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A triple basis set in Hylleraas coordinates is used to obtain improved variational bounds for the nonrelativistic energy and other properties of He, H^-, and Ps^-. The accuracy, numerical stability, and computational efficiency are compared with recent work based on quasirandom basis sets. The Kato cusp conditions are used to assess the accuracy of the wave functions at short distances.

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I. INTRODUCTION

Calculations of the ground-state energies for helium, H^-, and Ps^- by variational and other means continue to provide benchmark tests of the relative accuracies and efficiencies of various methods of calculation. Traditionally, the best results have been obtained by variational calculations in Hylleraas coordinates in which the trial wave function is written in the form

$$\Psi(\mathbf{r}_1,\mathbf{r}_2)=\sum_{i,j,k}^{i+j+k\leqslant\Omega} a_{ijk} r_1^i\, r_2^j\, r_{12}^k \exp(-\alpha r_1-\beta r_2) \pm(\text{exchange}), \quad (1)$$

where $r_{12}=|\mathbf{r}_1-\mathbf{r}_2|$ is the interelectron coordinate, the a_{ijk} are linear variational parameters determined by matrix diagonalization, and α and β are nonlinear scale factors that can be separately varied to minimize the energy. The size of the basis set is typically controlled by progressively increasing the value of Ω. Calculations of increasing size and sophistication (sometimes including logarithmic terms and fractional powers) have been done by many authors since the early days of quantum mechanics [1–13], resulting in progressively lower upper bounds on the ground-state energy. The best results so far have been obtained with ''double'' basis sets in which each combination of powers $\{i,j,k\}$ is included twice with different exponential scale factors α_1, β_1 and α_2, β_2 [10].

There has recently been considerable interest in a rather different kind of trial function which can be expressed in the form [14–17]

$$\Psi(\mathbf{r}_1,\mathbf{r}_2)=\sum_{i}^{N} a_i \exp(-\alpha_i r_1-\beta_i r_2-\gamma_i r_{12}) \pm(\text{exchange}), \quad (2)$$

where $\{\alpha_i, \beta_i, \gamma_i\}$ are triplets of numbers (including complex γ_i) that are chosen in a quasirandom fashion. The result is a kind of Monte Carlo calculation with a random distribution of exponential scale factors and no powers of the radial coordinates at all. Recent refinements to the quasirandom distribution of scale factors have yielded an improved upper bound to the ground-state energy of various three-body systems [16].

The purpose of this Brief Report is to extend our previous results for double basis sets in Hylleraas coordinates [10,13] to triple basis sets. For sufficiently large basis sets, it becomes advantageous to include each combination of powers $\{i,j,k\}$ in Eq. (1) three times with three independently optimized sets of exponential scale factors. This strategy yields better convergence and lower eigenvalues for a given size of basis set than simply increasing Ω to include more powers. The result is a new lowest upper bound for the ground state of three-body systems.

The stability and computational efficiency of the method will be compared with the quasirandom method. The Kato cusp conditions will be used to test the accuracy of the wave function itself at short distances. As has recently been discussed [18], an accurate variational energy does not necessarily guarantee an accurate wave function at short distances for the calculation of relativistic and QED effects.

II. CALCULATIONS

For a triple basis set, the complete trial function becomes

$$\Psi(\mathbf{r}_1,\mathbf{r}_2)=a_0\phi_0(Z,\mathbf{r}_1)\phi_0(Z-1,\mathbf{r}_2) + \sum_{p=1}^{3}\sum_{i,j,k}^{i+j+k\leqslant\Omega} a_{ijk}^{(p)} r_1^i\, r_2^j\, r_{12}^k \times \exp(-\alpha^{(p)} r_1-\beta^{(p)} r_2)\pm(\text{exchange}), \quad (3)$$

where the sum over p covers the three sets of nonlinear parameters $\alpha^{(1)}$, $\beta^{(1)}$, $\alpha^{(2)}$, $\beta^{(2)}$, and $\alpha^{(3)}$, $\beta^{(3)}$ for the asymptotic, intermediate, and short-range sectors, respectively. The screened hydrogenic term for nuclear charge Z is included for completeness on the right-hand side since it is important for Rydberg states. However, it makes little difference for the ground state of helium, and it must be omitted for the negative ions where it is not defined.

