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Uncertainty quantification molecular dynamics simulations

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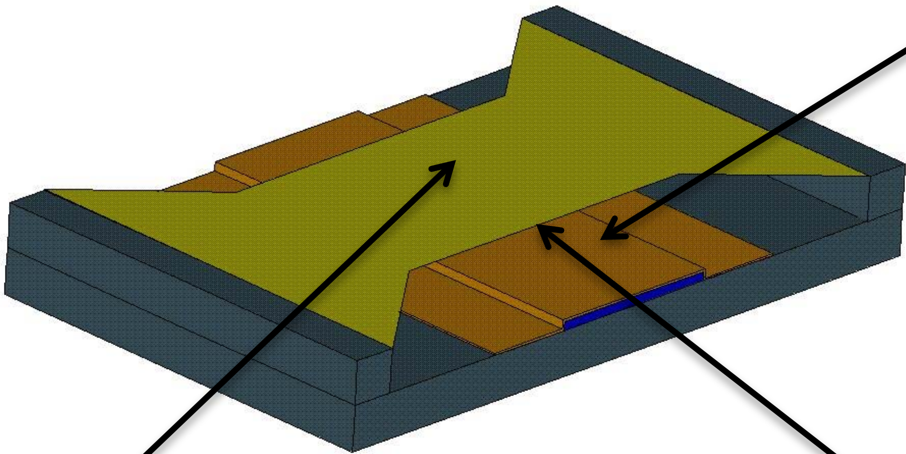
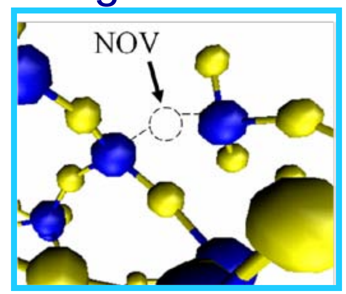
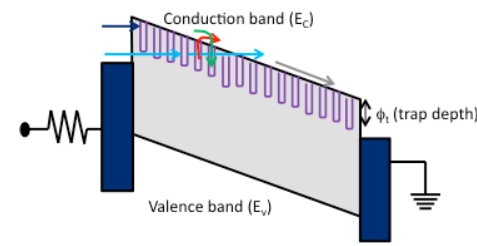
Atomistic simulations in PRISM



Dielectric Charging

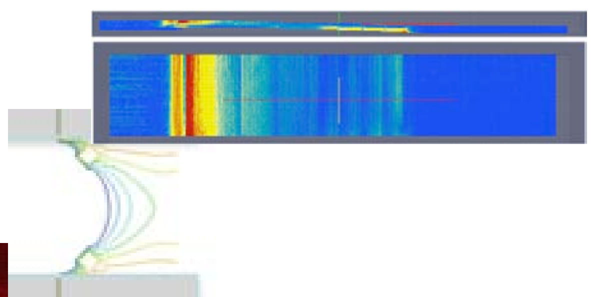
•Trapped charges cause uncontrollable changes in actuation voltage

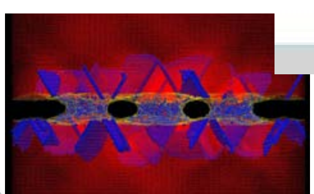




Solid mechanics

•Creep and stress relaxation, size effects

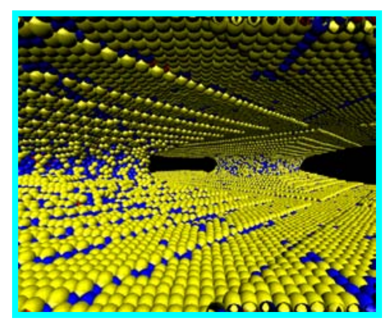


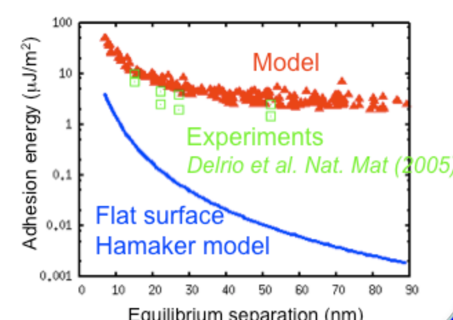


Defect properties: vacancies, dislocations, GBs

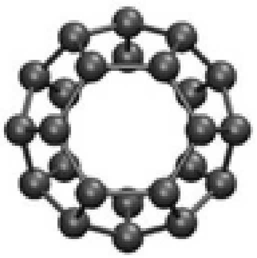
Contact physics

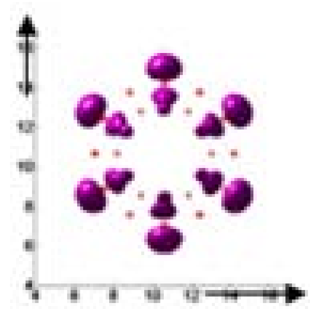
•Surface interaction, adhesion & stiction
•Contact hardness & strength
•Sub-surface defects & surface chemistry





Atomic level simulations of materials





Wave function of electrons

$$\psi(\{r_i\})$$

$$\left|\psi(\{r_i\})\right|^2$$

Probability density of finding electron 1 at r_1, electron 2 at r_2, etc.

