WP8:
Materials Science

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ICMAB

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==> ICMM

CIN2 CENTRE D'INVESTIGACIÓ EN NANOCIÈNCIA I NANOTECNOLOGIA CAMPUS UAB. BELLATERRA. BARCELONA

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UAM UNIVERSIDAD AUTONOMA DE MADRID

Jose M. Soler (IP)

Félix Ynduráin

CIC nanoGUNE nanoscience cooperative research center

Emilio Artacho

Fabiano Corsetti

BSC

CASE group

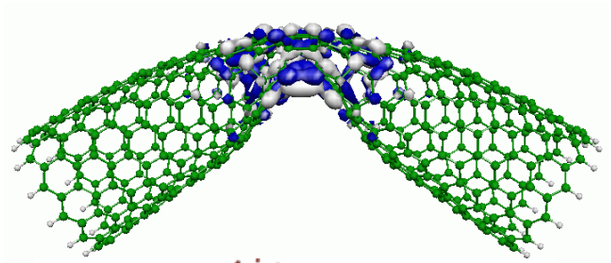
Georg Huhs

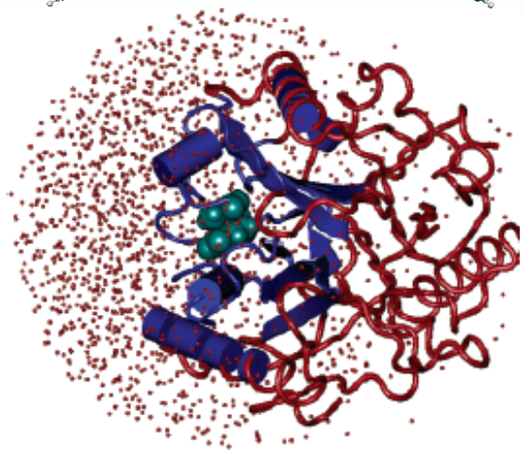
José María Cela

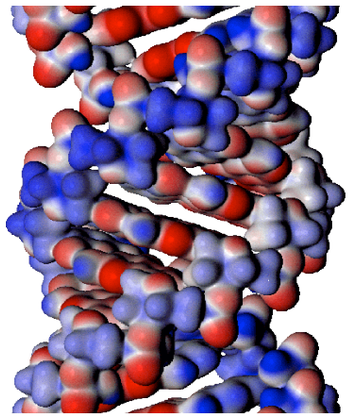
Rogeli Grima

First-Principles calculations at the atomic scale

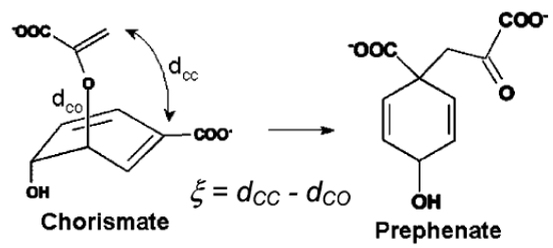
- Electronic Structure
- Atomic forces and stresses
- Molecular Dynamics

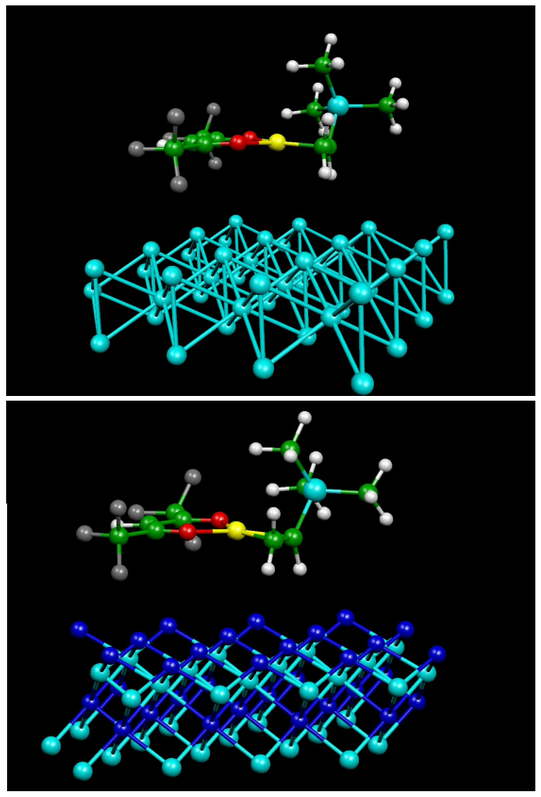






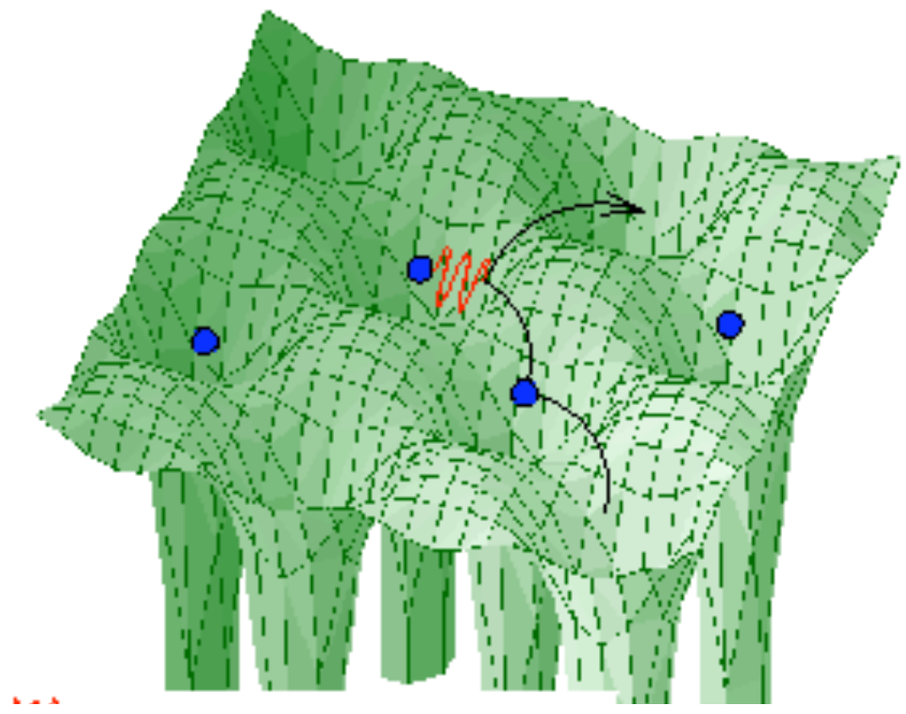
Scheme 1. Chorismate to Prephenate Conversion Reaction





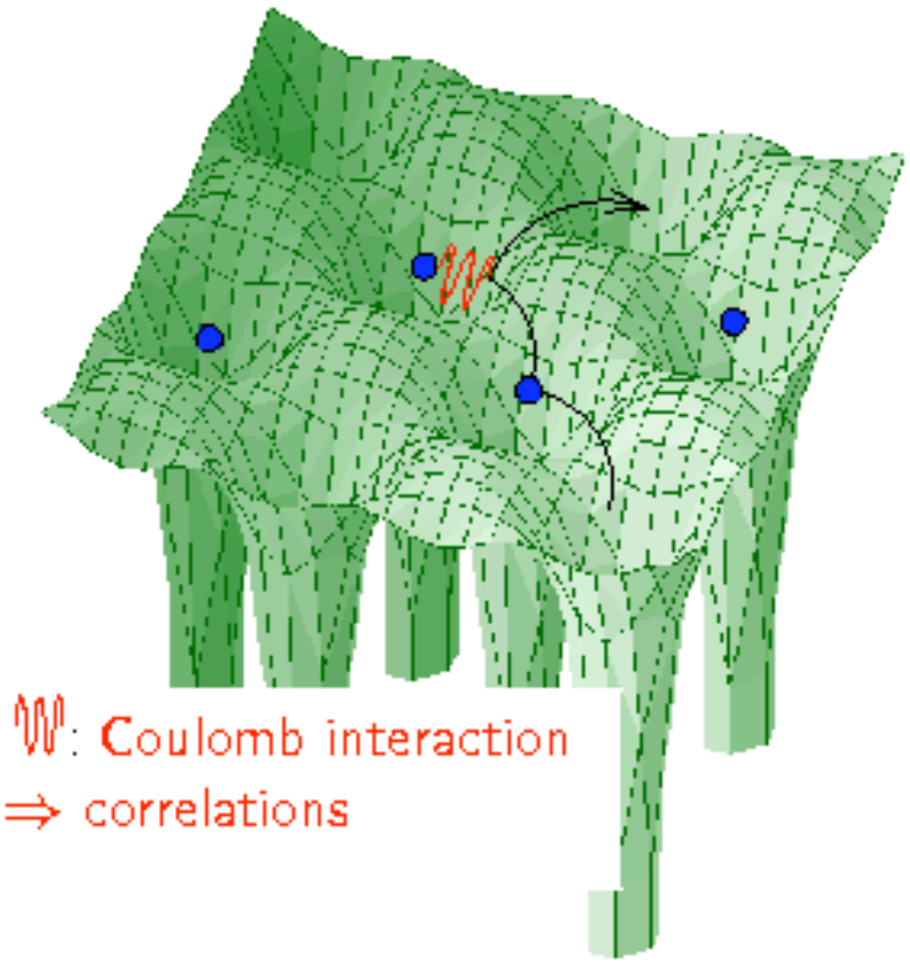
The "ultimate model" for electrons in a material

