

Molecular Dynamics Simulation of Fracture in Quartz

Part I: Introduction and Briefing

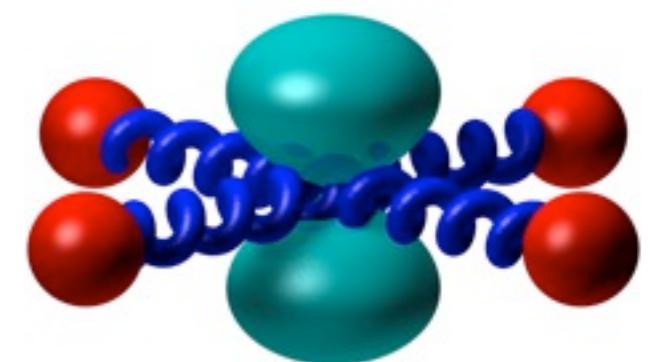
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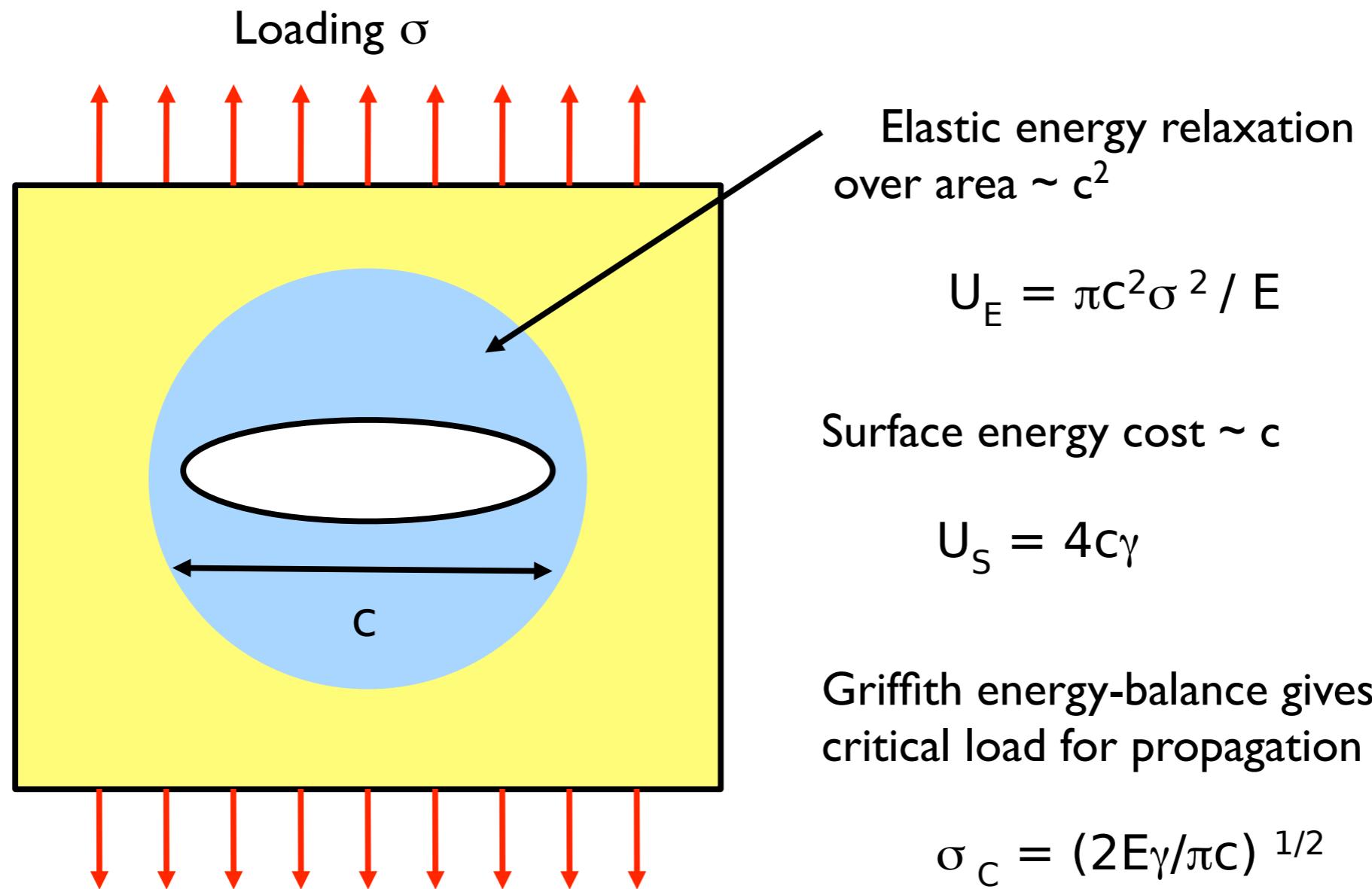
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Fracture Mechanics

- Continuum mechanics analysis of fracture dates back to Irwin and Griffith, beginning of 20th century
- For small cracks, energy balance idea due to Griffith shows that, for a given applied stress, cracks below a critical size will not grow



Fracture Mechanics

- Extension to general case: crack will propagate when

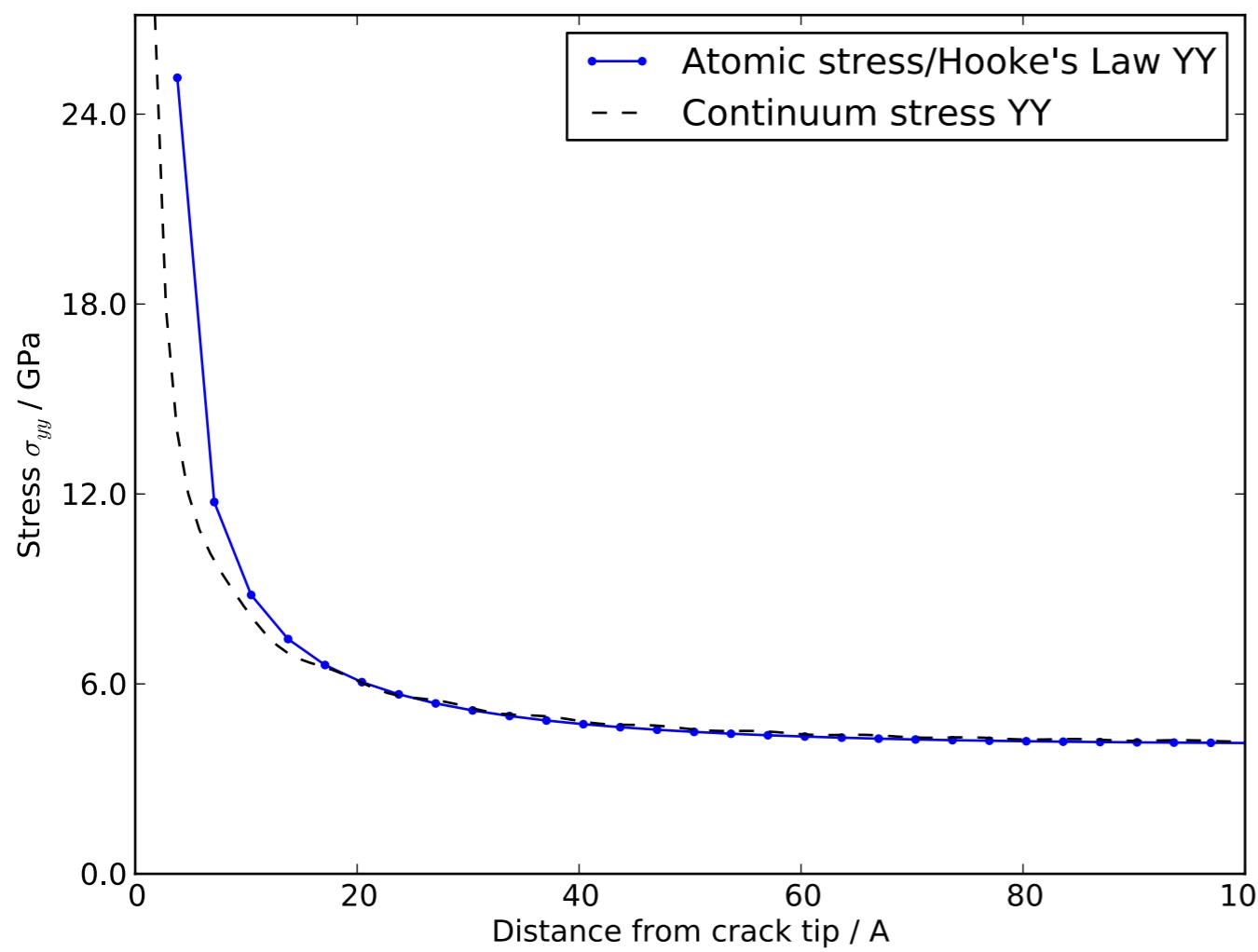
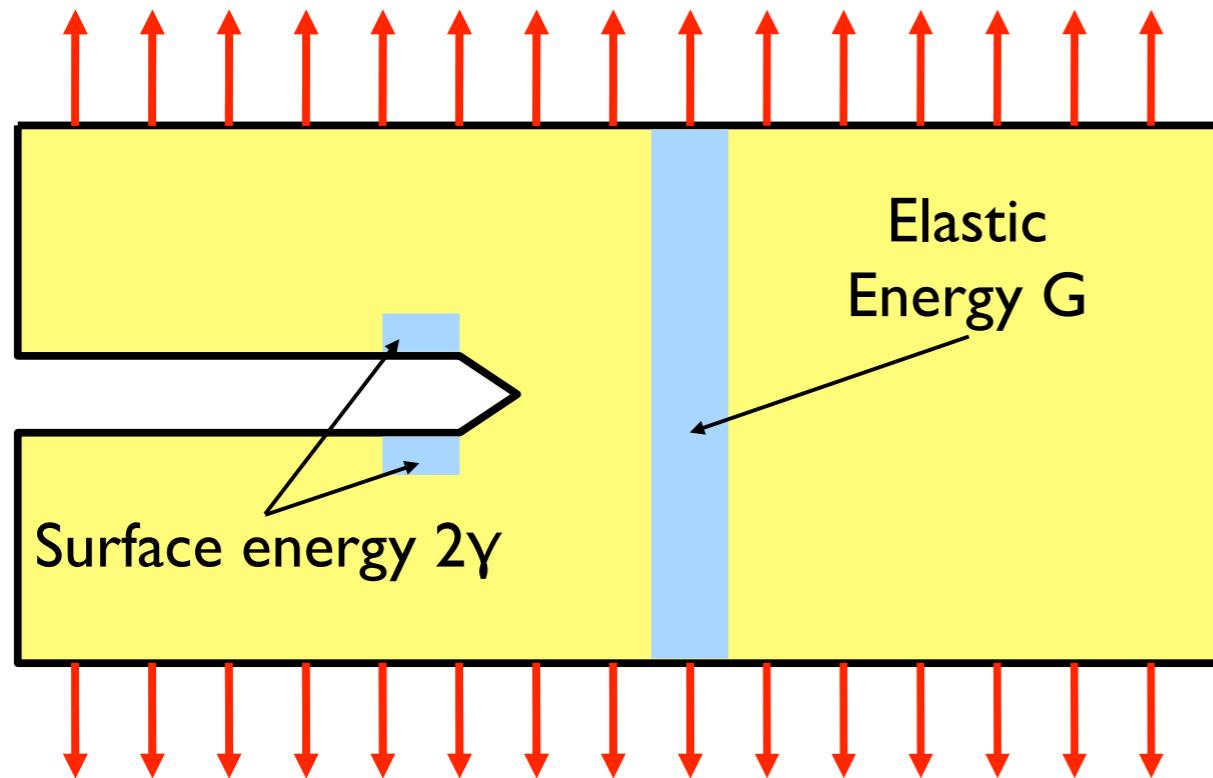
Energy release rate $G > 2\gamma$ Surface energy

