Interplay between crack propagation and surface reconstructions in Silicon (111) brittle fracture

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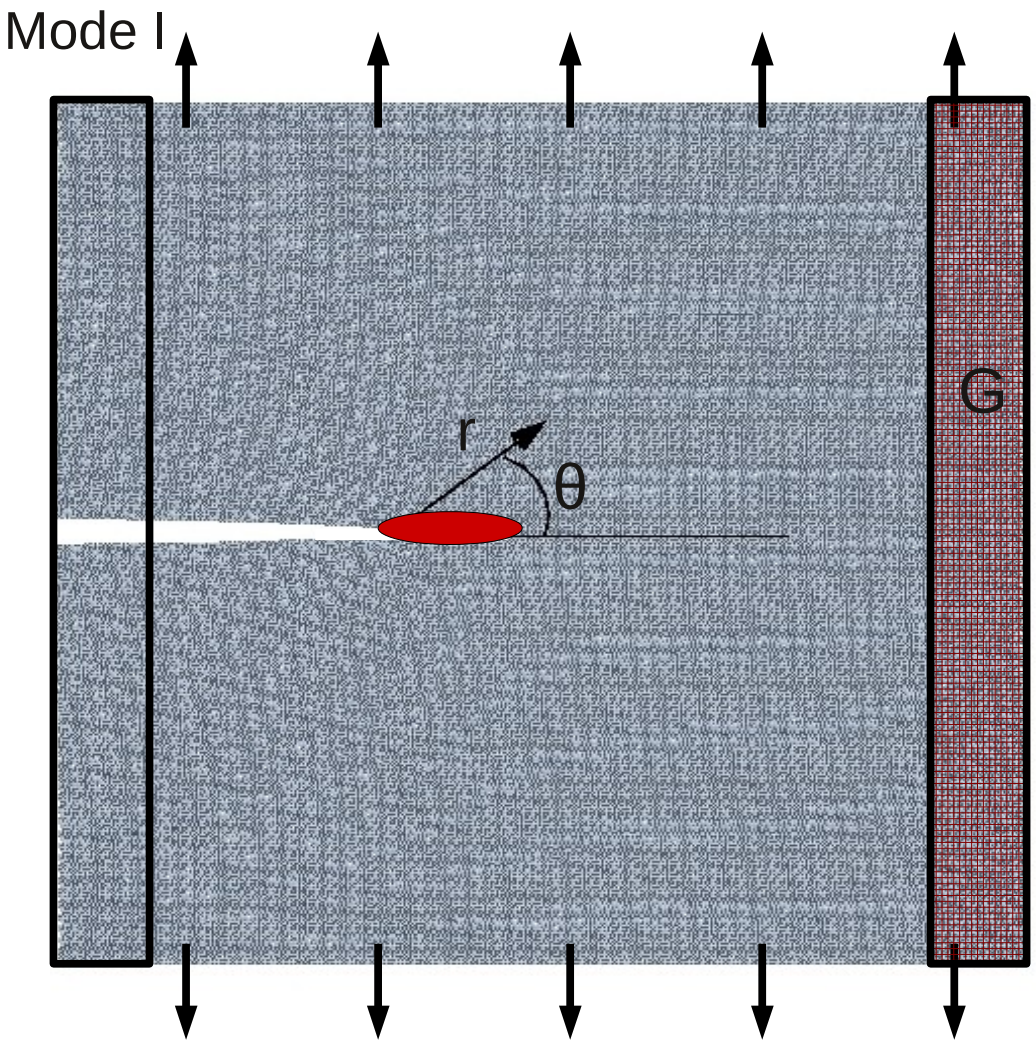
Problèmes de Fracture



Champ élastique longue porté :

$$u_i = K\sqrt{r}\, g_i(\theta) \qquad \sigma_{ij} = \frac{K}{\sqrt{r}} f_{ij}(\theta)$$

K : facteur d'intensité des contraintes

$$G = K^2 \frac{1-\nu^2}{E}$$

G : “énergie de fracture”

Critère de Griffith :

$G < 2\gamma_s$ fermeture

$G > 2\gamma_s$ ouverture

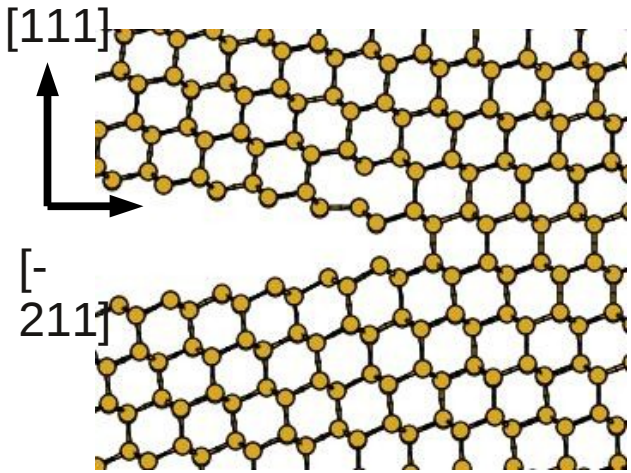
$G = 2\gamma_s$ pointe stable

Interaction pointe de fissure dynamique / Champ élastique → Système de grande taille

Description des ruptures de liens atomiques → Calculs quantiques

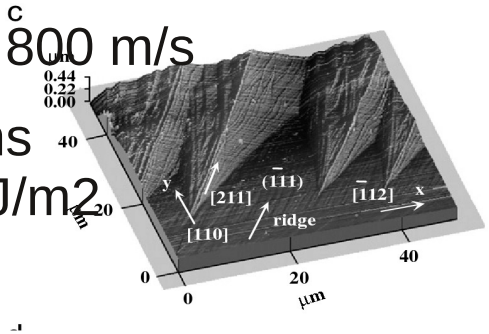
(111) Cleavage plane in Crystalline Silicon

(111)[-211]

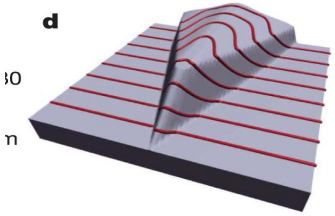


Stable dynamic crack above Vcrack ≈ 800 m/s

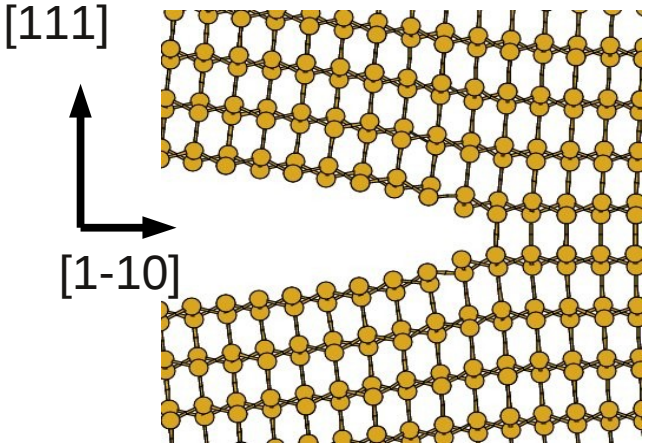
➔Fast propagation Vcrack 2000-3500 ms
"atomically" flat surfaces until G ≤ 16 J/m2
(J.Hauch et al. PRL 1999)



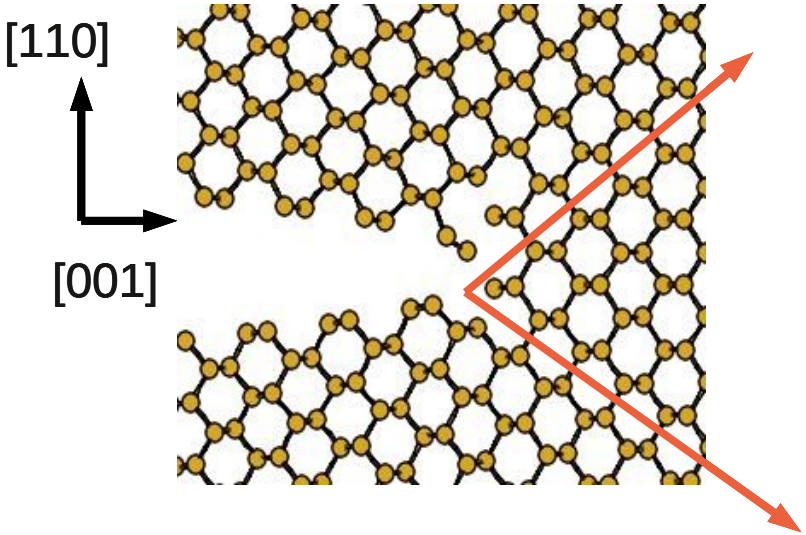
•At low crack velocities : macroscopic ridges due to low velocity crack tip reconstructions*(Sherman et al. J.Mech. Phys. Solids 2008, Kermode et al. Nature 2008)*



(111)[1-10]



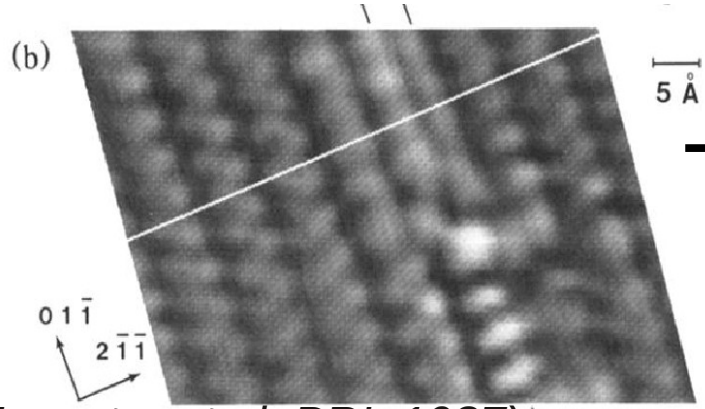
(110)[001]



Lattice trapping effect induced by the atomic-bond network

Some properties of (111) cleavage planes in Si :

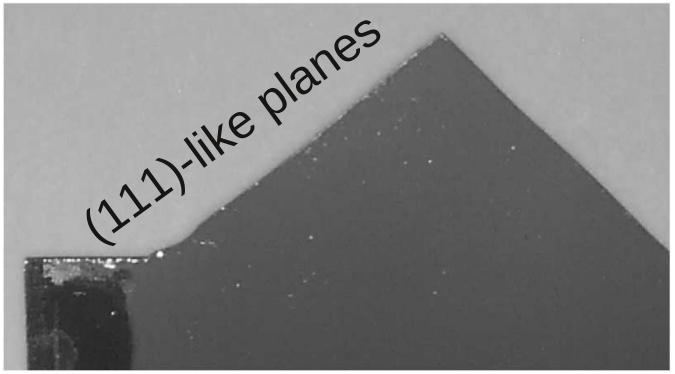
•(2X1) π-bonded reconstructions on both cleaved surfaces :



(Feenstra et al. PRL 1987)

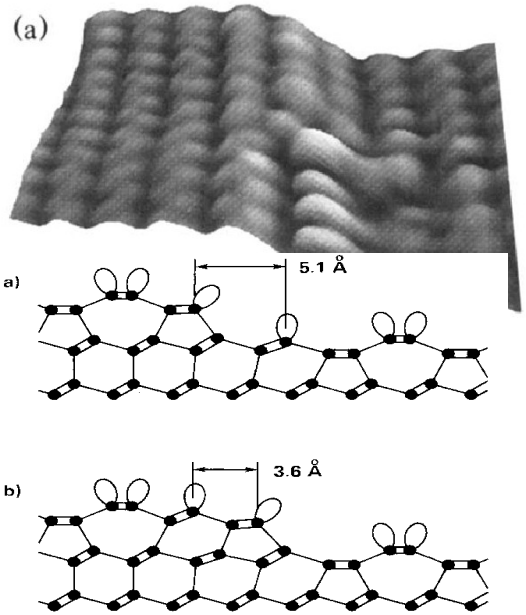
Not reproduced by simulations :
(2X1) π-bonded on one surface only !

•(111)[-211] system stable under shear :



(*Gumbsch et al. PRL 2000*)

•Presence of steps with specific symmetry :



Observed steps have
more dangling bonds !

•Description of surface reconstructions ?

•Stability under shear .vs. Step formation :
Apparent contradiction ?

LOTF SCHEME

A. De Vita, R. Car 1998
A. De Vita, G.Csanyi, T.albaret, J.Kermode, G. Moras (2004-2008)

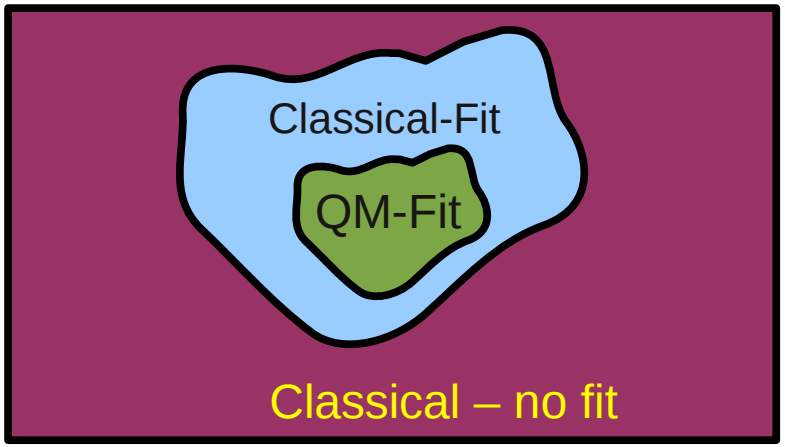
Describe the whole system with an adaptable classical potential :

$$V(c_{ij}, c_{ijk}, \ldots) \Rightarrow V(c_{ij}(r_{ij}, t), c_{ijk}(r_{ijk}, t), \ldots)$$

The potential "Learns On The Fly" from force fitting procedures in given regions of space

Goal : calculate "expensive" forces only where and when it is needed

Hybrid method in space :



...in time

C_{ij} parameters are usually updated periodically :

tn → Fit

tn + n_{int} dt → Fit

⇒ Speed up : n_{int} ~ 10

Propagation of coordinates

Extrapolation: n_{int} time steps at constant parameters

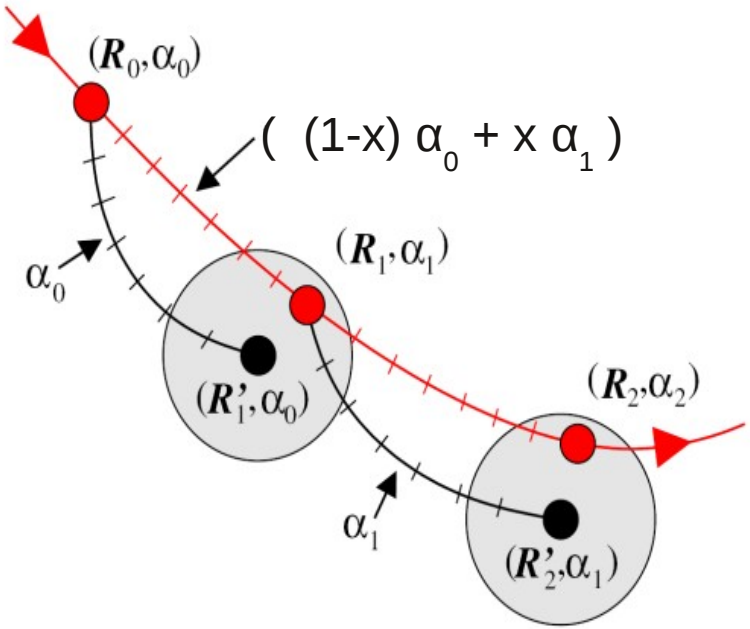
Selection: define the “Quantum atoms” and the fitting region

Force calculation : classical and quantum

Fit : adapt C_{ij} parameters of the potential to match the forces

Interpolation: interpolate parameters over n_{int} MD time steps

Extrapolation-Interpolation cycle :

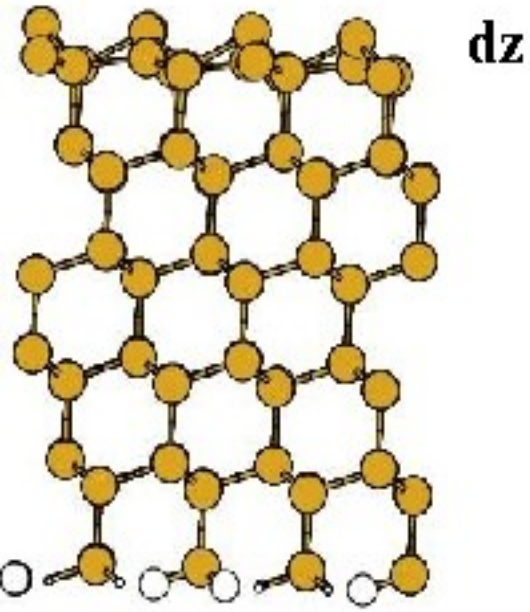


Tests statiques

Fit de a0, B0 et C44 pour accorder SW avec le potentiel liaisons fortes

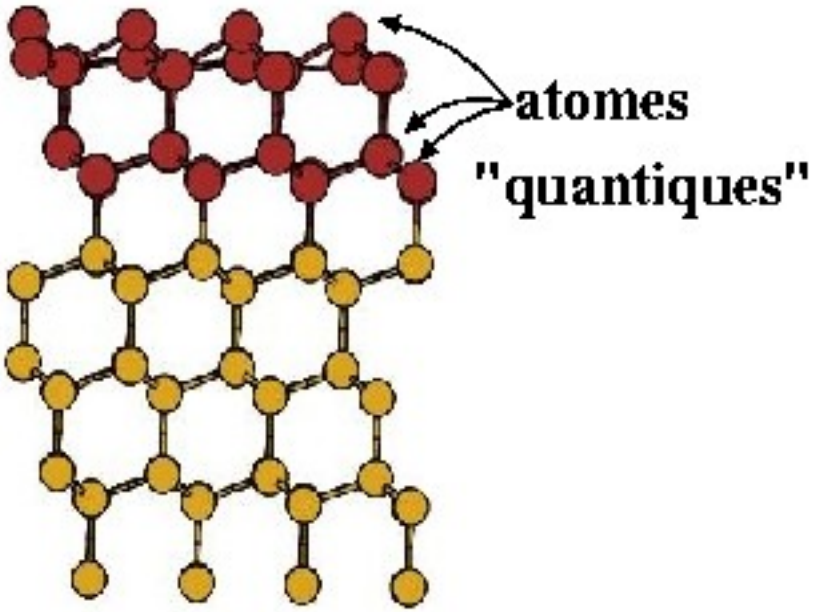
Surface Si(111)

