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Exchange effects in β decays of many-electron atoms

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The effect on the β spectrum of exchange between bound and continuum electrons is discussed for decay of a many-electron atom. Results of calculations of the exchange distortion of the β spectrum are presented using both a screened hydrogenic and a Hartree-Fock approximation for the electron wave functions. The results of the two approaches agree well and show that the inclusion of exchange leads to an enhancement of the β spectrum, particularly at low electron energy. This corrects an error in a previous calculation that found that exchange decreased the β intensity. For the low-energy β^- emitters ^{106}Ru and ^{241}Pu, the present calculation indicates that the enhancement due to exchange is of the order of several percent over much of the spectrum, becoming larger at very low electron energy. Exchange with 1*s* electrons dominates in the high-energy part of the spectrum, but exchange with *ns* ($n \geq 2$) electrons becomes significant in the low-energy region. The inclusion of exchange leads to an increase in the phase-space integral by 6.4% for ^{106}Ru and 7.5% for ^{241}Pu. Results are presented for exchange effects in other β spectra, including those of ^{14}C and ^{35}S, in which experimentally measured distortions have been interpreted as evidence for a heavy antineutrino of mass 17 keV. The distortions due to exchange are found to be significantly smaller than the measured distortions.

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I. INTRODUCTION

Interest in atomic effects on β-decay spectra has been revived recently because of the relevance to the interpretation of experiments that attempt to measure, or set limits on, the antineutrino mass from β spectra. Atomic distortion of the β spectrum has to be taken into account in order to extract any further distortion which may be due to antineutrino mass effects. Much theoretical work initially focused on the tritium β spectrum in the high-energy region to test the suggestion that the electron neutrino might have a mass of a few tens of eV [1–5]. More recently, attention has focused on the possibility of an antineutrino of rest mass 17 keV that may be emitted in 1% or 2% of β decays as first proposed by Simpson [6,7]. The effects of screening [8,9] and exchange [10] on the tritium β spectrum have been proposed as factors contributing to the experimentally measured distortion though these effects do not seem to account completely for the discrepancy. While the initial evidence for the 17-keV antineutrino came from distortions in the tritium β spectrum, there have been more recent reports of distortions in the β spectra of other isotopes, specifically ^{35}S [11,12] and ^{14}C [13], that also have been interpreted as supporting the heavy-neutrino hypothesis, though other groups have found no evidence for distortion of β spectra of ^{35}S [14] and ^{63}Ni [15].

The principal atomic effects on the β spectrum arise from screening of the β-particle wave function, exchange effects between bound and continuum electrons, and the distribution of electrons in final atomic states as a result of shakeup during the decay process. Screening effects in decays of many-electron atoms have been discussed by a number of authors [16,17]. In this article we examine exchange effects on the β spectra arising from decays of multielectron atoms. It is well known that the nonorthogonality between initial- and final-state orbitals leads to shakeup and shakeoff of atomic electrons in β^- decays. From this fact it follows that it is possible for the β electron to be created into an orbital of the daughter atom corresponding to one which was occupied in the parent atom, without violating the Pauli principle. β decay to a final state containing one continuum electron can then be viewed as occurring both by creation of the β electron in the continuum orbital (the direct process) or by creation into a bound orbital with an electron simultaneously making a transition to a continuum orbital of the daughter atom (the exchange process).

The effect of exchange on the β^- spectra of some low-energy β^- emitters has been estimated previously by Bahcall [18], who found that the amplitude for the exchange process interfered destructively with the direct process with the result that the inclusion of exchange between bound and continuum electrons led to a decrease in the probability of β^- emission, particularly at low electron energies. For ^{63}Ni (end-point energy, $E_0 = 67$ keV) it was predicted [18] that this effect would decrease the continuum decay constant by 0.5% relative to that calculated ignoring exchange. For ^{106}Ru (end-point energy, $E_0 = 39.4$ keV) and ^{241}Pu ($E_0 = 21$ keV) the exchange effect was predicted to reduce the decay constant by approximately 2%. Haxton [10] has performed a calculation of exchange effects for tritium decay. This indicated that exchange with 1s electrons enhanced the low-energy part of the β spectrum. This latter result would therefore appear to be in contradiction to Bahcall's earlier work though this point was not addressed in Haxton's article. One aim of the present article is therefore to resolve the discrepancy in the sign of the exchange terms. A second aim is to consider the effect of exchange with electrons other than 1s electrons. Bahcall argued that exchange processes involving 1s electrons should be considerably larger than those involving other electrons on the grounds that the size of the exchange process between a bound and continuum electron scales with the value of the bound electron wave function in the nuclear region where the β^- decay process occurs. However, this argument ignores the fact that the size of the exchange terms involving two electrons should also scale with the overlap integral between the bound and continuum wave functions. This overlap should be significantly larger for higher-lying bound s orbitals than for the 1s orbital. This would tend to offset the fact that the wave function for the 1s orbital in the nuclear region will be greater than for higher-lying s orbitals. Thus it is not clear that exchange with occupied higher-lying orbitals can be ignored. A secondary aim of this article is therefore to calculate the contribution due to exchange with bound electrons other than 1s electrons in decays of many-electron atoms. The exchange calculations in both [10] and [18] were based on ignoring screening effects on the electron wave functions. The validity of this approximation is not clear, particularly in the case of many-electron atoms, since the exchange terms which enter the expression for the intensity of the β spectrum at a particular energy depend upon overlap integrals between a bound electron wave function and a continuum electron wave function. These overlap integrals may receive significant contributions from regions of space in which screening effects play an important role. Thus a further aim of this paper is to evaluate the exchange effects on the spectrum using more accurate wave functions which take account of electron screening. Since the exchange process involves bound electrons there is a possibility that the process may be sensitive to the chemical state of the decaying atom. An additional aim of this paper is to estimate the magnitude of changes in the β-decay constant due to differences in the magnitude of the exchange process in different chemical states of the decaying atom.

II. THEORY

A. General formalism

We consider the β decay by an allowed transition of a general atom of nuclear charge $(Z-1)$ containing N electrons which are defined by the state label γ. The final state is a nucleus of charge Z, an antineutrino and $(N+1)$ electrons whose state label will be denoted γ'. The β decay process may be represented

$$(Z-1,A)+Ne^- \rightarrow (Z,A)+(N+1)e^- + \bar{\nu} \; . \quad (1)$$

In Ref. [19] we derived a general expression for the decay rate of a multielectron atom to a given final γ' state by an allowed β transition. In this work we focus on final states composed of at least one continuum electron and N other electrons. The dominant states produced in the β-decay process are those in which all these N other electrons are in bound states. If the different possible final states of the N bound electrons are denoted $\bar{\gamma}$ and the total number of electrons emitted with energies between E and $E+dE$ is denoted dI, then from Eq. (2.68) of [19], one has

$$\frac{dI}{dE}=\frac{C}{2(2J+1)}\sum_{\bar{\gamma}}\sum_{J'=J_{\min}}^{J+1/2}\sum_{\kappa_e=-1}^{1}\frac{W}{p}[W_0(\bar{\gamma})-W]^2\left|\langle N+1,\bar{\gamma},E,\kappa_e,J'\|\hat{b}^{\dagger}_{E,\kappa_e}\|N,\gamma,J\rangle U_{E,\kappa_e}(R)\right.$$
$$\left. +\sum_{A(\in\bar{\gamma})}\delta_{j_A 1/2}\langle N+1,\bar{\gamma},E,\kappa_e,J'\|\hat{b}^{\dagger}_{A}\|N,\gamma,J\rangle U_A(R)\right|^2 . \quad (2)$$

Here the operator $\hat{b}^{\dagger}_{E,\kappa_e}$ creates an electron in a continuum orbital of the final-state atom with kinetic energy E, total energy W, momentum p, and kappa quantum number κ_e. The energy of the electron, E, differs from its total energy W, merely in the omission of the rest mass term mc^2 so that $W=E+mc^2$. The quantity $U_{E,\kappa_e}(R)$ is one of the Dirac radial wave functions for this orbital and is defined such that $U_{E,\kappa_e}(R)=P_{Es'}(R)/R$ if $\kappa_e=-1$ and $U_{E,\kappa_e}(R)=Q_{E\bar{p}'}(R)/R$ if $\kappa_e=1$, where $P_{Es'}(R)$ is the large radial component of the Dirac wave function for an s-wave electron of kinetic energy E and $Q_{E\bar{p}'}(R)$ is the small radial component of the Dirac wave function for a $\bar{p}$-wave electron with this energy, evaluated at the nuclear radius R [19]. It should be noted that the $U_A(r)$ factors defined after Eq. (2.46) in Ref. [19] lack a factor of r in the denominator. The subsequent equations in [19] containing these factors are, however, correct as they stand. The quantity $U_A(R)$ is similarly defined as the appropriate component of the wave function for the subshell A belonging to the final $\bar{\gamma}$ state. The operator $\hat{b}^{\dagger}_{A}$ creates an electron in a final-state orbital with quantum numbers denoted by the label A. The quantities J and J' are the total angular momenta of the states γ and γ'. The reduced-matrix elements in Eq. (2) between the N-electron initial state γ and the $(N+1)$-electron final state γ' can be related to a matrix element between states of a given magnetic quantum number according to the Wigner-Eckart theorem as described in Ref. [19]. $W_0(\bar{\gamma})$ is the maximum β-particle energy (end-point energy) in the decay leading to the final state $\bar{\gamma}$ and is determined by energy conservation. The quantity $J_{\min}$, the minimum value of J', equals $J-\frac{1}{2}$ if $J\neq 0$ and equals $\frac{1}{2}$ if $J=0$. The factor C is a constant for a particular isotope, independent of electron energy.

