

Formalism

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Poisson Solver

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Electronic Structure Calculation Methods on Accelerators, Oak Ridge

Daubechies wavelets as a basis set for density functional pseudopotential calculation

T. Deutsch, L. Genovese, D. Caliste

L_Sim – CEA Grenoble

February 7, 2012

A basis for nanosciences: The BigDFT project

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BigDFT

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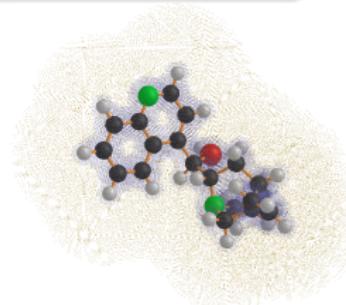
STREP European project: BigDFT(2005-2008)

Four partners, 15 contributors:

CEA-INAC Grenoble (T. D.), U. Basel (S. Goedecker),
U. Louvain-la-Neuve (X. Gonze), U. Kiel (R. Schneider)

Aim: To develop an ab-initio DFT code
based on **Daubechies Wavelets**
and *integrated in ABINIT (Gnu-GPL)*.

BigDFT 1.0 → January 2008



... Why have we done this?

- Test the potential advantages of a new formalism
- A lot of outcomes and interesting results
- A lot can be done starting from present know-how

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Next talk: Performances (MPI+OpenMP+OpenCL)

A DFT code based on Daubechies wavelets

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BigDFT: a PSP Kohn-Sham code

A Daubechies wavelets basis has unique properties for DFT usage

- Systematic, Orthogonal
- Localised, Adaptive
- Kohn-Sham operators are **analytic**

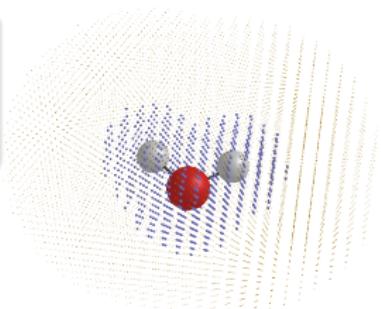
Short, Separable convolutions

$$\tilde{c}_\ell = \sum_j a_j c_{\ell-j}$$

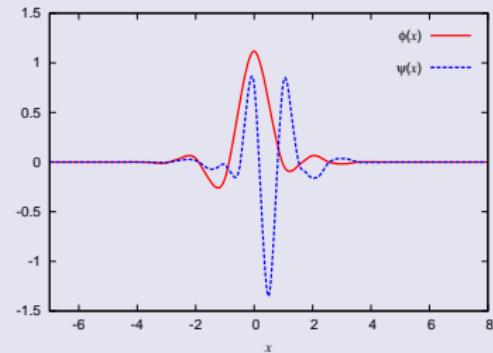
- Peculiar numerical properties

Real space based, highly flexible

Big & inhomogeneous systems



Daubechies Wavelets



A brief description of wavelet theory

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Two kind of basis functions

A Multi-Resolution real space basis

The functions can be classified following the resolution level they span.

Scaling Functions

The functions of low resolution level are a linear combination of high-resolution functions

The diagram shows a sequence of black dots representing a signal. A blue rectangular pulse is centered on the first dot. This pulse is followed by an equals sign. To the right of the equals sign, there are two terms separated by a plus sign. The first term is a red rectangular pulse centered on the third dot, with a label h_j above it. The second term is another red rectangular pulse centered on the fifth dot. This visualizes the equation:

$$\phi(x) = \sum_{j=-m}^m h_j \phi(2x - j)$$

Centered on a **resolution-dependent** grid: $\phi_j = \phi_0(x - j)$.

A brief description of wavelet theory

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Wavelets

They contain the DoF needed to complete the information which is lacking due to the coarseness of the resolution.

$$\dots \bullet \bullet \bullet = \frac{1}{2} \bullet \bullet + \frac{1}{2} \bullet \bullet \bullet$$

$$\phi(2x) = \sum_{j=-m}^m \tilde{h}_j \phi(x-j) + \sum_{j=-m}^m \tilde{g}_j \psi(x-j)$$

Increase the resolution without modifying grid space

SF + W = same DoF of SF of higher resolution

$$\psi(x) = \sum_{j=-m}^m \textcolor{red}{g}_j \phi(2x-j)$$

All functions have compact support, centered on grid points.

Wavelet properties: Adaptivity

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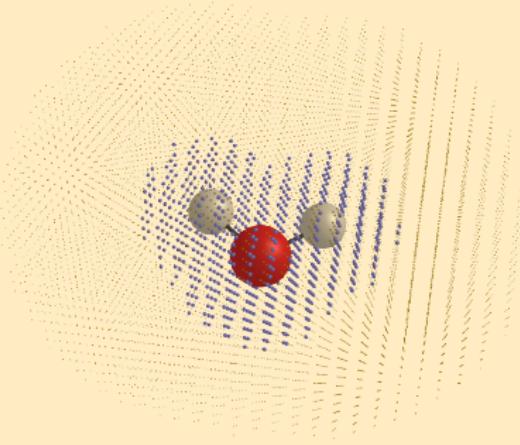
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Adaptivity

Resolution can be refined following the grid point.



The grid is divided in **Low** (1 DoF) and **High** (8 DoF) resolution points. Points of different resolution belong to **the same** grid. Empty regions must not be “filled” with basis functions.

