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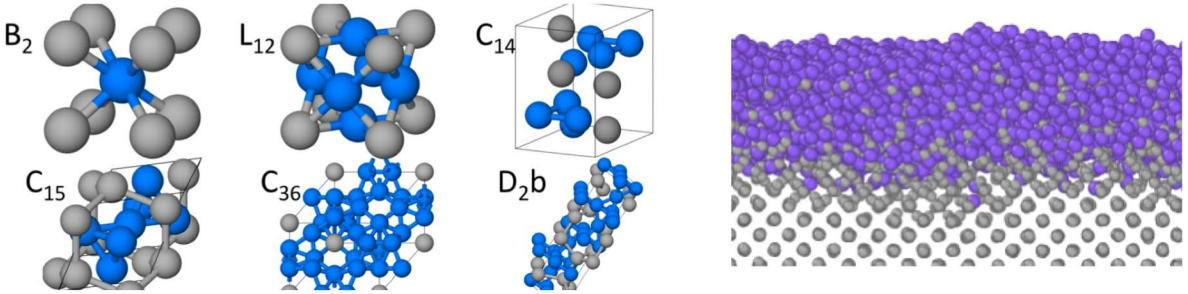

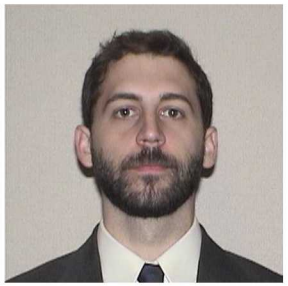

Predictive Atomistic Simulations of Materials using SNAP Machine-Learning Interatomic Potentials

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U.S. DEPARTMENT OF ENERGY

National Nuclear Security Administration

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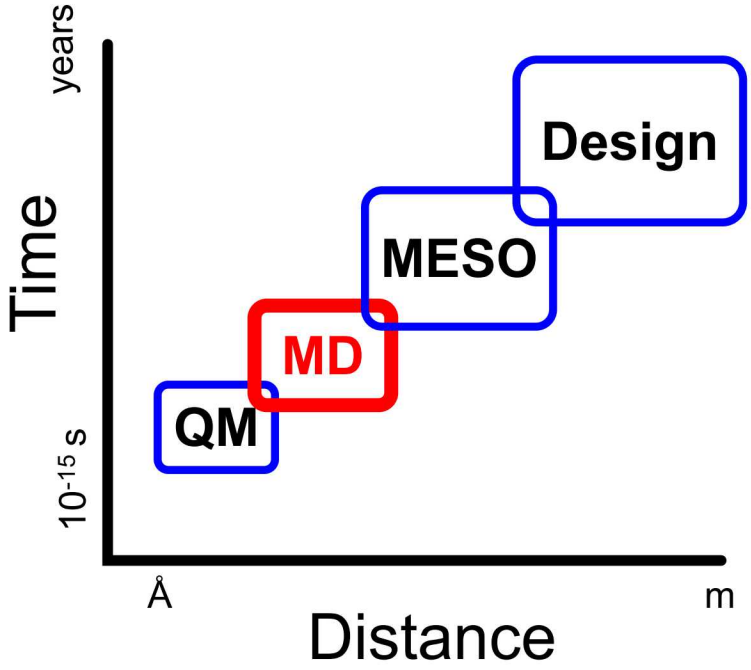
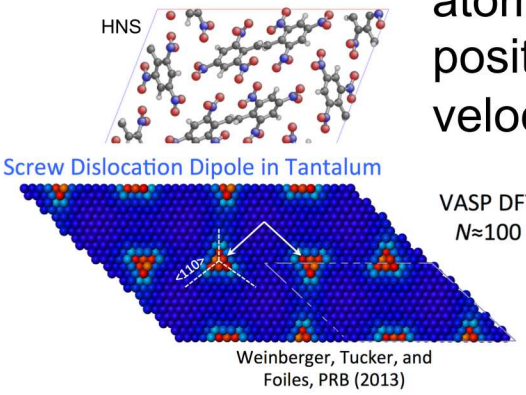
What is Molecular Dynamics Simulation?

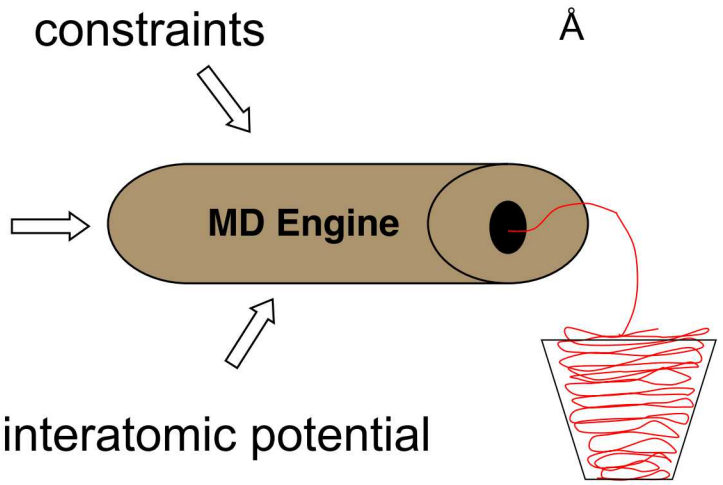
- Continuum models require underlying models of the materials behavior
- Quantum methods can provide very complete description for 100s of atoms
- Molecular Dynamics acts as the “missing link”
  - Bridges between quantum and continuum models
  - Moreover, extends quantum accuracy to continuum length scales; retaining atomistic information

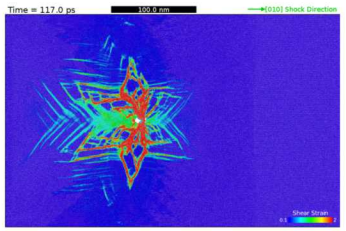




atoms,
positions,
velocities

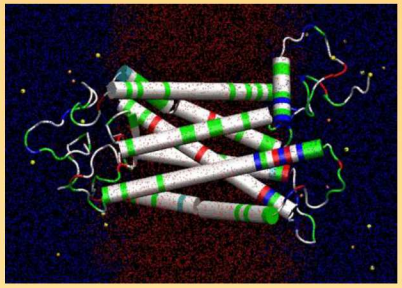
VASP DFT
$N \approx 100$

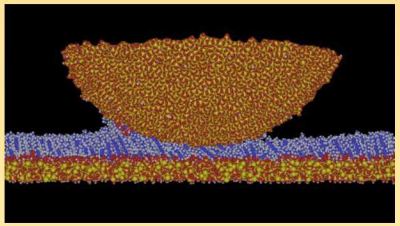




Positions, velocities and forces at many later times

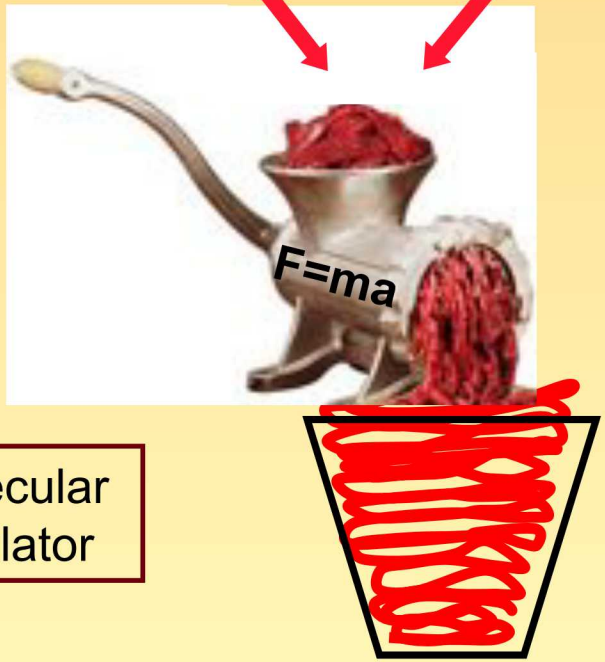
What is LAMMPS?