- Strain energy release rate G determines initiation and propagation of crack
- Continuum modelling has been very successful, but until relatively recently, little has been known about atomic scale mechanisms
- Can we simulate dynamic fracture with MD?
- Everything depends on the choice of potential

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

- For modelling fracture, potential must be:
 - Sophisticated enough to capture elastic and thermodynamic properties
 - But simple enough to compute for very large systems

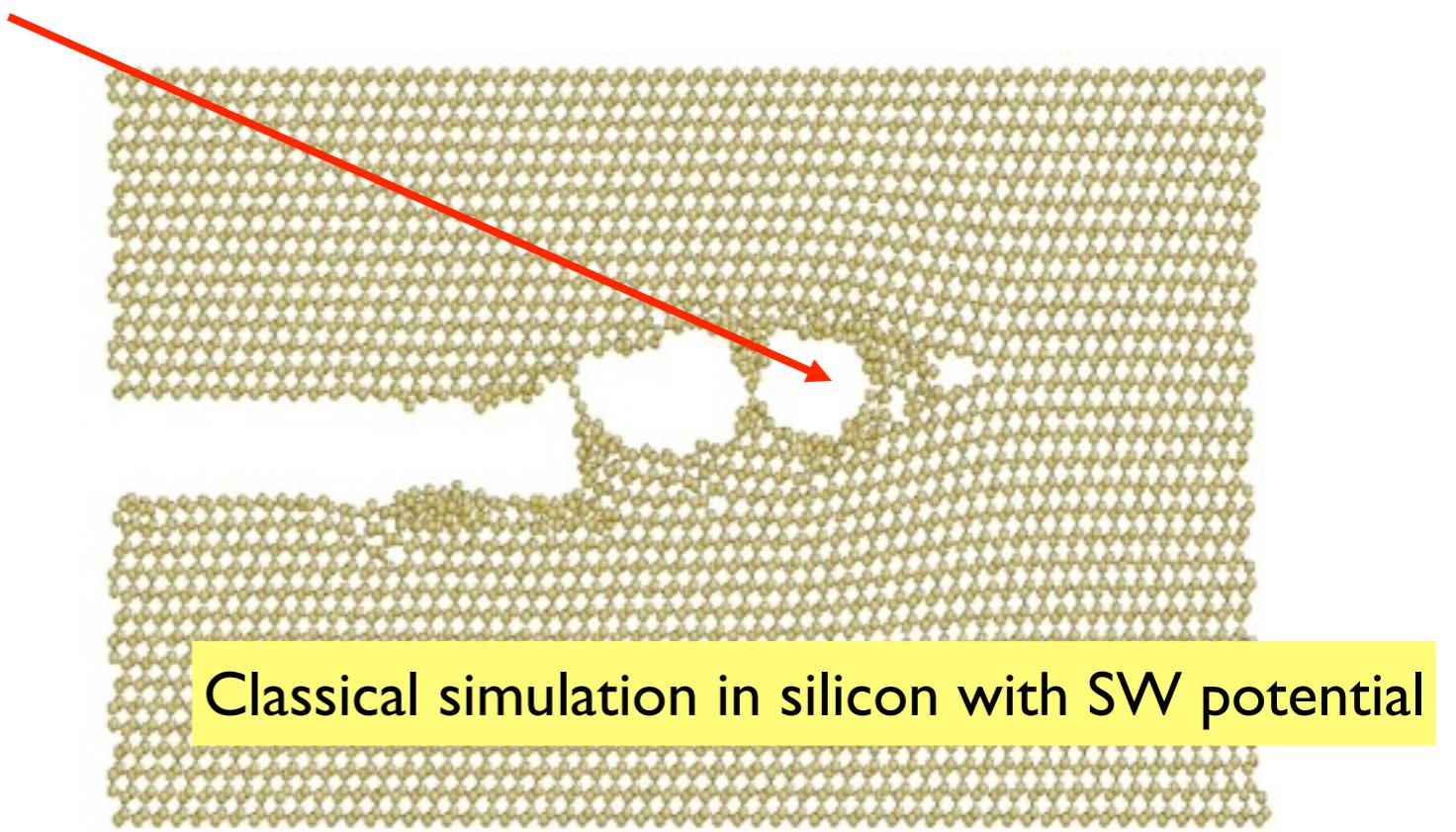
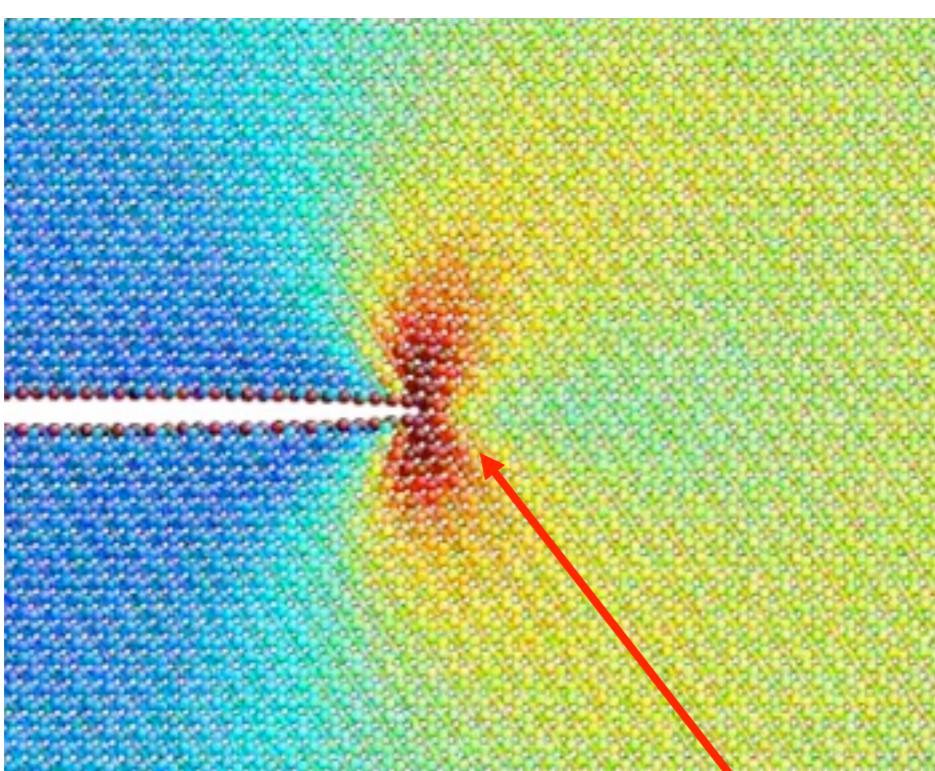
Simulating Fracture with Molecular Dynamics



