PHYSICAL REVIEW C VOLUME 5, NUMBER 6 JUNE 1972

Orbital-Electron-Capture Ratios in the Decay of ^{55}Fe†

J. G. Pengra,* H. Genz,‡ J. P. Renier, and R. W. Fink
School of Chemistry, Georgia Institute of Technology, Atlanta, Georgia 30332
(Received 7 February 1972)

The M/L and L/K orbital-electron-capture ratios in the decay of ^{55}Fe have been determined by introducing the radioactive compound ferrocene-^{55}Fe into a multiwire, wall-less anticoincidence proportional counter as part of the counting gas mixture. The results are $P_M/P_L = 0.157 \pm 0.003$ and $P_L/P_K = 0.117 \pm 0.001$ at $Z = 26$. These values are compared with various theoretical predictions and, together with previous experimental results on M/L capture ratios at low Z, they demonstrate that of all the combinations of theoretical approaches, the Watson-Freeman wave functions with the exchange corrections of Vatai are the only ones which give good agreement with experiment at low Z. All other theoretical combinations lie considerably higher than experiment in the region below $Z = 36$.

Fig. 1 & Source Preparation (Sect. 2 A) by Collé

1. INTRODUCTION

Previously we reported the determination of M/L orbital-electron-capture ratios in the allowed decays of ^{37}Ar ($Z = 18$)[1] and ^{71}Ge ($Z = 32$),[2] based on measurements of the radioactive gases in a wall-less, anticoincidence multiwire proportional counter and single-wire proportional counters. In the present work, the measurement of the M/L and L/K capture ratios in the decay of ^{55}Fe ($Z = 26$) is reported. These measurements are needed to evaluate which of the currently available sets of electron wave functions and which of the various approaches to the calculation of the electron-exchange–atomic-overlap correction factors best agree with experiment.

The decay of 2.7-yr ^{55}Fe is an especially favorable case for study, since it has a simple allowed decay scheme, a suitably long half-life, and forms a vaporous compound, ferrocene, which can be used in a proportional-counter gas. It has been pointed out[1,3] that only precision determinations of M/L capture ratios at low Z (<40) can distinguish among the various theoretical approaches.

The present results for ^{55}Fe again confirm agreement with the wave functions of Watson and Freeman[4] and the exchange correction of Vatai,[5] as was observed in the decays of ^{37}Ar and ^{71}Ge. The present experimental method is the same as that in the previous study of ^{71}Ge decay.[2]

2. EXPERIMENTAL

A. Source Preparation

The radionuclide ^{55}Fe, having a half-life of 2.7 yr, was obtained from New England Nuclear Corporation as $FeCl_3$ in dilute HCl and had been prepared by reactor irradiation of ^{54}Fe. It was aged to allow 45-day ^{59}Fe to decay. The ferrocene compound [bis-cyclopentadienyl-iron(II)] was prepared[6] by using the reaction between anhydrous radioactive $FeCl_2$ and sodium cyclopentadienide in tetrahydrofuran under an inert atmosphere. Figure 1 is a drawing of the glass apparatus used in the ferrocene synthesis[6] which was carried out in a water- and air-free atmosphere without having to separate or transfer the reactants. After removal of the solvent by evaporation under reduced pressure, the ferrocene was removed from the reaction mixture and collected by sublimation. The labeled $^{55}FeCl_2$ reactant was obtained by adding the $^{55}FeCl_3$ to electrolytic iron powder and reacting the mixture with HCl. The $^{55}FeCl_2$ was then dehydrated to anhydrous form by heating *in vacuo* at 160°C for 30 min. The final samples of ferrocene, containing about 2.5 μCi/mg of activity, were contained in an inert atmosphere with NaPb as a preservative against water and oxygen contamination prior to introduction into the counter.