Localization property, real space description

Optimal for big & inhomogeneous systems, highly flexible

Adaptivity of the mesh

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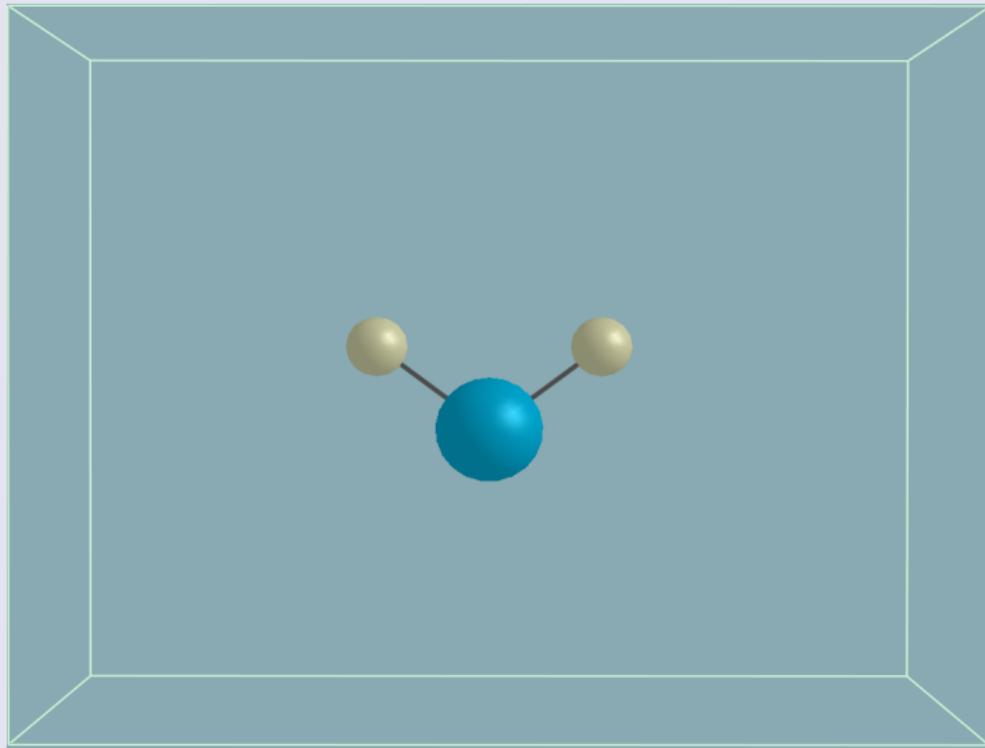
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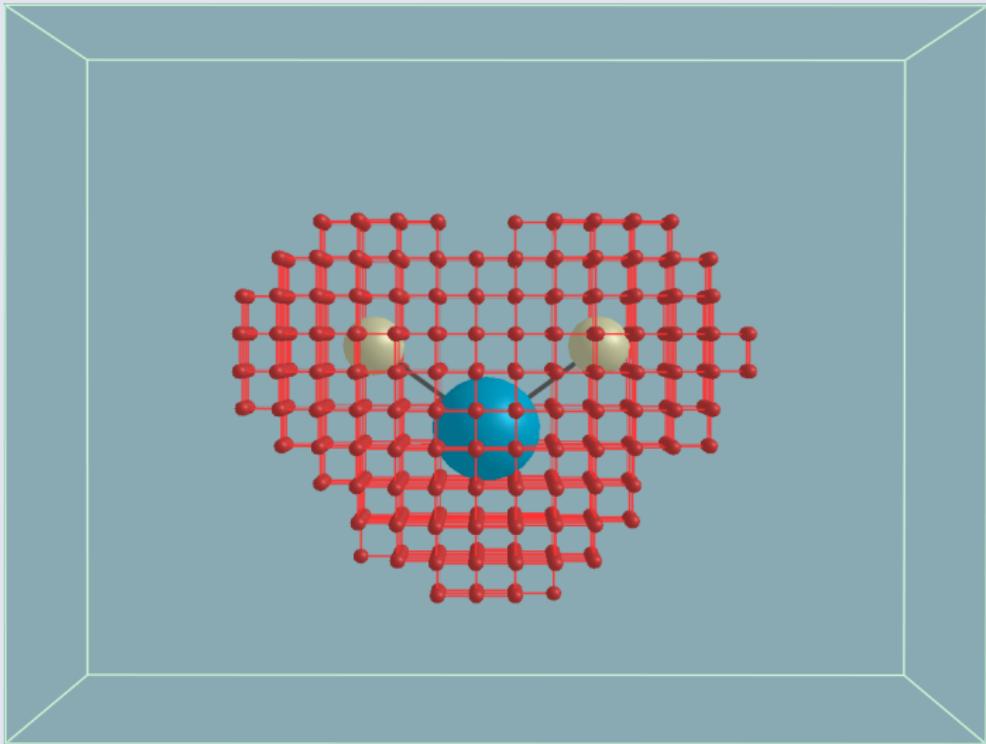
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Atomic positions (H_2O), *hgrid*



Adaptivity of the mesh

Fine grid (high resolution, `frmult`)



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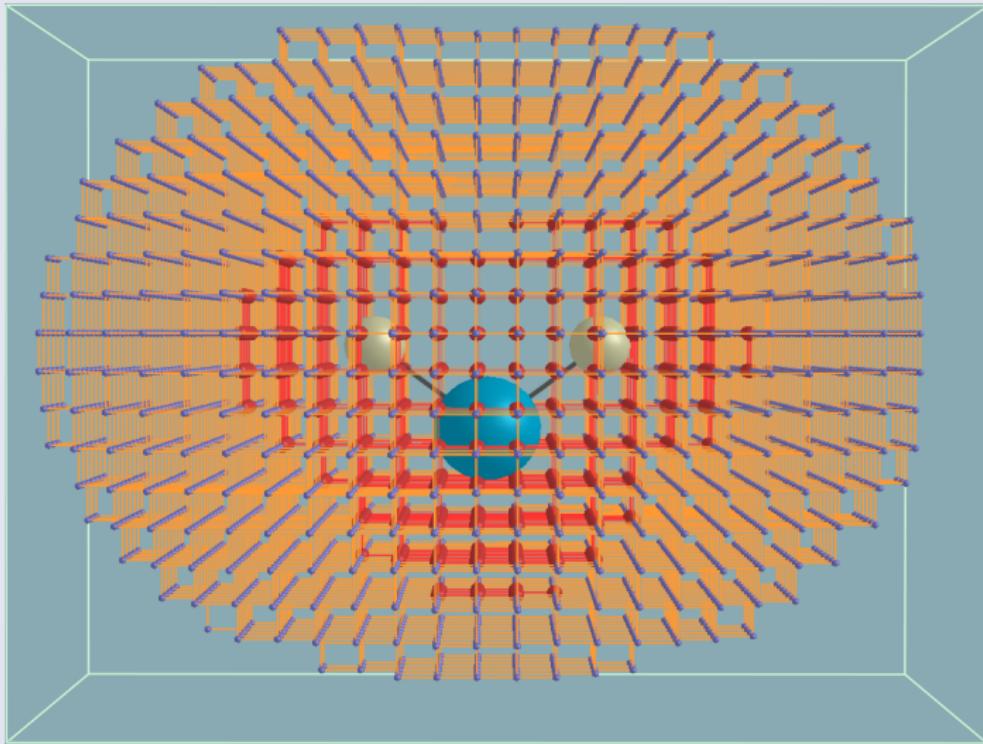
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Adaptivity of the mesh

Coarse grid (low resolution, `cramult`)



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BIG
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No integration error

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Orthogonality, scaling relation

$$\int dx \phi_k(x) \phi_j(x) = \delta_{kj} \quad \phi(x) = \frac{1}{\sqrt{2}} \sum_{j=-m}^m h_j \phi(2x - j)$$

The hamiltonian-related quantities can be calculated *up to machine precision in the given basis*.

