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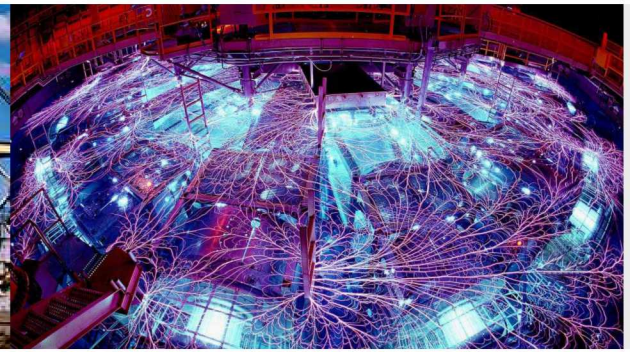

Scalable Machine Learned Interatomic Potentials With SNAP

Mitchell Wood, Mary Alice Cusentino and Aidan Thompson
Center for Computing Research, Sandia National Labs

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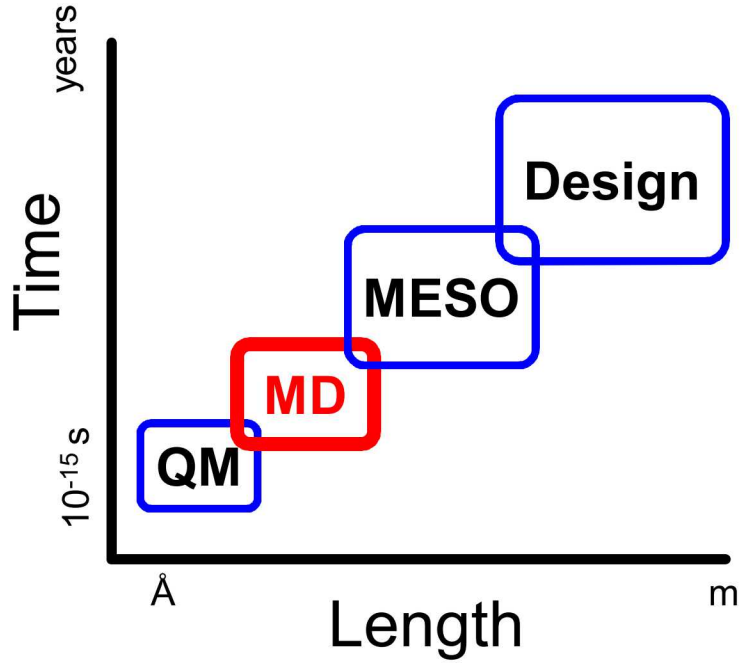
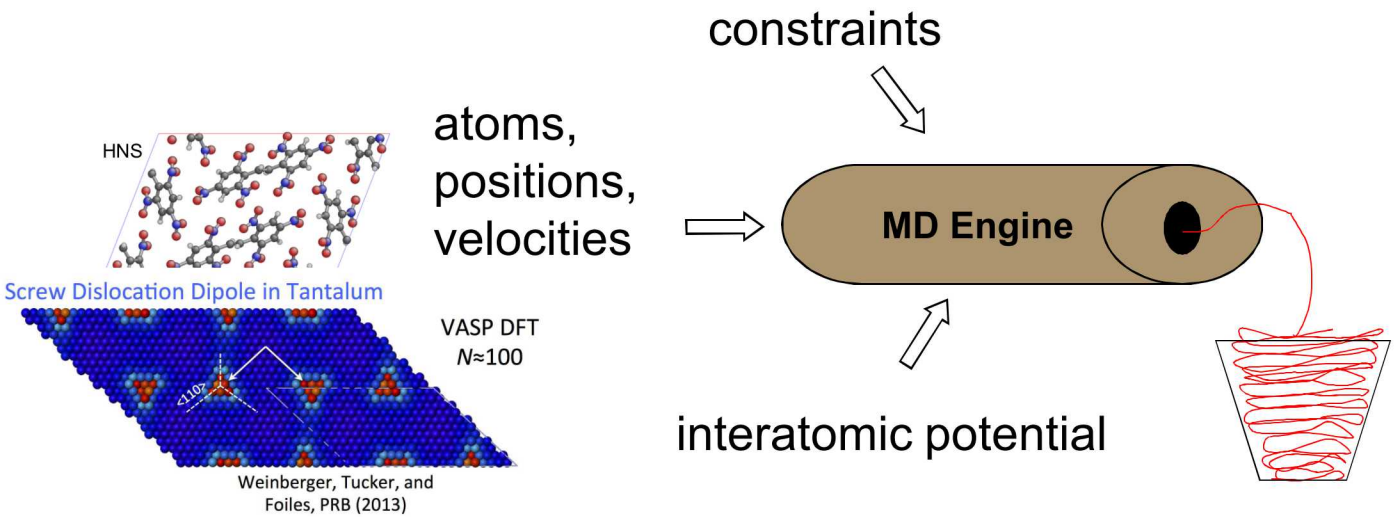
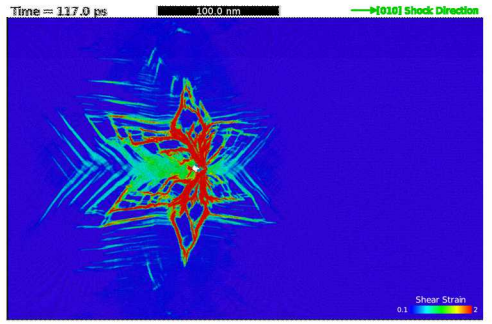
Molecular Dynamics Primer

- 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

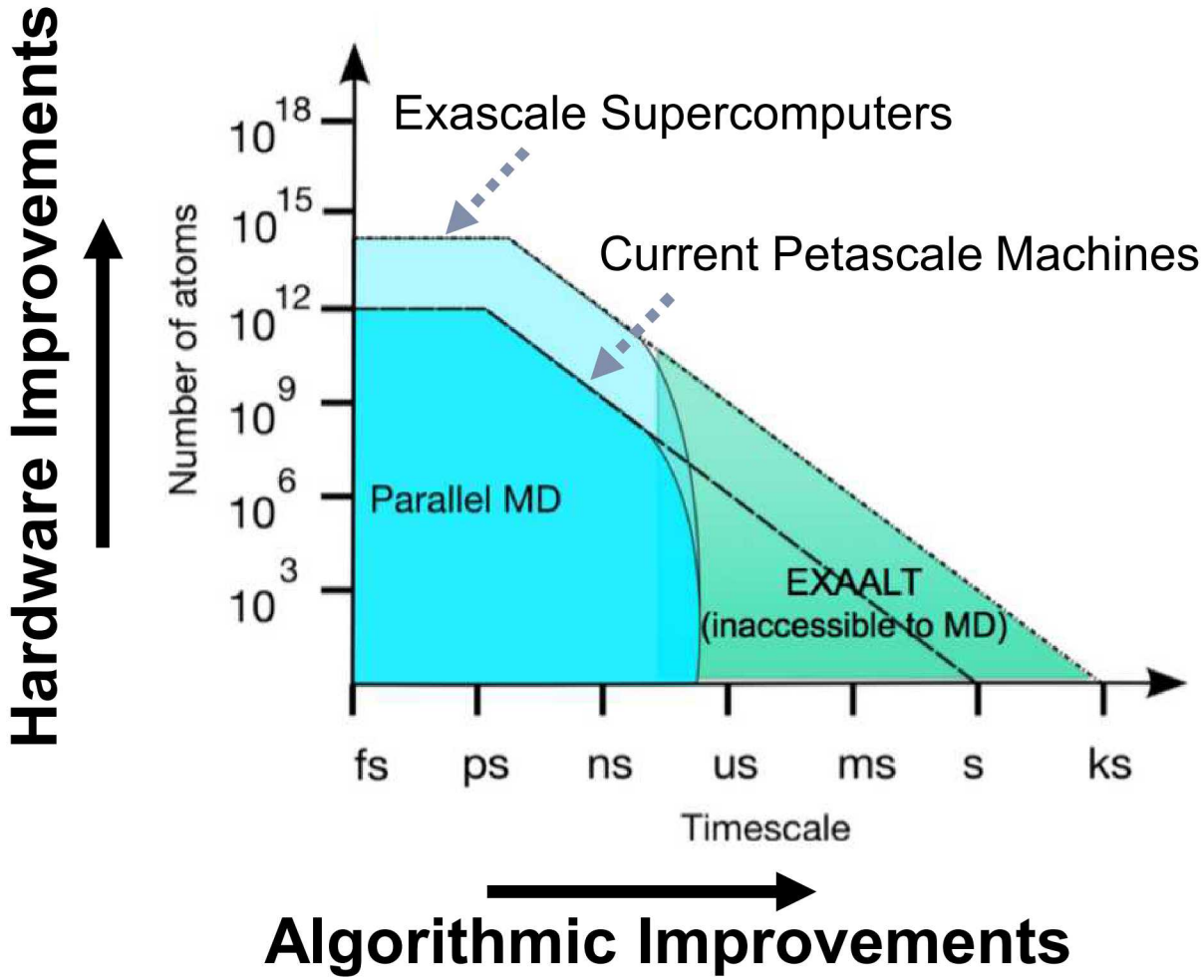




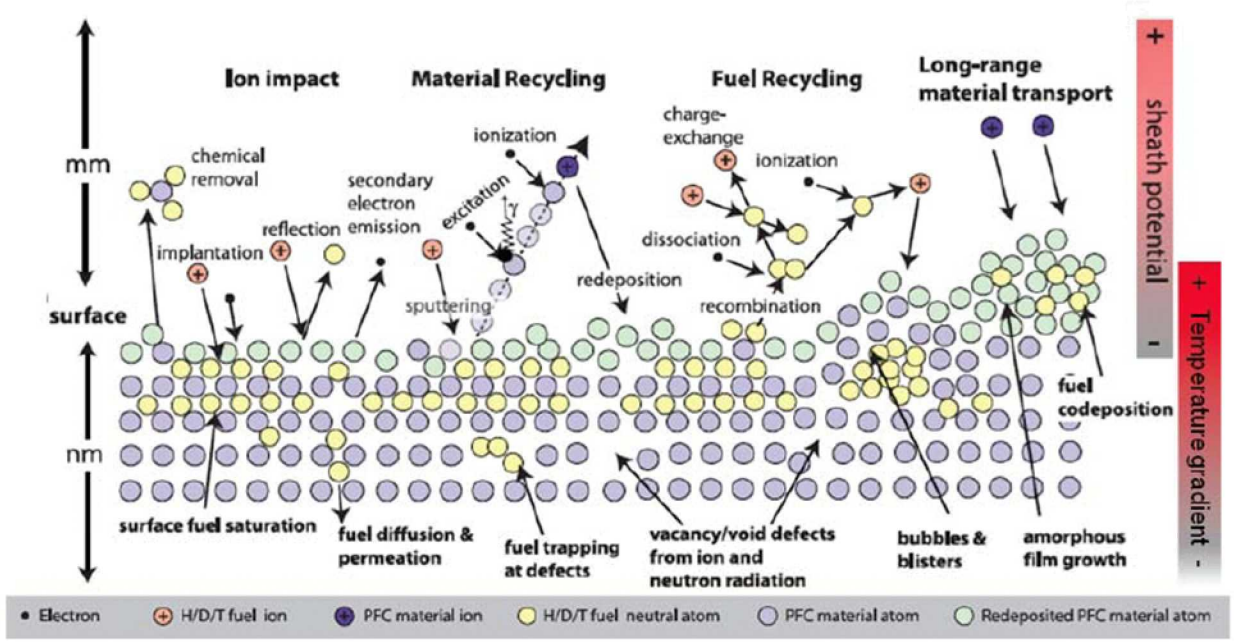


Positions, velocities and forces at many later times

Atomistic View of the (computational) World



- Key approximation of MD is the interatomic potential used
- Assumes all physics of the problem is contained on the Born-Oppenheimer potential energy surface



