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# Linear Scaling Quantum Monte Carlo

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# Comparison of Methods

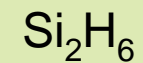
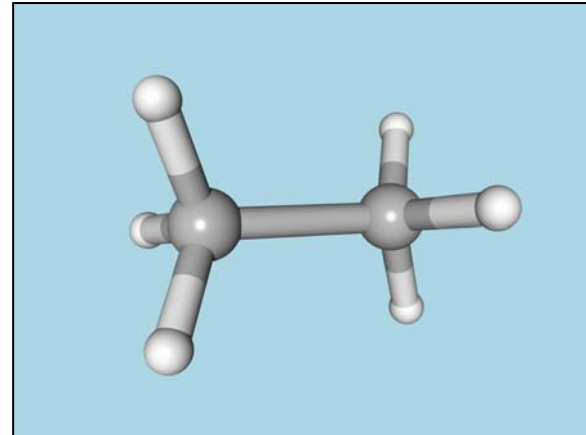
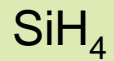
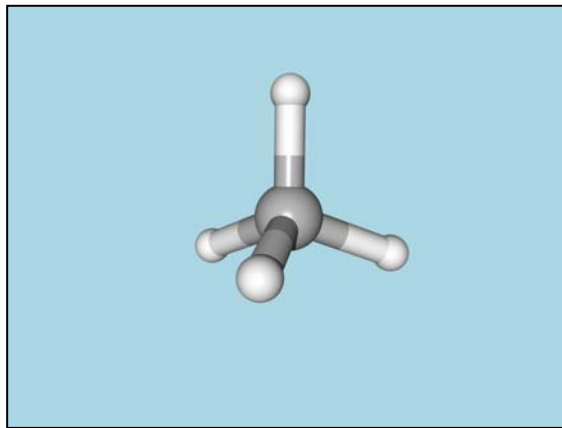
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Method	$E_{\text{corr}}$	$E_{\text{bind}}$ % errors	Scaling	Time for $C_{10}$
HF	0	$\approx 50\%$	$N^3$	14
LDA	N/A	15-25 %	$N^3$	1
VMC	85 %	2-10 %	$N^3$	16
DMC	$\approx 95\%$	1-4 %	$N^3$	300
CCSD(T)	$\approx 75\%$	10-15 %	$N^7$	1500

# Traditional View of QMC

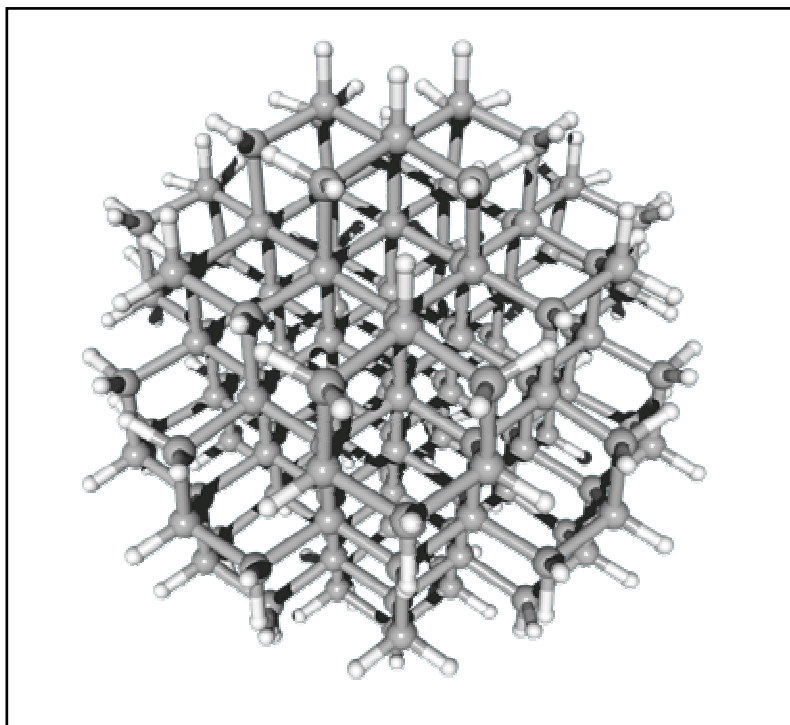
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*“...only applicable to atoms and small molecules....”*

