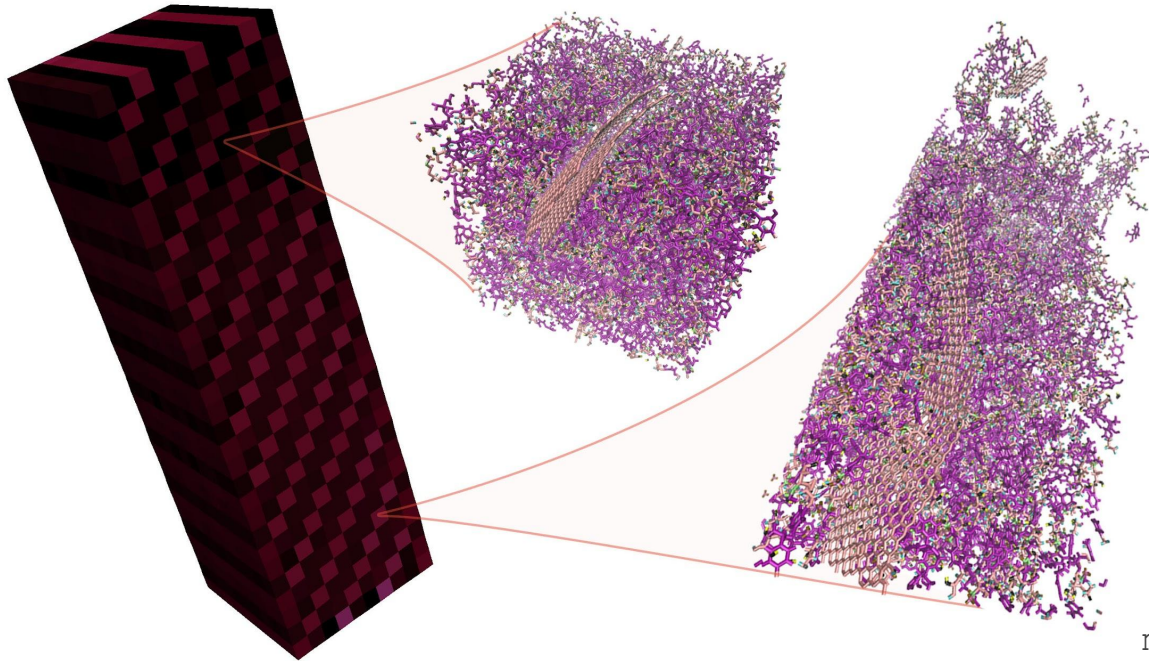


Towards high-fidelity materials property prediction from multiscale modeling and simulation



Maxime Vassaux
Institut de Physique de Rennes
Université Rennes 1

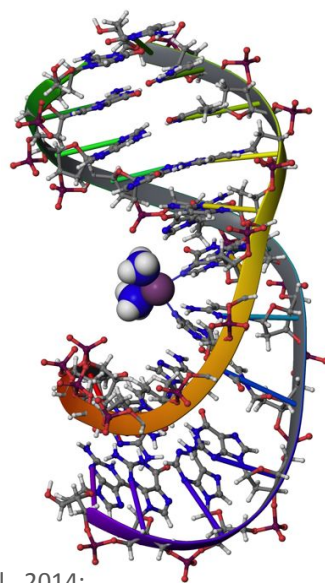
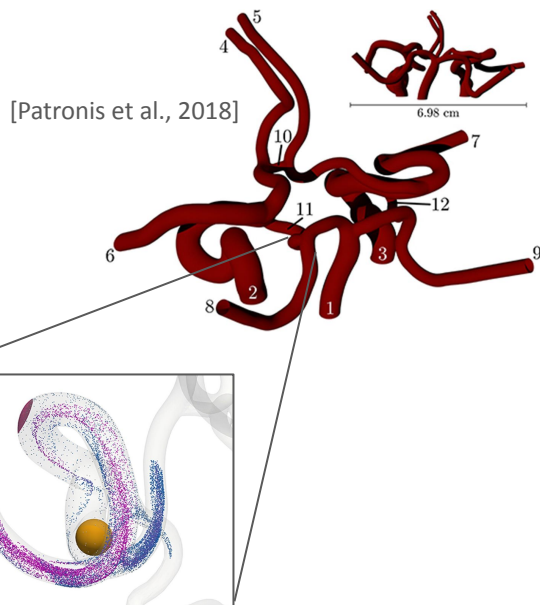


@mxvassaux

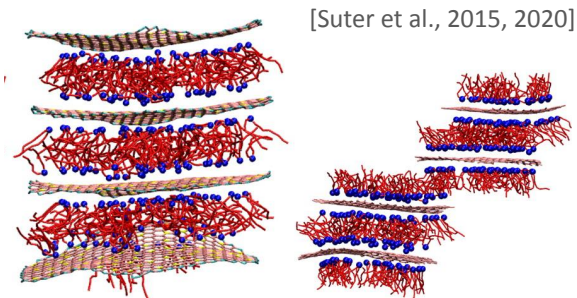
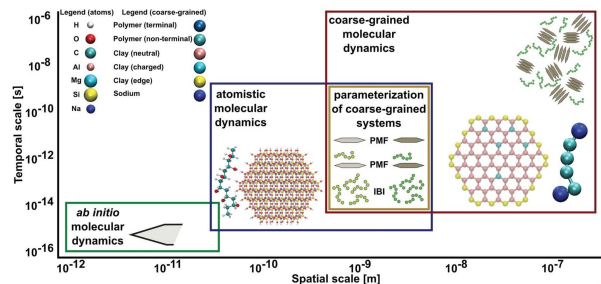
maxime.vassaux@univ-rennes1.fr

Centre for Computational Science at UCL (P.I.: Peter Coveney)

- HPC based on molecular dynamics and lattice Boltzmann: biomedicine, biomolecular, materials



[Jacquemin et al., 2014;
Gheorghiu et al., 2020]



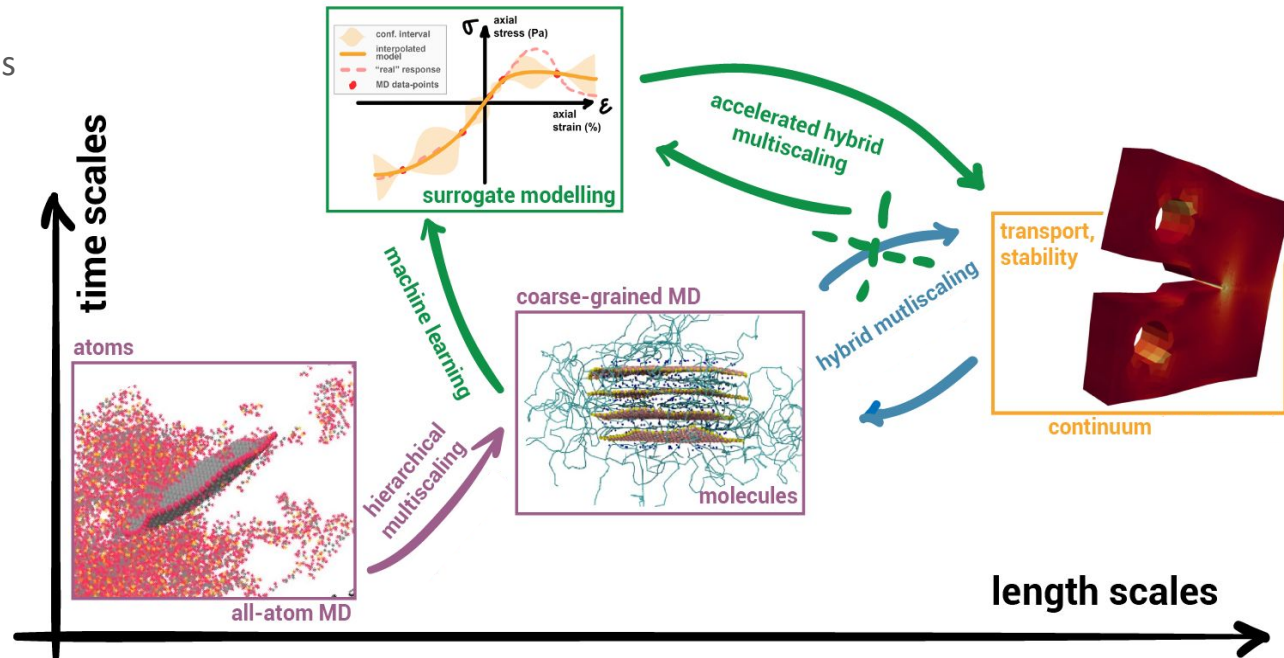
- Blood flow
 - particles diffusion
 - CG-MD → LB

- DNA nanopores
 - stability, transport
 - DFT → MM

- Polymer nanocomposites
 - material properties
 - DFT → AA-MD → CG-MD

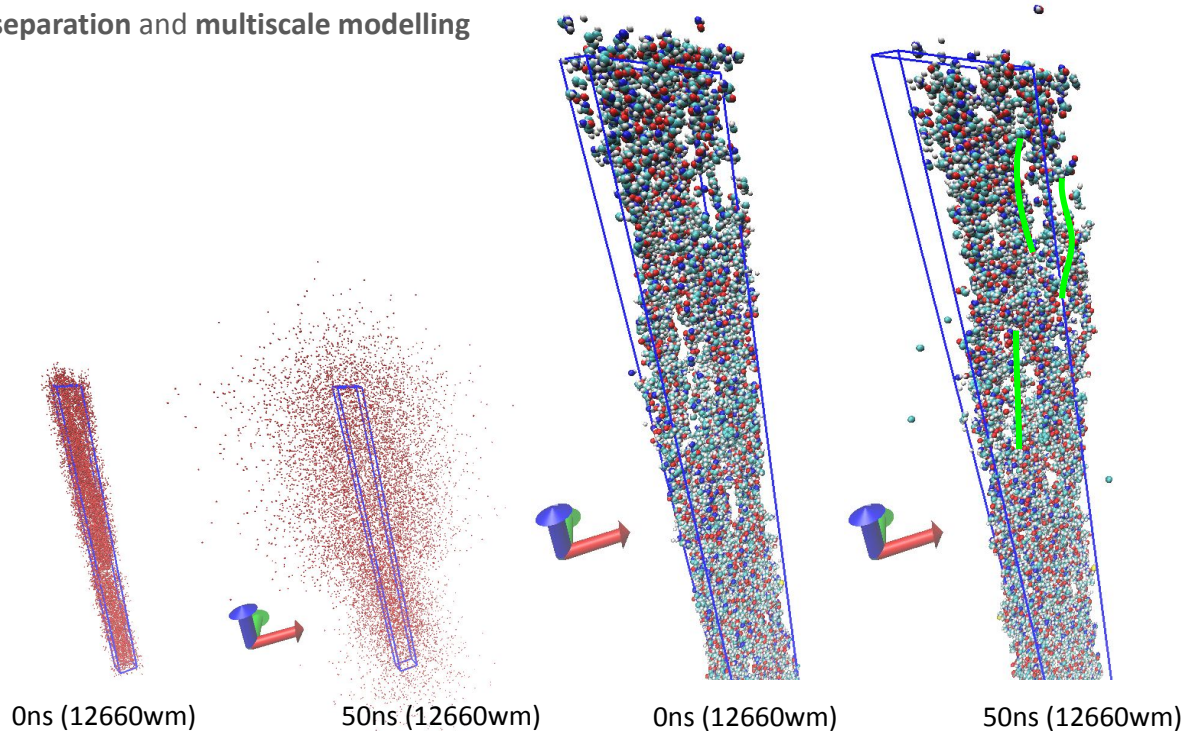
Towards actionable materials property prediction with chemical specificity

- **chemical specificity** from DFT/AA-MD simulations
 - **validation** against experiments needed
 - reaching **continuum**
 - making use of HPC resources
 - **certified** simulations
 - perspective: **reduced-order modelling**
- getting rid of biased **parameterisations**

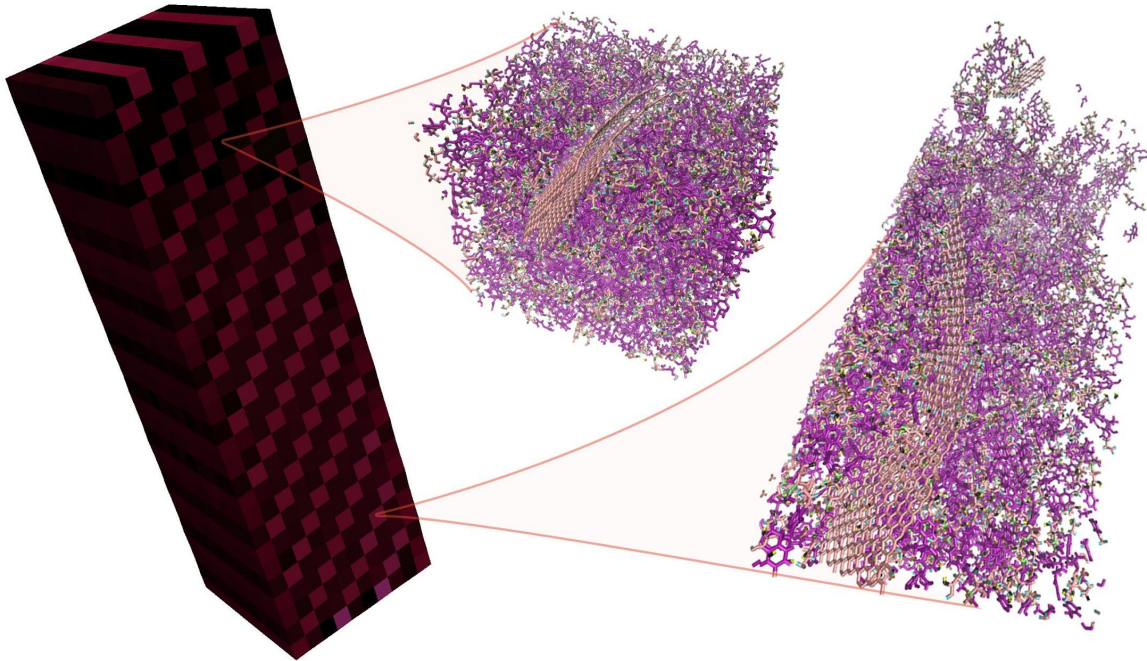


Single-scale all-atom molecular dynamics

- parameterisations (force fields) from DFT
- limited to **100,000s of atoms** and **10s of nanoseconds**
 - scale separation and multiscale modelling



Multiscale simulation methods



Molecular dynamics

- Newton's equations of motion, complex force field, **N-body dynamics**
- Lyapunov instability leads to **chaotic systems**
 - sensitive to **uncertainty**, need for **ensembles**

