Dual Kinetic Balance Approach to Basis-Set Expansions for the Dirac Equation

V. M. Shabaev,[1,2] I. I. Tupitsyn,[1] V. A. Yerokhin,[1,2] G. Plunien,[2] and G. Soff[2]

[1]*Department of Physics, St. Petersburg State University, Oulianovskaya 1, Petrodvorets, St. Petersburg 198504, Russia*
[2]*Institut für Theoretische Physik, TU Dresden, Mommsenstrasse 13, D-01062 Dresden, Germany*

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A new approach to finite basis sets for the Dirac equation is developed. It does not involve spurious states and improves the convergence properties of basis-set calculations. Efficiency of the method is demonstrated for finite basis sets constructed from B splines by calculating the one-loop self-energy correction for a hydrogenlike ion.

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At present, a great variety of calculations in atomic physics and quantum chemistry are based on finite basis sets. The first successful attempts to utilize finite basis sets in relativistic quantum mechanics were made many years ago [1–6]. Application of B splines for constructing basis sets [7–9] provided new impact to this field. Nowadays, B splines are widely employed in computational atomic and molecular physics [10,11].

In contrast to the nonrelativistic case, the use of B splines in the relativistic theory is generally accompanied by the occurrence of spurious states [9]. For the attractive Coulomb potential, spurious states appear for $\kappa > 0$ as the lowest bound states with nonphysical energies [$\kappa = (-1)^{j+l+1/2}(j + 1/2)$ is the quantum number determined by the angular momentum and the parity of the state]. The wave functions of these states oscillate rapidly and, therefore, in many cases they may be disregarded in practical atomic calculations [12,13]. However, since the presence of the spurious states disturbs the spectrum, it worsens the convergence properties of the basis-set calculations in some cases. In particular, this can be a reason of a poor convergence of the B-spline method in calculations of pure radiative corrections for low- and middle-Z systems [14]. To date, most of the QED calculations are performed by means of analytical or numerical representations for the Coulomb-Green function [15] or by the space discretization method [16], in which the spurious states are eliminated from the very beginning. One may expect that a proper solution of the problem of spurious states in the B-spline method would improve its convergence properties in calculations of radiative corrections.

A number of schemes for solving the problem of spurious states were presented previously [9,17–22]. Most of them are limited to a specific choice of finite basis sets (see the related discussion below) or require considerable modifications of the standard numerical procedure. For this reason, their applicability to calculations of QED effects has not yet been examined. We also find that treating the problem by a particular choice of the boundary conditions in the B-spline method [9], that in some cases simply moves the spurious states to the end of the spectrum, does not provide any improvement in calculations of radiative corrections.

In this Letter, we formulate a new method that solves the problem of spurious states and improves the convergence properties of basis-set calculations. The efficiency of the method is demonstrated for finite basis sets constructed from B splines by calculating the one-loop self-energy correction for a hydrogenlike ion.

For the case of a central field $V(r)$, the Dirac wave function is conveniently represented by

$$\psi(\mathbf{r}) = \frac{1}{r}\begin{pmatrix} G(r)\Omega_{\kappa m}(\mathbf{n}) \\ iF(r)\Omega_{-\kappa m}(\mathbf{n}) \end{pmatrix}, \tag{1}$$

where $\mathbf{n} = \mathbf{r}/r$. With this representation, the radial Dirac equations can be written as

$$H_\kappa \phi = E\phi, \tag{2}$$

where (in units: $\hbar = 1$)

$$H_\kappa = \begin{pmatrix} mc^2 + V & c[-\frac{d}{dr} + \frac{\kappa}{r}] \\ c[\frac{d}{dr} + \frac{\kappa}{r}] & -mc^2 + V \end{pmatrix} \tag{3}$$

and

$$\phi(r) = \begin{pmatrix} G(r) \\ F(r) \end{pmatrix} \tag{4}$$

is the two-component radial wave function. The scalar product of the two-component functions is defined by

$$\langle \phi_a | \phi_b \rangle = \int_0^\infty dr [G_a(r)G_b(r) + F_a(r)F_b(r)]. \tag{5}$$

