Modélisation atomistique de liquides à haute température

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cnrs dépasser les frontières

Journée de prospective sur la physicochimie des liquides à haute température
Paris, 29 nov 2024

Un grand merci à

- M. Benoit (Toulouse)
- L. Berthier (Paris)
- M. Boero (Strasbourg)
- T. Charpentier (CEA Saclay)
- J.M. Delaye (CEA Marcoule)
- R. Dupuis (Montpellier)
- N. Jakse (Grenoble)
- W. Kob (Montpellier)
- C. Massobrio (Strasbourg)
- M. Salanne (Paris)
- G. Tréglia (Marseille)

Sommaire

- Modélisation atomistique:
  approches, échelle de temps & longueur, complexité
- Modélisation de liquides: quelques exemples
  propriétés structurales, dynamiques, transport, etc
- Outils & moyens
  Codes de simulation, codes d'analyse de données
  Centres de calculs
  Plateformes de partage de données, potentiels, exemples de calcul
- Communauté de chercheurs
  oxydes (silicates, carbonates, ….), chalcogénures, liquides métalliques, liquides ioniques, ...

Computer simulations

(*in silico* experiments)

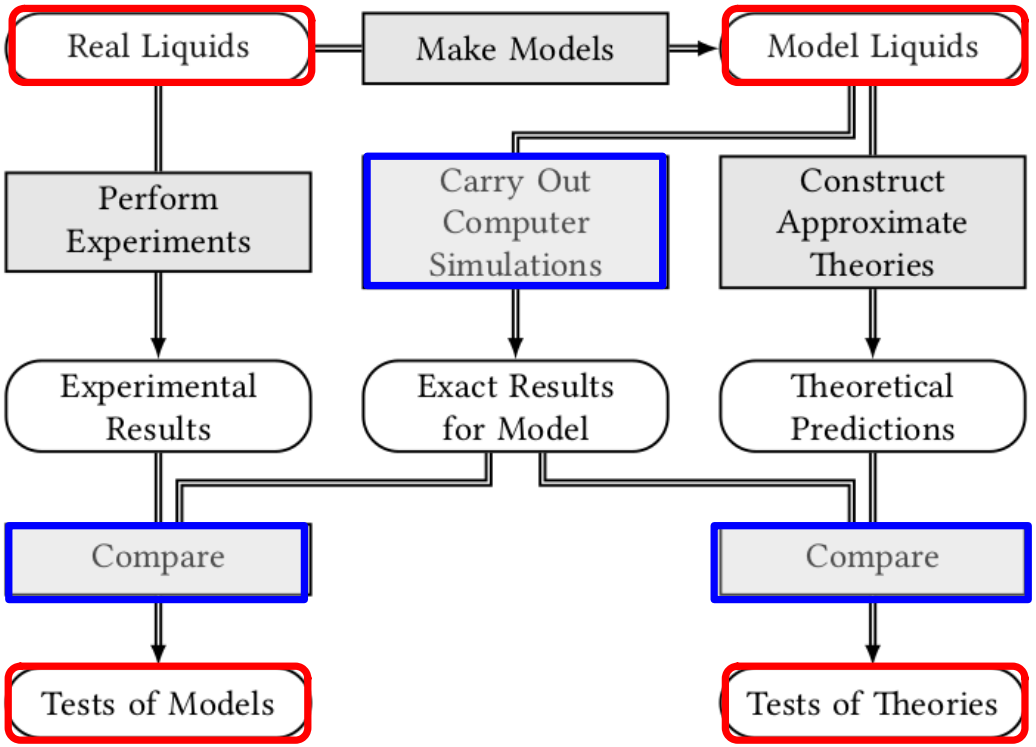


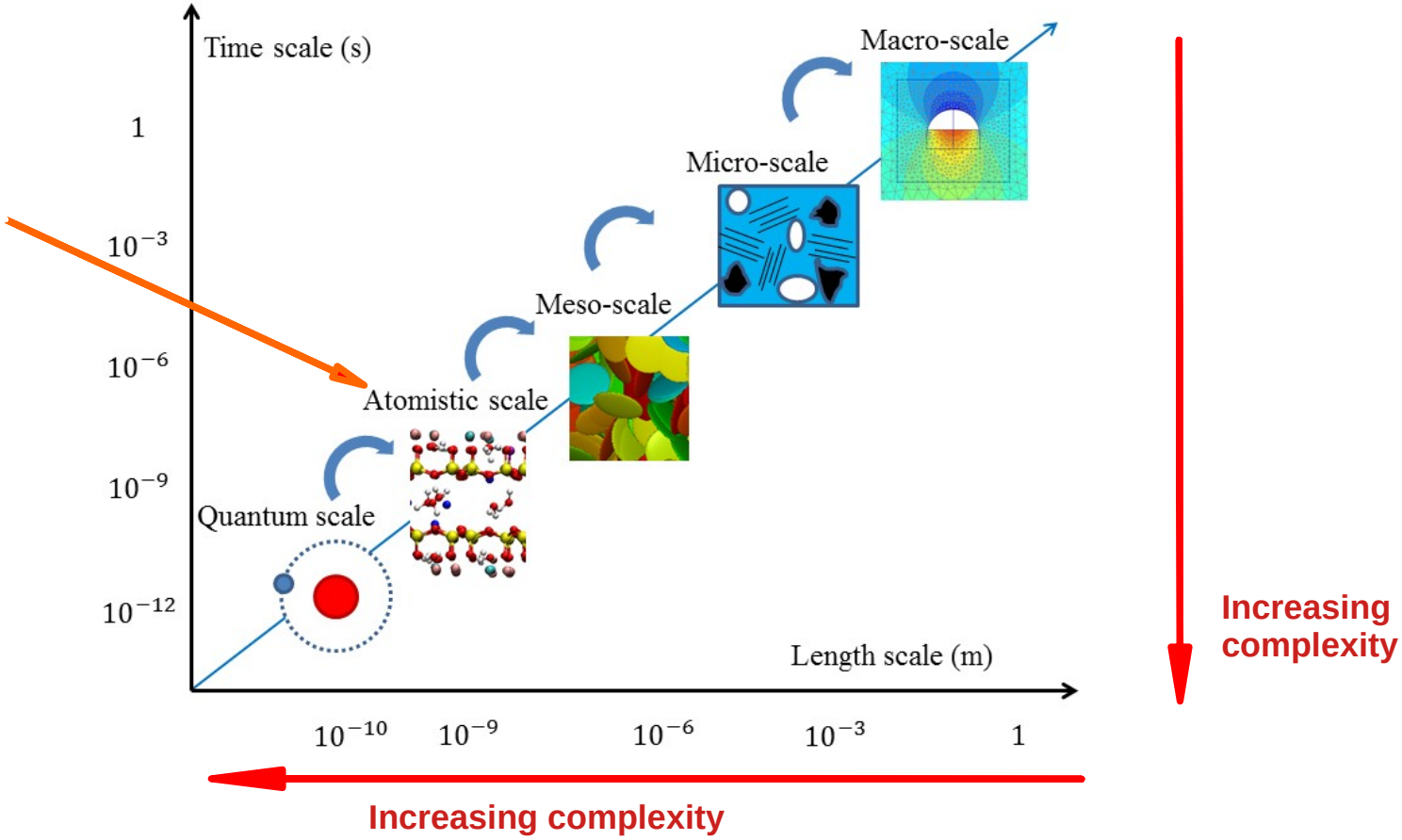
Fig. 1.2 The connection between experiment, theory, and computer simulation.

