slowly with energy and the angular distribution of neutrons to both the ground state and first excited states becomes quite isotropic at higher energies. The possibility of a direct-interaction mechanism has been suggested at lower energies[19] but the angular distributions, especially at high energies, show none of the strong forward or backward peaking usually associated with direct processes.

[19] H. R. Striebel, S. E. Darden, and W. Haeberli, Nucl. Phys. 6, 188 (1958).

ACKNOWLEDGMENTS

The authors wish to thank Professor H. H. Barschall for suggesting this experiment and for his advice and guidance. The help of Dr. C. H. Holbrow in collecting the data is also gratefully acknowledged.

PHYSICAL REVIEW VOLUME 129, NUMBER 6 15 MARCH 1963

Overlap and Exchange Effects in Beta Decay*

JOHN N. BAHCALL
California Institute of Technology, Pasadena, California
(Received 12 November 1962)

The change in nuclear charge by one unit in beta decay causes initial and final atomic states to overlap imperfectly. The effect of this imperfect overlap on the shape of allowed electron and positron emission spectra is calculated. The calculated change in the spectrum shape can be simulated by including the average excitation energy of the final atom in the energy balance. The inhibition, due to imperfect atomic overlap, of electron-capture rates, as well as total electron and positron-emission rates, is also determined. In all known cases, imperfect atomic overlap increases beta-decay lifetimes by at most a few percent and usually by an amount less than a few tenths of one percent. Antisymmetrization between decay and bound atomic electrons, in conjunction with the change in nuclear charge, gives rise to exchange effects in electron emission and electron capture. Due to exchange terms, the usual allowed electron spectrum is multiplied by a quantity that is of the order of $1-2Z^{-1}$ for energies less than the binding energy of a K electron in the initial atom. This exchange correction is negligible for higher energies of the emitted continuum electron. A simple approximate formula is derived that predicts the effect of exchange on L to K capture ratios; this formula predicts a 22% increase over the usual theoretical value for the L to K ratio of Ar^{37}. The Ar^{37} prediction is in excellent agreement with recent experiments and with a more complicated calculation by Odiot and Daudel. Exchange effects change total electron emission and electron capture rates by at most a few percent.

I. INTRODUCTION

HOW does the change in nuclear charge by one unit from initial to final atomic states affect beta decay? How much does the imperfect overlap of initial and final atomic states inhibit beta-decay rates? Does the possibility of exchange between bound and decay electrons significantly affect electron emission and electron capture probabilities? This paper is an attempt to answer the above questions.

Benoist-Gueutal[1] first emphasized that a correct specification of the initial and final states of a radioactive system must include a description of the atomic electrons. The overlap between initial and final atomic states is not equal to one since the initial and final states are eigenstates of zero-order Hamiltonians with different nuclear charges. Thus, one expects the theoretical decay rate to be decreased if atomic states are included in the description of the radioactive system. If this decrease were large, one would have to know the magnitude of the decrease in order to calculate nuclear matrix elements from experimentally determined parameters.

Benoist-Gueutal[1] estimated the effect of imperfect atomic overlap on the total electron capture rate of Be^7 by calculating the electron capture probability for various final atomic states. She concluded that the decrease in the total decay rate was between 0 and 30%; her calculation was limited by the lack of accurately known wave functions for an excited lithium atom. For heavier atoms, good atomic wave functions are even more difficult to obtain than for lithium. Moreover, the problem of evaluating the decay probability to all final states of a heavy atom is prohibitively complicated.

We calculate the effect of the change in nuclear charge by expanding the energy conserving delta function as a power series in the excitation energy of the final atom and then use closure to sum the beta-decay transition probability over all possible final atomic states. Explicit results are presented for allowed electron and positron emission and for allowed electron capture.

* Supported in part by the Joint Program of the Office of Naval Research and the U. S. Atomic Energy Commission, and in part by the National Aeronautics and Space Administration.
[1] P. Benoist-Gueutal, Ann. Phys. (Paris) 8, 593 (1953).

The allowed electron and positron spectrum shapes that are obtained by taking account of imperfect atomic overlap, but not exchange, can be simulated by including the average excitation energy of the final atom in the energy balance. The calculated change in the allowed spectrum shape due to imperfect overlap is small, in agreement with the expectation of some previous investigators.[1,2]

The fractional change in the total decay rate due to imperfect atomic overlap is, for electron and positron emission as well as electron capture, of the order of minus the average excitation energy of the final atom divided by the energy release of the process. That is,

$$\lambda^1/\lambda^0 \sim -\bar{E}_{ex}/E_{max}, \tag{1}$$

where λ^1/λ^0 is the fractional change in the total decay rate, $\bar{E}_{ex}$ is the average excitation energy of the final atom, and E_{max} is approximately equal to the difference of nuclear masses plus (for electron capture) or minus (for electron or positron emission) one electron mass.

This change in the total decay rate is less than 10% for all known radioactive decays and is less than a few tenths of one percent for most decays. It is completely negligible for Be^7. We conclude that it is not necessary to consider the imperfect overlap of atomic states in determining experimental nuclear matrix elements.

In electron emission, an exchange contribution to the decay probability results from the requirement that the final-state vector be antisymmetric under the interchange of the coordinates of any of the bound atomic electrons with the coordinates of the continuum electron. This exchange contribution would be zero if the initial bound electron states were orthogonal to the final continuum electron states. The effect of antisymmetrization is to multiply the allowed shape factor by a quantity that is of the order of $1-2Z^{-1}$ for energies less than one-half the binding energy of a K electron in the initial atom. This exchange correction is negligible for larger values of the energy of the continuum electron.

With respect to total decay rates, the overlap effect dominates for small Z and the exchange effect is more important for large Z.

Recent precision experiments[4] have shown that the Ar^{37} L-to-K capture ratio is about 22% larger than the value expected on the basis of the usual theory,[5,6] which does not include the whole atom in the description of initial and final atomic states. Following a suggestion by Benoist-Gueutal,[1,3] Odiot and Daudel[3] used wave functions for the whole atom to calculate the Ar^{37} L to K capture ratio. The prediction of Odiot and Daudel is in excellent agreement with the recent precision experiments,[4] provided that correlations due to the use of Hylleraas-like atomic wave functions do not play an important role in the decay process.[3,4]

We derive an approximate simple formula for the exchange correction to L to K ratios. This approximate formula yields results for Ar^{37} in agreement with experiment and with the more complicated calculation of Odiot and Daudel. All additional correlations are shown to be unimportant.

The effect of exchange on the total capture lifetimes of Be^7 and Ar^{37} is negligible and is at most a few percent in all other cases. Exchange effects are more important for L to K ratios than for total capture lifetimes because L capture is usually only a small part of the total capture probability and because the increase in the L-capture rate due to exchange is approximately cancelled by the decrease in the K-capture rate. For values of Z greater than 20 or 30, the effect of exchange on total electron capture probabilities is larger than the effect of imperfect overlap.

In Sec. II we discuss a formalism that is useful in isolating the effect of the change in nuclear charge from other small effects such as screening and finite nuclear size. We apply this formalism in Sec. III to the calculation of the effect of imperfect overlap on the allowed positron spectrum shape and on total positron decay rates. In Sec. IV we study the effect of exchange, as well as imperfect atomic overlap, on electron emission probabilities. We investigate, in Sec. V, the effect of exchange and imperfect overlap on total electron capture lifetimes and on L to K ratios.[6a]

II. ATOMIC HAMILTONIANS AND THE "GOLDEN RULE"

By choosing suitable unperturbed Hamiltonians for initial and final atomic states, we can separate the effect of the change in nuclear charge from other small effects such as electron screening,[5,7] direct collisions of emitted electrons with bound electrons,[8,9] and finite nuclear size.[5,10] These suitable Hamiltonians must include electrostatic interaction among all bound and continuum electrons that are present; they must also include a Coulomb potential due to a nucleus of finite size.

The Hamiltonian for the radioactive system is

$$H=H_0+H_\beta, \tag{2}$$

[2] H. M. Schwarz, Phys. Rev. **86**, 195 (1952); see also R. Serber and H. S. Snyder, *ibid.* **87**, 153 (1952).

