Multiscale Modelling Methods for Applications in Materials Science  CECAM JÜLICH, GERMANY

Introduction to Electronic Structure Calculations with BigDFT

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L_Sim - CEA Grenoble

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Objectives of the lesson

To know what are the main parameters in a DFT calculation
To run a calculation (i.e. the physicist problem)

1. Introduction

2. Running BigDFT
   - Atom positions
   - Basis set
   - Pseudopotential
   - Exchange-correlation
   - SCF Loop

3. Performances
   - Poisson Solver
   - Relaxation
   - High-Performance Computing (parallel and GPU)

4. Perspectives and Conclusion
   - Order N techniques (very large calculations)
   - Resonant States (Beyond DFT)
   - Conclusion
A basis for nanosciences: the BigDFT project

STREP European project: BigDFT(2005-2008)
Four partners, 15 contributors:

Aim: To develop an ab-initio DFT code based on Daubechies Wavelets, to be integrated in ABINIT, distributed freely (GNU-GPL license)

L. Genovese, A. Neelov, S. Goedecker, T. Deutsch, et al.,
“Daubechies wavelets as a basis set for density functional pseudopotential calculations”,
BigDFT version 1.7: capabilities

http://bigdft.org

- Free, surface and periodic boundary conditions
- Geometry optimizations (with constraints)
- Born-Oppenheimer Molecular Dynamics
- Saddle point searches (Nudged-Elastic Band Method)
- Vibrations
- External electric fields
- Unoccupied KS orbitals
- Collinear and Non-collinear magnetism
- All XC functionals of the ABINIT package
- Hybrid functionals
- Empirical van der Waals interactions (many flavors)
- Also available within the ABINIT package
Quantum Mechanics

Schrödinger equation for $N$ electrons in an external potential $V_{\text{ext}}(r)$:

$$\Psi(r_1, r_2, \ldots, r_N) = -\Psi(r_2, r_1, \ldots, r_N)$$

$$\mathcal{H} = -\frac{\hbar^2}{2m} \sum_i \nabla^2 r_i + \frac{1}{4\pi\varepsilon_0} \sum_{i\neq j} \frac{1}{|r_i - r_j|} + \sum_i V_{\text{ext}}(r_i)$$

$$\mathcal{H} \Psi(r_1, r_2, \ldots, r_N) = E \Psi(r_1, r_2, \ldots, r_N)$$

Ion-electron interactions:

$$V_{\text{ext}}(r) = -\frac{1}{4\pi\varepsilon_0} \sum_{\alpha} \frac{Z_{\alpha}}{|R_{\alpha} - r|}$$
The Hohenberg-Kohn theorem

**Schrödinger equation**

\[ H = \sum_{i=1}^{N} \left( -\frac{1}{2} \nabla^2_{r_i} + V_{\text{ext}}(r_i, \{R\}) \right) + \frac{1}{2} \sum_{i \neq j} \frac{1}{|r_i - r_j|} \]

Very difficult to solve for more than two electrons!

The fundamental variable of the problem is however not the wavefunction, but the electronic density

\[ \rho(r) = N \int dr_2 \cdots dr_N \psi^*(r, r_2, \cdots, r_N) \psi(r, r_2, \cdots, r_N) \]

**Hohenberg-Kohn theorem (1964)**

The ground state density \( \rho(r) \) of a many-electron system uniquely determines (up to a constant) the external potential. The external potential is a functional of \( \rho \):

\[ V_{\text{ext}}(r) = V_{\text{ext}}[\rho](r) \]
The Hohenberg-Kohn theorem

**Schrödinger equation**

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Density Functional Theory (DFT)

**Theorem:** The total energy of the ground state in an external potential $V_{ext}$ is a functional of the electronic density $\rho(r)$:

$$E[\rho(r)] = F[\rho(r)] + \int r V_{ext}(r)\rho(r)dr$$

**Variational principles:** $E[\rho_0(r)] \leq E[\rho(r)]$ \quad $\forall \rho(r)$

**Kohn-Sham Equation:** (approximation)

Having a one-electron hamiltonian in a mean field.

$$\left[-\frac{1}{2} \nabla^2 + V_{eff}(r)\right] \psi_i = \varepsilon_i \psi_i$$

$$\rho(r) = \sum_{i \text{ occupied}} |\psi_i(r)|^2$$

Laboratoire de Simulation Atomistique

http://inac.cea.fr/L_Sim

Thierry Deutsch
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$$\rho(r) = \sum_{\text{occupied}} |\psi_i(r)|^2$$
Kohn-Sham formalism

H-K theorem: \( E \) is an **unknown** functional of the density

\[ E = E[\rho] \rightarrow \text{Density Functional Theory} \]

**Kohn-Sham approach**

Mapping of an interacting many-electron system into a system with **independent particles** moving into an **effective potential**.

**Find a set of orthonormal orbitals** \( \Psi_i(r) \) that minimizes:

\[
E = -\frac{1}{2} \sum_{i=1}^{N/2} \int \psi_i^*(r) \nabla^2 \psi_i(r) dr + \frac{1}{2} \int \rho(r) V_H(r) dr + E_{xc}[\rho(r)] + \int V_{ext}(r) \rho(r) dr
\]

\[
\rho(r) = \sum_{i=1}^{N/2} \psi_i^*(r) \psi_i(r) \quad \nabla^2 V_H(r) = -4\pi \rho(r)
\]
Spirit of BigDFT

- Few input files (posinp.xyz, input.dft)
- Few input parameters
  ex. no parameters for parallel calculations
- Use input files with mandatory parameters
- Optional input files
  as input.kpt (for periodic systems)
  or input.perf (parameters for performance)
How to run BigDFT

Some pre-processing

```
bigdft-tool nprocs [name]
```

Help to find the optimal number of processors for the run that fit into the computer memory. Also useful to check that all input files are without errors.

