L/K-Capture Ratios, Mean L-Fluorescence Yields, and Transition Energies in Orbital Electron-Capture *

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INTRODUCTION

ADVANCES in the technique of proportional and scintillation spectrometry have extended our knowledge of orbital electron-capture. In a study of comparative half-lives (ft values), Major and Biedenharn[1] have summarized the data existing up to the middle of 1954.

The theoretical work of Marshak[2] and Rose and Jackson[3] has been extended by Brysk and Rose[4] in the light of present knowledge of beta-decay theory of forbidden transitions, with particular reference to capture of L-shell electrons.

In the interpretation of radiative electron-capture (inner bremsstrahlung) spectra, the capture of p-electrons appears to be significant.[5,6]

This review comprises a summary and analysis of the existing data on electron-capturing nuclides (up to May, 1955) whose decay schemes are relatively simple and well established. In particular, the primary concern is with the ratio of L-capture to K-capture both as a test of the theory of Marshak and of Brysk and Rose and as an application of the theory to the determination of transition energies in electron-capture and of L-fluorescence yields.

EXPERIMENTAL TECHNIQUES

Essentially three techniques have been applied to the determination of x-ray intensity and capture ratios. These are described below.

(A) Internal Source Spectrometry

The radioactive material is contained within the sensitive volume of the detector. Gas proportional counters have been used for the study of A^{37},[7] Kr^{79},[8] and Ge^{71} [9] (in the form of germane, GeH_4). In this method the prompt cascade of x-rays and Auger electrons, which follows each K-capture event, is integrated by the detector to give a single "K-line" in the pulse-height spectrum. An "L-line" arises from L-captures and from K-captures which are followed by escape of the K x-rays from the sensitive volume. The correction for K x-ray escape may be made small by suitable choice of detector material and size, and it can be calculated quite accurately from x-ray absorption data and K-fluorescence yields.[10] Since L x-rays and L-Auger electrons are totally absorbed, no correction need be made for L-fluorescence yield.

Scintillation crystals have been grown[11] containing radioactive I^{125} and Cd^{109}. In this type of experiment the amount of L-capture is obtained from the difference between the number of K x-rays and the total number of gamma-ray transitions on the assumption that one gamma ray accompanies each decay.

(B) External Source Spectrometry

The radioactive substance is placed outside of the sensitive volume, and corrections must be applied for source self-absorption and self-scattering, for differential air and window absorption, and for K and L fluorescence yields. One must also consider that a K-shell hole may be filled by an L-electron either by radiative transition ($K_\alpha = K-L_{II}, L_{III}$) or by Auger transition ($K-LL$, $K-LX$). The number of L-shell vacancies produced in this manner, n_{KL}, ranges from 1.36 at $Z=29$ to 0.75 at $Z=90$, as will be discussed below.

In case electron capture is followed by gamma emission, the conversion of the gamma rays must be taken into account.

Many electron-capturing nuclides of the heaviest elements have been investigated.[12,13] The intensities of the L x-rays have been measured carefully by use of proportional counters and a bent-crystal x-ray spectrometer.[12] However, most of these nuclides have low-energy gamma transitions which are highly converted in the L-shell so that the interpretation of x-ray intensities in terms of electron-capture ratios is tenuous. No attempt has been made to correlate these data with theory in this paper.

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(C) Absorption Experiments

Some absorption measurements of relative L and K x-ray intensities have been reported by Wilkinson and Hicks[14] and Chu.[15] These measurements have not been considered in this work. Most of the nuclides involved have hard gamma-rays and conversion electrons; their decay schemes and conversion coefficients are unknown, and there is always the possibility of low-energy (and highly converted) gamma transitions unresolved from the x-rays.

CALCULATED P_L/P_K RATIOS

Marshak[2] has shown that L-capture always accompanies K-capture and occurs with increasing probability as the transition energy becomes small and Z large. For allowed transitions the ratio of capture probabilities is given by

$$P_L/P_K=\left(\frac{q_L^2}{q_K^2}\right)\left(\frac{g_{L_I}^2}{g_K^2}\right)[1+f_{L_{II}}^2/g_{L_I}^2], \qquad (1)$$

where q_J is the neutrino energy for J-capture; g_J and f_J are the "large" and "small" components of the Dirac radial wave function in a Coulomb field, respectively. Marshak evaluated the appropriate ratios using relativistic wave-functions with Slater screening. He also indicated that for forbidden transitions the capture of electrons of $j>\frac{1}{2}$ might compete favorably under certain conditions.

