WAVELETS IN ELECTRONIC STRUCTURE CALCULATIONS

Claire Chauvin, *Ph-D student*

Condensed Matter Fundamental Research Department
- Grenoble, France -

Valérie Perrier, Stefan Goedecker

Physical context: electronic structure calculation

- Physical description.
- A non-linear problem.
- Algorithm.
- Issues.

Kind of wavelets used

Solving the self-consistent model

PHYSICAL DESCRIPTION

A SIMPLIFIED MODEL TO DETERMINE THE ELECTRONIC DENSITY

- Ω 3D domain with periodic or Dirichlet boundary conditions.
- Let M nuclei $\{Z_I,\ R_I\}_{1\leqslant I\leqslant M}$ supposed to be fixed. We want to characterize the behaviour of N associated electrons, by means of the electronic density:
  $\rho(\vec{r}) = \sum_{i=1}^{N} |\Psi_i(\vec{r})|^2$ such that $\int_{\Omega} \rho(\vec{r}) d\vec{r} = N$.
- The orbitals $\Psi_i \in H^2(\Omega)$ are solutions of the following non-linear system, associated to the N greatest energies ε_i:

$$\mathcal{H}\Psi_i = \varepsilon_i \Psi_i,$$

with $\mathcal{H} = -\frac{1}{2}\Delta + \mathcal{V}[\rho] = -\frac{1}{2}\Delta + V_{ext}(\vec{r}) + V_C[\rho(\vec{r})] + V_{xc}[\rho(\vec{r})]$

PHYSICAL DESCRIPTION

$$\mathcal{H} = -\frac{1}{2}\Delta + \mathcal{V}[\rho],$$
$$\mathcal{V}[\rho] = V_{ext}(\vec{r}) + V_C[\rho(\vec{r})] + V_{xc}[\rho(\vec{r})]$$

$\mathcal{V}$ is the potential; it describes the particle interactions, by means of the non-linear terms: the coulombial interaction V_C, and the correction V_{xc}.

$$V_{ext}(\vec{r}) = -\sum_I \frac{Z_I}{|\vec{R}_I - \vec{r}|} + \sum_{I,J} \frac{Z_I Z_J}{|\vec{R}_I - \vec{R}_J|},$$
$$V_C[\,\rho(\vec{r})] = \int_\Omega \frac{\rho(\vec{r}\,')}{|\vec{r} - \vec{r}\,'|} d\vec{r}\,',$$
$$V_{xc}[\,\rho(\vec{r})] = \mathbf{F}[\rho]$$

Example 1 *The hydrogen atom contains only one term:* $V_{ext}(\vec{r}) = -\frac{1}{|\vec{R}_I - \vec{r}|}$.

Remark 1 *The Coulombial interaction* V_C *is the solution of:*

$$\Delta V_C = -4\pi\,\rho$$

THE NON-LINEAR PROBLEM $\mathcal{H}\Psi_i = \varepsilon_i \Psi_i$

The objective is the minimization of the total energy, using the solutions $\{\varepsilon_i, \Psi_i\}$, which charaterizes the ground state of the system:

$$\mathcal{E}[\rho] = \sum_{i=1}^{N} \varepsilon_i + \mathbf{E}[\rho, V_C, V_{xc}]$$

with $||\Psi_i||_2 = 1$, $\forall\, i \ = \ 1, \ldots, N$.

$\Longrightarrow$ this non-linear problem is called a self-consistent system, which will be solved by the following iterative algorithm:

- At iteration k, by knowing $\tilde{\rho}^k$, we have to solve:

$$\mathcal{H}[\tilde{\rho}^k]\Psi_i^{k+1} = \varepsilon_i^{k+1}\Psi_i^{k+1}, \quad 1 \leqslant i \leqslant N$$

- We mix the last $s-1$ input $\tilde{\rho}^m$ to form a new $\tilde{\rho}^{k+1}$.

