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Translating Excited State Dynamics to Classical Interatomic Potentials

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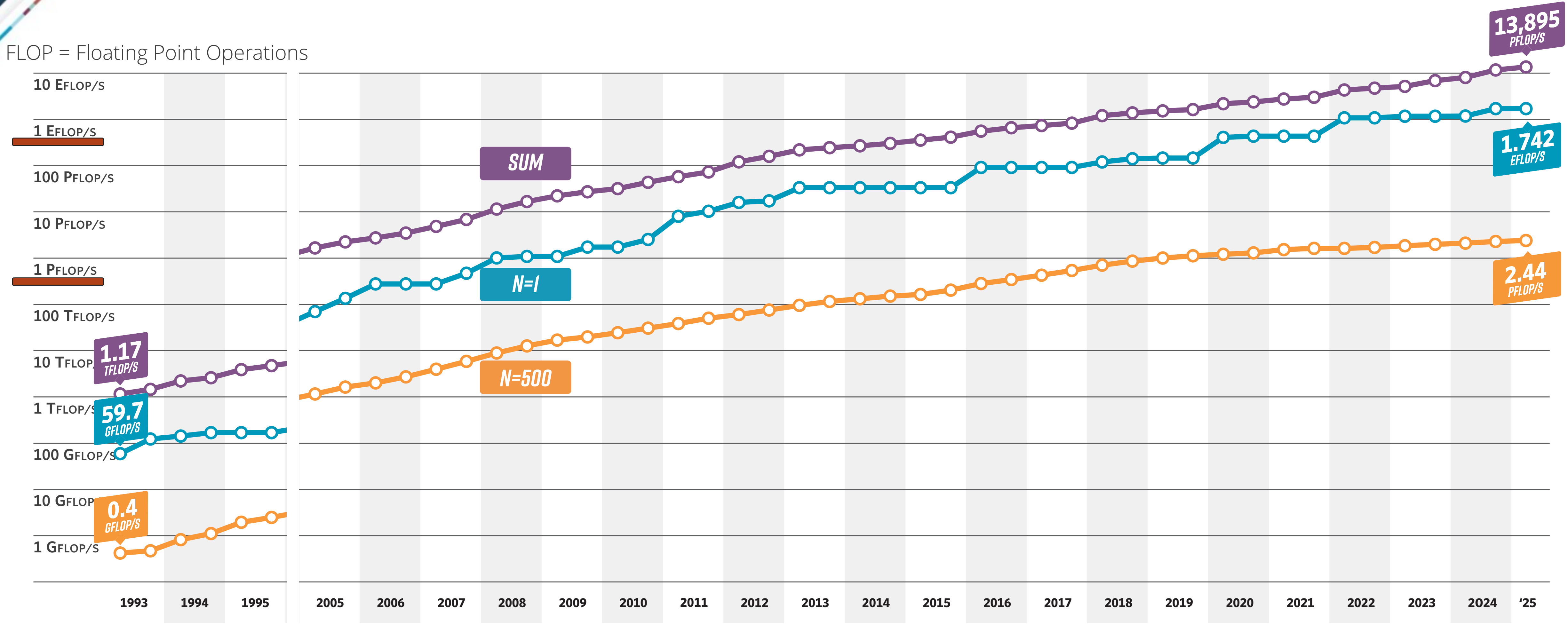
Center for Computing Research
Sandia National Laboratories, Albuquerque, NM

LAMMPS Workshop, Albuquerque, 2025

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

21st Century Theorist (?)

FLOP = Floating Point Operations

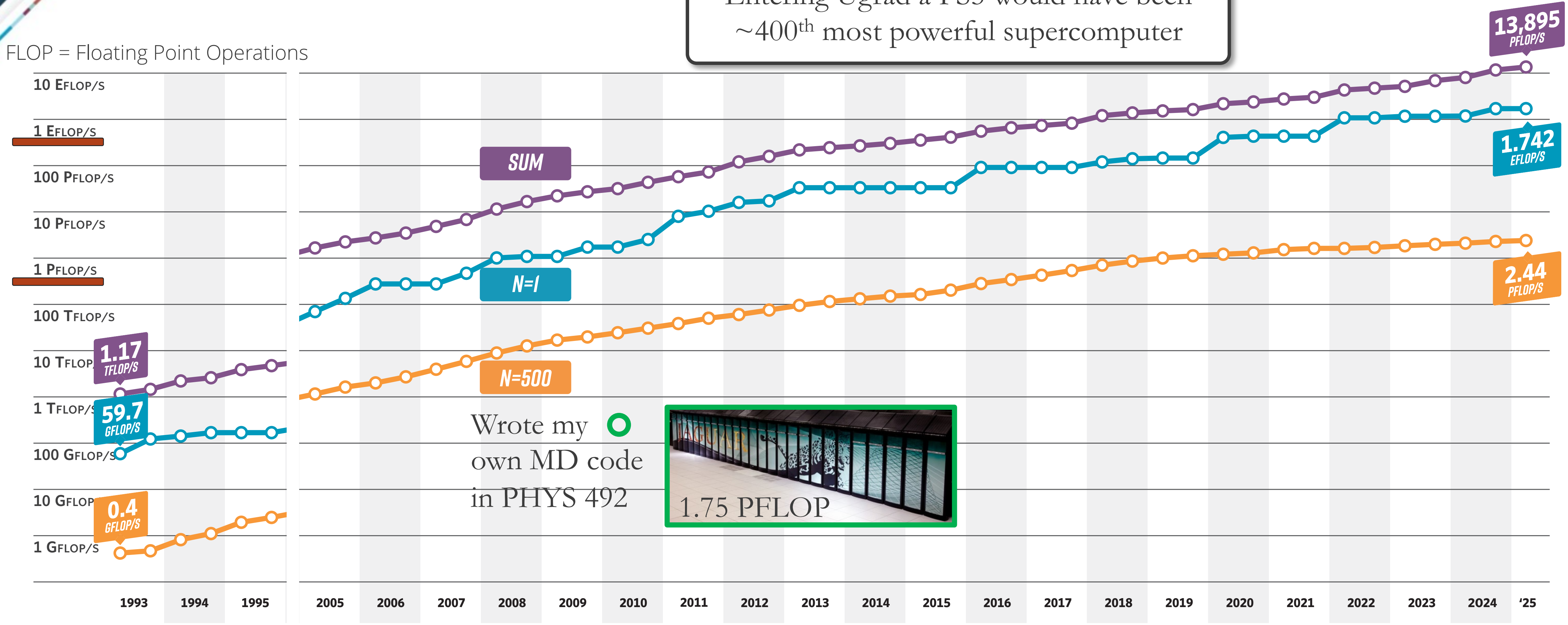


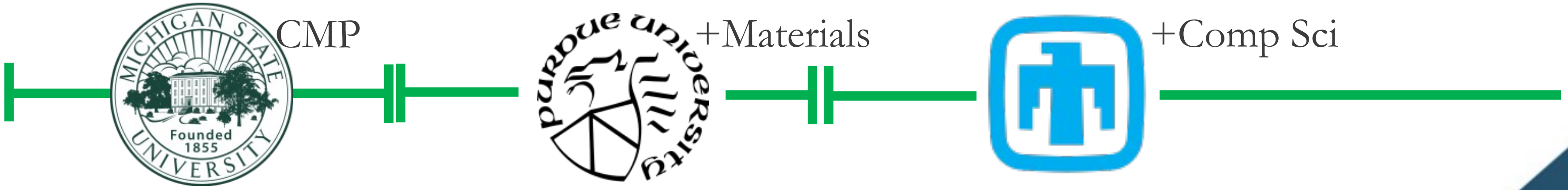
More powerful supercomputers are inevitable, but is our scientific usage of this technology keeping up?

My Perspective

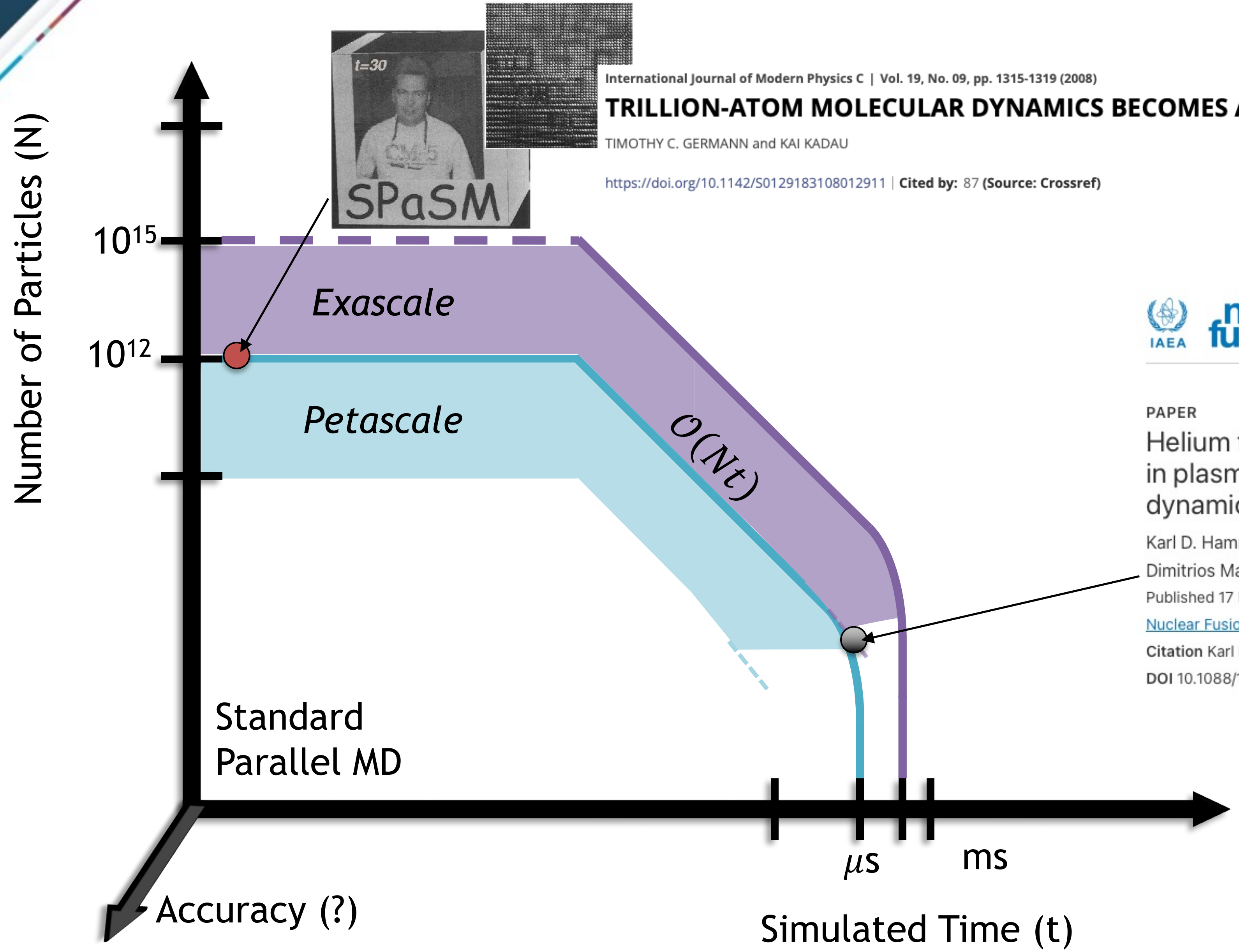
Entering Ugrad a PS5 would have been ~400th most powerful supercomputer

FLOP = Floating Point Operations





Frontiers of Molecular Dynamics



International Journal of Modern Physics C | Vol. 19, No. 09, pp. 1315-1319 (2008)

TRILLION-ATOM MOLECULAR DYNAMICS BECOMES A REALITY

TIMOTHY C. GERMANN and KAI KADAU

https://doi.org/10.1142/S0129183108012911 | **Cited by:** 87 **(Source: Crossref)**

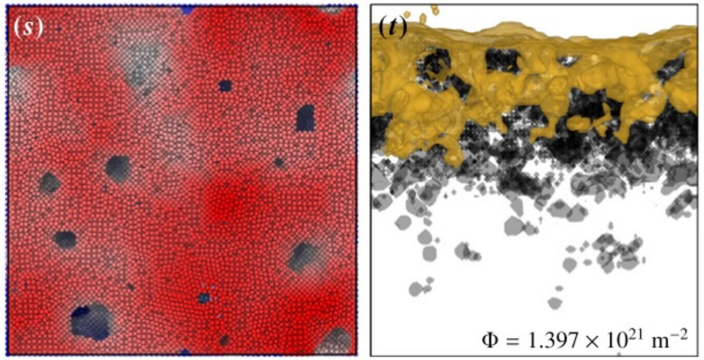
IAEA nuclear fusion

PAPER

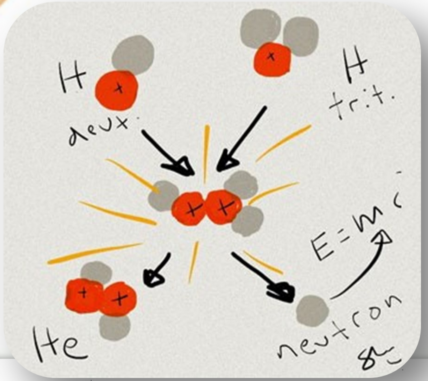
Helium flux effects on bubble growth and surface morphology in plasma-facing tungsten from large-scale molecular dynamics simulations

Karl D. Hammond[1,2], Ian V. Naeger[1], Wathsala Widanagamaachchi[8,3], Li-Ta Lo[4], Dimitrios Maroudas[5] and Brian D. Wirth[6,7]

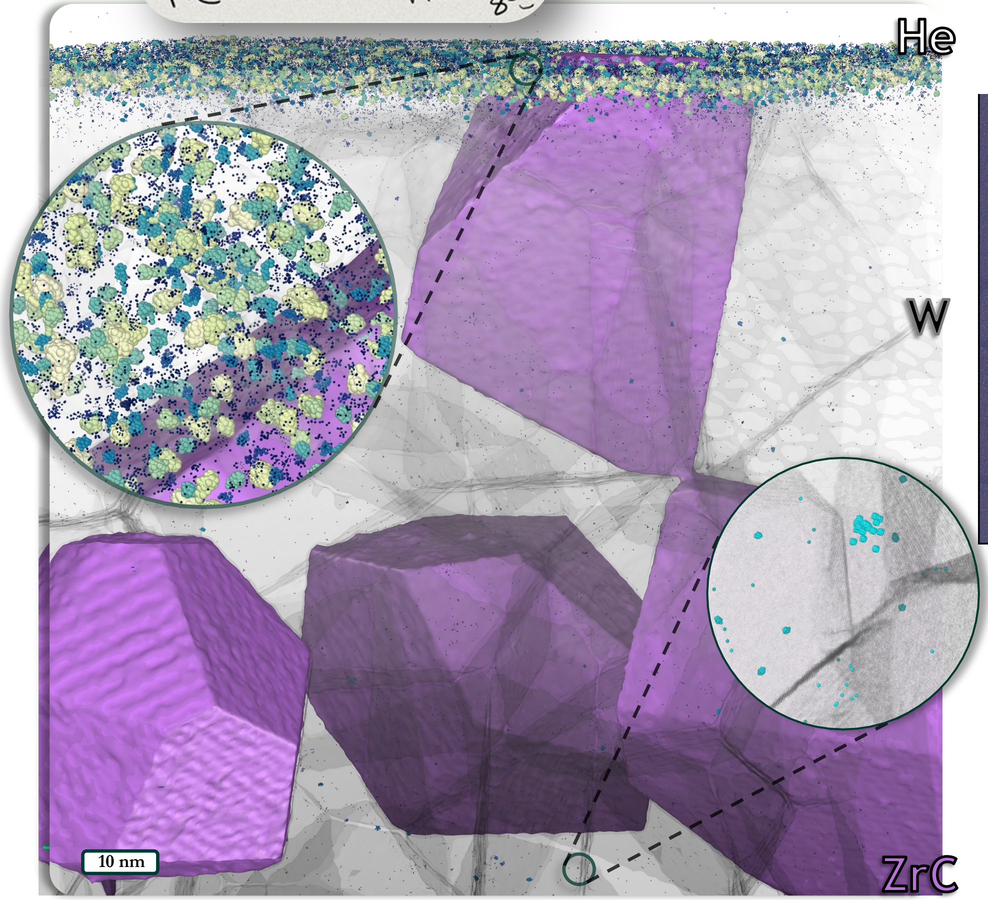
Published 17 May 2019 • © 2019 IAEA, Vienna
Nuclear Fusion, Volume 59, Number 6
Citation Karl D. Hammond *et al* 2019 *Nucl. Fusion* **59** 066035
DOI 10.1088/1741-4326/ab12f6



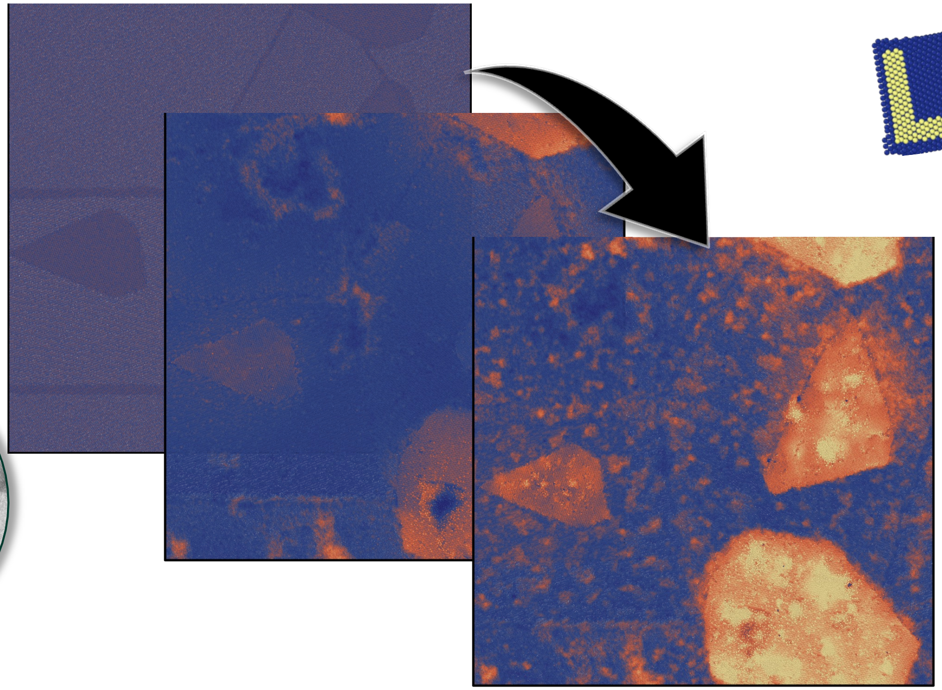
Exascale Molecular Dynamics



To operate safely and efficiently, fusion power plants must be designed with materials that can withstand the extreme heat and particle loads generated by the fusion process.



Challenge: Current approaches to material selection are too slow and expensive, because they rely on human physical intuition, domain expertise, and trial and error. Enter ML and Molecular Dynamics to help out.



gitlab.com/exaalt
github.com/lammps/lammps

Outcome: Demonstrated fast virtual assessment of new materials, from the training of accurate machine-learned models to large-scale, long-time exascale simulations

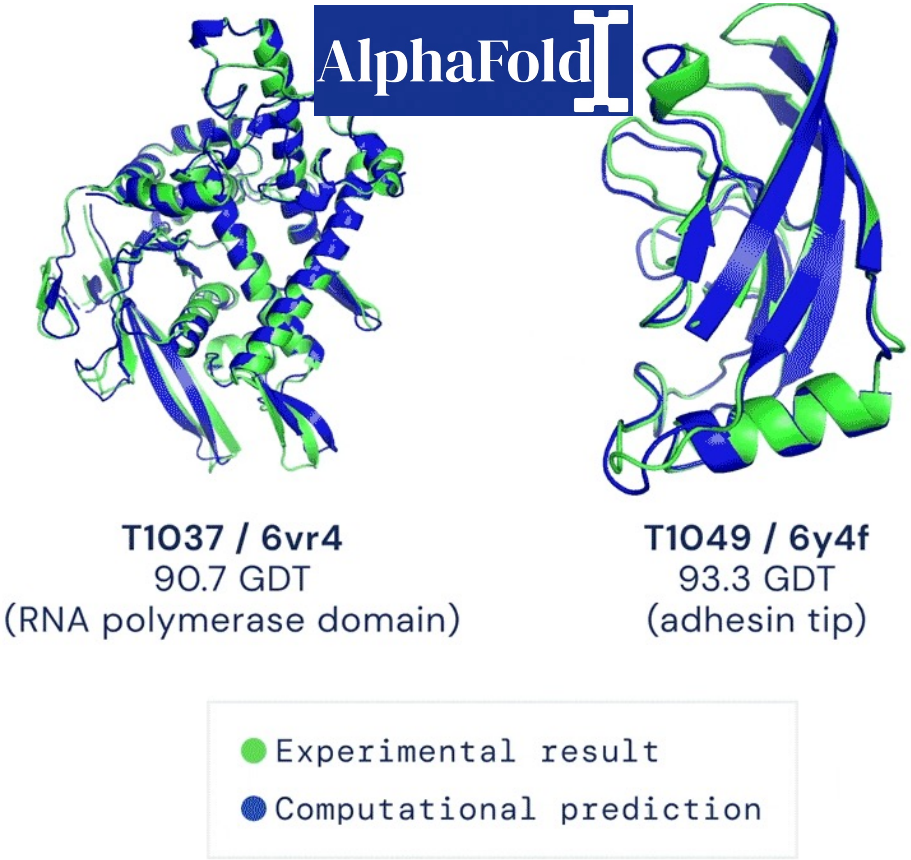
Captured surface erosion rates in a candidate material utilizing **4 Frontier-Days**.

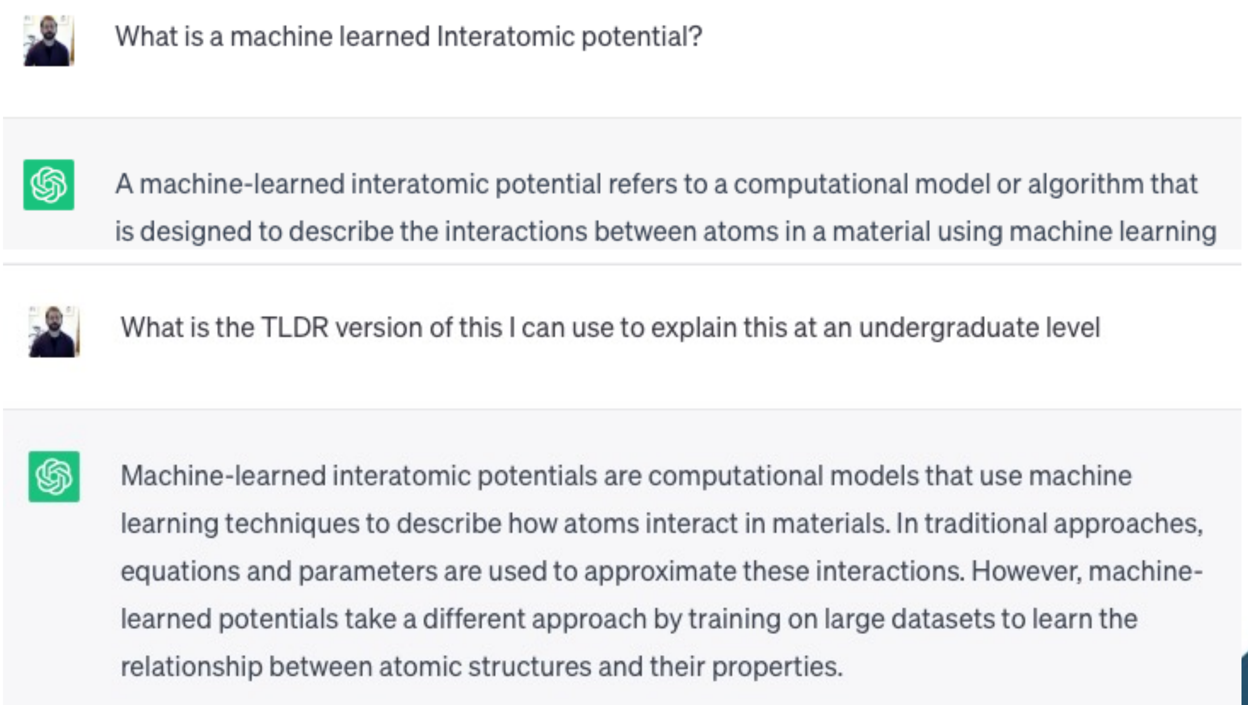
Exascale Molecular Dynamics

Observation 1: Machine learning has broken the traditional accuracy and cost tradeoff

Observation 2: Model development has been democratized, inviting broader uses of modeling / simulation methods

Claim: Exascale computing power will be used to push the frontiers of simulation accuracy not length/time




What is a machine learned Interatomic potential?

A machine-learned interatomic potential refers to a computational model or algorithm that is designed to describe the interactions between atoms in a material using machine learning

What is the TLDR version of this I can use to explain this at an undergraduate level

Machine-learned interatomic potentials are computational models that use machine learning techniques to describe how atoms interact in materials. In traditional approaches, equations and parameters are used to approximate these interactions. However, machine-learned potentials take a different approach by training on large datasets to learn the relationship between atomic structures and their properties.

MD Approximations Change Over Time

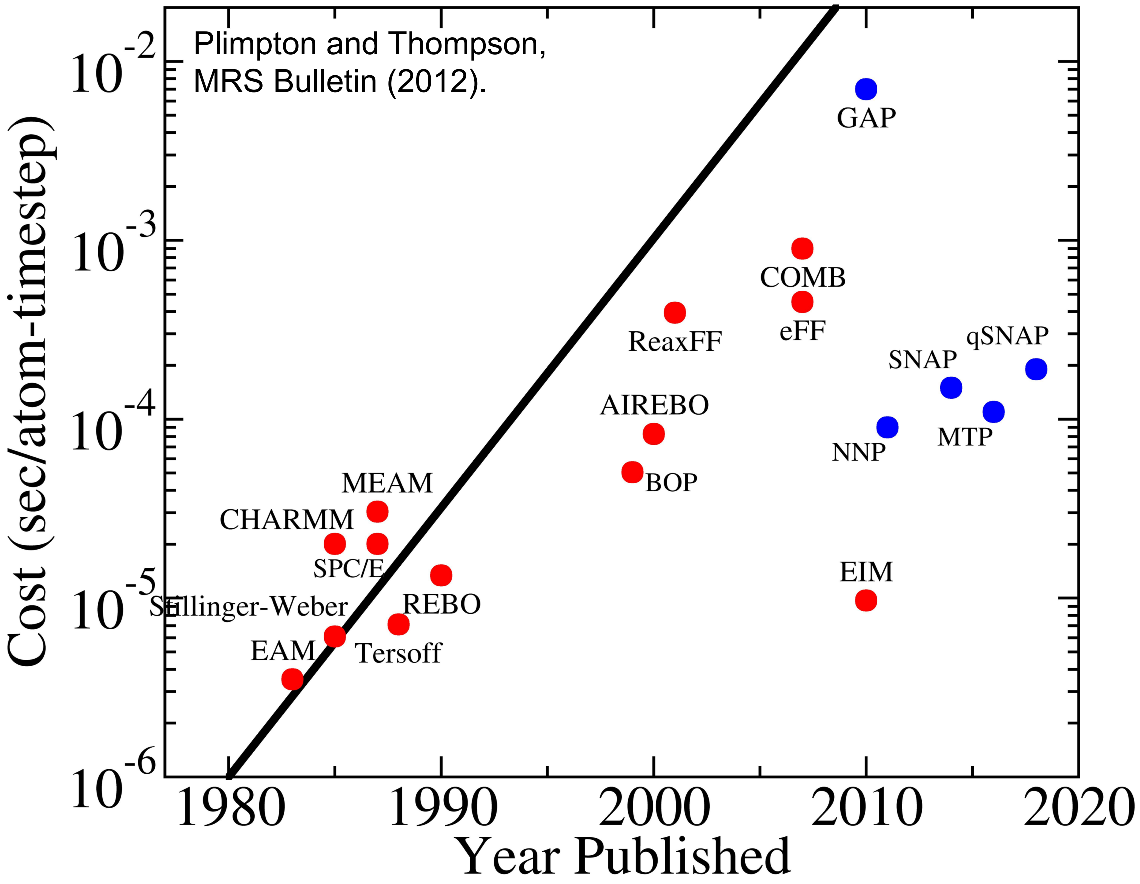
http://lammps.sandia.gov

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

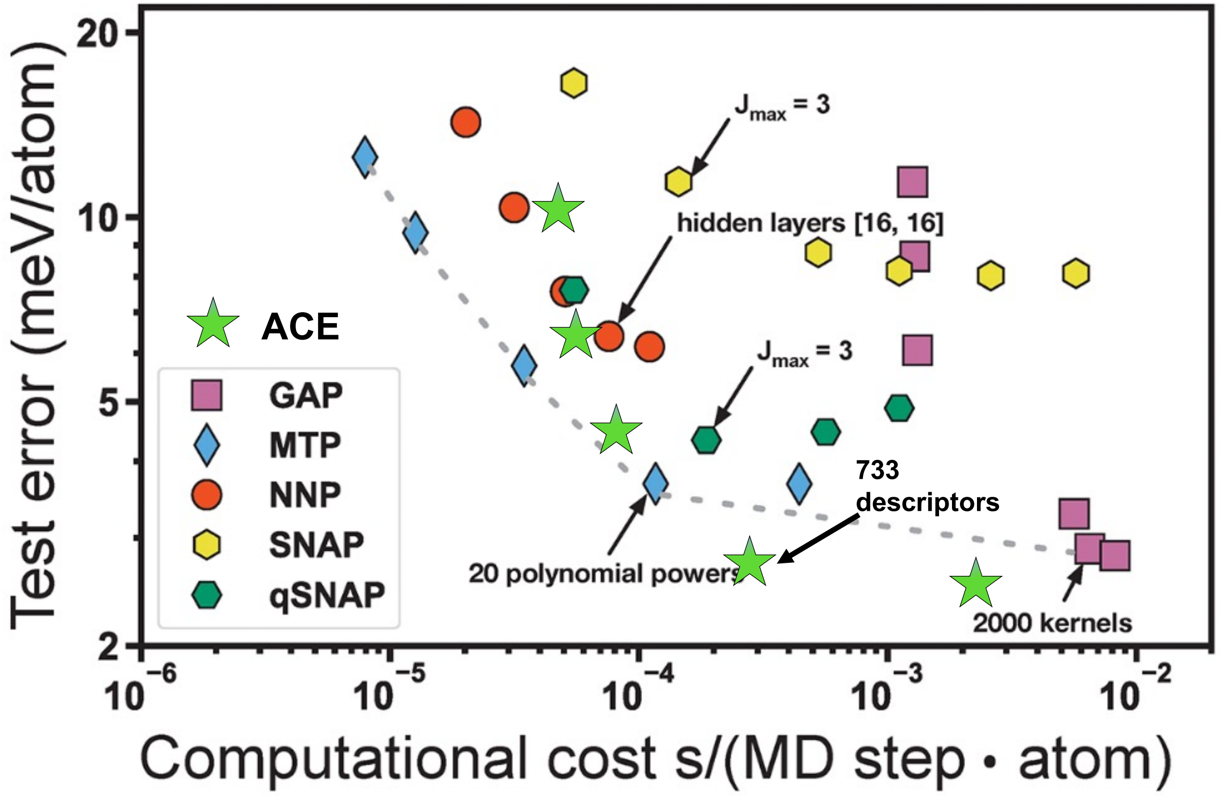
Manybody (1980s)
Stillinger-Weber, Tersoff, Embedded Atom Method

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

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



https://github.com/materialsvirtuallab/mlearn



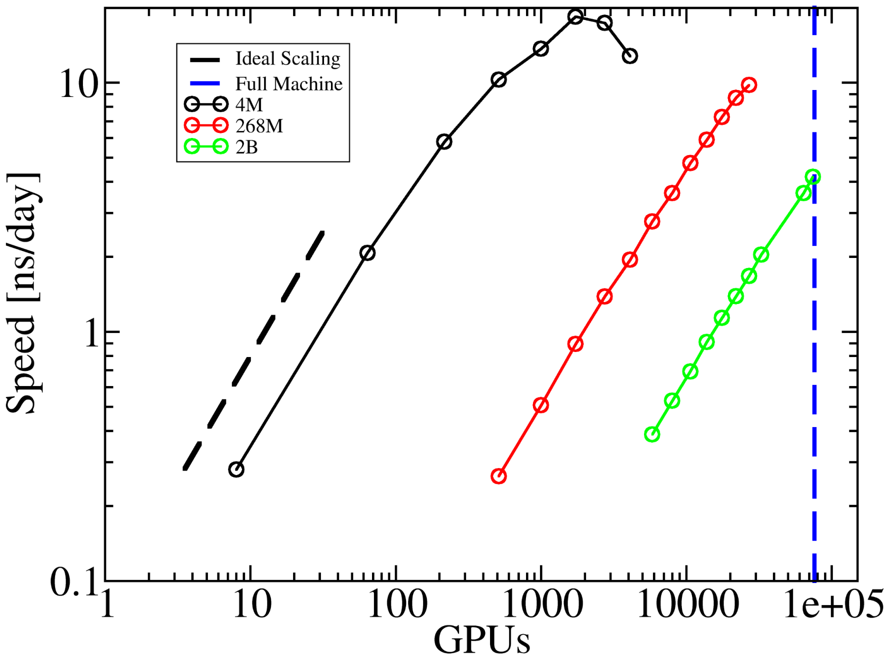
Exascale Ready Models

Performance Metrics

- Different descriptors, different computational cost.
- LAMMPS with ML-IAP are now a qualification metric for new exascale platforms!

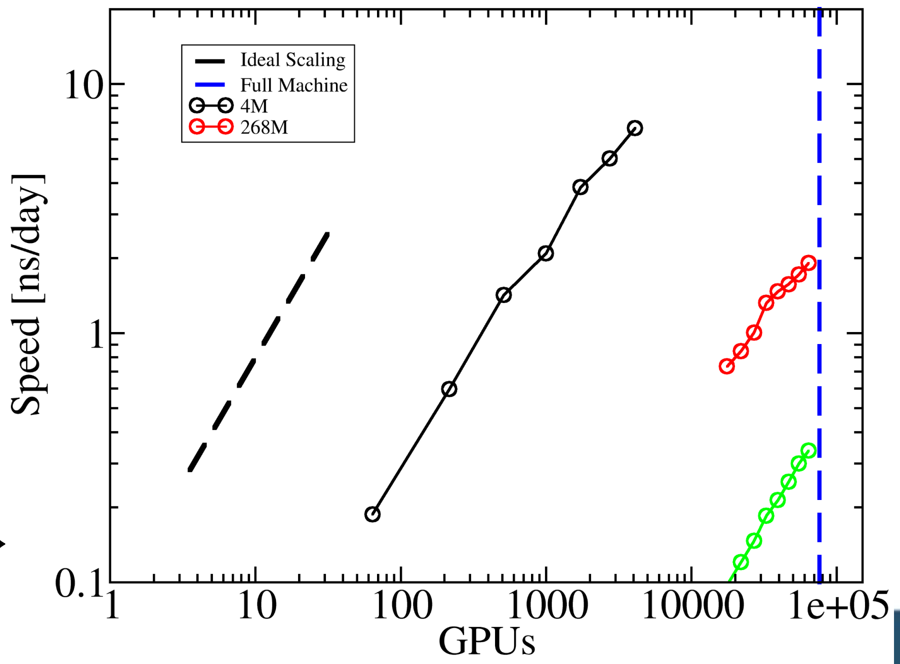
Oak Ridge National Laboratory

FRONTIER



←Bispectrum Components

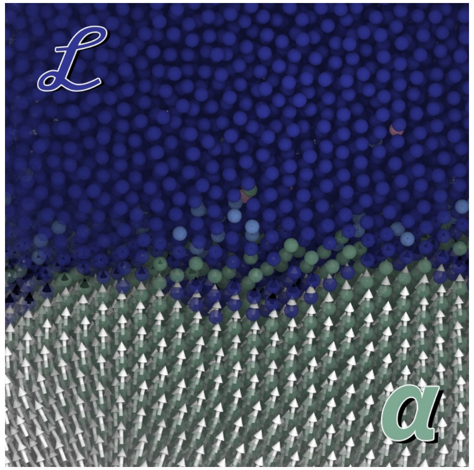
Atomic Cluster Expansion→



What is Next Gen MD?

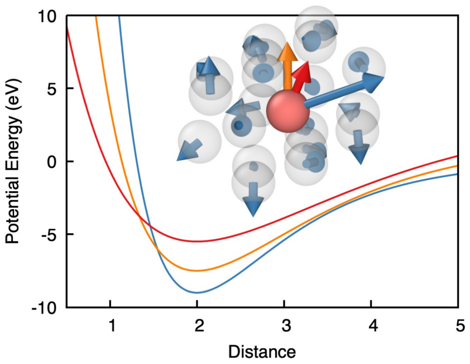
Magnetic Effects

- Available in LAMMPS, but developments needed for many-body spin interactions and other novel Hamiltonians



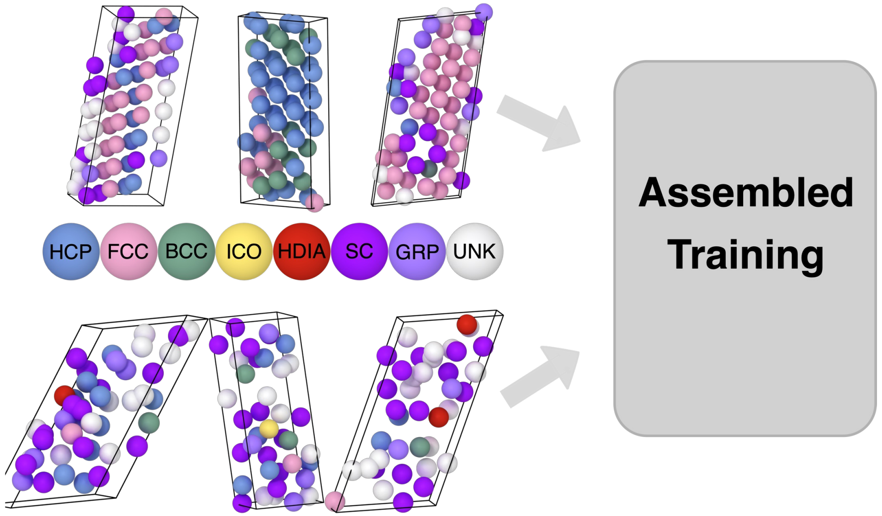
Electronic Excitations

- Aspirational, but achievable given the rapid advancements of ML-IAP



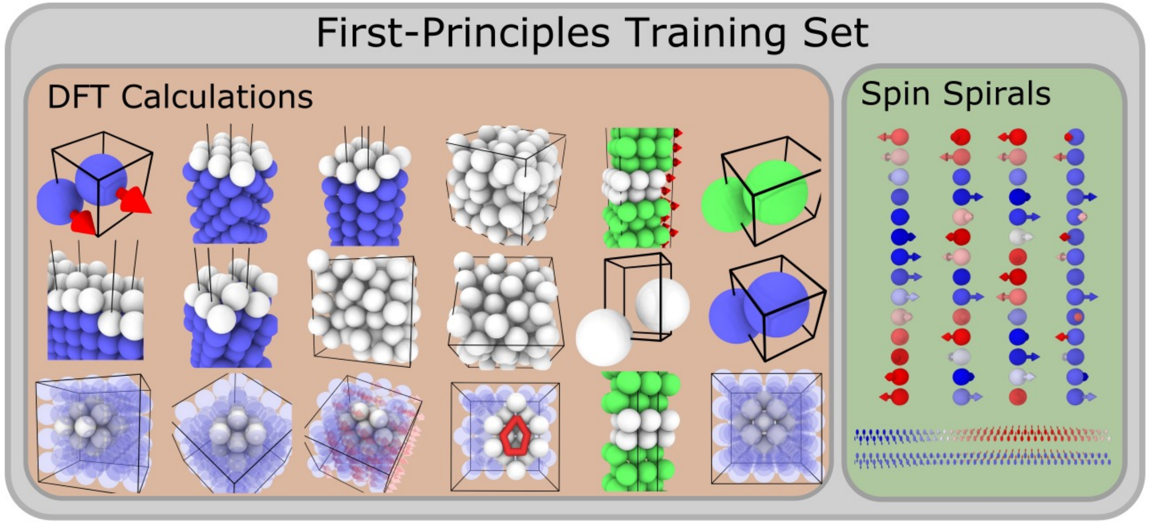
Novel Descriptor Usage

- What is the value added beyond accuracy of using a ML-IAP given the computational cost?
  - K > 0 : Minimize Local Entropy



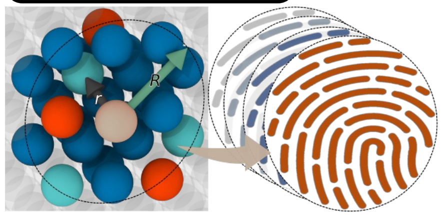
  - K < 0 : Maximize Local Entropy

Multi-Physics Molecular Dynamics



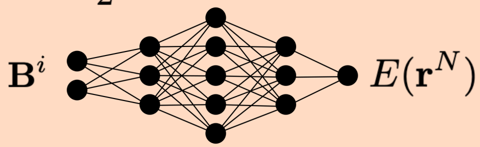
Atomic Interaction

$$\sum_i \frac{\mathbf{p}_i^2}{2m} + E(\mathbf{R})$$



SNAP Models

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

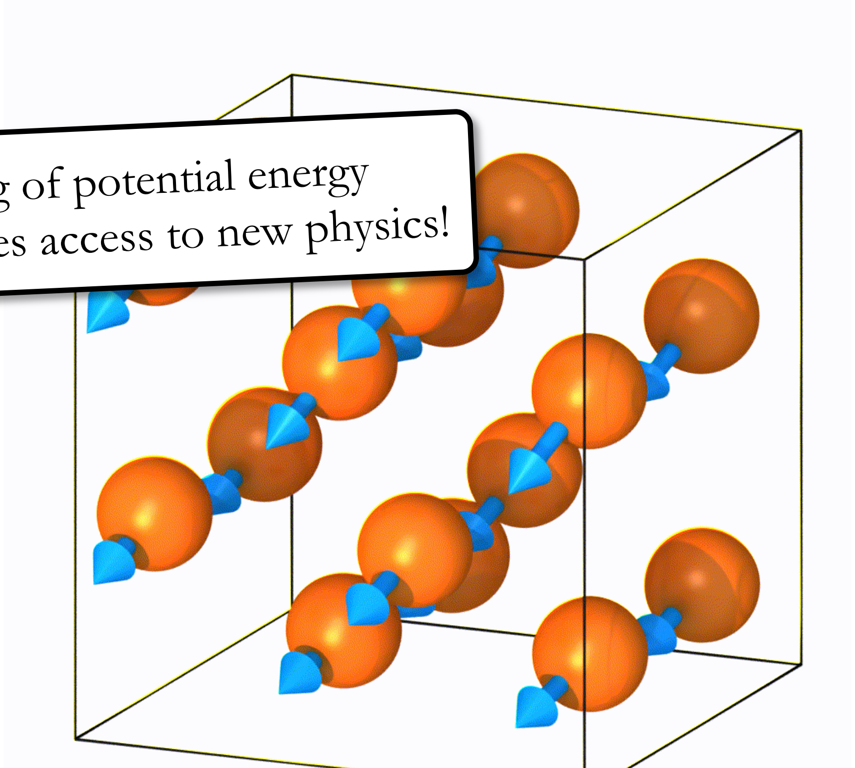


Spin Exchange Interaction

$$\sum_{i,j}^{N} J_{ij}(\mathbf{R})\left[\vec{s_i} \cdot \vec{s_j} - 1\right] - \sum_{i,j}^{N} K_{ij}(\mathbf{R})\left[\left(\vec{s_i} \cdot \vec{s_j}\right)^2 - 1\right]$$

Spin-Crystal Anisotropy Interaction

$$\frac{1}{2}\sum_{i,j=1,i\neq j}^{N} \left\{ \begin{array}{c} l_1(\mathbf{R})\left[(\boldsymbol{e}_{ij}\cdot \boldsymbol{s}_i)(\boldsymbol{e}_{ij}\cdot \boldsymbol{s}_j) - \frac{\boldsymbol{s}_i \cdot \boldsymbol{s}_j}{3}\right] + \\ q_1(\mathbf{R})\left[(\boldsymbol{e}_{ij}\cdot \boldsymbol{s}_i)^2 - \frac{\boldsymbol{s}_i \cdot \boldsymbol{s}_j}{3}\right]\left[(\boldsymbol{e}_{ij}\cdot \boldsymbol{s}_j)^2 - \frac{\boldsymbol{s}_i \cdot \boldsymbol{s}_j}{3}\right] \\ +q_2(\mathbf{R})\left[(\boldsymbol{e}_{ij}\cdot \boldsymbol{s}_i)(\boldsymbol{e}_{ij}\cdot \boldsymbol{s}_j)^3 + (\boldsymbol{e}_{ij}\cdot \boldsymbol{s}_j)(\boldsymbol{e}_{ij}\cdot \boldsymbol{s}_i)^3\right] \end{array} \right\}$$

