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Application of the Hylleraas-*B*-spline basis set: Static dipole polarizabilities of helium

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The Hylleraas-B-spline basis set is introduced for solving the Hamiltonian eigenvalue problem of a two-electron atomic system. We demonstrate this method by calculating the static dipole polarizabilities of the $1\,^1S$–$5\,^1S$, $2\,^3S$–$6\,^3S$ states of helium in both the length and velocity gauge, which shows a good agreement. Comparing to the traditional B-spline basis, the present approach can significantly improve the rate of convergence of energy eigenvalues. The final extrapolated values of the polarizabilities for these states are accurate to at least eight significant digits, indicating that this method can handle intermediate states, including the continuum, of the Hamiltonian sufficiently well.

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I. INTRODUCTION

Precise calculations and experimental measurements of physical properties of helium have attracted increasing interest. For instance, compared to hydrogen, the $2P$ state of helium has a much longer lifetime which can be used for a precise determination of the fine-structure constant [1–4]. Moreover, high accurate results of oscillator strengths and polarizabilities of helium can give a response of the atom to an external electric field and provide a new route to testing the theory of nonrelativistic quantum electrodynamics (NRQED) [5–7].

Being a Coulomb three-body system, the energy eigenvalue problem of helium does not have an analytic solution. To obtain a high precision approximate solution, the Rayleigh-Ritz variational method is usually adopted. The basic idea of variational calculation is to construct a convenient and efficient basis set that should contain some optimized parameters and should be nearly complete when the size of the basis set is sufficiently large. In 1929, Hylleraas made a successful attempt in calculation of the energy levels of helium by building a correlation between the two electrons explicitly in his basis set, called the Hylleraas basis set [8]; see the review [9] by Drake. Hylleraas basis sets are also capable of handling Rydberg states of helium [10]. A variant of Hylleraas-Gaussian basis can deal with low-lying energy levels of helium in strong magnetic field [11]. By explicitly using logarithmic factors and half-integer powers in the Hylleraas basis set, the variational upper bound of the ground state of helium can be calculated to 36 digits [12].

The variational method in Hylleraas coordinates mentioned above usually is not designed for a continuum state. When we want to calculate the polarizability of an atom, or calculate an energy level shift when we come to some higher-order QED corrections, all relevant continuum states should in principle be included in the calculation. For this situation, the concept of pseudostates is used; see [13]. First we put the atom in a non-penetrable box of limited size and thus the Hamiltonian spectrum is discrete. We then perform a calculation for a physical property of interest. When we increase the size of the box gradually, what we can expect is that the result we obtained should be progressively converged to the exact value of that property.

In order to utilize the pseudostates approach, we should construct a suitable basis set that is generated in a finite-size space. Among various basis sets used in atomic and molecular structure calculations, the B-spline basis set is the one that we will use [14,15]. As we know, a B-spline is highly localized in a finite-size box, which is ideal in implementing the concept of pseudostates. In fact, using B-spline basis sets, some calculations on the dipole polarizabilities of helium were performed [16,17]. Since a B-spine basis set is not capable of describing the wave-function behavior at two-electron coalescences, calculation of polarizabilities requires rather large partial-wave expansion. The results of adopting B-spline basis is usually less accurate than the Hylleraas-type basis. In order to enhance the computational power of a B-spline basis, we insert explicitly a correlation factor $r_{12} = |\mathbf{r}_1 - \mathbf{r}_2|$ in the traditional B-spline function. We call such generated basis function as a Hylleraas-B-spline; see (2) below. The Hylleraas-B-spline basis set not only inherits the virtues of the traditional B-spline but also has no numerical linear dependence problem. Because of the correlation factor appeared in the Hylleraas-B-spline basis, the rate of convergence in a partial-wave expansion should be improved greatly.

The paper is organized as follows. In Sec. II we first introduce the concept of Hylleraas-B-spline basis for the atomic system and its mathematical construction. We then present some basic formulas for calculating the dipole polarizability. Section III presents computational results of energy levels and dipole polarizabilities for the $1\,^1S$–$5\,^1S$ and $2\,^3S$–$6\,^3S$ states of helium using the Hylleraas-B-spline basis, including convergence studies against the length of partial-wave expansion and the size of the basis set. Finally, a summary is given in Sec. IV.