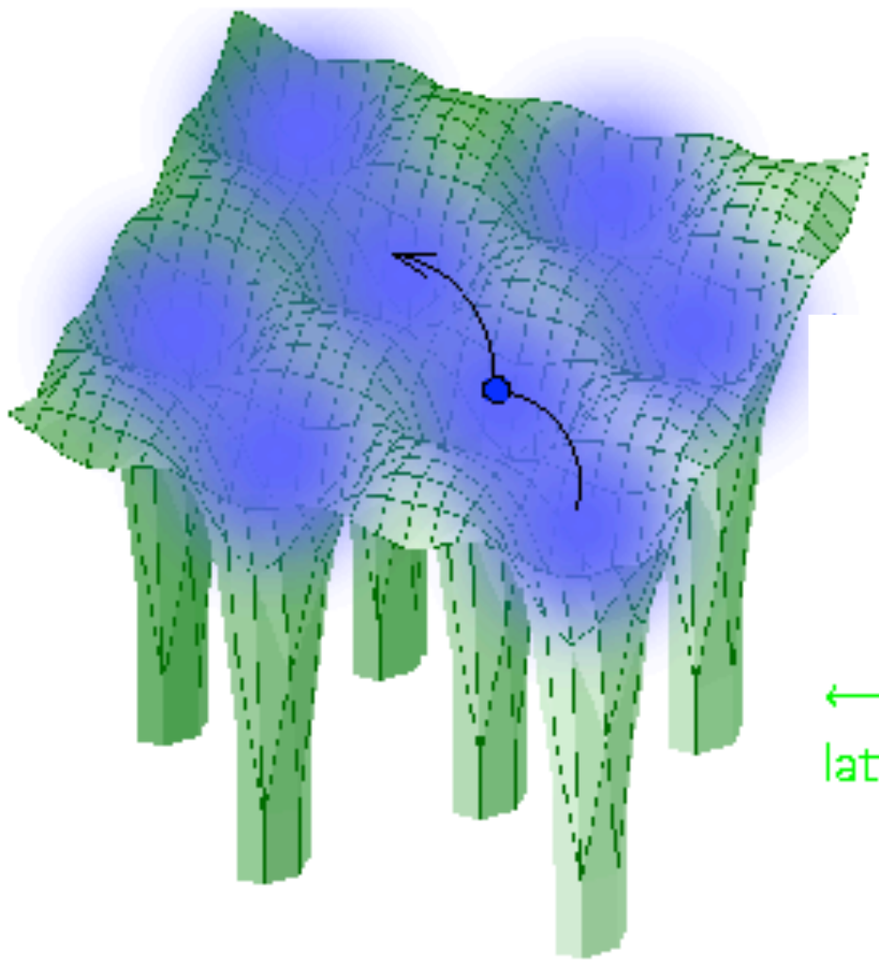
$$H = \sum_i \left[-\frac{\hbar^2 \Delta_i}{2m_e} + \sum_l \frac{-e^2}{4\pi\epsilon_0} \frac{Z_l}{|\mathbf{r}_i - \mathbf{R}_l|} ; \right] + \frac{1}{2} \sum_{i \neq j} \frac{e^2}{4\pi\epsilon_0} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$



$$\hat{H}\Psi = E\Psi \qquad \Psi(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n)$$

We could compute "everything"





Density-functional theory $\quad E = E[n] \quad n(\mathbf{r})$

$$\{-\nabla^2 + V_{\text{eff}}[n](\mathbf{r})\}\psi_i = \varepsilon_i \psi_i$$ One electron

$$V_{\text{eff}}[n](\mathbf{r}) = V_{\text{ext}}(\mathbf{r}) + V_{\text{H}}[n](\mathbf{r}) + V_{\text{xc}}[n](\mathbf{r})$$

The SIESTA method for ab-initio materials simulation

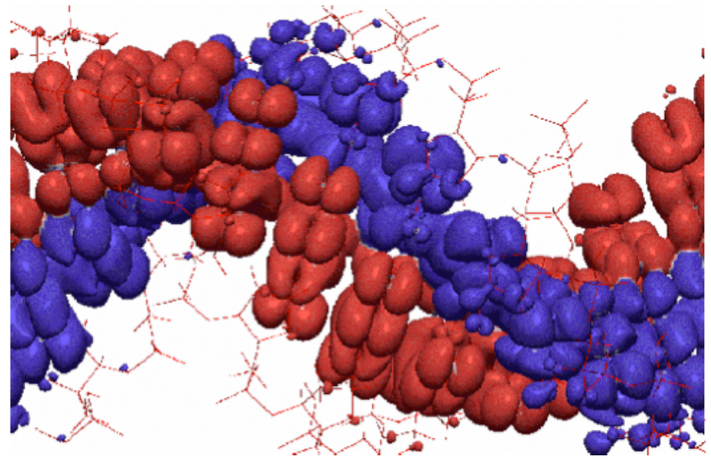
Soler, Artacho, García, Gale, Junquera, Ordejón, Sánchez-Portal

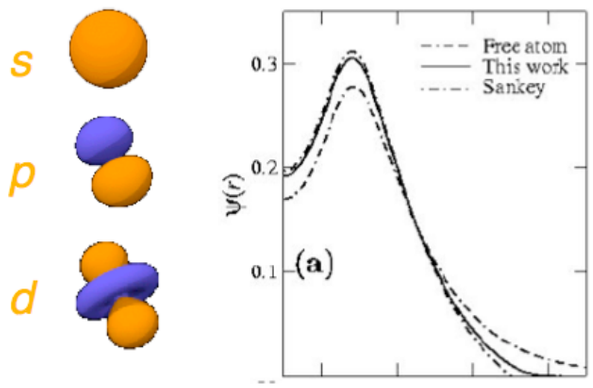
J. Phys. Cond. Matt (2002)

More than 3600 citations

More than 1000 active users

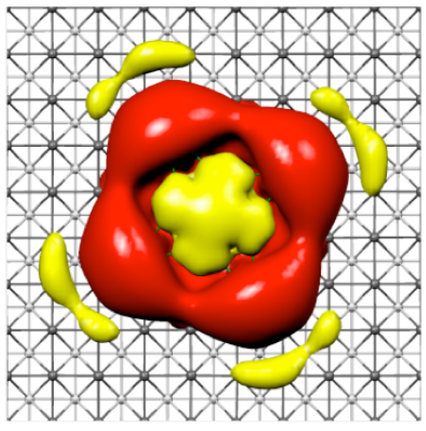
HOMO and LUMO of DNA
(800 atoms, small computer)
Linear-scaling calculation (1999)





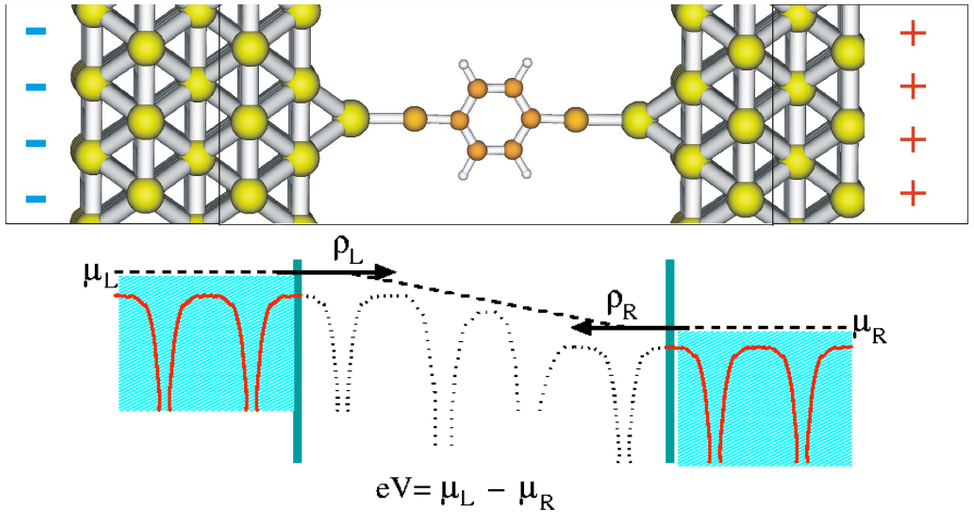
Localized atomic orbitals

Cu phthalocyanine
on Ag(001)
1400 atoms



TranSIESTA: Electronic transport

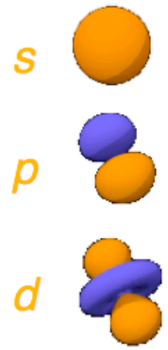
Brandbyge, Mozos, Ordejón, Taylor and Stokbro
Phys. Rev. B. (2002) --- 900 citations

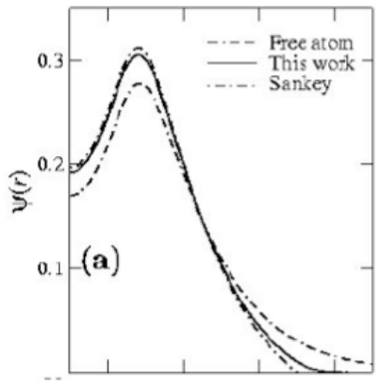


OBJECTIVES

- **Optimization of the SIESTA code**
- **Performance of a “Grand-Challenge” calculation**

$$\psi_i(\boldsymbol{r}) = \sum_\mu \phi_\mu(\boldsymbol{r}) c_{\mu i},$$





$$H^{\alpha\beta}_{\mu\nu} = \langle \phi_\mu | \hat{T} + \hat{V}^{KB} + V^{NA}(\boldsymbol{r}) + \delta V^H(\boldsymbol{r}) + V^{\alpha\beta}_{XC}(\boldsymbol{r}) | \phi_\nu \rangle$$

$$S_{\mu\nu} = \langle \phi_\mu | \phi_\nu \rangle$$

Hamiltonian

$$\sum_{\nu\beta} (H^{\alpha\beta}_{\mu\nu} - E_i S_{\mu\nu} \delta^{\alpha\beta}) c^\beta_{\nu i} = 0$$

