

Linear Scaling BigDFT

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Engineering and
Physical Sciences
Research Council

Outline

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Motivation

The Formalism

Support Functions

Density Kernel

Overall Algorithm

LS-BigDFT in Action

Scaling

Accuracy

Applications

1 Motivation

2 The Formalism

Support Functions

Density Kernel

Overall Algorithm

3 LS-BigDFT in Action

Scaling

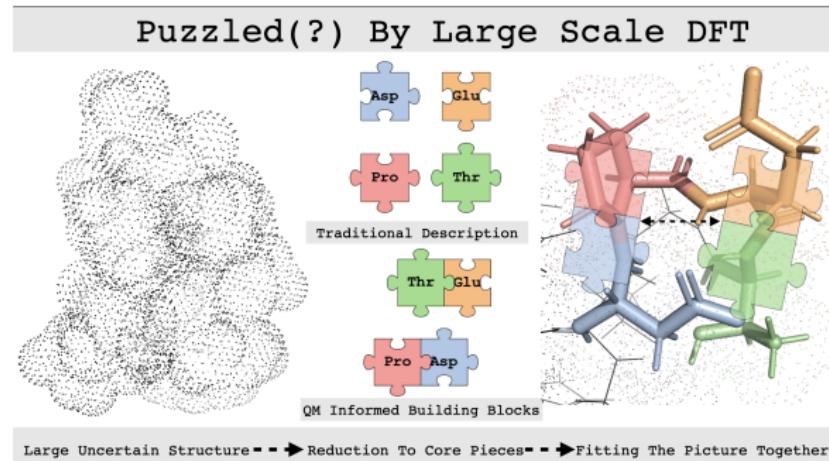
Accuracy

Applications

Why DFT with 1000s of Atoms?

Why do we Need QM for Large Systems?

- bridge lengthscale gap between QM and MM (validate empirical models, larger QM region etc)
- intrinsically QM quantities, e.g. electronic excitations
- new possibilities for simulating complex materials
- new regime for DFT → new ways of working



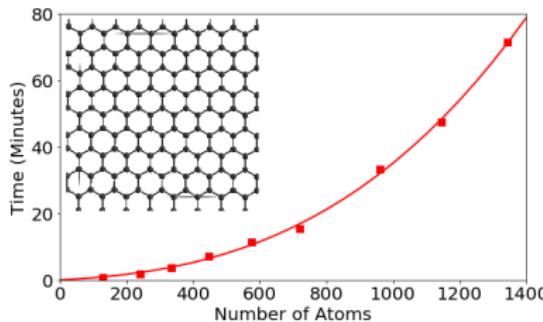
Large Scale QM Reviews: LER, Mohr, Huhs, Deutsch, Masella & Genovese WIREs Comput. Mol. Sci. 7, e1290 (2017); Dawson, Degomme, Stella, Nakajima, LER & Genovese WIREs Comput. Mol. Sci. 12, e1574 (2022)

The Cubic Scaling Limit

Scaling with System Size

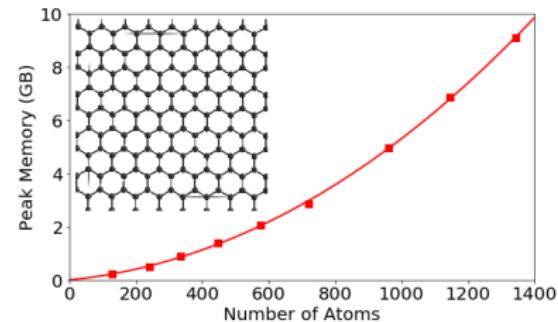
the operations **scale** differently:

- $\mathcal{O}(\mathcal{N} \log \mathcal{N})$: Poisson solver
 - $\mathcal{O}(\mathcal{N}^2)$: convolutions
 - $\mathcal{O}(\mathcal{N}^3)$: linear algebra
- and have different **prefactors**:
- $c_{\mathcal{O}(\mathcal{N}^3)} \ll c_{\mathcal{O}(\mathcal{N}^2)} \ll c_{\mathcal{O}(\mathcal{N} \log \mathcal{N})}$



Size Limitations

- up to ~ 1000 atoms thanks to wavelet properties and efficient **parallelisation**
- for bigger systems $\mathcal{O}(\mathcal{N}^3)$ **dominates**
- also reach memory limits
→ need a **new approach**



Nearsightedness and Locality

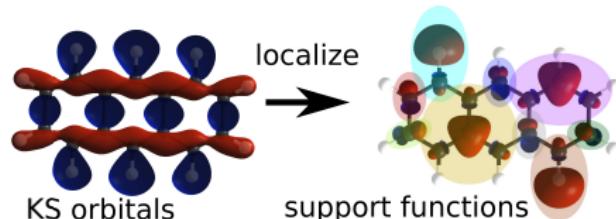
Nearsightedness

- the behaviour of large systems is **short-ranged** (nearsighted)
 - the density matrix, $\rho(\mathbf{r}, \mathbf{r}')$, decays exponentially in systems with a gap
- how can we exploit nearsightedness to treat large systems?