The main run

```
mpirun -np 8 bigdft [name] | tee [name].log
```

Number of MPI tasks : 8
OpenMP parallelization : Yes
Maximal OpenMP threads per MPI task : 1
Material acceleration : No #iproc=0

The outputs

- *log* is output on stdout, so redirect it.
- generated *data* are stored in a subdirectory called `data[~name]`.
The BigDFT input files (or not)

- **posinp.xyz**: the atomic coordinates and box definition.
- **psppar.Element**: pseudo-potentials for each element.
- **input.dft**: DFT related parameters;
- **input.geopt**: geometric relaxations and MD;
- **input.kpt**: the \( k \)-point mesh and band structure path;
- **input.mix**: diagonalisation mechanism;
- **input.sic, tddft, occ, perf**: other parameters;

Only the coordinates are mandatory.

All lines of different input files are mandatory and output on BigDFT log. **When BigDFT is run, all default values are output in files called default.xxx.**

**Naming scheme**

All input files may be renamed by providing a `name` argument to the command line i.e. `bigdft name`. 
Atomic positions: posinp.xyz or posinp.ascii

- **units** reduced, angstrom, bohr or atomic
- **Boundary conditions** periodic, surface

8 reduced
**periodic** 10.26085 10.26085
10.26085
Si 0.0.0.
Si 0.5 0.5 0.
Si 0.5 0.5
Si 0. 0. 0.5
Si 0.25 0.25 0.25
Si 0.75 0.75 0.25
Si 0.75 0.25 0.75
Si 0.25 0.75 0.75

---

BigDFT
http://bigdft.org

Introduction

BigDFT run
Atom positions

Basis set
Pseudopotential
XC
SCF Loop

Performances
Poisson Solver
Relaxation
HPC

Perspectives
Order N
Resonant states
Conclusion
Atomic positions: posinp.xyz or posinp.ascii

- **units** reduced, angstrom, bohr or atomic
- **Boundary conditions** periodic, surface

3 angstrom

**surface** 3.0 0.0 3.0
O 1.5 0.0 1.5
H .7285 0.620919 1.5
H 2.2715 0.620919 1.5
Atomic positions: posinp.xyz or posinp.ascii

- **units** reduced, angstroem, bohr or atomic
- **Boundary conditions** periodic, surface

5 atomic

Si 1.62 1.62 1.62
H 3.24 3.24 3.24
H 0 0 3.24
H 3.24 0 0
H 0 3.24 0
Atomic coordinate format

BigDFT uses $XYZ$ format plus additional modifications:

- **physical unit**, after the number of atoms on the first line put either *bohr* or *angstrom*.
- **boundary conditions**, on second line, start with one of freeBC, surface $X_{\text{lat}} 0 Z_{\text{lat}}$ or periodic $X_{\text{lat}} Y_{\text{lat}} Z_{\text{lat}}$.
- **frozen positions**, add a *f*, *fy* or *fxz* at the end of an atom line.
- **input polarisation**, add values at the end of an atom line.

On output, forces are added for each element at the end of the $XYZ$ file:

2 angstroem
freeBC
Na -1 0 0
Cl +1 0 0
forces
Na 2.654367E-2 0 0
Cl -2.654367E-2 0 0
Performing a DFT calculation

A self-consistent equation

\[ \rho(r) = \sum_i \psi_i^*(r) \psi_i(r), \text{ where } |\psi_i\rangle \text{ satisfies} \]

\[ \left( -\frac{1}{2} \nabla^2 + V_H[\rho] + V_{xc}[\rho] + V_{ext} + V_{pseudo} \right) |\psi_i\rangle = E_i |\psi_i\rangle, \]

(Kohn-Sham) DFT “Ingredients”

- A basis set for expressing the \(|\psi_i\rangle\)
- An XC potential, functional of the density
  several approximations exists (LDA, GGA, . . .)
- A choice of the pseudopotential (if not all-electrons)
  (norm conserving, ultrasoft, PAW, . . .)
- An (iterative) algorithm for finding the wavefunctions \(|\psi_i\rangle\)
- A (good) computer . . .
Basis sets in real space: Wavelet basis sets

Properties of wavelet basis sets:

- localized both in real and in Fourier space
- allow for adaptivity (for core electrons)
- are a systematic basis set

A basis set both adaptive and systematic, real space based
A brief description of wavelet theory

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

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

Centered on a resolution-dependent grid: \( \phi_j = \phi_0(x - j) \).
A brief description of wavelet theory

Wavelets

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

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

Increase the resolution without modifying grid space

\[ \psi(x) = \sum_{j=-m}^{m} g_j \phi(2x - j) \]

All functions have compact support, centered on grid points.
Adaptivity of the mesh

Adaptivity

Resolution can be refined following the grid point.

The grid is divided in:
- **Low** resolution pts (SF, 1 DoF)
- **High** resolution pts (SF + W, 8 DoF)

Points of different resolution belong to the same grid.

Data can thus be stored in *compressed form*.

Localization property

Empty regions must not be “filled” with basis functions. Optimal for big inhomogeneous systems, \( O(N) \) approach.
Adaptivity of the mesh

Atomic positions (H$_2$O), \texttt{hgrid}
Adaptivity of the mesh

Fine grid (high resolution, \textit{frmult})
Adaptivity of the mesh

Coarse grid (low resolution, \textit{crmult})
Defining the basis set in `input.dft`

Main specific variables to control the basis set

- `hgrid` the grid size
- `frmult` the expansion around atoms for fine grid
- `crmult` the expansion around atoms for coarse grid

Convergence properties

- Decreasing `hgrid` → more degrees of freedom.
- Increasing `crmult` → less box constraint.

It is important to improve both parameters together to avoid error on one hiding improvement made on the other one.
**input.dft:** `hgrid`, `crmult`, `frmult`

**hgrid**  Step grid: twice as bigger than the lowest gaussian coefficient of pseudopotential
Roughly 0.45 (only orthorhombic mesh)

**crmult**  Extension of the coarse resolution (factor 6.):
depends on the extension of the HOMO for the atom

**frmult**  Extension of the fine resolution (factor 8.):
depends on the pseudopotential coefficient

In practice, we use only `hgrid` from 0.3 to 0.55.
Wavelets: No integration error

Orthogonality, scaling relation

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

The hamiltonian-related quantities can be calculated \textit{up to machine precision} in the given basis.