Generalized eigenvalue problem

Output

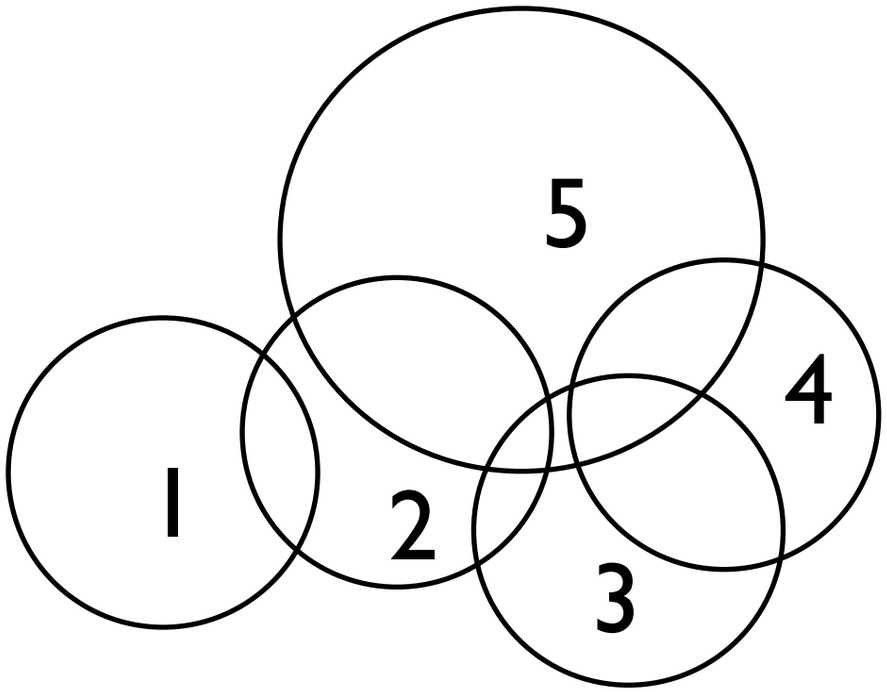
$$\rho(\boldsymbol{r}) = \sum_{\mu\nu} \rho_{\mu\nu} \phi^*_\nu(\boldsymbol{r}) \phi_\mu(\boldsymbol{r})$$

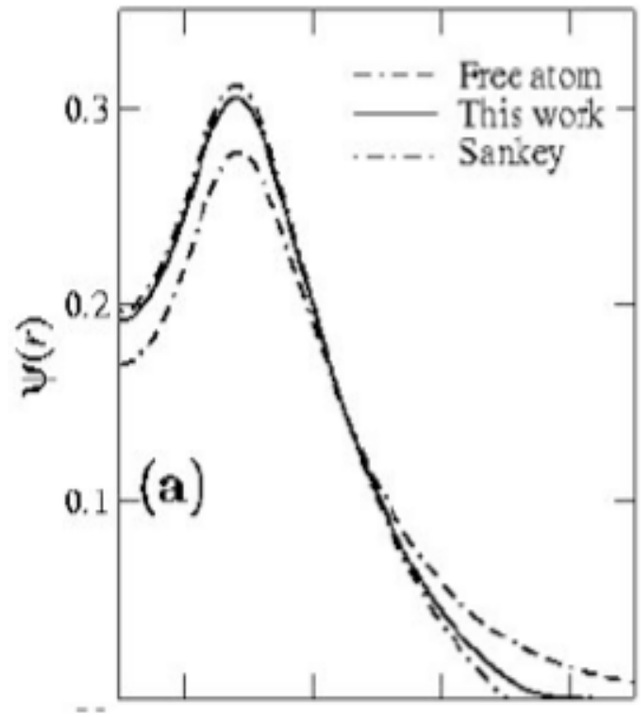
$$E^{BS} = \sum_i n_i \langle \psi_i | \hat{H} | \psi_i \rangle = \sum_{\mu\nu} H_{\mu\nu} \rho_{\nu\mu} = \mathrm{Tr}(H\rho)$$

$$\rho_{\mu\nu} = \sum_i c_{\mu i} n_i c_{i\nu}$$

Density matrix

Sparsity

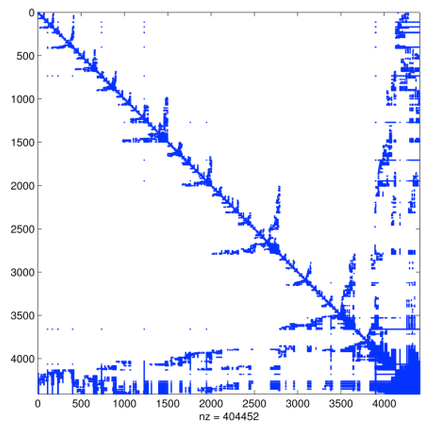




(Finite-range basis orbitals)

$S_{\mu\nu}$ and $H_{\mu\nu}$ are sparse

$\rho_{\mu\nu}$ is not strictly sparse but only a sparse subset is needed

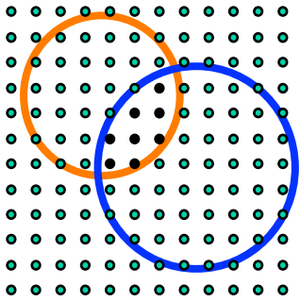


- **TASK 8.1: "Optimization of the SIESTA code"**
  - Massive parallelization for optimal performance in computers with thousands of processors (allowing the treatment of systems with many thousands of atoms)
  - Operation in MPI (present) and OpenMP (future) modes.

$$H_{\mu\nu}^{\alpha\beta} = \langle \phi_\mu | \hat{T} + \hat{V}^{KB} + V^{NA}(\boldsymbol{r}) + \delta V^{H}(\boldsymbol{r}) + V_{XC}^{\alpha\beta}(\boldsymbol{r}) | \phi_\nu \rangle$$

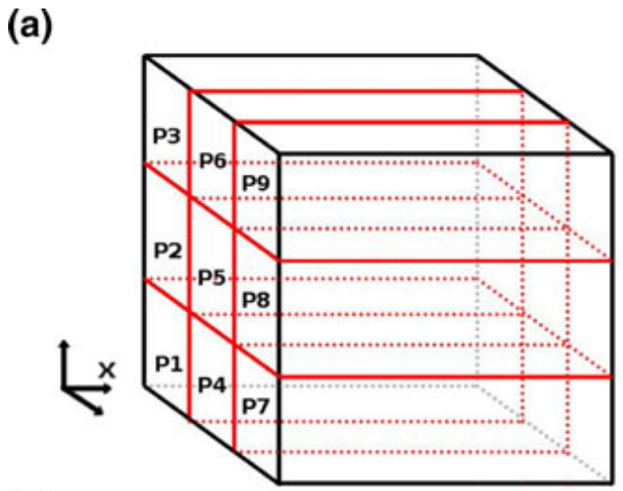
Hamiltonian

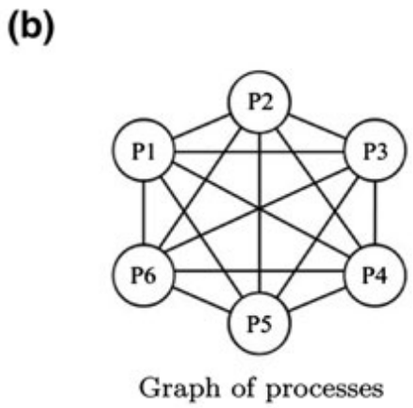
Improvement of load-balancing of real-space mesh operations