Computer simulations of liquids, Allen&Tildesley (2017)

- Established **tools to study complex systems** and get insight into their properties
- Dual role, bridge between
  - ➔ Models & experiments
  - ➔ Theories & models
- Simulations fill the gaps between real **experiments** and **theories**, and between **microscopic** features and **macroscopic** behavior

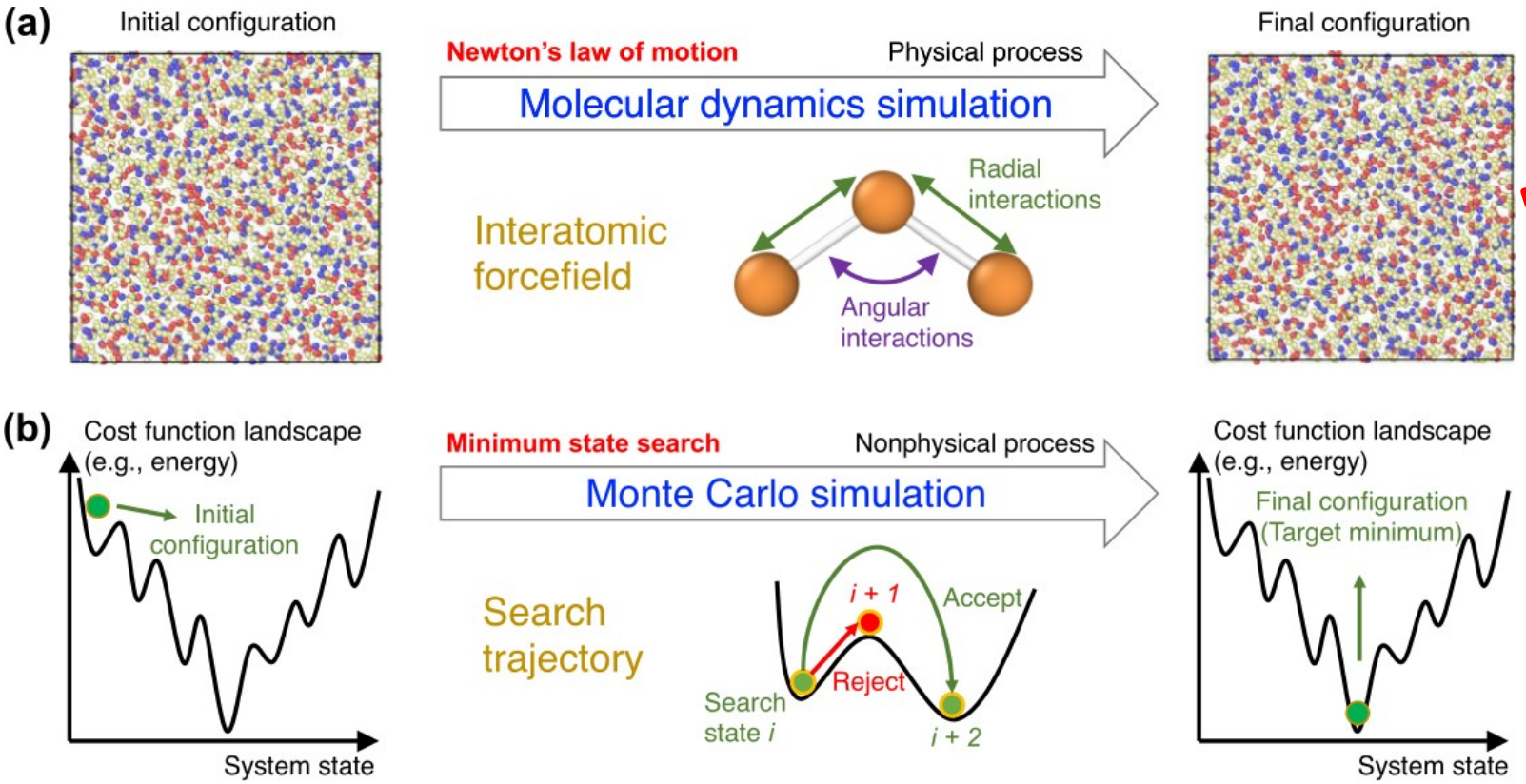
Time & length scales vs complexity

This talk



https://ajw-group.mit.edu/multiscale-modeling-clays & PhD Thesis, R. Brandelli Schaan, ENS Lyon 2022

Computer simulations at atomistic level



Main ingredient:

interaction model of the particles

2 possibilities: classical and *ab initio* simulations

Liu et al. "Challenges and opportunities in atomistic simulations of glasses: a review." Comptes Rendus. Géoscience (2022)

Classical *vs ab initio* approach

Classical approach

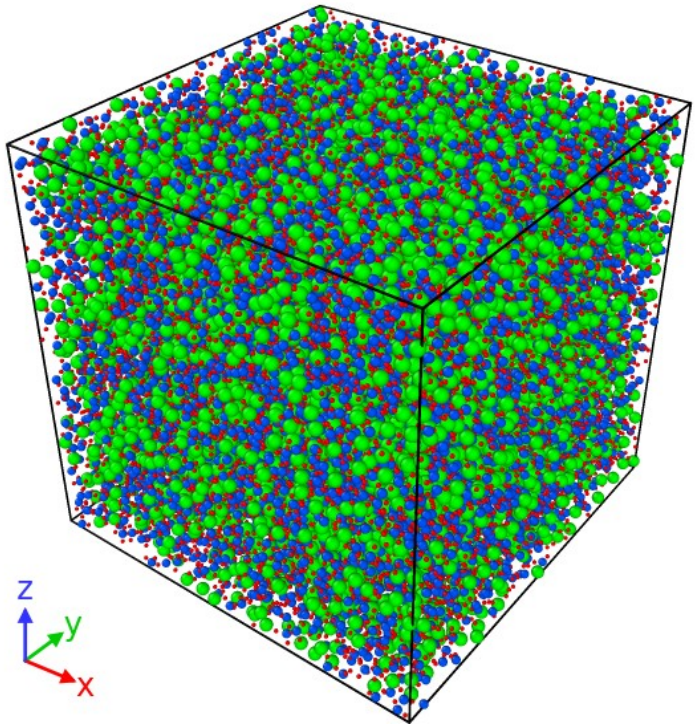
- Atoms considered as interacting point particles, electrons not explicitely taken into account
- One postulates an ansatz for the effective interatomic forces :
  - traditional potentials
  - Machine Learning potentials
- Balance between a simple and realistic description of the system under consideration

Ab initio approach

- Interatomic forces are calculated from the instantaneous positions of the ions and taking into account the first principles of quantum mechanics
- Universality (i.e. no ansatz for forces)
- It can handle relatively complex systems **but** it is computationally (**very) expensive**

Classical vs ab initio approach

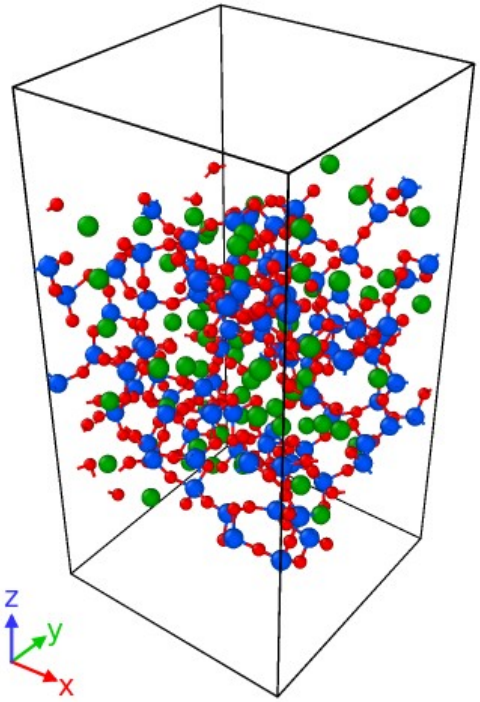
Classical MD:
600 000 atoms



Typical scales vs Approach	**Classical**	**Ab initio**
Nb of atoms	10,000 - millions	100 – 1,000
Length of the run	Nano- to µs	Tens of ps to few ns
Size of the simulation box	Nano to µm	Few nm