Rose and Jackson[3] have computed wave-function ratios using self-consistent (Hartree) wave functions and relativistic wave functions for a Thomas-Fermi atom with exchange. The ratio of $(g^2_{L_I}/g^2_K)$ is given graphically as a function of atomic number.

Brysk and Rose[4] have extended the previous work in the light of present knowledge of beta-decay theory of forbidden transitions. For the first-forbidden unique transition ($|\Delta J|=2$, yes), the expression for the capture ratio is particularly simple. They have also computed the values of the Dirac radial wave functions at the nuclear radius, and have given wave function ratios using relativistic wave functions corrected for the finite size of the nucleus, variations in electron wave functions over the nuclear volume, and screening. The results obtained for L_I/K capture are similar to those of Rose and Jackson. Using the wave functions and wave-function ratios given graphically by Brysk and Rose, one can compute L_{II} and L_{III} capture probabilities.

Capture ratios have been computed from the above theoretical results for those cases where the decay scheme is relatively simple and the transition energy known. Table I contains these computed ratios as well as experimental values for half-life, energy of the electron-capture transition, and branching ratio. Also tabulated is the nature of the transition according to the shell model, as given by King,[16] and the logarithm of the comparative half-life ($\log ft$). The latter is either taken from Major and Biedenharn[1] or calculated by their method or from the nomogram of Moszkowski.[17]

The relative probability of L_{III} capture has been calculated for the four cases where the spin change is believed to be greater than unity. The electron-capture transitions of K^{40}, Ca^{41}, and Tl^{204} involve changes of parity as well as changes of angular momentum of two units ($|\Delta J|=2$, yes), corresponding to first-forbidden unique type of beta decay. The shell model indicates that Ni^{59} undergoes a second-forbidden electron-capture transition ($|\Delta J|=2$, no); capture ratios were computed for mixtures of the A_{ij} and T_{ij} matrix elements (for the tensor interaction) and found to be appropriate for second-forbidden beta decay having $|\Delta J|=2$, no.[18] It appears that a sensitive experimental determination of the capture ratio of Ni^{59} might allow an estimate to be made of the ratio A_{ij}/T_{ij} for this transition.

COMPARISON OF THEORY WITH EXPERIMENT

In a few cases direct measurements have been made of the P_L/P_K capture ratio where the transition energy is known. These are given in Table II (a) along with the computed values transposed from Table I. In several other cases the energy is deduced from the measured capture ratio as shown in Table II (b). The experimental results have been recalculated by the present authors for Ge^{71} and Kr^{79} using K-fluorescence yields given by Broyles, Thomas, and Haynes[10] instead of those of Burhop[19] used in the original reports.

The theory is in relatively good agreement with the experiments on A^{37} and Kr^{79}, and in the case of K^{40} the agreement is not within the assigned error, because the experimental errors in the values used in the closed cycle calculation (see below) might lie mostly in one direction.

The energies deduced in Table II (b) lead to comparative half-lives (ft values) consistent with allowed transitions for Cd^{109}, I^{125}, and Ba^{133}, and with first-forbidden unique transitions for K^{40} (compare Ca^{41} and Tl^{204} in Table I). In the case of 9.5-year Ba^{133} the decay scheme is complex and the contribution of internal conversion uncertain. Langevin[20] suggests that the decay might proceed entirely by L-capture which would operate to reduce the comparative half-life bringing it into better agreement.