Wave function is obtained from the Schrodinger equation:

$$H(\{R_i\})\psi(\{r_i\};\{R_i\}) = E(\{R_i\})\psi(\{r_i\};\{R_i\})$$

Molecular dynamics simulations

MD involves numerically solving Newton's equations of motion

$$\dot{R}_i = \frac{P_i}{M_i}$$

$$\dot{P}_i = F_i$$

$$F_i = -\nabla_{R_i} E(\{R_i\})$$

Ab initio MD (forces & energy from QM)

MD with empirical potentials

Replace energy obtained from the Schrodinger equation $E(\{R_i\})$

with an empirical function that approximates its behavior:

Material specific force field parameters

$$U_{FF}(\{R_i\};\{C_i\})$$

Atomic positions

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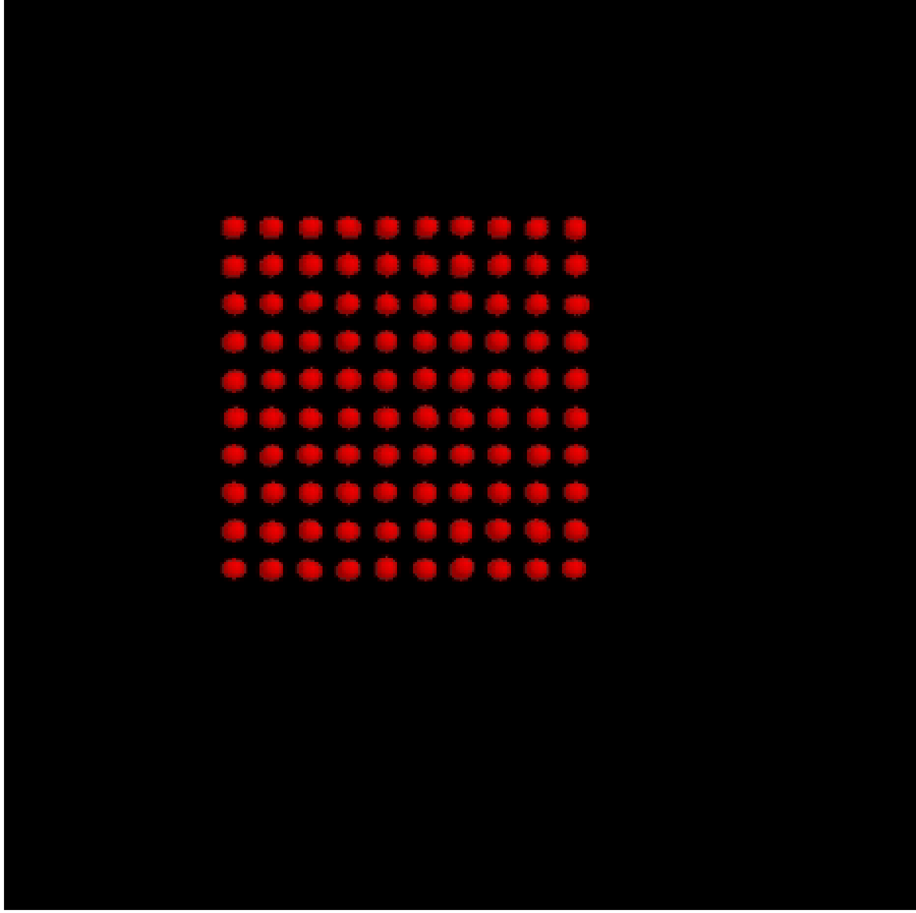
A simple MD run

