Dirac-Fock method for muonic atoms: Transition energies, wave functions, and charge densities

J. V. Mallow
Physics Department, Loyola University of Chicago, Chicago, Illinois 60626

J. P. Desclaux
Centre d'Etudes Nucléaires de Grenoble, Département de Recherche Fondamentale, CPN-85X, 38041 Grenoble Cédex, France

A. J. Freeman
Physics Department, Northwestern University, Evanston, Illinois 60201
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We have performed Dirac-Fock calculations for a variety of muonic atoms. The muon is explicitly included in the self-consistent field theory, and electron exchange is treated exactly. Wave functions and energies are obtained for a variety of sample systems ranging from light to heavy ions, using finite models of the nucleus. The problem of angular momentum coupling schemes for muonic atoms is discussed. Results presented include comparison of Dirac-Fock and Dirac-Slater models, the effect of the inclusion of the Breit operator and the Lamb shift as perturbations, contributions of electron screening and relaxation to muon transition energies, and the dependence of screening on muonic angular momentum.

I. INTRODUCTION

The study of muonic atoms has become, in the last decade, an important tool for understanding the detailed structure of nuclei, the interaction between the nucleus and the atomic electrons, magnetic properties, and even the possible limitations on the fundamental theory of quantum electrodynamics. The advent of high-intensity muon beams has made possible a wide range of experiments in atomic, nuclear, and solid-state physics. These investigations include transition energies between excited muonic states in atoms,[1] the use of the negative muon as a probe of distribution of nuclear charge[2] and spin,[3] and the study of magnetic properties of solids using such techniques as muon spin precession.[4] In addition, isotope and isomer shifts, as well as magnetic hyperfine constants, have been measured for a large number of muonic atoms.[5,6]

Until five years ago, available results from muonic atom experiments were explained on the basis of two-body (muon and nucleus) "hydrogenic" models, in which it was assumed that the muon was so close to the nucleus that the effect of the atomic electrons could be ignored.[7-10] These models were not entirely satisfactory, even for the older experiments, and are even less capable of explaining recent results where electron-muon interactions are pronounced, and where the experimental errors in transition energies are typically less than 15 eV. For example, the muonic x-ray transitions between highly excited states ($n_\mu \geq 5$, where n_μ is the muon principal quantum number) presently being measured by several groups, are strongly affected by electron densities in the regions in which they occur. Ponomarev[11] has shown that transition intensities are dependent on the atomic charge state, i.e., not only the core but the valence atomic electrons, and even on the details of molecular structure in polyatomic systems. As pointed out by Stroke,[12] the Bohr-Weisskopf hyperfine anomaly[13] is not adequately treated by two-body theory, since the nuclear and muonic charge distributions may severely modify the electronic wave functions, which in turn change the hyperfine structure. Yamazaki and co-workers at Berkeley have observed muon spin-rotation at the oxygen site in MnO, and attempted with some success to explain the resultant hyperfine field on the basis of simple overlap models with a single covalency parameter.[4] Hartmann and co-workers[14] have measured transitions from muonic p states with quantum numbers $2 \leq n_\mu \leq 20$.

On the theoretical side, considerably less work has been published. In the work of Fricke and Desclaux,[15] the Dirac-Fock (DF) method was used to study two-muonic atoms; in all the other calculations the Slater free-electron exchange approximation was employed. Vogel[16,17] has solved the Dirac equation for the muon in the field of the nucleus plus a screening field of the electrons. Both Fricke[18] and Vogel *et al.*[19] have carried out fully self-consistent Dirac-Slater calculations in which electrons and muon are included. The results between the self-consistent calculations and the previous ones agree well for low muon quantum number ($n_\mu < 8$). For highly excited muons

however, the muon-electron interactions are more complicated and so cannot be accounted for with a static screening model; the results are found to differ by as much as hundreds of eV.