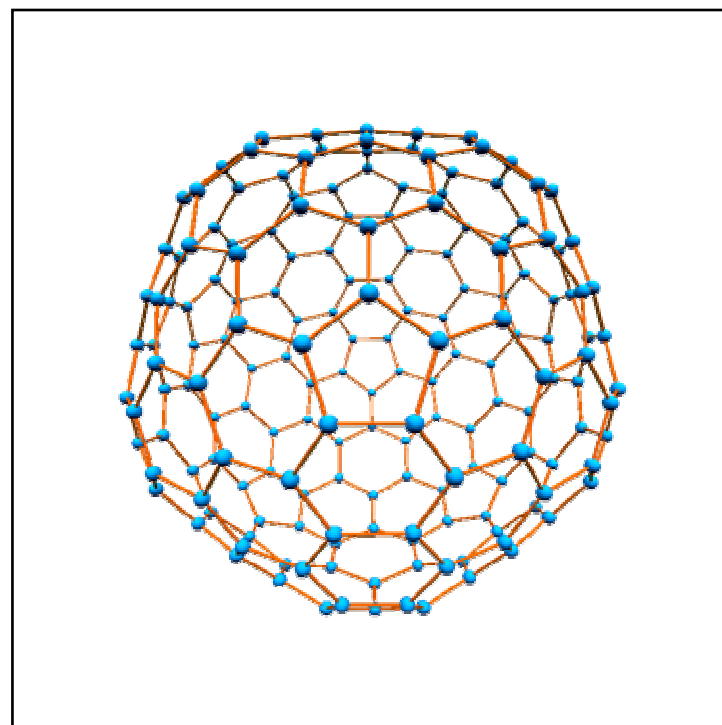


# State of the art QMC

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$\text{Si}_{148}\text{H}_{120}$



$\text{C}_{180}$

# Quantum Monte Carlo

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## Variational Monte Carlo

- Time independent Schrödinger equation

$$\hat{H} \Psi(\mathbf{R}) = E \Psi(\mathbf{R}) \quad , \quad E \approx \frac{\langle \Psi_T(\mathbf{R}) \hat{H} \Psi_T(\mathbf{R}) \rangle}{\langle \Psi_T(\mathbf{R}) \Psi_T(\mathbf{R}) \rangle}$$

- Sample configurations,  $\mathbf{R}$ , stochastically

## Diffusion Monte Carlo

Imaginary time dependent Schrödinger equation

$$\frac{d \Phi(R, t)}{dt} = \hat{H} \Phi(R, t)$$

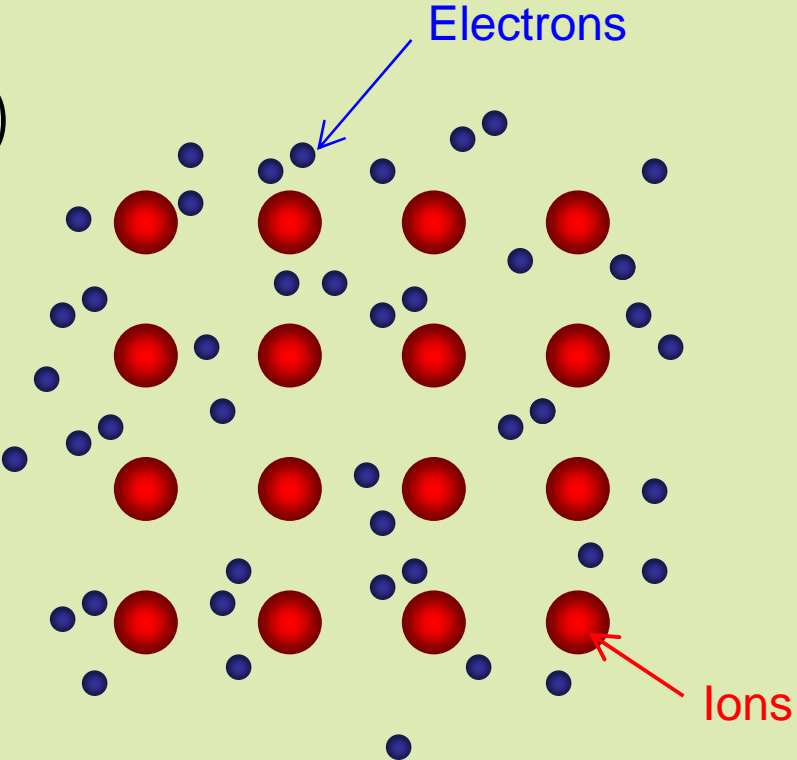
Ground state is projected out in long time limit

→ Provides near exact solution (95% correlation energy)

# Variational Monte Carlo

$\mathbf{R} = (r_1, r_2, r_3, \dots, r_N)$   
Sampled from  $|\Psi_T|^2$

$$E = \frac{1}{N_C} \sum_i^{N_C} \underbrace{\frac{\hat{H} \Psi_T(\mathbf{R}_i)}{\Psi_T(\mathbf{R}_i)}}_{\text{Local Energy}}$$



The diagram illustrates a molecular system. Large red spheres represent ions, and small blue spheres represent electrons. A blue arrow points to a blue sphere labeled "Electrons", and a red arrow points to a red sphere labeled "Ions". The ions are arranged in a regular grid, while the electrons are scattered throughout the space.

