Growth and self-organization of nanostructures using BART (BigDFT+ART Nouveau)

Eduardo Machado-Charry, Luigi Genovese, Damien Caliste,
Pascal Pochet and Normand Mousseau

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Multi Scale Design of nanomaterials with simulations on hybrid architectures: (the muscade project)

MUSCADE SYNOPSIS:

Software engineering

Boost *ab initio* package

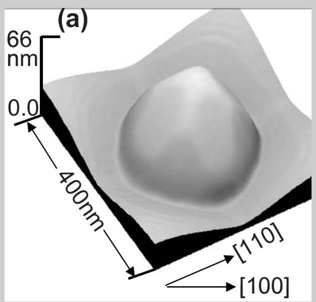
Algorithms for rare event

ab initio based exploration

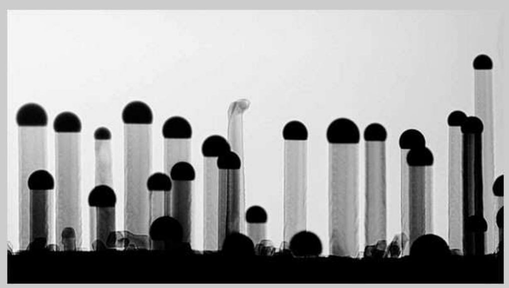
Simulation tools for growth study in nanosciences

Application targets

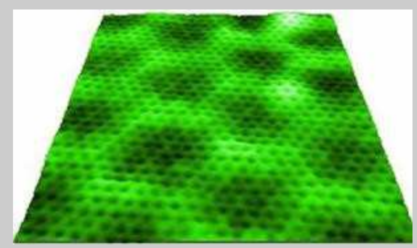
Ge QD on Si



Si Nanowire



Graphene on SiC



Experimental validation

Outline

OUTLINE

① BigDFT

OUTLINE

① BigDFT
② **ART**

OUTLINE

① BigDFT
② **ART**
③ Vacancy on silicon: an example

OUTLINE

① BigDFT
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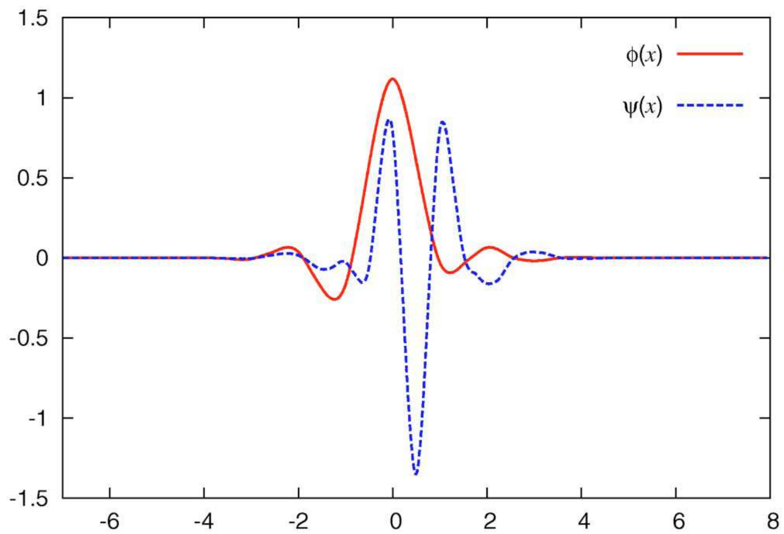
③ Vacancy on silicon: an example
④ Ongoing applications

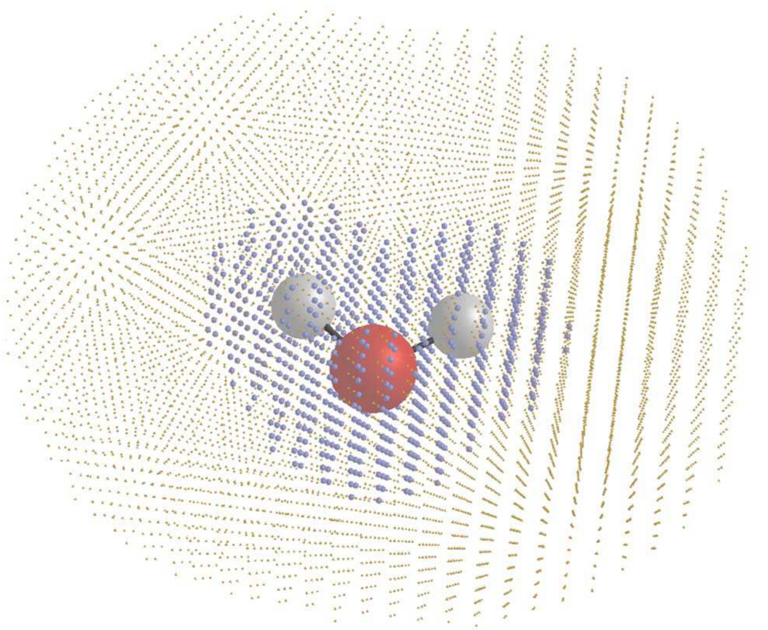
Outline

① BigDFT
② **ART**
③ Vacancy on silicon: an example
④ Ongoing applications
⑤ Final remarks

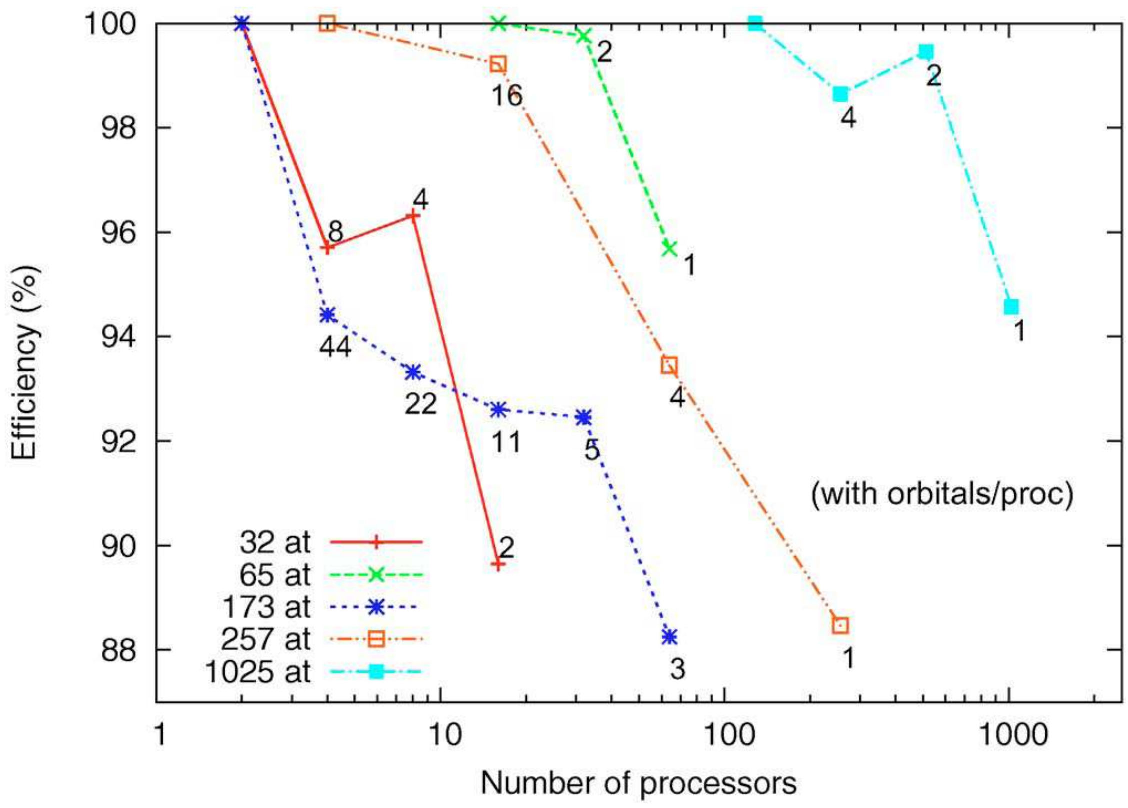
BigDFT

A DFT massively parallel electronic structure code





- Localised in real and fourier space
- (Bi-)orthogonal basis
- systematic
- Basic function can be placed only where are needed
- Adaptive:
  low and high resolution
- no numerical approximation to **H** related quantities
- pseudopotentials