II. THEORETICAL METHOD

A. Hylleraas-*B*-spline

In the center-of-mass frame, after eliminating the global motion of the system, the nonrelativistic Hamiltonian of atomic

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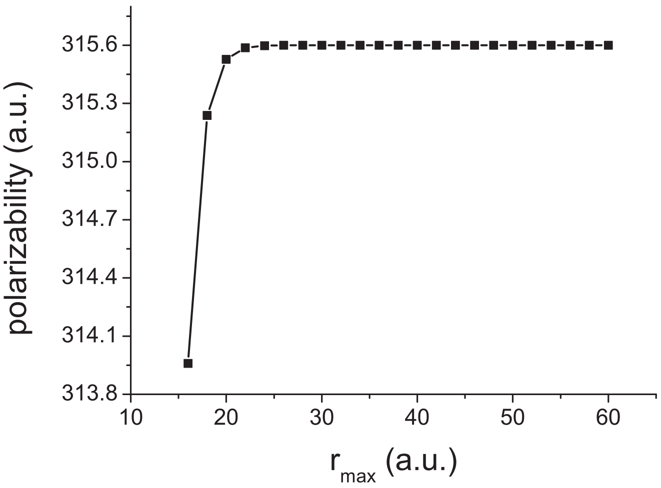

FIG. 1. Tendency of the polarizability for the 2 ^3S state of helium as the size of box $r_{\max}$ increases. Here we fix $l_{\max} = 1$ and $N = 50$. The black points represent the calculated results. Units are a.u.

helium can be written in the form

$$H = \sum_{i=1}^{2}\left(-\frac{1}{2\mu}\nabla_i^2 - \frac{2}{r_i}\right) - \frac{m_e}{M}\,\nabla_1\cdot\nabla_2 + \frac{1}{r_{12}}, \quad (1)$$

where m_e is the electron mass, M is the nuclear mass, $\mu = m_eM/(m_e + M)$ is the reduced mass between the electron and nucleus, r_1 and r_2 are the two electron distances relative to the nucleus, and r_{12} is the distance between the two electrons. We set $M = \infty$ in the present work.

The Hylleraas-B-spline variational basis set is constructed in the finite domain $(0,r_{\max})$ according to

$$\Phi^{(k)}_{ijcl_1l_2} = B_{i,k}(r_1)B_{j,k}(r_2)r_{12}^c\,\Lambda^{LM}_{l_1l_2}(\hat{\mathbf{r}}_1,\hat{\mathbf{r}}_2) \pm \text{exchange}\,, \quad (2)$$

where $\Lambda^{LM}_{l_1l_2}$ is the vector coupled product of angular momenta l_1 and l_2 for the two electrons

$$\Lambda^{LM}_{l_1l_2}(\hat{\mathbf{r}}_1,\hat{\mathbf{r}}_2) = \sum_{m_1m_2}\langle l_1l_2m_1m_2 \mid LM\rangle Y_{l_1m_1}(\hat{\mathbf{r}}_1)Y_{l_2m_2}(\hat{\mathbf{r}}_2)\,, \quad (3)$$

$B_{i,k}(r)$ is a B-spline [18] with i and k being the serial number and the spline order, respectively, and r_{12}^c is the correlation factor. The shape of $B_{i,k}(r)$ depends on the nondecreasing knot sequence $\{t_k\}$ (see below) and the spline order.

TABLE II. Convergence study of the ground-state energy of helium as the total number of B-splines N increases under fixed $l_{\max} = 4$. The number in parentheses of the extrapolated value is the computational uncertainty. Units are a.u.

N	Number of terms	$E(N)$	$R(N)$
20	2100	−2.9037176344643	
25	3250	−2.9037240799982	
30	4650	−2.9037243503318	23.842
35	6300	−2.9037243734819	11.677
40	8200	−2.9037243764457	7.8110
45	10350	−2.9037243769088	6.3989
50	12750	−2.9037243769997	5.0961
Extrap.		−2.9037243771(2)	

The B-splines satisfy the following recursion relation:

$$B_{i,k}(r) = \frac{r - t_i}{t_{i+k-1} - t_i}B_{i,k-1}(r) + \frac{t_{i+k} - r}{t_{i+k} - t_{i+1}}B_{i+1,k-1}(r), \quad (4)$$

together with the definition of the B-spline of order 1

$$\begin{cases} B_{i,1}(r){=}1, & t_i \leqslant r < t_{i+1}, \\ B_{i,1}(r){=}0, & \text{otherwise}. \end{cases} \quad (5)$$

In order to satisfy the boundary conditions of the wave functions, the knot sequence should be arranged as

$$\begin{cases} t_i = 0, & i = 1,2,\ldots,k-1, \\ t_i = r_{\max}\dfrac{e^{\gamma\left(\frac{i-k}{N-k+1}\right)} - 1}{e^{\gamma} - 1}, & i = k,k+1,\ldots,N, \\ t_i = r_{\max}, & i = N+1,\ldots,N+k-1\,, \end{cases} \quad (6)$$

where $\gamma = \tau\, r_{\max}$ with τ being an adjustable parameter to adjust the knot sequence, and N is the total number of B-splines. If $r_{\max}$, the knot sequence, the order k, and N are given, the basis set of Hylleraas-B-spline is completely constructed.