# Scaling of different terms

$$\text{CPU time} = \alpha N^2 + \beta N^3$$

$N^3$  terms

- Elements of  $D$  (95%)
- Value of the  $D$

$N^2$  terms

- electron-electron, ion-ion
- Jastrow function

Local Energy

$$\frac{\hat{H} \Psi_T(\mathbf{R}_i)}{\Psi_T(\mathbf{R}_i)}$$

$$\hat{H} = \sum_i^N -\frac{1}{2} \nabla_i^2 + \sum_{i>j}^N \frac{1}{|r_i - r_j|} - \sum_i \sum_{\alpha} \frac{Z_{\alpha}}{|r_i - R_{\alpha}|} + \sum_{\alpha\beta} \frac{Z_{\alpha}}{|R_{\alpha} - R_{\beta}|}$$

$$\Psi_T(\mathbf{R}_i) = D^{\uparrow} D^{\downarrow} \exp \left[ \sum_i^N \chi(r_i) - \sum_{i<j}^N u(r_{ij}) \right]$$

# Elements of the determinant

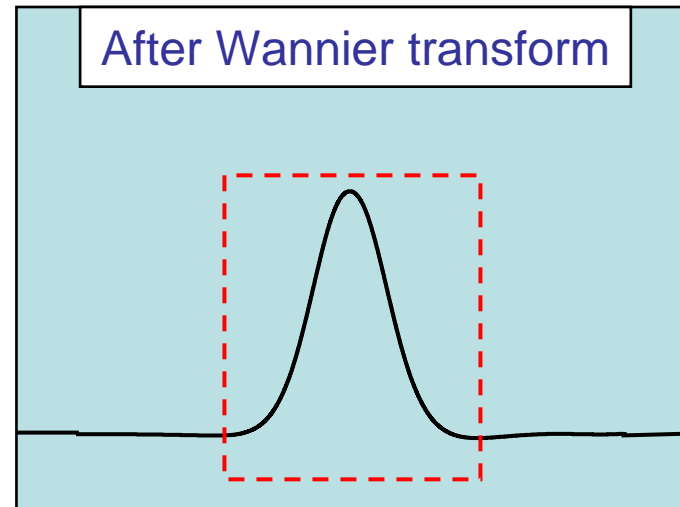
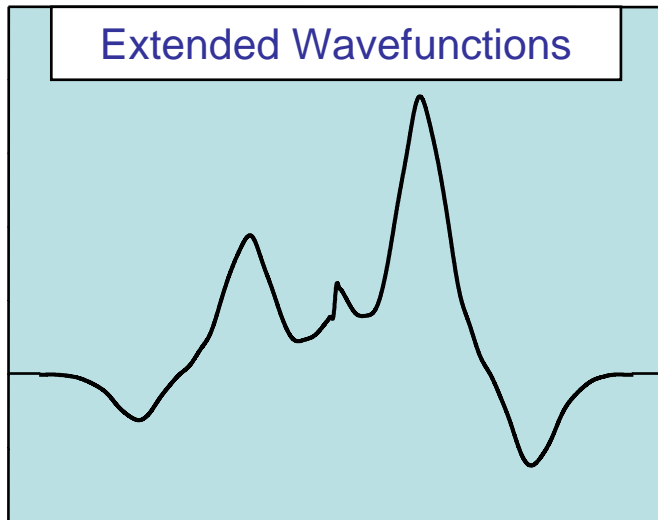
$$\Psi = \underbrace{\begin{pmatrix} \phi_1(\mathbf{r}_1) & \phi_1(\mathbf{r}_2) & \dots \\ \phi_2(\mathbf{r}_1) & \phi_2(\mathbf{r}_2) & \dots \\ \vdots & \vdots & \ddots \\ & & \phi_N(\mathbf{r}_N) \end{pmatrix}}_{\substack{\text{Slater Determinant} \\ O(N^3)}} \cdot \underbrace{\exp \left[ \sum_i^N \chi(\mathbf{r}_{iI}) - \sum_i^N u(\mathbf{r}_{ij}) \right]}_{\substack{\text{Jastrow correlation function} \\ O(N^2)}}$$

$N^3$  scaling for all the elements of determinant

- N electrons
- N orbitals,  $\phi$ , for each electron
- $O(N)$  basis functions in each orbital



# Localized Wannier functions



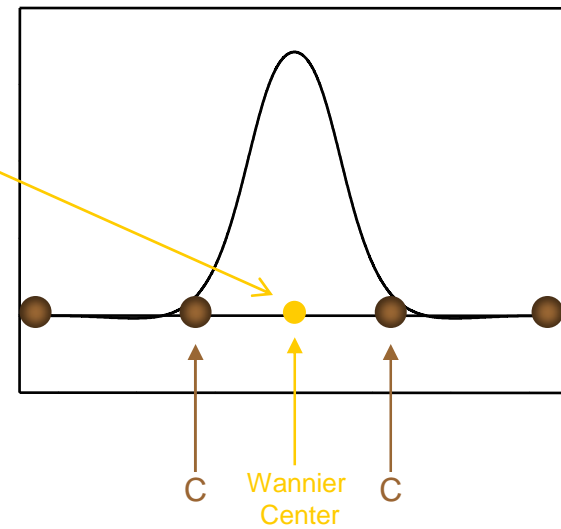
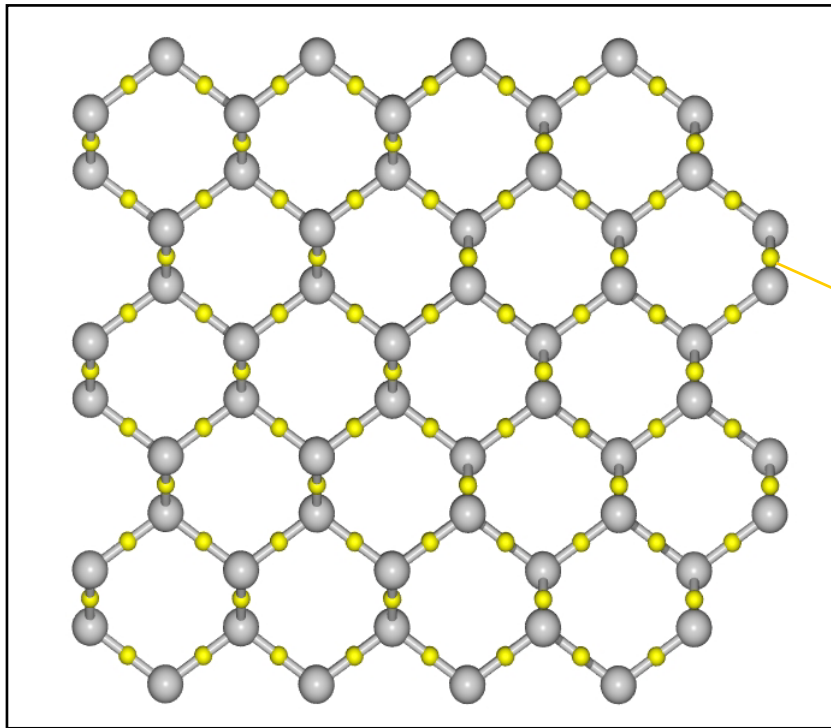
1. Apply Unitary Transform to DFT wavefunctions