- Usual MD tradeoff:
 - Speed: 100 000s of atoms for 100s of picoseconds
 - Accuracy: classical potential or quantum mechanics?
- Boundaries must be far enough away not to affect results
- Thin strip geometry: analytically solvable
- Use classical potential to calculate strains and stresses
- With big enough system, can match linear elasticity up to ~ 10 nm from tip
- This is “process zone” where interesting things happen

Simulating Fracture with Molecular Dynamics

- Classical potentials generally aren't accurate enough to correctly predict brittle fracture
- Tend to overestimate lattice trapping, leads to ductile fracture: rough surfaces, blunt tip, crack arrest



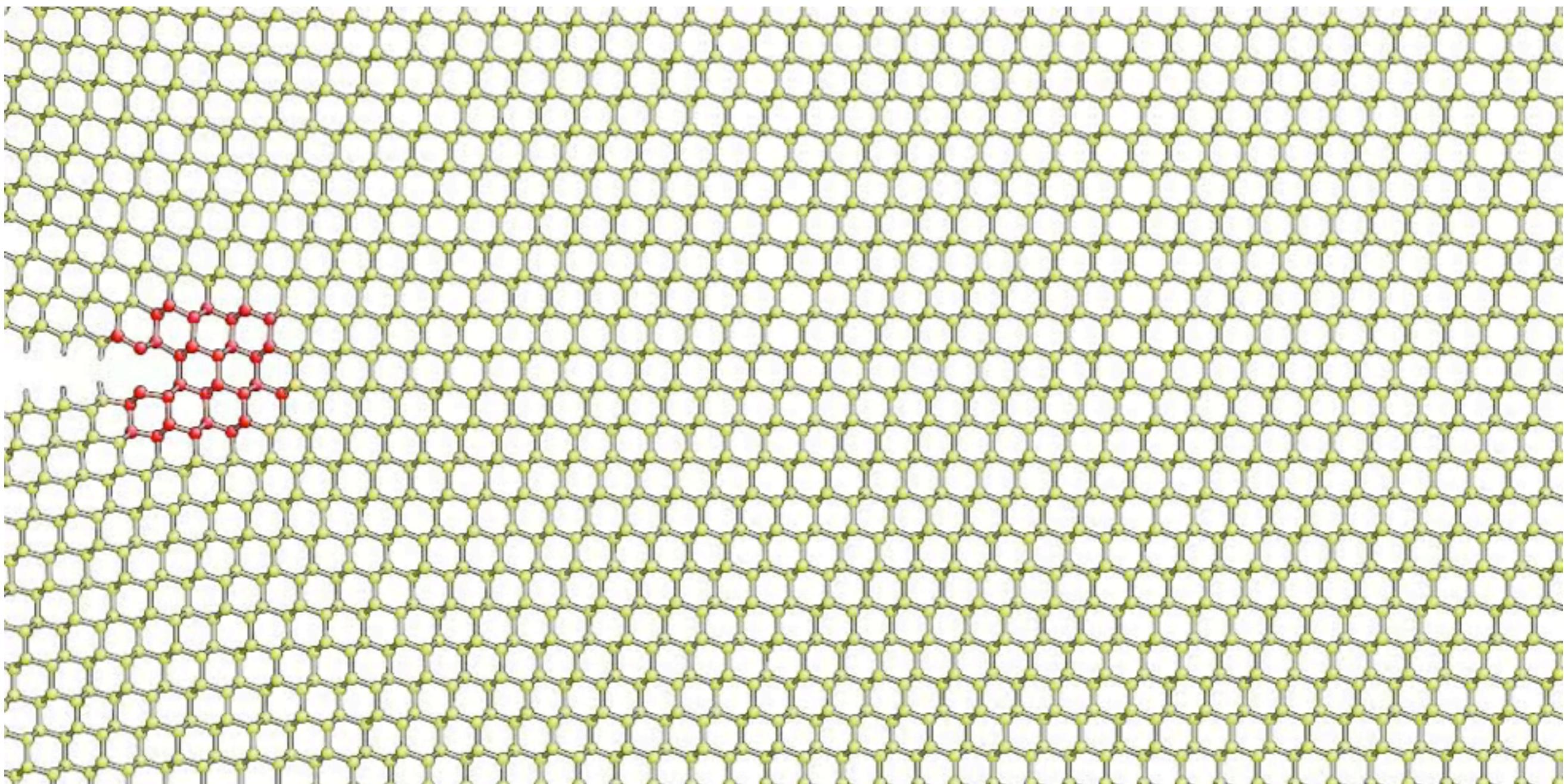
Classical stress field diverges at tip

Anharmonic bond stretching and rupture –
need Quantum Mechanics

Fracture is truly multi-scale problem, bidirectional coupling:
100 000s of atoms contribute to elastic relaxation

QM/MM modelling of Silicon Fracture

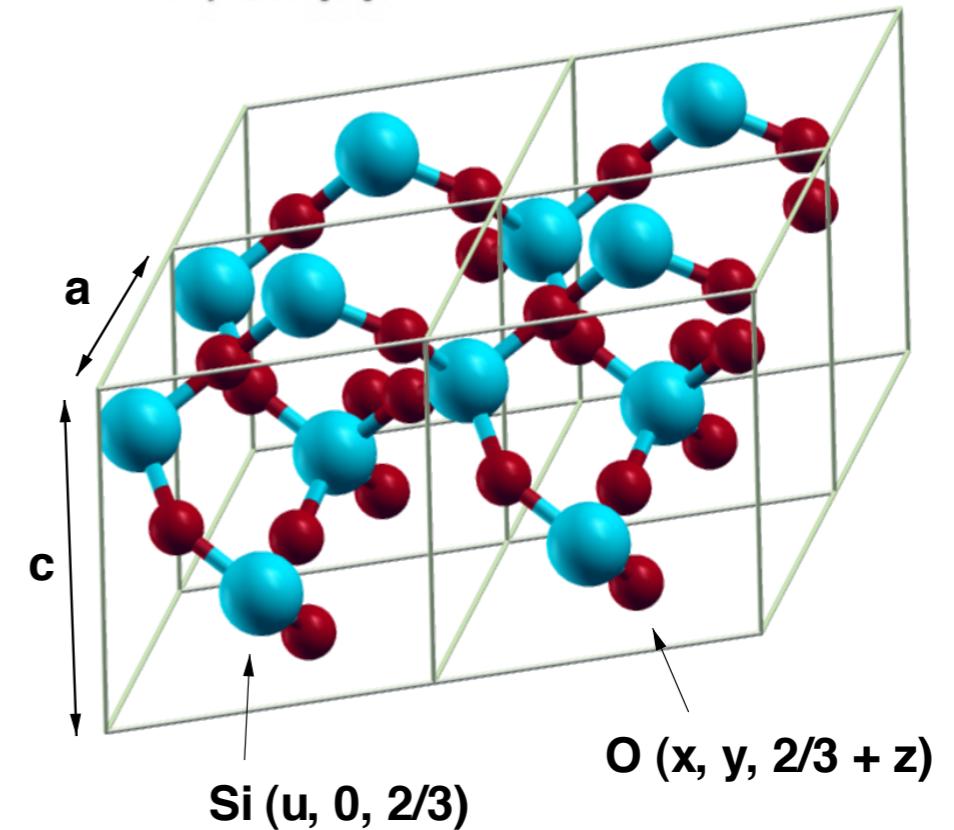
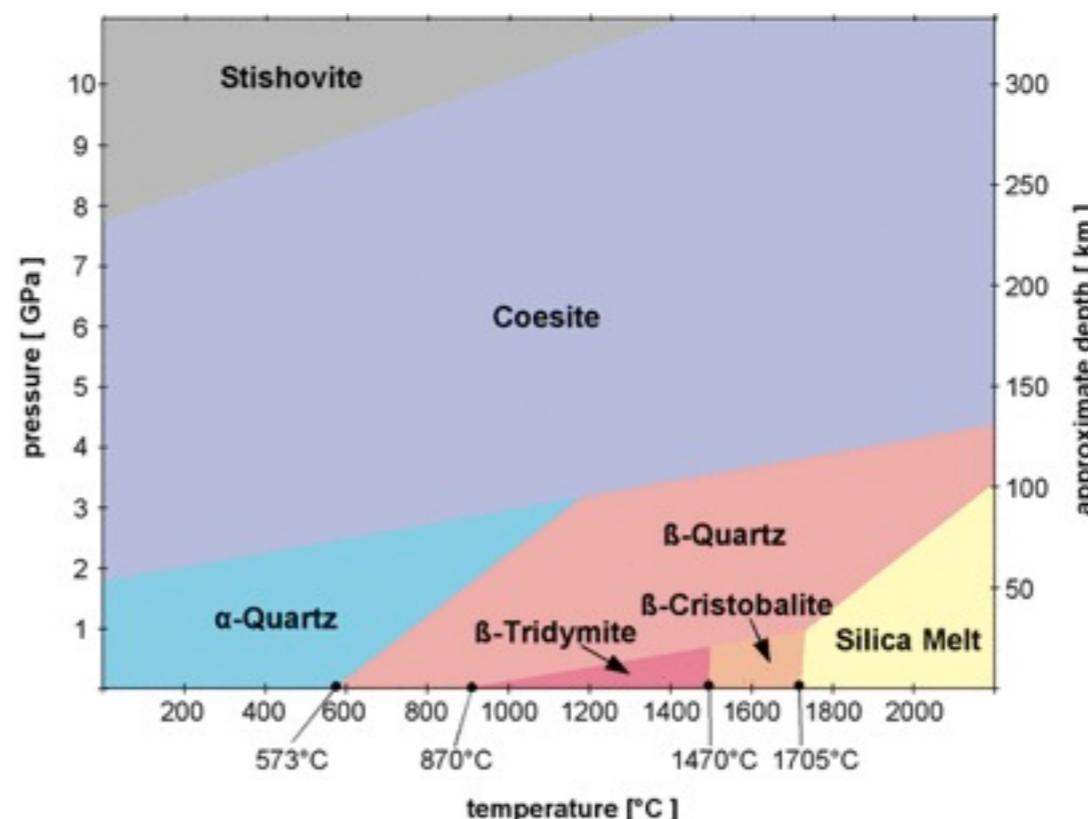
- Concurrent modelling: only use accuracy where it's needed
- 'Learn on the Fly' allows a small, movable QM region centred on crack



