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Uniform electron gases. III. Low-density gases on three-dimensional spheres

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By combining variational Monte Carlo (VMC) and complete-basis-set limit Hartree-Fock (HF) calculations, we have obtained near-exact correlation energies for low-density same-spin electrons on a three-dimensional sphere (3-sphere), i.e., the surface of a four-dimensional ball. In the VMC calculations, we compare the efficacies of two types of one-electron basis functions for these strongly correlated systems and analyze the energy convergence with respect to the quality of the Jastrow factor. The HF calculations employ spherical Gaussian functions (SGFs) which are the curved-space analogs of Cartesian Gaussian functions. At low densities, the electrons become relatively localized into Wigner crystals, and the natural SGF centers are found by solving the Thomson problem (i.e., the minimum-energy arrangement of n point charges) on the 3-sphere for various values of n. We have found 11 special values of n whose Thomson sites are equivalent. Three of these are the vertices of four-dimensional Platonic solids — the hyper-tetrahedron ($n = 5$), the hyper-octahedron ($n = 8$), and the 24-cell ($n = 24$) — and a fourth is a highly symmetric structure ($n = 13$) which has not previously been reported. By calculating the harmonic frequencies of the electrons around their equilibrium positions, we also find the first-order vibrational corrections to the Thomson energy. © *2015 AIP Publishing LLC.* [http://dx.doi.org/10.1063/1.4929353]

I. INTRODUCTION

In a recent series of papers,[1–8] we have shown that the behavior of electrons in the (flat) Euclidean space $\mathbb{R}^D$ is surprisingly similar to the behavior in the (curved) D-dimensional manifold $\mathbb{S}^D$, the surface of a $(D + 1)$-dimensional ball.[9] By exploiting this similarity between electrons on a line[10,11] and electrons on a ring,[6–8,12] we have constructed a new type of correlation functional (the generalized local-density approximation) for density functional theory (DFT) calculations and we have shown that this new functional yields accurate correlation energies in a variety of one-dimensional systems.[7,8]

However, our ultimate goal is to construct improved functionals[13] for three-dimensional (3D) systems and, to this end, we seek accurate correlation energies from the spherical space $\mathbb{S}^3$ (henceforth called a glome), which is the surface of a four-dimensional (4D) ball. Electrons in $\mathbb{R}^3$ or $\mathbb{S}^3$ enjoy three degrees of translational freedom but the properties of n electrons on a glome (n-glomium) have hitherto received little attention.[4,14–17]

An n-glomium is defined by the number n of electrons and the glome radius R. Its electron density is

$$\rho = n/(2\pi^2 R^3) \tag{1}$$

but it is often measured via the Wigner-Seitz radius[18]

$$r_s = \left(\frac{3\pi}{2n}\right)^{1/3} R, \tag{2}$$

which measures the average distance between neighbouring electrons. High-density systems (which are weakly correlated) have small r_s values while low-density systems (which are strongly correlated) have large r_s values. In the present study, we focus our attention on low-density glomiums with $2 \leq n \leq 48$.

In the low-density regime, the Coulomb energy (which decays as R^{-2}) dominates over the kinetic energy (which decays as R^{-1}) and the n electrons localize onto particular points on the glome that minimize their (classical) Coulomb repulsion. These minimum-energy configurations are called Wigner crystals[19] and, if all of its sites are topologically equivalent, we will call it a uniform lattice.

The work is organized as follows. In Sec. II, we study the Thomson problem on a glome and discuss the uniform solutions. In Sec. III, we calculate the harmonic vibrational energy of the electrons as they oscillate around their lattice positions. In Secs. IV and V, respectively, we report Hartree-Fock (HF), near-exact and correlation energies of n-glomium at various densities. Unless otherwise stated, all energies are reduced (i.e., per electron). Atomic units are used throughout.

II. THE THOMSON PROBLEM

What arrangement of n unit point charges on a unit D-sphere minimizes their classical Coulomb energy? This generalizes a question posed by Thomson as he devised the "plum pudding" model of atomic structure.[20] Although the model was abandoned long ago, the Thomson problem continues to intrigue mathematicians and has resurfaced in many fields of science: surface ordering of liquid metal drops confined in Paul

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traps,[21] fullerenes-like molecules,[22] arrangements of protein subunits on spherical viruses,[23] or multielectron bubbles in liquid helium.[24,25]

The Thomson problem on a 1-sphere (i.e., a ring) is trivial and the solutions consist of charges uniformly spaced around the ring. The problem on a 2-sphere is challenging and, although it has been studied numerically up to large values of n,[26–28] mathematically rigorous solutions[29] have been established only for $n \in \{2, 3, 4, 5, 6, 12\}$.