The present experimental accuracy makes it worthwhile to consider in detail all the approximations used in the theoretical calculations. This paper[20] presents a fully self-consistent DF method for determining the structure of muonic atoms and considers the quantum electrodynamic corrections to the electronic energy both with respect to muon binding energies, and muon x-ray energies. We also compare the case of exact exchange between electrons (as treated in the DF scheme) with the Slater free-electron exchange approximation when the muon is explicitly included in the self-consistent-field (SCF) procedure.

II. THEORETICAL METHOD

The energy eigenvalue equation for a many-electron atom is

$$H_1\Psi = E\Psi , \tag{1}$$

where, in the relativistic case, H_1 is the usual approximate Breit-Dirac Hamiltonian:

$$H_1 = \sum_j [\alpha \cdot c\vec{p}_j + \beta E_0 + V_n(r_j)] + \sum_{i<j}\left(\frac{1}{r_{ij}} + H_B(i,j)\right). \tag{2}$$

The summations run from 1 to N, where N is the number of electrons. The first summation in the one-electron Hamiltonian includes kinetic energy, spin-orbit interaction, and the electron-nucleus Coulomb interaction. Note that the nuclear potential V_n is not restricted to that arising from a nuclear point-charge distribution, but can account for the more physical finite-nuclear-charge distribution which removes the singularity on the s and $p_{1/2}$ orbitals at $r=0$. The $1/r_{ij}$ term is the electron-electron Coulomb repulsion and $H_B(i,j)$ is the Breit operator which takes into account magnetic interaction and retardation. The Breit interaction is used only as a first-order perturbation to partially correct for the relativistic interaction between the electrons. Also, as a perturbation for heavy atoms we have considered the Lamb-shift correction, calculated as the expectation value of the Uehling[21] potential for the vacuum polarization and the hydrogenic results of Mohr,[22] with a screening constant determined from the expectation value of r, for the self-energy.

Antisymmetrization of the total wave function, the central-field approximation, and application of the variational principle then lead to a series of coupled integro-differential equations for the one-electron radial functions:

$$\frac{d}{dr}\begin{pmatrix} P_i(r) \\ Q_i(r) \end{pmatrix} = \begin{pmatrix} -\kappa_i/r & 2c + (1/c)[\epsilon_i - V_i(r)] \\ -(1/c)[\epsilon_i - V_i(r)] & \kappa_i/r \end{pmatrix} \times \begin{pmatrix} P_i(r) \\ Q_i(r) \end{pmatrix} + \begin{pmatrix} X_{Q_i}(r) \\ X_{P_i}(r) \end{pmatrix}. \tag{3}$$

P_i and Q_i are the large and small parts of the radial function, $V_i(r)$ includes the Coulomb interactions between the ith electron and the nucleus, and between the ith electron and the other electrons. X_{P_i} and X_{Q_i} are the exchange interactions with the ith electron. ϵ_i is the one-electron energy and κ_i is the quantum number

$$\kappa = \begin{cases} l & \text{if } j = l - \frac{1}{2} \\ -(l+1) & \text{if } j = l + \frac{1}{2} . \end{cases} \tag{4}$$

The radial functions $P_i(r)$ and $Q_i(r)$ are varied until a self-consistency criterion is met.

If we now add to the N-electron atom a negative muon in a bound state, we add to the atomic Hamiltonian those terms which correspond to the muon's kinetic energy and its Coulomb interaction with the nucleus and the electrons:

$$H_\mu = H_1 + \alpha \cdot c\vec{p}_\mu + \beta E_0(\mu) + V_n(r_\mu) + \sum_{k=1}^{N} \frac{1}{r_{\mu k}} . \tag{5}$$

The effect on the resultant DF system of equations is to include an equation for the muon in the average field of the nucleus and the electrons, and to add a muon-electron Coulomb interaction term to the one-electron equations. Since muon and electron are distinct particles, there is no muon-electron exchange interaction. The method of solution of these equations is the same as for ordinary atoms.