JR Kermode et al. Nature **455** 1224 (2008)

Motivation for studying fracture of oxides

- Mining is among most energy intensive of human activities
- Environmental impact of traditional techniques is increasingly severe
- Surprisingly little is understood about fundamental fracture processes in oxide materials
- Industrially funded project to apply what we've learned in silicon to study fracture of oxide materials, starting with silica
- Fundamental building block of rock-forming materials
- Most polymorphs built up from tetragonal units
- Low T and P : quartz

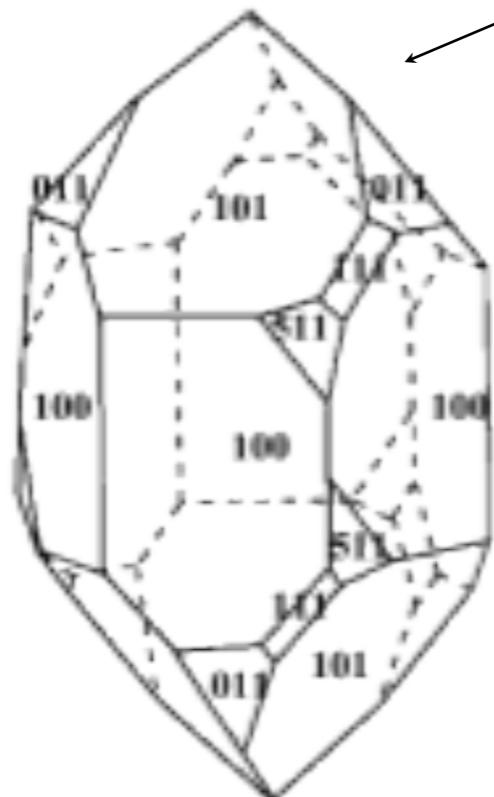


Fracture of quartz: cleavage planes

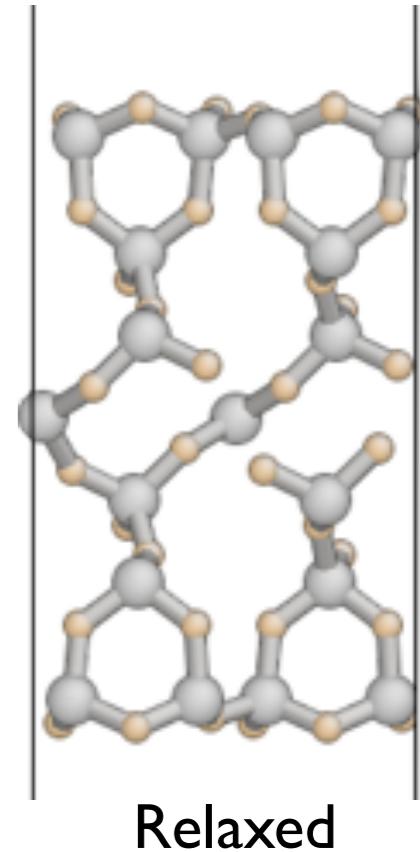
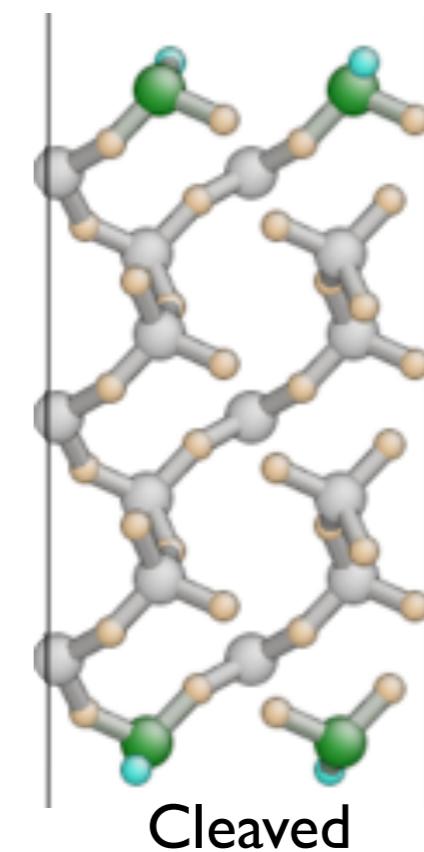
In experiments, α -quartz is seen to exhibit *conchoidal fracture*

- Most brittle materials have a preferred cleave plane
 - In conchoidal fracture, fracture toughness nearly equal across a fairly wide range of planes
 - Several planes obtained, with frequency proportional to Boltzmann factors of respective surface energies
 - Important planes include basal plane (0001), *m*-plane (10-10) and the positive (10-11) and negative (10-1-1) rhombohedral planes
 - Today we'll look at fracture on basal plane (0001)

Reproduced from Murashov et al, J. Phys. Chem. (2005)

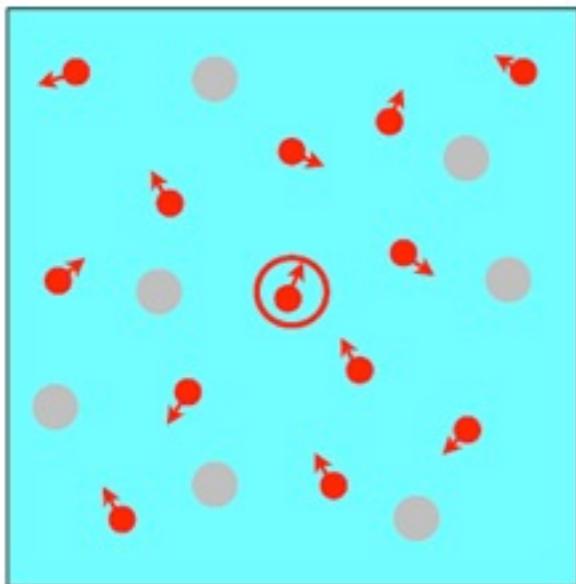


Basal plane (0001)



Modelling fracture of silicates

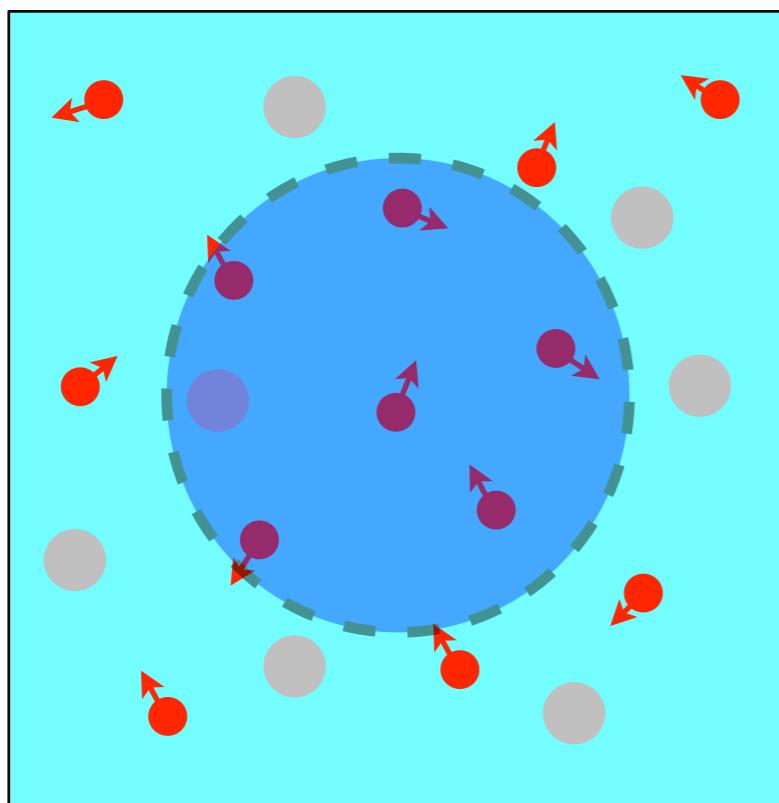
First requirement is good potential representation