optimisation de géométrie



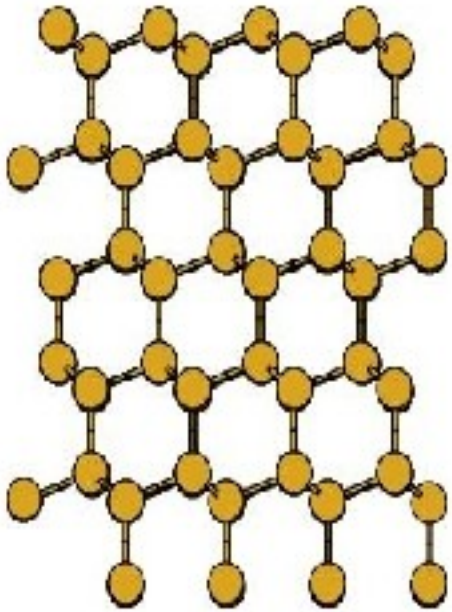
Liaisons fortes

dz = 0.9 A



Lotf

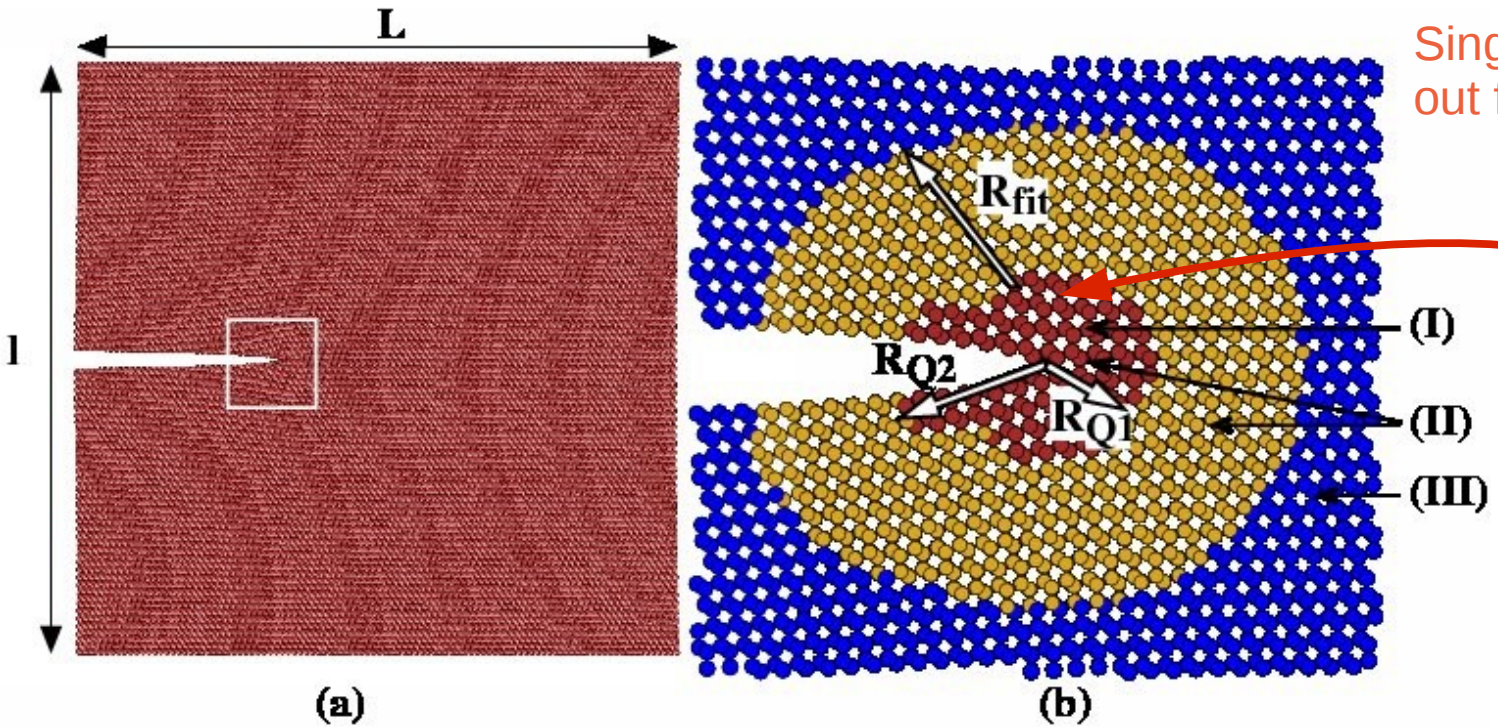
dz = 0.8 A



SW

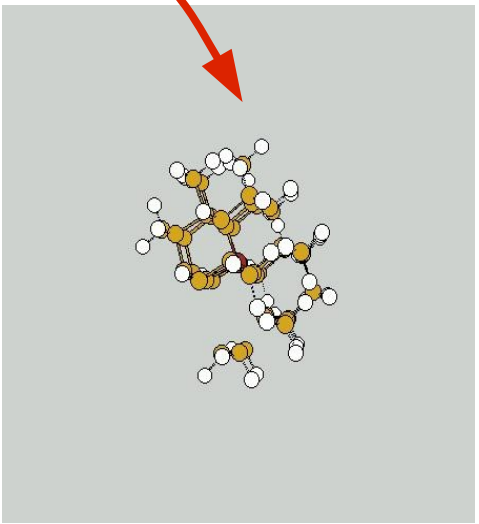
dz = 0 A

Application to the fracture in Silicium



Single cluster is carved out for each “quantum” atom

Easy parallelisation !

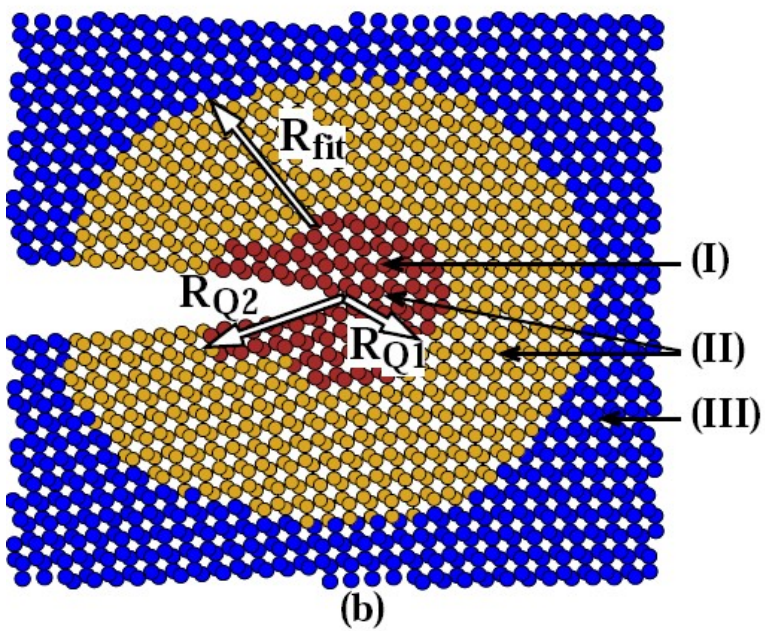


- L ≈ l ≈ 70 nm, front direction : 2 unit cells (≈8 A)
- ~200000 atoms, ~ 2000 fitted, ~300 quantum
- Classical Potential : Stillinger-Weber (3 adjustable parameters)
- QM systems : H-terminated clusters (100-300 atoms)
- QM calc. : DFT (SIESTA), SZP basis, LDA, 50 Ryd.
- The classical potential reproduces the lattice parameter, B0 and C44 given by the quantum potential (SIESTA)

Stillinger-Weber potential :

SW (2body): $$V_2(r) = A\left(\frac{B}{r^4} - 1\right)\exp\left(\frac{1}{r - rc}\right)$$

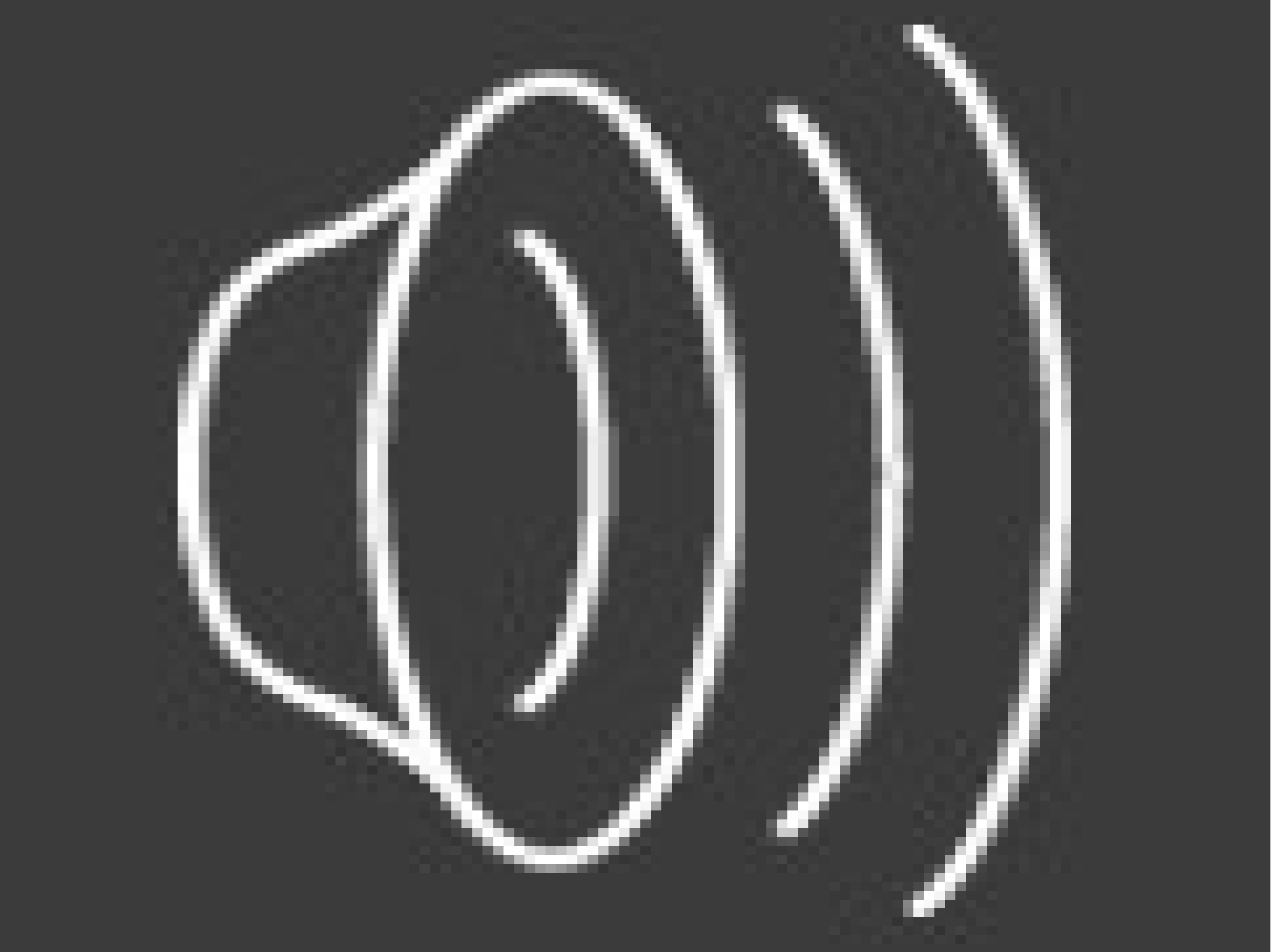
SW (3body): $$V_3(r_{ij}, r_{ik}) = \lambda \exp\left[\frac{\gamma}{(r_{ij} - rc)} + \frac{\gamma}{(r_{ik} - rc)}\right]\left(\cos(\theta_{ijk}) + \frac{1}{3}\right)^2$$

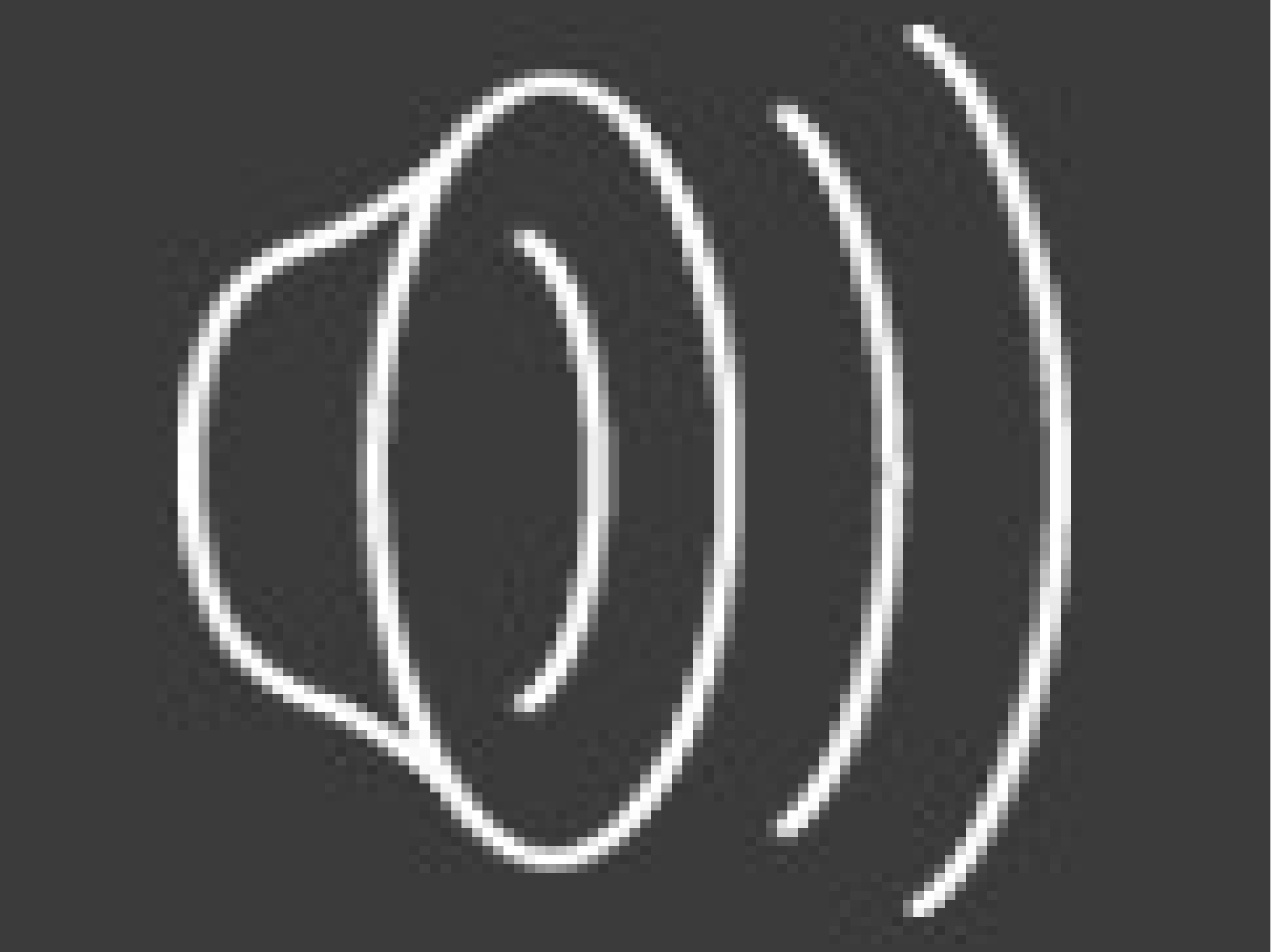


	R_{Q1}(Å)	R_{Q2}(Å)	R_{fit}(Å)	R_c(Å)
$LOTF - SIESTA$	10	16.5	23	6.6

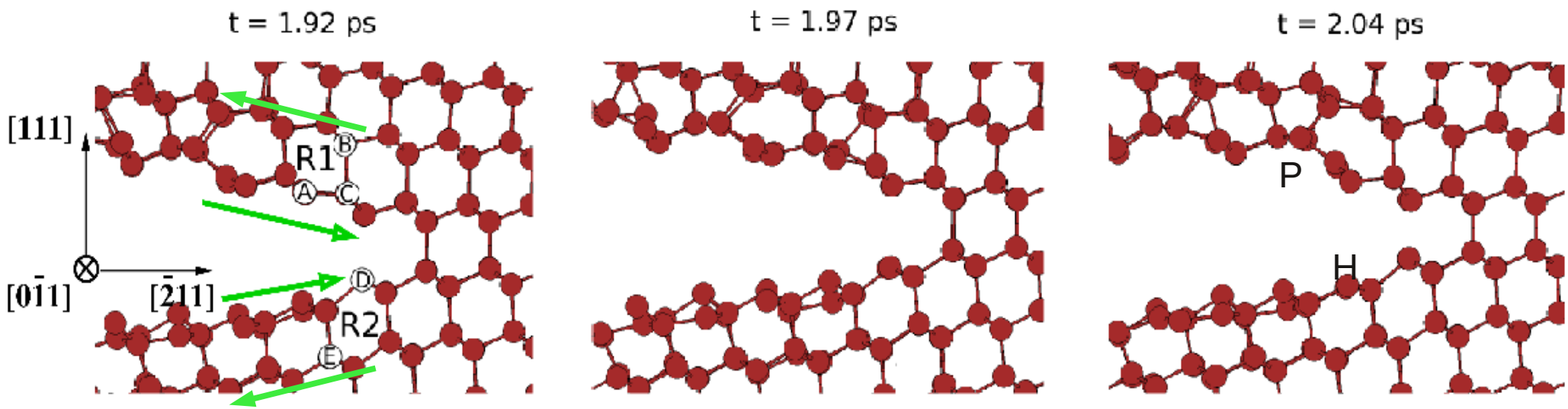
	A (SW unit)	B (SW unit)	λ (SW unit)	$\cos(\theta)$
original SW	7.0496	0.60222	21.0	$-\frac{1}{3}$
reference SW	7.0267	0.5948	34.3112	$-\frac{1}{3}$

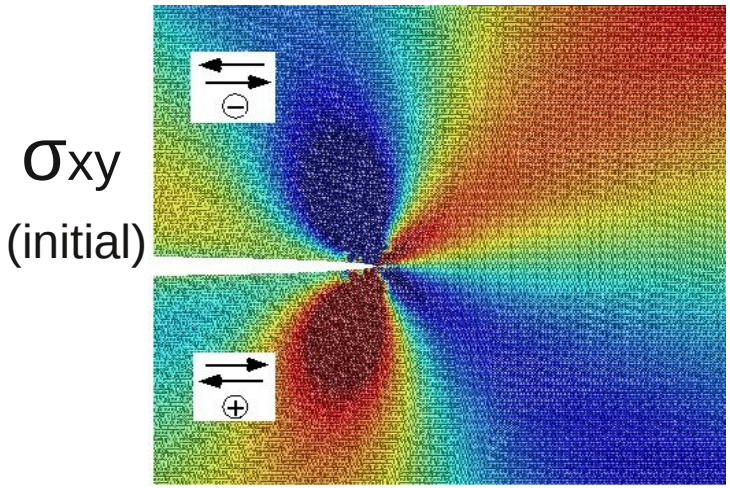
	a_0(Å)	$C_{11}(GPa)$	$C_{12}(GPa)$	$C_{44}(GPa)$
$Exp.$	5.431	166	64	80
$SIESTA(SZP)$	5.421	171	67	81
reference SW	5.421	184	60	81

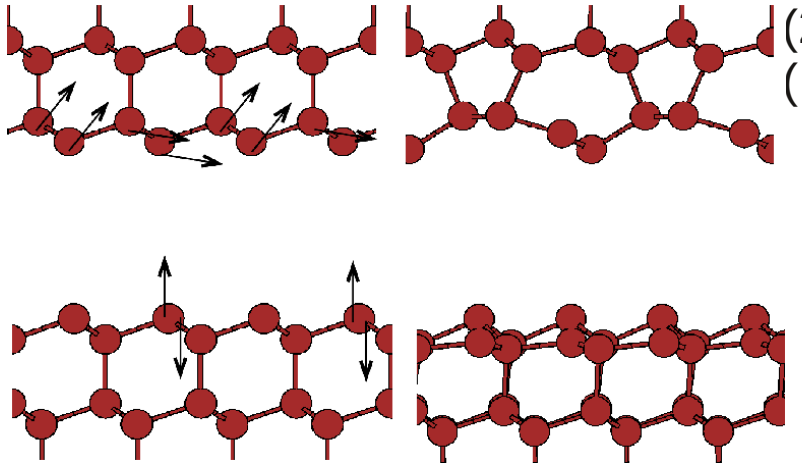




LOTF / Si(111)[-211]
initial propagation (1000ms-1 ≤ v ≤ 2000 ms-1)







(2x1) Pandey reconstruction
(π bonds along [0-11])