The large discrepancies indicated in Table II (a) for Pd^{103} and Ge^{71} may not be real; in the case of Pd^{103},

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TABLE I. Theoretical electron-capture ratios for cases where the transition energy is known and decay scheme is simple.[a]

1 Z	A	2 Class	3 Half-life	Reference[b]	4 E_{EC} and Method (Mev)	Reference[b]	5 Transition	% EC	Reference[b]	6 Log ft	7 Theoretical capture ratios P_{L_I}/P_K	$P_{L_{II}}/P_K$	$P_{L_{III}}/P_K$	P_L/P_K	8 Remarks
4 Be	7	A	52.9±0.2 days	S4	0.864±0.002 (p,n) 0.386±0.002 cc	R3 T1	$g(p_{3/2}-p_{3/2})$ $e(p_{3/2}-p_{1/2})$	88 12	D3 D3	3.33 3.48	0.04 0.04	<0.0001 <0.0001		0.04 0.04	
11 Na	22	A	2.6_0 yr	L5	1.564±0.005 β^+	M1	$e(3+-2+)$	10	S5	7.4	0.065	<0.0001		0.065	
18 A	37	A	34.1±0.3 days	W1	0.815±0.005 (p,n), IB	R3, E1, A1	$g(d_{3/2}-d_{3/2})$	100		5.1	0.0815	0.0002		0.082	See Table II (a)
19 K	40	B	1.3×10^9 yr	H1	0.048±0.021 cc	M11, J3, G3, F2	$e(4-$ to $2+)$	11	H1	12.5	0.107	0.0003	0.107	0.213	See Table II (a)
20 Ca	41	C	$\sim1.2\times10^5$ yr	B7	0.440±0.020 (p,n)	R3	$g(f_{7/2}-d_{3/2})$	100		10.8	0.087	0.0003	0.0013	0.089	
23 V	49	A	334±20 days	L10	0.617±0.020 (p,n), IB	T2, H3	$g(f_{7/2}-f_{7/2})$	100		6.4	0.088	0.0003		0.088	
24 Cr	51	B	27.8±0.3 days	L9	0.745±0.006 (p,n) 0.422±0.007 cc	R3 B10	$g(f_{7/2}-f_{7/2})$ $e(f_{7/2}-f_{5/2})$	90.2 9.8	B10 B10	5.4 5.8	0.088 0.088	0.0003 0.0003		0.088 0.088	
25 Mn	51	B	45.2 min	K2	3.18±0.05 β^+	K2	$g(f_{5/2,7/2}-f_{7/2})$	~3	F1	5.4	0.088	0.0004		0.088	
25 Mn	52	B	5.72±0.02 days	B1	1.598±0.015 β^+	P3	$e_3(6+-6+)$	65	G2	5.6	0.088	0.00045		0.088	
25 Mn	54	B	324±11 days	S7	0.368±0.007 (p,n), cc	L8, D1	$e(2,3+-2+)$	~100		5.9	0.090	0.00045		0.090	
26 Fe	55	A	2.94±0.03 yr	B9	0.218±0.005 IB, (p,n)	P1, K3, E1	$g(p_{3/2}-f_{5/2})$	100		5.9	0.096	0.0005		0.097	
27 Co	58	B	72±4 days	L6	1.494±0.006 β^+	C1	$e(2+-2+)$	85	G2	6.59	0.092	0.0005		0.092	
28 Ni	59	A	8×10^5 yr	W3	1.065±0.030 IB, (p,n)	M8, E1	$g(p_{3/2}-f_{7/2})$	100		11.9	0.091	0.0006	0.0004	0.092	See text
32 Ge	71	B C	11.4±0.1 days ~14 days	M7 L2	0.235±0.012 IB, (p,n) 0.170±0.020 IB	S1, T2, L2 L2	$g(p_{1/2}-p_{3/2})$?	100 ?		4.3 (4.1)	0.105 0.106	0.0006 0.0006		0.106 0.107	See Table II (a)
36 Kr	79	B	34.5±0.2 hr	R1	1.617±0.006 β^+	B2	$e(g_{7/2}-p_{3/2})$	~90	B2	5.41	0.100	0.001		0.101	See Table II (a)
39 Y	88	B	105 days	D4	0.960±0.020 β^+, cc	H1	$e_2(4-$ to $3-)$	~100		6.9	0.104	0.001		0.105	
40 Zr	89	B	79.3 hr	S6	1.927±0.010 β^+	H1	$e(g_{9/2}-g_{9/2})$	75	G1	6.1	0.104	0.001		0.105	
42 Mo	91	B	15.7±0.3 min	A3	4.7±0.2 β^+	D5	$g(g_{9/2}-g_{9/2})$	~5	F1	5.8	0.105	0.002		0.107	
42 Mo	93	C	>2 yr	H1	0.49±0.04 (p,n)	P2	$g(_{5/2,7/2}-g_{9/2})$	100		>7.3	0.114	0.002		0.116	P_L/P_K calculated for allowed transition
46 Pd	103	B	17.0 days	M6	$0.494^{+0.027}_{-0.012}$ IB	R4	$e(_{5/2,7/2}-g_{7/2})$	100		5.7	0.118	0.002		0.120	See Table II (a)
55 Cs	131	A	9.9±0.1 days	H1	0.352±0.010 IB	S2, R5	$g(d_{5/2}-d_{3/2})$	100		5.5	0.156	0.004		0.160	See Table III
59 Pr	140	B	3.4 min	D2	3.25±0.02 β^+	B8	$g(1+-0+)$	~50	F1	4.3	0.126	0.004		0.130	
61 Pm	145	C	16 days	L7	1.47±0.05 β^+	L7	$g(_{5/2,7/2+}-f_{7/2})$	~99	F1	7.2	0.133	0.005		0.138	P_L/P_K calculated for allowed transition
81 Tl	204	B	4.0±0.1 yr	H2	0.335±0.010 IB	M4	$g(2-$ to $0+)$	~2.5	M4	10.3	0.394	0.0316	0.098	0.524	See Table III
91 Pa	229	C	1.5 days	H1	0.37 cc	S3	$g(p_{3/2}-?)$	>99	M9	6.8	0.31	0.034		0.34	P_L/P_K calculated for allowed transition
93 Np	233	C	35 min	M2	1.09 cc	S3	$g(_{5/2}?-d_{5/2})$	>99	M2	5.2	0.206	0.025		0.23	P_L/P_K calculated for allowed transition to ground state
93 Np	235	C	410 days	J1	0.170 cc	S3	$g(f_{5/2}-d_{5/2})$	100?	J1	7.2	1.265	0.152		1.42	P_L/P_K calculated for allowed transition to ground state
94 Pu	234	B	8.5 hr	P4	0.210 cc	S3	$g(0+$ to $0,1-)$	96	H1	4.47	0.728	0.091		0.82	P_L/P_K calculated for allowed transition to ground state