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

Exact evaluation of kinetic energy

Obtained by convolution with filters:

\[ f(x) = \sum_{\ell} c_{\ell} \phi_{\ell}(x), \quad \nabla^2 f(x) = \sum_{\ell} \tilde{c}_{\ell} \phi_{\ell}(x), \]
\[ \tilde{c}_{\ell} = \sum_{j} c_{j} a_{\ell-j}, \quad a_{\ell} \equiv \int \phi_0(x) \partial_x^2 \phi_{\ell}(x), \]
Wavelets: 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
- Non-local pseudopotentials
Performing a DFT calculation

A self-consistent equation

\[ \rho(r) = \sum_i \psi_i^*(r) \psi_i(r), \quad \text{where } |\psi_i\rangle \text{ satisfies} \]

\[ \left( -\frac{1}{2} \nabla^2 + V_H[\rho] + V_{xc}[\rho] + V_{ext} + V_{pseudo} \right) |\psi_i\rangle = E_i |\psi_i\rangle, \]

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Gaussian type separable Pseudopotentials (HGH)

Local part

\[ V_{loc}(r) = \frac{-Z_{ion}}{r} \text{erf}\left[\frac{r}{\sqrt{2}r_{loc}}\right] + \exp\left[-\frac{1}{2} \left(\frac{r}{r_{loc}}\right)^2\right] \]

\[ \left\{ C_1 + C_2 \left(\frac{r}{r_{loc}}\right)^2 + C_3 \left(\frac{r}{r_{loc}}\right)^4 + C_4 \left(\frac{r}{r_{loc}}\right)^6 \right\} \]

Nonlocal (separable) part \( H(\mathbf{r}, \mathbf{r}') \)

\[ H^{sep}(\mathbf{r}, \mathbf{r}') = \sum_{i=1}^{2} \sum_{m} Y_{s,m}(\hat{\mathbf{r}}) \, p_i^s(r) \, h_i^s \, p_i^s(r') \, Y_{s,m}^*(\hat{\mathbf{r}}') \]

\[ + \sum_{m} Y_{p,m}(\hat{\mathbf{r}}) \, p_1^p(r) \, h_1^p \, p_1^p(r') \, Y_{p,m}^*(\hat{\mathbf{r}}') \]

\[ p_1^l(r) = \sqrt{2} \frac{r^l e^{-\frac{1}{2} \left(\frac{r}{r_l}\right)^2}}{r_l^{l+\frac{3}{2}} \sqrt{\Gamma(l + \frac{3}{2})}} \]

\[ p_2^l(r) = \sqrt{2} \frac{r^{l+2} e^{-\frac{1}{2} \left(\frac{r}{r_l}\right)^2}}{r_l^{l+\frac{7}{2}} \sqrt{\Gamma(l + \frac{7}{2})}} \]
All-electron accuracy with Norm Conserving PSP

Non-linear core correction added to HGH PSP


- Simple analytic form (a single gaussian as $\rho_c$)
- Same hardness as HGH
  - a systematic localized basis is **fundamental**

Accuracy considerably improved

Example: G2-1 test set (Atomization Energies)

<table>
<thead>
<tr>
<th>kcal/mol</th>
<th>MAD</th>
<th>RMSD</th>
<th>MSD</th>
<th>maxAD</th>
<th>minAD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Old HGH</td>
<td>6.85</td>
<td>9.13</td>
<td>-6.76</td>
<td>23.94</td>
<td>0.10</td>
</tr>
<tr>
<td>NLCC-HGH</td>
<td>0.51</td>
<td>0.63</td>
<td>0.16</td>
<td>1.50</td>
<td>0.03</td>
</tr>
<tr>
<td>PAW Paier</td>
<td>0.46</td>
<td>0.56</td>
<td>-0.43</td>
<td>1.13</td>
<td>0.01</td>
</tr>
<tr>
<td>$\Delta$ AE (geopt)</td>
<td>0.29</td>
<td>0.70</td>
<td>-0.29</td>
<td>4.21</td>
<td>0.00</td>
</tr>
</tbody>
</table>

AE accuracy for quantities in different environments

Bond lengths, Pressure (Bulk systems), Dispersion-corrected interaction energies, . . .
Pseudopotential file (as `psppar.N`)

- No need if you use LDA or PBE functionals
  Default pseudopotentials inside BigDFT!

- Put in the same directory as the calculation.

- Large library:

- Accurate and transferable
Performing a DFT calculation

A self-consistent equation

\[ \rho(r) = \sum_i \Psi_i^*(r) \Psi_i(r), \text{ where } |\psi_i\rangle \text{ satisfies} \]

\[ \left( -\frac{1}{2} \nabla^2 + V_H[\rho] + V_{xc}[\rho] + V_{ext} + V_{pseudo} \right) |\psi_i\rangle = E_i |\psi_i\rangle, \]

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Exchange-Correlation Energy \( (\text{input.dft}) \)

\[
E_{xc} [\rho] = K_{\text{exact}} [\rho] - K_{\text{KS}} [\rho] + V_{e-e \text{exact}} [\rho] - V_{\text{Hartree}} [\rho]
\]

- \( K_{\text{KS}} [\rho] \): Kinetic energy for a non-interaction electron gas
- Kohn-Sham formalism is exact (mapping of an interacting electron system into a non-interacting system)

Many types from

- ABINIT: Use same codes
  (ixc: LDA=1, PBE=11, see manual)
- libXC: ‘code for exchange’ ‘code for correlation’
  (−101130 see manual)
- Hybrid functionals from libXC

\[ \text{Laboratoire de Simulation Atomistique} \quad \text{http://inac.cea.fr/L_Sim} \quad \text{Thierry Deutsch} \]
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Kohn-Sham Equations: Operators

Apply different operators

Having a one-electron hamiltonian in a mean field.

\[
\left[ -\frac{1}{2} \nabla^2 + V_{\text{eff}}(r)(r) \right] \psi_i = \varepsilon_i \psi_i
\]

\[
V_{\text{eff}}(r) = V_{\text{ext}}(r) + \int_{\mathcal{V}} \frac{\rho (r')}{|r - r'|} \, dr' + \mu_{\text{xc}}(r)
\]