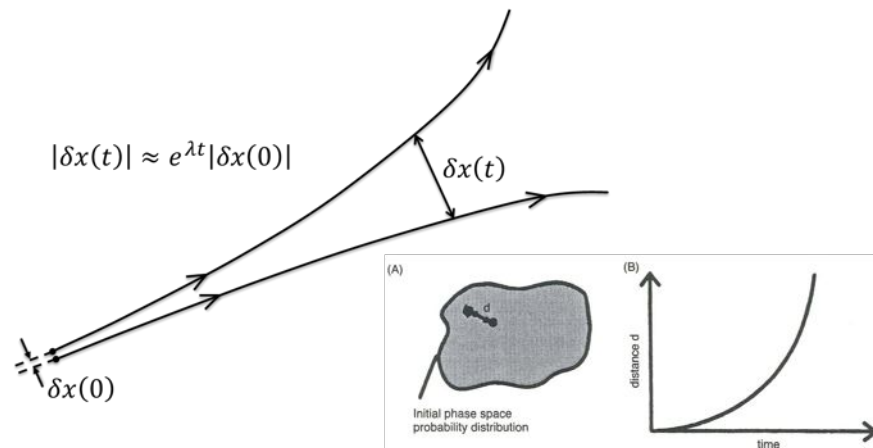
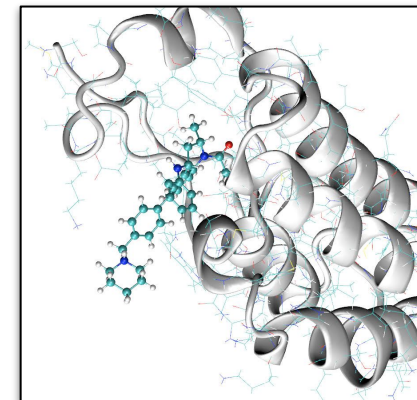
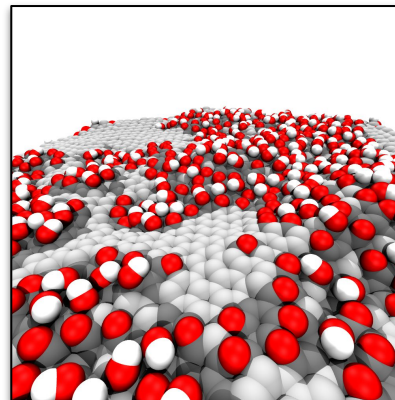
$$\mathbf{F} = m \frac{d^2 \mathbf{r}}{dt^2}$$

$$\mathbf{F} = -\nabla V(\{\mathbf{R}\})$$

$$V(\{\mathbf{R}\}) = \sum_{\text{bonds}} K_b (b - b_0)^2 + \sum_{\text{angles}} K_\theta (\theta - \theta_0)^2$$

$$+ \sum_{\text{dihedrals}} K_\chi (1 + \cos(n\chi - \chi_0)) + \sum_{\text{impropers}} K_\psi (\psi - \psi_0)^2$$

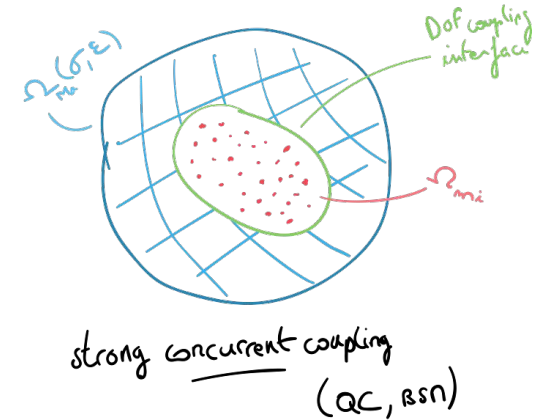
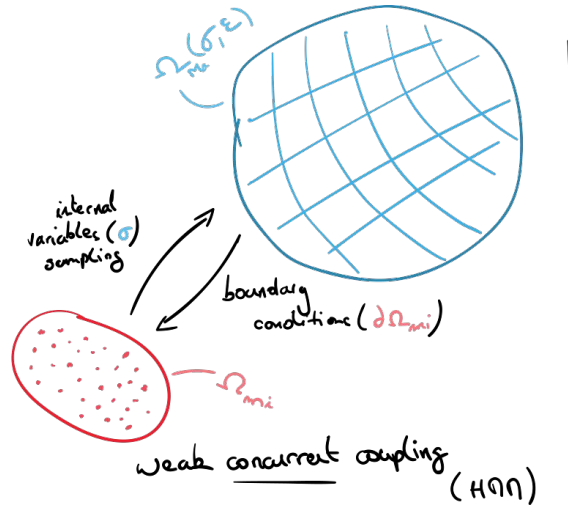
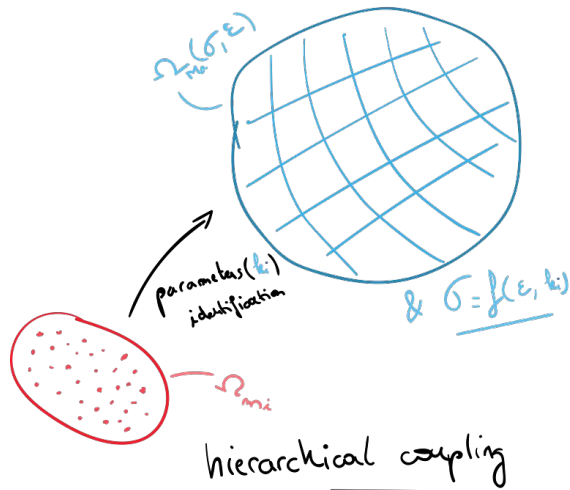
$$+ \sum_{\text{nonbond}} \left[\left(\frac{A_{ij}}{r_{ij}} \right)^{12} - \left(\frac{B_{ij}}{r_{ij}} \right)^6 + \frac{q_i q_j}{\epsilon r_{ij}} \right]$$



[Boghosian, Coveney and Wang. *Adv. Theor. Simul.* \(2019\) 2.12](#)
[Wan, et al. *J. Chem. Theory Comput.* \(2015\) 11, 7, 3346–3356](#)
[Suter et al... *Adv. Mat.* \(2020\) 32.36](#)

Scale-bridging or coupling scales

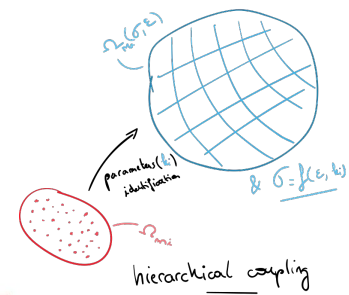
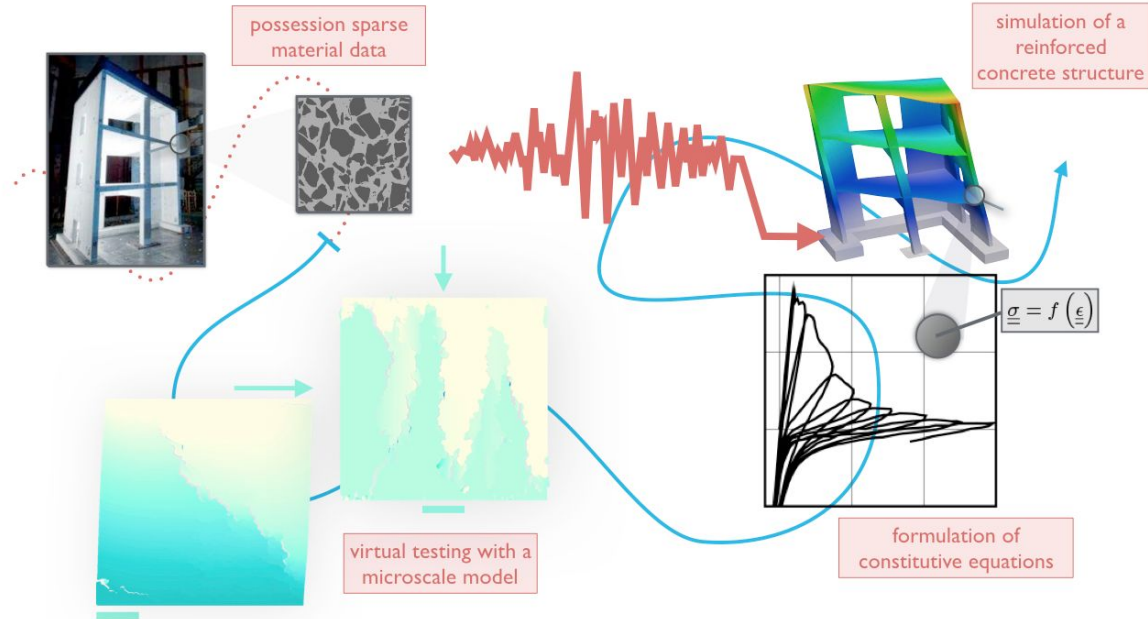
- different feedback frequency and resolution
 - hierarchical
 - semi-concurrent
 - concurrent



[Shenoy et al. J. Mech. Phys. Solids \(1999\) 47.3](#)
[E et al. Phys. Rev. B: Condens. Matter \(2003\) 67.9](#)

Hierarchical multiscaling

- continuum constitutive law parameterisation
- coarse-graining from all-atom molecular dynamics (IBI, STM, ...)



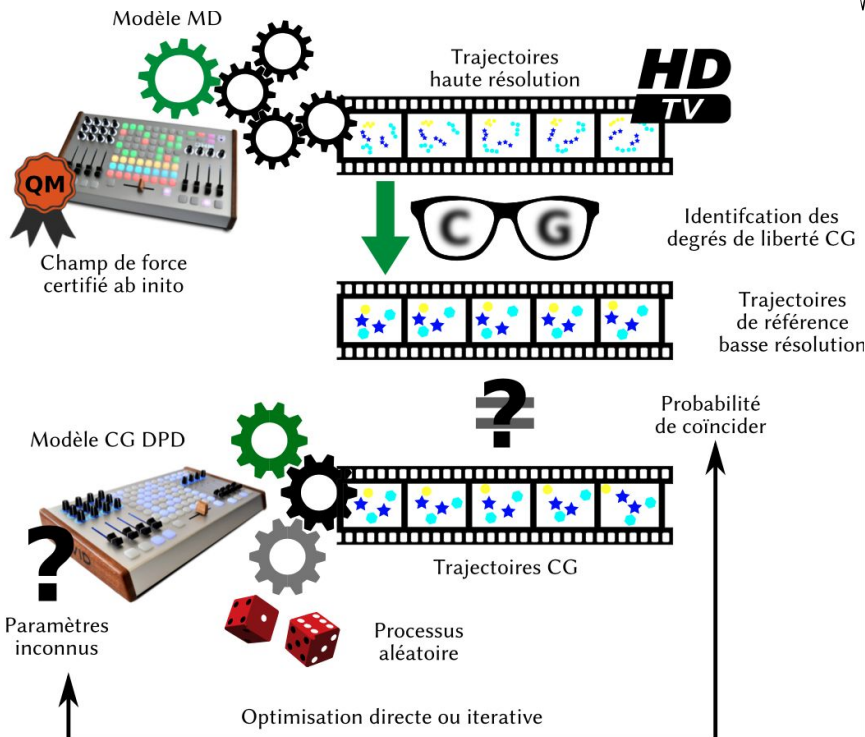
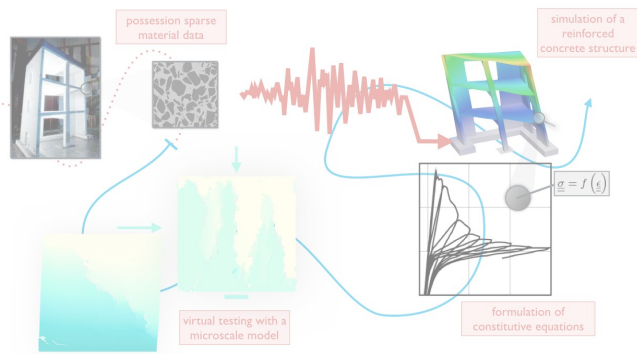
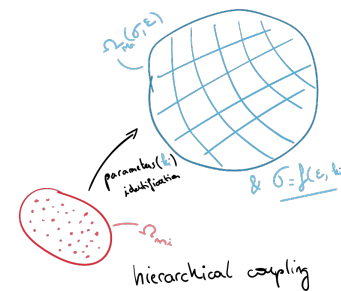
[Vassaux, et al. Cem. Concr. Compos. 70 \(2016\)](#)

[Vassaux, et al. Int. J. Numer. Anal. Methods Geomech. 39.7 \(2015\)](#)

[Vassaux, et al. Eng. Fract. Mech. 149 \(2015\)](#)

Hierarchical multiscaling

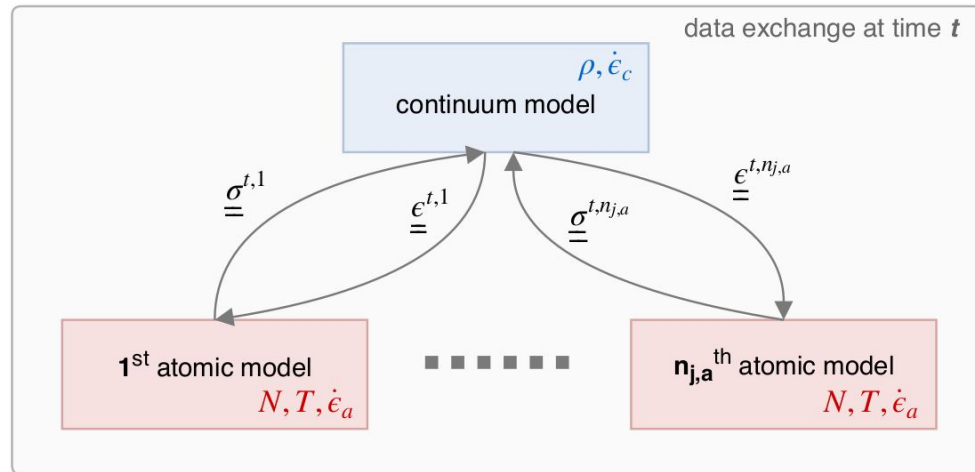
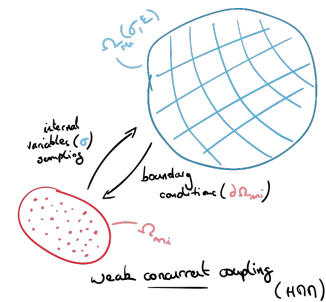
- continuum **constitutive law** parameterisation
- **coarse-graining** from all-atom molecular dynamics (IBI, STM, ...)



