Ingénierie quantique pour la fonctionnalisation des matériaux

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Comprendre et optimiser la fonctionnalité des matériaux

Design de la solidification

Compréhension des premiers stades de la cristallisation et de l'amorphisation:

Identification d'un ordre local orientationnel dépendant de la directionnalité des liaisons chimiques et de la compacité pour le contôle des conditions de solidification.

Concept de structures locales favorisées en lien avec la composition chimique du système

Applications en cours:

propriétés dynamiques (viscosité, diffusivité)
(ANR Strudylis,)
Matériaux à structure complexe (amorphes, quasicristaux, HEA,..)
(GDR SAM (coll. Nancy))

Moyens :

Locaux/CIMENT/GENCI

Design de nanostructures

Aide à la fabrication et au design de ces objets en répondant aux besoins en

- caractérisation : détermination de la structure interne via les calculs

- Identification et contrôle des paramètres influants sur les propriétés recherchées.

- possibilité de tester et d'optimiser la réponse de la fonction visée en fonction des paramètres influants.

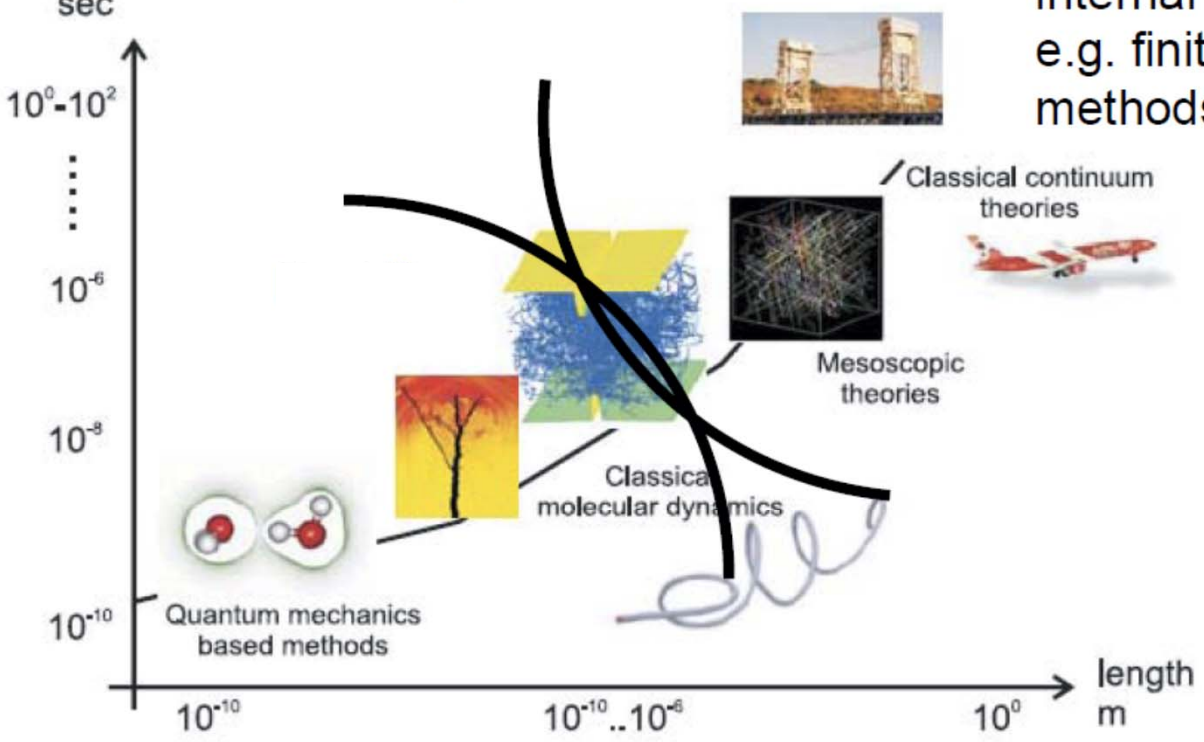
Applications en cours :

Nanoparticules pleines et creuses de Pt (1 ANR :SIMAP/LEPMI/CEA, labex CEMAM).
Matériaux inovants pour le stockage de l'énergie : batteries sodium (dépôt ANR int. SIMAP/CEA/ NanYang univ.) et batteries polymères (coll. LEPMI).