The accuracy is only limited by the basis set ($O(h_{\text{grid}}^{14})$)

Exact evaluation of kinetic energy

Obtained by convolution with filters:

$$f(x) = \sum_{\ell} c_{\ell} \phi_{\ell}(x), \quad \nabla^2 f(x) = \sum_{\ell} \tilde{c}_{\ell} \phi_{\ell}(x),$$

$$\tilde{c}_{\ell} = \sum_j c_j \mathbf{a}_{\ell-j}, \quad \mathbf{a}_{\ell} \equiv \int \phi_0(x) \partial_x^2 \phi_{\ell}(x),$$

Separability in 3D

The 3-dim scaling basis is a *tensor product decomposition* of 1-dim Scaling Functions/ Wavelets.

$$\phi_{j_x,j_y,j_z}^{e_x,e_y,e_z}(x,y,z) = \phi_{j_x}^{e_x}(x)\phi_{j_y}^{e_y}(y)\phi_{j_z}^{e_z}(z)$$

With (j_x, j_y, j_z) the node coordinates,
 $\phi_j^{(0)}$ and $\phi_j^{(1)}$ the SF and the W respectively.

Gaussians and wavelets

The separability of the basis allows us to save computational time when performing scalar products with separable functions (e.g. gaussians):

- Initial wavefunctions (input guess)
- Poisson solver (tensor decomposition of $\frac{1}{r}$)
- Non-local pseudopotentials

Wavelet families used in BigDFT code



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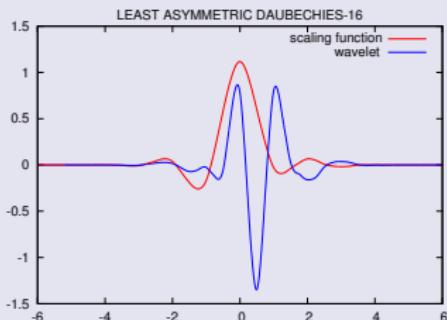
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Daubechies $f(x) = \sum_{\ell} c_{\ell} \phi_{\ell}(x)$

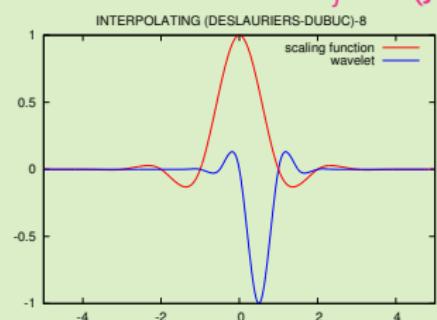
Orthogonal $c_{\ell} = \int dx \phi_{\ell}(x) f(x)$



Used for **wavefunctions**,
scalar products

Interpolating $f(x) = \sum_j f_j \phi_j(x)$

Dual to dirac deltas $f_j = f(j)$



Used for **charge density**,
function products

“Magic Filter” method (A. Neelov, S. Goedecker)

The **passage** between the two basis sets can be performed without **losing accuracy**.

Operations performed



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The SCF cycle

Orbital scheme:

- Hamiltonian
- Preconditioner

Coefficient Scheme:

- Overlap matrices
- Orthogonalisation

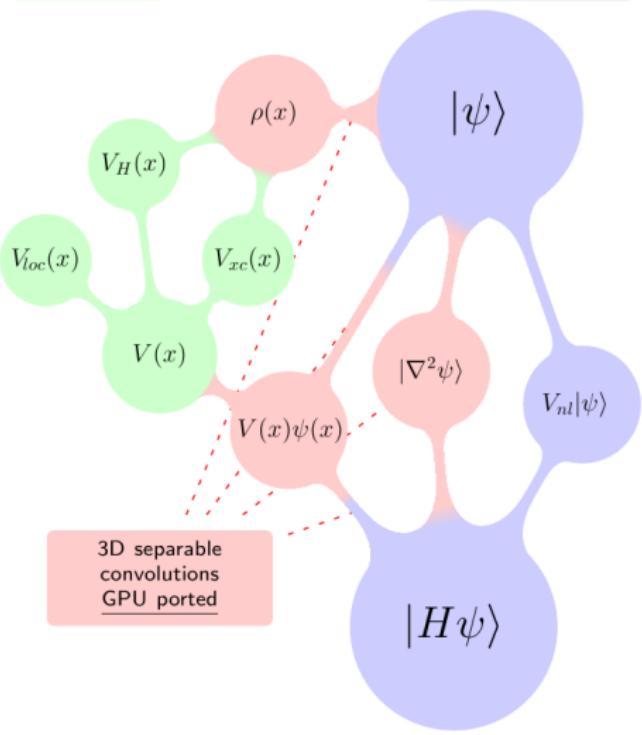
Comput. operations

- Convolutions
- BLAS routines
- FFT (Poisson Solver)

Why not GPUs?

Real Space

Daub. Wavelets



Kohn-Sham Equations: Computing Energies

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Calculate different integrals

$$E[\rho] = K[\rho] + U[\rho]$$

$$K[\rho] = -\frac{2 \hbar^2}{2 m_e} \sum_i \int_{\mathcal{V}} dr \psi_i^* \nabla^2 \psi_i$$

$$U[\rho] = \int_{\mathcal{V}} dr V_{ext}(r) \rho(r) + \underbrace{\frac{1}{2} \int_{\mathcal{V}} dr dr' \frac{\rho(r)\rho(r')}{|r-r'|}}_{Hartree} + \underbrace{E_{xc}[\rho]}_{exchange-correlation}$$

We minimise with the variables $\psi_i(r)$ and f_i

with the constraint $\int_{\mathcal{V}} dr \rho(r) = N_{el}$.