Support Functions (SFs)

write **extended KS orbitals** in terms of **localised SFs** ($\{\phi_\alpha(\mathbf{r})\}$):

$$\Psi_i(\mathbf{r}) = \sum_{\alpha} c_i^{\alpha} \phi_{\alpha}(\mathbf{r})$$



Density Matrix Formulation

Avoiding Diagonalisation

- express $\rho(\mathbf{r}, \mathbf{r}')$ in terms of **density kernel** $K^{\alpha\beta}$ and **SFs**:

$$\begin{aligned}\rho(\mathbf{r}, \mathbf{r}') &= \sum_i f_i |\Psi_i(\mathbf{r})\rangle\langle\Psi_i(\mathbf{r}')| \\ &= \sum_{\alpha, \beta} |\phi_\alpha(\mathbf{r})\rangle K^{\alpha\beta} \langle\phi_\beta(\mathbf{r}')|\end{aligned}$$

- no explicit reference to KS orbitals – avoid diagonalisation
 - orthogonality requirement translates to idempotency:
- $$\langle\Psi_i|\Psi_j\rangle = \delta_{ij} \rightarrow \mathbf{K} = \mathbf{KSK}$$
- the density is found via $n(\mathbf{r}) = \rho(\mathbf{r}, \mathbf{r})$

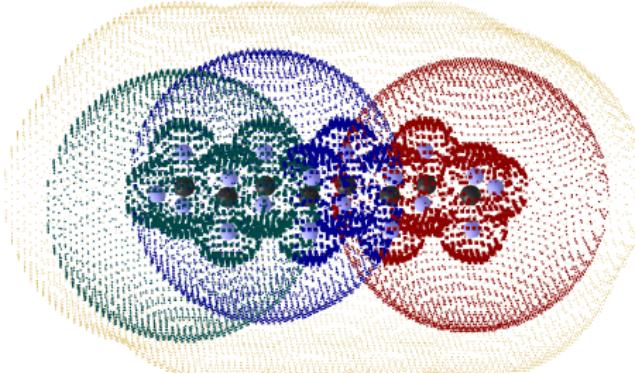
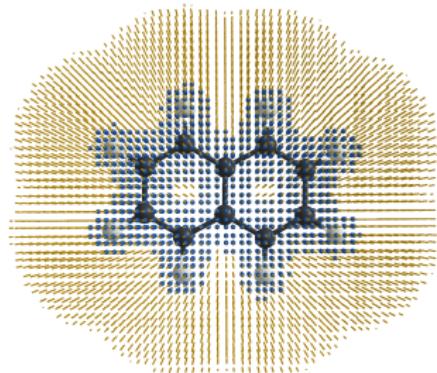
Matrix Expressions

$$\begin{aligned}H_{\alpha\beta} &= \langle\phi_\alpha|\hat{H}|\phi_\beta\rangle; & S_{\alpha\beta} &= \langle\phi_\alpha|\phi_\beta\rangle \\ E &= \text{Tr}(\mathbf{KH}); & N &= \text{Tr}(\mathbf{KS})\end{aligned}$$

Support Functions

SF Properties

- atom-centred
- strictly localised ($\sim 6 - 8 a_0$ radius) – user defined (systematic convergence)
- numerical functions expanded in wavelets \rightarrow 2 levels of basis ('contracted wavelets')
- (typically) minimal (1 SF per H, 4 per C/N/O...)
- quasi-orthogonal



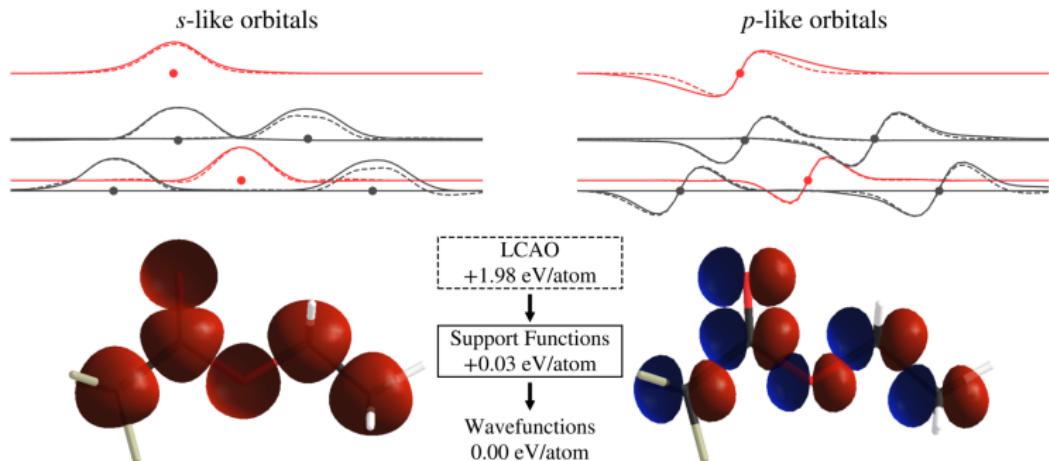
Support Function Optimisation

How to Have an Accurate Minimal Basis?