If the reduced-matrix element terms in Eq. (2) involving the quantities $\hat{b}^{\dagger}_{A}$ are ignored then Eq. (2) gives the standard expression for the β intensity ignoring exchange effects. The term involving $\hat{b}^{\dagger}_{E,\kappa_e}$ is therefore called the direct term since it is associated with the probability for creating an electron directly into the continuum s and $\bar{p}$ orbitals of energy E with the atomic electrons effectively playing the role of spectators in the decay process. The terms in Eq. (2) involving $\hat{b}^{\dagger}_{A}$ are called exchange terms and are associated with the probability for an electron being created in the subshell A of the daughter atom with an electron simultaneously making a transition from an initial-state bound orbital (having the same quantum numbers as the orbital in which the electron is created) to a continuum orbital of energy E. The reduced-matrix elements in Eq. (2) can be evaluated using the methods described in section j of [19] where the case of reduced-matrix elements involving only bound states was considered. We consider two particular cases: an initial state containing one electron and a general closed-shell initial state.

B. One-electron initial states

As a simple test case we consider an initial state with the configuration $1s^1$ in which case $J=\frac{1}{2}$. Orbital-orthogonality considerations show that the only nonzero contributions to the sum over $\bar{\gamma}$ in Eq. (2) come from states having the electronic configuration ns',E,κ_e so that the label $\bar{\gamma}$ reduces to the principal quantum number n. Each configuration ns',E,κ_e gives rise to two final states which differ only in the value of J' which can be 0 or 1. The expression for dI/dE obtained from Eq. (2) by considering all these possible final states is

$$\begin{aligned}\frac{dI}{dE}=CR^{-2}\sum_{n}\frac{W}{p}[W_0(ns')-W]^2\{&\langle Z,ns'|Z-1,1s\rangle^2[P^2_{Es'}(R)+Q^2_{E\bar{p}'}(R)]\\&-\langle Z,ns'|Z-1,1s\rangle\langle Z,Es'|Z-1,1s\rangle P_{Es'}(R)P_{ns'}(R)\\&+\langle Z,Es'|Z-1,1s\rangle^2P^2_{ns'}(R)\}\ .\end{aligned}\tag{3}$$

This reduces to the result given by Haxton [10] for tritium decay if the nonrelativistic limit is taken, in which case $Q_{E\bar{p}'}(R)$ is zero.

C. General many-electron initial states containing closed s and $\bar{p}$ subshells

In the following analysis we consider decay of an initial state containing doubly occupied s or $\bar{p}$ subshells and possibly also containing partially filled subshells built from orbitals of individual angular momentum (j) greater than $\frac{1}{2}$. The initial and final electron states will be described in the single manifold (a manifold consists of all the different relativistic configurations that reduce to the same nonrelativistic configuration in the formal limit that the velocity of light tends to infinity) approximation. These states can each be factorized into a product of two kets, one describing the state of the s and $\bar{p}$ electrons and one describing the state of electrons with $j>\frac{1}{2}$. This factorization then allows the sum over the many possible final states which may be produced in the decay to be performed using a closure relation for the final-state wave functions, as described below.

In the following discussion the state labels u and v will be used to define states of the s and $\bar{p}$ electrons while the state labels a and b will be used to define the states of the electrons with $j>\frac{1}{2}$. An initial state $|N,\gamma,J,M\rangle$ can thus be written as $|\gamma_a,J,M\rangle|\gamma_u\rangle$ $(\equiv|\gamma_a,\gamma_u,J,M\rangle)$ where the state $|\gamma_u\rangle$, consisting of only filled subshells each containing two s or two $\bar{p}$ electrons, necessarily has zero total angular momentum so that the total angular momentum (J) and its z component (M) for the state $|N,\gamma,J,M\rangle$ are the same as those of the state $|\gamma_a,J,M\rangle$ of the electrons having $j>\frac{1}{2}$. The only final states generated with significant probability by allowed β decays are those containing the same number of electrons having $j>\frac{1}{2}$ as are present in the initial state. Furthermore the total angular momentum of these $j>\frac{1}{2}$ electrons in the final state must be the same as that (J) of these electrons in the initial state. The state of all the s and $\bar{p}$ electrons in the final state containing a continuum electron of energy E with angular momentum equal to $\frac{1}{2}$ will be denoted $\langle\gamma'_v,E,\kappa_e,J'_s,M'_s|$ with γ'_v denoting the state of the s and $\bar{p}$ electrons, excluding the continuum electron and J'_s and M'_s being the total angular momentum and its z component for all the s and $\bar{p}$ electrons, including the continuum electron. Using the result of Eq. (A5) in the Appendix, the wave functions for the electrons with $j>\frac{1}{2}$ can be factored out of the reduced-matrix elements in Eq. (2). After performing the sum over J' in Eq. (2) one obtains

$$\begin{aligned}\frac{dI}{dE}=\frac{C}{2}\sum_{\gamma'_b}\langle\gamma'_b,J|\gamma_a,J\rangle^2\sum_{\gamma'_v}\sum_{\kappa_e=-1}^{1}\frac{W}{p}[W_0(\gamma'_b,\gamma'_v)-W]^2\Bigg[&\langle\gamma'_v,E,\kappa_e,J'_s\|\hat{b}^{\dagger}_{E,\kappa_e}\|\gamma_u\rangle U_{E,\kappa_e}(R)\\&+\sum_{A(\in\gamma'_v)}\langle\gamma'_v,E,\kappa_e,J'_s\|\hat{b}^{\dagger}_{A}\|\gamma_u\rangle U_A(R)^2\Bigg]\ ,\end{aligned}\tag{4}$$

where the quantity $\langle \gamma'_b,J|\gamma_a,J\rangle$ is the overlap between the initial and final state of the $j>\frac{1}{2}$ electrons. The label $\overline{\gamma}$ in Eq. (2) has become the double (γ'_b,γ'_v) in Eq. (4). Both reduced-matrix elements in Eq. (4) involve the wave functions for just the s and $\overline{p}$ electrons. From the analysis in the Appendix, the angular momentum J'_s must equal $\frac{1}{2}$. The reduced-matrix elements involving the operator $\hat{b}^{\dagger}_{E,\kappa_e}$ which appear in the direct terms can then be expressed in terms of orbital overlaps according to [19]

$$\langle \gamma'_v,E,\kappa_e,\tfrac{1}{2}\|\hat{b}^{\dagger}_{E,\kappa_e}\|\gamma_u\rangle=-\sqrt{2}\langle\gamma'_v|\gamma_u\rangle \ . \tag{5}$$

The reduced-matrix elements involving the operator $\hat{b}^{\dagger}_A$ which appear in the exchange terms can be evaluated for any given γ'_v and any orbital A. However, in decays of a many-electron initial state one particular γ'_v state dominates the sum over γ'_v in Eq. (4). This is the state in which the s and $\overline{p}$ orbitals have the same occupancy as in the initial state. This final state, which will be called the core state of the s and $\overline{p}$ electrons, will be written γ'_u where the label u indicates the quantum numbers of the electrons are the same as in the initial state γ_u. For decay of the ions of ^{106}Ru and ^{241}Pu that forms a major focus of this work, we have shown by explicit computation of the overlaps $\langle\gamma'_u|\gamma_u\rangle$ that the state γ'_u is produced in more than 99.5% of decays so that less than 0.5% of s and $\overline{p}$ electrons are excited in the decay process. This may be compared with the corresponding probability for decay to the final state of the $j>\frac{1}{2}$ electrons in which these electrons are in orbitals that have the same quantum numbers as those in the initial state. If this final state is denoted γ'_a, computation of the overlaps $\langle\gamma'_a,J|\gamma_a,J\rangle$ shows that the probability of decay to the γ'_a state is 75–80 %, implying that electrons with $j>\frac{1}{2}$ are excited in 20–25 % of decays. Thus in the following we focus on exchange terms in the γ'_u state which gives the dominant contribution to the exchange effects on the β spectrum and sum over all the γ'_b states using closure. For the γ'_u state, the reduced-matrix element in the exchange terms is given by [19]

$$\langle\gamma'_u,E,\kappa_e,\tfrac{1}{2}\|\hat{b}^{\dagger}_A\|\gamma_u\rangle=\sqrt{2}\langle\gamma'_u(A\rightarrow E,\kappa_e)|\gamma_u\rangle \ , \tag{6}$$