Direct Minimisation: Flowchart

$$\left\{ \psi_i = \sum_a c_a^i \phi_a \right\}$$

Basis: adaptive mesh ($0, \dots, N_\phi$)

Orthonormalized



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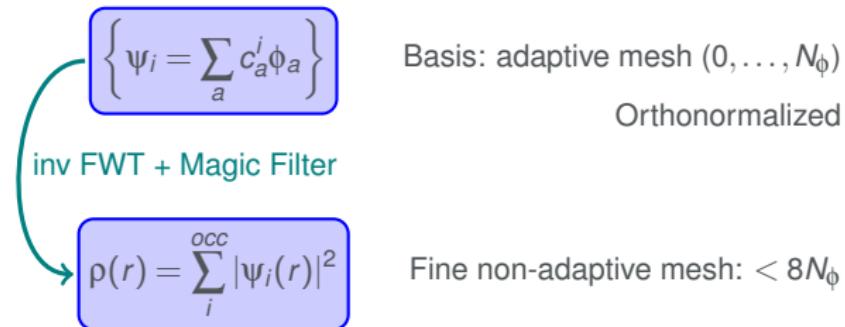
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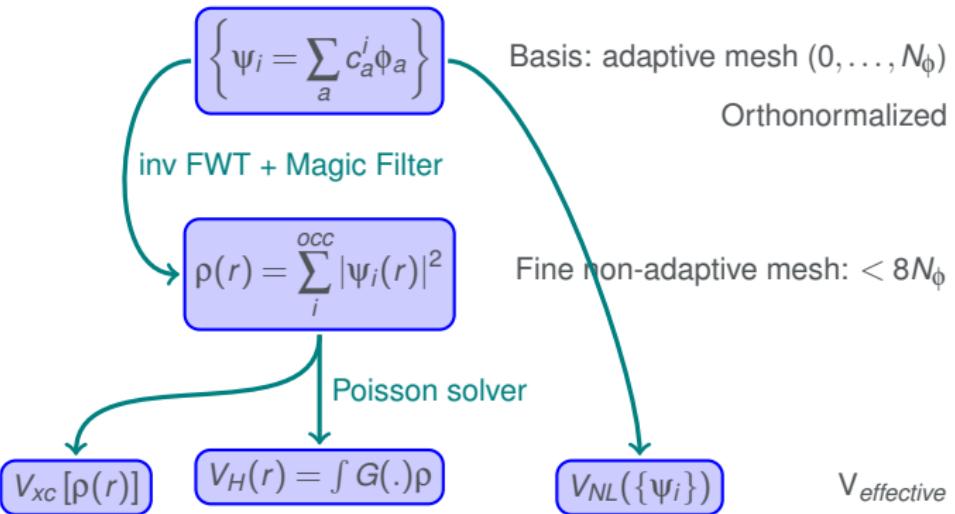
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Direct Minimisation: Flowchart



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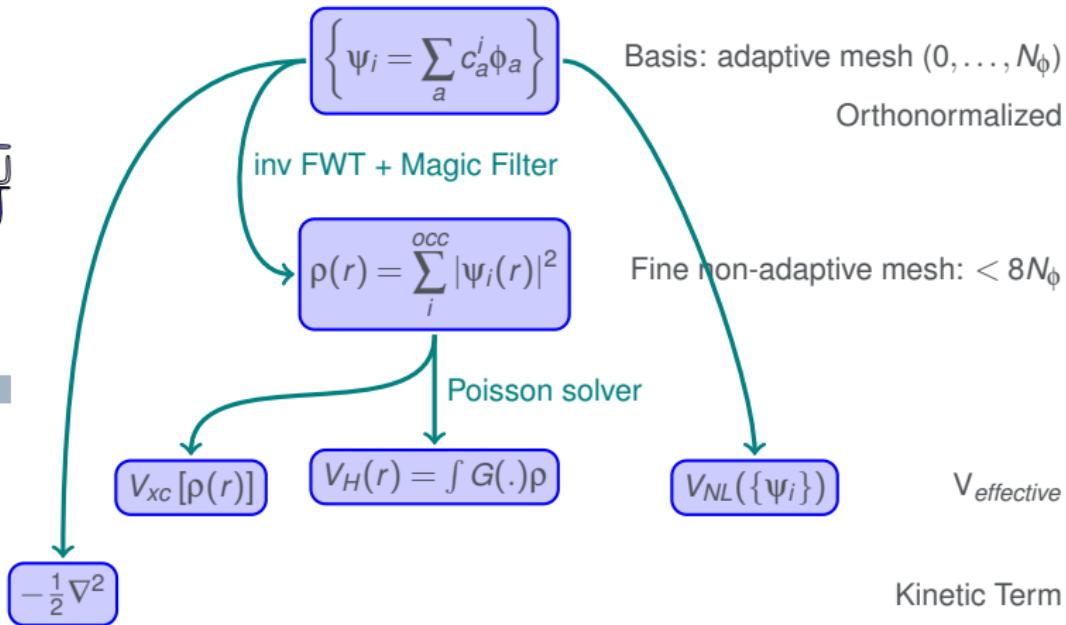
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Björn
Dierckx

