Exchange and Overlap Effects in Electron Capture and in Related Phenomena*

JOHN N. BAHCALL
California Institute of Technology, Pasadena, California
(Received 20 May 1963)

Theoretical expressions are presented for L/K electron capture ratios and electron capture-positron emission ratios that include the effects of electron exchange and imperfect atomic overlap. The role of electron exchange and imperfect atomic overlap in the determination of fluorescence yields is also discussed. In order to facilitate experimental comparisons, numerical values are presented for the exchange and overlap corrections to L/K ratios and electrons capture-positron emission ratios; these corrections were calculated with the analytic Hartree-Fock wave functions of Watson and Freeman. The corrected L/K ratios are in good agreement with the precisely measured L/K ratios, showing that overlap and exchange corrections explain the systematic disagreement between the predictions of the usual electron capture theory and experiment.

I. INTRODUCTION

THE usual theory of allowed electron capture yields a simple expression[1,2] for the ratio of the capture probability of an L electron to the capture probability of a K electron. This theoretical expression, which was first obtained by Marshak[3,4], appeared for many years to be consistent with measurements of L/K electron capture ratios. However, after Drever *et al.*[5] developed an essentially wall-less multiwire proportional counter that could be used with gaseous sources, a number of experimental groups built and used multiwire counters to obtain precise experimental values for L/K capture ratios. The results of these precise measurements disagreed systematically[6] with the predictions of the usual theory of allowed electron capture, even when screening, nuclear size, and relativistic effects were taken into account.[1]

In the first papers of this series, I and II,[7] the present writer developed the suggestion of Benoist-Gueutal[8] and Daudel[9] that atomic variables be included in the description of a radioactive system. For electron emission and electron capture, corrections were found to the usual beta-decay theory; these corrections arise from imperfect atomic overlap and electron exchange. We estimated, in I, some one-electron exchange integrals and showed that exchange corrections eliminated the systematic disagreement between theory and experiment for L/K ratios. In II we presented the formal theory of allowed electron and positron emission and electron capture that is appropriate when atomic variables are included in the description of the radioactive system. In III[10] the effect of imperfect atomic overlap and electron exchange on M/L capture ratios was calculated using analytic Hartree-Fock wave functions obtained by Watson and Freeman.[11,12] The M_{I} and L_{I} electron-capture amplitudes were also presented in a form that clarifies the physical origin of overlap and exchange corrections.

In Sec. II of this paper, theoretical expressions are presented for L/K capture ratios and electron-capture–positron-emission ratios that include both overlap and exchange effects. Overlap effects are usually less important numerically than exchange effects, and, hence, we frequently describe corrections that are due to both imperfect atomic overlap and electron exchange by the phrase "exchange corrections." The exchange-corrected L/K ratio given in Sec. II reduces, within a few percent, to the formula previously used in I and II. We also discuss in Sec. II the effect of imperfect overlap and electron exchange on the determination of fluorescence yields of isotopes that decay by electron capture. We present in Sec. III numerical values of typical one-electron overlap and exchange integrals and also tabulate numerical values of exchange corrections for electron-capture–positron-emission ratios and L/K ratios. All numerical results were obtained with the analytic Hartree-Fock wave functions of Watson and Freeman.[11,12] In Sec. IV, the nine precisely measured L/K capture ratios are compared with the predictions of the usual theory and the predictions of the exchange-corrected theory. We conclude that overlap and exchange corrections remove the systematic discrepancy between the usual theory and experiment and also give rise to predictions that agree well with measurements of individual capture ratios. Only Zn^{65} has a measured[13]

* Supported in part by the Office of Naval Research and in part by the National Aeronautics and Space Administration.

[1] H. Brysk and M. E. Rose, Rev. Mod. Phys. **30**, 1169 (1958).

[2] M. A. Preston, *Physics of the Nucleus* (Addison-Wesley Publishing Company, Inc., Palo Alto, California, 1962).

[3] R. E. Marshak, Phys. Rev. **61**, 431 (1942).

[4] An interesting historical discussion of electron capture is continued in the review article by R. Bouchez and P. Depommier, Rept. Progr. Phys. **23**, 395 (1960).

[5] R. W. P. Drever, A. Moljk, and S. C. Curran, Nucl. Instr. **1**, 41 (1957); R. W. P. Drever and A. Moljk, Phil. Mag. **2**, 427 (1957).

[6] B. L. Robinson and R. W. Fink, Rev. Mod. Phys. **32**, 117 (1960).