The Thomson problem on a 3-sphere (i.e., a glome) seeks the global minimum $\varepsilon_0 = V(\mathbf{x}_0)$ of

$$V(\mathbf{x}) = \sum_{i<j}^{n} r_{ij}^{-1}, \tag{3}$$

where $\mathbf{x}$ describes the positions of the n charges on the glome and r_{ij} is the Euclidean distance between charges i and j, measured through the unit glome. It has attracted much less attention[30,31] than the $D = 2$ problem.

We performed a numerical study to determine the values of $n \leq 50$ for which the Thomson minimum-energy configuration on a glome is uniform. Although it would be more suitable to use a global optimization, because we consider relatively small numbers of electrons, we adopt the following computational strategy: for each n, we generated randomly at least 1000 distinct initial structures and minimized their energy using local optimization algorithms, as implemented in the MATHEMATICA software package.[32] Our numerical experiments indicate that there are eleven uniform lattices. Their energy ε_0 and principal moments of inertia are listed in Table I and their Cartesian coordinates are in the supplementary material.[33] The set $\{2, 3, 4, 6, 8, 12, 24\}$ of n values for uniform $D = 2$ Thomson lattices[17] is a subset of the $D = 3$ set but we do not understand the reasons for this.

The principal moments of inertia of a lattice indicate the degree of its symmetry. Generalizing the standard notation for 3D structures,[34] we define a "hyperspherical top" as a 4D structure with four equal moments; the extremely symmetrical $n = 5, 6, 8, 10, 13, 24$, and 48 lattices are of this type. We define

a "spherical top" as a lattice in which three of the four moments are equal; the $n = 2$ (prolate) and $n = 4$ (oblate) lattices are of this type. We define a "symmetric top" as a lattice in which the moments form two pairs; the $n = 3$ and $n = 12$ lattices are of this type.

The glome lattices for $n = 2$ (a diameter) and $n = 3$ (equilateral triangle) are the same as on a 1-sphere and 2-sphere. The glome lattice for $n = 4$ (regular tetrahedron) is the same as on a 2-sphere. The $n = 5$ lattice is a regular hyper-tetrahedron (also called a regular simplex[35]), a 4D Platonic solid with ten equal side lengths. The $n = 6$ lattice is the union of an equilateral triangle in the wx-plane and another such triangle in the yz-plane. The $n = 8$ lattice is a hyper-octahedron (or 16-cell), a 4D Platonic solid with vertices at ± 1 on each of the four Cartesian axes. The $n = 10$ lattice is the union of a regular pentagon in the wx-plane and another such pentagon in the yz-plane, while the $n = 12$ lattice is the union of two perpendicular triangular prisms. The $n = 13$ lattice is peculiar to the $D = 3$ Thomson problem and, to the best of our knowledge, has not been previously described. The $n = 24$ lattice is the 24-cell, a 4D Platonic solid with no analogue in 3D. It is the union of a hyper-octahedron and a hyper-cube. The $n = 48$ lattice is peculiar to the $D = 3$ Thomson problem.

III. HARMONIC VIBRATIONAL ENERGY

The energy E_W of an n-electron Wigner crystal on a glome can be estimated by solving the Schrödinger equation in the harmonic potential

$$V_2(\mathbf{x}) = V(\mathbf{x}_0) + \frac{1}{2}(\mathbf{x} - \mathbf{x}_0)^T \cdot \mathbf{H} \cdot (\mathbf{x} - \mathbf{x}_0), \tag{4}$$

where $\mathbf{H}$ is the $3n \times 3n$ second-derivative (Hessian) matrix

$$H_{ij} = \left. \frac{\partial^2 V(\mathbf{x})}{\partial t_i \partial t_j} \right|_{\mathbf{x}=\mathbf{x}_0} \tag{5}$$

and the t_i are suitable tangential coordinates. The square roots of the Hessian eigenvalues are the harmonic frequencies ω_i and

TABLE I. Non-reduced Thomson energy ε_0 and principal moments of inertia I_k of the Thomson lattices on a unit glome.