\(E[\rho]\) can be expressed by the orthonormalized states of one particule: \(\psi_i(r)\) with the fractional occupancy number \(f_i\) \((0 \leq f_i \leq 1)\):

\[
\rho(r) = \sum_i f_i |\psi_i(r)|^2
\]
Kohn-Sham Equations: Computing Energies

Calculate different integrals

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

\[ K[\rho] = -\frac{1}{2} \frac{\hbar^2}{m_e} \sum_i \int_V dr f_i \psi_i^* \nabla^2 \psi_i \]

\[ U[\rho] = \int_V dr \, V_{ext}(r) \rho(r) + \frac{1}{2} \int_V dr \, dr' \frac{\rho(r) \rho(r')}{|r - r'|} + E_{xc}[\rho] \]

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

with the constraint \( \int_V dr \rho(r) = N_{el} \).
KS Equations: Self-Consistent Field

Set of self-consistent equations:

\[
\begin{aligned}
\left\{ -\frac{\hbar^2}{2m_e} \nabla^2 + V_{\text{eff}} \right\} \psi_i &= \varepsilon_i \psi_i \\
V_{\text{eff}}(r) &= V_{\text{ext}}(r) + \int_{\mathcal{V}} dr' \frac{\rho(r')}{|r-r'|} \quad \text{(Hartree)} + \frac{\delta E_{xc}}{\delta \rho(r)} \quad \text{(exchange–correlation)}
\end{aligned}
\]

and:

\[\rho(r) = 2 \sum_i f_i |\psi_i(r)|^2\]
KS Equations: Self-Consistent Field

Set of self-consistent equations:

\[
\begin{aligned}
\left\{- \frac{1}{2 m_e} \nabla^2 + V_{\text{eff}} \right\} \psi_i = \varepsilon_i \psi_i
\end{aligned}
\]

with an effective potential:

\[
V_{\text{eff}}(r) = V_{\text{ext}}(r) + \int_{\mathbb{V}} \frac{\rho(r')}{|r - r'|} \text{d}r' + \frac{\delta E_{\text{xc}}}{\delta \rho(r)}
\]

and:

\[
\rho(r) = 2 \sum_i f_i |\psi_i(r)|^2
\]

Poisson Equation:

\[
\Delta V_{\text{Hartree}} = \rho
\]

(Laplacian: \( \Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \))
KS Equations: Self-Consistent Field

Set of self-consistent equations:

\[
\left\{ -\frac{1}{2} \frac{\hbar^2}{m_e} \nabla^2 + V_{\text{eff}} \right\} \psi_i = \varepsilon_i \psi_i
\]

with an effective potential:

\[
V_{\text{eff}}(r) = V_{\text{ext}}(r) + \int_V dr' \frac{\rho(r')}{|r - r'|} \quad \text{Hartree}
\]

and:

\[
\rho(r) = 2 \sum_i f_i |\psi_i(r)|^2
\]

Poisson Equation: \( \Delta V_{\text{Hartree}} = \rho \) (Laplacian: \( \Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \))
KS Equations: Self-Consistent Field

Set of self-consistent equations:

\[
\left\{- \frac{1}{2} \frac{\hbar^2}{m_e} \nabla^2 + V_{\text{eff}} \right\} \psi_i = \varepsilon_i \psi_i
\]

with an effective potential:

\[
V_{\text{eff}}(r) = V_{\text{ext}}(r) + \int_{\mathcal{V}} \frac{\rho(r')}{|r - r'|} \, dr' \quad \text{[Hartree]}
\]

and:

\[
\rho(r) = 2 \sum_i f_i |\psi_i(r)|^2
\]

Poisson Equation:

\[
\Delta V_{\text{Hartree}} = \rho
\]

(Laplacian: \(\Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}\))

\[
\frac{\delta E_{\text{xc}}}{\delta \rho(r)}
\]

exchange–correlation
KS Equations: Self-Consistent Field

Set of self-consistent equations:

\[ \left\{ -\frac{1}{2m_e} \nabla^2 + V_{\text{eff}} \right\} \psi_i = \varepsilon_i \psi_i \]

with an effective potential:

\[ V_{\text{eff}}(r) = V_{\text{ext}}(r) + \int_{V} \frac{\rho(r')}{|r - r'|} \text{Hartree} \]

\[ \frac{\delta E_{xc}}{\delta \rho(r)} \text{ exchange–correlation} \]

and:

\[ \rho(r) = 2 \sum_i f_i |\psi_i(r)|^2 \]

Poisson Equation:

\[ \Delta V_{\text{Hartree}} = \rho \] (Laplacian: \( \Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \))
KS Equations: Self-Consistent Field

Set of self-consistent equations:

$$
\left\{ -\frac{1}{2} \frac{\hbar^2}{m} \nabla^2 + V_{\text{eff}} \right\} \psi_i = \varepsilon_i \psi_i
$$

with an effective potential:

$$
V_{\text{eff}}(r) = V_{\text{ext}}(r) + \int_{\mathcal{V}} \frac{\rho(r')}{|r-r'|} \, dr' \quad \text{(Hartree)}
$$

and:

$$
\rho(r) = 2 \sum_i f_i |\psi_i(r)|^2
$$

Poisson Equation:

$$
\Delta V_{\text{Hartree}} = \rho \quad \text{(Laplacian: } \Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2})
$$

Poisson Solver
Relaxation
HPC
Direct Minimisation: Flowchart

\[ \psi_i = \sum_a c_i^a \phi_a \]

Basis: adaptive mesh \((0, \ldots, N_\phi)\)

Orthonormalized
Direct Minimisation: Flowchart

\[ \psi_i = \sum_a c^i_a \phi_a \]

Basis: adaptive mesh \((0, \ldots, N_\phi)\)

Orthonormalized

\[ \rho(r) = \sum_i^{occ} |\psi_i(r)|^2 \]

Fine non-adaptive mesh: \(< 8N_\phi\)
Direct Minimisation: Flowchart

\[ \psi_i = \sum_a c_i^a \phi_a \]

