Molecular dynamics with on-the-fly machine learning of quantum mechanical forces

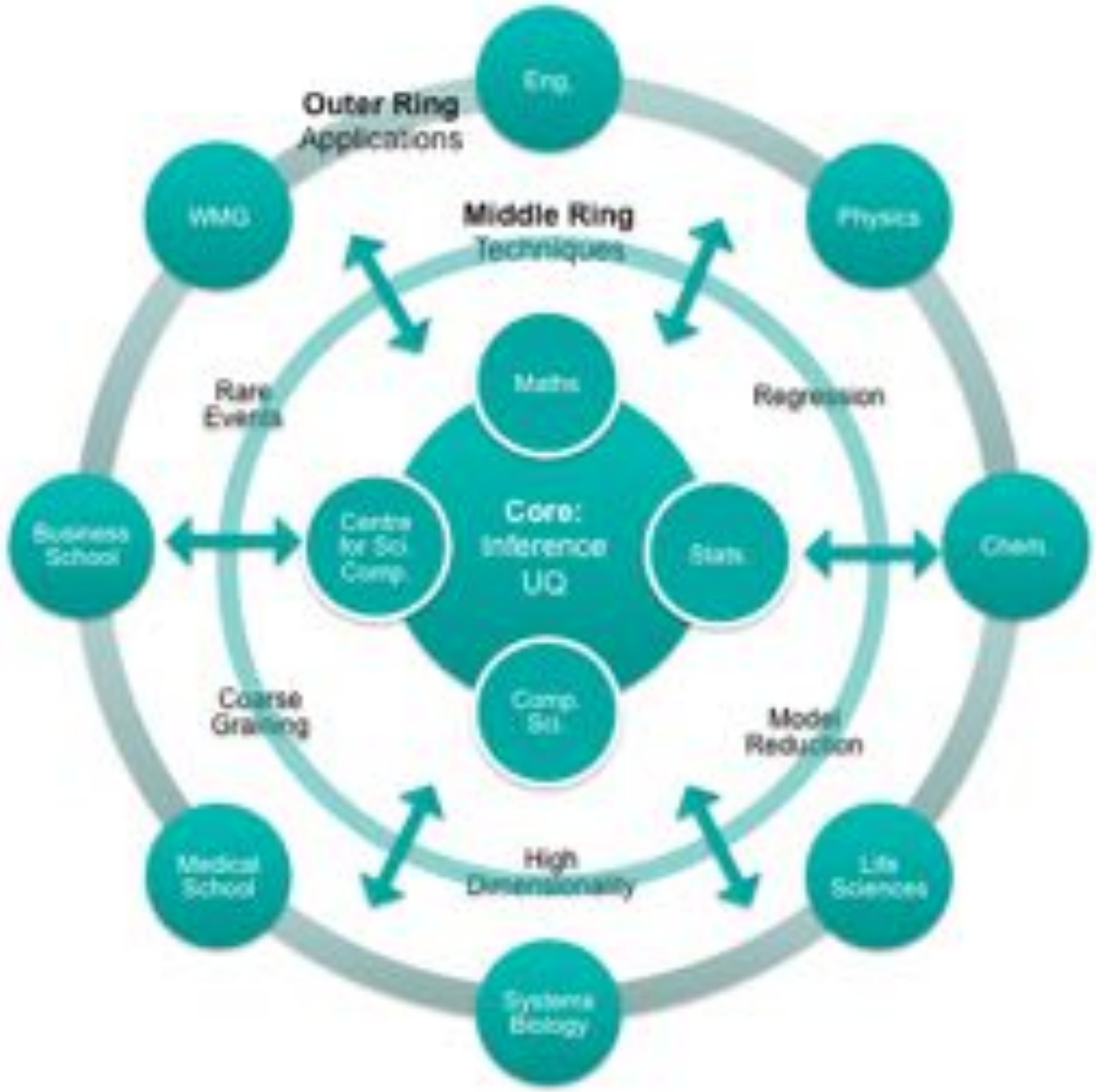
James Kermode
Warwick Centre for Predictive Modelling
School of Engineering
University of Warwick
www.warwick.ac.uk/wcpm

Warwick Centre for Predictive Modelling Seminar Series
5th March 2015

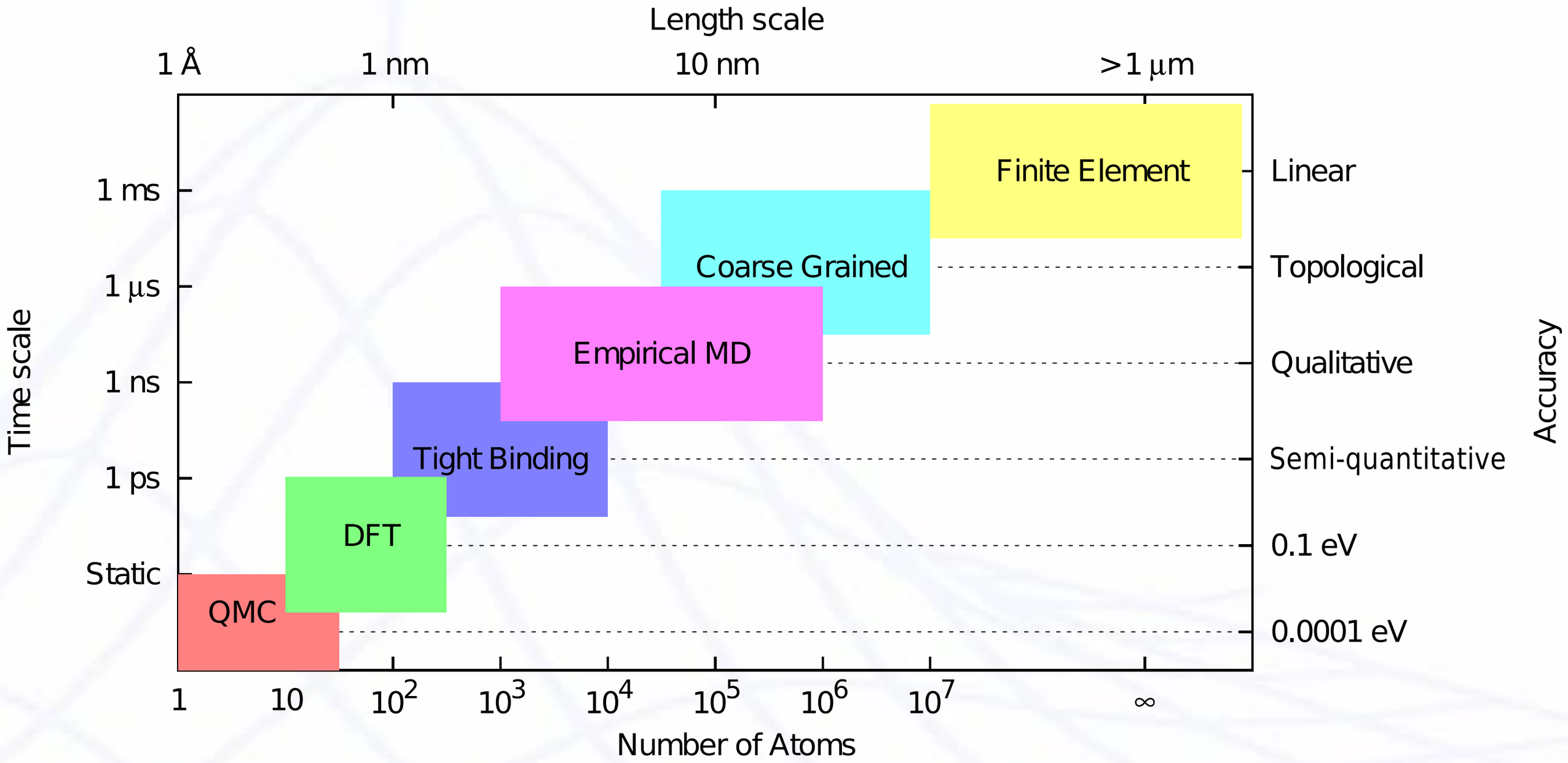
WCPM

WARWICK

Warwick Centre for Predictive Modelling



Multiscale Materials Modelling



Accuracy & transferability

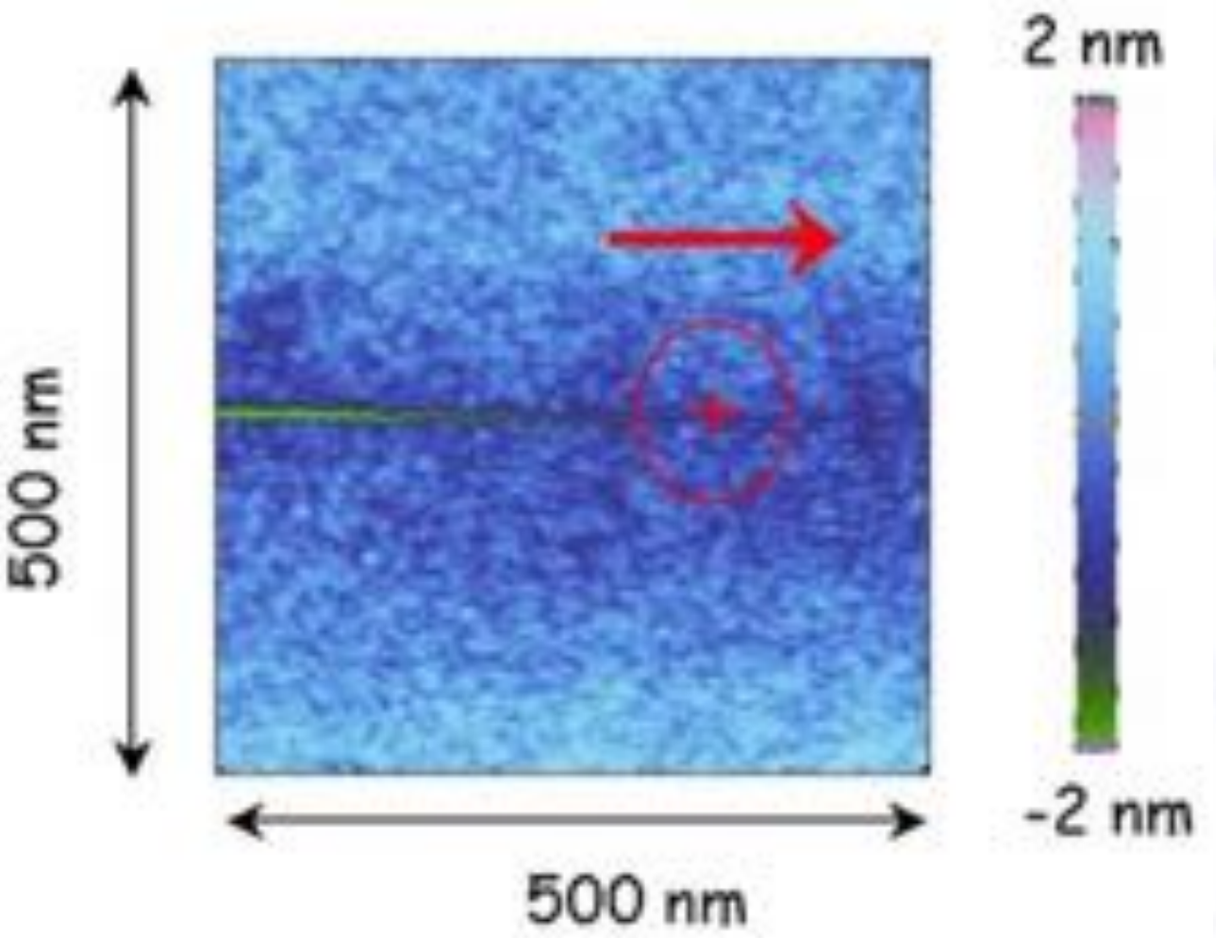
Materials failure process:
complex chemistry *and* large systems

Matching Atomistic and Continuum

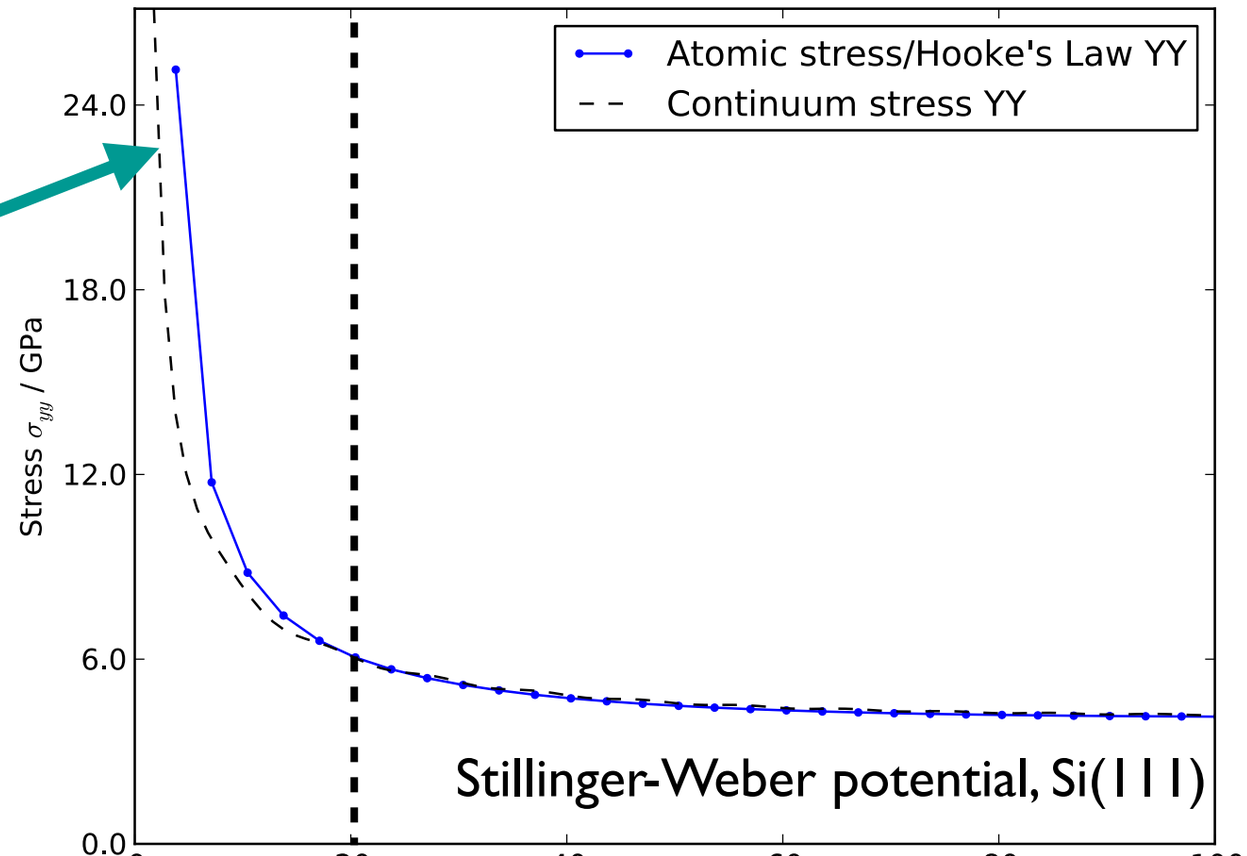
Cracks in ideal brittle materials are atomically sharp