Note.—Footnotes for Table I on opposite page.

because of the contribution made by the internal conversion of the 40-kev isomeric state in the daughter; and in the case of Ge^{71}, because of reported isomerism.[21]

The electron-capture transition energy used in computing the P_L/P_K ratio in Table I for K^{40} was deduced from a closed cycle[22] consisting of the beta-decay energy of K^{40}, 1.325 ± 0.015 Mev,[23] the $Ca^{40}-A^{40}$ mass difference, 0.199 ± 0.015 mmu$=0.185\pm0.014$ Mev,[24] and the energy of the first excited state of A^{40}, 1.459 ± 0.007

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[a] Column 2 is an estimate of the relative reliability of the experimental information; transitions in the A category are well established, while those in the C category remain to be confirmed. This classification follows that of Major and Biedenharn (see reference 1).

Column 4 gives the energy of the transition indicated in column 5 and the method by which it was determined. The following nomenclature is used: (p,n), reaction threshold measurement; β^+, positron decay energy; IB, inner bremsstrahlung endpoint plus K-electron binding energy; cc, closed cycle calculation, which may include energies of alpha particles, beta decay, or gamma rays, and, in the case of K^{40}, mass spectrometer data.

Column 5 lists the type of transition involved and the branching ratio for electron-capture. g and e refer to ground-state and first-excited state transitions, respectively (e_i means ith excited state).

Column 7 gives the theoretical capture ratios for L subshells and the total capture ratio. $P_{L_{III}}/PK$ is given only for cases where $|\Delta J|\geq 2$ (see text).

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W3. H. W. Wilson, Phys. Rev. **82**, 548 (1951).

TABLE II (a). Experimental determinations of orbital electron capture ratio for cases where transition energy is known.

Z	A	Experimental P_L/P_K	Ref.[a]	Theoretical P_L/P_K (Table I)	Remarks
18 A	37	0.08 to 0.09	P5	0.082	
19 K	40	1.35±0.23	H4	$0.21^{+0.33}_{-0.06}$	See Table II (b).
32 Ge	71	0.19±0.03	L3	0.106	Experimental results recalc. using $\omega_K=0.50$.[b] Decay scheme in question (L2).
36 Kr	79	0.096±0.030	R2, L1	0.101	Experimental results recalc. using $\omega_K=0.63$.[b]
46 Pd	103	0.79±0.15	A2	0.120	Conversion of the 40 kev *IT* daughter in question.