2. Truncate Wannier states to reduce computational cost

$$\phi_{\text{Wannier}}^j = \sum_i U_{ij} \phi_{\text{LDA}}^i$$

Minimize  $\left[ \langle (\mathbf{r} - \mathbf{R}_n)^2 \rangle \right]^{\frac{1}{2}}$  N. Mazari and D. Vanderbilt, Phys. Rev. B **56**, 12847 (1997)

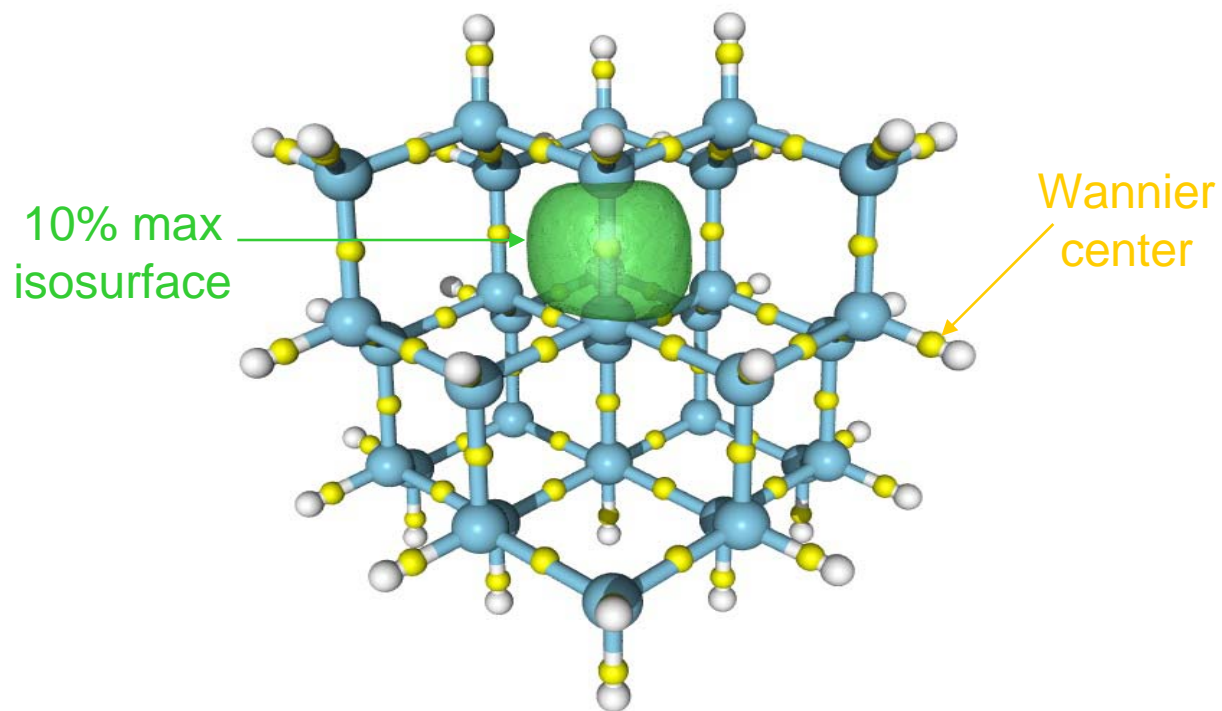