Use stat mech to relate microscopic world to thermodynamical properties

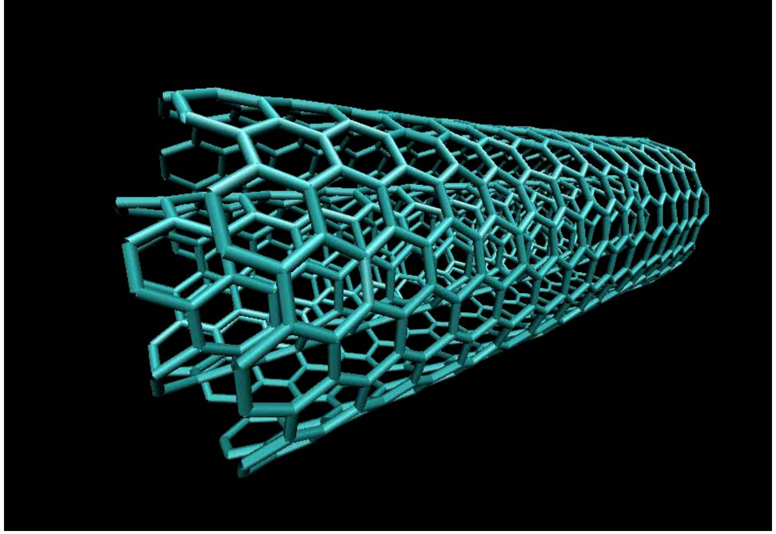


Temperature ~ kinetic energy per DoF

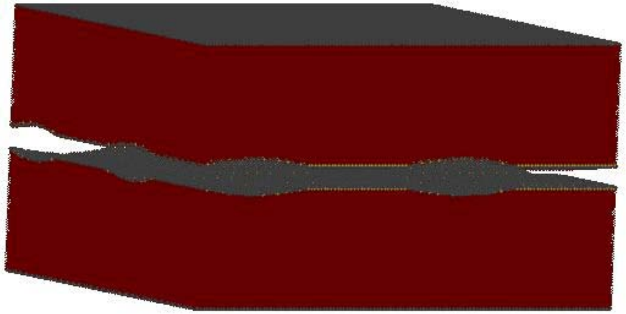
$$\langle K \rangle = \frac{3N}{2} kT$$

Stress tensor from the virial theorem

$$\sigma_{\alpha\beta} = \frac{1}{V} \sum_{i<j} F_{ij}^{\alpha} R_{ij}^{\beta} + \frac{1}{3V} \sum_{i} M_i \langle V_i^{\alpha} V_i^{\beta} \rangle$$

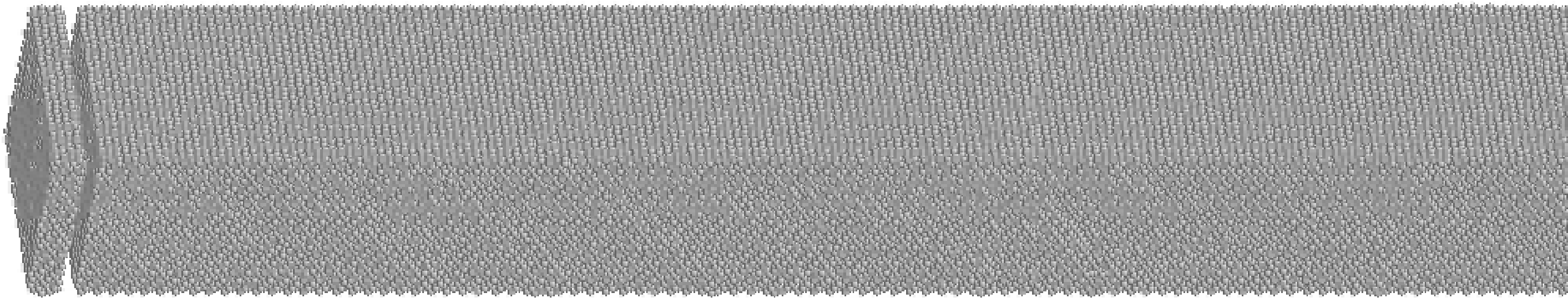


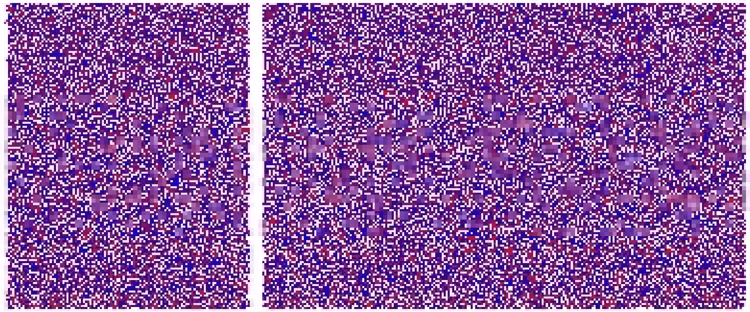
Contacts



Friction in CNTs

Shocks in molecular crystals





Spall failure

Interatomic potentials and uncertainties in MD

Newton's equations of motion

$$\dot{r}_i = \frac{p_i}{m_i}$$

$$\dot{p}_i = F_i$$

Force field form & parameters:
Largest source of epistemic uncertainties in MD simulations

$$F_i = -\nabla_{r_i} U\left(\{r_i\};\{C_i\}\right)$$

Interatomic potential for fcc metals

Lennard Jones:

$$U\left(\{R_i\};\varepsilon,\sigma\right) = \sum_{i<j} 4\varepsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right]$$

Very simple, not appropriate for metals

EAM (Sutton Chen)

$$U\left(\{R_i\};\varepsilon,\sigma,c\right) = \sum_{i<j}\phi\left(R_{ij}\right) + \sum_i F\left(\rho_i\right)$$

$$\varepsilon\sum_i\left[\sum_{j<i}\left(\frac{\sigma}{R_{ij}}\right)^{10} - c\sqrt{\sum_{j\neq i}\left(\frac{\sigma}{R_{ij}}\right)^{5}}\right]$$

How can we quantify this uncertainties?

