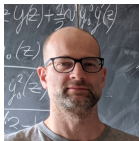


Wigner localization in one, two and three dimensions: an *ab initio* approach.

Miguel Escobar Azor.



with thanks to:



Arjan Berger



Stefano Evangelisti



Alfredo Sánchez de Merás

www.daltonprogram.org

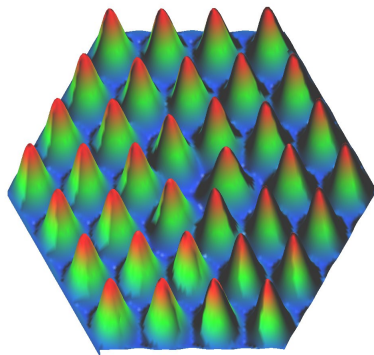
Wigner localization in two and three dimensions: An *ab initio* approach

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Miguel Escobar Azor, Estefania Alves, Stefano Evangelisti and J. Arjan Berger

What is a Wigner Crystal?

Almost a century ago E. Wigner predicted that a system consisting of interacting electrons in a neutralizing uniform background would, at sufficiently low densities, crystallize with electrons sharply localized around equilibrium positions.



Wigner crystallization in 2D. From P. D. Drummond et al., J. of Math. Phys. 54, 042107 (2013)

What is a Wigner Crystal?

For a system constituted of free electrons, two different limits can be observed, a high- and a low-density regimes.

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- High (electron) density (small r_s)
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 - Fermi liquid = delocalized state
- Low (electron) density (large r_s)
 - The electronic repulsion energy, proportional to r_s^{-1} , dominates the kinetic energy (strongly-correlated regime).
 - Wigner crystal = electrons localize on lattice sites (Wigner localization).

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Basis set

$$g_\mu(\mathbf{r} - \mathbf{R}_\mu) = \left(\frac{2\alpha}{\pi}\right)^{\frac{3}{4}} e^{-\alpha|\mathbf{r}-\mathbf{R}_\mu|^2}$$

M -identical 1s Gaussian orbitals evenly distributed on a regular d -dimensional grid having a common α exponent.

- Confine the electrons to a d -dimensional space with a positive background.

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- Use of Periodic boundary conditions (PBC).

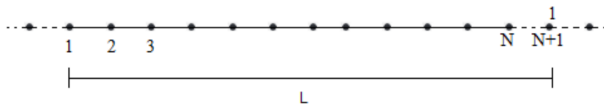
A Clifford torus is a flat, closed d -dimensional real Euclidean space embedded in a d -dimensional complex Euclidean space. The approach consists in creating a d -dimensional regular supercell and then modify its topology into the topology of a d -dimensional torus by identifying and "gluing" opposite sides.

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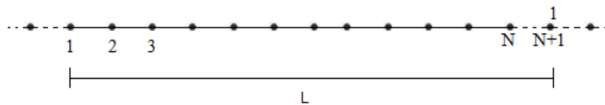
Geodesic distance defined as the length of the shortest path between two points on the surface of the torus.

Clifford Boundary Conditions. Defining the distance.

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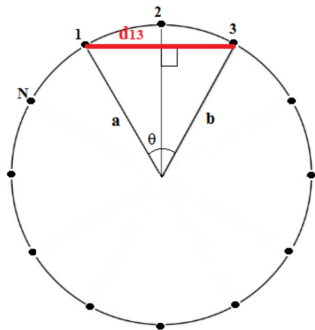
$$r_{12}^{geo} = \sqrt{\sum_{i=1}^d r_{12}^2(i)}$$

$$r_{12}(i) = \begin{cases} |r_1(i) - r_2(i)| & \text{if } |r_1(i) - r_2(i)| \leq \frac{L}{2} \\ L - |r_1(i) - r_2(i)| & \text{if } |r_1(i) - r_2(i)| > \frac{L}{2} \end{cases}$$

Euclidean (or straight-line) distance, defined as the length of the shortest possible path between two points in the embedding space of the Clifford torus.

Clifford Boundary Conditions. Defining the distance.