# Wannier functions in bulk diamond



- Two Wannier functions per atom
- Located in the center of the C-C bonds

# Wannier functions in $\text{Si}_{35}\text{H}_{36}$

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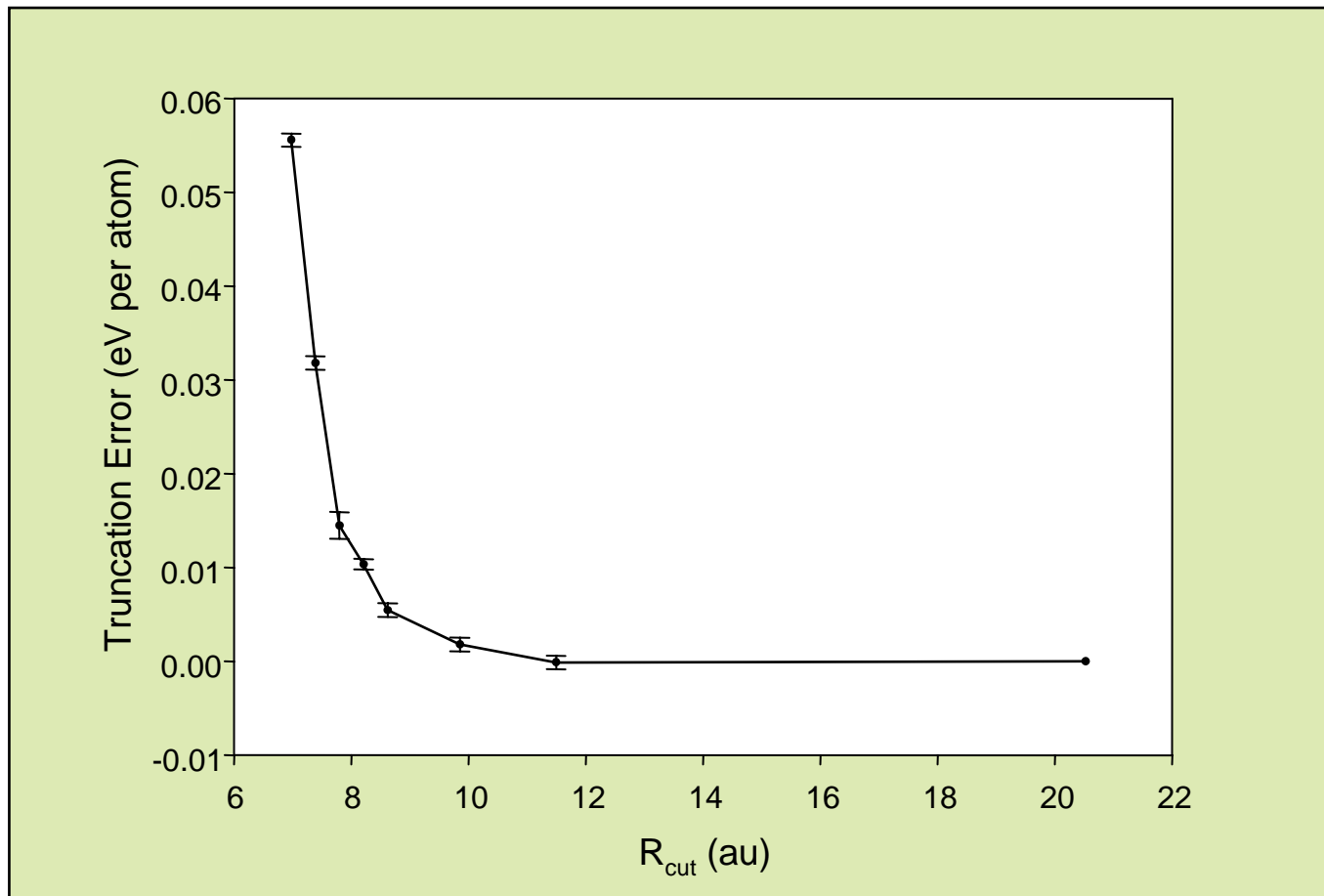


Wannier functions provide chemical insight.

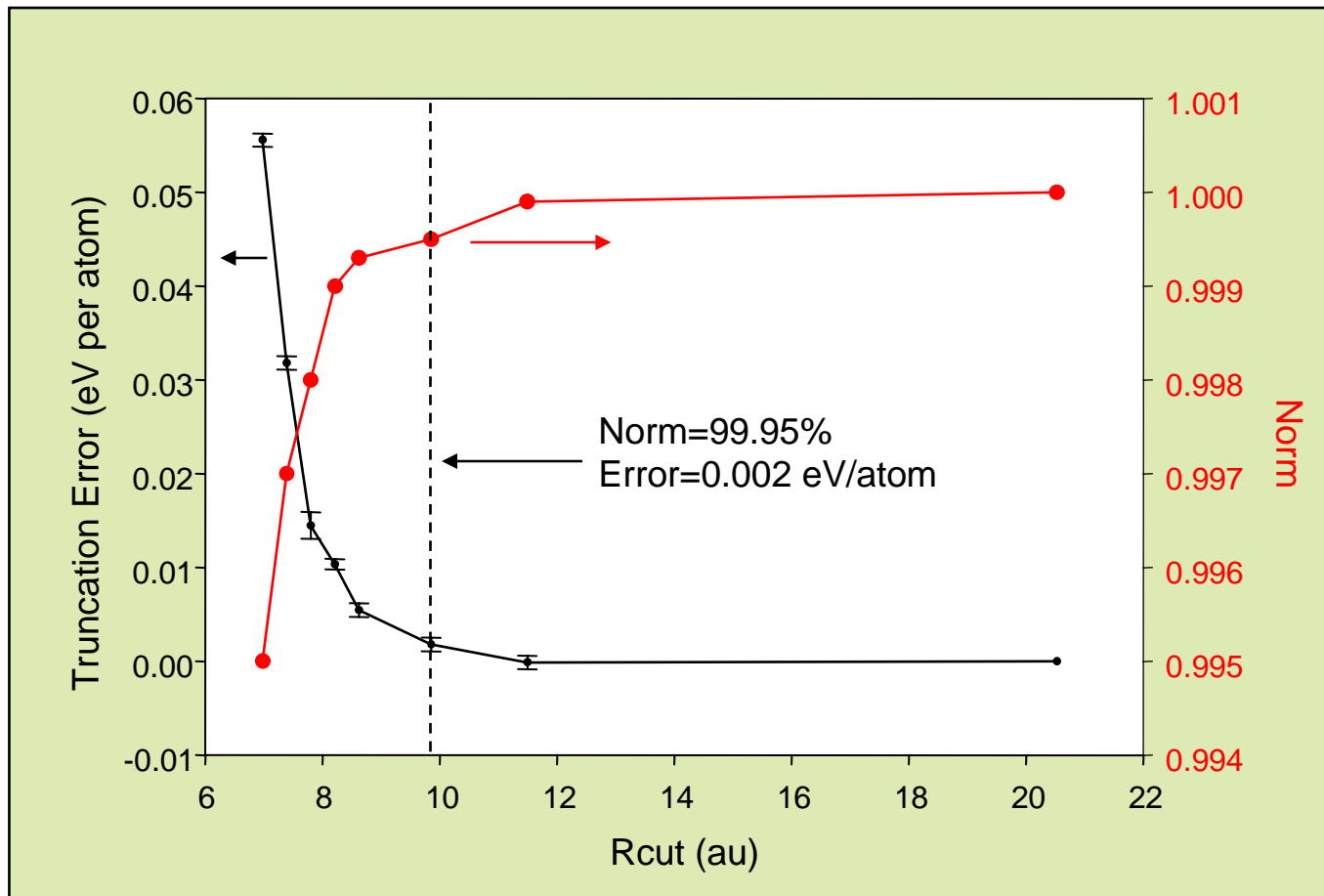


# Effect of truncation in $\text{SiH}_4$

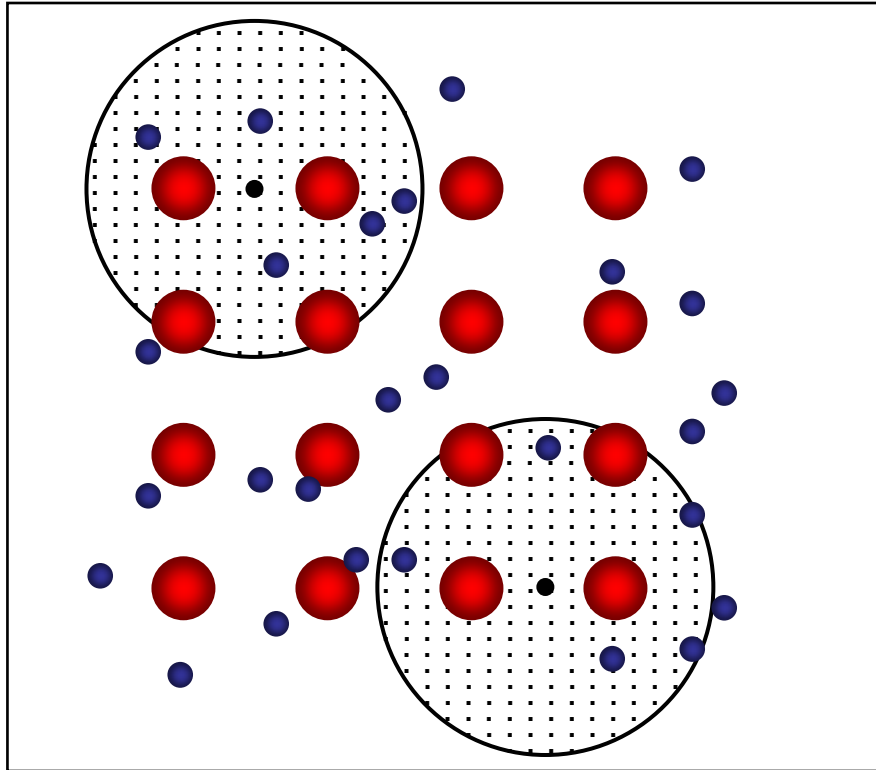
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# How can we predict a good $R_{\text{cut}}$ ?



# Real space grid for Wannier functions



- Take advantage of the *localized* nature of the Maximally Localized Wannier orbitals - expand in a *localized* basis.
- Could use Gaussians – we find real space grid to be superior.
- $O(N)$  memory requirement for the spline grids.