[7] J. N. Bahcall, Phys. Rev. Letters **9**, 500 (1962), hereafter called I; J. N. Bahcall, Phys. Rev. **129**, 2683 (1963), hereafter called II.

[8] P. Benoist-Gueutal, Compt. Rend. **230**, 624 (1950); Ann. Phys. (N. Y.) **8**, 593 (1953).

[9] S. Odiot and R. Daudel, J. Phys. Radium **17**, 60 (1956).

[10] J. N. Bahcall, Phys. Rev. **131**, 1756 (1963), hereafter called III.

[11] R. E. Watson, Technical Report Number **12**, 1959, Solid-State and Molecular Theory Group, MIT, Cambridge, Massachusetts; R. E. Watson, Phys. Rev. **118**, 1036 (1960).

[12] R. E. Watson and A. J. Freeman, Phys. Rev. **123**, 521 (1961); *ibid.* **124**, 1117 (1961).

[13] A. G. Santos-Ocampo and D. C. Conway, Phys. Rev. **128**, 258 (1962).

L/K capture ratio that differs from the exchange-corrected ratio by more than the usual systematic uncertainties of about 4%. It would be useful to repeat the Zn^{65} measurement with greater precision in order to clarify this discrepancy.

II. THEORETICAL EXPRESSIONS

A. L/K Capture Ratios

The theoretical $L_{\rm I}$ to K capture ratio obtained by generalizing[7] the usual theory of allowed electron capture to include atomic variables in the initial and final states of the radioactive system is

$$\frac{\lambda_{L\rm I}}{\lambda_K}=\left|\frac{q(2s')f(2s')}{q(1s')f(1s')}\right|^2, \tag{1}$$

where $q(2s')$ and $q(1s')$ are the neutrino energies[14] for $L_{\rm I}$ and K capture, respectively, and $f(2s')$ and $f(1s')$ are the amplitudes for the production of a hole in the final $2s'$ or $1s'$ shell. The amplitudes are

$$f(2s')=\langle 1s'|1s\rangle\langle 3s'|3s\rangle\psi_{2s}(0) -\langle 1s'|2s\rangle\langle 3s'|3s\rangle\psi_{1s}(0) -\langle 3s'|2s\rangle\langle 1s'|1s\rangle\psi_{3s}(0), \tag{2a}$$

$$f(1s')=\langle 2s'|2s\rangle\langle 3s'|3s\rangle\psi_{1s}(0) -\langle 2s'|1s\rangle\langle 3s'|3s\rangle\psi_{2s}(0) -\langle 3s'|1s\rangle\langle 2s'|2s\rangle\psi_{3s}(0). \tag{2b}$$

In Eqs. (2), we have omitted constants that are the same for both amplitudes. The atomic matrix elements $\langle ns'|ms\rangle$ represent the overlap of the ms wave function of an electron in the initial atom with the ns' wave function of an electron in the final atom. The $\psi_{ms}(0)$ are one-electron wave functions, evaluated at the nucleus, of electrons in the initial atom.

An $L_{\rm I}$ capture can occur in three important ways that are experimentally indistinguishable: (a) annihilation of a $2s$ electron with the $1s$ and $3s$ electrons appearing in the final $1s'$ and $3s'$ states; (b) annihilation of a $1s$ electron with a $2s$ electron jumping into the final $1s'$ shell; (c) annihilation of a $3s$ electron with a $2s$ electron jumping into the final $3s'$ shell. The three processes (a)–(c) correspond to the three terms in the $L_{\rm I}$ capture amplitude, $f(2s')$; the usual theory[1,2] only considers process (a). The minus signs in the amplitude $f(2s')$ occur because (b) and (c) differ from (a) only in the exchange of a single electron. The amplitude for K capture, $f(1s')$, can be interpreted in a similar way.

The assumptions that underlie the derivation of Eqs. (2) were discussed in II and III.

Equations (1) and (2) can be written in the form

$$\frac{\lambda^{L_I}}{\lambda_K}=\left(\frac{\lambda^{L_I}}{\lambda_K}\right)^0 X^{L/K}, \tag{3a}$$

where

$$\left(\frac{\lambda^{L_I}}{\lambda_K}\right)^0=\left|\frac{q(2s')\psi_{2s}(0)}{q(1s')\psi_{1s}(0)}\right|^2 \tag{3b}$$

is the usual[1,2] $L_{\rm I}$ to K capture ratio and

$$X^{L/K}=\left|\frac{\langle 1s'|1s\rangle}{\langle 2s'|2s\rangle}\right|^2\left|\frac{1-\dfrac{\langle 1s'|2s\rangle}{\langle 1s'|1s\rangle}\dfrac{\psi_{1s}(0)}{\psi_{2s}(0)}-\dfrac{\langle 3s'|2s\rangle}{\langle 3s'|3s\rangle}\dfrac{\psi_{3s}(0)}{\psi_{2s}(0)}}{1-\dfrac{\langle 2s'|1s\rangle}{\langle 2s'|2s\rangle}\dfrac{\psi_{2s}(0)}{\psi_{1s}(0)}-\dfrac{\langle 3s'|1s\rangle}{\langle 3s'|3s\rangle}\dfrac{\psi_{3s}(0)}{\psi_{1s}(0)}}\right|^2 \tag{3c}$$

is the exchange correction[15] to the usual L to K capture ratio. Numerical values for $(\lambda_{L_I}/\lambda_K)^0$ have been tabulated by several authors[1,16]; numerical values for $X^{L/K}$ are given in Table III of Sec. III of this paper.

In order to compare Eqs. (3) with experimentally observed L/K capture ratios, the small probability for decay by capture of a $p_{1/2}$ electron should be taken into account. Thus, we write for the total L/K capture ratio:

$$\frac{\lambda_L}{\lambda_K}=\left(\frac{\lambda_{L_I}}{\lambda_K}\right)^0\left[X^{L/K}+\frac{L_{\rm II}}{L_{\rm I}}\right]. \tag{4}$$

The quantity $L_{\rm II}/L_{\rm I}$ has been calculated by several authors.[1,16,17] For Z less than 40, $L_{\rm II}/L_{\rm I}$ is well represented[1,16,17] by the Coulomb expression

$$L_{\rm II}/L_{\rm I}\cong\tfrac{3}{16}(\alpha Z_{\rm eff})^3, \tag{5}$$

where $Z_{\rm eff}$ is the screened nuclear charge.

B. Electron-Capture to Positron-Emission Ratios

The arguments presented in II can also be used to predict the effect of exchange on electron-capture to positron-emission ratios. We find, for example, that

$$\frac{\lambda_K}{\lambda_{\beta^+}}=\left(\frac{\lambda_K}{\lambda_{\beta^+}}\right)^0 B_K, \tag{6a}$$

where $(\lambda_K/\lambda_{\beta^+})^0$ is the usual[1,2] K-capture to positron-emission ratio,

$$\left(\frac{\lambda_K}{\lambda_{\beta^+}}\right)^0=\frac{2\pi^2[q(1s')]^2|\psi_{1s}(0)|^2}{f(W_0,-Z)}, \tag{6b}$$

[14] It is shown in II that the binding energy of the $2s'$ electron [$1s'$ electron] in the *final* atom should be used when calculating $q(2s')[q(1s')]$.

[15] Expression (3c) actually includes both overlap and exchange effects, but the overlap integrals are all very nearly equal to unity (see Table I, Sec. III). Hence $X^{L/K}$ can be written, as in I and II, in an approximate form that does not contain any overlap integrals. Thus, we use the simple phrase "exchange correction" in referring to $X^{L/K}$ and similar corrections.

[16] I. M. Band, L. N. Zyrianova, and Iu. P. Suslov, Izv. Akad. Nauk SSSR Ser. Fiz. **22**, 952 (1958).

[17] A. H. Wapstra, G. J. Nijgh, and R. L. Lieshout, *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959).

and

$$B_K=\left|\frac{f(1s')}{\psi_{1s}(0)}\right|^2 \tag{6c}$$

is the appropriate exchange correction. Numerical values for B_K are given in Table III of Sec. III.

Expressions similar to Eqs. (6) were given in III for L to β^+-emission ratios.

For M to β^+-emission ratios, we find

$$\frac{\lambda_{M_I}}{\lambda_{\beta^+}}=(\lambda_{M_I}/\lambda_{\beta^+})^0 B_{M_I}, \tag{7a}$$

where

$$B_{M_I}=\left|\frac{f(3s')}{\psi_{3s}(0)}\right|^2 \tag{7b}$$

and

$$\frac{\lambda_M}{\lambda_{\beta^+}}=\left(\frac{\lambda_{M_I}}{\lambda_{\beta^+}}\right)^0\left[B_{M_I}+\frac{M_{\rm II}}{M_{\rm I}}\right]. \tag{7c}$$

In Eq. (7b), $f(3s')$ is the $M_{\rm I}$ capture amplitude which was defined explicitly in III. Numerical values for B_{M_I} are given in Table IV of Sec. III. The quantity $M_{\rm II}/M_{\rm I}$, which represents the small probability for capture of a $p_{1/2}$ electron, is approximately equal to the tabulated[1,16,17] quantity $L_{\rm II}/L_{\rm I}$.