Starting point: TS force field with self-consistent dipoles

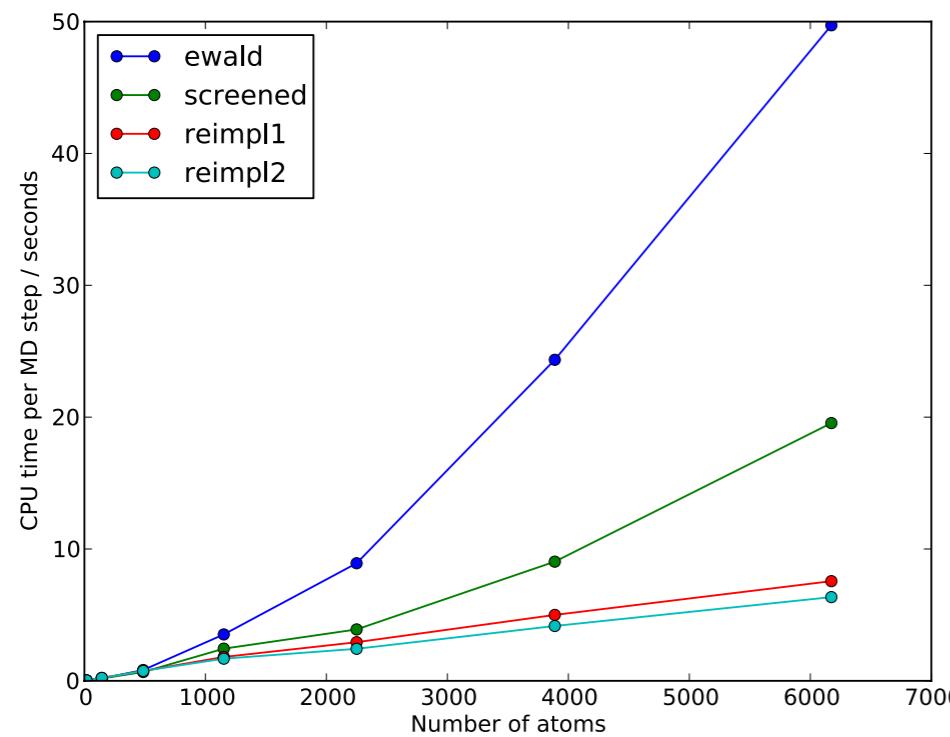
$$\mathbf{p}_i = \alpha_i \mathbf{E}_i (\{\mathbf{r}_j\}, \{\mathbf{p}_j\})$$

Short range (Yukawa): $\frac{q_i q_j}{r_{ij}} \rightarrow \frac{q_i q_j}{r_{ij}} e^{-\alpha r}$

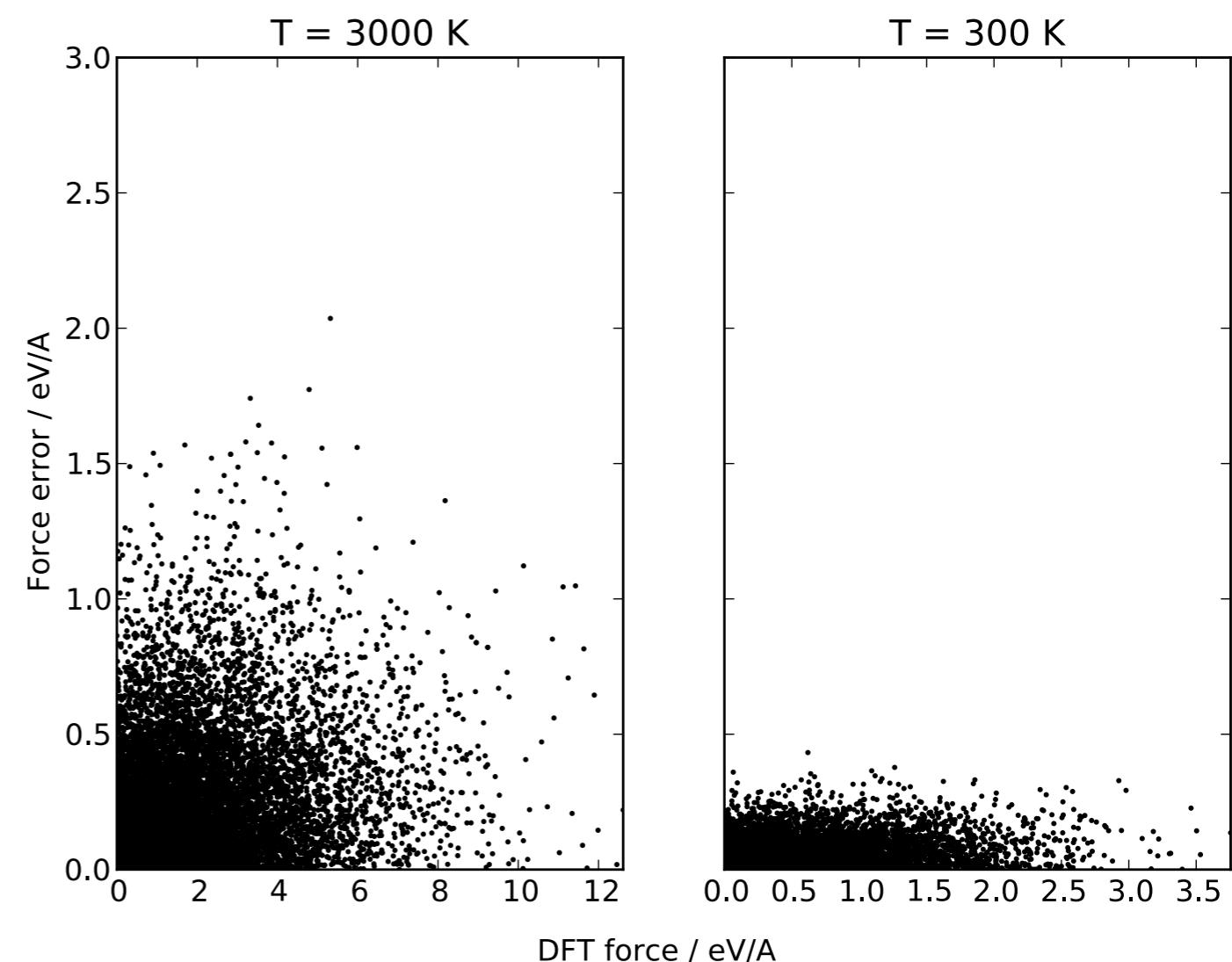
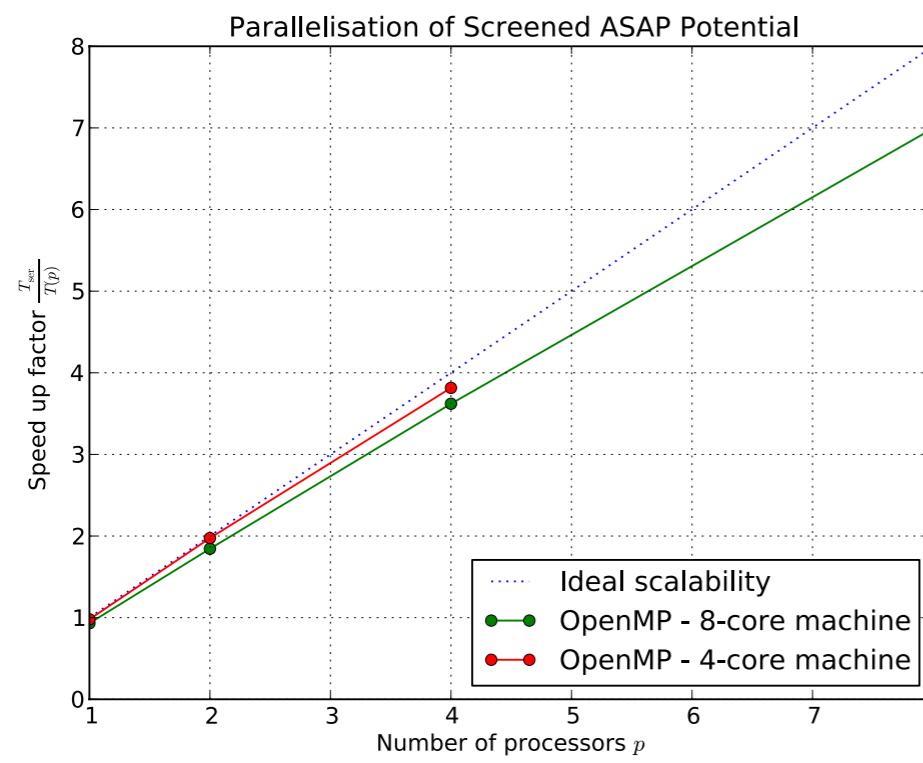


- Dipoles now only “see” other dipoles within distance $\sim 1/\alpha$
- Corresponds to physical screening processes
- This makes algorithm linear scaling, and suitable for parallelisation
- We had to refit potential parameters