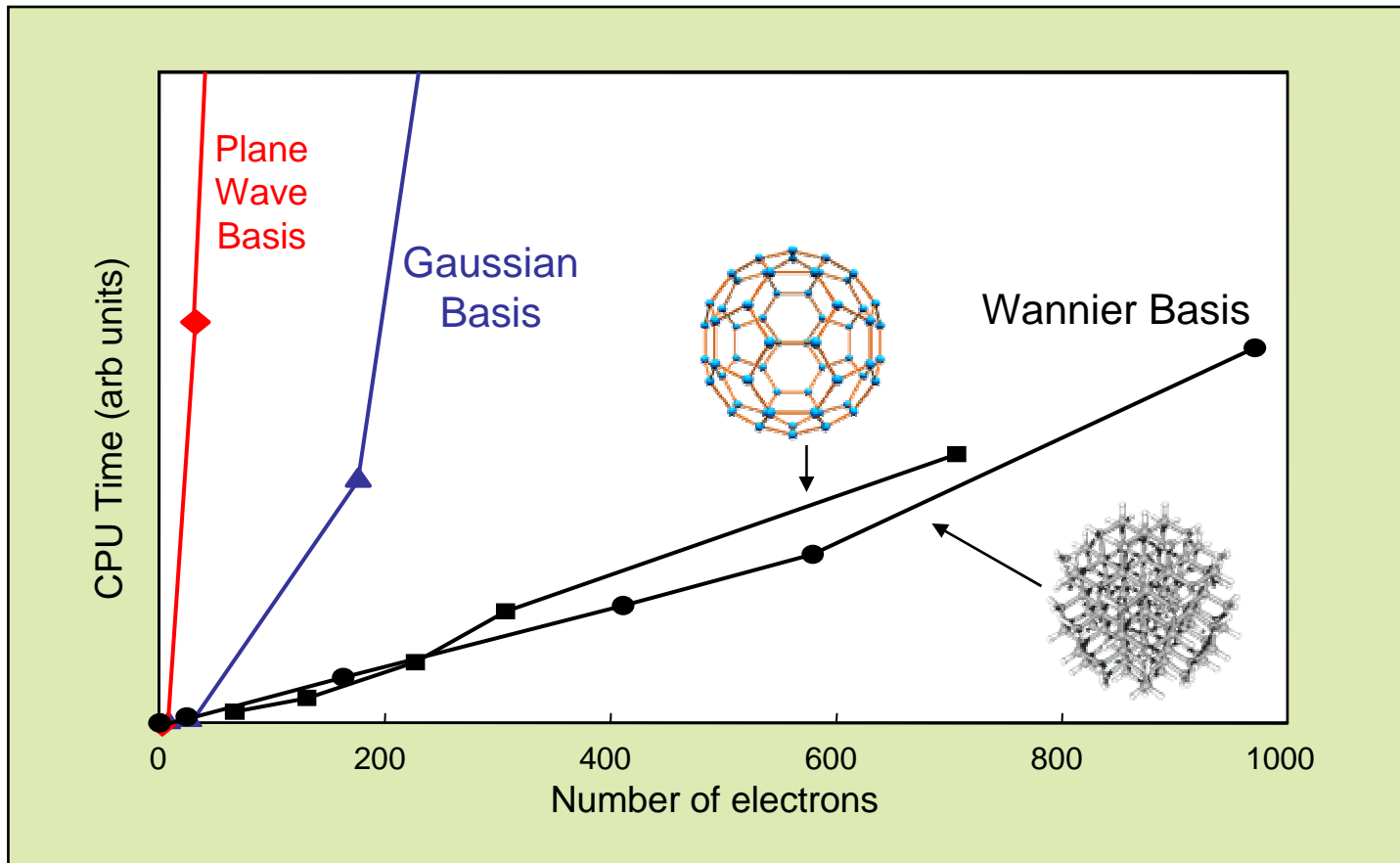
# $N^3 \rightarrow N$ scaling

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	Original $N^3$ Scaling	New $O(N)$ Scaling
Number of electrons	$N$	$N$
Number of orbitals per electron	$N$	const.
Number of basis functions per orbital	$\propto N$	const.



# Near Linear Scaling Achieved



A.J. Williamson, R. Hood, and J. Grossman, Phys. Rev. Lett. **87**, 246406 (2001)

# How linear is it ?

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- For 1000 electrons, 10% is spent in  $N^2$  and  $N^3$  routines
- Need to work on these beyond ~2000 electrons