[Kempfer, et al. ACS Omega. 4 \(2019\)](#)

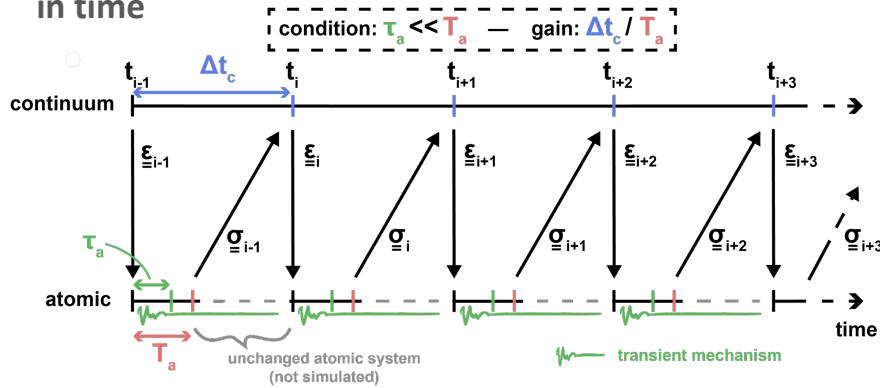
Heterogeneous multiscale workflow

- **weakly** or **semi-concurrent** coupling
 - **robustness** due to single-scale independent models
 - highly **scalable for HPC**
- **I/O** of the **atomistic** and **continuum** model simulations:
 - bottom-up **AND** top-down data exchange
- **no more parameterisation!**

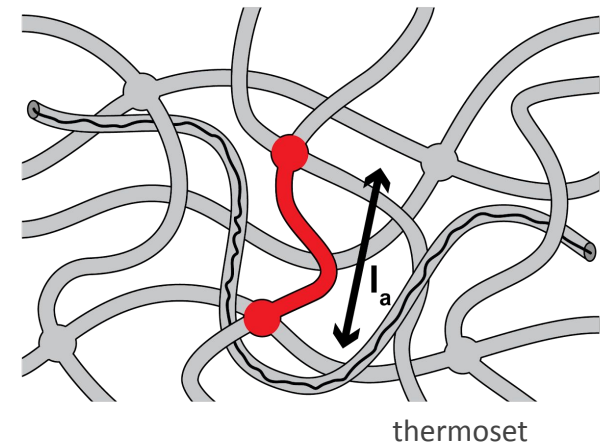
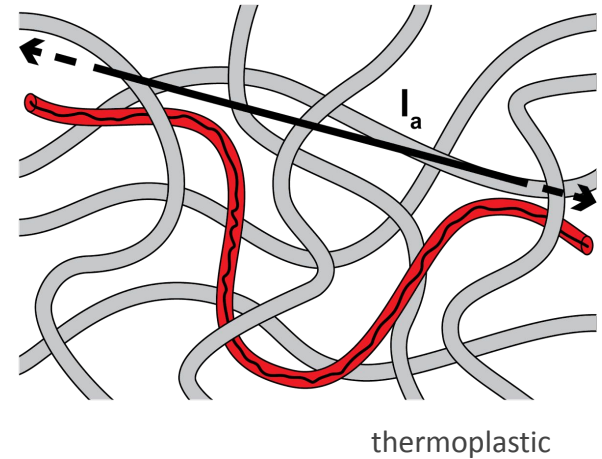
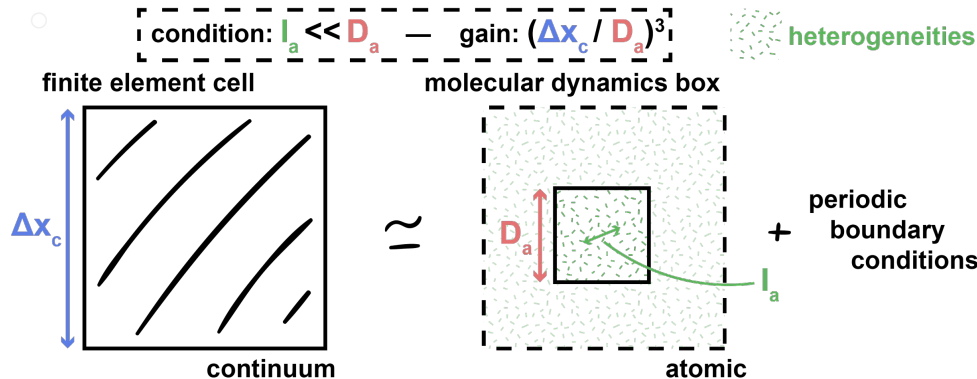


Scales separation and computational gain

- in time

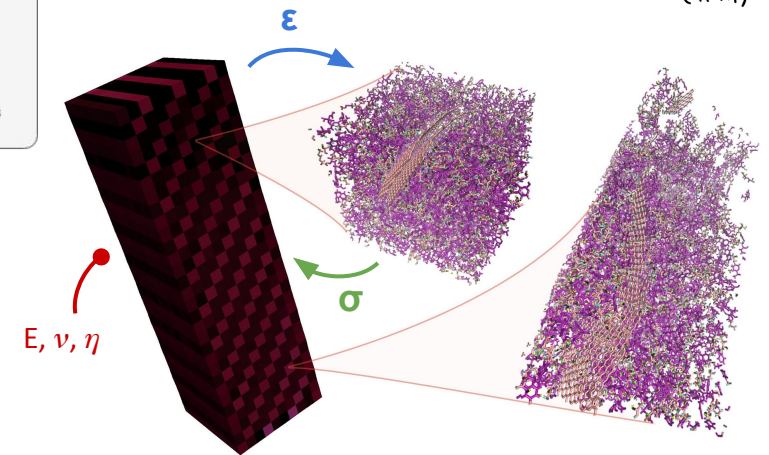
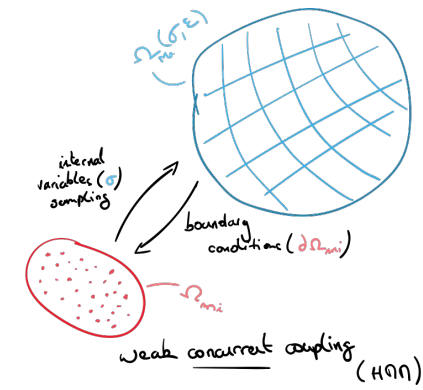
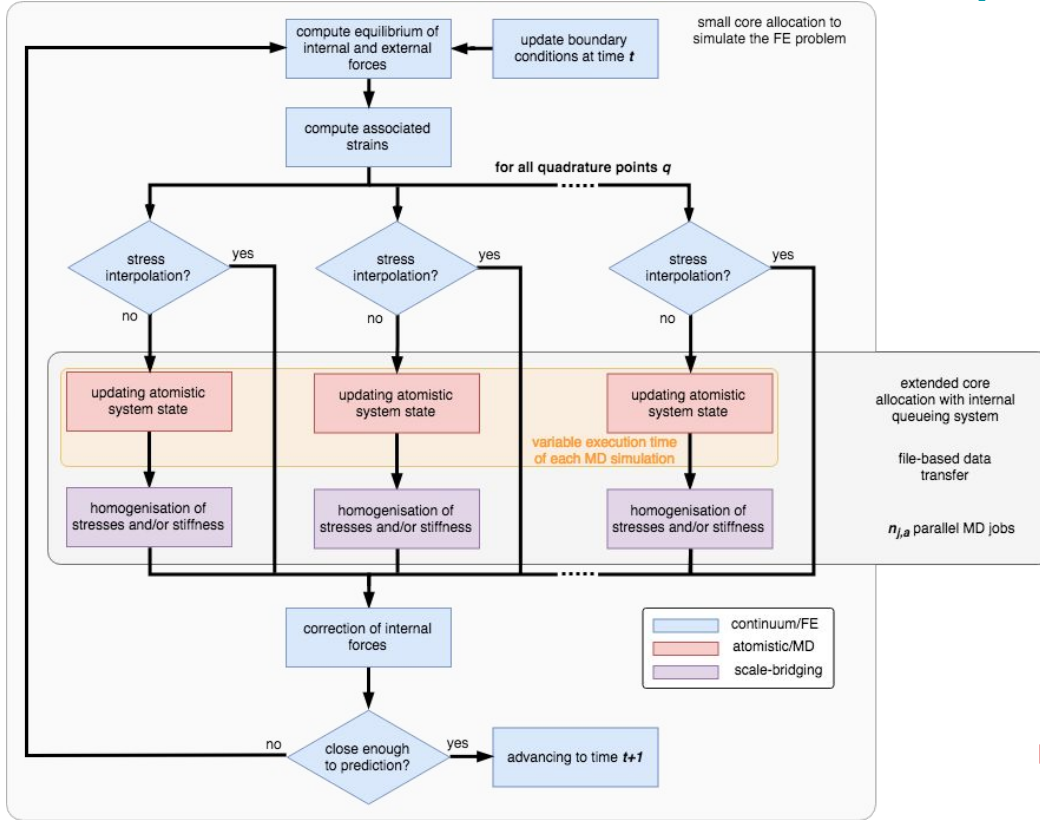


- in space



Heterogeneous multiscale workflow

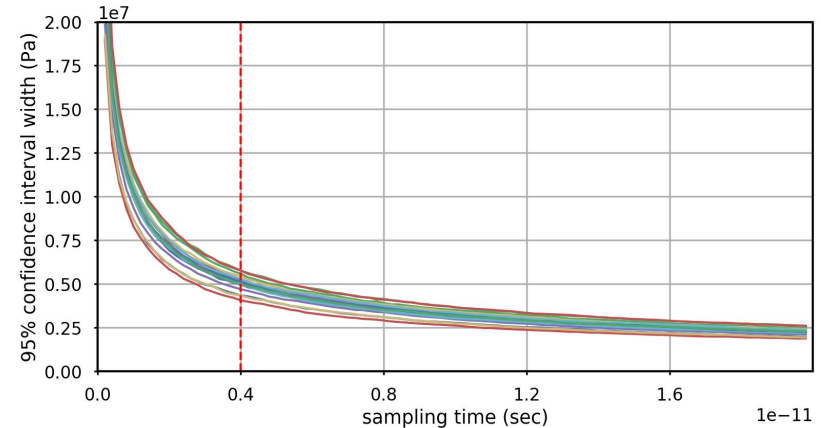
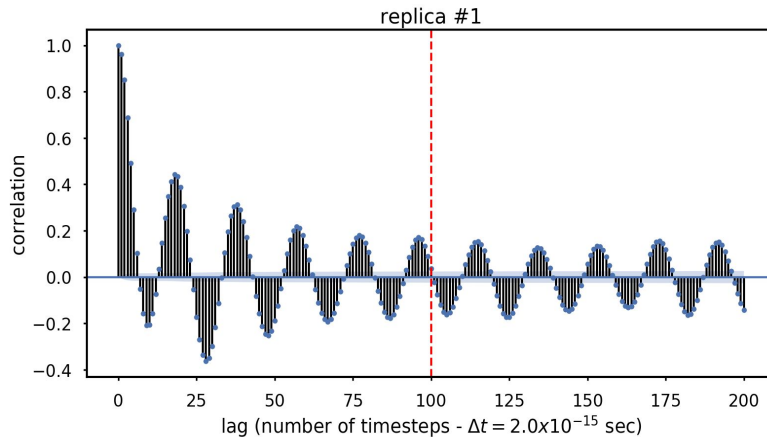
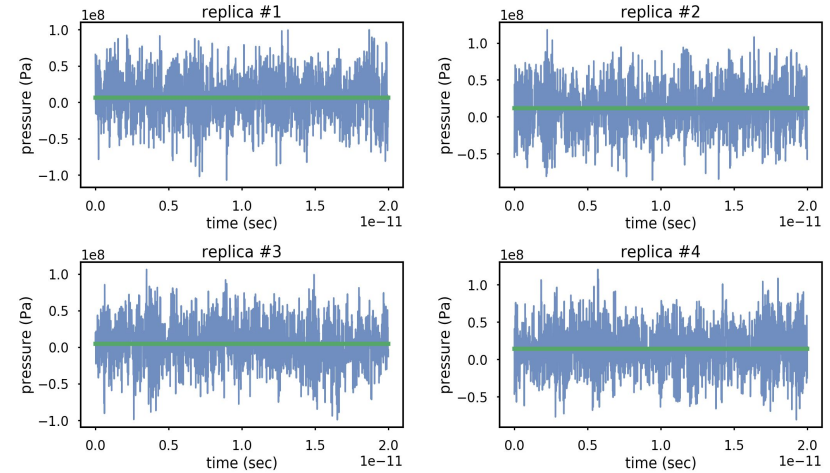
SCEMa (Simulation Coupling Environment for Materials):
<https://github.com/UCL-CCS/SCEMa>



Vassaux, Richardson and Coveney. *Philos. Trans. R. Soc. London, Ser. A.* (2019) 377.2142

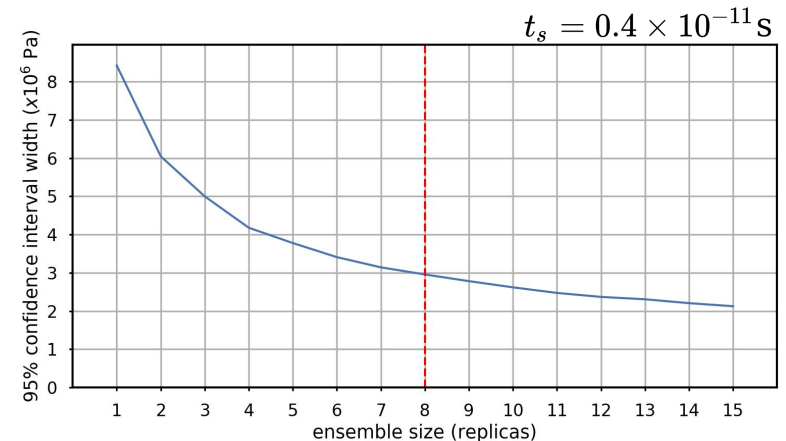
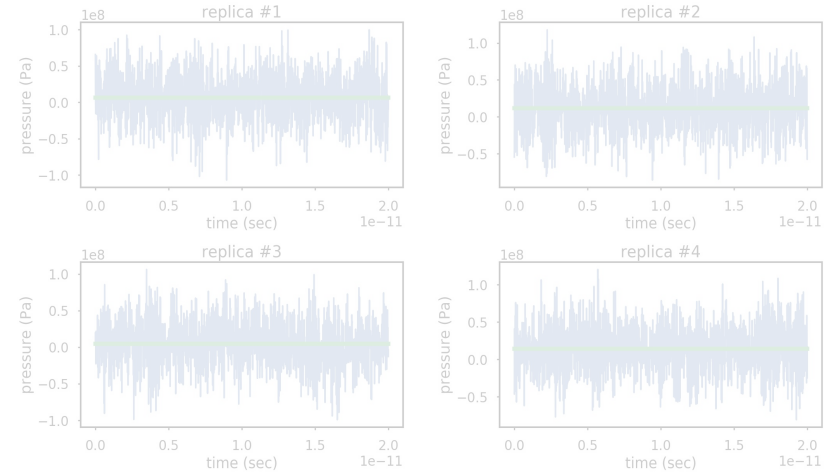
Data transfer between scales