The vapor pressure of ferrocene is about 0.2 mm at room temperature and increases rapidly with temperature. The melting point is 173.5°C and the compound sublimes below this temperature. Ferrocene is chemically very stable under 200°C. It turned out that the specific activity was too low for optimum performance in the counter, and the use of larger quantities resulted in a loss of activity to the counter walls. In order to decrease the rate of loss of ferrocene activity, the counter was wrapped in Teflon insulation and was surrounded with a heating tape, in order to maintain a temperature of between 40 and 65°C. Heating the counter walls in this way led to an increase in activity, indicating that some ferrocene had been vaporized from the walls. During a typical run, therefore, the temperature was increased gradually (typically by 1°C per day) to yield a fairly constant activity in the center counter. The time

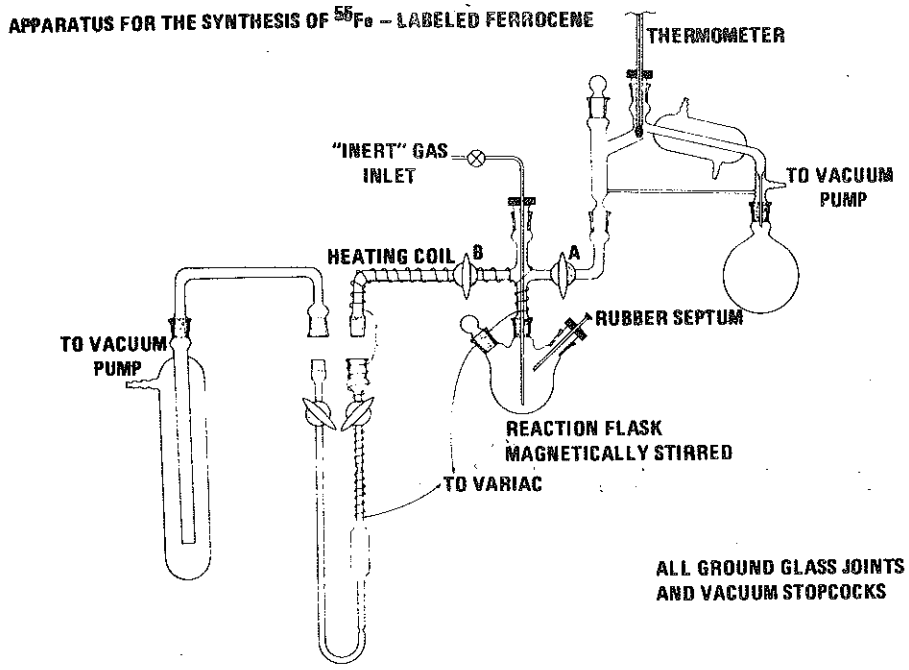

FIG. 1. Apparatus for the synthesis of radioactive ferrocene ^{55}Fe. The vacuum lines were double-trapped with condensers cooled in dry-ice–acetone baths. After assembly, the apparatus was flamed and pumped to remove absorbed moisture. The synthesis is accomplished in an inert atmosphere without having to separate or transfer the reactants. The yields of ferrocene with respect to iron activity were in the range of 75 to 92%.

dependence of the background due to condensation of ferrocene activity on the walls is negligible during a typical run, since the total activity in gaseous form changed during a typical run by no more than 0.5%. A correction was made for the slight decrease in gaseous activity, but the increase in the background was negligible and its uncertainty is included in the total error.

B. Electronic System

The counter and electronic system have been described in detail previously.[2] In the present work, it was necessary to monitor the activity of the ferrocene because of its loss in time. This was done by monitoring the counting rate of the center counter and normalizing the individual runs to the same monitor rate. This monitor system used K-peak pulses from the first stage in the Tennelec TC-200 amplifier. The pulses were passed through an amplifier and a single-channel analyzer, set to pass the K peak through the window to a gate and a scaler. The gate was controlled by pulses originating in the ring counter.

C. Spectral Measurements

Typical M, L, and K spectra taken in separate runs at the same gas gain ($\approx 10^4$), but with different electronic gains, are shown in Figs. 2 and 3. To obtain reasonable statistics, these runs required 10–12, 3–4, and 1 h, respectively, for the M, L, and K spectra. Such long counting times were required because of the large deadtime (≈60% in the M region) and the low counting rate (70 dis/sec total in the central counter). The long deadtimes arise primarily from the paralysis times used on the center and the ring counters.[2] Spectra were taken with several independent runs and dif-

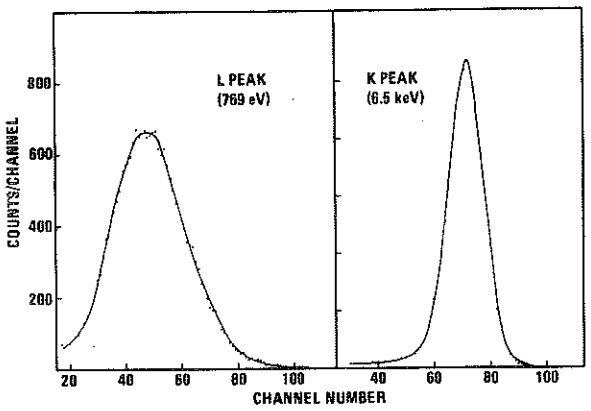

FIG. 2. Typical L and K spectra of ^{55}Fe after subtraction of background, and in the case of the L peak, subtraction of K degradation (see Ref. 2). The K peak exhibits a typical resolution of 16% full width at half maximum and the L peak of ≈55%.

ferent counter fillings. The background was determined by filling the counter with the same gas mixture, but omitting the radioactive source before and after each counter filling. An external ^{57}Co source of 14.4-keV γ rays was used to calibrate for identical gas gain.

D. Evaluation of the Intensity Ratios N_L/N_K and N_M/N_L

The computer program for the ^{71}Ge problem described in detail previously[2] was used in the present work. The N_L/N_K and N_M/N_L peak intensity ratios must be corrected for three effects: (1) the probability that a photon strikes a central-counter cathode wire (P_1); (2) the probability (P_2) that an x ray passes from the center counter through the ring counter undetected; and (3) the probability (P_3) that an x ray escapes through the ends of the counter. These probabilities are dependent on photon energy, gas pressure and mixture, and counter dimensions. For the L/K capture ratio, it can be shown that

$$\frac{P_L}{P_K}=\frac{N_L}{N_K}(1-\omega_K P_\alpha)-\omega_K P_\alpha k_\alpha\,, \tag{1}$$

where ω_K is the K-shell fluorescence yield of the daughter ($Z=25$); $P_\alpha=P_1+P_2+P_3$ for $K\alpha$ x rays, and k_α is the fraction of $K\alpha$ x rays in the K series of the daughter.

For the M/L capture ratio, it can be shown[2] that

$$\frac{P_M}{P_L}=\frac{N_M}{N_L}\left(1+\frac{P_K}{P_L}\omega_K P_\alpha k_\alpha\right)-\frac{P_K}{P_L}\omega_K P_\beta(1-k_\alpha)\,, \tag{2}$$

where $P_\beta=P_1+P_2+P_3$ is computed for the $K\beta$ x-rays of the daughter ($Z=25$). The quantities P_α and P_β are calculated following a procedure outlined by Vatai and are listed in Table I, along with the other quantities taken from the literature and the results of the present experiments.

The principal source of error in the M/L capture ratio is the uncertainty in the N_M/N_L intensity ratio and in the value of k_α. The errors in N_M/N_L and N_L/N_K are probable errors based on counting statistics and are propagated quadradically when subtracting backgrounds and degradation tails.

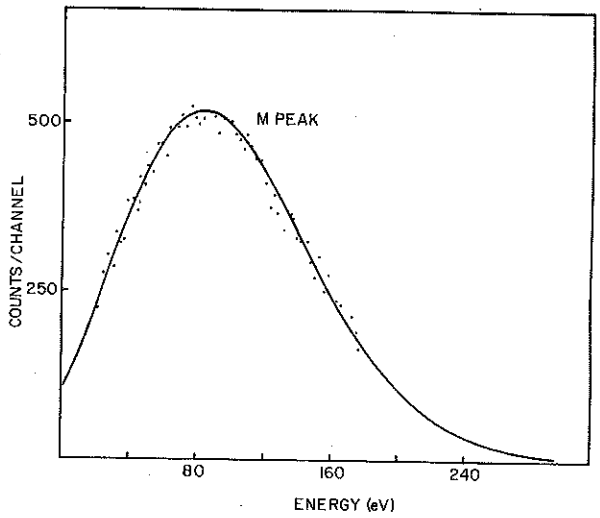

FIG. 3. A typical M spectrum after subtraction of background and degradation tails from the K and L peaks. The curve is a fit to a Poisson distribution with a mean value of $m=3.1$ (see Ref. 2). The resolution of the M peak at a mean energy of 84 eV is ≈160%.