EXISTENCE OF EIGENVALUES

$$\mathcal{H}[\rho^k]\Psi_i^{k+1} = \varepsilon_i^{k+1}\Psi_i^{k+1}$$

Theorem 1 *Virial: For a potential behaving like $\frac{1}{|r|}$, we have the following identity:*

$$\mathcal{E}_t = -\mathcal{E}_k = \frac{1}{2}\mathcal{E}_p$$

Result of eigenspace existence:

- $\mathcal{H}$ self-adjoint, but bounded.
- estimation

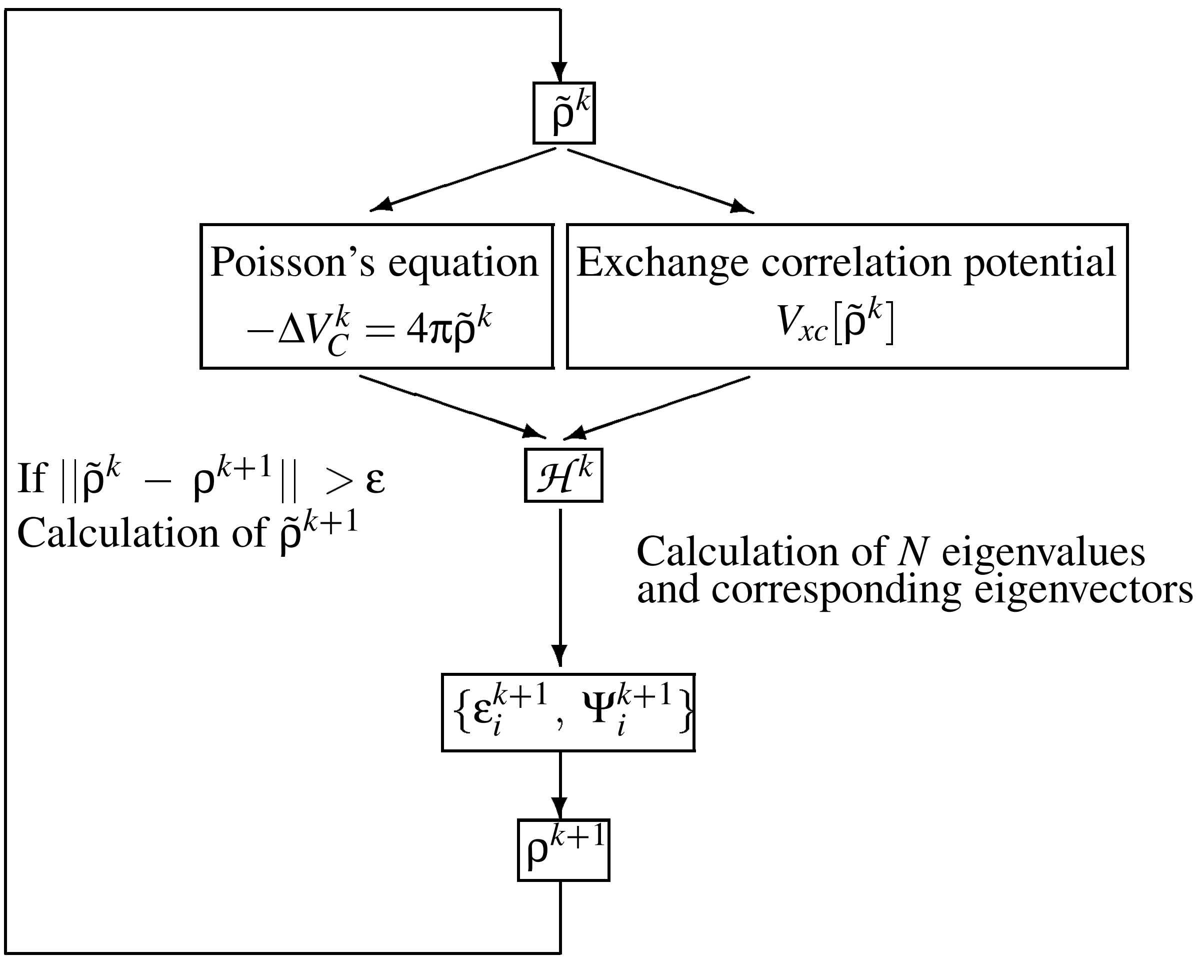
THE SELF-CONSISTENT PART

$\rho^k \Longleftarrow \tilde{\rho}^{k+1}$

Let $\tilde{\rho}^{k+1},\ \rho^k,\ ,\ldots,\rho^{k-s+2}$ the last $s-1$ input densities plus the current output density.