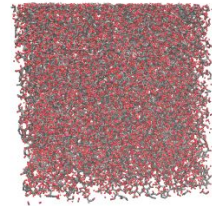
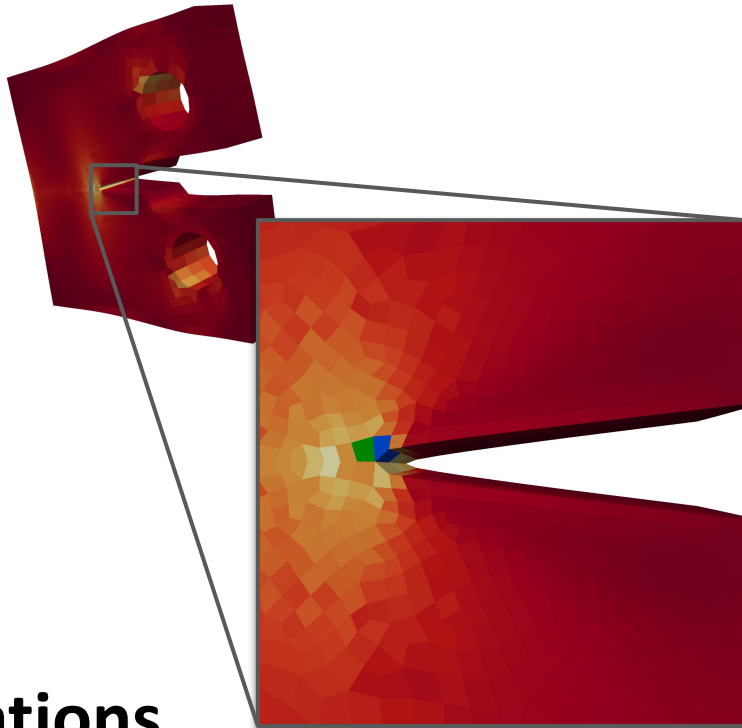
- **proper stress sampling**
 - how **long**? how many **replicas**?
 - target: optimal **uncertainty** to **cost** ratio
 - stabilisation of mean
 - reduced confidence interval
 - time sampling using **bootstrapping**
 - decorrelated subsamples?
- subsampling time \gg **autocorrelation** length



Data transfer between scales

- **proper stress sampling**
 - how **long**? how many **replicas**?
 - target: optimal **uncertainty** to **cost** ratio
 - stabilisation of mean
 - reduced confidence interval
 - time sampling using **bootstrapping**
 - decorrelated subsamples?
→ subsampling time \gg autocorrelation length
 - ensemble sampling using **bootstrapping**
 - independent replicas of the same material
 - similar “global” properties
 - e.g mass, cross-link density
 - **same building process**

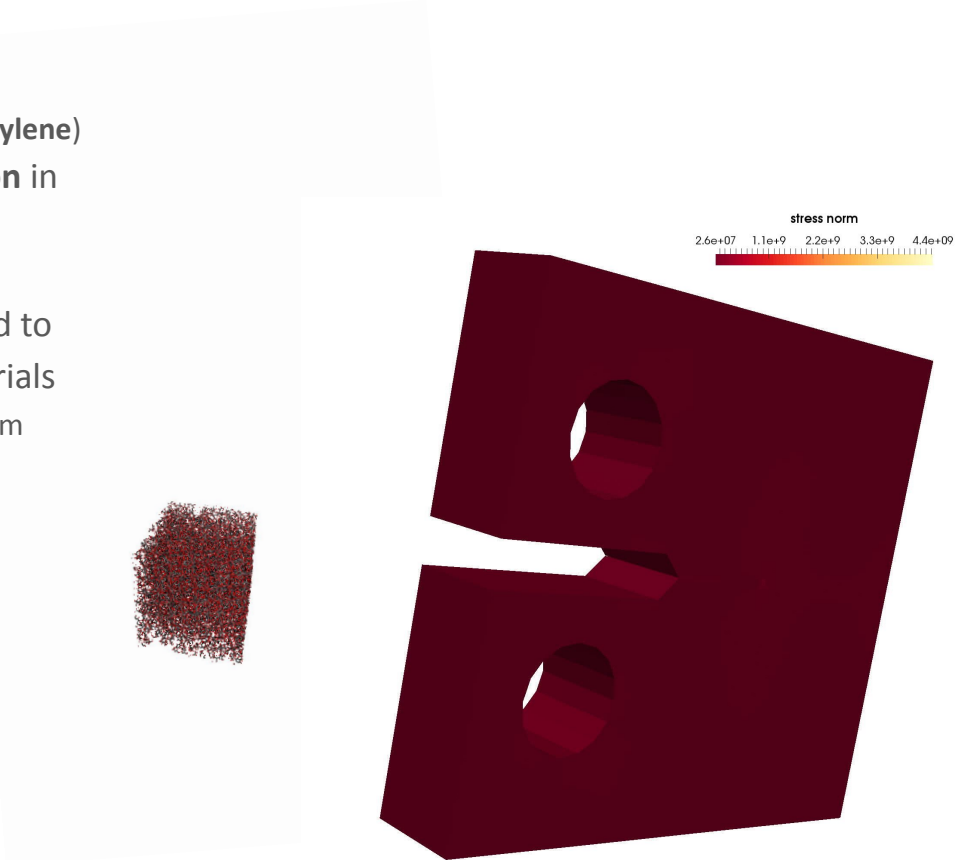
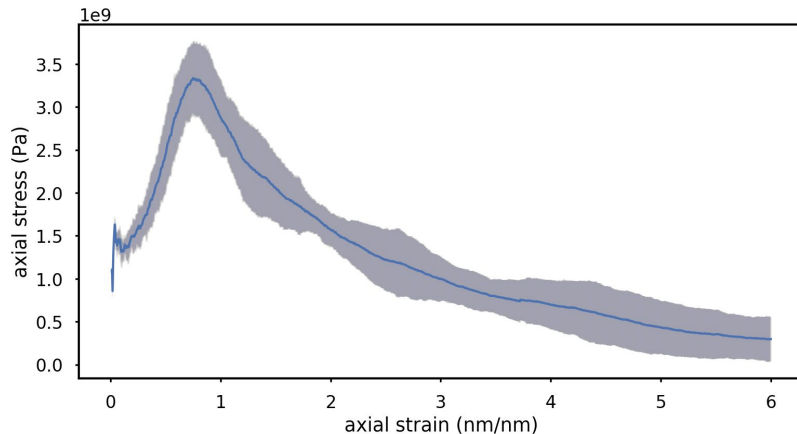




**Applications
to polymers and nanocomposites**

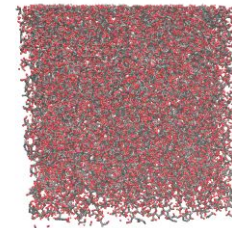
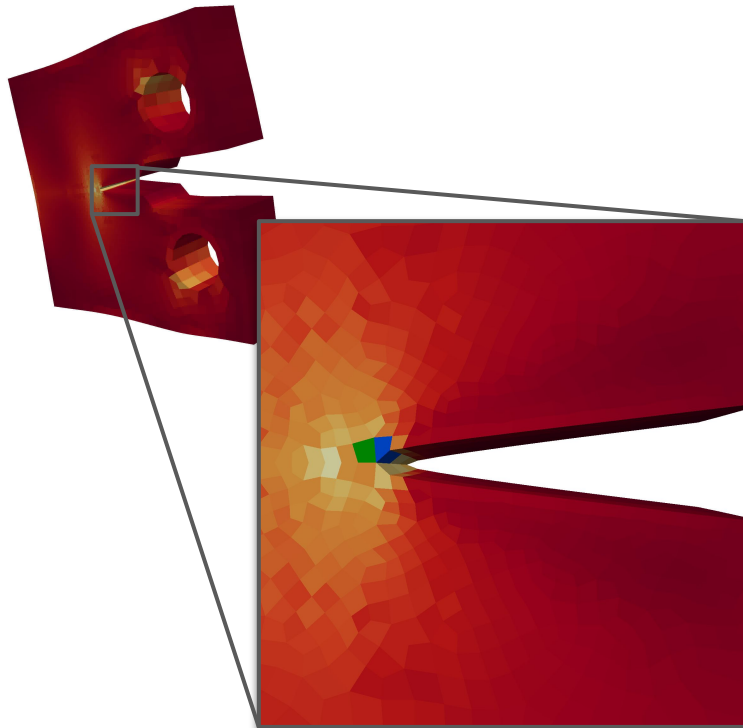
Fracture toughness of an epoxy resin

- assess fracture toughness prediction
 - from chemical structure using MD+OPLS (polyethylene)
- single-scale approaches assume a **uniaxial tension** in the region of interest (where strain localizes)
 - measure dissipated energy at complete failure
- standardized “compact-tension” test (ASTM) used to derive the **fracture toughness** of structural materials
 - high strain **localization** at the tip of **notched** system



Fracture toughness of an epoxy resin

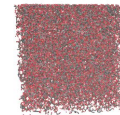
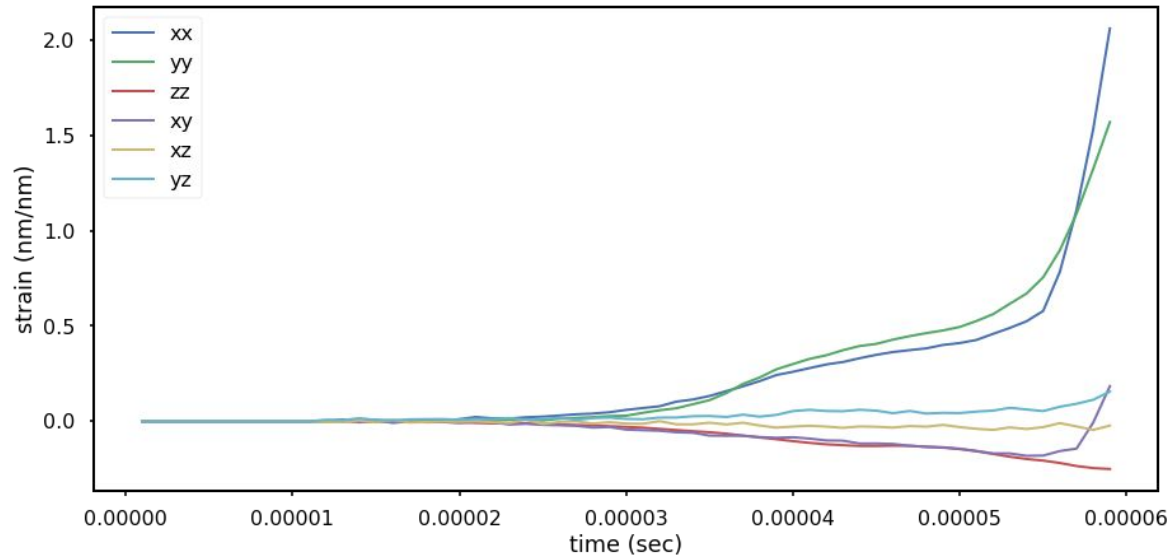
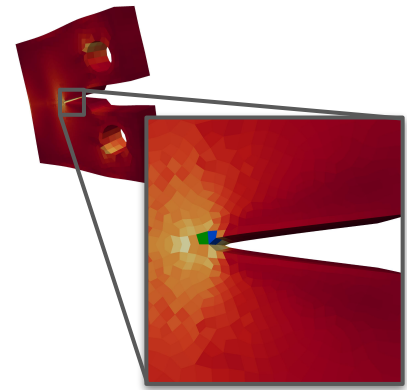
- what is the actual mechanical state at the crack-tip?
 - focus on the cells in the fracture process zone



“constrained” cell

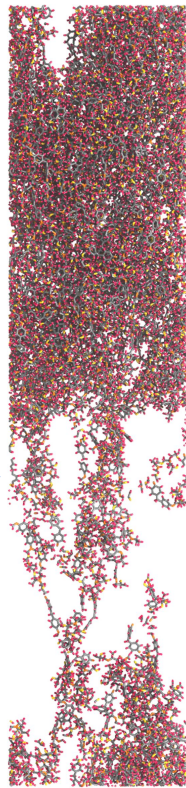
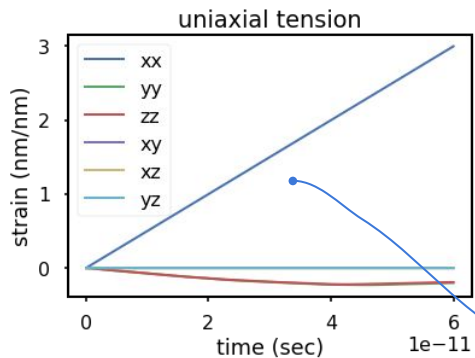
Fracture toughness of an epoxy resin

- what is the actual mechanical state at the crack-tip?
 - focus on the cells in the fracture process zone
- highly complex (**3D, nonlinear**) mechanical (stress/strain) state
 - bi-axial tension + shear