n	ε_0	I_1	I_2	I_3	I_4
2	$1/2$	0	2	2	2
3	$3/\sqrt{3}$	$3/2$	$3/2$	3	3
4	$6/\sqrt{8/3}$	$8/3$	$8/3$	$8/3$	4
5	$10/\sqrt{5/2}$	$15/4$	$15/4$	$15/4$	$15/4$
6	$9/\sqrt{2} + 6/\sqrt{3}$	$9/2$	$9/2$	$9/2$	$9/2$
8	$24/\sqrt{2} + 4/2$	6	6	6	6
10	$\dfrac{10}{2\sin(\pi/5)} + \dfrac{25}{\sqrt{2}} + \dfrac{10}{2\sin(2\pi/5)}$	$15/2$	$15/2$	$15/2$	$15/2$
12[a]	$\dfrac{6}{2c} + \dfrac{12}{\sqrt{3}s} + \dfrac{12}{\sqrt{2+2s^2}} + \dfrac{24}{\sqrt{2-s^2}} + \dfrac{12}{\sqrt{4-s^2}}$	$6(1+c^2)$	$6(1+c^2)$	$6(1+s^2)$	$6(1+s^2)$
13	$\displaystyle\sum_{k=1,2,4} \dfrac{26}{\sqrt{2-2\cos(k\pi/13)\cos(5k\pi/13)}}$	$39/4$	$39/4$	$39/4$	$39/4$
24	$96/1 + 72/\sqrt{2} + 96/\sqrt{3} + 12/2$	18	18	18	18
48	$\dfrac{24}{2} + \dfrac{240}{\sqrt{2}} + \dfrac{48}{\sqrt{2\pm\sqrt{2}}} + \dfrac{96}{\sqrt{2\pm\sqrt{2}/2}} + \dfrac{96}{\sqrt{2\pm\sqrt{6}/2}} + \dfrac{96}{\sqrt{2\pm(\sqrt{3}-1)/2}} + \dfrac{96}{\sqrt{2\pm(\sqrt{3}+1)/2}}$	36	36	36	36

[a] $c = \cos\theta$, $s = \sin\theta$ and $\theta = 0.793\,553\,668\,5\ldots$.

TABLE II. Reduced Thomson energy E_0, harmonic vibrational energy E_1, near-exact energy E, Hartree-Fock energy E_{HF}, and correlation energy E_c (in mE_h) for various n and r_s.

	r_s	20	50	100	150
	E_0	16.633	6.653	3.327	2.218
	E_0+E_1	23.068	8.281	3.902	2.531
$n=2$	E	23.928	8.385	3.924	2.540
	E_{HF}	24.911	8.795	4.100	2.643
	$-E_c$	0.983	0.410	0.176	0.103
	E_0	33.557	13.423	6.711	4.474
	E_0+E_1	41.074	15.324	7.384	4.840
$n=3$	E	41.783	15.405	7.400	4.847
	E_{HF}	43.811	16.079	7.664	5.000
	$-E_c$	2.028	0.674	0.264	0.153
	E_0	48.507	19.403	9.701	6.468
	E_0+E_1	57.190	21.600	10.478	6.890
$n=4$	E	57.155	21.550	10.461	6.882
	E_{HF}	59.886	22.358	10.757	7.046
	$-E_c$	2.731	0.808	0.296	0.164
	E_0	62.009	24.804	12.402	8.268
	E_0+E_1	71.916	27.310	13.288	8.750
$n=5$	E	71.038	27.119	13.238	8.728
	E_{HF}	74.240	27.985	13.537	8.888
	$-E_c$	3.202	0.866	0.299	0.160
	E_0	75.564	30.226	15.113	10.075
	E_0+E_1	85.823	32.821	16.030	10.575
$n=6$	E	85.406	32.682	15.986	10.553
	E_{HF}	88.283	33.530	16.293	10.720
	$-E_c$	2.877	0.848	0.307	0.167
	E_0	99.390	39.756	19.878	13.252
	E_0+E_1	110.951	42.681	20.912	13.815
$n=8$	E	110.614	42.620	20.915	13.823
	E_{HF}	112.893	43.234	21.115	13.927
	$-E_c$	2.279	0.614	0.200	0.104
	E_0	122.336	48.934	24.467	16.311
	E_0+E_1	133.974	51.879	25.508	16.878
$n=10$	E	133.522	51.767	25.485	16.874
	E_{HF}	136.154	52.479	25.727	17.002
	$-E_c$	2.632	0.712	0.242	0.128
	E_0	143.339	57.336	28.668	19.112
	E_0+E_1	155.615	60.441	29.766	19.710
$n=12$	E	154.713	60.269	29.740	19.707
	E_{HF}	157.560	61.001	29.970	19.822
	$-E_c$	2.847	0.732	0.230	0.115
	E_0	153.600	61.440	30.720	20.480
	E_0+E_1	165.909	64.554	31.821	21.079
$n=13$	E	164.804	64.322	31.767	21.061
	E_{HF}	167.947	65.126	32.033	21.196
	$-E_c$	3.143	0.804	0.266	0.135
	E_0	252.272	100.909	50.454	33.636
	E_0+E_1	265.600	104.280	51.647	34.285
$n=24$	E	264.917	104.138	51.624	34.280
	E_{HF}	267.464	104.793	51.831	34.387
	$-E_c$	2.547	0.655	0.207	0.107