## Remaining $N^3$ and $N^2$ terms:

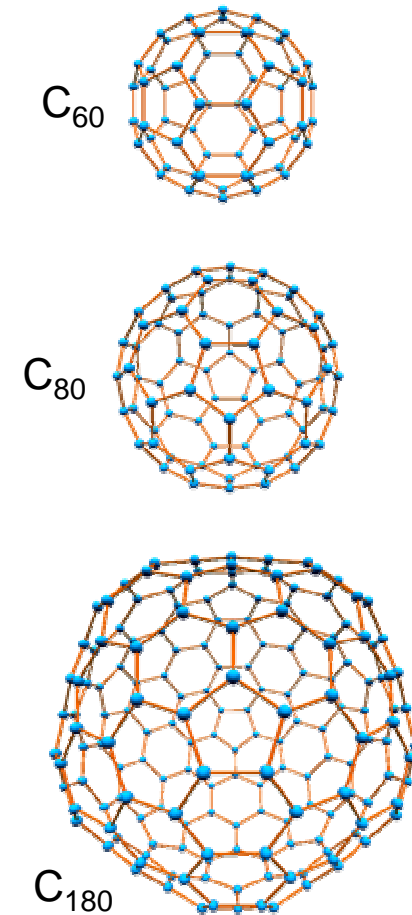
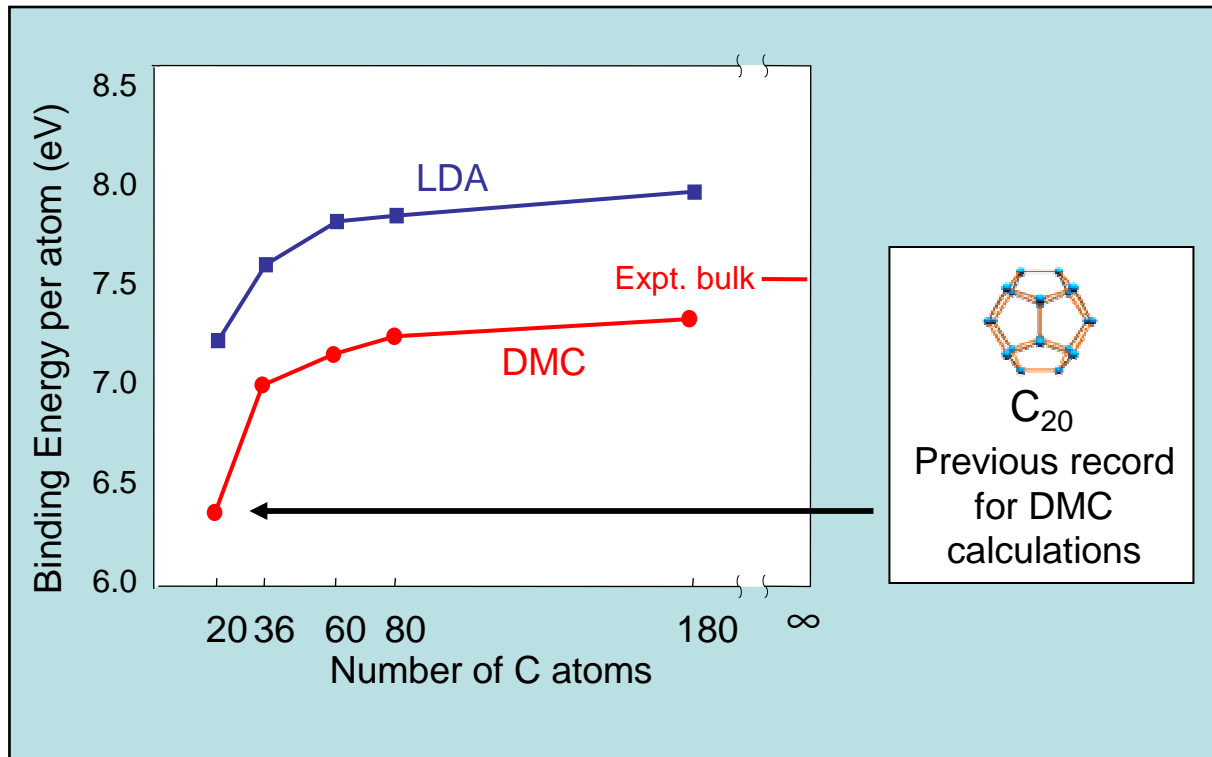
- Calculating the value of the determinant ( $N^3$ )
- Calculating electron-electron and electron-ion distances ( $N^2$ )

# Scaling of Actual Calculations

$$E_{\text{mean}} = \frac{1}{N_C} \sum_i^{N_C} \frac{\hat{H} \Psi_T(\mathbf{R}_i)}{\Psi_T(\mathbf{R}_i)} \quad \sigma_{\text{mean}} = \frac{\sigma_{\text{intrinsic}}}{\sqrt{N_C}} \rightarrow \propto \sqrt{N}$$

	$N_C$	Cost per config.	Total Cost	Original cost
<b>Total Energy</b> (Optical gaps)	$\propto N$	$N$	$\propto N^2$	$\propto N^4$
<b>Per atom</b> (Binding energies)	$\propto N^{-1}$	$N$	$\propto N^0$	$\propto N^2$

# Binding Energy of Carbon Fullerenes



- More accurate binding energies for fullerenes
- Cost of DMC calculation is independent of size!

# A work in progress....

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- O(N) evaluation of the *value* of the determinant.
- Can this apply to metals?
- Are non-orthogonal Wannier functions useful?

$$\varphi_{\text{Wannier}}^j = \sum_i U_{ij} \varphi_{\text{LDA}}^i$$

# How is this different to $O(N)$ density functional codes?

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- Similar to introducing sparsity into the overlap matrix in density functional approaches, but
- No self-consistency required:
  - No conservation of charge problem
  - No noise introduced into the Hamiltonian
- Inverting overlap matrix introduces large overhead for linear scaling in DFT, here there is no additional overhead.

# Summary

- A simple transformation of the input single particle orbitals yields localized orbitals which can be truncated to introduce sparsity into the Slater determinant.
- These truncated orbitals can be stored on a cubic spline grid.
- Resulting algorithm scales nearly linearly up to 1000 electrons.
- Successfully applied to fullerenes, silicon clusters and bulk diamond.