where $\langle\gamma'_u(A\rightarrow E,\kappa_e)|$ is the Fock-space bra generated from $\langle\gamma'_u|$ by replacing the orbital belonging to subshell A and having $m_j=\frac{1}{2}$ by the continuum $m_j=\frac{1}{2}$ orbital with kinetic energy E and kappa quantum number κ_e. When the reduced-matrix elements are expressed in terms of orbital overlaps by means of Eqs. (5) and (6), the exchange terms are seen to contain a phase factor of opposite sign to that of the direct term. This phase factor arises from the antisymmetrization of the electron wave functions. The overlap in Eq. (6) can be written as a determinant whose elements are one-electron overlaps. The main contribution to the value of the determinant comes from the leading diagonal. In particular, terms involving products of two or more one-electron overlaps between an initial-state bound orbital and a final-state continuum orbital are very small. Thus to a good approximation we have

$$\langle\gamma'_u(A\rightarrow E,\kappa_e)|\gamma_u\rangle=\langle\gamma'_u|\gamma_u\rangle\frac{\langle E,\kappa_e|a_A\rangle}{\langle a'_A|a_A\rangle} \ , \tag{7}$$

where $\langle E,\kappa_e|a_A\rangle$ is a one-electron overlap between an initial-state orbital belonging to subshell A and a final-state continuum orbital of kinetic energy E and kappa quantum number κ_e. In this and all subsequent one-electron overlaps appearing below, the initial- and final-state orbitals are assumed to have the same magnetic quantum number which is omitted for convenience. The γ'_b and γ'_v states satisfy the closure relations

$$\sum_b\langle\gamma'_b,J|\gamma_a,J\rangle^2=1 \ , \tag{8}$$

$$\sum_v\langle\gamma'_v,J|\gamma_u,J\rangle^2=1 \ . \tag{9}$$

When Eqs. (5)–(7) are substituted in Eq. (4) the above closure results can be used to perform all the sums over γ'_b as well as that over γ'_v in the direct terms, at the same time replacing $W_0(\gamma'_b,\gamma'_v)$, which is only a weak function of γ'_b and γ'_v, by an average end-point energy $\overline{W}_0$, as described in Refs. [19] and [20]. In the sum over γ'_v, the exchange terms for states other than the core state γ'_u will be neglected since, for the reasons given above, this state is by far the most important final state of the s and $\overline{p}$ electrons and exchange terms in this state will therefore be dominant. Then, noting that to a very good approximation $\langle\gamma'_u|\gamma_u\rangle=1$ and $\langle a'_A|a_A\rangle=1$ (except for $Z\cong 1$), one has

$$\frac{dI}{dE}=C\frac{W}{p}(\overline{W}_0-W)^2\times\sum_{\kappa_e=-1}^{1}\left[U_{E,\kappa_e}(R)-\sum_{A(\in\gamma)}\langle E,\kappa_e|a_A\rangle U_A(R)\right]^2 \ , \tag{10}$$

where the summation over A is over orbitals which are occupied in the initial state γ. If the terms involving $U_A(R)$ are neglected, Eq. (10) yields the standard result for the β spectrum intensity with no exchange (denoted by the superscript NE):

$$\frac{dI^{\mathrm{NE}}}{dE}=CR^{-2}\frac{W}{p}(\overline{W}_0-W)^2[P^2_{Es'}(R)+Q^2_{E\overline{p}'}(R)] \ . \tag{11}$$

The β spectrum intensity calculated including the exchange terms can then be written in terms of the intensity calculated ignoring exchange terms:

$$\frac{dI}{dE}=\frac{dI^{\mathrm{NE}}}{dE}[1+\eta^T_{\mathrm{ex}}(E)] \ , \tag{12}$$

where the factor $\eta^T_{\mathrm{ex}}(E)$ describes the modification to the β spectrum intensity due to the inclusion of exchange effects. This factor will be called the total exchange factor and from Eqs. (10)–(12) it is given by

$$\eta^T_{\mathrm{ex}}(E)=f_s(2T_s+T_s^2)+(1-f_s)(2T_{\overline{p}}+T_{\overline{p}}^2) \ , \tag{13}$$

where f_s is given by

$$f_s=\frac{P^2_{Es'}(R)}{P^2_{Es'}(R)+Q^2_{E\bar{p}'}(R)}\ , \tag{14}$$

and T_s and $T_{\bar{p}}$ are sums of exchange terms given by

$$T_s=-\sum_{ns'(\in\gamma)}\langle Es'|ns\rangle\frac{P_{ns'}(R)}{P_{Es'}(R)}\ , \tag{15}$$

and

$$T_{\bar{p}}=-\sum_{n\bar{p}'(\in\gamma)}\langle E\bar{p}'|n\bar{p}\rangle\frac{Q_{n\bar{p}'}(R)}{Q_{E\bar{p}'}(R)}\ . \tag{16}$$

If exchange effects are ignored, then for those decays producing a continuum electron of energy E, f_s represents the fraction of β decays in which this electron emerges in an s state. In the following discussion, the focus will be on exchange terms involving s electrons and exchange effects involving $\bar{p}$ electrons will be ignored. The contribution to the total decay rate made by $\bar{p}$-wave decays is relatively small except in very-high-Z decays and therefore $\bar{p}$-wave exchange should be considerably less important than s-wave exchange.

It will be convenient to consider the separate contributions that different ns subshells make to the exchange factor. We therefore define a subshell exchange factor $\eta^{ns}_{\text{ex}}(E)$ for a particular subshell at a given electron energy by

$$\eta^{ns}_{\text{ex}}(E)=f_s\left[-\left[2\langle Es'|ns\rangle\frac{P_{ns'}(R)}{P_{Es'}(R)}\right]+\left[\langle Es'|ns\rangle\frac{P_{ns'}(R)}{P_{Es'}(R)}\right]^2\right]\ . \tag{17}$$

The total exchange factor can be written in terms of the subshell exchange factors according to

$$\eta^{T}_{\text{ex}}(E)=\sum_{ns}\eta^{ns}_{\text{ex}}(E)+\sum_{\substack{ms,ns\\(m\neq n)}}\left[\langle Es'|ms\rangle\frac{P_{ms'}(R)}{P_{Es'}(R)}\right]\times\left[\langle Es'|ns\rangle\frac{P_{ns'}(R)}{P_{Es'}(R)}\right]\ . \tag{18}$$

Since the exchange terms are generally quite small compared to unity, the products of terms in large parentheses in Eq. (18) are considerably smaller than the quantity $\eta^{ns}_{\text{ex}}(E)$. Thus the total exchange factor is given, to a good approximation, by the sum of subshell exchange factors defined by Eq. (17).

Since we also wish to quantify exchange effects on the phase-space integral due to exchange with different subshells, we define quantities called subshell exchange contributions by

$$f^{ns}_{\text{ex}}=\int\frac{dI^{\text{NE}}}{dE}\eta^{ns}_{\text{ex}}(E)dE\left[\int\frac{dI^{\text{NE}}}{dE}dE\right]^{-1}\ . \tag{19}$$

The subshell exchange contribution represents the fractional change in the phase-space integral for the decay as a result of the inclusion of exchange with ns electrons. In a similar way, the total exchange contribution is defined by

$$f^{T}_{\text{ex}}=\int\frac{dI^{\text{NE}}}{dE}\left[\sum_{ns}\eta^{ns}_{\text{ex}}(E)\right]dE\left[\int\frac{dI^{\text{NE}}}{dE}dE\right]^{-1}\ , \tag{20}$$

and represents the fractional change in the phase-space integral as a result of the inclusion of exchange with all bound s electrons.

D. Nonrelativistic approximations

Generally, relativistic effects on the electron wave functions are very important in the β-decay process because the $U_{E,\kappa_e}(E)$ factors, which determine the direct contribution to the decay process, involve the electron wave function at the nuclear radius. However, one may expect the exchange terms T_s to be less sensitive to relativistic effects because they involve ratios of electron wave functions in the nuclear region in which relativistic effects should largely cancel out. Also, the overlaps should not be strongly dependent upon relativistic effects. In many cases it may therefore be convenient to evaluate the exchange effects on the β spectrum by calculating $\eta^{ns}_{\text{ex}}(E)$ using a nonrelativistic approximation for the wave functions in Eq. (15) but retaining a relativistic treatment of the factor f_s. Using point nucleus unscreened Dirac Coulomb wave functions for $P_{Es'}(R)$ and $Q_{E\bar{p}'}(R)$, it can be shown that f_s is well approximated by the quantity $(W+\gamma_0)/(2W)$ with $\gamma_0=[1-(\alpha Z)^2]^{1/2}$ where α is the fine-structure constant. For low-energy decays f_s can vary from a value close to unity in a low-Z atom to a value of 0.85 in a high-$Z\simeq95$ atom. If the effect of exchange on the (minor) $\bar{p}$ contribution to the β intensity is ignored, then in this nonrelativistic approximation, the total exchange factor is given by

$$\eta^{T}_{\text{ex}}(E)=(2T_s+T_s^2)(W+\gamma_0)/2W\ . \tag{21}$$

The validity of this nonrelativistic approximation for the exchange terms is examined for several cases in Sec. III by comparison with exchange terms calculated using fully relativistic wave functions.