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Force field parameterization and predictions

Find "optimal" FF parameter set in terms of a set of materials properties:

M_i^{tar} Training set of experimental or *ab initio* properties

Error function to be minimized:

Target value

FF prediction

$$X(\{C_i\}) = \sum_i \frac{\left(M_i^{tar} - M_i^{FF}(\{C_i\})\right)^2}{2\sigma_i^2} w_i$$

Weight of property *i*

Standard deviation

with respect to $\{C_i\}$

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Example: force field for Nickel

Target properties from experiments (training set)

Lattice parameter: $0.352 \pm 0.001 nm$

Cohesive energy: $-4.45 \pm 0.05 eV$

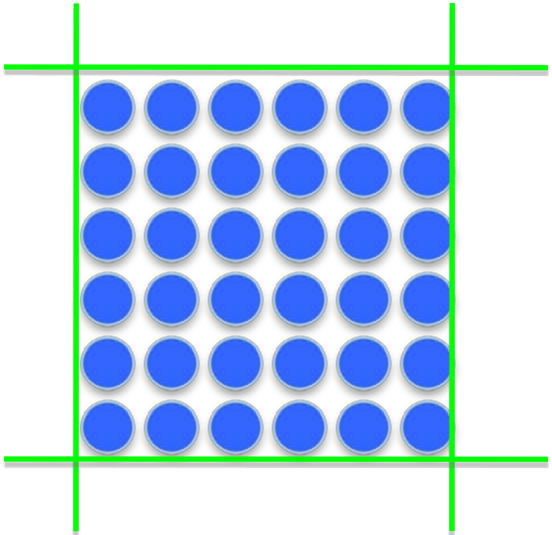
Bulk modulus: $187 \pm 5 GPa$

Surface energy: $2.3 \pm 0.05 J/m^2$

Prediction:

Vacancy formation energy:

Exp: $1.55 - 1.85 eV$



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Force field optimization via simulated annealing

Explore FF parameter space & optimize them via Monte Carlo

1. Start with initial guess for parameters

2. Generate new parameter set randomly from current set

$$\{C_i^{new}\} \leftarrow \{C_i + \delta_i\}$$

3. Compute change in cost function:

$$\Delta X = X(\{C_i\}) - X(\{C_i^{new}\})$$

4. If ΔX
   - ≤0: accept new FF parameter set $\{C_i\} \leftarrow \{C_i^{new}\}$
   - >0: accept new FF parameter set with probability: $p = \exp(-\Delta X / T)$

Metropolis Monte Carlo

- Start at a high “temperature” (T) to explore parameter space
  - Generates the canonical distribution: $p(\{C_i\}) \propto \exp\left[-X(\{C_i\})/T\right]$
  - Minimizes free energy: $F = \langle X(\{C_i\})\rangle - T S$
- “Cool down” system to obtain minimum E parameter set: $\{C_i\}_{opt}$