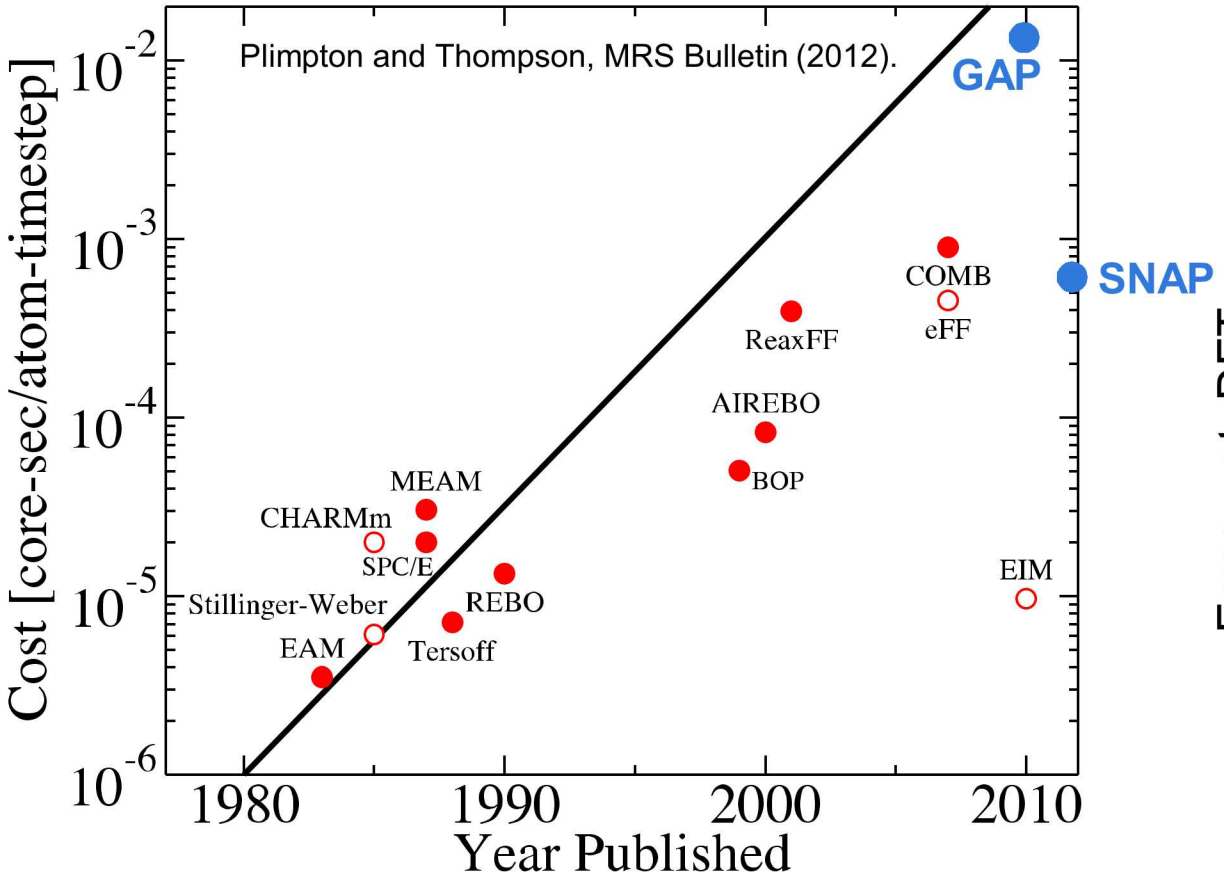
MD Approximations Change Over Time

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

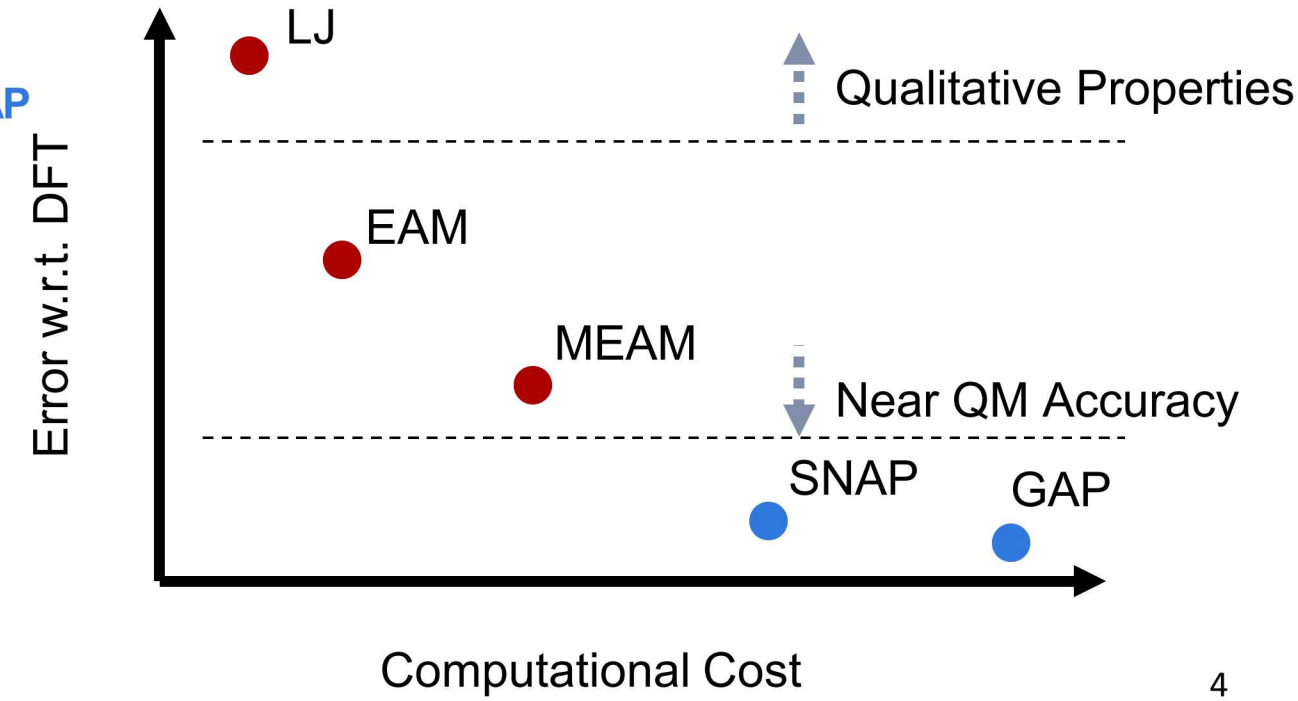
Manybody (1980s)
Stillinger-Weber, Tersoff, Embedded Atom Method

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

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



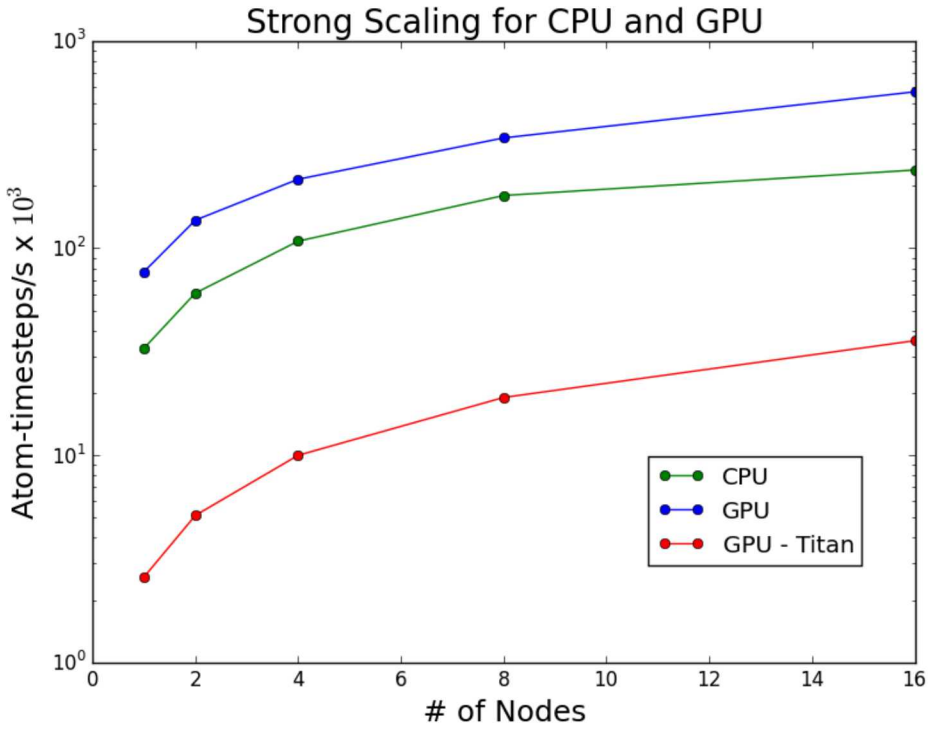
- **Resources are limited, which is your best choice?**



Computing Environments Change Over Time

Sandia National Laboratories

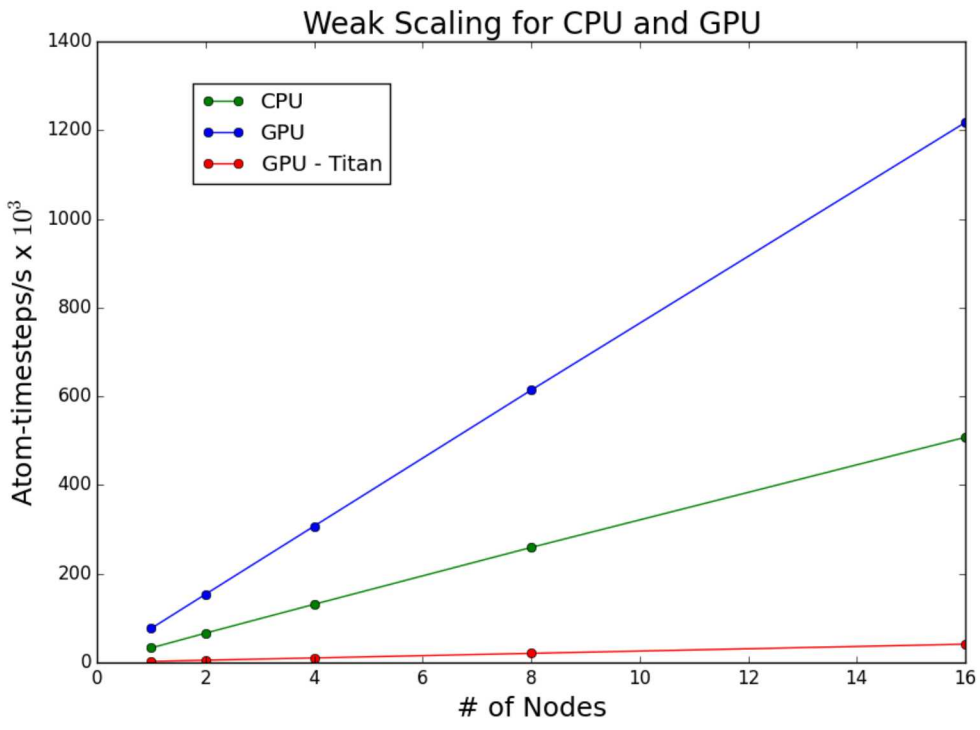
- SNAP will be Exascale ready on all hardware via KOKKOS implementation
- Performance on modern GPUs (P100's) far exceeds previous leadership platforms (Titan, K20X's)



4 P100 GPUs

Intel Broadwell

1 K20X GPU



Best Practices for Interatomic Potentials?

Sandia National Laboratories

Generating interatomic potentials is essentially a black box, this is true for all potential types. Plenty of progress to apply known optimization tools to the fitting, GA being the most common.

- Which training data tells us the most about a material?
- How much training data until we can say we are at DFT-level of accuracy?

Transferrable, User Generated Training

- **Better for distribution to a broad audience.**
- **User interaction with training sets and objectives is very high.**
- Accuracy is potentially sacrificed for stability.

Slower, safer but still looks like black magic to outside viewers.

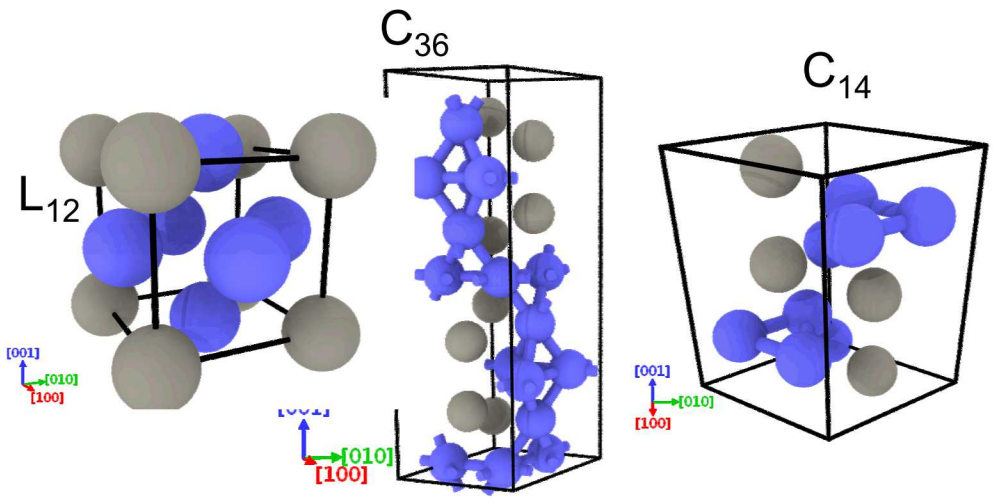
Less-Transferrable, Learn On-The-Fly

- Best when the target application is well known.
- **Hands off approach to parameterization.**
- **Accuracy is completely unknown for untrained systems.**

Potentially faster and the intended use is well known to end users

SNAP - A Look Under the Hood

Set of Descriptors



- Local density around each atom expanded in 4D hyperspherical harmonics
- **Bispectrum coefficients are a superset of the bond-orientational order parameters, in 4D space.**
- Preserve universal physical symmetries: invariance w.r.t. rotation, translation, permutation

Model Form

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

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

Regression Method

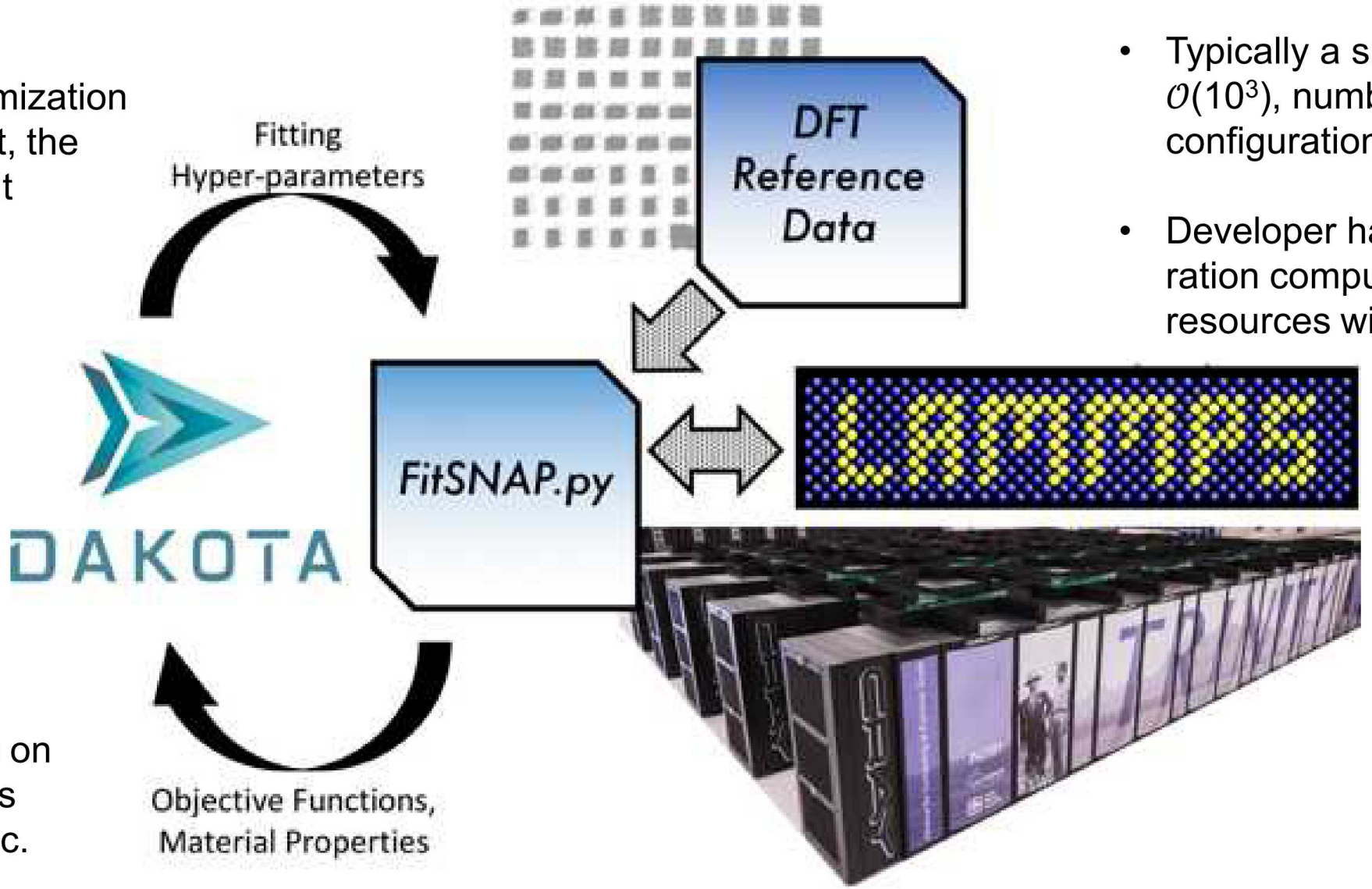
- β vector fully describes a SNAP potential, found through a weighted linear regression.
- Decouples MD speed from training set size

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

Weights Set of Descriptors DFT Training

Semi- Automated Generation Interatomic Potentials

- Even if the optimization routine is robust, the process still isn't transparent.



- Typically a small, of $\mathcal{O}(10^3)$, number of configurations.
- Developer has to ration compute resources with DFT

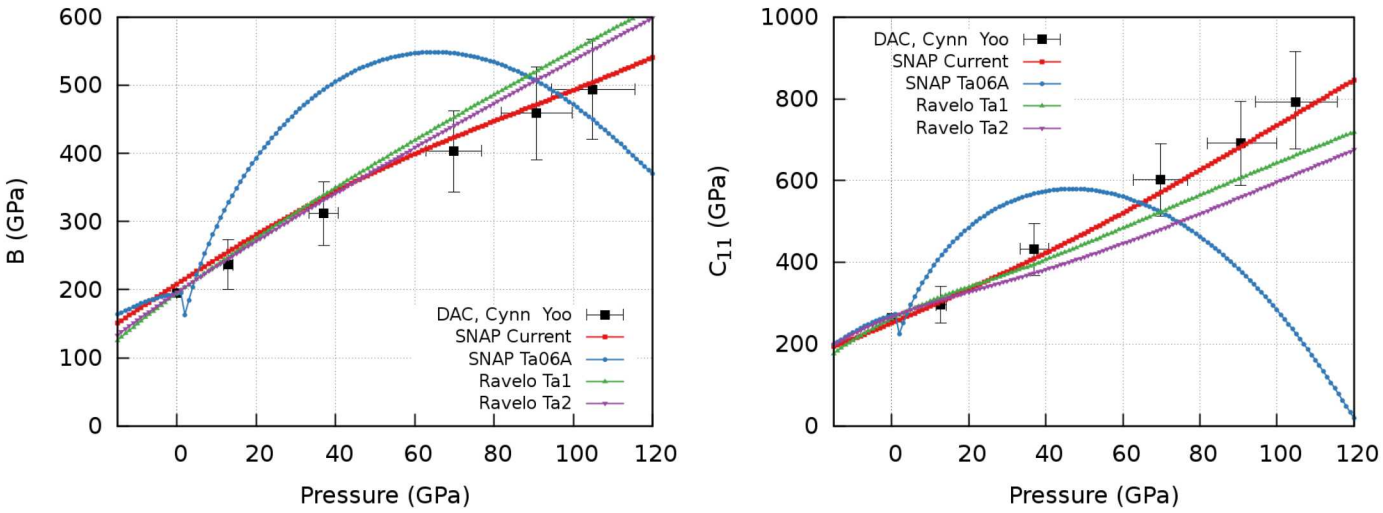
- The importance on each objective is part of the magic.

