z \leq 30$ , the chief source of uncertainty lies in the K-fluorescence yield; if  $\omega_K$  were known to one percent, then the uncertainty in  $n_{KL}$  would be halved. If in addition the ratio of the intensities of the  $K_{\alpha}$  and  $K_{\beta}$  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  $Z \leq 50$ ; for  $30 \leq Z \leq 50$ , improved Auger yield measurements are also required. For  $Z \leq 30$ , 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 nth forbidden transitions with maximum spin change (the so-called unique transitions). The only nuclides repeated in Table I are  $Np^{235}$  and  $K^{40}$ , 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,<sup>2</sup> one might examine the nonrelativistic wave functions for a hydrogenlike nonrelativistic wave functions for a hydrogenlik<br>atom.<sup>12</sup> The s-electron wave-functions are proportion 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  $\dot{p}$  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<br>others.<sup>13</sup> Following the latter, we have calculated the others. Following the latter, we have calculated the total contribution to s-electron capture and p-electron

<sup>&</sup>lt;sup>12</sup> See for instance L. Pauling and E. B. Wilson, Jr., *Intro-*<br>duction to Quantum Mechanics (McGraw-Hill Book Company,<br>Inc., New York, 1935), pp. 135–139.<br><sup>13</sup> D. R. Hartree, Proc. Roy. Soc. (London) **A141**, 282

<sup>(1933),</sup>  $(Cl^-, Cu^+)$ ; D. R. Hartree, Phys. Rev. 46, 737 (1934), (Hg<sup>+</sup>); D. R. Hartree, Proc. Roy. Soc. (London) A143, 506 (1934), (Cs<sup>+</sup>, K<sup>+</sup>); D. R. Hartree and W. Hartree, Proc. Roy. Soc. (London) A143, 506 (1934), (Cs<sup>+</sup>



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capture from shells of  $n \geq 2$ . 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 Rose' 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 mercury<sup>14</sup> 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. Comyarative Half-Life

As before, the logarithms of the comparative halflives  $(\log ft)$  have been obtained from the nomograms of Moszkowski<sup>15</sup> or by the method given by Major and Biedenharn<sup>16</sup> in the case of low-energy transitions.

Davidson<sup>17</sup> 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 Biedenharn'6 and by Brysk and Rose':

 $ft = f_0t = (\pi/2)g_K^2 q_K^2 [1 + (P_{LM}.../P_K)]t,$ 

 $f_1t = (\pi/24) g_K^2 q_K^4 [1+(P_{LM}.../P_K)]t,$ and

$$
f_2t = (\pi/2160)g_K^2 q_K^6 [1 + (P_{LM}.../P_K)]t. \quad (5)
$$

For completeness we give here the  $\log f_1 t$  values for the first forbidden unique transitions listed in reference 1: Ca<sup>41</sup>,  $\log f_1 t = 9.4$ ; T<sup>204</sup>,  $\log f_1 t = 8.4$ . These two transitions, the ground state transitions of As<sup>74</sup>, Rb<sup>84</sup>, and  $I^{126}$ , and the weak inner transition in  $Sm^{145}$  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,<sup>18</sup> although  $Ca<sup>41</sup>$  is at the upper limit. Feenberg,<sup>18</sup> although Ca<sup>41</sup> is at the upper limit

The  $\log f_2 t$  value for the second forbidden unique transition of  $Po^{209}$  is also near the values calculated by Davidson<sup>17</sup> for Be<sup>10</sup> and Na<sup>22</sup>. On the basis of comparative half-life, one is tempted to classify the electroncapture of Bi<sup>208g</sup> as second forbidden  $(\Delta I=2, n_0)$ , 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 Ni<sup>59</sup> ( $\Delta I = 2$ , no; log ft= 11.9). For forbidden transitions with  $\Delta I \geq 2$ , 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  $Rose<sup>2</sup>$  in which  $g$  and  $f$ , the large and small part of the Dirac wave function, are mixed.

(i) There is no effect on the  $L_I/K$  ratio for two reasons. In the first place,  $(f_{L_I}/f_K)^2 = (g_{L_I}/g_K)^2$ . 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  $L_{III}/L_{I}$  ratio also depends on the matrix elements, and precise determinations of both the capture energy and the  $L_{III}/L_{I}$  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 Ni<sup>59</sup>, Tc<sup>97g</sup>, 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 <sup>15</sup> cases. Table II corresponds to Table II $(a)$  of reference 1 and includes the cases mentioned therein with the exception of Pd<sup>103</sup>; the latest experimental results on Pd<sup>103</sup> make the situation appear inconclusive at this time. '

<sup>&</sup>lt;sup>14</sup> D. F. Mayers, Proc. Roy. Soc. (London) A241, 93 (1957), (Hg+, relativistic). "S.A. Moszkowski, Phys. Rev. 82, <sup>35</sup> (1951);reproduced in

part in reference (S14), pp. 597–600.  $\frac{1}{16}$ . K. Major and L. C. Biedenharn, Revs. Modern Phys. 26,

<sup>321</sup> (1954).