As an aid in understanding some of the problems encountered in the study of muonic atoms, we were guided by our experience in performing atomic calculations. We have recently used both restricted and unrestricted forms of theory to calculate isomer shifts and core-polarization hyperfine fields, respectively, in heavy (high-Z) atoms.[23,24] In Dirac theory, the one-electron orbitals are jj coupled; to obtain appropriate LS or intermediate coupling states for the total atom, it is necessary to construct a linear combination of jj wave functions. The various one-electron orbitals are restricted in the sense that the radial parts of orbitals of different magnetic quantum number m_j are the same. As has been shown for Hartree-Fock theory,[25] this restriction is appro-

TABLE I. Electron screening contributions to muonic transition energies [Δ_{SCF} calculations].

Muon transition	ΔE_{tot} (eV) $Ba^{+1}+\mu^-$	$Ba^{+56}+\mu^-$	ΔE_{scr} (eV)	ΔE_{scr} [a]
$8d_{3/2}$-$4f_{5/2}$	415 006	415 392	386	400
$14f_{5/2}$-$8d_{5/2}$	92 642	93 769	1127	1243
	$Br^{+1}+\mu^-$	$Br^{+35}+\mu^-$		
$14d_{5/2}$-$8p_{3/2}$	35 723	36 355	632	690
$15d_{5/2}$-$8p_{3/2}$	37 880	38 612	732	...

[a] Reference 17.

priate for calculations which involve energies and charge rather than spin densities.

For muonic atoms, the situation is similar. Energies, charge densities, and wave functions were obtained using the m_j-restricted muonic DF scheme. As is well-known for ordinary atoms, *total* energies of the various states are affected very little by the m_j restriction on the orbitals. In particular, the restriction on m_j introduces much less error than choosing the incorrect coupling scheme (e.g., the use of a single jj determinant). We need emphasize that it is possible for us to calculate muonic x-ray transition energies and matrix elements for systems exhibiting jj, LS, or intermediate coupling, using the multiconfiguration method. Thus we are able, in a completely *ab initio* manner, to investigate the effect of choosing various coupling schemes.

III. RESULTS AND DISCUSSION

Table I shows electron screening effects for two sample systems: muonic Ba and Br. Screening is calculated by first performing separate (DF) calculations on the neutral and bare muonic atom, columns 2 and 3, respectively, for each of the muon states in column 1. The transition energies in columns 2 and 3 are differences of total energies of the various muonic excited states; the screening in column 4 is the difference in transition energies between bare and neutral atom.

For the bare muonic-atom calculations, we have solved the one-muon Dirac equation in the field of a finite nucleus. For the neutral atoms, we have approximated the actual intermediate coupling state by a mixed-configuration state.[26] In all of our calculations, we assume a spherical nuclear-charge distribution of radius $R = 1.2\,A^{1/3}$ F, except for nuclei for which Fermi-distribution data are available[5]; in these latter cases, we describe the nucleus by a Fermi charge distribution using the tabulated parameters. Our results for total energies are converged to better than 15 eV in all cases, and to better than 5 eV in most. Since the expectation value of the muon's orbital radius is scaled down from the electron Bohr radius by n_μ^2/m_μ, where n_μ is the muon principal quantum number and m_μ is the reduced muon mass $\simeq 200 m_{e^-}$, the muon orbit coincides with the electron K shell for $n_\mu \simeq 14$. Conservatively then, one should expect to see inner, as well as outer, screening of the muon by electrons for $n_\mu \gtrsim 8$. Our results for Ba in Table I illustrate these screening effects rather vividly. For the transition $8d_{3/2}$-$4f_{5/2}$, our exact screening results (column 4) agree to within 14 eV with the outer-screening calculations.[17] For the transition $14f_{5/2}$-$8d_{3/2}$, however, the $14f_{5/2}$ screening is certainly not merely outer screening, and our results are 116 eV lower than the results of the outer screening calculation in column 5.

Results for a lighter ion, Br, also show the importance of calculating muon and electron orbitals self-consistently, especially for transition states in the K-shell region. For the $14d_{5/2}$-$8p_{3/2}$ transition, the exact screening calculation is 58 eV lower than the outer-screening approximation. Also shown are our results for the $15d_{5/2}$-$8p_{3/2}$ transition, some 100 eV above the $14d_{5/2}$-$8p_{3/2}$ transition.