[3] P. Benoist-Gueutal, Compt. Rend. **230**, 624 (1950); S. Odiot and R. Daudel, J. Phys. Radium **17**, 60 (1956).

[4] See, for example, A. G. Santos-Ocampo, and D. C. Conway, Phys. Rev. **120**, 2196 (1960); C. Manduchi and G. Zannoni, Nuovo Cimento **22**, 462 (1961); P. W. Dougan, H. W. D. Ledingham, and R. W. P. Drever, Phil. Mag. **7**, 475 (1962).

[5] H. Brysk and M. E. Rose, Rev. Mod. Phys. **30**, 1169 (1960).

[6] R. Bouchez and P. Depommier, Rept. Progr. Phys. **23**, 395 (1960).

[6a] The exchange correction to the L-to-K capture ratio has now been calculated for a number of decays; this correction removes a systematic discrepancy between precision experiments and the usual electron capture theory. See J. Bahcall, Phys. Rev. Letters **9**, 500 (1962).

[7] J. R. Reitz, Phys. Rev. **77**, 10 (1950). Other useful references are given in this paper.

[8] A. Migdal, J. Phys. (U.S.S.R.) **4**, 449 (1941).

[9] E. L. Feinberg, J. Phys. (U.S.S.R.) **4**, 424 (1941).

[10] M. E. Rose and D. K. Holmes, Phys. Rev. **83**, 190 (1951).

where the unperturbed Hamiltonian H_0 includes nuclear, atomic, and neutrino parts:

$$H_0 \equiv H_N + H_A + H_\nu. \tag{3}$$

The beta-decay interaction is described by H_β. The total Hamiltonian H operates on state vectors that specify all relevant nuclear, atomic, and leptonic variables.

The usual derivations[11] of "The Golden Rule"[12] make use of time-dependent perturbation theory and require that H_0 be the same for initial and final states. Since the total number of nucleons is unchanged by beta decay, the nuclear Hamiltonian H_N, when written in isotopic spin notation, is the same in initial and final states. The number of electrons, however, changes in electron capture or electron emission as does also the electron-nucleus Coulomb interaction. We want H_0 to provide for the electrostatic interaction among all electrons that are present.

This can be done conveniently by writing H_A in second-quantized form. We have

$$H_A = \sum_{i,j} h_{i,j} a_i^\dagger a_j + \tfrac{1}{2}\alpha \sum_{ij,kl} u_{ij,kl} a_i^\dagger a_j^\dagger a_l a_k, \tag{4}$$

where $a_i^\dagger$ (a_j) are operators that create (destroy) electrons in the single-particle states i (or j). The matrix elements in Eq. (4) are defined by

$$h \equiv \boldsymbol{\alpha}\cdot\mathbf{p} + \beta - \alpha N_p/r, \tag{5a}$$

with

$$h_{i,j} \equiv (\varphi_i, h\varphi_j), \tag{5b}$$

and

$$u_{ij;kl} \equiv \left(\varphi_i(1)\varphi_j(2), \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|}\varphi_k(1)\varphi_l(2)\right). \tag{6}$$

The quantity h is the single-particle Dirac operator and is written, for simplicity, with a Coulomb interaction due to a point nucleus. The operator N_p is the proton number operator and is equal to Z in the initial state and $Z\pm1$ in the final state; α is the fine structure constant. We use throughout this paper units in which $\hbar = m_e = c = 1$. The mutual electrostatic interaction between electrons is described by u and the second term of Eq. (4). The electrostatic interaction between, for example, an emitted electron and any bound electron is automatically included in (4). In applications, we shall use wave functions that treat the mutual electrostatic interaction among electrons by a self-consistent field approximation.

Equation (4) is valid for any complete set of one-particle basis functions $\varphi_i(\mathbf{x})$; we shall find different sets are appropriate for electron capture and for electron emission. We shall also see (Sec. V) that the second-quantized version of H_A is most convenient for the computation of exchange terms in electron capture.

Positron decay can be treated by adding to H_A, given by Eq. (4), a term containing the Dirac positron Hamiltonian plus a term describing the electrostatic interaction of the positron with all electrons that are present. The positron terms are identical with the corresponding electron terms except for a change in sign of the electrostatic contributions and the replacement of electron creation and annihilation operators by positron creation and annihilation operators.

The neutrino field is easily written in second-quantized form[13] and will not be discussed here.

The atomic Hamiltonian given in Eq. (4) can be used for both initial and final states. It will be convenient to refer to the version of H_A for which N_p is equal to the initial nuclear charge as the initial atomic Hamiltonian and to refer to the version for which N_p is equal to the final nuclear charge as the final atomic Hamiltonian.

The usual derivations of the "Golden Rule" are valid if the total Hamiltonian is chosen as described above. The partial decay rate from an initial state i to a final state f is then given by

$$\lambda = 2\pi |\langle f|H_\beta|i\rangle|^2 \delta(E_i - E_f). \tag{7}$$

The initial and final atomic states are included in $|i\rangle$, $|f\rangle$ and the initial and final atomic energies are included in E_i, E_f. In order to find total decay probabilities, λ of Eq. (7) must be summed over all possible final atomic states and averaged over initial atomic states.

III. POSITRON EMISSION

A. General Results

Let the initial state of a positron-emitting atom be represented by

$$|i\rangle \equiv |G; k\rangle \tag{8}$$

and the final state by

$$|f\rangle \equiv |A'; e^+; \nu; k'\rangle, \tag{9}$$

where $|G\rangle$ is the state vector of the initial atom in its ground state and $|A'\rangle$ is the state vector for the final atom in any one of its possible states. Final states in which some of the originally bound electrons are shaken off[8,9,14] are included in the set $\{A'\}$. The initial and final nuclear variables are represented by k and k' and the positron and neutrino variables are represented by e^+ and ν.

[11] See, for example, H. S. Snyder, Phys. Rev. **83**, 1154 (1951), or L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1949).

[12] E. Fermi, *Nuclear Physics* (University of Chicago Press, Chicago, 1950), rev. ed., p. 142. Fermi uses "Golden Rule No. 2" to refer to the transition probability formula.

[13] The second-quantized notation introduced in this section is explained and justified by S. S. Schweber, *An Introduction to Quantum Field Theory* (Row, Peterson and Company, Evanston, Illinois, 1961), Chap. 6, p. 121.

[14] See, for example, J. S. Levinger, Phys. Rev. **90**, 11 (1953), and A. Winther, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **27**, No. 3 (1952).

The allowed positron spectrum can be calculated in the usual way[15] using Eqs. (7), (8), and (9). We find

$$\lambda(p)=\frac{G_V^2\xi}{4\pi^3}p^2dp\int_0^\infty dp\ q^2M, \tag{10}$$

where G_V is the vector coupling constant, ξ is the usual allowed combination of nuclear matrix elements,[15]

$$\xi\equiv\langle 1\rangle^2+C_A^2/C_V^2\langle\sigma\rangle^2, \tag{11}$$

and

$$M\equiv\sum_{A',\sigma'}\langle A';e^+|\psi_e(0)|G\rangle(1+\gamma_5)\times\langle G|\psi_e^+(0)|A';e^+\rangle\delta(E_i-E_f). \tag{12}$$

In Eq. (12), $\psi_e(0)$ is the electron field operator evaluated at the nucleus; ψ_e destroys electrons and creates positrons. The summation in Eq. (12) is over all final states, A', of Z electrons in the presence of a nucleus of charge $Z-1$ and over the spin projection σ' of the emitted positron.

The difference between initial and final energies of the radioactive system is given by the following equation:

$$E_i-E_f=W_0-W-q+E(G)-E(A')=0. \tag{13}$$

In Eq. (13), W_0 is the difference between initial and final *nuclear* masses, W is the total electron energy including its rest mass, q is the neutrino energy, and $E(G)$ and $E(A')$ are the initial and final atomic energies. As used here, $E(G)$ and $E(A')$ represent only atomic binding energies and do not contain the rest masses of the bound electrons.