↓ ↓

Shear stress relaxation

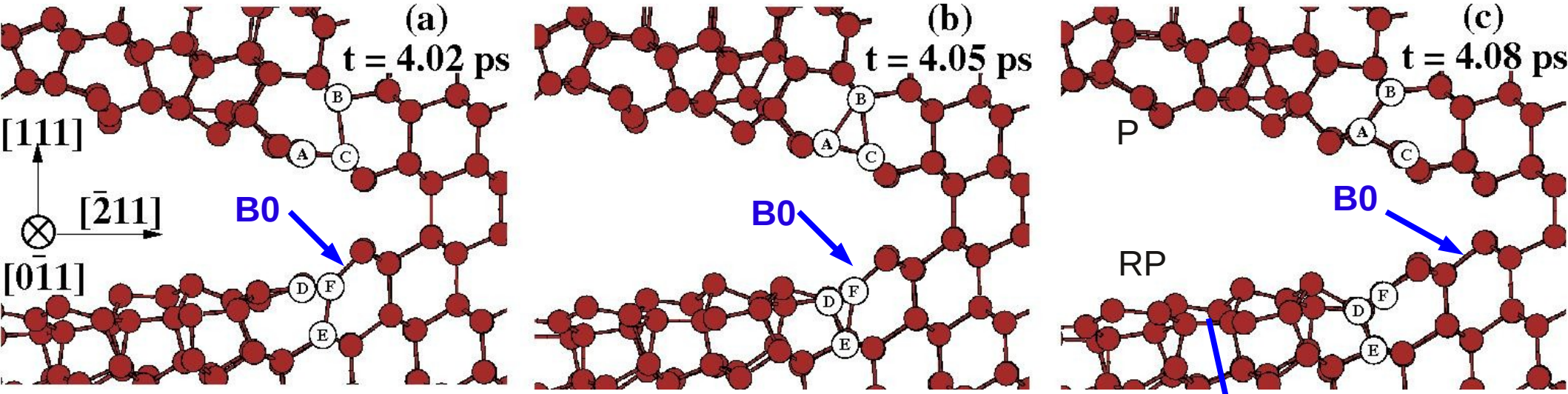
Haneman reconstruction

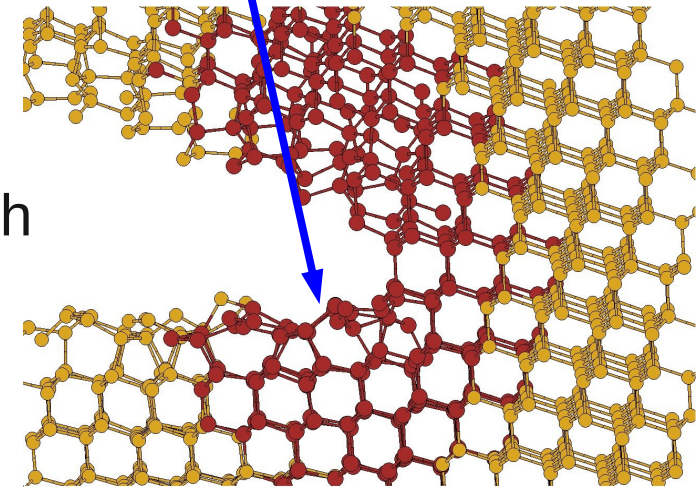
↓ ↓

No shear stress relaxation

- Atomic structures consistent with the shear stress at the tip
- Not in full agreement with experiments ...
- Results similar to previous calculations (20 000 atoms, *Hoshi et al PRB 2005*)

LOTF / Si(111)[-211]
2nd propagation regime (2000 ms-1/3000ms-1)



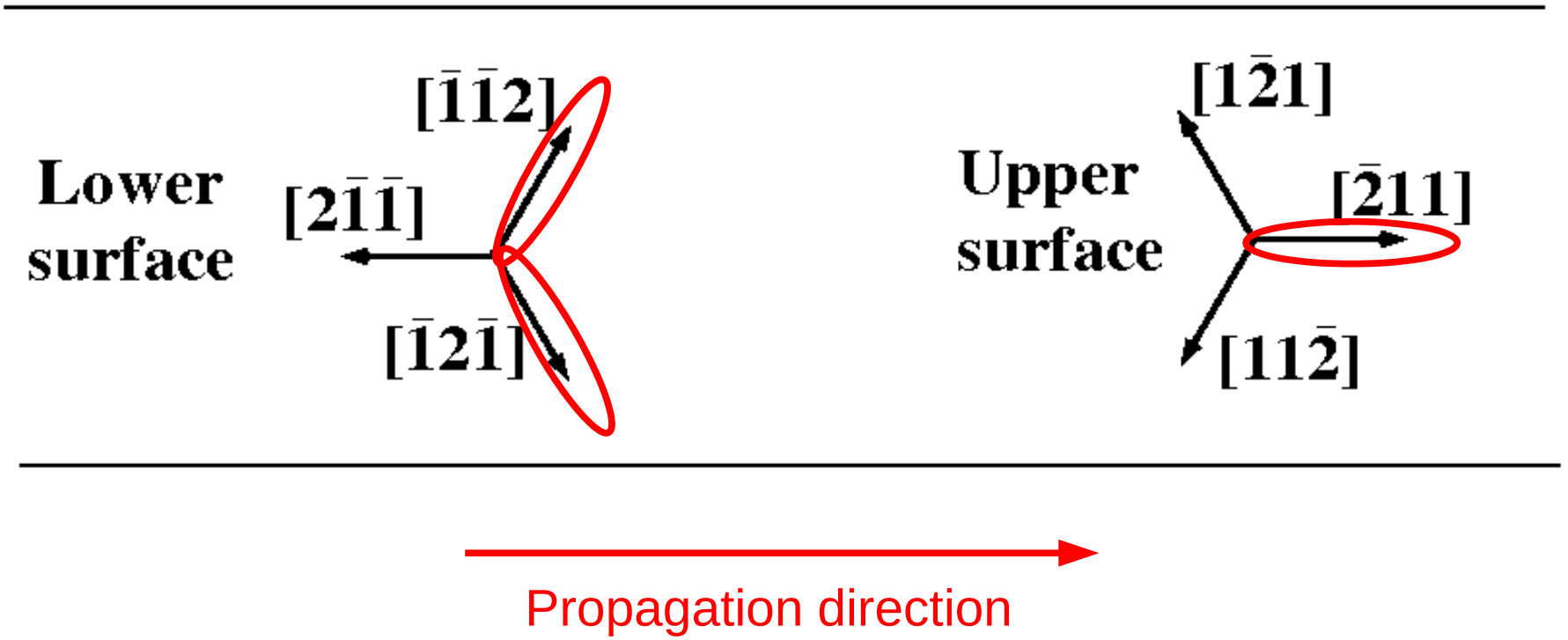


- (2X1)π-bonded reconstructions obtained on both surfaces : regular and rotated by $60^{0.}$
- Surface structures are asymmetric but consistent with both experiments and shear stress field at the tip
- Shear stress relaxation occurs on both surfaces, B0 bond is "protected" by the specific reconstruction mechanism on the lower surface

→ The surface reconstructions provide a mechanism to stabilize the (111)[-211] crack system

Stress compliant π-bonded reconstructions

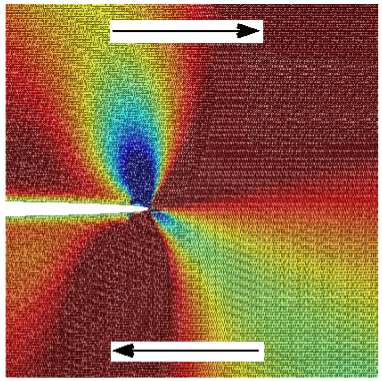
Main relaxation directions for the possible π-bonded reconstructions :

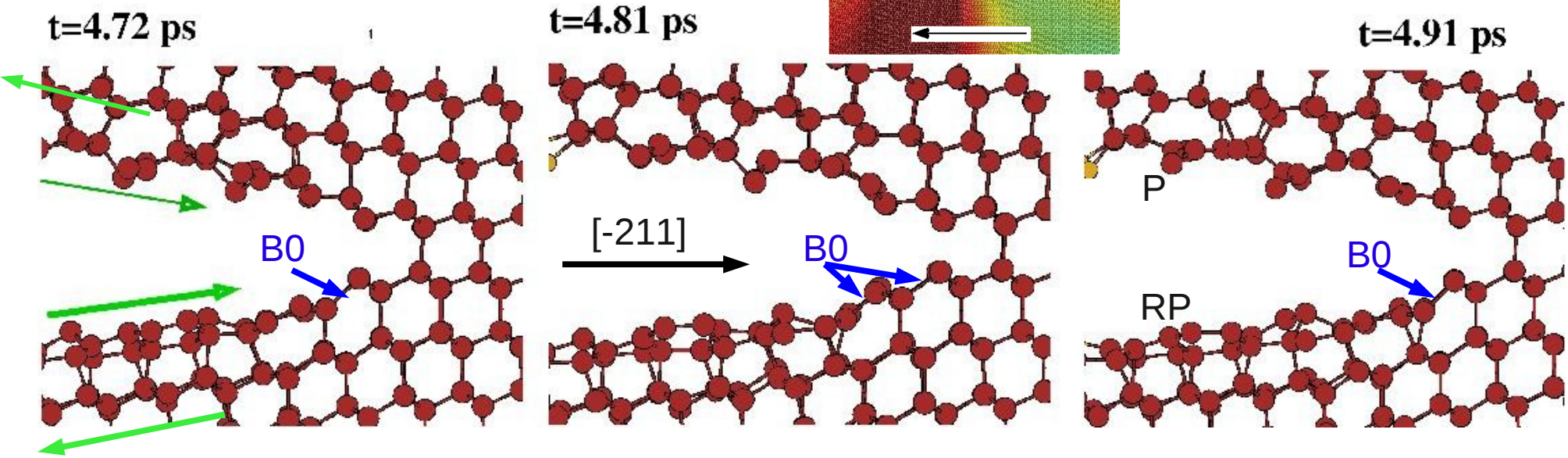


Si(111)[-211] under mixed mode I-II

(1.2% shear strain imposed on the top and lower planes (19° /[111]), G= 7.6 G/m2)

“Positive” Shear :



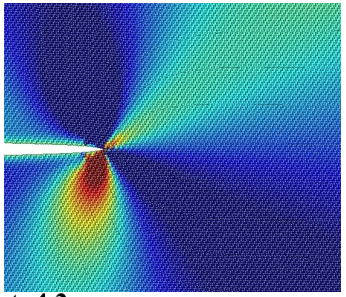


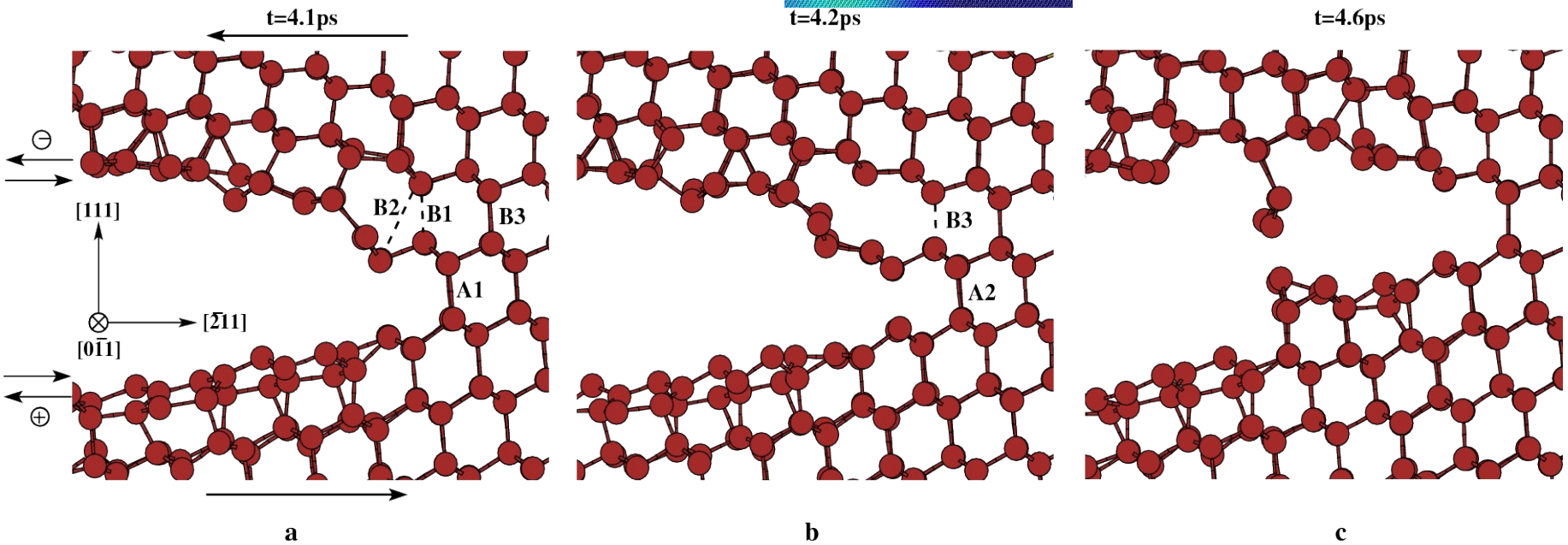
- No step formation, propagation similar as pure mode I
- Positive shear stress favours the “rotated” π-bonded reconstruction, this reconstruction “protects” bond B0 which in turn favours a [-211] propagation

Si(111)[-211] under mixed mode I-II

(1.2% shear strain imposed on the top and lower planes (19° /[111]), G= 7.6 G/m2)

“Negative” shear :

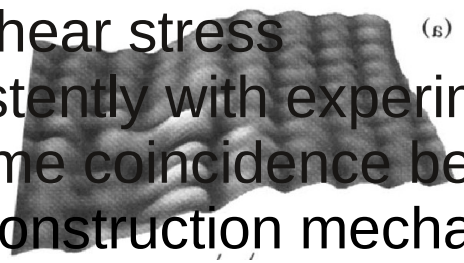




•the reconstruction mechanism on the upper surface is easily perturbated by a negative shear stress

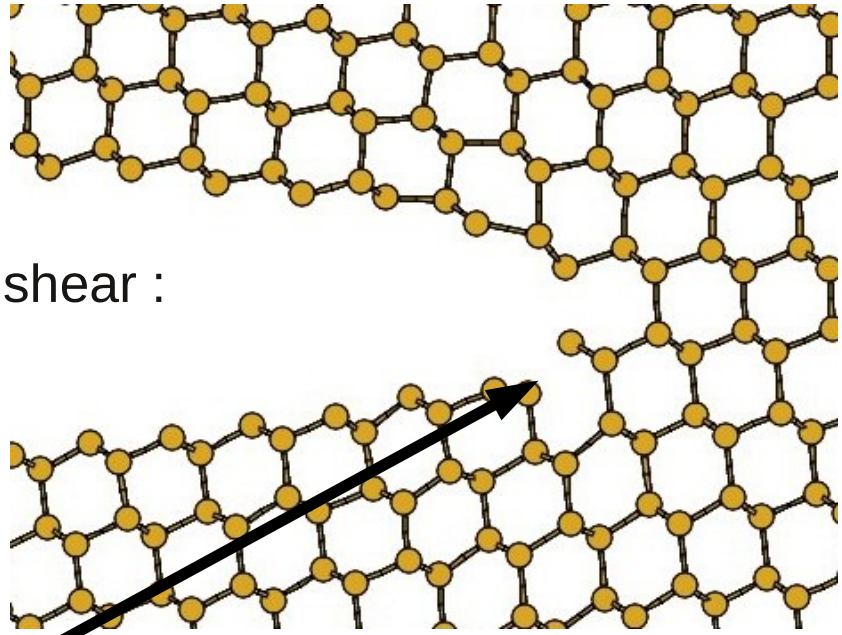
•[2-1-1] steps are produced consistently with experiments

•steps occur depending on the time coincidence between regular bond breaking and the reconstruction mechanism

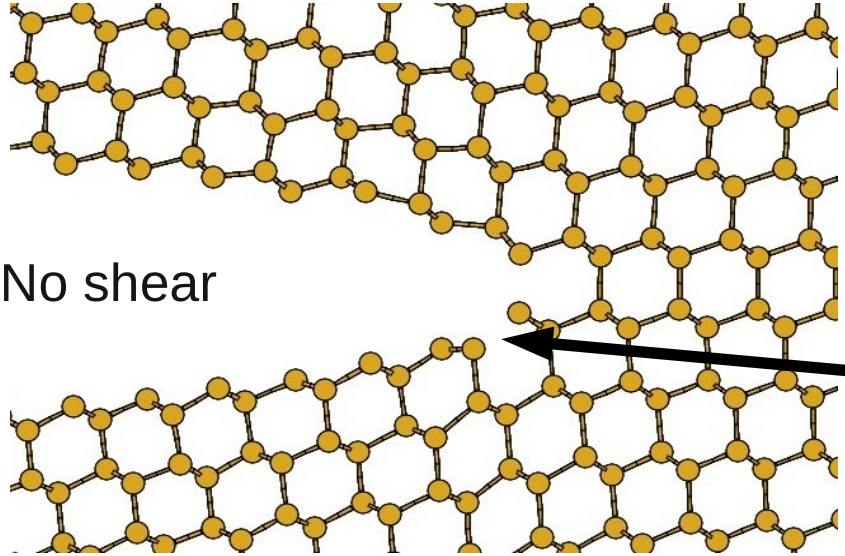


[-211]

Classical Simulations
Si(111)[-211] :



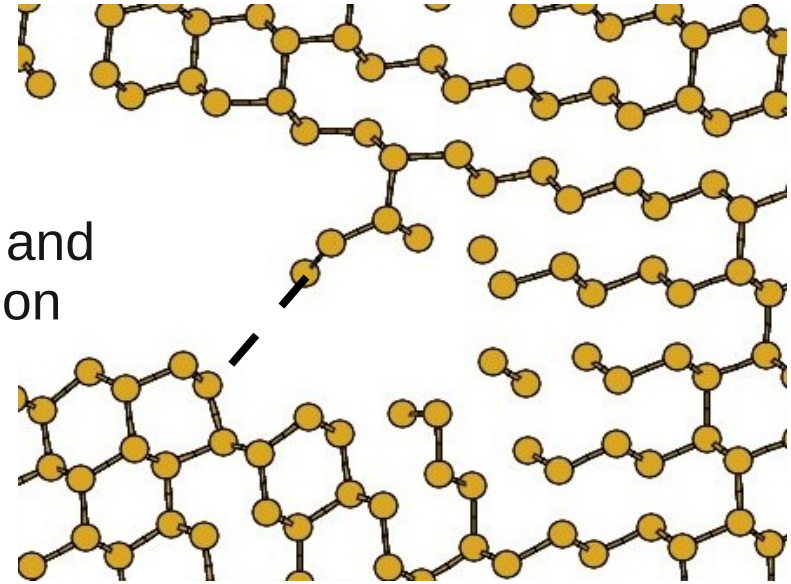
Negative shear :
No step



No shear

B0 bond

Positive shear :
B0 bond breaking and
[-211] step formation



Results opposite to experiments

Conclusions

- (2X1) π-bonded asymmetric reconstrustructions are obtained on the two cleavage planes

- These structures are consistent with experimental observations and with the shear stress field at the tip

- The associated displacements contribute to reduce the shear stress field in the tip vicinity which provides a stabilization mechanism for the Si-(111)[-211] crack propagation

- The asymmetric surface structures leads to an asymmetric response respect to a shear perturbation : straight propagation or formation of [2-1-1] steps

→ Explains both the observed predominant [-211] step structures seen in the experiments and the high stability of this crack system under shear