The possibility that in the Poisson distribution of the M capture events, Fig. 3, some M capture transitions produce zero ion pairs and hence no counter pulse has been taken into account by fitting the spectral data with a Poisson distribution which has a nonzero intercept *after* subtraction of the degradation tails due to K and L capture events, as discussed in Ref. 2. This nonzero intercept fully corrects for M capture events giving zero ion pairs. This is true in the previous measurements of the M/L capture ratio in ^{71}Ge decay[2]; in the decay of ^{37}Ar, this effect is taken into account by the single-electron distribution from ultraviolet photons used to fit the M capture spectrum.[1]

E. Results for P_L/P_K and P_M/P_L

The present result for the L/K capture ratio in the decay of ^{55}Fe is

$$P_L/P_K=0.117\pm0.001\,,$$

where the error limit represents the probable error with no estimate for possible systematic errors. This value is larger than the previously re-

TABLE I. Experimental results obtained with the multiwire proportional counter from ^{55}Fe decay.

P_α	P_β	k_α	ω_K	N_L/N_K	N_M/N_L	P_L/P_K	P_M/P_L
0.021 [a]	0.035 [a]	0.89 [b]	0.314 [c]	0.124	0.160	0.117 ± 0.001	0.157 ± 0.003

[a] E. Vatai, private communication.
[b] J. S. Hansen, H. U. Freund, and R. W. Fink, Nucl. Phys. A142, 604 (1970).
[c] W. Bambynek, B. Crasemann, R. W. Fink, H. U. Freund, H. Mark, R. E. Price, P. V. Rao, and C. D. Swift, Rev. Mod. Phys. (to be published).

ported value of 0.106 by Scobie, Moler, and Fink[7] and by Manduchi and Zannoni.[8] It should be pointed out, however, that in these earlier measurements, contributions due to the degradation tails of the K and L peaks have not been taken into account. In the present work, the contributions due to degradation of L and K capture events amount to 7.4 and 3.3% of the total activity, respectively. If this effect is not taken into account, the value of P_L/P_K obtained in the present work decreases to 0.111, instead of 0.117, and agrees within the error limits with the previously reported[7,8] results for ^{55}Fe. It is believed, however, that the degraded L and K events have to be included in the total peak intensities (see discussion in Ref. 2), and thus the higher value of P_L/P_K is reported here. The higher value also agrees well with theoretical calculations summarized by Genz.[3]

The result from the present work for the M/L capture ratio in ^{55}Fe decay is

$$P_M/P_L = 0.157 \pm 0.003 ,$$

where the error limit represents the probable error with no attempt to include possible systematic errors. No previous measurements of the M/L capture ratio exist for ^{55}Fe decay.

3. DISCUSSION AND COMPARISON WITH THEORY

A thorough investigation of allowed orbital-electron-capture probabilities and ratios, including atomic variables, has been carried out theoretically in the range $14 \leq Z \leq 37$ by Bahcall,[9] who found that imperfect atomic overlap between initial and final states is small and largely cancels out in ratios of capture probabilities. However, an electron-exchange correction was found to make a significant contribution in the capture ratios, being largest for low Z and for the M shell. The theory as given by Bahcall[9] leads to the expressions for the ratios of allowed orbital-electron-capture probabilities:

$$\frac{P_L}{P_K} = \left| \frac{q_{(2s')}\psi_{2s(0)}}{q_{(1s')}\psi_{1s(0)}} \right|^2 \left(X^{L/K} + \frac{P_{L_2}}{P_{L_1}} \right), \tag{3}$$

and

$$\frac{P_M}{P_L} = \left| \frac{q_{(3s')}\psi_{3s(0)}}{q_{(2s')}\psi_{2s(0)}} \right|^2 X^{M/L} , \tag{4}$$

where the primed quantities refer to the final state and unprimed to the initial state; $q_{(1s')}$, $q_{(2s')}$, $q_{(3s')}$ are the respective neutrino-emission energies for K, L_1, and M_1 electron capture; $\psi_{ns(0)}$ are the respective electron wave functions evaluated at the nucleus taking into account finite nuclear size; and $X^{L/K}$ and $X^{M/L}$ are the electron-exchange–atomic-overlap corrections, as defined by Bahcall.[9] The term P_{L_2}/P_{L_1} takes into account the small probability in allowed transitions of capture of $2p_{1/2}$ electrons.