Since only the positron part of ψ_e contributes to M, it is easy to show that

$$M=2F(-Z,W)\sum_{A'}\delta(E_i-E_f)|\langle A'|G\rangle|^2, \tag{14}$$

where $F(-Z,W)$ is the familiar Fermi function for positrons.[15] If the energy conserving delta function in Eq. (14) were independent of A', we could immediately use closure to reduce M to the usual expression

$$M^0=2F(-Z,W)\delta(E_i^0-E_f^0). \tag{15}$$

Equation (15) leads to the usual allowed positron spectrum shape[15] when M^0 is inserted in Eq. (10).

Thus the effect we are investigating appears in the formalism as a consequence of energy conservation. In order to separate out phase-space dependence upon the atomic energy release, we introduce the difference,

$$\Delta E=E(G)-E(G'), \tag{16}$$

of the ground-state energies of initial and final atoms. The total energy difference can now be written

$$E_i-E_f=(E_i^0-E_f^0)+[E(G')-E(A')], \tag{17}$$

where

$$E_i^0-E_f^0=W_0-W-q+\Delta E, \tag{18}$$

and $E(G')-E(A')$ is the negative of the excitation energy of the final atom. The maximum positron kinetic energy, $E_{\max}$, obtained from equating $E_i^0-E_f^0$ to zero is

$$E_{\max}=W_0+\Delta E-1. \tag{19}$$

The quantity $E_{\max}+2mc^2$ is usually called the Q value for positron decay.[16]

Using the definitions for the energy differences adopted in Eqs. (17) and (18), we can write

$$\delta(E_i-E_f)=\delta(E_i^0-E_f^0)+\delta'(E_i^0-E_f^0)[E(G')-E(A')]+\delta''(E_i^0-E_f^0)[E(G')-E(A')]^2/2+\cdots, \tag{20}$$

where $\delta'(x)$ is the derivative of the Dirac delta function with respect to x. The Taylor series expansion of the delta function is easily justified along the lines described, for example, by Lighthill[17] or by showing that for specific spectrum shapes Eq. (20) is equivalent to the binomial expansion of the neutrino energy defined by Eq. (13).

When the Taylor series expansion of the delta function is substituted in the definition of M, the sum over all final atomic states can be carried out by means of the closure relation

$$\sum_{A'}|\langle A'|G\rangle|^2=1. \tag{21}$$

We find for the spectrum shape

$$\lambda(p)\cong\lambda^0(p)[1+\lambda^1(p)/\lambda^0(p)], \tag{22}$$

where

$$\lambda^0(p)\equiv(G_V^2\xi/2\pi^3)dp\ p^2F(-Z,W)q_0^2 \tag{23}$$

is the usual allowed positron spectrum and

$$\lambda^1(p)/\lambda^0(p)\equiv-(2/q_0)\sum_{A'}|\langle A'|G\rangle|^2[E(A')-E(G')] \tag{24}$$

is the fractional correction to the usual spectrum. In the above equations, q_0 is the neutrino energy obtained by setting $E_i^0-E_f^0$ equal to zero in Eq. (18). The summation over A' represents the average excitation energy of the final atom.[1,2,18]

Only the first two terms in the Taylor series expansion of the delta function were included in the derivation of Eqs. (22) to (24). The justification for the omission of higher order terms will become obvious when the magnitude of the first-order correction is calculated for typical cases.

The fractional correction to the total positron decay

[15] E. J. Konopinski, Ann. Rev. Nucl. Sci. **9**, 99 (1959).

[16] F. Ajzenberg-Selove and T. Lauritsen, Nucl. Phys. **11**, 1 (1959).

[17] M. J. Lighthill, *Introduction to Fourier Analysis and Generalised Functions* (Cambridge University Press, London, 1958).

[18] R. F. Christy, Nucl. Phys. **22**, 301 (1961).

rate can be calculated by integrating Eq. (22) over all positron momenta. One finds

$$\lambda \cong \lambda^0[1+\lambda^1/\lambda^0], \tag{25}$$

where λ^0 is the usual total decay rate and

$$\lambda^1/\lambda^0 = -(R/E_{\max}) \times \sum_{A'} [E(A')-E(G')]|\langle A'|G\rangle|^2, \tag{26}$$

with

$$\frac{R}{E_{\max}} \equiv 2\frac{\int\int dpdq\, p^2q F(-Z,W)\delta(E_i^0-E_f^0)}{\int\int dpdq\, p^2q^2 F(-Z,W)\delta(E_i^0-E_f^0)}. \tag{27}$$

Since $E(A')$ is greater than or equal to $E(G')$, the imperfect overlap of atomic states decreases the decay probabilities $\lambda(p)$ and λ from their usual values $\lambda^0(p)$ and λ^0.

B. Evaluation of Fractional Corrections

The summation over A' can be performed in the usual way[1,2]:

$$\begin{aligned}\sum_{A'} [E(A')-E(G')]|\langle A'|G\rangle|^2 &= E(G)-E(G')+\langle G|\{H_0'-H_0\}|G\rangle \\ &= E(G)-E(G')+\langle G|\textstyle\sum_{i=1}^{Z}(\alpha/r_i)|G\rangle.\end{aligned} \tag{28}$$

In Eqs. (28), $H_0(H_0')$ is the initial (final) atomic Hamiltonian. Since[19,20]

$$\partial E(G)/\partial Z = -\langle G|\textstyle\sum_{i=1}^{Z}(\alpha/r_i)|G\rangle, \tag{29}$$

we find

$$\sum_{A'} [E(A')-E(G')]|\langle A'|G\rangle|^2 \cong -\tfrac{1}{2}[\partial^2 E(G)/\partial Z^2]. \tag{30}$$

The derivative of the ground-state energy with respect to Z, $\partial E(G)/\partial Z$, has been given for light atoms by Allard[21] in a modified form of the Fermi-Thomas result and, for heavy atoms, Foldy[20] has discussed the Hartree-Fock predictions of $\partial E(G)/\partial Z$. When the results of Allard and Foldy are differentiated with respect to Z, one finds

$$\begin{aligned}-\partial^2 E(G)/\partial Z^2 &= 49\, Z^{1/3}\text{ eV}, \quad Z<10 \\ &= 46\, Z^{2/5}\text{ eV}, \quad Z>10.\end{aligned} \tag{31}$$

Equation (31) is not valid for hydrogen, but should be accurate to 10 or 20% for other atoms.

Figure 1 was obtained from Eq. (31) by joining smoothly the two branches of $-\partial^2 E(G)/\partial Z^2$ vs Z at Z equal to ten. This figure shows that $-\partial^2 E(G)/\partial Z^2$ is a monotonically increasing function of Z that rises slowly from about 50 eV for the lightest atoms to about 300 eV for the heaviest atoms.

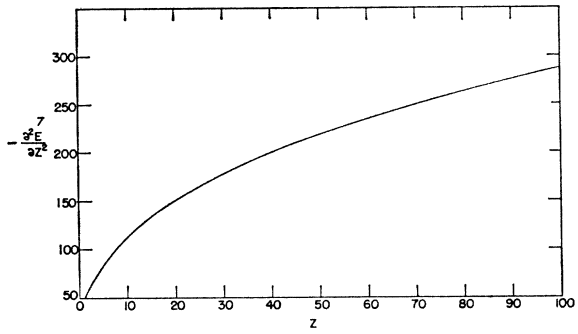

FIG. 1. Plot of $-\partial^2 E(G)/\partial Z^2$ vs Z.

The quantity R that occurs in Eqs. (26) and (27) can be calculated in two limiting cases; we find

$$\begin{aligned}R/E_{\max} &\cong 7/2 \quad \text{if } \langle 2\pi\alpha ZW/p\rangle_{\text{av}} \ll 1, \\ &\cong 2\pi\alpha Z(2/E_{\max})^{1/2} \quad \text{if } \pi\alpha Z(2/E_{\max})^{1/2} \gg 1.\end{aligned} \tag{32}$$

For actual positron decays, the two expressions given above for R do not differ much from each other. This shows that R is not a sensitive function of energy or nuclear charge in the region of interest.