Acknowledgements

- Delia Fernandez Torre (Universidad Autonoma de Madrid)
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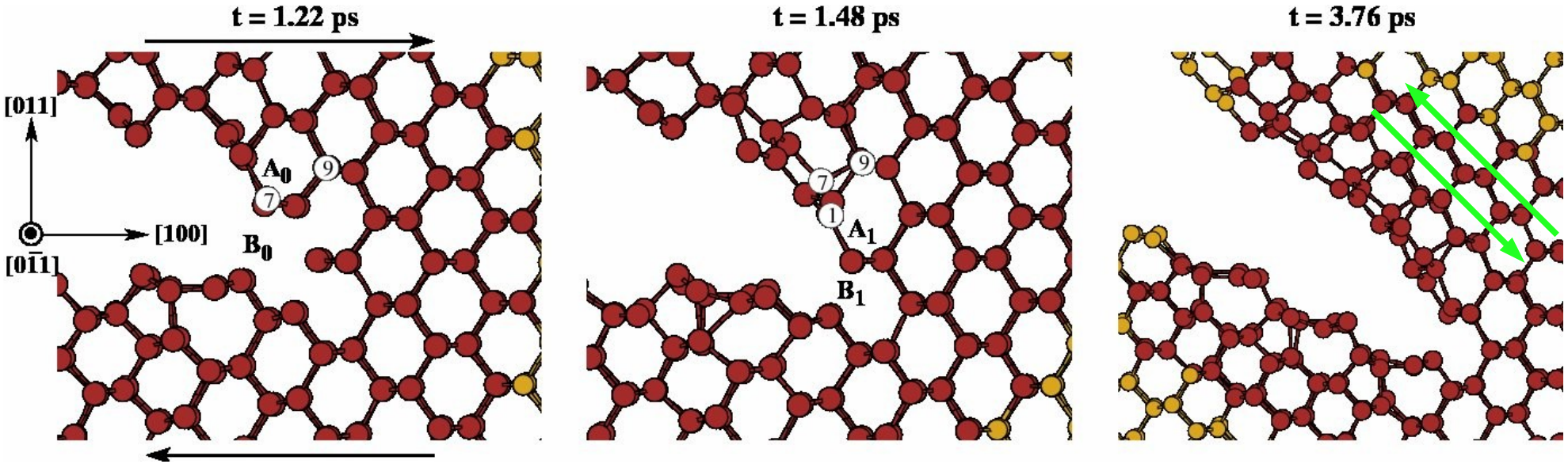
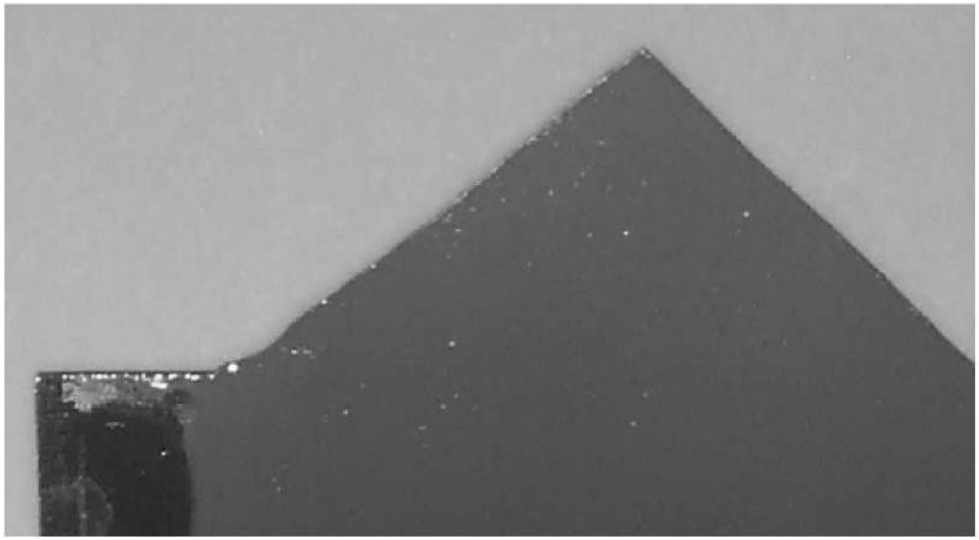
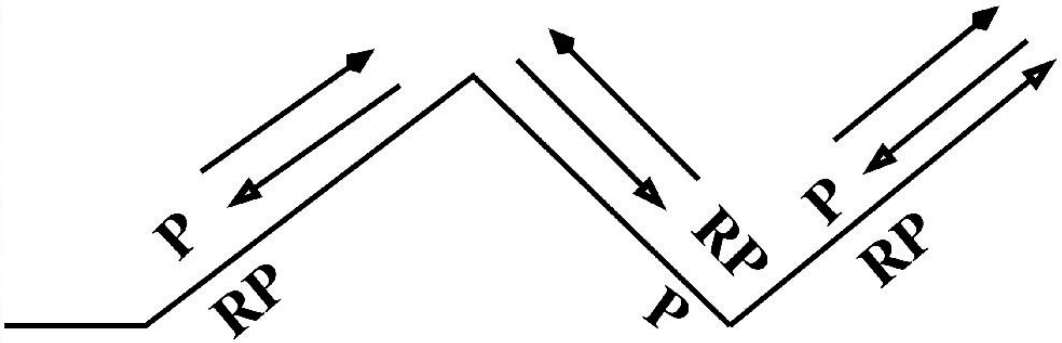
- Massimiliano Stengel (Barcelona)

Fernandez-Torre, Albaret, De Vita Phys.Rev.Lett. 2010

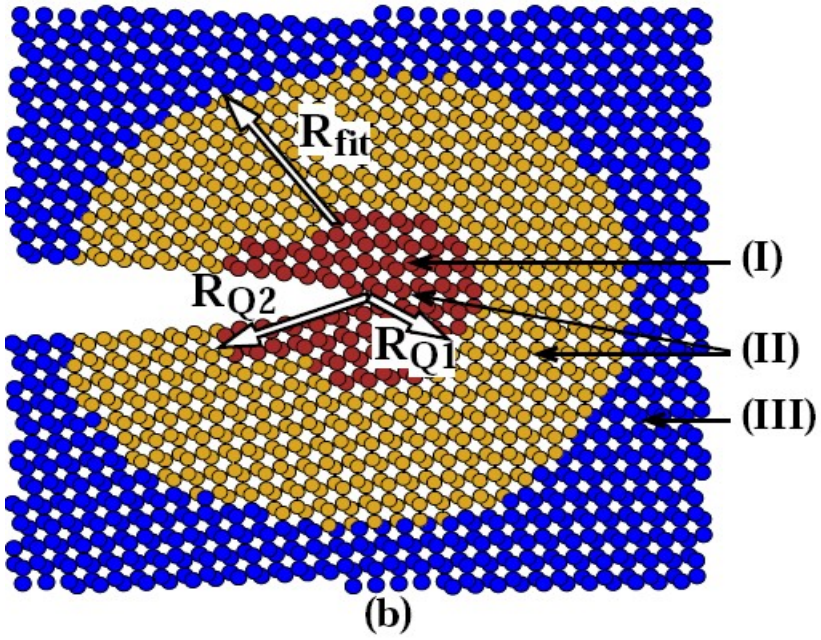
Si(011)[100] : mixed mode I-II





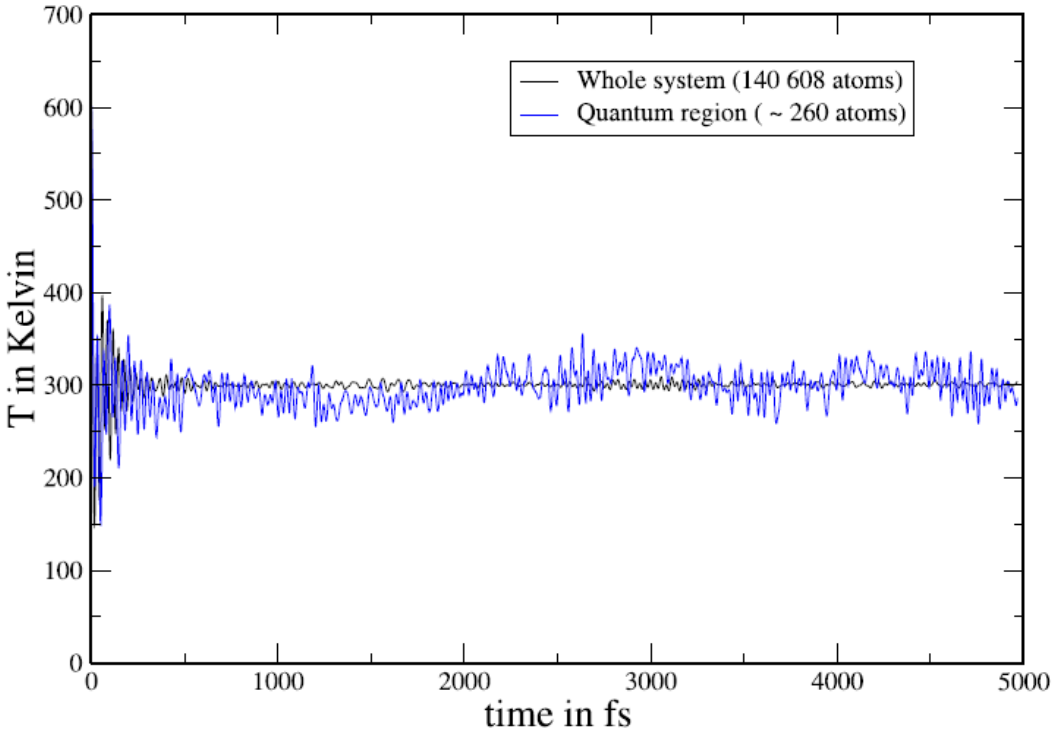


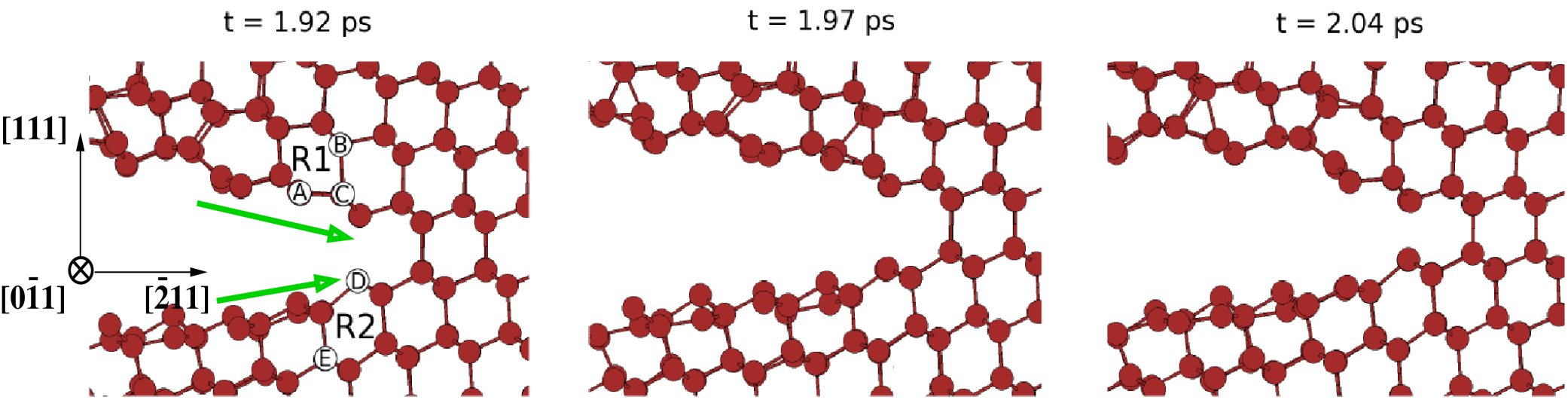
$$Min\left\{\sum_{i,x}\left|F_{ref}^{i,x}-F_{SW}^{i,x}\left(\{A\},\{B\},\{\cos\theta\}\right)\right|^2\right\}$$



- assure l'action/reaction
- répartition “homogène” des erreurs
- Précision du fit ≈ 10^{-3} eV/Å par atome
- Précision entre 2 fits ≈ 10^{-2} eV/Å par atome

Calcul “microcanonique”
positions initiales : cristal, 600K
zone quantique mobile (1000 m.s-1)



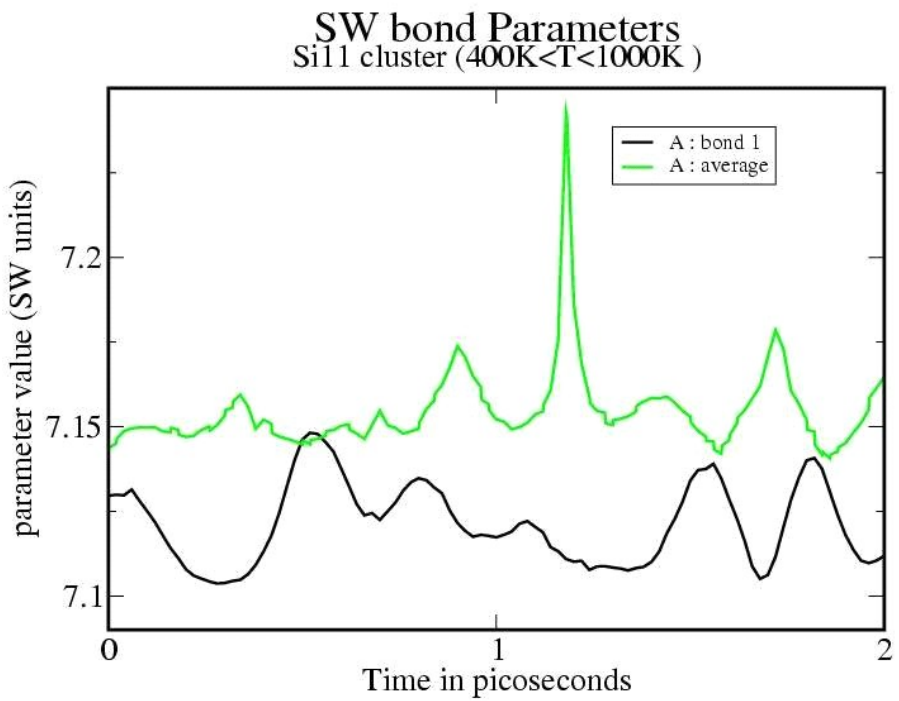

t = 1.92 ps
t = 1.97 ps
t = 2.04 ps
[111]
[0$\bar{1}$1]
[$\bar{2}$11]
R1
R2
A
B
C
D
E

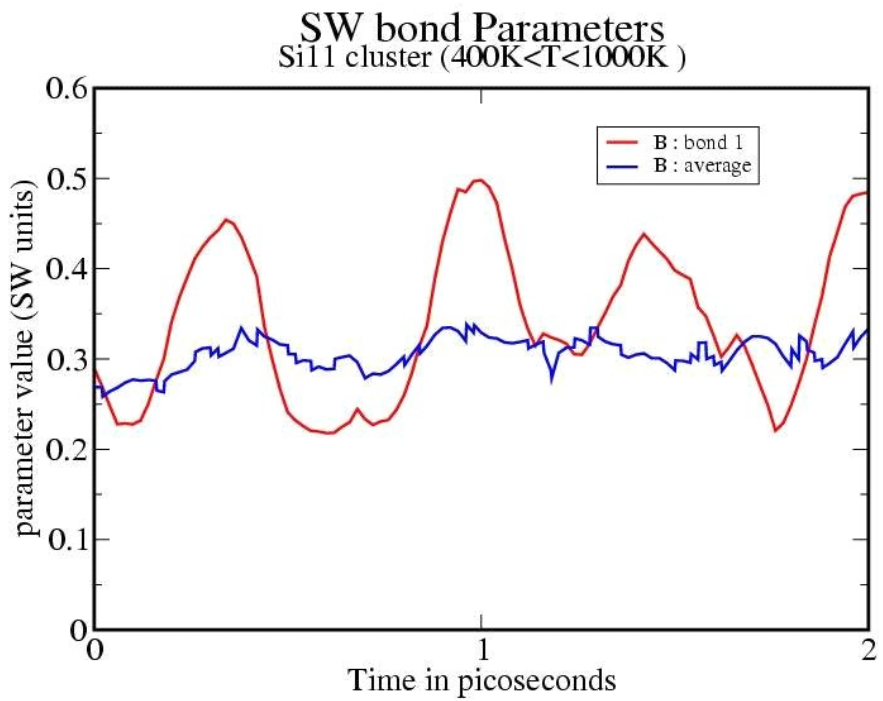
Evolution of the adaptable Parameters

SW original parameters : A = 7.05 B = 0.602 λ = 21

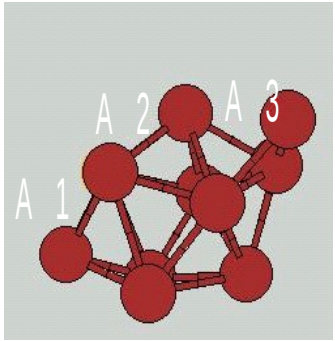
Sw parameters of reference : A = 7.11 B = 0.6 λ = 25.6

(this calculation)





Accuracy of the fit ?



Quantum forces are calculated by :

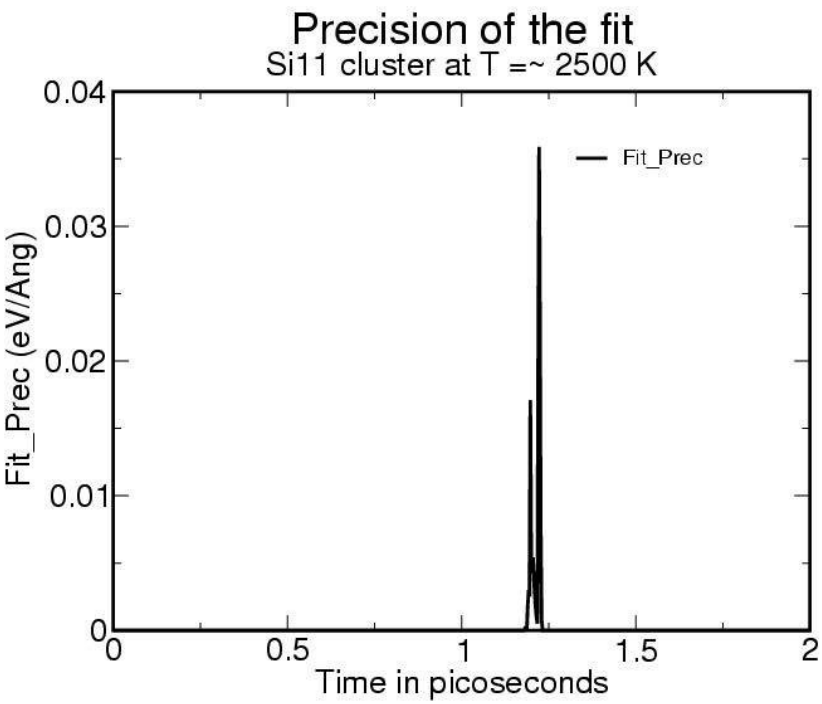
1) Non Self Consistent tight-binding model

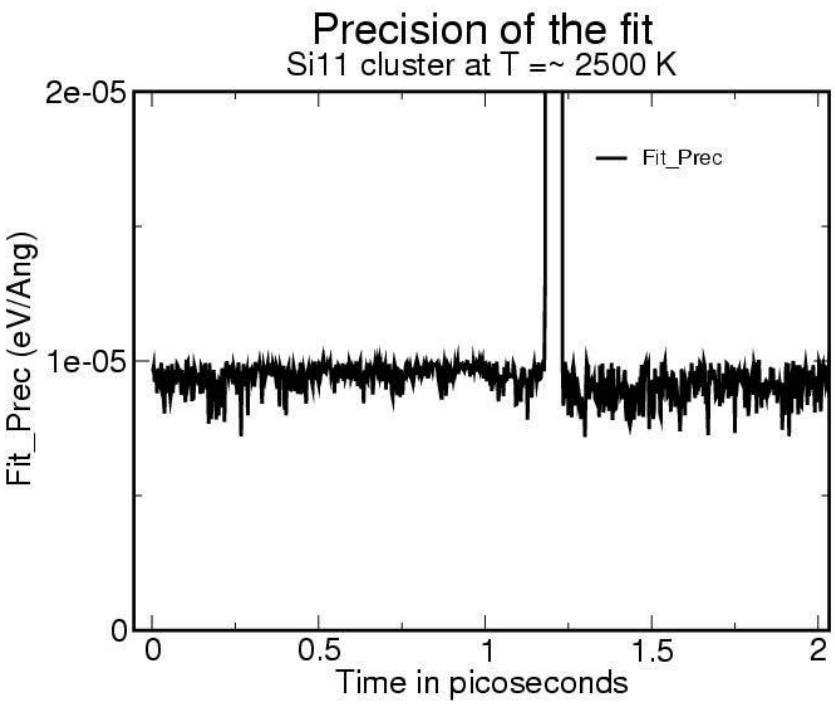
2) Ab-initio software (SIESTA)

Ai, Bi and Ci are local quantities

More variable parameters than forces to fit :

→ Minimum of $|F_q - F_{c\,adapt}|^2$ found through a damped dynamics in the parameters space

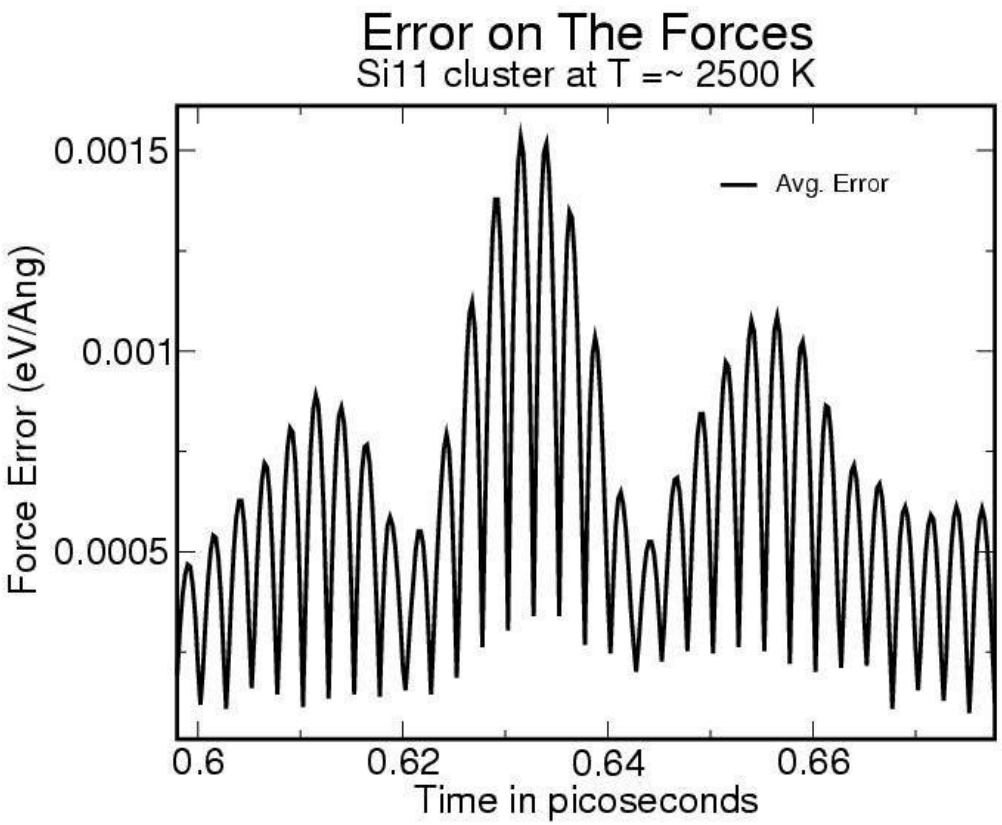




Accuracy of the Forces

T ~= 2500 K Force average : 1 eV/Ang

Biggest error : 0.12 ev/A for a force of 3 eV/A (5% of the force)



films !!!