E. Kurie plot

β spectra are commonly interpreted in terms of the Kurie plot. The Kurie function $K(E)$ is defined as

$$K(E)=\left[\left[\frac{dI}{dE}\right]_{\text{expt}}\left[\frac{W}{p}[P^2_{Es'}(R)+Q^2_{E\bar{p}'}(R)]\right]^{-1}\right]^{1/2}\ , \tag{22}$$

where $(dI/dE)_{\text{expt}}$ is the experimentally measured β^- intensity. If the theoretical intensity (dI/dE) in Eq. (12) is equal to the experimental intensity (which should be the case if antineutrino mass is zero) then, using Eqs. (11) and (22), $K(E)$ is given by

$$K(E)=\beta[1+\eta_{\rm ex}^{T}(E)]^{1/2}(E_0-E)\ , \tag{23}$$

where β is a constant independent of electron energy. A Kurie plot, which is a plot of $K(E)$ versus E, should thus be linear if the total exchange factor $\eta_{\rm ex}^{T}(E)$ is negligible or if it is independent of energy in the energy range considered. It is thus only through their energy dependence that exchange effects manifest themselves as a distortion of the Kurie plot. If the exchange factor is small compared to unity then the inclusion of exchange leads to multiplication of the Kurie function by a factor approximately equal to $1+\frac{1}{2}\eta_{\rm ex}^{T}(E)$.

III. EVALUATION OF THE EXCHANGE TERMS

A. General overview

The exchange factors $\eta_{\rm ex}^{ns}(E)$ and $\eta_{\rm ex}^{T}(E)$ given by Eqs. (17) and (18) involve the overlap $\langle Es'|ns\rangle$ between a bound s orbital wave function in the initial-state atom (nuclear charge $Z-1$) and a continuum s orbital wave function in the final-state atom (nuclear charge Z). This overlap requires knowledge of both wave functions over a wide region of space. Screening effects of atomic electrons on both wave functions may need to be taken into account since screening may be significant in the regions of space contributing to the overlap integral. Also care has to be taken to ensure that the final-state continuum electron wave function is orthogonal to the wave functions for the final-state bound orbitals which are eigenfunctions of the same Hamiltonian. In particular, in numerical calculations, care has to be taken to ensure that the orbital sets used to describe the initial and final states are calculated so that the overlap $\langle Es'|ns'\rangle$ (which should be identically zero by orbital orthogonality) is much smaller than $\langle Es'|ns\rangle$, otherwise the overlap $\langle Es'|ns\rangle$ may be significantly in error.

Two methods of calculating the exchange overlap integrals are described below. The first is based on a hydrogenic approximation for both bound and continuum wave functions. The second is based on a Hartree-Fock approximation for the electron wave functions.

B. Hydrogenic approximation

Within the hydrogenic approximation, two cases will be considered: one based on using eigenfunctions of the Schrödinger equation and the other on eigenfunctions of the Dirac equation. In the nonrelativistic case, the analytic forms of the bound and continuum hydrogenic electron wave functions are well known [21] and, moreover, matrix elements involving hydrogenic wave functions and radial operators (including the unit operator) can be expressed analytically in terms of hypergeometric functions [22]. If the effective nuclear charge of a hydrogenic bound wave function with principal quantum number n and angular momentum l is denoted Z_i and the effective nuclear charge of a hydrogenic continuum wave function of energy E and angular momentum l is denoted Z_f, then the overlap integral between these wave functions, $\langle Z_f,El|Z_i,nl\rangle$, can be shown from [22] to be given by

$$\begin{aligned}\langle Z_f,El|Z_i,nl\rangle=&\frac{(-1)^{n-l-1}(Z_i-Z_f)e^{2i(l+1-n)\arctan(p/\bar{Z}_i)}}{[(2l+1)!]p(p^2+\bar{Z}_i^{\,2})^{l+2}}2^{2l+3}(p\bar{Z}_i)^{l+3/2}|\Gamma(l+1-i(Z_f/p))|\\&\times e^{\pi Z_f/2p}e^{-(2Z_f/p)\arctan(p/\bar{Z}_i)}F(l+1+i(Z_f/p),l+1-n,2l+2,-4pi\bar{Z}_i/(\bar{Z}_i-ip)^2)\ ,\end{aligned} \tag{24}$$

where $\bar{Z}_i=Z_i/n$ and the continuum wave functions are assumed to be normalized on the energy scale according to

$$\int\psi_{El}(\mathbf{r})\psi_{E'l}(\mathbf{r})d\mathbf{r}=\delta(E-E')\ . \tag{25}$$

Setting $l=0$ and $n=1$ Eq. (24) yields

$$\begin{aligned}\langle Z_f,Es|Z_i,1s\rangle=&8(Z_i-Z_f)(Z_fZ_i^3)^{1/2}\\&\times e^{-(2Z_f/p)\arctan(p/Z_i)}(p^2+Z_i^2)^{-2}\\&\times(1-e^{-2\pi Z_f/p})^{-1/2}\ .\end{aligned} \tag{26}$$

This result for the overlap between a hydrogenic 1s wave function and a hydrogenic continuum electron wave function agrees with the result given in [2].

Setting $l=0$ and $n=2$ in Eq. (24) yields

$$\begin{aligned}\langle Z_f,Es|Z_i,2s\rangle=&8(Z_i-Z_f)(Z_fZ_i^3)^{1/2}\\&\times e^{-(2Z_f/p)\arctan(p/\bar{Z}_i)}\\&\times(p^2+\bar{Z}_i^{\,2})^{-3}(1-e^{-2\pi Z_f/p})^{-1/2}\\&\times[p^2+\bar{Z}_i(2Z_f-\bar{Z}_i)]\ .\end{aligned} \tag{27}$$

By virtue of the factor (Z_i-Z_f), the overlaps for both 1s and 2s orbitals are clearly seen to be of negative sign. The bound and continuum wave functions used in this calculation are defined with phases such that the large component of the nonrelativistic radial wave function is positive for small r. The quantities $P_{ns'}(R)$ and $P_{Es'}(R)$ in Eqs. (17) and (18) are therefore also positive. Taken together, these results for the signs of the quantities $\langle Es'|ns\rangle$, $P_{Es'}(R)$, and $P_{ns'}(R)$ show that the exchange terms involving 1s and 2s electrons must be of positive sign with the result that they interfere constructively with the direct term thereby leading to an enhancement of the β-decay rate. This result is the reverse of that reported by Bahcall [18]. An inconsistency in the wave functions used in the latter calculation would seem to explain the discrepancy. Evaluation of Eq. (24) with a range of values of n for $l=0$ and 1 shows that the overlaps $\langle Z_f,El|Z_i,nl\rangle$ are negative irrespective of the value of n. This implies the general result that the inclusion of exchange with s and $\bar{p}$ electrons tends to enhance the β-decay rate. Exchange terms involving two bound electrons arise in the theory of bound-state β decay [19].

Evaluation of these exchange terms for several decays also shows that the sign of the exchange terms are such that they tend to enhance the β-decay rate [19]. It should be emphasized that the above comments regarding the sign of the bound-continuum exchange terms refer to exchange terms in Eq. (2) in final states in which the s and $\bar{p}$ electrons are in orbitals which have the same quantum numbers as those in the initial state, though electrons with $j > \frac{1}{2}$ may be in excited orbitals. Exchange terms involving final states containing excited s or $\bar{p}$ electrons are not necessarily guaranteed to interfere constructively with the direct term. However, as indicated above, the latter final states are produced with very low probability in decays of medium- and high-Z atoms.