<sup>&</sup>lt;sup>17</sup> Jack P. Davidson, Jr., Phys. Rev. 82, 48 (1951).<br><sup>18</sup> Eugene Feenberg, *Shell Theory of the Nucleus* (Princeto:<br>University Press, Princeton, 1955), Chap. V.

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

<sup>&</sup>lt;sup>19</sup> Avignon, Michalowicz, and Bouchez, J. phys. et radium 16, 404 (1955).

z $\pmb{A}$	Experimental method and results Methoda References $P_L/P_K$			$P_{LM}/P_K$	Theoretical predictions $P_L/P_K$ $P_{LM}$ / $P_K$		Remarks
18 A 37	$B(P \text{ small})$	(L11 K10	$\substack{0.092 + 0.010 \ 0.102 - 0.005 \ 0.102 \pm 0.008}}$		0.082		h
19 K 40	$A^b$	H4					See Sec. IV for discussion
26 Fe 55	$B(P\rightarrow 0)$	S <sub>13</sub>	$0.108 + 0.006$		0.097		h
27 Co 57	c	M12		$0.20 \pm 0.13$		0.107	
32 Ge 71	$B(P\rightarrow 0)$	D6, D7 D <sub>9</sub>	$0.128 + 0.005$ -0.003 $0.116 + 0.005$		0.106		h This result is considered to supercede that given in reference D6
33 As 74	$B(P\rightarrow 0)$	S9	$0.085 + 0.020$		0.095		
36 Kr 79	$B(P \text{ not small})$ $B(P\rightarrow 0)$	L12 D <sub>9</sub>	$0.16 + 0.03$ $0.108 + 0.005$		0.101		i $\mathbf h$
37 Rb 84	C <sub>d</sub>	W <sub>6</sub>		$0.12 + 0.05$		0.121	Capture to 0.89-Mev level
38 Sr 85	C <sup>d</sup>	<b>B26</b>		$0.14 \pm 0.05$		0.130	Capture to 513-kev level; reported as $P_K = (88 \pm 4)\%$ ; theoretical value is $89\%$
53 I 126	$B(P\rightarrow 0)$ <sup>e</sup>	S <sub>12</sub>	$0.142 + 0.005$ -0.018		0.123		h
62 Sm 145	C <sup>f</sup> $\rm C^{d}$	<b>B25</b>		$0.20 \pm 0.02$ $0.6 \pm 0.1$		0.189 0.99 0.29	Capture to 61-key level Forbidden transition to 485-key level Allowed transition to 485-kev level
85 At 211	A	$_{\rm H8}$	0.143		$0.215 \pm 0.002$		See also (G6)
93 Np 235	A	∫H10 GG8	$30\pm2$ $36.7 + 4$		$11.3 + no$ limit -8.5		ĵ
94 Pu 237	A°	H14	$2.8 + 0.8$		$+20$ $3.0 - 1.2$		Capture to 59.6-key level; see Table III
97 Bk 245	C <sup>d</sup>	M13 $\sim 0.33$			$0.32 + 0.01$		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 coincidence spectrometry, respectively; P is the probability that a K x-ray escape from the counter in internal source spectrometry. The probability that a K

ment between theory and experiment. The apparent discrepancies in the cases of  $K^{40}$ , At<sup>211</sup>, and the inner branch of Sm<sup>145</sup> can in all likelihood be attributed to insufficiently precise data; and the result of Langevin for  $Kr^{79}$  (L12) depends critically upon the K-fluorescence yield. However, the precision measurements for  $A^{37}$ , Fe<sup>55</sup>, Ge<sup>71</sup>, Kr<sup>79</sup>, and I<sup>126</sup> are not in accord with the predictions of the theory of Brysk and Rose.' The six experimental results which are precise enough for critical comparison with the predictions of Brysk and Rose' are summarized in Fig. 3. The correction proposed by Odiot and Daudel<sup>20</sup> for  $A^{37}$  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.<sup>21</sup>

## A. Correlations

Odiot and Daudel<sup>20</sup> 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

<sup>&</sup>amp;0 S. Odiot and R, Daudel, J. phys, et radium 17, 60 (1956).