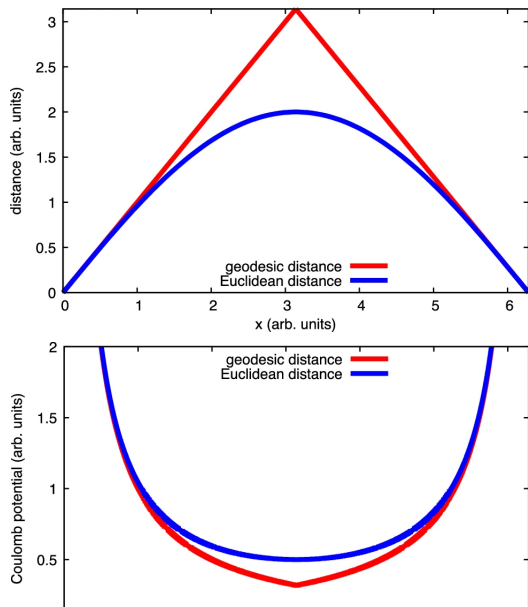
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$$d_{13}^{euc} = \frac{L}{\pi} \sin\left(\frac{\pi r_{13}}{L}\right)$$

$$d_{12}^{euc} = \frac{L}{\pi} \sqrt{\sum_{i=1}^d \sin^2\left(r_{12}(i) \frac{\pi}{L}\right)}$$

Clifford Boundary Conditions. Defining the distance. Geodesic vs Euclidean.



The derivative of the geodesical distance with respect to a Cartesian coordinate (i) is discontinuous in those points where $r_{12}(i) = \frac{L}{2}$. While the Euclidean distance is smooth and continuously differentiable.

- Primitive basis consisting of M -identical 1s orbitals.

$$g_{\mu}(\mathbf{r} - \mathbf{R}_{\mu}) = \left(\frac{2\alpha}{\pi}\right)^{\frac{3}{4}} e^{-\alpha|\mathbf{r}-\mathbf{R}_{\mu}|^2}$$

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- The eigenstates of \hat{H} can be chosen to be equal to the eigenstates of \hat{T}_R

$$\phi_{\mathbf{k}}(\mathbf{r}) = S_{\mathbf{k}}^{-1/2} \sum_{\mu \in CSC} e^{i\frac{2\pi}{m}\mathbf{k}\cdot\boldsymbol{\mu}} g_{\mu}(\mathbf{r} - \mathbf{R}_{\mu})$$

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- We solve the time-independent Schrödinger equation by employing an exact diagonalization approach

In the strong correlated limit we can Taylor expand the Coulomb potential in the Hamiltonian

$$\hat{H} = \underbrace{-\frac{1}{2} \sum_{1,2} \nabla_{\vec{r}_i}^2 + \hat{U}_0 + \hat{U}_1 + \hat{U}_2 + \hat{U}_3 + \hat{U}_4 + \dots}_{\hat{H}_0}$$

\hat{U}_0 is the classical energy of two electrons located at their equilibrium positions.

$$\hat{U}_0 = \frac{1}{r_{12}^{euc}} \rightarrow U_0 = \frac{\pi}{\sqrt{dL}}$$

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\hat{U}^2 represents the zero-point correction to the classical energy in the harmonic approximation. Equivalent to a quantum harmonic oscillator with harmonic frequency

$$\omega = \sqrt{\frac{2\pi^3}{(dL^2)^{\frac{3}{2}}}}$$

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The anharmonics corrections can be calculated using perturbation theory where the lowest-order correction to the energy consists of \hat{U}_3^2 and \hat{U}_4^1

$$U_4^1 = \frac{(6-d)\pi^2}{16L^2}$$

For two electrons confined into a Clifford supercell the diagonal elements of the 2-RDM are given by

