

Thinking outside the box: The uniform electron gas on a hypersphere

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We discuss alternative homogeneous electron gas systems in which a finite number n of electrons are confined to a D -dimensional sphere. We derive the first few terms of the high-density ($r_s \rightarrow 0$, where r_s is the Seitz radius) energy expansions for these systems and show that, in the thermodynamic limit ($n \rightarrow \infty$), these terms become identical to those of D -dimensional jellium. © 2011 American Institute of Physics. [doi:10.1063/1.3665393]

I. INTRODUCTION

The D -dimensional uniform electron gas (UEG), or D -jellium, is the foundation of most density functionals. It consists of interacting electrons in an infinite volume and in the presence of a uniformly distributed background positive charge. Traditionally, in its paramagnetic version, the system is constructed by allowing the number n of paired electrons in a D -dimensional box of volume V to approach infinity with $\rho = n/V$ held constant.^{1,2}

Using atomic units, the high-density ($r_s \rightarrow 0$, where r_s is the Seitz radius) expansion of the reduced energy (i.e., energy per electron) of D -jellium is

$$\varepsilon(D) = \varepsilon_T(D) + \varepsilon_X(D) + \varepsilon_C(D), \quad (1)$$

where ε_T and ε_X are kinetic^{3,4} and exchange^{5,6} energies

$$\varepsilon_T(D) = \frac{a_{-2}(D)}{r_s^2}, \quad \varepsilon_X(D) = \frac{a_{-1}(D)}{r_s}, \quad (2)$$

and ε_C is the correlation energy. After many important contributions,⁷⁻²⁶ it is known that, for 2- and 3-jellium, the correlation energy takes the form

$$\varepsilon_C(D) = \sum_{j=0}^{\infty} [a_j(D) + b_j(D) \ln r_s] r_s^j. \quad (3)$$

The constant term in Eq. (3) is usually decomposed as

$$a_0(D) = a_{0,J}(D) + a_{0,K}(D), \quad (4)$$

where $a_{0,J}$ is the direct (“ring-diagram”) contribution, and $a_{0,K}$ is the second-order exchange part. The first few a_j and b_j are known analytically or numerically for the important $D = 2$ and $D = 3$ cases (see Table I).

In this article, we introduce an alternative paradigm, in which the electrons are confined to a D -sphere, that is, the surface of a $(D + 1)$ -dimensional ball. These systems possess uniform densities, even for finite n , and because all points on a D -sphere are equivalent, their mathematical analysis is relatively straightforward.²⁷⁻³¹

Electronic properties of the UEG on a 2-sphere have been previously studied in modeling multielectron bubbles in liquid helium (see Ref. 32), and similarities between this system

and 2-jellium have been noticed by Longe and Bose.³³ However, the UEG on a 3-sphere has not been considered before, and this article presents the first study of correlation effects in a spherically confined three-dimensional UEG.

II. HARTREE-FOCK ENERGIES

A. Exact results for finite n

The orbitals for an electron on a D -sphere of radius R are the normalized hyperspherical harmonics $Y_{\ell\mu}$, where ℓ is the principal quantum number and μ is a composite index of the remaining quantum numbers.^{34,35} We confine our attention to systems in which every orbital with $\ell = 0, 1, \dots, L$ is occupied by two electrons, thus yielding an electron density that is uniform on the sphere (see Eq. (9) below). The resulting model is defined completely by the three parameters D, L , and R .

The volume of a D -sphere is

$$V = \frac{2\pi^{\frac{D+1}{2}}}{\Gamma(\frac{D+1}{2})} R^D, \quad (5)$$

where Γ is the gamma function,³⁶ the number of orbitals with quantum number ℓ is

$$n_\ell = \frac{(2\ell + D - 1)\Gamma(\ell + D - 1)}{\Gamma(D)\Gamma(\ell + 1)}, \quad (6)$$

and each of these has energy

$$\kappa_\ell = \frac{\ell(\ell + D - 1)}{2R^2}. \quad (7)$$

Because the total number of electrons is

$$n = 2 \sum_{\ell=0}^L n_\ell = 2 \frac{(2L + D)\Gamma(L + D)}{\Gamma(D + 1)\Gamma(L + 1)}, \quad (8)$$

it follows that the uniform electron density is

$$\begin{aligned} \rho &= \frac{n}{V} = \frac{\Gamma(D/2 + 1)}{\pi^{D/2} r_s^D} \\ &= \frac{(2L + D)\Gamma(L + D)}{\Gamma(D/2 + 1)\Gamma(L + 1)} \frac{1}{(4\pi R^2)^{D/2}}, \end{aligned} \quad (9)$$

and the Seitz radius is

$$r_s = \alpha_D \left[\frac{\Gamma(L + 1)}{(L + D/2)\Gamma(L + D)} \right]^{1/D} R \quad (10)$$