[a] See list of references in footnote b of Table I. [b] See reference 10 in text.

TABLE II (b). Experimental determinations of orbital electron capture ratios from which transition energy is calculated.

Z	A	Half-life	Ref.[a]	Experimental P_L/P_K	Ref.	Transition	Calculated Energy (Mev)	Log *ft*	Remarks
19 K	40	1.3×10^9 yr	H1	1.35±0.023	H4	$e(4-$ to $2-)$	0.019±0.002	10.3	Consistent with ($\|\Delta J\|=2$ yes)
48 Cd	109	470 days	G4	0.28±0.03	M5	$e(5/2,7/2-g_{7/2})$	$0.062^{+0.009}_{-0.005}$	4.1	See Table III
53 I	125	60.0 days	F4	0.23±0.03	M5	$e(d_{5/2}-d_{3/2})$	$0.100^{+0.020}_{-0.012}$	4.9_3	
56 Ba	133	~9.5 yr	H1	≥9	L4	$e_3(s_{\frac{1}{2}}-s_{\frac{1}{2}})$	≤0.040	5.6	Decay scheme complex and controversial (L4, H6).

[a] See list of references in footnote b of Table I.

Mev.[25] For this energy, 51±22 Kev, the capture ratio for second-forbidden unique ($|\Delta J|=2$, yes) transition lies between 0.54 and 0.15, $\left(0.21^{+0.33}_{-0.06}\right)$. Heintze[26] has measured the specific activity of potassium for the emission of argon K x-rays; after correcting for the K-fluorescence yield of argon, he finds that the specific activity for K-capture (K x-rays) is less than the specific activity for electron-capture (gamma rays). The excess of gamma rays is attributed to L-capture giving an experimental ratio of $P_L/P_K=1.35\pm0.23$. From this one may deduce that the transition energy is 19±2 kev. As mentioned previously, the comparative half-life for this energy is very similar to those for similar transitions in other nuclides.

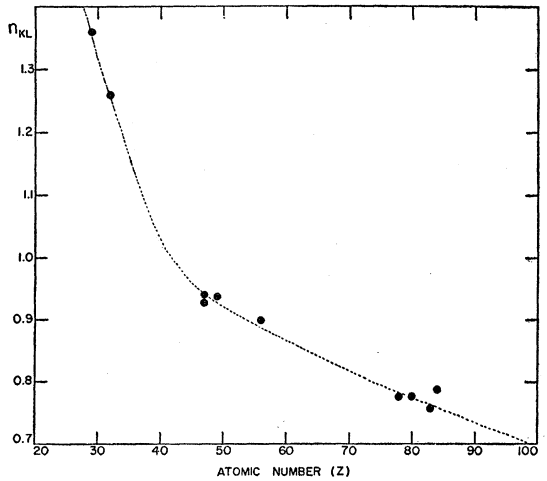

FIG. 1. $n_{KL}=\omega_{KL}+a_{KL}$. The number of L-shell vacancies produced in the filling of a K-shell vacancy.

[25] M. L. Good, Phys. Rev. **81**, 891 (1951).
[26] J. Heintze, Z. Naturforsch. **9a**, 469 (1954).

X-RAY INTENSITY RATIOS AND MEAN L-FLUORESCENCE YIELDS

The relative intensities of K and L x-rays have been measured for several electron-capturers for which the capture ratio is known, either from experiment or by computation from the transition energy. The x-ray intensity ratio I_L/I_K is related to the capture ratio P_L/P_K by the expression

$$I_L/I_K=\left(\frac{P_L}{P_K}+n_{KL}\right)\frac{\bar{\omega}_L}{\omega_K}, \qquad (2)$$

where $\omega_K=K$-fluorescence yield; $\bar{\omega}_L=$mean L-fluorescence yield,* and $n_{KL}=$number of L-shell vacancies

* For completeness, because of common usage, the following quantities are also defined: $\omega_{L\text{I}}$, $\omega_{L\text{II}}$, $\omega_{L\text{III}}$=fluorescence yields for the L_I, L_II, L_III subshells, respectively. (The mean L-fluorescence yield defined above is an average of these partial yields weighted in an essentially unknown manner. In general, these subshell yields differ from one another.) ω_{LK}=fluorescence yield from secondary L-vacancies (i.e., arising from K-vacancies). ω_{LL}=fluorescence yield from primary L-vacancies (i.e., arising from nuclear processes, L-capture or internal conversion in the L-shell).