By minimizing the residual of $\hat{\rho}^k = \sum_{i=1}^{s} \alpha_i \rho^{k-i}$, so that $\sum \alpha_i = 1$, we are ensured that we minimize the difference between ρ^k and $r\tilde{h}o^{k+1}$. (Pulay,1980, Bowler, 2001)

ALGORITHM



$\tilde{\rho}^k$
Poisson's equation
$-\Delta V_C^k = 4\pi\tilde{\rho}^k$
Exchange correlation potential
$V_{xc}[\tilde{\rho}^k]$
$\mathcal{H}^k$
If $||\tilde{\rho}^k - \rho^{k+1}|| > \varepsilon$
Calculation of $\tilde{\rho}^{k+1}$
Calculation of N eigenvalues
and corresponding eigenvectors
$\{\varepsilon_i^{k+1}, \Psi_i^{k+1}\}$
ρ^{k+1}

EXISTING METHODS

Let Ψ_i be decomposed on a basis: $\Psi_i = \sum_\alpha c_\alpha \phi_\alpha$.

- Gaussians (GAUSSIAN):

  For non periodic systems — redundant a lot of parameters

  Centered at the positions of the nuclei — no preconditioning for the Laplacian

- Plane waves - Fourier (CPMD, ab-init, VASP):

  For any kind of system — Very good solver for Poisson.

  High resolution for good approximation — *Not adaptive*

- Mixed basis: Gaussians + Plane waves (WIEN)

  Problems of domain decomposition — Heavy code

  The more widespread

- Wavelets: (???????)

  A new development — Adaptive

  More complex solver for Poisson's equation

`Physical context: electronic structure calculation`

`Kind of wavelets used`

- Three-dimensional biorthogonal MRA.
- Regularity and vanishing moments
- Approximation property

`Solving the self-consistent model`

3D BIORTHOGONAL MRA

- Let $\{V_j\}$ and $\{\tilde{V}_j\}$ be two biorthogonal MRA of $L^2(\mathbb{R})$, with scaling functions ϕ_λ and $\tilde{\phi}_\lambda$, satisfying:

$$< \phi_k, \tilde{\phi}_l >= \delta_{k,l}$$

- The associated biorthogonal wavelets $\{\psi_\lambda, \tilde{\psi}_\lambda\}$ generate basis of spaces $W_{j+1} = V_{j+1} \cap (\tilde{V}_j)^\perp$ and $\tilde{W}_{j+1} = \tilde{V}_{j+1} \cap (V_j)^\perp$. We thus get two L^2-stable multiscale decompositions in the basis

$$\{\phi_{j_0,k}\}_{k\in\mathbb{Z}} \cup \{\psi_{j,k},\, j \geqslant j_0,\; k \in \mathbb{Z}\}$$ and its dual.

3D BIORTHOGONAL MRA

$\Omega = [0,\ L[^3$. We consider the isotropic 3-dimensionnal MRA in the periodic case:

Definition 1 *Three-dimensional biorthogonal MRA on Ω:*

$$\mathbb{V}_j(\Omega) = V_j \otimes V_j \otimes V_j, \quad \tilde{\mathbb{V}}_j = \tilde{V}_j \otimes \tilde{V}_j \otimes \tilde{V}_j,$$

Let $\varepsilon = (\varepsilon_1, \varepsilon_2, \varepsilon_3)$, *with* $\varepsilon_i \in \{0,1\}$ *and* $\varepsilon \neq \{(0,0,0)\}$. *Taking* $W_j^0 = V_j$ *and* $W_j^1 = W_j$, *we introduce the wavelet isotropic space:*

$$\mathbb{W}_j(\Omega) = \oplus_{\varepsilon=\{0,1\}^3 \setminus \{(0,0,0)\}} \otimes_{m=1}^{3} W_j^{\varepsilon_m}$$

$\Longrightarrow$ Seven generating wavelets in 3D, products of 1D basis functions **of the same scale**.

This kind of basis leads to efficient Fast Wavelet Transform: with tabular structures adapted to the language (Fortran), we get with good performances.

Properties of the wavelets

- The MRA is of order m if $\mathbb{P}_{m-1} \subset V_j$. This condition is equivalent for $\tilde{\psi}$ to have m vanishing moments.
- If ϕ and $\tilde{\phi}$ are in L^2, and $\{V_j\}$ is an m-order MRA, then:

$$\forall f \in H^m,\ ||f - P_j f|| \leqslant C\, 2^{-js} ||f||_s,\ \forall s \leqslant m$$

- If $\phi \in H^m$, then $\forall f \in V_j$ we have:

$$||f||_s \leqslant C\, 2^{sj} ||f||,\ \forall s \leqslant m$$

- If these two conditions are fulfilled, then we have the equivalence between the H^s-norm of f and the l^2-convergence of its wavelet coefficients $2^{js} < f, \tilde{\psi}_{j,k} >$.