C. Interpretation

The results of the previous sub-sections show that

$$\begin{aligned}\lambda(p) &\cong \lambda^0(p)[1-(1/q_0)|\partial^2 E(G)/\partial Z^2|] \\ &\equiv \lambda^0(p)[1-2\bar{E}_{\text{ex}}/q_0],\end{aligned} \tag{33}$$

and

$$\lambda \cong \lambda^0[1-R\bar{E}_{\text{ex}}/E_{\max}], \tag{34}$$

where

$$q_0 \equiv W_0 - W + \Delta E, \tag{35}$$

and the average excitation energy of the final atom is[1,2,18]

$$\bar{E}_{\text{ex}} \cong -\tfrac{1}{2}\partial^2 E(G)/\partial Z^2. \tag{36}$$

The spectrum given by Eq. (33) is the same as would be obtained if one ignored atomic overlap and replaced, in the usual theory, ΔE by $\Delta E+\bar{E}_{\text{ex}}$. The first-order effect of the imperfect overlap of atomic states is thus a shift of the positron spectrum, by an amount equal to the average excitation energy of the final atomic states.

One may be tempted to ignore imperfect overlap and regard the spectrum shift by $\bar{E}_{\text{ex}}$ as due only to the decrease in available energy for the emitted positron when the excitation of the final atom is taken into account. This interpretation is not consistent, however, since the probability for a transition to an excited state of the final atom is zero if the imperfect overlap of atomic states is not considered. Moreover, the definition for $(E_i^0-E_f^0)$ that is given in Eq. (18) is the one that is actually used in tabulations.

If the conventional definition of $E_i^0-E_f^0$ that was given in Eq. (18) were changed by adding $\bar{E}_{\text{ex}}$ to ΔE, then the first-order correction to $\lambda(p)$ would be zero

[19] R. P. Feynman, Phys. Rev. **56**, 340 (1939).
[20] L. Foldy, Phys. Rev. **83**, 397 (1951).
[21] G. Allard, J. Phys. Radium **9**, 225 (1948).

and the lowest-order correction would be proportional to the second derivative of the energy-conserving delta function. In this case,

$$\lambda^2(p)/\lambda^0(p)\cong(1/6q_0^2)[\partial^3E(G)/\partial Z^3], \quad (37)$$

which is completely negligible.

D. Applications

The discussion in the previous subsection shows that the main effect of atomic overlap on the positron spectrum is a shift of the end-point energy by at most a few hundred electron volts. This shift is less than the experimental error in almost all current experiments and hence can be ignored.

Some approximate results for total positron lifetimes are shown in Table I. The results are approximate since neither of the extremes described by Eq. (32) applies to the cases considered and, thus, R was not determined accurately. Table I and Eq. (34) show that the overlap effect on total lifetimes is less than a few tenths of one percent for most positron decays. This shift in the positron lifetime is within current experimental accuracy but is far too small to be significant in the determination of experimental nuclear matrix elements. In particular, the important O^{14} matrix element is affected by less than one-tenth of one percent.

The overlap effect would be important only for very low-energy decays, lower energy decays than those listed in Table I. However, very low-energy positron decays are much less likely than electron capture because of the additional mc^2 of energy available in the electron-capture process. We see in Sec. V that the overlap effect is negligible in electron capture if positron emission is energetically possible. Thus the change in the total disintegration rate of a given nucleus is negligible for very low-energy positron decays.

However, it is conceptually possible to test the correctness of the formulas developed in this paper by measuring accurately the tiny positron to electron capture ratio for some very low-energy positron emitters. The required experimental accuracy in the ratio would be of the order of 1 or 2% and, hence, this experiment is difficult.

TABLE I. Overlap effect for some moderate energy positron decays.

Parent isotope	Daughter isotope	E_{max} (MeV)	$-\lambda_1/\lambda_0$ (%)
Zn^{65}	Cu^{65}	0.325	0.1
Br^{77}	Se^{77}	0.342	0.1
In^{114}	Cd^{114}	0.400	0.1
La^{135}	Ba^{135}	0.300±0.150	0.2±0.1
Pa^{230}	Th^{230}	0.400	0.1

IV. ELECTRON EMISSION

A. General Discussion

The calculation of the effect of the change in nuclear charge on electron-emission rates is similar to the previously described calculation for positron emission, except that the antisymmetrization of the final-state vector between bound and continuum electrons must be taken into account.

The final atomic Hamiltonian generates a complete set of state vectors that we shall denote by $|A';e_c\rangle$. We limit ourselves to final states that contain at least one continuum electron, since the probability for the creation of a bound electron by the beta-decay process is usually too small to be of importance in terrestrial experiments.[22–24] The treatment given below can easily be extended to include the possibility of bound-state beta decay if radioactive systems are ever found for which this process is important.

The electron spectrum, allowing for a change in nuclear charge, is given by

$$\lambda(p)=\frac{G_V^2\xi}{4\pi^3}dp\,p^2\int dq\,q^2M, \quad (38)$$

where now

$$M\equiv\sum_{A',\sigma'}\langle A';e_c|\psi_e^\dagger(0)|G\rangle(1+\gamma_5)\times\langle G|\psi_e(0)|A';e_c\rangle\delta(E_i-E_f). \quad (39)$$

Equations (38) and (39) are identical with the corresponding positron Eqs. (10) and (12) except for the replacement in M, of e^+ by e_c and ψ_e by $\psi_e^\dagger$.

The effect of the antisymmetrization of the final-state vector between bound and continuum electrons can be exhibited explicitly by writing ψ_e as a sum of bound plus continuum parts. Let[24]

$$\psi_e\equiv\psi_B+\psi_C+\text{positron operators}=\sum_{b'}a_{b'}\varphi_{b'}+\sum_{c'}a_{c'}\varphi_{c'}+\text{positron operators}, \quad (40)$$

where $\varphi_{b'}$, $\varphi_{c'}$ form a complete set of one-electron bound and continuum wave functions referring to some approximate form of the final atomic Hamiltonian. The positron part of ψ_e does not contribute to electron emission probabilities. Thus,

$$M\equiv M_{C-C}+M_{B-C}+M_{C-B}+M_{B-B}, \quad (41)$$

where M_{C-C} is the same as the positron M except for the obvious substitutions mentioned previously. The new terms are

$$M_{B-C}\equiv\sum_{A',\sigma'}\langle A';e_c|\psi_B^\dagger(0)|G\rangle(1+\gamma_5)\times\langle G|\psi_C(0)|A';e_c\rangle\delta(E_i-E_f) \quad (42)$$
$$=M_{C-B}^\dagger,$$

[22] R. Daudel, M. Jean, and M. Lecoin, J. Phys. Radium 8, 238 (1947); Compt. Rend. 225, 290 (1948); R. Daudel, P. Benoist, R. Jacques, and M. Jean, *ibid.* 224, 1427 (1947).

[23] See also the description by Benoist-Gueutal in reference 1 of the work of M. Jean.

[24] J. Bahcall, Phys. Rev. 124, 495 (1961).

and

$$M_{B-B}=\sum_{A',\sigma'}\langle A';e_c|\psi_B^\dagger(0)|G\rangle(1+\gamma_5)\times\langle G|\psi_B(0)|A';e_c\rangle\delta(E_i-E_f). \quad (43)$$

Since the final atomic Hamiltonian is not the same as the initial atomic Hamiltonian, the final continuum electron states are not orthogonal to the initial bound electron states. It is this fact, plus the antisymmetry of the wave functions, that permits M_{B-C}, M_{C-B}, and M_{B-B} to be nonvanishing. The lack of orthogonality between final continuum electron states and initial bound electron states also permits the shaking off of[8,9,14] bound electrons into the continuum after the beta decay has occurred.

The change in the electron spectrum from its usual shape can be separated into two parts, one part due to the change in M_{C-C} from its usual value and another part due to the nonvanishing of M_{B-C}, M_{C-B}, and M_{B-B}. In subsection (B), we summarize changes in decay probabilities due only to M_{C-C}; in subsection (C) we investigate changes in decay probabilities due to other parts of M. The results are combined and applied in subsection (D).