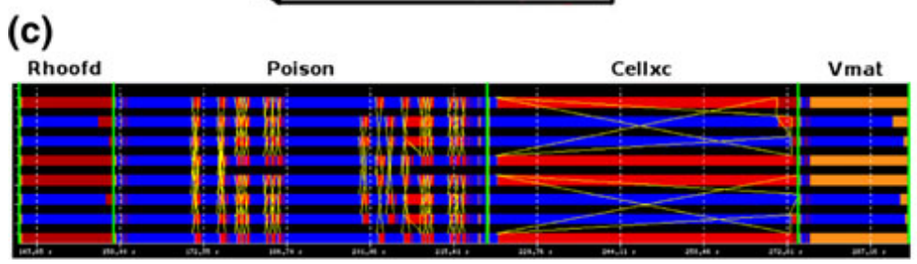


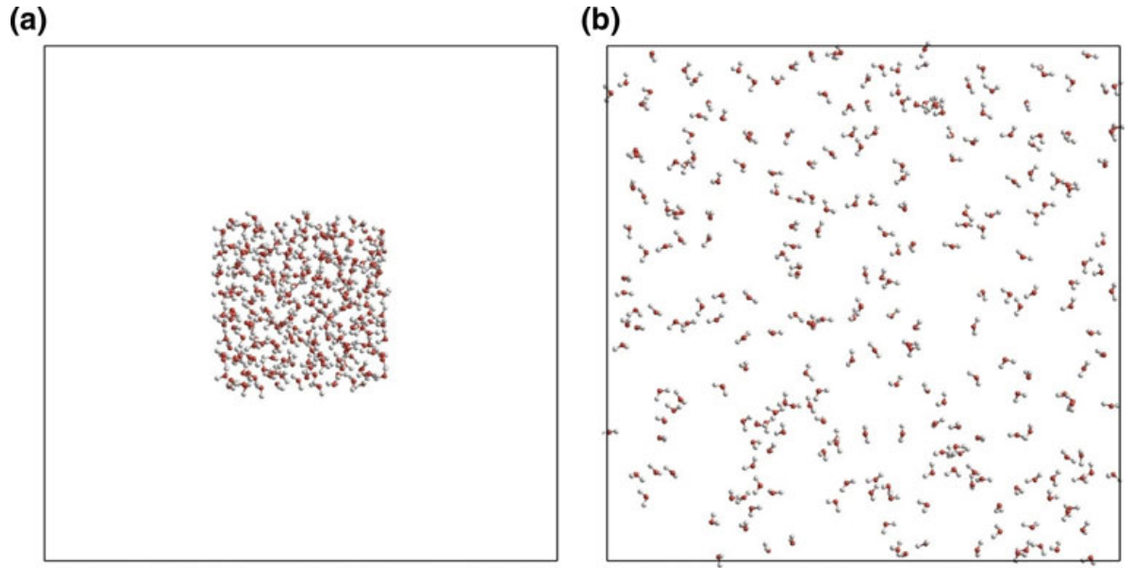
$$\sum_{\nu\beta} (H_{\mu\nu}^{\alpha\beta} - E_i S_{\mu\nu} \delta^{\alpha\beta}) c_{\nu i}^{\beta} = 0$$

Generalized eigenvalue problem

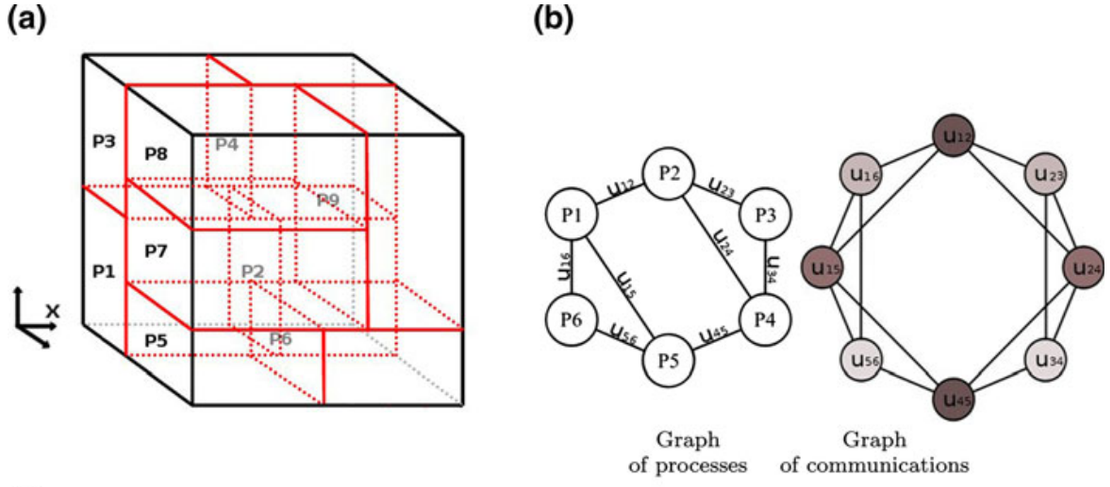


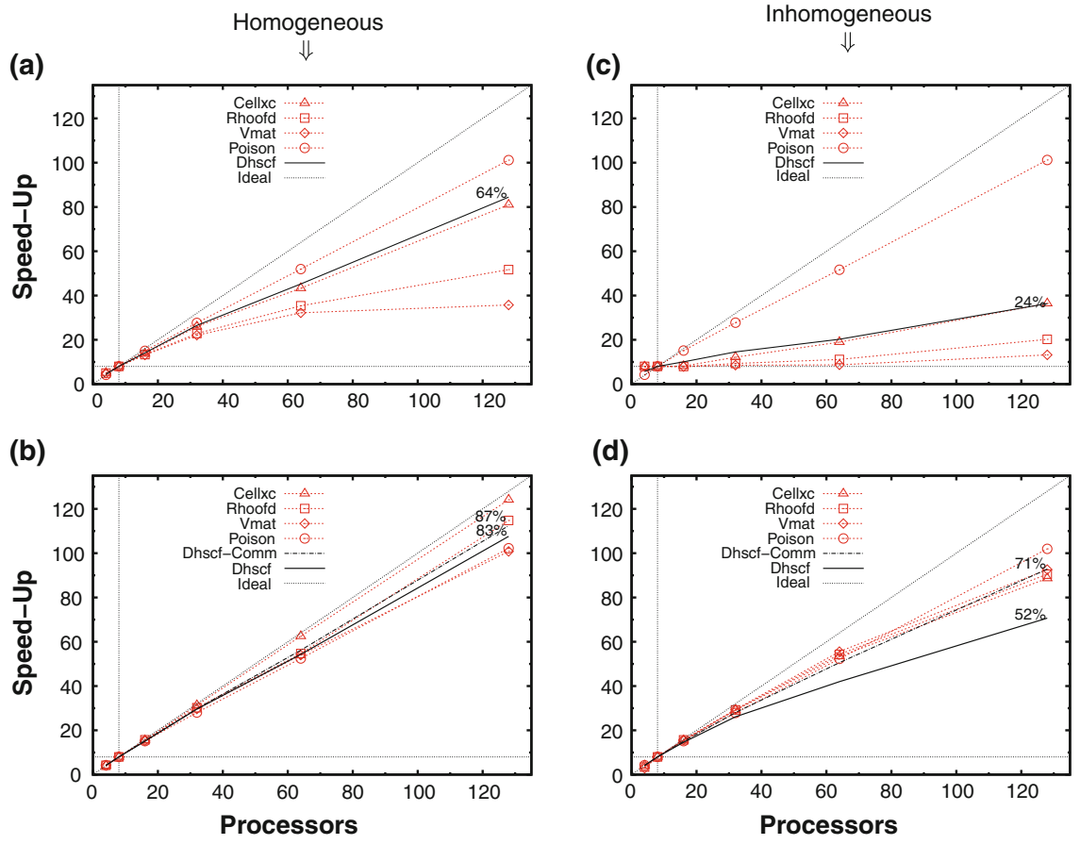






(Rogeli Grima, J.M. Cela, BSC)





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Optimization of the diagonalization: Ideas to beat ScaLapack

ScaLapack: Uses dense matrices; limited scalability

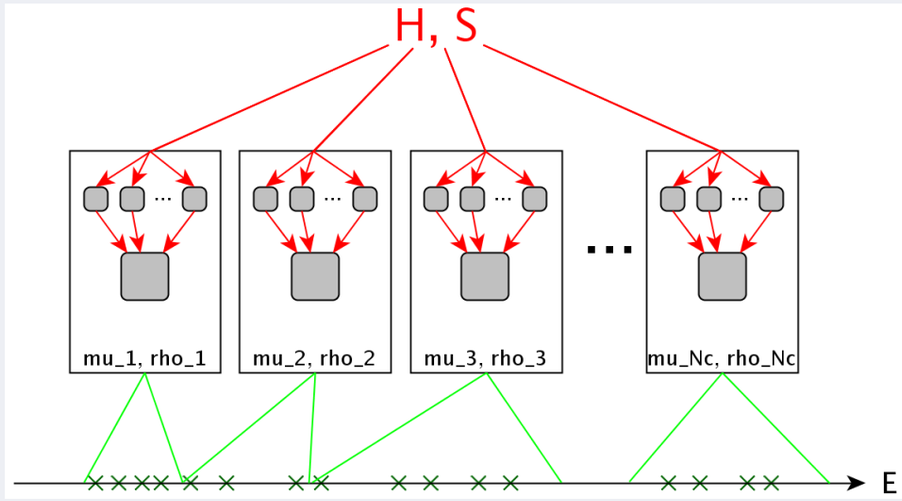
- Use an iterative method, exploiting the sparse character of the H and S matrices.
  - Krylov-type methods: Not appropriate when the number of eigenvectors to compute is relatively large.
- Split-spectrum methods for trivial parallelization at the top level.

