Dual kinetic balance approach to basis set expansions for the Dirac equation

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Abstract

A new approach to finite basis sets for the Dirac equation is developed. It solves the problem of spurious states and, as a result, 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.
At present, a great variety of calculations in atomic physics and quantum chemistry are based on finite basis sets. 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 (see [10,11] and references therein).

In contrast to the nonrelativistic case, the use of finite basis sets in the relativistic theory is generally accompanied by the occurrence of spurious states (see, e.g., [4,9]). For the attractive Coulomb potential, spurious states appear for \( \kappa > 0 \) as the lowest bound states with non-physical 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). For the point nucleus, their energies coincide with the physical energies of the lowest bound states with \( \kappa < 0 \). The wave functions of these states oscillate rapidly and, therefore, in many cases they may be disregarded in practical atomic calculations. However, since the presence of the spurious states disturbs the spectrum, it worsens the convergence properties of the basis set calculations in some cases. For this reason, despite of the early promising results [13,14], the finite basis set method has not been extensively employed in calculations of radiative corrections. To date, most of these calculations are performed by means of analytical or numerical representations for the Coulomb-Green function (see, e.g., [15–18]) or by the space discretization method [19–21], in which the spurious states are eliminated from the very beginning (see [19] for details).

The problem of spurious states is especially demanding in calculations of atoms in strong external fields and for molecules, where, generally speaking, they can not be selected as the lowest bound-state energies and, therefore, can not be eliminated without employing special methods. Furthermore, even in those cases where spurious states do not play any significant role, their presence generates difficulties for providing an adequate estimate of the accuracy of the calculations.

A number of proposals for solving the problem of spurious states were presented previously [22,9,23–25]. Our analysis of these methods indicates, however, that none of them can
be considered as completely satisfactory. We find that treating the problem by a particular choice of the boundary conditions in the B-spline method [9] does not provide any improvement in calculations of radiative corrections. The other methods either require considerable modifications of the standard numerical procedure or they are strongly limited to a specific choice of finite basis sets. Their applicability to calculations of radiative corrections thus remains questionable. For this reason, we suggest a new method for solving the problem of spurious states. 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

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

where \( n = r/r \). With this representation, the radial Dirac equations can be written as

\[
H_\kappa \phi = E \phi,
\]

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

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

and

\[
\phi(r) = \begin{pmatrix} G(r) \\ F(r) \end{pmatrix}
\]

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

\[
\langle \phi_a | \phi_b \rangle = \int_0^\infty dr \left[ G_a(r) G_b(r) + F_a(r) F_b(r) \right].
\]

The radial Dirac equations can be derived from an action principle \( \delta S = 0 \) with
\[ S = \langle \phi | H_{\kappa} | \phi \rangle - E \langle \phi | \phi \rangle , \quad (6) \]

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

\[ \phi(r) = \sum_{i=1}^{2n} c_i u_i(r) , \quad (7) \]

where the two-component functions \( u_i(r) \) are assumed to be square integrable, linearly independent, and satisfying proper boundary condition at \( r = 0 \). The variational principle reduces to the following algebraic equations

\[ \frac{dS}{dc_i} = 0, \quad i = 1, 2, ..., 2n . \quad (8) \]

This leads to a generalized eigenvalue problem

\[ K_{ik} c_k = E B_{ik} c_k , \quad (9) \]

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 demonstrate that the widely applied choice

\[ u_i(r) = \begin{pmatrix} \pi_i(r) \\ 0 \end{pmatrix}, \quad i = 1, ..., n, \quad (10) \]

\[ u_i(r) = \begin{pmatrix} 0 \\ \pi_{i-n}(r) \end{pmatrix}, \quad i = n + 1, ..., 2n \quad (11) \]

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, equation (9) takes the form

\[ (mc^2 + V - E)_{ik} p_k + c D_{ik} q_k = 0 , \quad (12) \]

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

where \( (\pm mc^2 + V - E), \ D, \) and \( D^\dagger \) are \( n \times n \) matrices with elements
\[(\pm mc^2 + V - E)_{ik} = \int_0^\infty dr \pi_i(r)(\pm mc^2 + V - E)\pi_k(r), \quad (14)\]

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

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

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

\[DD^\dagger P + 2m(mc^2 + V - E)P = 0. \quad (17)\]

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

\[C_{\kappa}C_{\kappa}^\dagger P = \epsilon P, \quad (18)\]

where \(\epsilon = [2m(E - mc^2) + m^2\lambda^2/\kappa^2]\) and the dependence of the \(C\) matrix on \(\kappa\) is explicitly indicated. Taking into account that \(C_{-\kappa} = -C_{\kappa}^\dagger\), we find that the corresponding equation for \(\kappa' = -\kappa\) can be written as

\[C_{\kappa}^\dagger C_{\kappa}P' = \epsilon' P'. \quad (19)\]

Multiplying Eq. \((18)\) with \(C_{\kappa}^\dagger\), we obtain 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

$$(d/dr + \kappa/r - m\lambda/\kappa)G(r) = 0,$$  \hspace{1cm} (20)

which in case of finite dimensions is equivalent to the equation $C^\dagger_\kappa P = 0$. Solving equation (20) yields $G(r) = A_0 r^{-\kappa} \exp(m\lambda/\kappa r)$. For $\kappa < 0$, this solution has the proper behaviour at $r \to 0$ and at $r \to \infty$. However, this does not hold for $\kappa > 0$. Concluding, in the approximation of finite dimensions, our proof clearly indicates the presence of spurious states with energy $E - mc^2 = -m\lambda^2/2\kappa^2$ corresponding to $\epsilon = 0$. It can be shown that this result remains valid for the full relativistic theory as well. 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. [23] "kinetically balanced" Slater type functions were employed. Within this method, for $\kappa > 0$ the lower components in equation (11) are replaced by functions $\rho_i(r)$ which, in the nonrelativistic limit, are related to the upper components $\pi_i(r)$ in equation (10) via

$$\rho_i(r) \approx (1/2mc)(d/dr + \kappa/r)\pi_i(r).$$  \hspace{1cm} (21)

This method provides a high accuracy in calculations of bound-state energies in atoms for both sign of $\kappa$, if an extended nuclear charge distribution is introduced and proper boundary conditions are implemented [23]. However, since the basis functions are "kinetically balanced" for positive energy states only, the application of this method to calculations of the QED corrections might be problematic [26]. The equivalent treatment of positive and negative energy states would provide, in particular, the well-known symmetry properties of the Dirac spectrum under the transformations $V \to -V$, $\kappa \to -\kappa$, and $G \leftrightarrow F$. It is evident that the "kinetically balanced" functions do not meet with this requirement.

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 so-called MIT boundary
condition [12]: $G(R) = F(R)$, 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 [27–31] and for relativistic calculations of the recoil effect [32–34]. However, its application to calculations of pure radiative corrections [13,14] was less successful, compared to the other methods [15–18,20,21]. 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 [8,9].

It is known (see, e.g., [14,10,23]) that the case of the pure Coulomb potential requires generally a 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

$$u_i(r) = \begin{cases} \frac{1}{2mc} \left( \frac{d}{dr} + \frac{\kappa}{r} \right) \pi_i(r), & i \leq n, \\ \frac{1}{2mc} \left( \frac{d}{dr} - \frac{\kappa}{r} \right) \pi_{i-n}(r), & i \geq n + 1, \end{cases}$$

where the linearly independent functions $\{\pi_i(r)\}_{i=1}^n$ are assumed to be square integrable and satisfying the proper boundary condition at $r = 0$. We state that this basis set satisfies the following requirements:

1. It is symmetric with respect to the replacement $\kappa \to -\kappa$ and the interchange of the upper and lower components.
2. The functions \( u_1, ..., u_n \) provide the correct relation between upper and lower components for \( |E - mc^2|, |V(r)| \ll 2mc^2 \), while the functions \( u_{n+1}, ..., u_{2n} \) do the same for \( |E + mc^2|, |V(r)| \ll 2mc^2 \).

3. Calculations utilizing the standard finite basis set determined by Eqs. (10) and (11) can be easily adopted when employing the basis (22)-(23).

4. No spurious states occur for attractive as well as for repulsive potentials.

The properties 1 - 3 follow immediately from definitions (22) and (23). 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

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

where

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

Eq. (24) takes the form of the ordinary Schrödinger equation with \( l = |\kappa| + (\kappa/|\kappa| - 1)/2 \) 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

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

where

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

Eq. (26) has also the form of the ordinary Schrödinger equation but with \( l' = |\kappa| - (\kappa/|\kappa| + 1)/2 \). It transforms into equation (24) under the replacements \( \kappa \to -\kappa, V \to -V, E \to -E \), \( Q \to P \) 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 4 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. Standard test calculations (see, e.g., [9]) show that this basis satisfies suitable completeness criteria as $n \to \infty$.

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 Refs. [35,36].

As to 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 1, we compare our results obtained for the many-potential term for $Z = 20$ employing the DKB basis set (22), (23) 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. The shell model for the nuclear charge distribution has been used with the radius $R = 3.478$ fm. In the basis set calculations, the contributions with $|\kappa| \geq 10$ were obtained by an extrapolation. Adding the zero- and one-potential terms to the many-potential term yields 0.06409 a.u. for the old basis, 0.06426 a.u. for the DKB basis, and 0.06425(1) a.u. for the Coulomb-Green function calculation. This comparison clearly demonstrates a significant improvement in accuracy, if the DKB basis is employed instead of the old one. More extensive calculations employing the DKB basis will be presented in forthcoming papers.

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TABLE I. The partial-wave contributions to the many-potential term (in a.u.) for the 1s state at $Z = 20$, obtained by the basis set methods and by the Coulomb-Green function (CGF) method. The number of the basis functions: $n = 60$. The shell model for the nuclear charge distribution has been used with $R = 3.478$ fm.

| Term  | Old basis  | DKB basis  | CGF method |
|-------|------------|------------|------------|
| $|\kappa| = 1$ | 0.848691   | 0.848750   | 0.848741   |
| $|\kappa| = 2$ | 0.020618   | 0.020662   | 0.020653   |
| $|\kappa| = 3$ | 0.005302   | 0.005331   | 0.005326   |
| $|\kappa| = 4$ | 0.002121   | 0.002139   | 0.002137   |
| $|\kappa| = 5$ | 0.001050   | 0.001062   | 0.001062   |
| $|\kappa| = 6$ | 0.000590   | 0.000597   | 0.000598   |
| $|\kappa| \leq 9$ | 0.879127   | 0.879303   | 0.879288   |
| $|\kappa| \geq 10$ | 0.000587   | 0.000585   | 0.000583   |
| Tot. many-pot. | 0.87971    | 0.87989    | 0.87987(1) |