<sup>»</sup> H. Brysk (private communication 1959).

the nucleus. (The latter is in a fashion taken into account by Brysk and Rose' 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 4Be' seems to be quite inaccessible to experimental verification; however, some of the other low-Z electroncapturers  $({}_9\text{F}^{18}, { }_{11}\text{Na}^{22}, { }_{13}\text{Al}^{26}, { }$  and  ${}_{17}\text{Cl}^{36}$ ) 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 ascribed<sup>11</sup> to  $_{32}Ge^{71}$ , it may perhaps account for the small discrepancies found in the recent precision experiments.

### B. Discussion of Cases in Table Il

Argon-37.—The effect of correlations has been cal-*Argon-37*.—The effect of correlations has been calculated only for this case,<sup>20</sup> 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  $Kr<sup>79</sup>$  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 R indicated 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 Cd<sup>109</sup>,  $I^{125}$ , and W<sup>181</sup> are merely repeated; there are new experimental results for Ba<sup>133</sup>, Os<sup>185</sup>, Au<sup>195</sup>, and T<sup>202</sup>, and twelve other radioactivities;  $K^{40}$  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  $K^{40}$ , Tc<sup>97g</sup>,  $La<sup>138</sup>$ , and  $Er<sup>165</sup>$ , 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  $Er^{165}$ is not understood;  $Tc^{97g}$  is a second forbidden transition  $(\Delta I=2, n_{0})$  and its comparative half-life is in the range of the beta decays listed by Feenberg<sup>18</sup>; 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 Pb<sup>202</sup>; 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



TABLE III. New decay energies from capture ratios.

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is inverted, and observed capture ratios are used to determine transition energies. In the cases of  $Bk^{245}$ ,  $Pu^{237}$ , and  $Np^{235}$ , 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.,  $Np^{235}$ .

Potassium-40.—The decay scheme and decay constants of naturally occurring  $K<sup>40</sup>$  have been the subject of an unusual amount of interest primarily because of its geochronological importance. The decay scheme suggested by Morrison<sup>22</sup> has been confirmed and is generally accepted. The nuclear spin of  $K^{40}$  has been measured<sup>23</sup> and is found to be  $4$ , and odd parity is assigned according to the shell model.<sup>24</sup> The ground states of Ca<sup>40</sup> and A<sup>40</sup> are of course 0+. The 1.328  $\pm 0.010$ -Mev beta decay of K<sup>40</sup> has a unique shape which is characteristic of a third forbidden transition which is characteristic of a third forbidden transition  $(\Delta I=4, \text{ yes})^{25}$  The electron capture of  $K^{40}$  proceed (almost) entirely through the first excited state of  $A<sup>40</sup>$ which is of course believed to be  $2+$ . Helm<sup>26</sup> 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  $A^{40} - (C_3H_4)$  and  $K^{40} - (C_3H_4)$  in a high-resolution mass spectrometer, and find that the mass difference corresponds to  $1505 \pm 3$  kev. It may be mentioned in passing that their result for the  $K^{40}$  –  $Ca^{40}$ mass difference is  $1320 \pm 3$  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  $1522 \pm 6$  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  $1462 \pm 5$  kev<br>has been adopted by Endt and Braams,<sup>27</sup> although it has been adopted by Endt and Braams,<sup>27</sup> although it may be noted that they frequently refer to the "level at  $1.48\pm0.02$  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  $43\pm8$  and  $60\pm8$  kev, respectively, for the capture energy of  $K^{40}$ .

~ P. Morrison, Phys. Rev. 82, <sup>209</sup> (1951).

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- $(1957).$

![](_page_8_Picture_1505.jpeg)

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<sup>&</sup>lt;sup>23</sup> Davis, Nagle, and Zacharias, Phys. Rev. 76, 1068 (1949); J.<br>R. Zacharias, Phys. Rev. 61, 270 (1942).<br><sup>24</sup> Reference 18, p. 32, for instance.<br><sup>25</sup> L. Feldman and C. S. Wu, Phys. Rev. 87, 1091 (1952).<br><sup>26</sup> R. H. Helm,

![](_page_9_Figure_1.jpeg)