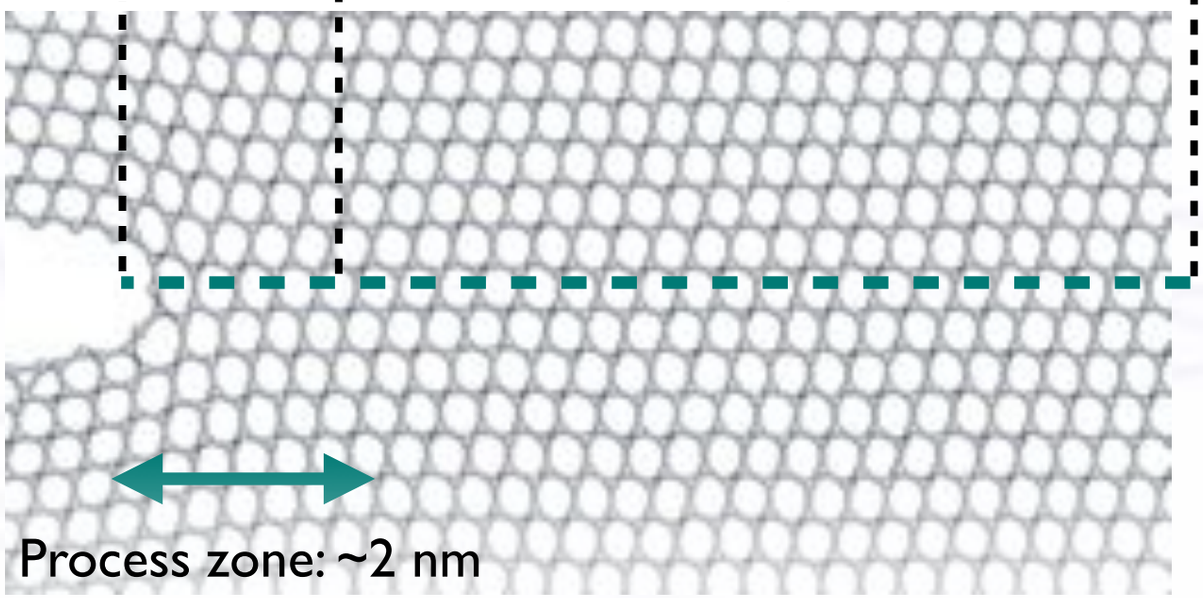
Divergence of stress field near a crack tip

$$\sigma \sim \frac{1}{\sqrt{r}}$$

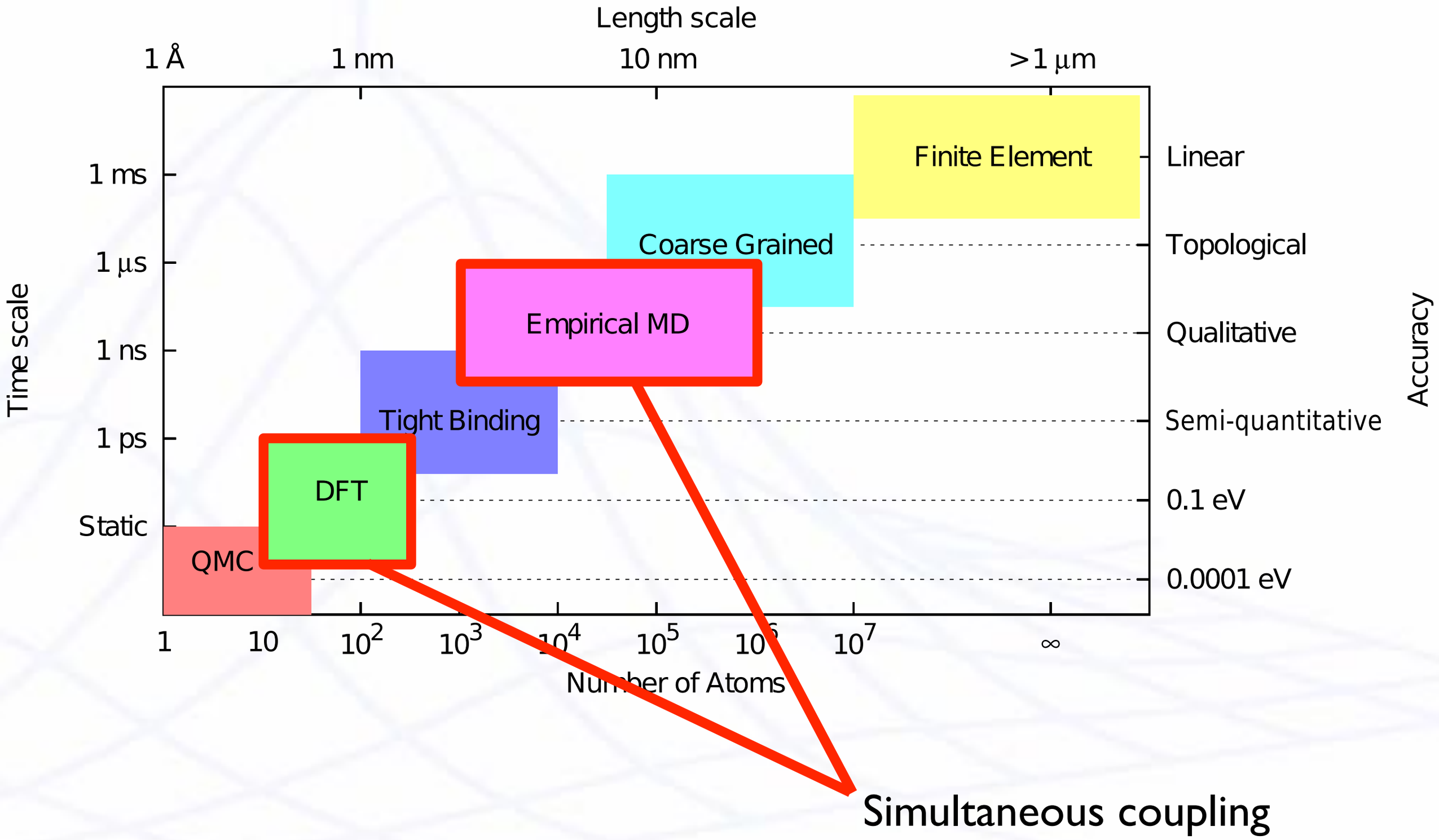


In situ AFM image of atomically sharp crack tip in silica (Image: C. Marlière)

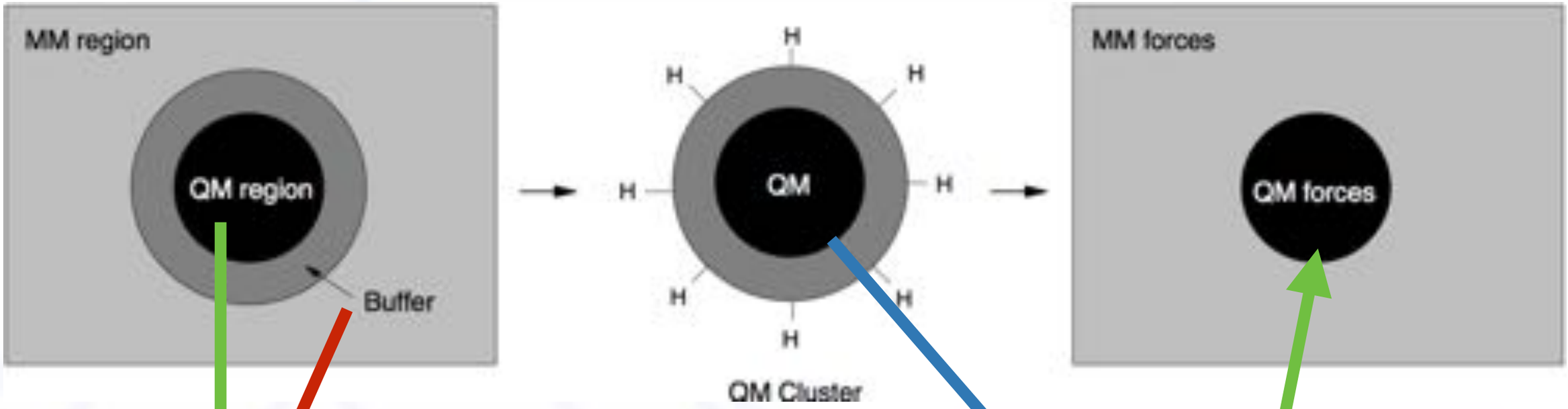


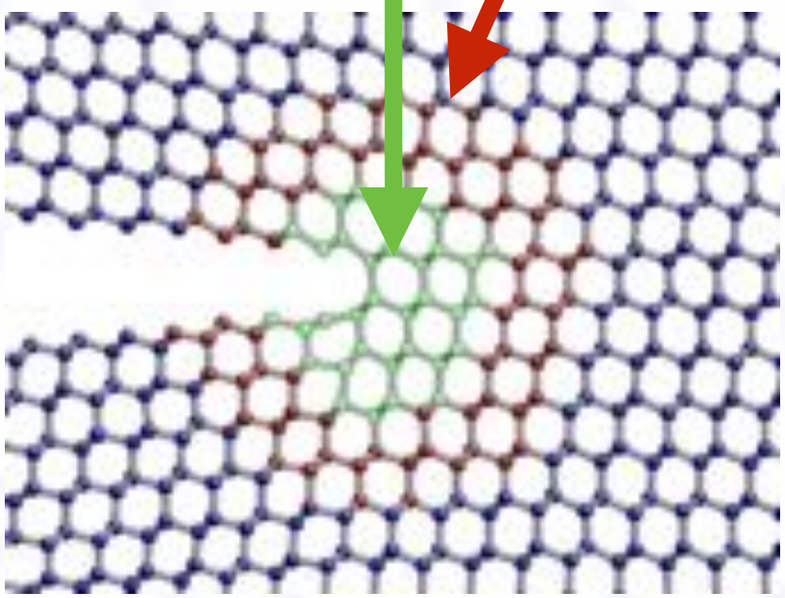


Multiscale Materials Modelling

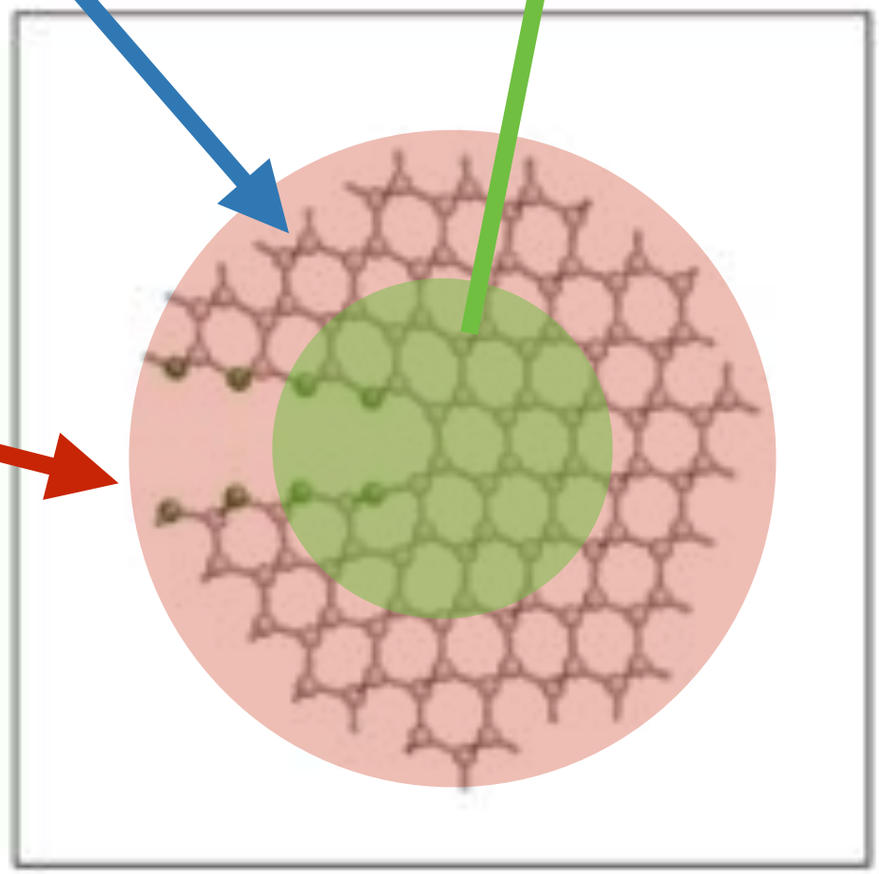


QM/MM – buffered force mixing



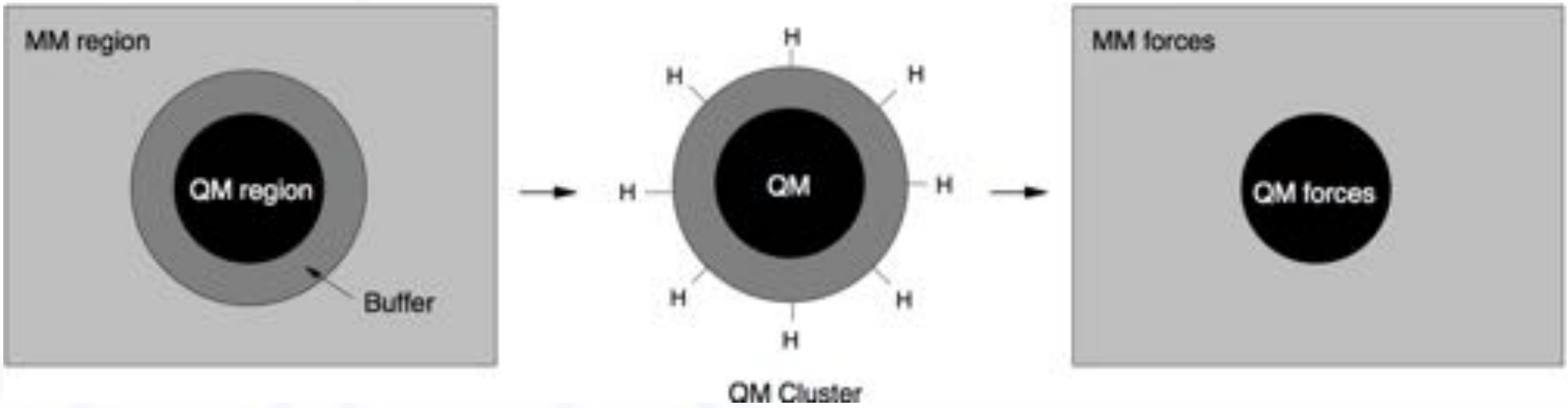


- Retain QM forces only in (green) core region.
- Throw away 'bad' QM forces on buffer region close to artificial surface

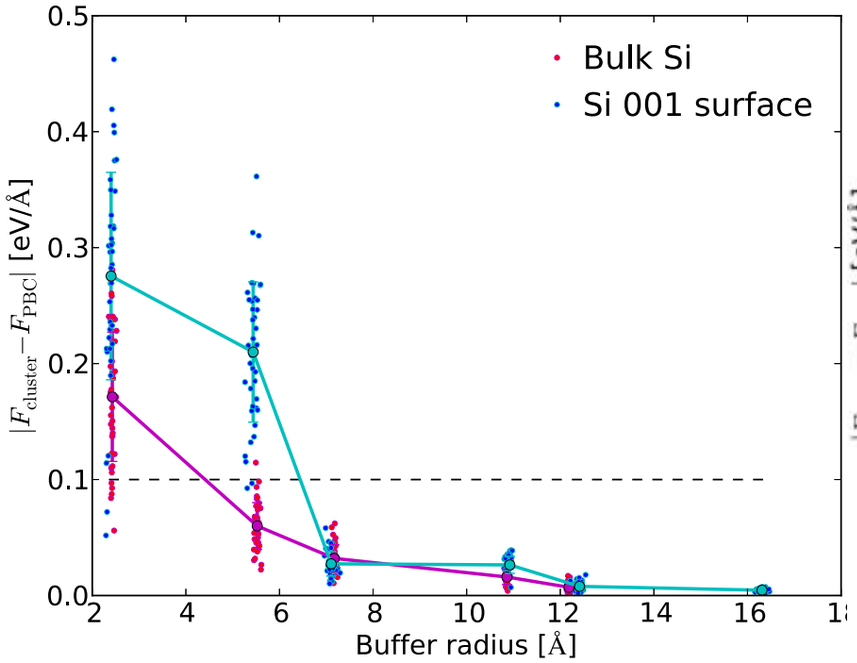


Abrupt force mixing and *extended buffer region* solve electronic termination problem, giving accurate QM forces.

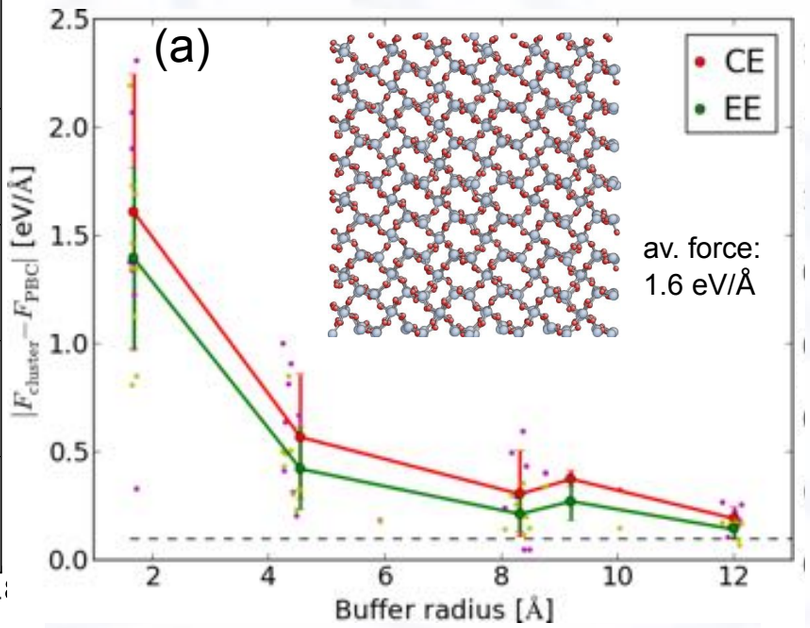
QM/MM – buffered force mixing



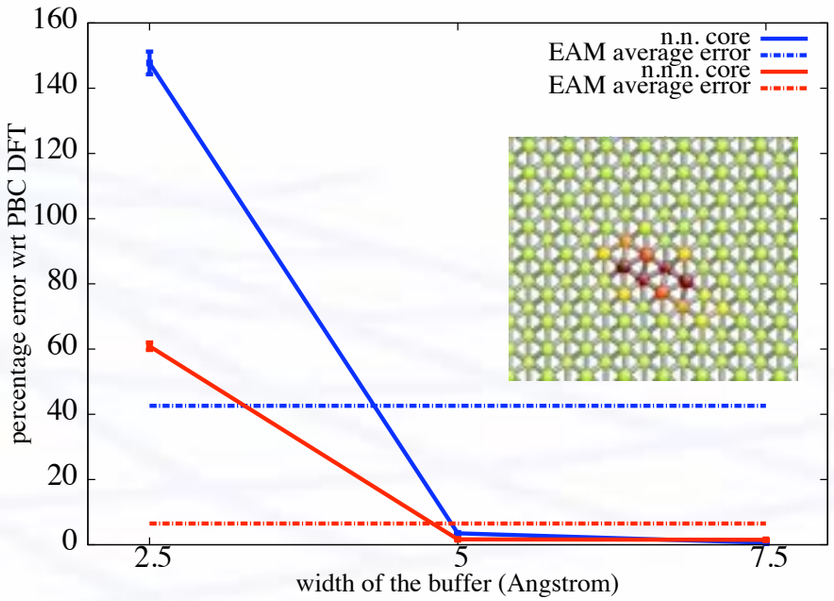
Covalent Materials | **Oxides** | **Metals**