Sugiura-Sakurai and FEAST algorithms

(Implementation and benchmarking: Georg Huhs, BSC)

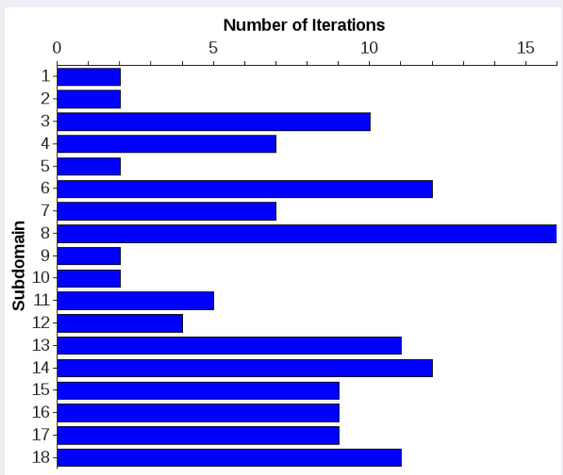
Idea

- Solving many (sparse) linear systems
- Massive parallelization due to three levels
- Reuse of data



Problems

- Less gain of reusing data than expected
- Load balancing problem

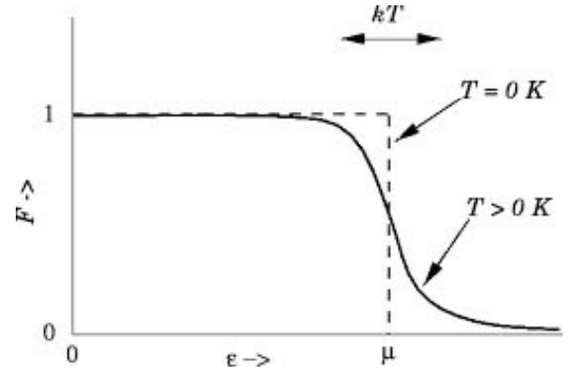


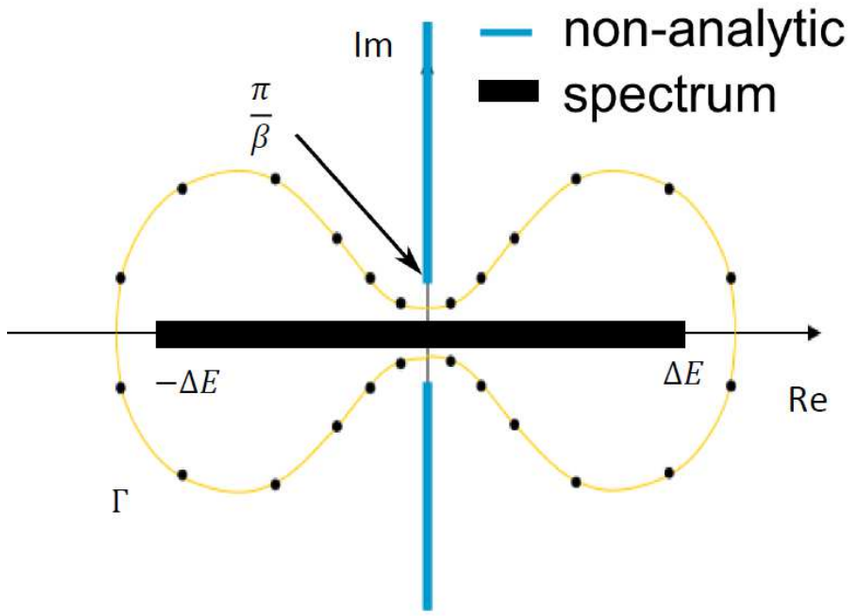
Alternative electronic-structure method
Pole Expansion plus Selected Inversion

$$\hat{\rho} = f_\beta(\hat{H} - \mu)$$

$$f_\beta(\epsilon_i - \mu) = \frac{2}{1 + \mathrm{e}^{\beta(\epsilon_i - \mu)}}$$

Fermi-Dirac function





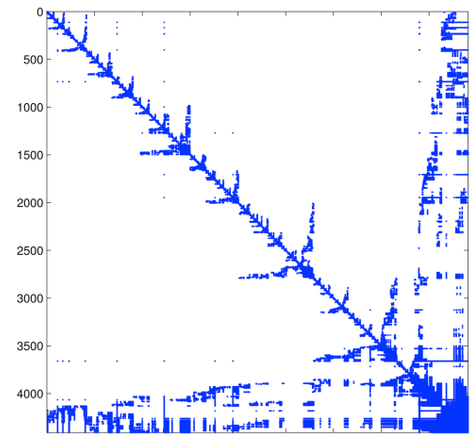
$$f_\beta(\epsilon_i - \mu) \approx Im \sum_{l=1}^{P} \frac{\omega_l}{\epsilon_i - (z_l + \mu)}$$

$$\hat{\rho} = \quad Im \left(\sum_{l=1}^{P} \frac{\omega_l}{H - (z_l + \mu)S} \right)$$

$$\hat{\rho} = \quad Im\left(\sum_{l=1}^{P}\frac{\omega_l}{H-(z_l+\mu)S}\right)$$

One inversion per pole

Only a limited number of elements needed in density matrix



Selected Inversion

$$A = \begin{pmatrix} a & b^T \\ b & \hat{A} \end{pmatrix}$$

$$(A =)LDL^T = \begin{pmatrix} 1 & \\ l & \hat{L} \end{pmatrix}\begin{pmatrix} \alpha & \\ & \hat{A}-bb^T/\alpha \end{pmatrix}\begin{pmatrix} 1 & l^T \\ & \hat{L} \end{pmatrix}$$

$$A^{-1} = \begin{pmatrix} \alpha^{-1}+l^TS^{-1}l & -l^TS^{-1} \\ -S^{-1}l & S^{-1} \end{pmatrix}$$

Pole Expansion plus Selected Inversion

(Lin Lin, Chao Yang, LBNL)

Trivially parallel over poles, with perfect load balancing

For sufficiently big problems

$$\begin{aligned} \text{(quasi-)1D:} \quad & \mathcal{O}(N) \\ \text{(quasi-)2D:} \quad & \mathcal{O}(N^{3/2}) \\ \text{3D:} \quad & \mathcal{O}(N^2) \end{aligned}$$

(Due to sparsity of the target density matrix)

Will beat Scalapack for large systems: It can use thousands of cores efficiently, with small memory footprint.

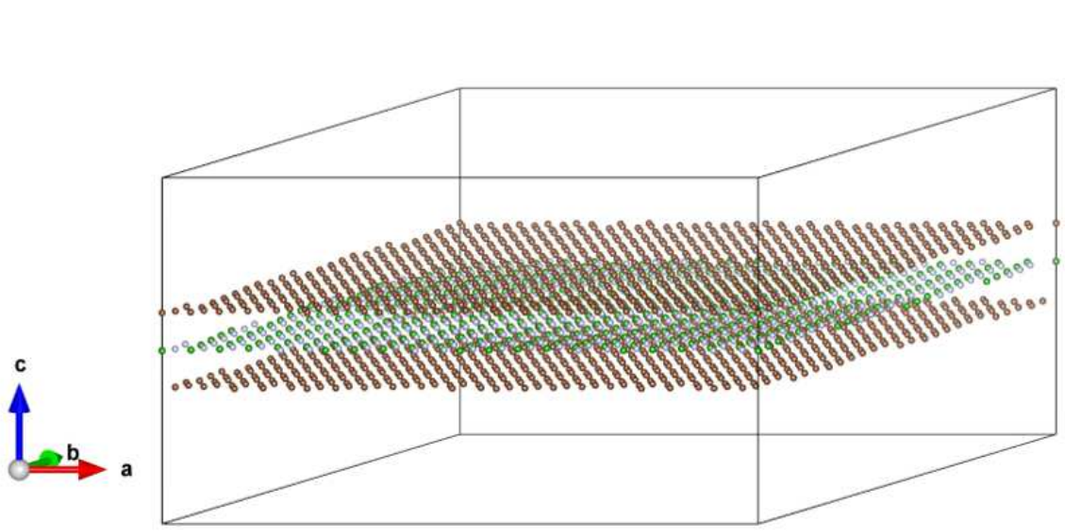
There is still scope for further optimization of the factorization and inversion operations

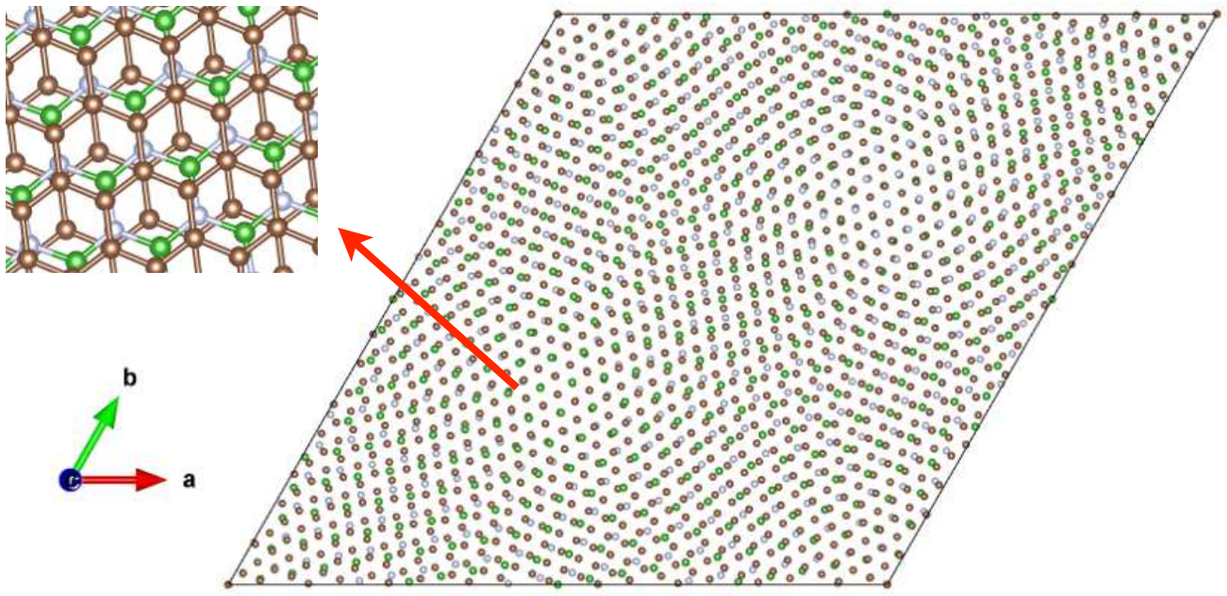
Pole Expansion plus Selected Inversion (PEXSI)