Plans de Clivage dans le Silicium

V_{crack} = 2000-3500 m/s

•Surfaces clivées
" planes "

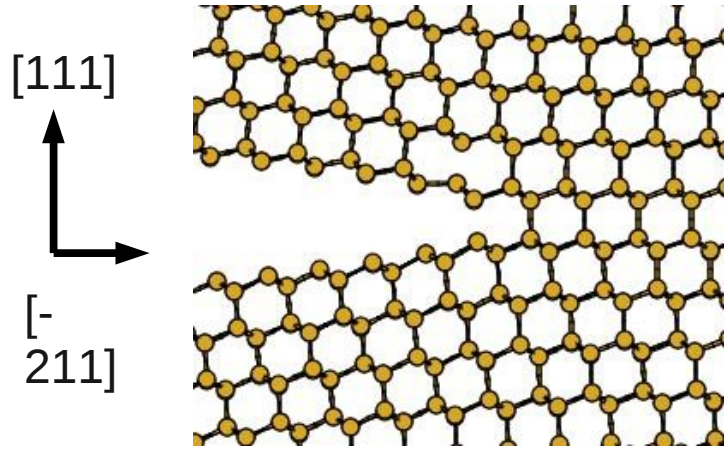
•Reconstruites (2x1)

•Marches "[2-1-1]"

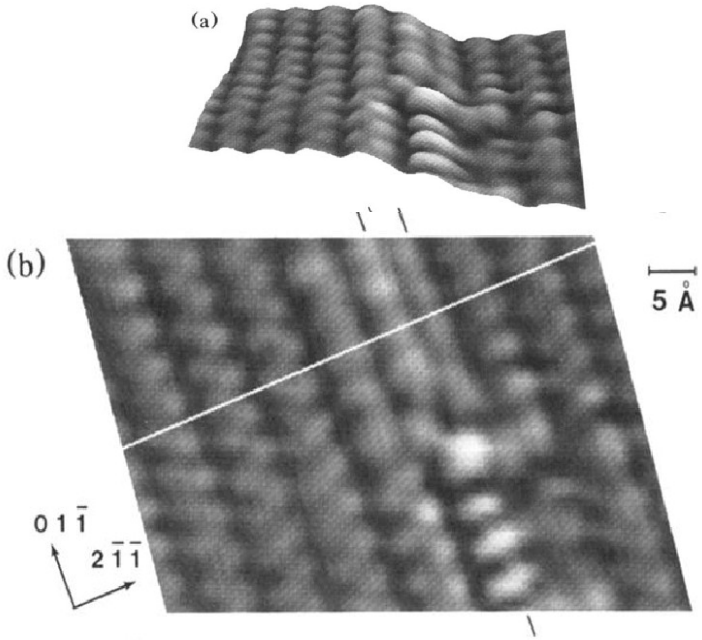
(Jusqu'à G<16 J/m2)

V_{crack} < 800 m/s : instabilité

(111) et (110)

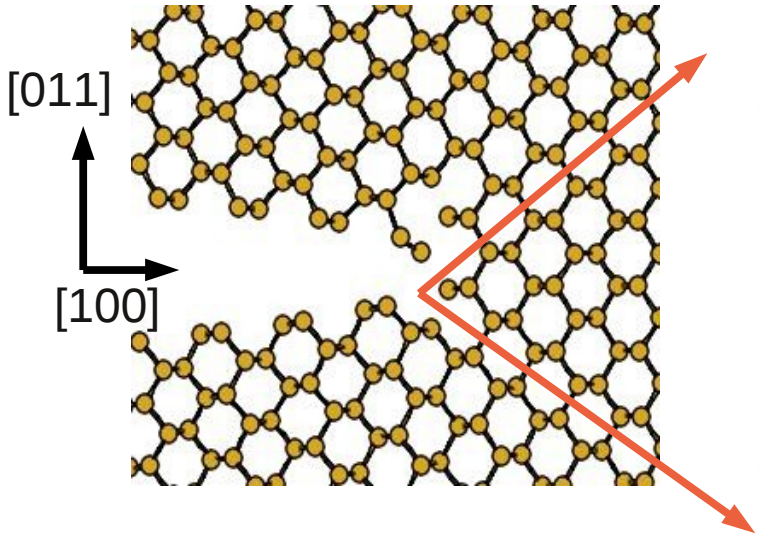


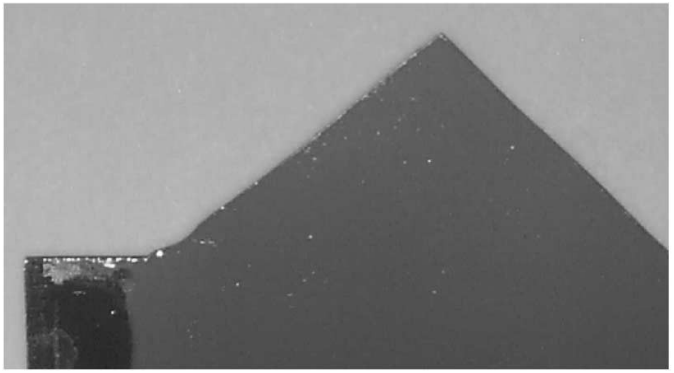
STM (Feenstra et al.)



•Propagation selon [100] à très faible V_{crack}

•Déviation sur des plans (111)



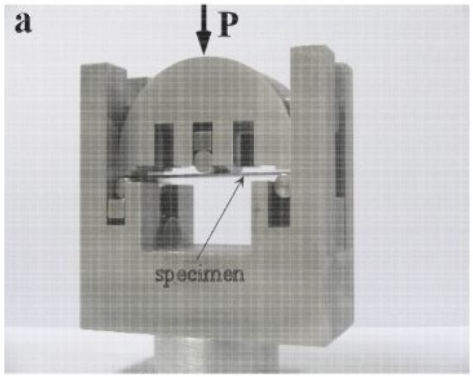


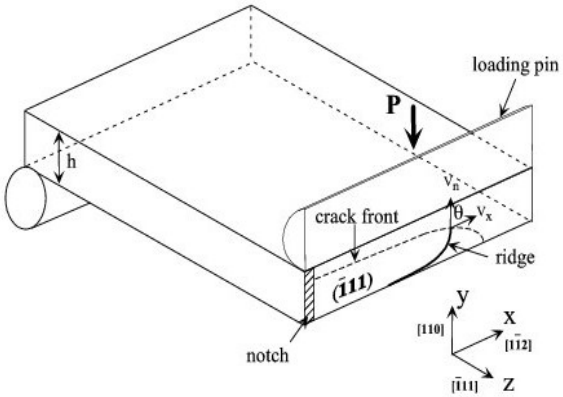
(Gumbsch et al. 2000)

•Système (111)[-211] stable sous cisaillement

Instabilité à faible vitesse de propagation
Orientation (111)[-211]

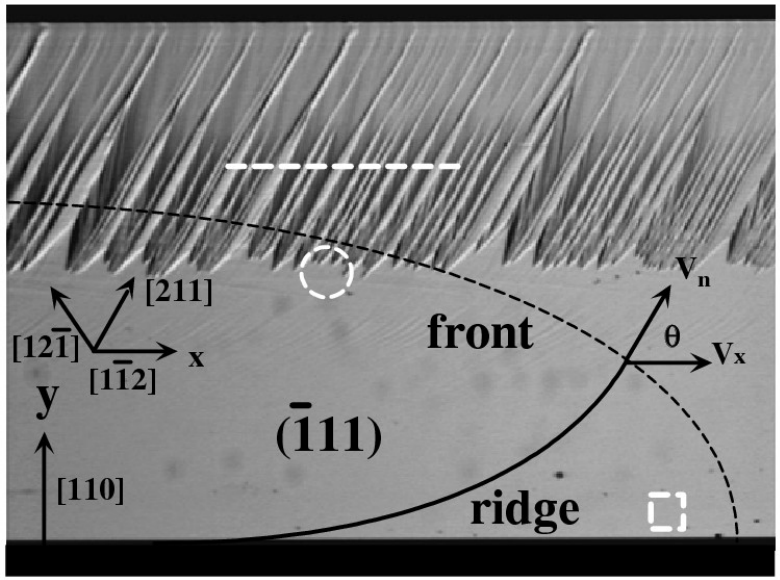
(Sherman 2000-2008)

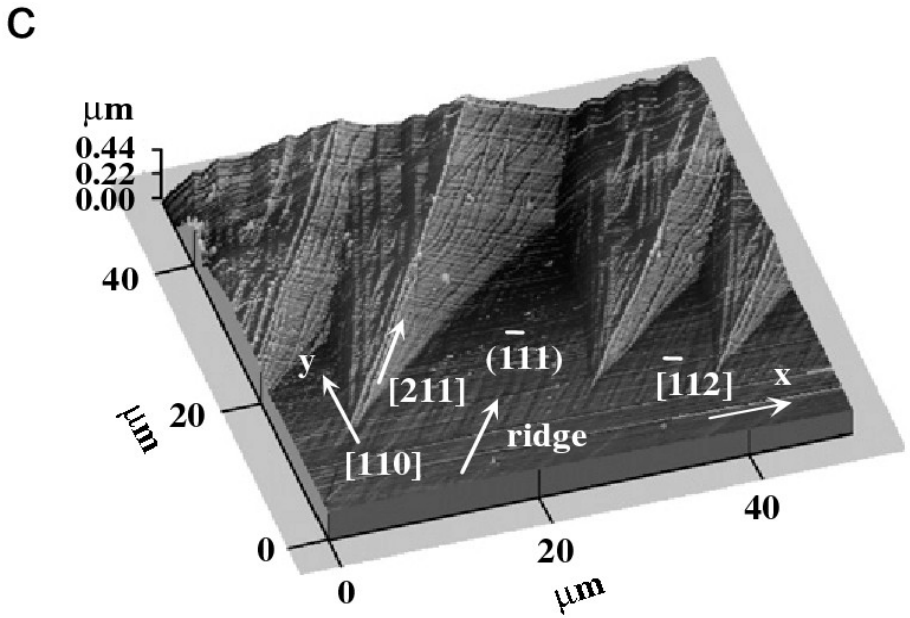




Formation d'arêtes :

Déviation systématique
du plan de fracture
à faible vitesse
(Vcrack < 800m/s)

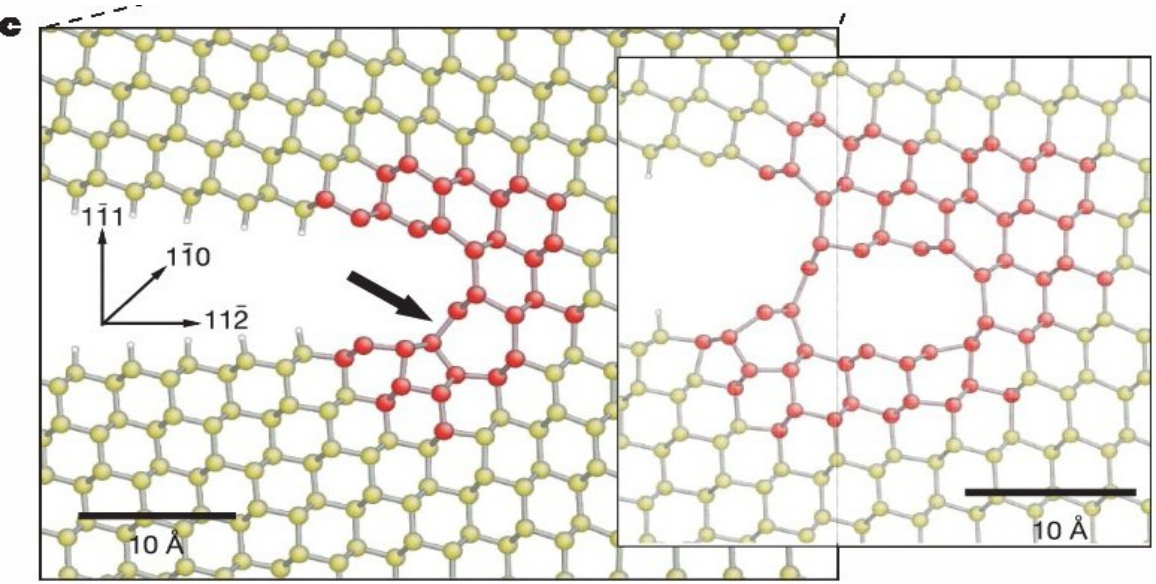




Instabilité à faible vitesse de propagation (111) [-211]

(Kermode 2008)

LOTF

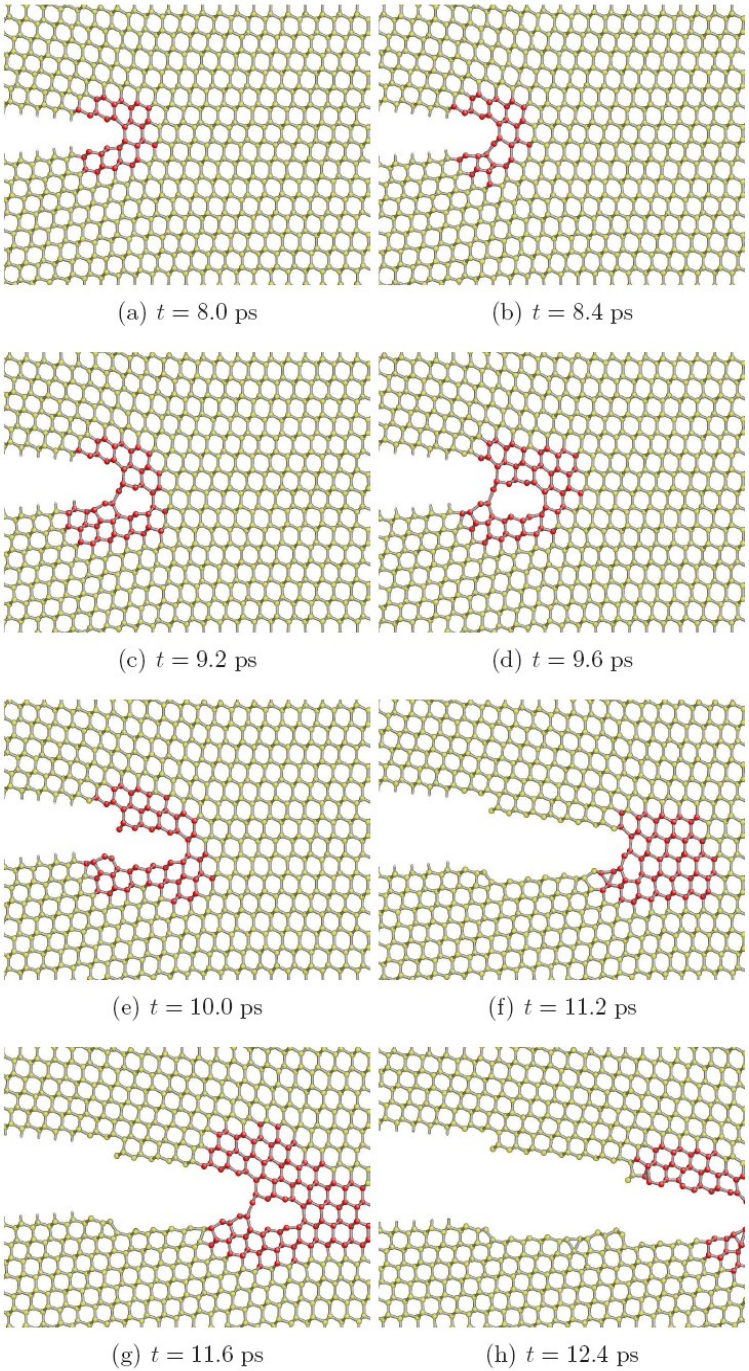


reconstruction de la pointe de fracture (anneaux 5/7)

→ Effet de piégeage par le réseau

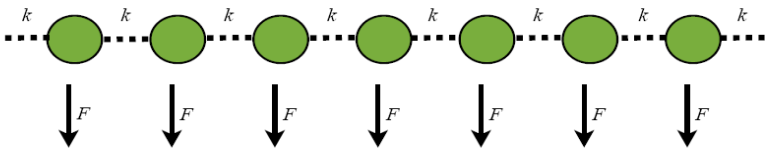
→ V_{crack} diminue

→ Changement systématique du plan de fracture



Instabilité à faible vitesse (111)[-211] : modèle mésoscopique

(Kermode 2008)

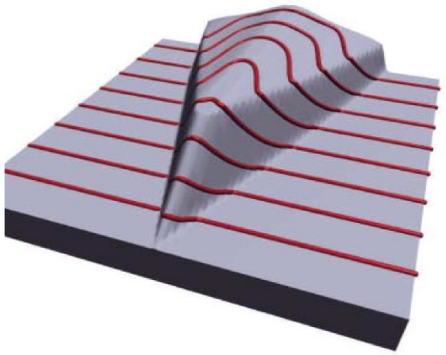


$$v_i(t+dt) = v_i(t) + dt/m\,\Big[\overbrace{f^0\,\hat{x} - b\,v_i(t)}^{\text{charge}} + \overbrace{k\,\big(r_{i+1}(t)+r_{i-1}(t)-2r_i(t)\big)}^{\text{tension de ligne}}\Big]$$

$$\underbrace{-\xi_i\,\tau/l\,\big(v_i^x(t)\big)^2\,\hat{x}}_{\text{reconstruction}} + \underbrace{\eta\,L/N\,\Sigma_j\,max(0, r_j - r_i)\,\hat{x}}_{\text{charge “accrue”}}$$

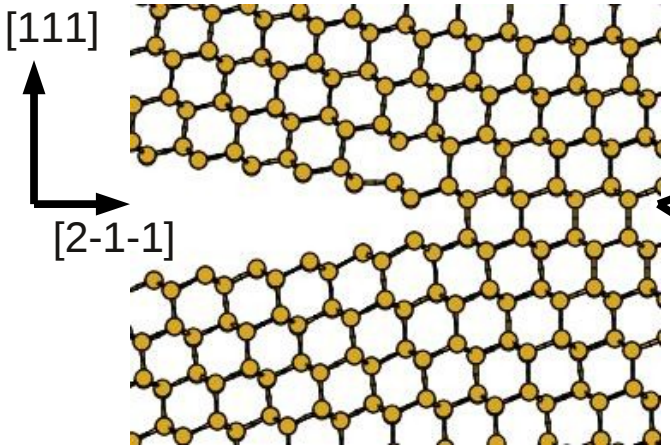
$$\xi_i = \frac{s}{1+\exp\big((v_i^x - v_0^x)/d\big)}$$