Multi scale view of materials

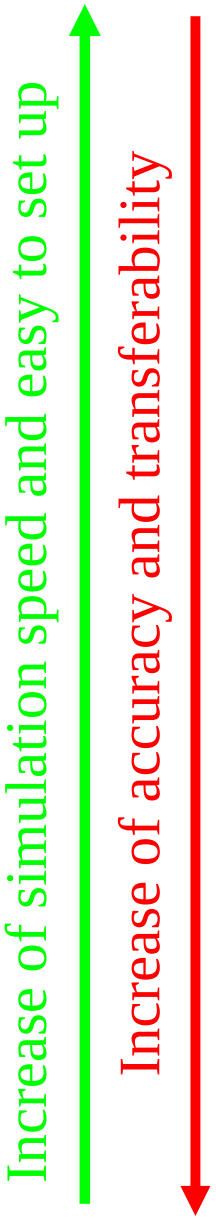
"molecular" (explicitly resolve molecules/atoms) Molecular Dynamics

"continuum" (matter infinitely divisible, no internal structure) e.g. finite element methods



"quantum" (explicitly resolve electrons); e.g. Density Functional Theory

Designing potentials



Put inside potentials as much physics as you need !

Empirical models: simple mathematical forms
parameters fitted on experimental data or ab initio calc.

- Pair potentials
- Multi-body potentials

Semi-empirical Models : functional forms taking more or less explicitly the electronic structure

- Tight Binding (TB)
- Embeded Atom Model (EAM, MEAM)
- etc…

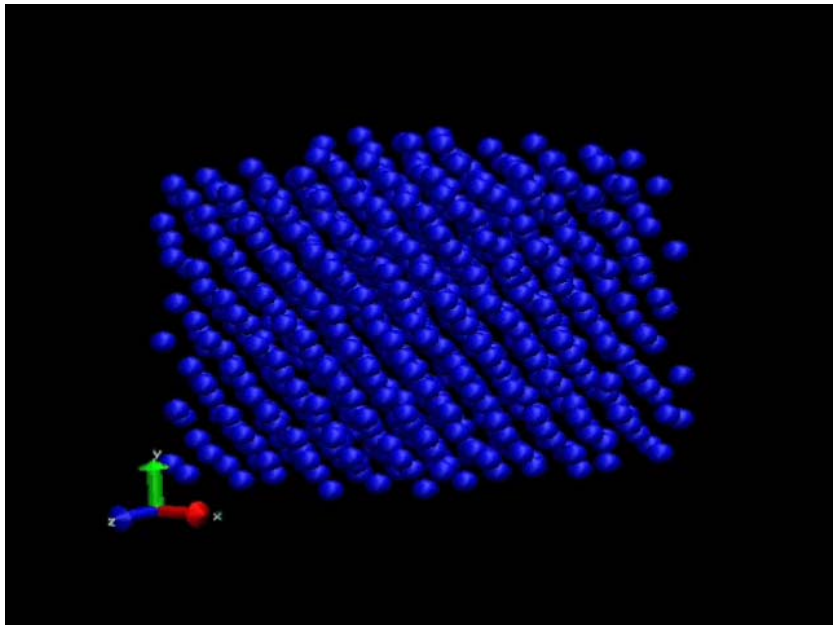
Density functional Theory : ab initio force calculations
The most powerfull approach !
BUT limited to small simulation cells and times

Molecular dynamics

- solving numerically Newton's equation of motion from the interaction between atoms

$$m\frac{d^2\vec{r}}{dt^2} = \vec{F}(\vec{r}) = -\vec{\nabla}V(\vec{r})$$

Time evolution of each atom $\begin{cases} \vec{r}(t) \\ \vec{v}(t) \end{cases}$



- In principle each atom interact with each other

→ It's a many body problem !!