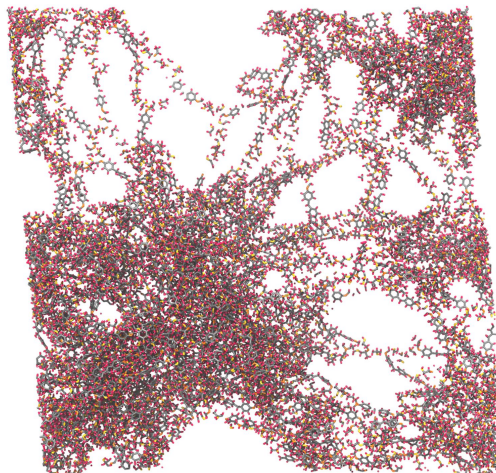


“constrained” cell

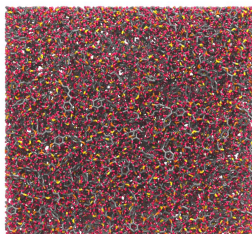
Fracture toughness of an epoxy resin



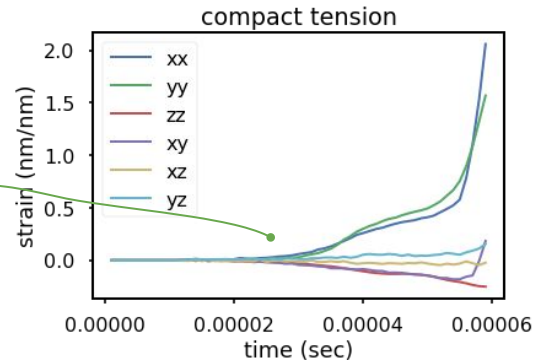
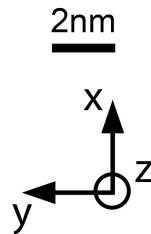
uniaxial tension



compact tension

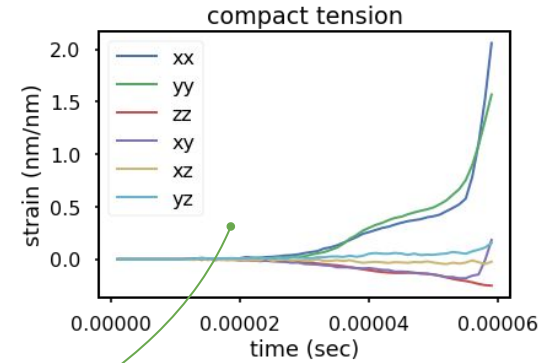
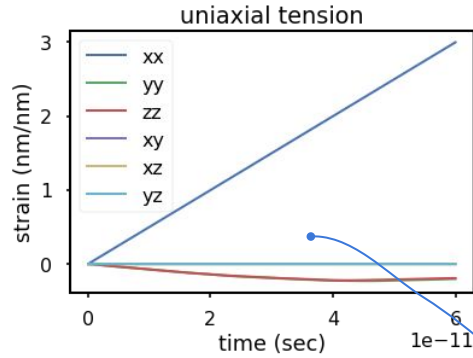


equilibrium state

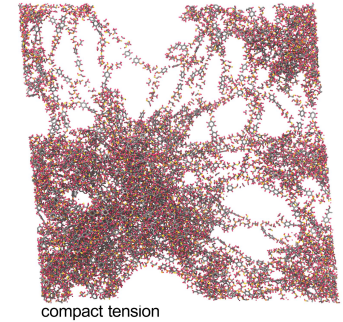
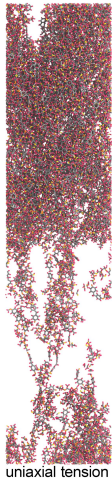
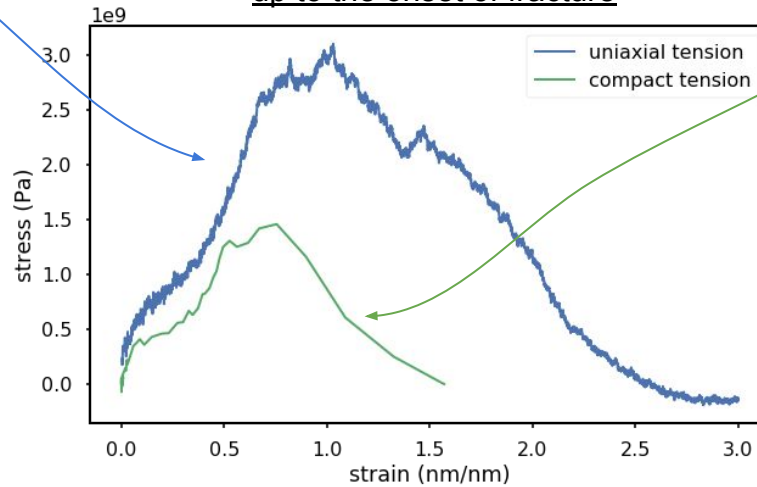


final local state of the epoxy resin at the onset of fracture

Fracture toughness of an epoxy resin

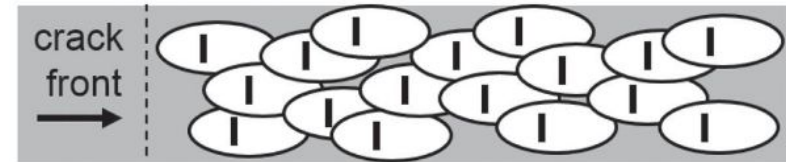
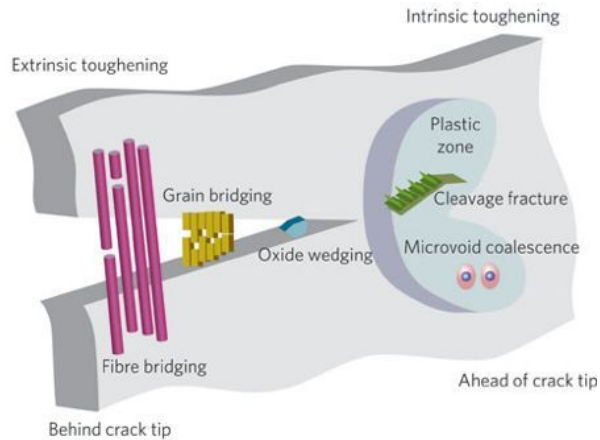


macroscopic behaviour of the epoxy resin up to the onset of fracture



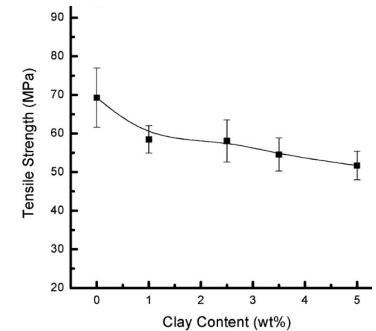
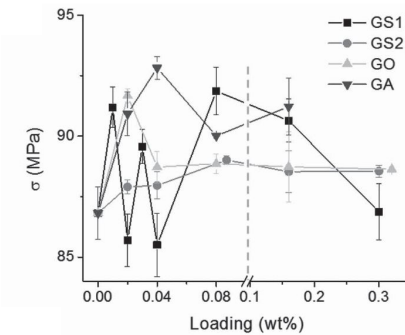
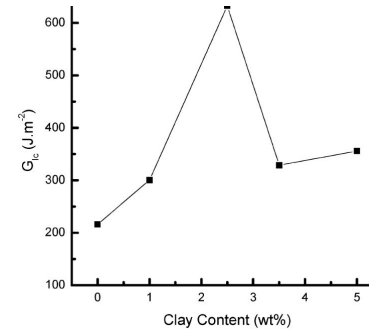
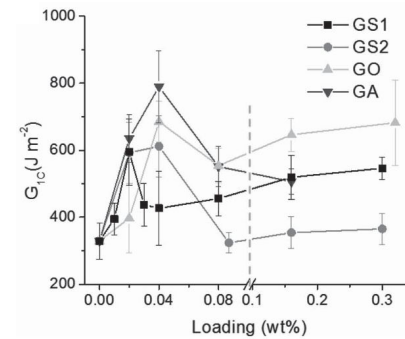
Enhancing epoxy resins with graphene nanoparticles

- two possible mechanisms
 1. crack deflection, pinning, bridging
 2. microcrack dispersion



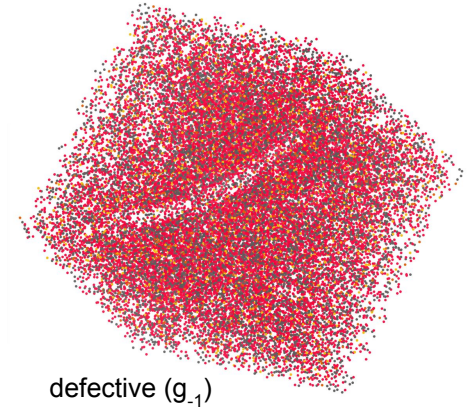
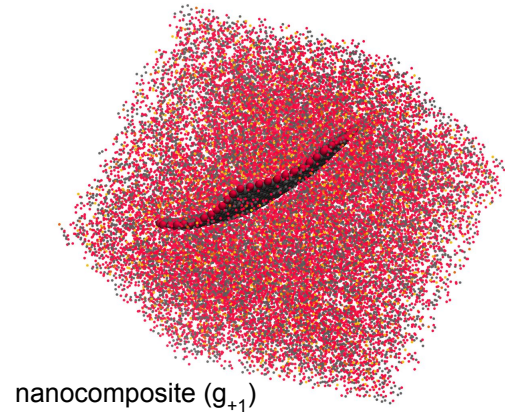
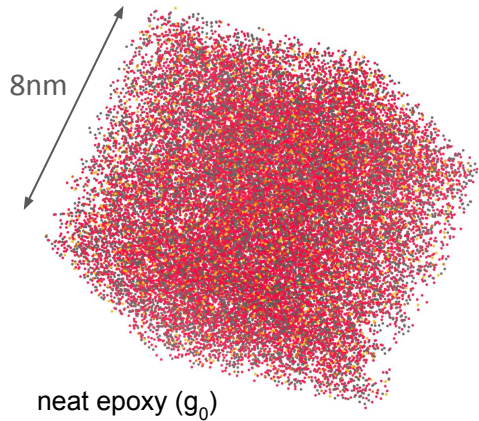
Enhancing epoxy resins with graphene nanoparticles

- two possible mechanisms
 1. crack deflection, pinning, bridging
 2. microcrack dispersion
 - experimentally (for a wide range of %wt):
 - fracture energy ↗
 - strength ↗
- ➔ mostly **unclear**, but may favor *microcrack* theory



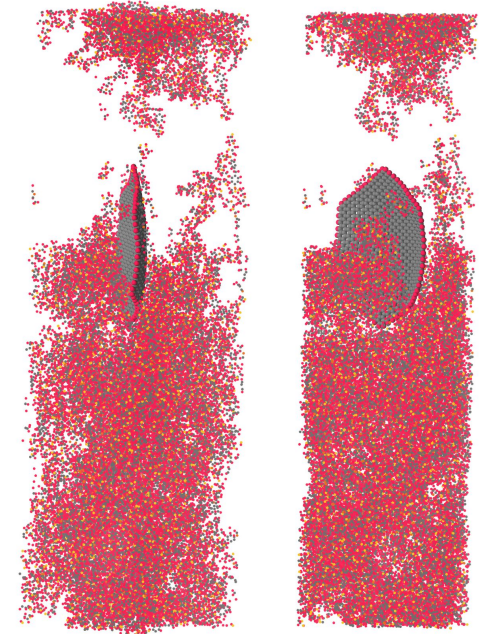
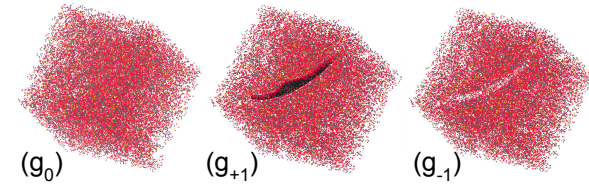
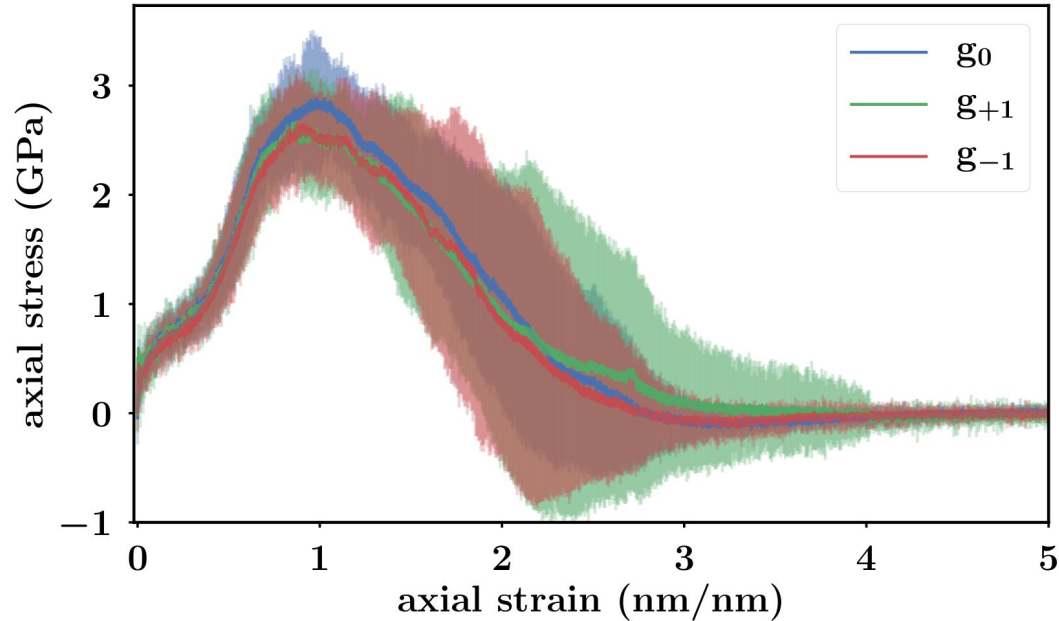
Atomistic modelling of graphene-epoxy nanocomposites

- molecular dynamics (LAMMPS) with ReaxFF (force field) simulations
- 92% crosslinked epoxy resins and 6nm-wide hexagonal graphene flakes
 - **defective**: graphene **present during polymerisation**, then removed



Atomistic modelling of graphene-epoxy nanocomposites

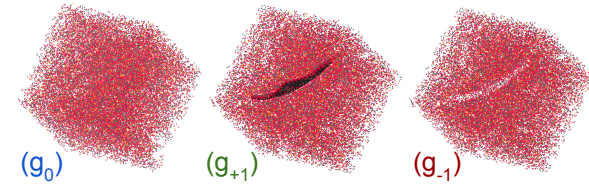
- molecular dynamics (LAMMPS [1]) with ReaxFF (force field [2]) simulations
- 92% crosslinked epoxy resins and 6nm-wide hexagonal graphene flakes



nanocomposite (g_{+1}) at 200% strain

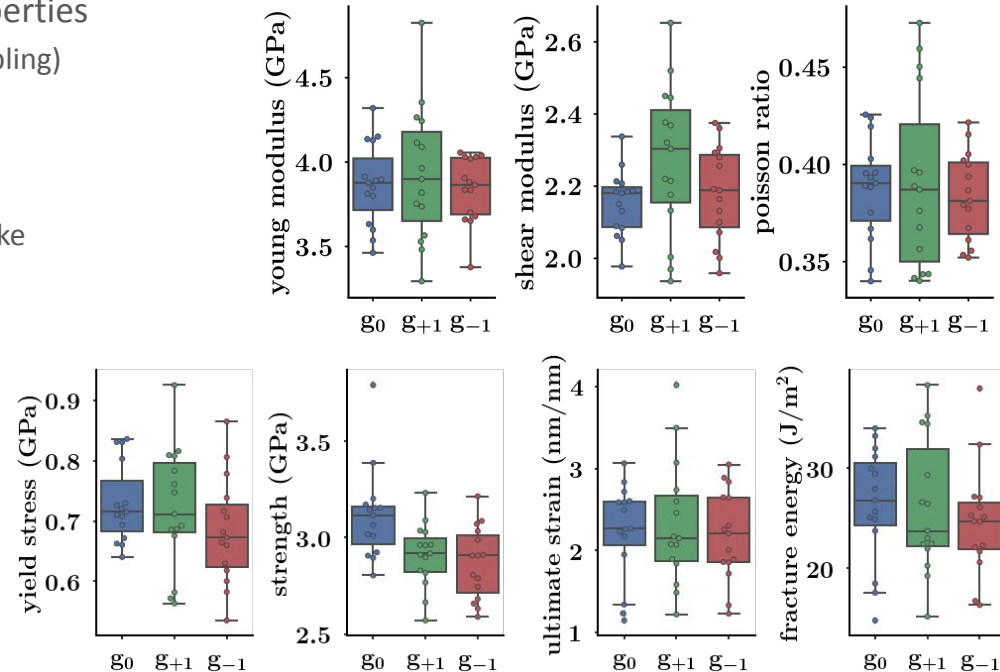
Atomistic modelling of graphene-epoxy nanocomposites