Silicon bulk and surface ~ 7 A



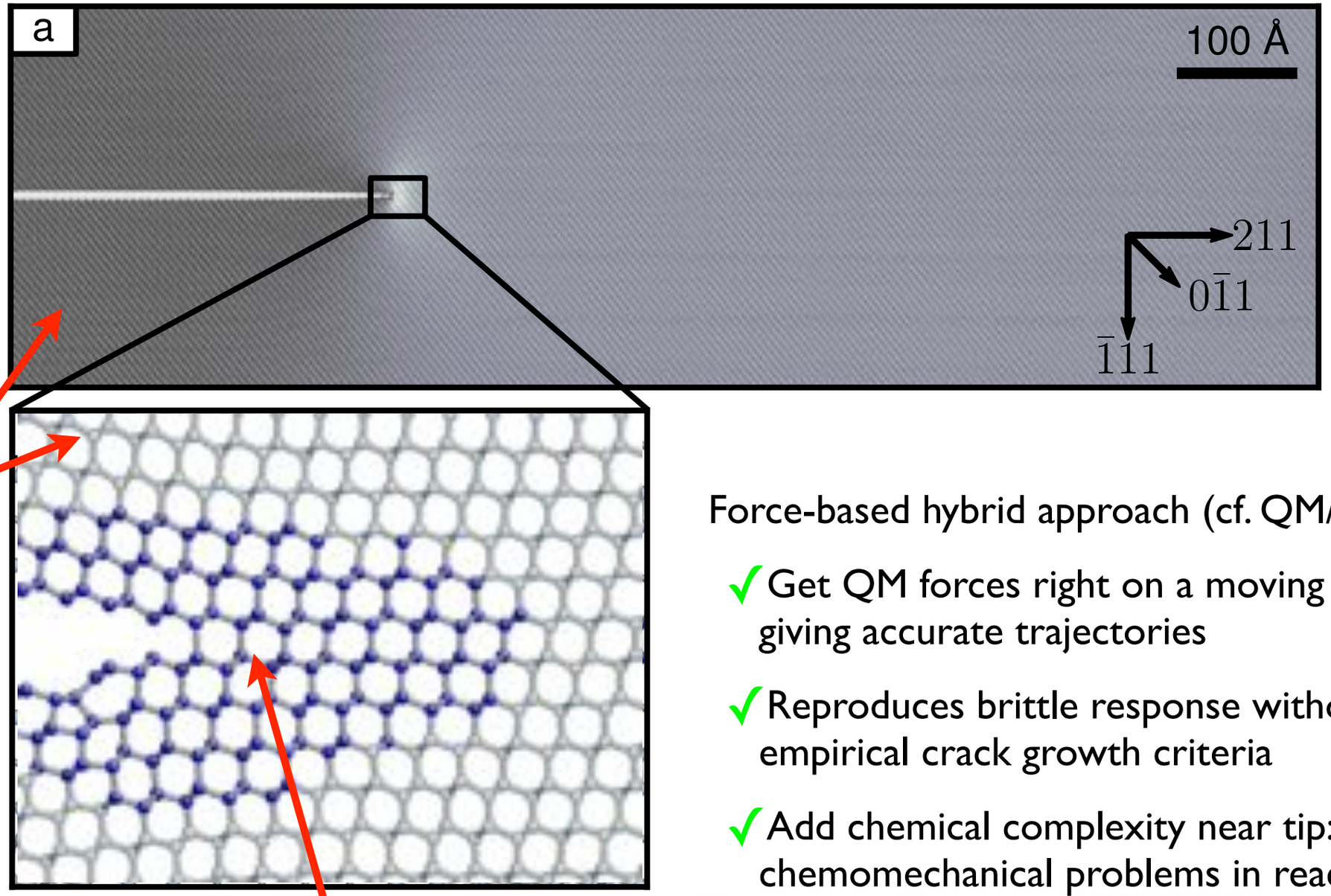
Silica (quartz, amorphous) ~ 10 A



Dislocation core in Ni ~ 5 A

N Bernstein et al. Rep. Prog. Phys. **72** 026501 (2009)
A Peguiron et al., J. Chem. Phys, **142** 064116 (2015)

‘Learn on the Fly’ scheme



Molecular Mechanics (empirical, fast)

Quantum Mechanics (accurate, slow)

Force-based hybrid approach (cf. QM/MM)

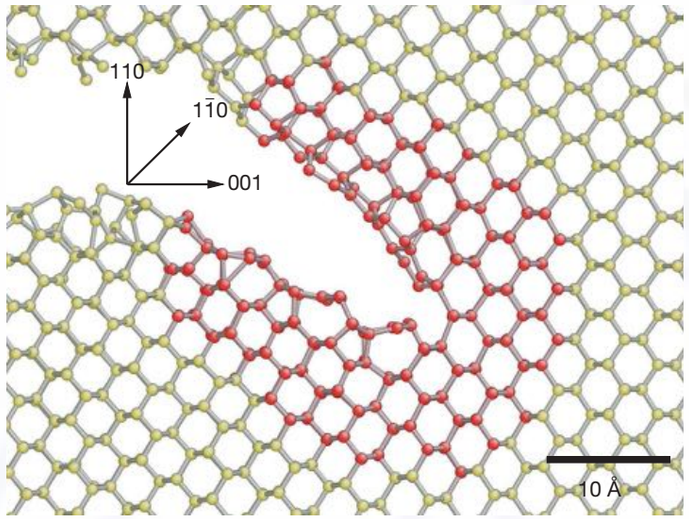
- ✓ Get QM forces right on a moving region, giving accurate trajectories
- ✓ Reproduces brittle response without any empirical crack growth criteria
- ✓ Add chemical complexity near tip: wide range of chemomechanical problems in reach

Code: http://www.libatoms.org

A. De Vita and R. Car, MRS Proc 491, 473 (1998)
G Csányi et al. Phys. Rev. Lett. 93 175503 (2004)

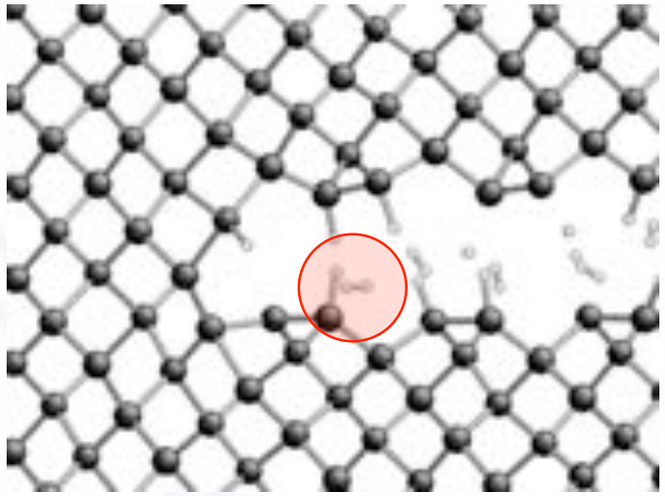
Recent Chemomechanical Applications

Dynamical instabilities in Si



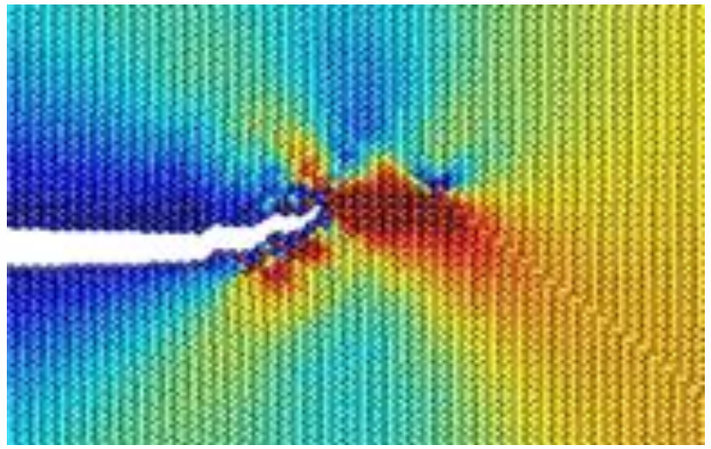
JR Kermode et. al.
Nature 455 1224 (2008)

H induced 'SmartCut'



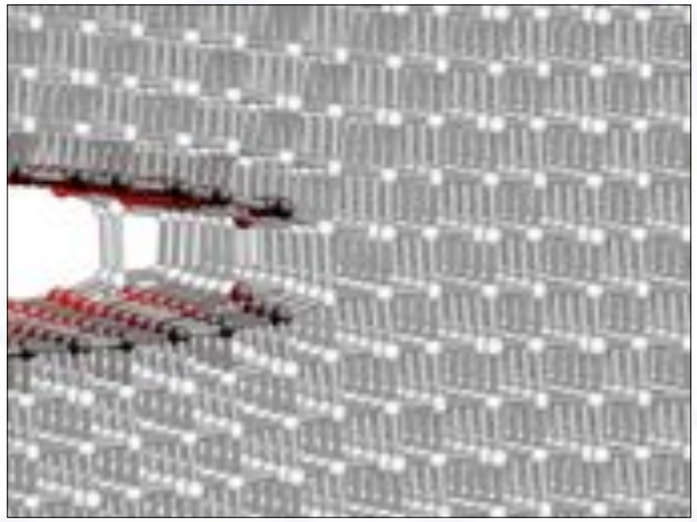
G Moras, L Ciachhi, C Elsässer,
P Gumbsch and A De Vita
Phys. Rev. Lett. 105, 075502 (2010)

Crack-dislocation interactions



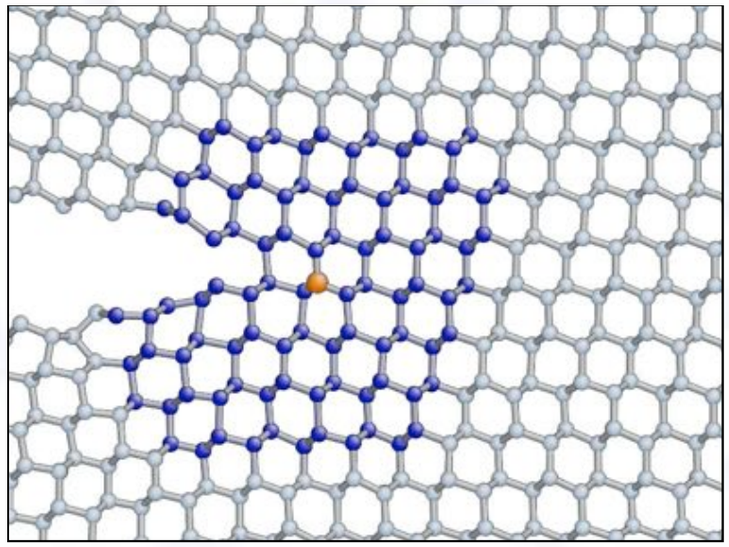
C Gattinoni, JR Kermode and
A De Vita, In prep

Three dimensional effects



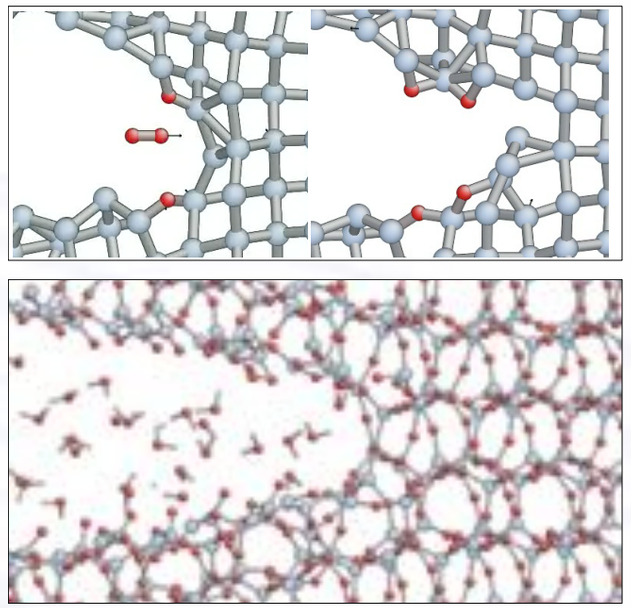
JR Kermode, A. Glazier, G Kovel, L Pastewka,
G Csányi, D Sherman and A. De Vita, In prep

Crack-impurity scattering

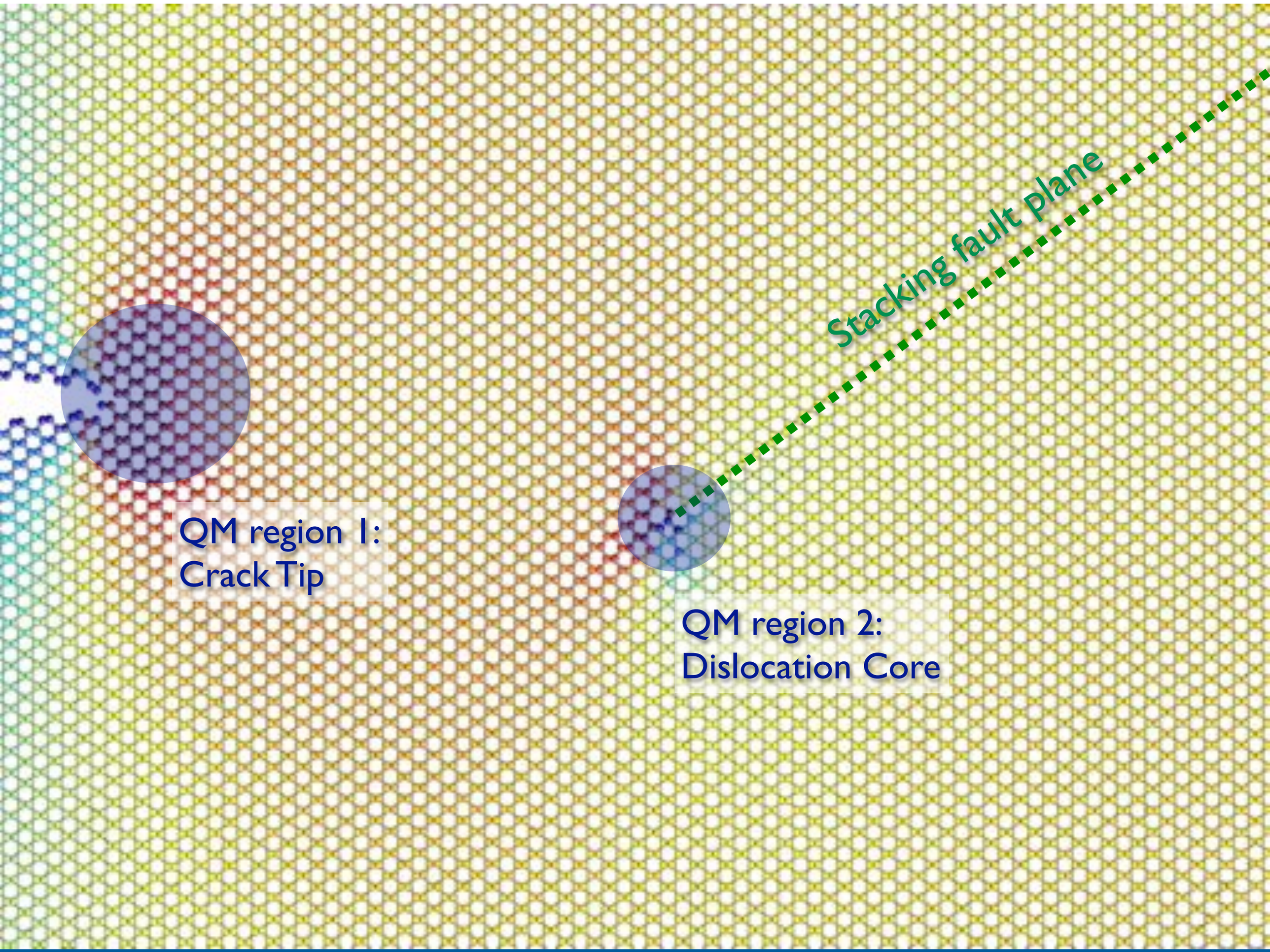


JR Kermode, L Ben-Bashat, F Atrash,
JJ Cilliers, D Sherman and A. De Vita.,
Nat. Commun. 4 2441 (2013)

Stress corrosion cracking

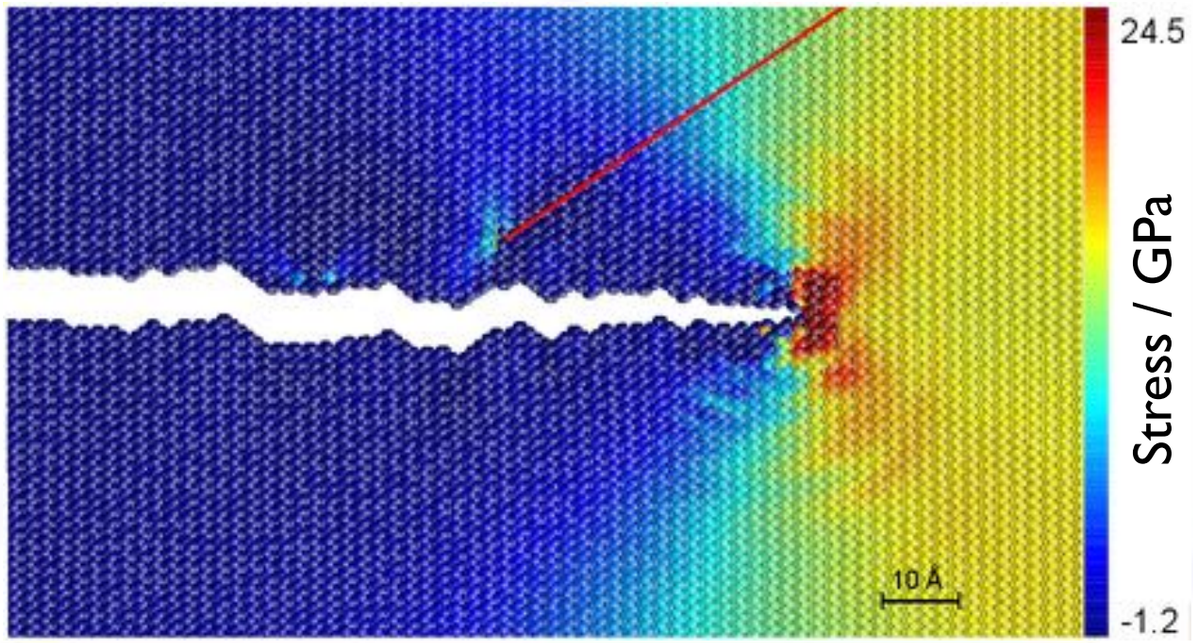


A Glazier, G Peralta, JR Kermode,
A De Vita and D Sherman,
Phys. Rev. Lett, 112 115501 (2014).