Basis: adaptive mesh \((0, \ldots, N_\phi)\)
Orthonormalized

\[ \rho(r) = \sum_i |\psi_i(r)|^2 \]

Fine non-adaptive mesh: \(< 8N_\phi\)

\[ V_{xc}[\rho(r)] \]

\[ V_H(r) = \int G(.) \rho \]

\[ V_{NL}(\{\psi_i\}) \]

\[ V_{\text{effective}} \]
Direct Minimisation: Flowchart

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Fine non-adaptive mesh: \(<8N_\phi\)

\[ \rho(r) = \sum_i |\psi_i(r)|^2 \]

Poisson solver

\[ V_{xc}[\rho(r)] \]

\[ V_H(r) = \int G(.) \rho \]

\[ V_{NL}(\{\psi_i\}) \]

\[ -\frac{1}{2} \nabla^2 \]

\[ V_{\text{effective}} \]

Kinetic Term

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Laboratoire de Simulation Atomistique
http://inac.cea.fr/L_Sim
Thierry Deutsch
Direct Minimisation: Flowchart

\[ \psi_i = \sum_a c^i_a \phi_a \]

\[ \rho(r) = \sum_i \text{occ} |\psi_i(r)|^2 \]

\[ V_{xc}[\rho(r)] \]

\[ V_H(r) = \int G(\cdot) \rho \]

\[ V_{NL}\{\psi_i\} \]

\[ -\frac{1}{2} \nabla^2 \]

\[ \text{FWT} \]

\[ \delta c^i_a = -\frac{\partial E_{total}}{\partial c^*_i(a)} + \sum_j \Lambda_{ij} c^j_a \]

\[ \Lambda_{ij} = \langle \psi_i | H | \psi_j \rangle \]

Basis: adaptive mesh \((0, \ldots, N_\phi)\)

Orthonormalized

Fine non-adaptive mesh: < \(8N_\phi\)

Poisson solver

Kinetic Term

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\[ V_H(r) = \int G(.) \rho \]

\[ V_{NL}\{\psi_i\} \]

\[ V_{effective} \]

Kinetic Term

\[ -\frac{1}{2} \nabla^2 \]

\[ \delta c_a^i = -\frac{\partial E_{total}}{\partial c_i^*(a)} + \sum_j \Lambda_{ij} c_a^j \]

preconditioning

\[ \Lambda_{ij} = \langle \psi_i | H | \psi_j \rangle \]

Steepest Descent, DIIS

\[ c_a^{new, i} = c_a^i + h_{step} \delta c_a^i \]

Stop when \(\delta c_a^i\) small

FWT

inv FWT + Magic Filter

Poisson Solver

Relaxation

HPC

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input.dft: idsx DIIS history

- Use DIIS algorithm
  Direct Inversion in the Iterative Subspace

- Use idsx=6 previous iterations Memory-consuming
  (decrease if too big)

- Steepest descent if idsx=0

- itermax=50 Specify maximum number of iterations
  Typically 15 iterations to converge

- gnrm_cv=1.e-4 convergence criterion
  Residue $|\delta \psi|^2 < gnrm_cv$
Description of the file *input.dft*

0.450 0.450 0.450
5.0 8.0
1
0 0.0
1 0
1.E-04
50 10
7 6
0
0 0 0
5.0 30
0 0 0
davidson treatment, # virtual orbitals, # plotted orbitals
disable the symmetry detection

hx, hy, hz
crmult, frmult, coarse and fine radius
ixc: XC parameter (LDA=1,PBE=11)
ncharge: charge of the system, Electric field
nspin=1 non-spin polarization, total magnetic moment
gnrm_cv: convergence criterion gradient
itermax, nrepmax
# CG (preconditioning), length of diis history
dispersion correction functional (0, 1, 2, 3)
InputPsiId, output_wf, output_grid
length of the tail, # tail CG iterations
T disable the symmetry detection
Description of the file `input.dft`

```
0.450 0.450 0.450 hx, hy, hz
5.0 8.0 crmult, frmult, coarse and fine radius
1
0 0.0 ncharge: charge of the system, Electric field
1 0 nspin=1 non-spin polarization, total magnetic moment
1.E-04
50 10 # CG (preconditioning), length of diis history
7 6 itermax, nrepxmax
0 dispersion correction functional (0, 1, 2, 3)
0 0 0 InputPsiId, output_wf, output_grid
5.0 30 length of the tail, # tail CG iterations
0 0 0 davidson treatment, # virtual orbitals, # plotted orbitals
T disable the symmetry detection
```
Description of the file input.dft

0.450 0.450 0.450 hx, hy, hz
5.0 8.0 crmult, frmult, coarse and fine radius
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Thierry Deutsch
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5.0 8.0 crmult, frmult, coarse and fine radius
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0 0.0 ncharge: charge of the system, Electric field
1 0 nspin=1 non-spin polarization, total magnetic moment
1.E-04 gnrm_cv: convergence criterion gradient
50 10 itermax, nreppmax
7 6 # CG (preconditioning), length of diis history
0 dispersion correction functional (0, 1, 2, 3)
0 InputPsild, output_wf, output_grid
0 0 0 # virtual orbitals, # plotted orbitals
davidson treatment, T disable the symmetry detection
5.0 30 length of the tail, # tail CG iterations
0 0 0
Description of the file `input.dft`

Description of the file `input.dft`

BigDFT
http://bigdft.org

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Thierry Deutsch
Description of the file input.dft