- molecular dynamics (LAMMPS [1]) with ReaxFF (force field [2]) simulations
- 92% crosslinked epoxy resins and 6nm-wide hexagonal graphene flakes



* analyse of the elastic, plastic and fracture properties

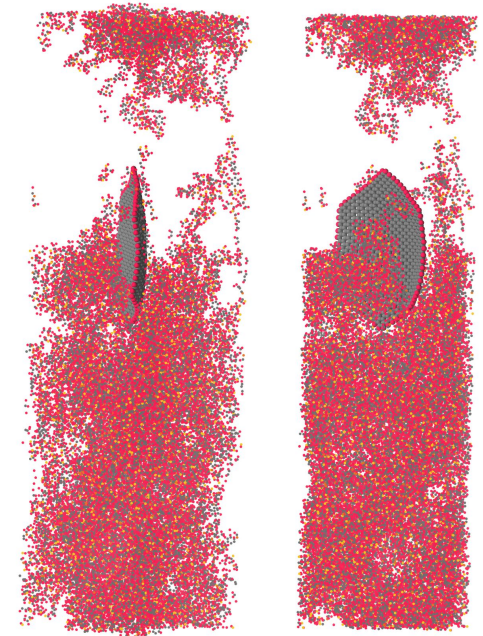
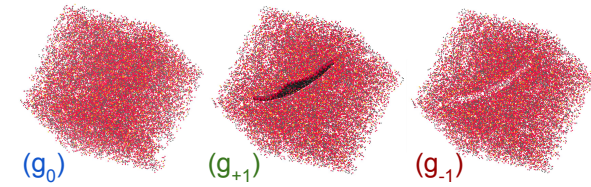
- increased variability (ensemble+direction sampling)
 → increased anisotropy
- **increased shear modulus**
- reduced strength
- hardly any **active** influence of the graphene flake



Atomistic modelling of graphene-epoxy nanocomposites

- molecular dynamics (LAMMPS [1]) with ReaxFF (force field [2]) simulations
- 92% crosslinked epoxy resins and 6nm-wide hexagonal graphene flakes

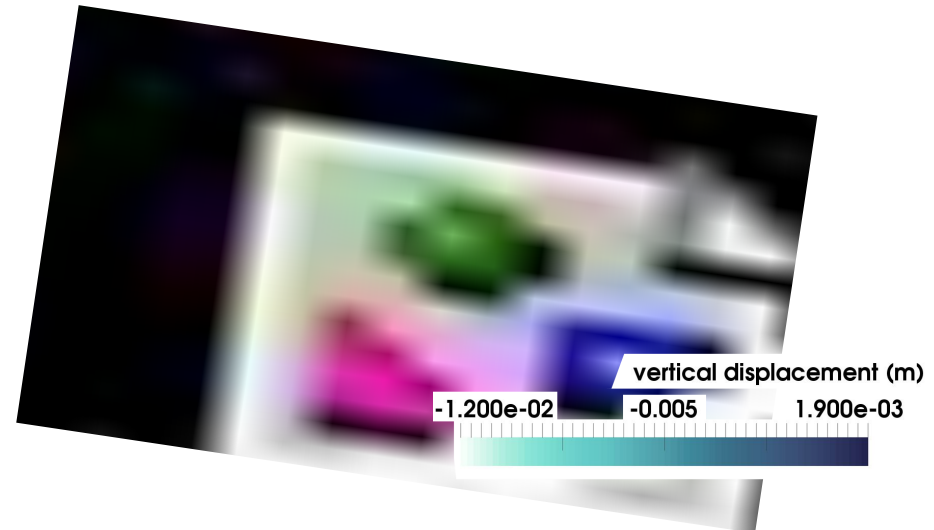
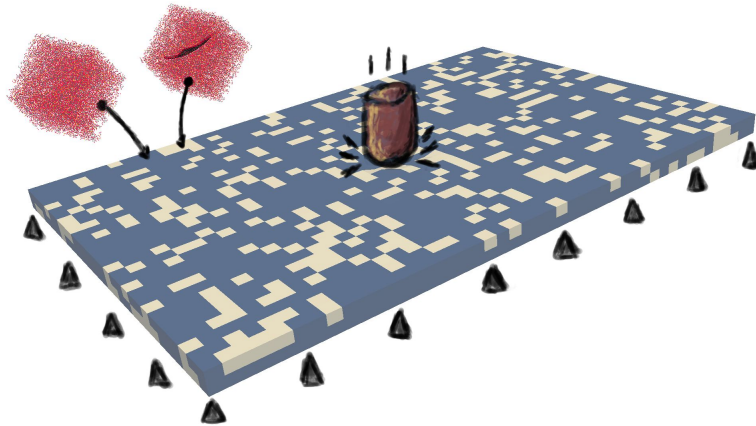
- * analyse of the elastic, plastic and fracture properties
 - increased variability (ensemble+direction sampling)
 - ⇒ increased anisotropy
 - increased shear modulus
 - reduced strength
 - small **active** influence of the graphene flake
- * isolated at such small scale, the flake is mostly an engineered defect
 - ↳ favors *microcrack* theory



nanocomposite (g_{+1}) at 200% strain

Multiscale modelling of graphene-epoxy nanocomposites

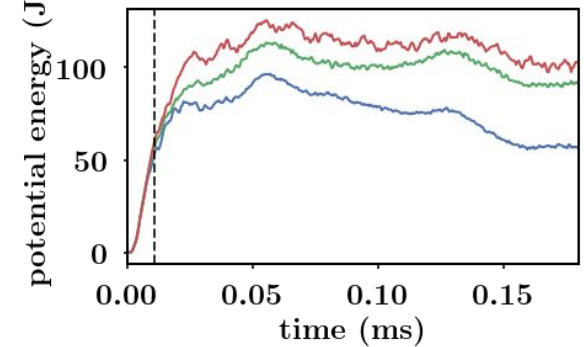
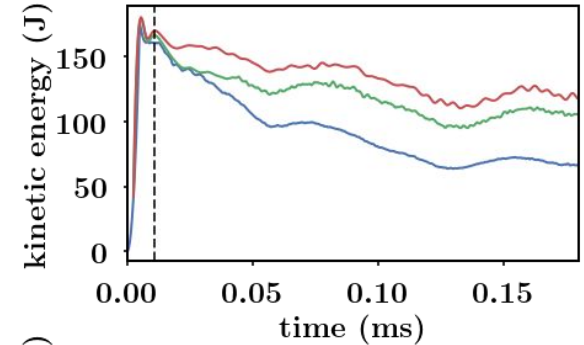
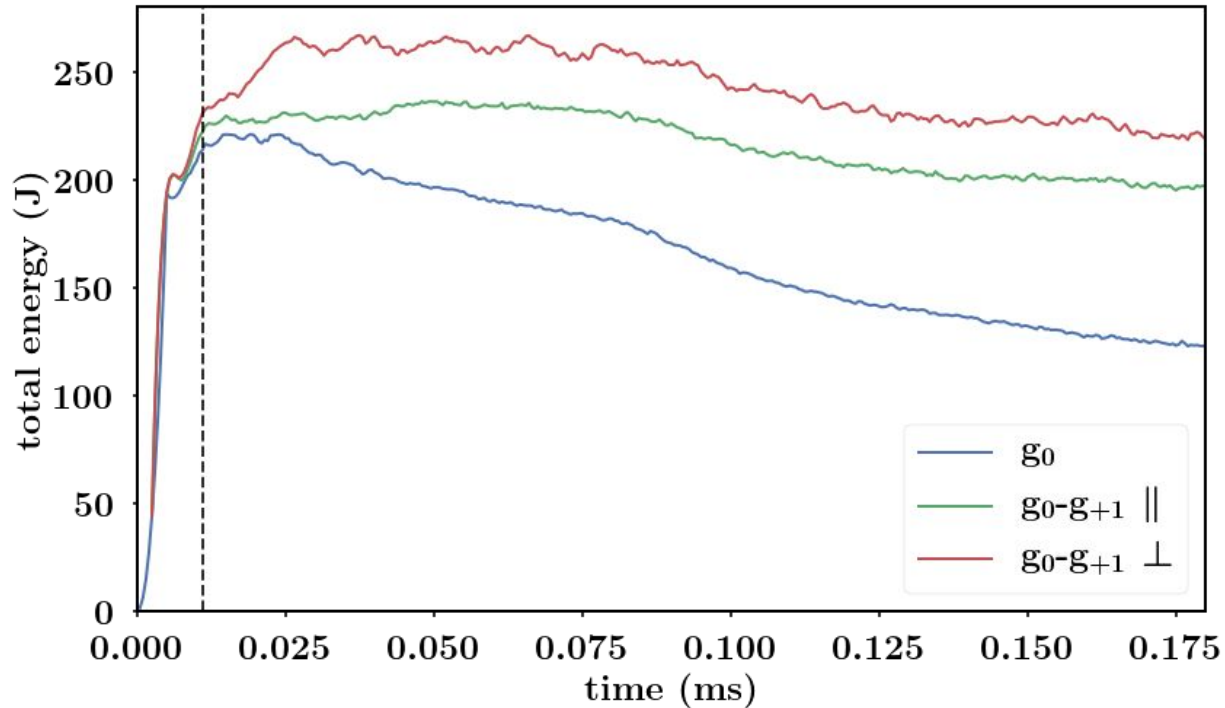
- heterogeneous multiscale model:
continuum (FE/deal.ii), atomistic (MD/LAMMPS)
 - combination of large number of atomistic systems via a dynamic continuum model
- high-velocity impact ($400\text{m}\cdot\text{s}^{-1}$) on shell of nanocomposite (1% wt)
 - 150mm×100mm×5mm shell oscillating for 0.2ms
 - **350000** (2700×500) **MD** simulations
 - FE simulation on 16 cores, 1 MD simulation on 64 cores
 - computational cost: **120,000 CPUhs**



[Bangerth, Hartmann, and Kanschä. ACM Trans. Math. Software.\(2007\) 33.4](#)
[Plimpton. J. Comput. Phys. \(1995\) 117.1](#)

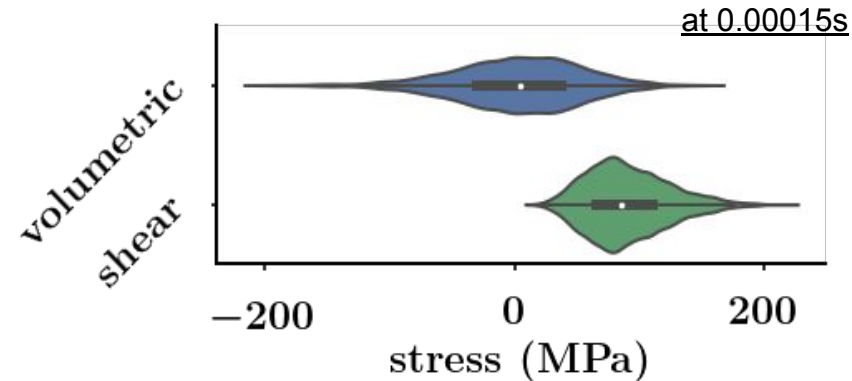
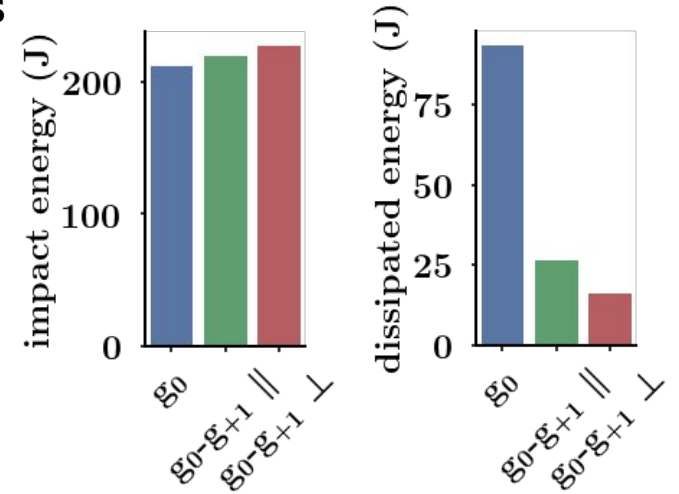
Multiscale modelling of graphene-epoxy nanocomposites

- evolution of the **shell energy** content during the impact



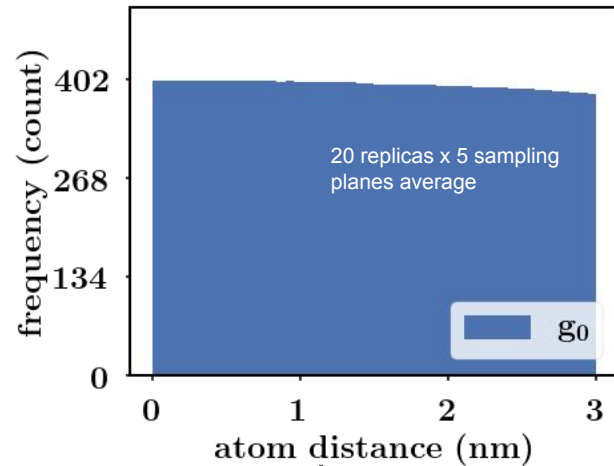
Multiscale modelling of graphene-epoxy nanocomposites

- evolution of the **shell energy** content during the impact
 - major reduction (↘ 70%) of energy dissipation by adding graphene nanoparticles
- analyse of local mechanical state
 - moderate stress amplitude
 - **no significant void growth**, or fracture via **bond dissociation**
 - dominance of **shear** over **volumetric** stresses
 - **non-existent force transfer** between polymer and graphene
 - **relaxation** of the flake
 - graphene **potential energy** ↘ 1% (relatively to total atomistic system pot. eng.)