Implementation of interface to SIESTA and benchmarking:
Alberto García (ICMAB) and Georg Huhs (BSC)

Development of heuristics to handle the variation of
the chemical potential during the self-consistent-field updates

Main test systems: quasi-2D C-BN structures





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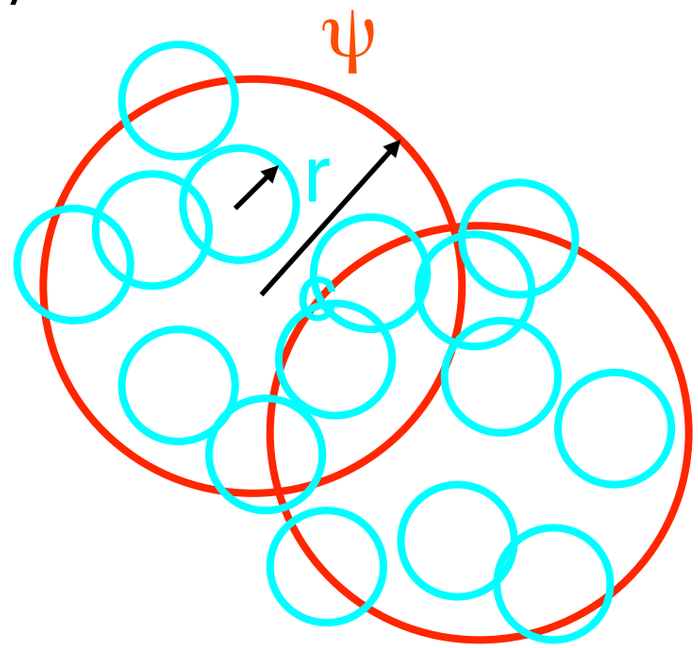
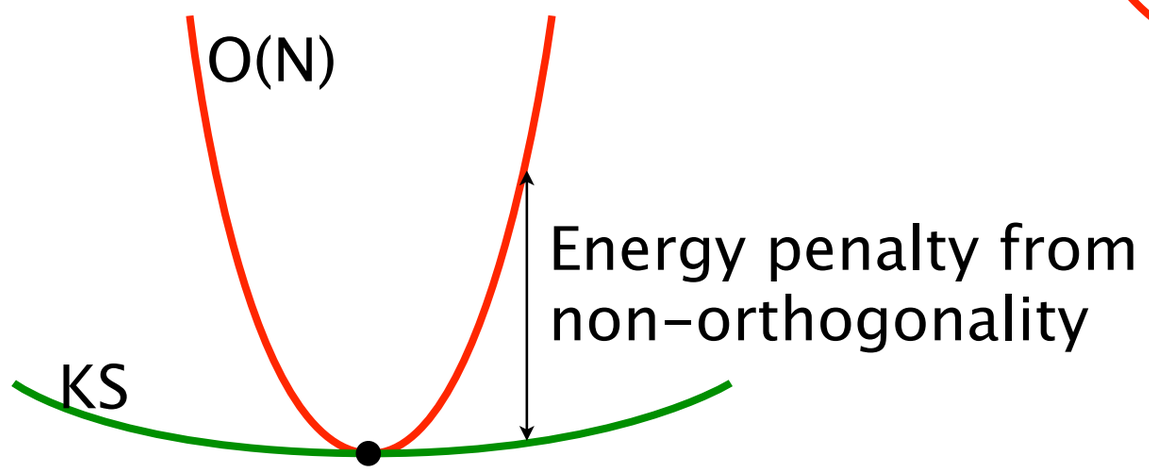
O(N) functional

- $S_{ij} = \langle \psi_i | \psi_j \rangle$, $|\psi'_k\rangle = \sum_j |\psi_j\rangle S_{jk}^{-1/2}$
- $E_{KS} = \mathrm{Tr}[S^{-1} H]$
- $E_{O(N)} = \mathrm{Tr}[(2I - S) H]$
- $E_{KS} = E_{O(N)}$ at the minimum



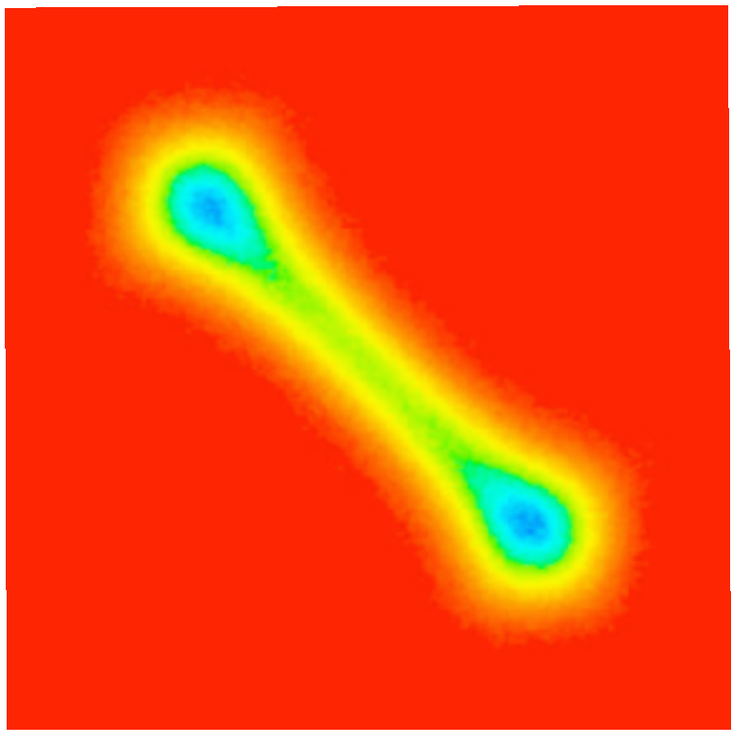


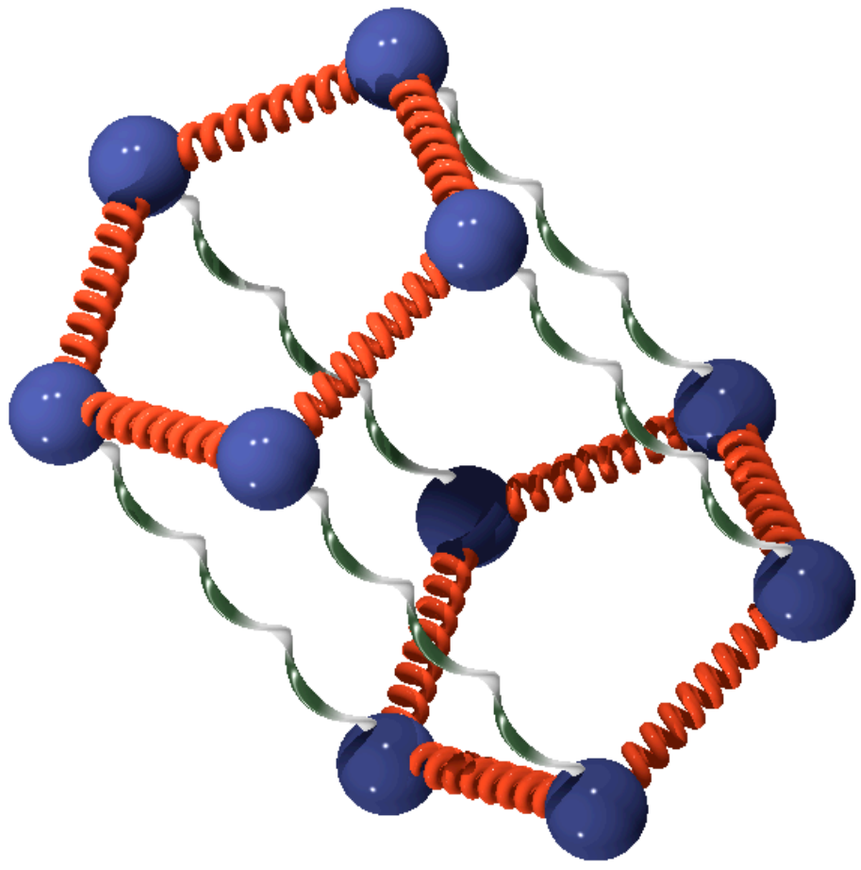
Optimization of sparse-matrix operation (CASE)
Easier case: Minimization without localization (CASE, F. Corsetti)

• TASK 8.2: "Tools for data handling and implementation of new simulation techniques"

- New library to process operational parameters in parallel (Raúl de la Cruz, BSC)
- Use of more efficient formats for data storage (HDF5, ongoing)
- Implementation of a mechanism to dispatch tasks in parallel, used to drive SIESTA from a Path-Integral program.