0.450 0.450 0.450  \quad hx, hy, hz
5.0  8.0       \quad crmult, frmult, coarse and fine radius
 1                \quad ixc: XC parameter (LDA=1, PBE=11)
0 0.0             \quad ncharge: charge of the system, Electric field
1 0               \quad nspin=1 non-spin polarization, total magnetic moment
1.0E-04           \quad gnrm_cv: convergence criterion gradient
50 10             \quad itermax, nrepsmax
7  6              \quad # CG (preconditioning), length of diis history
  0                \quad dispersion correction functional (0, 1, 2, 3)
0  0  0           \quad InputPsild, output_wf, output_grid
5.0  30           \quad length of the tail, # tail CG iterations
0  0  0           \quad davidson treatment, # virtual orbitals, # plotted orbitals
T                \quad disable the symmetry detection
Description of the file `input.dft`

```
0.450 0.450 0.450 # hx, hy, hz
5.0 8.0 # crmult, frmult, coarse and fine radius
1 # ixc: XC parameter (LDA=1,PBE=11)
0 0.0 # ncharge: charge of the system, Electric field
1 0 # nspin=1 non-spin polarization, total magnetic moment
1.E-04 # gnrm_cv: convergence criterion gradient
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50 10 # itermax, nrepmax
7 6 # CG (preconditioning), length of diis history
0 # dispersion correction functional (0, 1, 2, 3)
0 0 0 # InputPsiId, output_wf, output_grid
5.0 30 # length of the tail, # tail CG iterations
0 0 0 # davidson treatment, # virtual orbitals, # plotted orbitals
T # disable the symmetry detection
```
Description of the file `input.dft`

```
0.450 0.450 0.450  # hx, hy, hz
5.0 8.0            # crmult, frmult, coarse and fine radius
1                # ixc: XC parameter (LDA=1,PBE=11)
0 0.0            # ncharge: charge of the system, Electric field
1 0            # nspin=1 non-spin polarization, total magnetic moment
1.E-04           # gnrm_cv: convergence criterion gradient
50 10            # itermax, nrepmax
7 6            # CG (preconditioning), length of diis history
0            # dispersion correction functional (0, 1, 2, 3)
0 0 0            # InputPsiId, output_wf, output_grid
5.0 30            # length of the tail, # tail CG iterations
0 0 0            # davidson treatment, # virtual orbitals, # plotted orbitals
T            # disable the symmetry detection
```

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Description of the file input.dft

0.450 0.450 0.450 hx, hy, hz
5.0 8.0 crmult, frmult, coarse and fine radius
1 ixc: XC parameter (LDA=1,PBE=11)
0 0.0 ncharge: charge of the system, Electric field
1 0 nspin=1 non-spin polarization, total magnetic moment
1.E-04 gnrm_cv: convergence criterion gradient
50 10 itermax, nrepmax
7 6 # CG (preconditioning), length of diis history
dispersion correction functional (0, 1, 2, 3)
0 InputPsild, output_wf, output_grid
displacement correction
5.0 30 length of the tail, # tail CG iterations
do 0 0 0 davidson treatment, # virtual orbitals, # plotted orbitals
t1 disable the symmetry detection
Description of the file `input.dft`

```
0.450 0.450 0.450 hx, hy, hz
5.0 8.0 crmult, frmult, coarse and fine radius
1 ixc: XC parameter (LDA=1,PBE=11)
0 0.0 ncharge: charge of the system, Electric field
1 0 nspin=1 non-spin polarization, total magnetic moment
1.E-04 gnrm_cv: convergence criterion gradient
50 10 itermax, nrepmax
7 6 # CG (preconditioning), length of diis history
0 dispersion correction functional (0, 1, 2, 3)
0 0 0 InputPsiId, output_wf, output_grid
5.0 30 length of the tail, # tail CG iterations
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```
Description of the file `input.dft`

0.450 0.450 0.450  hx, hy, hz
5.0 8.0  crmult, frmult, coarse and fine radius
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0  dispersion correction functional (0, 1, 2, 3)
0 0 0  # virtual orbitals, # plotted orbitals
5.0 30  length of the tail, # tail CG iterations
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T  disable the symmetry detection
Description of the file `input.dft`