SNAP as a Many-Body Correction Term

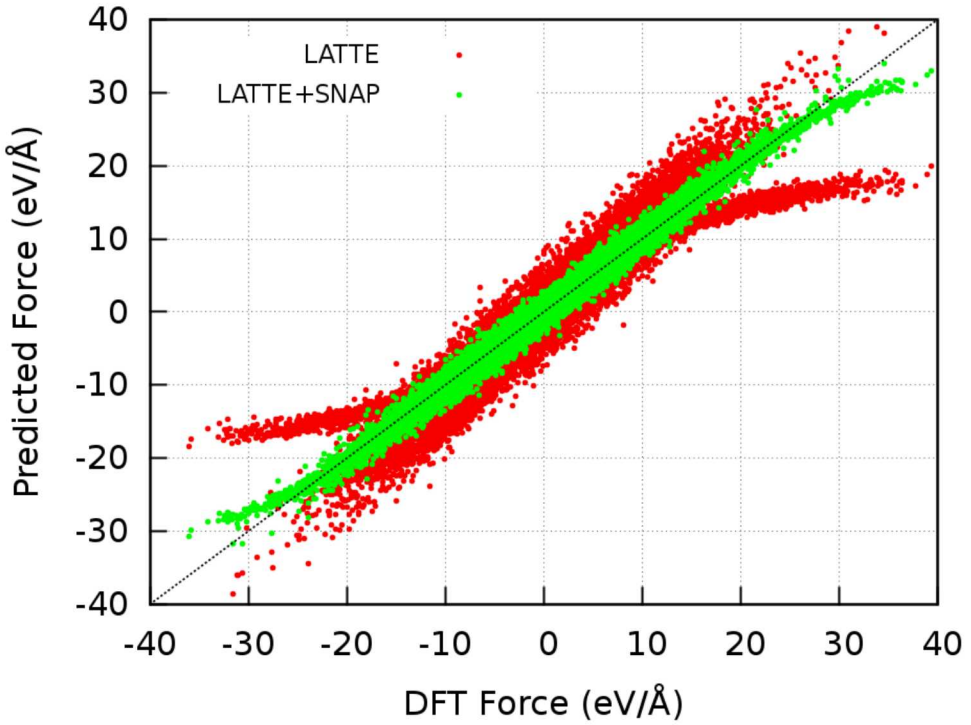
- Base potential is E_{ref}, typically ZBL is used to constrain at small atom separations.
- Could be Columbic, LJ, EAM, etc.

$$E(\mathbf{r}^N) = E_{ref}(\mathbf{r}^N) + E_{SNAP}(\mathbf{r}^N),$$

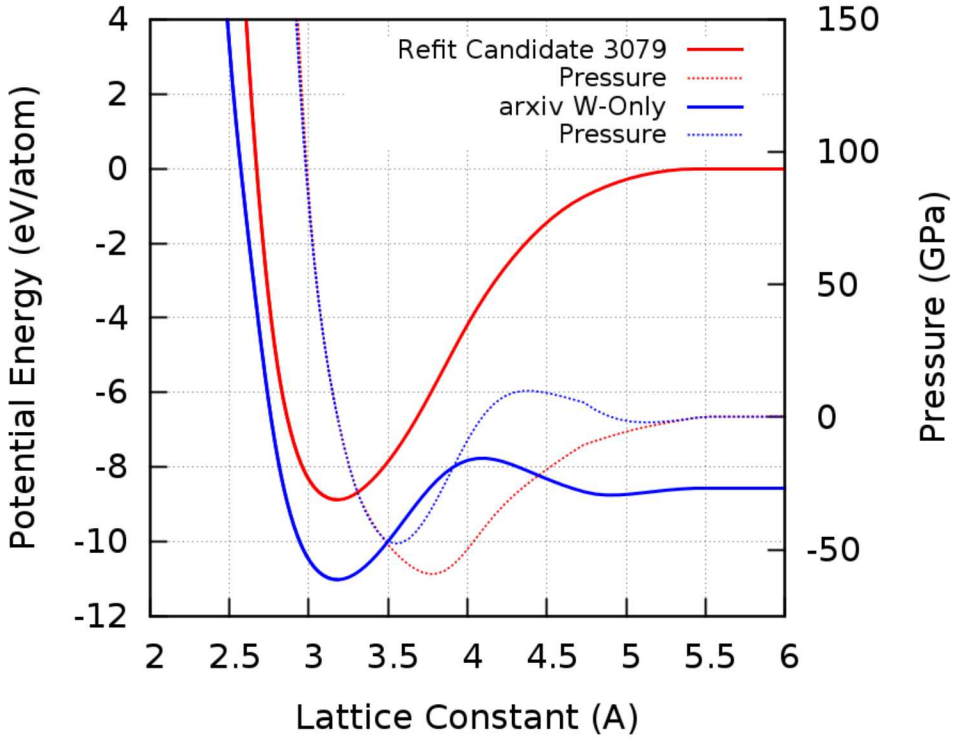
SNAP + EAM :



SNAP + Tight Binding :



Changing the Functional Form of SNAP

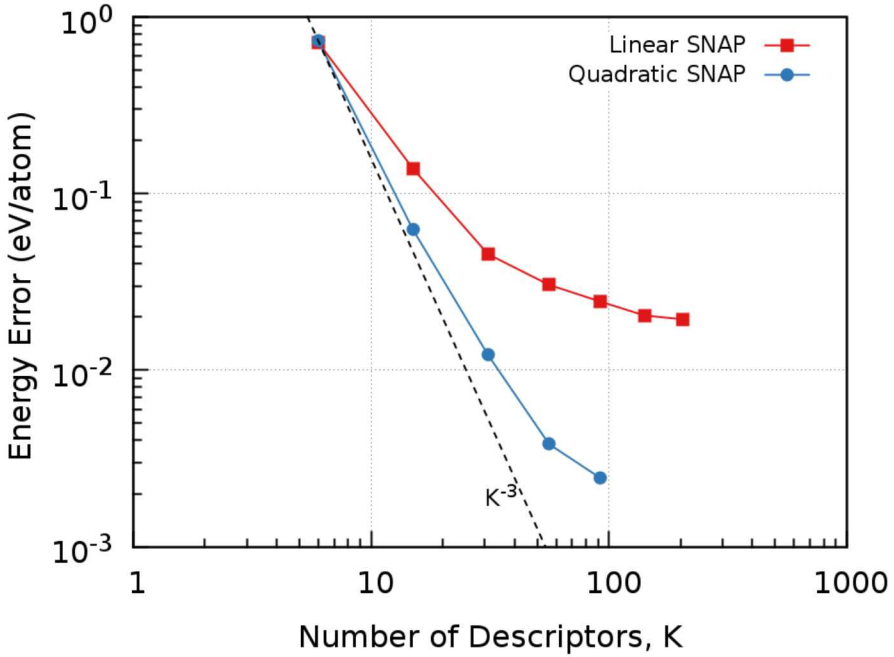


- Removing some just makes physical sense, i.e. cohesive energy of crystals.

- Bispectrum components are non-zero as r→∞, energy functional needed to be corrected

- When expanding the energy functional beyond linear order in the bispectrum, significant accuracy increase observed.

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

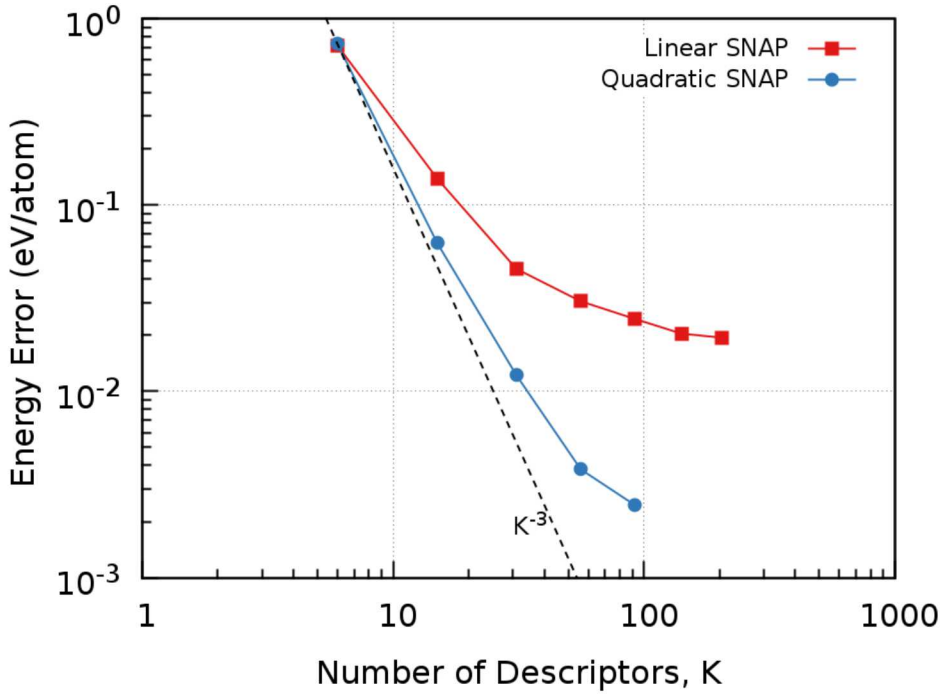


Quadratic SNAP

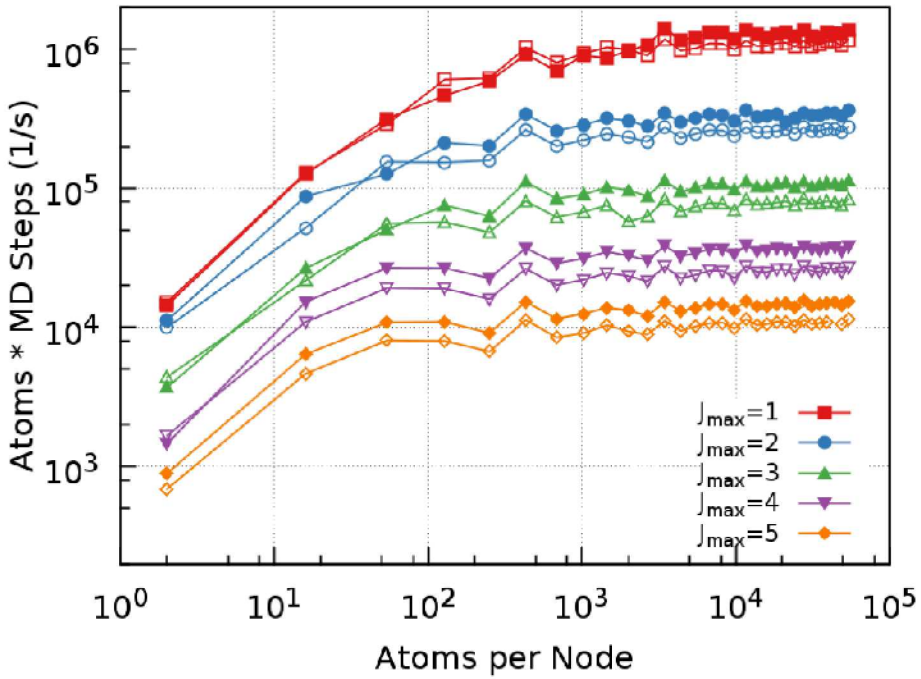
$$E^i_{SNAP} = \boldsymbol{\beta} \cdot \mathbf{B}^i + \frac{1}{2}(\mathbf{B}^i)^T \cdot \boldsymbol{\alpha} \cdot \mathbf{B}^i$$

- 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
- *C*an still use linear least squares (SVD)
- Number of columns will increase from *K* to K(*K*+1)/2



