Lessons from electron(s) on sphere(s)

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Mathematical Methods in Quantum Chemistry
Oberwolfach, Germany
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Arguments for high-impact journals

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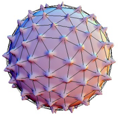

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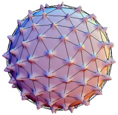

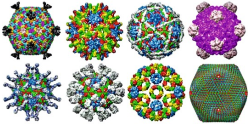

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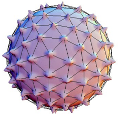
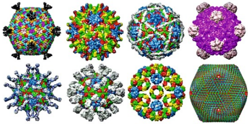
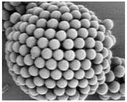

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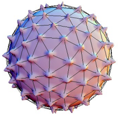

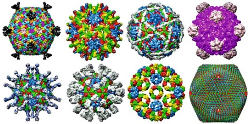

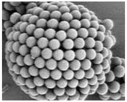

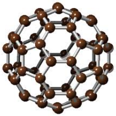

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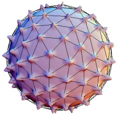

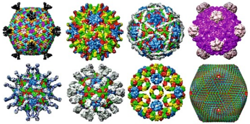

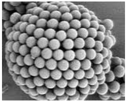

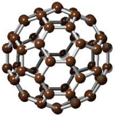

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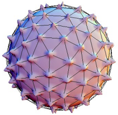

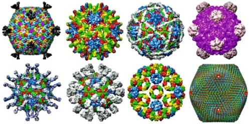

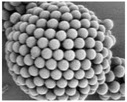

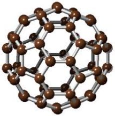

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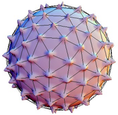
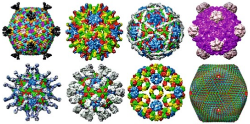
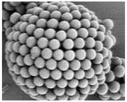
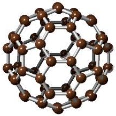

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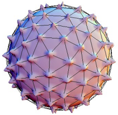

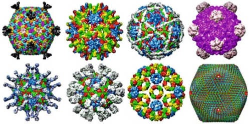

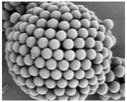

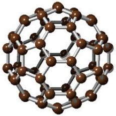

Our arguments...

- This is mathematically challenging
- This is actually related to "real" systems
- It yielded a number of unexpected discoveries

The spherium atom: electron(s) on a sphere of radius R

Loos & Gill Phys Rev A 79 (2009) 062517

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One electron on a sphere

$$\hat{H} = -\frac{1}{2}\nabla^2$$

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??? $\Rightarrow$ Exciting!!!

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Hamiltonian of the ground state

$$\boxed{\hat{H} = \left(\frac{r_{12}^2}{4R^2} - 1\right)\frac{d^2}{dr_{12}^2} + \left(\frac{3r_{12}}{4R^2} - \frac{1}{r_{12}}\right)\frac{d}{dr_{12}} + \frac{1}{r_{12}}}$$

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We seek polynomial solutions: $\Psi(\mathbf{r}_1, \mathbf{r}_2) = \sum_{\ell=0}^{\infty} c_\ell r_{12}^\ell$

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Analytical solutions

$$\begin{array}{lll} R = \sqrt{3}/2 & E = 1 & \Psi(\mathbf{r}_1, \mathbf{r}_2) = 1 + r_{12} \\ R = \sqrt{7} & E = 2/7 & \Psi(\mathbf{r}_1, \mathbf{r}_2) = 1 + r_{12} + \frac{5}{28} r_{12}^2 \\ \vdots & \vdots & \vdots \end{array}$$

Loos & Gill Phys Rev Lett 103 (2009) 123008

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Analytical solutions

$$\begin{array}{lll} R = \sqrt{10}/2 & E = 1/2 & \Psi(\mathbf{r}_1, \mathbf{r}_2) = 1 + \frac{1}{2} r_{12} \\ R = \sqrt{66}/2 & E = 2/11 & \Psi(\mathbf{r}_1, \mathbf{r}_2) = 1 + \frac{1}{2} r_{12} + \frac{7}{132} r_{12}^2 \\ \vdots & \vdots & \vdots \end{array}$$

Generalization to a D-dimensional space

First exact solutions for a D-sphere

State	D	$2R$	E	$\Psi(\mathbf{r}_1, \mathbf{r}_2)$
^1S	1	$\sqrt{6}$	2/3	$r_{12}(1 + r_{12}/2)$
	2	$\sqrt{3}$	1	$1 + r_{12}$
	3	$\sqrt{10}$	1/2	$1 + r_{12}/2$
	4	$\sqrt{21}$	1/3	$1 + r_{12}/3$
^3P	1	$\sqrt{6}$	1/2	$1 + r_{12}/2$
	2	$\sqrt{15}$	1/3	$1 + r_{12}/3$
	3	$\sqrt{28}$	1/4	$1 + r_{12}/4$
	4	$\sqrt{45}$	1/5	$1 + r_{12}/5$