B. Direct Emission

If only M_{C-C} contributes to M, then the calculation of the electron spectrum is identical with the previously described calculation of the positron spectrum and the fractional correction to the usual allowed spectrum is again given by Eq. (33). The fractional correction, analogous to Eq. (34), to the *total* electron decay rate is

$$\lambda^1/\lambda^0\cong(R'/2E_{\max})(\partial^2E/\partial Z^2) \quad (44a)$$

$$\equiv-(R'/E_{\max})\bar{E}_{\rm ex}, \quad (44b)$$

where R' is the only new quantity appearing in Eqs. (44). The ratio R' differs from the positron ratio R [Eq. (27)] only in that $F(-Z,W)$ is replaced everywhere by $F(+Z,W)$.

We can evaluate R' for the same limiting cases that were discussed in connection with R; we find:

$$R'/E_{\max}=7/2 \quad \text{if} \quad \langle 2\pi\alpha ZW/p\rangle_{\rm av}\ll 1, \quad (45a)$$

$$=3 \quad \text{if} \quad \pi\alpha Z(2/E_{\max})^{1/2}\gg 1. \quad (45b)$$

Equations (45) show that $R'/E_{\max}$ changes slowly with energy and nuclear charge. If the condition for the validity of Eq. (45a) is satisfied, then the ratio λ^1/λ^0 is the same for both electron and positron decays; this must be true since the Coulomb distortion of the continuum wave functions is negligible if (45a) obtains.

C. Exchange Emission

1. Approximate Expressions

We now investigate terms involving the bound-state part of the electron field operator, i.e., M_{B-C}, M_{C-B}, and M_{B-B}. These terms are easiest to interpret physically if we consider only the largest contributions,

$$M_{B-C}\cong-\delta(E_i^0-E_f^0)\langle e_c|1s\rangle|\langle G|G'\rangle|^2\langle 1s'|1s\rangle^{-1}\times\sum_{\sigma'}\varphi_{1s'}^\dagger(0)(1+\gamma_5)\varphi_C(0) \quad (46a)$$

$$\cong-2\delta(E_i^0-E_f^0)\langle e_c|1s\rangle\varphi_{1s'}^\dagger(0)\varphi_C(0) \quad (46b)$$

$$\cong M_{C-B}^\dagger, \quad (46c)$$

and, in the same approximation,

$$M_{B-B}\cong 2\delta(E_i^0-E_f^0)|\varphi_{1s'}(0)|^2|\langle e_c|1s\rangle|^2. \quad (47)$$

The overlap integral $\langle e_c|1s\rangle$ enters Eqs. (46) with a minus sign due to the antisymmetry of the wave functions. The way the minus sign arises can be seen most easily by representing $|G\rangle$ and $|G'\rangle$ by simple Slater determinants.

The $1s$ electrons provide the largest contribution to M_{B-C} and M_{B-B} because

$$|\varphi_{ns}(0)|^2\cong\pi^{-1}(\alpha Z/n)^3, \quad (48)$$

where n is the principal quantum number. We have neglected in Eqs. (46) and (47) the imperfect overlap of analogous atomic states, i.e., we have set $\langle G|G'\rangle$ and $\langle 1s|1s'\rangle$ equal to one. This approximation is justified, as we shall see later, since M_{B-C} and M_{B-B} are small themselves. We have also neglected relativistic effects since $\langle e_c|1s\rangle$ is negligible for continuum electron energies much in excess of the binding energy of a K electron.

The above expressions for M_{B-C} and M_{B-B} should be compared with the usual approximate expression for M_{C-C},

$$M^0\cong 2\delta(E_i^0-E_f^0)|\varphi_C(0)|^2=2\delta(E_i^0-E_f^0)F(Z,W)/V. \quad (49)$$

2. Interpretation

In the usual expression for M_{C-C} that is given by Eq. (49), $2|\varphi_C(0)|^2$ is a measure of the probability that a continuum electron e_c is created, at the nucleus, in the presence of the final atom. Similarly, $2|\varphi_{1s'}(0)|^2$, which appears in M_{B-B}, is a measure of the probability that a bound $1s'$ electron is created in the final atomic state. The term $|\langle e_c|1s\rangle|^2$ in M_{B-B} represents the probability that the initially present $1s$ electron is flipped into the final continuum state e_c, thus making room for the creation of the $1s'$ electron by the beta-decay process.

Hence, M_{B-B} represents the probability that an electron is created in the final $1s$ bound orbit while the initially present $1s$ electron is flipped, by the sudden change of nuclear charge, into the continuum. The process represented by M_{B-B} is an exchange effect since M_{B-B} (and M_{B-C}) would be zero if antisymmetrization between the bound and continuum electrons in $|A';e_c\rangle$ were not taken into account. It is for this reason that we call M_{B-B} and M_{B-C} exchange

terms[6a] and call M_{C-C}, which represents the creation of a continuum electron, a direct term.

The quantities M_{B-C} and M_{C-B} arise from the interference between the amplitude for the direct creation of a continuum electron and the amplitude for the creation of a bound electron with a continuum electron being shaken off.

3. Calculations

It is convenient to write

$$M \equiv M_{C-C}\left(1+2\frac{M_{B-C}}{M^0}\frac{M^0}{M_{C-C}}+\frac{M_{B-B}}{M^0}\frac{M^0}{M_{C-C}}\right) \quad (50a)$$

$$\cong M_{C-C}(1+2(M_{B-C}/M^0)+M_{B-B}/M^0), \quad (50b)$$

where M^0 is given by Eq. (49). From Eqs. (46), (47), and (49), we see that

$$M_{B-B}/M^0 \cong (M_{B-C}/M^0)^2, \quad (51)$$

and, therefore,

$$M \cong M_{C-C}(1+M_{B-C}/M^0)^2. \quad (52)$$

The ratio

$$M_{B-C}/M^0 = -\langle e_c|1s\rangle R_{1s}(0)(4\pi F/V)^{-1/2} \quad (53)$$

can be calculated approximately by assuming a Coulomb distorted plane wave for the wave function of e_c and a nonrelativistic bound Coulomb function for the radial function R_{1s}. The integrals can be calculated exactly using a method developed by Sommerfeld and Schur[25] in connection with the problem of K-shell photoionization. We find, with $|e_c\rangle$ normalized per unit energy:

$$\langle e_c|1s\rangle = (4\pi)^{-1/2}\langle g_{-1}|R_{1s}\rangle \quad (54a)$$

$$= [8F(Z+1, W)]^{1/2}x^{7/2}(\pi\alpha Z^2)^{-1}(1+x^2)^{-2} \times \exp[-(2y\cot^{-1}x)], \quad (54b)$$

where

$$x = \alpha Z p^{-1}, \quad (54c)$$

and

$$y = \alpha(Z+1)p^{-1}. \quad (54d)$$

The factor of $(4\pi)^{-1/2}$ in Eq. (54a) is due to the fact that $|1s\rangle$ projects out only the s-wave part of $|e_c\rangle$. Formulas (54) are in agreement with the result of Levinger[14] if one neglects the difference between x and y. It is necessary to multiply the right-hand side of Eqs. (54a) and (54b) by $(8\pi^3/Vp)^{1/2}$ in order to convert the normalization to one particle per unit volume. Equations (53) and (54) are written in a form that assumes $R_{1s}(0)$ and $\varphi_C(0)$ are real and positive; the ratio M_{B-C}/M^0 is independent of the initial relative phase of R_{1s} and φ_C.

[25] A. Sommerfeld and G. Schur, Ann. Physik 4, 409 (1930).

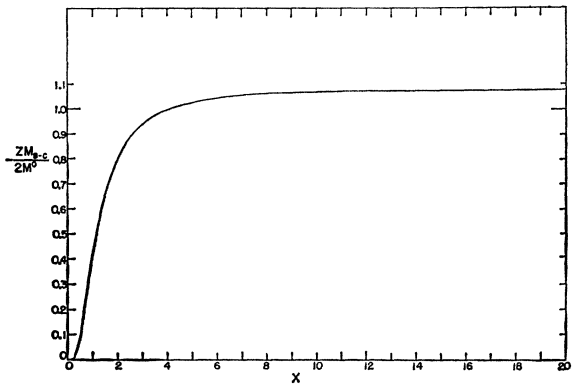

FIG. 2. Exchange contribution vs x.