$$\Gamma(\mathbf{r}_1, \mathbf{r}_2) = |\Psi(\mathbf{r}_1, \mathbf{r}_2)|^2$$

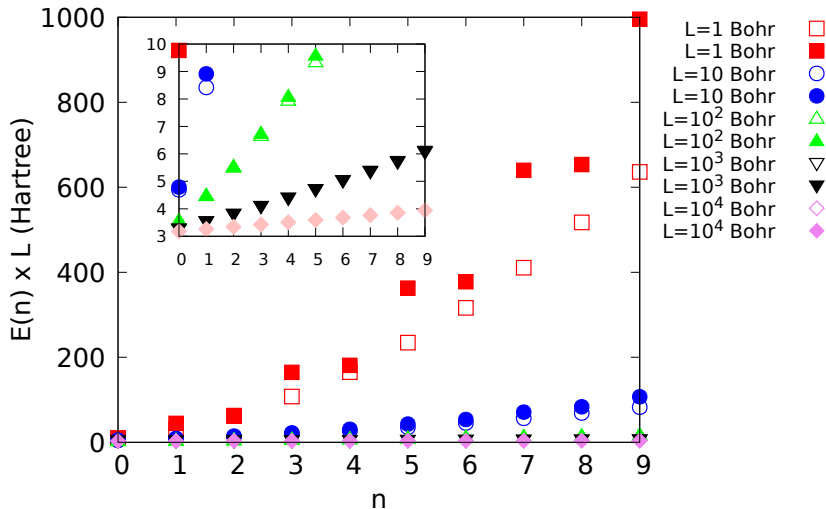
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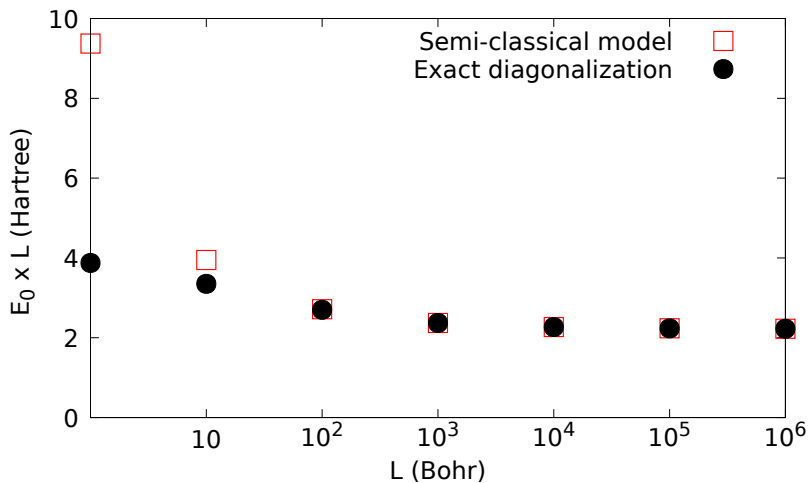
It is useful to express Γ into the gaussian basis set according to

$$\Gamma_{\mu,\nu} = \iint g_{\mu}(\mathbf{r}_1)g_{\nu}(\mathbf{r}_2)\Gamma(\mathbf{r}_1, \mathbf{r}_2)d\mathbf{r}_1d\mathbf{r}_2$$

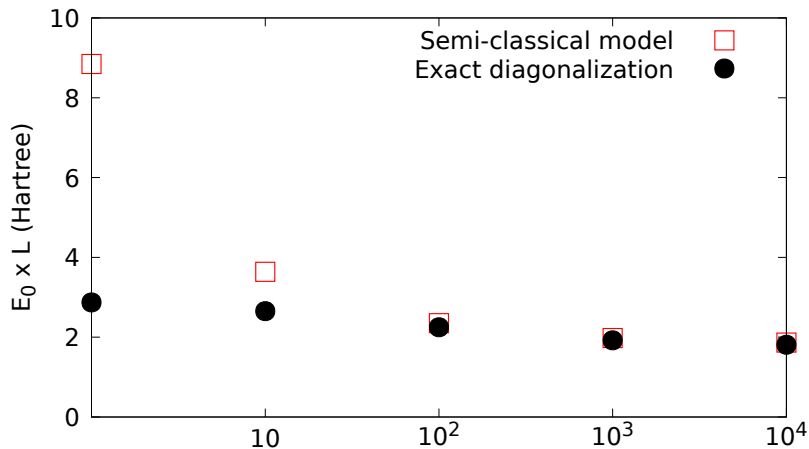
1-Clifford Torus Validation Semi-Classical model vs Quantum approach