Scaling, and force accuracy in quartz

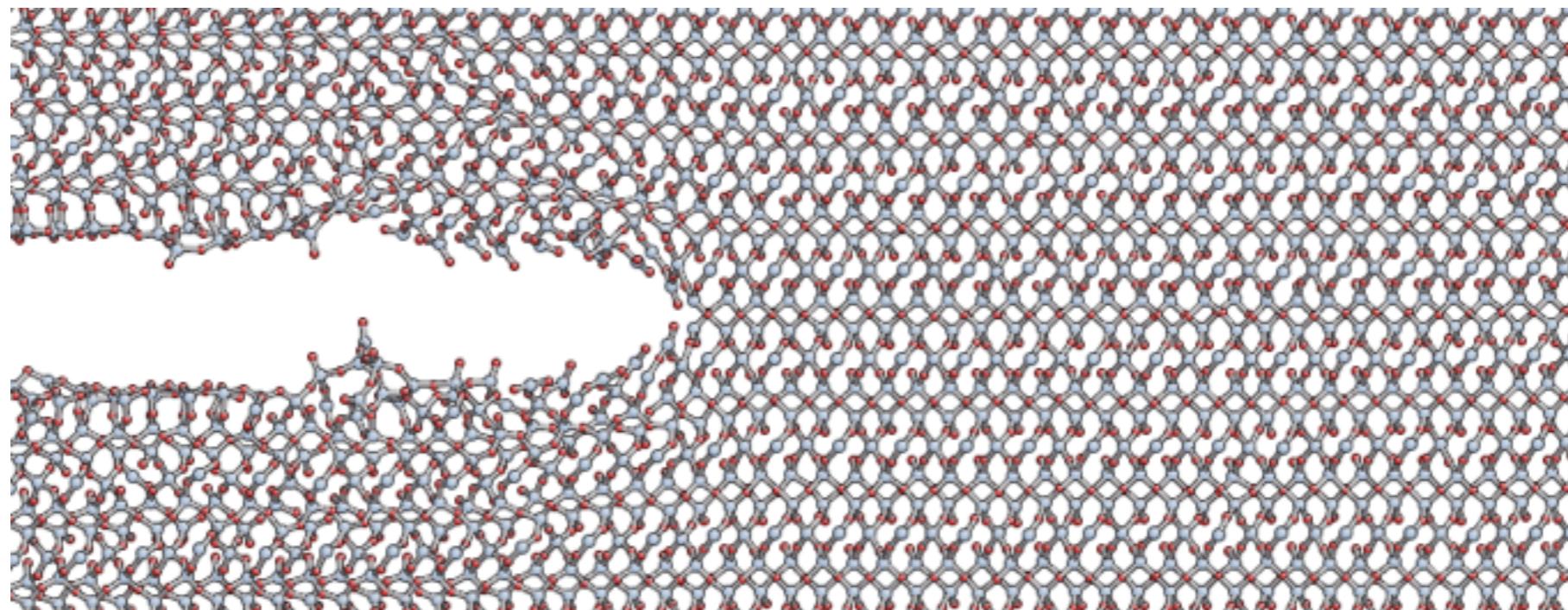


- Potential has been refitted from scratch to a new QM (LDA) database generated from molecular dynamics at $T \sim 500$ K
- Good fit with screening parameter $\alpha = 0.1 a_0^{-1}$, using a cutoff of $2/\alpha = 20 a_0 (\sim 10 \text{ \AA})$



Practical session

- Carry out classical simulations of fracture in quartz
- Make theoretical predictions, compare with simulation results
- We'll be using QUIP molecular dynamics code
- Ask me or one of other tutors if you have problems
 - Gianpietro Moras, Fouad Atrash and Giovanni Peralta
- Follow step-by-step online tutorial
 - <http://www.jrkermode.co.uk/quippy/adglass.html>



Molecular Dynamics Simulation of Fracture in Quartz

Part 2: Results and Conclusion

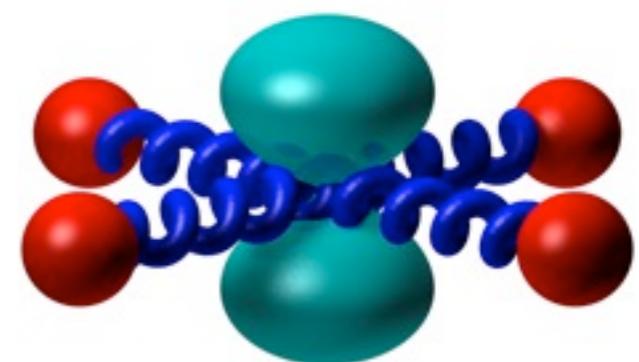
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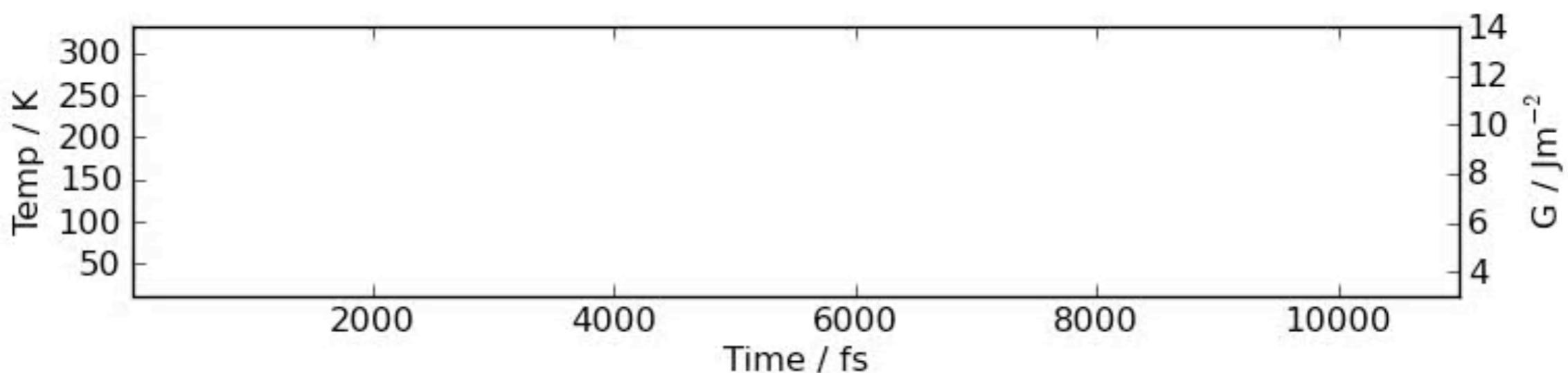
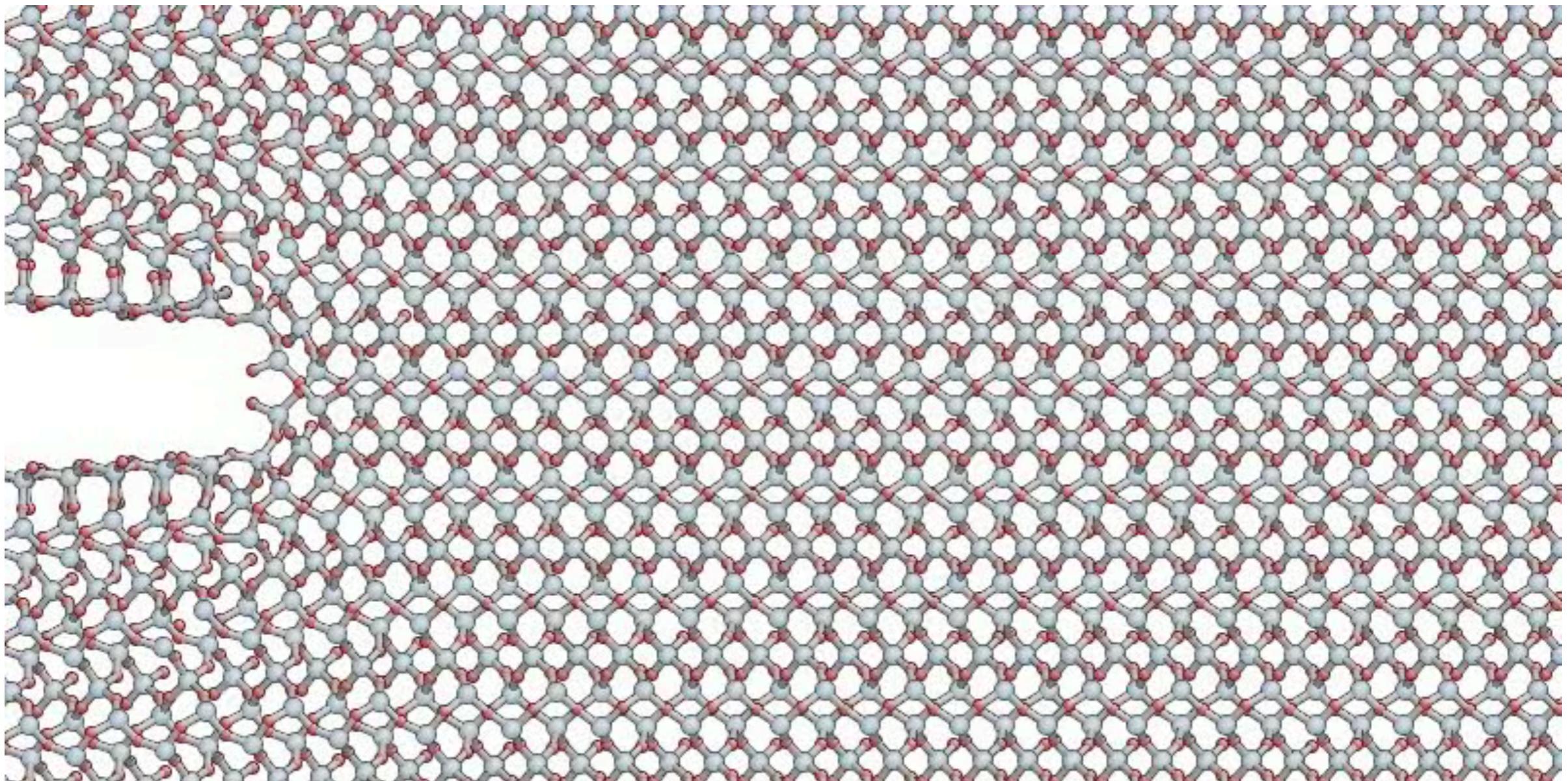