Sodium silicate Na_2O-3SiO_2

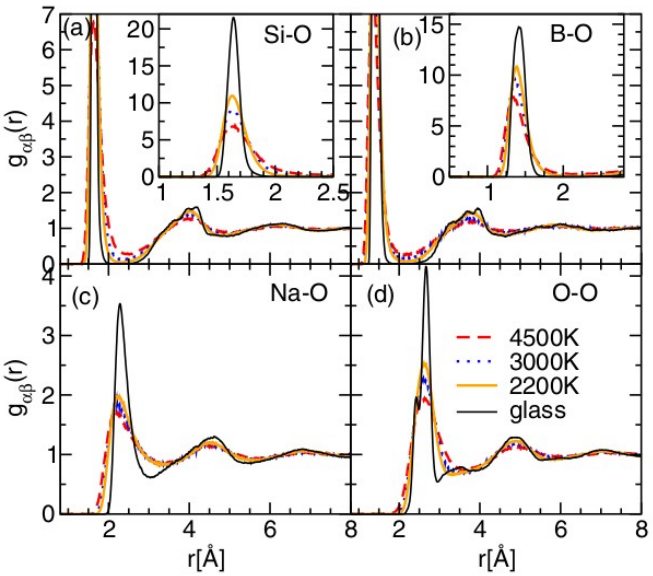
Ab initio MD:
420 atoms

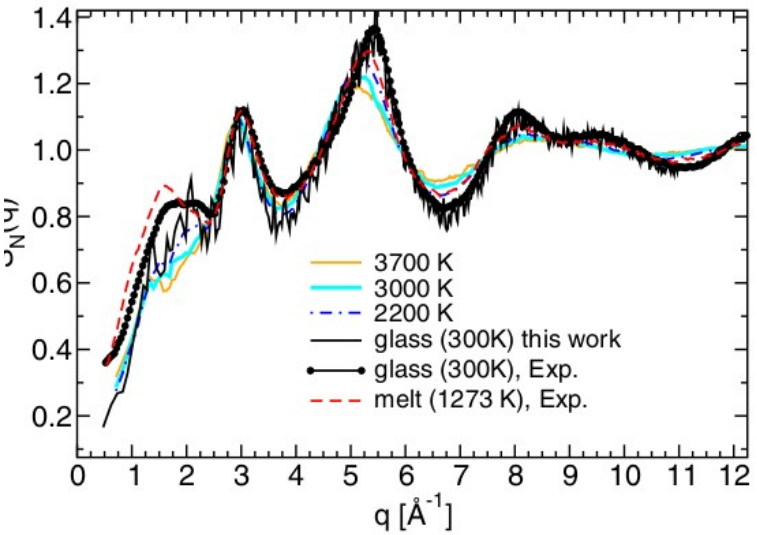


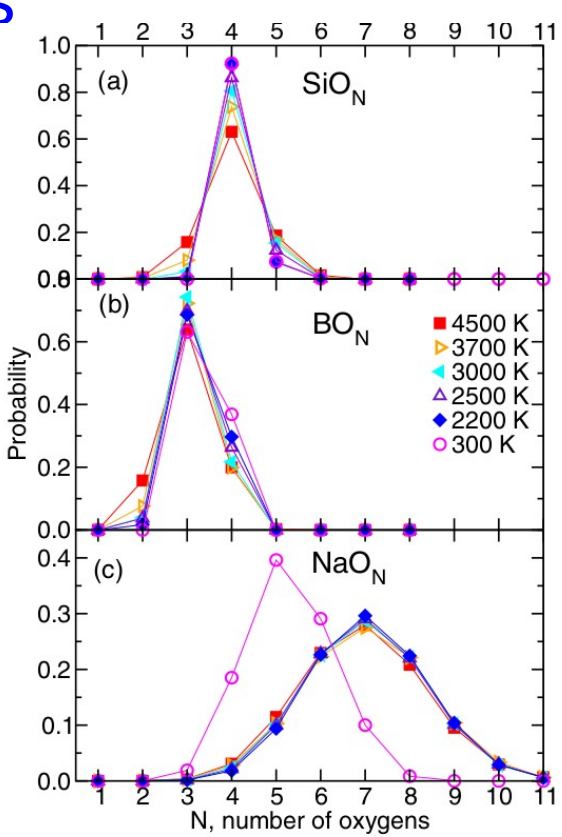
Atomistic simulations of liquids: some examples

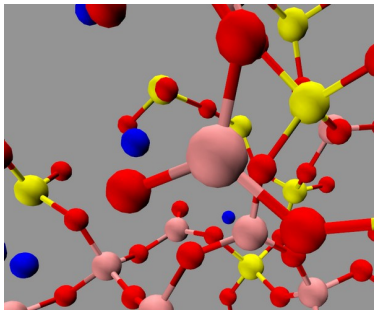
Disclaimer: choice of examples biaised by my own interests and expertise.

Sodium borosilicate liquid: structural properties







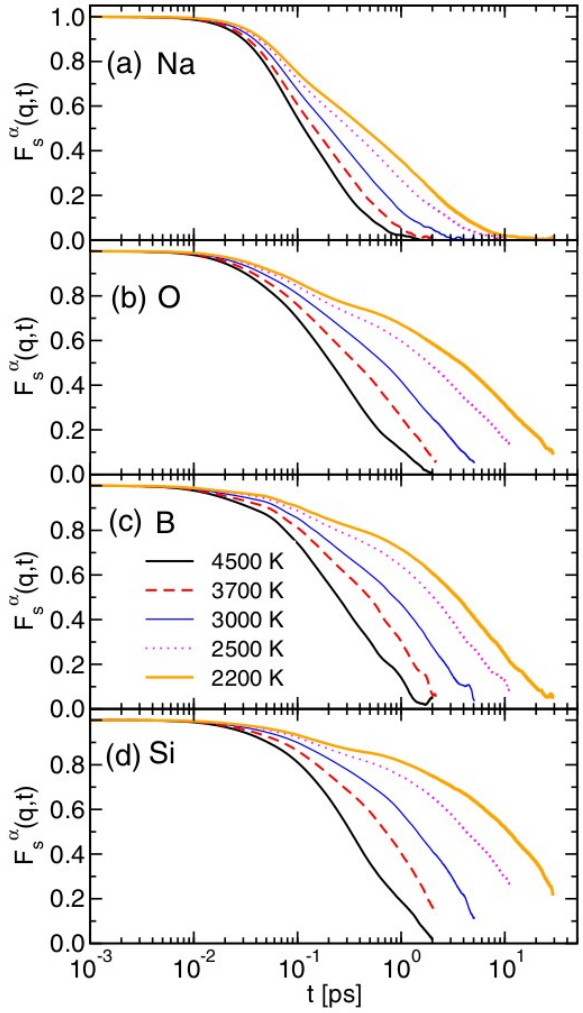


***Ab initio* MD study**

- Si-O & Na-O correlations expected behaviour with decreasing temperature
- B-O & O-O correlations more complex behavior with decreasing temperature
- Neutron structure factor: good agreement to exp. data