WAVELET FAMILIES CONSIDERED

- Daubechies wavelet families: orthogonal, with minimal support for a given number of vanishing moments.
- *Orthogonal Coiflets*: Primal scaling functions have also vanishing moments.
- *Biorthogonal interpolating wavelets*: the Deslauriers-Dubuc scaling function θ is associated to the Dirac function as dual scaling function. We can also construct regular one using the *lifting scheme* (Sweldens, 1996): we obtain a new wavelet with as many vanishing moments as we want.

Physical context: electronic structure calculation

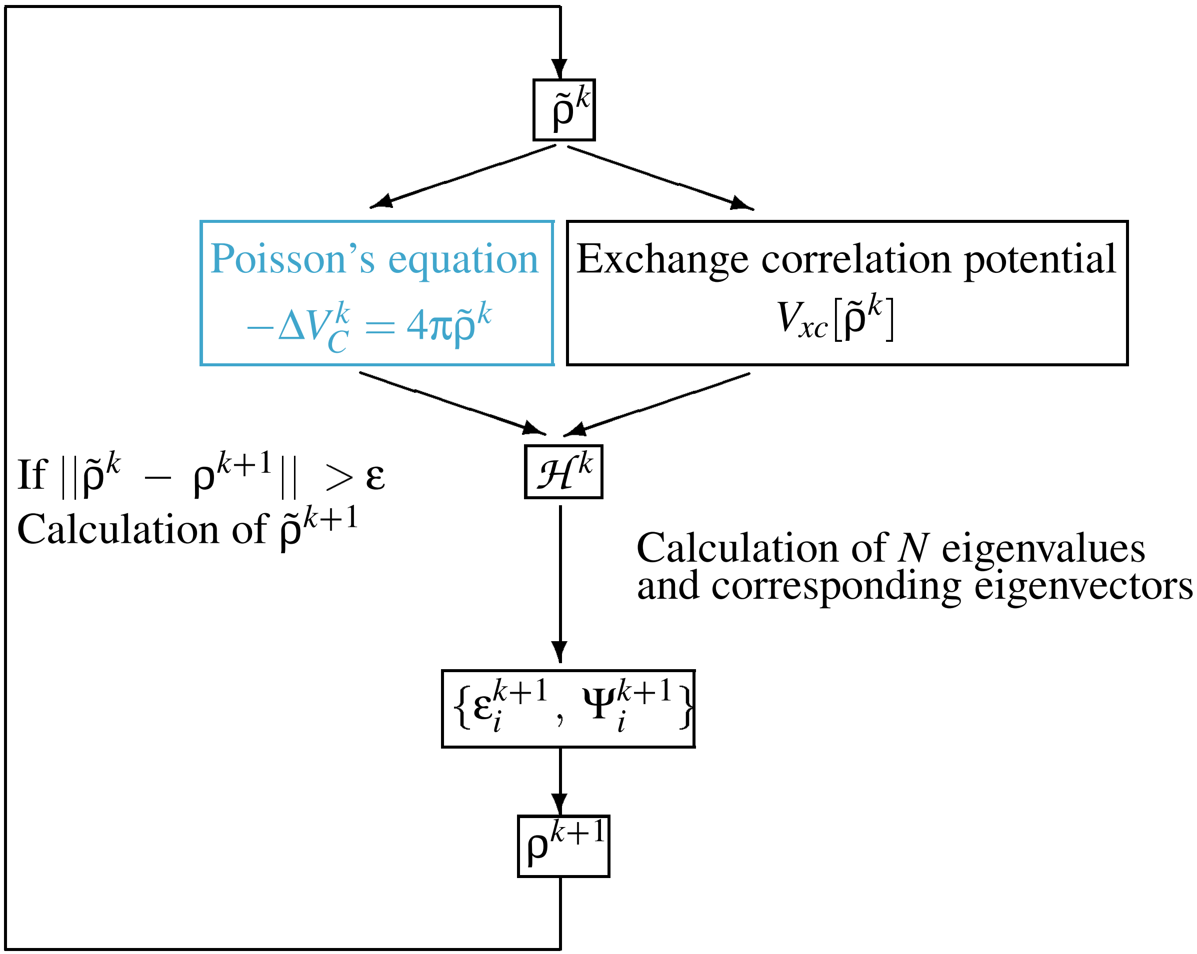
Kind of wavelets used

Solving the self-consistent model

- A solver for Poisson's equation.
- Test-case: the hydrogen atom and gaussian potentials.
- The self consistency.