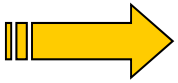
- Following the time evolution of the material
→ determination of the properties as a function of temperature and pressure

Computational strategies

- Atomistic simulations : ab initio calculations and classical Molécular Dynamics (MD)
- Three simulation techniques are illustrated here

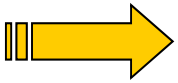
Full ab initio molecular dynamics mainly with VASP code :

1. MD simulation with ab initio forces determined at each time step from the instantaneous atomic configurations.

 Pure Elements, Boron, Au-Si, Al-Ni and AlCu Alloys

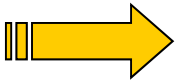
Hybrid simulation technique:

1. classical MD to bring the system in the desired states
2. ab initio MD from the generated configurations to refine the description

 Liquid-Liquid phase transition of Si

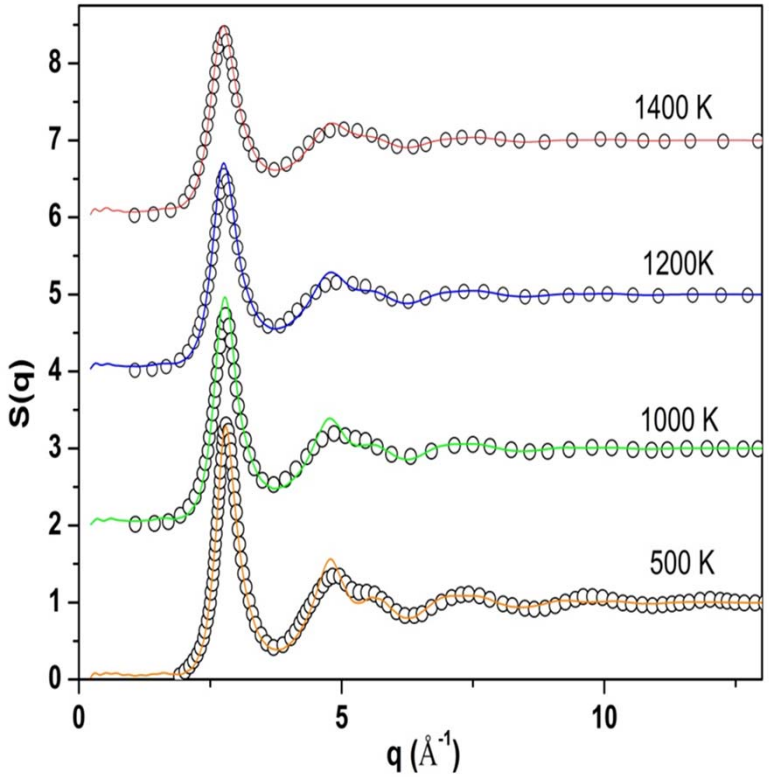
Force-Matching method :

1. ab initio MD on small cells for various states of the systems
   and extraction of the ab initio forces on each atoms (several conf.)
2. Fitting of a potential using
3. Classical MD simulation in states not accessible to the full ab initio MD