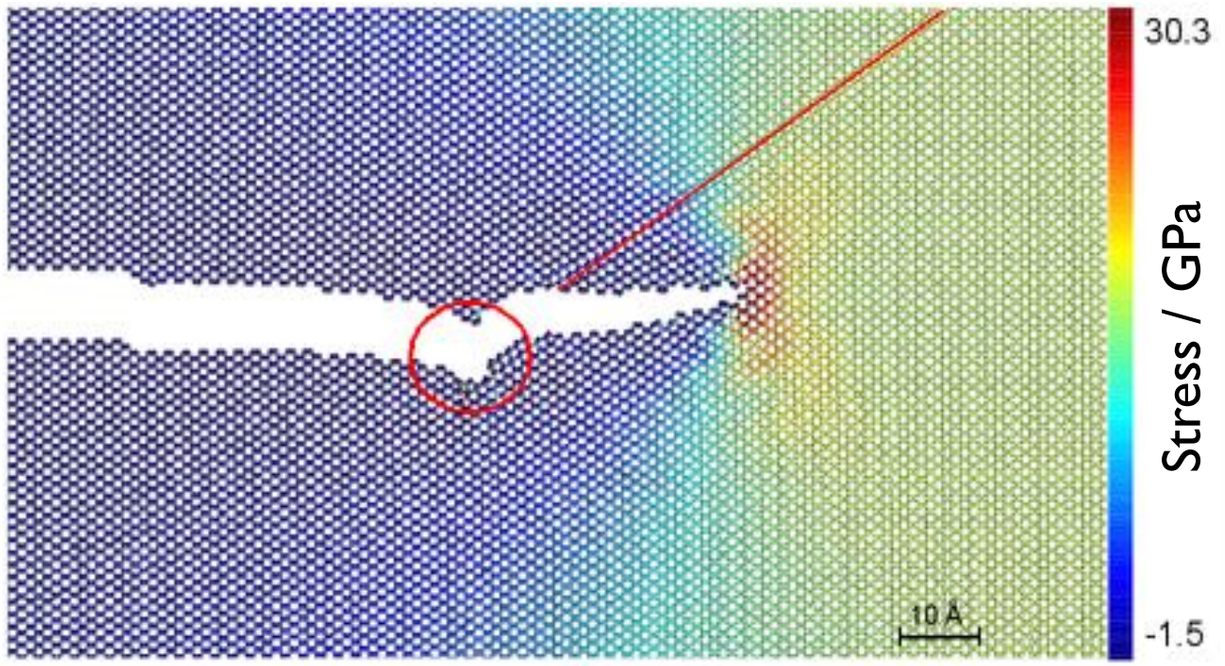

Stacking fault plane
QM region 1:
Crack Tip
QM region 2:
Dislocation Core

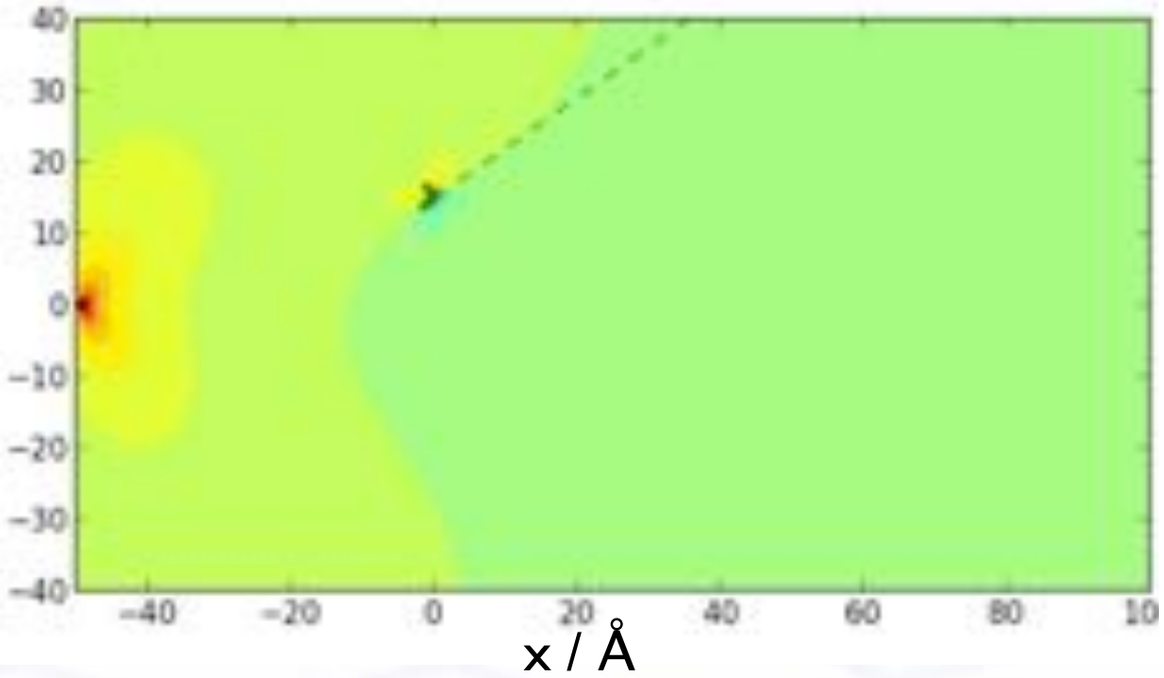
Crack/dislocation – coarse-graining

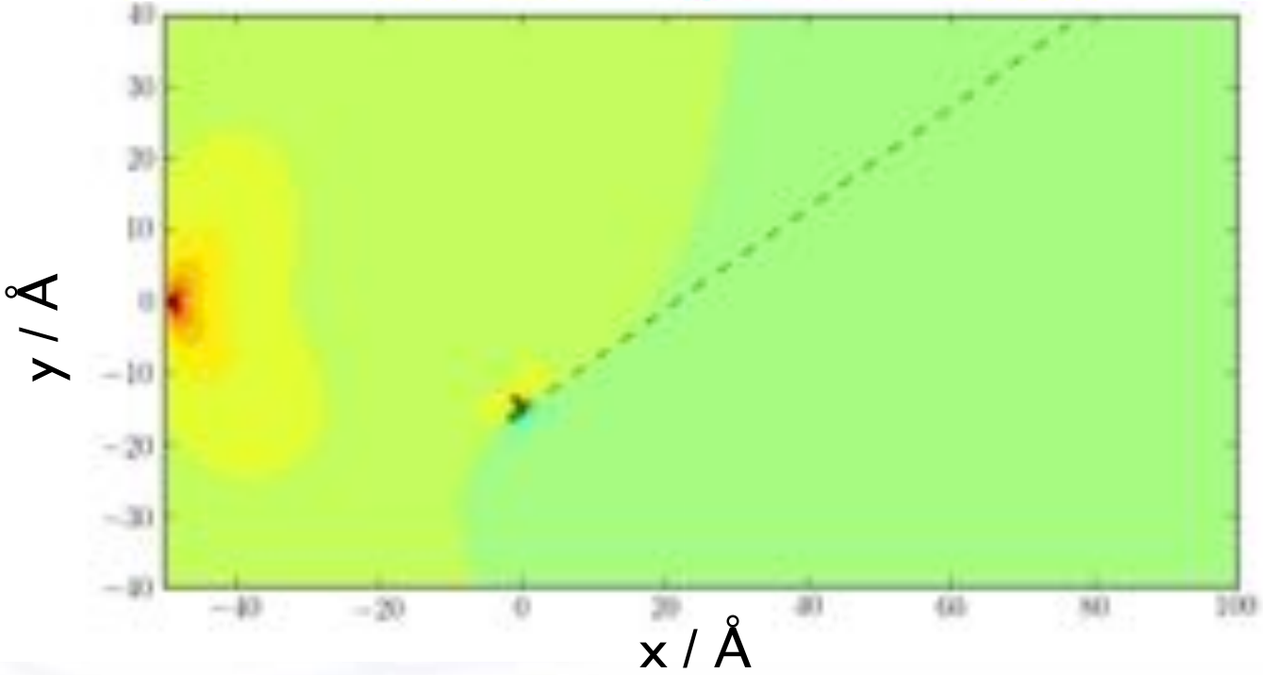
Dislocation core above crack



Dislocation core below crack



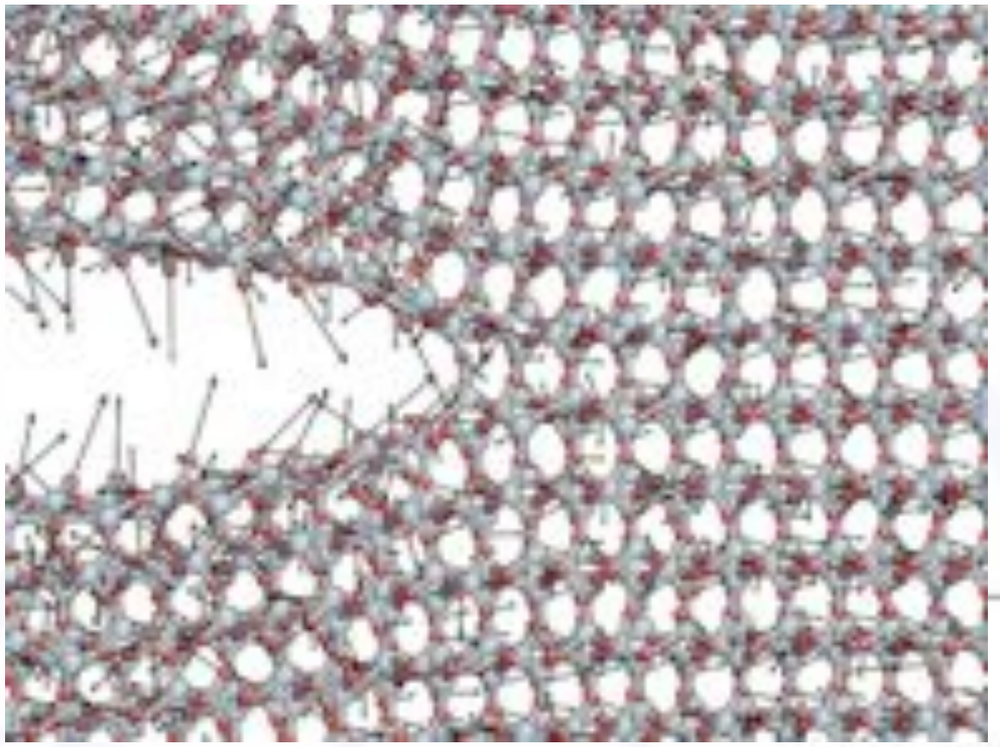




Quantifying loss of information during coarse-graining: uncertainty propagation from atomistic to continuum

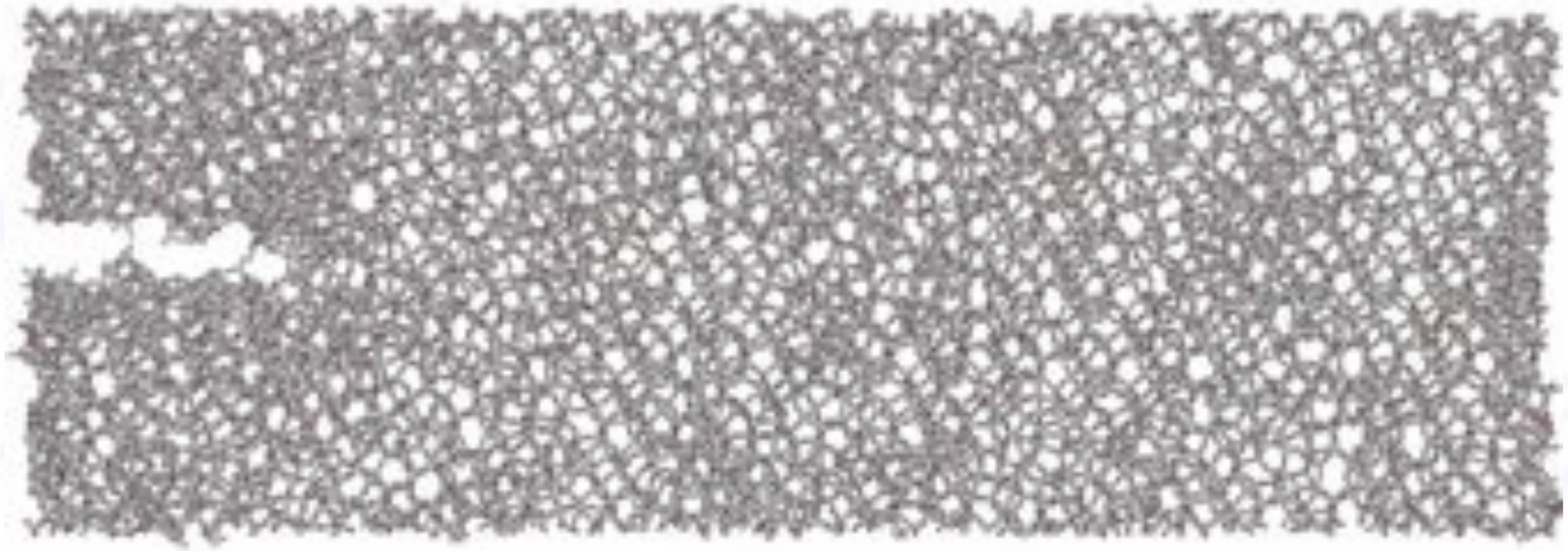
C. Gattinoni, J.R. Kermode and A. De Vita, in prep.

Microstructural Uncertainty



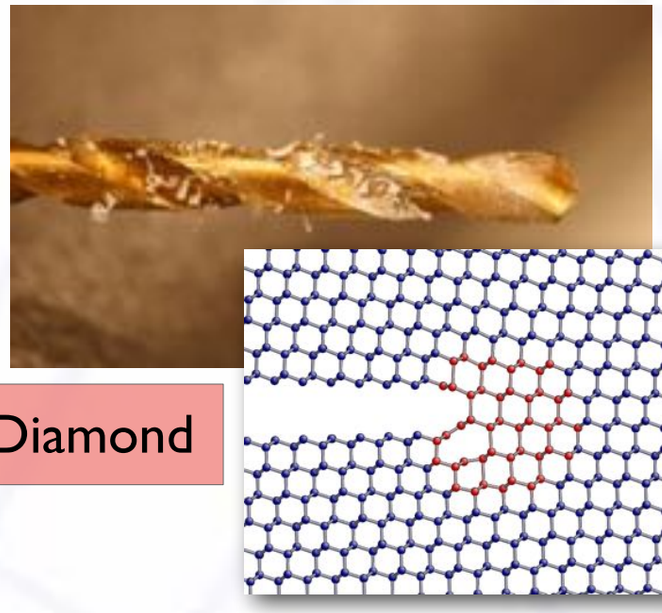
- Most of our work so far has only considered *deterministic* materials failure problems
- *Stochastic* microstructural effects are also extremely important
- e.g. fracture in glass, a prototypical amorphous material, where microstructural variation plays key role in determining fracture response

Goal: average over representative microstructural configurations to produce effective mean-field response. Probabilistic 'error bars' essential.