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TABLE I. High-density coefficients for D -jellium and the UEG on a D -sphere. β and ζ are the Dirichlet beta and Riemann zeta functions.³⁶

Coefficient	Term	$D = 2$	$D = 3$
a_{-2}	r_s^{-2}	1/2	$\frac{3}{10}(9\pi/4)^{2/3}$
a_{-1}	r_s^{-1}	$-\frac{4\sqrt{2}}{3\pi}$	$-\frac{3}{4\pi}(9\pi/4)^{1/3}$
b_0	$\ln r_s$	0	$(1 - \ln 2)/\pi^2$
a_0	r_s^0	$a_{0,J}$	$\ln 2 - 1$
		$a_{0,K}$	$\beta(2) - \frac{8}{\pi^2}\beta(4)$
			$\frac{\ln 2}{6} - \frac{3}{4\pi^2}\zeta(3)$
b_1	$r_s \ln r_s$	$-\sqrt{2}\left(\frac{10}{3\pi} - 1\right)$	+0.009229

with

$$\alpha_D = 2^{1-1/D}\Gamma^{2/D}(D/2 + 1). \quad (11)$$

Using the hyperspherical harmonic addition theorem,³⁵ one finds that the one-particle density matrix is

$$\begin{aligned} \rho_1(\mathbf{\Omega}_1, \mathbf{\Omega}_2) &= 2 \sum_{\ell=0}^L \sum_{\mu=1}^{n_\ell} Y_{\ell\mu}^*(\mathbf{\Omega}_1) Y_{\ell\mu}(\mathbf{\Omega}_2) \\ &= \rho \frac{\Gamma(D/2 + 1)\Gamma(L + 1)}{\Gamma(L + D/2 + 1)} P_L^{(D/2, D/2-1)}(\cos\theta), \end{aligned} \quad (12)$$

where $P_L^{(\alpha,\beta)}$ is a L th degree Jacobi polynomial.³⁶ The angle θ is that subtended by the electrons at the origin and is related to the interelectronic distance by the relation.³⁷

$$r_{12} \equiv |\mathbf{r}_1 - \mathbf{r}_2| = R\sqrt{2 - 2\cos\theta}. \quad (13)$$

The density matrix decays rapidly with interelectronic separation when L is large (Fig. 1), illustrating the ‘‘short-sightedness’’ of matter.^{38,39}

Many properties of the UEG on a D -sphere can be found from Eqs. (6) to (12). Its kinetic energy, for example, is

$$\varepsilon_T(D, L) = \frac{2}{n} \sum_{\ell=0}^L n_\ell \kappa_\ell = \frac{D}{2(D+2)} \frac{L(L+D)}{R^2}, \quad (14)$$

and it can be shown that its exchange energy is

$$\begin{aligned} \varepsilon_X(D, L) &= -\frac{1}{2n} \iint \frac{\rho_1(\mathbf{\Omega}_1, \mathbf{\Omega}_2)^2}{r_{12}} d\mathbf{\Omega}_1 d\mathbf{\Omega}_2 \\ &= -\frac{n}{2R} \frac{\Gamma(\frac{D+1}{2})}{\sqrt{\pi} \Gamma(\frac{D}{2})} \frac{D \Gamma(D-1)}{2L+D} \frac{\Gamma(L+\frac{3}{2})}{\Gamma(L+D-\frac{1}{2})} \\ &\quad \times {}_4F_3 \left[\begin{matrix} -L, & L+D, & \frac{D-1}{2}, & -\frac{1}{2}; & 1 \\ -L-\frac{1}{2}, & L+D-\frac{1}{2}, & \frac{D+2}{2} \end{matrix} \right], \end{aligned} \quad (15)$$

where ${}_4F_3$ is a generalized hypergeometric function.³⁶

B. The thermodynamic limit

The above expressions are exact for all L but, in the thermodynamic limit ($n, L \rightarrow \infty$), each simplifies significantly.

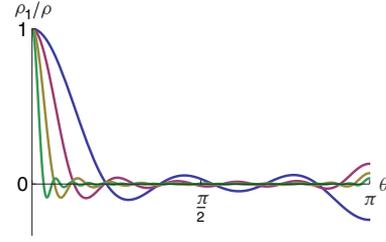


FIG. 1. The one-particle density matrix for the UEG on a 3-sphere. Plots for $L = 5$ (blue), $L = 10$ (red), $L = 20$ (yellow), and $L = 40$ (green).