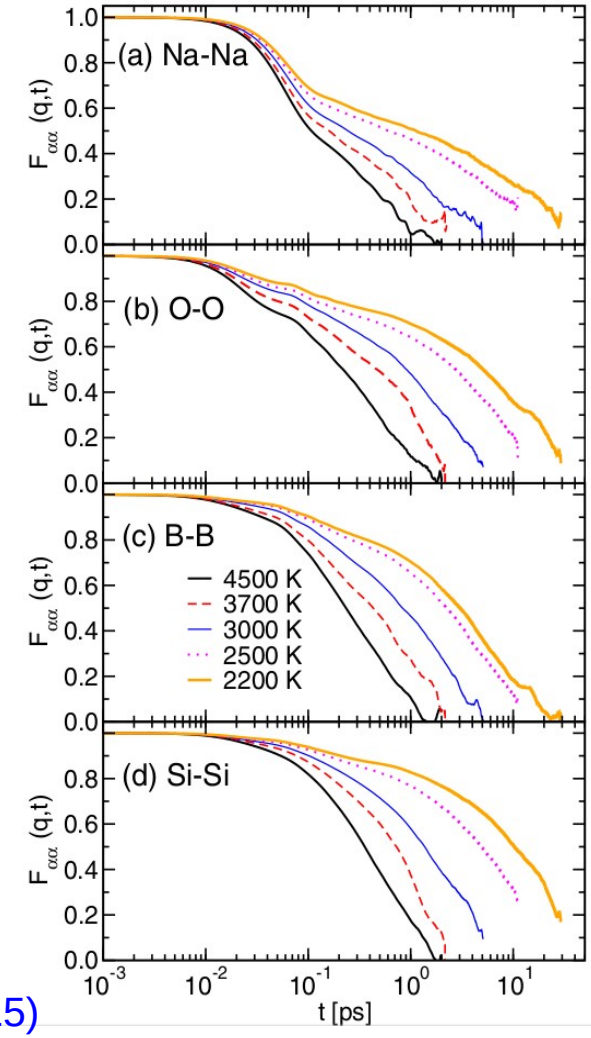
Pedesseau, Ispas, Kob, Phys.Rev B 91 (2015)

Sodium borosilicate liquid: dynamical properties



Intermediate scattering functions:
incoherent vs coherent

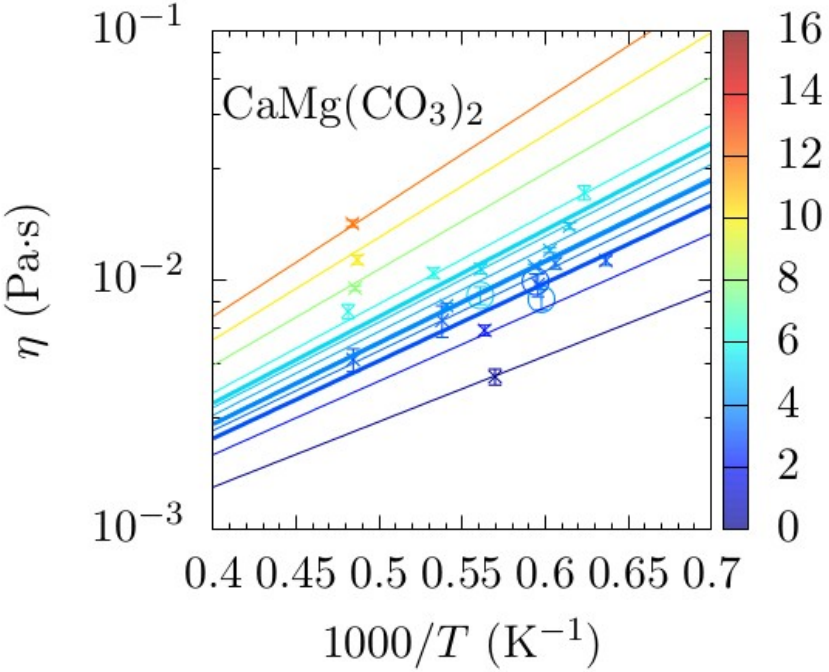
- Quick relaxation at high temperature (exponential decay & no species dependence)
- Two-step decay at lower temperatures
- Very similar correlators for Si, O & B (shape and relaxation time)
- Different trend for Na: “special sites” (once a site is vacated it is quickly occupied by another Na)
- Time scale to decorrelate of the “special sites” is the time needed by the matrix to rearrange

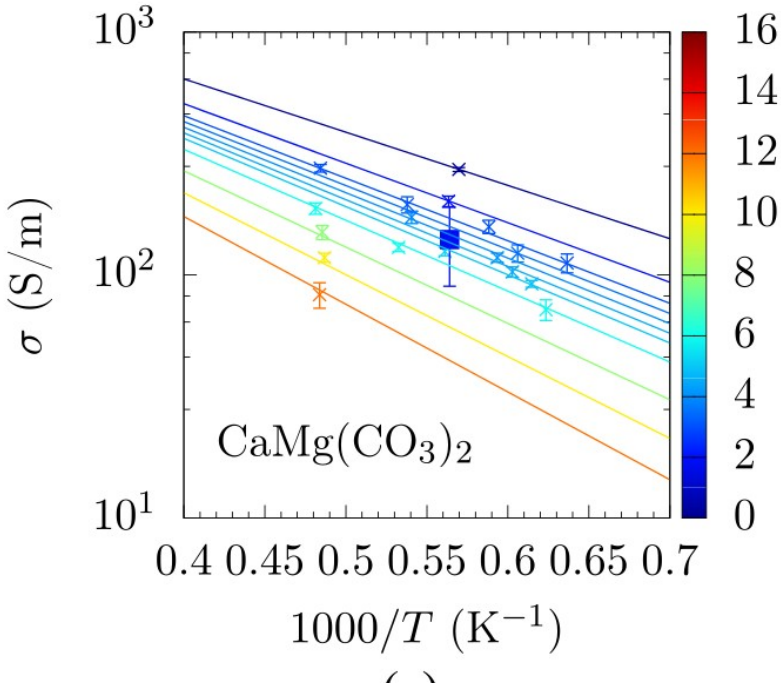


Pedesseau, Ispas, Kob, Phys.Rev B 91 (2015)

Molten carbonotes: transport properties

Classical MD simulations, pair potential

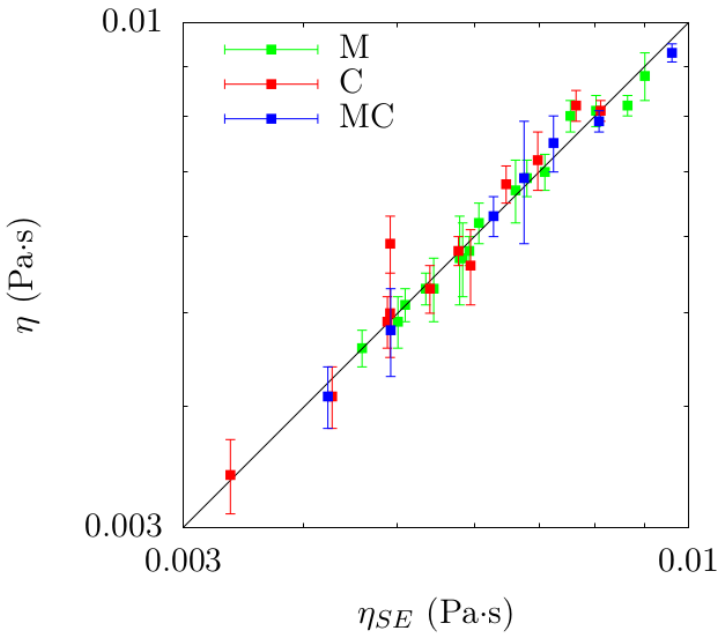


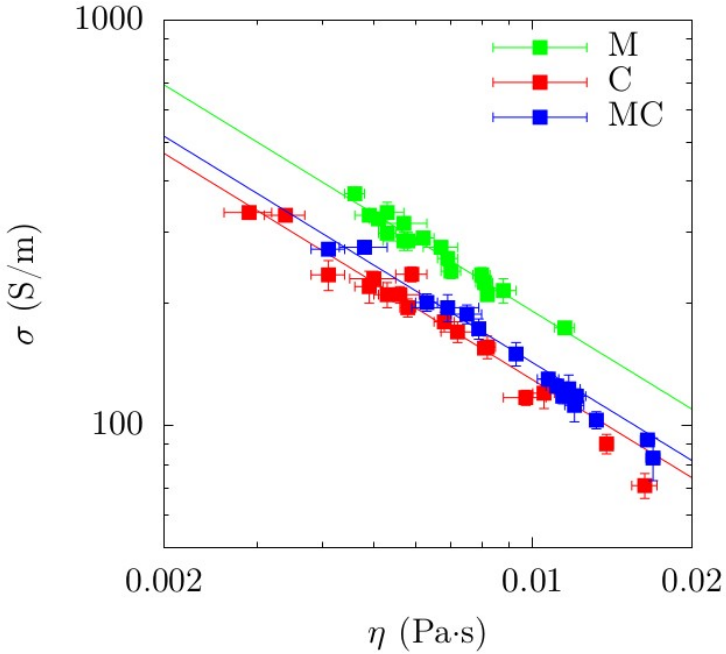


- Smooth dependence (Arrhenius like) with respect to T and P
- Non-negligible effect of the composition
- Insights into magmatic processes implying carbonatitic melts

Desmaele, Sator, Vuilleumier, Guillot, J. Chem Phys 150 (2019)

Molten carbonates: transport properties



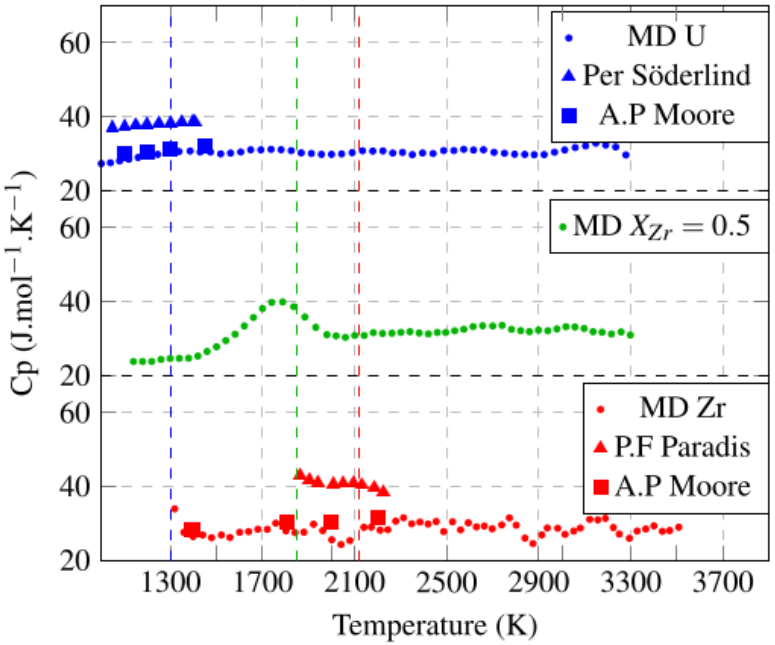


- Testing the reliability of the phenomenological equations (here Stokes-Einstein)
- Identification of empirical relationship between viscosity and thermal conductivity

Desmaele, Sator, Vuilleumier, Guillot, J. Chem Phys 150 (2019)

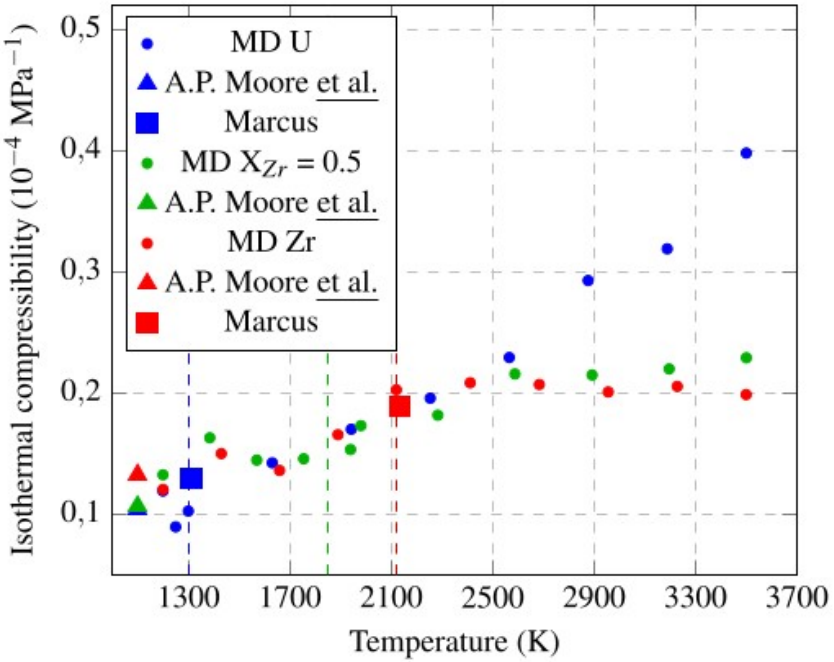
Liquid (U,Zr) mixture: thermophysical properties

Classical MD simulations, MEAM potential



Heat capacity:

- pure U and Zr equal to ~3R for both solid and liquid phase
- (U, Zr) mixture: different trend



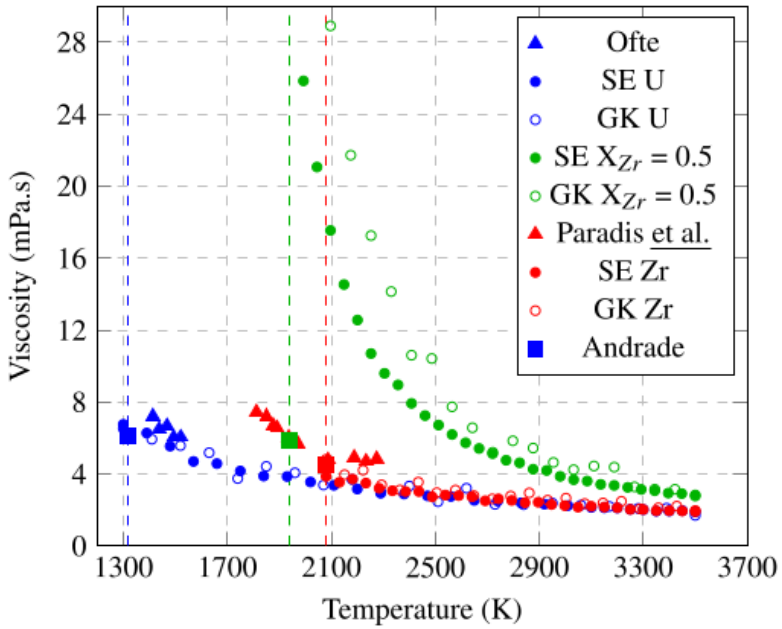
Isothermal compressibility:

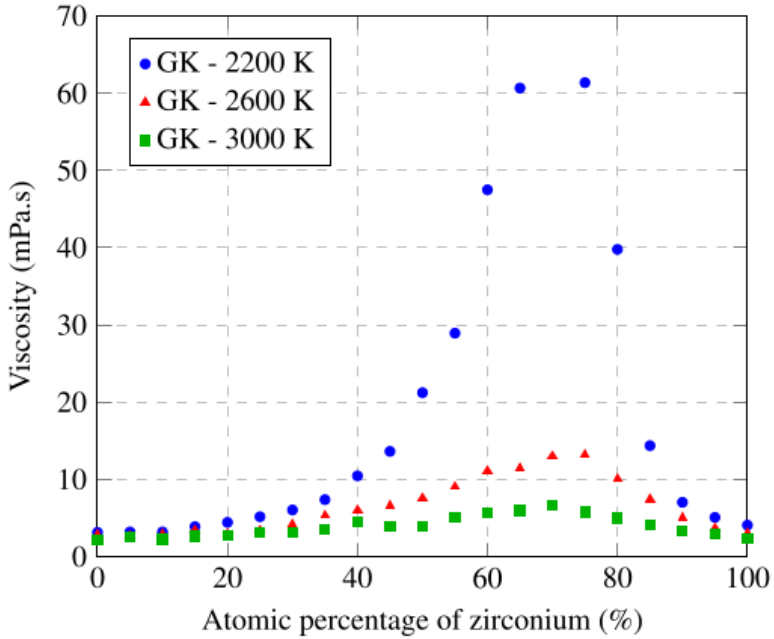
- Liquid phase: different trends
- difference not currently accounted for in nuclear accident scenario codes

Tranchida, Nicaud, Beeler, Bourasseau J. Chem Phys 160 (2024)

Liquid (U,Zr) mixture: thermophysical properties

Classical MD simulations, MEAM potential





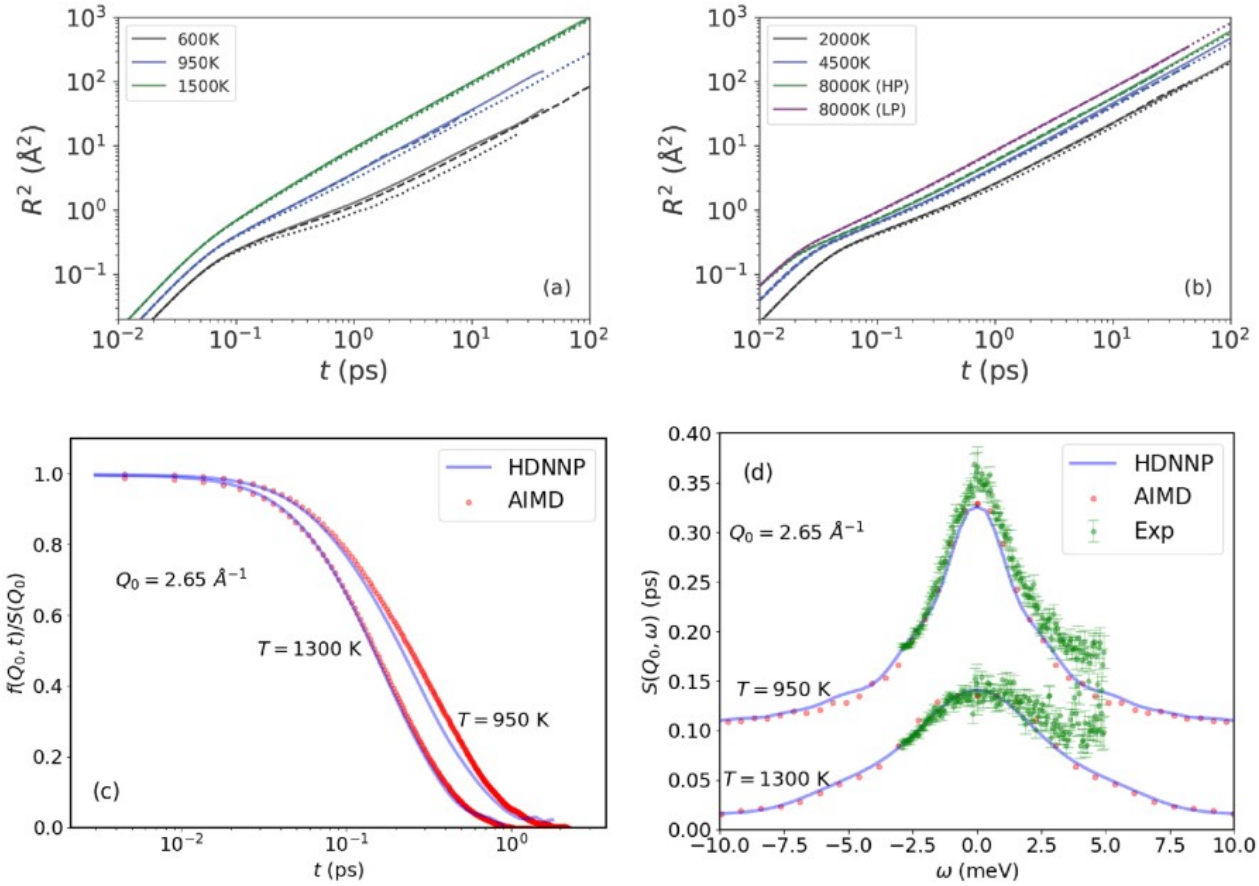
Viscosity:

- Pure U and Zr: phenomenological models work
- Breakdown of Stokes-Einstein relation for (U, Zr) mixture

- Large increase of the viscosity at intermediate (U, Zr) composition
- May have important consequences for large-scale nuclear accident scenario simulations

Tranchida, Nicaud, Beeler, Bourasseau J. Chem Phys 160 (2024)

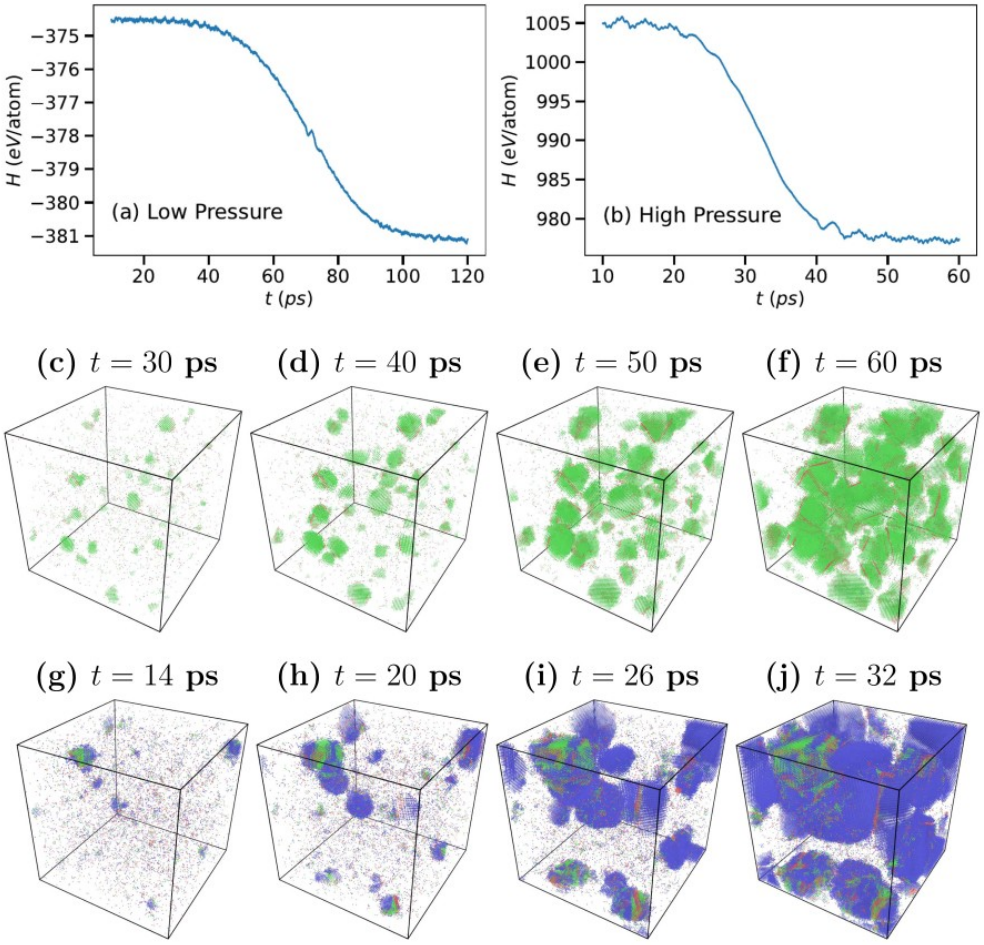
Machine learning (ML) interatomic potentials for Al: application to solidification



- **Diffusion:** ML potential reproduces the ab initio trends for all temperatures and pressures
- **Intermediate scattering function:** good agreement of ML results to AIMD ones
- Good match of the calculated dynamic structure factors (ML & ab initio) to exp. Data
- Predictive character of the ML potential for states not included in the training

Jakse et al. J. Phys.: Condens. Matter 35 (2023)

Machine learning interatomic potentials for Al: application to solidification



- Investigation of the homogeneous crystal nucleation on large scales with an ab initio accuracy
- A single step nucleation, with an emerging fcc ordering and hcp stacking fault defects
- A single step nucleation pathway with bcc nuclei observed at high pressure.

Jakse et al. J. Phys.: Condens. Matter 35 (2023)

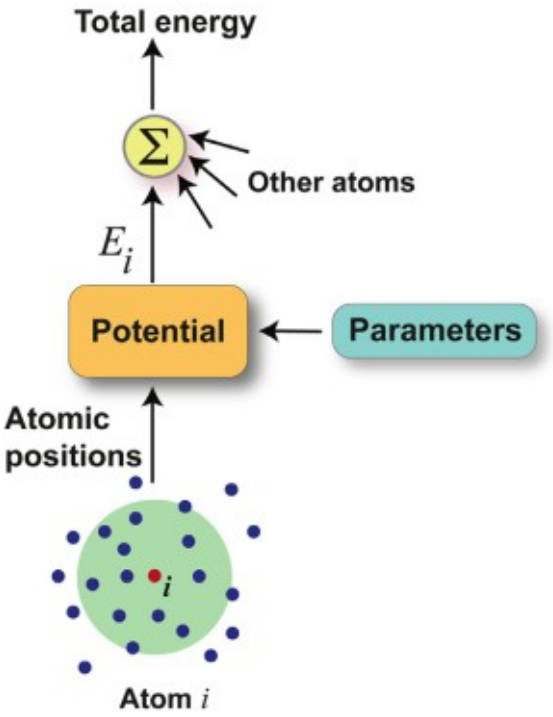
Machine learning interatomic potentials for materials

	Potential type		
	Traditional	ML	Physically-informed ML
Physical foundation	Strong	None	Strong
Number of fitting parameters	~ 10	$\gtrsim 10^3$	$\gtrsim 10^3$
Computational speed	Very high	Slower[a]	Slower[a]
Reference database	Small	Large	Large
Accuracy (interpolation)	Limited	~ 1 meV/atom	~ 1 meV/atom
Transferability (extrapolation)	Reasonable	Poor	Reasonable
Reliance on human expertise	Strong	Weaker[b]	Weaker[b]
Extension to chemistries	Challenge	Challenge	Challenge
Specific to class of materials?	Yes	No	No
Systematically improvable?	No	Yes	Yes

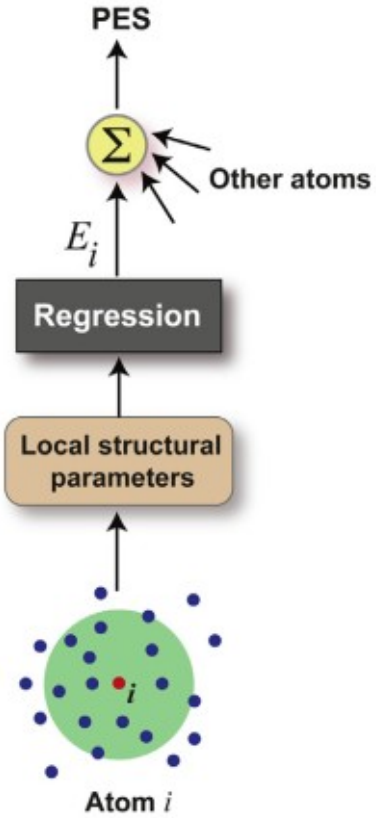
Mishin, Acta Materialia 214 (2021)

Machine learning interatomic potentials for materials

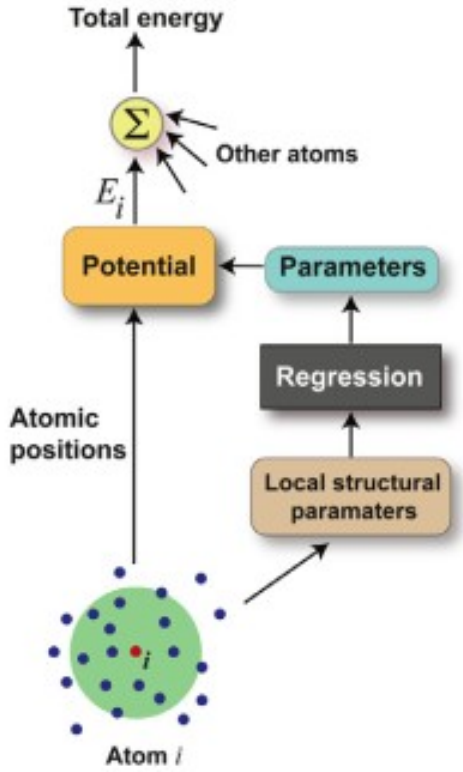
Traditional potentials



ML potentials

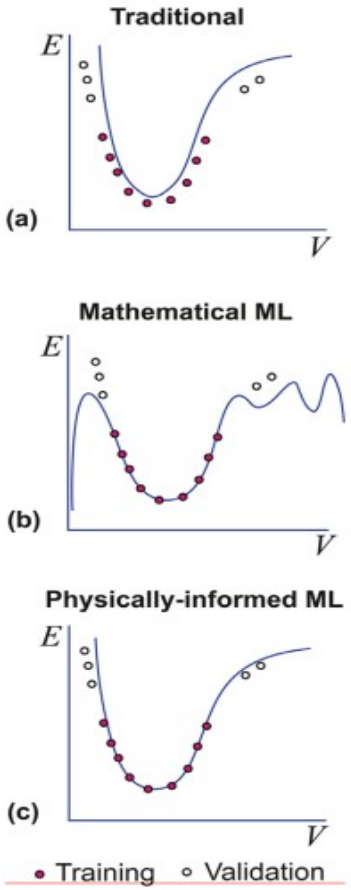


Physically informed ML potentials



Mishin, Acta Materialia 214 (2021)

Machine learning interatomic potentials for materials



Accuracy and transferability:
Predictions of the energy-volume (E-V) relation compared to DFT calculations

Mishin, Acta Materialia 214 (2021)

Codes utilisés

Approche ab initio: VASP, CP2K, CPMD, Quantum Espresso, Abinit, ….
Approche classique: LAMMPS, DL_POLY, ….
Codes spécifiques: ….
Analyse des données: logiciel Ovito, code maison rendus publiques, LAMMPS

Moyens de calcul

National: GENCI (Grand Equipement National de Calcul Intensif)
– 2 appels à projets/an + accès dynamiques
– 3 centres: IDRIS (Orsay), CINES (Montpellier), TGCC (Bruyères Le Châtel)
Mésocentres: e.g. CALMIP (Toulouse), Méso@LR (Montpellier), Lyon, Grenoble, Paris, Nantes, Strasbourg, ….
Cluster locaux: laboratoire ou équipe

Projet PRACE: accès aux centres HPC européens (Juelich, Barcelona, ...)