If all terms with $i+j+k\leqslant\Omega$ were included in each sector, then the number of terms per sector would be $(\Omega+1)(\Omega+2)(\Omega+3)/6$. However, since the optimized $\alpha^{(p)}$, $\beta^{(p)}$ pairs are nearly equal, terms with $i<j$ can and should be omitted in order to preserve numerical stability. The number of terms in the p th sector having $\Omega=\Omega_p$ is then the integer closest to

$$N(\Omega_p)=\tfrac{1}{12}(\Omega_p+1)(\Omega_p+\tfrac{7}{2})(\Omega_p+3) \quad (4)$$

TABLE I. Convergence study for the nonrelativistic energy of the ground state of $^{\infty}$He (infinite nuclear mass). Ω is the highest power in the triple basis set, and N is the total number of terms. Units are atomic units.

Ω	N	$E(\Omega)$	Ratio[a]
8	269	−2.903 724 377 029 560 058 400	
9	347	−2.903 724 377 033 543 320 480	
10	443	−2.903 724 377 034 047 783 838	7.90
11	549	−2.903 724 377 034 104 634 696	8.87
12	676	−2.903 724 377 034 116 928 328	4.62
13	814	−2.903 724 377 034 119 224 401	5.35
14	976	−2.903 724 377 034 119 539 797	7.28
15	1150	−2.903 724 377 034 119 585 888	6.84
16	1351	−2.903 724 377 034 119 596 137	4.50
17	1565	−2.903 724 377 034 119 597 856	5.96
18	1809	−2.903 724 377 034 119 598 206	4.90
19	2067	−2.903 724 377 034 119 598 286	4.44
20	2358	−2.903 724 377 034 119 598 305	4.02
Extrap.		−2.903 724 377 034 119 598 311(1)	
[b]	2200	−2.903 724 377 034 119 598 296	
[c]		−2.903 724 377 034 119 598 306(10)	
[d]	8066	−2.903 724 377 034 119 593 82	
[e]	24497	−2.903 724 377 034 119 589(5)	
[f]	476	−2.903 724 377 034 118 4	

[a]Ratio is the ratio of successive differences $[E(\Omega-1)-E(\Omega-2)]/[E(\Omega)-E(\Omega-1)]$.
[b]Korobov variational bound [16].
[c]Korobov extrapolation [16].
[d]Goldman variational bound [12].
[e]Bürgers *et al.* variational bound [11].
[f]Baker *et al.* variational bound [7].

for both even and odd Ω_p. In addition, we employed a form of truncation first suggested by Kono and Hattori [19] in which terms with $i+j+k+|i-j|>\Omega_p$ and $k\geqslant\overline{k}$ are omitted in sectors 2 and 3 (the intermediate and short-range sectors). This is not an absolute truncation—the omitted terms eventually reappear as Ω increases. With the definition $\overline{\Omega}_p=\Omega_p-\overline{k}$, the number of terms omitted for a given $\overline{\Omega}_p$ is

$$\overline{N}(\overline{\Omega}_p)=\begin{cases}\frac{1}{24}\overline{\Omega}_p(\overline{\Omega}_p+2)(\overline{\Omega}_p+4) & \text{for } \overline{\Omega}_p \text{ even}\\ \frac{1}{24}(\overline{\Omega}_p+1)(\overline{\Omega}_p+2)(\overline{\Omega}_p+3) & \text{for } \overline{\Omega}_p \text{ odd.}\end{cases} \tag{5}$$

For helium, we set $\Omega_1=\Omega_2=\Omega_3=\Omega$ and, after some experimentation, found that one can set $\overline{k}=4$ without significantly affecting the convergence. The total number of terms for the triple basis set is then $N_{\rm tot}=3N(\Omega)-2\overline{N}(\Omega-4)$. For H^- and Ps^- we found that relatively few terms are needed in sector 3, but that $\overline{k}$ should be increased to 7. We therefore set $\Omega_1=\Omega_2=\Omega$, $\Omega_3=\Omega-8$, and $\overline{k}=7$. The total number of terms is then $2N(\Omega)+N(\Omega-8)-\overline{N}(\Omega-7)-\overline{N}(\Omega-15)$.