The radial Dirac equations can be derived from an action principle $\delta S = 0$ with

$$S = \langle \phi | H_\kappa | \phi \rangle - E\langle \phi | \phi \rangle, \tag{6}$$

if proper boundary conditions for $G(r)$ and $F(r)$ are implemented. The functions $\phi(r)$ can be approximated by

$$\phi(r) = \sum_{i=1}^{2n} c_i u_i(r), \tag{7}$$

where the two-component functions $u_i(r)$ are assumed to be square integrable, linearly independent, and satisfying

proper boundary condition at $r = 0$ (see below). The variational principle reduces to the following algebraic equations:

$$dS/dc_i = 0, \qquad i = 1, 2, \ldots, 2n. \tag{8}$$

This leads to a generalized eigenvalue problem:

$$K_{ik}c_k = EB_{ik}c_k, \tag{9}$$

where $K_{ik} = (\langle u_i|H_\kappa|u_k\rangle + \langle u_k|H_\kappa|u_i\rangle)/2$, $B_{ik} = \langle u_i|u_k\rangle$, and the summation over repeated indices is implied.

Let us first prove that the widely applied choice

$$u_i(r) = \begin{pmatrix} \pi_i(r) \\ 0 \end{pmatrix}, \qquad i = 1, \ldots, n, \tag{10}$$

$$u_i(r) = \begin{pmatrix} 0 \\ \pi_{i-n}(r) \end{pmatrix}, \qquad i = n+1, \ldots, 2n, \tag{11}$$

where $\{\pi_i(r)\}_{i=1}^n$ are square integrable functions satisfying the boundary condition $\pi_i(0) = 0$, results in the occurrence of spurious states. In this case, Eq. (9) takes the form

$$(mc^2 + V - E)_{ik}p_k + cD_{ik}q_k = 0, \tag{12}$$

$$c(D^\dagger)_{ik}p_k + (-mc^2 + V - E)_{ik}q_k = 0, \tag{13}$$

where $(\pm mc^2 + V - E)$, D, and $D^\dagger$ are $n \times n$ matrices with elements

$$(\pm mc^2 + V - E)_{ik} = \int_0^\infty dr\pi_i(r)(\pm mc^2 + V - E)\pi_k(r), \tag{14}$$

$$D_{ik} = \int_0^\infty dr\pi_i(r)\left(-\frac{d}{dr} + \frac{\kappa}{r}\right)\pi_k(r), \tag{15}$$

$$(D^\dagger)_{ik} = \int_0^\infty dr\pi_i(r)\left(\frac{d}{dr} + \frac{\kappa}{r}\right)\pi_k(r), \tag{16}$$

and $p_i = c_i$, $q_i = c_{i+n}$ for $i = 1, 2, \ldots, n$. Let us consider the nonrelativistic limit ($c \to \infty$) and introduce vectors P and Q with components $\{p_i\}_{i=1}^n$ and $\{q_i\}_{i=1}^n$, respectively. Then Eq. (13) yields $Q = (1/2mc)D^\dagger P$. Substituting this expression into Eq. (12), we obtain

$$DD^\dagger P + 2m(mc^2 + V - E)P = 0. \tag{17}$$

For the pure Coulomb field, $V(r) = -\lambda/r$ ($\lambda > 0$), introducing the matrix $C_{ik} = D_{ik} - (m\lambda/\kappa)\delta_{ik}$, Eq. (17) reduces to

$$C_\kappa C_\kappa^\dagger P = \epsilon P, \tag{18}$$

where $\epsilon = [2m(E - mc^2) + m^2\lambda^2/\kappa^2]$ and the dependence of the C matrix on κ is explicitly indicated. For $\kappa' = -\kappa$, taking into account that $C_{-\kappa} = -C_\kappa^\dagger$, we find that the corresponding eigenvalue equation

$$C_{\kappa'}C_{\kappa'}^\dagger P' = \epsilon' P' \tag{19}$$

can be written as

$$C_\kappa^\dagger C_\kappa P' = \epsilon' P'. \tag{20}$$

On the other hand, multiplying Eq. (18) with $C_\kappa^\dagger$ yields

$$C_\kappa^\dagger C_\kappa (C_\kappa^\dagger P) = C_\kappa^\dagger C_\kappa \tilde{P} = \epsilon\tilde{P}, \tag{21}$$