TABLE II. (*Continued.*)

	r_s	20	50	100	150
	E_0	425.792	170.317	85.158	56.772
	E_0+E_1	439.690	173.833	86.401	57.449
$n=48$	E	438.667	173.681	86.406	57.474
	E_{HF}	441.542	174.334	86.581	57.547
	$-E_c$	2.875	0.653	0.175	0.073
Jellium	$-E_c$	6.839	3.607	2.080	1.499

one can then write

$$E_W = \frac{\varepsilon_0}{R} + \frac{\sum_{i=1}^{N_{vib}} \omega_i}{2R^{3/2}} + O(R^{-2}) \tag{6}$$

$$= E_0 + E_1 + O(R^{-2}). \tag{7}$$

One finds that, for $n > 3$ particles in $\mathbb{S}^3$, exactly $3n-6$ of the Hessian eigenvalues are non-zero and six vanish because they correspond to rotations on the glome. This is analogous to the familiar $3n-6$ rule[36] for non-linear molecules vibrating in $\mathbb{R}^3$. Numerical values of $E_0 + E_1$ for a range of n and r_s are presented in Table II.

IV. HARTREE-FOCK ENERGIES

We now turn to the *ab initio* treatment of n spin-up electrons on a glome, i.e., ferromagnetic n-glomium. We have performed HF calculations[37] in a basis of s-type spherical Gaussian functions (SGFs),[17]

$$G_\alpha^A(r) = \sqrt{\frac{\alpha}{2\pi^2 I_1(2\alpha)}} \exp(\alpha \, r \cdot A), \quad r \in \mathbb{S}^3, \tag{8}$$

where $A \in \mathbb{S}^3$ is the center of the SGF, α is the exponent, and I_1 is a modified Bessel function.[38] A SGF behaves like a Gaussian near A and is therefore a natural basis function for describing a localized electron. Moreover, the product of two SGFs is a third SGF which make them computationally attractive.[39] All the required one- and two-electron integrals can be found in Ref. 17.

Adopting the "Gaussian lobe" philosophy introduced by Whitten[40] many years ago, we use off-center s-type SGFs to mimic SGFs of higher angular momentum. The basis set consists of a grid of s-type SGFs with same exponent α clustered around each Thomson site (see Fig. 1). The complete basis set (CBS) HF energy is obtained by successively adding Level 0 (L0), Level 1 (L1), Level 2 (L2), and Level 3 (L3) functions to the basis set. In each calculation, we optimize the SGF exponent α and the nearest-neighbor distance δ using the Newton-Raphson optimization procedure. Our target accuracy was 1 microhartree (μE_h) per electron. The resulting HF energies for a range of n and r_s are shown in Table II.

Table III reports HF energies of n-glomium as the basis set is gradually improved. SGFs are optimal for localized electronic systems but become less efficient as the density increases so the convergence for $r_s > 20$ is always at least as fast as for $r_s = 20$. We therefore show results for $r_s = 20$, the most challenging case.
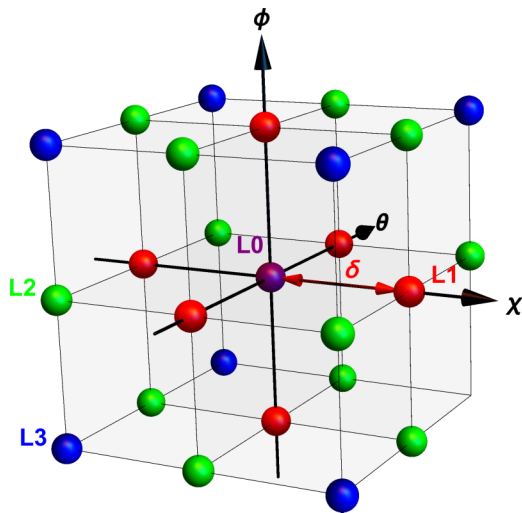
FIG. 1. Three-dimensional SGF grid used at each Thomson site in HF calculations. The Level 0 (L0), Level 1 (L1), Level 2 (L2), and Level 3 (L3) functions are represented in purple, red, green, and blue, respectively. δ is the shortest distance (around the glome) between two grid points.