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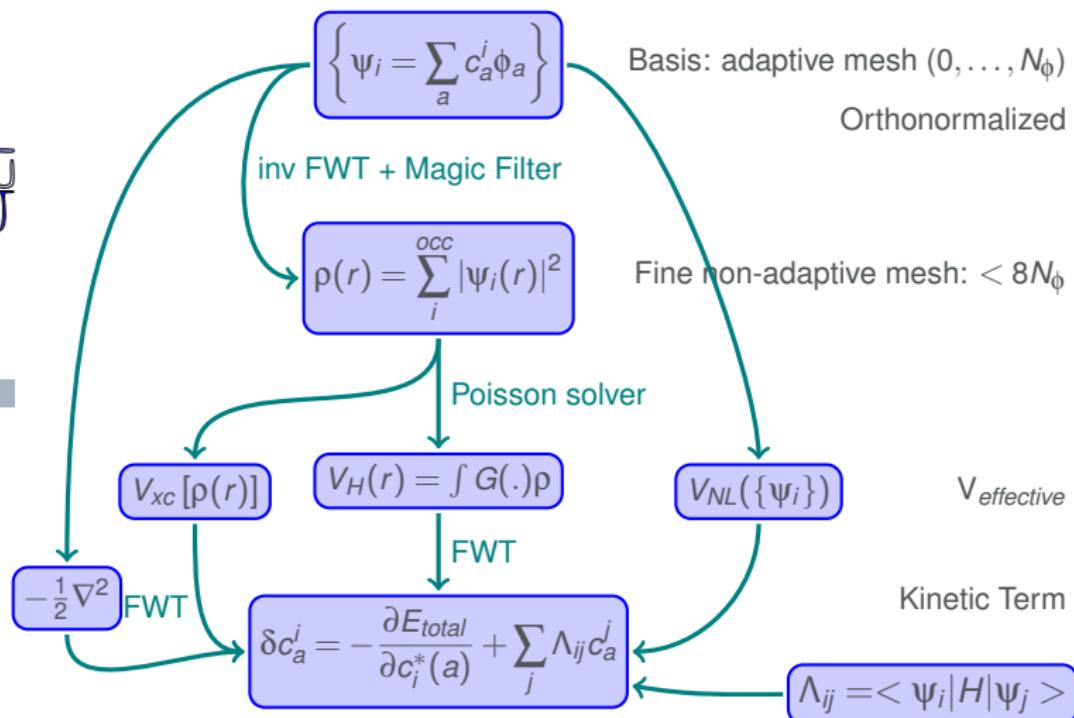
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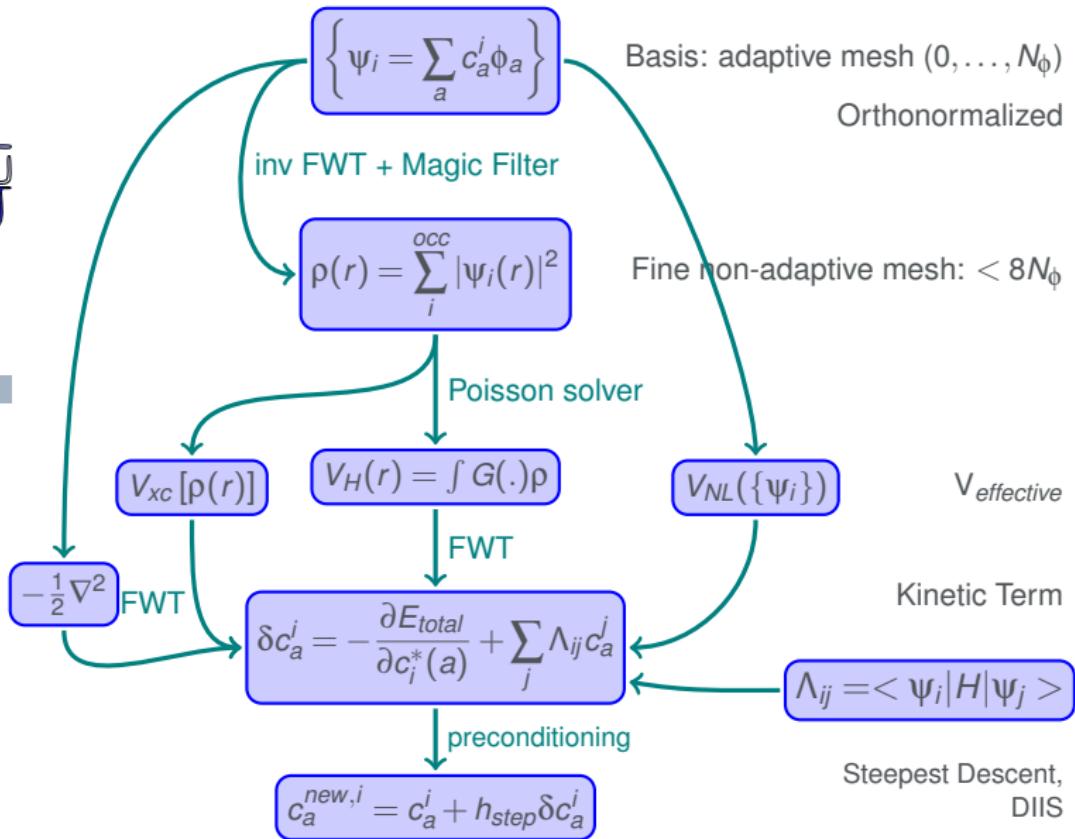
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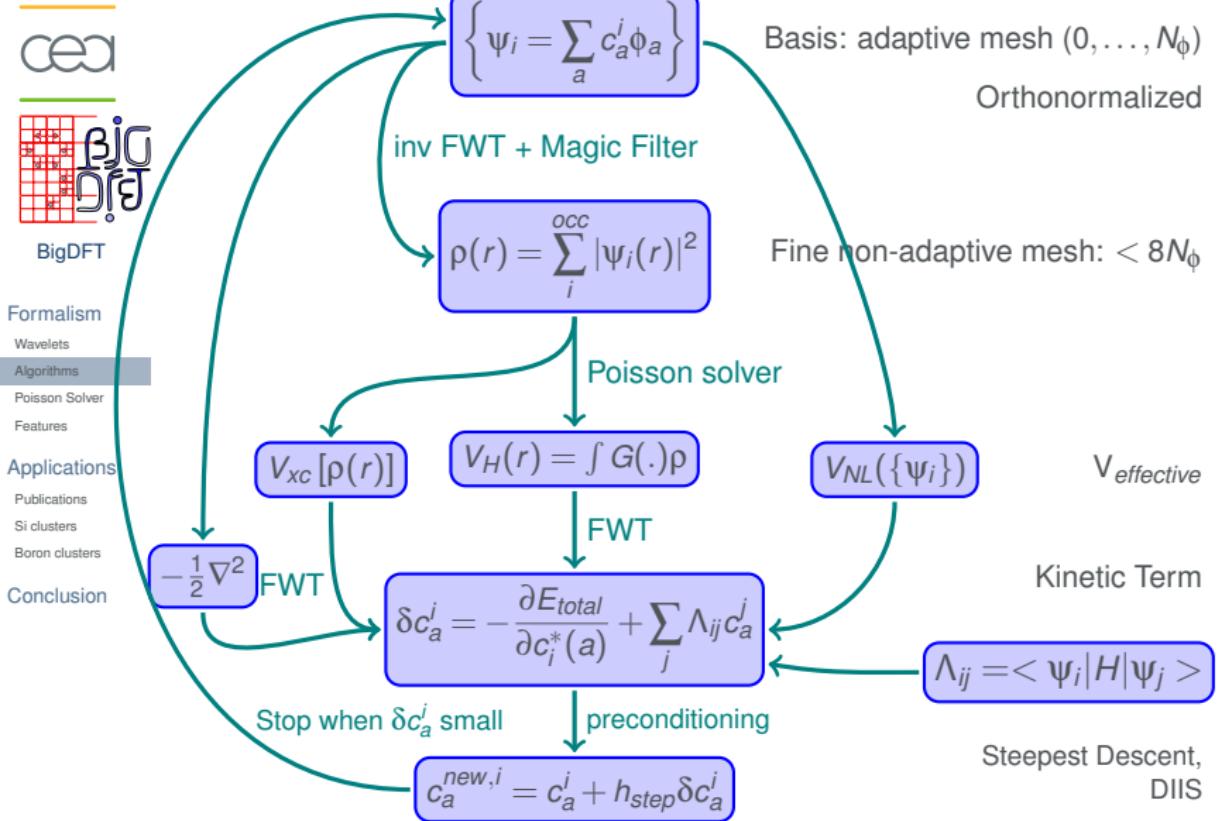
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Direct Minimisation: Flowchart



Poisson Solver using Green function



What is still missing is a method that can solve Poisson's equation with free boundary conditions for arbitrary charge densities.