where $\tilde{P} \equiv C_\kappa^\dagger P$. This implies that each nonzero eigenvalue of $C_\kappa C_\kappa^\dagger$ is an eigenvalue of $C_\kappa^\dagger C_\kappa$. Evidently, the inverse statement can be proven in a similar manner and the dimension of a nonzero eigenvalue subspace is the same for $C_\kappa C_\kappa^\dagger$ and $C_\kappa^\dagger C_\kappa$. Accordingly, the spectra of $C_\kappa C_\kappa^\dagger$ and $C_\kappa^\dagger C_\kappa$ may differ only by the dimension of the zero eigenvalue subspace. For finite matrices, the dimension of the subspace with $\epsilon = 0$ is the same for $C_\kappa C_\kappa^\dagger$ and $C_\kappa^\dagger C_\kappa$, since the total number of eigenvectors, as well as the dimension of the nonzero eigenvalues subspace, is the same for $C_\kappa C_\kappa^\dagger$ and $C_\kappa^\dagger C_\kappa$. Therefore, the finite matrices $C_\kappa C_\kappa^\dagger$ and $C_\kappa^\dagger C_\kappa = C_{-\kappa}C_{-\kappa}^\dagger$ have an identical spectrum. Conversely, we know that the exact analytical solution of the Dirac equation for the Coulomb potential yields different lowest bound-state energies for $\kappa < 0$ and $\kappa > 0$. This is due to the fact that within the exact (infinite dimension) treatment the subspace with $\epsilon = 0$ may have different dimensions for $\kappa < 0$ and $\kappa > 0$ cases. This can easily be checked by solving the equation

$$(d/dr + \kappa/r - m\lambda/\kappa)G(r) = 0, \tag{22}$$

which in case of finite dimensions is equivalent to the equation $C_\kappa^\dagger P = 0$. Solving Eq. (22) yields $G(r) = A_0 r^{-\kappa}\exp(m\lambda/\kappa r)$. For $\kappa < 0$, this solution has the proper behavior at $r \to 0$ and at $r \to \infty$. However, this does not hold for $\kappa > 0$. Therefore, the states with $\epsilon = 0$ and $\kappa > 0$, obtained in the finite dimension approximation, are spurious. They correspond to energy $E - mc^2 = -m\lambda^2/2\kappa^2$. This ends the proof. It is obvious that spurious states must occur for any other potential one is dealing with in atomic calculations.

To eliminate the spurious states, in Ref. [18] "kinetically balanced" Slater-type functions were employed. Within this method, for $\kappa > 0$ the lower components in Eq. (11) are replaced by functions $\rho_i(r)$ which, in the nonrelativistic limit, are related to the upper components $\pi_i(r)$ in Eq. (10) via

$$\rho_i(r) \approx (1/2mc)(d/dr + \kappa/r)\pi_i(r). \tag{23}$$

In Refs. [19,21], the basis set was constructed from Gaussian spinors that satisfy the boundary conditions associated with the finite nucleus. These functions automatically satisfy the kinetic-balance condition for a finite value of c and represent accurately the wave functions at the origin of the nucleus. Both methods provide a high accuracy in calculations of bound-state energies for extended nuclei. However, the applicability of these methods to calculations of the QED corrections has not yet been investigated [23].

In the original version of the B-spline method [8–10], to achieve that the first positive-energy states $\kappa > 0$ correspond to physical bound states, an additional term had to be introduced in the Hamiltonian, which formally implements the boundary condition [24]: $G(R) = F(R)$,

where R is the cavity radius, together with the condition $G(0) = 0$. However, since the presence of the additional term does not imply any practical advantages, it is usually omitted in calculations. Instead, the boundary conditions are generally implemented by eliminating the first and the last basis function, which are the only ones that do not vanish at $r = 0$ and $r = R$, respectively. This method was successfully employed for calculations of the two-photon exchange diagrams within the rigorous QED approach [12] and for relativistic calculations of the recoil effect [13]. However, its application to calculations of pure radiative corrections [14] was less successful, compared to the other methods [15,16]. We conjecture that this would not be the case if the spurious states were eliminated in a more natural manner than it was done in Refs. [8,9].