For a given value of r_s, the minimal-basis (L0) exponent α grows, i.e., the electrons become more localized, as n increases. The results of Table III show that L2 achieves μE_h accuracy for all n values and, indeed, L1 suffices for the largest n values. It is well known that, on a 2-sphere, the number of nearest neighbors around an electron approaches six (hexagonal lattice) for large n.[41] Similarly, on a glome, the number of nearest neighbors approaches eight (body-centered cubic lattice).[18] Thus, for large n, the density around each electron becomes approximately isotropic, and the L2 and L3 functions become largely superfluous.

V. NEAR-EXACT ENERGIES

To obtain near-exact energies for electrons on a glome, we have performed variational Monte Carlo (VMC) calculations.[42] The trial wave function is of the form

$$\Psi_T = \Psi_0 \, e^J, \qquad (9)$$

where Ψ_0 is a Slater determinant of either SGFs[17] or hyperspherical harmonics[43,44] (HSHs),

$$Y_{k\ell m}(\chi, \theta, \phi) = C_{k-\ell}^{\ell+1}(\cos\chi)\sin^\ell\chi \, Y_{\ell m}(\theta, \phi). \qquad (10)$$

C_k^ℓ is a Gegenbauer polynomial and $Y_{\ell m}$ is a spherical harmonic.[38] The Jastrow factor J is a symmetric function of the interelectronic distances r_{ij} containing two-body (2B), three-

body (3B), and four-body (4B) terms.[45,46] More details will be reported elsewhere.[47]

At low densities, the energy minimization procedure is unstable and the parameters of the Jastrow factor were therefore optimized by variance minimization using Newton's method.[48–51] In all calculations, the statistical error obtained by reblocking analysis[52,53] is under 1 μE_h. For small numbers of electrons, comparisons with extrapolated full configuration interaction (FCI) calculations[54,55] indicate that our VMC energies have sub-μE_h accuracy. They are reported in Table II for various r_s and n values.

Because we have observed that many-body effects are more important for small r_s, we have studied the convergence of the energy for $r_s = 20$ and various n values as 2B, 3B, and 4B terms are successively included. We found that the inclusion of 3B terms systematically lowers the energies by up to 20 μE_h per electron but that the inclusion of 4B terms offers less than 1 μE_h per electron. We therefore eschewed 4B terms in the calculations with $r_s > 20$.

When SGFs were used, the determinant Ψ_0 corresponds to a HF Level 0 calculation (i.e., a single SGF on each Thomson site) but the value of the SGF exponent was optimized to minimize the VMC energy. We found that the SGF basis is superior to the HSH basis for $n = 8, 10, 12,$ and 24, while the two basis sets yield identical energies for the other cases. 8-, 10-, 12-, and 24-glomium are "open-shell" systems, i.e., the highest occupied shell of HSHs is partially occupied and there are several low-lying determinants with significant weights. In contrast, SGFs at the Thomson lattice sites naturally describe the localized electrons and are particularly well suited to these systems. For 48-glomium, computational limitations precluded full exponent optimization and the results in Table II were therefore obtained with HSHs. They are probably less accurate than the other energies.

VI. DISCUSSION

By taking the difference between the CBS-HF energies of Sec. IV and the VMC energies of Sec. V, we have obtained the near-exact correlation energies E_c of n-glomium for various r_s and n values. Our results are reported in Table II.