Neglecting the difference between Z and $Z+1$ in the final answer, we find:

$$M_{B-C}/M^0 \cong -8Z^{-1}x^4(1+x^2)^{-2}\exp[-(2x\cot^{-1}x)]. \quad (55)$$

It is useful to note that x is the square root of the ratio of the K-electron binding energy in the initial atom to the kinetic energy of the final continuum electron, i.e.,

$$x \equiv (E_K/E)^{1/2}. \quad (56)$$

The ratio $-ZM_{B-C}/2M^0$ is plotted in Fig. 2. This ratio is approximately one for continuum electron energies less than one-half the binding energy of a K electron in the initial atom and decreases very rapidly for larger values of the energy of the continuum electron.

The allowed statistical spectrum is thus multiplied by a factor that is approximately[26]

$$1-(2/Z)\theta(E_K/2-E), \quad (57)$$

which is due to exchange decays. The factor (57) results in a change, $\Delta\lambda$, of the total decay rate that is given by

$$-\Delta\lambda/\lambda^0 \cong (2/Z)[1-(1-E_K/2E_{\max})^2] \quad \text{if } E_K/2E_{\max} \leq 1, \quad (58a)$$

$$\cong 2/Z \quad \text{if } E_K/2E_{\max} \geq 1. \quad (58b)$$

For a fixed value of $E_{\max}$, $-\Delta\lambda/\lambda^0$ increases with Z for light nuclei because E_K is of the order of $10Z^2$ eV. The K-binding energy can exceed two $E_{\max}$ for heavy nuclei and, in this case, $-\Delta\lambda/\lambda^0$ decreases with Z.

It is important to note that the low-energy electron emission probability is decreased, due to exchange, by an amount that is much greater than the well-known probability that an electron will be shaken off, after the decay process occurs, with a significant amount of energy.[14]

[26] This factor underestimates the correction somewhat since it neglects the effect of exchange between e_c and other s electrons. The function $\theta(x)$ is equal to zero for x negative and is equal to one for x positive.

D. Applications

The theoretical electron spectrum, including both overlap and exchange effects, is

$$\lambda(p)\cong\lambda^0(p)[1+\lambda^1(p)/\lambda^0(p)+\Delta\lambda(p)/\lambda^0(p)], \quad (59)$$

where

$$\lambda^1(p)/\lambda^0(p)\cong-2\bar{E}_{ex}/q_0, \quad (60)$$

and

$$\Delta\lambda(p)/\lambda^0(p)\cong-2Z^{-1}\theta(E_K/2-E). \quad (61)$$

The overlap effect given by Eq. (60) is important near the high-energy end of the electron spectrum and the exchange effect given by Eq. (61) is important for energies less than one-half the binding energy of the K electron in the initial atom.

The existence of the exchange process could be investigated by looking for deviations from the usual allowed shape in the low-energy part of an allowed electron spectrum. In order that an experimentally accessible part of the spectrum be affected, an isotope of rather high Z would have to be studied; an accuracy of the order of 1% would then be required in order to detect the predicted deviation from the usual allowed shape.[27] Ideally, one would want to study an isotope whose allowed low-energy electron decay is uncontaminated by internal conversion electrons or competing branches. Unfortunately, such ideal isotopes are rare and, moreover, there are well-known experimental difficulties in making accurate measurements on the low-energy part of the beta spectrum.

The percentage change in the total decay rate, λ^1/λ^0, due to the overlap effect is tabulated in Table II for some low-energy electron decays. These approximate values have been calculated by making use of Eq. (60) and Fig. 1. The results show that the overlap effect can change allowed beta-decay lifetimes by as much as 2%, although a more typical value is less than a few tenths of one percent.

Approximate values for the effect of exchange on these total decay rates are also listed in Table II; the exchange effects were calculated by means of Eqs. (58). In deriving Eqs. (58), a nonrelativistic wave function was used for the bound $1s$ electron and this usually leads to errors of the order of α^2Z^2 [28]; the values of $\Delta\lambda/\lambda^0$ for high Z are, therefore, very crude. A number of low-energy electron decays that are known to have maximum electron energies less than two E_K are not included in Table II because it was not established that they were allowed decays.

TABLE II. Overlap and exchange effects for some low-energy electron decays.

Parent nucleus	Daughter nucleus	Q (MeV)	$-\lambda_1/\lambda_0$ (%)	$-\Delta\lambda/\lambda_0$
C^{14}	N^{14}	0.1567	0.1	0.0
Si^{32}	P^{32}	0.100±0.50	0.2±0.1	0.3±0.2
Ni^{63}	Cu^{63}	0.0669	0.4	0.5
Ru^{106}	Rh^{106}	0.039	0.9	2
Yb^{177}	Lu^{177}	0.160	0.2	1
Os^{191}	Ir^{191}	0.143	0.3	1
Pu^{241}	Am^{241}	0.021	2	2

Table II shows that the exchange effect on *total* lifetimes is usually greater than the overlap effect. Since the total transition probability is

$$\lambda\cong\lambda^0[1+\lambda^1/\lambda^0+\Delta\lambda/\lambda^0], \quad (62)$$

the net result of including both exchange and overlap effects is to decrease the total theoretical transition probability by a small amount.

V. ELECTRON CAPTURE

A. General Considerations

The probability per unit of time that an atom will capture any of its electrons and leave the daughter atom in the final state $|A'\rangle$ is given by

$$\lambda(A')=G_V^2\xi(2\pi)^{-1}q^2(A')M^\dagger(A')(1+\gamma_5)M(A'), \quad (63)$$

where

$$M(A')\equiv\langle A'|\psi_e(0)|G\rangle, \quad (64)$$

and

$$q(A')=W_0+1+(E(G)-E(A')-1). \quad (65)$$

The state vector $|A'\rangle$ refers to the final system of $Z-1$ electrons associated with a nucleus of charge $Z-1$; the energies $E(G)$ and $E(A')$ *include* in this section the masses of the electrons.

In order to evaluate $M(A')$, we write ψ_e as a sum of bound plus continuum parts. Then

$$M(A')\equiv\langle A'|\psi_B(0)|G\rangle+\langle A'|\psi_C(0)|G\rangle \quad (66a)$$

$$=\sum_{b'}\langle A'|a_{b'}|G\rangle\varphi_{b'}(0)+\sum_{c'}\langle A'|a_{c'}|G\rangle\varphi_{c'}(0). \quad (66b)$$

The functions $\varphi_{b'}$, $\varphi_{c'}$ form a complete set of one-electron wave functions for the initial atomic Hamiltonian.

The expression (66b) for $M(A')$ can be evaluated easily only if $|G\rangle$ is represented by Slater determinants of one-electron states, the same one-electron states that are described by $\varphi_{b'}$, $\varphi_{c'}$. If this assumption is not made, all terms of Eq. (66b) will contribute and one has to calculate an infinite series.

If $|G\rangle$ can be represented as a single Slater determinant of one-electron states, then

$$M(A')=\sum_{b=1}^{Z}\langle A'|a_{b'}|G\rangle\varphi_{b'}(0). \quad (67)$$

Additional bound states must be included in the

[27] The deviations that have been investigated by Langer and his coworkers are unrelated to the effect being discussed here, since the Langer deviations occur for both electron and positron emission and at higher energies than we are considering. See, for example, J. H. Hamilton, L. M. Langer, and D. R. Smith, Phys. Rev. 123, 189 (1961).

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

summation over b' if $|G\rangle$ is represented as a sum of several Slater determinants.

The binding energy of a K electron is of the order of 100 keV for the heaviest atoms and, hence, cannot be ignored even in zero order calculations. We define, therefore,

$$q(A')\equiv q(1s')+\Delta q(A'), \tag{68a}$$

where

$$q(1s')\equiv W_0+E(G)-E(G')-\epsilon(1s'), \tag{68b}$$

and

$$\Delta q(A')\equiv E(G')-E(A')+\epsilon(1s'). \tag{68c}$$

In Eqs. (68), $\epsilon(1s')$ is the (positive) binding energy of an electron in the K shell of the final atom. The binding energy of an electron in the *final* atom appears in Eqs. (68) because the hole left by electron capture is in the final atom. In almost all cases, the capture of a $1s$ electron is the most probable mode of decay and thus Eqs. (68) are useful definitions for the purpose of calculating λ^1/λ^0 by a closure approximation. The energy difference $-\Delta q(A')$ is the quantity most analogous to the atomic excitation energy that appeared in our discussion of electron and positron emission.