Quantum effects described by Path-Integral method
Isomorphism to a classical system
with replicated particles

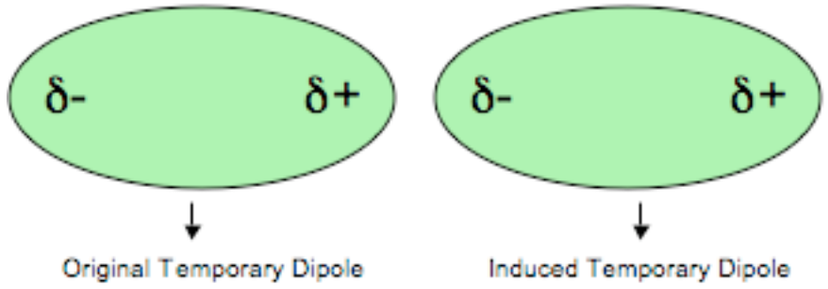




Each calculation is independent
The method is thus trivially parallelizable

JM. Soler (UAM), Rafael Ramírez (ICMM)

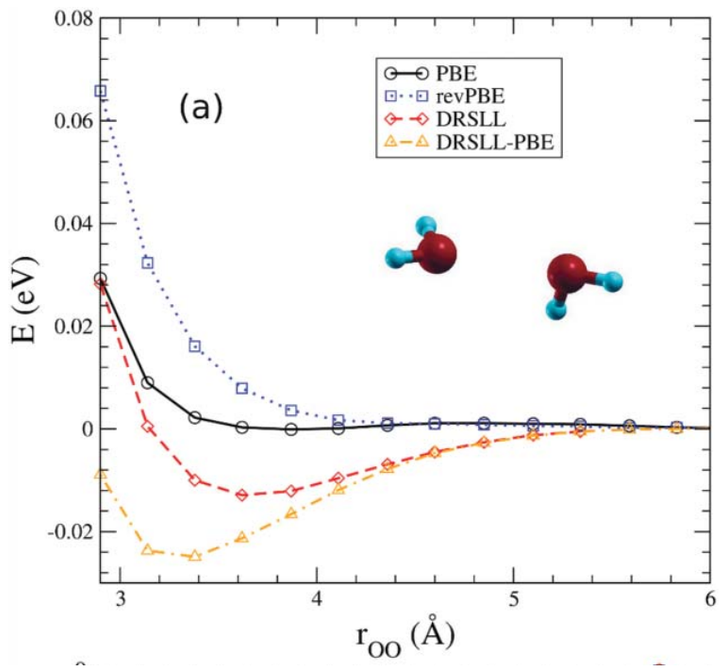
van der Waals interactions



Non-local functional, implemented efficiently in SIESTA
(JM Soler, UAM)

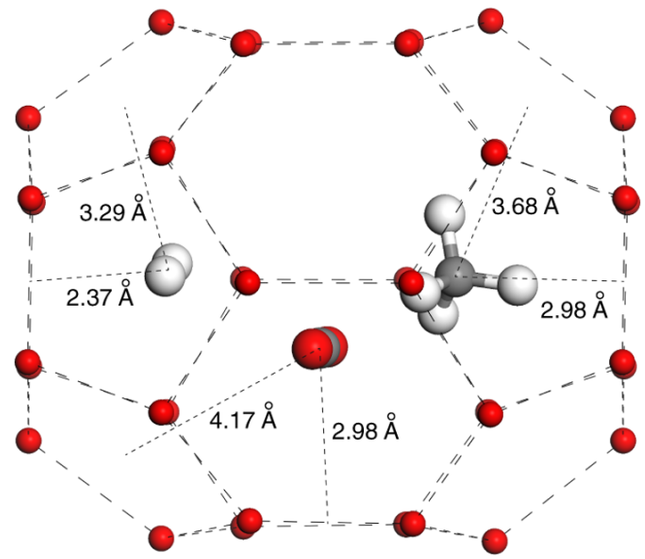
Residual interactions in water

Roman-Perez, Moaied, Soler, Yndurain. PRL (2010)

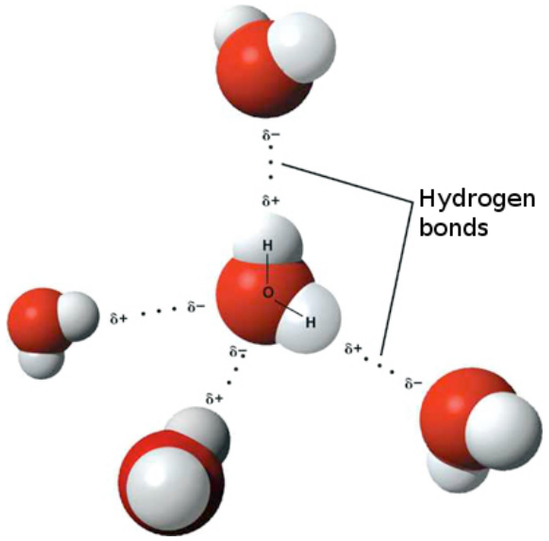


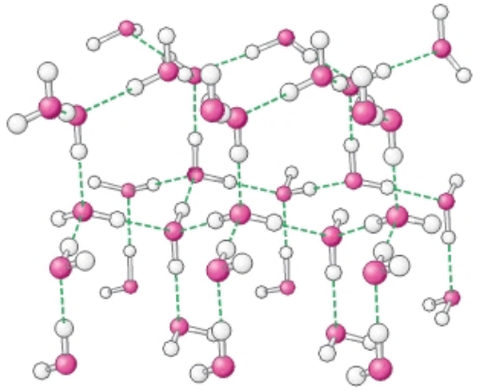
Stability of clathrate hydrates

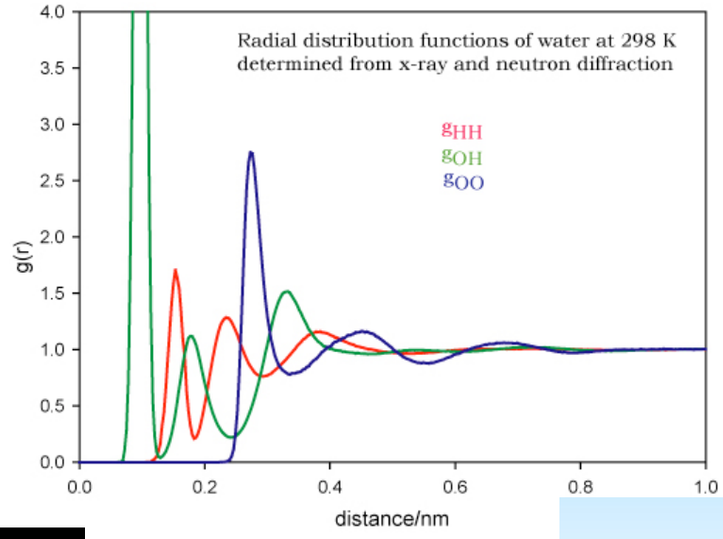
Wang, Roman-Perez, Soler, Artacho, Fernandez-Serra, JCP (2011)

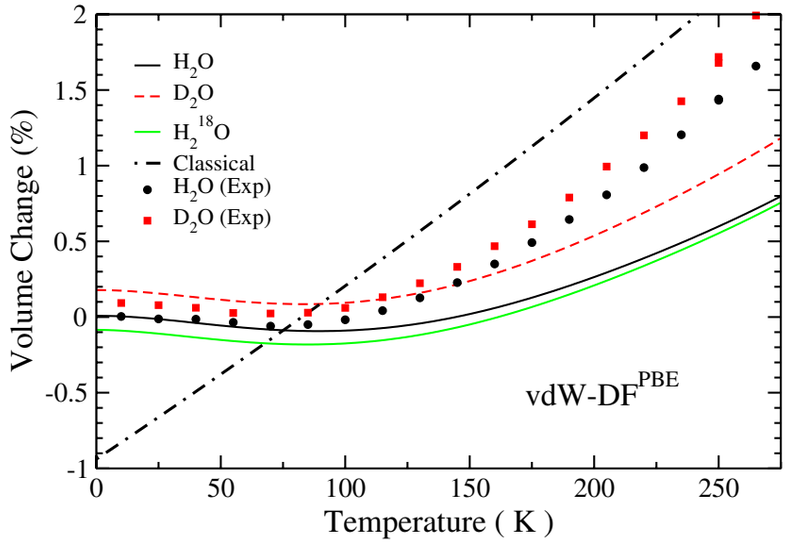


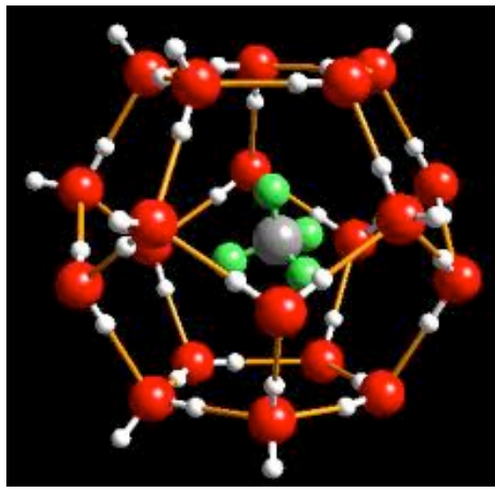
Quantum and van der Waals effects in hydrogen-rich systems: water and clathrate hydrates