C. Fluorescence Yields

If one knows what fraction of the total number of electron captures by a radioactive isotope produce a vacancy in a given electron shell, one can determine the fluorescence yield for this shell by measuring the ratio of the appropriate x-ray production rate to the total electron-capture rate. Taylor and Merritt[18] have recently applied this method to the determination of the K-fluorescence yields of Cr^{51}, Mn^{54}, and Zn^{65}.

In order to predict theoretically what fraction of the total number of electron captures produce a vacancy in, for example, the K shell, one must take account of exchange effects. Using the arguments given in II, we find

$$\frac{\lambda_K}{\lambda_{\rm total}}=\left(\frac{\lambda_K}{\lambda_{\rm total}}\right)^0 B_K, \tag{8a}$$

where

$$\left(\frac{\lambda_K}{\lambda_{\rm total}}\right)^0=\frac{2|\psi_{1s}(0)q(1s')|^2}{\Sigma_n|\psi_{ns}(0)q(ns')|^2} \tag{8b}$$

is the usual[1,2] theoretical ratio of K-capture probability to total-capture probability and B_K is the exchange correction defined by Eq. (6c). Similar expressions, involving B_{L_I} and B_{M_I}, obtain for the theoretical ratios of $L_{\rm I}$ and $M_{\rm I}$ capture ratios to total-capture ratios.

III. NUMERICAL VALUES

In Table I we list some typical values of one-electron overlap integrals; these integrals were calculated with the analytic Hartree-Fock wave functions of Watson and Freeman.[11,12] The fact that these overlap integrals are all very nearly equal to unity justifies the assumption that the core s electrons are inert if they are not captured; this assumption was used in II and III to derive the form of the exchange corrections.

TABLE I. One-electron overlap integrals. All wave functions are assumed to have the same phase at the origin.

$Z_{\rm final}$	$\langle 1s'\|1s\rangle$	$\langle 2s'\|2s\rangle$	$\langle 3s'\|3s\rangle$
15	0.998	0.995	0.987
21	0.999	0.997	0.995
25	0.999	0.998	0.997
30	1.000	0.999	0.997
35	1.000	0.999	0.998

In Table II we list some typical values of one-electron exchange integrals. The five values of $\langle 1s'|2s\rangle$ and $\langle 2s'|1s\rangle$ that were obtained in I with numerical Hartree-Fock wave functions are in good agreement with the exchange integrals of Table II, obtained with the Watson-Freeman wave functions.

The smooth dependence upon Z of both overlap and exchange integrals should be noted. As Z increases, all overlap integrals approach unity and all exchange integrals approach zero. This mathematical behavior reflects the fact that the fractional change in Z, which is $1/Z$, decreases as Z increases; hence, the resemblance between initial and final atomic states increases as Z increases.

TABLE II. One-electron exchange integrals. All wave functions are assumed to have the same phase at the origin.

$Z_{\rm final}$	$-\langle 1s'\|2s\rangle$	$+\langle 2s'\|1s\rangle$	$-\langle 1s'\|3s\rangle$	$+\langle 3s'\|1s\rangle$	$-\langle 2s'\|3s\rangle$	$+\langle 3s'\|2s\rangle$
15	0.0321	0.0287	0.0082	0.0072	0.0542	0.0441
21	0.0240	0.0223	0.0073	0.0069	0.0413	0.0368
25	0.0205	0.0193	0.0065	0.0062	0.0343	0.0314
30	0.0173	0.0165	0.0056	0.0054	0.0287	0.0267
35	0.0150	0.0143	0.0051	0.0049	0.0254	0.0238

In calculating the overlap and exchange integrals given in Tables I and II, we have assumed that all one-electron wave functions have the same phase at the origin. All physically significant quantities are of course unaffected by the phase convention. One can easily see, for example, that $X^{L/K}$, defined by Eq. (3c), and B_K, defined by Eq. (6c), are independent of all phase conventions.