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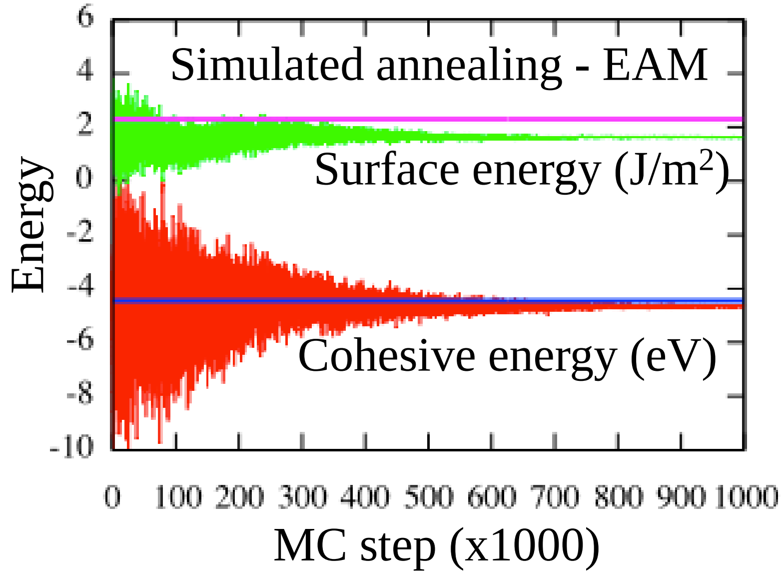
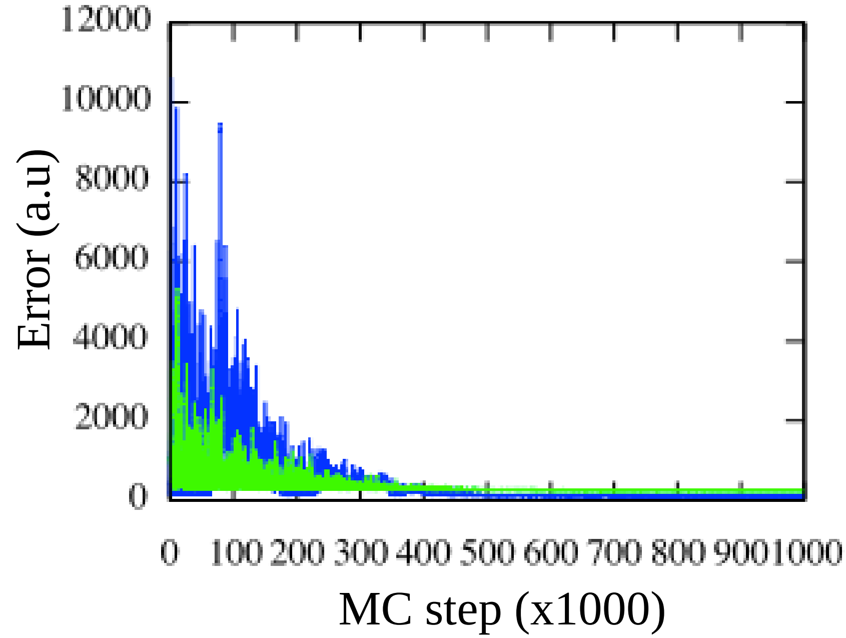
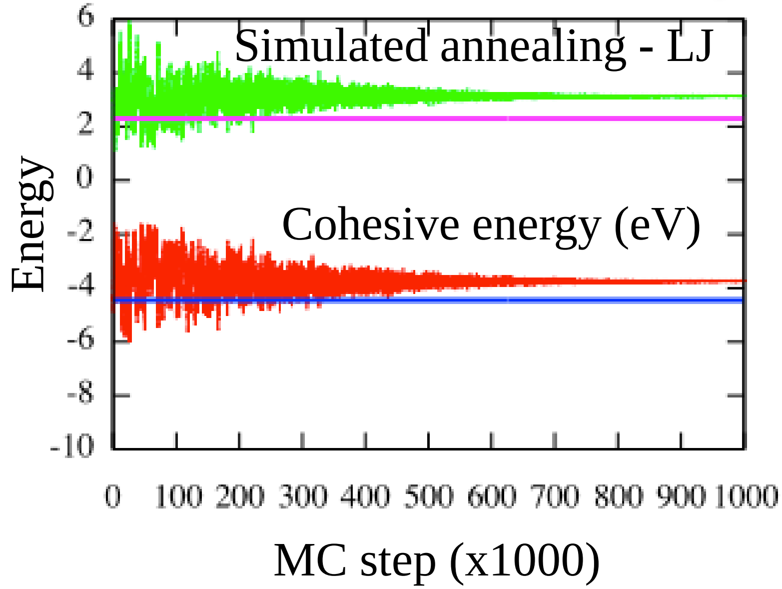
Strachan et al. PRB (1999); Strachan et al. MSMSE (2004)

Simulated annealing: optimal FF





Decreasing temperature



Predictions by optimized potential:

- EAM: 1.35 eV
- LJ: 3.76 eV
- Experiments: 1.55-1.85 eV

How to quantify uncertainties in $\{C_i\}$?

Assume training data in uncorrelated and each follow a normal distribution:

Probability of materials properties taking values $\{M_i\}$ is:

$$\prod_i \exp\left[-\left[\frac{\left(M_i^{tar}-M_i\right)^2}{2\sigma_i^2}\right]\right]$$

Probability of FF parameter set $\{C_i\}$ reproducing the training data:

$$P\left(\{C_i\}\right) \propto \prod_i \exp\left[-\left[\frac{\left(M_i^{tar}-M_i^{FF}\left(\{C_i\}\right)\right)^2}{2\sigma_i^2}\right]\right]$$

Generate an ensemble of potentials with this distribution

How to quantify uncertainties in $\{C_i\}$?

2. Generate new parameter set randomly from current set

$$\{C_i^{new}\} \leftarrow \{C_i + \delta_i\}$$

3. Compute change in cost function:

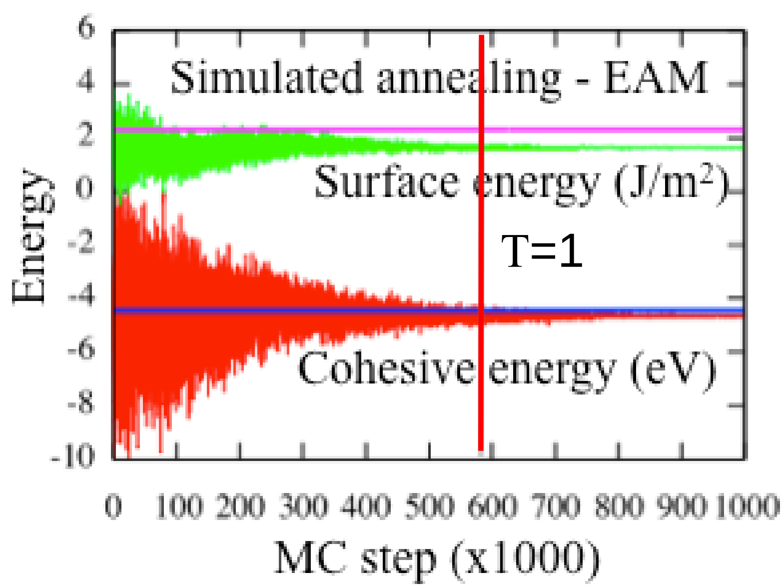
$$\Delta X = X(\{C_i\}) - X(\{C_i^{new}\})$$

4. If ΔX
- ≤ 0: accept new FF parameter set $\{C_i\} \leftarrow \{C_i^{new}\}$
- >0: accept new FF parameter set with probability: $p = \exp(-\Delta X / T)$

Metropolis Monte Carlo

Produces parameters with probability

$$p(\{C_i\}) \propto \exp\left[-X(\{C_i\})/T\right]$$



Frederiksen et al. PRL, 93, 165501 (2004)

Ensemble of FFs at T=1 does not reproduce all the target properties

Increase T until all properties are described by the ensemble

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Quantifying uncertainties in $\{C_i\}$

$$P(\{C_i\}) \propto \prod_i \exp\left[-\left[\frac{\left(M_i^{tar} - M_i^{FF}(\{C_i\})\right)^2}{2\sigma_i^2}\right]/T\right]$$

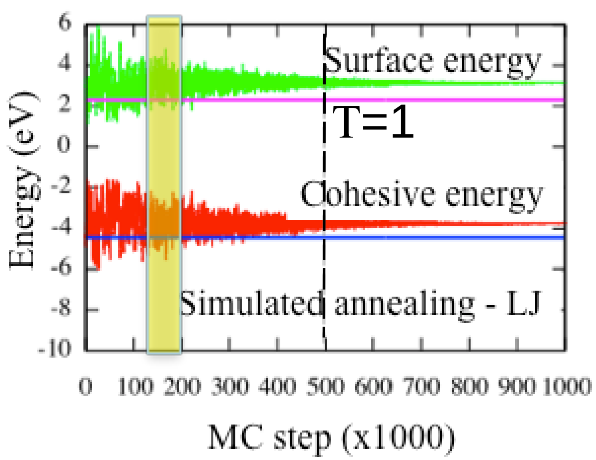
Ensemble of FF parameters for T=1: product of PDFs of target properties having values given by FF

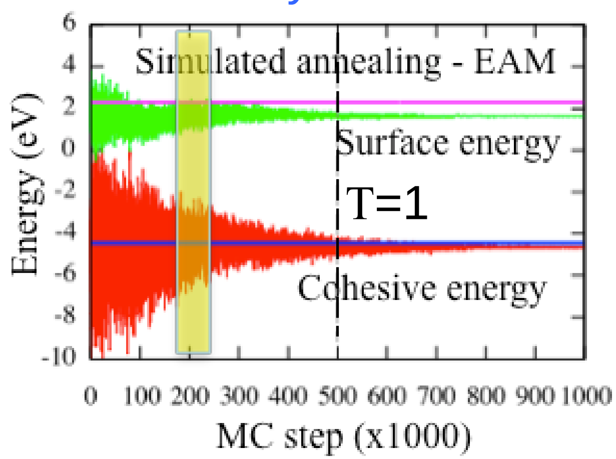
Ensemble of FF parameters spans all properties including standard deviation

Done: T=1

Ensemble of parameters does not describe all properties

Increase temperature until all properties are described by ensemble





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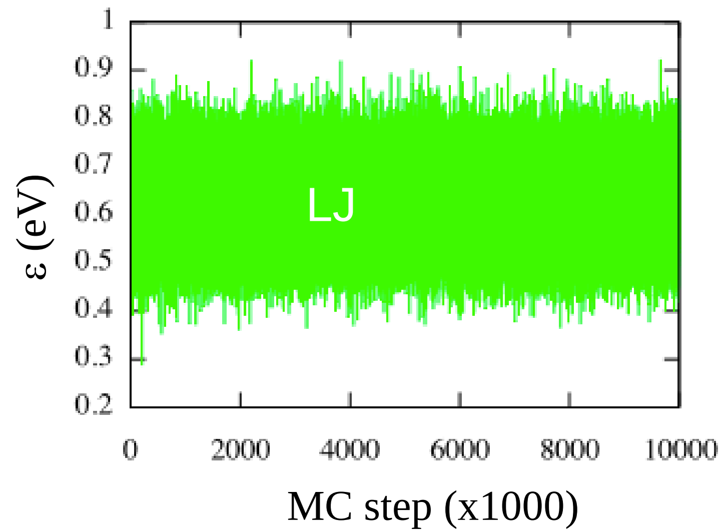
Ensemble of FFs predictions