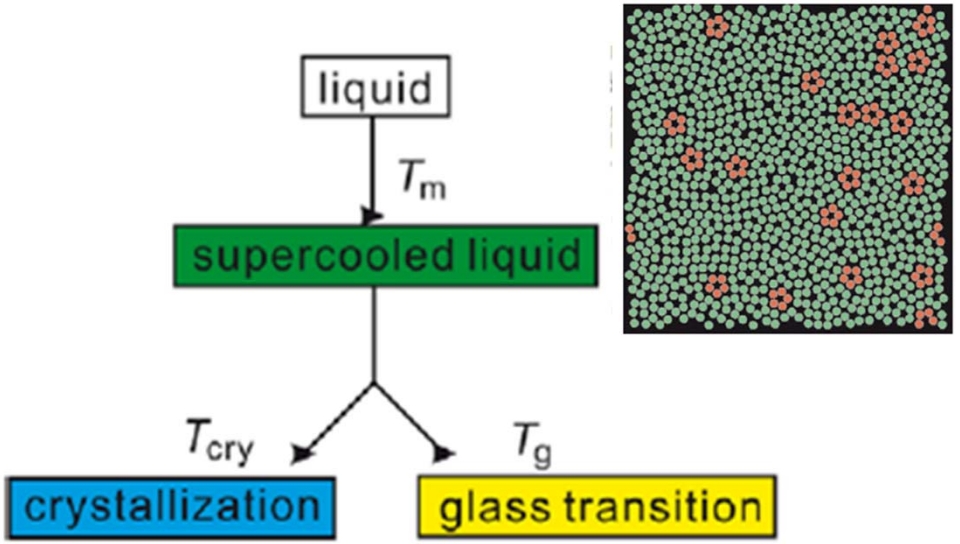
 Cu-Zr binary metallic glasses and Calcium Aluminosilicate glasses

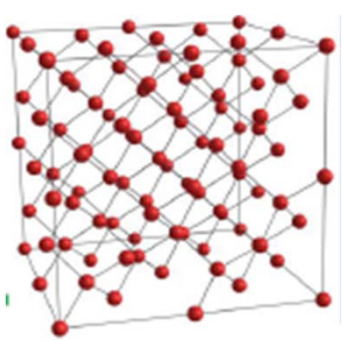
Design de la solidification : verres métallique massifs Cu-Zr