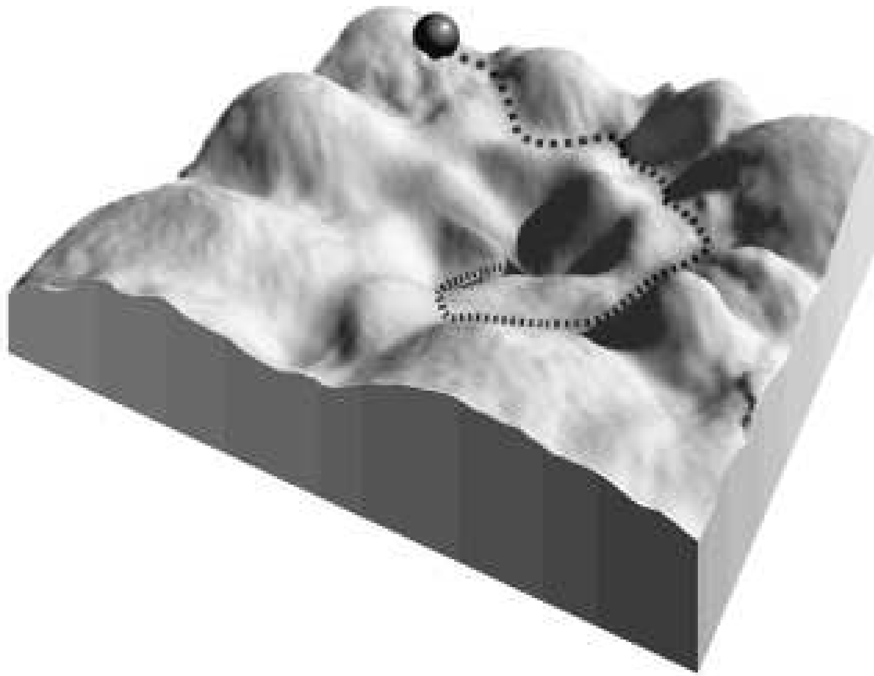
PERFORMANCE:

- localisation and orthogonality → data locality and high efficiency.
- Efficiency of the order of 90%, up to thousands of processors.
- Hybrid architectures (GPU)

FURTHER INFORMATION:

- Interfaced with ABINIT
- HOMEPAGE: http://inac.cea.fr/L_Sim/BigDFT/index.html
- Genovese *et al.*, J. Chem. Phys. **129**, 014109 (2008)

Activation Relaxation Technique



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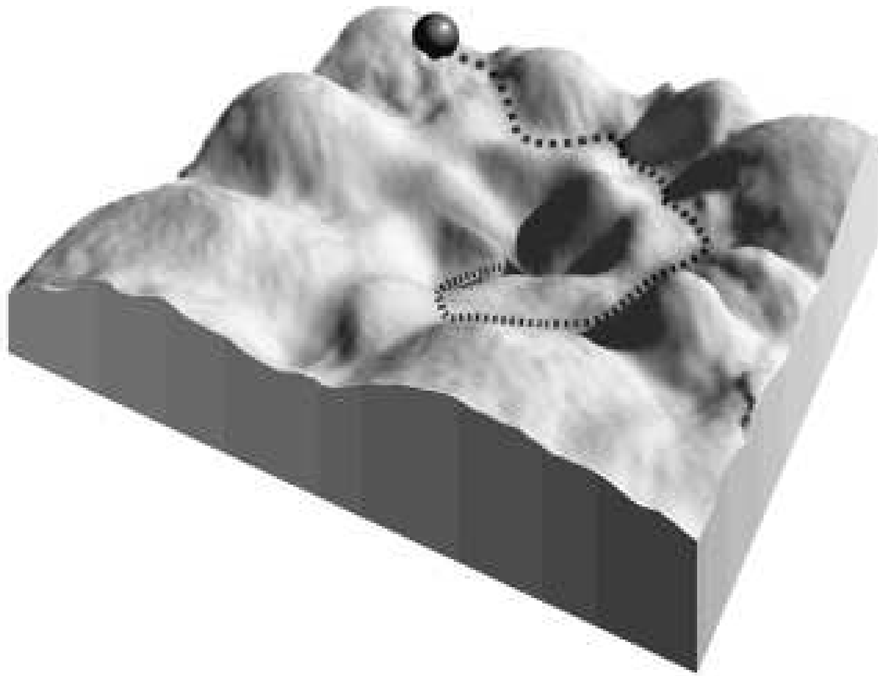
Generic problem

How to explore the space of variables of a high dimensional cost function?



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ART nouveau

Zero temperature :

Samples the local minima only, going through common saddle point, generating a continuous trajectory.

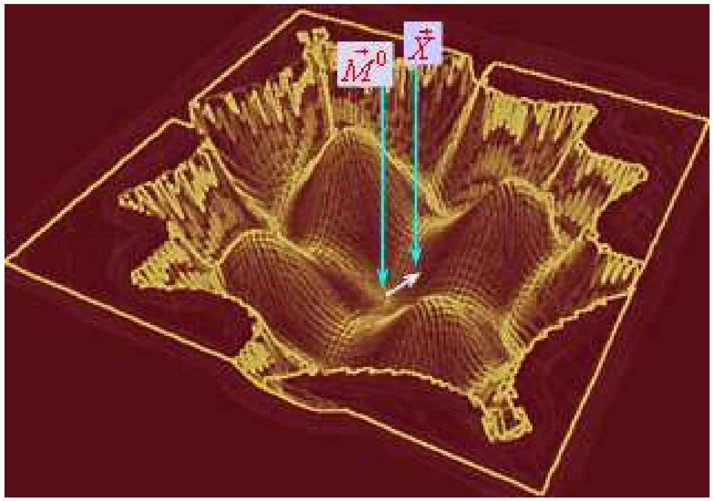
Barkema *et al*, PRL 1996; Malek *et al*, PRB 2000,

Doye *et al*, Z Phys B 1997; Henkelman and Jonson, 1999

Some applications of ART nouveau:

- **Ab initio calculation of defects diffusion mechanisms in Silicon, GaAs**
  El-Mellouhi *et al* - PRB (2004, 2005), J. Appl. Phys.(2006); Malouin, PRB (2007)
- **Amorphous silicon - structure, relaxation and activated mechanisms**
  Barkema, *et al* PRL (1996,1998), PRE (1998), PRB (2000, 2001,2003)
- **Amorphous gallium arsenide - structural properties**
  Lewis *et al* PRL (1997), PRB (1997), *et al* JPhys:CondMatt (2004)
- **Silica glass - structural properties, activated mechanisms**
  Barkema *et al* JCP (2000)
- **Lennard-Jones clusters and glasses**
  Brébec *et al* PRB (2000), Def. Diff. Forum (2001)
- **Protein folding**
  Derreumaux *et al* J. Mol. Graph. (2001), JCP (2003), Proteins (2004); St-Pierre *et al* (2008)
- **Protein aggregation**
  Boucher *et al* - JACS (2004), Biophys. J., Structure (2004), JCP (2005), Accounts Chem. Res.(2005), Proteins(2006), JCP (2006,2007)

1. Leave the harmonic basin



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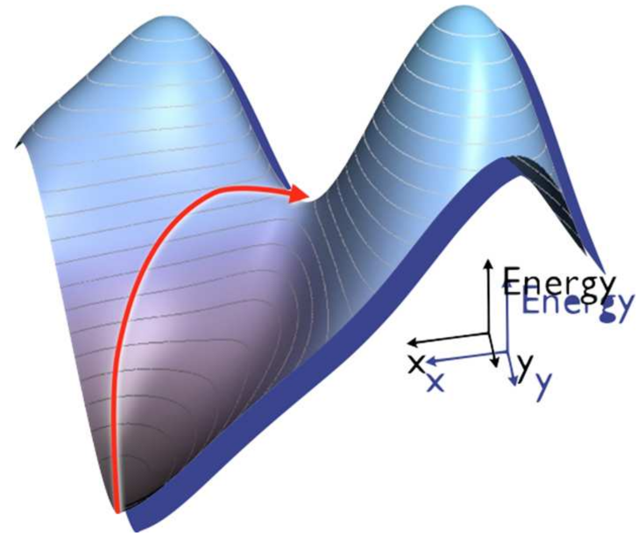
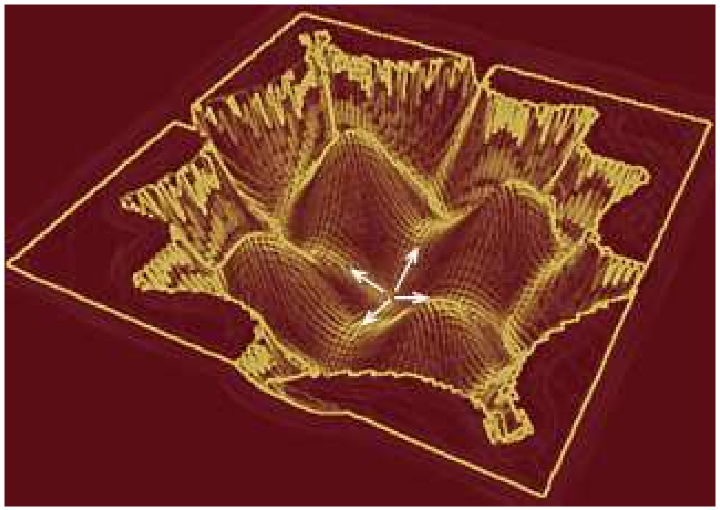