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Classical fracture simulations of quartz

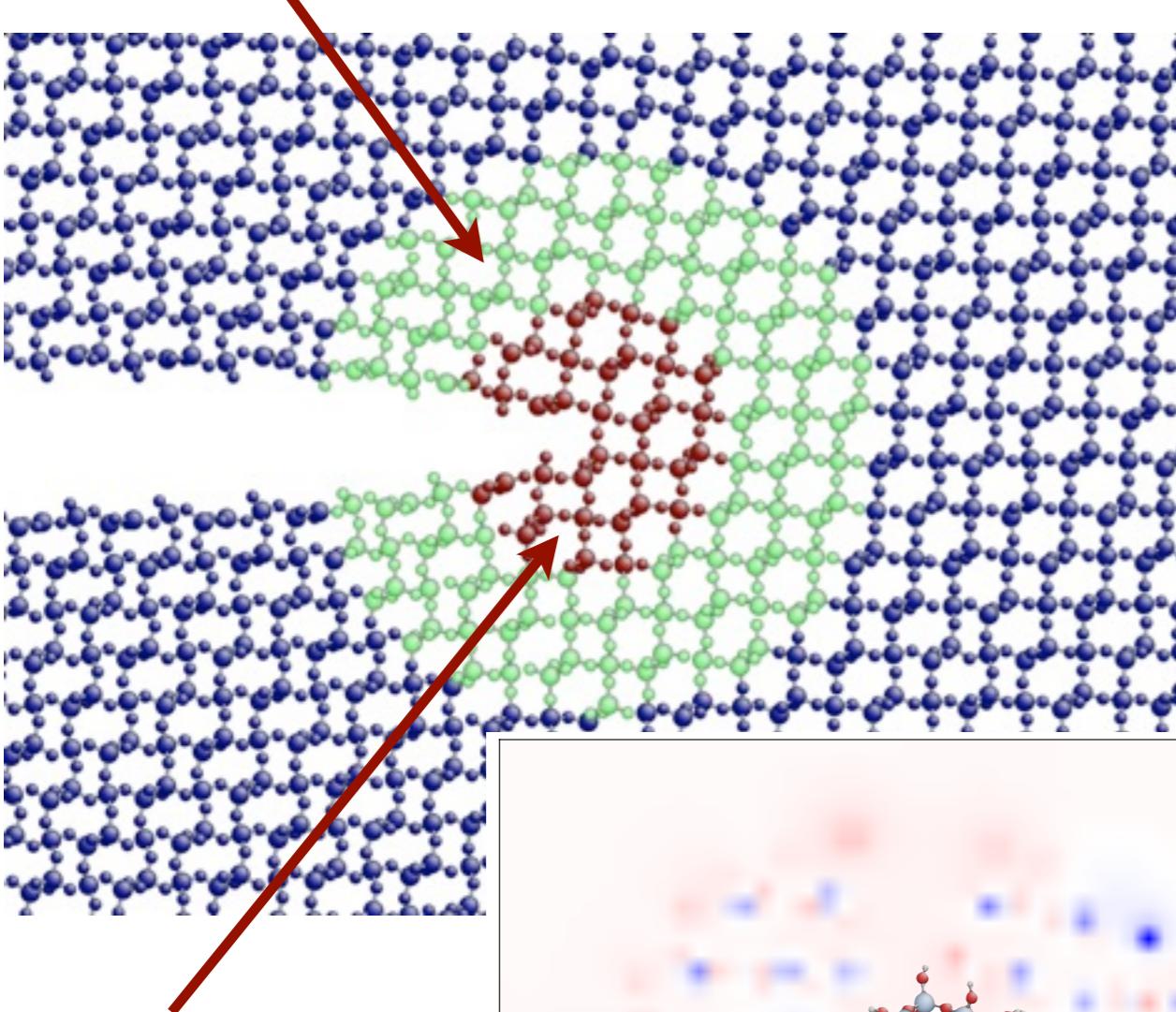


Overcoming Limitations

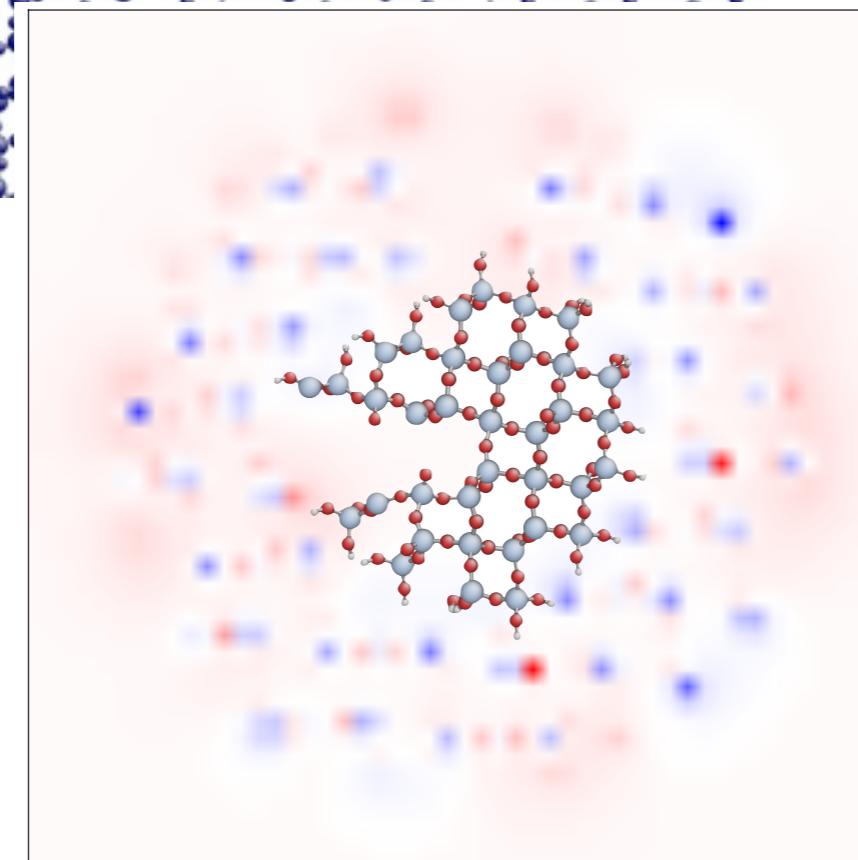
- System modelled is very small
- Applied loading is very large so fracture is fast
 - With larger computer and more time, can increase to 100 000s of atoms for 100s of picoseconds
 - Can converge to close to experimental fracture toughness
- Classical description of bond-breaking is fairly simplistic
 - To remove this limitation, need to embed a QM description near crack tip region
 - Essential to describe chemo-mechanical processes like stress corrosion

QM/MM electrostatic embedding

Electrostatic embedding region

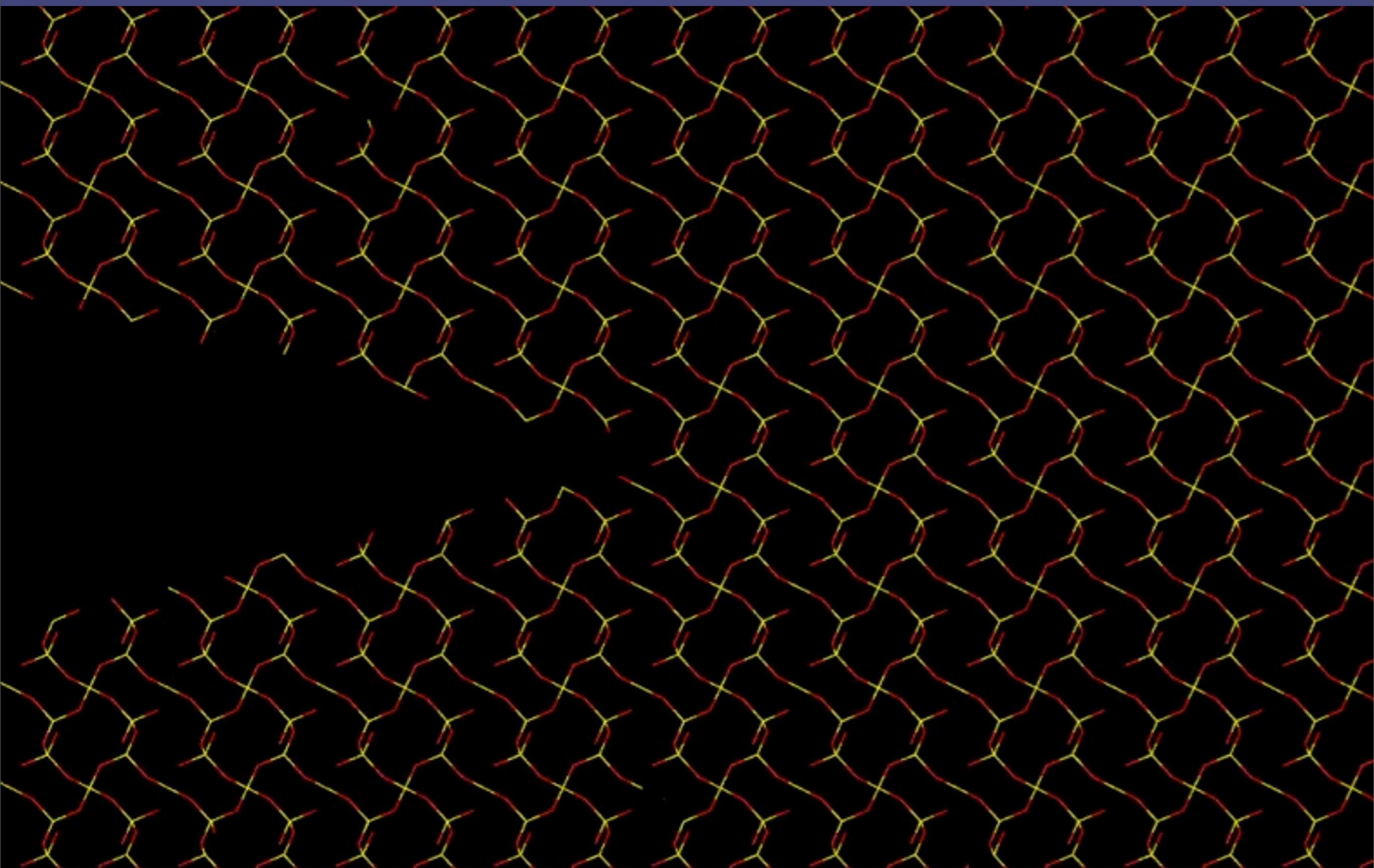


QM core region



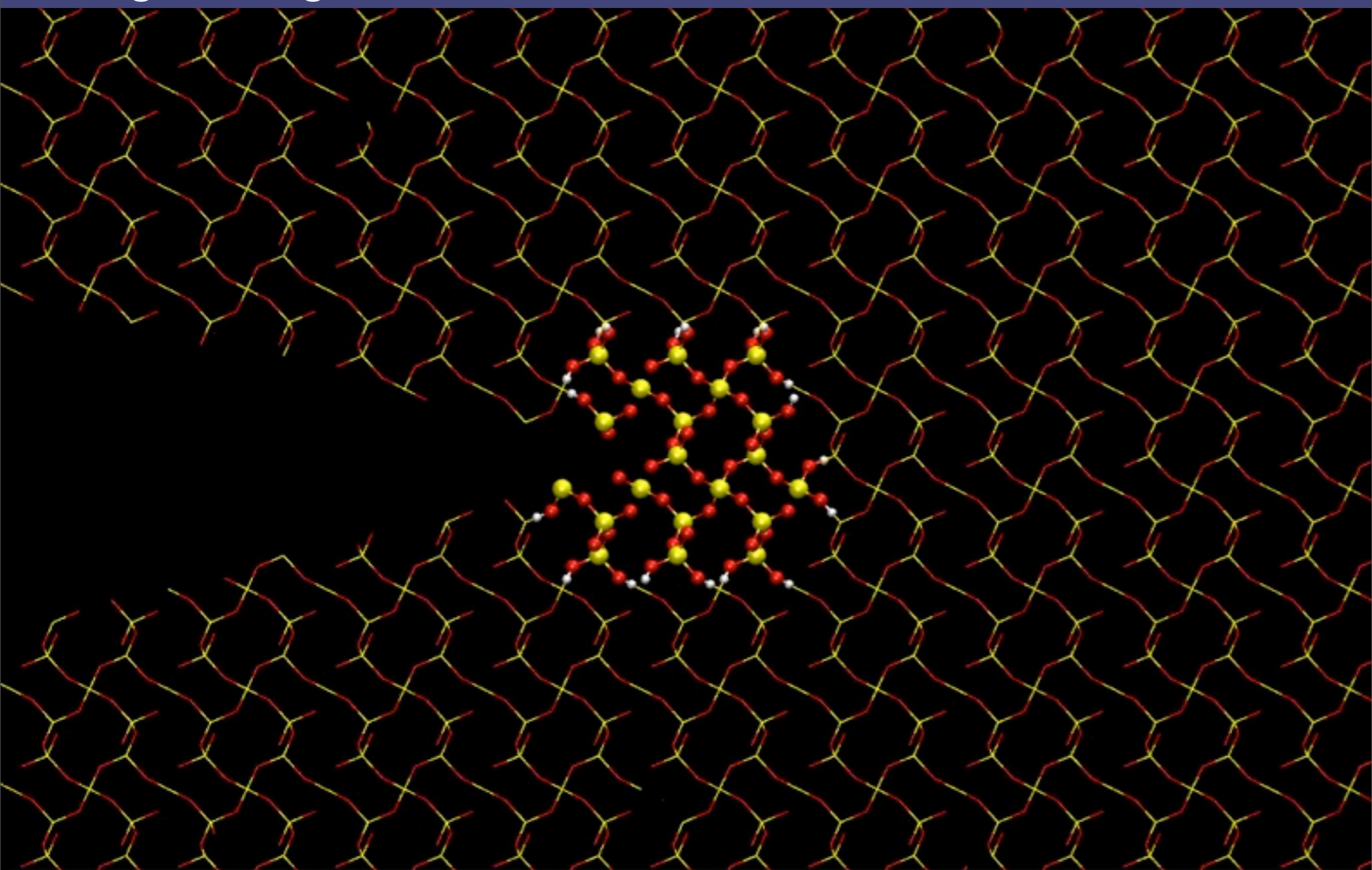
- ‘Learn on the Fly’ technique has traditionally used mechanical embedding with a to give accurate forces on central atoms
- Ionic nature of bonding here leads to longer range forces
- Embed QM region in full electrostatic potential due to all charges and dipoles within ~1 nm shell
- Modify QM external potential and forces accordingly

Putting it all together



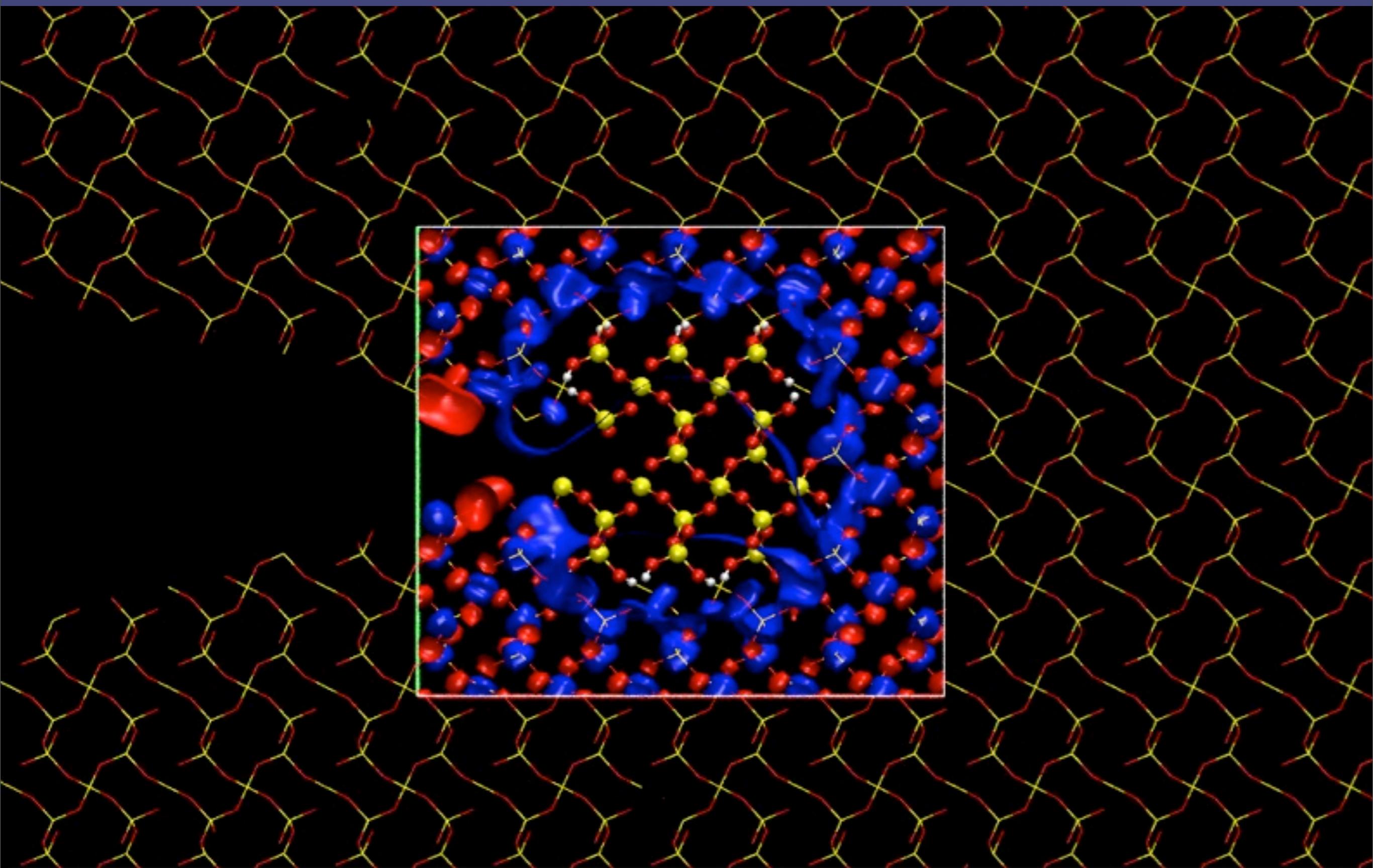
(I) Identify crack tip

Putting it all together



(2) Carve quantum mechanical cluster

Putting it all together



(3) Evaluate electrostatic potential on grid within cluster

Acknowledgments

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