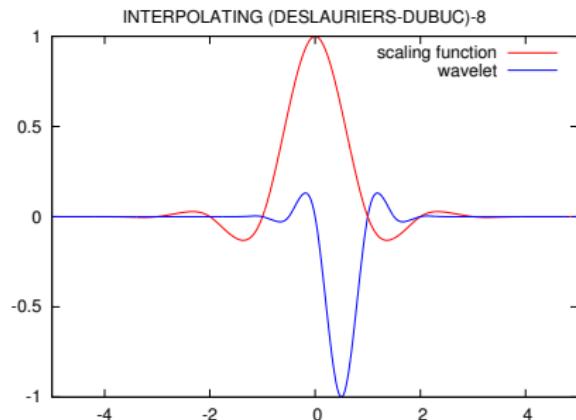
$$V(\mathbf{r}) = \int \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}'$$

Real Mesh (100^3): 10^6 integral evaluations in each point!

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Interpolating $f(x) = \sum_j f_j \phi_j(x)$
Dual to dirac deltas $f_j = f(j)$

The expansion coefficients
are the point values on a
grid.



Poisson Solver using interpolating scaling function

The Hartree potential is calculated in the interpolating scaling function basis using a tensor decomposition of $\frac{1}{r}$.

Poisson solver with interpolating scaling functions

On an uniform grid it calculates: $V_H(\vec{j}) = \int d\vec{x} \frac{\rho(\vec{x})}{|\vec{x} - \vec{j}|}$

- ✓ Very fast and accurate, optimal parallelization
- ✓ Can be used independently from the DFT code
- ✓ Integrated quantities (energies) are easy to extract
- ✗ Non-adaptive, needs data uncompression

Explicitly free boundary conditions

No need to subtract supercell interactions

→ charged systems.

Poisson Solver for surface boundary conditions

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A Poisson solver for surface problems

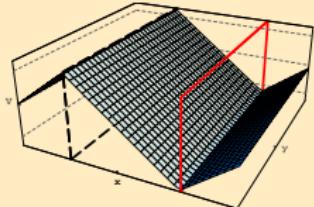
Based on a mixed reciprocal-direct space representation

$$V_{p_x, p_y}(z) = -4\pi \int dz' G(|\vec{p}|; z - z') \rho_{p_x, p_y}(z'),$$

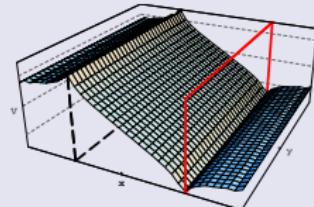
- ✓ Can be applied both in real or reciprocal space codes
- ✓ No supercell or screening functions
- ✓ More precise than other existing approaches

Example of the plane capacitor:

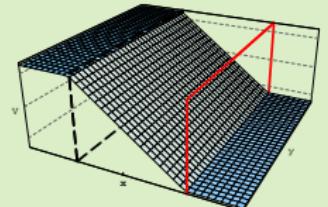
Periodic



Hockney



Our approach



Basis set features

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BigDFT features in a nutshell

- ✓ Arbitrary absolute **precision** can be achieved
Good convergence ratio for real-space approach ($O(h^{14})$)
- ✓ Optimal usage of the degrees of freedom (**adaptivity**)
Optimal speed for a systematic approach (**less memory**)
- ✓ Hartree potential accurate for **various boundary conditions**
Free and Surfaces BC Poisson Solver
(present also in CP2K, ABINIT, OCTOPUS)
- ✗ Data repartition is suitable for optimal scalability
Simple communications paradigm, **multi-level parallelisation** possible (and implemented)

Improve and develop know-how

Optimal for *advanced* DFT functionalities in HPC framework



Optimal for isolated systems

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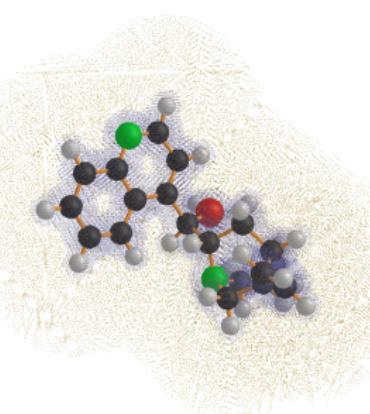
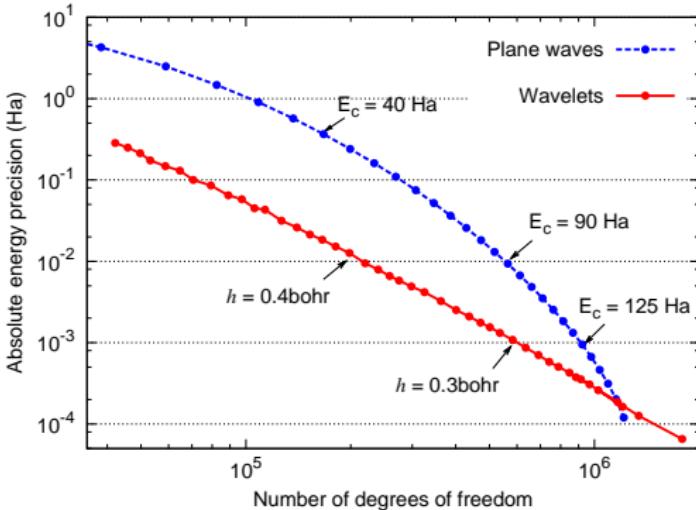
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Test case: cinchonidine molecule (44 atoms)



Allows a systematic approach for molecules

Considerably faster than Plane Waves codes.

10 (5) times faster than ABINIT (CPMD)