In order to evaluate the exchange terms using overlap integrals given by Eqs. (24)–(27), it is necessary to assign values to the effective nuclear charges Z_i and Z_f in such a way that they yield reasonable estimates of the overlap integrals between screened atomic orbitals. That is, the values of Z_i and Z_f should be chosen in order to yield values for the hydrogenic wave functions which are reasonable approximations to the wave functions for the states $\langle Z,El'|$ and $|Z-1,nl\rangle$ in the regions of space which contribute significantly to the overlap $\langle Z,El'|Z-1,nl\rangle$. In this work Z_i and Z_f have been chosen in two different ways, one taking account of electron screening effects, the second ignoring such effects completely. In the former method, denoted the screened hydrogenic approximation, Z_i and Z_f were chosen for each initial state orbital ns by matching the well-known result [23] for the mean radius of a hydrogenic orbital of effective nuclear charge Z_{eff},

$$\langle Z_{\text{eff}},nl|r|Z_{\text{eff}},nl\rangle=\frac{3n^2}{2Z_{\text{eff}}}-\frac{l(l+1)}{2Z_{\text{eff}}} , \tag{28}$$

to mean radii for ns orbitals calculated in the Hartree-Fock approximation. Specifically, Z_i and Z_f were calculated according to

$$\langle Z_i,ns|r|Z_i,ns\rangle=\langle r\rangle_{Z-1,\gamma,ns} , \tag{29a}$$

and

$$\langle Z_f,ns|r|Z_f,ns\rangle=\langle r\rangle_{Z,\gamma^c,ns} . \tag{29b}$$

Here the mean radii on the left of these expressions are calculated using Eq. (28) while the mean radii on the right-hand side refer to mean radii calculated in the Hartree-Fock approximation. $\langle r\rangle_{Z-1,\gamma,ns}$ is the mean radius of the ns orbital in the initial state γ of the N-electron atom with nuclear charge $Z-1$. $\langle r\rangle_{Z,\gamma^c,ns}$ is the mean radius of the ns orbital in the core state (denoted γ^c) of the N-electron atom with nuclear charge Z in which all the electrons have the same orbital occupation numbers as in the initial state γ. This procedure for calculating Z_i and Z_f aims at ensuring that the hydrogenic continuum orbital $\langle Z_f,Es|$ is very nearly orthogonal to the true final-state bound orbital $\langle Z,ns'|$. The values of Z_i and Z_f are based on matching values of mean radii, rather than other radial expectation values, in order that the hydrogenic orbitals $\langle Z_f,ns|$ and $|Z_i,ns\rangle$ provide reasonable estimates of the true orbital wave functions $\langle Z,ns'|$ and $|Z-1,ns\rangle$ in the regions contributing significantly to the overlap $\langle Z,Es'|Z-1,ns\rangle$. It should be noted that results are fairly insensitive to the choice of the particular expectation value used to fix Z_i and Z_f by matching to Hartree-Fock wave functions. Thus, consider, for example, the case of decay of the ion $^{106}\text{Ru}^{1+}$ in the ground state [Kr]$4d^7$. ^{106}Ru is chosen as an example since it is considered to be representative of a medium-Z low-energy β decay. The core final state is Rh^{2+} [Kr]$4d^7$. Table I shows values of the mean radii of the s orbitals of these Ru^{1+} and Rh^{2+} states calculated in the Hartree-Fock approximation together with values of effective nuclear charges obtained by fitting according to Eq. (29). The Hartree-Fock wave functions were generated using the GRASP code [24] with the velocity of light set to a large value which yields the nonrelativistic limit. For both γ and γ^c states, all configurations arising from the d^7 manifold were included in the calculation. Although the effective nuclear charges which fit the Hartree-Fock expectation values differ significantly from the full nuclear charge, particularly for high n, it is clear that for a given orbital, the difference between the effective nuclear charge in Rh^{2+} and Ru^{1+} is very close to unity. Table II shows the results of a similar analysis based on matching the hydrogenic mean inverse radius to the Hartree-Fock mean inverse radius. For a hydrogenic orbital the mean inverse radius is given by $\langle Z_i,nl|r^{-1}|Z_i,nl\rangle=Z_i/n^2$ [23]. Again the differences in effective nuclear charge between Ru^{1+} and Ru^{2+} are seen to be very close to unity. Furthermore these results for Z_i and Z_f based on matching the mean inverse radii are very similar to those calculated by matching the mean radii. The results based on matching the mean radii are preferred because the mean radius receives the most significant contributions at larger radial distances

TABLE I. Mean radii of s orbitals in Ru^{1+} and Rh^{2+}, calculated in the Hartree-Fock approximation, and associated effective nuclear charges (Z_i and Z_f) of hydrogenic s orbitals calculated according to Eqs. (28) and (29).

n	$\langle r\rangle_{\text{Ru}^{1+},ns}$ (a.u.)	Z_i	$\langle r\rangle_{\text{Rh}^{2+},ns}$ (a.u.)	Z_f
1	0.034 624	43.322	0.033 845	44.319
2	0.150 601	39.841	0.146 993	40.818
3	0.412 044	32.764	0.400 326	33.723
4	1.056 522	22.716	1.012 668	23.700

TABLE II. Mean inverse radii of s orbitals in Ru^{1+} and Rh^{2+}, calculated in the Hartree-Fock approximation, and associated effective nuclear charges (Z_i and Z_f) of hydrogenic s orbitals.

n	$\langle r^{-1}\rangle_{\mathrm{Ru}^{1+},ns}$ (a.u.)	Z_i	$\langle r^{-1}\rangle_{\mathrm{Rh}^{2+},ns}$ (a.u.)	Z_f
1	43.4823	43.482	44.4806	44.481
2	9.8650	39.460	10.1089	40.436
3	3.4731	31.258	3.5782	32.204
4	1.2736	20.377	1.3316	21.306

than the mean inverse radius and in a similar region of space to that which is expected to give the dominant contributions to the overlaps $\langle Es'|ns\rangle$.

The overlaps of the type just discussed may be sensitive to screening effects particularly for medium- to high-Z atoms. On the other hand, the ratios of electron wave functions evaluated at the nuclear radius that appear in Eqs. (17) and (18) should be considerably less sensitive to electron screening effects. Therefore, in calculations of exchange factors in the hydrogenic approximation, the components $P_{ns'}(R)$ and $P_{Es'}(R)$, evaluated at the nuclear radius, which appear in Eq. (17) have been approximated by the value appropriate for a completely unscreened nonrelativistic hydrogenic wave function.

Thus far the use of hydrogenic electron wave functions has been considered within a nonrelativistic approximation. However, in order to evaluate the validity of this nonrelativistic approximation, calculations have also been performed employing eigenfunctions of the Dirac equation but using a similar method to that described above for nonrelativistic wave functions. The relativistic counterpart of Eq. (28) is given in Ref. [25]. Values for Z_i and Z_f have been obtained by matching mean radii for relativistic hydrogenic wave functions to the mean radii of orbitals in the γ and γ^c states calculated using GRASP [24]. Again all configurations corresponding to a given manifold were included in the calculation. The values of Z_i and Z_f thus generated were used to calculate overlaps $\langle Z_f,Es|Z_i,ns\rangle$ by direct numerical integration using the well-known expressions for the relativistic hydrogenic bound and continuum electron wave functions [26]. The values of $P_{ns'}(R)$ and $P_{Es'}(R)$ used to calculate the exchange terms in Eq. (17) were obtained using completely unscreened hydrogenic Dirac wave functions. The results of a comparison between relativistic and nonrelativistic approaches are presented below.

C. Hartree-Fock approximation

Wave functions for bound electronic states can be readily calculated in the Hartree-Fock approximation. Solving the Hartree-Fock equations with both bound and continuum electrons is less straightforward. For the present purpose it has been found convenient to solve the Hartree-Fock equations for the continuum electron using a fixed core approximation for the bound electrons. The computational implementation of this method has been described by Bates [27]. With this approach, orthogonality integrals of the kind $\langle Z,El|Z,nl\rangle$ can be forced to be zero (to within a given numerical accuracy) using the method of Lagrange undetermined multipliers.

In order to generate the continuum wave functions used in the calculation of the exchange factor, the wave functions for a given $\bar{\gamma}$ state were first generated using the GRASP code [24] as described above. These were then used, after interpolation to a suitable grid, as the frozen core input used to solve the Hartree-Fock equations for the $(N+1)$-electron system composed of the $\bar{\gamma}$ state together with the continuum electron, according to the method described in [27].

The method has been found to be practical for relatively low energies ($E < 50$ keV). At higher energies the oscillatory nature of the continuum electron wave function leads to very large radial grids being required to adequately represent the wave functions, with consequent numerical difficulties. In the calculations reported below the continuum wave functions were evaluated at 30 intervals between the maximum and minimum energies.

IV. RESULTS

A. ^{106}Ru decay

1. Hydrogenic approximation

We consider decay of Ru^{1+} [Kr]$4d^7$ to final states containing one continuum electron and the Rh^{2+} [Kr]$4d^7$ ion. Exchange with s electrons may involve $1s$, $2s$, $3s$, or $4s$ electrons since all these orbitals are filled in the initial state. The effective nuclear charges used in the calculation of the overlap integrals in the nonrelativistic screened hydrogenic approximation are shown in Table I and have been discussed in the preceding section. The $1s$, $2s$, $3s$, and $4s$ subshell exchange terms evaluated in the hydrogenic approximation using Eqs. (17) and (24) are plotted in Fig. 1. Exchange with $1s$ electrons constitutes the most significant contribution at energies from 40 keV down to approximately 1 keV. However, the $2s$ contribution is non-negligible and amounts to approximately 25% of the $1s$ contribution over much of the energy range above 10 keV, becoming even more important at low energies. The energy dependence of the $1s$ exchange factor is rather weak. The exchange factors are, however, seen to become increasingly energy dependent for higher principal quantum number. The total exchange factor varies from approximately 2% at 40 keV to 7% at 5 keV. This would suggest that the exchange process may significantly modify the β intensity, particularly at low

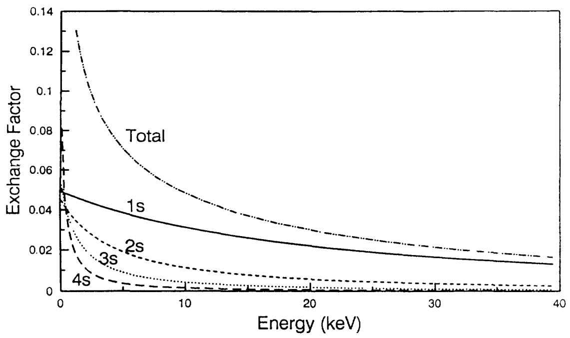

FIG. 1. Exchange factors for 1*s*, 2*s*, 3*s*, and 4*s* subshells and total exchange factor for ^{106}Ru decay, calculated in the screened hydrogenic approximation.

energy and thereby also significantly increase the phase-space integral for the decay.

The results obtained above in the screened hydrogenic approximation are compared in Fig. 2 with those obtained using completely unscreened hydrogenic electron wave functions. The result for the 1*s* subshell exchange using unscreened wave functions agrees well with the result obtained using the screened hydrogenic wave functions. For subshells with larger principal quantum numbers (n), the difference between the subshell exchange factors calculated in the two approximations tends to increase as n increases. However, except at very low electron energy, the contribution to the total exchange factor made by a given subshell decreases as n increases. The net result is that the total exchange factor calculated in the unscreened hydrogenic approximation also agrees well with the screened hydrogenic result, except at very low electron energy (see Fig. 2).

2. *Hartree-Fock approximation*

For ^{106}Ru^{1+} decay, Fig. 3 shows subshell exchange factors for the 1*s* and 2*s* subshells, evaluated in the Hartree-Fock approximation using the method described in Sec. III. The results agree very closely at high electron energies. At low energies the results calculated in the Hartree-Fock approximation are larger than the results calculated in the screened hydrogenic approximation. The agreement of the two results over much of the spectrum indicates the usefulness of the screened hydrogenic approximation.

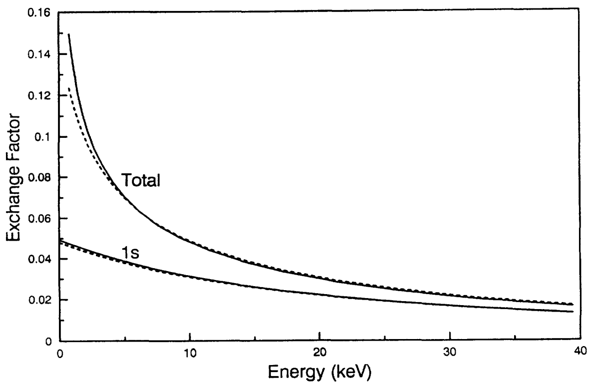

FIG. 2. Comparison of 1*s* subshell and total exchange factors calculated with screened hydrogenic (solid line) and unscreened hydrogenic (dashed line) nonrelativistic wave functions.

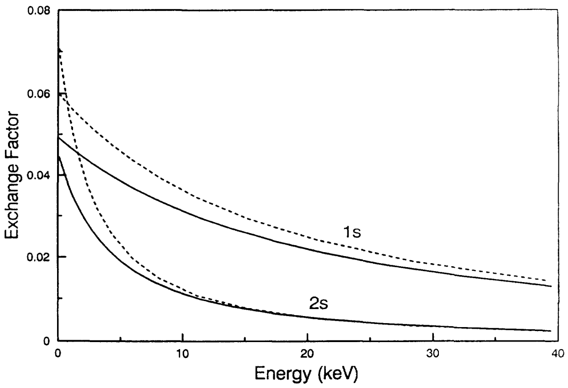

FIG. 3. Comparison of 1*s* and 2*s* subshell exchange factors for ^{106}Ru^{1+} decay. The solid lines were obtained using the screened hydrogenic approximation. The dashed lines were obtained using the Hartree-Fock approximation.

B. Comparison of relativistic and nonrelativistic results

Thus far the exchange factors have been calculated using the nonrelativistic approximation described in Sec. II D. In order to examine the validity of this approximation we consider the relativistic effects on the 1*s* exchange contribution for decays of two isotopes, ^{106}Ru and ^{241}Pu. The 1*s* exchange factor is chosen since this orbital makes the largest contribution to the exchange factor. The decay of ^{106}Ru is chosen as representative of a medium-Z, low-energy β emitter and the decay of ^{241}Pu as representative of a high-Z, low-energy β emitter. ^{241}Pu decays by a first forbidden transition to ^{241}Am. However, since the decay is one for which the ξ approximation holds good [28,29], the expression for the decay rate is the same as that for an equivalent allowed β emitter with the only difference that the β intensity is multiplied by an energy-independent constant. The theory presented above for allowed transitions can therefore be applied straightforwardly to ^{241}Pu decay. It should be noted that Ref. [29] used an alternative form of the ξ approximation in which the decay rate expression contains the small components of s orbitals instead of their large components as used here (and also the large components of $\bar{p}$ orbitals instead of their small components). This latter approach is formally more exact but gives numerical results which do not differ significantly from the ξ approximation as applied here.

The Dirac-Fock mean radii for the 1*s* orbitals of Ru^{1+} and Rh^{2+} were found to be 0.033 427 and 0.032 618 au, respectively, using the GRASP code [24]. Using the relativistic analog of Eq. (24) given in Ref. [25] yields the effective nuclear charges $Z_i=43.339$ and $Z_f=44.337$.

TABLE III. Mean radii of s orbitals in Pu^{3+} $[\mathrm{Rn}]5f^5$ and Am^{4+} $[\mathrm{Rn}]5f^5$, calculated in the Dirac-Fock approximation, and associated effective nuclear charges (Z_i and Z_f) of hydrogenic s orbitals.

n	$\langle r\rangle_{\mathrm{Pu}^{3+},ns}$ (a.u.)	Z_i	$\langle r\rangle_{\mathrm{Am}^{4+},ns}$ (a.u.)	Z_f
1	0.013 226	93.222	0.013 015	94.213
2	0.054 675	90.485	0.053 772	91.505
3	0.142 357	84.050	0.140 079	85.108
4	0.308 900	72.601	0.303 613	73.695
5	0.643 961	56.388	0.630 206	57.527
6	1.425 111	37.452	1.372 235	38.860

For Pu^{3+} and Am^{4+} the Dirac-Fock mean radii of the 1s orbitals and associated values of Z_i and Z_f are shown in Table III; these compare with the results $Z_i=93.222$ and $Z_f=94.213$ calculated from the Hartree-Fock wave functions for Pu^{1+} and Am^{2+} using Eq. (29). The 1s exchange factors for Ru^{1+} and Pu^{3+}, calculated in both relativistic and nonrelativistic screened hydrogenic approximations, are shown in Fig. 4. For Ru^{1+} the nonrelativistic approximation is seen to be a very good one with agreement to within a few percent over the whole spectrum. Since relativistic effects on the ns ($n > 1$) exchange factors will certainly be no larger than those on the 1s exchange factor, this result indicates that the nonrelativistic approximation (as described in Sec. II D) applied to a low- or medium-Z decay is certainly satisfactory. For Pu^{3+} decay the nonrelativistic approximation is seen to be less satisfactory and underestimates the 1s exchange factor by approximately 50% at 40 keV.

C. $^{241}\mathrm{Pu}$ decay

Based on the results of the preceding section, accurate calculation of exchange effects in $^{241}\mathrm{Pu}$ decay should be based on a relativistic description of the electron wave functions. The results of a calculation of exchange factors for $^{241}\mathrm{Pu}^{3+}$ using a fully relativistic screened hydrogenic approximation are shown in Fig. 5. The Dirac-Fock mean radii of s orbitals $^{241}\mathrm{Pu}^{3+}$ $[\mathrm{Rn}]5f^5$ and the core state of $^{241}\mathrm{Am}^{4+}$ $[\mathrm{Rn}]5f^5$ are shown in Table III, together with the associated effective nuclear charges. From Fig. 5 it can be seen that the 1s subshell again makes the single largest contribution to the total exchange factor over most of the spectrum but contributions from 2s, and higher-lying s orbitals are quite significant. The 1s exchange factor is almost independent of energy over the whole β spectrum of $^{241}\mathrm{Pu}^{3+}$.

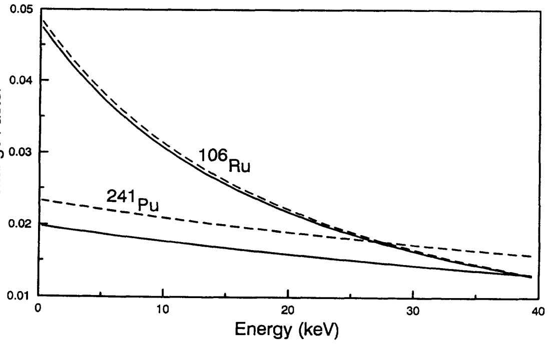

FIG. 4. Comparison of relativistic (dashed line) and nonrelativistic (solid line) 1s exchange factors for $^{106}\mathrm{Ru}^{1+}$ and $^{241}\mathrm{Pu}^{3+}$ decay, calculated in the screened hydrogenic approximation.