For example,

$$n \rightarrow \frac{4}{\Gamma(D+1)} L^D, \quad (16)$$

$$\rho \rightarrow \frac{2}{\Gamma(D/2+1)} \frac{L^D}{(4\pi R^2)^{D/2}}, \quad (17)$$

$$r_s \rightarrow \alpha_D \frac{R}{L}, \quad (18)$$

$$\rho_1 \rightarrow \rho \frac{\Gamma(D/2+1) J_{D/2}(L\theta)}{(L\theta/2)^{D/2}}, \quad (19)$$

where J_n is the n th-order Bessel function.³⁶ We note that Eq. (19) reduces to the usual density matrices in 2-jellium⁴⁰ and 3-jellium.⁵ The kinetic and exchange energies become

$$\varepsilon_T(D) = + \frac{D}{2(D+2)} \frac{\alpha_D^2}{r_s^2}, \quad (20)$$

$$\varepsilon_X(D) = - \frac{2D}{\pi(D^2-1)} \frac{\alpha_D}{r_s}. \quad (21)$$

Equations (20) and (21) yield the two terms in Eq. (2) and are identical to the D -jellium expressions. Particular cases are given in Table I. These results were originally discovered by Glasser and Boersma,⁴¹ and Iwamoto⁴² for D -jellium, but our derivation for the UEG on a D -sphere is more compact than theirs.

III. CORRELATION ENERGIES

A. Exact results for finite n

We now turn our attention to the study of the correlation energy of the spherically confined UEGs. By applying perturbation theory to UEG on a 2-sphere, we find that the reduced energy coefficient corresponding to the lowest order ring-diagram contribution is

$$\begin{aligned} a_{0,J}(2, L) &= \frac{2}{n} \sum_{i,j}^{\text{occ}} \sum_{a,b}^{\text{virt}} \frac{\langle ij|ab \rangle^2}{\kappa_i + \kappa_j - \kappa_a - \kappa_b} \\ &= \frac{1}{n} \sum_{i,j=0}^L \sum_{a,b=L+1}^{\infty} \frac{(2i+1)(2j+1)(2a+1)(2b+1)}{\kappa_i + \kappa_j - \kappa_a - \kappa_b} \\ &\quad \times \sum_{\ell} \frac{1}{2\ell+1} \begin{pmatrix} i & \ell & a \\ 0 & 0 & 0 \end{pmatrix}^2 \begin{pmatrix} j & \ell & b \\ 0 & 0 & 0 \end{pmatrix}^2, \end{aligned} \quad (22)$$

where $\langle ij|ab \rangle$ are two-electron integrals and the brackets are 3j symbols.³⁶ For the UEG on a 3-sphere, the coupling coefficient in SO(4) is much simpler than in SO(3) (Ref. 43) and the energy coefficient from the lowest order ring-diagram is

$$c_{0,J}(3, L) = \frac{1}{n} \sum_{i,j=0}^L \sum_{a,b=L+1}^{\infty} \frac{(i+1)(j+1)(a+1)(b+1)}{\kappa_i + \kappa_j - \kappa_a - \kappa_b} \times \sum_{\ell} \frac{(2/\pi)^2}{(\ell + \frac{1}{2})^2 (\ell + \frac{3}{2})^2}, \quad (23)$$

where the sum over ℓ respects the same restrictions as in the 3j symbols in Eq. (22).

The second-order exchange part for the UEG on a 2-sphere is

$$a_{0,K}(2, L) = \frac{1}{n} \sum_{i,j}^{\text{occ}} \sum_{a,b}^{\text{virt}} \frac{\langle ij|ab \rangle \langle ba|ij \rangle}{\kappa_a + \kappa_b - \kappa_i - \kappa_j} = \frac{1}{n} \sum_{i,j=0}^L \sum_{a,b=L+1}^{\infty} \frac{(2i+1)(2j+1)(2a+1)(2b+1)}{\kappa_a + \kappa_b - \kappa_i - \kappa_j} \times \sum_{\ell_1, \ell_2} (-1)^{\ell_1 + \ell_2} \begin{pmatrix} i & \ell_1 & a \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} j & \ell_1 & b \\ 0 & 0 & 0 \end{pmatrix} \times \begin{pmatrix} i & \ell_2 & b \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} j & \ell_2 & a \\ 0 & 0 & 0 \end{pmatrix} \left\{ \begin{matrix} i & \ell_1 & a \\ j & \ell_2 & b \end{matrix} \right\}, \quad (24)$$