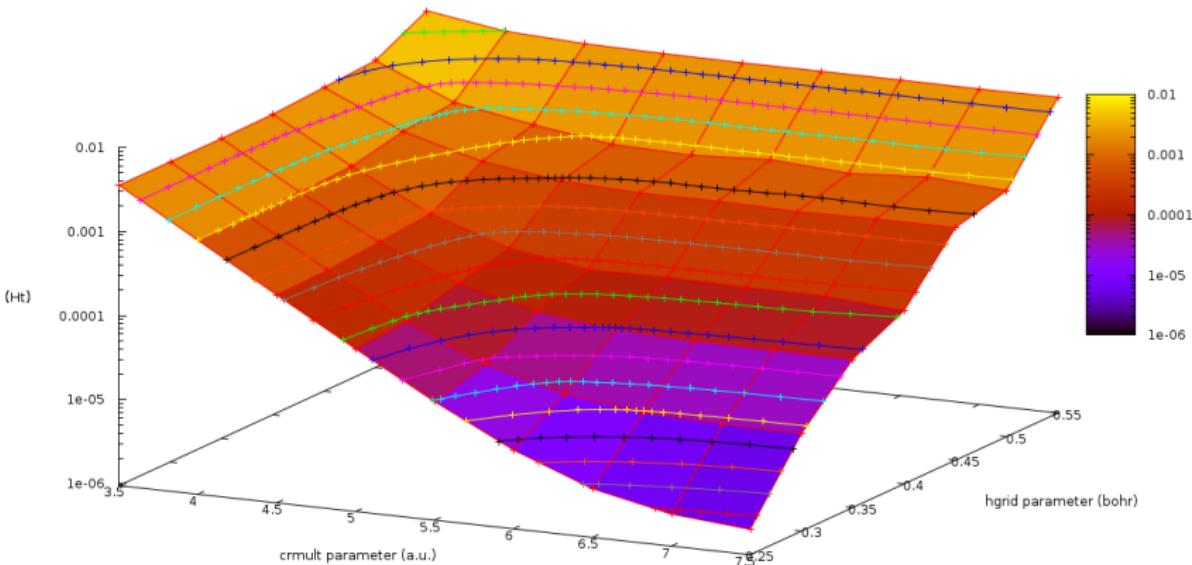
Charged systems can be treated *explicitly* with the same time

Systematic basis set

Two parameters for tuning the basis

- The grid spacing h_{grid}
- The extension of the low resolution points crmult

Convergence of a methane molecule



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BigDFT version 1.6.0: (ABINIT-related) capabilities

http://inac.cea.fr/L_Sim/BigDFT

- Isolated, surfaces and 3D-periodic boundary conditions (k-points, **symmetries**)
- All XC functionals of the ABINIT package (libXC library)
- Hybrid functionals, Fock exchange operator
- Direct Minimisation and **Mixing routines (metals)**
- Local geometry optimizations (with constraints)
- External electric fields (surfaces BC)
- Born-Oppenheimer MD, ESTF-IO
- Vibrations
- Unoccupied states
- Empirical van der Waals interactions
- Saddle point searches (NEB, Granot & Bear)
- All these functionalities are GPU-compatible

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The vessel: Ambitions from BigDFT experience

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Reliable formalism

- Systematic convergence properties
- Explicit environments, analytic operator expressions

State-of-the-art computational technology

- Data locality optimal for operator applications
- Massive parallel environments
- Material accelerators (GPU)

New physics can be approached

- Enhanced functionalities can be applied relatively easily
- Beyond the DFT approximations
- Order N method (in progress)

Production version (robustness)

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- ① Energy Landscape of Fullerene Materials: A Comparison of Boron to Boron Nitride an Carbon, PRL 106, 2011
- ② Low-energy boron fullerenes: Role of disorder and potential synthesis pathways, PRB 83, 2011
- ③ Optimized energy landscape exploration using the ab initio based activation-relaxation technique, JCP 135, 2011
- ④ The effect of ionization on the global minima of small and medium sized silicon and magnesium clusters, JCP 134, 2011
- ⑤ Energy landscape of silicon systems and its description by force fields, tight binding schemes, density functional methods, and quantum Monte Carlo methods, PRB 81, 2010
- ⑥ First-principles prediction of stable SiC cage structures and their synthesis pathways, PRB 82, 2010
- ⑦ Metallofullerenes as fuel cell electrocatalysts: A theoretical investigation of adsorbates on C₅₉Pt, PCCP 12, 2010
- ⑧ Structural metastability of endohedral silicon fullerenes, PRB 81, 2010
- ⑨ Adsorption of small NaCl clusters on surfaces of silicon nanostructures, Nanotechnology 20, 2009
- ⑩ Ab initio calculation of the binding energy of impurities in semiconductors: Application to Si nanowires, PRB 81, 2010

Metal doped silicon clusters

Are **fullerene** like Si_{20} cages ground state configurations?



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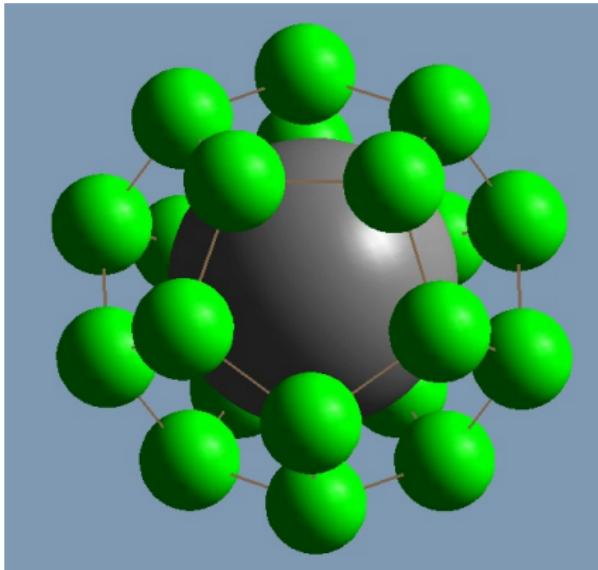
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Collaboration with the group of S. Goedecker, Basel University

Ground state structures of metal doped Si clusters

Never a fullerene configuration!

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Ba



Ca



Cr



Cu



K



Na



Pb



Rb



Sr



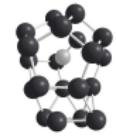
Ti



V



Zr



Phys. Rev. B 82 201405(R) 2010

Is it possible to have Boron fullerenes?

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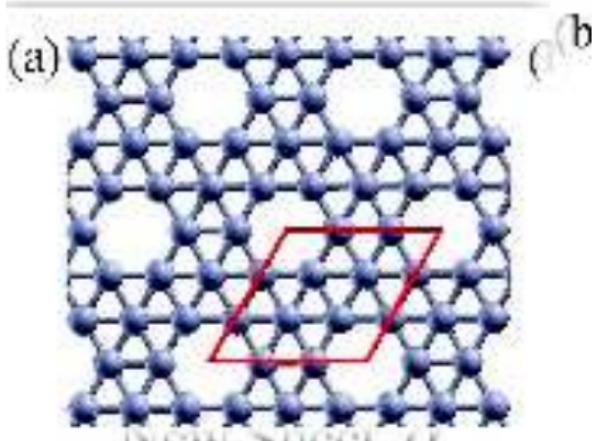
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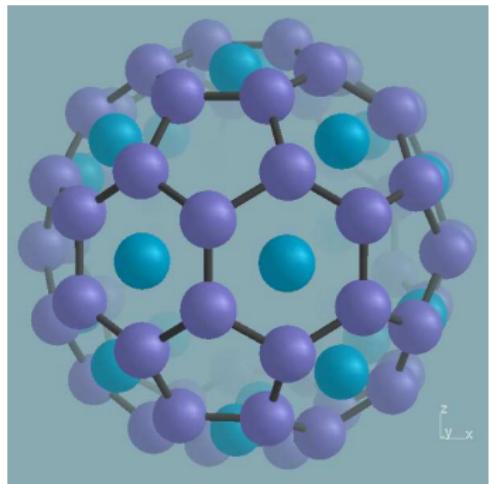
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Boron sheet (Ismail-Beigi)