In our first discussion, I, of L/K ratios, we set all overlap integrals equal to unity and neglected exchange effects between $3s$ electrons and $1s$ or $2s$ electrons. The results of Table I and II can be used to show that these approximations are accurate to about 2%.

We have also calculated overlap and exchange integrals for all values of Z from 13 to 37, except for Z equal to 18, 19, 20, and 29. These one-electron integrals

[18] J. G. V. Taylor and Janet S. Merritt (private communication).

TABLE III. Theoretical exchange corrections. See Section II for definitions of the exchange corrections. Values marked by an asterisk were interpolated using Eq. (10).

Z	Element	B_K	B_{L_I}	B_{M_I}	$X^{L/K}$
14	Si	0.924	1.199	1.804	1.298
15	P	0.939	1.193	1.711	1.271
16	S	0.947	1.181	1.639	1.248
17	Cl	0.954	1.172	1.579	1.228
18	Ar	0.959	1.162	1.530	1.212
19	K	0.963*	1.153*	1.489*	1.197*
20	Ca	0.966*	1.145*	1.454*	1.184*
21	Sc	0.969*	1.137*	1.423*	1.173*
22	Ti	0.970	1.128	1.399	1.162
23	V	0.973	1.122	1.375	1.154
24	Cr	0.974	1.117	1.354	1.146
25	Mn	0.976	1.112	1.335	1.139
26	Fe	0.977	1.107	1.317	1.133
27	Co	0.978	1.103	1.302	1.127
28	Ni	0.980	1.099	1.288	1.122
29	Cu	0.981	1.096	1.275	1.117
30	Zn	0.981*	1.090*	1.266*	1.112*
31	Ga	0.981	1.087	1.256	1.108
32	Ge	0.982	1.083	1.247	1.104
33	As	0.982	1.080	1.238	1.100
34	Se	0.983	1.078	1.230	1.096
35	Br	0.983	1.075	1.222	1.093
36	Kr	0.984	1.072	1.215	1.090
37	Rb	0.984	1.070	1.208	1.087

may be useful for other problems in which the nuclear charge changes by one unit. All overlap and exchange integrals that we have calculated are available upon request.

In Table III, we list values for the exchange corrections B_K, B_{L_I}, B_{M_I}, and $X^{L/K}$. The unstarred values of the exchange corrections were calculated from the one-electron overlap and exchange integrals obtained with the Watson and Freeman wave functions. The starred values of the exchange corrections given in Table III were calculated from the following formulas:

$$B_K=1-0.929Z^{-1}+20.98Z^{-2}-316.5Z^{-3}; \quad (9a)$$

$$B_{L_I}=1+1.695Z^{-1}+43.33Z^{-2}-387.1Z^{-3}; \quad (9b)$$

$$B_{M_I}=1+7.362Z^{-1}-12.47Z^{-2}+934.1Z^{-3}; \quad (9c)$$

$$X^{L/K}=1+2.810Z^{-1}+13.76Z^{-2}+75.2Z^{-3}. \quad (9d)$$

The coefficients in formulas (9) were obtained by a least-squares fit of the unstarred values of the exchange corrections in Table III. Arguments of the kind given by Layzer[19] suggest the theoretical justification for expanding the exchange corrections in an inverse power series in Z. The least-squares formulas (9) actually reproduce all the unstarred values given in Table III with a maximum error of 0.2%. Since the difference between any exchange correction and unity is small for large Z, we believe that formulas (9) can be used, with a conservative estimate of the uncertainties, to calculate exchange corrections that are accurate to 2%.

The numerical values of B_{L_I} are greater than unity because constructive interference occurs between processes (a) and (b) of Sec. II. The amplitude for process (c), which interferes destructively with the amplitudes for (a) and (b), is small in absolute magnitude. The numerical values of B_K, for entirely analogous reasons, are less than unity because of destructive interference between the processes corresponding to the first two terms of the K-capture amplitude, $f(1s')$, of Eq. (2b).