where the curly brackets denote 6j symbols,³⁶ and for the UEG on a 3-sphere, we found

$$a_{0,K}(3, L) = \frac{1}{n} \sum_{i,j=0}^L \sum_{a,b=L+1}^{\infty} \frac{(i+1)(j+1)(a+1)(b+1)}{\kappa_a + \kappa_b - \kappa_i - \kappa_j} \times \sum_{\ell_1, \ell_2} \frac{(2/\pi)(\ell_1 + 1)}{(\ell_1 + \frac{1}{2})(\ell_1 + \frac{3}{2})} \frac{(2/\pi)(\ell_2 + 1)}{(\ell_2 + \frac{1}{2})(\ell_2 + \frac{3}{2})} \times \left\{ \begin{matrix} i & \ell_1 & a \\ j & \ell_2 & b \end{matrix} \right\}, \quad (25)$$

where we have used the SO(4) version of the 6j symbols.⁴³ Numerical values for finite L are given in Table II.

TABLE II. Numerical values of $a_{0,J}(2, L)$, $a_{0,K}(2, L)$, $c_{0,J}(3, L)$, and $a_{0,K}(3, L)$ for various L .

L	n	UEG on a 2-sphere		UEG on a 3-sphere		
		$a_{0,J}(2, L)$	$a_{0,K}(2, L)$	n	$c_{0,J}(3, L)$	$a_{0,K}(3, L)$
0	2	-0.2274	+0.1137	2	-0.0476	+0.0238
1	8	-0.2534	+0.1111	10	-0.0717	+0.0231
2	18	-0.2677	+0.1118	28	-0.0897	+0.0231
3	32	-0.2762	+0.1124	60	-0.1038	+0.0233
4	50	-0.2816	+0.1128	110	-0.1154	+0.0234
⋮	⋮	⋮	⋮	⋮	⋮	⋮
∞	∞	-0.3069	+0.1144	∞	-∞	+0.0242

B. The thermodynamic limit

In the thermodynamic limit ($n \rightarrow \infty$), Eq. (22) becomes

$$a_{0,J}(2) = - \int_0^{\infty} \frac{d\ell}{\ell} \int_0^1 i \, di \int_0^1 j \, dj \int_{\max(1, |\ell-i|)}^{\ell+i} a \, da \int_{\max(1, |\ell-j|)}^{\ell+j} \times b \, db \frac{\mathcal{J}_{i,\ell,a} \mathcal{J}_{j,\ell,b}}{a^2 + b^2 - i^2 - j^2}, \quad (26)$$

where

$$\mathcal{J}_{i,\ell,a} = \frac{2}{\pi} \frac{1}{\sqrt{(i+\ell+a)(i+\ell-a)(\ell+a-i)(a+i-\ell)}} \quad (27)$$

comes from the asymptotic expansion of the 3j symbol.⁴⁴ Defining $a^2 = i^2 + \ell^2 - 2i\ell \cos \theta_1$ and $b^2 = j^2 + \ell^2 - 2j\ell \cos \theta_2$ to transform the a and b integrals into θ_1 and θ_2 integrals, and then renaming i, j , and ℓ as p_1, p_2 , and q , we obtain

$$a_{0,J}(2) = - \frac{1}{4\pi^3} \int \frac{d\mathbf{q}}{q^2} \int_{|p_1|<1} \int_{\substack{|p_2|<1 \\ |p_2+q|>1}} \frac{d\mathbf{p}_1 d\mathbf{p}_2}{q^2 + \mathbf{q} \cdot (\mathbf{p}_1 + \mathbf{p}_2)}, \quad (28)$$

which is precisely the lowest order ring-diagram contribution of 2-jellium.¹¹ This integral can be solved¹⁶ to yield

$$a_{0,J}(2) = \ln 2 - 1. \quad (29)$$

One also finds that the higher order ring-diagram contributions are identical in 2-jellium and in the UEG on a 2-sphere and the resummation technique¹¹ yields

$$b_0(2) = 0, \quad b_1(2) = -\sqrt{2} \left(\frac{10}{3\pi} - 1 \right). \quad (30)$$

For large L , the sums in Eq. (23) can be replaced by integrals and one finds and the leading order of (23) is

$$c_{0,J}(3, L) \sim - \frac{3}{\pi^2} \int_{1/L}^1 d\ell \int_0^{\infty} dt \left[\int_{1-\ell}^1 \int_1^{i+\ell} \frac{ai}{\ell^2} e^{-(a^2-i^2)t} da \, di \right]^2 = \frac{1 - \ln 2}{\pi^2} \ln \frac{1}{L} + O(L^0) = \frac{1 - \ln 2}{\pi^2} \ln r_s + O(r_s^0). \quad (31)$$