Initial positions and velocities

Interatomic potential



- Biomolecules
- Polymers (soft materials)
- Materials science (hard materials)
- Mesoscale to continuum

Large-scale Atomic/Molecular Massively Parallel Simulator

⇨

Positions and velocities at later times

Thanks to ~~Aidan Thompson~~ Mike Chandross

MD Approximations Change Over Time

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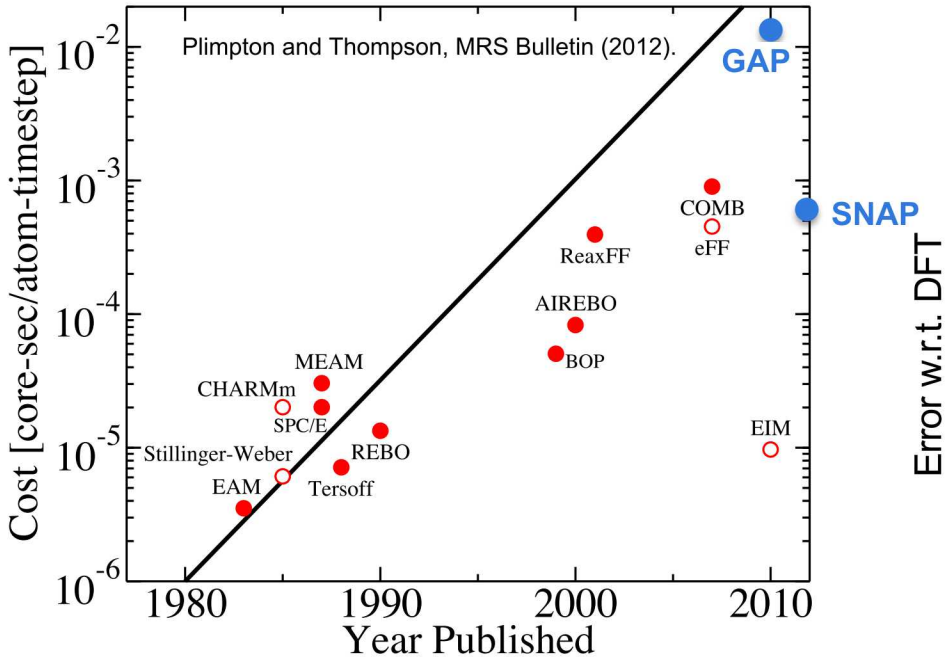
Twobody (B.C.)
Lennard-Jones, Hard Sphere, Coulomb, Bonded

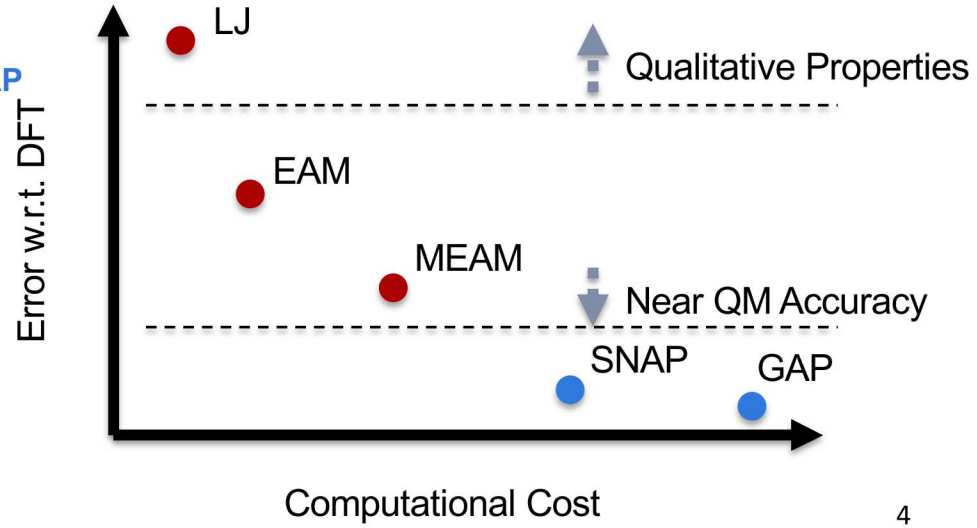
Manybody (1980s)
Stillinger-Weber, Tersoff, Embedded Atom Method

Advanced (90s-2000s)
REBO, BOP, COMB, ReaxFF

Big Data / Deep / Machine Learning (2010s)
GAP, SNAP, NN, …



- **Resources are limited, which is your best choice?**

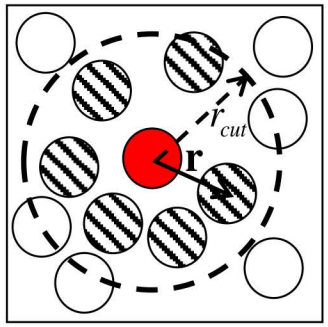


Bispectrum Components as Descriptor

- Neighbors of each atom are mapped onto unit sphere in 4D

$$(\theta_0, \theta, \phi) = \left(\theta_0^{max}\, r/r_{cut}\,, \cos^{-1}(z/r), \tan^{-1}(y/x)\right)$$

- Expand density around each atom in a basis of ***4D hyperspherical harmonics***,
- Bispectrum components of the 4D hyperspherical harmonic expansion are used as the geometric descriptors of the local environment
  - Preserves universal physical symmetries
  - Rotation, translation, permutation
  - Size-consistent



$$u^j_{m,m'} = U^j_{m,m'}(0,0,0) + \sum_{r_{ii'}<R_{cut}} f_c(r_{ii'}) w_i U^j_{m,m'}(\theta_0, \theta, \phi)$$

$$B_{j_1,j_2,j} = \sum_{m_1,m_1'=-j_1}^{j_1} \sum_{m_2,m_2'=-j_2}^{j_2} \sum_{m,m'=-j}^{j} (u^j_{m,m'})^* H^{jmm'}_{j_1 m_1 m_1' \; j_2 m_2 m_2'} \; u^{j_1}_{m_1,m_1'} u^{j_2}_{m_2,m_2'}$$

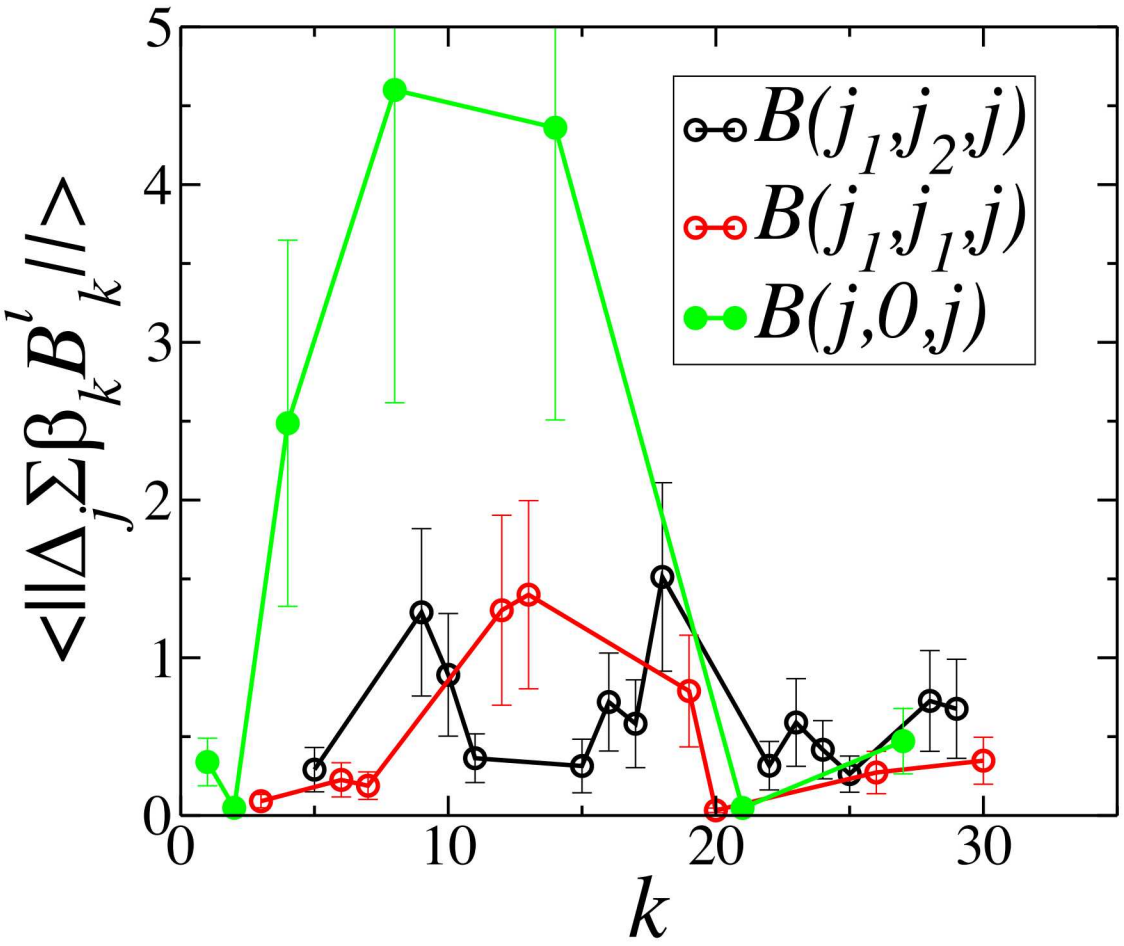
Symmetry relation:

$$\frac{B_{j_1,j_2,j}}{2j+1} = \frac{B_{j,j_2,j_1}}{2j_1+1} = \frac{B_{j_1,j,j_2}}{2j_2+1}$$