EAM

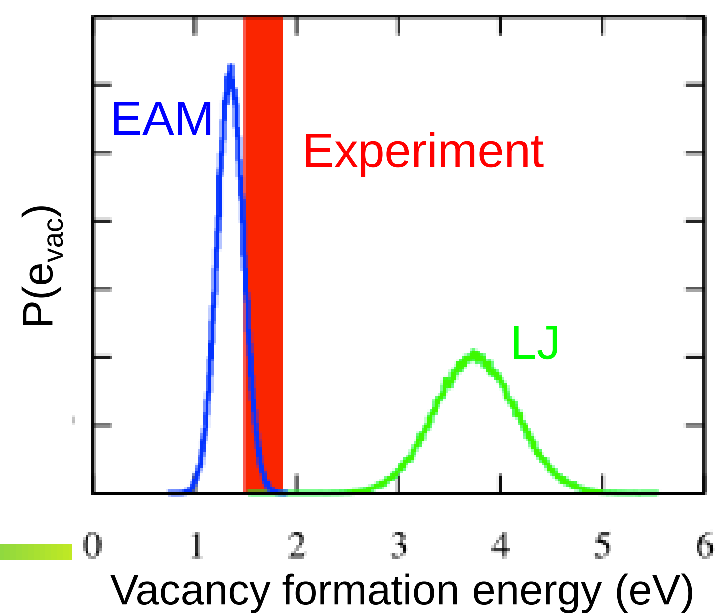
ε (eV)

MC step (x1000)



$$p(\{C_i\}) \propto \exp\left[-X(\{C_i\})/T\right] =$$

$$\prod_i \exp\left[-\left[\frac{\left(M_i^{FF}(\{C_i\}) - M_i^{tar}\right)^2}{2\sigma_i^2}\right]/T\right]$$



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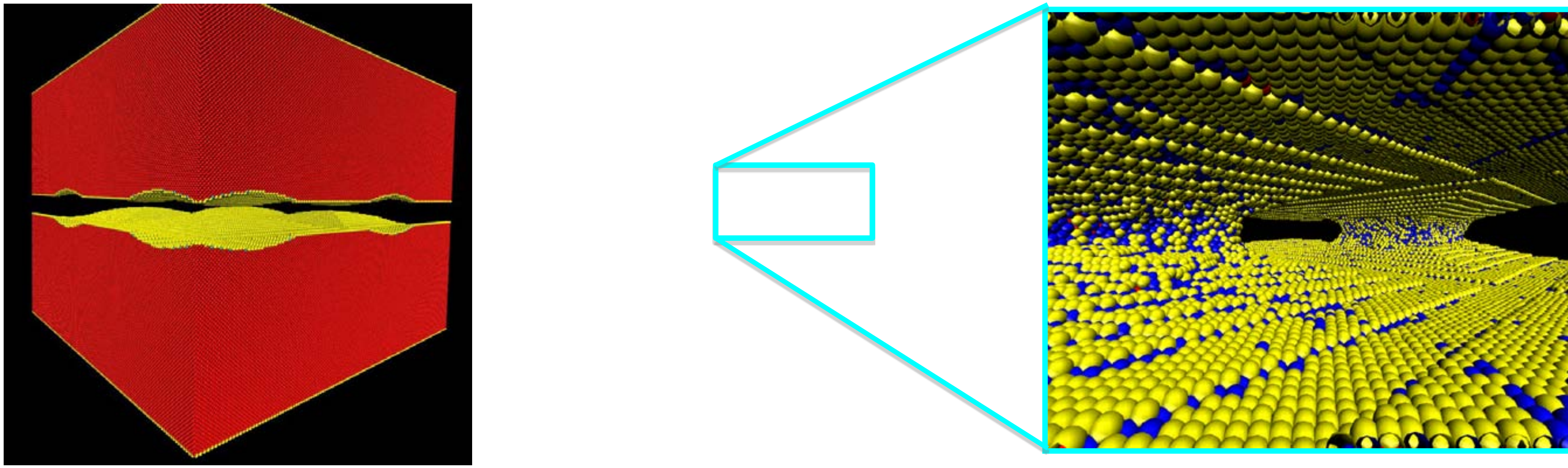
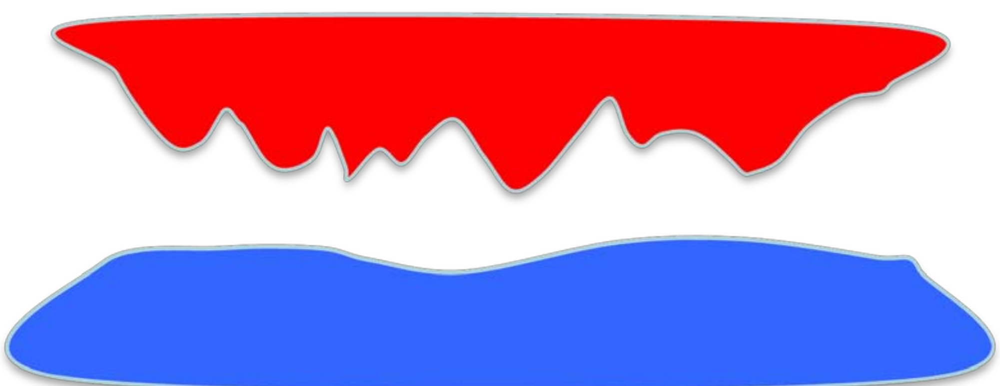
Atomistic contact simulations



For a given surface asperity (distribution of heights and peak to peak separations)

- What is the minimum pull-out force as a function of applied closing force?
- What sub-surface defects are generated in metal and dielectric?