B_{80} (G. Szwacki)



Lower energy B_{80} cage structures

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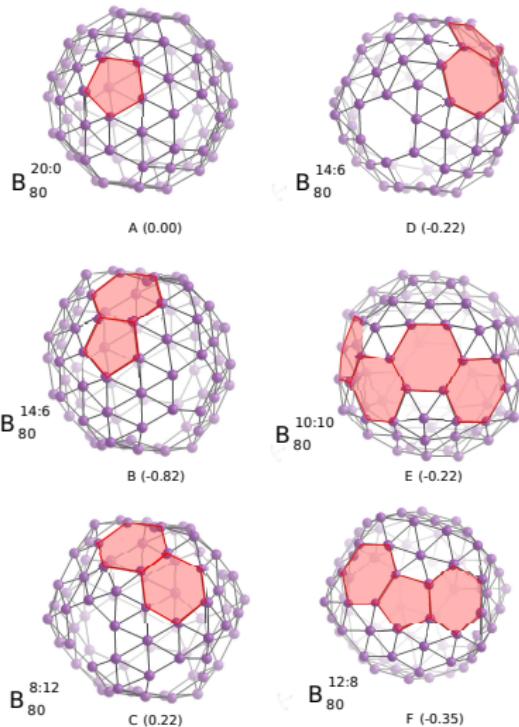
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Phys. Rev. B 83, 081403(R) 2011

Lowest energy B₈₀ cluster

Not a cage!

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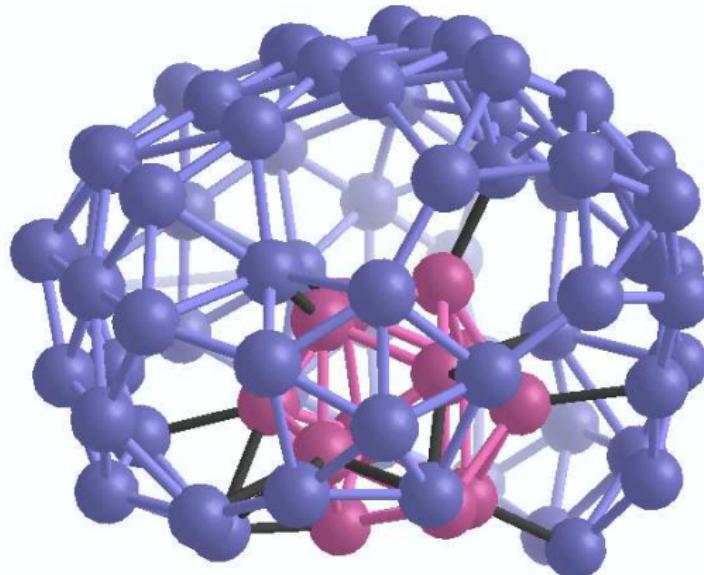
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Phys. Rev. Lett. 106, 225502 (2011)



Laboratoire de Simulation Atomistique http://inac.cea.fr/L_Sim

T. Deutsch, L. Genovese, D. Caliste

The configurational energy spectrum of B_{80} and C_{60}



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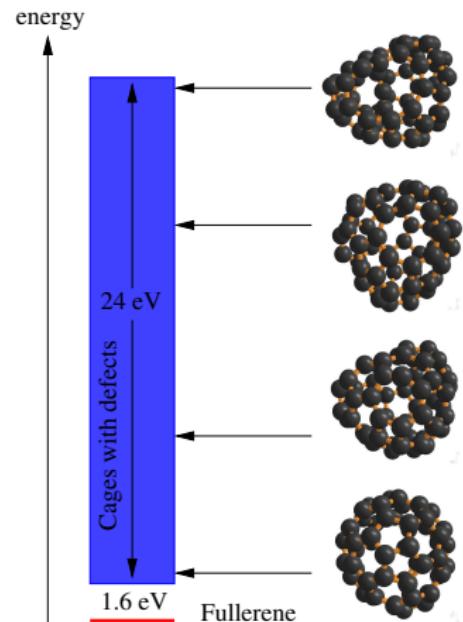
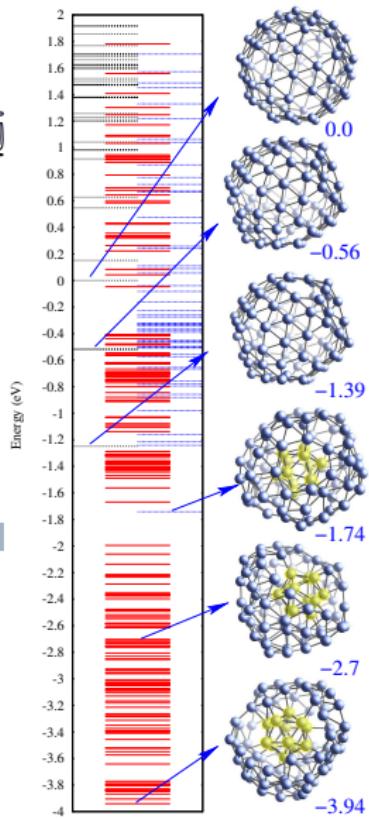
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BigDFT code: a modern approach for nanosciences

- ✓ Flexible, reliable formalism (wavelet properties)
- ✓ Easily fit with massively parallel architecture
- ✓ Open a path toward the diffusion of Hybrid architectures

Development in progress

- ✓ Wire boundary conditions and non-orthorhombic system (full and robust DFT code)
- ✓ PAW pseudopotentials (smoother pseudopotentials)
- ✓ Order N methods (localization regions): large system, QM/QM scheme for embedding systems

BigDFT tutorial on Wednesday

Hands-on, Basic runs, Scalability, GPUs

Acknowledgments



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Poisson Solver

Features

Applications

Publications

Si clusters

Boron clusters

Conclusion

CEA Grenoble – L_Sim

L. Genovese, D. Caliste, B. Videau, P. Pochet, M. Ospici, I. Duchemin, P. Boulanger, E. Machado-Charry, S. Krishnan, F. Cinquini

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And other groups

- Montreal University:
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- European Synchrotron Radiation Facility:
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- ABINIT community (BigDFT inside, strong reusing)