It follows that

$$b_0(3) = \frac{1 - \ln 2}{\pi^2}, \quad (32)$$

and thus the logarithmic divergence of the correlation energy in the UEG on a 3-sphere is exactly the same as in 3-jellium. One notes that the result (Eq. (31)) can be derived for any value of the radius R . The latter divergence, contrary to some claims in the literature, does not result from the long-range part of the Coulomb operator but from its short-range part.^{45,46} The observation of the same divergence in the UEG on a 3-sphere—where the interelectronic distance can never exceed $2R$ —also demonstrates this.

Proceeding similarly to the $D = 2$ case, it can be shown that, in the thermodynamic limit, Eq. (23) becomes

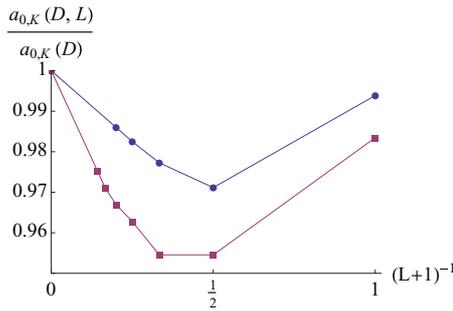


FIG. 2. $\frac{a_{0,K}(D,L)}{a_{0,K}(D)}$ as a function of $(L+1)^{-1}$ for $D=2$ (blue dots) and $D=3$ (red squares).

identical to the expression of the second-order ring-diagram in 3-jellium

$$c_{0,J}(3) = -\frac{3}{16\pi^5} \int \frac{dq}{q^4} \int_{\substack{|p_1| < 1 \\ |p_1+q| > 1}} \int_{\substack{|p_2| < 1 \\ |p_2+q| > 1}} \frac{dp_1 dp_2}{q^2 + \mathbf{q} \cdot (\mathbf{p}_1 + \mathbf{p}_2)}, \quad (33)$$

where the excitation vector q has the domain $\sqrt{r_s} < q < \infty$.⁸ Moreover, the higher order ring-diagram contributions are also identical in 3-jellium and in the UEG on a 3-sphere. Using the resummation technique,^{8,13} it follows that⁴⁷

$$a_{0,J}(3) = -0.071099. \quad (34)$$

For $D=2$ and 3, we have not been able to prove the equivalence of the second-order exchange contributions in D -jellium and in the UEG on a D -sphere, but the numerical results in Table II and Fig. 2 suggest that, in the thermodynamic limit, $a_{0,K}(2) \approx +0.11$ and $a_{0,K}(3) \approx +0.024$, which may be compared with the known 2-jellium and 3-jellium values^{9,12}

$$a_{0,K}(2) = \beta(2) - \frac{8}{\pi^2} \beta(4) = +0.114357, \quad (35)$$

$$a_{0,K}(3) = \frac{\ln 2}{6} - \frac{3}{4\pi^2} \zeta(3) = +0.024179, \quad (36)$$

where β and ζ are the Dirichlet beta and Riemann zeta functions.³⁶

IV. DISCUSSION

Uniform electron gases on a D -sphere are an attractive generalization of D -jellium and, as we have shown, one can derive compact expressions for the first few terms of the high-density energy expansions for both finite and infinite systems. Although UEGs on a D -sphere are physically different from D -jellium, we have shown that, in the thermodynamic limit, the first few terms of their high-density energy expansions are identical and we conjecture that the high-density expansions are identical to all orders.

Recent calculations on the Thomson problem suggest that the leading term of the low-density (large- r_s) energy expansions in 2-jellium and in the UEG on a 2-sphere are also identical.⁴⁸ Moreover, because the Thomson problem is trivial for $D=1$, it is actually possible to show the strict equality

of the leading term of the low-density energy expansions in 1-jellium and in the UEG on a ring (1-sphere).⁴⁹

Although it is pleasing to know that the spherical and conventional gases become equivalent in the thermodynamic limit, we believe that it is even more important to recognize that they are *not* equivalent for finite n . Equations (14) and (15) predict significantly different kinetic and exchange energies from Eqs. (20) and (21) when n is small. Moreover, combining the information from the high- and low-density regimes, one can easily construct local-density approximation-type correlation functionals for finite systems using interpolation functions.^{14,15,50} We believe that the UEG on a D -sphere will be useful in the future development of correlation functionals within density-functional theory.⁵¹

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