Closing

Small contact area and large stress: Plastic deformation -> increase in contact area until stress equals the materials hardness

Opening

Size dependent strength of bridges

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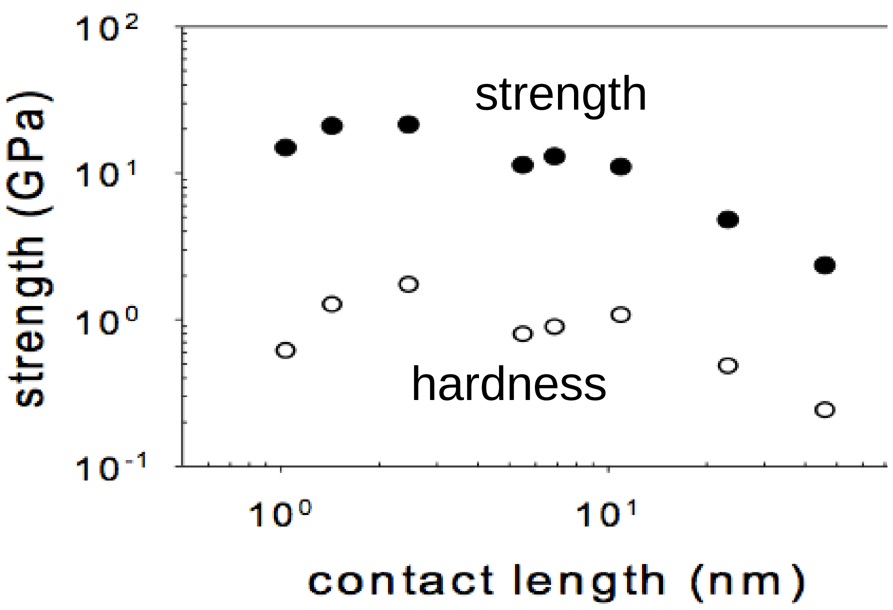
Kim and A. Strachan, Physical Review Letters 104, 215504 (2010).

Initial UQ in large-scale MD

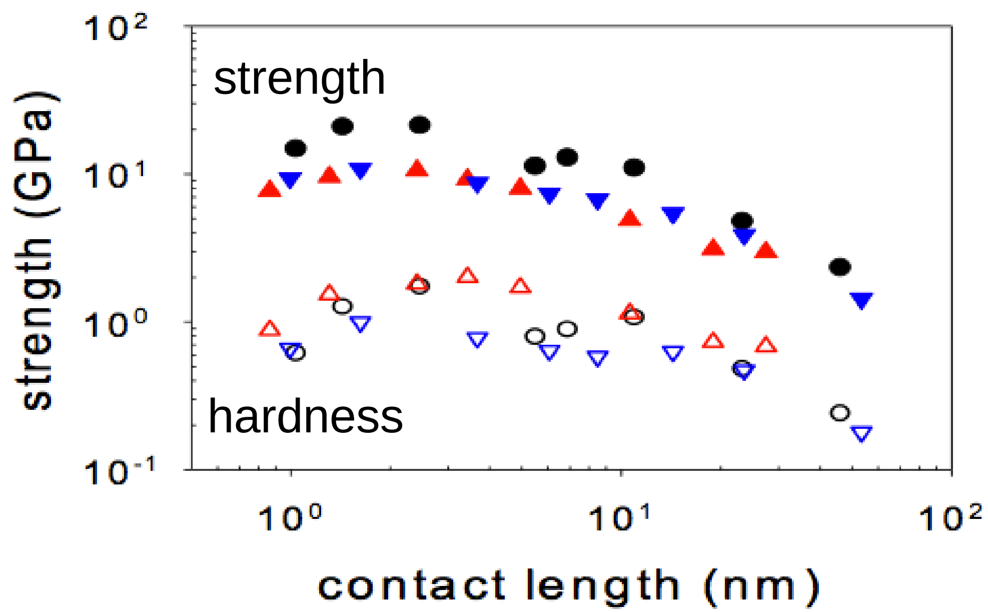
PDF of FF parameters is difficult to utilize for large-scale simulations

Current state-of-the-art FF results



•Force field overestimates binding

Decrease energy scales by 50%



•Initial UQ calculation leads to range properties

Summary

Uncertainties in force field parameters

- Monte Carlo procedure to generate an ensemble of FF parameters
  - PDF of parameter set associated with PDF of resulting properties
  - Ensemble describes all training set properties
- Overly simple FF ➔ large variation in properties ➔ uncertain prediction
- Future work
  - Correlated uncertainties in input data
  - Apply approach to PRISM device materials
  - Automatic model form uncertainty? Aidan Thompson @ SNL
  - *Ab initio* calculations – Peter Schultz @ SNL

- For large-scale MD simulations
  - Minimum-maximum approach as initial step
- Couple FF parameter uncertainty to other aleatoric uncertainties
- Propagate across scales