Various calculations of the electron wave-function densities for K, L, and M shells as a function of Z have been reported since the early results of Brysk and Rose.[10] The calculations of Bahcall[9] were based on the nonrelativistic Hartree-Fock wave functions of Watson and Freeman.[4] For low Z, calculations of K-, L-, and M-shell wave-function densities at the nucleus have been made by Behrens and Jänecke[11] by solving the Dirac equation for the bound electrons with the Hartree-Fock potential for $Z \leq 36$. Finite-nuclear-size effects were included by treating the nucleus as a uniformly charged sphere with a sharp surface, as was done also by Suslov,[12] who used the nonrelativistic self-consistent-field potential of Herman and Skillman for $Z \leq 72$. The M_1/L_1 wave-function densities of Martin and Blichert-Toft[13] are not comparable to the others, because they included contributions from all shells higher than the M_1 subshell.

Several theoretical approaches to the calculation of the electron-exchange–overlap factors $X^{L/K}$ and $X^{M/L}$ have been made.[5,9,13-15] A critical comparison among these approaches has been carried out[3] and reported elsewhere.[16] Faessler *et al.*[14] recalculated the Bahcall[9] exchange-overlap factors for light atoms using the Hartree-Fock-Slater functions of Herman and Skillman and find very good agreement (±0.5%) at $Z = 18$ for $X^{L/K}$ and agreement of the order of 1–2% for $X^{M/L}$. The exchange factor calculated by Martin and Blichert-Toft[13] exhibits a systematic discrepancy in $X^{L/K}$ with respect to Bahcall,[9] ranging from 7% at $Z = 14$ to 2% at $Z = 37$, and in $X^{M/L}$, of 15.4% at $Z = 14$ to 4.8% at $Z = 37$. The latter large deviation is due to the M-shell capture probability, which deviates by 20% from Bahcall's values.[9] Suslov[15] has calculated $X^{L/K}$ and $X^{M/L}$ according to Bahcall's method, but with wave functions obtained by numerical integration of Dirac's equation using a nonrelativistic self-consistent Hartree-Fock-Slater potential for $14 \leq Z \leq 73$, and finds values of $X^{L/K}$ nearly identical to those of Bahcall[9] below $Z = 37$, and within 0.5% for $X^{M/L}$.

The approximations made in the calculation of exchange and overlap integrals by Bahcall[9] were first critically examined by Vatai.[5,17] Bahcall[9] used the ground-state Hartree-Fock wave functions for the initial and final atoms. In reality the final atom is not in the ground state after the electron-capture process, but in an excited state with a vacancy in one of the inner shells, K, L, or M. The Hartree-Fock wave functions for the excited states are different from those of the ground state.

TABLE II. M/L orbital-electron-capture ratios and comparison with theory in the region $14 \leq Z \leq 36$. Neutrino energies were calculated from $Q_{EC} = 813.8 \pm 0.7$, 231.3 ± 1.3, and 233.0 ± 3.0 keV for the respective decays of ^{37}Ar, ^{55}Fe, and ^{71}Ge.

						Theory				
Z	Nuclide	P_M/P_L Experimental	Ref.	q_M^2/q_L^2	$\frac{P_M/P_L}{q_M^2/q_L^2}$	$\lvert\psi_M/\psi_L\rvert^2$, Vatai $X^{M/L}$	Watson-Freeman $X^{M/L}$, Bahcall	$\lvert\psi_M/\psi_L\rvert^2$, Suslov $X^{M/L}$, Vatai	$\lvert\psi_M/\psi_L\rvert^2$, Behrens and Jänecke $X^{M/L}$, Vatai	$X^{M/L}$, Bahcall
18	^{37}Ar	$0.104^{+0.006}_{-0.003}$	1	1.00	0.104	0.111	0.128	0.129		
26	^{55}Fe	0.157 ± 0.003		1.01	0.156	0.152	0.162	0.163		
32	^{71}Ge	0.162 ± 0.003	2	1.01	0.161	0.163	0.172	0.169	0.183	0.193

The neglect of this "rearrangement" has been pointed out by Vatai,[5,17] who suggested that the inclusion of this effect could improve the agreement between experimental and theoretical values of M/L capture ratios.