- Random deformation (local or global)
- Follow this direction until an eigenvalue become negative

2. Converging to a saddle point



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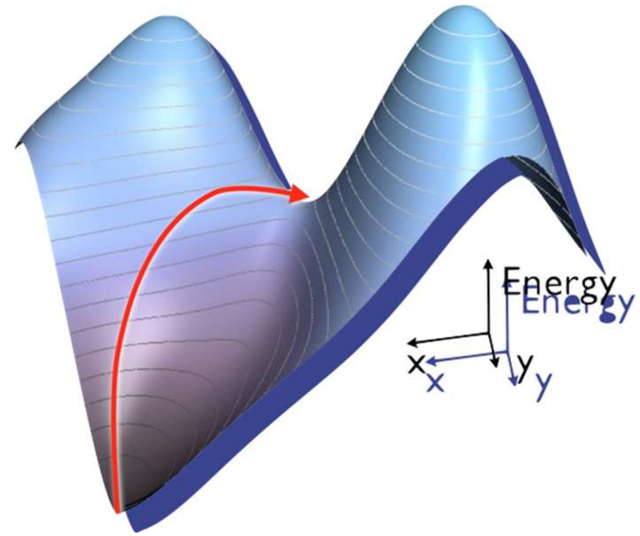
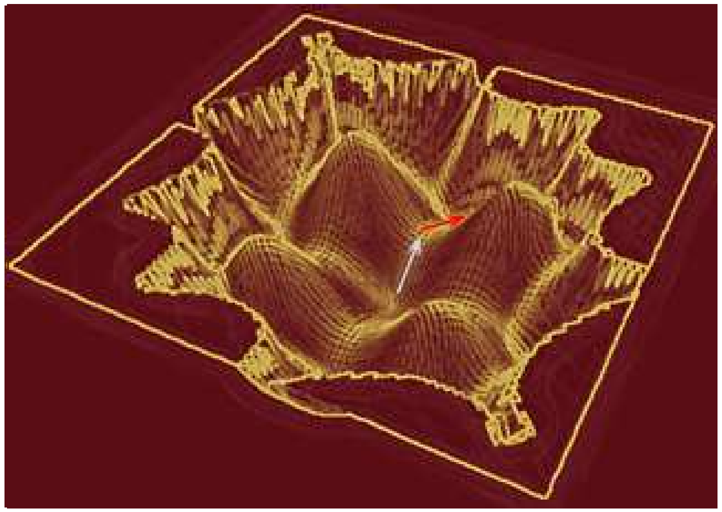


- Push the configuration up along the corresponding direction
- Convergence: Zero total force & negative eigenvalue
- Warning: The path is not the MEP

3. Relax to a new local minimum



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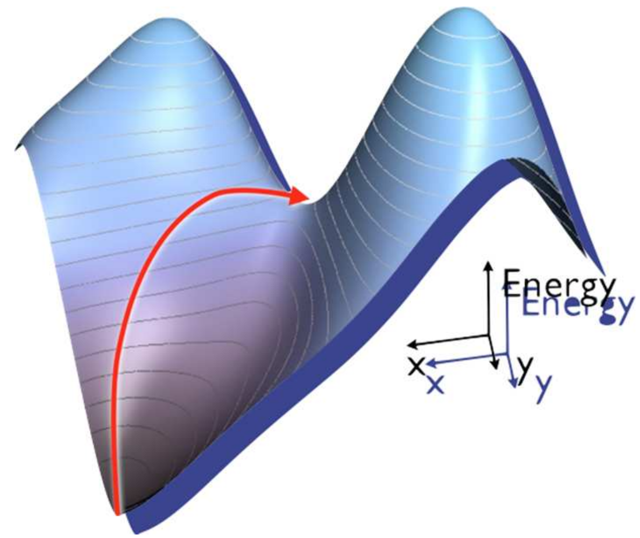
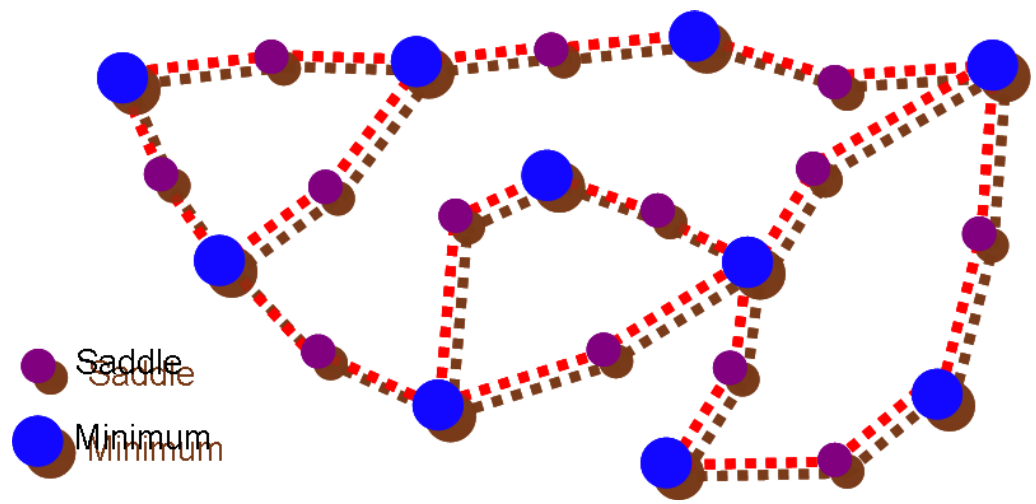


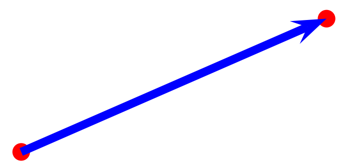
- Push the configuration slightly over the saddle point
- Relaxation

- Not sensitive to the real space complexity of activated jump nor the height of the activation energy barrier, which can be very high.
- Not biased toward pre-determined mechanisms.
- Seems to sample all classes of events (ergodic).
- Events are reversible.
- Events are accepted or rejected based on a standard Metropolis test.



Some details of the step:

$$q_{i+1} = q_i + \Delta\widehat{v}$$



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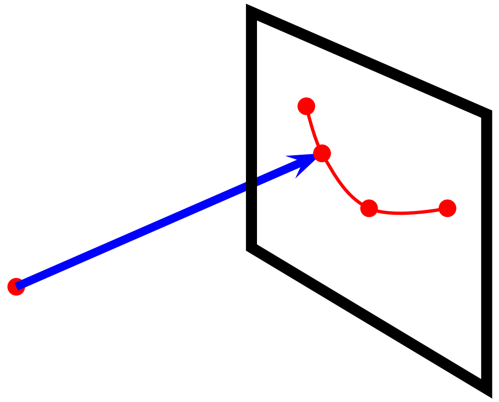