It is known (see, e.g., Refs. [10,14,18]) that the case of the pure Coulomb potential requires generally special care in implementing finite basis-set methods. This is due to the singularity of the Coulomb potential at $r \to 0$. However, in practical calculations it is standard to modify the potential to account for the finite nuclear size, which eliminates this problem. For this reason and for simplicity, we restrict our consideration to the case of a finite nuclear-charge distribution, bearing in mind that the limit of a point nucleus can be treated by extrapolating a series of calculations for extended nuclei to vanishing nuclear size. For extended nuclei, we propose to employ the following basis set:

$$u_i(r) = \begin{pmatrix} \pi_i(r) \\ \frac{1}{2mc}\left(\frac{d}{dr} + \frac{\kappa}{r}\right)\pi_i(r) \end{pmatrix}, \qquad i \le n, \tag{24}$$

$$u_i(r) = \begin{pmatrix} \frac{1}{2mc}\left(\frac{d}{dr} - \frac{\kappa}{r}\right)\pi_{i-n}(r) \\ \pi_{i-n}(r) \end{pmatrix}, \qquad i > n, \tag{25}$$

where the linearly independent functions $\{\pi_i(r)\}_{i=1}^{n}$ are assumed to be square integrable and to provide the boundary condition: $F(0) = 0$ for $\kappa < 0$ and $G(0) = 0$ for $\kappa > 0$. This basis set has the form of solutions of a four-component generalization of the Schrödinger equation which simultaneously describes the nonrelativistic electron and nonrelativistic positron [25]. We state that this basis set satisfies the following requirements: (i) It is symmetric with respect to the replacement $\kappa \to -\kappa$ and the interchange of the upper and lower components. (ii) The functions $u_1, \ldots, u_n$ provide the correct relation between upper and lower components for $|E - mc^2|$, $|V(r)| \ll 2mc^2$, while the functions $u_{n+1}, \ldots, u_{2n}$ do the same for $|E + mc^2|$, $|V(r)| \ll 2mc^2$. (iii) Calculations utilizing the standard finite basis set determined by Eqs. (10) and (11) can be easily adopted when employing the basis (24) and (25). (iv) No spurious states occur for attractive potentials nor for repulsive potentials. The properties (i)–(iii) follow immediately from definitions (24) and (25). The absence of spurious states can be explained as follows. Performing similar steps as for the derivation of Eq. (17), for $|E - mc^2| \ll 2mc^2$ we obtain

$$\frac{1}{2m} LP + (V + mc^2 - E)P = 0, \tag{26}$$

where

$$L_{ik} = \int_0^\infty dr \pi_i(r)\left(-\frac{d}{dr} + \frac{\kappa}{r}\right)\left(\frac{d}{dr} + \frac{\kappa}{r}\right)\pi_k(r). \tag{27}$$

Equation (26) takes the form of the ordinary Schrödinger equation with $l = |\kappa + 1/2| - 1/2$ in the finite basis representation. As is known, it generates no spurious states. The region $|E + mc^2| \ll 2mc^2$, where spurious states may exist for repulsive potentials and for $\kappa < 0$, can be considered similarly. In this case, we obtain the equation

$$\frac{1}{2m} MQ + (-V + mc^2 + E)Q = 0, \tag{28}$$

where

$$M_{ik} = \int_0^\infty dr \pi_i(r)\left(\frac{d}{dr} + \frac{\kappa}{r}\right)\left(-\frac{d}{dr} + \frac{\kappa}{r}\right)\pi_k(r). \tag{29}$$

Equation (28) has also the form of the ordinary Schrödinger equation but with $l' = |\kappa - 1/2| - 1/2$. It transforms into Eq. (26) under the replacements $\kappa \to -\kappa$, $V \to -V$, $E \to -E$, and $Q \to P$ (charge conjugation symmetry) and does not generate any spurious states. This is a consequence of the equivalent treatment of the positive and negative energy states. For this reason, the new basis may be termed conventionally as *dual kinetic-balance* (DKB) basis.