```
0.450 0.450 0.450       hx, hy, hz
5.0  8.0          crmult, frmult, coarse and fine radius
1                         icx: XC parameter (LDA=1, PBE=11)
0  0.0                  ncharge: charge of the system, Electric field
1  0                  nspin=1 non-spin polarization, total magnetic moment
1.E-04                                   gnrm_cv: convergence criterion gradient
50  10                          itermax, nrepmax
7  6            # CG (preconditioning), length of diis history
0                           dispersion correction functional (0, 1, 2, 3)
0  0  0                   InputPsiId, output_wf, output_grid
5.0  30                        # CG iterations
0  0  0                    length of the tail, # tail CG iterations
0  0  0                   davidson treatment, # virtual orbitals, # plotted orbitals
T                           disable the symmetry detection
```
Basis sets: wavelets

\[
- \frac{1}{2} \nabla^2 + V(r) + \mu_{xc}(r) \psi_i^k = \epsilon_i^k \psi_i^k
\]

- self-consistent (SCF)
- Harris-Foulkes functional
- relativistic
- non-relativistic
- periodic
- non-periodic
- \( N^3 \) scaling
- \( O(N) \) methods

- pseudopotential
- norm-conserving
- all electrons
- PAW

- Hybrid functionals
- beyond LDA
- GW
- LDA, GGA
- LDA+U

- atomic orbitals
- Gaussians
- Slater
- numerical
- plane waves
- augmented
- real space
- finite difference
- Wavelet

- non-spin polarized
- spin polarized
- non-collinear
- collinear

- Basis set
- Pseudopotential
- XC
- SCF Loop

- Performances
- Poisson Solver
- Relaxation
- HPC

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Optimal for isolated systems

Test case: cinchonidine molecule (44 atoms)

![Graph showing absolute energy precision vs number of degrees of freedom for Plane waves and Wavelets.]

- Plane waves
  - $E_c = 125$ Ha
  - $h = 0.3$ bohr
- Wavelets
  - $E_c = 90$ Ha
  - $h = 0.4$ bohr
  - $E_c = 40$ Ha
  - $h = 0.3$ bohr

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

- The grid spacing $h_{\text{grid}}$
- The extension of the low resolution points $c_{\text{rmult}}$

Convergence of a methane molecule
Comparison with a code based on gaussian basis set

The Daubechies expansion of a gaussian is known → Results from gaussian codes can be imported in BigDFT

Gaussian code (CP2K) test, H$_2$O and DZVP basis

- BigDFT Input Guess Energy
- CP2K Imported energy
- 6-31G Conv energy
- BigDFT converged energy
- Wavelet Conv energy
- BigDFT with finite-size corrections
Poisson Solver using interpolating scaling function

The Hartree potential is calculated in the **interpolating scaling function** basis.

**Poisson solver with interpolating scaling functions**

From the density $\rho(j)$ on an **uniform** grid it calculates:

$$V_H(r) = \int \frac{\rho(r')}{|r - r'|} dr'$$

- ✔ 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**).
A Poisson solver for surface problems

Based on a mixed reciprocal-direct space representation

\[ V_{px,py}(z) = -4\pi \int dz' G(|\vec{p}|; z - z') \rho_{px,py}(z), \]

- 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**
input.geopt: Relaxation or molecular dynamics

DIIS
500       ncount_cluster: max steps during geometry relaxation
1.d0 1.d-3 frac_fluct: geometry optimization stops if force norm less than frac_fluct of noise
0.0d0 maxval: maximum value of atomic forces
4.d0 5 randdis: random amplitude for atoms
betax: steepest descent step size, diis history

Different algorithms:
- SDCG: Steepest Descent Conjugate Gradient
- DIIS: Direct Inversion in the iterative subspace
- VSSD: Variable Size Steepest Descent
- LBFGS: Limited BFGD
- AB6MD: Use molecular dynamics routines from ABINIT 6
input.geopt: Relaxation or molecular dynamics

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input.geopt: Relaxation or molecular dynamics

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**Different algorithms:**

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- AB6MD: Use molecular dynamics routines from ABINIT 6
High-Performance Computing: Massively parallel

**Two kinds of parallelisation**
- By orbitals (Hamiltonian application, preconditioning)
- By components (overlap matrices, orthogonalisation)

**A few (but big) packets of data**
More demanding in bandwidth than in latency
- No need of fast network
- Optimal speedup (eff. $\sim 85\%$), also for big systems

**Cubic scaling code**
For systems bigger than 500 atomes (1500 orbitals): orthonormalisation operation is predominant ($N^3$)
Orbital distribution scheme

Used for the application of the hamiltonian

The hamiltonian (convolutions) is applied separately onto each wavefunction

\[ \psi_1 \quad \psi_2 \quad \psi_3 \quad \psi_4 \quad \psi_5 \]

\{ MPI 0 \quad MPI 1 \quad MPI 2 \}
Coefficient distribution scheme

Used for scalar product & orthonormalisation

BLAS routines (level 3) are called, then result is reduced

Communications are performed via MPI_ALLTOALLV
Performance in parallel (BigDFT)

Distribution per orbitals (wavelets)
Nothing to specify in input files
**GPU-ported operations in BigDFT (double precision)**

**Convolutions (OpenCL rewritten)**

GPU speedups between 10 and 20 can be obtained for different sections.

**Linear algebra (CUBLAS library)**

The interfacing with CUBLAS is immediate, with considerable speedups.
BigDFT code on Hybrid architectures

BigDFT code can run on hybrid CPU/GPU supercomputers

In multi-GPU environments, **double precision** calculations

**No Hot-spot operations**

Different code sections can be ported on GPU up to 20x speedup for some operations, 7x for the full parallel code
OpenCL acceleration:

Add this line in `input.perf`:

```
accel OCLGPU
```

See the web page


for more information about all keywords (verbosity, input guess, ...)
associated to the performance of the BigDFT code.

The log file gives the list of possible keywords.
BigDFT: Future developments

\[-\frac{1}{2} \nabla^2 + V(r) + \mu_{xc}(r) \psi_i^k = \varepsilon_i^k \psi_i^k\]

- pseudopotential
- norm-conserving
- PAW
- Hybrid functionals
- beyond LDA
- GW
- LDA, GGA
- LDA+U

- self-consistent (SCF)
- Harris-Foulkes functional
- relativistic
- all electrons
- non-relativistic
- non-periodic
- periodic
- \(N^3\) scaling
- \(O(N)\) methods

- non-spin polarized
- spin polarized
- non-collinear
- collinear
- atomic orbitals
- plane waves
- real space
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- finite difference
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- collinear
- atomic orbitals
- plane waves
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- numerical
- finite difference
- Wavelet
Linear scaling version (order N)

16 min of CPU time:
- 1000 atoms with the cubic scaling,
- 8000 atoms with the linear scaling one

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Localization regions (L. Ratcliff, P. Boulanger)

Wavelets have compact support

- The behaviour of large systems is short-ranged
- Wavelets are ideal to implement nearsightedness principle
- The hamiltonian can be expressed analytically
- We can adapt the basis based on system’s properties

Reduce DoF

The domain can be partitioned in localization regions

What about KS orbitals?
Improving the locality of the description

Localized description of the system

KS orbitals expressed by *adaptive support functions*:

\[ \Psi_i(r) = \sum_\alpha c_{i\alpha} \phi_\alpha(r) \]

With \( \phi_\alpha \) given in wavelets on the localization regions

We can therefore optimize both \( c_{i\alpha} \) and \( \phi_\alpha \), e.g. as in ONETEP, Conquest, SIESTA . . . or BigDFT.

Localized form for the Density and the Hamiltonian

\[
\rho(r) = \sum_i f_i \Psi_i(r) \Psi_i^*(r) = \sum_{\alpha,\beta} \phi_\alpha(r) K^{\alpha\beta} \phi_\beta^*(r)
\]

\[
H = \sum_{\alpha,\beta} \phi_\alpha(r) H^{\alpha\beta} \phi_\beta^*(r)
\]
Overall scheme

Initial Guess:
Atomic Orbitals

Basis optimization:
\[
\min_{\phi_\alpha} \sum_{\alpha,\beta} K_{\alpha\beta} \langle \phi_\alpha | H_{KS} | \phi_\beta \rangle
\]
or
\[
\min_{\phi_\alpha} \sum_{\alpha} \langle \phi_\alpha | H_{\alpha} | \phi_\alpha \rangle
\]

Kernel Optimization:
Diagonalize Hamiltonian
Update potential

Forces:
Derivative basis functions

Two-step optimization scheme
- The $\phi_\alpha$ minimize the “trace” of a confining KS hamiltonian
- Coefficients minimize KS energy

High quality results
- Good precision
- No need of Pulay forces due to basis completeness!
Nearsightedness and wavelets: it works (very) well!
- Convergence is not altered by system size
  - Basis set is physically meaningful
- KS operators are expressed in this basis without truncation
  - Basis set close to completeness
- Different schemes can be implemented with this basis set: $O(N)$, embedding, QM/MM, TB, . . .

Considerable gain
Example: 256 MPI tasks
- Crossover point at few hundred atoms
  - Lower with Geometry optimization (easier restart)

<table>
<thead>
<tr>
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<td>0</td>
<td>0</td>
</tr>
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<td>0.5</td>
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</tr>
<tr>
<td>4000</td>
<td>6</td>
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<td>2</td>
</tr>
</tbody>
</table>

Order N: Results and performances

- Convergence is not altered by system size
  - Basis set is physically meaningful
- KS operators are expressed in this basis without truncation
  - Basis set close to completeness
- Different schemes can be implemented with this basis set: $O(N)$, embedding, QM/MM, TB, . . .

Considerable gain
Example: 256 MPI tasks
- Crossover point at few hundred atoms
  - Lower with Geometry optimization (easier restart)
### Accuracy for sample systems

Energy and forces with accuracy of systematic approach!

<table>
<thead>
<tr>
<th>Chemical Formula</th>
<th>Energy (Ha)</th>
<th>Forces (Ha/bohr)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Linear</strong></td>
<td><strong>Cubic</strong></td>
<td><strong>(L - C)/at</strong></td>
</tr>
<tr>
<td>C\textsubscript{300}H\textsubscript{602}</td>
<td>-2062.98</td>
<td>-2062.98</td>
</tr>
<tr>
<td>H\textsubscript{2}O</td>
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<td>-412.03</td>
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<tr>
<td>C\textsubscript{19}H\textsubscript{22}N\textsubscript{2}O</td>
<td>-157.04</td>
<td>-157.04</td>
</tr>
<tr>
<td>B\textsubscript{80}</td>
<td>-225.68</td>
<td>-225.69</td>
</tr>
</tbody>
</table>
Charged systems

Basis represents physical degrees of freedom

$\Delta$-SCF straightforwardly implemented in this formalism

Increasing chain length of ladder polythiophene

Charge-Constrained DFT is under implementation
What about wavelets for Post-DFT (excited states)?

Initial work: excited states in isolated systems
- Free boundary conditions are explicitly implemented
- The number of interesting states increases with the simulation domain (continuum collapse)

Problem in TD-DFT and GW for large isolated systems

Resolvent reads

\[ G(\omega) = \sum_{i=1}^{\# \text{bound states}} \frac{|\psi_i(\psi_i|}{\omega - E_i} + \int dk \frac{|\psi_k(\psi_k|}{\omega - E_k} \]

Sum Over States is out of reach ... or not?
Resonant States: Motivation

Simplest example: **cross section** computation

A function of the resolvent: $S(\omega) = -\frac{1}{\pi} \text{Im} \langle \psi_0 | \hat{O}^\dagger G(\omega) \hat{O} | \psi_0 \rangle$

Is it possible to associate to each peak a state

- **discrete** (metastable),
- physically justified?

... Yes, but ...
The complex scaling allows this!

### Resonant States

#### Rigorous definition of the Green’s function:

\[
G(\omega) = \sum_i \frac{|\psi_i(\omega)|^2}{\omega - E_i} + \sum_{r=1}^{\# \text{res. states}} \frac{|\psi_r(\omega)|^2}{\omega - E_r} + \ldots
\]

- Complex energy → The resonant states explode!
- The “interesting” states are not in $L^2$ space (norm not finite)

### The complex scaling


\[
x \rightarrow xe^{i\theta} \Rightarrow \hat{H} \rightarrow \hat{H}_\theta \equiv -\frac{e^{-2i\theta}}{2} \nabla^2 + V(xe^{i\theta})
\]

With this transformation (some) resonant states become $L^2$!

### So far a big problem

Not possible for numerical (e.g. DFT) potentials!
Complex Scaling Method in Wavelets

Recent work arxiv:1303.6439
Wavelets for complex scaling
- Can scale any 3D ab-initio (numerical) potential
- Provide the results on a numerical grid
- Very high precision

Ongoing work
- A important bottleneck has been removed
- Resonant states extraction procedure
- Usage in conjunction with $\phi_\alpha$
Extracting information coming from resonant states

A meaningful one-particle excitations basis

- Unified description of bounds and metastable states
- Encode system’s physical (metastable) excitations
- Appear as eigenstates of a complex-rotated hamiltonian

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

Interest - driven developments

Unprecedented reliability for some approaches
  → new things can be done

Wavelet properties open new investigation directions:
  - Traditional DFT functionalities have been implemented
    ... and counting ...
  - The formalism is explicit and simplifies some
    approaches
  - A different way of thinking to a development problem

Future directions

- Improve locality of big systems (embedding QM/QM,
  use of Tight-Binding method)
- Usage of θ-resonant states for electronic properties
- Not to mention HPC involvement...
Conclusion

- **Functionality**: Mixed between physics and chemistry approach

- Order N (real space basis set as wavelets)

- Multi-scale approach (more than 1000 atoms feasible for a better parametrization)

- Beyond DFT (TD-DFT and GW methods with the resonant states)

- **Numerical experience**:
  - One-day simulation
  - Better exploration of atomic configurations
  - Molecular Dynamics
    - (4s per step for 32 water molecules)
Acknowledgments

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Order N methods  S. Mohr, L. Ratcliff, P. Boulanger (Néel Institute, CNRS)

Exploring the PES  N. Mousseau (Montreal University)

Resonant states  A. Cerioni, A. Mirone (ESRF, Grenoble), I. Duchemin (CEA)

Optimized convolutions  B. Videau (LIG, computer scientist, Grenoble)

Applications  P. Pochet, E. Machado, S. Krishnan, D. Timerkaeva (CEA-Grenoble)
Hands on

See


- First runs with BigDFT
  - Basis-set convergence
  - Acceleration example on different platforms: *Kohn-Sham DFT Operation with GPU acceleration*