We have also investigated screening effects for different muon angular momentum states of the

TABLE II. Electron screening dependence on angular momentum state, for a muon in the electron K-shell region.

State	ϵ_μ (eV) $Ba^{+1}+\mu^-$	$Ba^{+56}+\mu^-$	E_{scr} (eV)
$8s_{1/2}$	−123 573	−132 478	8905
$8p_{1/2}$	−129 314	−138 266	8952
$8d_{5/2}$	−129 345	−138 309	8964
$8f_{7/2}$	−129 089	−138 071	8982

TABLE III. Relaxation effects in muonic Δ_{SCF} calculations. ϵ_μ is the one-muon energy in an SCF calculation; E is the total energy from the same calculation; $\Delta\epsilon_\mu$ is the difference in one-muon energies obtained from separate SCF calculations for two states; ΔE is the total energy difference between these two states; $\Delta\epsilon_\mu - \Delta E$ gives an estimate of the relaxation of orbitals in transitions between two states.

Muon orbital	One-muon energy ϵ_μ (eV)	Total energy E	$\Delta\epsilon_\mu$	ΔE	$\Delta\epsilon_\mu - \Delta E$
		Case 1: $O^{+1}+\mu^-$			
$1s_{1/2}$	1.7717×10^5	1.7929×10^5			
$4p_{3/2}$	1.0671×10^4	1.2651×10^4	166 500	166 632	−1312
$9s_{1/2}$	1.7371×10^3	3.7284×10^3	8 934	8 928	+6
$14p_{3/2}$	5.2846×10^2	2.5319×10^3	1 209	1 197	+12
$20p_{3/2}$	1.6503×10^2	2.1738×10^3	363	358	+5
		Case 2: $Br^{+1}+\mu^-$			
$8p_{3/2}$	4.9387×10^4	1.2019×10^5			
$15d_{5/2}$	1.1530×10^4	8.2310×10^4	37 857	37 878	−21
$15p_{3/2}$	1.1517×10^4	8.2325×10^4	13	−15	+28
		Case 3: $Ba^{+1}+\mu^-$			
$4f_{5/2}$	5.4452×10^5	7.6605×10^5			
$8d_{3/2}$	1.2983×10^5	3.5104×10^5	414 692	415 006	−314
$14f_{5/2}$	3.7247×10^4	2.5840×10^5	92 582	92 642	−60

same principal quantum number. Typical results are shown in Table II. We have subtracted one-muon energies for bare and neutral Ba to obtain the screening, which, not surprisingly, increases monotonically with l. It is clear from the table that in order to obtain the correct screening corrections for muonic transition energies to within present experimental errors, one must correctly identify the angular momentum states involved in the transition. Transition moments, even more than energies, are extremely sensitive to the details of the state wave function, and are the subject of investigations to be reported on later.

In Table III, we display the results of our investigation of the validity of Koopmans' theorem for muonic transitions. For our results in Table I, we took total energy differences; in Table II, we considered one-muon energies. Table III exhibits explicitly the effect of relaxation on transition energies for muonic oxygen, bromine, and barium. It is clear from column 6 that relaxation effects vary both in magnitude and sign. In particular, for transitions involving low-lying states with large absolute energies, relaxation effects are correspondingly large. Thus, Δ_{SCF} calculations of muonic transition energies are essential.

TABLE IV. Charge densities and energies of $1s$ electrons in excited muonic atoms. $\phi^{e^-}_{1s}(0)$ is the value of the $1s$ orbital at $r=0$, $4\pi|\phi^{e^-}_{1s}(0)|^2$ is the probability density of the $1s$ electron at the nucleus ($r=0$), and $\epsilon^{e^-}_{1s}$ is the $1s$ electron energy.