The Watson–Freeman wave functions, which were used in our present calculations, do not take account of relativistic and nuclear size effects. Complete wave functions that take account of relativistic, nuclear size, and screening effects have been calculated for only a few heavy atoms; hence, we cannot compute overlap and exchange integrals that include relativistic and nuclear size effects. However, we can easily see that relativistic and nuclear size effects are relatively unimportant for exchange effects. We let

$$X^{L/K}=1+\Delta X, \quad (10)$$

where ΔX approaches zero for large Z. Relativistic effects[20] will change ΔX by terms of order $(\alpha Z_{\text{eff}})^2$, i.e.,

$$X^{L/K}\cong 1+(\Delta X)_{\text{N-R}}[1+0(\alpha^2 Z_{\text{eff}}^2)], \quad (11)$$

where $1+(\Delta X)_{\text{N-R}}$ is the nonrelativistic value of $X^{L/K}$. Since $(\Delta X)_{\text{N-R}}$ is small for large Z, relativistic effects are only important when the net effect of exchange is relatively unimportant. Similar remarks apply to relativistic effects on B_K, B_{L_I}, and B_{M_I}. Nuclear size effects, like relativistic effects, are significant only for large Z (small ΔX) and are, therefore, not very important for exchange corrections. However, mutual electrostatic interactions among the atomic electrons are appreciable for small Z (large ΔX) and the Hartree-Fock wave functions of Watson and Freeman do take account of these interactions.

We have discussed in III some additional reasons for believing that the Watson–Freeman wave functions provide a good basis set for calculating overlap and exchange corrections.

IV. EXPERIMENTAL TESTS

We compare in Table IV the nine precisely measured L/K capture ratios with the capture ratios predicted by the usual theory and those predicted by the exchange-corrected theory. In the second column of Table IV, we list the appropriate electron binding energy corrections; the atomic mass differences used in computing the neutrino energies, q, were obtained from the *Nuclear Data Sheets*.[21] Note that the effect of electron binding energy is as large as 8% for Ge^{71}. For those isotopes for which more than one allowed capture contributes to the observed L/K ratio, we have computed the electron

[19] D. Layzer, Ann. Phys. (N. Y.) **8**, 271 (1959).

[20] D. Layzer and J. Bahcall, Ann. Phys. (N. Y.) **17**, 177 (1962).

[21] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington 25, D. C.).

TABLE IV. Comparison of theoretical and experimental L/K capture ratio.

Isotope	$\left(\frac{q(2s')}{q(1s')}\right)^2$	Usual theoretical ratio [Eq. (13)]	Exchange-corrected ratio [Eq. (4)]	Observed ratio	Number of precision experiments
Ar^{37}	1.006	0.0820	0.099	0.100 ±0.003	4
Cr^{51}	1.014[a]	0.0882	0.101	0.1026±0.0004	1
Mn^{54}	1.020	0.0898	0.102	0.098 ±0.006	1
Fe^{55}	1.051	0.0936	0.106	0.106 ±0.003	2
Co^{57}	1.017	0.0915	0.103	0.099 ±0.011	1
Co^{58}	1.008	0.0907	0.102	0.107 ±0.004	1
Zn^{65}	1.041[a]	0.0970	0.108	0.119 ±0.007	1
Ge^{71}	1.083	0.103	0.114	0.1175±0.002	2
Kr^{79}	1.021[a]	0.102	0.111	0.108 ±0.005	1

[a] Average calculated with Eq. (12).

binding energy correction form the following formula:

$$\left(\frac{q(2s')}{q(1s')}\right)_{\text{av}}^2=\Sigma_i\Gamma(i)\left(\frac{q(2s')}{q(1s')}\right)_i^2, \tag{12}$$

where $\Gamma(i)$ is the branching ratio for the ith mode of decay. The usual theoretical L/K capture ratios, which are listed in column three, were computed from the following equation:

$$\left(\frac{\lambda_L}{\lambda_K}\right)_{\text{usual}}=\left(\frac{\lambda_{L\text{I}}}{\lambda_K}\right)^0\left[1+\frac{L_{\text{II}}}{L_{\text{I}}}\right]. \tag{13}$$

The values of $|\psi_{2s}(0)/\psi_{1s}(0)|^2$ and $L_{\text{II}}/L_{\text{I}}$ that were used in computing the usual L/K ratios were taken from the tabulation by Wapstra *et al.*[17] of the results of Brysk and Rose.[1] The Brysk-Rose values of $|\psi_{2s}(0)/\psi_{1s}(0)|^2$ differ by at most 3% from the values computed with the Watson–Freeman wave functions. We calculated the exchange-corrected ratios, column four, from Eq. (4) and Table III. The maximum difference between the exchange-corrected L/K ratios given in Table IV and the exchange-corrected ratios estimated in I is 3%. The observed ratios that are given in column five, except for Kr^{79}, were taken from the tabulation by Moler and Fink[22]; the ratio for Kr^{79} was taken from Robinson and Fink.[6] Only precision measurements obtained with multiwire proportional counters are listed in Table III. For those isotopes whose L/K ratio has been measured in more than one precision experiment, we have averaged the experimental results and listed the spread among the experimental results as the experimental uncertainty. Moler and Fink[22] present the results of the individual measurements. It is interesting to note that the spread among the individual experimental values[22] is of the order of a few percent, although some observers report experimental errors of only a few tenths of a percent. In column six, we list the number of precision experiments that have been reported for each isotope.