Drautz showed that Steinhardt, SOAP, SNAP and many other descriptors are all special forms of the atomic cluster expansion (*Phys.Rev.B* 2019)

Effect of High-Order Bispectrum Components

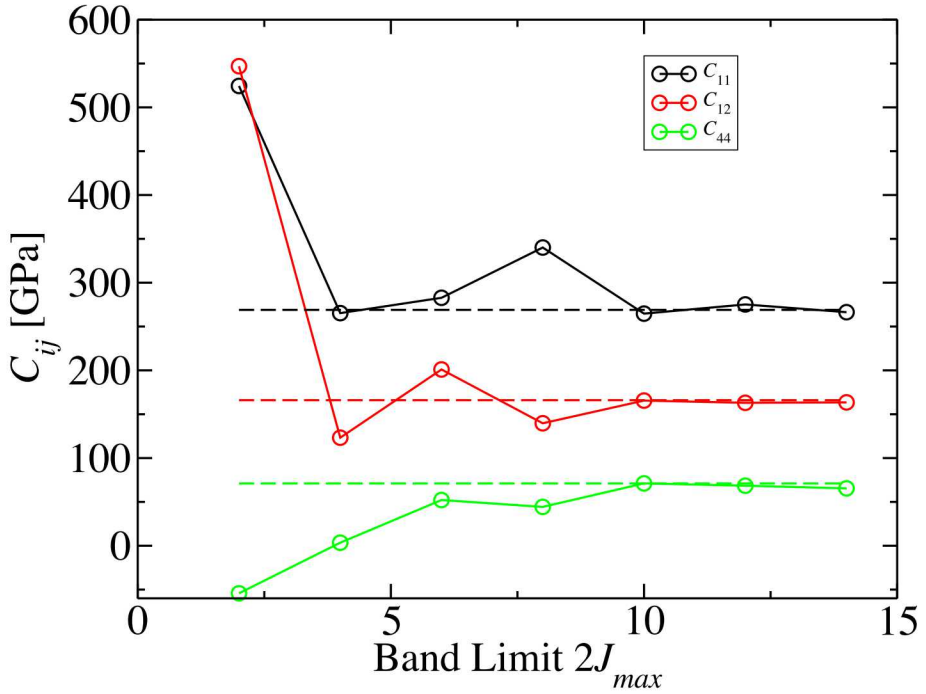
- MD simulation of molten tantalum using SNAP Ta06A potential
- Magnitude of average force contributed by each bispectrum component



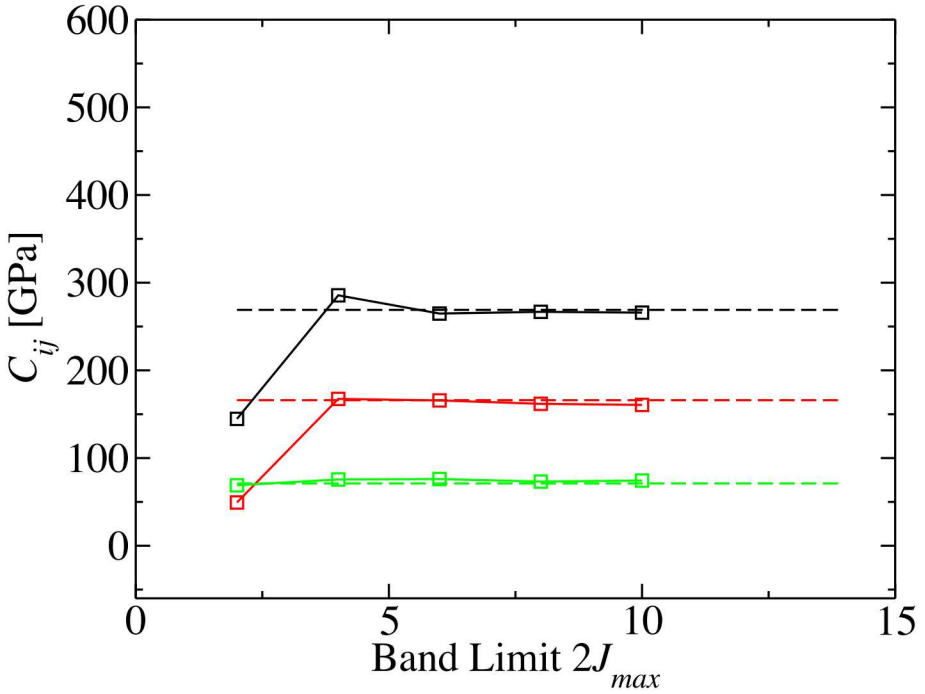
Effect of High-Order Bispectrum Components

- Elastic constants for tantalum versus band limit

Linear SNAP



Quadratic SNAP



ML-IAP Definition

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Model Form

- Energy of atom i expressed as a basis expansion over K components of the bispectrum (B_k^i)

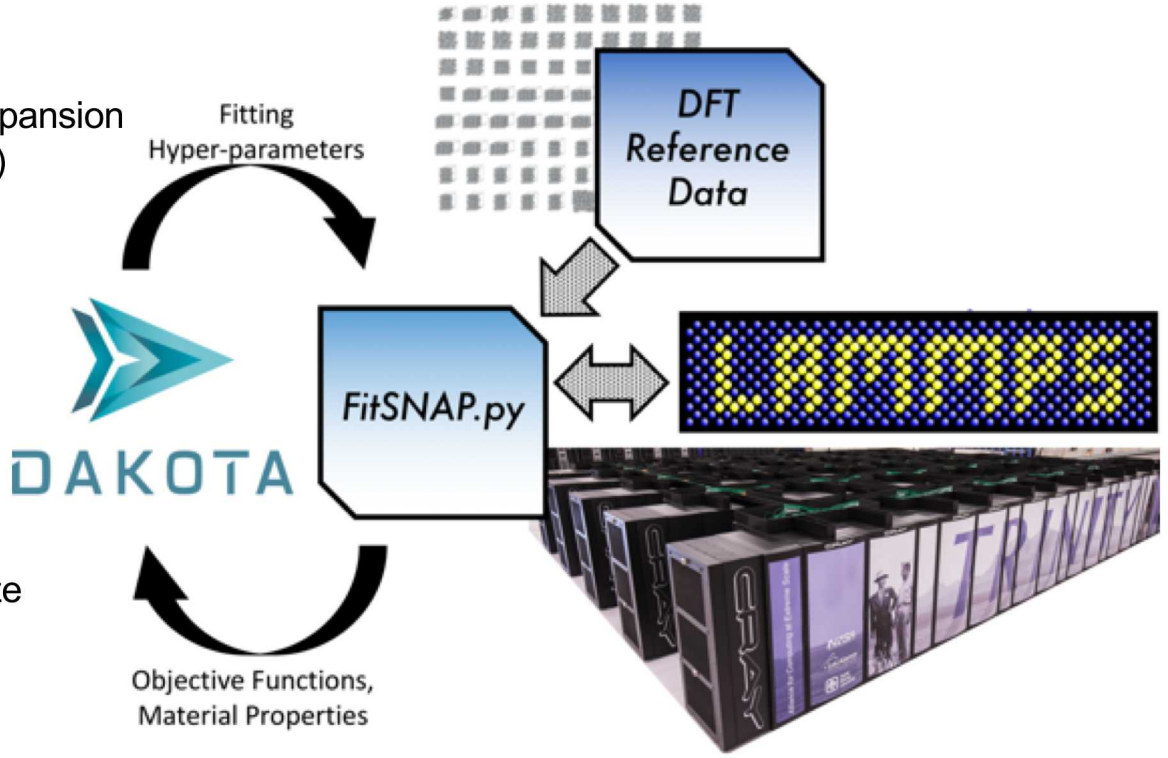
$$E_{SNAP}^i = \beta_0 + \sum_{k=1}^{K} \beta_k (B_k^i - B_{k0}^i)$$

Regression Method

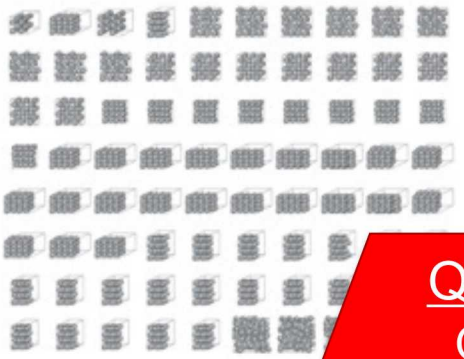
- β vector fully describes a SNAP potential
- Decouples MD speed from training set size

$$\min(||\mathbf{w} \cdot D\boldsymbol{\beta} - T||^2 - \gamma_n \, ||\boldsymbol{\beta}||^n)$$

Weights Set of Descriptors DFT Training



SNAP Fitting Process
FitSnap.py



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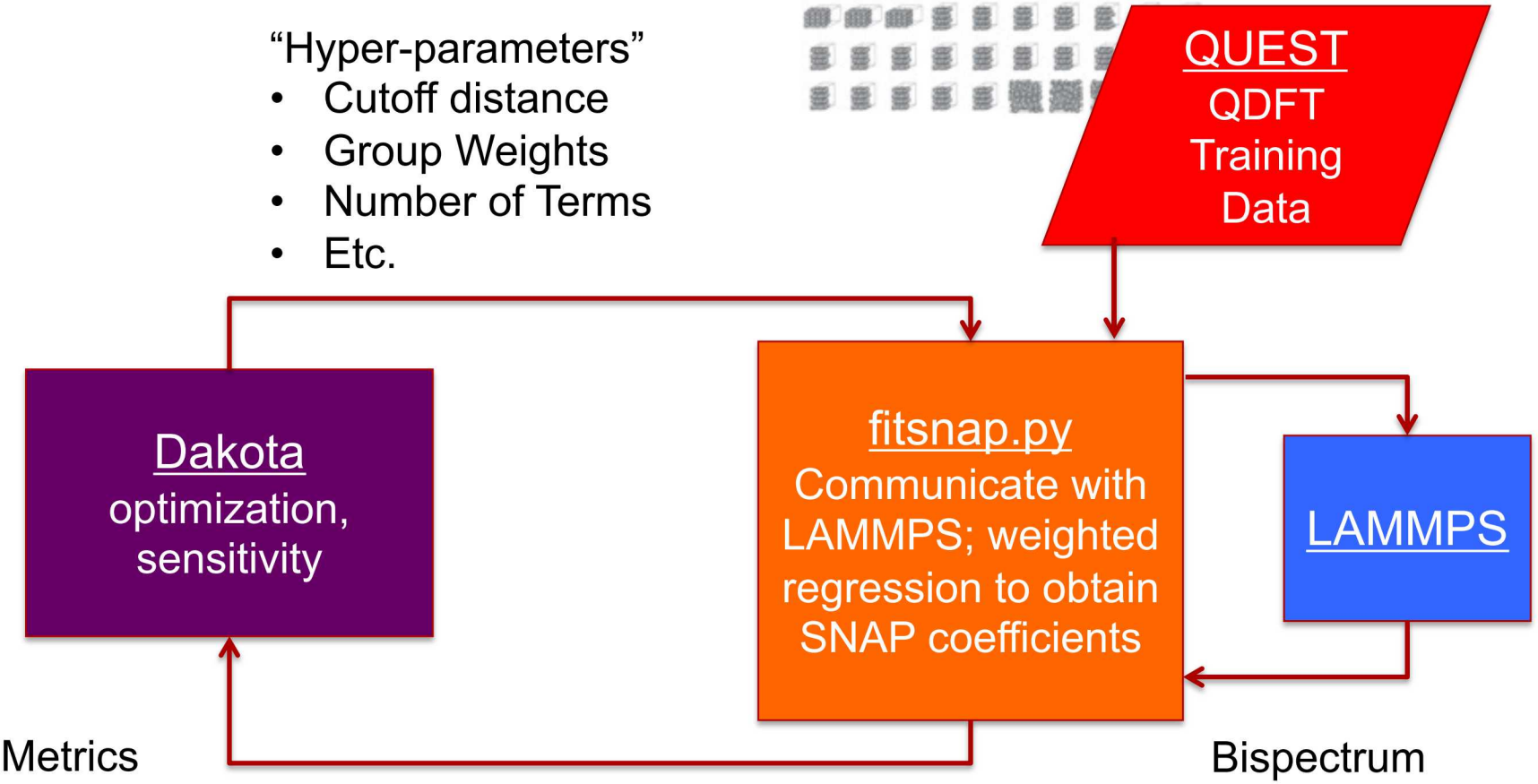
"Hyper-parameters"
- Cutoff distance
- Group Weights
- Number of Terms
- Etc.



Metrics
- Force residuals
- Energy residuals
- Elastic constants
- Etc.

Bispectrum components & derivatives, reference potential

Outline of Current Research Areas

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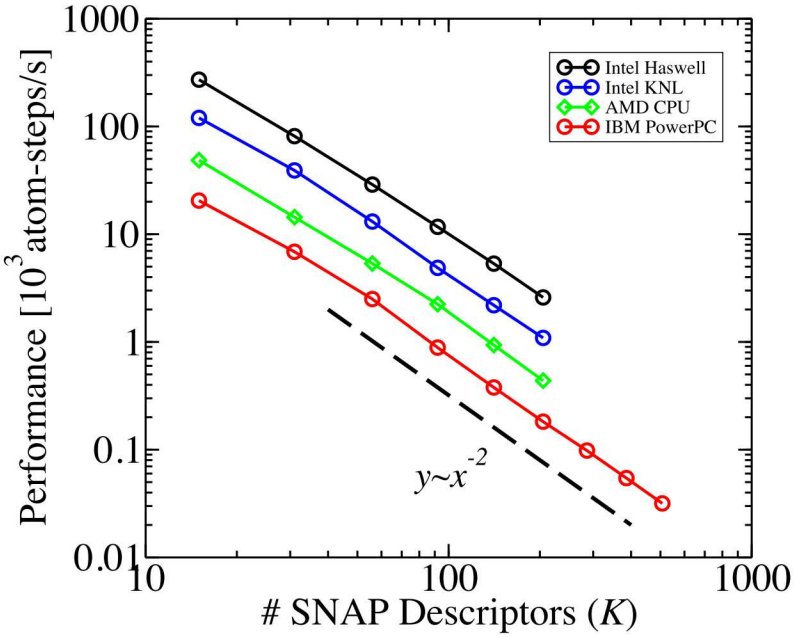
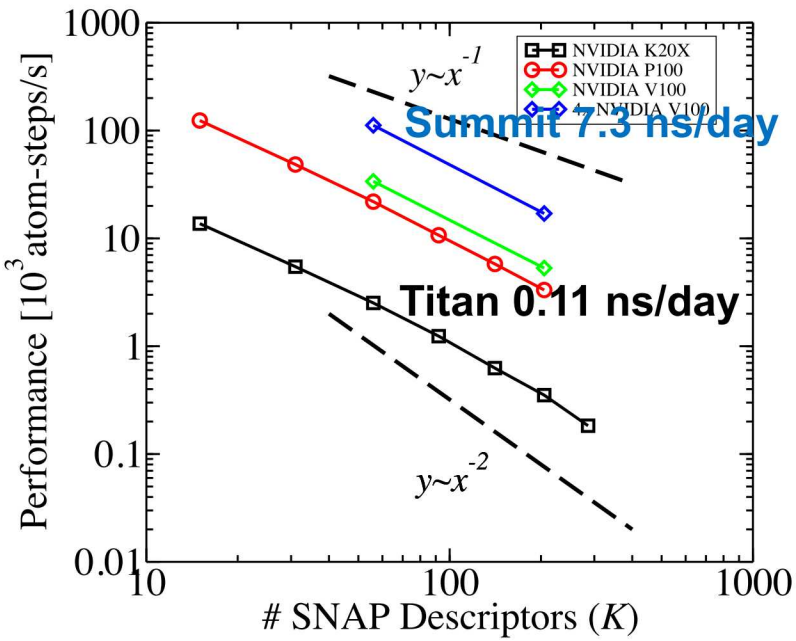
- **SNAP Computational Speed in LAMMPS**
  - SNAP with OpenMP (Good for CPUs)
  - **SNAP with KOKKOS (CPU, GPU,...) [ECP CoPA project]**
  - Exploring new GPU algorithms [NERSC/NESAP]
- **Plasma-Facing Materials (SciDAC-4)**
  - **Tungsten/Beryllium (complete)**
  - W/Be/H (in progress)
  - Nitrogen, Neon (future work)
- **Phase Transitions in Extreme Environments**
- **Radiation Damage in III-V Semiconductors**
  - **New Multi-element SNAP formulation**
- **SNAP Accuracy**
  - **Quadratic SNAP**
  - SNAP + Neural Networks
  - Better descriptors

Adding Descriptors Increases Cost A Lot

CPU



GPU



- Benchmarks for Exascale Computing Project
- Short MD simulation of BCC tungsten @ 300K
- GPU and KNL use the LAMMPS Kokkos package
- 2000 atoms, 1 node

Tungsten and Beryllium as Plasma-Facing Material

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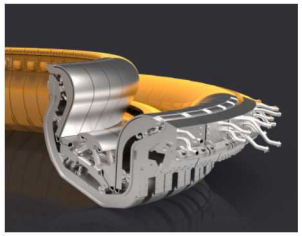
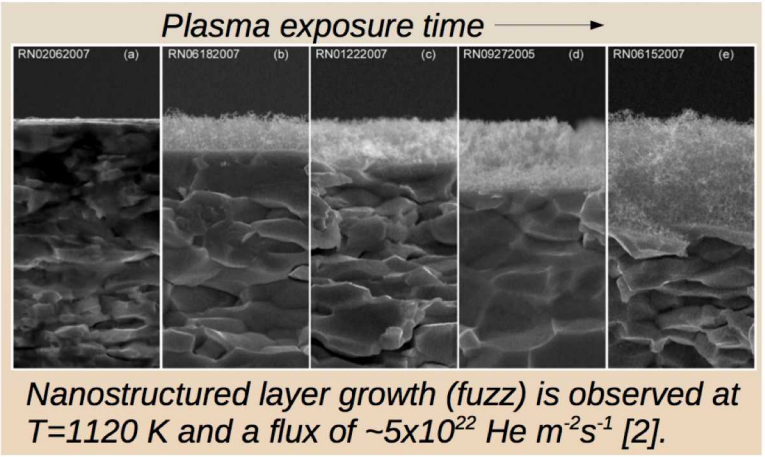
First Wall
PFM = Beryllium

ITER fusion reactor:

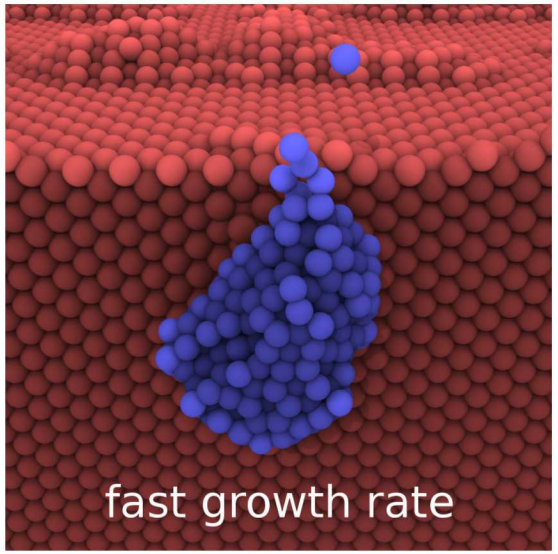
- Putting the sun in a box
- Plasma-facing material is tungsten
- Exposed to He and H at elevated temperature
- Fuzz buildup limits power output and useful life of divertor elements

Divertor
PFM = Tungsten
10-20 MW/m²





Nanostructured layer growth (fuzz) is observed at T=1120 K and a flux of ~5x10^{22} He m^{-2}s^{-1} [2].



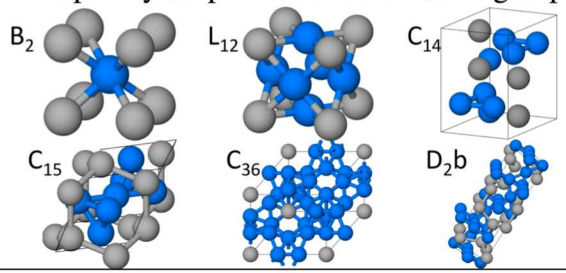
Luis Sandoval, Blas Uberuaga, Danny Perez, Art Voter, Phys. Rev. Lett. (2015)

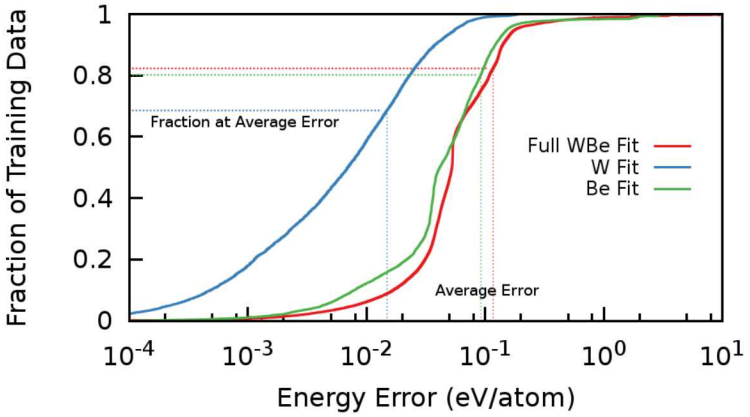
Training SNAP for Transferability – Tungsten+Beryllium

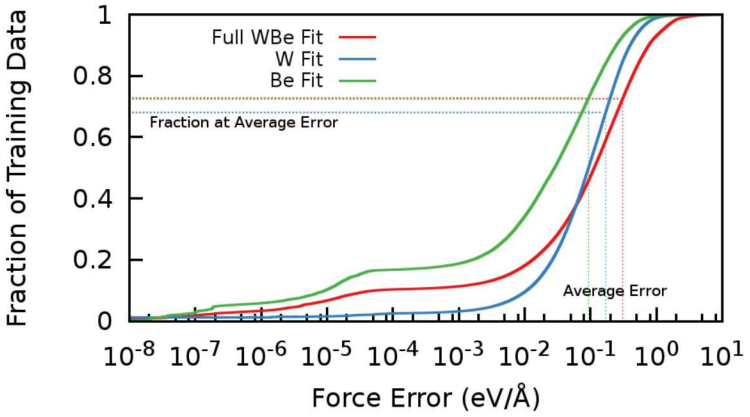
- Making a multi-element SNAP potential does sacrifice some accuracy from either pure component fit.
- Looking at which training data was weighted heavily

Description	N_E	N_F	σ_E	σ_F
W-Be:				
Elastic Deform†	3946	68040	$\mathbf{3 \cdot 10^5}$	$2 \cdot 10^3$
Equation of State†	1113	39627	$2 \cdot 10^5$	$4 \cdot 10^4$
DFT-MD†	3360	497124	$7 \cdot 10^4$	$6 \cdot 10^2$
Surface Adhesion	381	112527	$2 \cdot 10^4$	$\mathbf{9 \cdot 10^4}$

† Multiple crystal phases included in this group:

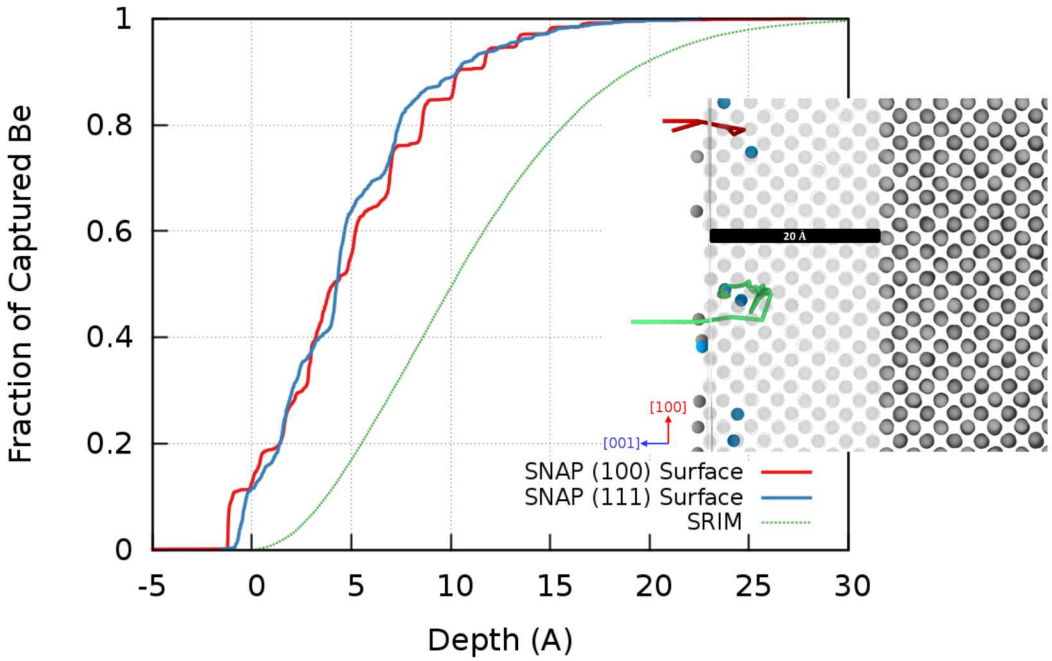






Single Implantation Simulations

- MD depth profile is more shallow than binary collision models predict
- Capture rate is lower than BC model at 40% (versus 60%)



Defect Type	Percent of Implanted Be (100) Surface	(111) Surface
[111] Dumbbell	41.2	23.9
Substitution	22.2	34.6
[100] Surf. Hollow Site	12.3	8.3
Tetrahedral Interstitial	10.4	12.4
[110] Dumbbell	8.4	11.3
Octahedral Interstitial	5.3	4.1
Other	0.4	2.8
Surf. Bridge Site	0.03	2.6

Defect Type	Formation Energy (eV) DFT	SNAP	BOP
[111] Dumbbell	4.30	3.66	0.67
Substitution	3.11	3.29	-2.00
[100] Surf. Hollow Site	-1.05	-1.39	-3.52
Tetrahedral Interstitial	4.13	4.20	-0.28
[110] Dumbbell	4.86	4.29	-0.03
Octahedral Interstitial	3.00	5.11	0.34
[100] Surf. Bridge Site	1.01	0.44	-1.30

Cumulative Energetic Implantation

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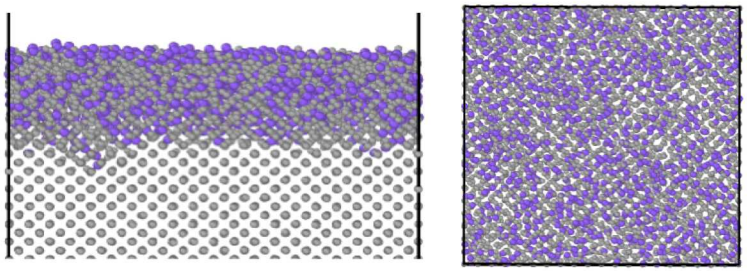
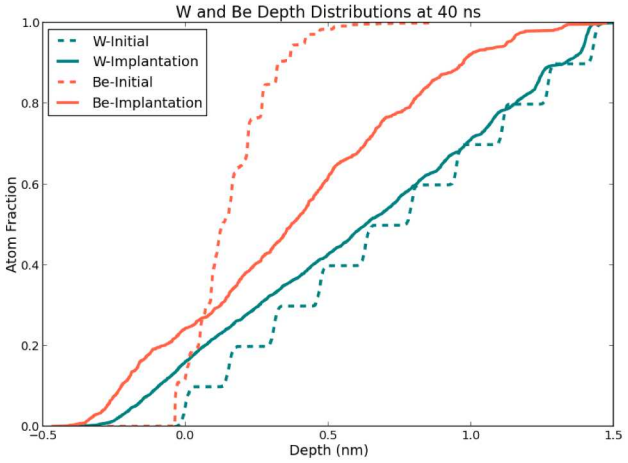
4000 atoms, 1.1 x 10^{20} m^{-2}
35% Retention

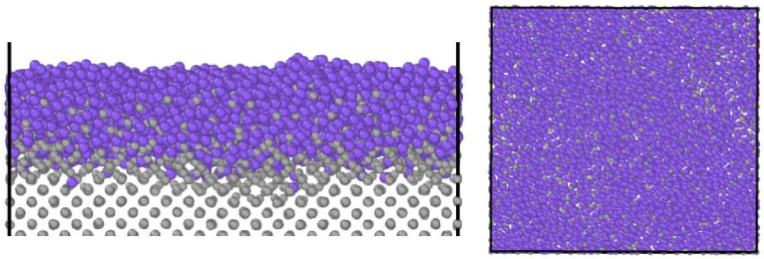


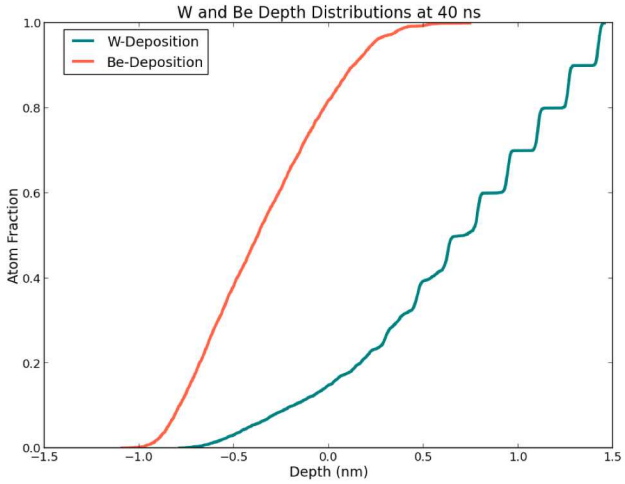


- 75 eV cumulative Be implantation in W
- Implanted every 10 ps
- 1000 K, (100) surface, 6 nm x 6 nm x 12 nm box
- Initially Be implants into material and resides at defects like <111> dumbbell or substitutional sites
- At higher fluences, layer becomes amorphous
  - Exchange of W and Be with no obvious crystal structure
- Amorphous layer extends from about 1.5 nm in the surface to 0.2-0.3 nm above the original surface
- Tungsten loses crystal structure
- Be penetrates deeper (diffusion?)

Cumulative Thermal Deposition

4000 atoms, 1.1 x 10^{20} m^{-2}





- Be now randomly placed on surface with no energy
- Rest of simulation parameters are the same
- Initially Be resides at hollow sites but once those become occupied, Be starts to exchange with W
- Similar results of amorphous layer that forms at higher fluences
- Layer is thicker, extending from 0.5 nm below the surface to 1 nm above the surface
- Be mainly remains near surface
- Almost 20% of the W in the first 1.5 nm is located above the original surface

Can We Improve Multi-Element SNAP?

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Current Multi-element SNAP

Elemental Weight

$$u_{jmm'} = U_{jmm'}(0,0,0) + \sum_{\substack{r_{i'j} < R_{cut}}} f_c(r_{i'}) w_\delta U_{jmm'}(\theta_0, \theta, \phi)$$

$$B_{j_1,j_2,j} = \sum_{m_1,m_1'=-j_1}^{j_1} \sum_{m_2,m_2'=-j_2}^{j_2} \sum_{m,m'=-j}^{j} (u^j_{m,m'})^* H^{jmm'}_{\substack{j_1 m_1 m_1' \\ j_2 m_2 m_2'}} u^{j_1}_{m_1,m_1'} u^{j_2}_{m_2,m_2'}$$

Explicit Multi-Element SNAP

Elemental Basis Function

$$u^\delta_{jmm'} = U_{jmm'}(0,0,0) + \sum_{\substack{r_{ii'} < R_{cut} \\ i' \in \delta}} f_c(r_{ii'}) w_\delta U_{jmm'}(\theta_0, \theta, \phi)$$

Three-Element Bispectrum Component

$$B^{\beta\gamma\delta}_{j_1 j_2 j} = \sum_{m_1,m_1'=-j_1}^{j_1} \sum_{m_2,m_2'=-j_2}^{j_2} \sum_{m,m'=-j}^{j} (u^\beta_{jmm'})^* H^{jmm'}_{\substack{j_1 m_1 m_1' \\ j_2 m_2 m_2'}} u^\gamma_{j_1 m_1 m_1'} u^\delta_{j_2 m_2 m_2'}$$

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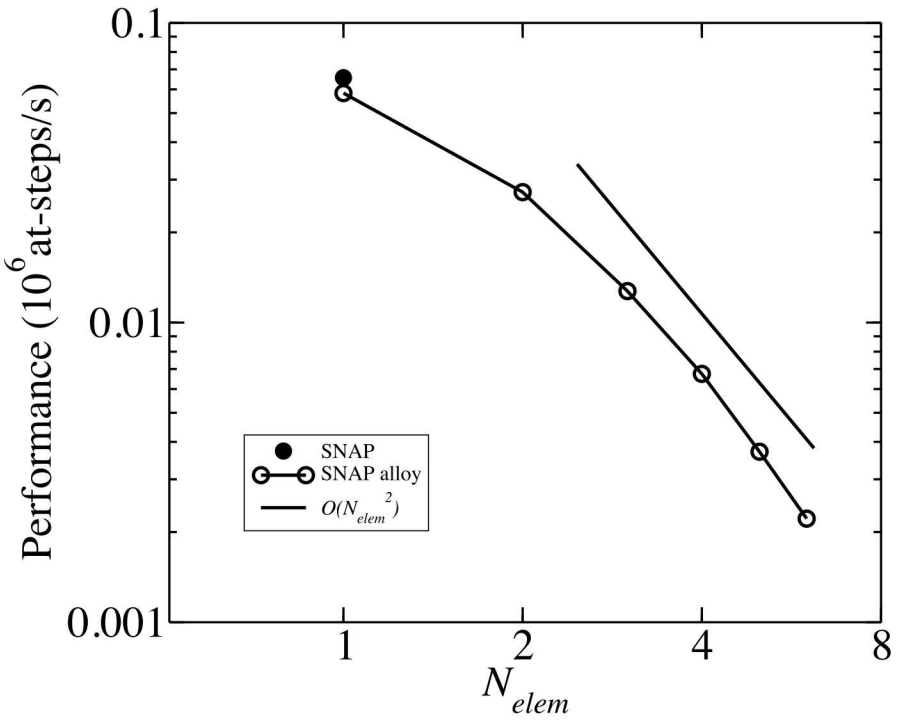
Computational Cost of Multi-Element

- Cost dominated by dBi/dRj
- For each bispectrum component there are Nelem^3 labellings
- But neighbor j only contributes to the ones that have at least one label matching element of j
- Nelem^2 labellings of remaining two labels



Indium Phosphide SNAP Potential

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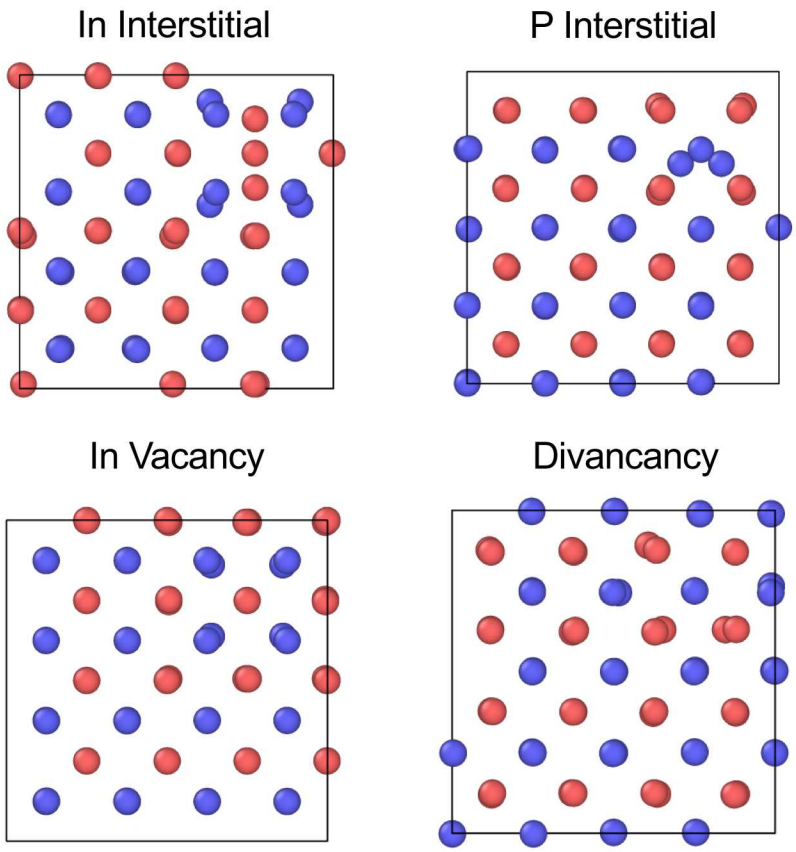
- Interest in studying radiation damage in semiconductors such as indium phosphide
  - Potential needs to be able to reproduce dynamics of a high energy collision cascade
- For radiation damage, accurately reproducing defect formation energies is crucial
- SNAP needs to not only reproduce these important parameters but also be able to distinguish between 2 element types
- Initial multi-element version of SNAP replicated InP properties fairly well but failed in reproducing the defect formation energies
  - Difference between SNAP and DFT was greater than 1 eV for some defects
- Preliminary results of new multi-element alloy formulism of SNAP is showing much improvement



InP SNAP Preliminary Results

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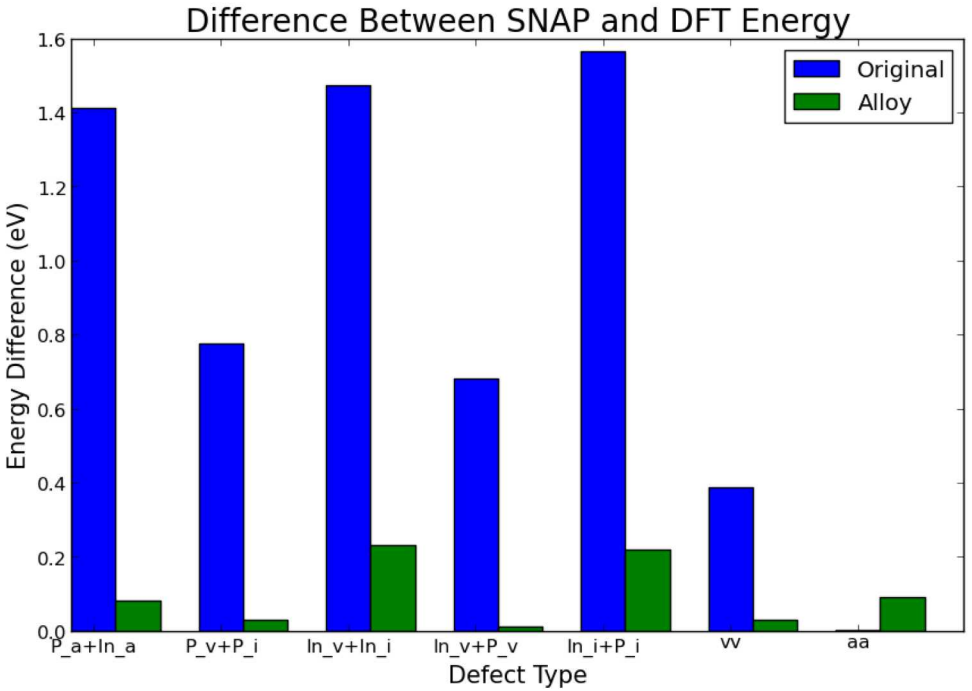
- Compare original and alloy version of SNAP for InP
- Compare defect formation energies between SNAP and DFT
  - Defect formation energies were chosen to conserve stoichiometry
- Previous multi-element version of SNAP failed to reproduce defect formation energies
  - Still over 1 eV difference from DFT for best potential
- New alloy multi-element version of SNAP performs much better
  - Defect formation energies show much reduced error from DFT
  - Largest difference id 0.23 eV



InP Elastic Constants

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	Exp. *	SNAP
C11 (Gpa)	101.1	99.5
C12 (Gpa)	56.1	53.6
C44 (Gpa)	45.6	16.7
Bulk Modulus (Gpa)	71.1	66.8
Shear Modulus (Gpa)	22.5	17.7, 26.1
Poisson Ratio	0.36	0.33

- Elastic constants for InP are fairly well reproduced
- More consistent with expected values compared to previous iterations of this potential

* D.N. Nichols, D.S. Rimai, and R.J. Sladek Solid State Commun. 667, 36 (1980)

What About Adding Quadratic Terms?

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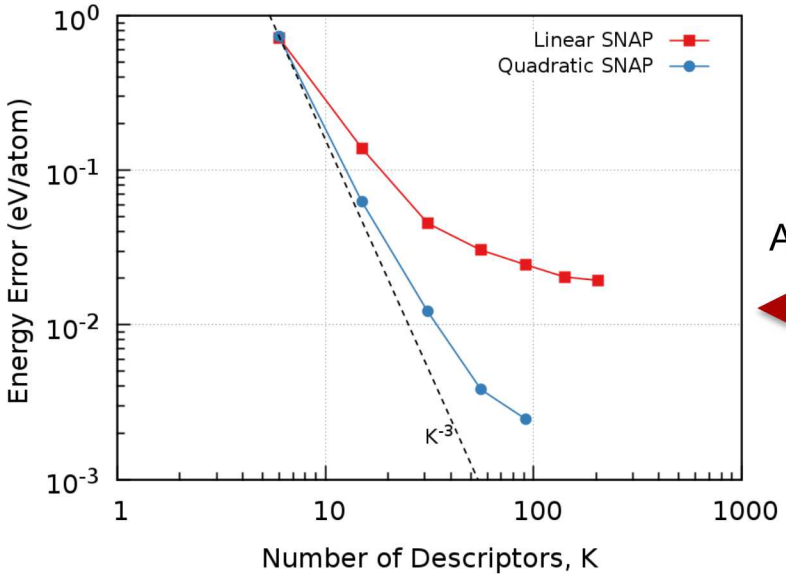
$$E^i_{SNAP} = \boldsymbol{\beta} \cdot \mathbf{B}^i + \frac{1}{2}(\mathbf{B}^i)^T \cdot \boldsymbol{\alpha} \cdot \mathbf{B}^i$$

- Linear terms are 4-body
- Quadratic terms are 7-body
- Number of linear coefficients grows as $O(J^3)$
- Number of quadratic coefficients grows as = $O(J^6)$

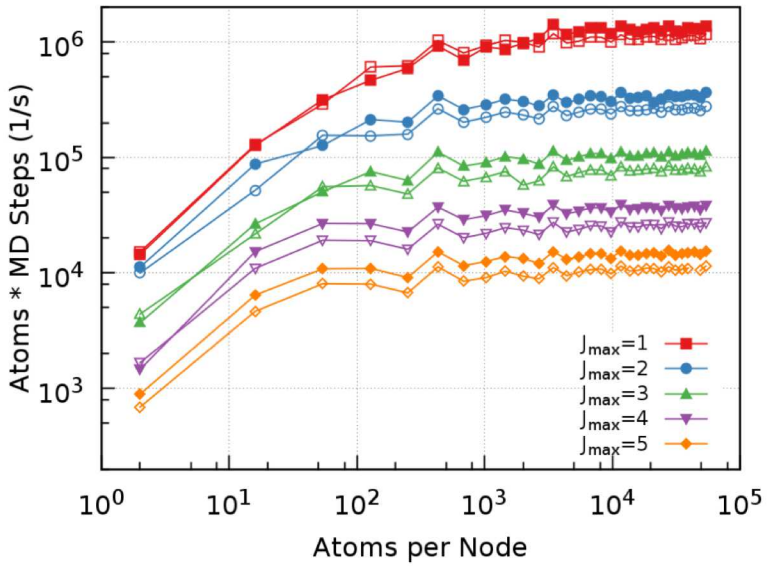
- Energy, force, stress remain **linear** in β and α
- Can still use linear least squares (SVD)
- Number of columns will increase from K to $K(K+1)/2$

Wood and Thompson, *J. Chem.Phys.*, March, 2018
Also https://arxiv.org/abs/1711.11131

SNAP Tantalum



Accuracy-speed tradeoff



Quadratic SNAP – Cross Validation

- Concerned with overfitting now that there are MANY more free parameters during the fit.
- (Training Points) : (Descriptors) still >> 1 for assembled training sets

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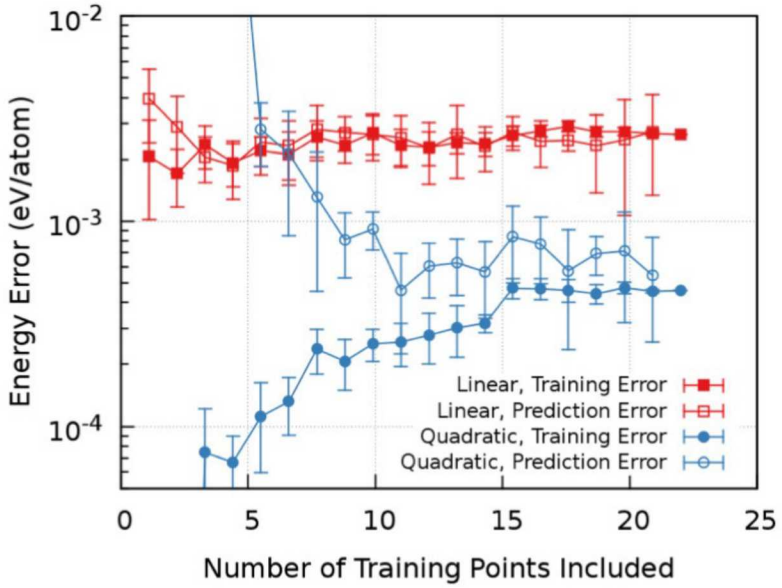

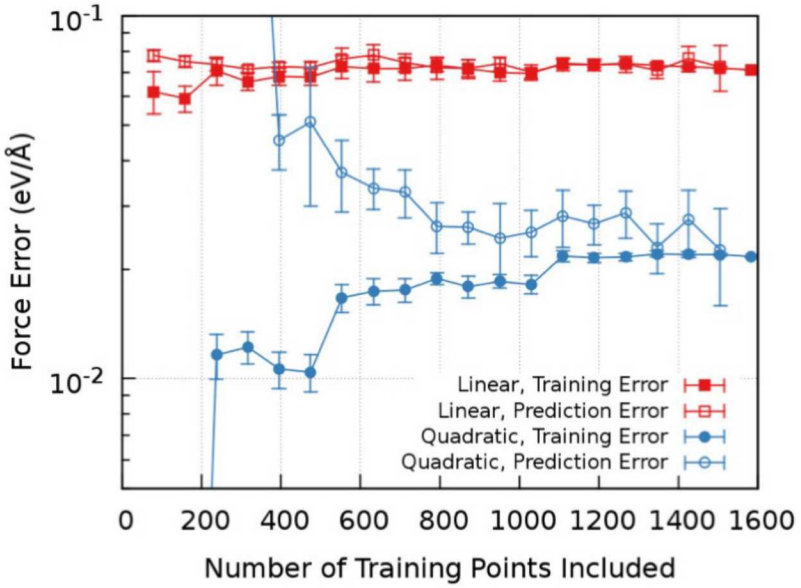

Conclusions

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- Molecular dynamics is a powerful tool for exploring materials behavior
- Can access physics, chemistry, length, and timescale that is inaccessible to other methods (continuum, quantum)
- Applications are driving demand potentials of greater accuracy
- We have built a system for converting large amounts of quantum calculations into SNAP potentials for large-scale MD simulations of complex materials
- SNAP strikes a good balance between speed, accuracy, and robustness
- We still don't really understand why ML potentials work or don't work
- Still lots of room for improvement

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