In choosing i,j,c,l_1,l_2 in (2), we restrict c to be less than 2. Thus a polynomial of $\cos\theta$ from the correlation factor will not directly present in basis functions, where θ is the angle between $\mathbf{r}_1$ and $\mathbf{r}_2$. This choice of c will not be in conflict with the freedom of choosing l_1 and l_2. We list these parameters

TABLE I. Convergence study of the ground-state energy of helium as the total number of B-splines N and the partial-wave expansion length $l_{\max}$ increase. Units are a.u.

$N \backslash l_{\max}$	1	2	3	4
20	−2.9036	−2.90371	−2.90371	−2.90371
25	−2.903723	−2.903723	−2.903723	−2.9037240
30	−2.9037241	−2.90372430	−2.90372434	−2.90372435
35	−2.90372425	−2.90372436	−2.903724371	−2.903724373
40	−2.903724265	−2.903724375	−2.903724375	−2.9037243764
45	−2.903724267	−2.9037243766	−2.9037243767	−2.90372437690
50	−2.9037242683	−2.90372437687	−2.90372437696	−2.903724376999

TABLE III. Comparison of the energies of the five lowest singlet and triplet states of helium. Numbers in parentheses of the extrapolated values are the computational uncertainties. Units are a.u.

State	This work	Ref. [21]	Ref. [22]	Ref. [19]
$1\,^1S$	−2.9037243771(2)	−2.9035774		−2.9037243770341195
$2\,^1S$	−2.14597404608(4)	−2.1459649	−2.1459740292	−2.145974046054419(6)
$3\,^1S$	−2.06127198976(3)	−2.0612681	−2.0612719720	−2.061271989740911(5)
$4\,^1S$	−2.03358671705(3)	−2.0335850	−2.0335866995	−2.03358671703072(1)
$5\,^1S$	−2.02117685159(3)	−2.021175	−2.0211768309	−2.021176851574363(5)
$2\,^3S$	−2.17522937826(4)	−2.1752288	−2.175229378176	−2.17522937823679130
$3\,^3S$	−2.068689067469(3)	−2.0686888	−2.068689067283	−2.06868906747245719
$4\,^3S$	−2.0365120831(1)	−2.0365120	−2.036512082933	−2.03651208309823630(2)
$5\,^3S$	−2.0226188723(1)	−2.0226188	−2.022618871382	−2.02261887230231227(1)
$6\,^3S$	−2.0153774530(1)		−2.015377452422	−2.01537745299286219(3)

below:

$$\begin{aligned} i &= 1,2,\ldots,j,\\ j &= 1,2,\ldots,N,\\ c &= 0,1, \qquad\qquad (7)\\ l_1 &= 0,1,\ldots,l_{\max},\\ l_2 &= 0,1,\ldots,l_{\max}. \end{aligned}$$

Since the wave function must be antisymmetric against the permutation between the two electrons, any term that makes the norm of $\Phi^{(k)}_{ijcl_1l_2}$ to be zero must be eliminated.

B. Polarizability

The averaged dipole polarizability for an atom is defined by

$$\overline{\alpha}_1 = \sum_{n\neq 0} \frac{\overline{f^{(1)}_{n0}}}{(E_n - E_0)^2}, \tag{8}$$

where $\overline{f^{(1)}_{n0}}$ is the averaged dipole oscillator strength

$$\overline{f^{(1)}_{n0}} = \frac{2}{3(2L_0+1)}(E_n - E_0)\left|\left\langle \psi_0 \left\| \sum_i \mathbf{r}_i \right\| \psi_n \right\rangle\right|^2. \tag{9}$$

In the above, the summation i runs over all the electrons in the atom, L_0, E_0, and ψ_0 are, respectively, the total angular momentum, the energy, and the corresponding wave function for the state of interest, and E_n and ψ_n are for the nth intermediate state. The above length-gauge oscillator strength can also be expressed in the velocity gauge

$$\overline{f^{(1)}_{n0}} = \frac{2}{3(2L_0+1)(E_n - E_0)}\left|\left\langle \psi_0 \left\| \sum_i \mathbf{p}_i \right\| \psi_n \right\rangle\right|^2. \tag{10}$$

Ideally, these two gauges would give rise to identical results for a dipole polarizability if all eigenvalues and eigenfunctions involved are exact. Thus the following quantity η may be used as a measure for the quality of our calculations

$$\eta = 2\frac{|a-b|}{a+b}, \tag{11}$$

where a and b stand for the results from the two gauges.