- start with atomic orbital guess for the SFs
- use a gradually decreasing confining potential:

$$\hat{H}_\alpha = \hat{H} + c_\alpha(\mathbf{r} - \mathbf{R}_\alpha)^4$$

- optimise SFs *in situ* by minimising E (subject to constraints)
→ minimal, localised basis with high accuracy of wavelets



Density Kernel Optimisation

Three Methods for Obtaining \mathbf{K} :

- **Diagonalisation**

- good for small systems and benchmarking/debugging

- **Direct Minimisation**

- work directly with the KS coefficients
- can include (low energy) unoccupied states

- **Fermi Operator Expansion (FOE)**

- density matrix is expressed in terms of \mathbf{H} , i.e. $\mathbf{K} = f(\mathbf{H})$
- use a Chebyshev polynomial expansion
- uses a (small) finite temperature – works for metals
- implemented in CHESS (Chebyshev Sparse Solvers) library

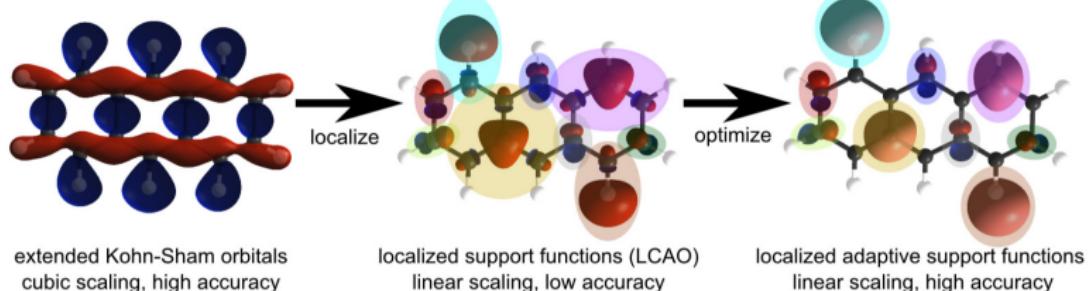
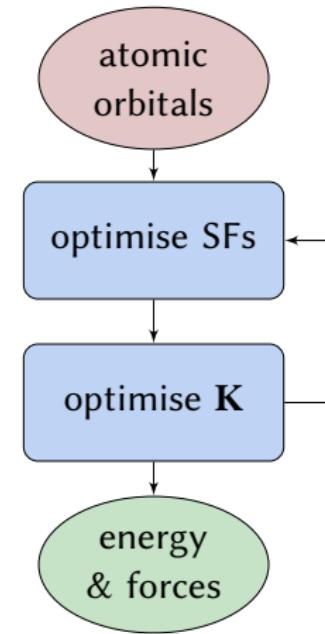
Kernel Truncation

- simple distance criterion $K_{\alpha\beta} = 0$ if $|R_\alpha - R_\beta| > K_{\text{cut}}$

The Algorithm

Calculation Steps

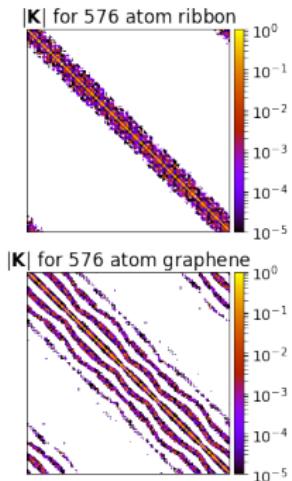
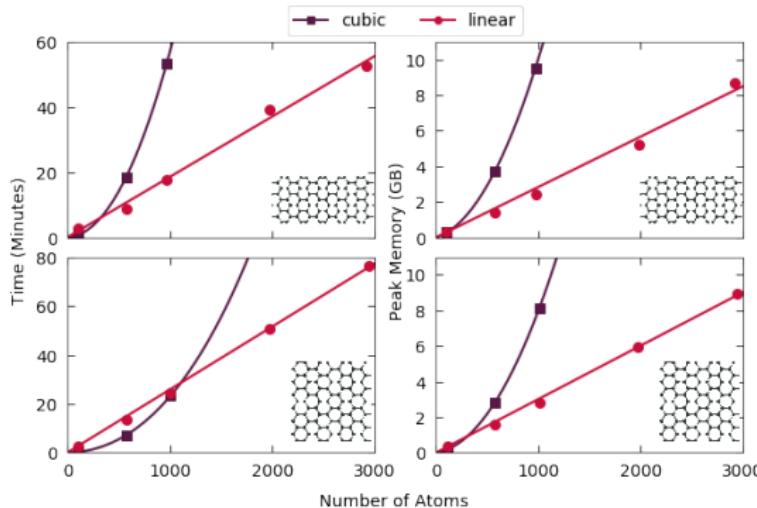
- minimise energy with respect to **both SFs and K**
- **accurate forces** – geometry optimisations, MD
- one-off diagonalisation for (occupied) KS energies → densities of states