TABLE III. Experimental determinations of x-ray intensities in electron capture from which mean L-fluorescence yields are calculated.

Z	A	I_L/I_K	Ref.[a]	P_L/P_K	Ref.	n_{KL}	ω_K	Calcd. $\bar{\omega}_L$	Remarks
48 Cd	109	0.0505±0.0003	B3	0.28±0.03	M5	0.93	0.83	0.029±0.003	Corrected for internal conversion of 87 kev IT (A4, B11)
55 Cs	131	0.121±0.008	F3	0.160	Table I	0.89	0.87	0.10±0.01	
81 Tl	204	0.33±0.05	J2	0.524	Table I	0.78	0.95	0.24±0.05	
83 Bi	210							0.30±0.05	See reference 29
								0.38±0.49	See reference 29a

[a] See list of references in footnote b of Table I.

TABLE IV. Experimental determinations of x-ray intensity ratios in electron capture with capture ratios calculated using estimated $\bar{\omega}_L$ (Fig. 2) and transition energies deduced from capture ratios.

Z	A	Half-life (days)	Ref.	Transition	I_L/I_K	Ref.	ω_K	(Est.) $\bar{\omega}_L$	n_{KL}	P_L/P_K	E_{EC} (Mev)	Log ft	Remarks
$_{74}W^{181}$		140	W2	$g(\frac{1}{2}, 3/2-g_{7/2})$	0.39±0.01	B5	0.93	0.22±0.05	0.81	0.83±0.35	0.110^{+27}_{-12}	6.4	
								0.15		1.54	0.092	6.4	See B5
$_{76}Os^{185}$		97	K1	$e_2(\frac{1}{2}-$ to $d_{5/2})$	0.40±0.05	M10	0.94	0.23±0.05	0.80	0.8±0.4	<0.07	5.8	[b]
$_{79}Au^{195}$		180	S8	$e_2(3/2-f_{5/2})$	0.408±0.016	B4	0.95		0.78	0.58±0.14	0.148^{+28}_{-12}	6.9	[c]
$_{81}Tl^{202}$		12.5	M3	$e(2-$ to $2+)$	0.39	H5	0.95	0.25±0.05	0.78	0.70±0.10	0.15±0.01	5.33	[c]

[a] See list of references in footnote b of Table I. [b] Transition to upper state is pure L-capture. [c] Calculated for allowed transition.

produced in the filling of a K-shell vacancy,

$$n_{KL}=\omega_K\left(\frac{I_{K\alpha}}{I_K}\right)+a_K\frac{2(K-LL)+(K-LX)}{\Sigma \text{ Augers}}$$

(this sum might be broken down further into subshell components); $I_{K\alpha}/I_K$=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 ratio $(I_{K\alpha}/I_K)$ has been computed from data given in Compton and Allison,[27] K-fluorescence yields are given by Broyles, Thomas, and Haynes,[10] and partial Auger yields have been computed from the literature.[28] The values of n_{KL} computed for $Z=29$, 32, 47, 49, 56, 78, 80, 83, and 84 are plotted in Fig. 1 and a smooth curve has been drawn. Appropriate values of n_{KL} are entered in Table III together with observed intensity ratios, capture ratios, and K-fluorescenc-yields. Mean L-fluorescence yields $\bar{\omega}_L$ have been computed and also entered in Table III. The value of $\bar{\omega}_L$ found by Damon and Edwards[29] for the conversion of the 47.5-Kev gamma ray of RaD(Pb^{210}) is also entered.[29a]