III. RESULTS AND DISCUSSIONS

Figure 1 presents the tendency of the polarizability of the $2\,^3S$ state of helium as the size of the box $r_{\max}$ increases. Here we set the partial-wave expansion length $l_{\max}$ to be 1, the number of B-splines N to be 50, and the parameter τ to be 0.038. From Fig. 1, we can see that as $r_{\max}$ reaches about 30 a.u. the curve becomes flat. In our calculation, however, we set $r_{\max} = 200$ a.u., because some of the states under consideration are quite diffuse.

A. Energy levels

Table I shows a convergence study for the helium ground-state energy as the number of B-splines N and the partial-wave expansion length $l_{\max}$ increase progressively. It can be seen that fixing $l_{\max}$ to be 4 should be sufficient to guarantee the

TABLE IV. Convergence study of the static dipole polarizability for the $2\,^3S$ state of helium in the length gauge as the total number of B-splines N and the partial-wave expansion length $l_{\max}$ increase. Units are a.u.

$N \backslash l_{\max}$	1	2	3	4
20	315.5	315.6	315.6311	315.637
25	315.601	315.6311	315.6315	315.6316
30	315.6004	315.63148	315.631475	315.63148
35	315.600336	315.63147	315.6314722	315.631473
40	315.6003313	315.631465	315.6314723	315.6314726
45	315.6003316	315.6314644	315.6314724	315.6314724
50	315.600331943	315.631464209	315.631472397	315.631472384

TABLE V. Convergence study of the static dipole polarizability for the $2\,^3S$ state of helium in the velocity gauge as the total number of B-splines N and the partial-wave expansion length $l_{\max}$ increase. Units are a.u.

$N \backslash l_{\max}$	1	2	3	4
20	315.3	315.5	315.61	315.62
25	315.5550	315.62	315.6311	315.6313
30	315.55591	315.63140	315.63143	315.63146
35	315.55590	315.631455	315.63146	315.6314723
40	315.555913	315.631452	315.631471	315.6314724
45	315.555916	315.6314508	315.6314722	315.631472364
50	315.55591865	315.631450648	315.631472353	315.63147236368

partial-wave convergence at the level of about 10 significant digits. Table II displays the results for fixed $l_{\max} = 4$, as N increases, where the ratio $R(N)$, defined by

$$R(N) = \frac{E(N-5) - E(N-10)}{E(N) - E(N-5)}, \qquad (12)$$

could be regarded as an indicator for the rate of convergence of the energy. The energy extrapolated to $N = \infty$ is estimated by assuming a constant $R(N)$ for all N greater than the largest N (denoted by $N_{\max}$) used in the calculation [19]

$$E(\infty) = E(N_{\max}) + \frac{E(N_{\max}) - E(N_{\max} - 5)}{R(N_{\max}) - 1}. \qquad (13)$$

The uncertainty is taken to be the difference between $E(\infty)$ and $E(N_{\max})$. The extrapolated value for the ground state is thus $E = -2.9037243771(2)$, which has three significant digits more precise than $E = -2.903724268$ obtained from the traditional B-spline basis [20] using $l_{\max} = 16$. In fact in the present approach, such a level of accuracy can be achieved by only using $l_{\max} = 1$.

Table III is a comparison of our results for the five lowest singlet and triplet states with other calculations. According to the benchmark values [19] in the fifth column, our results are substantially more precise than the results using the traditional B-spline basis [21] listed in the third column, and comparable to the values using the exponentially correlated Hylleraas basis [22] listed in the fourth column. This comparison shows that the current Hylleraas-B-spline basis could be considered as another effective method to calculate energy levels of high principal quantum number.

TABLE VI. Relative difference η for the static dipole polarizability of the $2\,^3S$ state of helium in both the length and velocity gauge as the total number of B-splines N and the partial-wave expansion length $l_{\max}$ increase.