In order to include rearrangement in the Bahcall correction, Faessler *et al.*[14] calculated the self-consistent single electron orbits for the excited state with a hole in the K, L, or M shell using the Herman-Skillman wave functions, and found that the rearrangement effect reduces the correction factors $X^{L/K}$ and $X^{M/L}$. The differences are important, however, only for very light nuclei (e.g., ^{7}Be). For $23 \leq Z \leq 36$, the difference in $X^{L/K}$ is less than 1% and almost negligible at $Z = 36$, while $X^{M/L}$ is lowered by 1–2% in the region $14 \leq Z \leq 36$. Thus, the theoretical values of P_L/P_K and P_M/P_L are brought slightly closer to experiment, but not enough to remove the disagreement.

Vatai[5] not only included rearrangement effects, but also calculated the contribution of each possible final state involving the exchange overlap of all inner shell s, p, and d electrons. He used the wave functions of Watson and Freeman[4] to describe the initial state and as a basis set for the calculation of the final state by first-order perturbation theory according to the self-consistent-field method. For $X^{L/K}$, Vatai's results[5] lie only slightly lower (1% at $Z = 26$) than those of Bahcall,[9] but agree extremely well with Faessler's calculation.[14] However, for $X^{M/L}$, Vatai's values[5] are

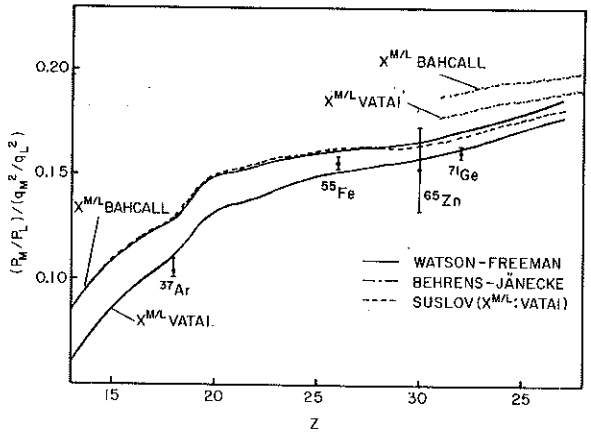

FIG. 4. Comparison of low-Z precision M/L orbital-electron-capture ratios with theoretical results based on the wave functions of Watson-Freeman (see Ref. 4), Behrens-Jänecke (Ref. 11), and Suslov (Ref. 12) with exchange corrections either of Bahcall (Ref. 9) or Vatai (Ref. 5). It is clear that the only combination of theoretical results which agrees with experiment in the region of $Z = 18$ to 36 is that based on the Watson-Freeman wave functions corrected by the Vatai exchange factor (see discussion in the text).

considerably smaller (6.7% at $Z=26$) than those of Bahcall.[9]

In order to compare the present experimental ratio $P_L/P_K = 0.117 \pm 0.001$ with the theoretical predictions, the experimental values are customarily divided by the neutrino-energy factor, which in ^{55}Fe decay has a value of 1.052 for L/K capture, since $Q_{EC}=231$ keV. The present work therefore gives $(P_L/P_K)/(q_L{}^2/q_K{}^2)=0.111 \pm 0.001$, while the theoretical calculation, based on the wave-function-density ratio from any of the above theoretical calculations,[4,11-13] but not including Brysk and Rose,[10] together with the exchange-correction factor $X^{L/K}$ of Bahcall[9] or Vatai[5] gives values of 0.109 or 0.108, respectively. Thus, the L/K capture ratio measurement is *not* sufficiently sensitive to distinguish among the various combinations of theoretical wave-function densities and exchange-correction calculations.