Toward a self-consistent resolution

ALGORITHM



About the preconditionning in Galerkin method

Condition number of Galerkin stiffness matrices (1D) $\int \nabla\psi_\alpha \nabla\psi_\beta$.

	Daubechies 6		Daubechies 10		Interpolating		Dual lifted	
Length filter	6		10		6		8	
PREC?	No	Yes	No	Yes	No	Yes	No	Yes
128	5818	11.51	4310	4.92	119359	38	140	9.14
1024	372356	13.04	275864	4.98	61112070	47	1120	9.29

Preconditionning using the exact diagonal.

The length filter N satisfies $N = 2d$, where d is the vanishing moment of the dual wavelet.

About the preconditionning in Petrov-Galerkin method

Condition number of Petrov Galerkin 1D stiffness matrices $\int \nabla\psi_\alpha \nabla\tilde{\psi}_\beta$.

d	Interpolating 6			Interpolating 8	
$\tilde{d}$	2-lifted		4-lifted	2-lifted	4-lifted
PREC?	No	Yes			
128	4511	5.68	5.01	4.56	4.23
1024	288759	5.99	5.18	4.58	4.23

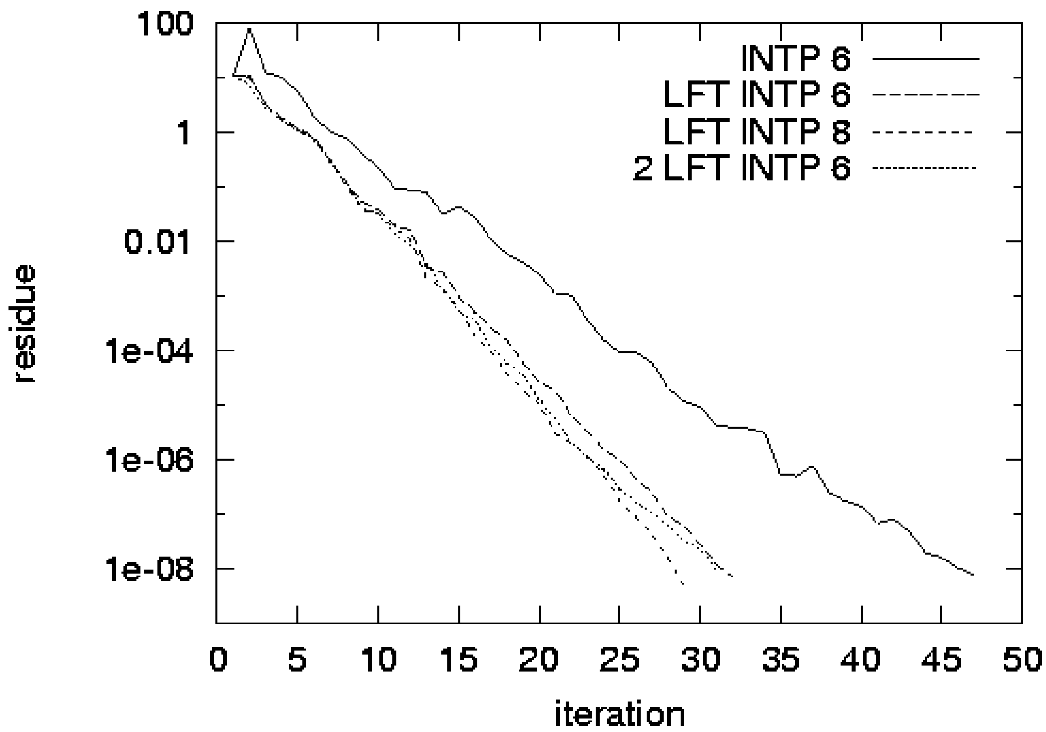
Preconditionner: exact wavelet diagonal.

Conditionning number calculated using the Singular Value Decomposition.