If one uses a single-particle representation of $|G\rangle$, the total electron capture rate can be written

$$\lambda=G_V^2\xi(2\pi)^{-1}\sum_{b_1,b_2,A'} q^2(A')\varphi_{b_1}^{\dagger}(0)(1+\gamma_5)\varphi_{b_2}(0) \times\langle G|a_{b_1}^{\dagger}|A'\rangle\langle A'|a_{b_2}|G\rangle. \tag{69}$$

Let

$$q(b')\equiv W_0+E(G)-E(G')-\epsilon(b'), \tag{70}$$

where $\epsilon(b')$ is the binding energy of an electron in the single-particle state b' of the final atom. Then

$$\lambda\cong\lambda^0[1+\lambda^1/\lambda^0+\Delta\lambda/\lambda^0], \tag{71a}$$

where

$$\lambda^0\equiv G_V^2\xi(2\pi)^{-1}\textstyle\sum_b q^2(b')|\varphi_b(0)|^2, \tag{71b}$$

$$\lambda^1\equiv G_V^2\xi\pi^{-1}q(1s')\textstyle\sum_b|\varphi_b(0)|^2[-\epsilon(1s')+\epsilon(b') +\sum_{A'}\Delta q(A')\langle G|a_b^{\dagger}|A'\rangle\langle A'|a_b|G\rangle], \tag{72}$$

and

$$\Delta\lambda\equiv q(1s')G_V^2\xi\pi^{-1}\sum_{b_1,b_2,A'}{}' \varphi_{b_1}^{\dagger}(0)\varphi_{b_2}(0)\Delta q(A') \times\langle G|a_{b_1}^{\dagger}|A'\rangle\langle A'|a_{b_2}|G\rangle. \tag{73}$$

The quantity λ^0 is the zero-order total electron-capture rate when the binding energy of an electron in the final atom is used in defining $q(b')$.

In deriving Eqs. (70) to (72), we have used the approximation

$$q^2(1s')-q^2(b')\cong 2q(1s')[\epsilon(b')-\epsilon(1s')]. \tag{74}$$

In subsection B, we calculate the fractional effect, λ^1/λ^0, of imperfect atomic overlap on total capture rates and in subsection C we calculate the fractional effect, $\Delta\lambda/\lambda^0$, of exchange terms. The results for total capture rates are summarized in subsection D. The exchange correction to the usual L to K capture ratio is calculated in subsection E; the calculated exchange correction is in excellent agreement with the experimental values for the Ar^{37} L to K ratio.

B. Atomic Overlap

The second term in the expression for λ^1, Eq. (72), can be simplified by applying closure; we find

$$\textstyle\sum_{A'}\Delta q(A')\langle G|a_b^{\dagger}|A'\rangle\langle A'|a_b|G\rangle =E(G')-E(G)+\epsilon(1s') +\langle G|a_b^{\dagger}a_bH_0-a_b^{\dagger}H_0'a_b|G\rangle, \tag{75}$$

where H_0, H_0' are the atomic Hamiltonians for initial and final states, respectively. The most convenient method for evaluating the expectation value that appears in Eq. (75) is to use the second quantized representation of the atomic Hamiltonians given in Eqs. (4) to (6). After some calculation, one finds

$$\langle G|a_b^{\dagger}a_bH_0-a_b^{\dagger}H_0'a_b|G\rangle =E_b^0+\partial E(G)/\partial Z+(1/r)_{b,b} +\textstyle\sum_{b'}\langle b'b|1/r_{12}|b'b\rangle, \tag{76}$$

where E_b^0 is the Dirac energy of the one-electron state $|b\rangle$ and $|bb'\rangle$ is the antisymmetric two-electron state formed from $|b\rangle$ and $|b'\rangle$.

Equation (76) can be greatly simplified by noting that[29]

$$E_b^0+\textstyle\sum_{b'}\langle b'b|1/r_{12}|b'b\rangle\cong-\epsilon(b)+1, \tag{77}$$

and

$$-\epsilon(b)+(1/r)_{b,b}\cong-\epsilon(b'). \tag{78}$$

Equation (78) follows from the Feynman identity given in Eq. (29).

From Eqs. (76) to (78), one finds

$$\langle G|a_b^{\dagger}a_bH_0-a_b^{\dagger}H_0'a_b|G\rangle\cong 1-\epsilon(b')+\partial E(G)/\partial Z. \tag{79}$$

With the help of Eqs. (71), (72), (75), and (79), one can then show that

$$\lambda^1/\lambda^0\cong[q(1s')]^{-1}\partial^2E(G)/\partial Z^2. \tag{80}$$

Equation (80) gives the effect on total electron capture rates of imperfect atomic overlap and is very similar to Eqs. (34) and (44) which give the overlap effect on total positron and electron rates.

C. Exchange Capture

The quantity $\Delta\lambda$ arises from exchange effects. The exchange origin of $\Delta\lambda$ can be established by examining the approximate contribution to $\Delta\lambda$ of the term in which, for example, b_1 represents the $1s$-electron state and b_2 represents the $2s$ state. In this case,

$$\lambda(1s,2s)\propto\varphi_{1s}^{\dagger}(0)\varphi_{2s}(0)\langle 2s'|1s\rangle. \tag{81}$$

The term $\lambda(1s,2s)$ represents the interference between the amplitude for the production of a hole in the K'

[29] J. C. Slater, Phys. Rev. 34, 1293 (1929).

shell by the destruction of a $1s$ electron in the initial atom [factor $\varphi_{1s}^{\dagger}(0)$] and the amplitude for the same process occurring by the destruction of a $2s$ electron [factor $\varphi_{2s}(0)$] with a $1s$ electron being flipped into the $2s'$ shell by the change of nuclear charge (factor $\langle 2s'|1s\rangle$). A similar term $\lambda(2s,1s)$ represents the interference between the amplitude for the production of a $2s'$ hole by the destruction of a $2s$ electron and the amplitude for the same result occurring by the destruction of a $1s$ electron with the $2s$ electron jumping into the $1s'$ level.

One can show, by the same procedure that was used to derive Eqs. (75) and (76), that

$$\Delta\lambda\cong q(1s')G_V^2\xi\pi^{-1}\sum_{b_1,b_2}{}'\,\varphi_{b_1}^{\dagger}(0)\varphi_{b_2}(0)\times[(1/r)_{b_1,b_2}+\textstyle\sum_{b'}\langle b'b_2|1/r_{12}|b'b_1\rangle]. \quad (82)$$

If we retain only the largest term in $\Delta\lambda$ and in λ^0, we find

$$\frac{\Delta\lambda}{\lambda^0}\cong\frac{4}{q(1s')}\frac{R_{2s}(0)}{R_{1s}(0)}\times\left[\left(\frac{1}{r}\right)_{2s,1s}+\sum_{b'}\left\langle b'2s\left|\frac{1}{r_{12}}\right|b'1s\right\rangle\right]. \quad (83)$$

The bracketed terms in Eq. (83) can be evaluated approximately by making use of screened hydrogenic wave functions. One finds that the term involving r_{12}^{-1} is negligible but that

$$(1/r)_{2s,1s}\cong 6Z \text{ eV}. \quad (84)$$

By making the additional approximation,

$$R_{2s}(0)/R_{1s}(0)\cong 3^{-1}, \quad (85)$$

we find

$$\Delta\lambda/\lambda^0\sim 8[q(1s')]^{-1}Z \text{ eV}. \quad (86)$$

D. Applications

The total electron capture rate is

$$\lambda\cong\lambda^0[1+\lambda^1/\lambda^0+\Delta\lambda/\lambda^0]. \quad (87)$$

Overlap and exchange effects are of the opposite sign and, hence, they partially cancel each other in the total capture rate. A comparison of Eqs. (80), (86), and Fig. 1 shows that the overlap effect predominates for small Z and the exchange effect is more important for large Z. They are roughly equal for Z of the order of 20. The net effect on total lifetimes does not exceed a few percent for electron captures with $q(1s')$ greater than or of the order of 50 keV.