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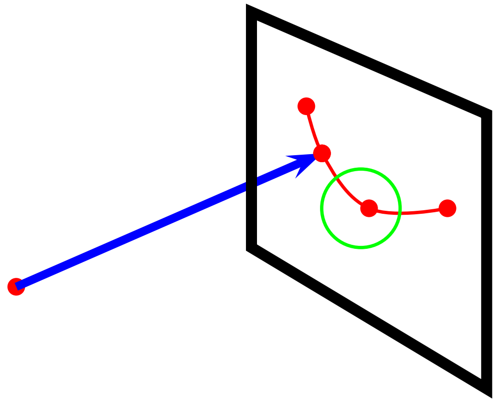
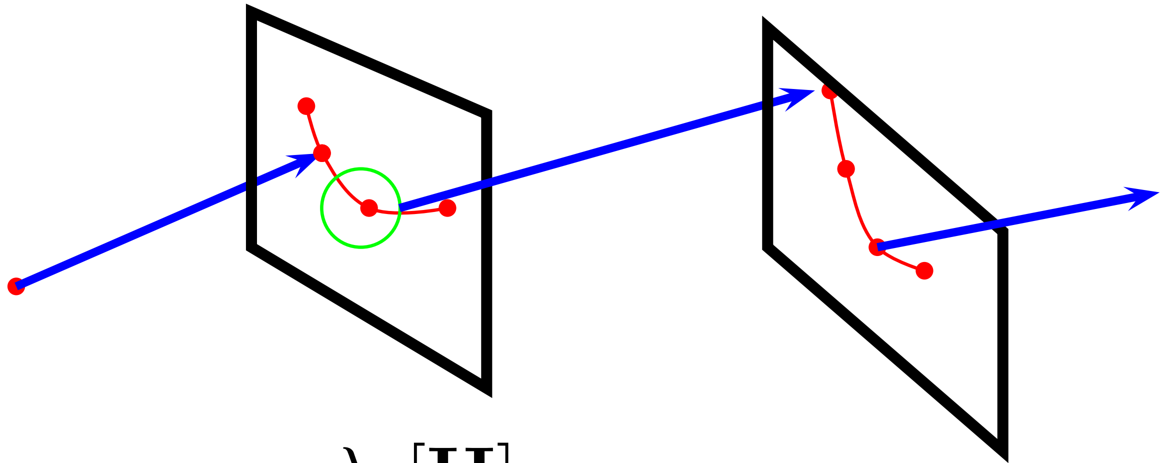


$$\lambda_1[\mathbf{H}]$$

$$\widehat{v}_1[\mathbf{H}]$$

$$q_{i+1} = q_i + \Delta\widehat{v}$$

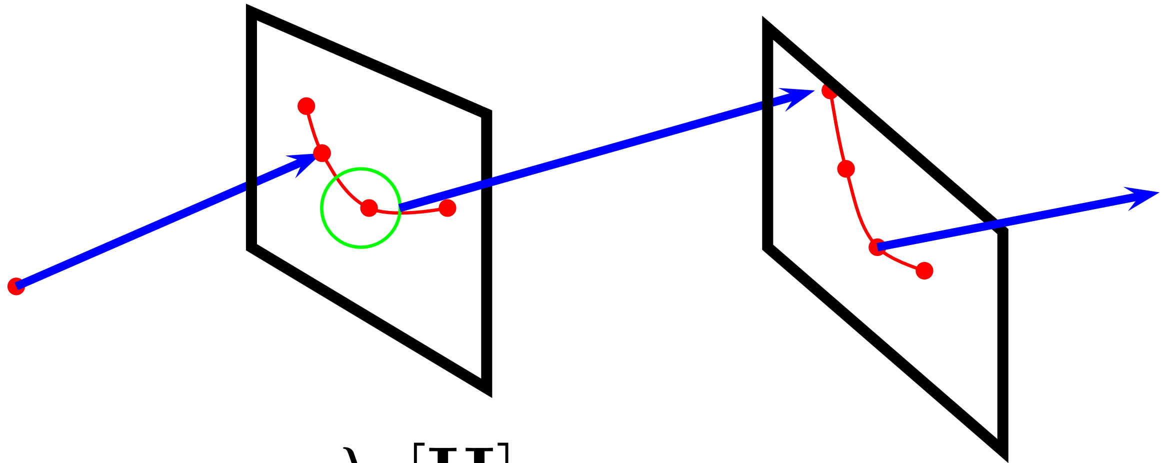


$\lambda_1[\mathbf{H}]$

$\widehat{v}_1[\mathbf{H}]$

How to get λ_1 ?

$$q_{i+1} = q_i + \Delta\widehat{v}$$



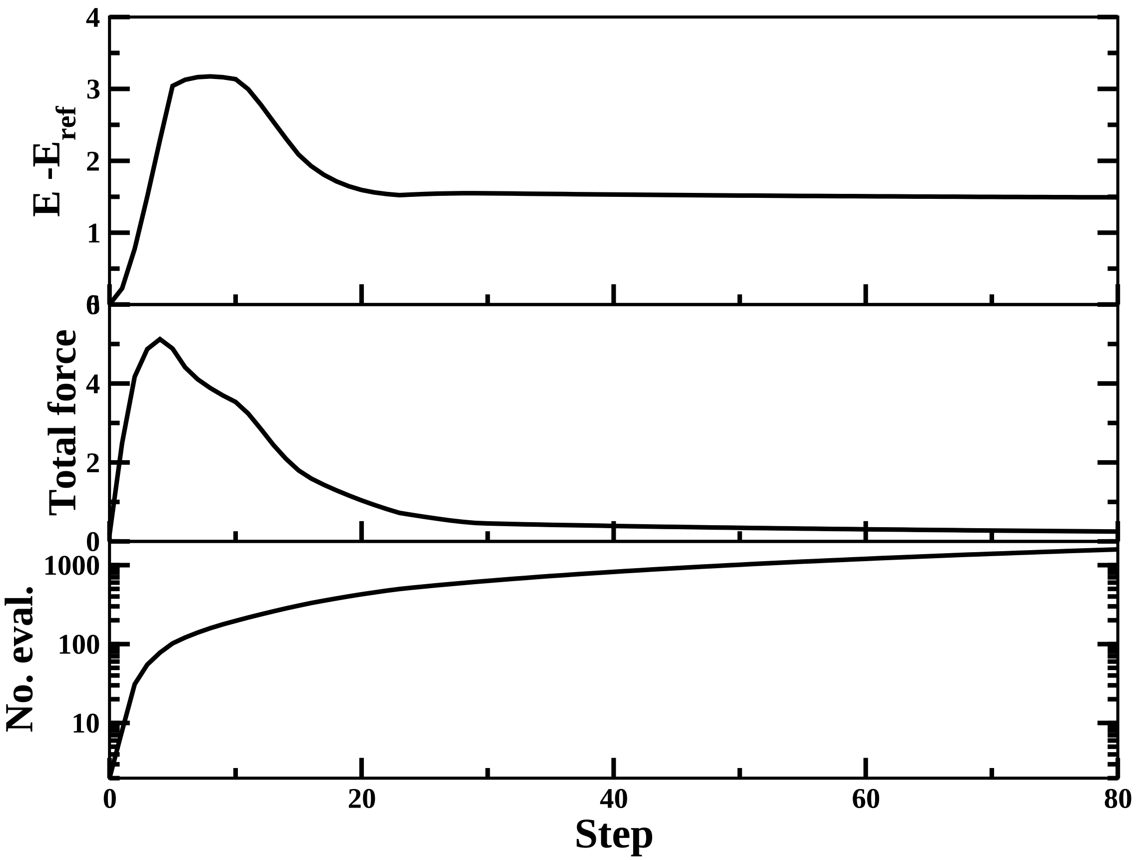
$$\lambda_1[\mathbf{H}]$$

$$\widehat{v}_1[\mathbf{H}]$$

We use the lanczos method

$\mathbf{H}_{3N\times 3N} \longrightarrow \mathbf{L}_{l\times l}$ Typically, $15 < l < 20$