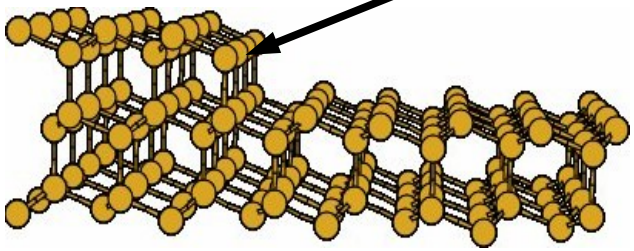
$$r_i(t+dt) = r_i(t) + dt\,v_i(t)$$



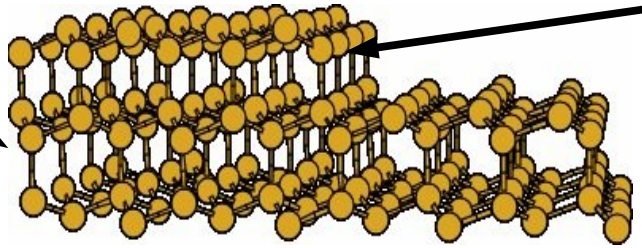
Correspond à l'instabilité observée à faible vitesse pour ce système

Marches sur les surfaces clivées (111)[-211]



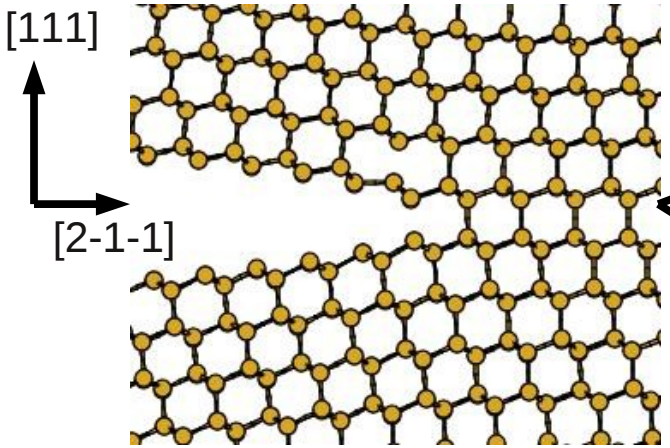


Marche descendant selon [2-1-1]

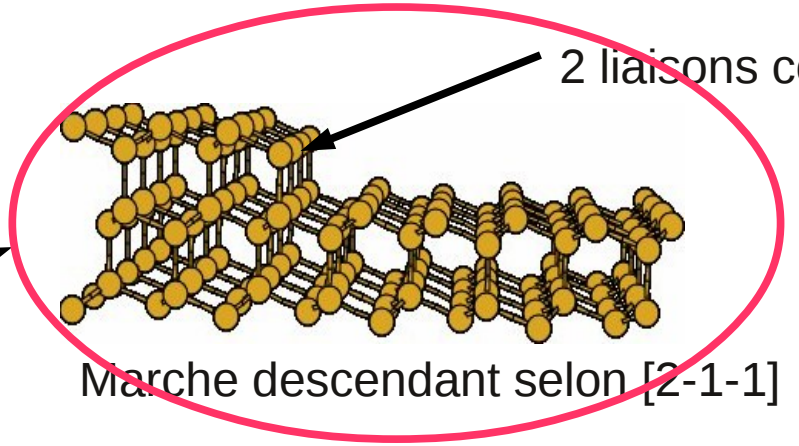


Marche descendant selon [-211]

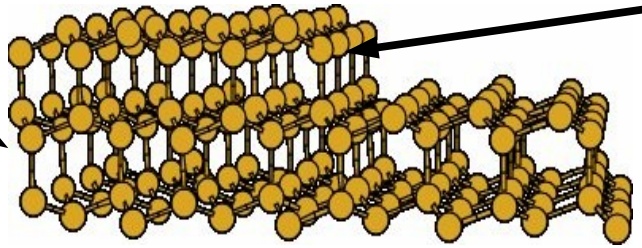
Marches sur les surfaces clivées (111)[-211]



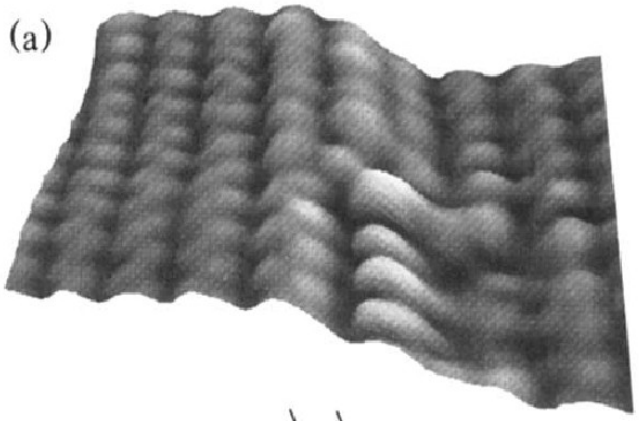
(STM Feenstra 1987)

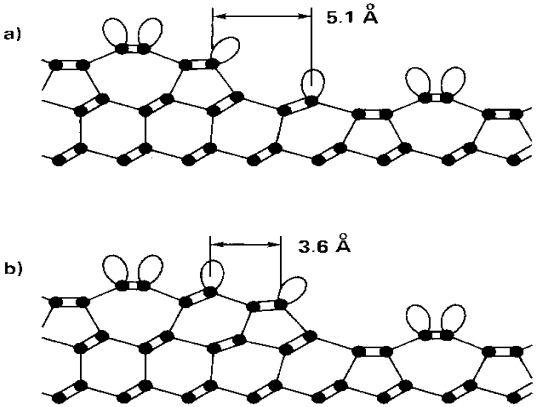


Marche descendant selon [2-1-1]

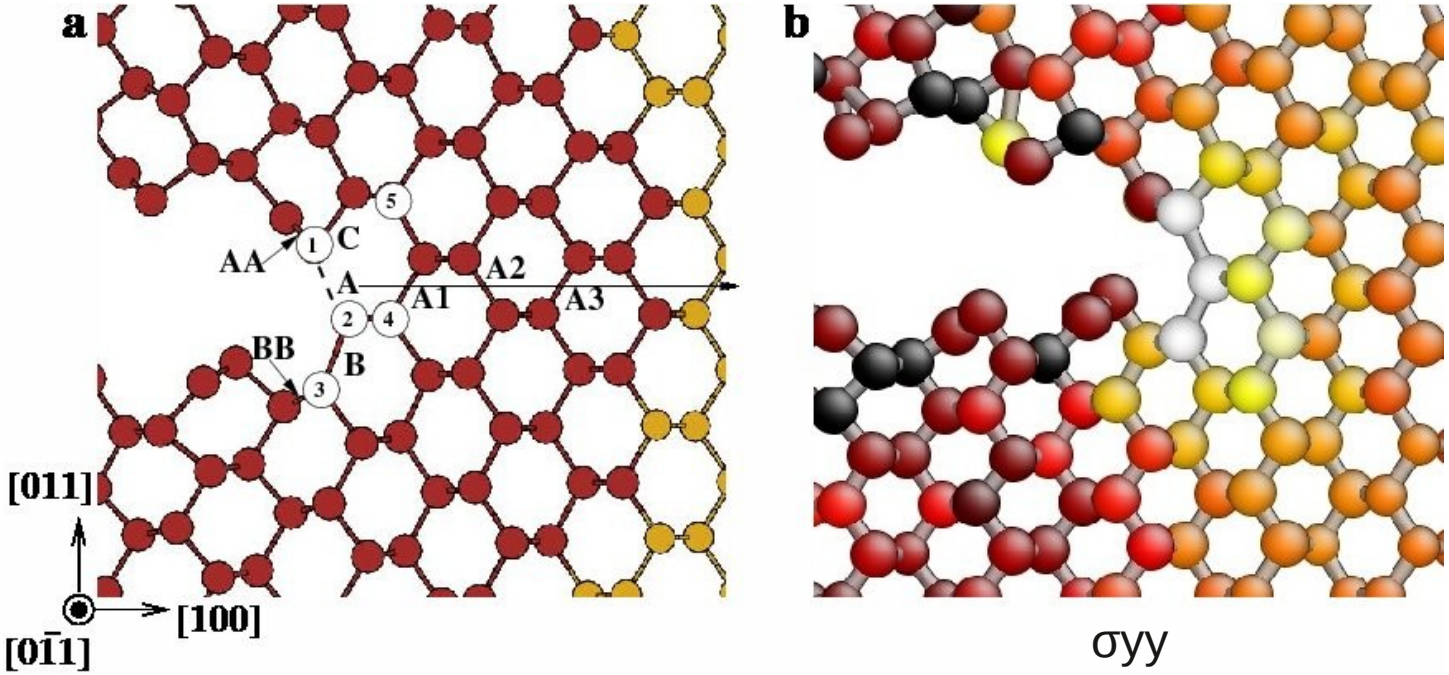


Marche descendant selon [-211]





Si(011)[100] : Mode I



<u>Compétition entre les liens A et B en pointe de fracture</u> :

Lien <u>A est plus faible</u> à cause de son environnement chimique :

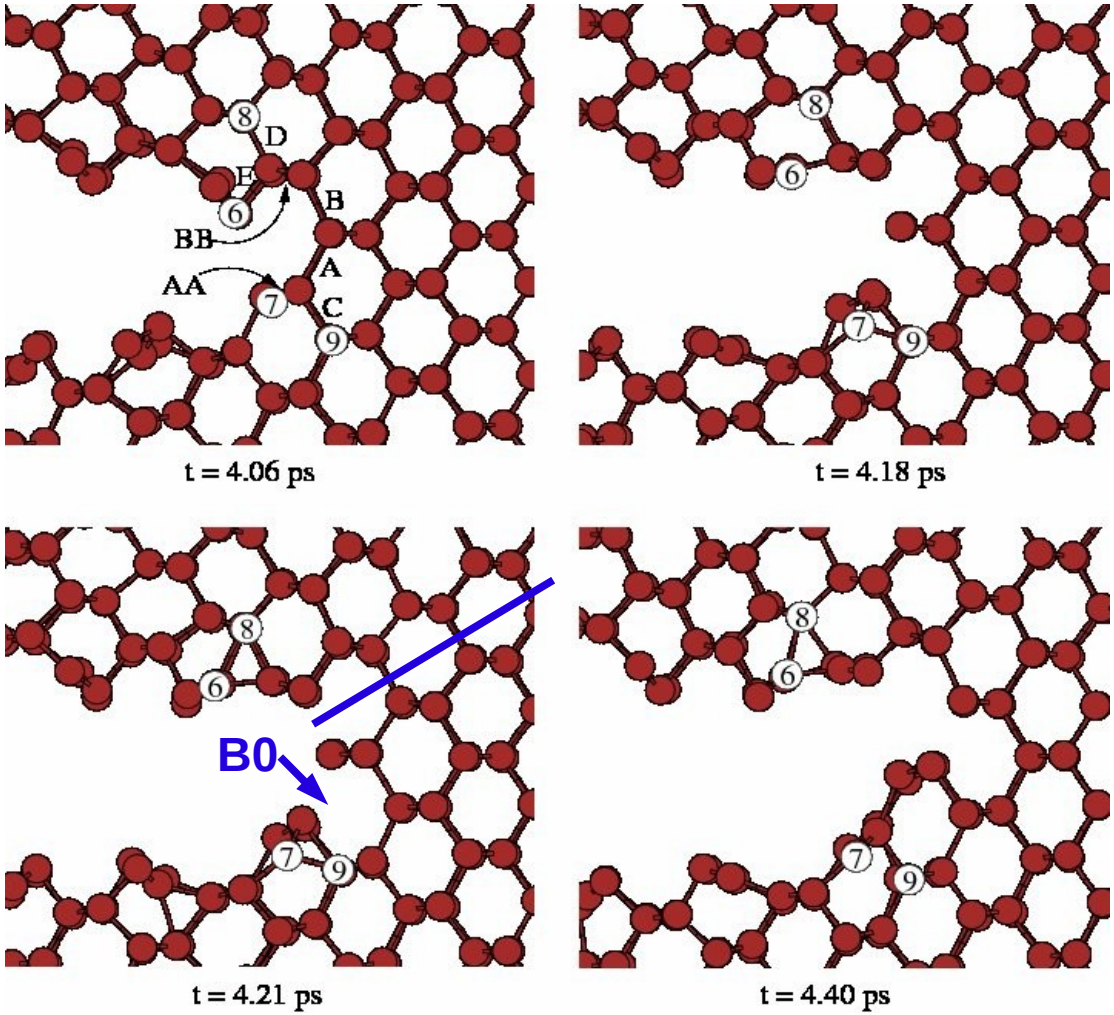
les liaisons avec les atomes sous coordinés sont plus fortes (distances, Mulliken)

•confirmé par calcul ab-initio, Gumbsch-Pérez 2000

→ Effets cinétiques favorisent la rupture succéssive de liens de type A

→ Propagation initiale selon [100]

Si(011)[100] : Mode I



Vitesse plus élevée : (2000ms-1) relaxations atomiques permettent des re-hybridations entre surface et sous-surface qui renversent la stabilité des liens A et B

→ Création de marches en traction pure

→ Mécanisme similaire aux mécanismes de reconstruction (111)

Modifications to study Au

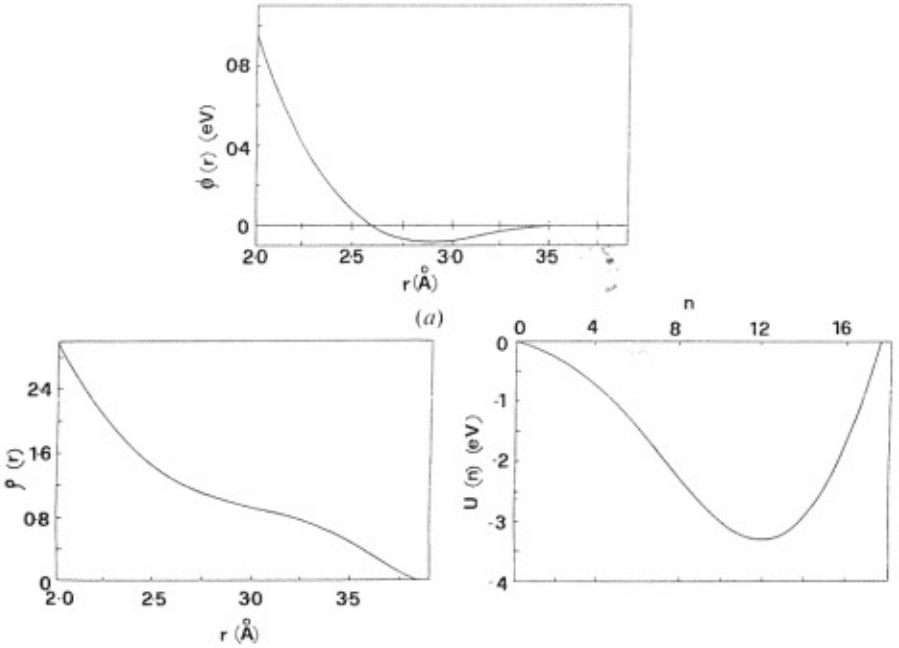
Glue EAM potential by Ercolessi et al. (1988):

$$V=\sum_{i>j}\phi(r_{ij})+\sum_{i}U(n_i)$$

$$n_i=\sum_{j\neq i}\rho(r_{ij})$$

$$\rho(r_{ij})\approx atomic\ density$$

$$n_i\approx coordination$$



We add two parameters to do the fit:

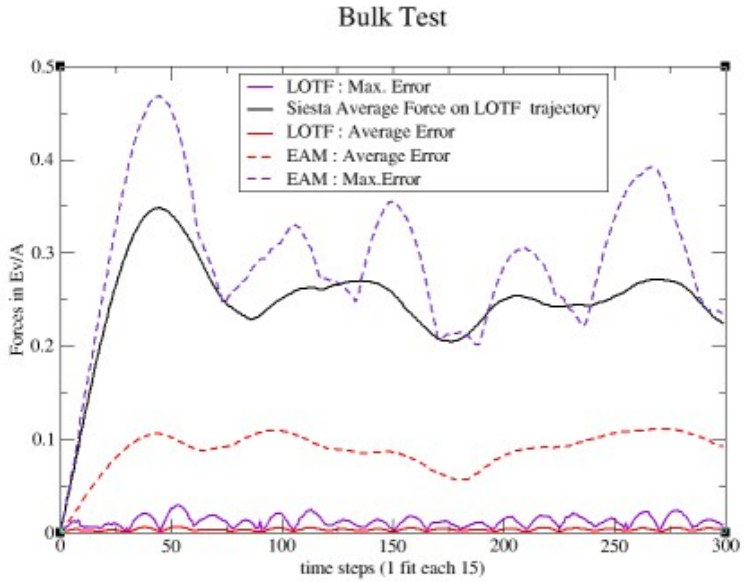
$$\phi(r_{ij})\Rightarrow A_{ij}\phi_{B_{ij}}(r_{ij})$$

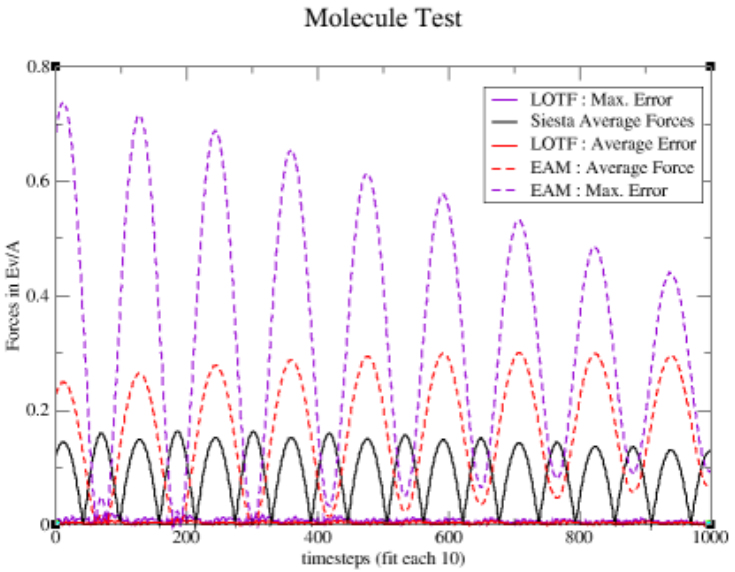
$$A_{ij}=scale\ factor$$

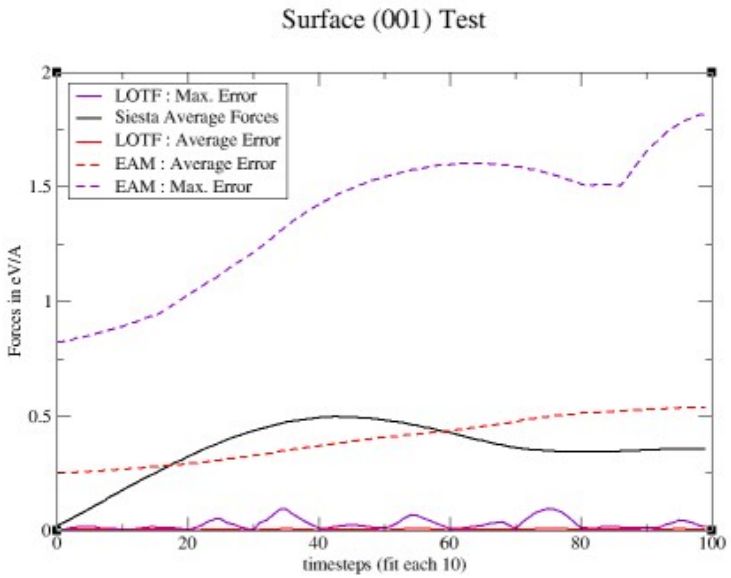
$$B_{ij}=position\ of\ the\ minimum$$

Fitting tests

And... we fit!