The observed L/K ratios exceed the usual theoretical ratios by 6 to 22% for the nine isotopes listed in Table IV. Moreover, the disagreement between the usual theory and experiment decreases with increasing atomic number. This systematic disagreement between the usual theory and experiment was first pointed out by Robinson and Fink.[6]

Table IV shows that the exchange-corrected ratios are in good agreement with the observed ratios.

The most significant test of the exchange-corrected theory is to compare the fractional differences between the measured and the usual theoretical L/K capture ratios with $X^{L/K}-1$. This comparison isolates the purely exchange contributions to the capture ratios. We therefore plot in Fig. 1 the experimental values, versus atomic number, of

$$X_{\text{exp}}{}^{L/K}-1\equiv\frac{(\lambda_L/\lambda_K)_{\text{obs}}-(\lambda_L/\lambda_K)_{\text{usual}}}{(\lambda_L/\lambda_K)^0}, \tag{14}$$

where $(\lambda_L/\lambda_K)_{\text{obs}}$ is the observed L/K ratio. According to the usual theory, $X_{\text{exp}}{}^{L/K}-1$ should be equal to zero. According to the exchange-corrected theory,

$$X_{\text{exp}}{}^{L/K}-1=X^{L/K}-1, \tag{15}$$

where $X^{L/K}$ is the theoretical L/K exchange correction whose numerical values are given in Table III and Eq. (9d). The curve in Fig. 1 was obtained from the theoretical values of $X^{L/K}$ given in Table III and Eq. (9d). Figure 1 shows that the exchange-corrected theory explains both the general trend and the individual values of the disagreement between the usual theory and experiment. Of the nine isotopes listed in Table IV and Fig. 1, only Zn^{65} has a measured L/K ratio that differs from the corresponding exchange-corrected ratio by more than the usual systematic uncertainties of about 4%. It would be useful to repeat the Zn^{65} measurement with greater precision in order to clarify this discrepancy.

An additional test of the exchange-corrected theory

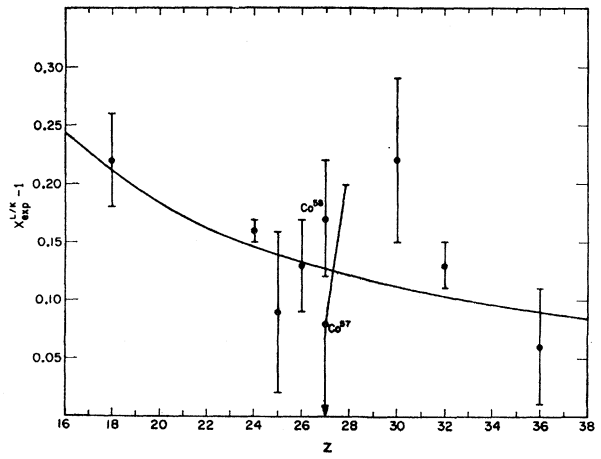

FIG. 1. Comparison of experimental and theoretical L/K exchange corrections. Experimental points are $X_{\text{exp}}{}^{L/K}-1$. The smooth curve represents the theoretical exchange correction $X^{L/K}-1$.

[22] R. B. Moler and R. W. Fink, Phys. Rev. **131**, 821 (1963).

that could also be made is the measurement of the L/K ratio for V^{49}. The usual theoretical L/K ratio for V^{49} is 0.087; the exchange-corrected ratio is 0.101.

ACKNOWLEDGMENTS

I am grateful to B. A. Zimmerman and Dr. P. A. Seeger for valuable help in programming the computations for the California Institute of Technology IBM 7090. I have profited by many discussions with members of the Kellogg Radiation Laboratory and am particularly indebted to Dr. R. W. Kavanagh for his patient explanations of experimental problems and techniques. I am grateful to a number of workers, especially Dr. R. W. Fink, Dr. B. L. Robinson, and Dr. J. G. V. Taylor, for informing me of their results prior to publication.