Having constructed the basis set, the principal computational step is to solve the generalized eigenvalue problem $\mathbf{H}-E\mathbf{O}=0$, where $\mathbf{H}$ is the Hamiltonian matrix and $\mathbf{O}$ is the overlap matrix. The matrix elements of $\mathbf{H}$ can be easily calculated from the explicitly Hermitian form given by Eq. (11.33) of Ref. [20]. The optimization of the $\alpha^{(p)}$, $\beta^{(p)}$ is efficiently accomplished by simultaneously calculating the energy derivatives

$$\frac{\partial E}{\partial\alpha^{(p)}}=-2\frac{\langle\Psi|(H-E)r_1|\Psi^{(p)}\rangle}{\langle\Psi|\Psi\rangle}, \tag{6}$$

where $\Psi^{(p)}$ denotes the part of the wave function that depends explicitly on $\alpha^{(p)}$, and similarly for the $\beta^{(p)}$ derivative. There is no contribution from terms arising from $\partial a_{ijk}^{(q)}/\partial\alpha^{(p)}$ or $\partial a_{ijk}^{(q)}/\partial\beta^{(p)}$ because of the variational stability of the wave function. The final step is then to change the $\alpha^{(p)}$s and $\beta^{(p)}$s in the directions indicated by the derivatives, resolve the generalized eigenvalue problem, recalculate the derivatives, and locate their zeros by Newton's method.

All calculations were done in quadruple precision (about 32 decimal digit) arithmetic on a Compaq alpha workstation. For the largest basis sets (about 2300 terms), a complete iteration, incuding the calculation of derivatives, takes about 1 h. Since good starting values for the $\alpha^{(p)}$ and $\beta^{(p)}$ are always available from previous calculations with smaller basis sets, only a few iterations are required.

III. RESULTS

Tables I, II, and III show the convergence pattern for the ground states of $^{\infty}$He, $^{\infty}H^-$, and Ps^-, and comparisons with other calculations. The numbers in the last column of each table give the values of the ratios of successive differences defined by

$$R(\Omega)=\frac{E(\Omega-1)-E(\Omega-2)}{E(\Omega)-E(\Omega-1)}. \tag{7}$$

If $R(\Omega)$ were a constant, then the series would be a geometric series with the limit

$$E(\infty)=E(\Omega)+\frac{E(\Omega)-E(\Omega-1)}{R-1}. \tag{8}$$

Since the actual values of $R(\Omega)$ show some scatter and tend to decrease with Ω, we fit them to the functional form a/Ω^b and sum the series of differences numerically to obtain the extrapolated value. The uncertainty is derived from the uncertainty in the values of the fitting parameters a and b. In each of the three cases, the largest basis set gives the lowest upper bound obtained so far, and the extrapolated result is more accurate than Korobov's result [16] by about an order of magnitude. However, it is satisfying that all the results agree to within their estimated accuracies, even though they were obtained with quite different strategies for the construction of basis sets.

The complete wave functions can be immediately regenerated from the values of the optimized scale factors $\alpha^{(p)}$ and $\beta^{(p)}$ listed in Table IV for $^{\infty}$He and Ps^-. The optimization produces a natural partition of the basis set into three distinct

TABLE II. Convergence study for the nonrelativistic energy of $^{\infty}H^{-}$ (infinite nuclear mass). Units are atomic units.

Ω	N	$E(\Omega)$	Ratio
10	324	−0.527 751 016 537 120 298 160	
11	411	−0.527 751 016 543 123 297 506	
12	512	−0.527 751 016 544 190 011 531	5.63
13	630	−0.527 751 016 544 351 706 935	6.60
14	764	−0.527 751 016 544 373 661 892	7.36
15	918	−0.527 751 016 544 376 556 281	7.59
16	1089	−0.527 751 016 544 377 083 777	5.49
17	1283	−0.527 751 016 544 377 173 607	5.87
18	1495	−0.527 751 016 544 377 191 103	5.13
19	1733	−0.527 751 016 544 377 195 175	4.30
20	1990	−0.527 751 016 544 377 196 198	3.98
21	2276	−0.527 751 016 544 377 196 503	3.34
Extrap.		−0.527 751 016 544 377 196 613(22)	

distance scales with $\alpha^{(1)}$, $\beta^{(1)}$ describing the asymptotic behavior of the wave function, $\alpha^{(2)}$, $\beta^{(2)}$ the intermediate range behavior, and $\alpha^{(3)}$, $\beta^{(3)}$ the short-range behavior. Note that the latter two continue increasing approximately linearly with Ω such that the function $r^{\Omega}\exp[-\alpha(\Omega)r]$ peaks at about the same distance $r=\Omega/\alpha(\Omega)$, independent of Ω. These sets of functions can be thought of as spreading inwards to describe complex correlation effects at progressively shorter distance scales. This linear increase with Ω is essential in order to avoid problems with numerical linear dependence in the basis set. Provided that this precaution is observed, the method has good numerical stability with standard quadruple precision arithmetic. In comparison, the quasirandom method used by Korobov [16] and Frolov [21] required up to 60-figure extended precision arithmetic in order to maintain numerical stability.