Accuracy-speed tradeoff

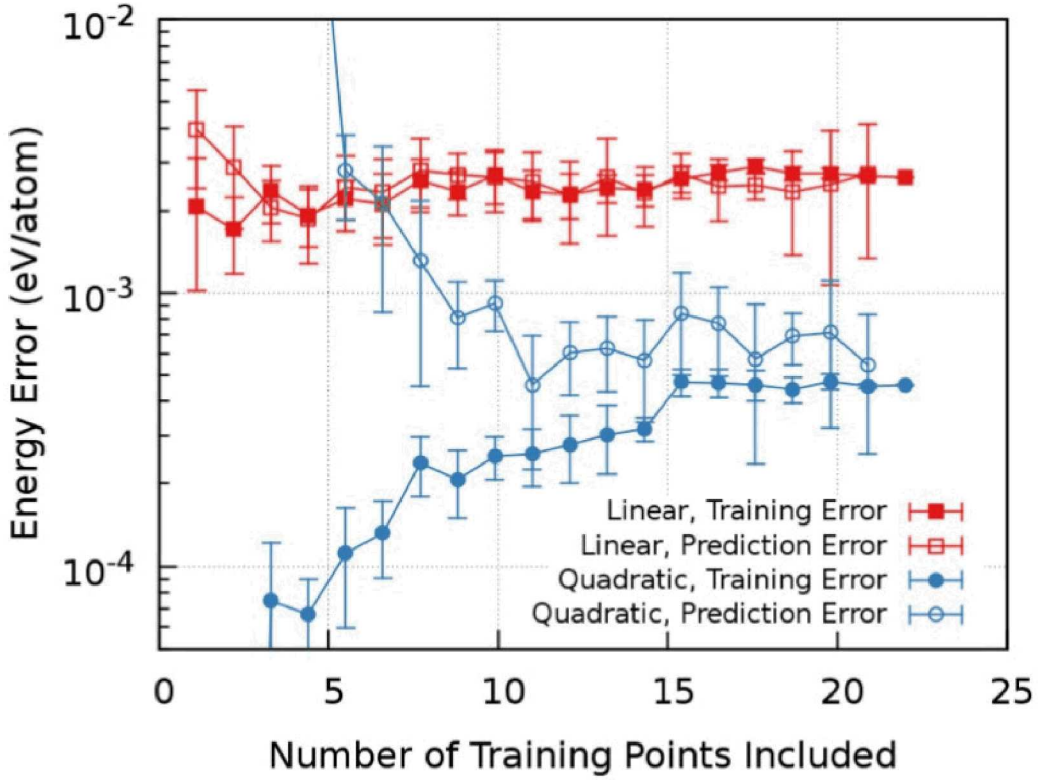


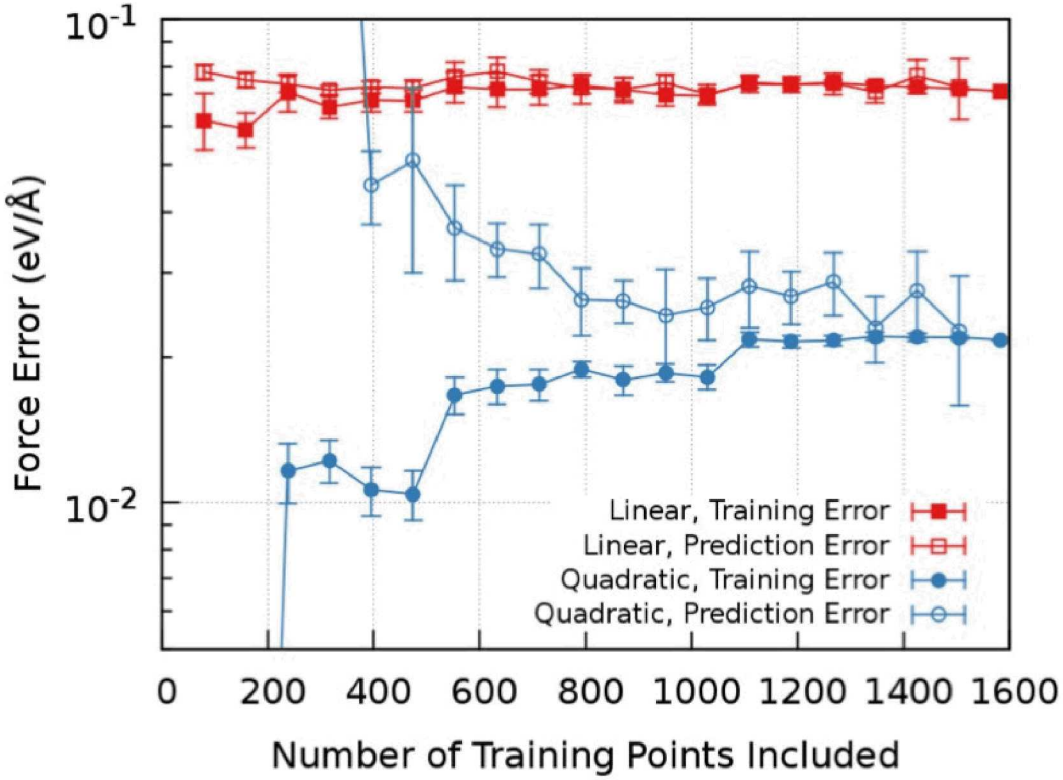
Wood and Thompson, *J. Chem.Phys.*, March, 2018 Also https://arxiv.org/abs/1711.11131

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

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

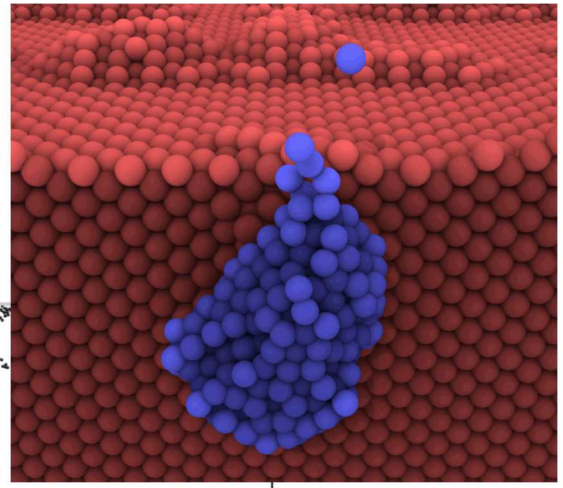


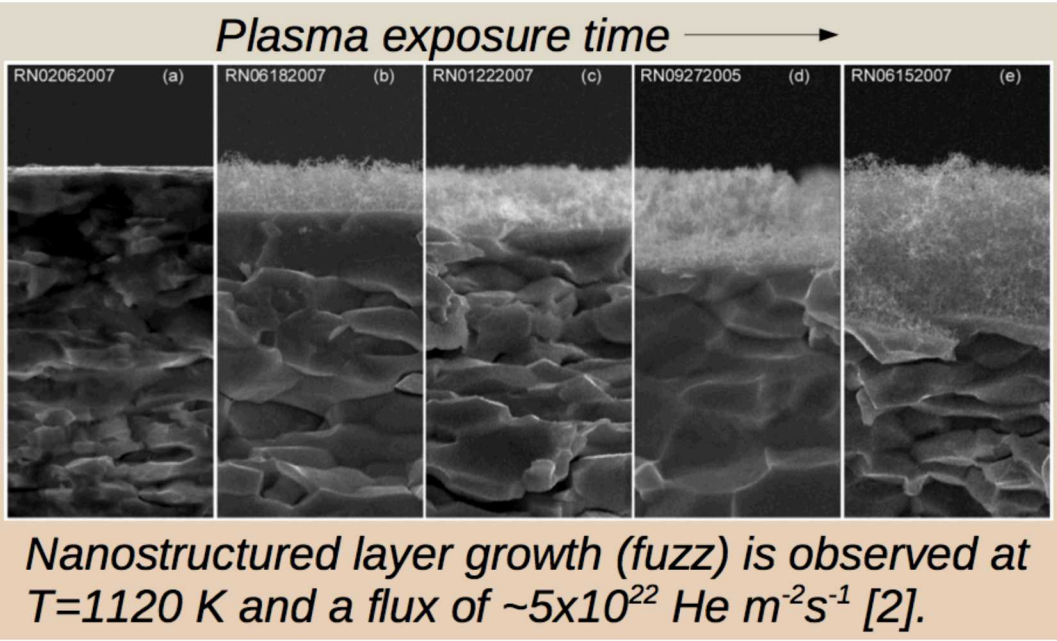


Plasma-Facing Materials

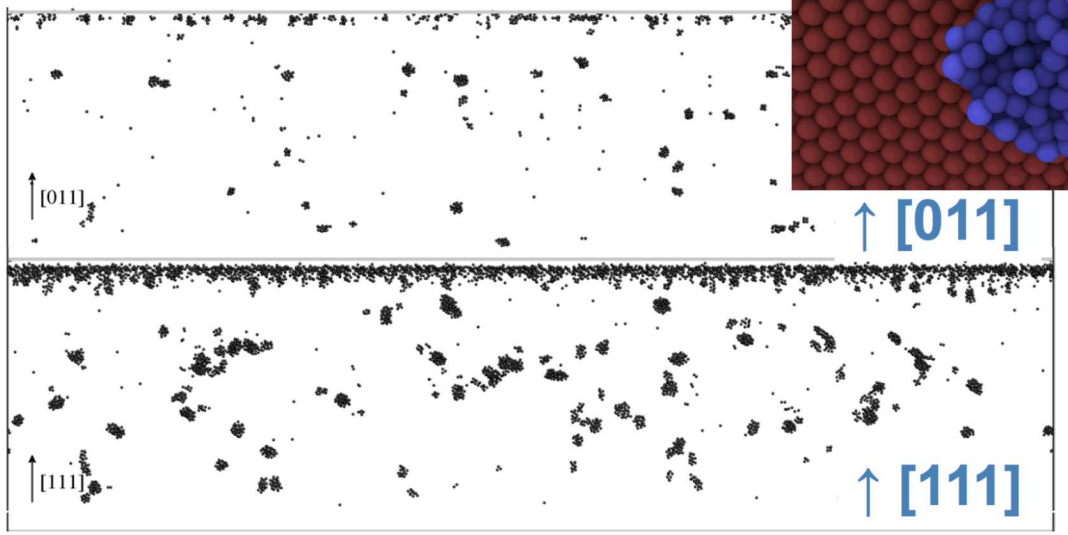
ITER fusion reactor:

- Plasma-facing material is tungsten
- Exposed to He and H at elevated temperature
- Fuzz buildup limits power output and useful life of diverter elements





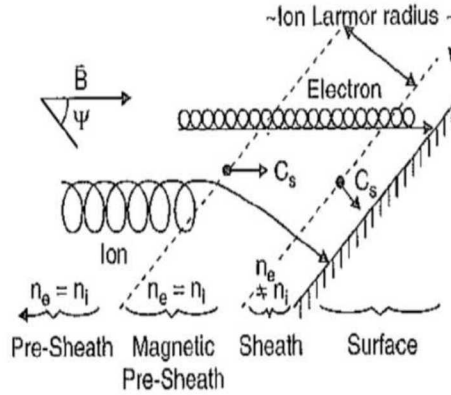
Nanostructured layer growth (fuzz) is observed at T=1120 K and a flux of ~5x10^{22} He m^{-2}s^{-1} [2].



Luis Sandoval, Blas Uberuaga, Danny Perez, Art Voter, Phys. Rev. Lett. (2015)
Sefta, F., Hammond, K. D., Juslin, N., & Wirth, B. D. (2013). *Nuclear Fusion*, *53*(7), 073015.

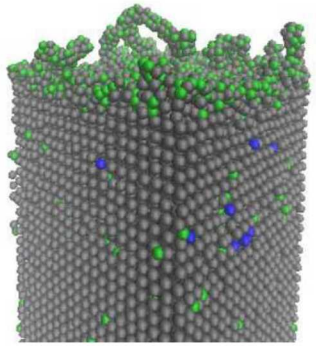
Atomistic Simulation Efforts

- Objective is to develop coupled simulation capability across three distinct spatial regions:
  1. **Edge/Scrape-off-layer** of the plasma, with sheath effects (W,Be + He,H)
  2. **Near Surface Material** response to plasma exhaust
  3. **Structural Materials** response to intense, 14MeV peaked neutron spectrum



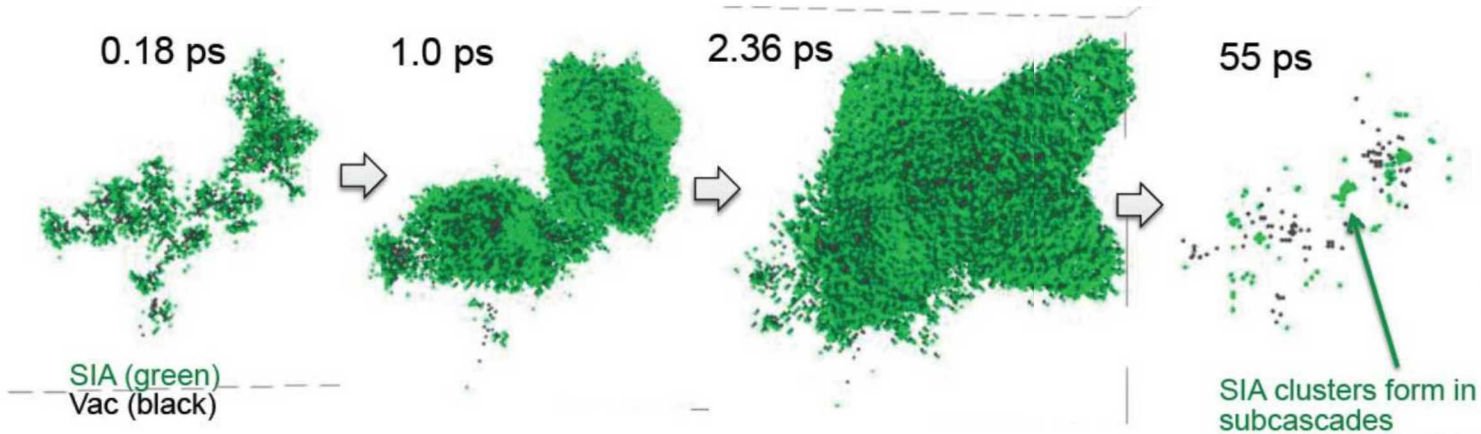
Plasma Edge SOL Sheath: heat & particle flux, recycling, etc.

~mm



Material surface: erosion (impurity & dust), T retention, surface evolution

nm to μm



Material bulk: neutron damage & transmutation

μm to mm

Training SNAP for Transferability – Tungsten

- In many cases we want a general use potential that many users can apply to their research needs.

Elastic Deformations
- ~2200 configurations

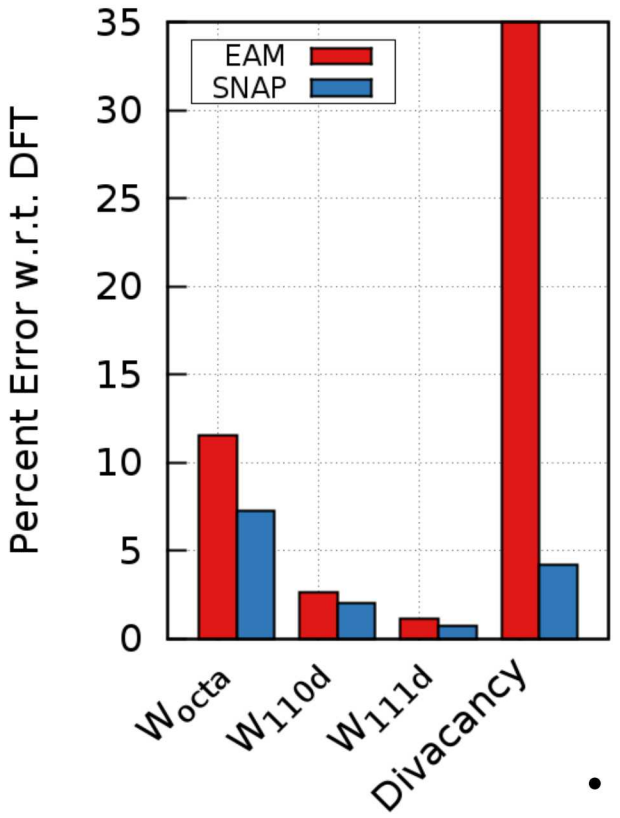
Defects (Interstitials, Vacancies, Dislocations)
- ~600 configurations

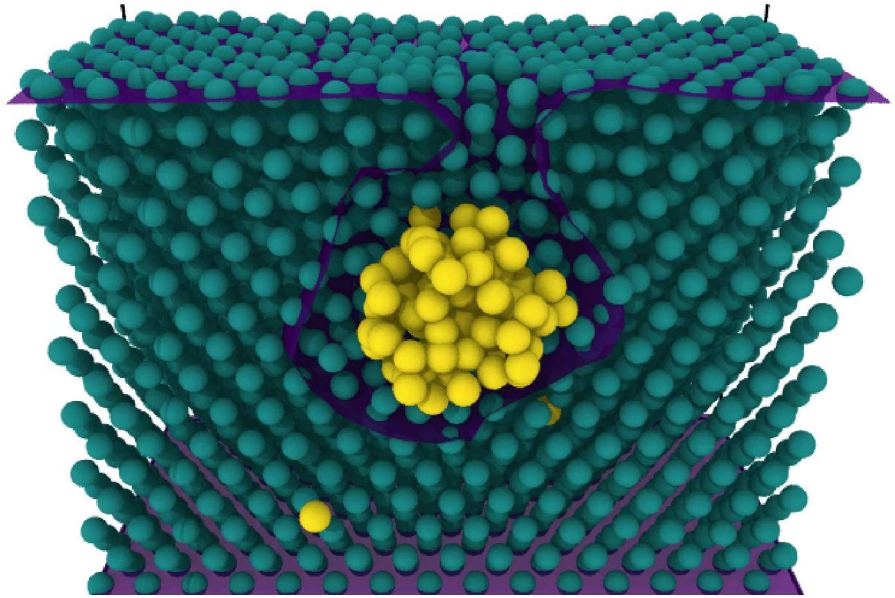
Free Surfaces
- ~200 configurations

DFT-MD Trajectories (Solid and Liquids)
- ~100 configurations

Gamma Surface
- ~7000 configurations

- ‘Plug-and-Play’ capability with existing W-He and He-He potentials. Easily fix shortcomings of existing tungsten EAM potentials.