Target Application Areas

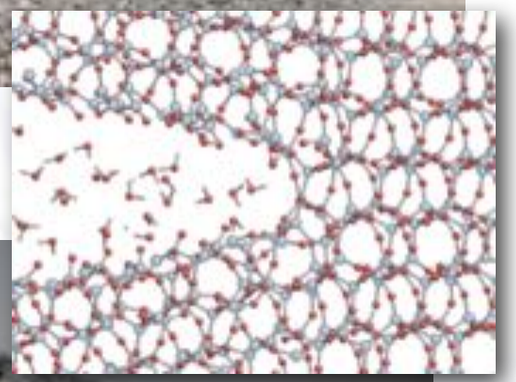
Covalent Materials

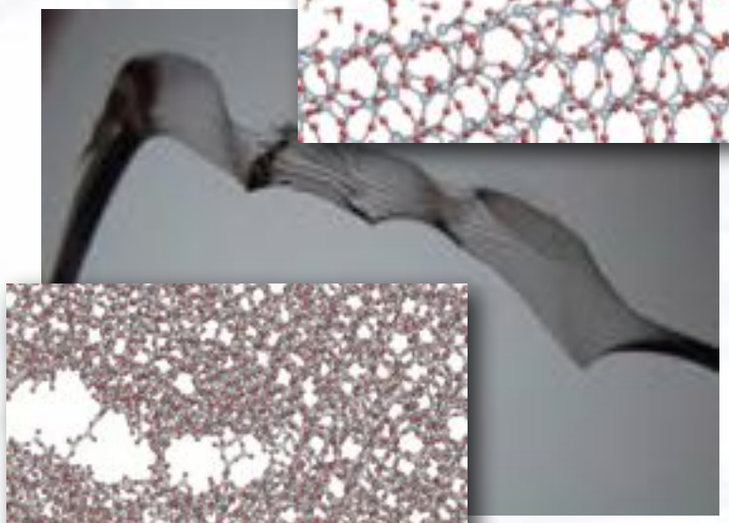


Diamond

Oxides

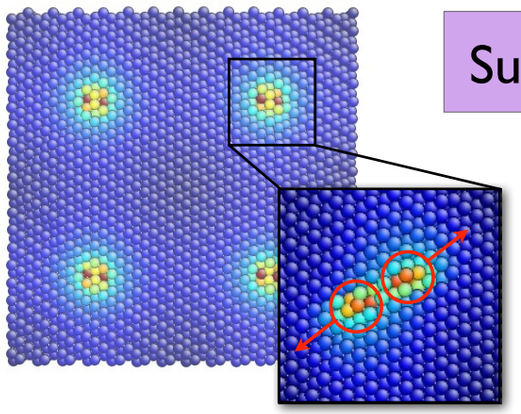
Rocks



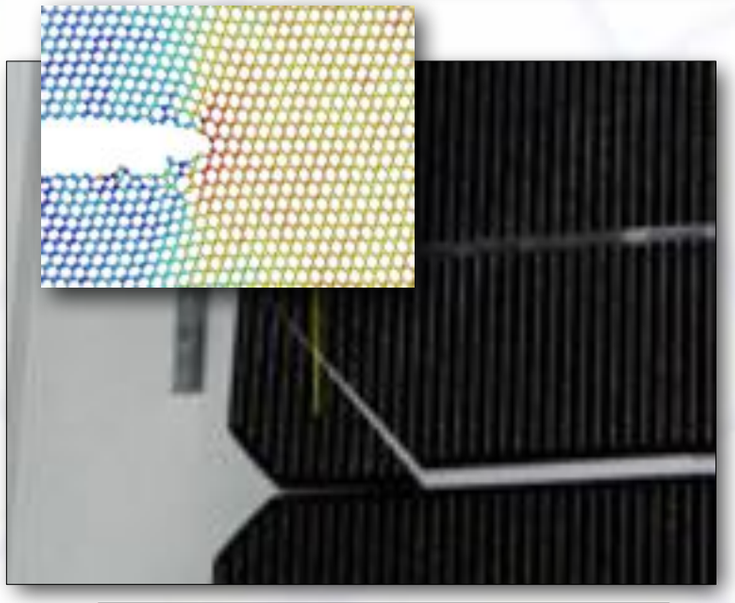


Glass

Metals

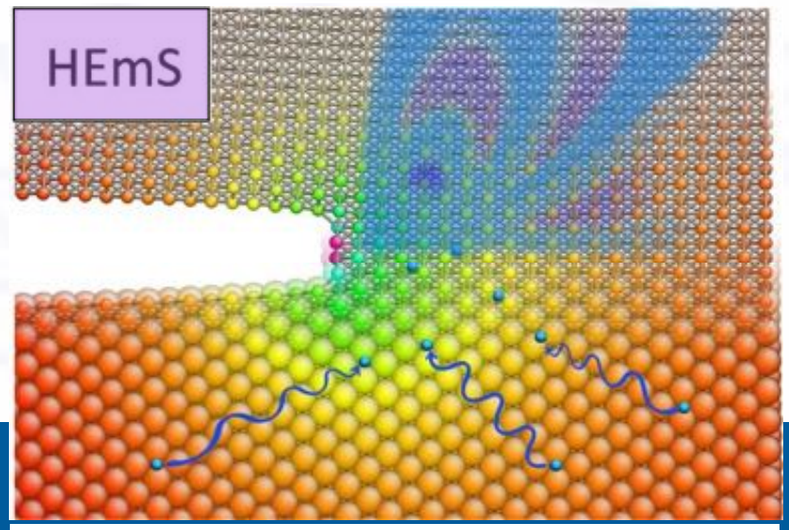


Superalloys



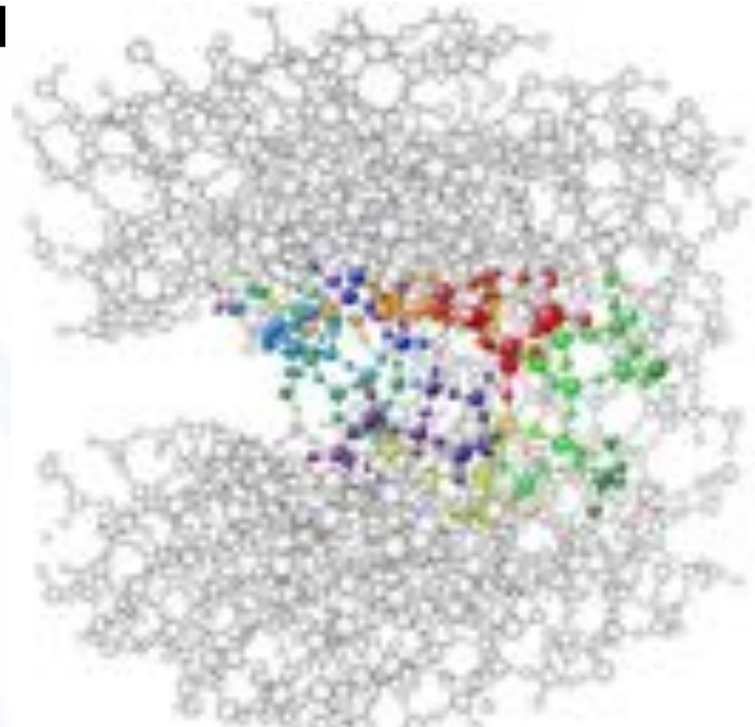
Silicon Photovoltaics

HEmS



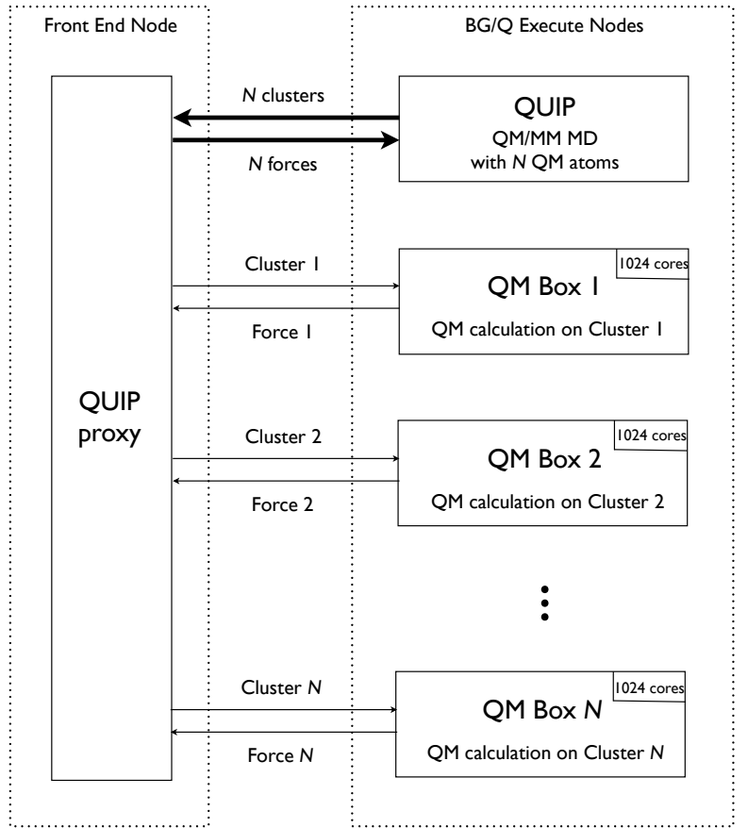
HPC Scalability

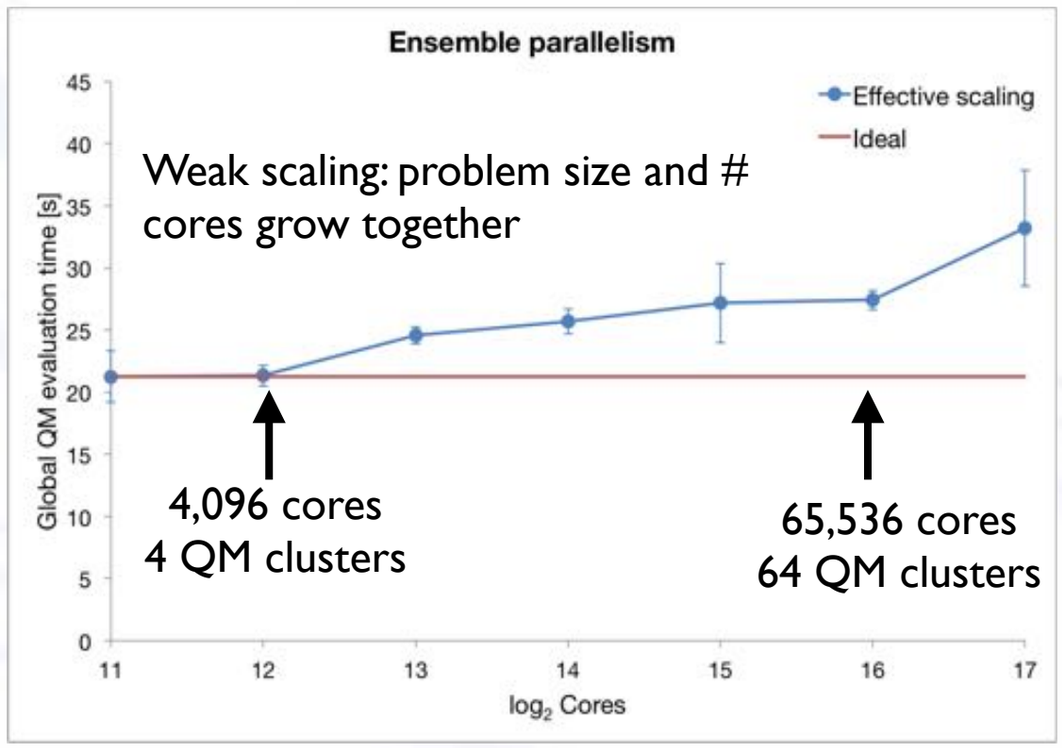
- Force-mixing allows work of QM calculations to be distributed
- Automatically split large QM regions into smaller convex clusters by graph partitioning
- Individual QM calculations run in parallel on ~1024 cores each
- Scalable up to ~100,000 core Petascale machines



U.S. DEPARTMENT OF ENERGY
INCITE
LEADERSHIP COMPUTING

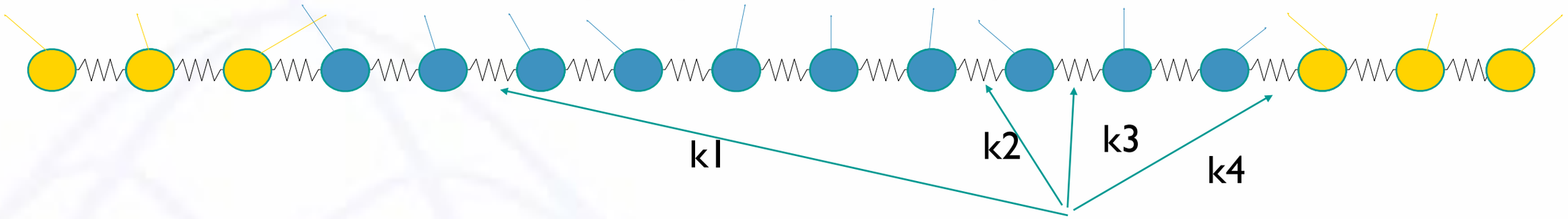
PRACE





‘Learn on the Fly’ predictor/corrector

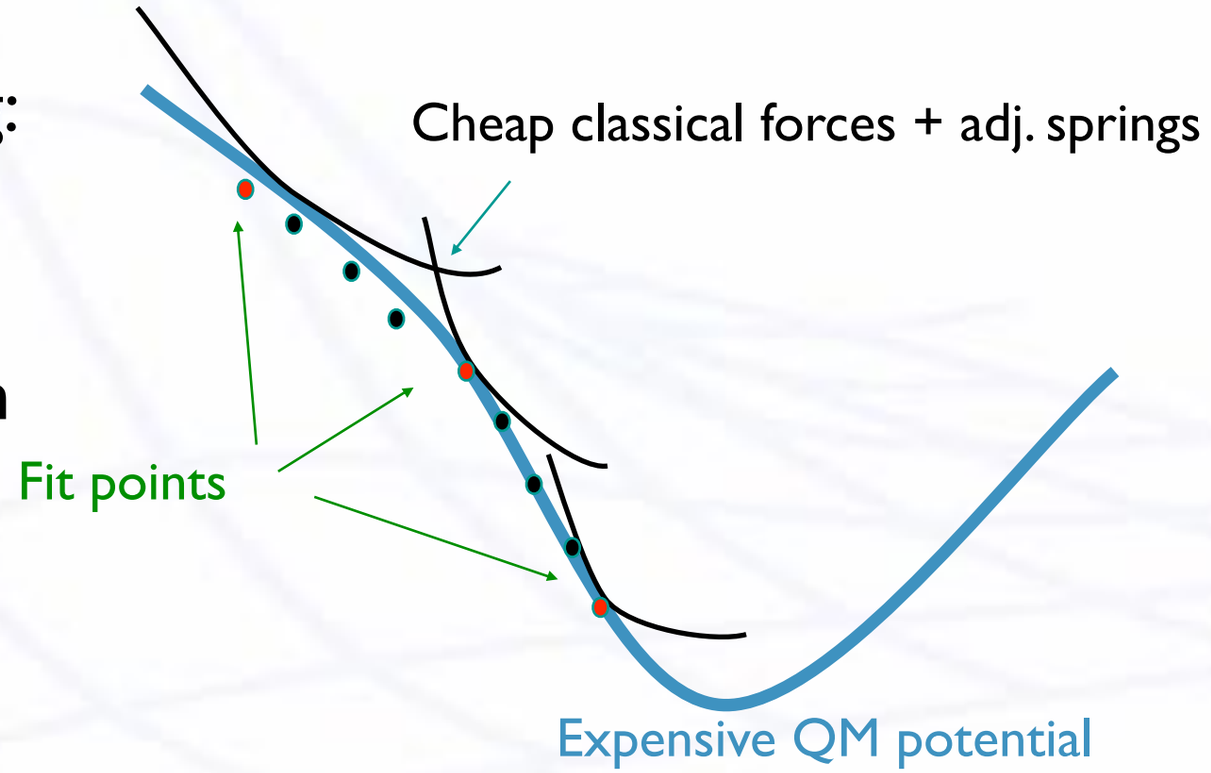
‘Learn on the Fly’ scheme adds ‘springs’ tuned to reproduce QM forces



$$U(\mathbf{R}) = U_{classical}(\mathbf{R}) + \sum_{\text{springs } n} \alpha_n r_n$$

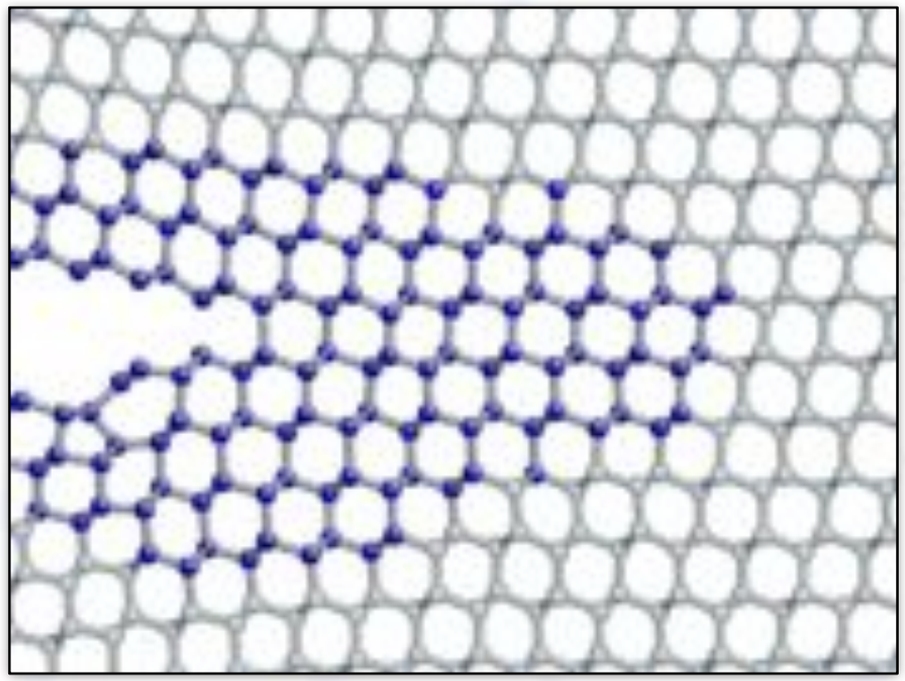
LOTF vs. buffered force mixing:

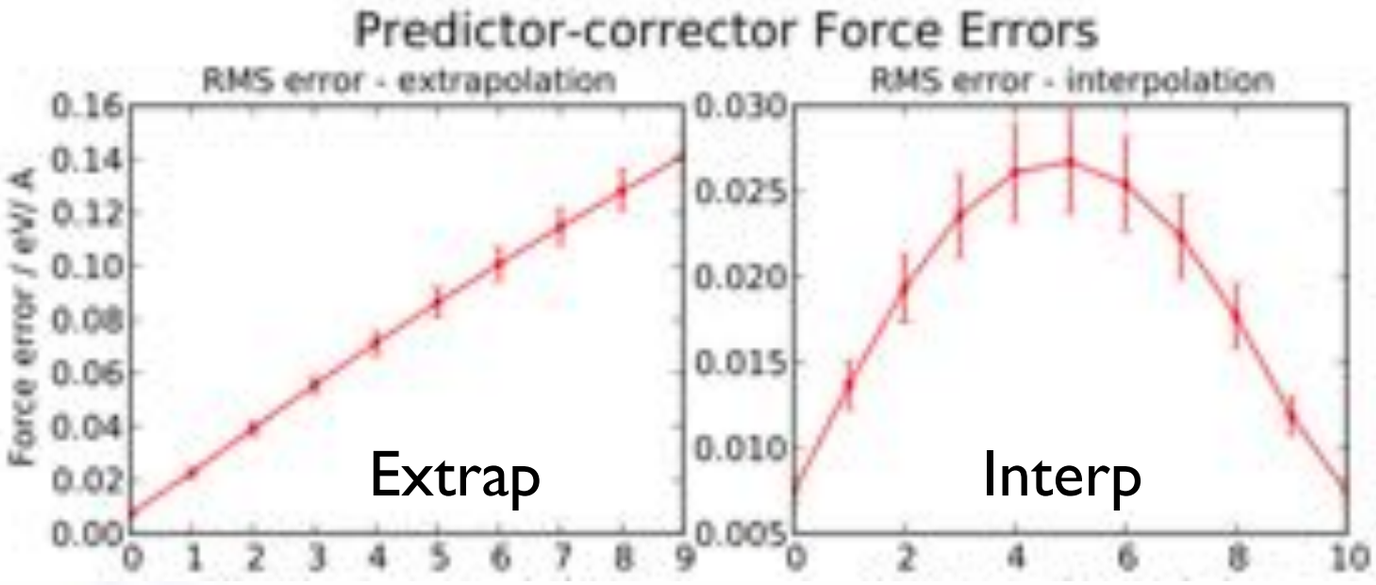
- ✓ Both give correct trajectories
- ✓ LOTF allows speed up with predictor/corrector dynamics

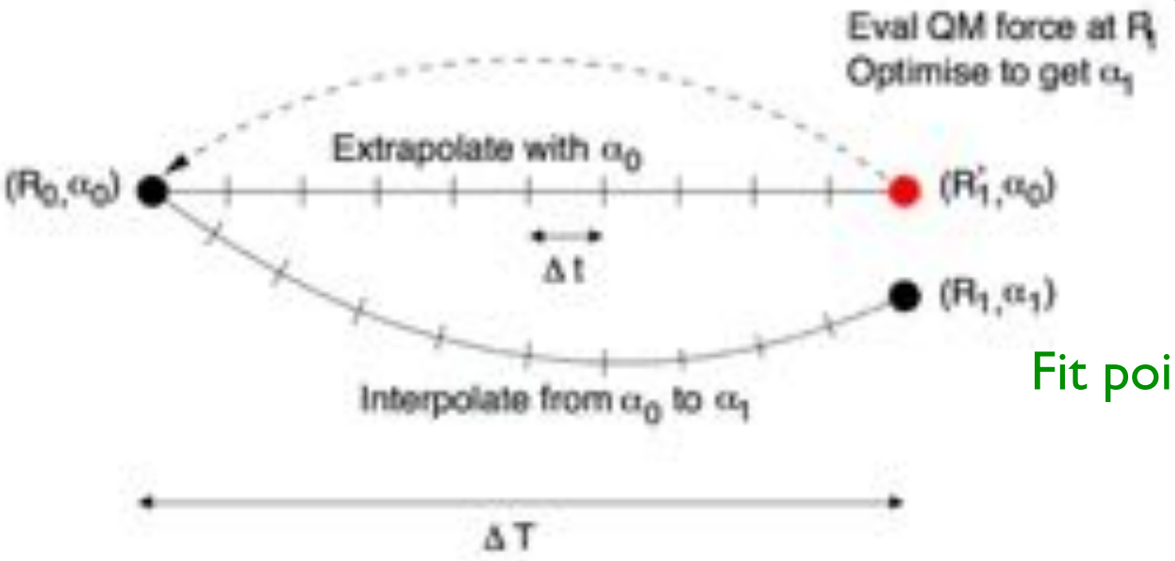


G Csányi et al. Phys. Rev. Lett. 93 175503 (2004)
N Bernstein et al. Rep. Prog. Phys. 72 026501 (2009)

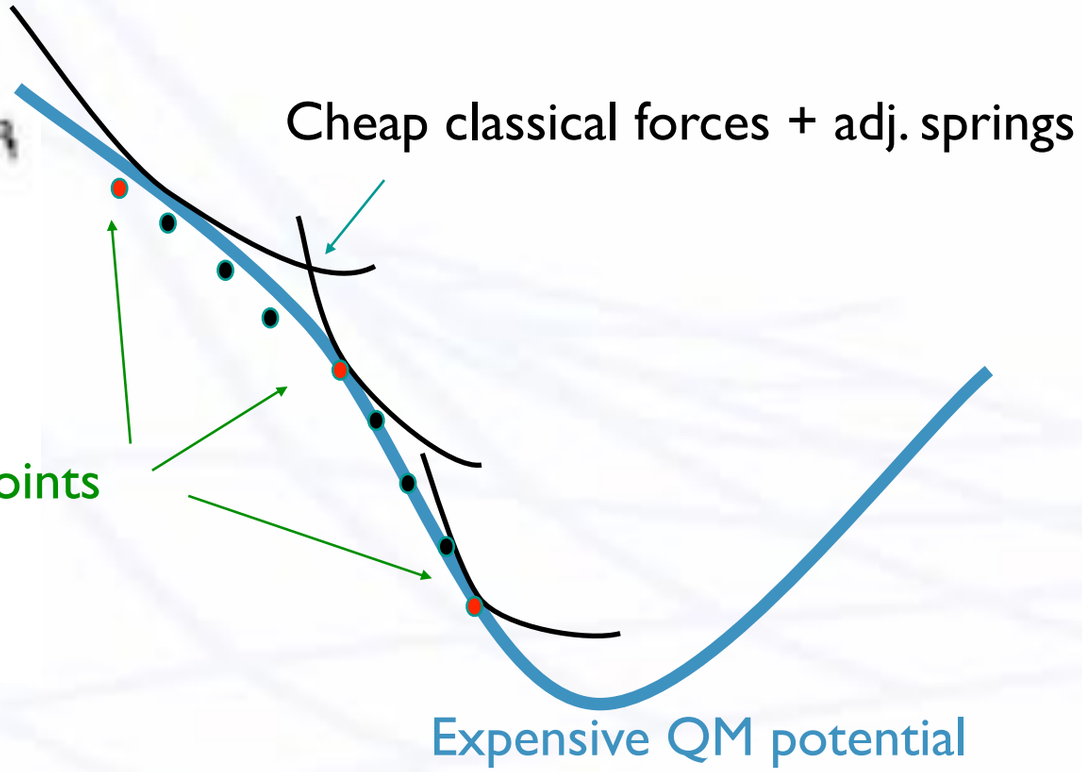
‘Learn on the Fly’ predictor/corrector







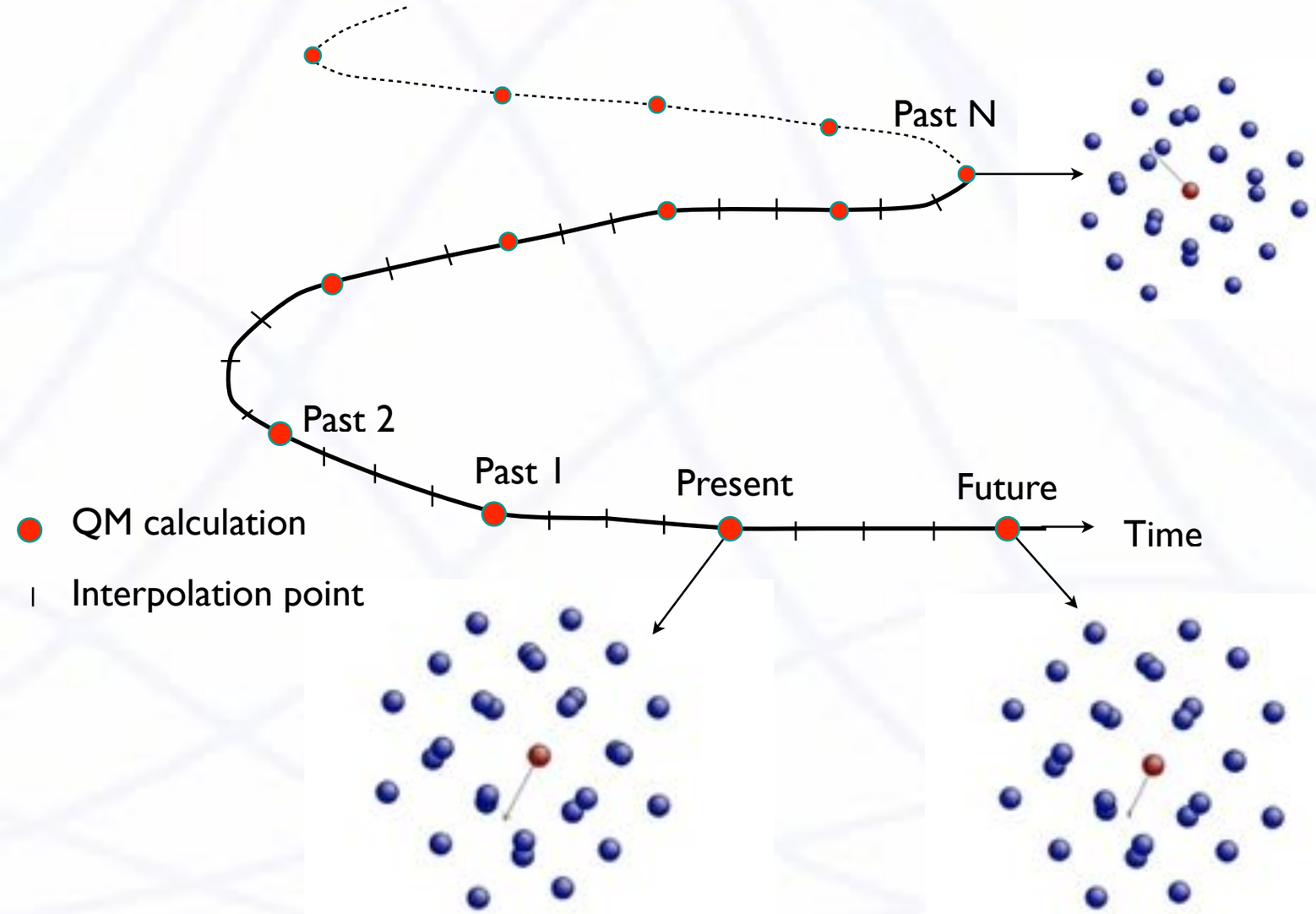
Typically $\Delta T \sim 10\,\Delta t$



G Csányi et al. Phys. Rev. Lett. 93 175503 (2004)
N Bernstein et al. Rep. Prog. Phys. 72 026501 (2009)

Towards Optimal Information Efficiency

- Recent trend of automatic, non-parametric force field construction using Machine Learning techniques (e.g. NNs [1], GAPs [2]), extending force-matching approach
- *Our Idea*: rather than learning whole PES once-and-for-all, use same technique, Gaussian Process regression, to predict QM forces as a function of local atomic environment
- Keeping "in touch" with QM reduces risk of extrapolating outside of fit domain



Accelerate QM/MM with ML-based scheme where forces on atoms are:

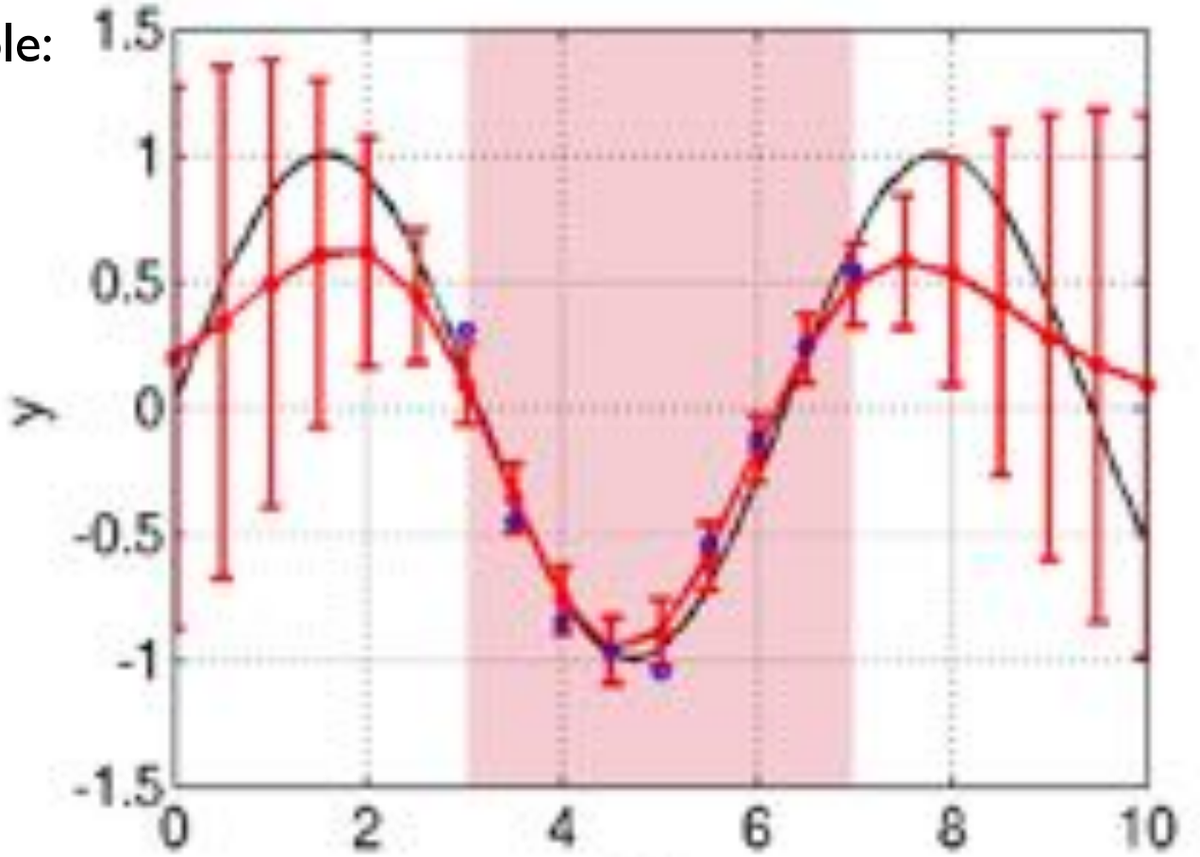
- *Either* predicted using Gaussian Process regression
- *Or* computed on-the-fly and added to growing ML database

Z Li, JR Kermode and A De Vita, Phys. Rev. Lett, In Press (2015)

Gaussian Process Regression

Gaussian Process (GP) regression: use Bayesian inference to compute most likely function values given data and a prior distribution for model parameters

1D Example:



$$\text{Posterior} \quad p(\text{parameters}|\text{data}) \propto L(\text{data}|\text{parameters})\, p(\text{parameters})$$

Likelihood · Prior

Black: function f(x)

Blue: observations
yi = f(xi)

Red: GP mean, std. dev.

Covariance (smoothness of prior)

$$C_{mn} = \exp\left(-\frac{d_{mn}^2}{2\sigma_{\text{cov}}^2}\right) + \delta_{mn}\sigma_{\text{err}}^2.$$

Posterior mean $y_{N+1} = \mathbf{k}^T\mathbf{C}^{-1}\mathbf{y}$

Posterior variance $\sigma_{y,N+1}^2 = \kappa - \mathbf{k}^T\mathbf{C}^{-1}\mathbf{k}$

Covariance matrix **C**
Feature vector **k**
Self-covariance κ

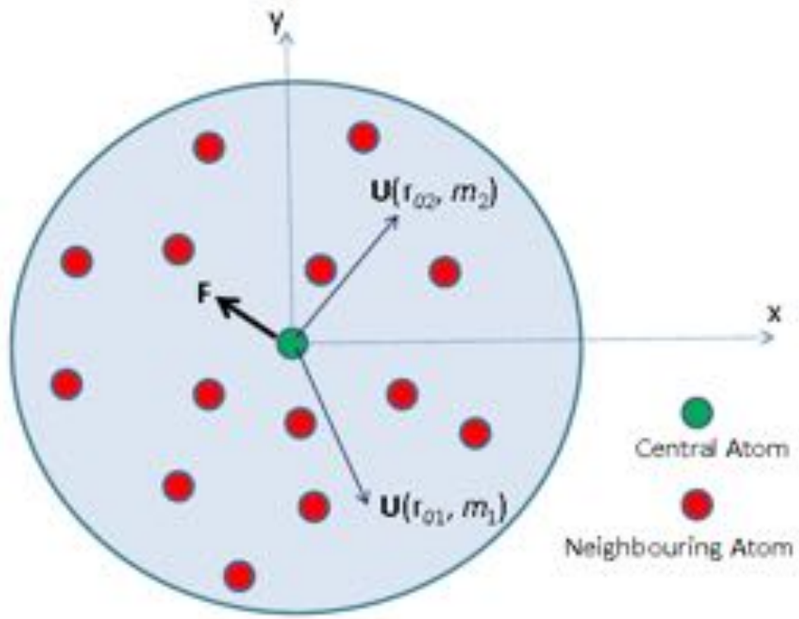
Describing Atomic Environments

Application to force learning: compute *most probable* force given database of atomic environments and associated QM forces (plus a prior assumption that forces vary smoothly as atoms move).

Requires a measure of "similarity for force learning" of two atomic configs, invariant to *translation*, *permutation* and *rotation*. Complication: forces are vectors!