D. Effect of exchange on the Kurie plot

Figure 6 shows the effect of exchange on the Kurie plots for decay of $^{106}\mathrm{Ru}^{1+}$ and $^{241}\mathrm{Pu}^{3+}$. The Kurie plots including exchange were calculated according to Eq. (23) using the Hartree-Fock approximation for the electron wave functions for $^{106}\mathrm{Ru}^{1+}$ and the relativistic screened hydrogenic approximation for $^{241}\mathrm{Pu}^{3+}$, implemented as described in the preceding section. The inclusion of exchange leads to distortion of the Kurie plot from the straight line expected in the case of no exchange. The distortion becomes larger at lower electron energy. However, from the end-point energy to an energy of about 5 keV the deviation from linearity is a few percent at most in both cases. Deviations of this magnitude appear to be too small to observe in the existing data [30–32] on the β spectra of these isotopes.

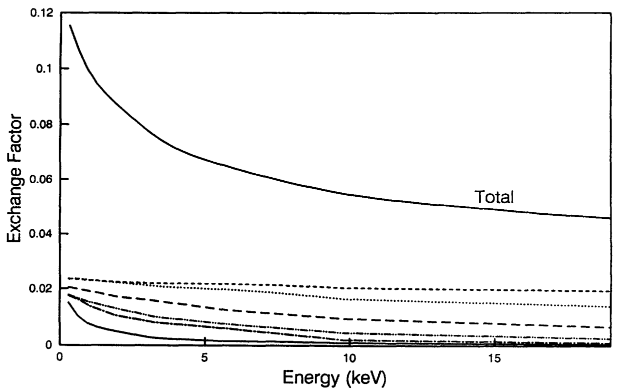

FIG. 5. Exchange factors for $^{241}\mathrm{Pu}^{3+}$ decay calculated in the screened hydrogenic approximation using relativistic wave functions. The short-dashed line is for the 1s subshell, the dotted line for the 2s subshell, the long-dashed line for the 3s subshell, the chain-dotted line for the 4s subshell, the chain-dashed line for the 5s subshell, and the solid line for the 6s subshell. Also shown is the total exchange factor.

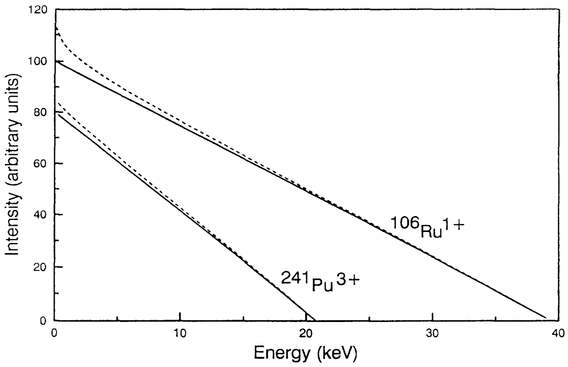

FIG. 6. Effect of exchange on the Kurie plots for $^{106}\mathrm{Ru}^{1+}$ decay and $^{241}\mathrm{Pu}^{3+}$. The solid lines are the Kurie plots assuming exchange effects are negligible while the dotted lines are calculated including exchange with *s* electrons. For $^{106}\mathrm{Ru}$ the results were calculated in the Hartree-Fock approximation while for $^{241}\mathrm{Pu}$ the results were calculated using the screened hydrogenic approximation with effective nuclear charges obtained by matching to Dirac-Fock *s* orbital wave functions.

E. Effect of exchange on the phase-space integral

Integrating the β spectrum from zero energy to the end point using Eqs. (19) and (20) yields the exchange contributions to the phase-space integrals for $^{106}\mathrm{Ru}^{1+}$ and $^{241}\mathrm{Pu}^{3+}$ decay shown in Table IV. Exchange with 1*s* electron is the single most important contribution, but the sum of exchange contributions from *ns* ($n > 1$) electrons is of comparable magnitude to the 1*s* exchange contribution. The total enhancement in the phase-space integral due to exchange is approximately 6.3% for $^{106}\mathrm{Ru}^{1+}$ decay and 7.5% for $^{241}\mathrm{Pu}^{3+}$ decay. This would suggest that exchange effects may significantly modify the phase-space integrals of low-energy β emitters and should be taken account of if an accuracy in the value for the phase-space integral of order 10% or better is required.

F. Application to β decays of other nuclei

There have been recent reports that experimental β spectra of $^{14}\mathrm{C}$ [13] and $^{35}\mathrm{S}$ [11,12] deviate from the theoretical spectra in a region approximately 17 keV from the end point. These deviations have been interpreted as evidence for a heavy antineutrino of mass 17 keV which was first postulated to explain an apparent distortion of the tritium β spectrum [6,7]. Since it may be of interest to define the atomic distortion of the β spectrum very precisely for these isotopes, exchange effects have been calculated using the nonrelativistic hydrogenic approximation described above. Figure 7 shows the total exchange factors for $^{14}\mathrm{C}$ and $^{35}\mathrm{S}$ calculated in this way. The exchange process enhances the β spectrum by approximately 0.003% for $^{14}\mathrm{C}$ and 0.05% for $^{35}\mathrm{S}$ in the region 17 keV from the end point. The energy dependence of the exchange factor is, however, rather weak with the result that exchange effects need not be considered at the level of accuracy necessary for the interpretation of the experimental data on the 17-keV antineutrino.

TABLE IV. Exchange contributions to the phase-space integral for $^{106}\mathrm{Ru}^{1+}$ and $^{241}\mathrm{Pu}^{3+}$ decays.

	Exchange contribution (%)	
Orbital	$^{106}\mathrm{Ru}^{1+}$	$^{241}\mathrm{Pu}^{3+}$i
1*s*	3.31	2.19
2*s*	1.57	2.01
3*s*	0.89	1.42
4*s*	0.57	0.94
5*s*		0.56
6*s*		0.34
Total	6.3	7.5

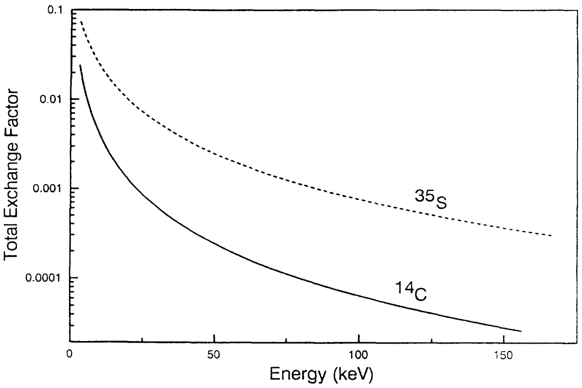

FIG. 7. Total exchange factors for $^{14}\mathrm{C}$ and $^{35}\mathrm{S}$ calculated using the unscreened hydrogenic approximation.

In order to provide some estimate of exchange effects for general β emitters, we have performed calculations of exchange effects in the unscreened hydrogenic approximation for a range of initial nuclear charges from 10 to 90 and electron energies up to 200 keV. The total exchange factors are plotted in Fig. 8 as a function of energy. In all cases exchange with occupied *s* orbitals up to

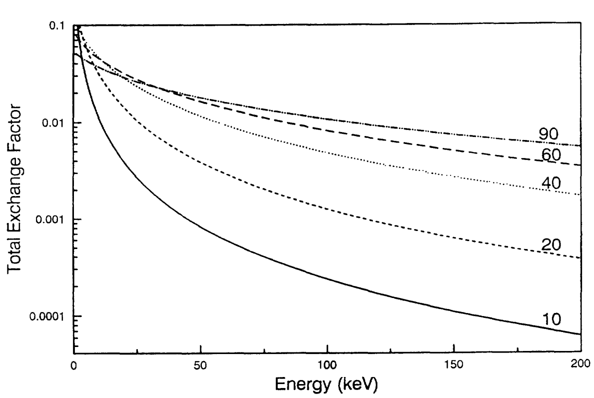

FIG. 8. Total exchange factors for decay of isotopes of different initial nuclear charge, Z_i, calculated in the unscreened hydrogenic approximation. Exchange with occupied subshells up to and including the 3*s* subshell is included.

and including the 3*s* orbital was included. On the basis of the previous discussion, this approach should yield the dominant contributions to the exchange effects. The exchange effects were truncated at the 3*s* subshell since the unscreened hydrogenic approximation is not likely to be a good approximation for high-*n* orbitals particularly in medium- or high-*Z* atoms and the exchange factors for 4*s* and higher subshells are negligible. Except at very low energy, the total exchange factors are seen to become progressively larger as the nuclear charge increases. Clearly the total exchange factor decreases with increasing energy and thus exchange effects are entirely negligible for β emitters with end-point energies greater than about 100 keV, irrespective of the *Z* value.