- (Above) Interested in He-bubble bursting

Training SNAP for Transferability – Beryllium

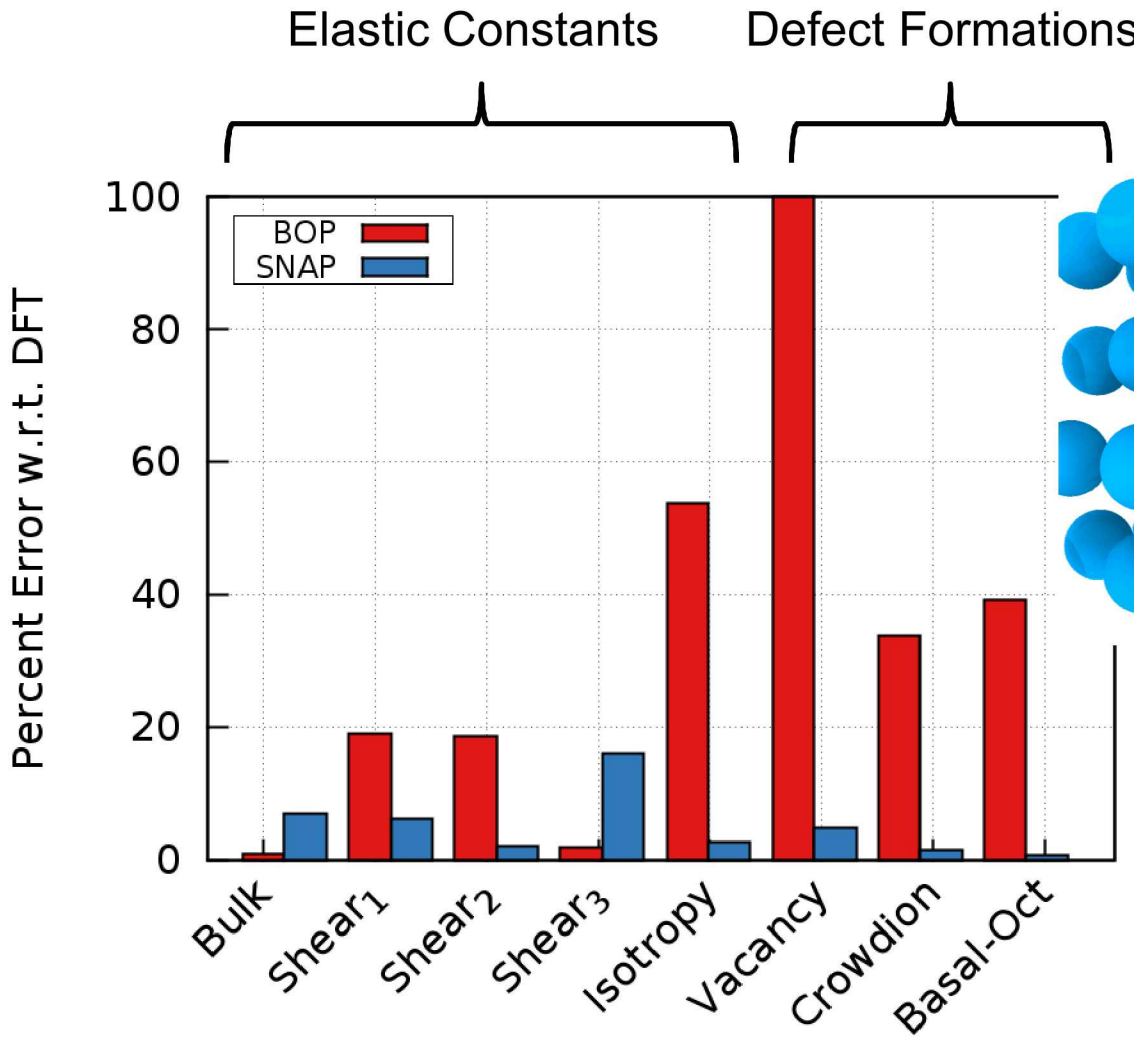
- Few existing Be potentials, challenging material system.
- Significant improvement over existing bond order potentials (BOP)

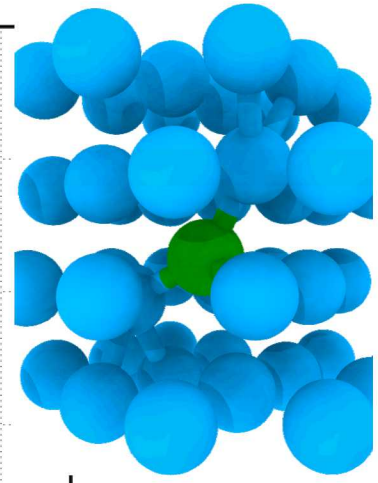
Elastic Deformations (HCP, FCC, BCC)
- ~4500 configurations

Defect Relaxations (Interstitials, Vacancies)
- ~170 configurations

Free Surfaces
- ~100 configurations

DFT-MD Trajectories (Solid and Liquids)
- ~1100 configurations





SNAP relaxed crowdion defect

BOP: C Björkas *et al* 2010 *J. Phys.: Condens. Matter* **22** 352206

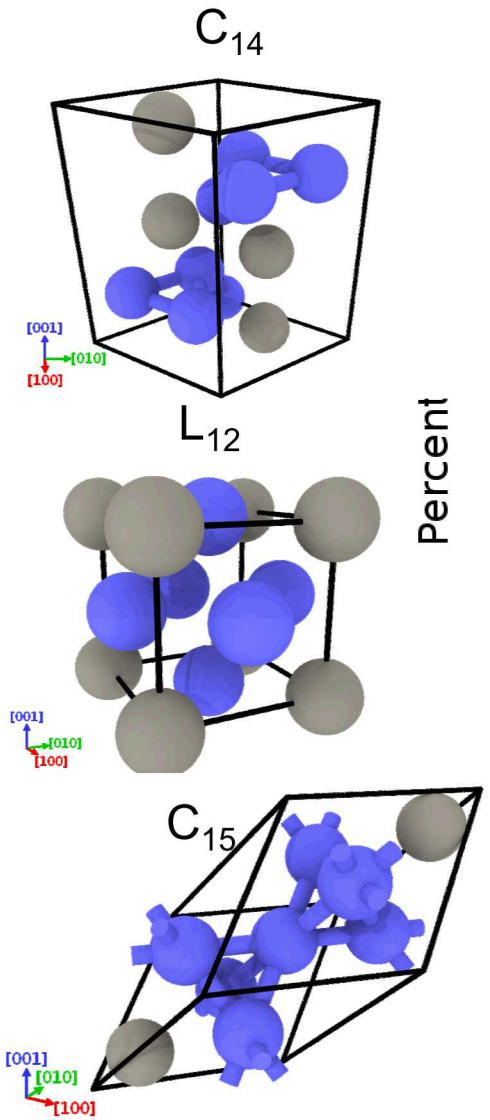
Training SNAP for Transferability – Tungsten+Beryllium

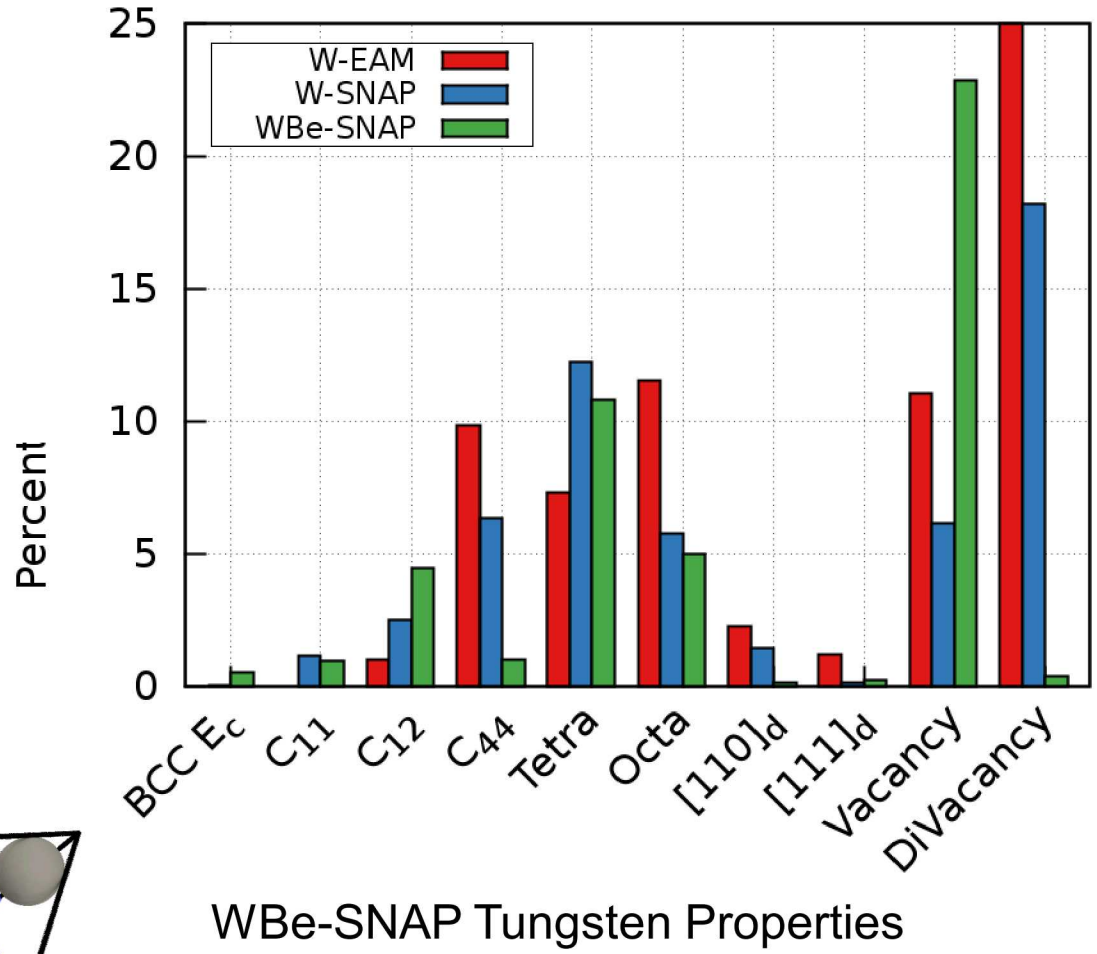
- Making a multi-element potential means sacrificing some accuracy from either pure component form.
- So far the focus of the joint potential has been on ordered phases of WBe

Elastic Deformations
- ~5400 configurations

DFT-MD Trajectories
- ~3500 configurations

Surface Adhesion
- ~400 configurations





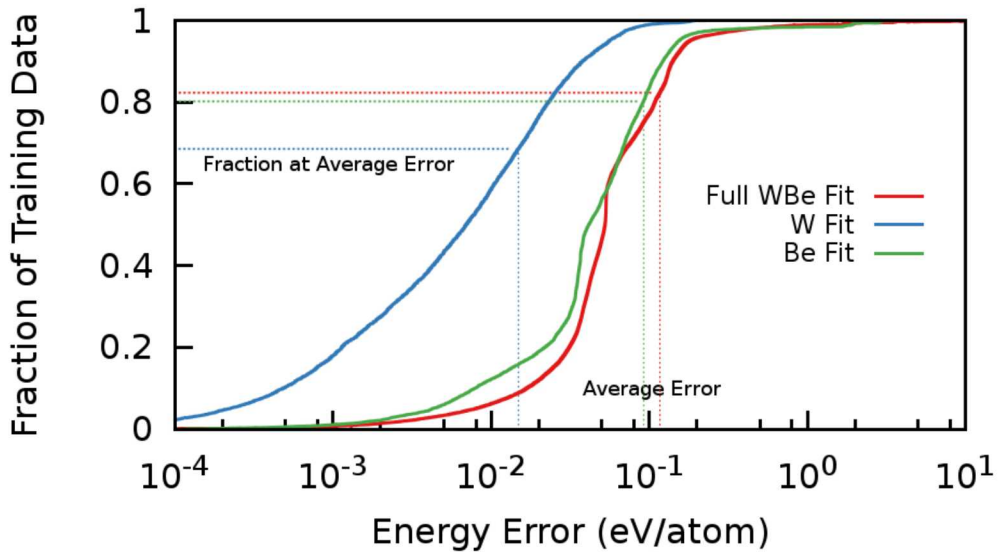
WBe-SNAP Tungsten Properties

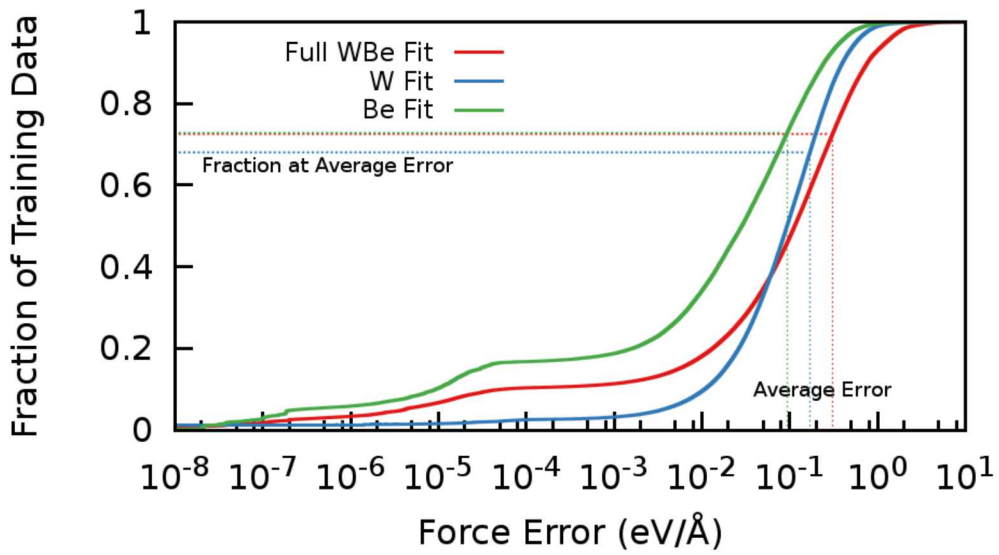
Training SNAP for Transferability – Tungsten+Beryllium

- Making a multi-element SNAP potential does sacrifice some accuracy from either pure component fit.
- No curation of the training data was done to remove ‘bad actors’

Fit weight applied to Energy/Forces

Description	N_E	N_F	σ_E	σ_F
W-Be:				
AIMD	3360	497124	$7 \cdot 10^4$	$6 \cdot 10^2$
Elastic Deform	3946	68040	$\mathbf{3 \cdot 10^5}$	$2 \cdot 10^3$
Equation of State	1113	39627	$2 \cdot 10^5$	$4 \cdot 10^4$
Surface Adhesion	381	112527	$2 \cdot 10^4$	$\mathbf{9 \cdot 10^4}$
Total:	8800	717318		