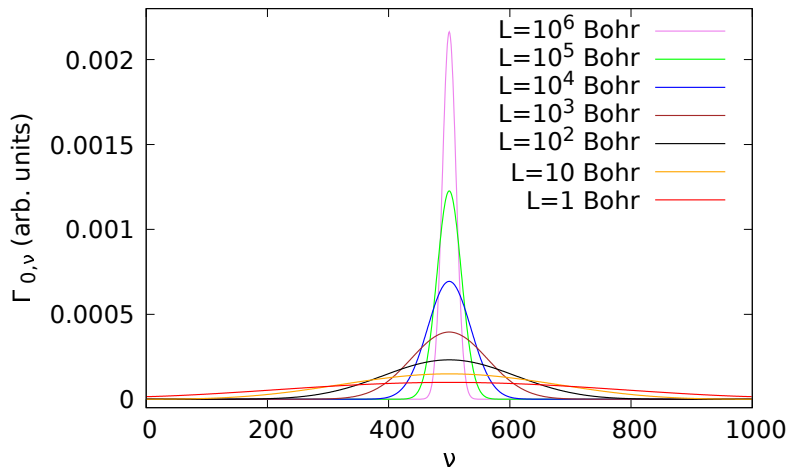
2-Clifford Torus Validation Semi-Classical model vs Quantum approach



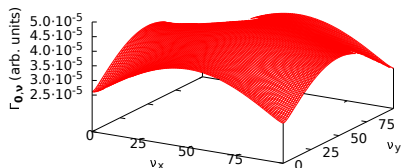
3-Clifford Torus Validation Semi-Classical model vs Quantum approach



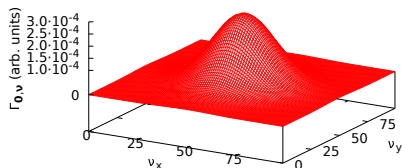
2-RDM 1-Clifford Torus



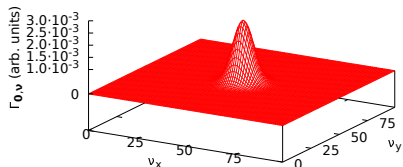
$L=1$ Bohr



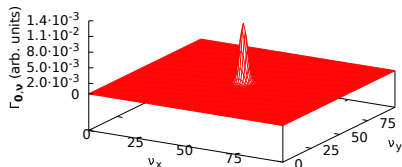
$L = 10^2$ Bohr



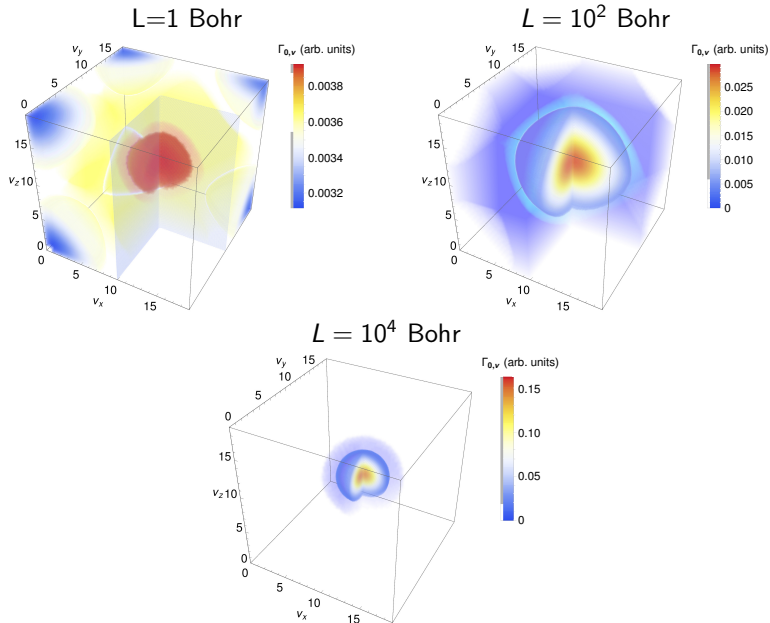
$L = 10^4$ Bohr



$L = 10^6$ Bohr



2-RDM 3-Clifford Torus



We have presented an accurate and numerically efficient approach to study Wigner localization in systems of various dimensions. Its main features are:

- The application of Clifford periodic boundary conditions with a renormalized distance to describe the Coulomb potential
- The use of M -identical gaussian basis functions evenly distributed on a regular grid inside a Clifford supercell.

- The generalization of our approach to more than 2 electrons in order to study Wigner crystals.
- The implementation of our approach in Hartree-Fock (HF) theory and post-HF *ab initio* approaches such as coupled-cluster theory.
- The inclusion of ions in our approach which will allow the study of the solid state.
- The development of specific Gaussian basis set adapted to the topology of the systems (Toroidal Gaussians).

Thanks for your attention!