Exceptional service in the national interest

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SNAP: Spectral neighbor analysis method for automated generation of quantum-accurate interatomic potentials for LAMMPS

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August 2015 LAMMPS Users' Workshop and Symposium

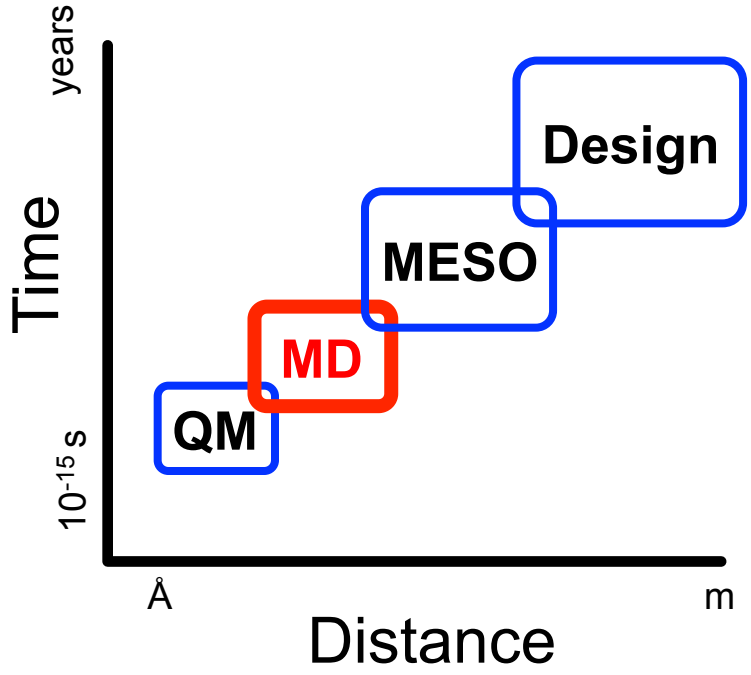
SAND 2014-1297C

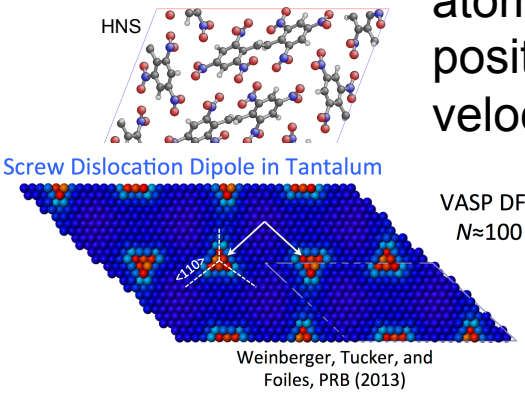
U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

Why Use 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

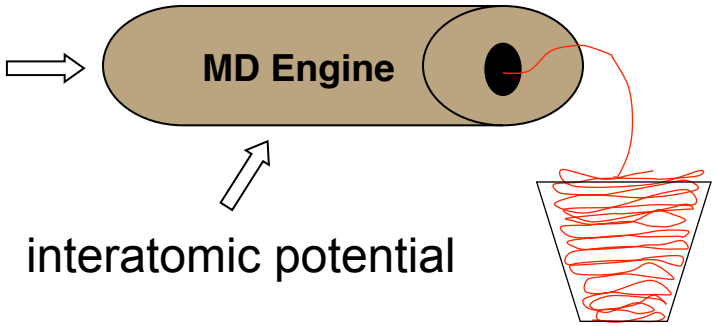




VASP DFT
$N \approx 100$

Weinberger, Tucker, and Foiles, PRB (2013)

atoms, positions, velocities



Positions, velocities and forces at many later times

Example: Plasticity in BCC Metals

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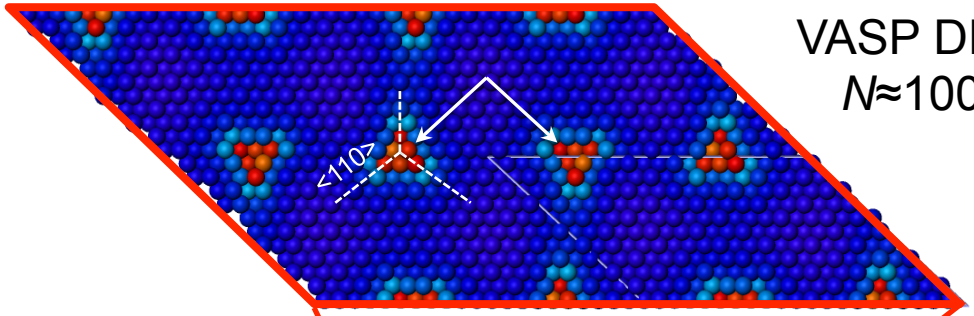
Screw Dislocation Motion in BCC Tantalum



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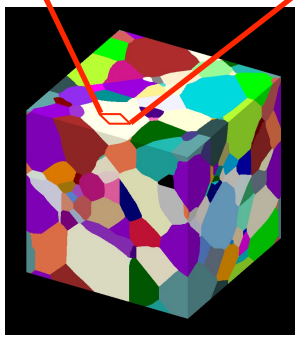
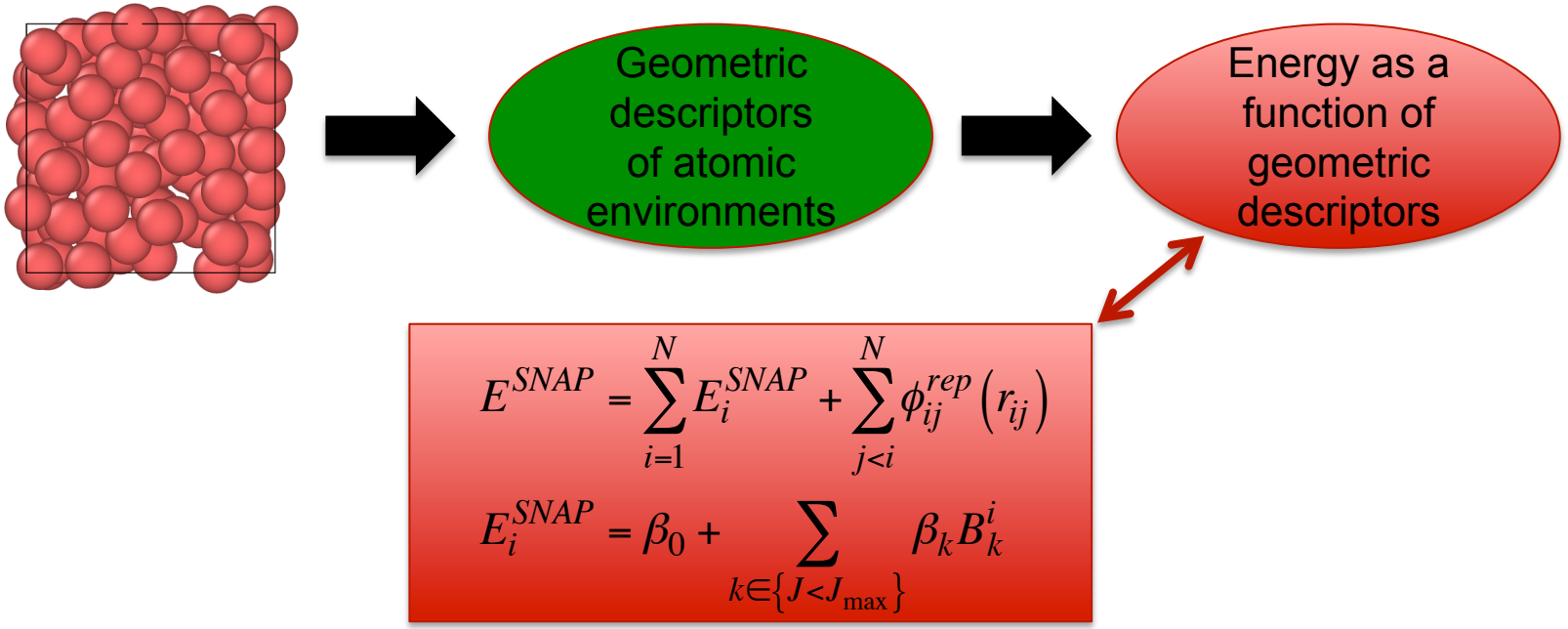
Weinberger, Tucker, and Foiles, PRB (2013)

Polycrystalline Tantalum Sample



LAMMPS MD
$N \approx 10^8$

SNAP: Spectral Neighbor Analysis Potentials



- **GAP (Gaussian Approximation Potential):** Bartok, Csanyi et al., *Phys. Rev. Lett, 2010.* Uses 3D neighbor density bispectrum and **Gaussian process regression**.
- **SNAP (Spectral Neighbor Analysis Potential):** Our SNAP approach uses GAP's neighbor bispectrum, but replaces Gaussian process with **linear regression**.
  - More robust
  - Lower computational cost
  - Decouples MD speed from training set size
  - Enables large training data sets, more bispectrum coefficients
  - Straightforward sensitivity analysis

Bispectrum Components as Descriptor

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

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

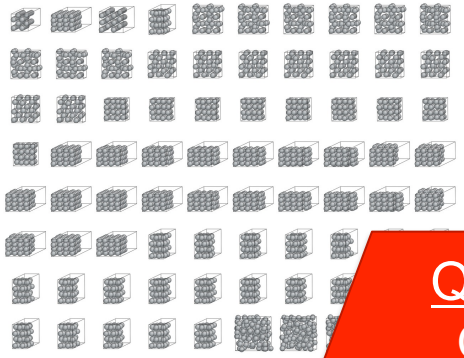
- Expand density around each atom in a basis of ***4D hyperspherical harmonics***,
- Bispectrum components of the 4D hyperspherical harmonic expansion are used as the geometric descriptors of the local environment
  - Preserves universal physical symmetries
  - Rotation, translation, permutation
  -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

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

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

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

SNAP Fitting Process
FitSnap.py



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

QUEST
QDFT
Training
Data

Dakota
optimization,
sensitivity

fitsnap.py
Communicate with LAMMPS; weighted regression to obtain SNAP coefficients

LAMMPS

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

Bispectrum components & derivatives, reference potential

Ta SNAP potential was fit to a DFT-based training set containing 'usual suspects'

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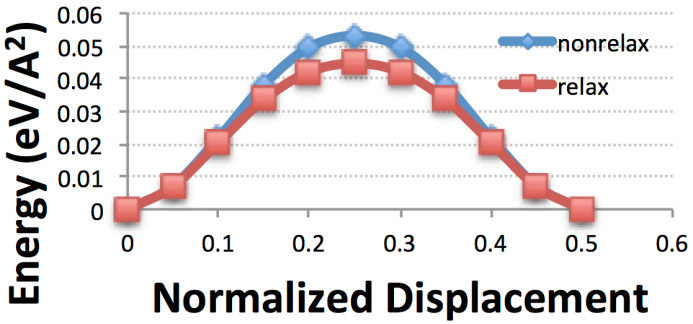
For each configuration in training set, fit total energy, atomic forces, stress

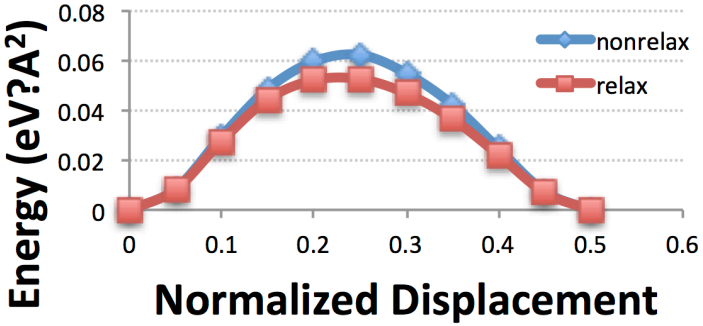
- Equilibrium lattice parameter
- Elastic constants (C_{11}, C_{12}, and C_{44}) and bulk modulus (B)
- Free surface energies: (100), (110), (111), and (112)
- Generalized planar stacking fault curves: {112} and {110}
- Energy-Volume (Contraction and Dilation) - BCC, FCC, HCP, and A15
- Lattices with random atomic displacements
- Liquid structure

Example: DFT-based Generalized Stacking Fault Energies

(110)

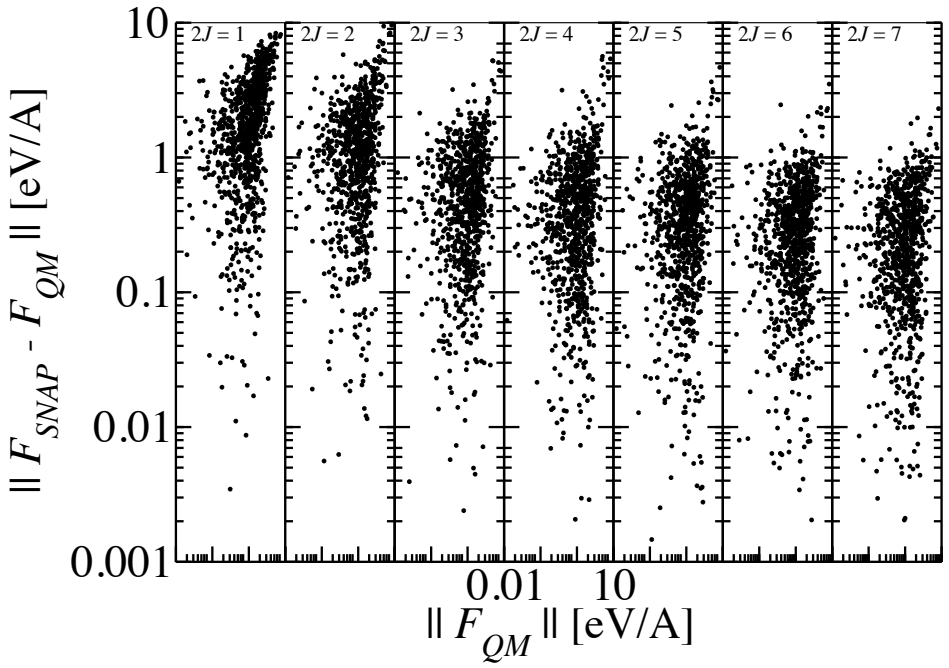


(112)



Effect of Higher-order Bispectrum Components

- Liquid force errors decrease with increasing J
- Diminishing returns beyond J = 7/2



2*J*	N	Ferr
1	2	2.09
2	5	1.39
3	8	0.66
4	14	0.53
5	20	0.44
6	30	0.35
7	40	0.30

SNAP potential yields good agreement with DFT results for some standard properties

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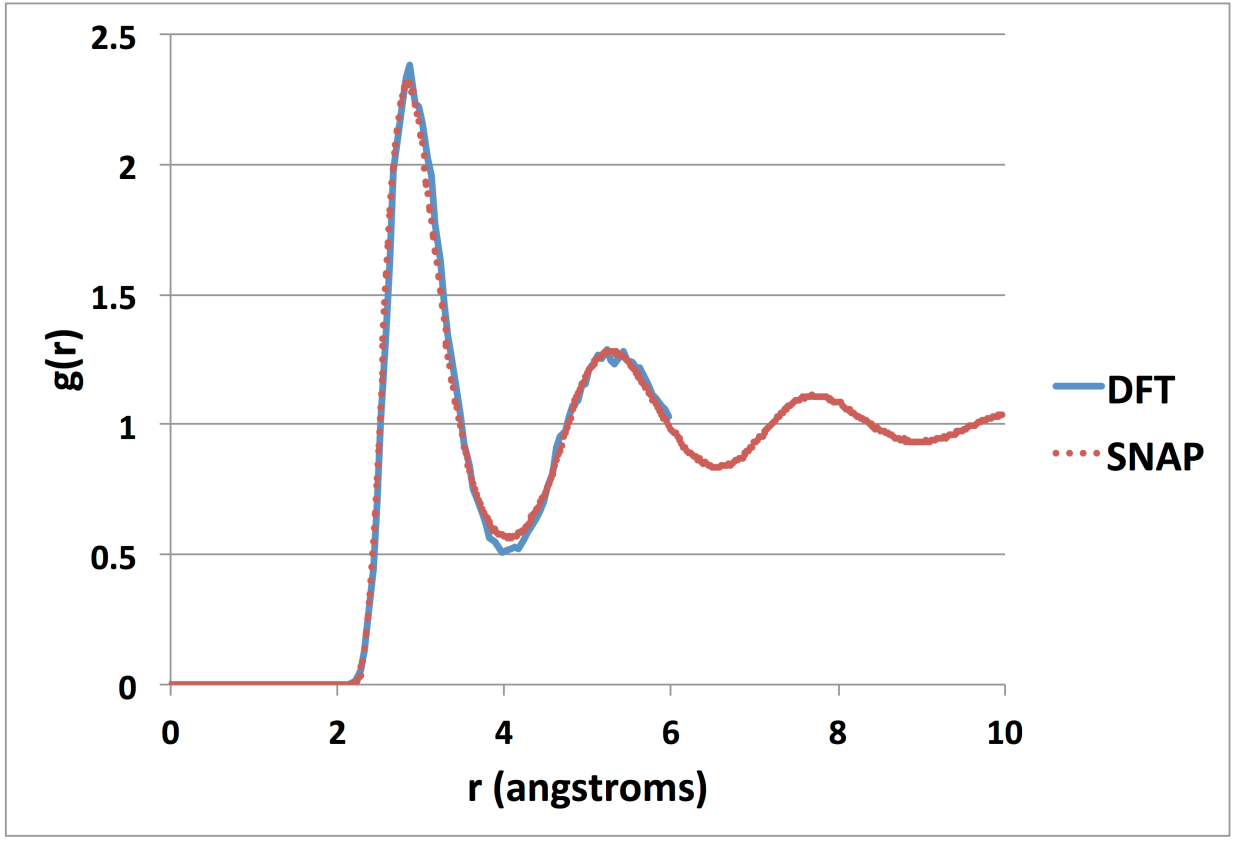
	DFT	SNAP	Zhou (EAM)	ADP
Lattice Constant (Å)	3.320	3.316	3.303	3.305
B (Mbar)	1.954	1.908	1.928	1.971
C' = (1/2)(C_{11} – C_{12}) (Mbar)	50.7	59.6	53.3	51.0
C_{44} (Mbar)	75.3	73.4	81.4	84.6
Vacancy Formation Energy (eV)	2.89	2.74	2.97	2.92
(100) Surface Energy (J/m^2)	2.40	2.68	2.34	2.24
(110) Surface Energy (J/m^2)	2.25	2.34	1.98	2.13
(111) Surface Energy (J/m^2)	2.58	2.66	2.56	2.57
(112) Surface Energy (J/m^2)	2.49	2.60	2.36	2.46
(110) Relaxed Unstable SFE (J/m^2)	0.72	1.14	0.75	0.58
(112) Relaxed Unstable SFE (J/m^2)	0.84	1.25	0.87	0.74

Liquid structure: SNAP and DFT are in excellent agreement



Liquid pair correlation function, g(r) computed at 3250 K (~melting point) and experimental density

- DFT: 100 atoms, 2 picoseconds
- SNAP: 1024 atoms, 200 picoseconds

SNAP potentials predict correct Peierls barrier for Ta screw dislocations

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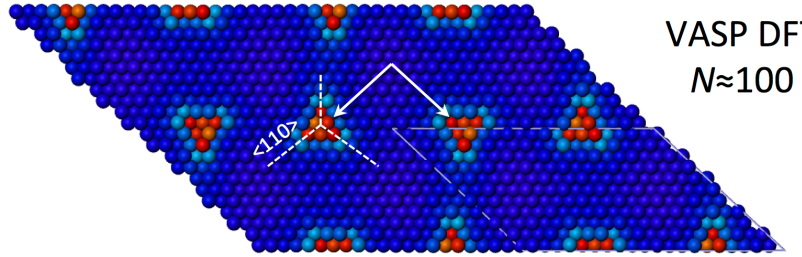
- Peierls barrier is the activation energy to move a screw dislocation
- Many simple interatomic potentials incorrectly predict a metastable state
  - Leads to erroneous dynamics
- SNAP potential agrees well with DFT calculations
  - Future work will explore dislocation dynamics based on this potential

Thompson et al. arxiv.org/abs/1409.3880
J. Comp. Phys. (2015)

Screw Dislocation Dipole in Tantalum



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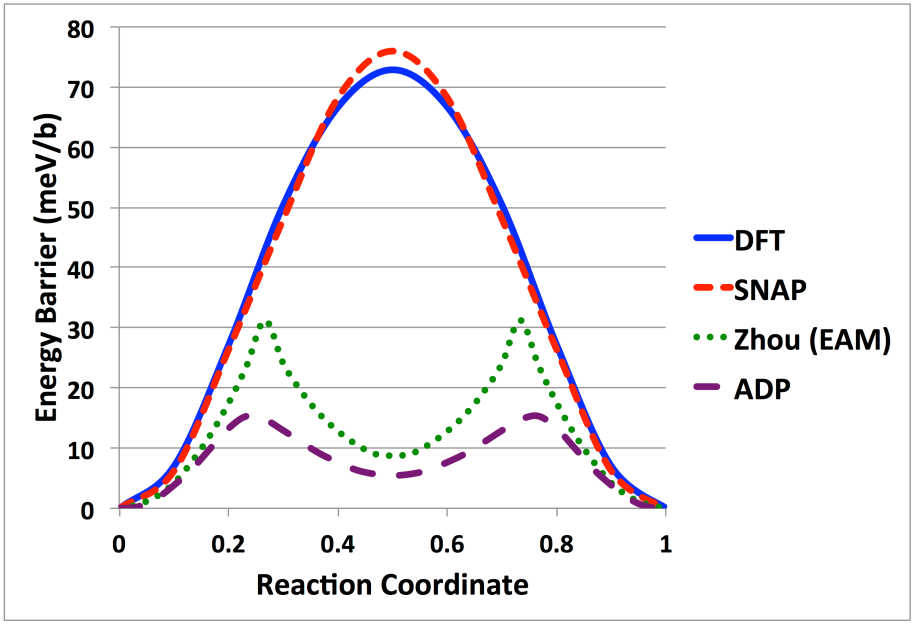
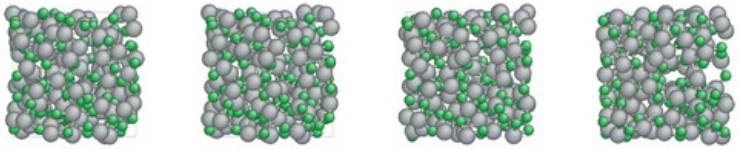
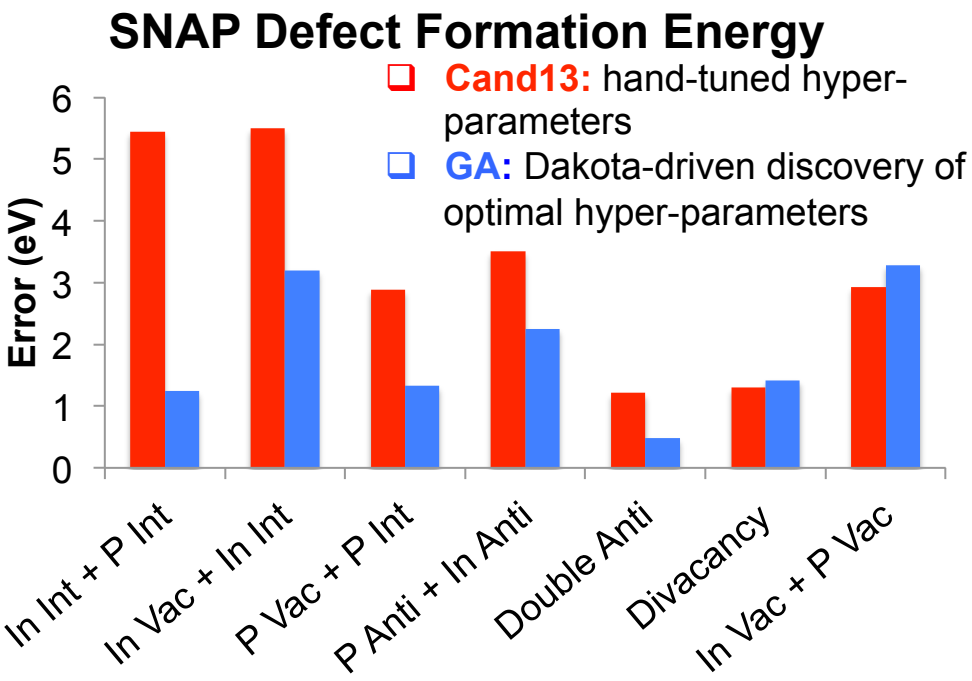


SNAP Indium Phosphide





Additional Challenges

- Two elements
- Different atom sizes
- Diverse structures
- Defect formation energies
- Sensitive to curvature

Innovations

- Differentiate elements by: density weight, linear coefficients, neighbor cutoff
- Trained against relaxed defect structures
- Trained against deformed defect structures

Result (so far)

- Good overall fit
- Defect energy error > 1 eV

SNAP Silica: Promising Start

(Stan Moore, Paul Crozier, Peter Schultz)

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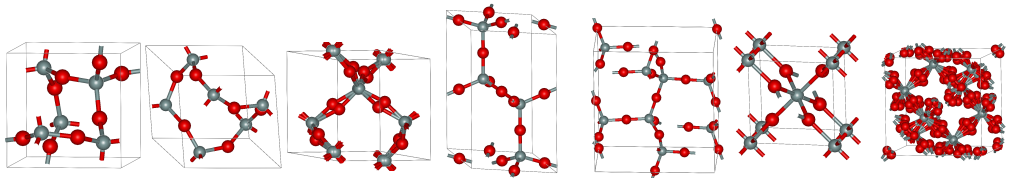
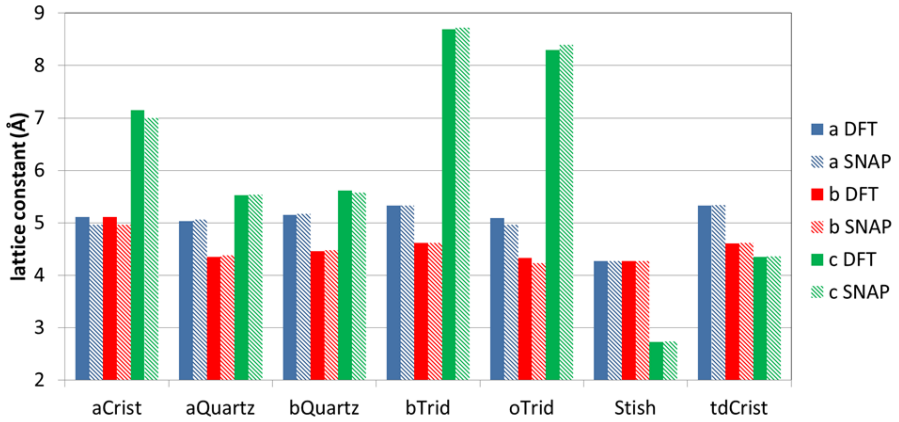
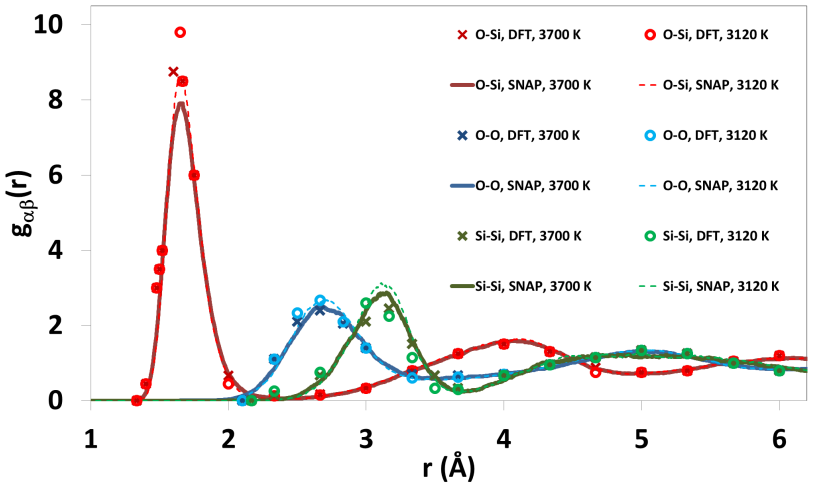
Less than 3% error in predicted lattice parameters of 7 crystal polymorphs





Good agreement with QM liquid structure for SiO2



Additional Challenges

- Electrostatics
- Started with no training data
- Goal: quantum-accurate prediction of Si/SiO2 interface

Innovations

- Generated training data adaptively, on-the-fly
- Added fixed point charges, long-range electrostatics

Result (so far)

- Good agreement with QM for SiO2 crystal polymorphs
- Good agreement with QM liquid structure for SiO2

Conclusions

- SNAP is a new formulation for interatomic potentials
  - Geometry described by bispectrum components
  - Energy is a linear regression of bispectrum components
- Works well for Ta
  - Liquid structure
  - Peierls barrier for screw dislocation motion
- Ongoing work
  - Extension to binary systems: InP, SiO2, TaO_x
- SNAP Ta potential published
  - arxiv.org/abs/1409.3880
  - J. Comp. Phys. (2015)
- SNAP Ta available in LAMMPS

Primary Collaborators
Laura Swiler
Stephen Foiles
Garritt Tucker

Additional Collaborators
Christian Trott
Peter Schultz
Paul Crozier
Stan Moore
Adam Stephens

FitSnap.py: Robust Software Framework

Key advantages of fitsnap.py

- Minimal file I/O
- Use of NumPy/SciPy
- Caching and reuse of data
- File-based input
- Supports parallel LAMMPS