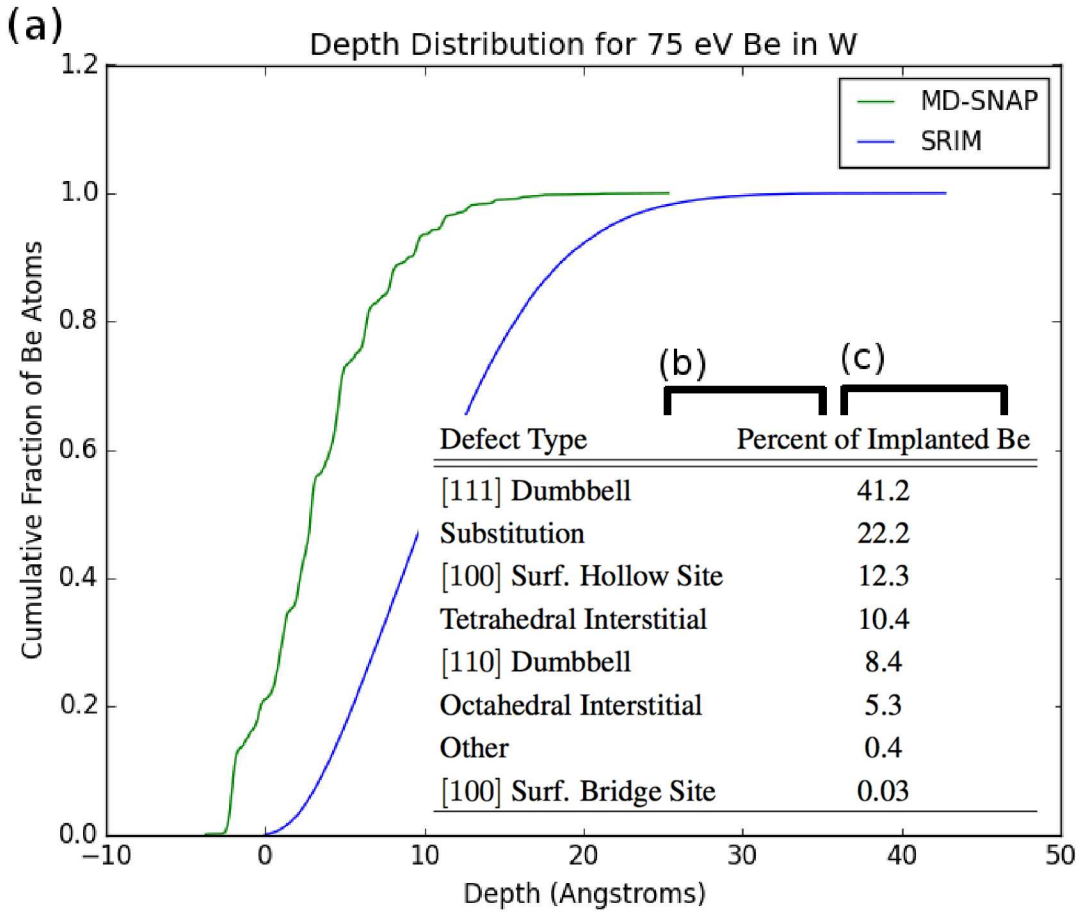
Out of Training Predictions

- Only one other W-Be potential available in LAMMPS, a Bond Order Potential (BOP) from Björkas *et al* 2010 J. Phys.: Condens. Matter

Defect Type	Formation Energy (eV)		
	DFT	SNAP	BOP
Tetrahedral Interstitial	4.13	4.20	-3.92
Octahedral Interstitial	3.00	5.11	-3.29
Substitution	3.11	3.29	-2.00
[111] Dumbbell	4.30	3.66	-3.20
[110] Dumbbell	4.86	4.29	-3.66
[100] Surf. Hollow Site	-1.05	-1.39	-3.52
[100] Surf. Bridge Site	1.01	0.44	-1.30

Much closer to DFT predictions

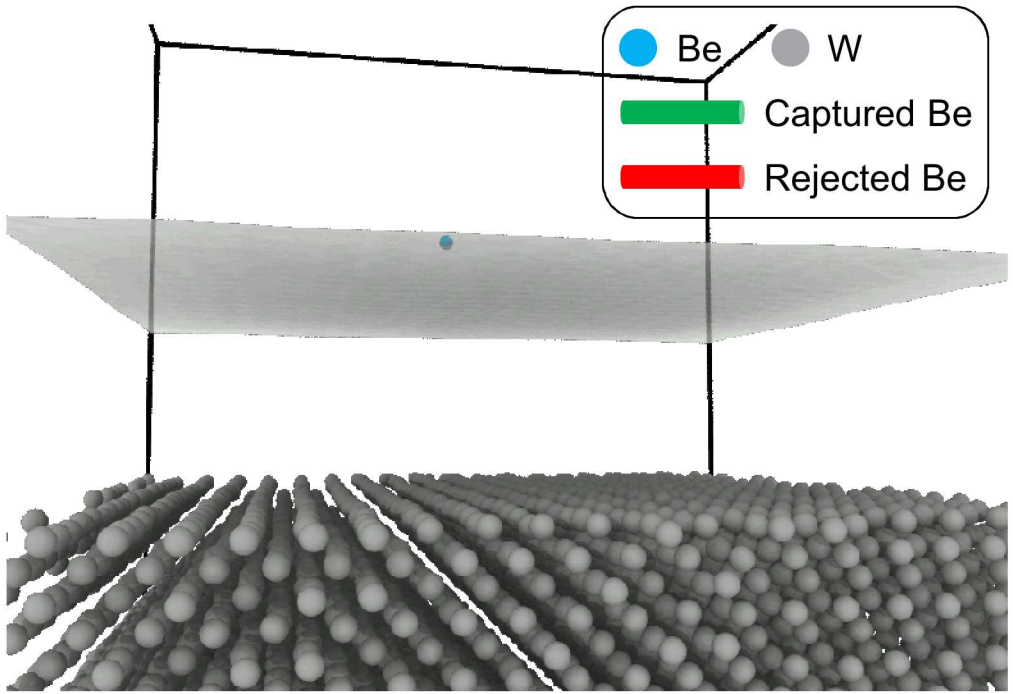
Energetically favored interstitial defects

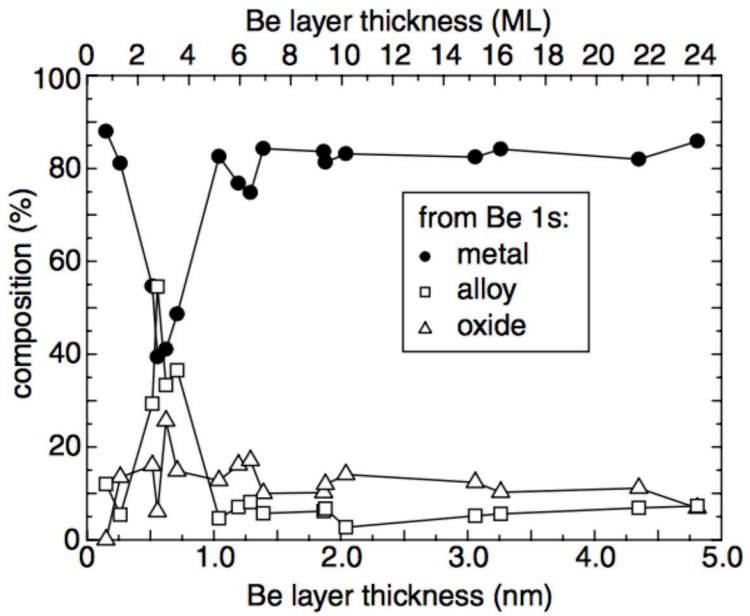


*~60% of total Be is not captured in the W matrix

Scrape-off Layer: Be Deposition on W Diverter

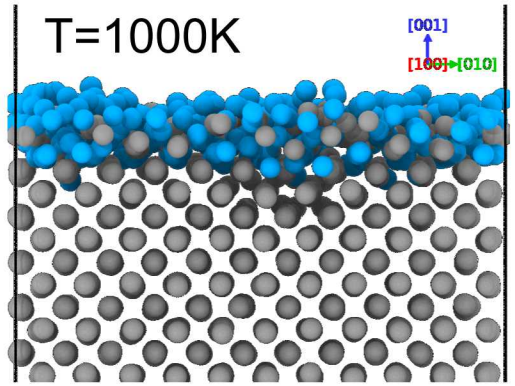
1) Accumulation of Be at W-surfaces
2) Formation of Be-rich intermetallic phases (WBe_2, WBe_{12})
3) Intermetallic phases have lowered melting temperatures
4) Degradation of diverter rapidly quenches plasma

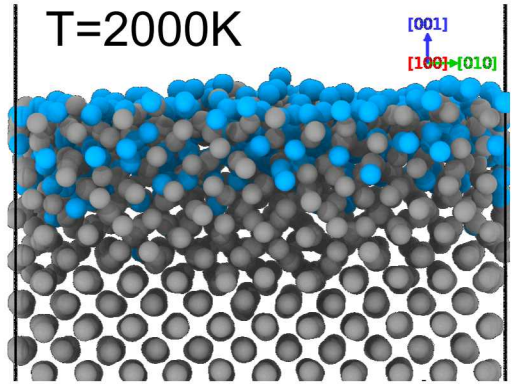




Wiltner, Linsmeier Journal of Nuclear Materials 337–339 (2005) 951–955

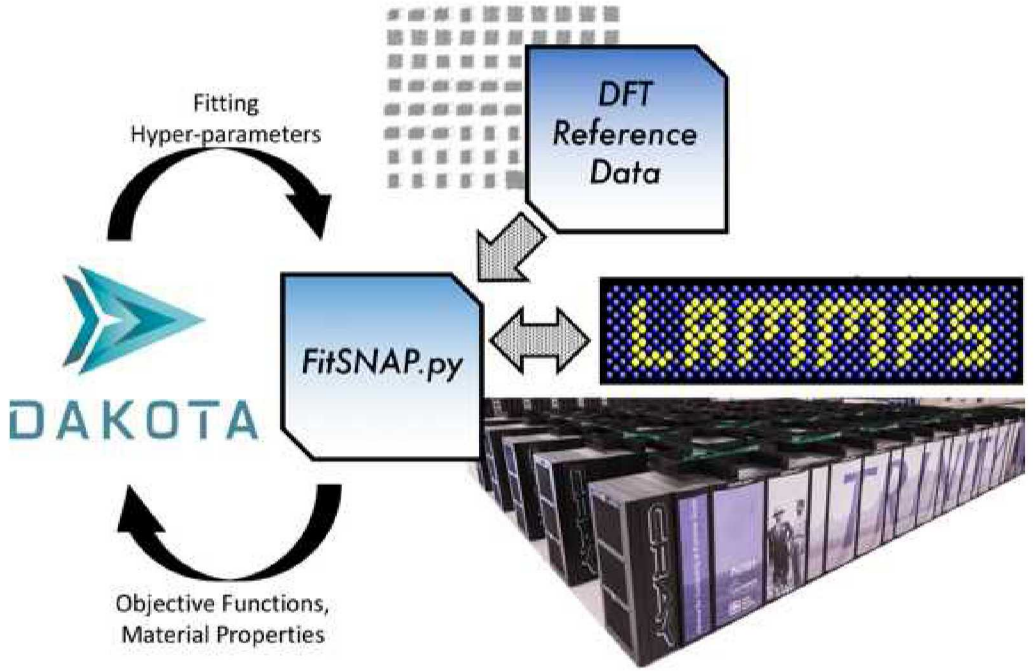
- Be film persists at high temperature:





Conclusions/Path Forward

- SNAP development is targeting a space where few other potentials exist, or where traditional energy functionals lack quantitative accuracy.
- Biggest worry about generated potentials is how ‘robust’ they are.
- Understanding the pitfalls in a potential is far more valuable than knowing its strengths
- Questions?



Extra Slide Content

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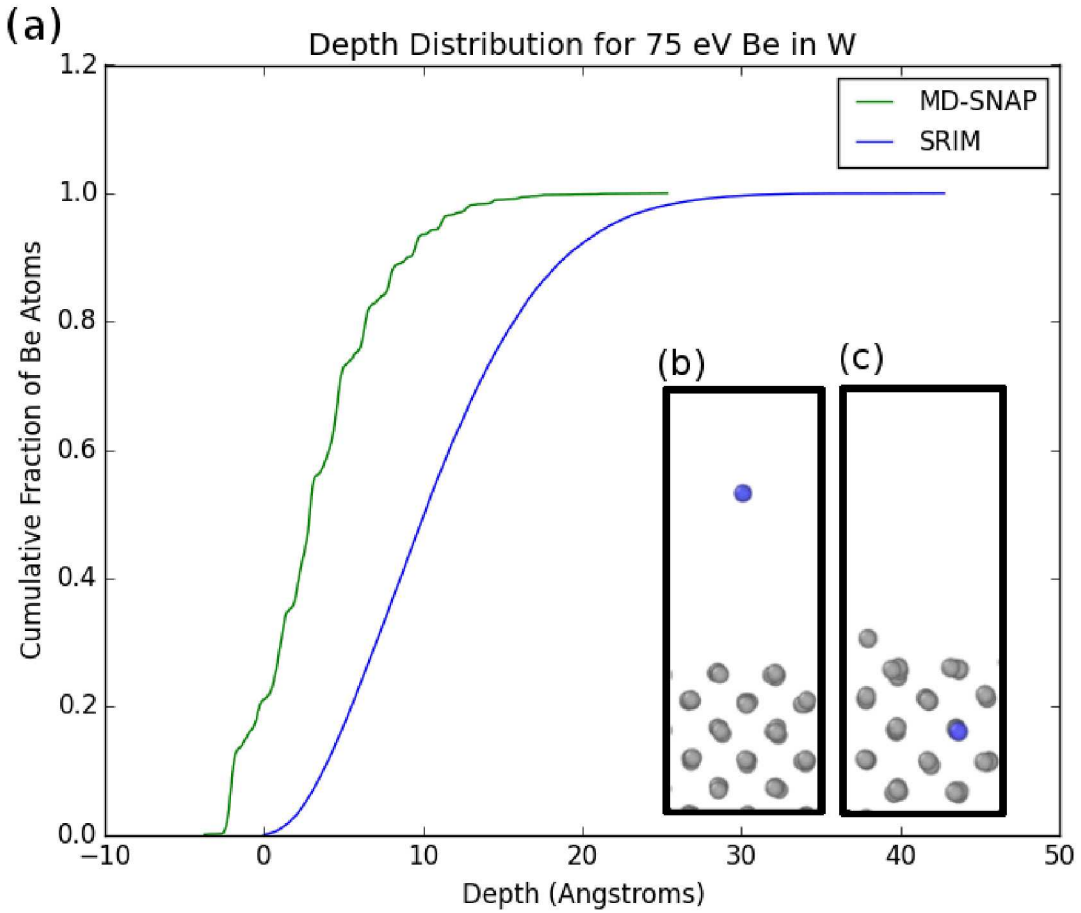
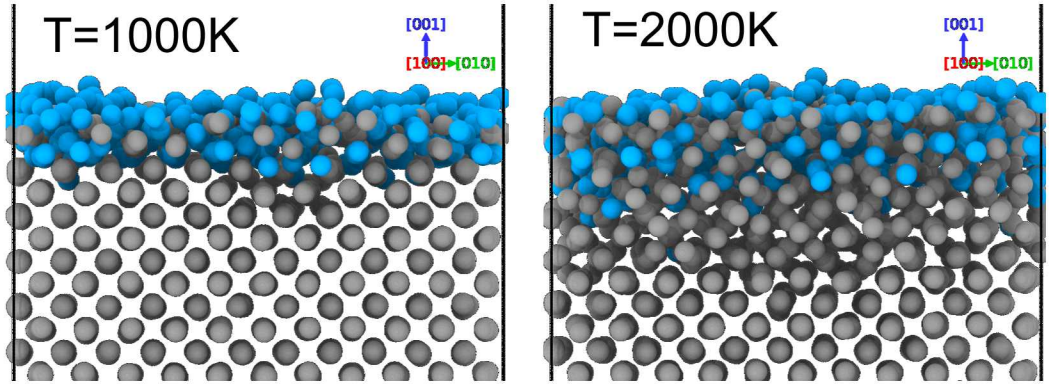
7) Figure and Table