From Sparsity to Linear Scaling

Sparse Matrices

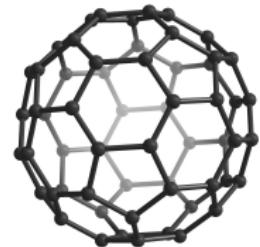
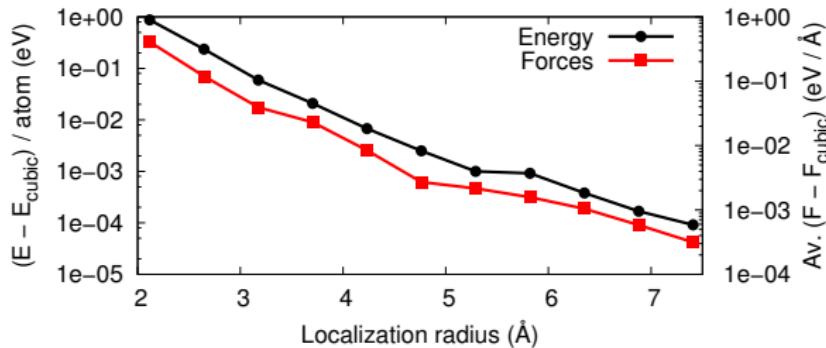
- strict localisation leads to sparse matrices
- sparsity depends on size, dimensionality, SF radii → crossover
- performance also depends on band gap



Accuracy

Controlling the Accuracy

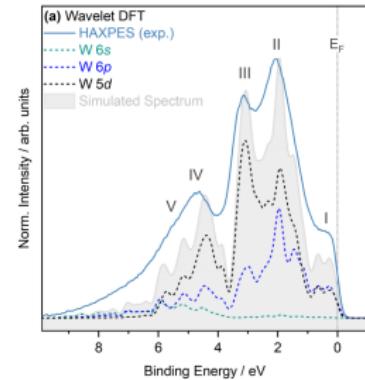
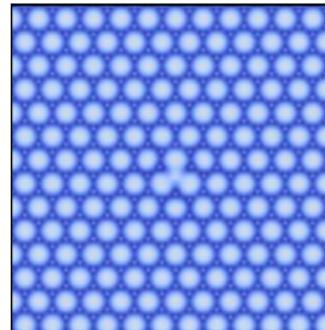
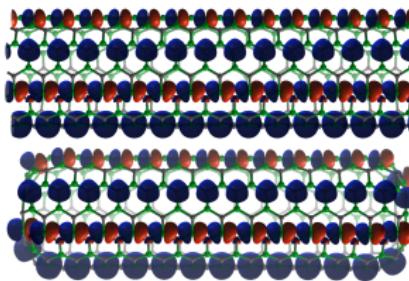
- SF localisation radii are the key parameters
- can choose different values for different atomic species
- can also use **robust profiles**
- energies and forces converge to cubic scaling values



Applicability

Functionalities

- same structure as cubic scaling code – just import a profile
- same range of boundary conditions
- not all functionalities available in LS-BigDFT: hybrid functionals, TDDFT...



SiC Nanotubes: LER & Genovese, J. Phys.: Condens. Matter **31**, 285901 (2019)

Defective Graphene LER & Genovese, in Theory and Simulation in Physics for Materials Applications, edited by Levchenko, Dappe & Ori, Springer International Publishing (2020)

Bulk W: Kalha, LER, Gutiérrez Moreno, Mohr, Mansinen, Fernando, Thakur, Lee, Tseng, Nunney, Kahk, Lischner & Regoutz, Phys. Rev. B **105**, 045129 (2022)

Simulating Thousands of Atoms

Beyond 50,000 Atoms

- DNA:
 - LS behaviour for many thousands of atoms
- Covid spike:
 - LS-BigDFT with implicit solvent
 - ~52,000 atoms in ~6 hours on 128 nodes (16,384 cores)
 - new opportunities for analysis...