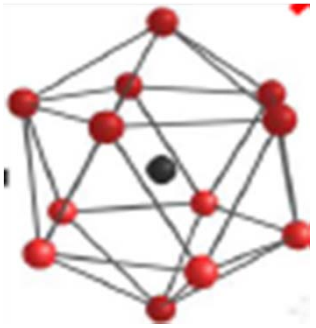
Alliage binaire $Cu_{64}Zr_{36}$



La dist. d'ordre local cristallin et icosaédrique





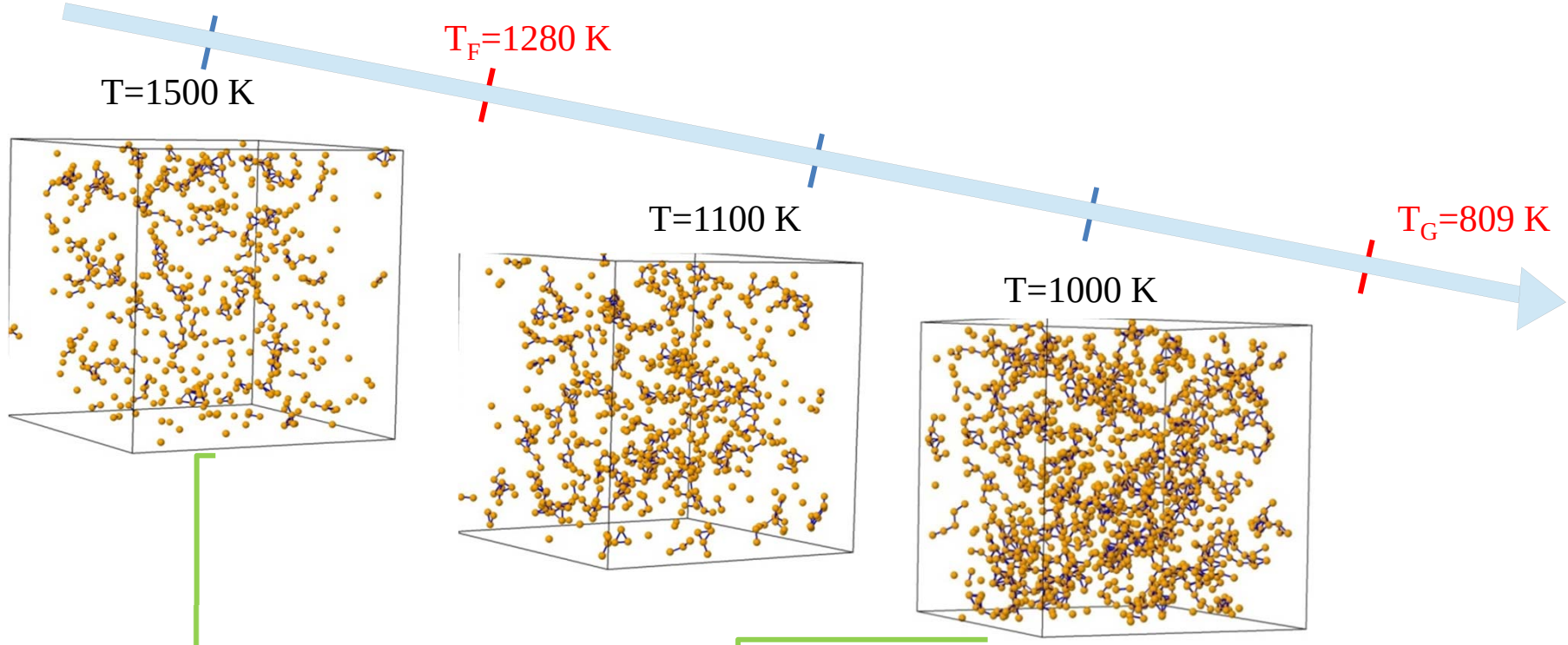


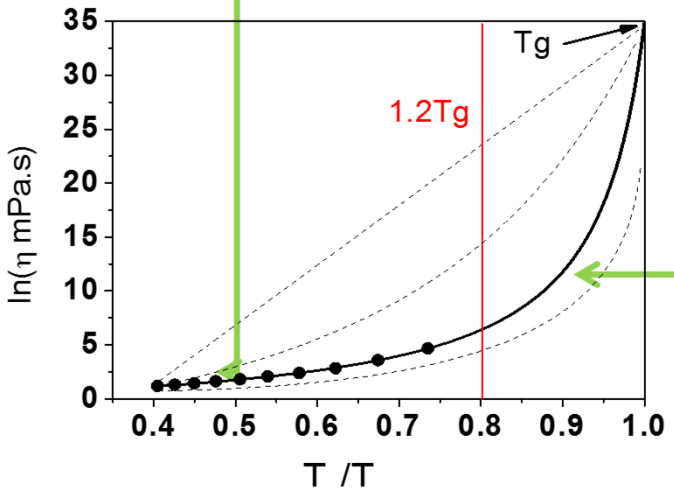
Le facteur de structure exp donne l'ordre local moyenné.

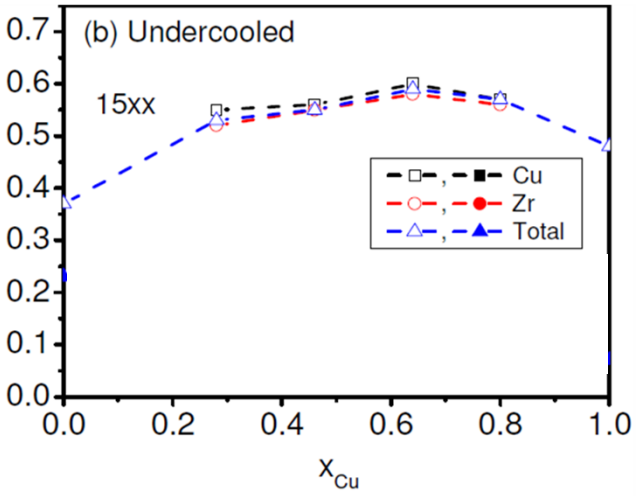
Les calculs fournissent un facteur comparable