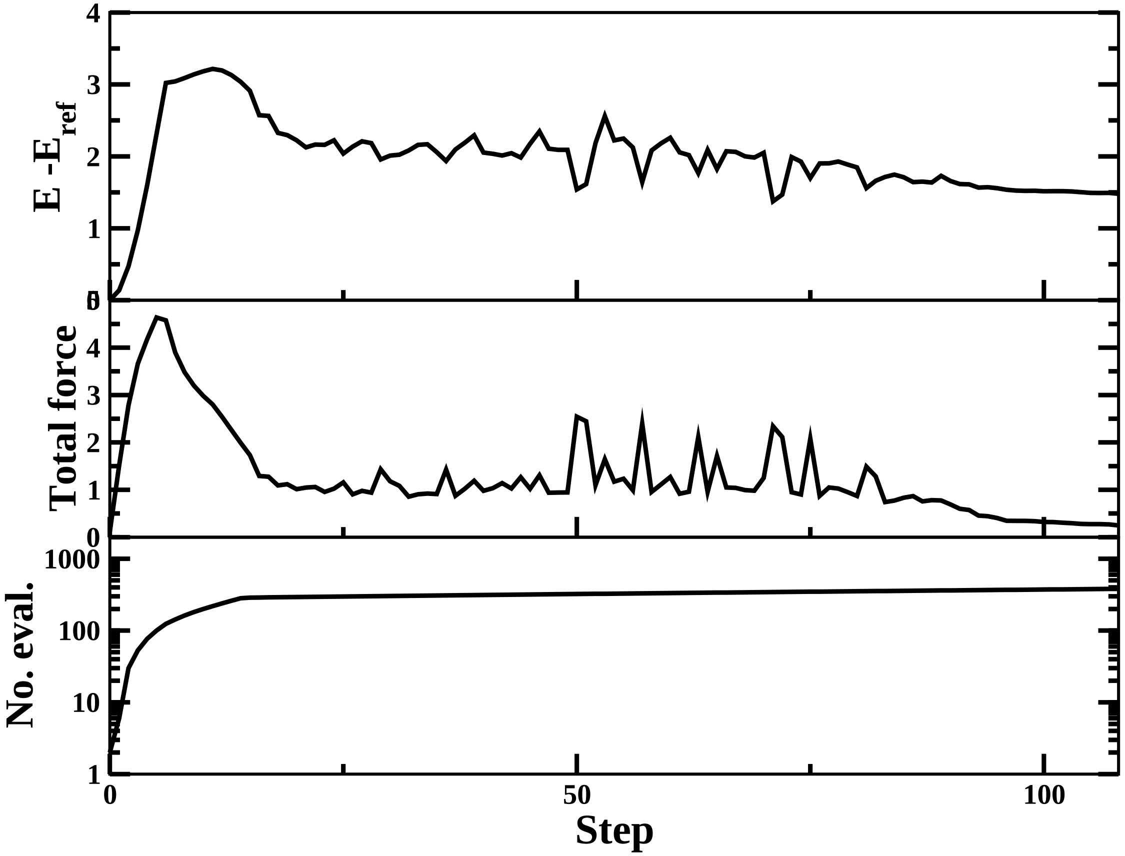
Amorphous silicon (SW semiempirical potential)



Statistics

• attempted event	2889
• eval/attempted event	1789
• successful events	**1000**
• eval/per successful event	5168
• % of events which lead **back to the minimum**	**5.15**
• **eval/per new event**	**5450**

Amorphous silicon (SW semiempirical potential)



Statistics

- attempted event 2709
- eval/attempted event 448
- successful events **1000**
- eval/per successful event 1214
- % of events which lead **back to the minimum** **35.1**
- **eval/per new event** **1872**

Amorphous silicon (SW semiempirical potential)

pros:

A remarkable improvement in the efficiency

cons:

DIIS detects all critical points including **shoulders**

Statistics

• attempted event	2709
• eval/attempted event	448
• successful events	**1000**
• eval/per successful event	1214
• % of events which lead **back to the minimum**	**35.1**
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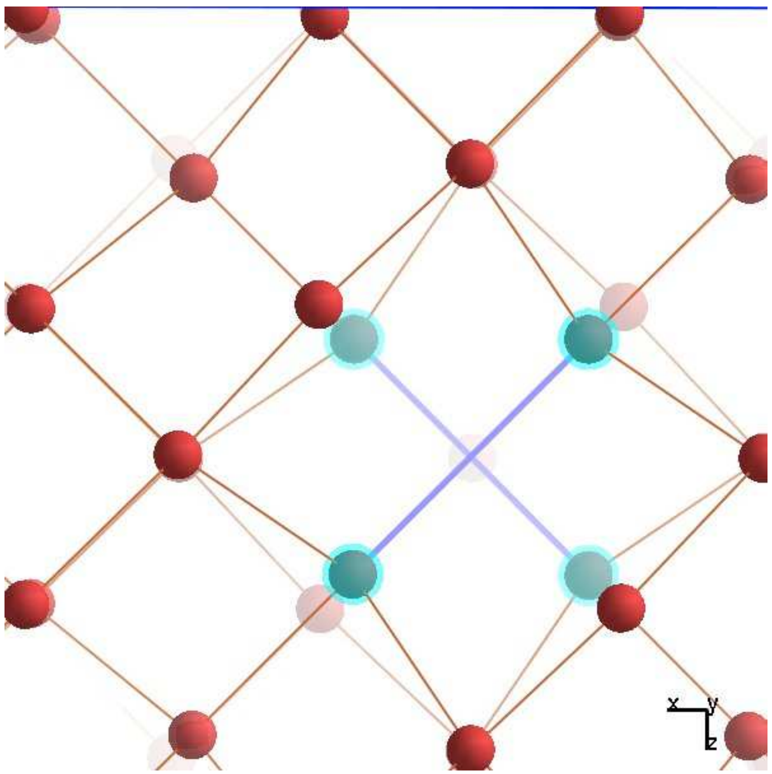
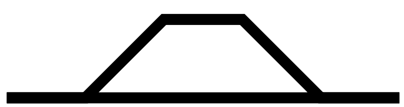
Vacancy in silicon

VACANCY IN SILICON:



$$v_y \longrightarrow T(v_y)$$

$$E_b = 0.34eV$$



System size: 215 atoms

Previous ab initio work

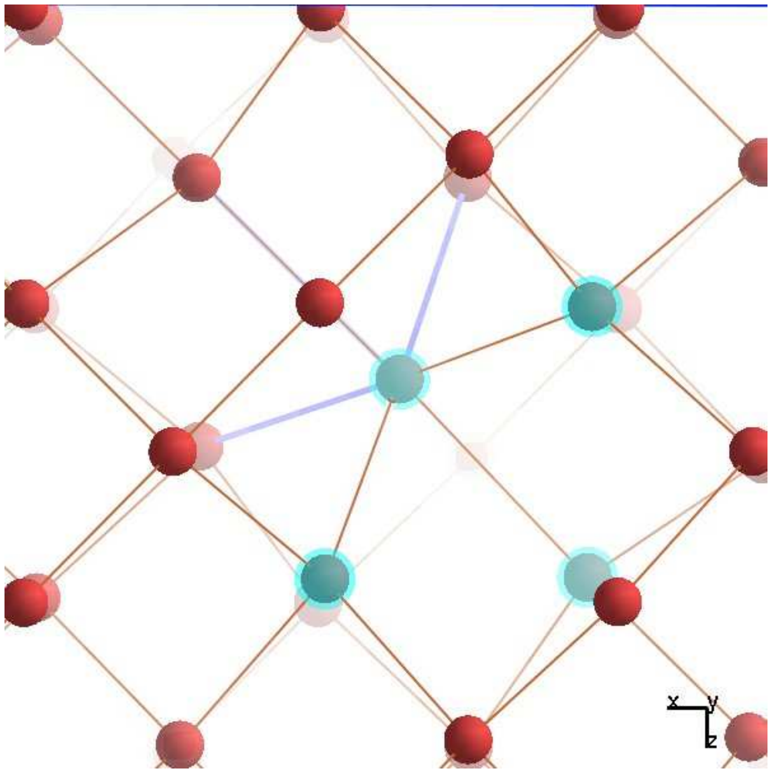
- El-Melluhi *et al.*

  PRB **70**, 205202 (2004)
- (with 4 CPU)
- ~ 700 force evaluations/event
- ~ 0.33 events/day

This work

- 430 CPU (1 orbital/proc)
- ~ 300 force evaluations/event

  ~ 5 events/day



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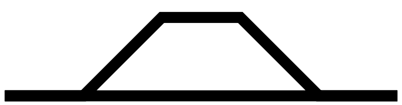