Conclusion:

In Petrov-Galerkin Method

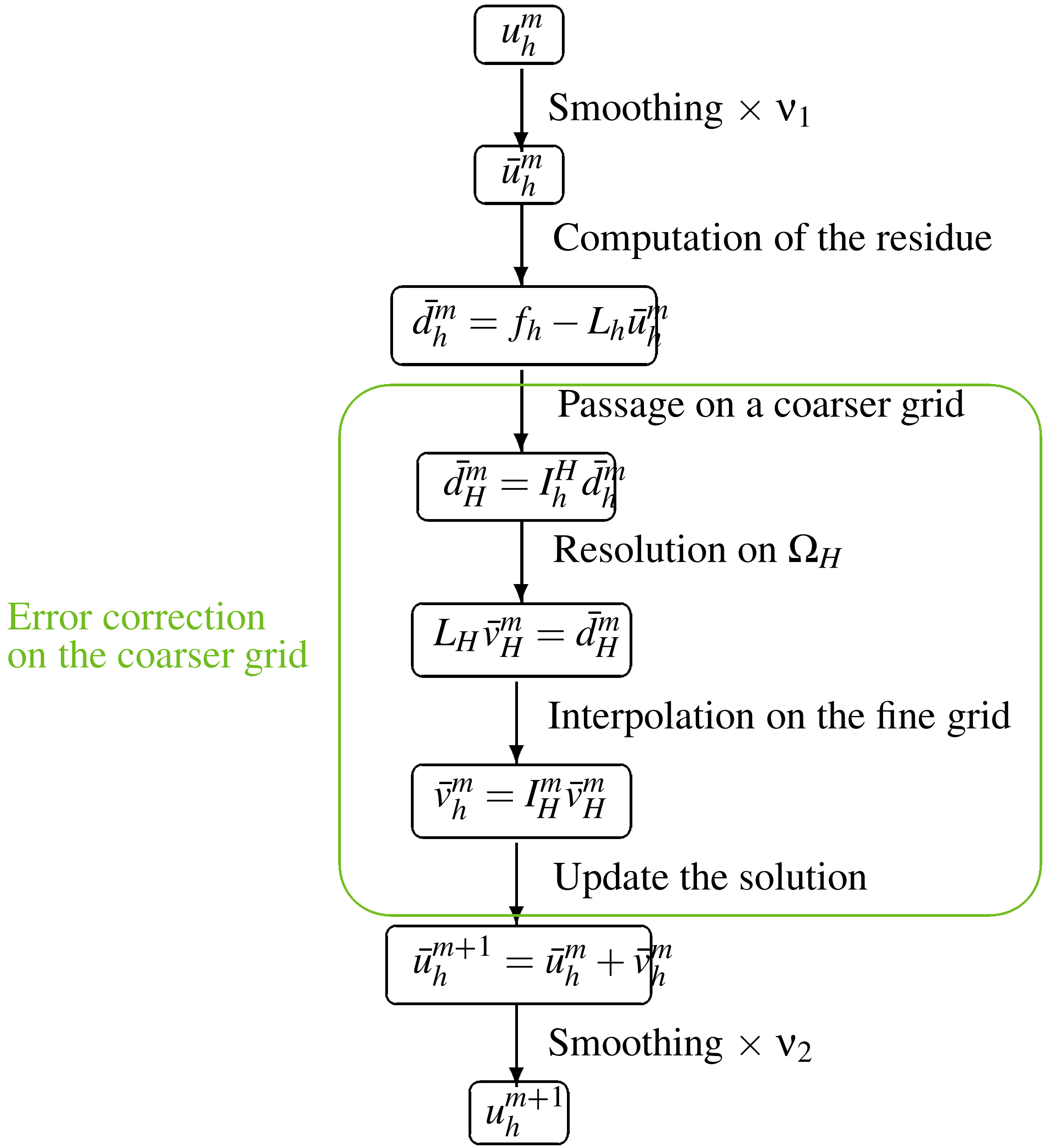
Error versus number of GMRES iterations, for a 3D resolution, in Petrov Galerkin approximation (64^3 points), using the exact diagonal preconditionning.



$H = 2^{j-1}$ and $h = 2^j$.