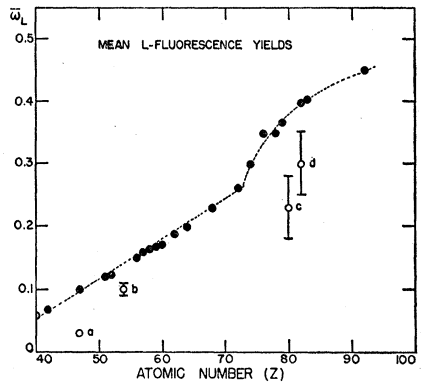

FIG. 2. Mean L-fluorescence yields. The full circles are values obtained by H. Lay, Z. Phys. **91**, 533 (1934), using a photographic technique and x-ray excitation. Points a–d have been deduced from nuclear data as follows: a Cd^{109} EC—Bertolini, Bisi, Lazzarini, and Zappa, Nuovo cimento **11**(9), 539 (1954). b Cs^{131} EC—R. W. Fink and B. L. Robinson, Phys. Rev. **98**, 1293 (1955). c Tl^{204} EC—H. Jaffe, UCRL-2537 (1954) (unpublished). d Pb^{210} (RaD)—Internal Conversion. P. R. Damon and R. R. Edwards, Phys. Rev. **95**, 1698 (1954). (See reference 29a.)

[27] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), second edition.

[28] J. F. Perkins and S. K. Haynes, Phys. Rev. **92**, 687 (1953) (Cu). M. Ference, Phys. Rev. **51**, 727 (1937) (Ge). Huber, Humbel, Schneider, and de Shalit, Helv. Phys. Acta. **25**, 3 (1952) (Ag, Cd). Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953) (In, Ba, Hg). Steffen, Huber, and Humbel, Helv. Phys. Acta. **22**, 167 (1949) (Pt). C. D. Ellis, Proc. Roy. Soc. (London) **A139**, 336 (1933) (Bi). R. W. Hoff, University of California Radiation Laboratory Report, UCRL-2325 (1953), unpublished thesis, (Po).

[29] P. E. Damon and R. R. Edwards, Phys. Rev. **95**, 1698 (1954).

[29a] *Note added in proof.*—Ross, Cochran, Hughes, and Feather [Proc. Phys. Soc. (London) **A68**, 612 (1955)], have examined critically all existing experimental data on the fluorescence yields of the L-levels of bismuth excited by internal conversion (RaD) and by soft x-rays. They conclude that for bismuth the value of $\bar{\omega}_L$ "probably lies between 0.38 and 0.49." They also have shown that most of the transitions from the L_I subshell (for which the

In Fig. 2 the mean L-fluorescence yields are plotted against Z. The results of Lay[30] obtained by a photographic method for fluorescent excitation are also shown. (A break is indicated at $Z\sim73$; beyond this point Coster-Krönig transitions contribute. A Coster-Krönig transition is an Auger transition of the type ($L_{\mathrm{I}}-L_{\mathrm{III}}M_{\mathrm{IV,V}}$) which shifts L_{I} vacancies to the L_{III} shell for which the partial fluorescence yield is greater.)

If a systematic error in the work of Lay is excluded, Fig. 2 implies that the weighting of the subshell contributions to the mean L-fluorescence yield is fundamentally different for different methods of excitation, and that $\bar{\omega}_L$ lies lower for nuclear excitation than for x-ray excitation (except see reference 29a).

SOME TRANSITION ENERGIES

The intensities of the L and K x-rays have been reported for several electron-capturing nuclides. These are given in Table IV together with K-fluorescence yields, n_{KL}, and mean L-fluorescence yields estimated from Fig. 2. Capture ratios, transition energies, and comparative half-lives have been computed for these cases and entered in the table. The four cases are discussed individually below.

(a) The nuclide W^{181} has been reported[31] to exhibit pure electron-capture with no gamma rays; from x-ray intensity measurements the capture energy is interpreted to be 92 kev (corresponding to $\bar{\omega}_L=0.15$). From Fig. 2 one estimates $\bar{\omega}_L=0.22\pm0.05$ leading to a capture ratio of 0.83, and an energy of 110 kev. The previously reported weak gamma rays[32] at 136.5 and 152.5 kev are believed to be the result of impurities.