The validity of statement (iv) has also been proven by numerical calculations with $\pi_i(r) = B_i(r)$, where $B_i(r)$ are the B splines defined on the interval $(0, R)$ as in Ref. [9]. The first and the last spline function have been omitted. For $|\kappa| = 1$, this provides the boundary condition at the origin, required above. To obey the same boundary conditions for $|\kappa| \ge 2$, strictly speaking, one should remove the second spline as well. Our calculations show, however, that this has practically no effect on the results. We have found that removing the last two splines, which would implement the boundary condition $G(R) = F(R) = 0$, neither affects the results.

To test the accuracy of the computed wave functions in the nuclear region, we have calculated the nuclear size correction for the uniform sphere and the Fermi model for the nuclear-charge distribution. For $Z = 80$ and the uniform sphere model, our result for the $1s$ binding energy, obtained with 30 spline functions, coincides within nine digits with the result obtained by the Gaussian basis-set method [21]. For $Z = 92$ and the Fermi model ($\langle r^2 \rangle^{1/2} = 5.8604$ fm), we have obtained the nuclear size correction to be 198.81, 37.77, and 4.42 eV for $1s$, $2s$, and $2p_{1/2}$ states, respectively. These values coincide with the exact ones within all digits indicated and were obtained with 30 spline functions, whereas a typical number of functions used in practical

calculations is 2 or 3 times larger. We have also found that the DKB functions mimic equally well the behavior of both radial Dirac components at the origin of an extended nucleus. For instance, for the $1s$ state of Hg^{79+}, using 60 basis functions and the sphere model with $\langle r^2\rangle^{1/2} = 5.4359$ fm (it corresponds to the nuclear radius used in Ref. [21]), we get at $r = 0.1$ fm: $G(r)/r = 2.1768$ and $F(r)/r^2 = -34.81$ (in units: $\hbar = c = m = 1$). These values are in very good agreement with the exact ones, $G(r)/r = 2.1769$ and $F(r)/r^2 = -34.83$, and with the values $[G(r)/r]_{r=0}$ presented in Ref. [21].

Finally, let us consider the calculation of the one-loop self-energy (SE) correction to the ground-state energy of a hydrogenlike ion employing the new basis set. Generally, the SE correction is expanded into the zero-, one-, and many-potential terms. The ultraviolet divergences in the zero- and one-potential terms and in the counterterm cancel each other and their evaluation can be performed according to the formulas presented in Ref. [26]. As for the many-potential term, although it does not contain any ultraviolet divergences, its calculation is most difficult since it involves the summation over the whole Dirac-Coulomb spectrum. In Table I, we compare our results obtained for $Z = 20$ employing the DKB basis set (24), (25) with $\pi_i(r) = B_i(r)$, the old basis (10), (11) with the same $\pi_i(r)$, and the results of a calculation using the analytical representation for the Coulomb-Green function. This comparison clearly demonstrates a significant improvement in accuracy, if the DKB basis is employed instead of the old one.

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TABLE I. The partial-wave contributions to the self-energy correction (in atomic units) for the $1s$ state of hydrogenlike calcium ($Z = 20$), calculated by different basis-set methods and by the Coulomb-Green function (CGF) method. The results are obtained with 60 basis functions and for the shell model for the nuclear-charge distribution with $R = 3.478$ fm. In the basis-set calculations, the part with $|\kappa| \geq 10$ is obtained by an extrapolation.

Term	Old basis	DKB basis	CGF method
$\|\kappa\| = 1$	0.848 691	0.848 750	0.848 741
$\|\kappa\| = 2$	0.020 618	0.020 662	0.020 653
$\|\kappa\| = 3$	0.005 302	0.005 331	0.005 326
$\|\kappa\| = 4$	0.002 121	0.002 139	0.002 137
$\|\kappa\| = 5$	0.001 050	0.001 062	0.001 062
$\|\kappa\| = 6$	0.000 590	0.000 597	0.000 598
$\|\kappa\| \leq 9$	0.879 127	0.879 303	0.879 288
$\|\kappa\| \geq 10$	0.000 587	0.000 585	0.000 583
Total many potential	0.879 71	0.879 89	0.879 87(1)
Zero and one potential	−0.815 626	−0.815 626	−0.815 626
Total SE	0.064 09	0.064 26	0.064 25(1)

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