TABLE III. Convergence study for the nonrelativistic energy of Ps^{-}, with mass polarization (specific mass shift) included. Units are atomic units.

Ω	N	$E(\Omega)$	Ratio
10	324	−0.262 005 070 206 699 500 141	
11	411	−0.262 005 070 227 775 783 440	
12	512	−0.262 005 070 232 069 520 272	4.91
13	630	−0.262 005 070 232 832 810 937	5.63
14	764	−0.262 005 070 232 959 187 559	6.04
15	918	−0.262 005 070 232 977 195 563	7.02
16	1089	−0.262 005 070 232 979 519 328	7.75
17	1283	−0.262 005 070 232 980 001 224	4.82
18	1495	−0.262 005 070 232 980 080 426	6.08
19	1733	−0.262 005 070 232 980 101 597	3.74
20	1990	−0.262 005 070 232 980 106 481	4.33
21	2276	−0.262 005 070 232 980 107 412	5.25
22	2528	−0.262 005 070 232 980 107 627	4.33
Extrap.		−0.262 005 070 232 980 107 696(12)	
Korobov [16]		−0.262 005 070 232 980 107 4	
Extrap. [16]		−0.262 005 070 232 980 107 7(3)	

TABLE IV. Optimized scale factors for $^{\infty}$He and Ps^{-}. Units are $\mu Z/(ma_0)$, where a_0 is the Bohr radius, Z is the nuclear charge, and μ is the reduced electron mass.

Ω	$\alpha^{(1)}$	$\beta^{(1)}$	$\alpha^{(2)}$	$\beta^{(2)}$	$\alpha^{(3)}$	$\beta^{(3)}$
			$^{\infty}$He			
8	1.217 77	1.200 01	1.850 16	1.979 43	4.252 38	4.312 99
9	1.236 88	1.195 68	2.182 50	1.996 95	6.101 87	5.002 38
10	1.239 62	1.218 20	2.446 11	2.137 63	5.865 17	5.932 25
11	1.275 02	1.227 72	2.610 05	2.267 21	6.407 23	6.437 19
12	1.292 48	1.239 26	2.753 48	2.455 20	6.890 81	6.894 04
13	1.312 07	1.237 79	3.065 98	2.573 49	9.144 84	9.166 08
14	1.326 60	1.285 16	2.883 97	3.171 26	11.193 73	11.806 03
15	1.344 79	1.288 21	2.961 36	3.415 83	12.262 94	12.341 19
16	1.363 22	1.289 98	3.104 55	3.797 91	14.283 26	15.384 64
17	1.382 93	1.300 11	3.309 33	4.076 78	17.851 99	18.233 89
18	1.385 38	1.299 74	3.456 05	4.364 01	19.090 64	21.023 99
19	1.400 27	1.299 38	3.865 36	4.671 14	22.556 58	24.475 34
20	1.464 90	1.328 37	3.996 03	4.773 19	25.820 43	26.855 77
			Ps^{-}			
10	0.980 16	0.585 21	1.801 09	1.825 99	7.329 04	8.990 72
11	0.997 19	0.589 42	1.810 85	2.070 86	6.306 88	8.208 07
12	1.024 35	0.603 03	1.884 22	2.074 95	7.436 22	5.615 36
13	1.037 41	0.604 80	1.963 75	2.263 43	8.901 18	8.781 13
14	1.035 58	0.607 73	2.210 21	2.295 53	9.506 35	9.430 18
15	1.055 91	0.610 78	2.235 47	2.367 37	11.442 20	11.435 18
16	1.067 87	0.612 37	2.455 20	2.317 69	11.564 88	11.550 54
17	1.098 08	0.619 87	2.482 91	2.352 36	11.466 37	11.700 68
18	1.113 71	0.630 00	2.565 25	2.498 72	13.624 88	14.222 84
19	1.164 67	0.643 19	2.807 25	2.611 76	15.991 27	15.816 77
20	1.157 96	0.640 01	2.807 31	2.713 50	15.779 60	16.359 92
21	1.184 57	0.656 01	2.681 27	2.694 34	16.530 15	17.160 83
22	1.192 26	0.663 33	2.861 82	2.879 15	17.552 19	18.039 55