(b) The K and L x-rays of Os^{185} have been observed by Miller and Wilkinson.[33] Two gamma rays at 648 and 878 kev are associated with the decay and were found not to be in coincidence. Weak conversion electrons corresponding to the "stopover" transition have been reported,[34] but the gamma ray has not been observed. On the assumption of little or no "stopover" transition, it is impossible to satisfy both the observed gamma-ray intensity ratio and the x-ray intensity ratio, I_L/I_K, except by assuming that the transition to the second excited state is pure L-capture, thus setting an upper limit of about 70 kev for the energy of this transition.

(c) By means of a subtle interpretation of the pulse-height spectrum of Au^{195} in a proportional counter, Bisi and Zappa[35] concluded that the capture ratio for transitions to the second excited state is 0.58, from which a transition energy of 148 Kev is deduced.

(d) In connection with a study of Pb^{202}, Huizenga and Stevens[36] have reported for Tl^{202} an x-ray intensity ratio $I_K/I_L=2.6$. Neglecting the weak conversion of the 435-kev gamma ray (about 2.5%[37]), and the possibility of ground-state transitions, one deduces a capture energy of about 150 Kev for this nuclide.

SUMMARY AND CONCLUSIONS

The theory of orbital capture appears to be in relatively good agreement with experiment in those cases where there is no question about the decay scheme (2 cases). The theory has been applied to the determination of: (1) transition energies where P_L/P_K is known (4 cases); (2) mean L-fluorescence yields where both the transition energy and x-ray intensities are known (3 cases); and (3) transition energies where x-ray intensities are known and mean L-fluorescence yields estimated (4 cases). Theoretical capture ratios have been computed and tabulated for nuclides whose transition energies are known and decay schemes simple (28 cases).

It is clear from the lack of good experimental cases that more refined orbital capture data are required. In this respect the internal source technique should be more widely applied because the interpretation does not require a knowledge of L-fluorescence yields.

It is indicated that mean L-fluorescence yields depend upon the method of excitation; various modes of excitation result in different relative populations of L-shell vacancies. The relative contribution of the L-subshells to the mean L-fluorescence yields can be determined only by studying the L x-ray fine structure with instruments of sufficiently high resolution (e.g., bent crystal spectrometer).

Note added in proof.—Recently there has been much discussion[38] concerning the charge distribution on the Cl^{37} recoil ions from electron capture in A^{37}, and the relation to the L/K capture ratio via theoretical Auger transition probabilities. The charge distributions observed by Kofoed-Hansen and Snell and Pleasonton do not appear to be consistent with the 8 to 9% L/K capture ratio in A^{37} (reference 7).

We feel that the experimental results are probably valid and that the difficulty lies in the use of an inadequate theory of Auger transition probabilities. It has been suggested by Daudel [J. phys. radium **16**, 515 (1955)] that the neglect in the present Auger theory of the correlations among the positions of the electrons may lead to large errors.

fluorescence yield is small) are of the Coster-Kronig type, and that most of the L-fluorescence x-rays arise from the L_{III} subshell. Consequently, in this region $\bar{\omega}_L\sim\bar{\omega}_{L_{\mathrm{III}}}$, regardless of the primary excitation. The value for Tl^{204} (point c, Fig. 2) was obtained by neglecting Coster-Kronig transitions and it is probably much too low. On the other hand, Coster-Kronig transitions do not occur for $Z<73$ and the values for points a and b are undoubtedly correct.

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[31] Bisi, Terrani, and Zappa, Nuovo cimento **1(10)**, 651 (1955).
[32] Cork, Nester, Le Blanc, and Brice, Phys. Rev. **92**, 119 (1953).
[33] M. M. Miller and R. G. Wilkinson, Phys. Rev. **83**, 1050 (1951).
[34] J. B. Swan and R. D. Hill, Phys. Rev. **88**, 831 (1952).
[35] A. Bisi and L. Zappa, Nuovo cimento **12**, 539 (1954).
[36] J. R. Huizenga and C. M. Stevens, Phys. Rev. **96**, 548 (1954).
[37] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).
[38] O. Kofoed-Hansen, Phys. Rev. **96**, 1045 (1954); A. H. Snell and F. Pleasonton, Phys. Rev. **97**, 246 (1955); R. A. Rubenstein and J. N. Snyder, Phys. Rev. **99**, 189 (1955); P. Radvanyi, J. phys. radium **16**, 509 (1955); and A. Winther, J. phys. radium **16**, 562 (1955).