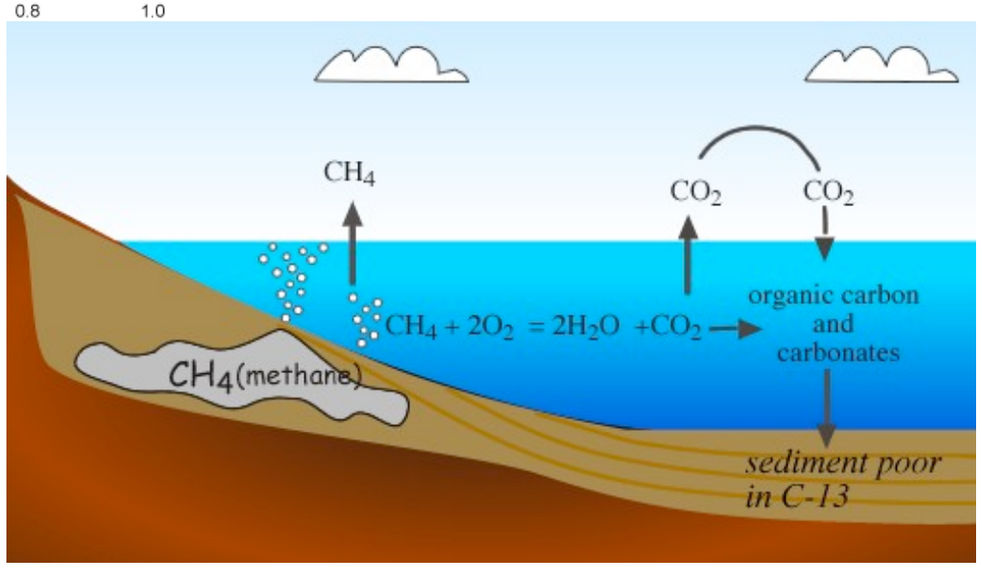






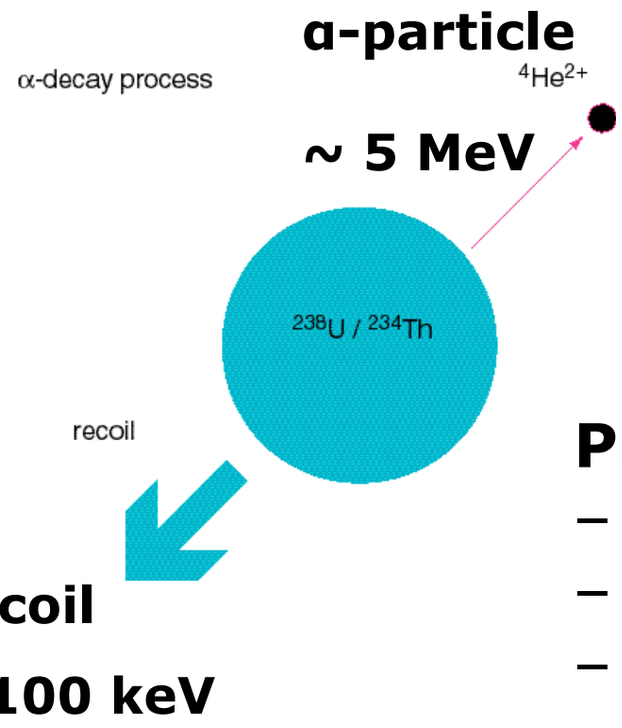


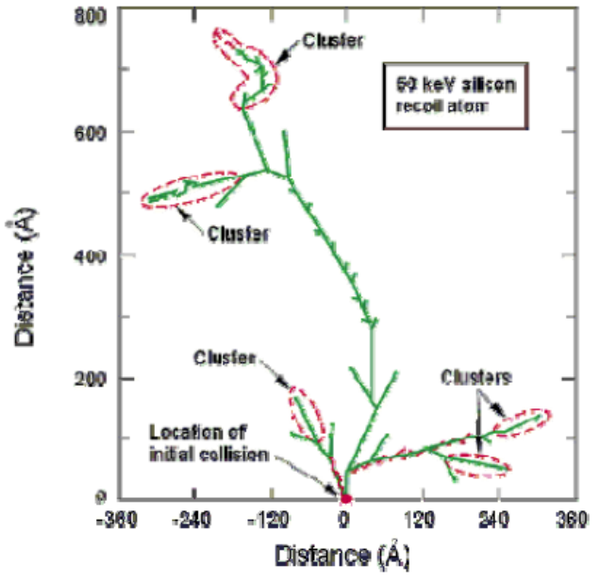




- **TASK 8.3: "Preparation and Execution of Radiation Damage simulations"**

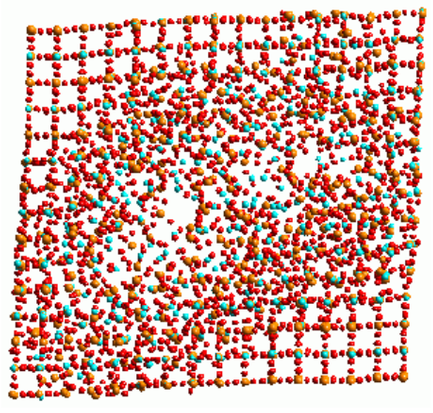
α-decay process





Produces:

- Amorphization
- Swelling
- Fragilization
- Fractures
- …

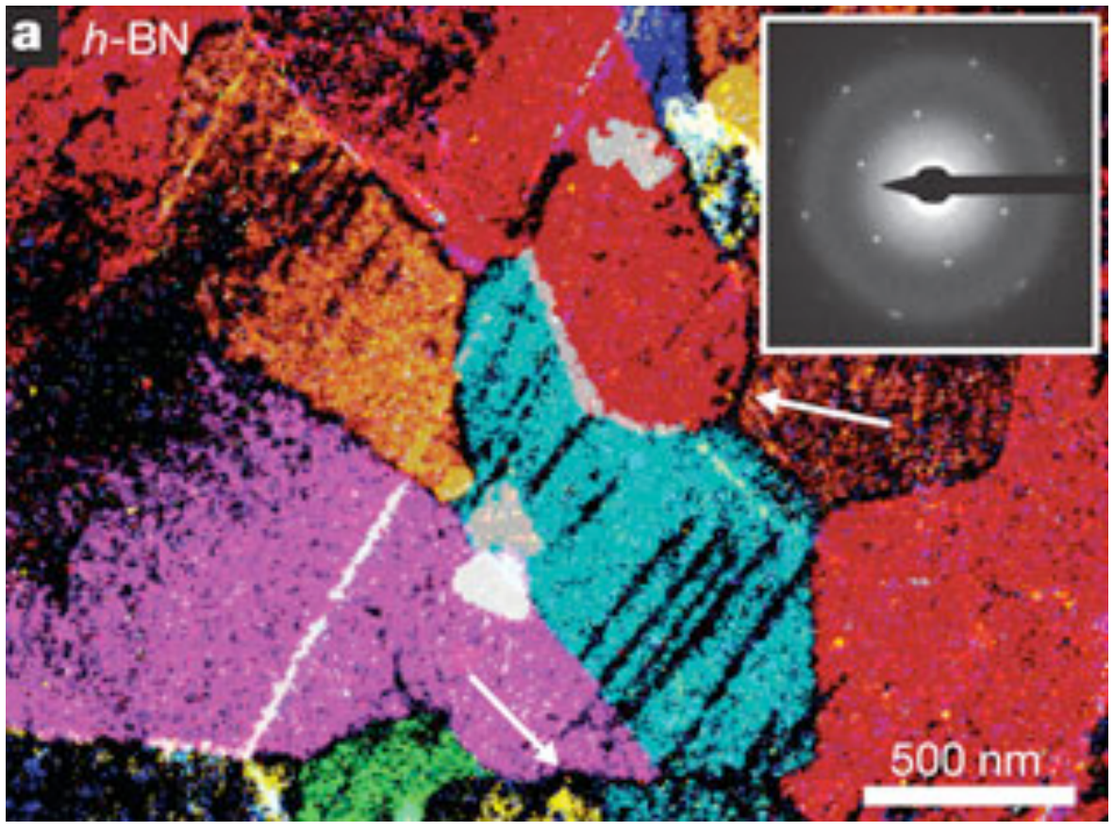


- Simulation of "atomic" collisions.
- Energy loss due to electron heating effects (coupled electron-ion dynamics)

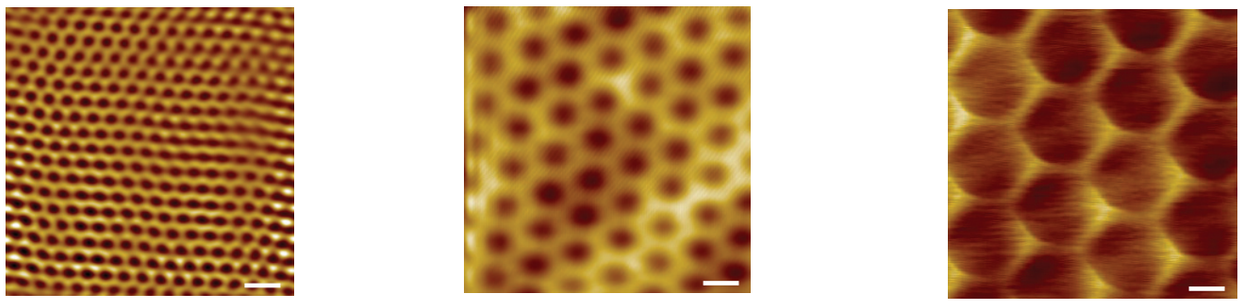
Thousands of atoms, and long times, needed in the simulation

Quasi two-dimensional systems

Lateral graphene-BN heterostructures



Moiré patterns of graphene over hex-BN



Ongoing work

- Final refinements to PEXSI method to allow the performance of converged large scale calculations, including molecular dynamics simulations.
- Completion of the upgrade of the I/O subsystem in SIESTA.
- Further work on sparse matrix library to streamline the coding of the O(N) method.

Collaborations

- Optimization of SIESTA: BSC-CASE.
- Pole-Expansion plus Selected-Inversion method: Lin Lin, Chao Yang (L. Berkeley Lab)
- Quasi-2D graphene-based systems: Miguel Pruneda, Rafael Martínez (CIN2, Barcelona)
- Radiation damage (D. Sánchez-Portal, centro mixto CSIC-UPV (San Sebastián) and M. Pruneda, CIN2)
- Structure of water and quantum effects: M.V. Fernández-Serra, Stony Brook U, R. Ramírez, ICMM.
- Work towards GPU implementation: NVIDIA