The atomic wave functions used in calculating $(1/r)_{2s,1s}$ are not accurate enough to provide more than an order of magnitude estimate of $\Delta\lambda$ and hence no detailed results are presented for the net effect of overlap and exchange on total capture rates. Accurate calculations could be performed by numerically integrating Hartree-Fock wave functions, but such elaborate calculations do not seem justified on the basis of the order of magnitude estimates provided by Eqs. (80) and (86).

Equations (80) and (86) are accurate enough, however, to show that overlap and exchange effects on the total lifetime of Be^7 are less than a tenth of one percent, since $q(1s')$ is several hundred keV for both Be^7 decay branches. This result is consistent with a recent analysis of the experimental Gamow-Teller matrix elements for Be^7 capture.[30] Benoist-Gueutal[1] estimated by another method that Be^7 capture would be inhibited by an amount less than or equal to 34% due to atomic overlap. Equations (80) and (86) also show that overlap and exchange effects have a negligible influence on the total electron capture rate of Ar^{37} for which $q(1s')$ is 814 keV.

E. L to K Ratios

1. General Discussion

The K-capture transition probability is given by Eq. (68) when the summation over A' is limited to states that have at least one hole in their K shells. The L_{I} transition probability is obtained by summing Eq. (68) over states A' that have filled K shells but no more than one $2s$ electron. Final atomic states that are missing both a K and an L_{I} electron give rise to very small transition probabilities; they can also be discriminated against experimentally.

A particular set of states dominates the K-capture summation. These states, which we represent by $|A'(1s')\rangle$, contain only one $1s'$ electron but otherwise have the same inner electron configuration as that of the initial atom. The outer electrons can be in any of the available one-electron states, including continuum states. The states $|A'(2s')\rangle$ dominate the L_{I} capture summation; these states contain only one $2s'$ electron but otherwise their inner electron configuration is the same as that of the initial atom. We neglect, for simplicity, L_{II} and higher captures; such captures are rare in the cases in which we are most interested.

We want to derive a simple expression for the L to K capture ratio including exchange. In order to do this, we make use of the following approximate formula:

$$\sum_{\{A'(2s')\}}\langle G|a_{1s}^{\dagger}|A'(2s')\rangle\langle A'(2s')|a_{2s}|G\rangle\cong-\langle 2s|1s'\rangle\langle 1s'|1s\rangle|\langle 1s'|1s\rangle\langle 2s'|2s\rangle|^2. \quad (88)$$

The overlap integral $\langle 2s|1s'\rangle$ enters Eq. (88) with a minus sign due to the antisymmetry of the total wave functions.

In deriving Eq. (88), we have made use of the completeness of the outer electron states $\{A'(2s')\}$. A relation similar to Eq. (88) in which the position of

[30] J. N. Bahcall, Phys. Rev. 128, 1297 (1962).

every $1s$ and $2s$ are interchanged relative to (88) can also be proved. We also note that

$$\langle 1s'|1s\rangle/\langle 2s'|2s\rangle \cong 1. \tag{89}$$

Using the approximations given in Eqs. (88) and (89), we find from Eq. (68) that

$$\frac{\lambda_{LI}}{\lambda_K}=\left(\frac{\lambda_{LI}}{\lambda_K}\right)^0\left[\frac{1-[2R_{1s}(0)/R_{2s}(0)]\langle 1s'|2s\rangle}{1-[2R_{2s}(0)/R_{1s}(0)]\langle 2s'|1s\rangle}\right], \tag{90}$$

where the usual capture ratio is given by[5,6]

$$(\lambda_{LI}/\lambda_K)^0=[q^2(2s')/q^2(1s')][R_{2s}^2(0)/R_{1s}^2(0)]. \tag{91}$$

In writing Eq. (91), we have made use of the usual convention that all $R_{ns}(0)$ and $R_{ns'}(0)$ are real. The exact expression obtained from Eq. (73) is, of course, independent of all phase conventions.

2. *L to K Applications*[6a]

Simple numerical integrations of the nonrelativistic Hartree-Fock wave functions[31] for the Ar and Cl atoms yield

$$\langle 1s'|2s\rangle=-0.0292, \tag{92a}$$

and

$$\langle 2s'|1s\rangle=+0.0252. \tag{92b}$$

The same set of Hartree-Fock wave functions yield the value:

$$R_{1s}(0)/R_{2s}(0)=3.522. \tag{93}$$

Also

$$q^2(2s')/q^2(1s')=1.007. \tag{94}$$

Substituting the above numbers in Eq. (90), we find for the predicted L_I to K ratio

$$\begin{aligned}\lambda_{LI}/\lambda_K &\cong (0.0813)(1.219),\\ &=0.099.\end{aligned} \tag{95}$$

We have used nonrelativistic Hartree-Fock wave functions to evaluate $R_{1s}(0)/R_{2s}(0)$, since nonrelativistic Hartree-Fock functions were used to evaluate the overlap integral $\langle 1s'|2s\rangle$. However, relativistic effects for the overlap integral should be of the order[28] of α^2Z^2 and, hence, only amount to 1 or 2% of this already small quantity. If one uses the relativistic value[32] for $R_{1s}(0)/R_{2s}(0)$ and the nonrelativistic overlap integral, one finds

$$\lambda_{LI}/\lambda_K \cong 0.100. \tag{96}$$

This theoretical value is probably accurate to better than 2%, since Eqs. (88) and (89) are well satisfied for argon.

The above results are in excellent agreement with the recent precision measurements of the L- to K capture ratio of Ar^{37}; the precision measurements yield the value[4] 0.100 ± 0.003.

If we add the exchange corrections to L and K capture that are given by Eqs. (90) and (92), we find an exchange correction to the total capture rate of about $0.004q^2$. The contribution of states other than $|A'(1s')\rangle$ and $|A'(2s')\rangle$ cancels this small residual exchange effect and makes the net exchange effect proportional to q [see Eq. (73)]. Since the magnitude of the correction due to states other than $|A'(1s')\rangle$ and $|A'(2s')\rangle$ is small, we were justified in ignoring these states in our discussion of the L to K ratio. An explicit calculation using the appropriate Hartree-Fock wave functions shows that Eq. (89) is accurate to a few tenths of one percent for the Ar^{37} decay.

Since the sum over all final states except $|A'(1s')\rangle$ and $|A'(2s')\rangle$ only yields a term of the order of a few tenths of one percent of the main term, any additional correlations due to the use of Hylleraas wave functions[3,4] must be small for Ar^{37} electron capture.

The exchange correction to the usual capture ratio decreases with Z, because $\langle 1s'|2s\rangle$ and $R_{1s}(0)/R_{2s}(0)$ decrease in magnitude with Z. This result also appears to be in agreement with experiment.[33] Detailed calculations are underway to determine accurately the effect of exchange for a number of isotopes whose L- to K capture ratios have been measured precisely.

Odiot and Daudel[3] first calculated, by using wave functions referring to the whole atom, the correction due to electron correlations for the Ar^{37} L to K ratio. They predicted an L to K ratio of 0.10, which has since been experimentally verified.

ACKNOWLEDGMENTS

I am grateful to Professor R. F. Christy for helpful suggestions. It is a pleasure to acknowledge stimulating conversations with Professor R. W. Kavanagh, Dr. F. C. Michel, Dr. S. B. F. Wahlborn, and Dr. M. Wellner. I am indebted to Professor R. B. Curtis for an enlightening discussion of the Golden Rule. The numerical calculations were performed by Barbara A. Zimmerman. I am grateful for her valuable assistance.

[31] D. R. Hartree, Proc. Roy. Soc. (London) **A156**, 45 (1933); D. R. Hartree and W. Hartree, *ibid.* **A166**, 450 (1938).

[32] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).

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