Mais fournissent en plus →

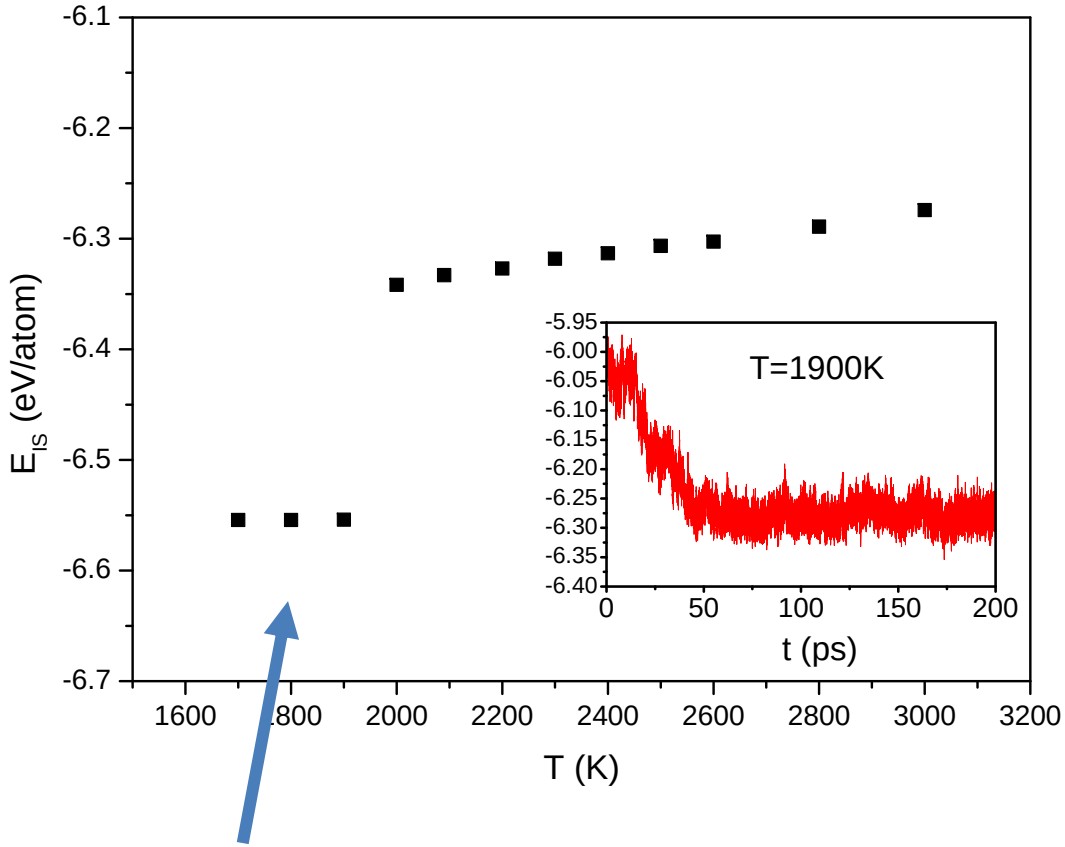
verres métallique massifs Cu-Zr

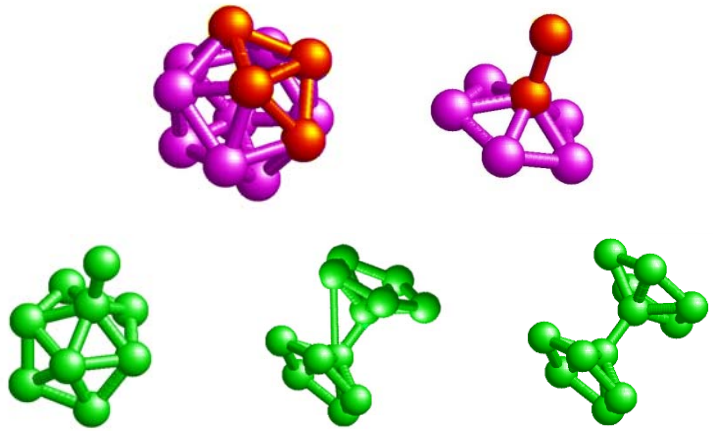


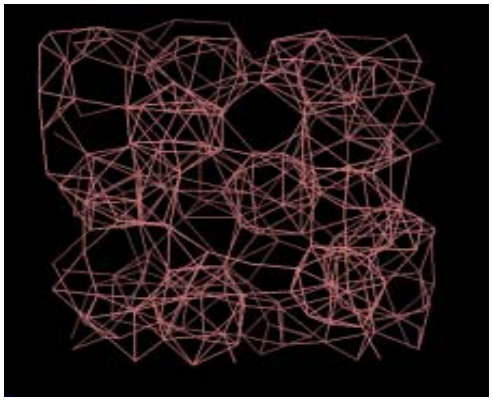


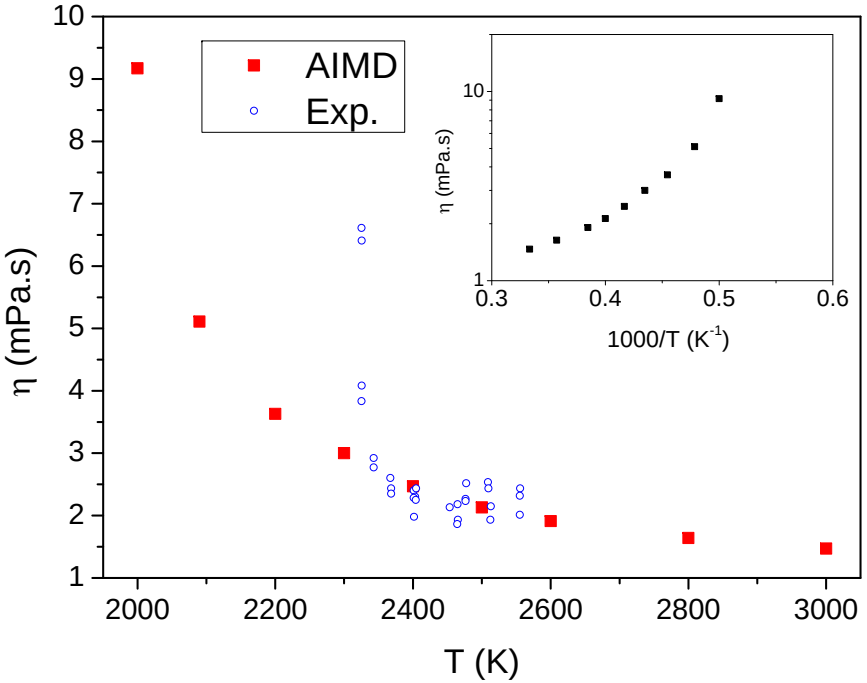


Crystallisation of boron









Liquid silicon: Liquid-liquid phase transition

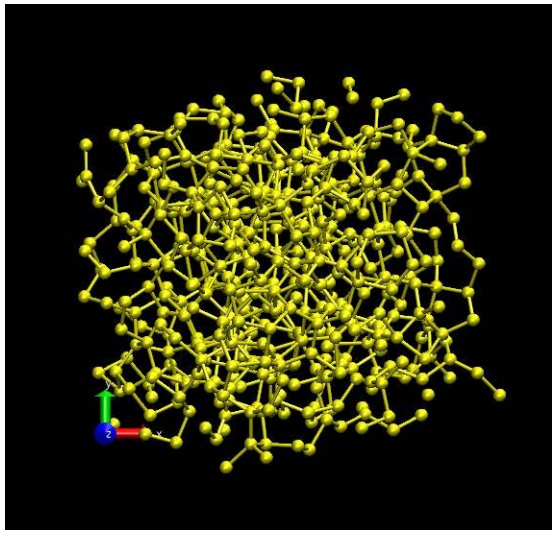
Molecular dynamics simulations (N=512)

- Empirical potential: Stillinger-Weber
  PRB, 31, 5262 (1985)

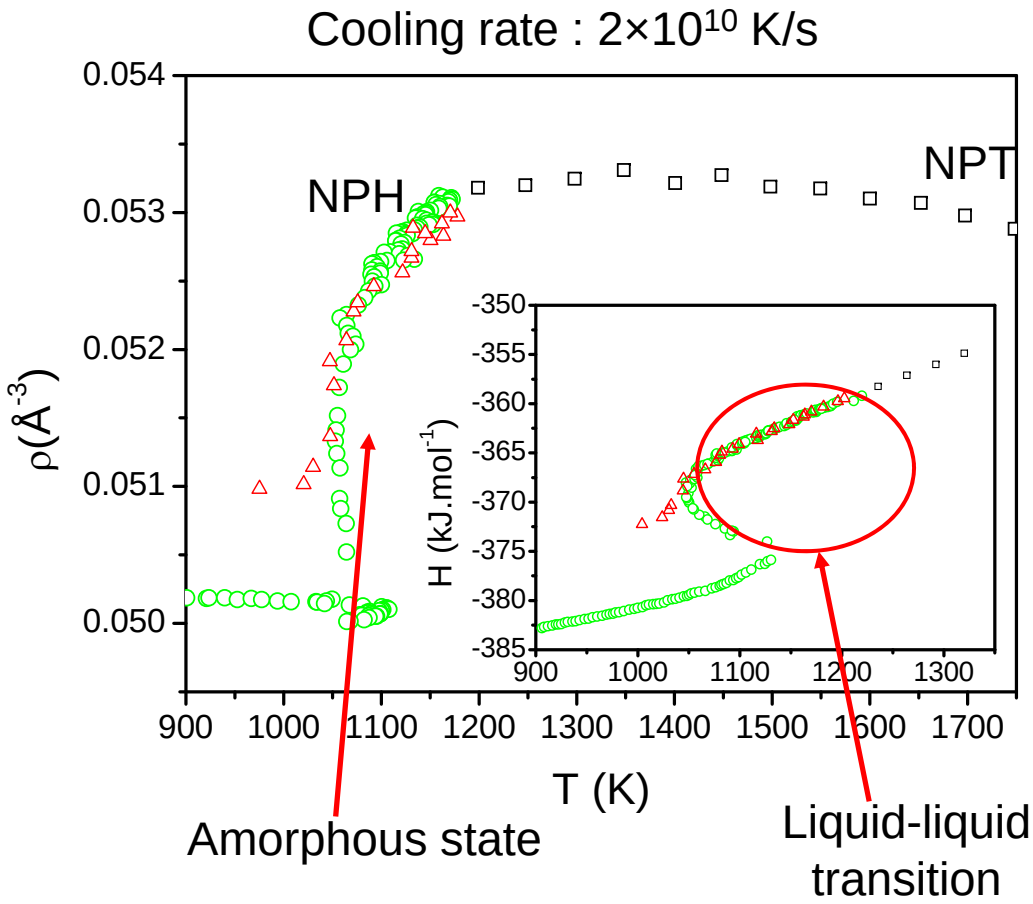
- Pb : slight change of parameters :
  disappearance of the LLT
  Beaucage and Mousseau, JPCM 17 (2005)

- Full *ab initio* MD : too heavy !
  (low cooling rate)

→ Combining classical and *ab initio* MD



LDL snapshot



Sastry and Angell, Nature Mat (2004)

NJ et AP, PRL **99**, 205702 (2007)

Conclusion

- Calculations made possible with CIMENT(Froggy:equip@meso +Nanostar) and GENCI

- Ab initio molecular dynamics (AIMD):
  - gives an accurate description of the structural and dynamical properties : viscosity and diffusion which are important for the solidification process.
  - capture the cristallisation (liquid boron)
  - necessary to capture the bond-orientational order and liquid favored structures

- Hybrid strategy consisting in combining AIMD and classical MD:
  - capture the structural and dynamic evolution upon undercooling and in the vicinity of the glass transition (Cu-Zr).
  - Liquid-liquid phase change in undercooled silicon

- Liquid is not a random disordered, homogeneous state but has local structural order or locally favored structures characterized by bond orientational order.

- Understanding the local structural ordering in liquids is important for solidification processes

Thank for your attention