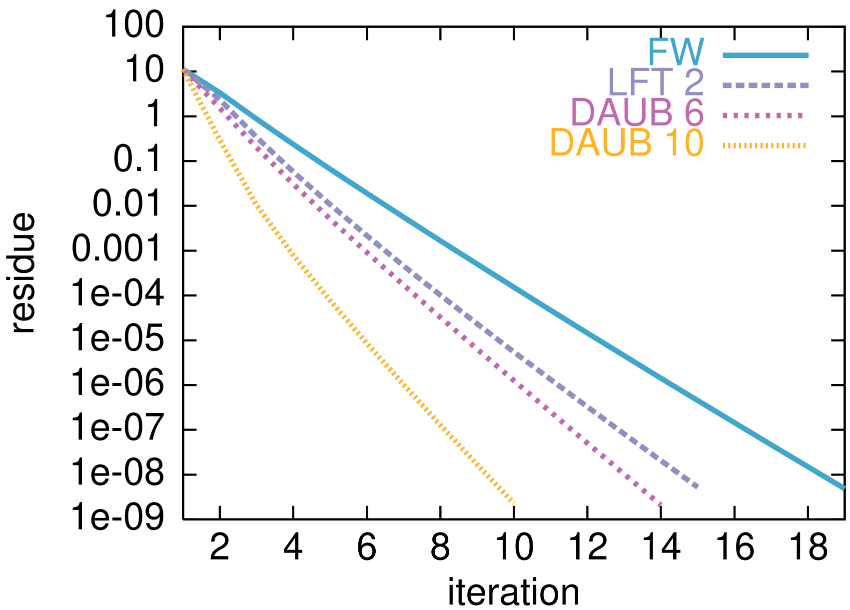
Smoother: Gauss Seidel

$I_h^H = P_{j-1}, I_H^h = P_j$.

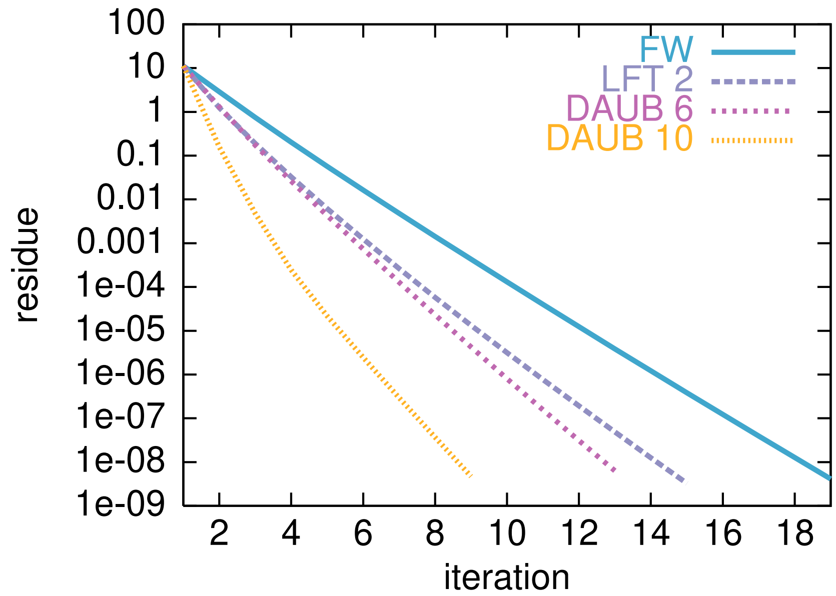


A SOLVER FOR POISSON'S EQUATION
A WAVELET-MG METHOD

Convergence rates for the standard V-cycle, and the modified V-cycle. Fine grid composed of 64^3 points, and four levels of resolution. Differential operator discretized with a 6-order finite differences.



Standard multigrid



Wavelet-multigrid

THE NON SELFCONSISTENT MODEL

We have to find the N greatest eigenvalues ε_i and the associated eigenvectors Ψ_i of the problem:

$$\mathcal{H}\Psi_i^{k+1} = \varepsilon_i \Psi_i^{k+1},$$
$$\mathcal{H} = -\frac{1}{2}\Delta + \mathcal{V}$$

to form the density: $\rho = \sum_{i=1}^{N} |\Psi_i|^2$.

ε_i is composed of the kinetic energy $\int \nabla\Psi_i \nabla\Psi_i$ and the potential energy $\int \Psi_i \mathcal{V} \Psi_i$.