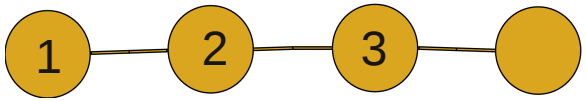




Cluster structures

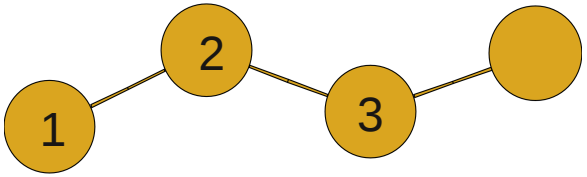
Relaxation of Au_4 clusters with LOTF, SIESTA, and the glue potential

L4	Glue	LOTF	SIESTA
d_{12}(Å)	2.68	2.53	2.52
d_{23}(Å)	2.61	2.56	2.55



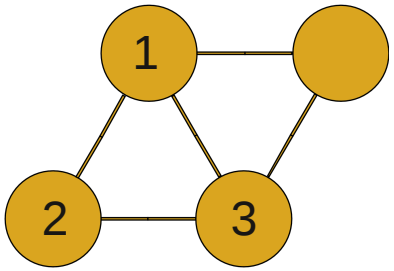
L4

Z4	Glue	LOTF	SIESTA
d_{12}(Å)	2.68	2.51	2.51
d_{23}(Å)	2.62	2.54	2.54
α(°)	127.0	133.0	127.0



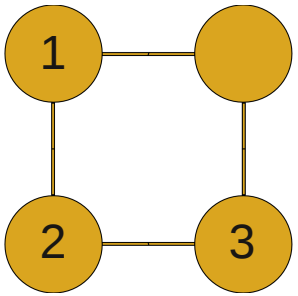
Z4

P4-1	Glue	LOTF	SIESTA
d_{12}(Å)	-	2.65	2.65
d_{23}(Å)	-	2.64	2.64
α(°)	-	59.7	60.0



P4-1

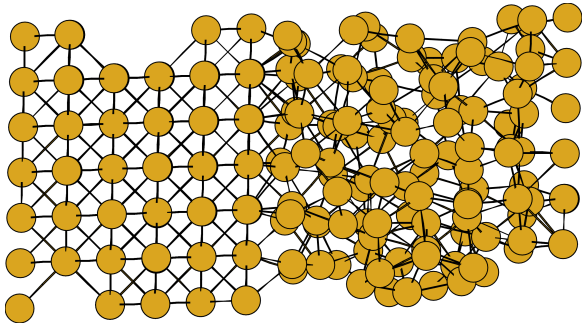
P4-2	Glue	LOTF	SIESTA
d_{12}(Å)	1.85	2.60	2.60
d_{23}(Å)	1.73	2.60	2.60
α(°)	90.0	90.0	90.0



P4-2

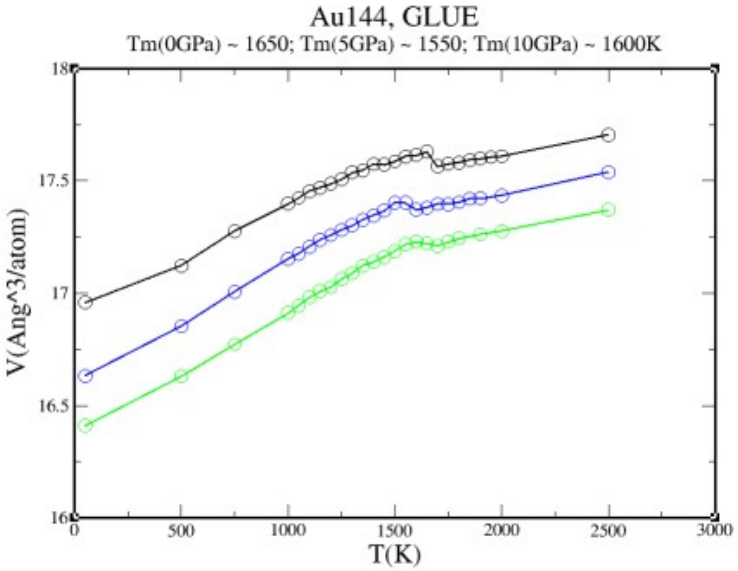
Melting temperature (glue potential)

Melting simulations using the 2-phase method (Belonoshko et al, 1994)

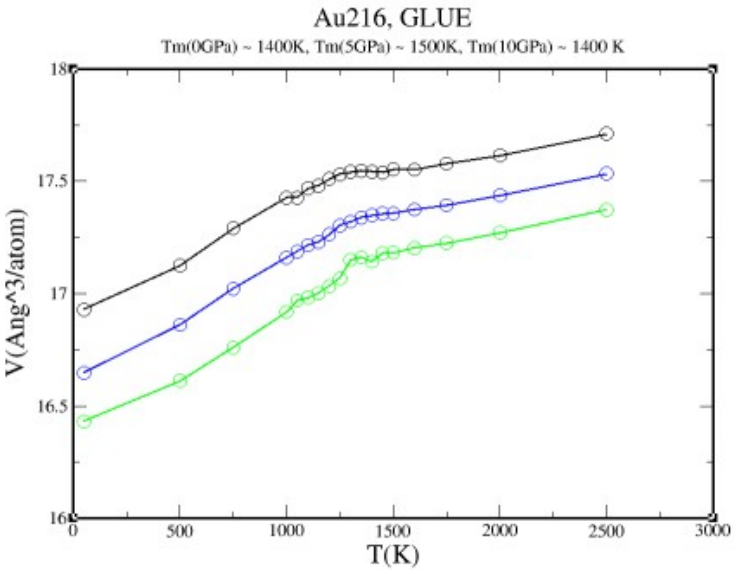


To avoid overheating

Molecular dynamics methodology: thermostat (rescaling T) and barostat (with damping term)

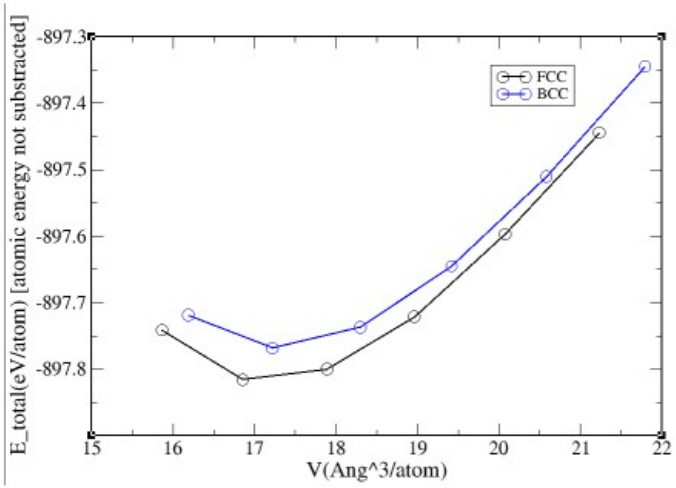


(1) Size dependent results (~1000 atoms is enough)



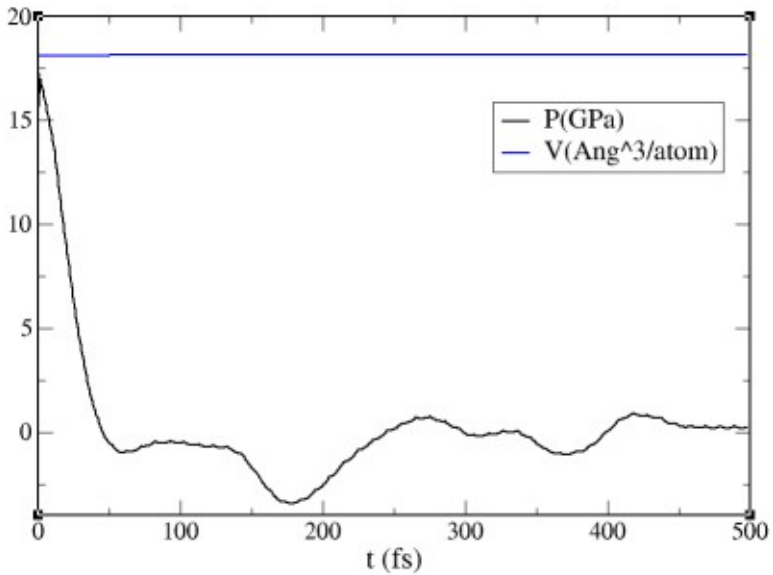
(2) The volume of the liquid is badly represented with the glue!

Volume of the liquid



256 atoms is enough to have the right FCC – BCC behavior at T = 0 K

Simulation of the liquid at 1773 K with 256 atoms



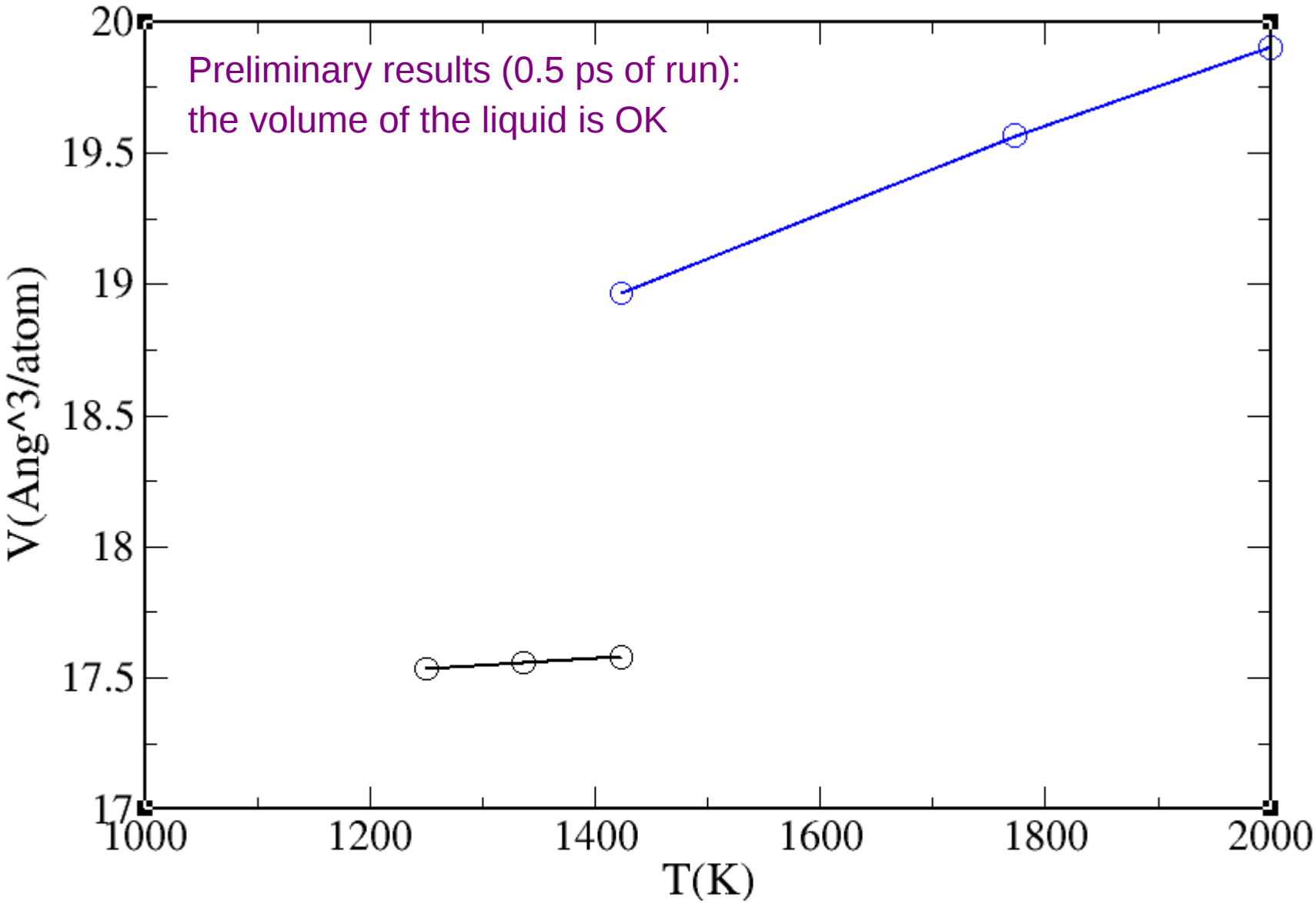
The volume of the liquid is still too small!

Why??

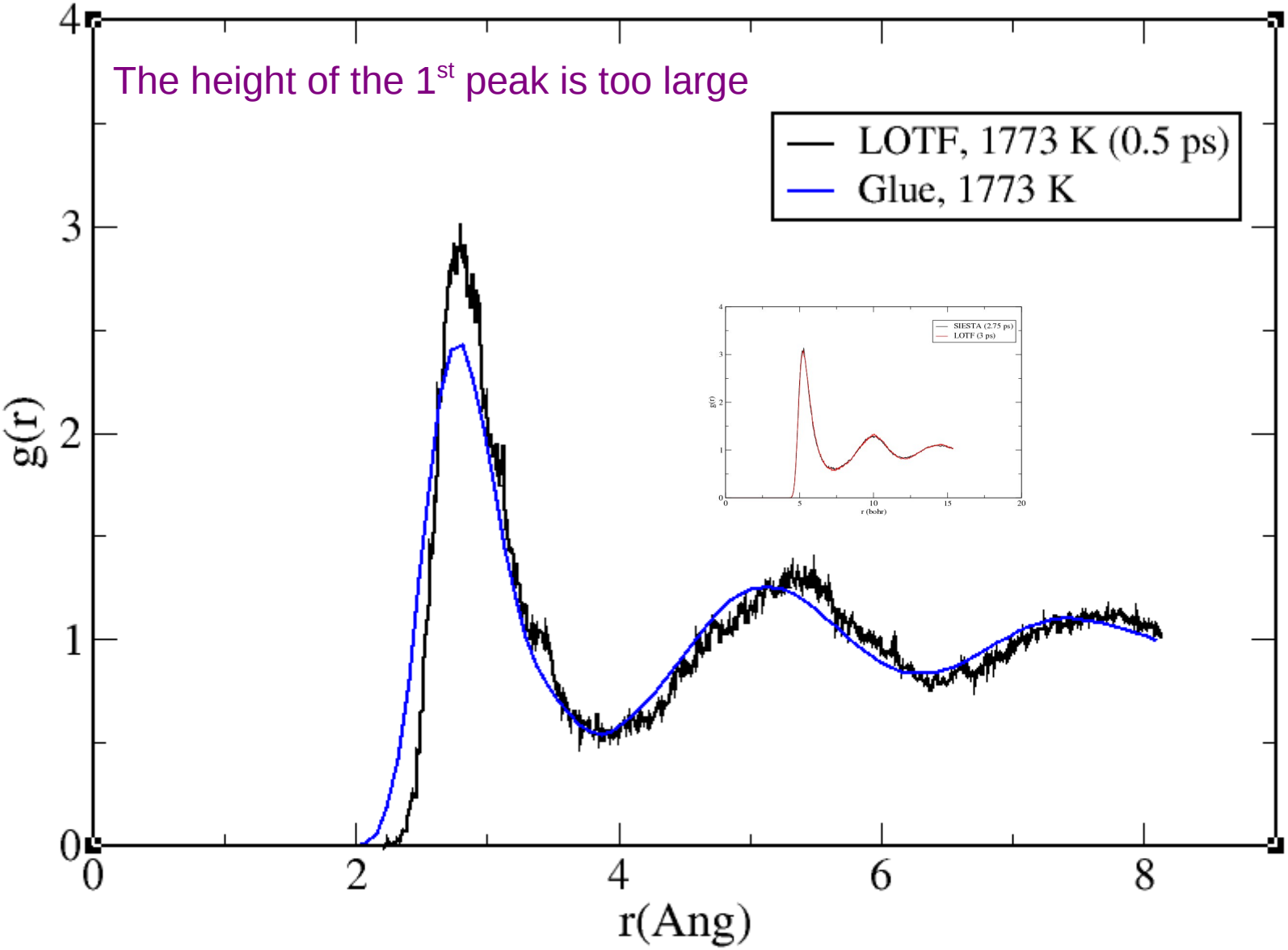
We are fitting only the QM forces, and the pressure out of the resulting parameterization is too small (inherited from the glue behavior?)

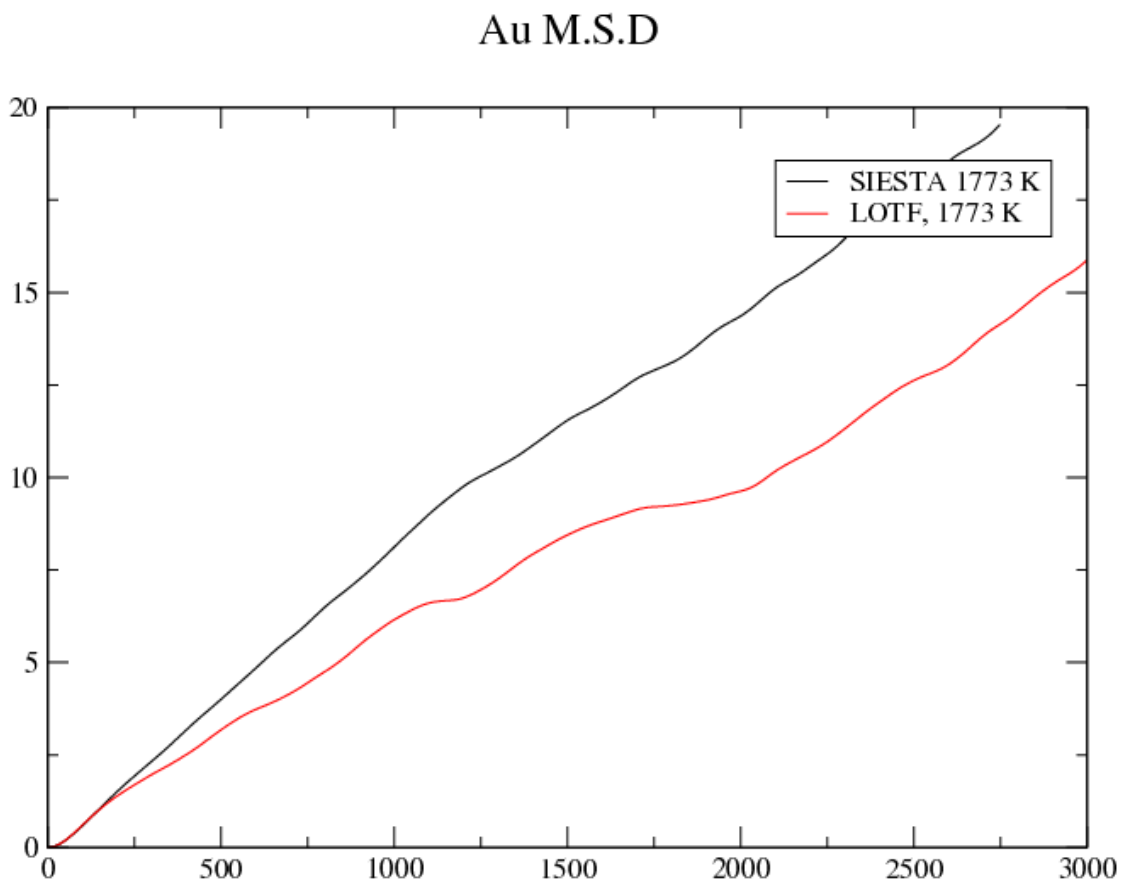
Fitting stress

What if we fit the stress together with the forces?



Preliminary results for liquid Au




Au M.S.D
SIESTA 1773 K
LOTF, 1773 K
0
5
10
15
20
0
500
1000
1500
2000
2500
3000

Conclusions

- Méthode QM/MM LOTF appliquée à la fracture dans Si :

➢ Intègre champ élastique à longue porté et donne une précision suffisante en pointe de fracture : *Fracture fragile*

➢ Instabilités à faible vitesse du système de fracture (111)[-211], formation préférentielle de marches de type [2-1-1]

➢ Instabilité du système (011)[100]

➢ Lien entre les *reconstructions de surface, contraintes* à la pointe et *instabilités*

– LOTF : adapté aux problèmes de *structure* intrinsèquement multi-échelles

➢ Frontières dynamiques, QM/MM en *temps* et *espace*

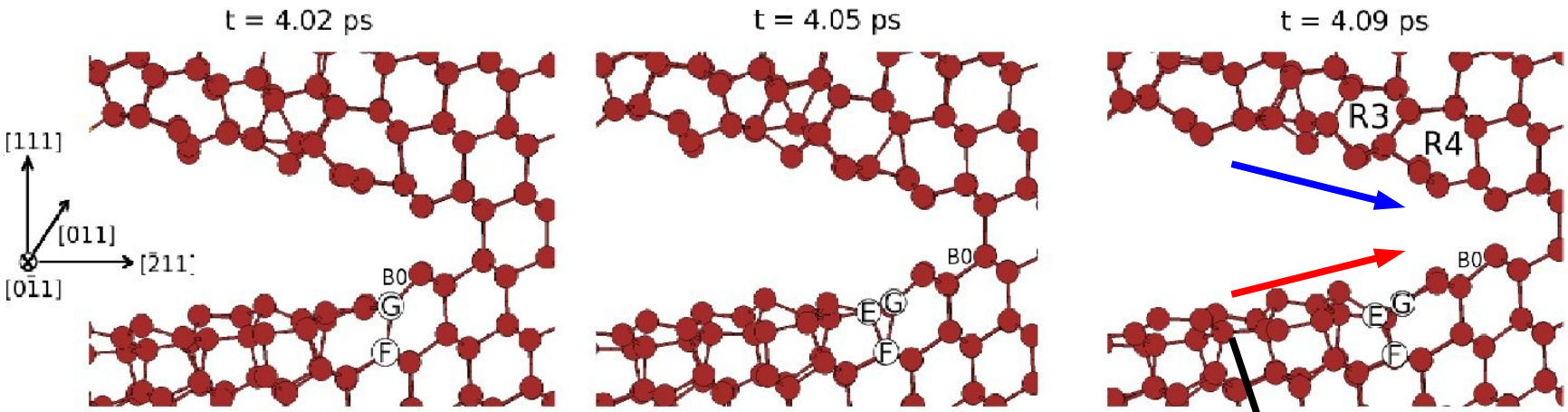
- Développements / Perspectives :

➢ Autres éléments (Au, Si-H smart-cut Moras, De Vita, fracture SiO2 ...)

