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SNAP: Automated Generation of High-Accuracy Interatomic Potentials using Quantum Data

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U.S. DEPARTMENT OF ENERGY
NNSA National Nuclear Security Administration

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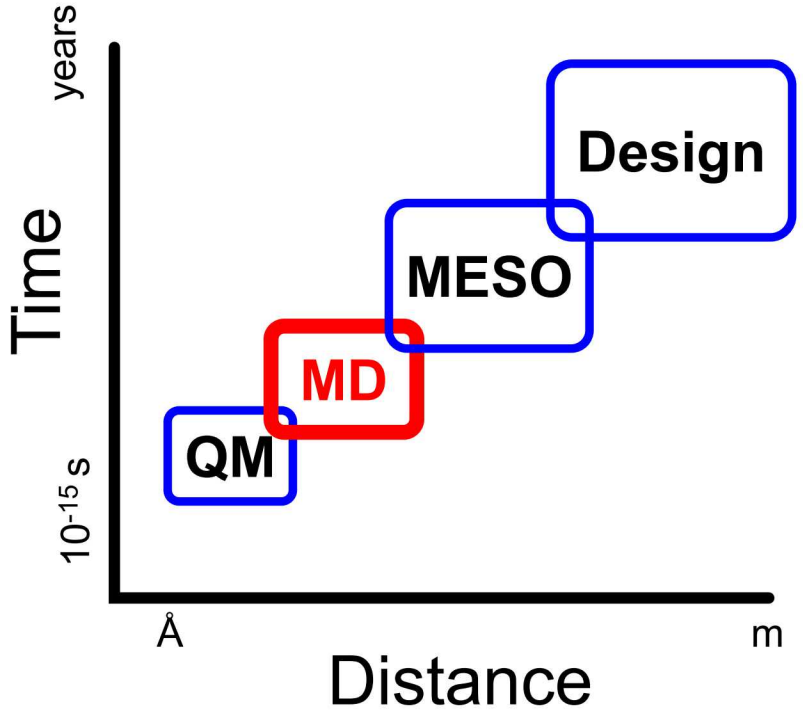
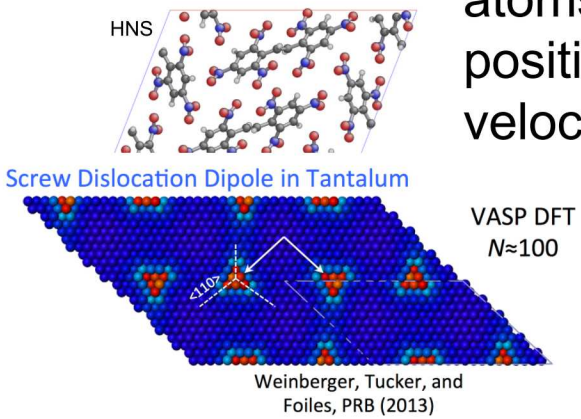
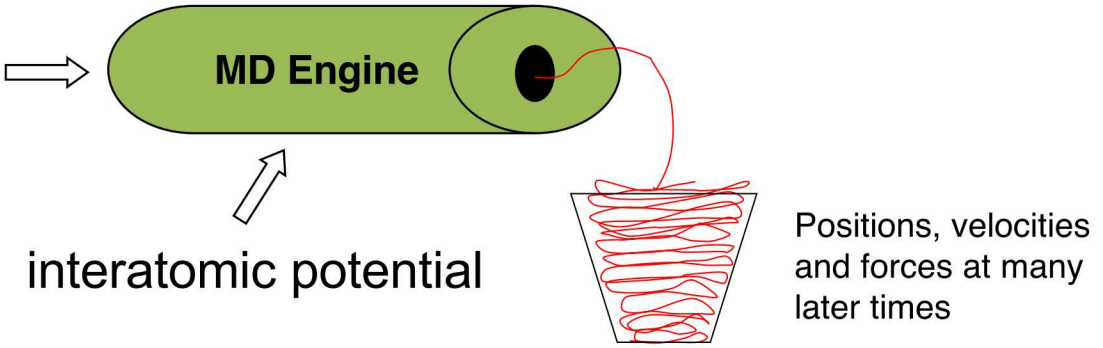
What is Molecular Dynamics Simulation?

- 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

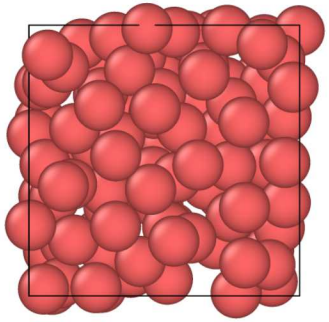




atoms,
positions,
velocities



SNAP: Spectral Neighbor Analysis Potentials



Geometric descriptors of atomic environments

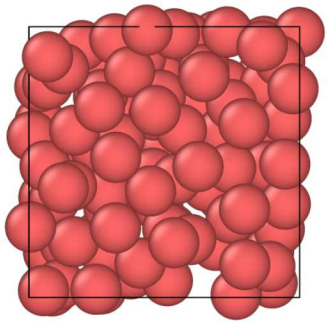
Energy as a function of geometric descriptors

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

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

- **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

SNAP Quantum-Accurate Interatomic Potentials



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- **GAP (Gaussian Approximation Potential):** Bartok, Csanyi et al., *Phys. Rev. Lett, 2010.* Uses 3D neighbor density bispectrum
- **SNAP (Spectral Neighbor Analysis Potential):** Our SNAP approach (Thompson *et al., J.Comp.Phys, 2015*) combines the neighbor bispectrum with weighted linear regression
  - Robust
  - Decouples MD speed from training set size
  - Enables large training data sets, more bispectrum coefficients
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Bispectrum Components

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

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

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

		LJ	SNAP	SNAP/LJ
Data	kBytes/atom	1	1	1
Computation	MFlOps/atom-step	0.001	10	10,000
Min *N/P*	Atom/node	100	1	1/100
Max Speed	Step/Sec	10,000	1,000	1/10

SNAP potentials predict correct Peierls barrier for Ta screw dislocations

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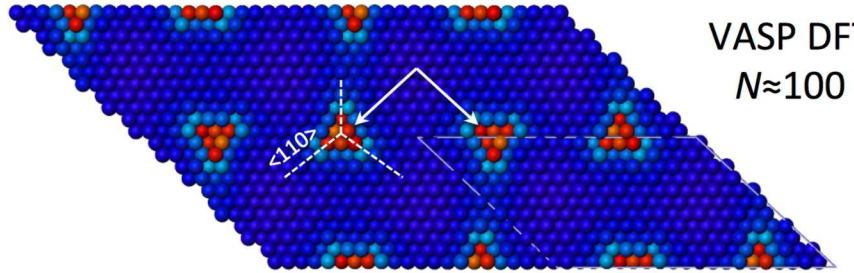
- Peierls barrier is the activation energy to move a screw dislocation
- Many simple interatomic potentials incorrectly predict a metastable state
- Leads to erroneous dynamics
- SNAP potential agrees well with DFT calculations

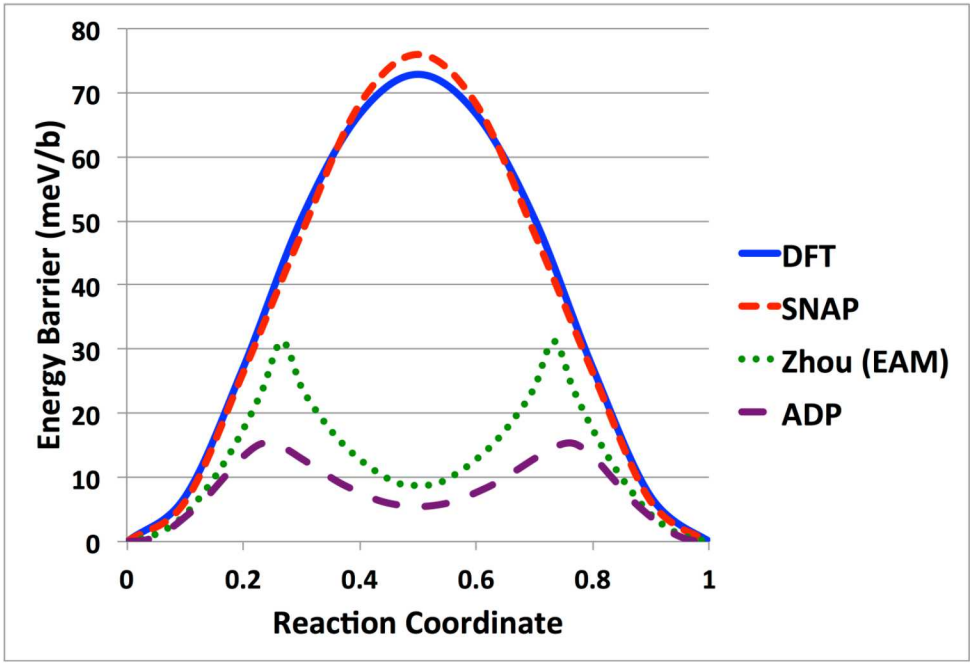
Thompson et al. arxiv.org/abs/1409.3880
J. Comp. Phys. (2015)

Screw Dislocation Dipole in Tantalum



VASP DFT
$N \approx 100$

Weinberger, Tucker, and Foiles, PRB (2013)



SNAP Potentials for W-He

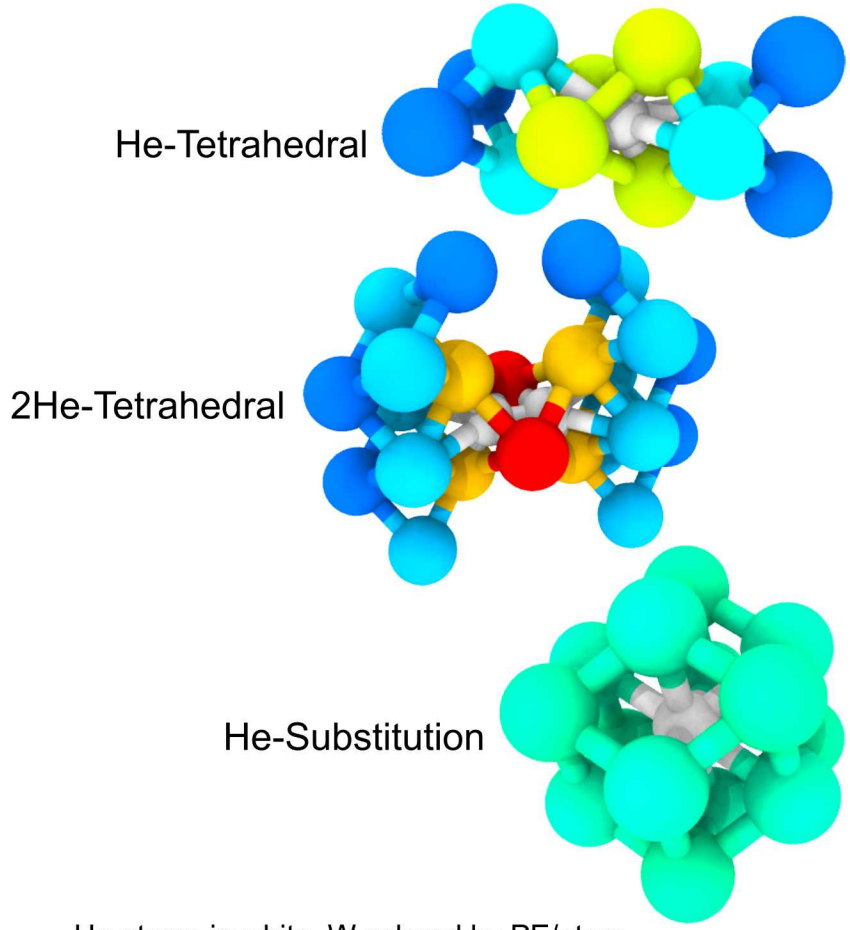
Transferrable, User Generated Training

2911 Pure W Training Data +

250 x Elastic Deformation w/ 1 octahedral He
250 x Elastic Deformation w/ 1 tetrahedral He
50 x EoS w/ 1 octahedral He
50 x EoS w/ 1 tetrahedral He
227 x Random displacements w/ 1,2 or 4 He

Objective Functions in Dakota:
- Energy Error w.r.t. DFT
- Force Error w.r.t. DFT
- Tungsten Structure Properties (a_0, elastic const.)
- Tungsten Defects (same as before)
- Helium substitution, Tetra, Octa and Tetra-Tetra binding

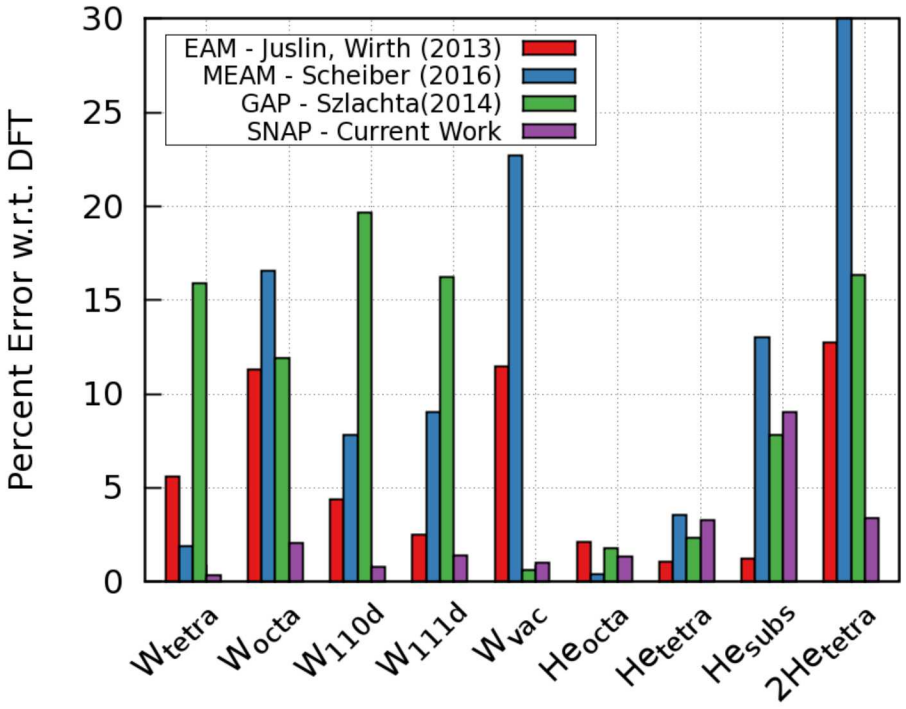
*Reference values taken from Brian's paper



He atoms in white, W colored by PE/atom

Comparison of W EAM, MEAM and SNAP + He pair

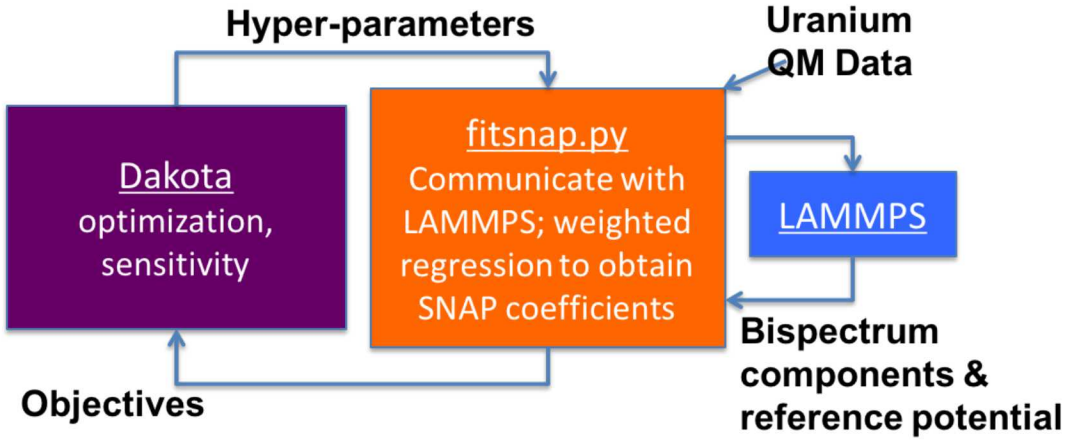
	DFT[a]	EAM[b]	MEAM[c]	GAP[d]	SNAP[e]
$E_{W-Tetra}$	11.1	10.4	11.3	12.8	11.1
E_{W-Octa}	11.7	10.4	13.6	13.1	11.5
$E_{W-[110]d}$	9.8	10.3	9.1	11.8	9.8
$E_{W-[111]d}$	9.6	9.8	10.4	11.1	9.7
$E_{W-vacancy}$	3.3	3.7	4.0	3.3	3.2
$E_{W-divacancy}$[f]	0.1	-0.4	-0.2	0.4	0.1
$E_{He-Tetra}$	6.2	6.2	6.4	6.3	6.4
$E_{He-Octa}$	6.4	6.2	6.4	6.3	6.3
$E_{He-Subs}$	4.7	4.8	5.3	4.3	4.3
$E_{2He-Tetra}$[g]	1.0	0.9	0.6	0.8	1.0



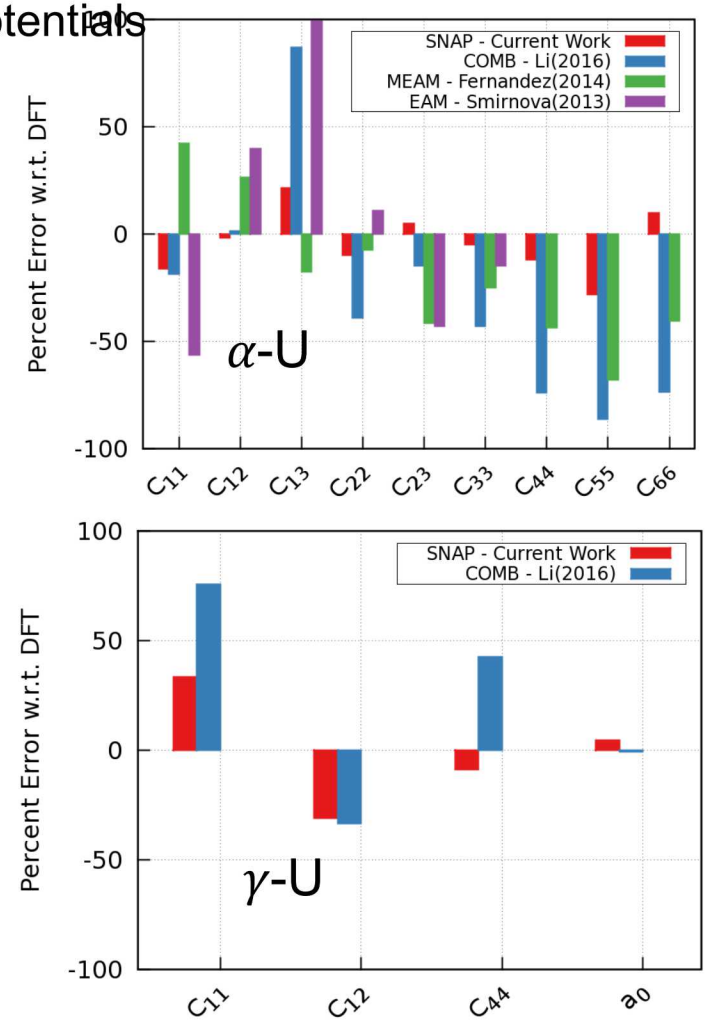
- Percent error is with respect to DFT values reported from *Becquart, Domain, Nucl. Instrum. Methods Phys. Res. B 255 (1) (2007) 23– 26.*
- Difference in He-Oct and He-Tet is tricky to handle in all potentials, should be -0.22eV