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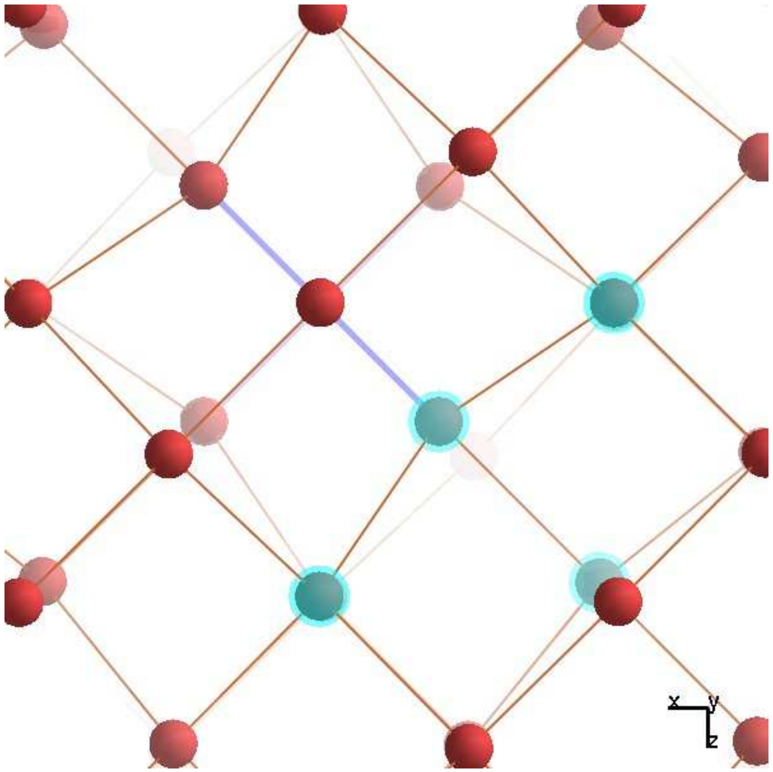

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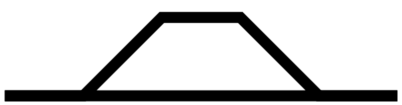

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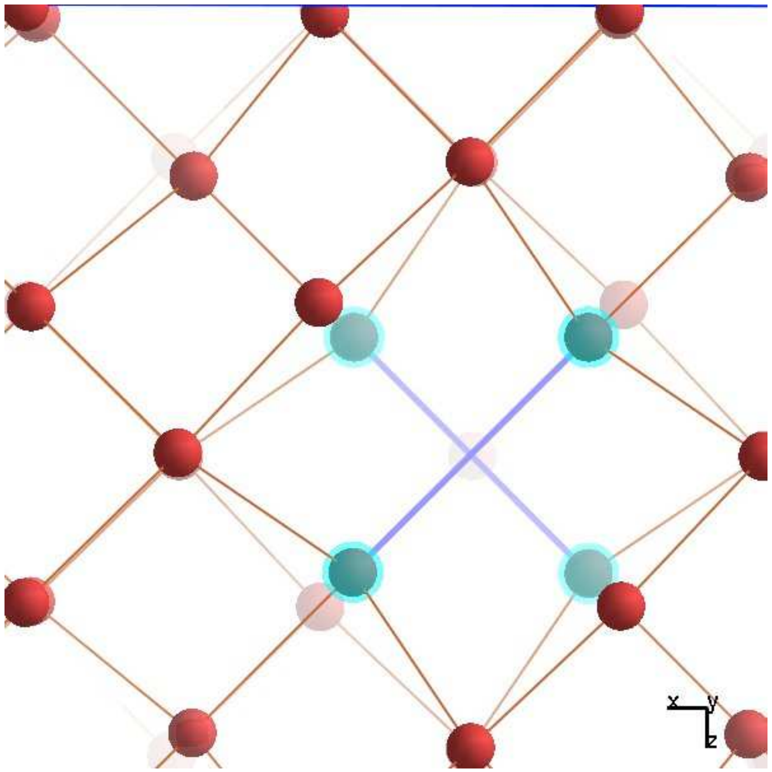
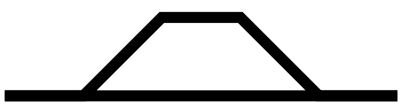


$$v_y \longrightarrow v_z$$

$$E_b = 0.27 eV$$



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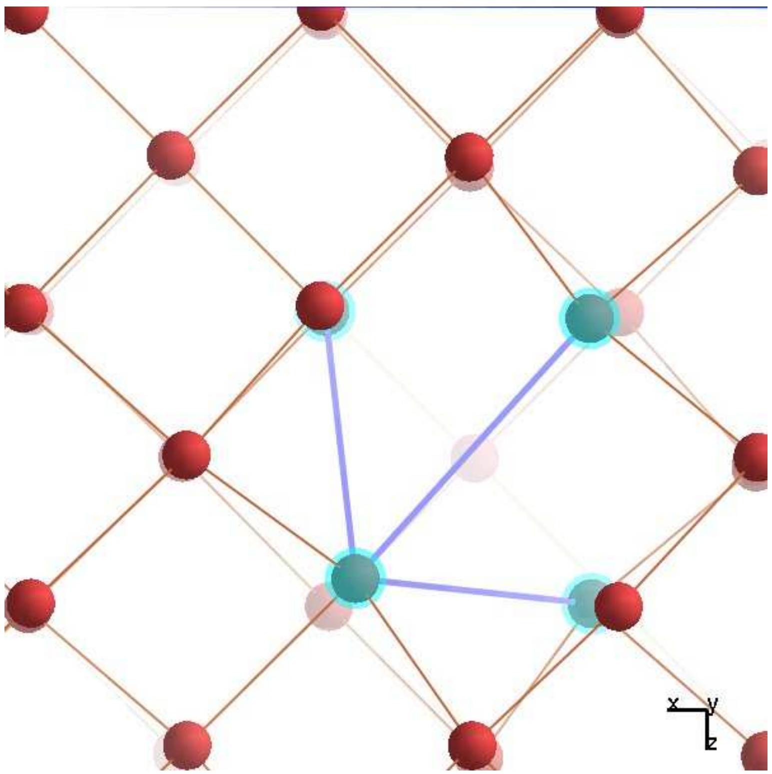

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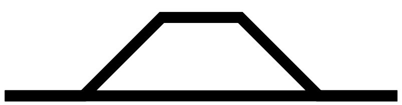

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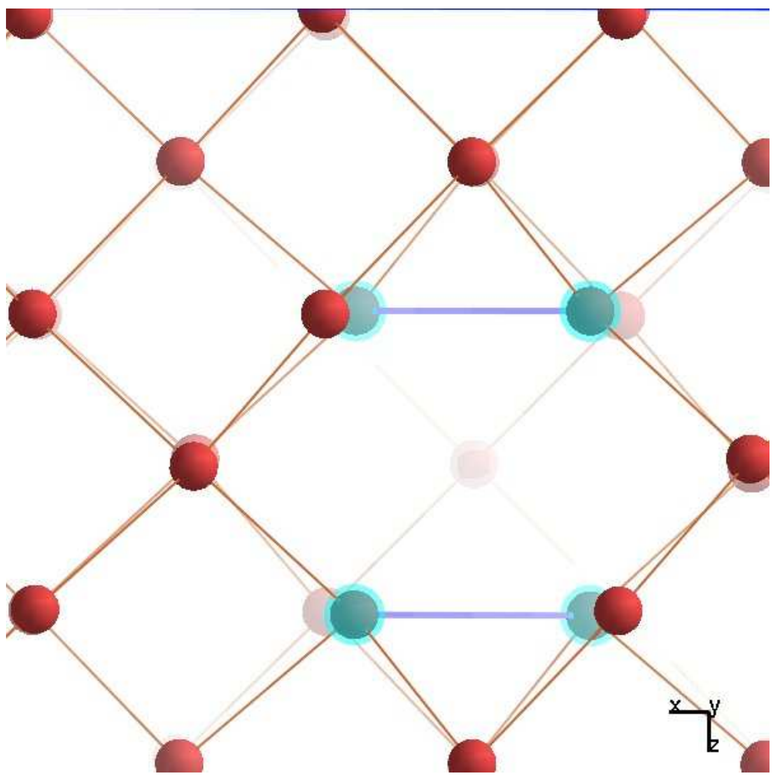

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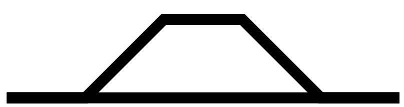