$N \backslash l_{\max}$	1	2	3	4
20	6×10^{-4}	1×10^{-4}	6×10^{-5}	3×10^{-5}
25	1×10^{-4}	4×10^{-6}	1×10^{-6}	9×10^{-7}
30	1×10^{-4}	2×10^{-7}	1×10^{-7}	6×10^{-8}
35	1×10^{-4}	5×10^{-8}	1×10^{-8}	4×10^{-9}
40	1×10^{-4}	5×10^{-8}	2×10^{-9}	6×10^{-10}
45	1×10^{-4}	5×10^{-8}	6×10^{-10}	1×10^{-10}
50	1×10^{-4}	5×10^{-8}	1×10^{-10}	6×10^{-11}

B. Dipole polarizabilities

The dipole polarizability of the $2\,^3S$ state in both the length and velocity gauge is given in Tables IV and V, respectively. The relative difference η between these two gauges is given in Table VI. Table VII summarizes the results for the five lowest singlet and triplet states, together with other calculations. The procedure of extrapolation we used in Table VII is the same as in Table III.

Table VI shows that the difference of results between the two gauges becomes smaller and smaller with increasing N and $l_{\max}$. When $N = 50$ and $l_{\max} = 4$, the relative difference reaches 6×10^{-11}. On the other hand, the maximum value of η in the whole calculation is only 9×10^{-10}. Using the B-spline configuration interaction method, Zhang *et al.* [17] calculated the dipole polarizability for the $2\,^3S$ state of helium with the result 315.6315(2). In their calculations, the partial-wave expansion was terminated at $l_{\max} = 10$. In our Hylleraas-B-spline approach, however, the same level of accuracy can be reached at $l_{\max} = 2$, as can be seen from Tables IV and V.

As shown in Table VII, compared with the traditional B-spline basis calculation [23] in the fourth column, a much higher accuracy for the dipole polarizabilities has been achieved by us. From the table one can see that the excellent agreement between the length and velocity gauge at the level of at least eight significant digits has been obtained for the dipole polarizabilities. Compared to the precision results from Hylleraas basis sets [24] in the fifth column of the table, there is a good agreement for $1\,^1S$, $2\,^1S$, $3\,^1S$, $2\,^3S$, and $3\,^3S$. However, some discrepancies appear for the rest of states. Such a high degree of agreement between the two gauges probably implies that our results are likely reliable. As the principal quantum number increases, very little loss of significant digits in our results is detected. Therefore, the Hylleraas-B-spline approach holds potential for efficient and precise calculation of polarizabilities of a two-electron atomic system in a Rydberg state.

IV. SUMMARY

In this paper, we have introduced the method of Hylleraas-B-spline basis by coupling the correlation factor $r_{12} = |\mathbf{r}_1 - \mathbf{r}_2|$ with the traditional B-spline basis. This basis is capable of describing the two-electron coalescences. As we have shown in this paper, the Hylleraas-B-spline basis cannot only improve computational accuracy and efficiency for excited states of helium compared to the traditional

TABLE VII. Comparison of the static dipole polarizabilities for the five lowest singlet and triplet states of helium. Numbers in parentheses are computational uncertainties. Units are a.u.

State	This work (length gauge)	This work (velocity gauge)	Ref. [23]	Ref. [24]
$1\,^1S$	1.3831921742(3)	1.3831921744(1)	1.38328	1.38319217440(5) [26]
$2\,^1S$	800.3162331(1)	800.3162332(1)	800.306	800.31633(7)
$3\,^1S$	16887.18563(1)	16887.18564(1)		16887.17(1)
$4\,^1S$	135851.5811(1)	135851.5812(1)		135851.430(1)
$5\,^1S$	669586.064(1)	669586.0653(1)		669585.8982(2)
$2\,^3S$	315.63147233(5)	315.63147236(1)	315.630	315.63147(1)
$3\,^3S$	7937.585923(3)	7937.5859256(5)		7937.58(1)
$4\,^3S$	68650.20892(2)	68650.20897(4)		68650.061(2)
$5\,^3S$	351796.228(1)	351796.2291(1)		351796.060(2)
$6\,^3S$	1314954.976(1)	1314954.979(3)		1314954.806(3)

B-spline basis, but can also offer a feasible way to overcome the ground-state difficulty of using the traditional B-spline-type basis. Furthermore, the Hylleraas-B-spline basis is very suitable for dealing with problems involving the continuum.

We have demonstrated our method by calculating energy levels and dipole polarizabilities of helium in various S states at the level of at least 10 and eight significant digits, respectively, which would not be easy by using the traditional B-spline basis. Our results are comparable with the doubled Hylleraas basis approach, in particular for dipole polarizabilities. Our method is applicable to two-electron atomic systems in Rydberg states.

It would be interesting to apply the Hylleraas-B-spline method to calculate higher-pole polarizabilities, the Bethe logarithms of quantum electrodynamic corrections, and the blackbody radiation shifts [25] for a two-electron atomic system.

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