While we have shown that, in 1D, the correlation energy is a smooth and monotonic function of n,[6,7] the situation is rather different in 3D. As shown in Figure 2 (where have plotted E_c as a function of n), the reduced correlation energies do not change monotonically as n increases. Instead, they initially increase and reach a maximum at $n = 5$ or $n = 6$. Beyond $n = 5$, they oscillate and tend to decrease slowly with n, especially at

TABLE III. HF energies (in mE_h) for ferromagnetic n-glomium with $r_s = 20$. There are M basis functions per Thomson site.

| Basis set		Number of electrons n										
Level	M	2	3	4	5	6	8	10	12	13	24	48
0	1	24.983	43.939	60.016	74.277	88.345	112.911	136.249	157.626	167.991	267.468	441.543
1	7	24.917	43.816	59.890	74.253	88.290	112.893	136.169	157.606	167.951	267.464	441.542
2	19	24.911	43.811	59.886	74.240	88.283	112.893	136.154	157.560	167.947	267.464	441.542
3	27	24.911	43.811	59.886	74.240	88.283	112.893	136.154	157.560	167.947	267.464	441.542
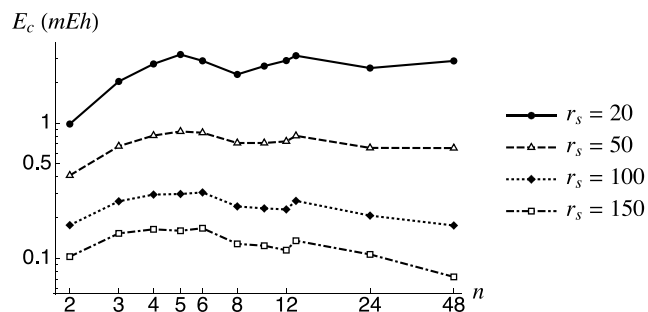
FIG. 2. Reduced correlation energy E_c (in mE_h) as a function of n for various r_s.

very low densities. The oscillations are probably due to "shell effects" which originate from partially filled energy level in open-shell systems (see above). Such shell effects are also observed in 2D.[17] For fixed n, our numerical results show that E_c decreases as $r_s^{-3/2}$ for large r_s. This is expected due to the cancellation of the leading term (proportional to r_s^{-1}) in the exact and HF energies expansion at large r_s (see Eq. (6)). We have also reported the correlation energies[56–58] of the jellium model (which corresponds to $n \to \infty$) in Table II. Exact jellium energies are available in many previous papers[56–63] but fully relaxed HF energies[64,65] are rare. As a result, most of the jellium correlation energies in the literature are based on unrelaxed HF energies and are consequently significantly larger than ours.

The harmonically corrected Thomson energy ($E_0 + E_1$) is usually higher than the exact energy but $n = 2$ and $n = 3$ are exceptional cases. At very low densities, however, it always approximates the exact energy well. Including the first anharmonic correction E_2 would probably yield even better estimates.[66,67]

VII. CONCLUSION

The goal of this work was to generate benchmark correlation energies for the development of improved correlation functionals for DFT calculations on 3D systems. To achieve this, we have studied the correlation energies of low-density spin-polarized electron gases on a glome.

First, we looked at the Thomson problem on a glome, using numerical optimization algorithms to locate uniform lattices, i.e., those in which all the lattice sites are equivalent, for $n \le 50$. We found eleven uniform lattices. We also noted that three of these uniform lattices correspond to well-known 4D Platonic solids: the hyper-tetrahedron ($n = 5$), the hyper-octahedron ($n = 8$), and the 24-cell ($n = 24$). Moreover, we have pointed out the highly symmetric case of $n = 13$ and we stressed that this polychoron has not been previously described anywhere in the literature. By taking into account the quantum oscillation of the electrons around their equilibrium positions, we obtained the harmonic vibrational contribution to the (classical) Thomson energy. As expected, the sum of the Thomson and harmonic energies is a very good approximation of the exact energy at very low density.

Moreover, by systematically increasing the number of s-type SGF basis functions around each Thomson site, we

obtained the CBS HF energy of n-glomium for a range of densities ($20 \le r_s \le 150$). In general, the convergence analysis reveals that only L2 calculations are required to converge the HF energies to microhartree accuracy. However, we note that as we move into the high-density regime, more s-type SGF functions are needed to reach the CBS limit. In this regime, HSHs might constitute a more suitable one-electron basis set. We will investigate this in a forthcoming paper.

The near-exact energies were obtained using highly accurate VMC stochastic calculations using HSH and SGF one-electron basis sets. We have shown that four-body terms in the Jastrow factor have insignificant effect on the energy. The energies obtained using both basis sets are in very good agreement, except for 8-, 10-, 12-, and 24-glomium where the SGF basis is superior.

The present work is a significant step towards the construction of correlation functionals for molecules and solids. The next step is to generate accurate correlation energies within the high density regime, and for partially polarized systems. This work is underway and our results will be reported elsewhere.

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