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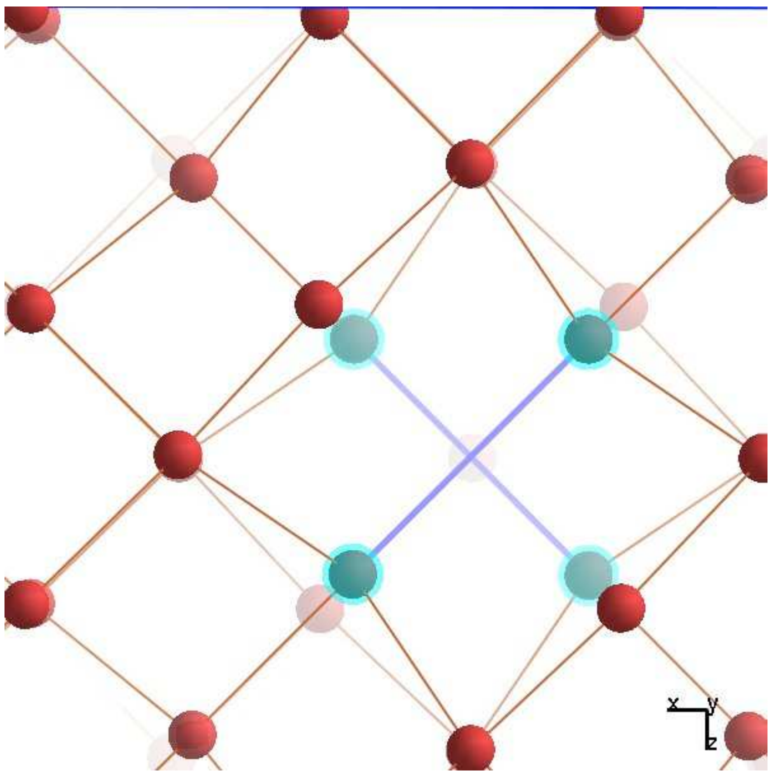

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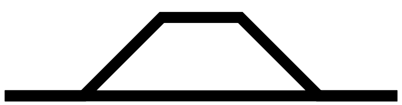

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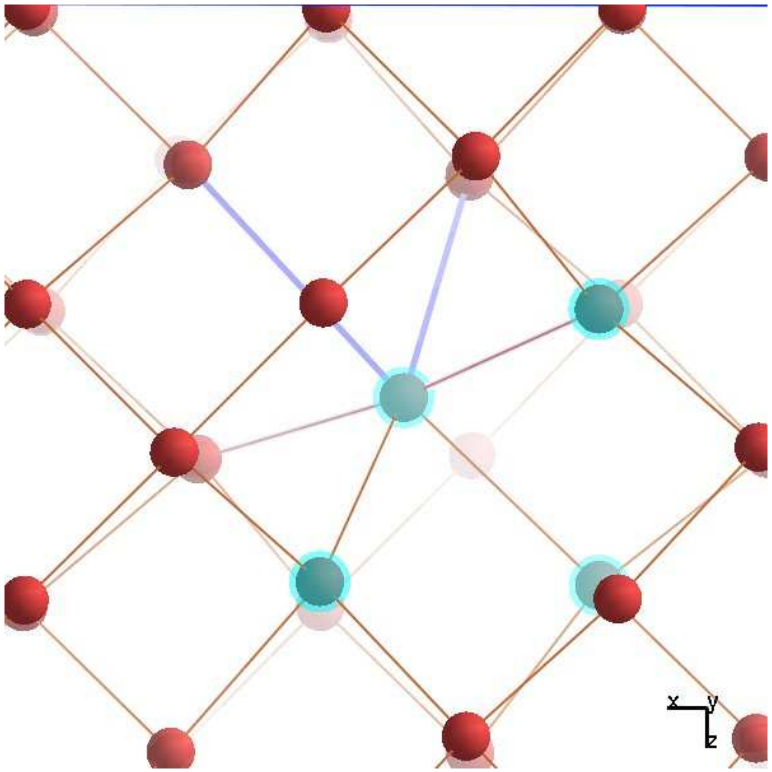

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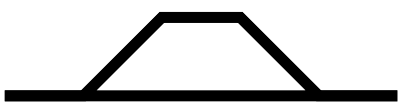

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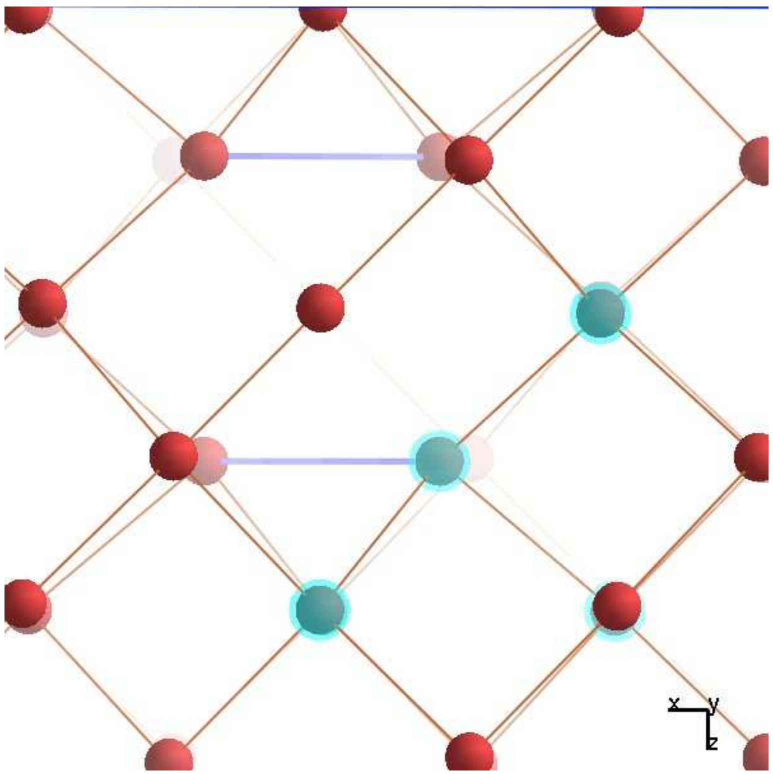

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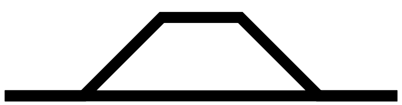

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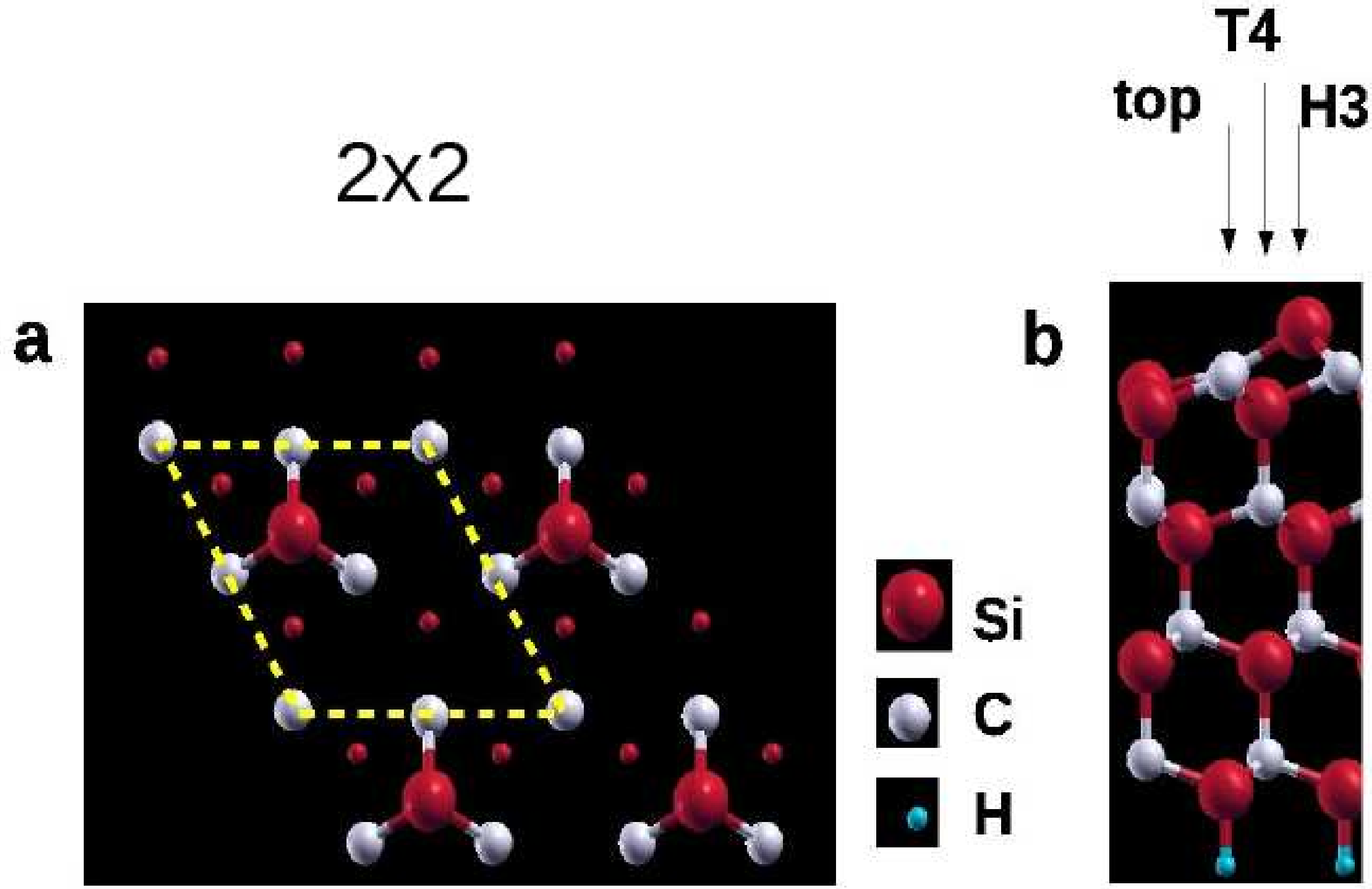
Ongoing work