G. Chemical effects on the exchange process

Since the exchange process involves bound electron wave functions it may possibly be sensitive to the chemical state of the element undergoing β decay and it may therefore be interesting to consider to what extent the half-life for β decay will be modified in different chemical states as a result of exchange. An estimate of the chemical effect on the exchange process has been made by comparing the exchange contributions calculated in the screened hydrogenic approximation for two chemical states of ^{106}Ru. One state chosen was the Ru^{1+} $[\mathrm{Kr}]4d^7$ ion, on which calculations were reported above. The second state was the Ru^{2+} $[\mathrm{Kr}]4d^6$ ion which decays to a core final state Rh^{3+} $[\mathrm{Kr}]4d^6$. Hartree-Fock mean radii of the *s* orbitals of these states of Ru^{2+} and Rh^{3+} are shown in Table V, together with the values of Z_i and Z_f calculated from Eq. (28). Using the nonrelativistic screened hydrogenic approximation, the phase-space integral for $^{106}\mathrm{Ru}^{2+}$ was found to be larger than that for $^{106}\mathrm{Ru}^{1+}$ by 0.0026% as a result of differences in exchange effects in these two ions. Thus chemical effects on the exchange process are very weak. This result can be compared with a calculation [19] of other chemical effects on the decay constant of ^{106}Ru. Besides exchange, there are three other principal factors which lead to a chemical dependence of the half-life or decay constant: changes in atomic electron binding energies which determine the end-point energy, changes in the bound-state β-decay rate, and changes in atomic electron screening of the β-particle wave function [19,33]. In Ref. [19] it was shown that the first two effects are dominant and individually result in fractional changes in the total decay constant of order 0.1% between chemical states of ^{106}Ru differing by one unit of charge. However, these effects tend to partially cancel one another with the result that the net chemical effects are somewhat less than 0.1%. Thus (ignoring exchange effects) the total decay constant of $^{106}\mathrm{Ru}^{2+}$ was found to be larger than that of $^{106}\mathrm{Ru}^{1+}$ by approximately 0.03%. The change in decay constant of ^{106}Ru due to chemical effects on the exchange process reported here is thus considerably smaller than the change due to these other chemical effects on the decay process.

V. CONCLUSION

A general expression has been derived to describe the effect of exchange between bound and continuum electrons on the β spectrum of a multielectron atom. If a screened hydrogenic approximation is used to describe the electron wave functions then the effect of exchange due to a given electron subshell can be expressed in a simple analytical form. This shows that, for the dominant atomic final states produced in the decay, exchange with bound *s* electrons necessarily leads to an enhancement of the β-decay rate. This result is the opposite of that reported in a previous calculation which appears to contain an error in the evaluation of the overlap integral between bound and continuum electron wave functions.

The low-energy β decay of ^{106}Ru has been used as an example for the detailed comparison of exchange effects calculated in different approximations. The results obtained with a screened hydrogenic approximation, with effective nuclear charges chosen to reproduce Hartree-Fock mean orbital radii, agree well with those calculated using a full Hartree-Fock approximation for the electron wave functions. Relativistic effects on the exchange process have been shown to be negligible for low- and medium-*Z* decays but become significant for high-*Z* decays. The simple analytic results obtained in the hydrogenic approximation thus represent a convenient way of calculating exchange effects in a general β spectrum of a low- or medium-*Z* beta emitter.

Detailed calculations on the β spectra of $^{106}\mathrm{Ru}^{1+}$ and $^{241}\mathrm{Pu}^{3+}$ have shown that the inclusion of exchange leads to a distortion of the Kurie plot, but this distortion is very small except at very low electron energies, essentially because the exchange effects are not strongly energy dependent except at low electron energy. However, exchange effects on the phase-space integrals are considerably more significant. For ^{106}Ru, integrating over the β spectrum shows that the inclusion of exchange leads to an increase in the phase-space integral for the decay of

TABLE V. Mean radii of *s* orbitals in Ru^{2+} and Rh^{3+}, calculated in the Hartree-Fock approximation, and associated effective nuclear charges (Z_i and Z_f) of hydrogenic *s* orbitals calculated according to Eqs. (28) and (29).

n	$\langle r \rangle_{\mathrm{Ru}^{2+},ns}$ (a.u.)	Z_i	$\langle r \rangle_{\mathrm{Rh}^{3+},ns}$ (a.u.)	Z_f
1	0.034 624	43.322	0.033 845	44.320
2	0.150 600	39.841	0.146 992	40.819
3	0.412 020	32.765	0.400 272	33.727
4	1.050 732	22.841	1.005 635	23.866

6.3%. For the first forbidden β decay of ^{241}Pu, the exchange effect is found to increase the phase-space integral by 7.5%. These results suggest that if the absolute theoretical β intensity of a low-energy β emitter (with end-point energy less than approximately 100 keV) is required to an accuracy of 10% or better, then exchange effects should be taken into consideration.

Results have also been presented for exchange effects on the β spectra of ^{14}C and ^{35}S, in which recently reported spectral deviations have been interpreted as evidence for a heavy neutrino of mass 17 keV. Exchange effects in the region 17 keV from the end point would appear to enhance the intensity of the spectrum by approximately 0.003% for ^{14}C and 0.05% for ^{35}S. The energy dependence of the exchange process is relatively weak, however, with the result that the shape of the β spectrum is not significantly affected at the level of significance relevant for the determination of antineutrino mass effects.

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APPENDIX

Consider an overall $(N+1)$-electron final state $|N+1,\bar{\gamma},E,\kappa_e,J',M'\rangle$ as defined in Sec. II. This can be constructed by vector coupling the states of the s and $\bar{p}$ electrons (denoted $|\gamma_v',E,\kappa_e,J_s',M_s'\rangle$) to the states of the electrons with $j>\frac{1}{2}$ (denoted $|\gamma_b',J,M_b'\rangle$ where M_b' is the z component of the total angular momentum of these electrons) according to

$$|N+1,\bar{\gamma},E,\kappa_e,J',M'\rangle \equiv |N+1,\gamma_b',J,\gamma_v',E,\kappa_e,J_s',J',M'\rangle \tag{A1}$$

$$=\sum_{M_s'}\langle J,J_s',M'-M_s',M_s'|J',M'\rangle|\gamma_b',J,M'-M_s'\rangle|\gamma_v',E,\kappa_e,J_s',M_s'\rangle\ , \tag{A2}$$

where $\langle J,J_s',M'-M_s',M_s'|J',M'\rangle$ is a vector coupling coefficient [34]. The reduced-matrix elements in Eq. (2) arise from writing the Wigner-Eckart theorem in the form used by Racah [35], thus being greater by a factor of $(2J'+1)^{1/2}$ than the more conventional ones of [34]. After using the appropriate relation on p. 152 of [34] suitably modified to take account of this difference, the reduced-matrix elements in Eq. (2) can be written in the form

$$\langle N+1,\bar{\gamma},E,\kappa_e,J'\|\hat{b}_G^{\dagger}\|N,\gamma,J\rangle \equiv \langle N+1,\gamma_b',J,\gamma_v',E,\kappa_e,J_s',J'\|\hat{b}_G^{\dagger}\|\gamma_a,\gamma_u,J\rangle\ , \tag{A3}$$

$$=(-1)^{J+J'+J_s'}[(2J+1)(2J'+1)]^{1/2}\begin{Bmatrix} J' & J & J_s' \\ 0 & J_s' & J \end{Bmatrix}\langle\gamma_b',J|\gamma_a,J\rangle\langle\gamma_v',E,\kappa_e,J_s'\|\hat{b}_G^{\dagger}\|\gamma_u\rangle\ , \tag{A4}$$

where G denotes any s or $\bar{p}$ subshell. The M quantum numbers on the overlaps $\langle\gamma_b',J|\gamma_a,J\rangle$ have been suppressed for convenience since the overlap $\langle\gamma_b',J,M'|\gamma_a,J,M\rangle$ vanishes unless $M'=M$ in which case the result is independent of M. On simplifying the 6j symbol and noting that $J'=J\pm\frac{1}{2}$, one has

$$\langle N+1,\bar{\gamma},E,\kappa_e,J'\|\hat{b}_G^{\dagger}\|N,\gamma,J\rangle=[(2J'+1)/2]^{1/2}\langle\gamma_b',J|\gamma_a,J\rangle\langle\gamma_v',E,\kappa_e,J_s'\|\hat{b}_G^{\dagger}\|\gamma_u\rangle\ . \tag{A5}$$

This procedure has thus allowed the wave functions for the $J>\frac{1}{2}$ electrons to be factored out of the reduced-matrix elements. Since the total angular momentum of the state γ_u is zero and the operator $\hat{b}_G^{\dagger}$ creates an electron with $j=\frac{1}{2}$, the reduced-matrix element in Eq. (A5) is zero unless $J_s'=\frac{1}{2}$. It should be noted that this result has been obtained using a different order of the angular-momentum coupling of the electrons from that used to derive Eq. (2.68) of [19]. However, it is straightforward to show that Eq. (A5) is valid even if the order of coupling of the angular momenta is changed.

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