Atom	Muon orbital	$4\pi\lvert\phi^{e^-}_{1s}(0)\rvert^2$ (a.u.)	$\epsilon^{e^-}_{1s}$ (eV)
$O^{+1}+\mu^-$	$4p_{3/2}$	1309	427
	$9s_{1/2}$	1534	445
	$14p_{3/2}$	1785	409
	$20p_{3/2}$	1891	538
$Br^{+1}+\mu^-$	$8p_{3/2}$	2.343×10^5	12 824
	$15d_{5/2}$	2.448×10^5	13 139
$Ba^{+1}+\mu^-$	$4f_{5/2}$	1.631×10^6	36 173
	$8d_{5/2}$	1.678×10^6	36 336
	$14f_{5/2}$	1.725×10^6	36 813

Energies and charge densities in the nuclear region for $1s$ electrons in several muonic atoms with the muon in a variety of excited states are presented in Table IV. One may think of these quantities as a measure of the screening of the $1s$ electrons by the muon. Not surprisingly, the state of higher muon quantum number screens the nucleus less efficiently, and thus yields higher charge densities of the $1s$ electrons at the nucleus, as well as greater (more negative) one-electron energies. Of course, the actual values of $|\phi^{e^-}_{1s}(0)|^2$ and $\epsilon^{e^-}_{1s}$ depend on the precise overlaps of muonic and electronic orbitals.

The percentage difference in $1s$ screening between different muonic states decreases rapidly with increasing Z. For muonic oxygen $|\phi^{e^-}_{1s}(0)|^2$ and $\epsilon^{e^-}_{1s}$ increase by 16% and 10%, respectively, as n_μ goes from 9 to 14. Similar changes in n_μ

TABLE V. Total energies, and muon binding energies ($E_{B\mu}$) in muonic oxygen. All values are obtained in eV. Energies are obtained in the Dirac states ($\rho^{1/3}$ exchange) and Dirac-Fock (exact exchange) schemes.

	Dirac Slater	Dirac Fock
O^{+1}	1997.6	2021.2
$O^{+1}+\mu(9s)$	3708.5	3728.7
$O^{+1}+\mu(14p_{3/2})$	2510.9	2532.3
$E_{B\mu}(9s)$	1710.9	1707.5
$E_{B\mu}(14p_{3/2})$	513.3	510.1

TABLES VI. Total energies and muon binding energies (in eV) for lead.

	Dirac-Slater	Dirac-Fock	
		a	b
Pb^{+1}	568 833	569 131	568 066
$Pb^{+1}+\mu(7s)$	910 667	910 969	909 950
$Pb^{+1}+\mu(12s)$	679 825	680 122	679 090
$E_{B\mu}(7s)$	341 834	341 838	341 884
$E_{B\mu}(12s)$	110 992	110 991	111 024

[a] Unperturbed Dirac-Fock.
[b] Breit operator and Lamb shift included.

for muonic bromine produce changes in charge density and energy of 4% and 2%, respectively; for barium, the changes are 3% and 1%, respectively. The more tightly bound muons in the heavier atoms are correspondingly closer to the nucleus; thus, their screening of the 1s electrons is proportionally less sensitive to the actual muon state.

Finally, we present in Tables V and VI a comparison between the Dirac-Slater and DF calculations. This permits us to judge the importance of treating the exchange terms exactly in the SCF calculations. As can be seen, although the total energies are rather different, the muon binding energy is practically unchanged in the two calculations. Note that the binding energy of the muon is calculated as the difference between total energies and not taken as the one-particle eigenvalue (Koopmans' theorem). Comparing, in Table VI, the two sets of DF results, (a) unperturbed and (b) including the Breit operator and Lamb-shift correction, it can be seen that these higher-order relativistic corrections are by no means negligible compared to present experimental accuracy. It must be noted that in these cases the muon is either well inside the electron K shell ($n_\mu=7$) or almost in the same space region ($n_\mu=12$), and consequently the muon screens the nuclear charge for the K-shell electrons compared to what they experience in the absence of the muon. As the higher-order relativistic corrections have a Z^4 dependence, the change in the screening substantially modifies the value of the correction.

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