- Calculation of the Potentiel Energy.
- Example of simulation: the hydrogen case.
- Error in terms of the family.

THE NON SELFCONSISTENT MODEL
CALCULATION OF THE POTENTIEL ENERGY

$$\mathcal{E}_p = \int_\Omega \Psi_i \mathcal{V} \Psi_i$$

Ψ_i is developed on an orthogonal basis of $\mathbb{V}_j$, whereas $\mathcal{V}$ is expand in terms of interpolating wavelets $\mathbb{V}'_j$.

$$\Psi_i(r) = \sum_k c_{j,k} \phi_{j,k}(r),$$
$$V(r) = \sum_l v_{j,l} \theta_{j,l}(r)$$

Two possibilities:

- Evaluation of $\int \phi_k(r)\theta_l(r)\phi_m(r)dr$
- Calculate $|\Psi_i|^2$ on the physical grid made of $(2^{j+1})^3$ points.

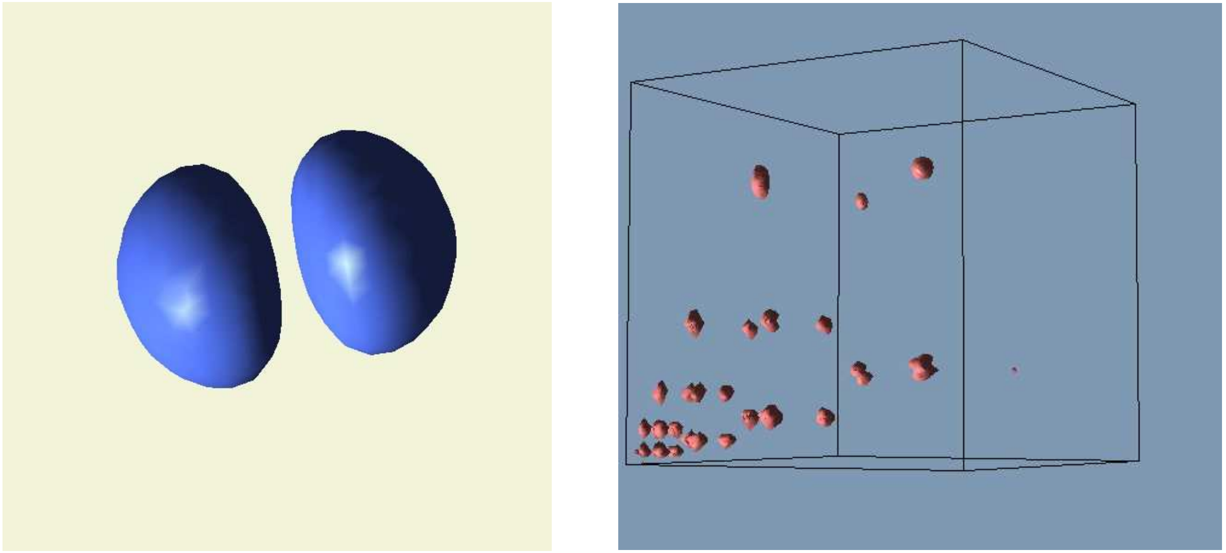
Error in function of wavelet families

- One nuclear of charge $Z = 1$, positionned at $R \in \Omega$.
  $\mathcal{V} = \mathcal{V}(\vec{r}) = -\frac{1}{|R-r|}$.
- The exact energy of the electron is -0.5.

Density	Interpolating 8	Coifman 2	Daubechies 8
Daubechies 8	-.49944	-.49937	-.49929
Daubechies 6	-.49890	H	H
Coifman1	-.49943	-.49938	
Coifman2	-.49947	-.49942	

Eigenvalue calculated for 64^3 points. Row: orthogonal filter used to expand eigenfunction. Column: filter used to expand the density in the $(2^{j+1})^3$ real space grid.

EXAMPLE OF A SIMULATION
THE POTENTIAL IS A GAUSSIAN.



3D isosurface of the second eigenfunction density $|\Psi_2|^2$ and its wavelet coefficients greatest than 10^{-2}.

For such an orbital, 44% of the wavelets coefficients are greatest than 10^{-10}.

FORECASTS AND PERSPECTIVES

- Compute the self-consistent scheme.
- Test the convergence of the algorithm.
- Introduce physical potentials for an atom.
- Computation with several atoms.
- Devise and develop a wavelet adaptive scheme.