Epitaxial graphene on SiC($000\bar{1}$)(C-face):



L. Magaud

PRB **79**, 161404 (2005), Phys. Stat. Sol. RRL **3**, 172 (2009)

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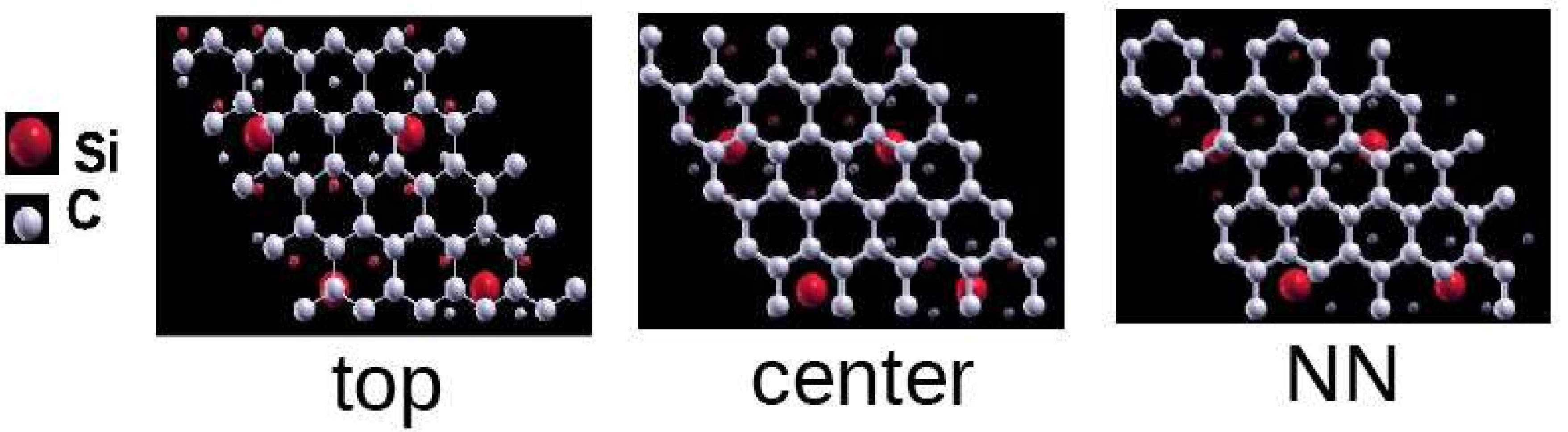


L. Magaud

PRB **79**, 161404 (2005), Phys. Stat. Sol. RRL **3**, 172 (2009)

Final remarks

- BART : An efficiency tool for growth study in nanosciences.
- Further efficiency improvement using GPU
- Need of further improvements in the DIIS method

THE LANCZOS METHOD:

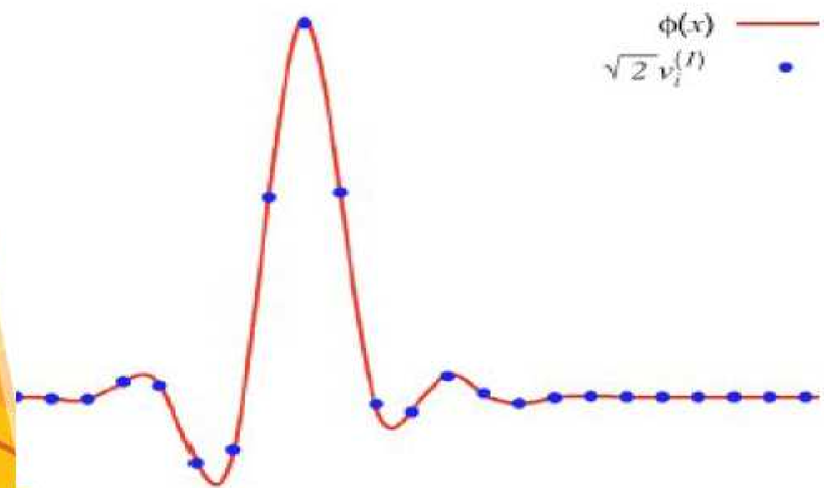
$$\begin{aligned} H|x_0> &= a_o|x_0> + b_1|x_1> \\ H|x_1> &= a_1|x_1> + b_1|x_0> + b_2|x_2> \\ H|x_2> &= a_2|x_2> + b_2|x_1> + b_3|x_3> \\ &\vdots \\ H|x_{l-1}> &= a_{l-1}|x_{l-1}> + b_{l-1}|x_{l-2}> + b_1|x_1> \\ H|x_1> &= a1|x_l> + b_l|x_{k-1}> \end{aligned} \tag{1}$$

$$L_l = \begin{pmatrix} a_0 & b_1 & & & & \\ b_1 & a_1 & b_2 & & & \\ & & & \ddots & & \\ & & & & b_{l-1} & a_{l-1} & b_l \end{pmatrix} \lambda_1[\mathbf{H}] \Rightarrow \lambda_1[\mathbf{L_l}](l \longrightarrow 3N).$$

Theory of Wavelets

- Scaling relation (SF)

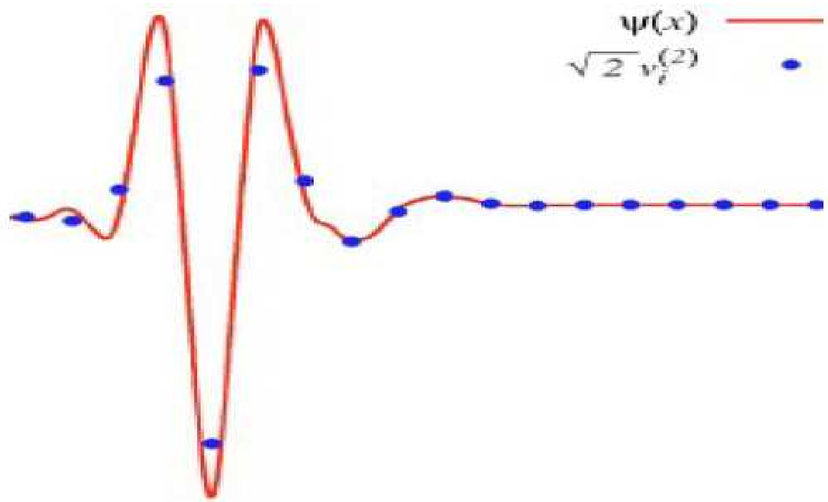
$$\phi(x) = \sum_{j=-m}^{m} h_j \, \phi(2x - j)$$



Theory of Wavelets

- Wavelets- nothing but more details

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



- Both have compact support and centered on grid

Particular application dictates which wavelets is selected

Wavelets in Action

The KS orbitals are expressed in ***Daubechies wavelet basis***

$$\Psi(\mathbf{r}) = \sum_{i_1,i_2,i_3} s_{i_1,i_2,i_3}\,\phi_{i_1,i_2,i_3}(\mathbf{r}) + \sum_{j_1,j_2,j_3} \sum_{\nu=1}^{7} d^{\nu}_{j_1,j_2,j_3}\,\psi^{\nu}_{j_1,j_2,j_3}(\mathbf{r})$$

where in practice,