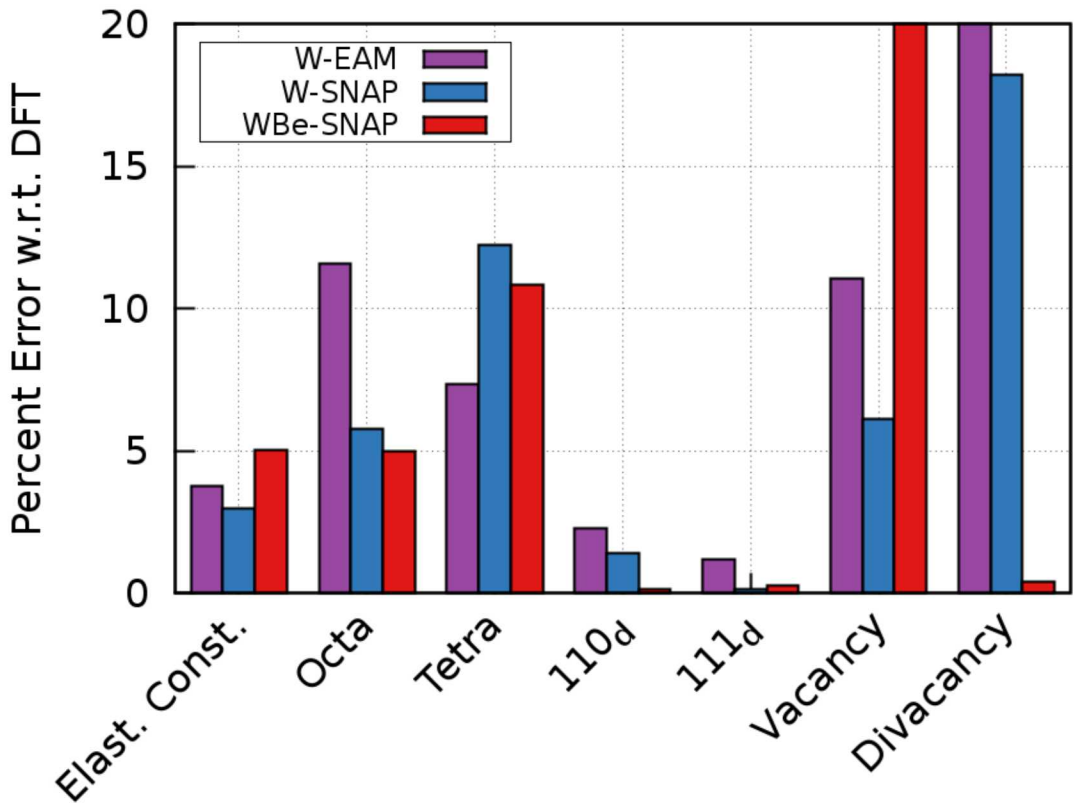
(a) Cumulative depth distribution plot for SNAP and SRIM
(b) Atomistic snapshot of initial position of atoms
(c) Atomisitic snapshot of final position of atoms after single Be Implantation, Be located at substitutional site and the displaced tungsten atom is now on the surface as an adatom

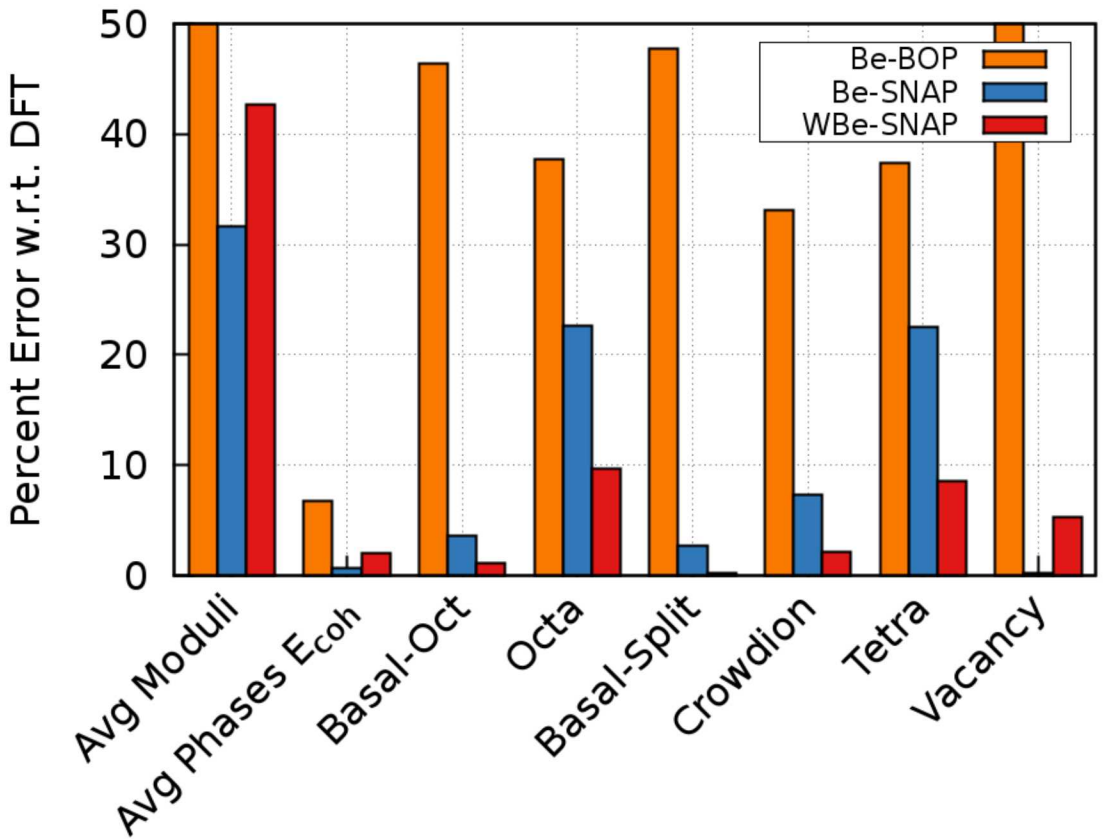


		Formation Energy (eV/f.u.)		
Phase	Composition	DFT	BOP	SNAP
B2	WBe	0.67	-2.20	0.30
C14	WBe_2	-0.87	-4.20	-1.27
C15	WBe_2	-0.92	-4.19	-1.15
C36	WBe_2	-0.90	-4.20	-1.22
L12	WBe_3	-0.51	-4.58	-0.15
D2b	WBe_{12}	-0.96	-6.69	0.34

*~60% of total Be is not captured in the W matrix

Description	N_E	N_F	σ_E	σ_F	Description	N_E	N_F	σ_E	σ_F	Description	N_E	N_F	σ_E	σ_F
W:					Be:					W-Be:				
Self-Interstitials	1 5	5805	$5 \cdot 10^{-2}$	$8 \cdot 10^{2}$	Surfaces	90	17280	$1 \cdot 10^{3}$	$4 \cdot 10^{5}$	AIMD	3360	497124	$7 \cdot 10^{4}$	$6 \cdot 10^{2}$
Dislocations	98	39690	$3 \cdot 10^{0}$	$9 \cdot 10^{4}$	Self-Interstitials	179	137931	$3 \cdot 10^{2}$	$4 \cdot 10^{5}$	Elastic Deform	3946	68040	$\mathbf{3 \cdot 10^{5}}$	$2 \cdot 10^{3}$
Liquids	27	3120	$4 \cdot 10^{-3}$	$3 \cdot 10^{2}$	AIMD	909	130896	$\mathbf{2 \cdot 10^{5}}$	$2 \cdot 10^{6}$	Equation of State	1113	39627	$2 \cdot 10^{5}$	$4 \cdot 10^{4}$
Divacancy	39	6084	$1 \cdot 10^{0}$	$1 \cdot 10^{3}$	Elastic Deform	4594	43260	$1 \cdot 10^{5}$	$\mathbf{1 \cdot 10^{7}}$	Surface Adhesion	381	112527	$2 \cdot 10^{4}$	$\mathbf{9 \cdot 10^{4}}$
Equation of State	125	3468	$1 \cdot 10^{-1}$	$6 \cdot 10^{4}$	Equation of State	502	5418	$6 \cdot 10^{4}$	$3 \cdot 10^{6}$					
Γ-Surface	6183	328338	$1 \cdot 10^{0}$	$\mathbf{1 \cdot 10^{6}}$	Stacking Faults	6	864	$3 \cdot 10^{0}$	$2 \cdot 10^{6}$					
Γ-Surf.+Vacancy	750	105750	$4 \cdot 10^{-1}$	$\mathbf{3 \cdot 10^{6}}$	Liquids	75	57600	$7 \cdot 10^{1}$	$7 \cdot 10^{5}$					
AIMD	60	23040	$3 \cdot 10^{0}$	$1 \cdot 10^{4}$										
Elastic Deform	2000	6000	$5 \cdot 10^{0}$	$6 \cdot 10^{4}$										
Surfaces	180	334818	$\mathbf{1 \cdot 10^{5}}$	$3 \cdot 10^{5}$										
Monovacancy	420	183054	$2 \cdot 10^{3}$	$1 \cdot 10^{5}$										
Total	9897	1039167				6355	393249				8800	717318		


Percent Error w.r.t. DFT
20
15
10
5
0
W-EAM
W-SNAP
WBe-SNAP
Elast. Const.
Octa
Tetra
110d
111d
Vacancy
Divacancy


Percent Error w.r.t. DFT
50
40
30
20
10
0
Be-BOP
Be-SNAP
WBe-SNAP
Avg Moduli
Avg Phases Ecoh
Basal-Oct
Octa
Basal-Split
Crowdion
Tetra
Vacancy

Fully-Automated Generation of SNAP

Sandia National Laboratories

Par-Splice

ECP EXASCALE COMPUTING PROJECT

- Simulation hyper parameters (T,P, …)
- Resource management

Trajectory Farm

VASP

- Generates new training configurations, returns SNAP potentials

DAKOTA

FitSNAP.py

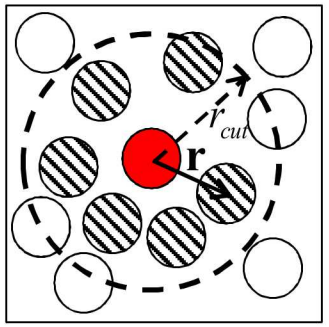
- SNAP fit hyper parameters (i.e. r_{cut})
- Min regression / prediction errors
- Genetic Algorithm

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

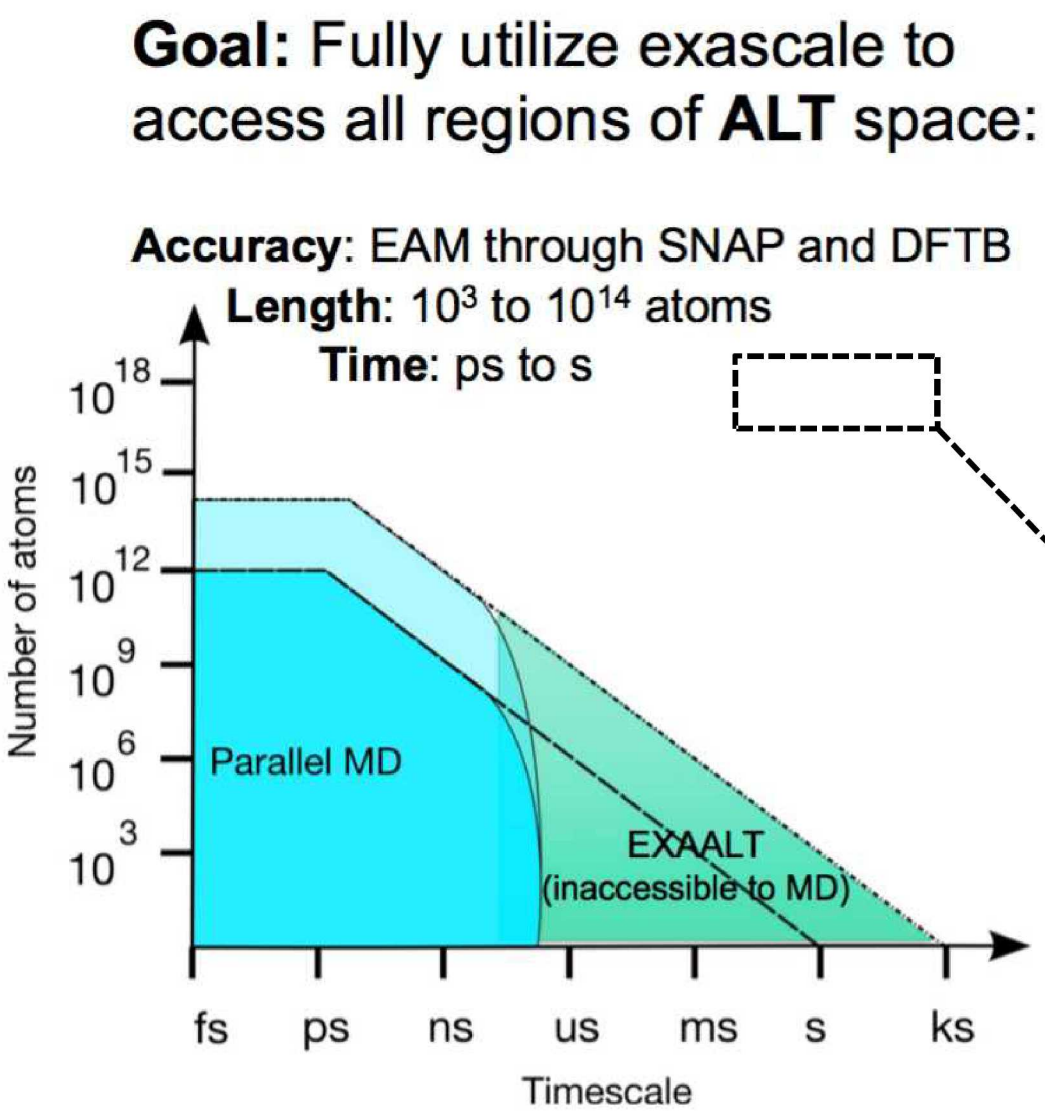


$$u^j_{m,m'} = U^j_{m,m'}(0,0,0) + \sum_{r_{ii'}<R_{cut}} f_c(r_{ii'}) w_i U^j_{m,m'}(\theta_0, \theta, \phi)$$

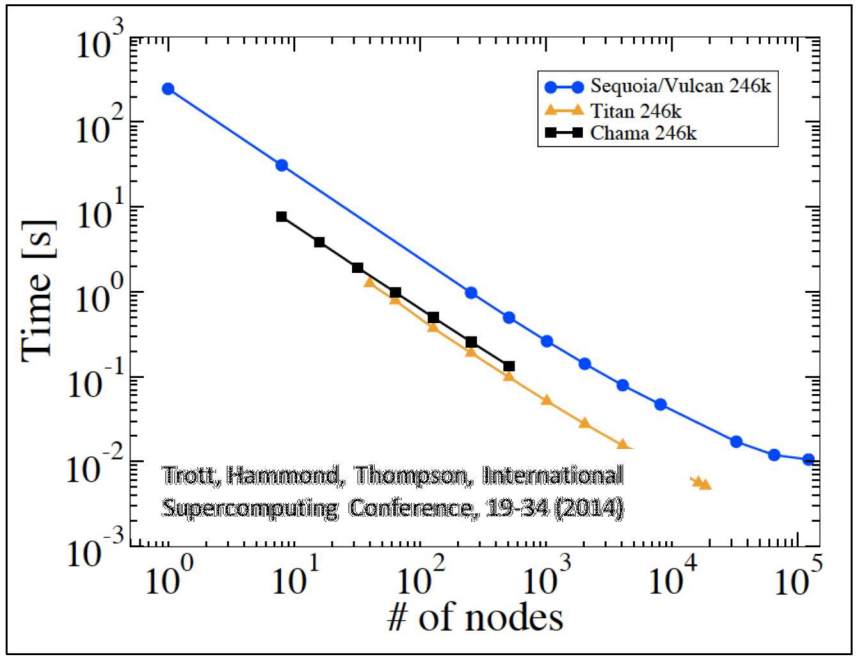
$$B_{j_1,j_2,j} = \sum_{m_1,m_1'=-j_1}^{j_1} \sum_{m_2,m_2'=-j_2}^{j_2} \sum_{m,m'=-j}^{j} (u^j_{m,m'})^* H^{jmm'}_{j_1 m_1 m_1' \; j_2 m_2 m_2'} \, u^{j_1}_{m_1,m_1'} u^{j_2}_{m_2,m_2'}$$