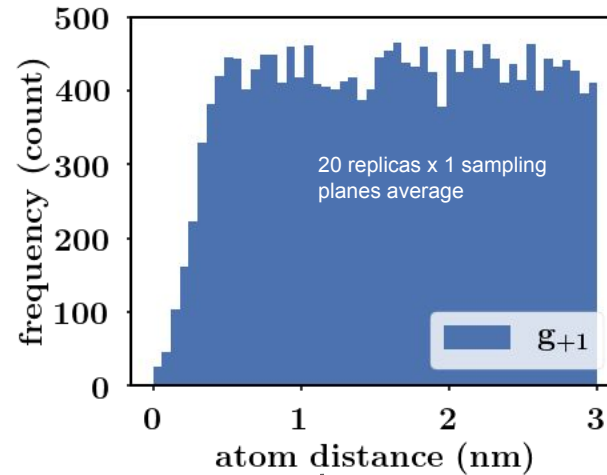


Multiscale modelling of graphene-epoxy nanocomposites

- what are suitable **candidate interaction mechanisms** between graphene and epoxy resin?
 - explaining the **absence of force transfer** and the **reduced friction/conformational changes**
 - ? **passive mechanism** e.g. polymer **network disruption** during thermosetting (curing)
 - ? **active mechanism** e.g. **geometric constraint** limiting polymer mobility (reptation)
- density profile of carbon atoms of the polymer network in the atomistic systems
 - **no local or global densification**



→ random sampling direction

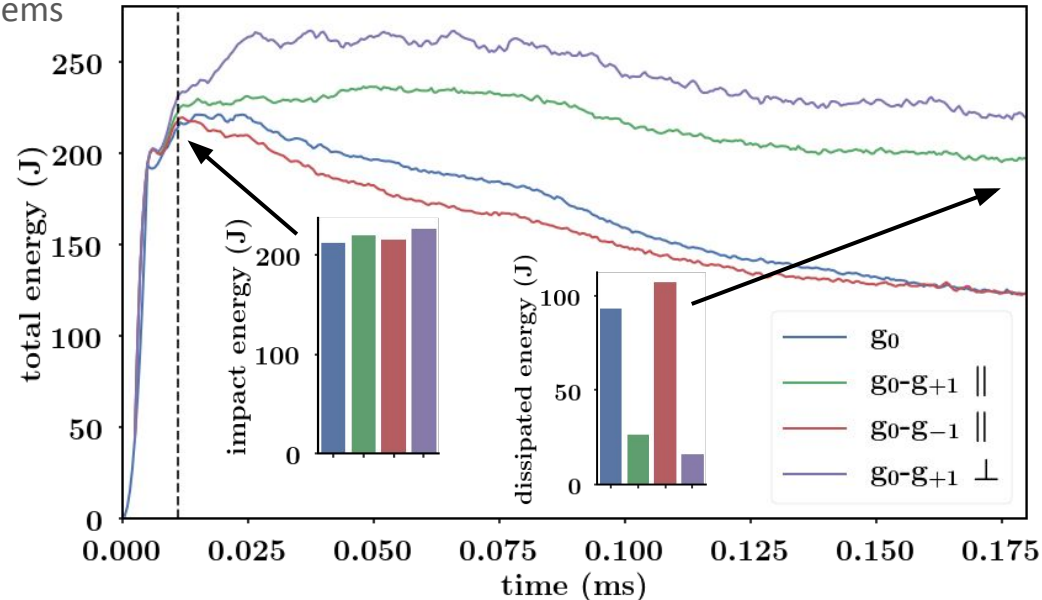


→ (to orthogonal direction to graphene plane)

[Vassaux, et al. Adv. Theor. Simul. \(2019\) 2.5](#)

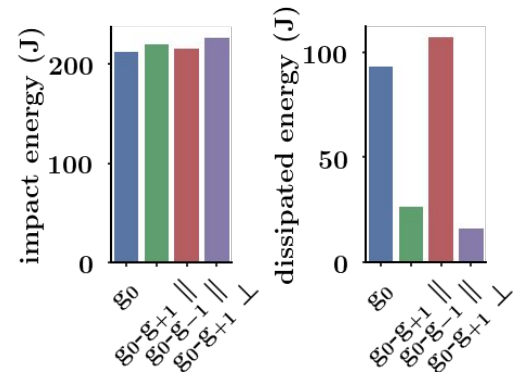
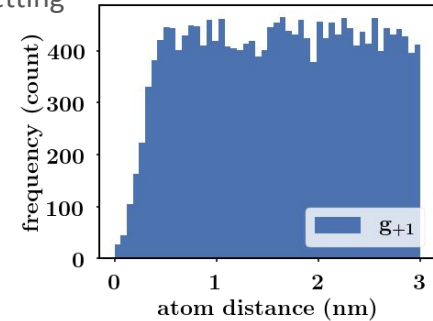
Multiscale modelling of graphene-epoxy nanocomposites

- what are suitable **candidate interaction mechanisms** between graphene and epoxy resin?
 - ⇒ explaining the **absence of force transfer** and the **reduced friction/conformational changes**
 - ? **passive mechanism** e.g. polymer **network disruption** during thermosetting (curing)
 - ? **active mechanism** e.g. **geometric constraint** limiting polymer mobility (reptation)
- density profile of carbon in the atomistic systems
 - ⇒ **no local or global densification**
- simulation using “**defective**” (g_{-1}) systems
 - ⇒ **loss of reduction** of energy dissipation



Multiscale modelling of graphene-epoxy nanocomposites

- what are suitable **candidate interaction mechanisms** between graphene and epoxy resin?
 - ⇒ explaining the **absence of force transfer** and the **reduced friction/conformational changes**
 - ? **passive mechanism** e.g. polymer **network disruption** during thermosetting
 - ? **active mechanism** e.g. **geometric constraint** limiting polymer mobility
 - density profile of carbon in the atomistic systems
 - ⇒ **no local or global densification**
 - simulation using “**defective**” (g_{-1}) systems
 - ⇒ **loss of reduction** of energy dissipation
- ➔ favors the **geometric constraint** hypothesis.
- at low-amplitude
 - with volume-constant/shear deformation (nanoconfinement)



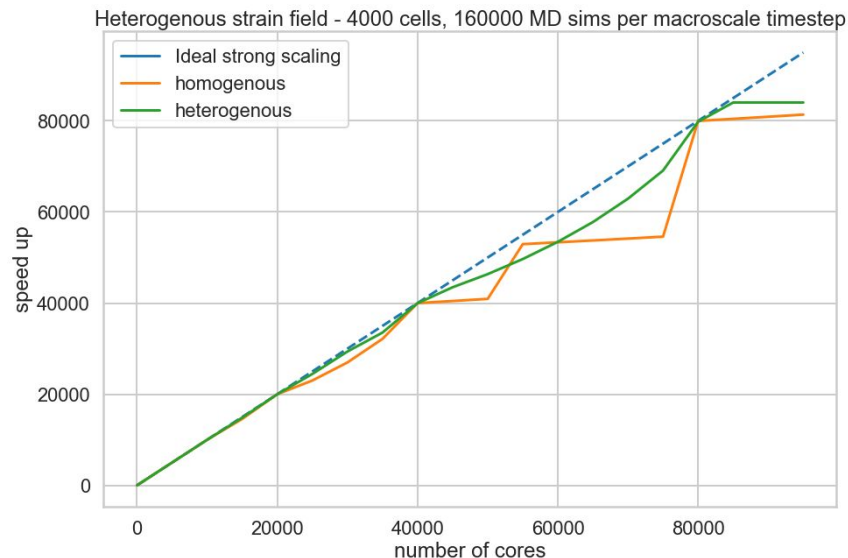
High-performance computing and scalability of multiscale simulations





Benchmarking and scalability

- benchmark and optimisation
 - POP-CoE (Brian Wylie, FZ Juelich)
 - potential for **exascale** application
- a concurrent model coupling asynchronously
 - **embarrassingly parallel** application
- computational costs
 - 0.1 core hour per microscale simulation
 - 10,000 cells x 8qps/cell x 5 replicas/qp
 - 400,000 independent microscale simulations per macroscale simulation timestep

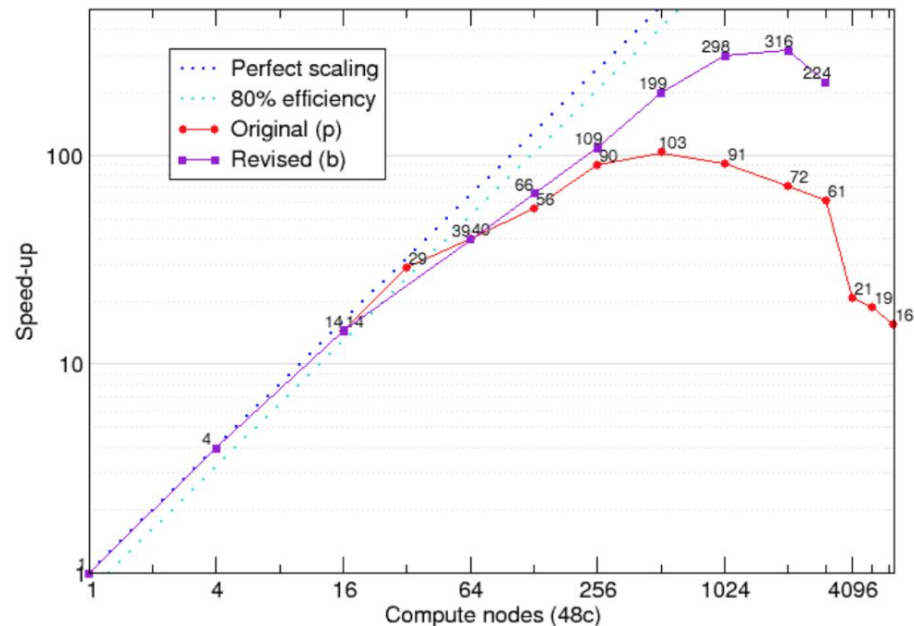
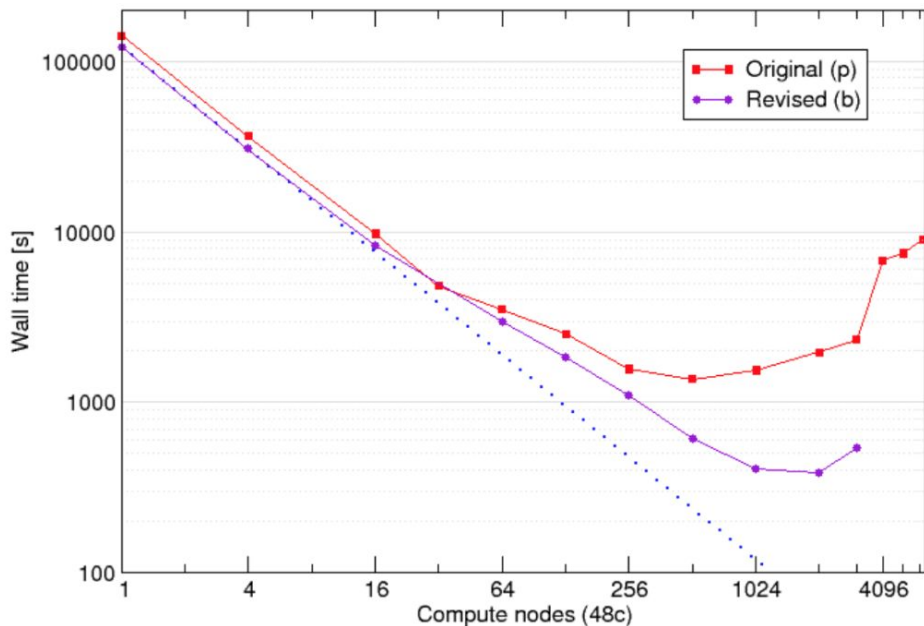


theoretical speed-up w.r.t
single core performance



Benchmarking and scalability

- **Original version** ran successfully up to 6400 nodes, but **peak speed-up 512** compute nodes
- **Revised version** executes faster, and continues to **scale well to 1024** (peak at 2048) compute nodes
 - 300x speed-up for 1024 nodes compared to a single compute node (previous best 100x)



Our current infrastructure *playground*:

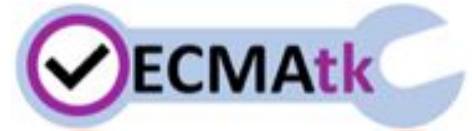
- Tier-0 and Tier-1 supercomputers
 - Summit (ORNL) - #2*
 - Frontera (TACC) - #8
 - Marconi-100 (CINECA) - #9
 - SuperMUC-NG (LRZ) - #13
 - MareNostrum (BSC) - #37
 - Scafell Pike (STFC) - #247
 - Archer (EPCC) - #386
 - Eagle (PSNC)
 - Kathleen (UCL, RITS)



*June 2020 Top 500 rankings

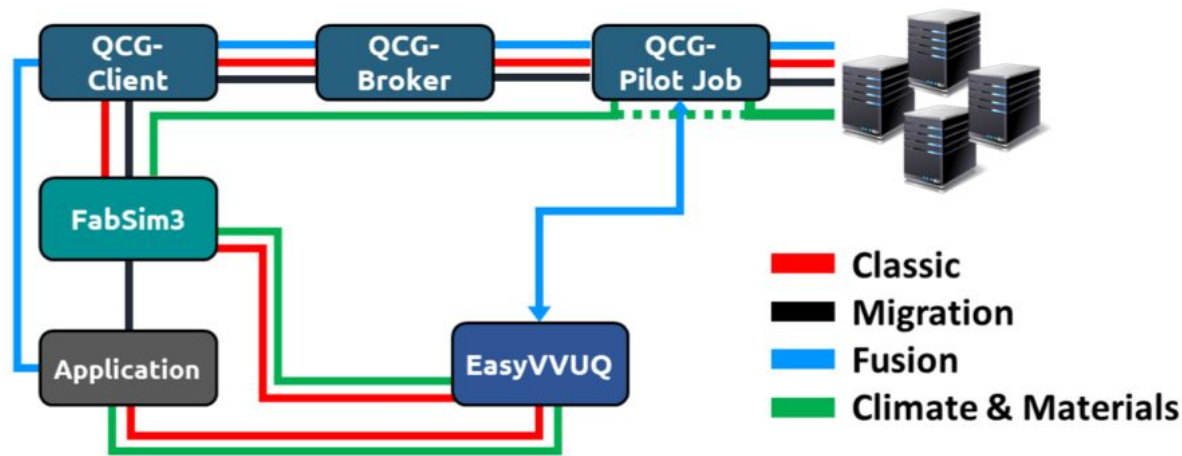
Uncertainty quantification: VECMAtk — a generic toolkit for (VV)UQ