FIG. 4. (top) Comparative half-life of  $K^{40}$  vs capture energy for<br>a second forbidden unique transition. (bottom) Electron capture<br>ratio vs transition energy for  $K^{40}$ . (a)  $L/K$  ratio, Brysk and Rose<br>(reference 2); (b ratio including effects of correlations, Odiot and Daudel (reference 20);  $(d) LM/K$  ratio including correlations (reference 20 and Sec.  $\overrightarrow{HA}$ . (Curves b and c are omitted from the drawing but are approximately uniformly spaced between curves a and d.) [The spin of  $K^{40}$  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 ratio<sup>2</sup>; (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 Daudel<sup>20</sup> for  $A^{37}$ ; 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,  $P_{LM}/P_K$  $= 1.35 \pm 0.25$ , 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 experiment<sup>28</sup> and found  $\omega_K = 0.13$ . The transition energy inferred from Heintze's capture ratio is  $20\pm3$  kev. Although the fluorescence yields obtained by Heintze<sup>28</sup> for higher-Z noble gases are a trifle larger than those reported by others<sup>10</sup> 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 K<sup>40</sup>. Feenberg<sup>18</sup> finds that first forbidden unique beta transitions have 7.5  $\approx$  log  $f_1 t \approx 9.5$ , 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  $20\pm3$  kev as the electron-capture energy, and 1505  $\pm 3$  kev (*MS*) or 1522 $\pm 6$  kev (*p*,*n*) for the mass difference; then the separation between the ground state and the first excited state of  $A^{40}$  is  $1485 \pm 5$  kev (*MS*) or  $1502 \pm 7$  kev  $(p,n)$ . The former value is not badly in disaccord with most of the measurements of the K<sup>40</sup> gamma-ray energy.

*Lanthanum-138*.—The nuclear spin of La<sup>138</sup> has been measured<sup>29</sup> to be 5, and even parity is assigned by the shell model. La<sup>138</sup> undergoes a beta transition to the first excited state of Ce<sup>138</sup> at 0.81 Mev, and an electroncapture transition to the first excited state of Ba<sup>138</sup> at 1.43 Mev (T3). The gamma rays are assigned by their appearance in the spectra of Pr<sup>138</sup> and Cs<sup>138</sup>, respectively; furthermore, in the elucidation of the decay scheme of La<sup>138</sup>, 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<sup>138</sup> for barium  $K$  x-rays and for gamma rays, and found the capture ratio to be  $P_{LM}$ .../ $P_K = 1.4$  $\pm 0.25$ . 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  $185 \pm 15$  kev. However we are unable to include the contribution of  $d$  electrons, and this transition energy may be too small. Log  $f_2t$  is 12.9, and it is comparable to other transitions of this class.<sup>17</sup>

Samarium-145.—As a final indication of the applicability of electron-capture theory, we turn to the experiments (B25) on the decay scheme of Sm<sup>145</sup>. 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

<sup>&</sup>lt;sup>28</sup> J. Heintze, Z. Physik 143, 153 (1955).

<sup>&</sup>lt;sup>29</sup> P. B. Sogo and C. D. Jeffries, Phys. Rev. 99, 613 (1955).

![](_page_10_Picture_1504.jpeg)

TABLE V. Recent measurements of L-fluorescent vields.

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 (FS). 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  $K_{\alpha}$ and  $K_{\beta}$  x-ray intensities. These new results are summarized in Table V; and these values, as well as all other available nuclear data on  $\omega_L$ , 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  $L_I - L_{III} M_{IV, V}$ 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  $Z \nless 73$ , and other Coster-Krönig transitions which transfer vacancies between the  $L$  subshells occur only to a very small extent.<sup>30</sup> to a very small extent.

As pointed out previously,<sup>1</sup> the mean  $L$ -fluorescence yield is an average of the fluorescence yields of the  $L$ subshells, weighted in a manner which is not at all well known, and weighted differently for different modes of excitation. Ross, Cochran, Hughes, and Feather<sup>31</sup> 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  $RaD(Pb^{210})$ . This analysis is summarized briefly by Wapstra, Nijgh, and Van Lieshout.<sup>4</sup> Unfortunately there is very little detailed data except in the region around  $Z=82$ , and we can only echo the call<sup>31</sup> for more precise and more extensive experimental measurements in this field.

![](_page_10_Figure_11.jpeg)

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 Cd<sup>109</sup> (B3), (b) Xe<sup>131</sup>, from EC in Cs<sup>131</sup> (F3), (c) Eu<sup>157</sup> from decay of Gd<sup>153</sup> (B13), (d) Hf<sup>179</sup>, from EC in Ta<sup>179</sup> (B15), (e) Hg<sup>204</sup>, from *EC* in Tl<sup>204</sup> (S10), (f) Bi<sup>210</sup>, from decay of Pb<sup>210</sup>  $(RaD)$  (F5), (g) Tl<sup>208</sup>, from alpha decay of Bi<sup>212</sup>(ThC) (W9) (see also B22),  $(h)$  Bi<sup>212</sup>, from decay of Pb<sup>212</sup>(ThB) (W9) (see also  $B22$ ), (i) Ra<sup>226</sup>, from alpha decay of Th<sup>230</sup>(Io) (B23), (j) Hg<sup>19</sup> from decay of Au<sup>199</sup> (H16). Open triangles are the data from  $(L15)$ .

<sup>&</sup>lt;sup>30</sup> E. H. S. Burhop, The Auger Effect (Cambridge University

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