In Table II a comparison is given between theory and the experimental M/L capture ratios for $14 \leq Z \leq 37$, as obtained by this group. The experimental ratios have been divided by the appropriate neutrino-energy factors. For the theoretical calculations the wave functions by Watson and Freeman[4] for $13 \leq Z \leq 37$ and those by Behrens and Jänecke[11] for $30 \leq Z \leq 37$ have been compared. The correction factors $X^{M/L}$ have been taken from Bahcall[9] and Vatai,[5] as shown in Fig. 4. In addition, the calculations by Suslov[12] are plotted in Fig. 4, after correction by Vatai's exchange calculation.[5] If the wave-function densities of Suslov[12] are corrected for $X^{M/L}$ by other than the Vatai results,[5] they will lie systematically higher.

4. CONCLUSION

From Fig. 4 it can be observed that the agreement between experiment and exchange-corrected theory for M/L capture ratios has been improved since the last summary[16] and that in the decay of the three low-Z nuclides ^{37}Ar ($Z=18$), ^{55}Fe ($Z=26$), and ^{71}Ge ($Z=32$), the experimental M/L capture ratios agree within the error limits with theoretical predictions using the wave functions of Watson and Freeman[4] and the exchange corrections by Vatai,[5] which are also based on Watson-Freeman wave functions. This combination of the Watson-Freeman wave functions with the Vatai exchange correction is the only one which gives good agreement with experimental M/L capture ratios at low Z. All other theoretical combinations lie considerably higher than experiment in the region below $Z=36$.

5. ACKNOWLEDGMENT

We are indebted to R. P. Colle for devising and carrying out the improved synthesis of the radioactive ferrocene samples and for Fig. 1.

---

†Work supported in part by the U. S. Atomic Energy Commission.

*Present address: Department of Physics, Whitman College, Walla Walla, Washington.

‡Present address: Ruhruniversität Bochum, Physikdepartment, Bochum, Germany.

[1]J. P. Renier, H. Genz, K. W. D. Ledingham, and R. W. Fink, Phys. Rev. 166, 935 (1968).

[2]H. Genz, J. P. Renier, J. G. Pengra, and R. W. Fink, Phys. Rev. C 3, 172 (1971).

[3]H. Genz, Ph.D. thesis, Emory University, 1971 (unpublished).

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

[5]E. Vatai, Nucl. Phys. A156, 541 (1970).

[6]R. P. Colle, Georgia Tech (to be published).

[7]J. Scobie, R. B. Moler, and R. W. Fink, Phys. Rev. 116, 657 (1959); R. B. Moler, Ph.D. thesis, University of Arkansas, 1960 (unpublished).

[8]C. Mansuchi and G. Zannoni, Nuovo Cimento 27, 251 (1963).

[9]J. N. Bahcall, Phys. Rev. 132, 362 (1963); 131, 1756 (1963).

[10]H. Brysk and M. E. Rose, Rev. Mod. Phys. 30, 1169 (1958); ORNL Report No. ORNL-1830, 1955 (unpublished).

[11]H. Behrens and J. Jänecke, *Landolt-Börnstein: New Series* (Springer, Berlin, 1969), Group I, Vol. 4.

[12]Yu. P. Suslov, Izv. Akad. Nauk SSSR, Ser. Fiz. 34, 2223 (1970) [transl.: Bull. Acad. Sci. USSR, Phys. Ser. 34, 2223 (1970)].

[13]M. J. Martin and P. H. Blichert-Toft, Nucl. Data A8, 1 (1970).

[14]A. Faessler, E. Huster, O. Krafft, and F. Krahn, Z. Physik 238, 352 (1970).

[15]Yu. P. Suslov, Izv. Akad. Nauk SSSR, Ser. Fiz. 34, 91 (1970) [transl.: Bull. Acad. Sci. USSR, Phys. Ser. 34, 91 (1970)].

[16]H. Genz, in Proceedings of the International Conference on Inner Shell Ionization Phenomena, Atlanta, April, 1972 (North-Holland, Amsterdam, to be published).

[17]E. Vatai, in *Proceedings of the International Conference on Electron Capture and Higher Order Processes in Nuclear Decay, Debrecen* (Eötvös Lorand Physical Society, Budapest, Hungary, 1968), Vol. 1, p. 71.

[18]R. W. Fink, Phys. Rev. 180, 1220 (1969).