PHYSICAL REVIEW VOLUME 132, NUMBER 1 1 OCTOBER 1963

Lifetime Measurements on the First Excited States of O^{17} and F^{18}†

J. LOWE* AND C. L. MCCLELLAND‡
Brookhaven National Laboratory, Upton, New York
(Received 8 May 1963)

The mean life of the first excited state of O^{17} at 871 keV has been measured by a pulsed Van de Graaff beam technique. A value of $\tau_m = (0.263 \pm 0.008)$ nsec was found. Similar measurements on the first excited state of F^{18} at 940 keV established an upper limit to the mean life: $\tau_m < 0.2$ nsec.

I. INTRODUCTION

In a shell-model description, O^{17} consists of a single neutron outside a closed-shell O^{16} core. The $\frac{5}{2}+$ ground state and $\frac{1}{2}+$ first excited state of O^{17} correspond, respectively, to the $1d_{5/2}$ and $2s_{1/2}$ states of the odd neutron. The 871 keV, $E2$ gamma decay of the first excited state is on this model, therefore, a single neutron transition outside the core. The lifetime for this decay is of interest theoretically,[1] since the odd neutron makes no contribution to the matrix element for the transition, which takes place only through contributions from the O^{16} core. Many attempts have been made to calculate the core contributions, using both the shell model,[2] and also the weak-coupling collective model.[1] The importance of a measurement of this lifetime arises partly from the possibility of a direct experimental test of the theoretical work for this relatively simple nucleus, and also from the consequent availability of an empirical value of the O^{16} core contribution, for use in the interpretation of lifetime measurements in neighboring nuclei in the $1d-2s$ shell.

Several measurements of the lifetime for this transition have been published,[3–5] but in only two cases were the measurements made by direct observation of the exponential decay of the level using electronic timing. Lifetimes measured by this method are not subject to the uncertainties and systematic errors associated with recoil or centroid shift techniques. However, the two published measurements in which the decay was observed directly[4,5] differ substantially. Also, one of the measurements involved delayed coincidence techniques, in which the identification of the gamma responsible for the observed decay is somewhat less certain. The present paper reports a further measurement of this lifetime using pulsed beam techniques.

A detailed study of the properties of the low-lying states of F^{18} has been published by Kuehner, Almqvist, and Bromley.[6] Kuehner *et al.* compared their experimental data with the shell-model predictions for this nucleus calculated by Elliott and Flowers[7] and by Redlich,[8] and found that the shell model can give a reasonably satisfactory account of the levels below 3 MeV. Kuehner *et al.* set an upper limit of 5 nsec on the lifetime of the 3+ first excited state at 940 keV for $E2$ gamma decay to the 1+ ground state. An identical upper limit for this lifetime was set by Allen *et al.*[9] and

† Work performed in part under the auspices of the U. S. Atomic Energy Commission. A preliminary report of part of this work is given in Bull. Am. Phys. Soc. **7**, 340 (1962); **7**, 419 (1962).

* Present address: Physics Department, Birmingham University, Birmingham, England.

‡ On leave from Brookhaven National Laboratory. Present address: Arms Control and Disarmament Agency, Washington, D. C.

[1] B. J. Raz, Phys. Rev. **120**, 169 (1960). This paper reviews other theoretical work on gamma transitions in nuclei in this mass region.

[2] For example, G. Barton, Nucl. Phys. **11**, 466 (1959); G. Barton, D. M. Brink, and L. M. Delves, *ibid.* **14**, 256 (1959).

[3] For example, J. Thirion and V. L. Telegdi, Phys. Rev. **92**, 1253 (1953).

[4] J. V. Kane, R. E. Pixley, R. B. Schwartz, and A. Schwarzschild, Phys. Rev. **120**, 162 (1960).

[5] N. H. Gale, J. B. Garg, and J. M. Calvert, Nucl. Phys. **38**, 222 (1962).

[6] J. A. Kuehner, E. Almqvist, and D. A. Bromley, Phys. Rev. **122**, 908 (1961).

[7] J. P. Elliott and B. H. Flowers, Proc. Roy. Soc. **A229**, 536 (1955).

[8] M. G. Redlich, Phys. Rev. **95**, 448 (1954); **110**, 468 (1958).

[9] K. W. Allen, D. Eccleshall, and M. J. L. Yates, Proc. Phys. Soc. (London) **74**, 660 (1959).