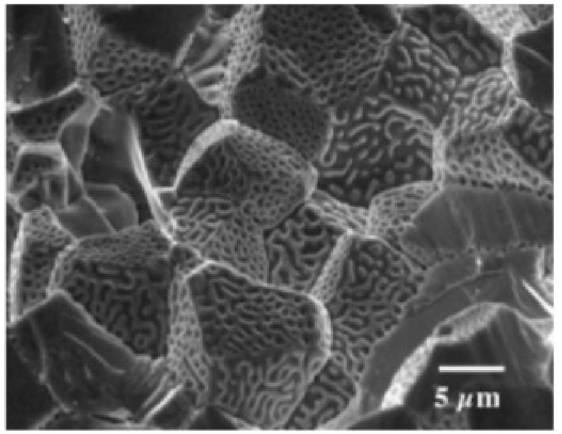
Symmetry relation: $\dfrac{B_{j_1,j_2,j}}{2j+1} = \dfrac{B_{j,j_2,j_1}}{2j_1+1} = \dfrac{B_{j_1,j,j_2}}{2j_2+1}$

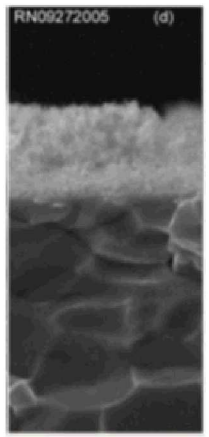
Design Philosophy for Multiscale, SNAP



- Extensions of parallel MD through improvement of the accuracy of MD potentials







- Bonus points if the MD potential scales well on leadership computing platforms

Training SNAP for Transferability – WBe

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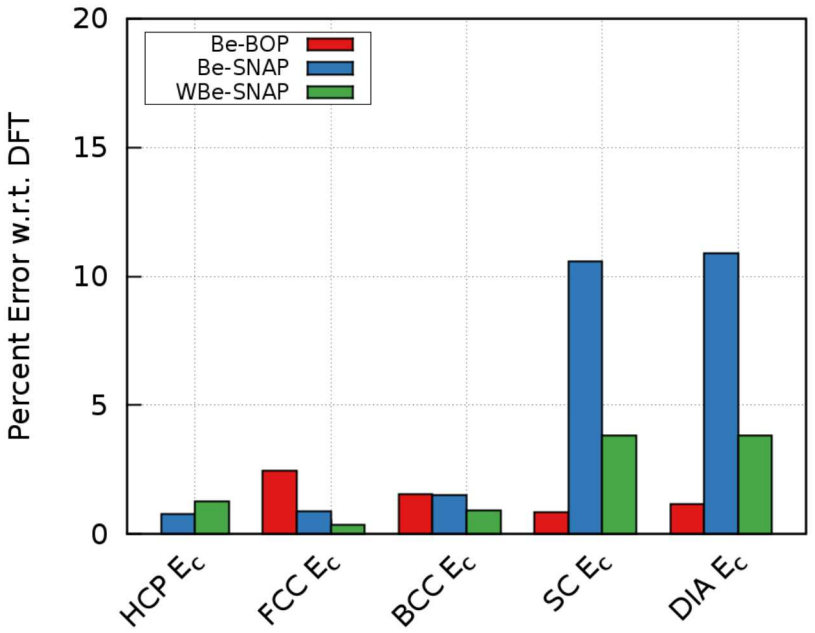
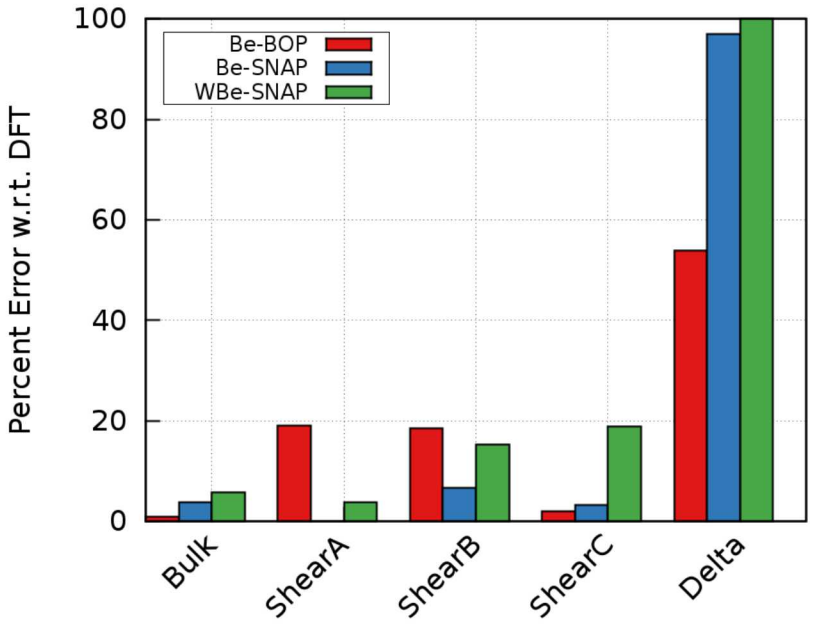
Candidate 18584
Predicts the correct WBe intermetallic phases
(stable Laves phases, unstable B2 and L12)

Key drawbacks are Be-elastic and W-vacancy properties.

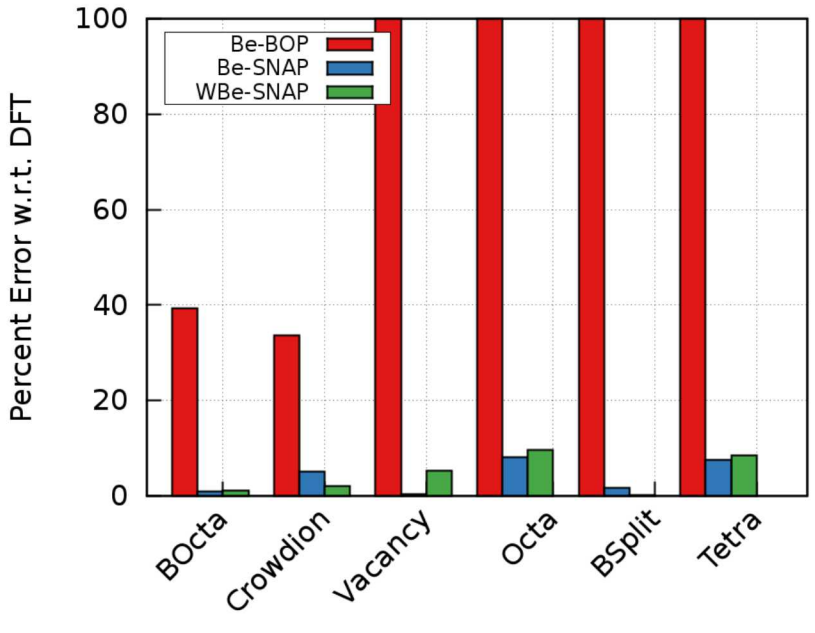
Be Phase Stability



Be Elastic Moduli



Be Defect Formation



Training SNAP for Transferability – Tungsten

Sandia National Laboratories

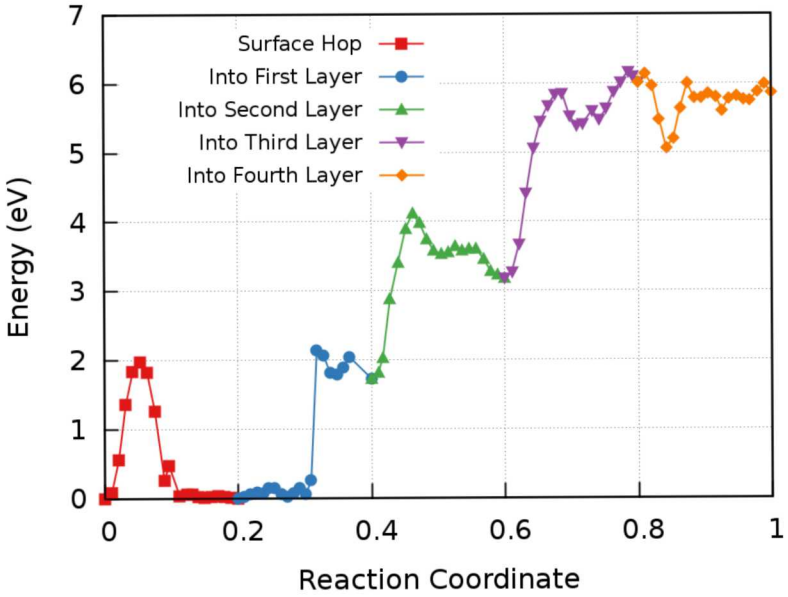
	DFT	EAM	SNAP
a_0	3.1803	3.165	**3.1797**
C_{11}	517	517	**523**
C_{12}	198	200	**193**
C_{44}	142	156	**151**

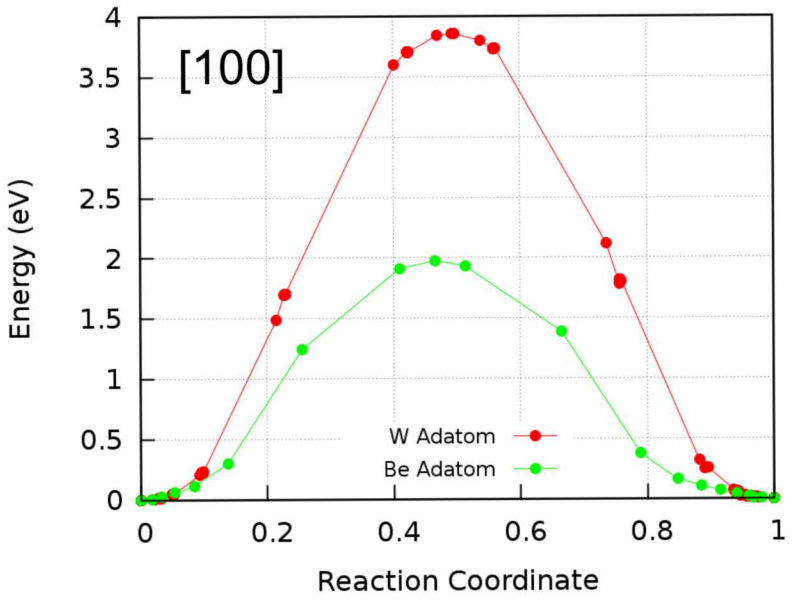
	DFT	EAM	SNAP
He-Tet	6.2	6.2	**6.2**
He-Oct	6.4	6.3	**6.5**
He-Sub	4.7	4.7	**4.2**
2HeTet	1.0	0.9	**0.7**

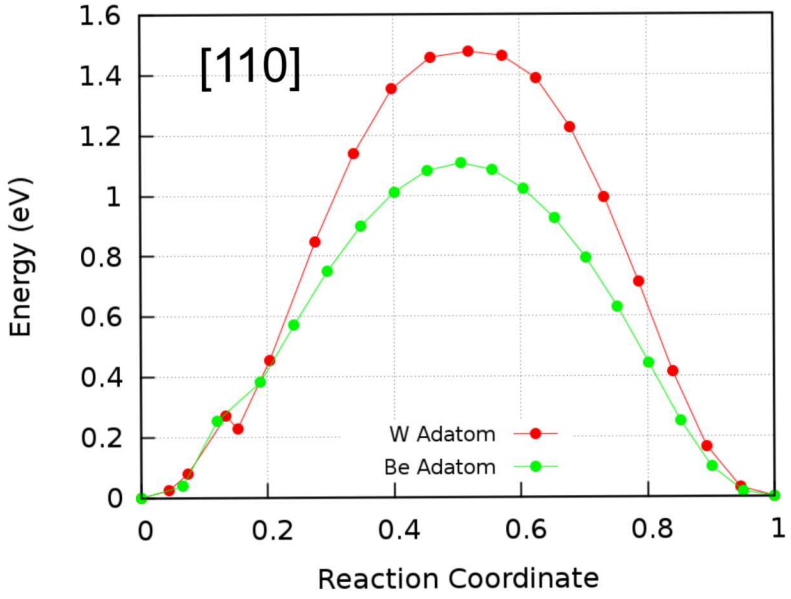
- W-He and He-He terms are using JW tables

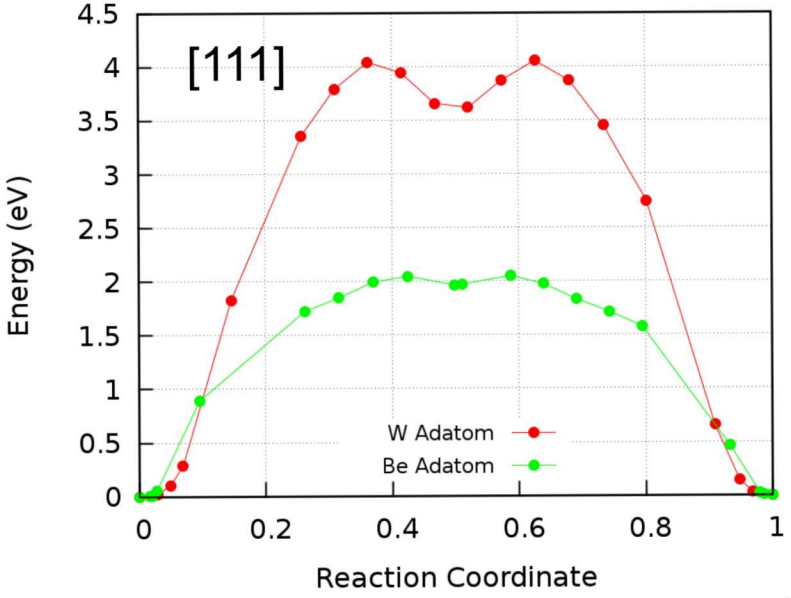
	$\Delta E_{fcc\text{-}bcc}$	$\Delta E_{hcp\text{-}bcc}$	$\Delta E_{sc\text{-}bcc}$	$\Delta E_{A15\text{-}bcc}$	$\Delta E_{dia\text{-}bcc}$
EAM	0.155	0.155	1.37	0.216	3.38
SNAP	0.478	0.461	1.86	0.103	3.97

	DFT (eV)	EAM (eV)	EAM (%)	SNAP (eV)	SNAP (%)
Tetra	11.1	10.24	**7.34**	9.70	**12.22**
Octa	11.7	10.35	**11.57**	11.03	**5.77**
[110]d	9.8	10.06	**2.26**	9.70	**1.43**
[111]d	9.6	9.44	**1.18**	9.56	**0.13**
Vac	3.3	3.63	**11.05**	3.07	**6.14**
Divac	0.1	-0.432	**460.2**	0.098	**18.21**


Surface Hop
Into First Layer
Into Second Layer
Into Third Layer
Into Fourth Layer
Energy (eV)
Reaction Coordinate


[100]
W Adatom
Be Adatom
Energy (eV)
Reaction Coordinate


[110]
W Adatom
Be Adatom
Energy (eV)
Reaction Coordinate


[111]
W Adatom
Be Adatom
Energy (eV)
Reaction Coordinate