Loos & Gill Phys Rev Lett 103 (2009) 123008; Mol Phys 108 (2010) 2527

Uniform electron gas in Flatland

The 2D- and 3D-jellium model

Giuliani & Vignale, Quantum theory of electron liquid

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- Uniform electron density at all points only if $n \to \infty$ and $V \to \infty$
- Characterized by one parameter: the Seitz radius r_s

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Uniform electron gas in Sphereland

We fill each (hyper)spherical harmonic $Y_{\ell m(n)}$ up to $\ell = L$ with one up- and one down-electron

Loos & Gill Phys Rev B (submitted) arXiv:1101.3131

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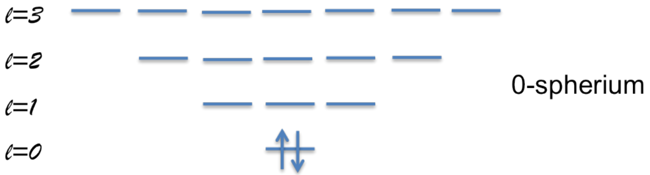

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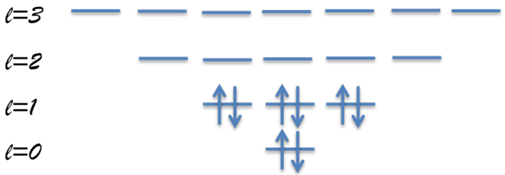

1-spherium

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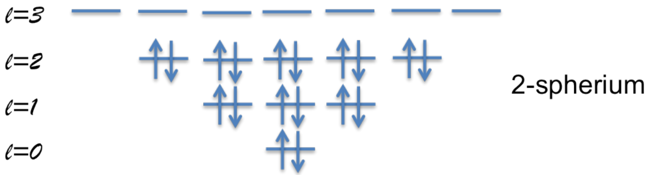

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L-Spherium

$$\boxed{\sum_{m=-\ell}^{+\ell} |Y_{\ell m}(\theta,\phi)|^2 = \frac{2\ell+1}{4\pi}}$$

$$n = 2(L+1)^2$$

$$\rho = \frac{(L+1)^2}{2\pi R^2} = \frac{1}{\pi r_s^2}$$

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L-Glomium

$$\boxed{\sum_{m=0}^{\ell} \sum_{n=-m}^{+m} |Y_{\ell mn}(\chi,\theta,\phi)|^2 = \frac{(\ell+1)^2}{2\pi^2}}$$

$$n = 2(L+1)(L+3/2)(L+2)/3$$

$$\rho = \frac{(L+1)(L+2)(2L+3)}{6\pi^2 R^3} = \frac{3}{4\pi r_s^3}$$

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High-density ($r_s \to 0$) limit: L-spherium vs. 2D-jellium

$$\bar{E}_{\text{2D-jell}}(r_s) = \frac{\varepsilon_{-2}}{r_s^2} + \frac{\varepsilon_{-1}}{r_s} + (\varepsilon_{0,\text{J}} + \varepsilon_{0,\text{K}}) + \lambda_1 r_s \ln r_s + O(r_s)$$

$\varepsilon_{-2} =$

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$$\varepsilon_{-2} = + \frac{L(L+2)}{2(L+1)^2}$$

$$\varepsilon_{-1} = - \frac{1}{\sqrt{2}} F \left[\begin{array}{cccc} -L, & L+2, & \frac{1}{2}, & -\frac{1}{2} \\ -L-\frac{1}{2}, & L+\frac{3}{2}, & 2 & \end{array} \right]$$

$$\varepsilon_{0,\text{J}} = - \frac{2}{n} \sum_{ij}^{\text{occ}} \sum_{ab}^{\text{virt}} \frac{\langle ij|ab\rangle^2}{\kappa_a + \kappa_b - \kappa_i - \kappa_j}$$

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$$\lambda_1 = \text{(resummation)}$$

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$$\lambda_1 = (\text{resummation}) \xrightarrow[L\to\infty]{} - \sqrt{2}\left(\frac{10}{3\pi} - 1\right)$$

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$$\lambda_1 = \text{(resummation)} \xrightarrow[L\to\infty]{} - \sqrt{2}\left(\frac{10}{3\pi} - 1\right) \quad \checkmark$$

High-density ($r_s \to 0$) limit: L-glomium vs. 3D-jellium

$$\bar{E}_{\text{3D-jell.}}(r_s) = \frac{\varepsilon_{-2}}{r_s^2} + \frac{\varepsilon_{-1}}{r_s} + \lambda_0 \ln r_s + (\varepsilon_{0,\text{J}} + \varepsilon_{0,\text{K}}) + O(r_s \ln r_s)$$

ε_{-2}

ε_{-1}

λ_0

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Conjecture: high-density expansions identical to all order!!

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- They might be more convenient models for DFT functional development