Accomplishment: Ensemble of Uranium Potentials

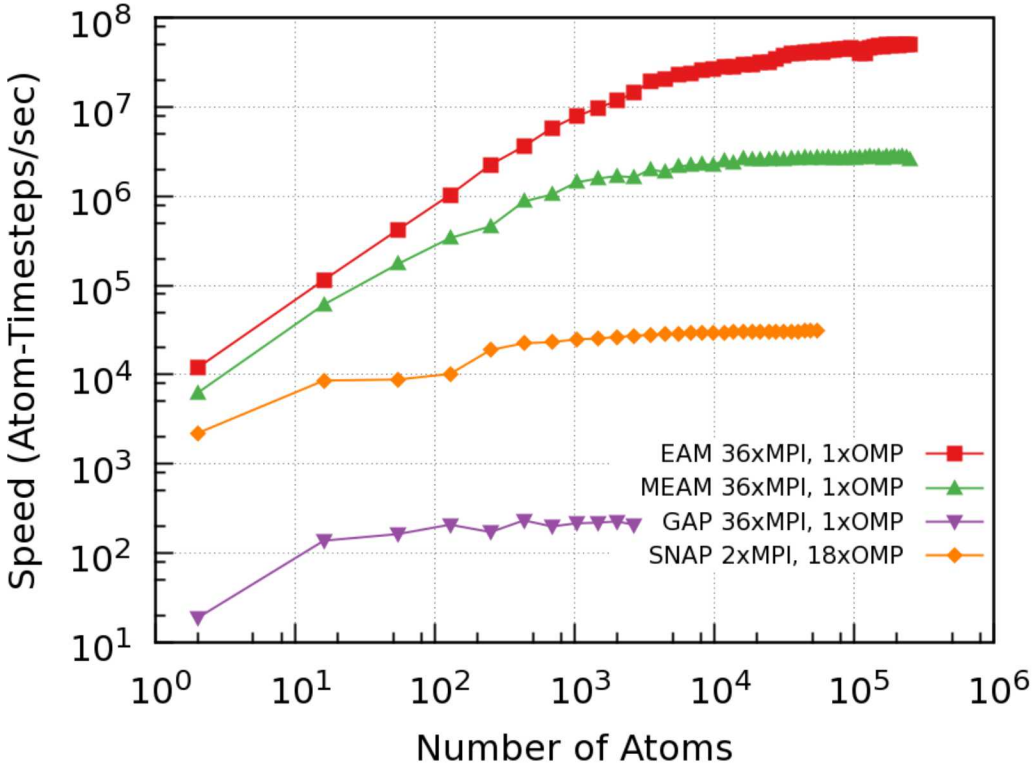
- **Generating interatomic potentials is essentially a black box, this is true for all potential types. We aim to improve the transparency and transferability of this process.**
- SNAP fitting process generates ensemble of potentials
- Evaluation against multiple objectives identifies Pareto-optimal potentials, subject to further screening
- **Sub-set of SNAP potential ensemble out-perform previous published potentials on elastic constants**



Comparison of elastic constants predictions for α-U and γ-U phases: representative SNAP potential against previous published potentials



Computational Cost of Tungsten Potentials



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Conclusions

- Highly complex potentials becoming more prevalent
- High Flops/atom allows excellent strong scaling
- Many-core and GPU platforms require exploiting low-level parallelism
- Extensive code modification required for each new platform
- Algorithmic improvements also important
- SNAP code and potentials available at http://lammps.sandia.gov