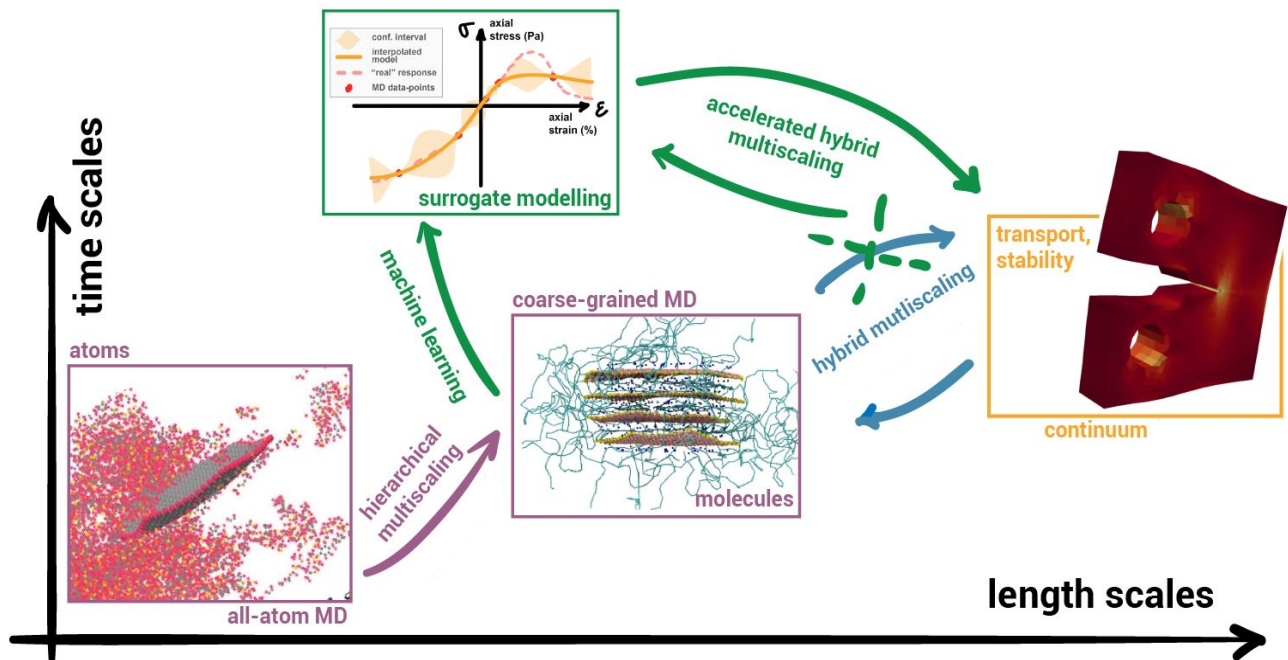
- **UQ** is essential for **actionable simulations** and **requires HPC**
- **Objective:** develop a **VVUQ** toolkit for potential exascale **multiscale** applications (VECMa)
 - identified UQ patterns
 - fully automated generation, management, and execution of **UQ campaigns on Supercomputers**



VECMa: <https://www.vecma.eu/>

VECMa Toolkit: <https://www.vecma-toolkit.eu/>

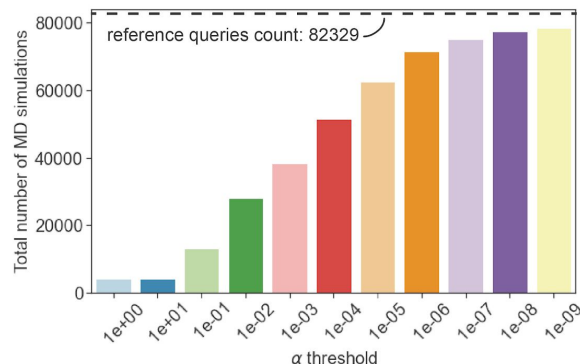
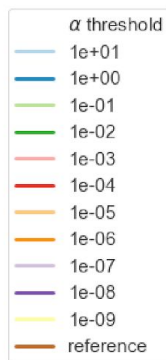
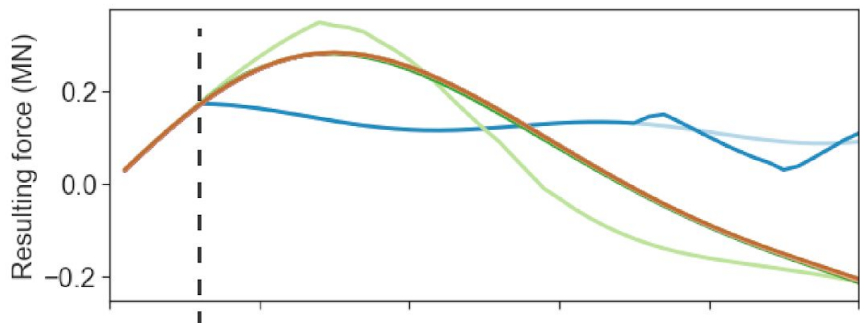
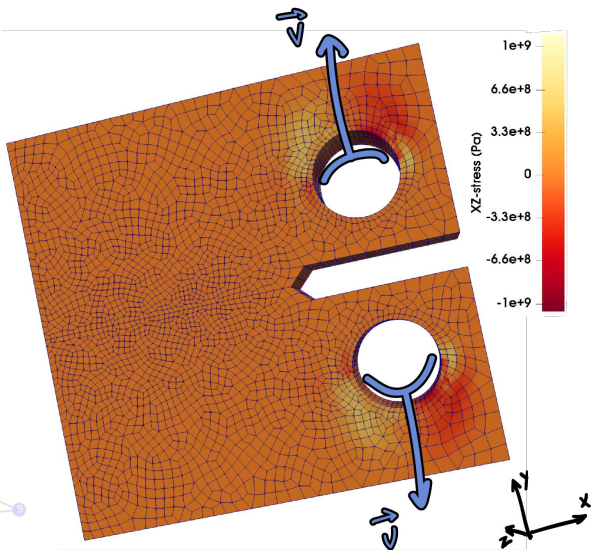
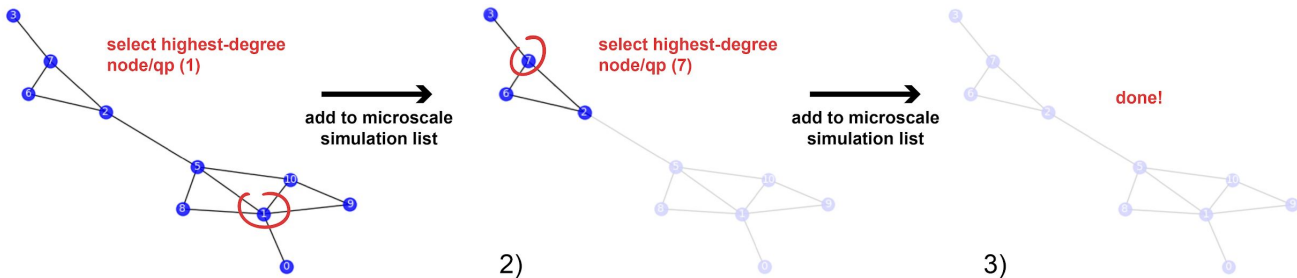




**Perspective:
 Automated parameterisation**

Highly redundant simulations

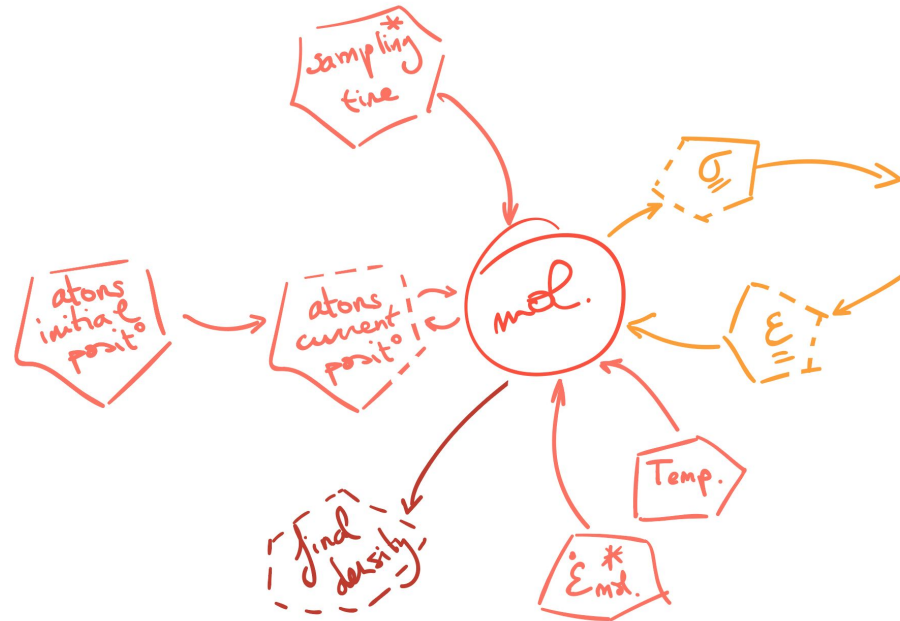
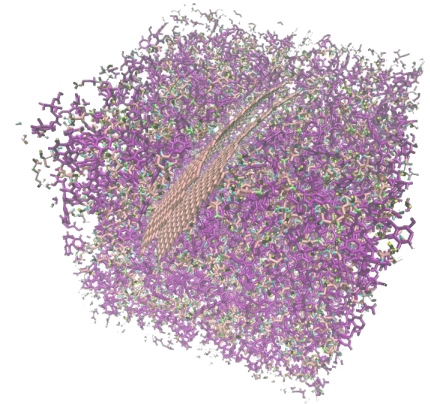
- acceleration of multiscale simulations using graph-based clustering
- compute similarity of strain histories
- graph-based clustering: **3-fold reduction** MD simulations count!



[Vassaux, et al. Adv. Theor. Simul. \(2020\) 4.2](#)

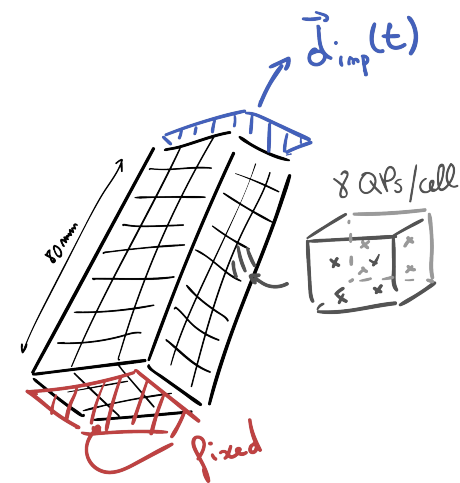
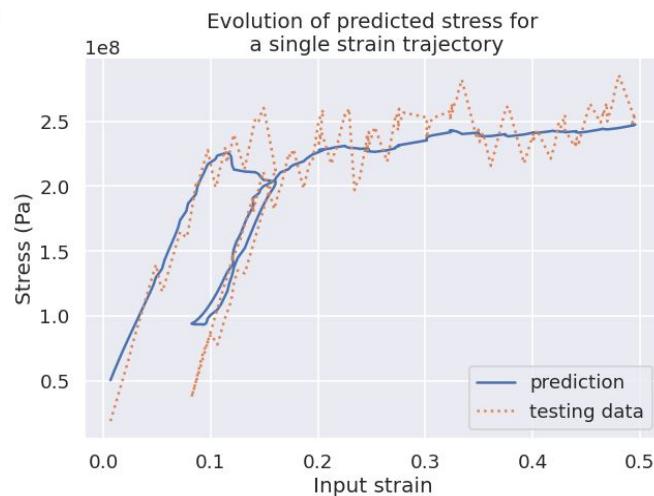
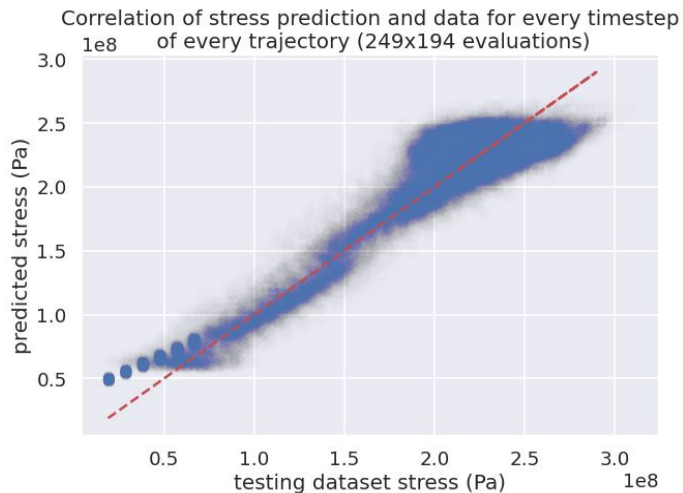
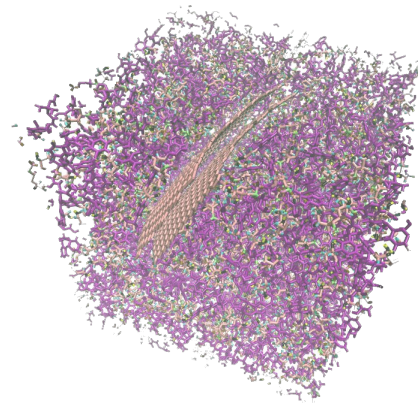
Surrogate modelling of molecular dynamics

- remove parameterisation **bias**
- **surrogate model** from an Artificial Neural Network
- reduce I/O of the molecular model to the 4 tensors (3x3)
 - inputs
 - current strain
 - previous stress
 - previous strain
 - outputs
 - current stress



Surrogate modelling of molecular dynamics

- surrogate model from an Artificial Neural Network
- reduce I/O of the molecular model to the 4 tensors (3x3)
 - **1000x speed-up** of stress query



Conclusions

- implementation of **parameterisation-free multiscaling**
- application of multiscale modelling
 - **fracture** and **viscoelasticity** of **polymer** nanocomposites
 - hydration and dynamics of **self-assembled proteins**
- **highly-scalable** method suited for **HPC**
- reduced-order modelling and model-form UQ for online learning of **unbiased parameterisation**

- Special thanks
 - Robert C. Sinclair (former UCL PhD)
 - Robin A. Richardson (former UCL PDRA, now Netherlands eScience Center)
 - Peter V. Coveney (Dept. Chem., UCL)
 - Wouter Edeling (Centrum Wiskunde & Informatica, Amsterdam)
 - Franck Artzner (IPR, CNRS)
 - Aziz Ghoufi (IPR, UR1)