The Kato cusp conditions provide a useful test of the accuracy of the variational wave function near the electron-electron and electron-nucleus coalescence points. With the definition

$$\nu_{ij}=\frac{\left\langle \delta(\mathbf{r}_{ij})(\partial/\partial r_{ij})\right\rangle}{\langle \delta(\mathbf{r}_{ij})\rangle} \tag{9}$$

the exact cusp values are [22]

$$\nu_{ij}^{(0)}=q_i q_j \frac{m_i m_j}{m_i+m_j}, \tag{10}$$

where q_i and q_j are the charges and m_i and m_j the masses of the particles. The quantity $C_{ij}=1-\nu_{ij}/\nu_{ij}^{(0)}$ then measures the relative departure of the calculated value from the exact value. The results for the electron-nucleus cusp are $C_{\mathrm{en}}=0.1$, 0.9, and 2 ppb (parts per billion) for He, H^{-}, and Ps^{-}, respectively. For the electron-electron cusp, the values are $C_{\mathrm{ee}}=8$, 100, and 200 ppb, respectively. The values for Ps^{-} are about a factor of 100 smaller than those reported by Frolov [23]. They indicate that the present wave functions

TABLE V. Expectation values of various operators for Ps^- in atomic units.

Operator	Expectation value
$\langle 1/r_1^2 \rangle$	0.279 326 542 225 011(6)
$\langle 1/r_1 \rangle$	0.339 821 023 059 220 350(25)
$\langle r_1 \rangle$	5.489 633 252 359 448 7(27)
$\langle r_1^2 \rangle$	48.418 937 226 238 5(5)
$\langle 1/r_{12}^2 \rangle$	0.036 022 058 454 577(10)
$\langle 1/r_{12} \rangle$	0.155 631 905 652 480 400(25)
$\langle r_{12} \rangle$	8.548 580 655 099 182 7(7)
$\langle r_{12}^2 \rangle$	93.178 633 847 981 1(4)
$\langle \mathbf{r}_1 \cdot \mathbf{r}_2 \rangle$	1.829 620 302 247 297(8)
$\langle 1/r_1 r_2 \rangle$	0.060 697 690 288 582 15(5)
$\langle 1/r_1 r_{12} \rangle$	0.090 935 346 529 989 425(13)
$\langle \delta(r_1) \rangle$	0.020 733 198 005 165(13)
$\langle \delta(r_{12}) \rangle$	0.000 170 996 756 79(10)

are well suited to the calculation of expectation values for the highly singular operators appearing in relativistic and QED corrections.

Finally, the expectation values of various operators are listed in Table V for Ps^-, including their uncertainties obtained from the convergence pattern with increasing Ω. In several cases, the last one or two figures quoted by Frolov [23] are in disagreement with the present results. However, no uncertainty estimates are given by Frolov. A similar table of high precision expectation values for $^\infty$He and $^\infty H^-$ has been published previously [20].

IV. DISCUSSION

The results presented here demonstrate that a triple basis set in Hylleraas coordinates is capable of exceeding the accuracy of recent calculations for three-body systems based on quasirandom Monte Carlo methods, while using basis sets of about the same size. The excellent numerical stability resulting from the use of multiple basis sets obviates the need for extended precision arithmetic beyond standard quadruple precision, at least up to current levels of accuracy. In addition to its numerical stability, the current method is computationally much more efficient than the ''booster'' form of the quasirandom method recently employed by Frolov [17], and it provides a well-defined convergence pattern that can be used to assess the accuracy of the results. In addition, the small table of optimized scale factors in Table IV provides sufficient information to regenerate the entire sequence of progressively larger wave functions.

The 22-figure accuracy of the present nonrelativistic eigenvalues of course goes well beyond the accuracy warranted by the uncertainty in the Rydberg constant itself. However, this extraordinary accuracy is a consequence of the variational stability of the energy eigenvalue. As illustrated by the Kato cusp conditions, other quantities are typically accurate to less than half as many significant figures, and it is for the determination of these and other quantities related to relativistic and QED effects that the present results are physically important.

Note added in proof. We have recently learned of two new calcuations for helium by V.I. Korobov (unpublished) and J.H. Sims and S.A. Hagstrom (unpublished). The former obtains an improved variational bound by extending the work in [16] to larger basis sets.

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