Internal vectors ($i = 1 \ldots k$)

$$\mathbf{V}_i = \sum_{q=1}^{N_{\mathrm{neighb}}} \hat{\mathbf{r}}_q \exp\left[-\left(\frac{r_q}{r_{\mathrm{cut}}(i)}\right)^{p(i)}\right]$$



Feature matrix

$$X_{ij} = \mathbf{V}_i \cdot \hat{\mathbf{V}}_j = (V A^T)_{ij}$$

$$V^T = \begin{pmatrix} | & | & | \\ \mathbf{V}_1 & \ldots & \mathbf{V_k} \\ | & | & | \end{pmatrix} \quad A^T = \begin{pmatrix} | & | & | \\ \hat{\mathbf{V}}_1 & \ldots & \hat{\mathbf{V}}_k \\ | & | & | \end{pmatrix}$$

Distance metric $$d^2_{mn} = \frac{1}{k}\sum_{i,j=1}^{k}\left[\frac{X^m_{ij}}{\chi_i} - \frac{X^n_{ij}}{\chi_i}\right]^2$$

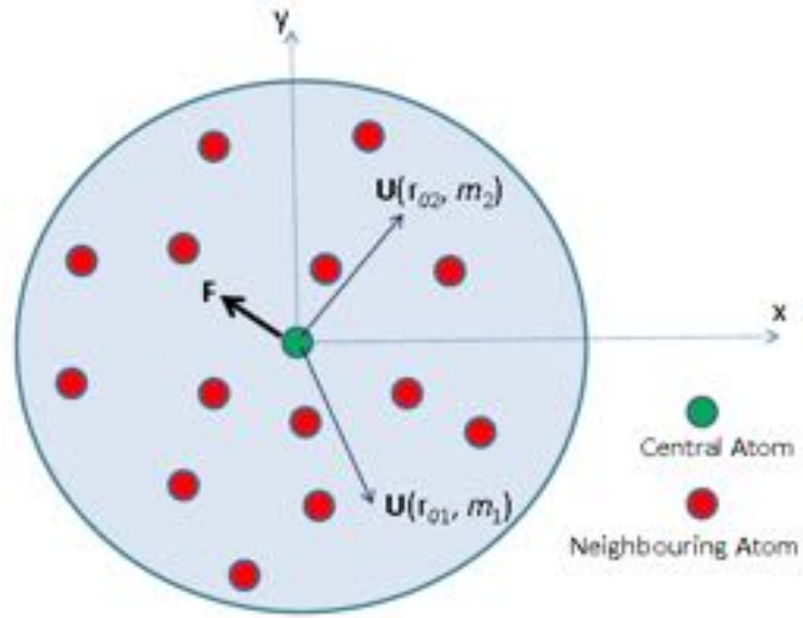
Scale factors over database:

$$\chi^2_i = \sum_{n,m=1}^{N}\sum_{j=1}^{k}\frac{(X^m_{ij} - X^n_{ij})^2}{N^2}.$$

Covariance $$\mathrm{Cov}(m,n) = \exp\left(-\frac{d^2_{mn}}{2\sigma^2_{\mathrm{cov}}}\right) + \delta_{mn}\sigma^2_{\mathrm{err}}$$

"Curse of dimensionality" – QM is long range

- Forces depend mostly on atoms within cutoff ~10 A from central atom
- Project target forces into internal coordinate system (dim. k ~ 10-20)
- Perform GP regression there
- Overdetermined system - transform back to find 'best' absolute Cartesian reference frame in least squares sense

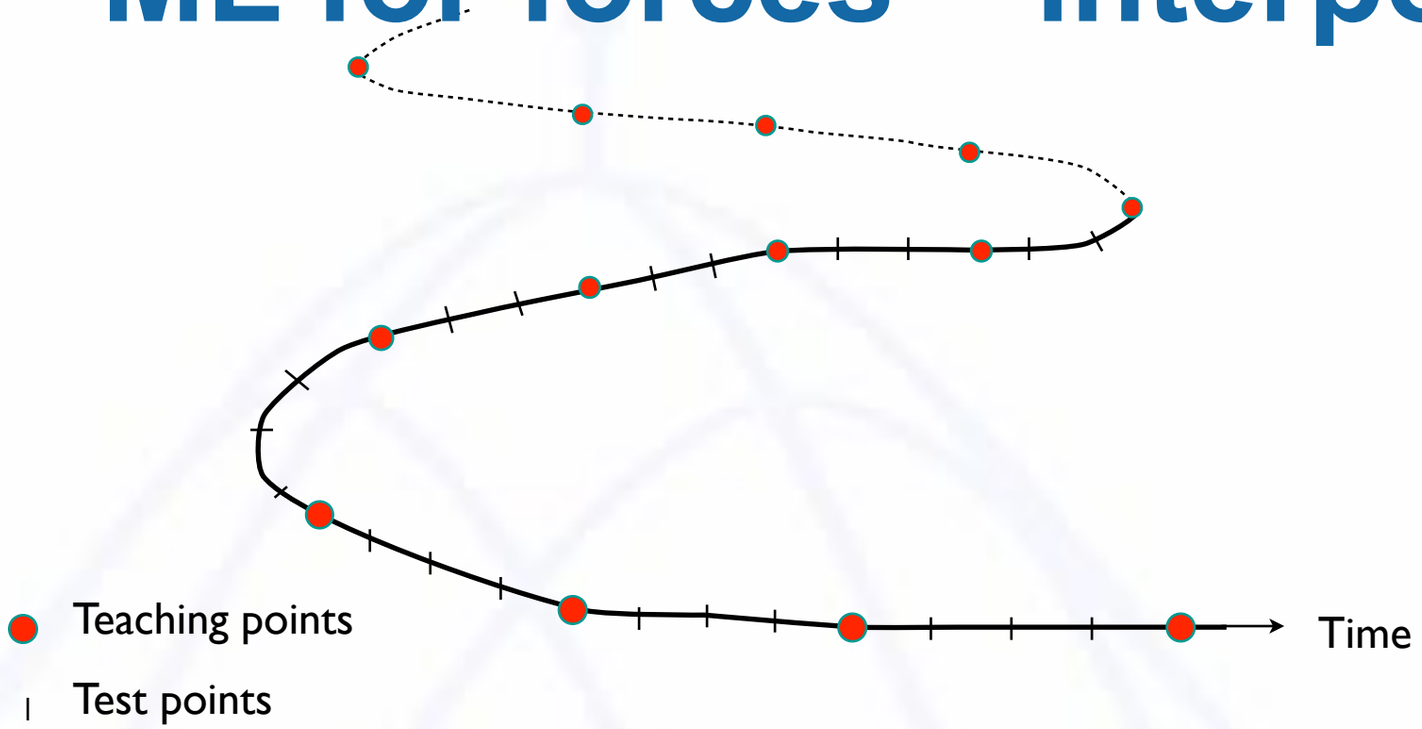


$$\mathcal{F} = A\mathbf{F}$$

$$A^+ = \left(A^T A\right)^{-1} A^T$$

$$\mathbf{F} = A^+ \mathcal{F}$$

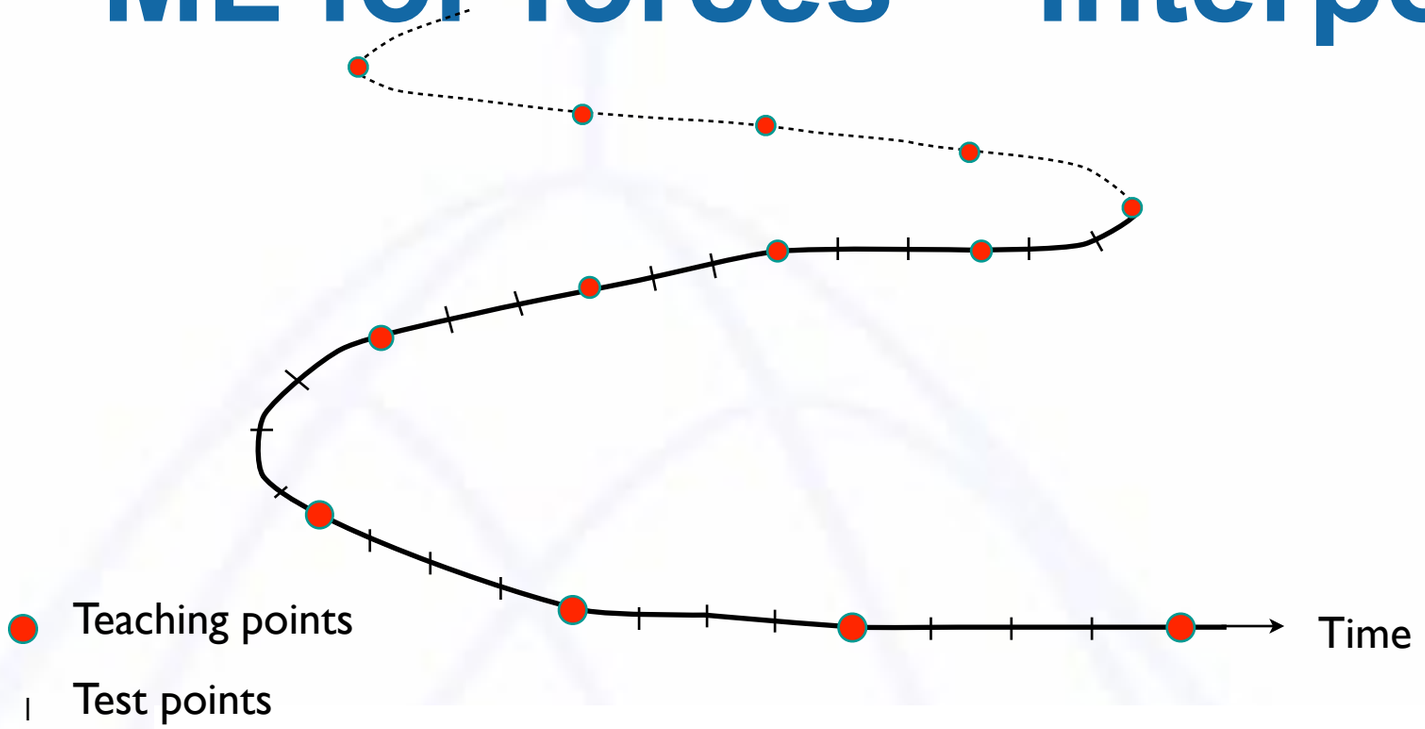
ML for forces – interpolation accuracy

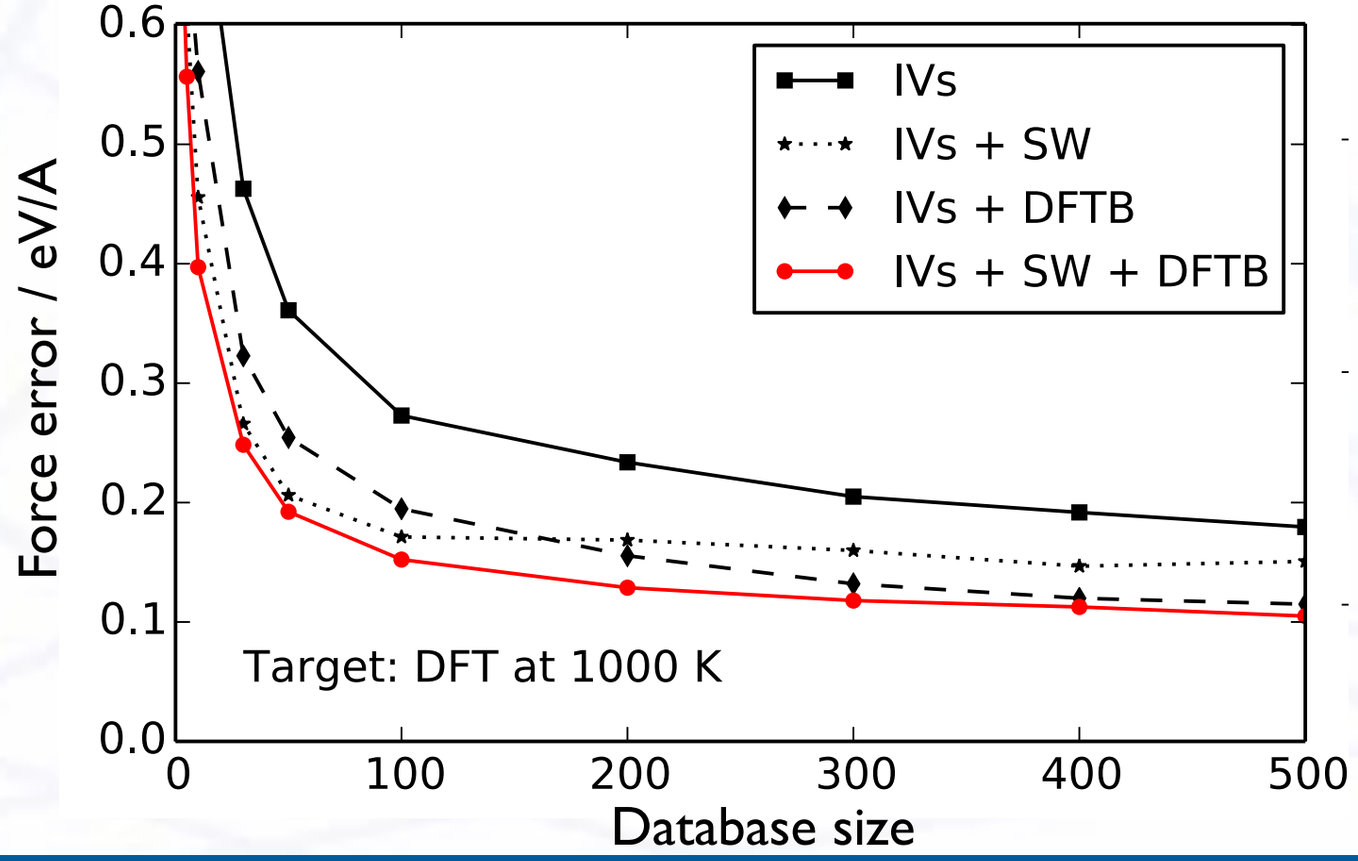


Teaching database built from ~2000 configs at 20 fs intervals along a reference DFT MD trajectory in Si at 1000 K

Measure *interpolation* ability: test configs midway between teaching points

ML for forces – interpolation accuracy





Teaching database built from ~2000 configs at 20 fs intervals along a reference DFT MD trajectory in Si at 1000 K

Measure *interpolation* ability: test configs midway between teaching points

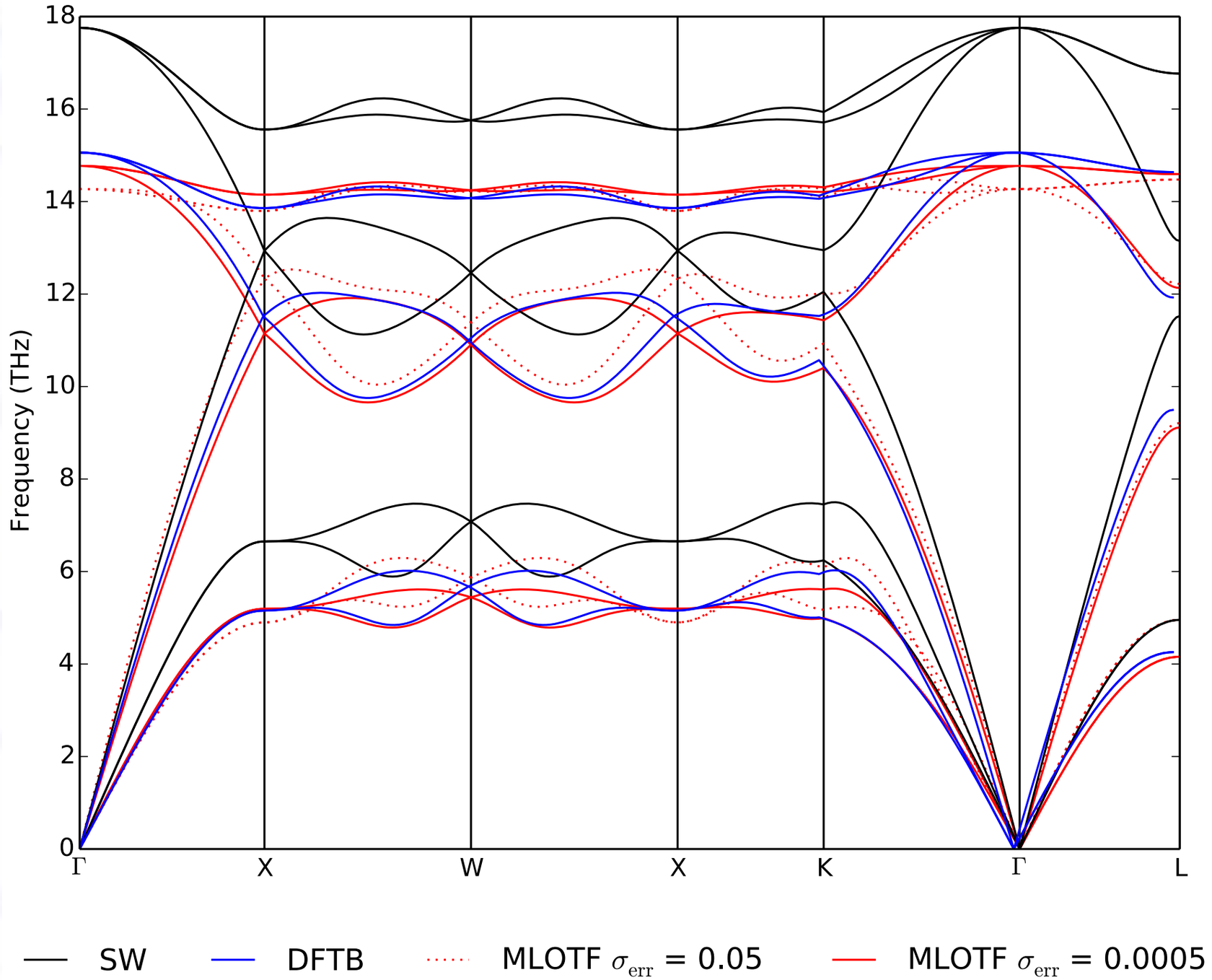
Sorting by distance and *selecting* only ~500 most relevant environments sufficient to predict accurate forces at test points