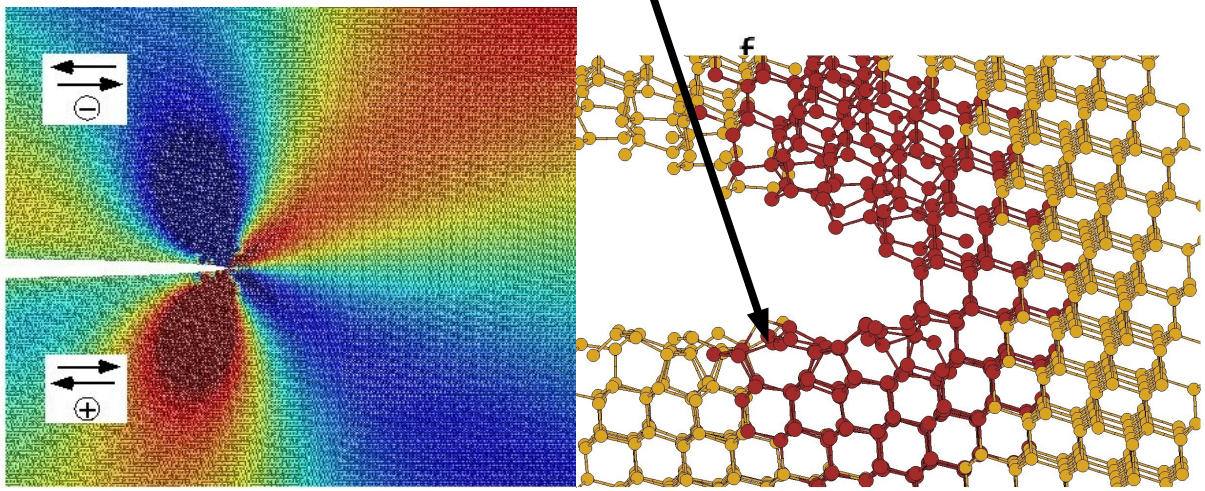
➢ Systèmes à l'équilibre (technique d'interpolation)

LOTF / Si(111)[-211]

2000 ms-1 < V_{crack} < 3000 ms-1



d

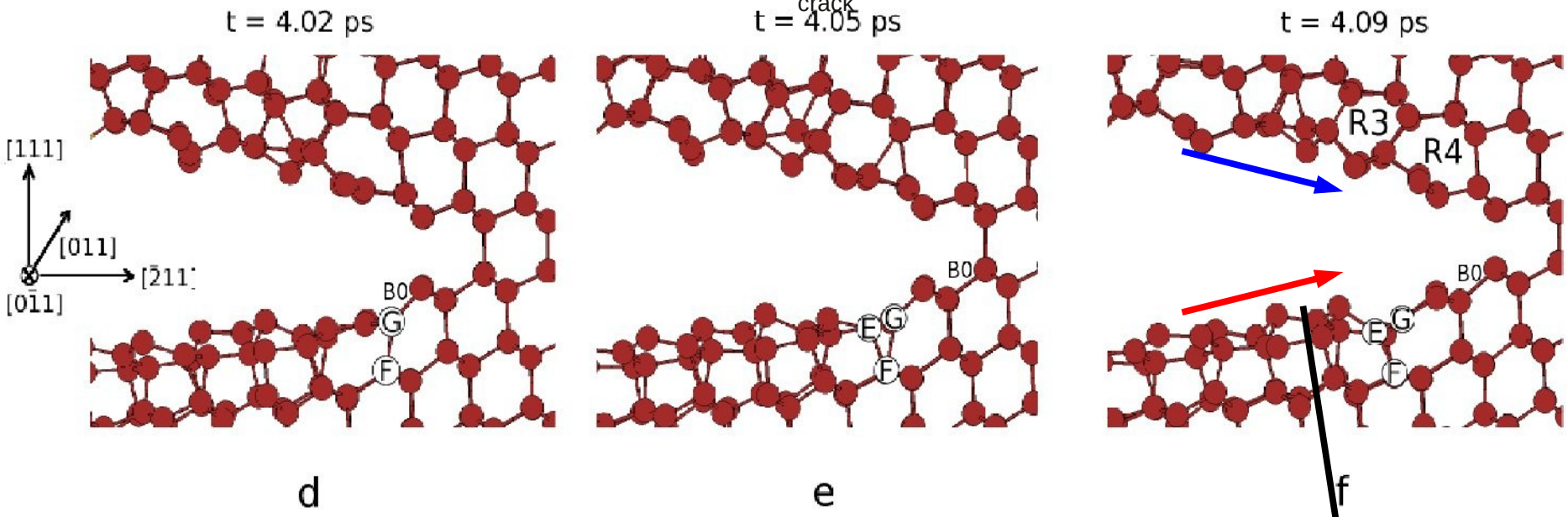


Reconstructions (2x1) de Pandey avec des chaines d'atomes sous-coordinés

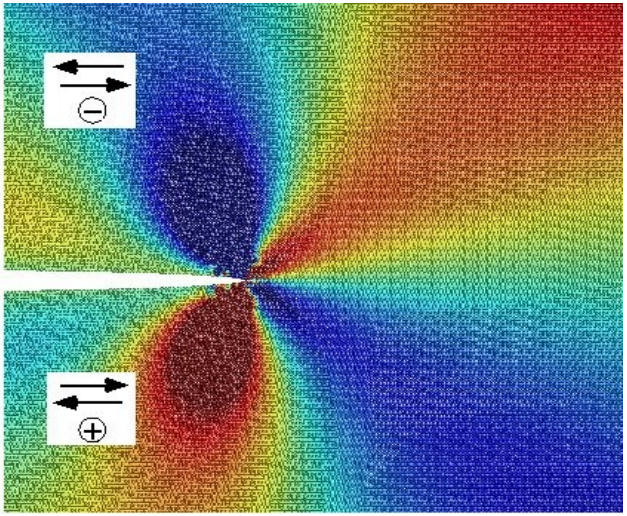
- Cisaillement positif : pas de marche, propagation [2-1-1]
- Direction de propagation [-211] ”protégée” par la reconstruction de Pandey

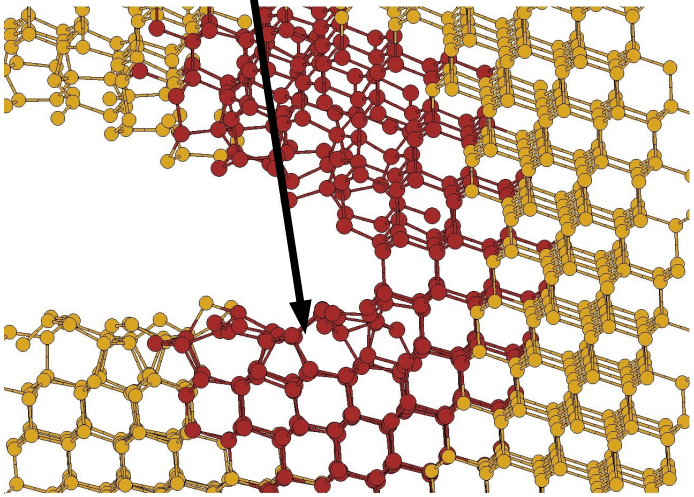
LOTF / Si(111)[-211]

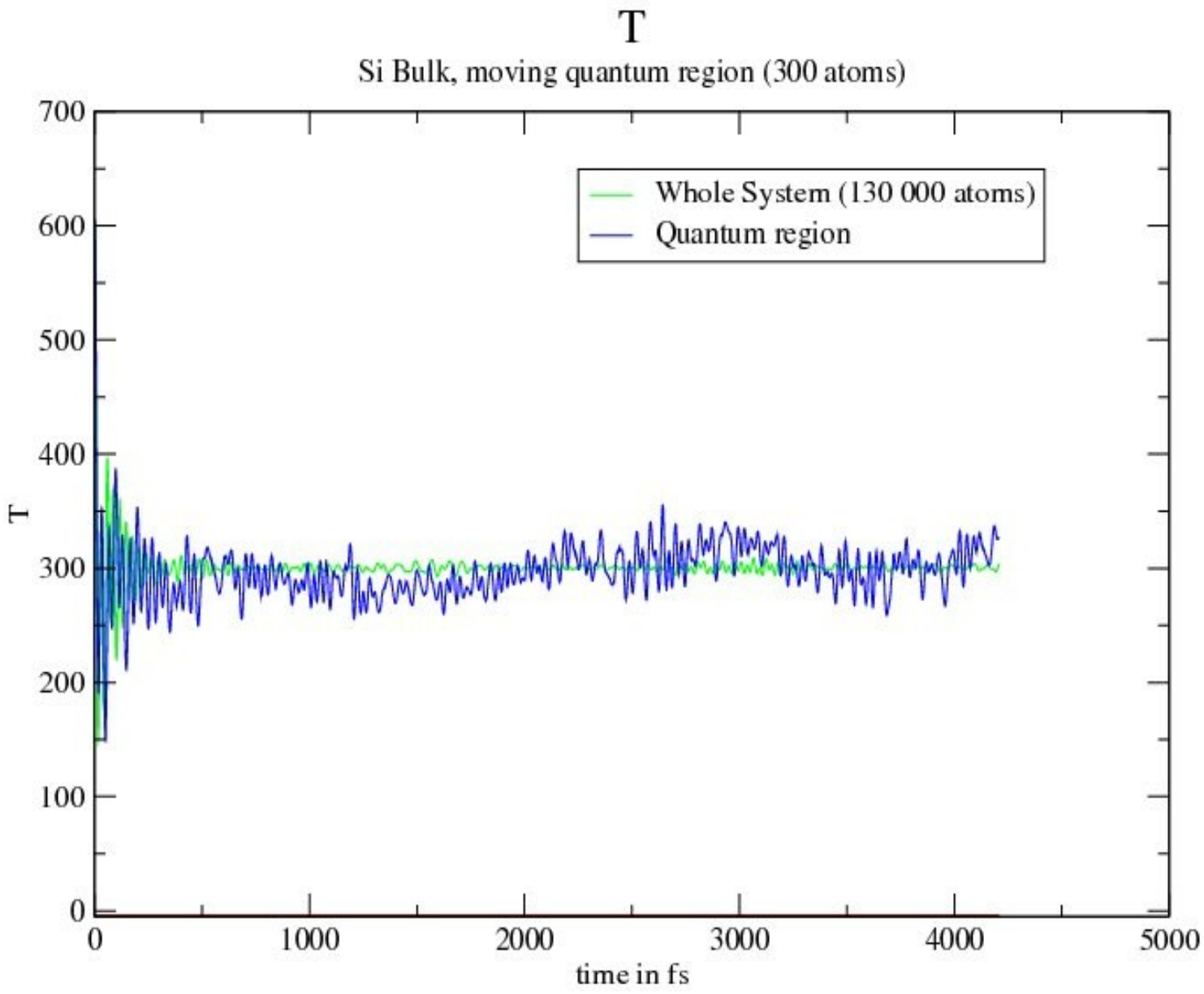
2000 ms-1 < V_{crack} < 3000 ms-1



Reconstructions (2x1) de Pandey avec des chaines d'atomes sous-coordinés



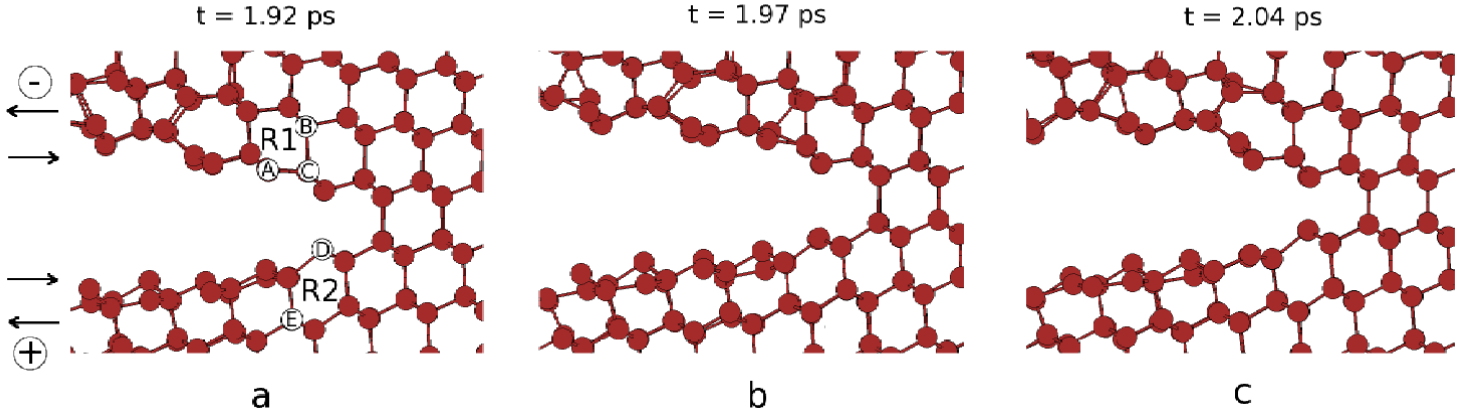


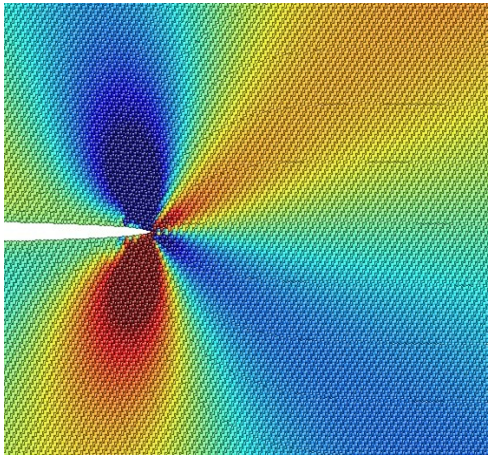

T
Si Bulk, moving quantum region (300 atoms)
Whole System (130 000 atoms)
Quantum region
T
time in fs
0
100
200
300
400
500
600
700
0
1000
2000
3000
4000
5000

Si(111)[-211]

Propagation forcée à plus grande vitesse en utilisant le potentiel classique , G=7.4 J/m2

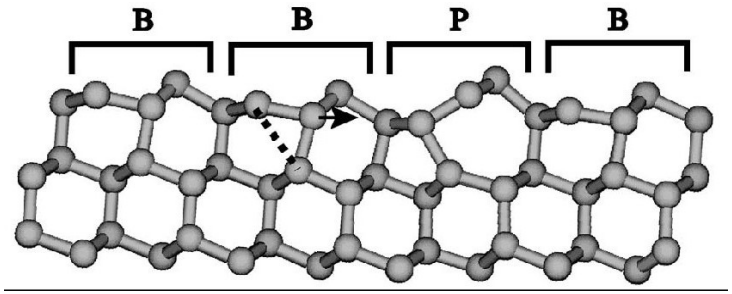
1000 ms-1 < V_{crack} < 2000 ms-1





σxy

2 types de reconstruction de surface :



<u>Surface supérieure (-1-1-1)</u> :

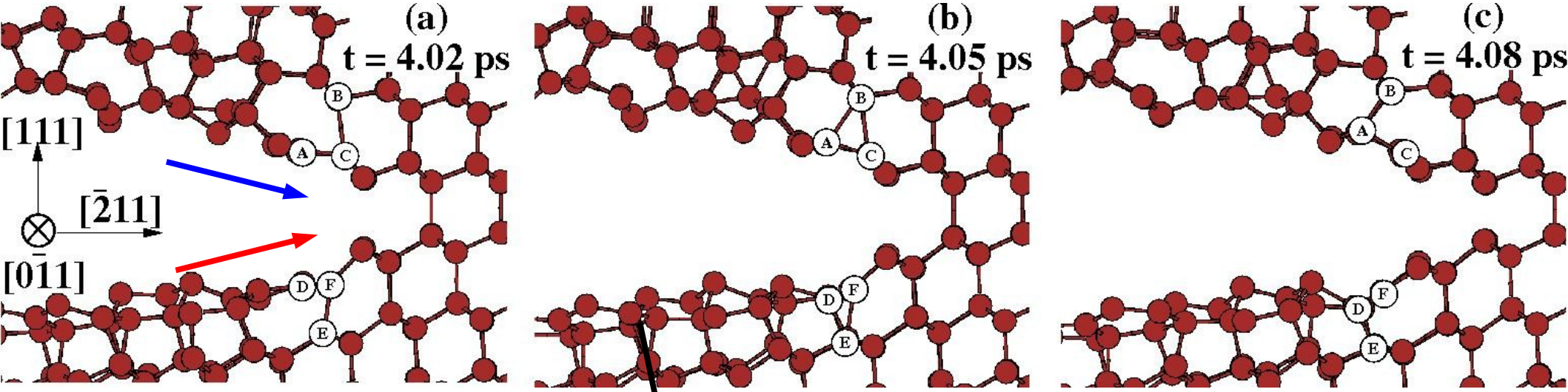
(2x1) Pandey avec des chaines d'atomes sous coordinés parallèles au front

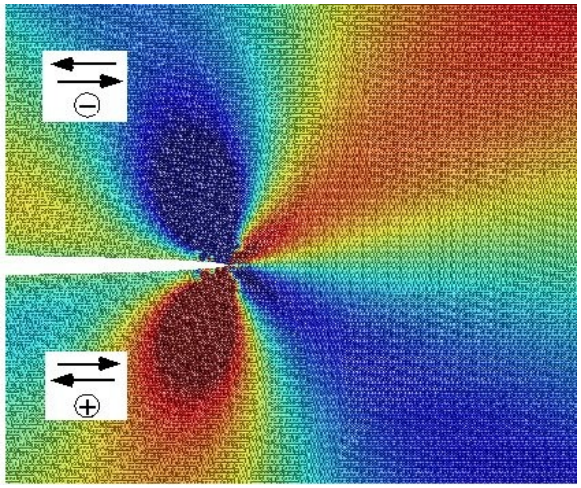
<u>Surface inférieure (111) :</u>

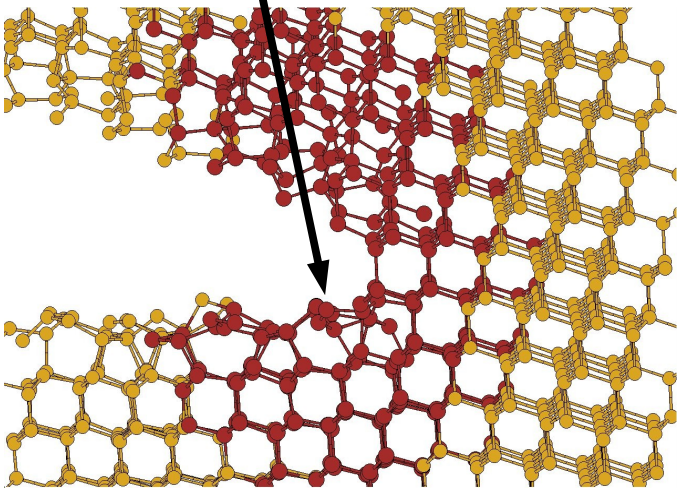
reconstructions de Haneman (1x2) ou (2x1)

LOTF / Si(111)[-211]

2000 ms-1 < V_{crack} < 3000 ms-1







•Reconstructions (2x1) de Pandey formant des chaines d'atomes sous-coordinés

•Structures cohérentes avec les contraintes en pointe de fissure

• Cisaillement positif : pas de marche, propagation [2-1-1]

•Direction de propagation [-211] ”protégée” par la reconstruction de Pandey