Miscellaneous

- **FAIRmat: Findable, Accessible, Interoperable, Re-usable**

A data infrastructure for condensed-matter physics and the chemical physics of solids *www.fairmat-nfdi.eu/fairmat*

- **OpenKIM** - a curated repository of interatomic potentials *openkim.org*

- ML potential repositories: see *Mishin, Acta Materialia 214 (2021)*

- **MATSCI**.org: is a community forum for the discussion of anything materials science, with a focus on computational materials science research

MATSCI COMMUNITY DISCOURSE

- **CECAM** *www.cecam.org*

cecam
Centre Européen de Calcul Atomique et Moléculaire

Modélisation atomistique de liquides en France

Liste préliminaire (ie. très loin d'être exhaustive)

- Oxydes (silicates, carbonates, …)
  - M. Micoulaut, M. Salanne, N. Sator, R. Vuilleumier (Paris), T Charpentier (CEA), JM Delaye (CEA), N Jakse (Grenoble), W. Kob (Montpellier)
- Liquides métalliques:
  - N. Jakse, JF Wax (Metz), J. Lam (Lille), O. Politano (Dijon), C. Bichara (Marseille)
- Liquides ioniques, électrolytes:
  - M Salanne, B. Rotenberg, G. Jeanmairet (Paris), M Boero & G Ori (Strasbourg)

    A. Padua (Lyon), JF Dufrêche, M. Duvail, B. Siboulet (ICSM Marcoule), C. Merlet (Toulouse)S. Mossa (Grenoble), A. Rakhmatulin (Orléans)

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Modélisation atomistique de liquides en France

Liste préliminaire (ie. très loin d'être exhaustive)

- Chalcogénures:

  C. Massobrio, M Boero & G Ori (Strasbourg), M Micoulaut

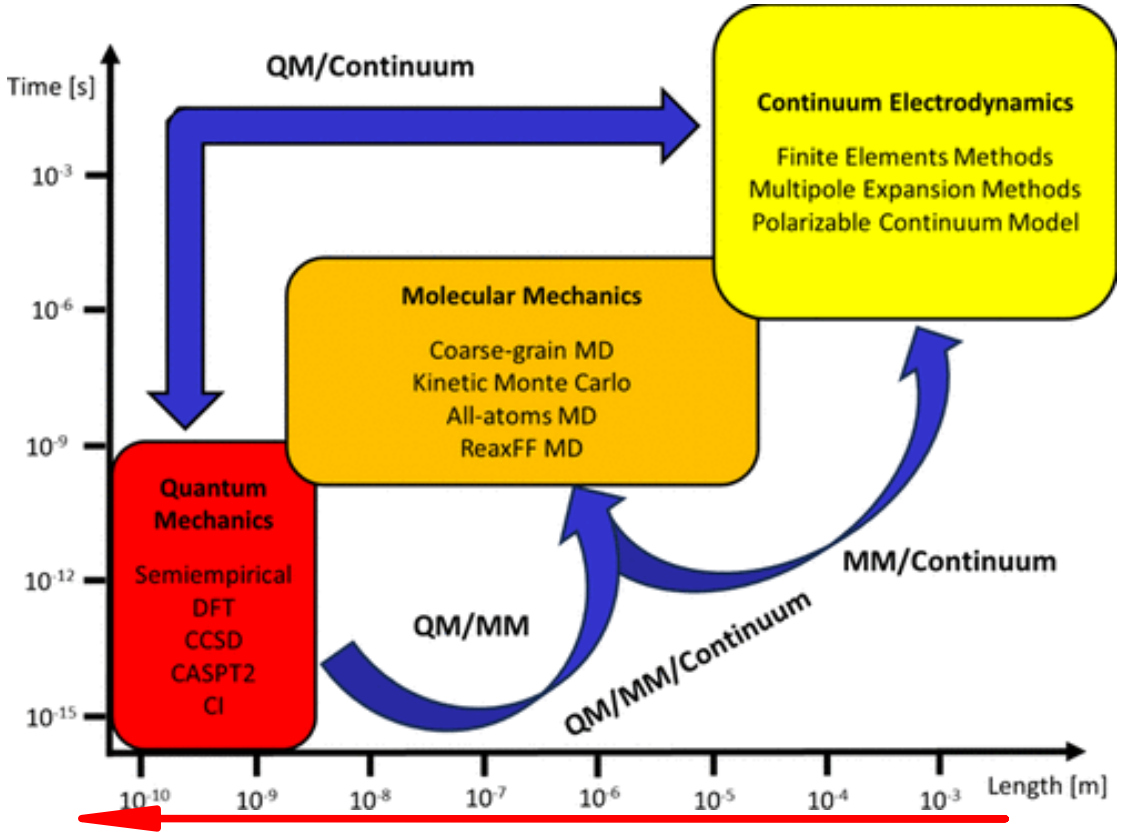
- Thiosilicate & sélenosilicate:
  - M Micoulaut (Paris)
- Géopolymètres:
  - R Dupuis, R Pellenq (Montpellier), A Poulesquen (CEA Marcoule)
- Géochimie:
  - M Meheut, M. Benoit (Toulouse), R. Caracas (Paris)
- Combustibles fondus:
  - E. Bourasseau, J. Tranchida, J Bouchet (CEA Cadarache)
- Séparations des actinides en phase liquide:
  - D. Guillaumont, D. Moreno Martinez, P. Guilbaud (CEA Marcoule)

Modélisation atomistique de liquides en France

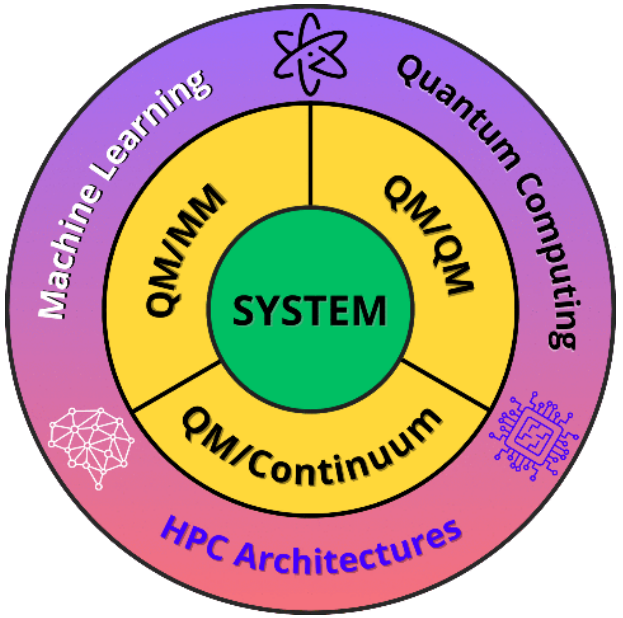
Liste en construction (ie. très loin d'être exhaustive)

- Systèmes similaires (liquides moléculaires):
  - A. France-Lanord, M Saitta, F Pietrucci, G. Ferlat, A. Seitsonen, R. Vuilleumier (Paris), C. Millet (Nancy), JM Leyssale (Bordeaux),
- Spectroscopie vibrationnelle des liquides:
  - M-P Gaigeot (Evry), D. Laage (Paris)
- Aspects méthodologiques:
  - L. Berthier (Paris), JL Barrat, B. Coasne (Grenoble), W. Kob (Montpellier), G. Foffi, F. Smallenburg (Orsay)
  - D. Borgis (Paris), D. Lesnicky, A. Carof (Nancy)
- …….

Back to time and length scales vs methods and their coupling



Increasing complexity



Capone et al. ACS Phys. Chem (2024)

Merci pour votre attention!

Help: compléter & corriger la liste des chercheurs/groupes travaillant sur la modélisation de liquide en France