Augmenting descriptor IVs with classical potential force vectors significantly improves accuracy

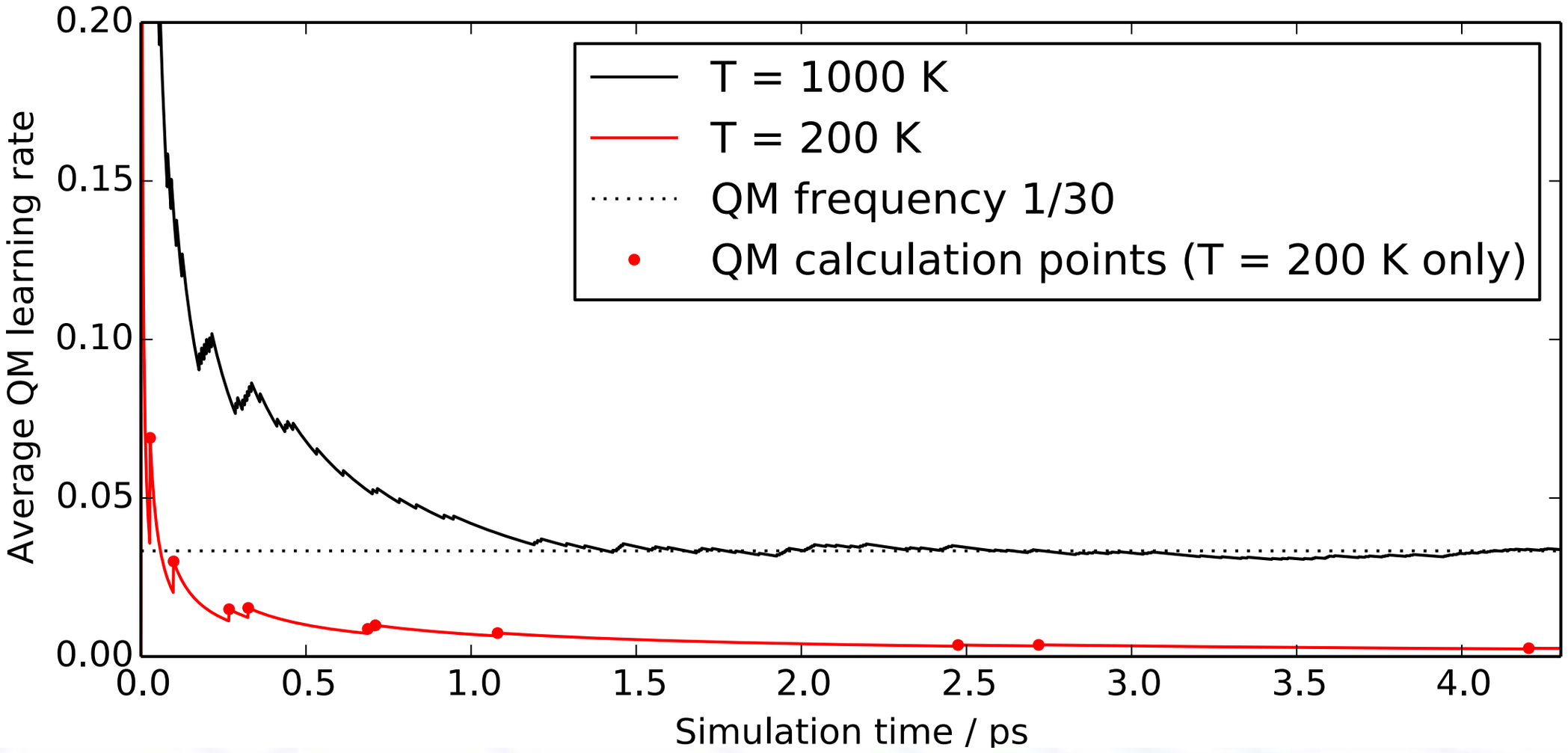
~6% force error wrt DFT

High Accuracy Forces: Phonon Spectrum

Computed using forces learnt from 300 K MD trajectory (DFTB)

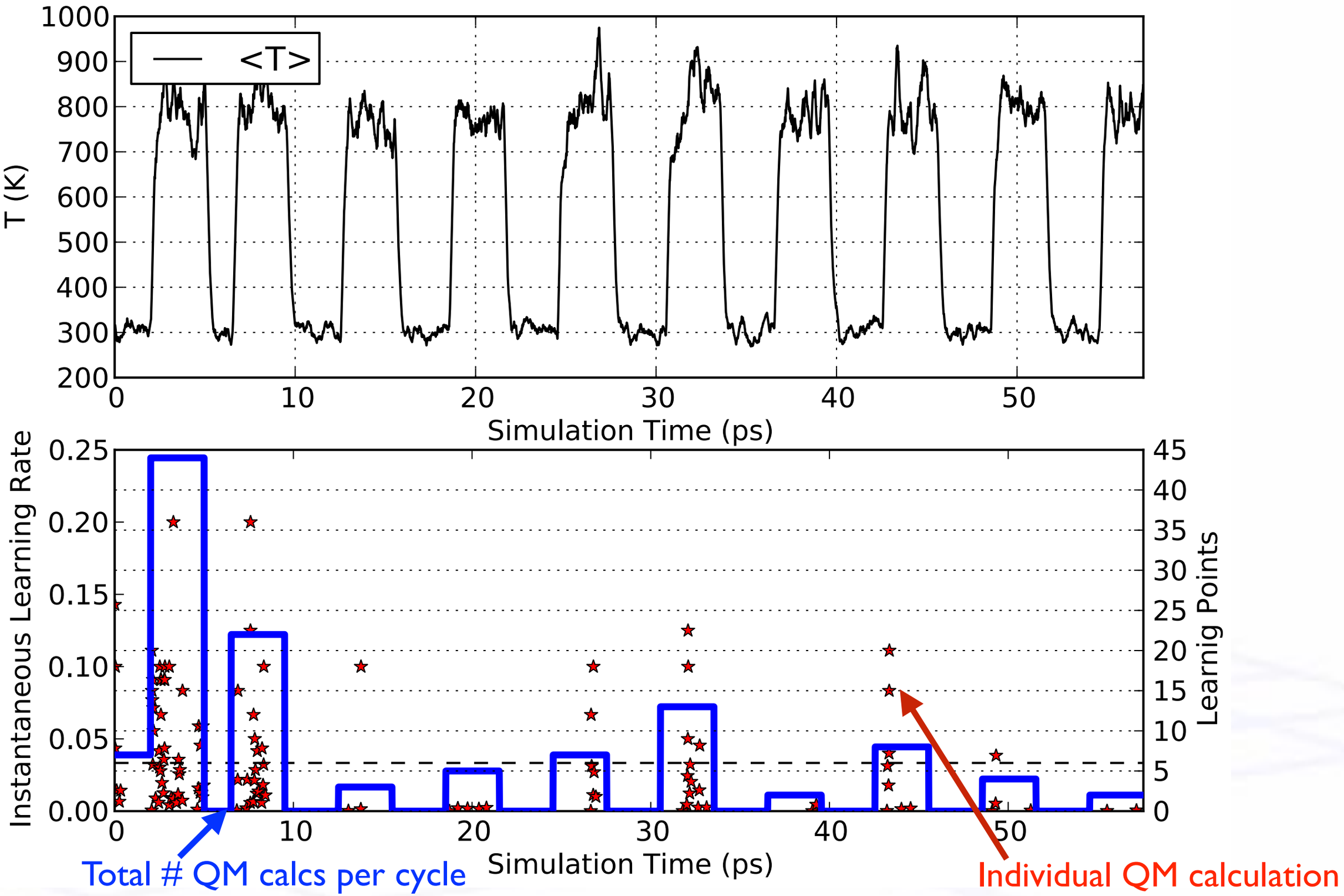


On-the-fly Machine Learning of QM Forces



- 64 atom bulk Si test system; QM forces evaluated every step for testing
- Configs added to database when average force error > 0.1 eV/A
- *Learning rate* provides measure of complexity for MD force learning - 'chemical novelty'
- *T* = 200 K – long periods (> 1 ps) where no QM is necessary
- *T* = 1000 K – QM calculation rate of 1/30 sufficient after initial phase

Quantifying Chemical Novelty



Z Li, JR Kermode and A De Vita, Phys. Rev. Lett, In Press (2015)

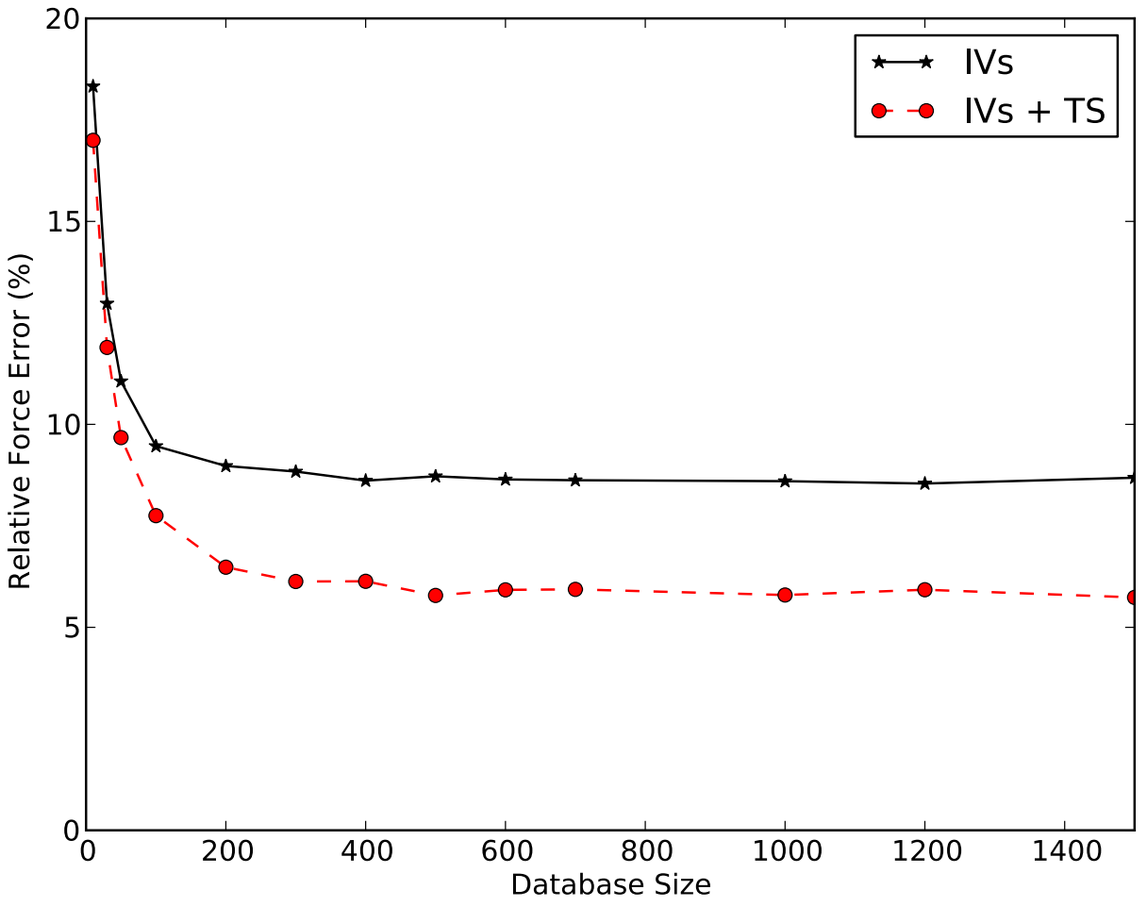
Extension to Multi-species Systems

Internal vectors

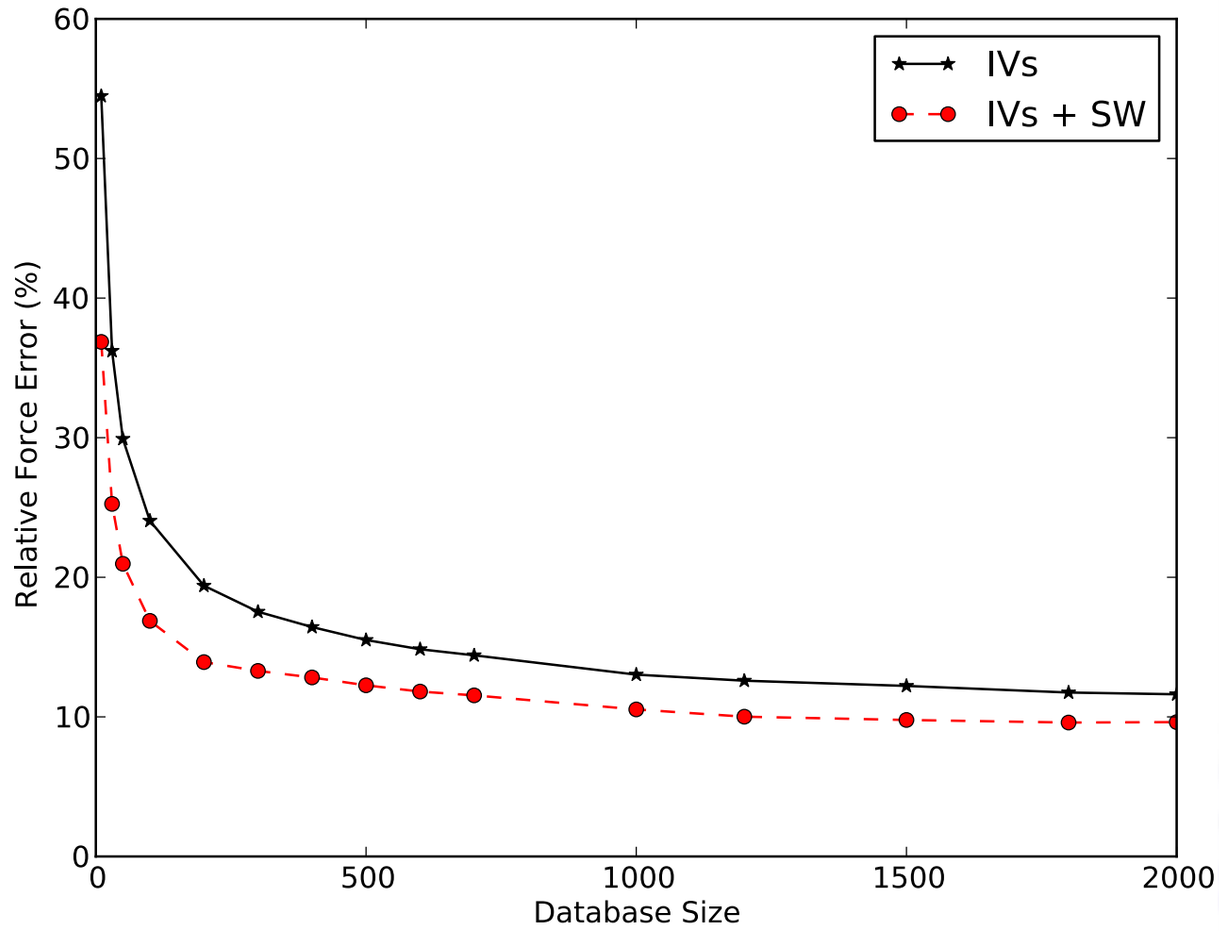
$$\mathbf{V}_i^{(s)} = \sum_{q=1}^{N_{\text{neighb}}} \hat{\mathbf{r}}_{sq} \exp\left[-\left(\frac{r_{sq}}{r_{\text{cut}}(i)}\right)^{p(i)}\right] \delta_{Z_s,Z_q}$$

Block feature matrix

$$\mathbf{X} = \left(\begin{array}{c|c} \mathbf{V}_{Z_1}\mathbf{A}_{Z_1}^T & \mathbf{V}_{Z_1}\mathbf{A}_{Z_2}^T \\ \hline \mathbf{V}_{Z_2}\mathbf{A}_{Z_1}^T & \mathbf{V}_{Z_2}\mathbf{A}_{Z_2}^T \end{array}\right)$$



Amorphous SiO_2, target DFT



SiC, target DFTB

Summary

- **Hybrid QM/MM approaches allow 'chemomechanical' materials failure problems to be modelled with QM precision**
  - Addresses lengthscale limitations of QM approaches e.g. DFT
  - Practical: e.g., experimentally verifiable predictions of fracture phenomena
  - Timescale limitations – prohibitive to model many important processes
- **On-the-fly Machine Learning of QM forces improves timescales accessible**
  - Target optimal information efficiency – only do QM when necessary
  - Need representation of atomic environments suitable for force learning
  - Progressively fewer QM calculations are required when same chemical process is encountered again
  - Chemical 'novelty' and uncertainty can be quantified
- **Next Steps**
  - Improved atomic environment descriptors, e.g. vector covariance kernel
  - Use GP variance predictions to quantify uncertainties
  - Integrate on-the-fly ML and hybrid QM/MM approaches
  - Propagation of uncertainties through coarse-graining

Acknowledgments

Federico Bianchini, Marco Caccin,
Zhenwei Li and Alessandro De Vita

Albert Bartok-Partay, Gábor Csányi and Mike Payne

Anke Peguiron, Gianpietro Moras, Lars Pastewka and Peter Gumbsch

Gaurav Singh and Robert Zimmerman

Chiara Gattinoni

Noam Bernstein

Financial support and supercomputing resources: