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Atomistic Materials Simulation Using Quantum-Accurate Interatomic Potentials

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IPAM, February 25, 2015, SAND 2014-1297C

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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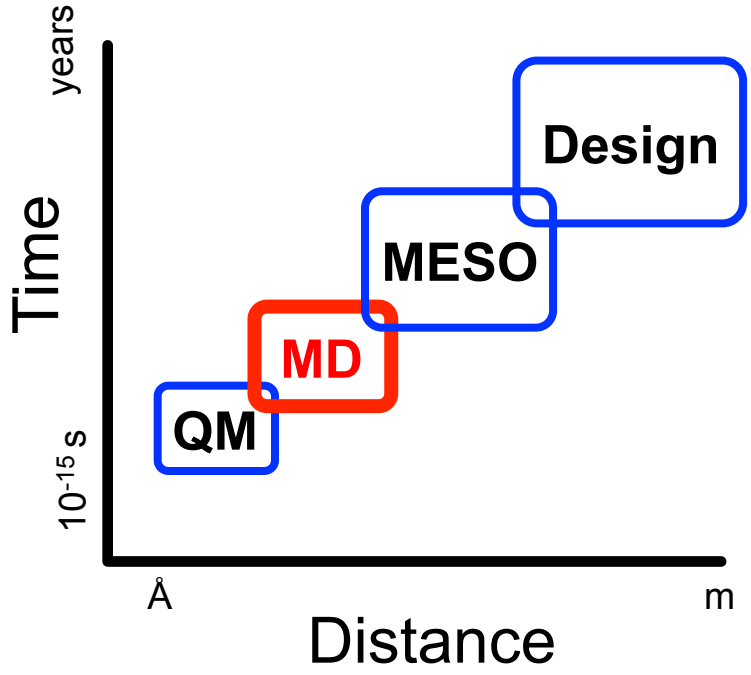
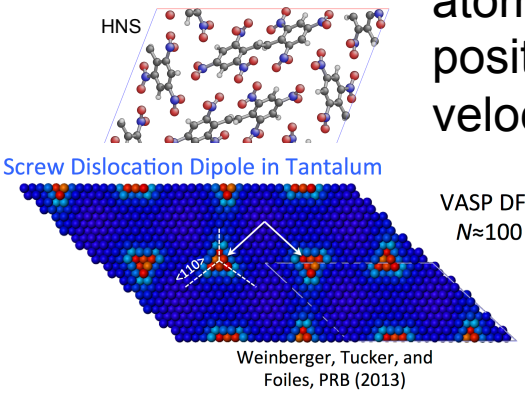
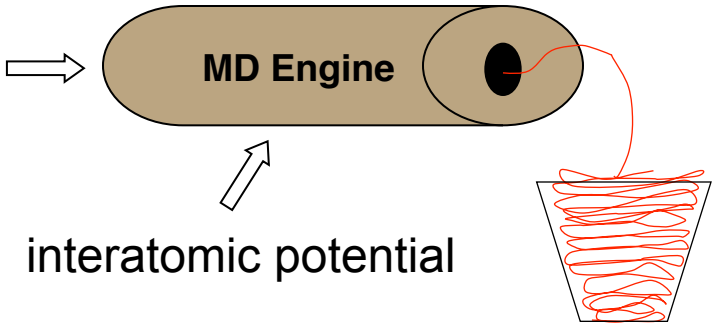
What is Molecular Dynamics Simulation?

- 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



Positions, velocities and forces at many later times

Example: Plasticity in BCC Metals

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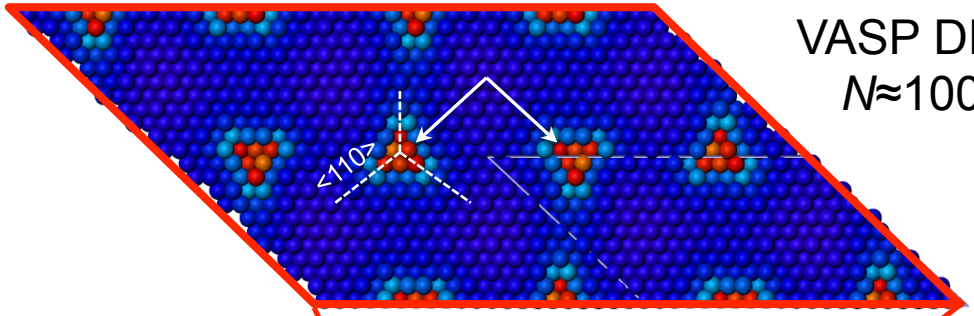
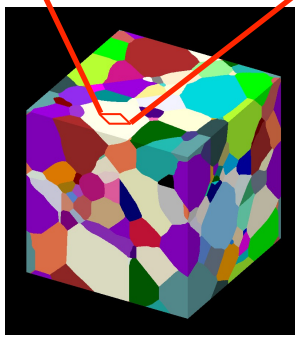
Screw Dislocation Motion in BCC Tantalum



VASP DFT
$N \approx 100$

Weinberger, Tucker, and Foiles, PRB (2013)

Polycrystalline Tantalum Sample



LAMMPS MD
$N \approx 10^8$

What is LAMMPS?

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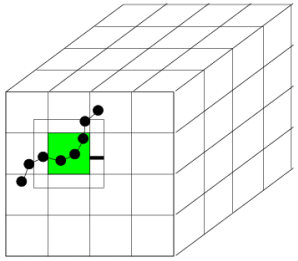
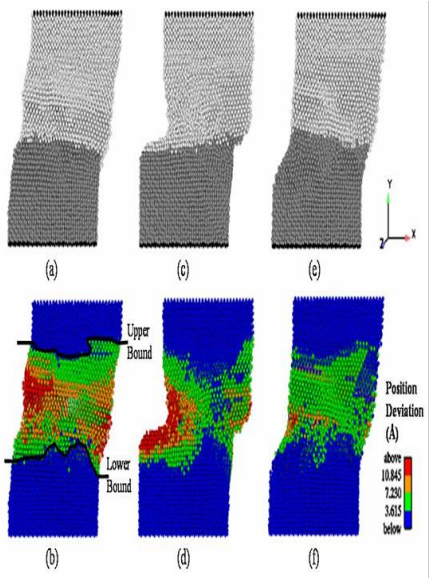
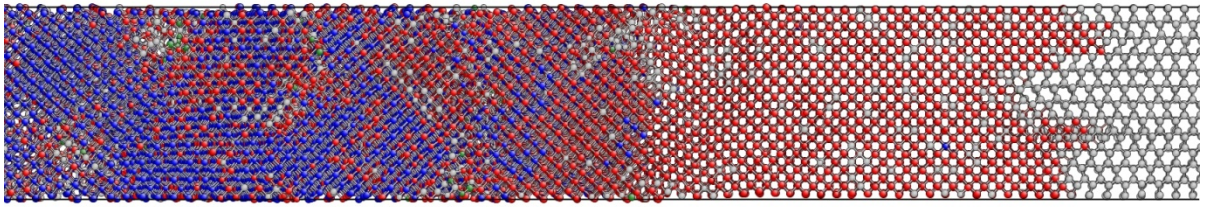
(Large-scale Atomic/Molecular Massively Parallel Simulator)

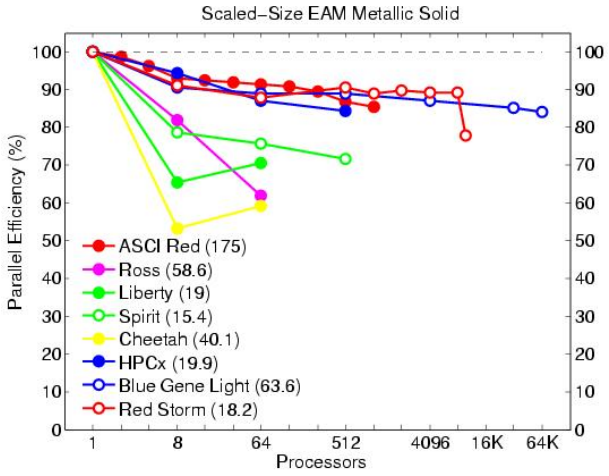
http://lammps.sandia.gov

- Classical MD code.
- Open source, highly portable C++.
- Freely available for download under GPL.
- Easy to download, install, and run.
- Well documented.
- Easy to modify or extend with new features and functionality.
- Active user's e-mail list with over 650 subscribers.
- More that 1000 citations/year
- Users' workshops: 2010, 2011, 2013, 2015
- Spatial-decomposition of simulation domain for parallelism.
- Energy minimization via conjugate-gradient relaxation.
- Atomistic, mesoscale, and coarse-grain simulations.
- Variety of potentials (including many-body and coarse-grain).
- Variety of boundary conditions, constraints, etc.









MD is Big Consumer of Computer Time

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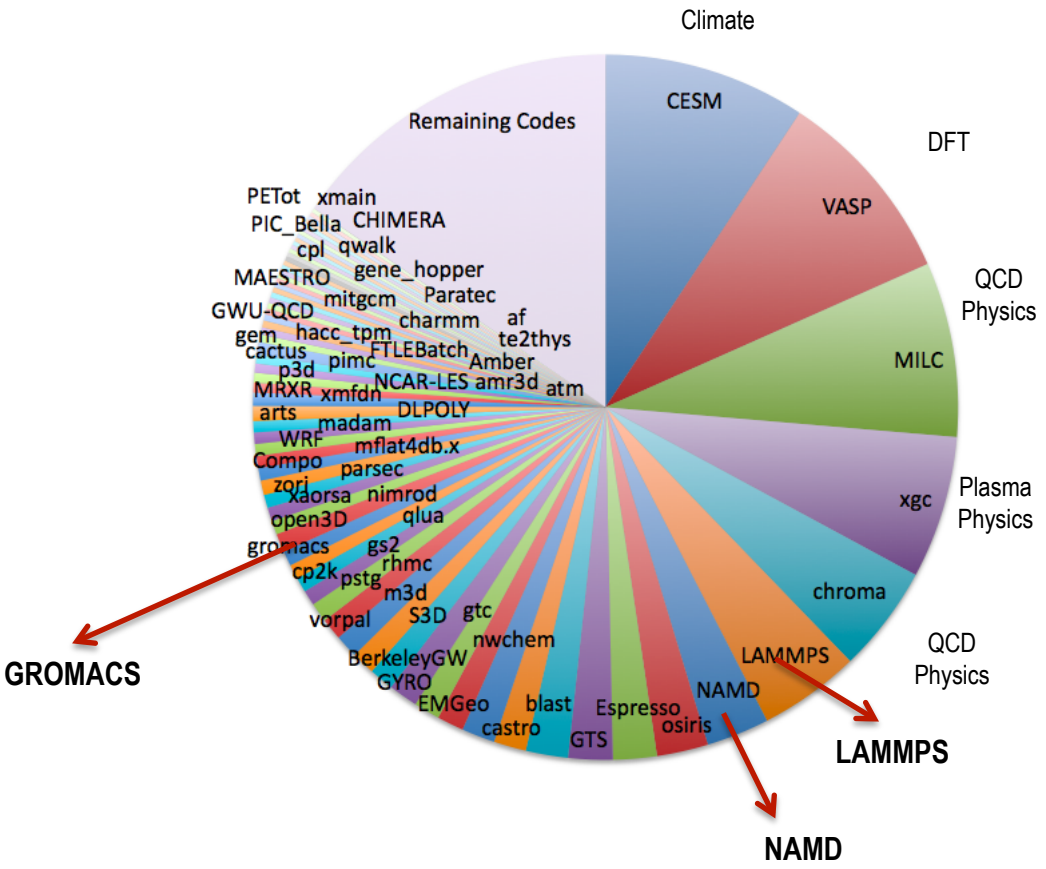

2012 Top Application Codes at NERSC

Exponential Growth in Use of MD

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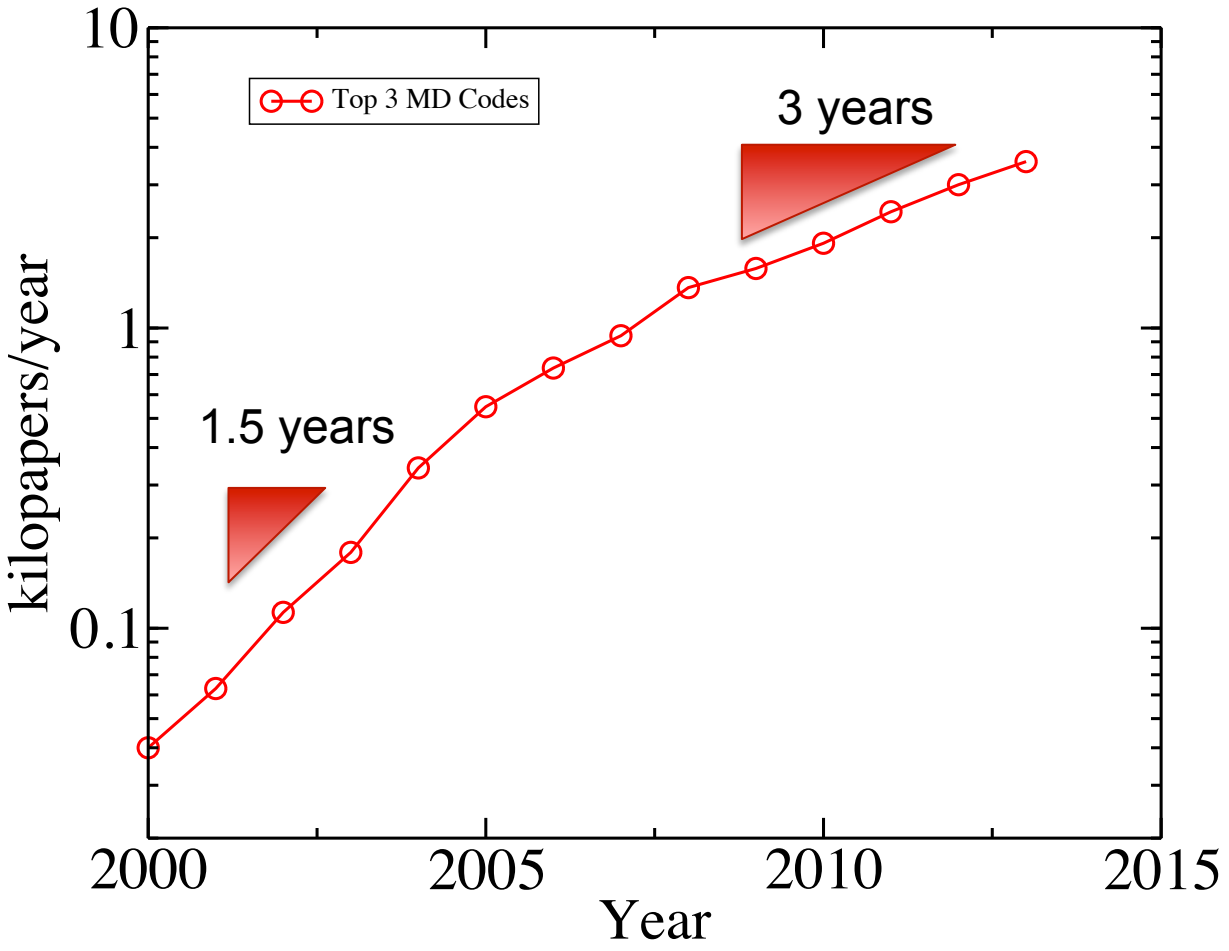
Papers citing LAMMPS, NAMD, GROMACS
Measured using Web of Science citation reports



Historical Development for Potentials

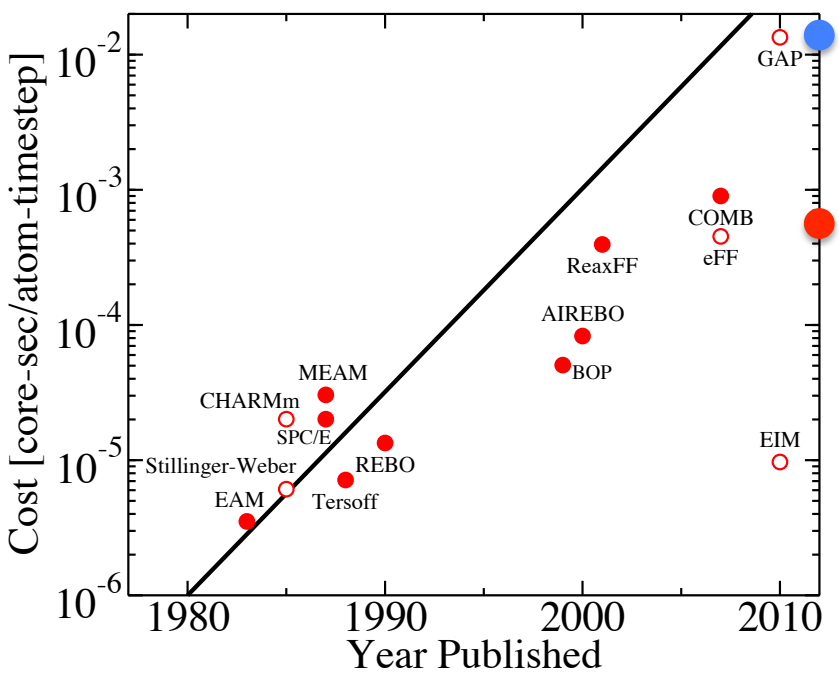
Twobody (B.C.)
Lennard-Jones
Hard Sphere
Coulomb
Bonded

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

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

Machine Learning (2010s)
GAP, SNAP, NN,…

Moore's Law for Interatomic Potentials
Plimpton and Thompson, MRS Bulletin (2012).



Two philosophical extremes in the development of interatomic potential models

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The Force!

- Functional forms based on fundamental understanding of electronic origins of bonding
  - Bond Order Potentials (BOP)
  - Model Generalized Pseudopotential Theory (MGPT)
  - COMB
  - ReaxFF
  - …
- Gives confidence that it will interpolate/extrapolate reasonably

Luke: ***Is the dark side stronger?***
Yoda: ***No, no no. Quicker, easier, more seductive.***

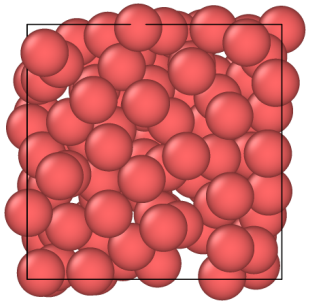
The Dark Side!

- Empirical fit of a flexible functional form
  - Gaussian Approximation Potentials (GAP)
  - Spectral Neighbor Analysis Potential (SNAP) - this work
  - …
- Replaces the need for intuition/art with extensive computation
  - Automate the fitting process?
  - Apply across multiple materials classes?

Darth Vader: ***You underestimate the power of the dark side!***

Borrowed from **Stephen Foiles**

SNAP: Spectral Neighbor Analysis Potentials



Geometric descriptors of atomic environments

Energy as a function of geometric descriptors

$$E^{SNAP} = \sum_{i=1}^{N} E_i^{SNAP} + \sum_{j<i}^{N} \phi_{ij}^{rep}\left(r_{ij}\right)$$

$$E_i^{SNAP} = \beta_0 + \sum_{k \in \{J<J_{\max}\}} \beta_k B_k^i$$

- **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

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

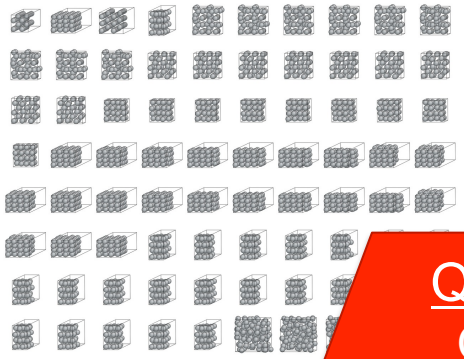
- Expand density around each atom in a basis of ***4D hyperspherical harmonics***,
- Bispectrum components of the 4D hyperspherical harmonic expansion are used as the geometric descriptors of the local environment
  - Preserves universal physical symmetries
  - Rotation, translation, permutation
  -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

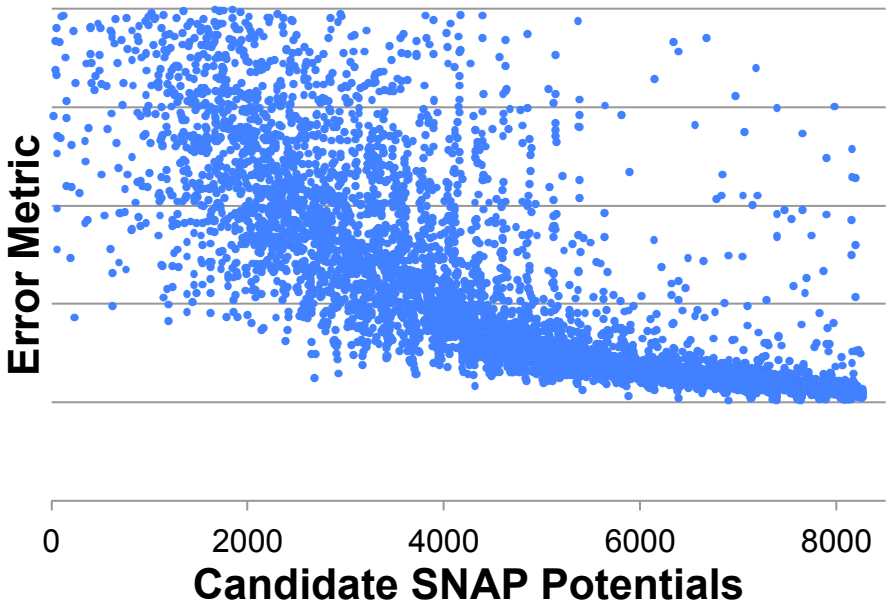
Metrics
- Force residuals
- Energy residuals
- Elastic constants
- Etc.

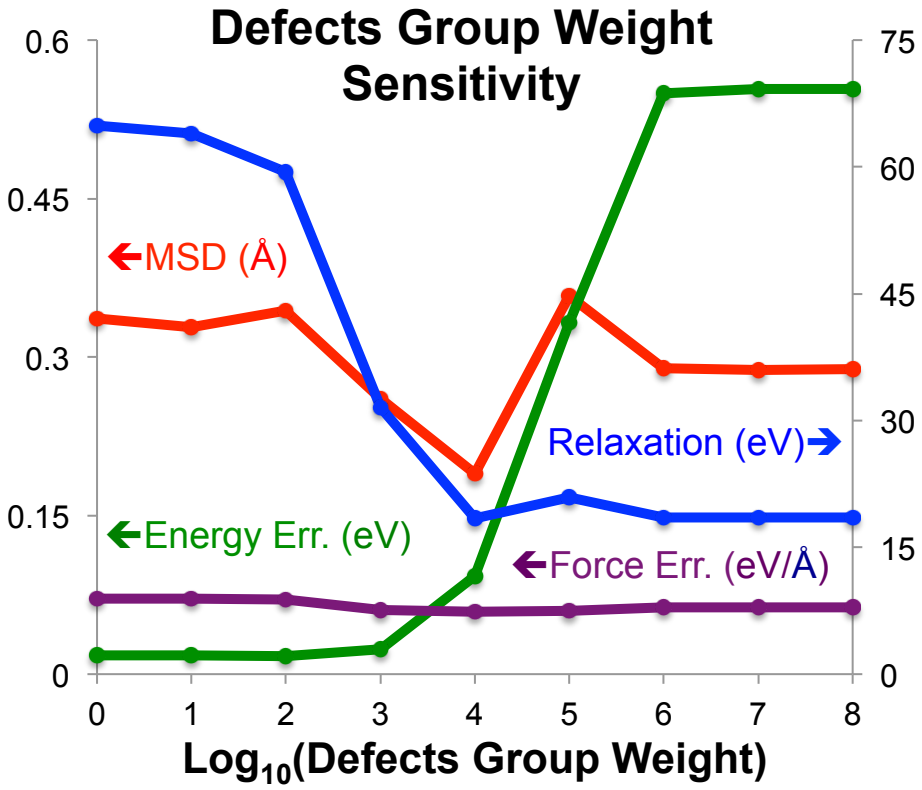
Bispectrum components & derivatives, reference potential

FitSnap.py: Robust Software Framework

- Fitting previously based on a fragile collection of shell scripts, Python scripts, and C++ code.
- All scripts and codes were brought under version control and systematically combined and re-written into a unified tool: fitsnap.py.

Hyper-parameter Optimization





Key advantages of fitsnap.py

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

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

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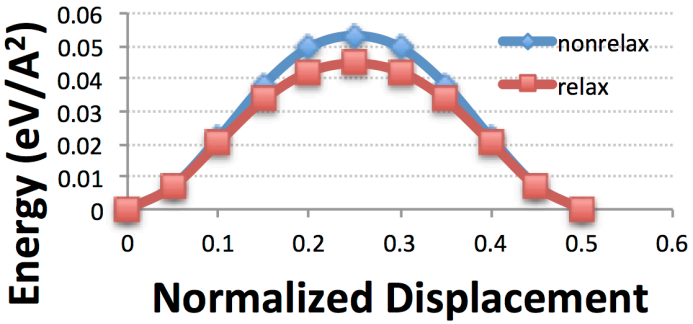
For each configuration in training set, fit total energy, atomic forces, stress

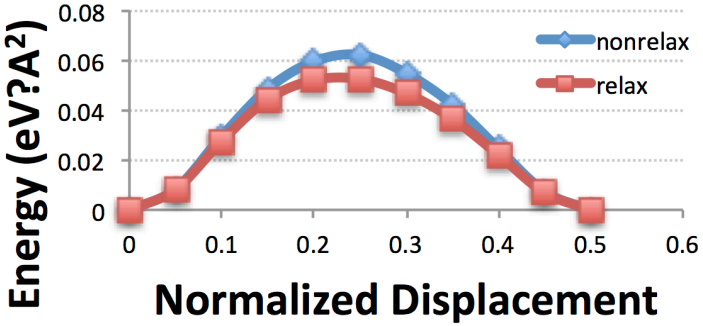
- Equilibrium lattice parameter
- Elastic constants (C_{11}, C_{12}, and C_{44}) and bulk modulus (B)
- Free surface energies: (100), (110), (111), and (112)
- Generalized planar stacking fault curves: {112} and {110}
- Energy-Volume (Contraction and Dilation) - BCC, FCC, HCP, and A15
- Lattices with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

Example: DFT-based Generalized Stacking Fault Energies

(110)

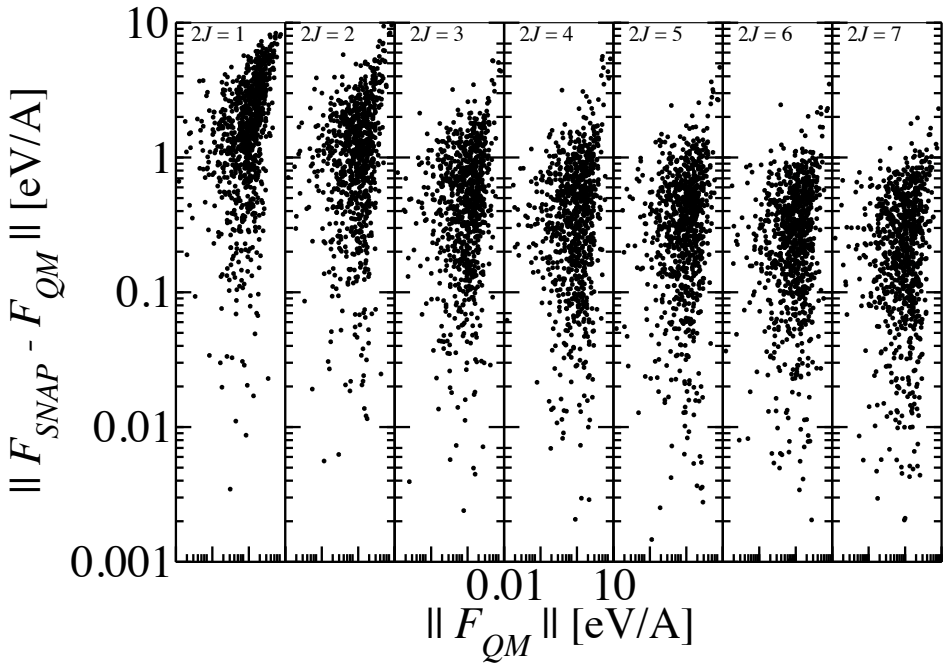


(112)



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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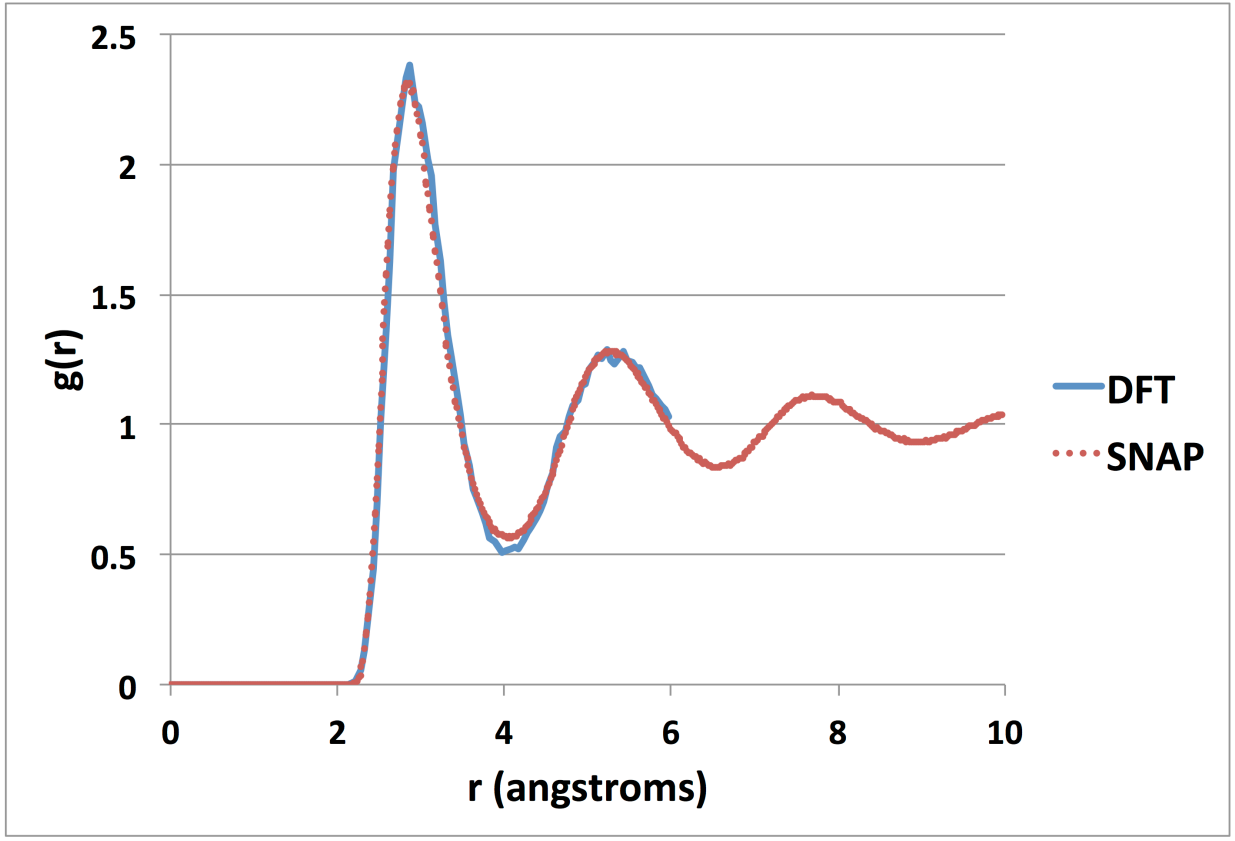
	DFT	SNAP	Zhou (EAM)	ADP
Lattice Constant (Å)	3.320	3.316	3.303	3.305
B (Mbar)	1.954	1.908	1.928	1.971
C' = (1/2)(C_{11} – C_{12}) (Mbar)	50.7	59.6	53.3	51.0
C_{44} (Mbar)	75.3	73.4	81.4	84.6
Vacancy Formation Energy (eV)	2.89	2.74	2.97	2.92
(100) Surface Energy (J/m^2)	2.40	2.68	2.34	2.24
(110) Surface 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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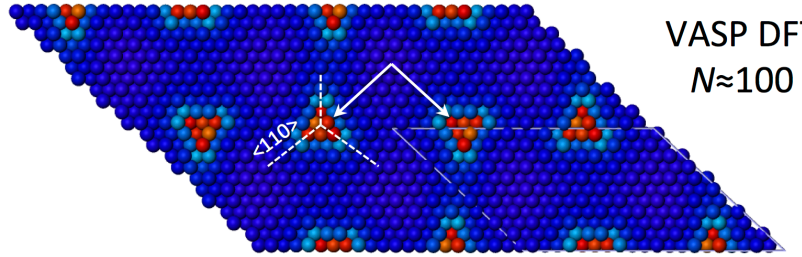
- Peierls barrier is the activation energy to move a screw dislocation
- Many simple interatomic potentials incorrectly predict a metastable state
  - Leads to erroneous dynamics
- SNAP potential agrees well with DFT calculations
  - Future work will explore dislocation dynamics based on this potential

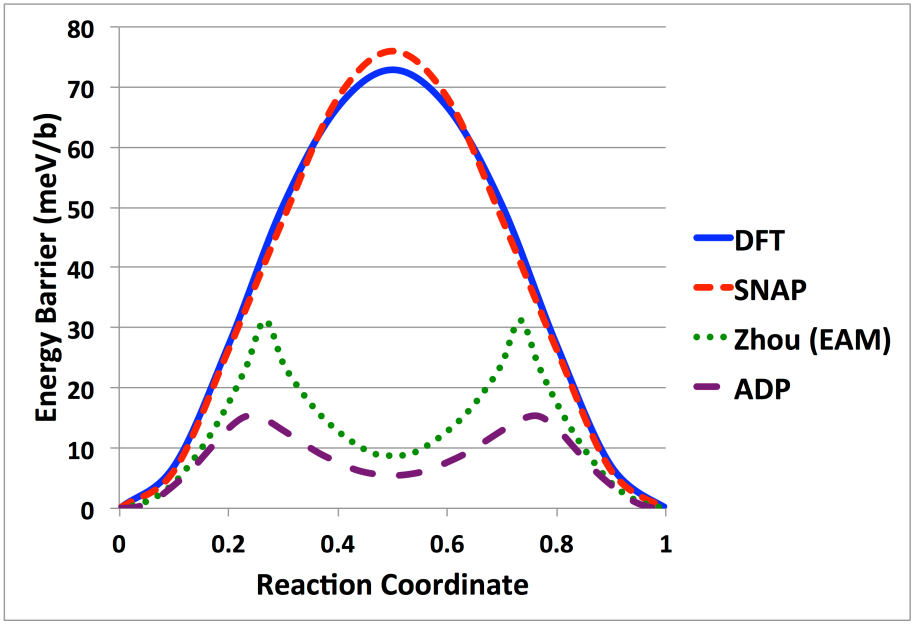
Thompson et al. arxiv.org/abs/1409.3880
J. Comp. Phys. (2015)

Screw Dislocation Dipole in Tantalum

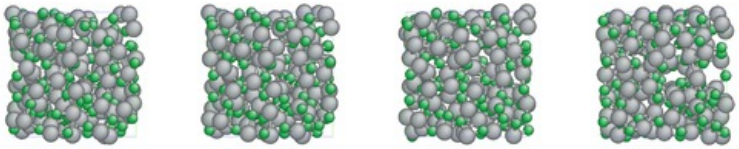


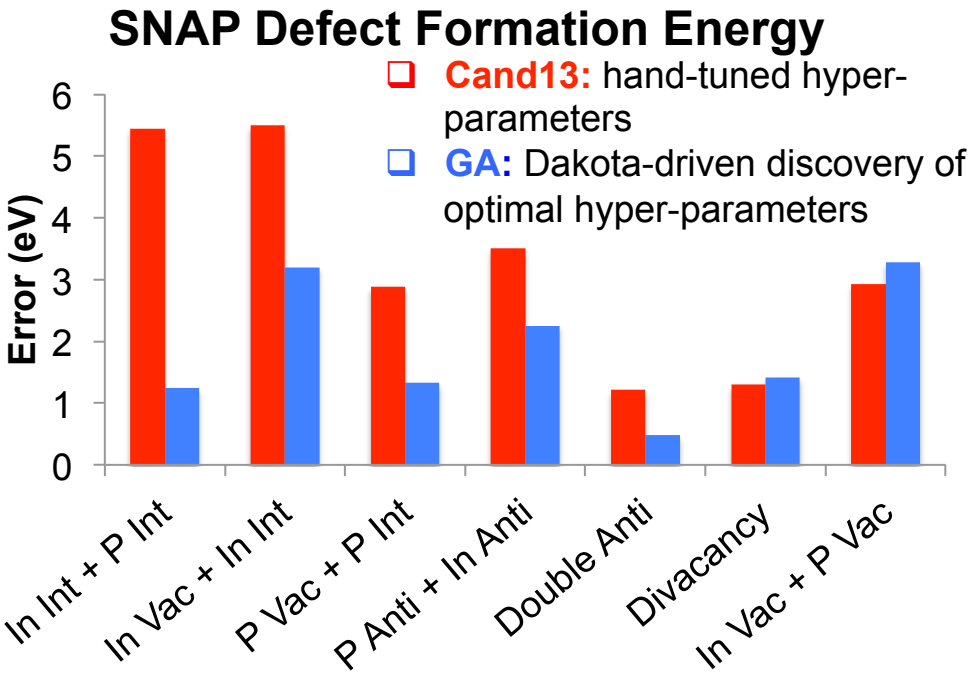
VASP DFT
$N \approx 100$

Weinberger, Tucker, and Foiles, PRB (2013)



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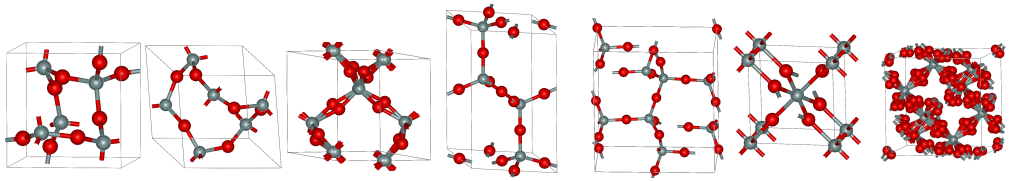
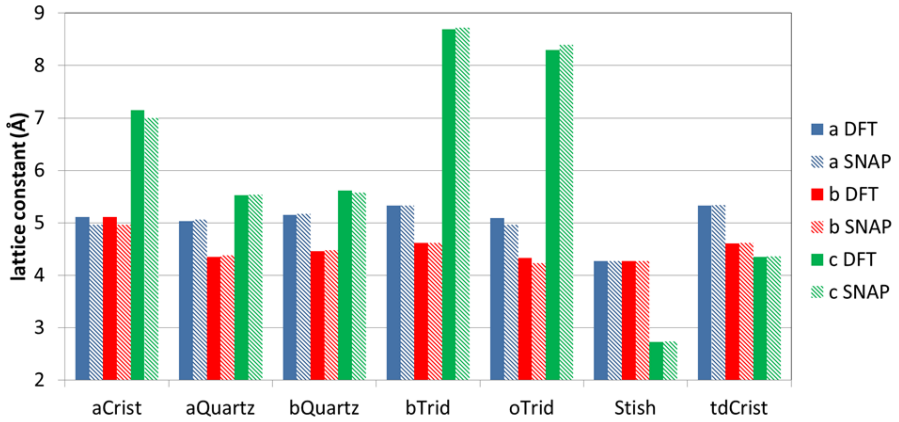
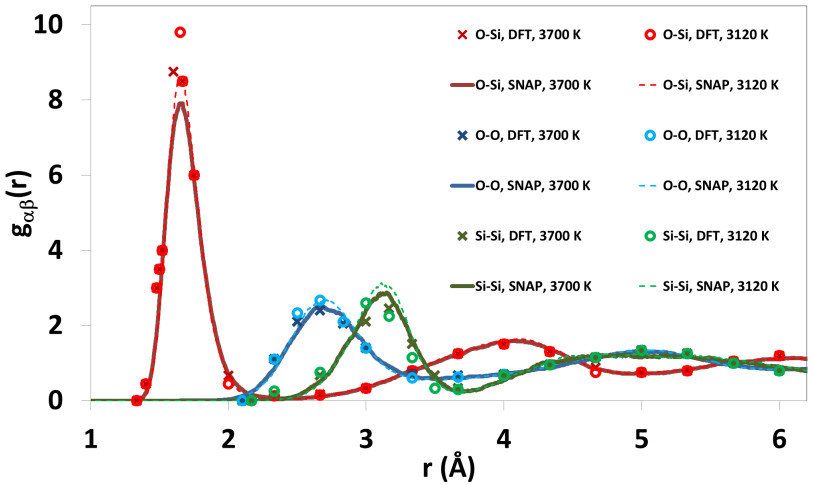
Less than 3% error in predicted lattice parameters of 7 crystal polymorphs





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
- As of yesterday, GAP is also in the LAMMPS download

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Christian Trott
Peter Schultz
Paul Crozier
Stan Moore
Adam Stephens