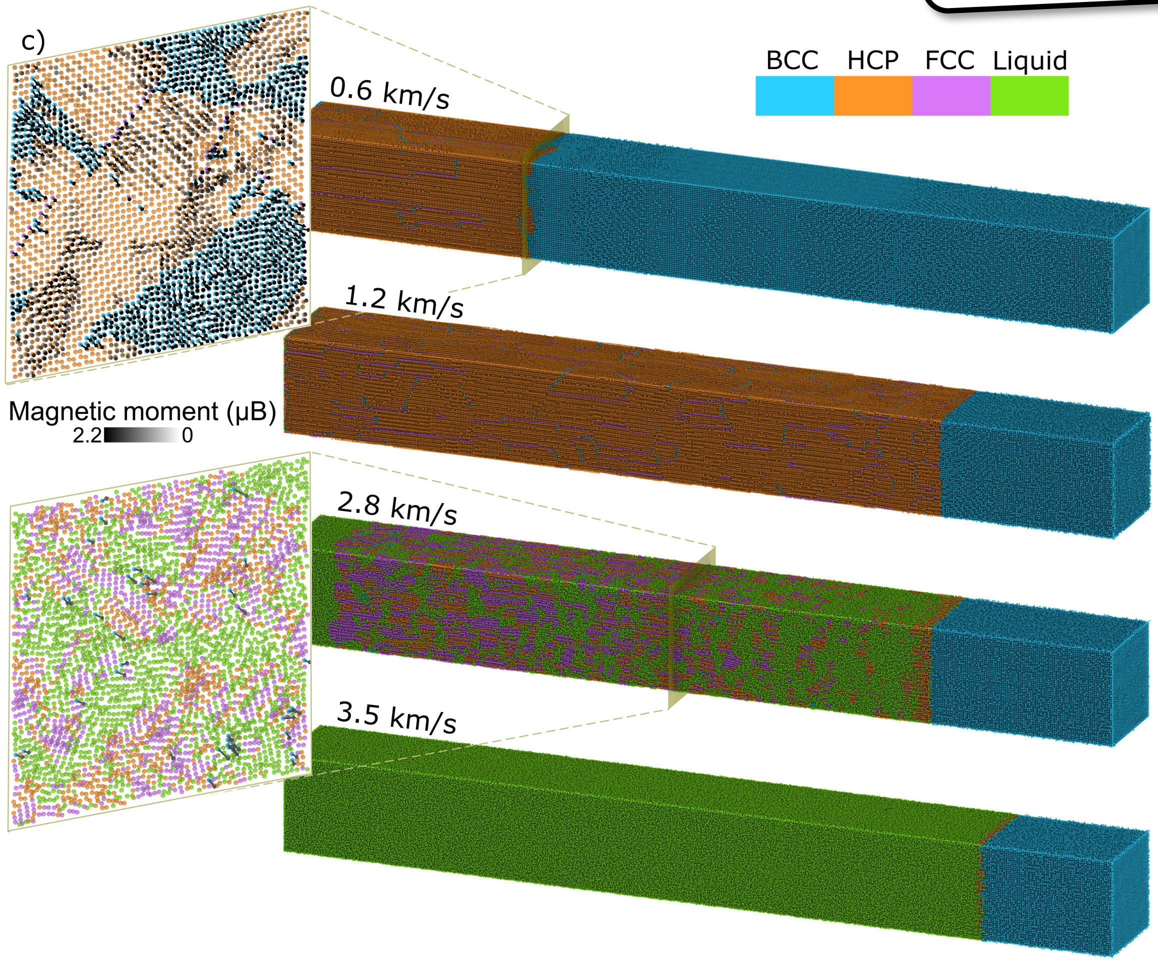


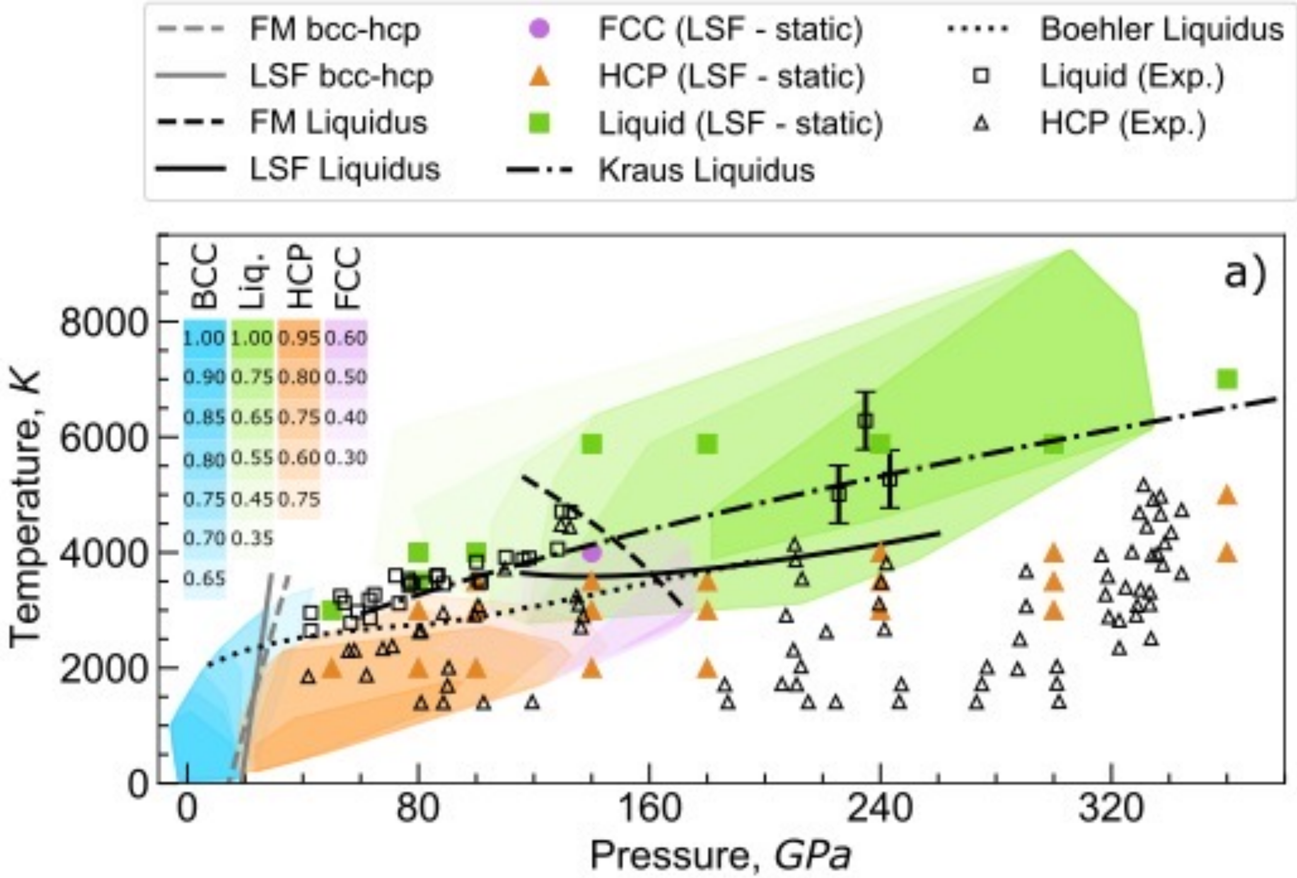
Layering of potential energy surfaces gives access to new physics!

Atomic Insight Into the Earths' Core

"Probing iron in Earth's core with molecular-spin dynamics"
Nikolov, S … Wood, M. PNAS 2024

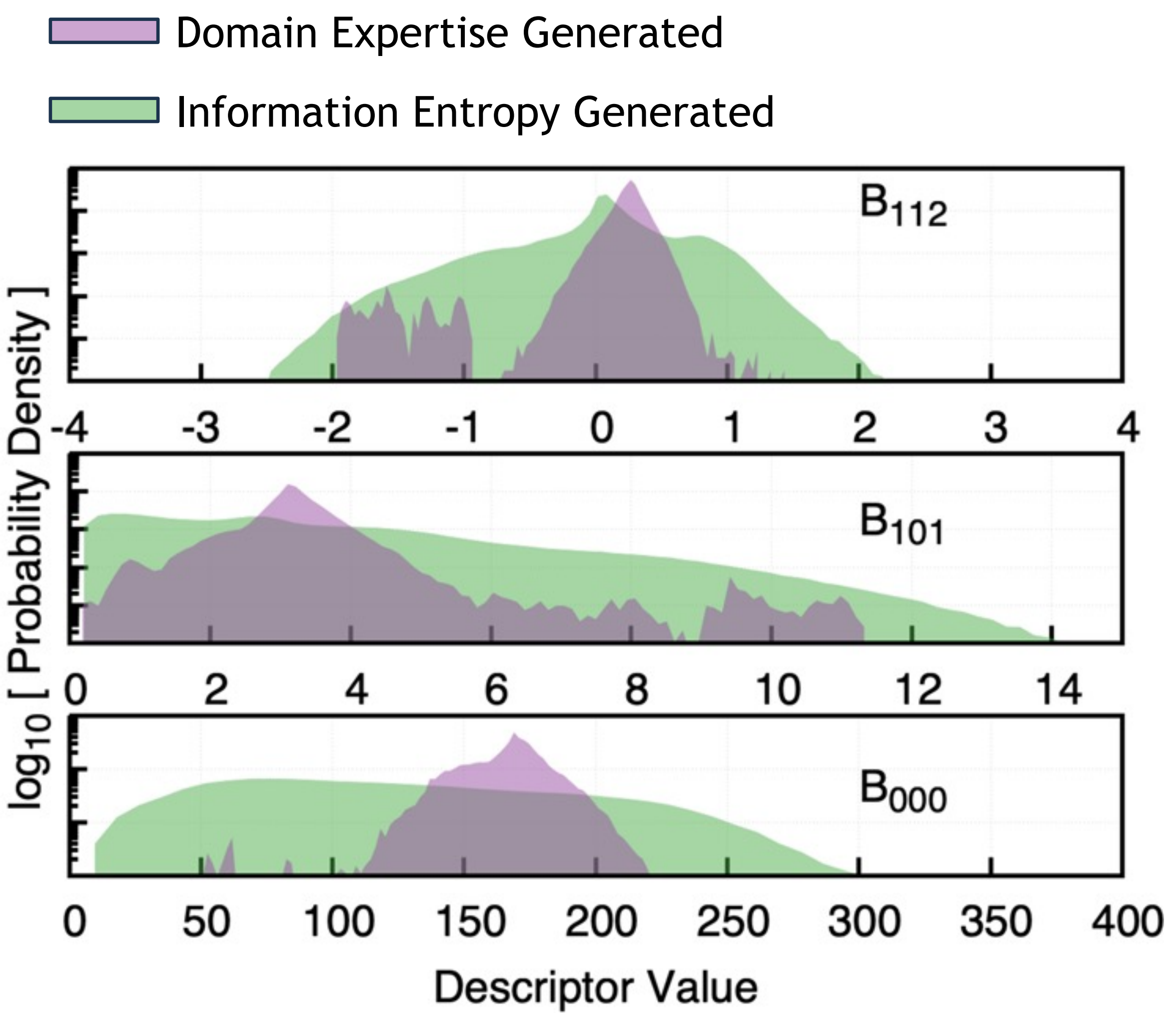
Magnetic and structural transitions captured in one MD simulation → Efficient sampling of dynamic phase diagram!

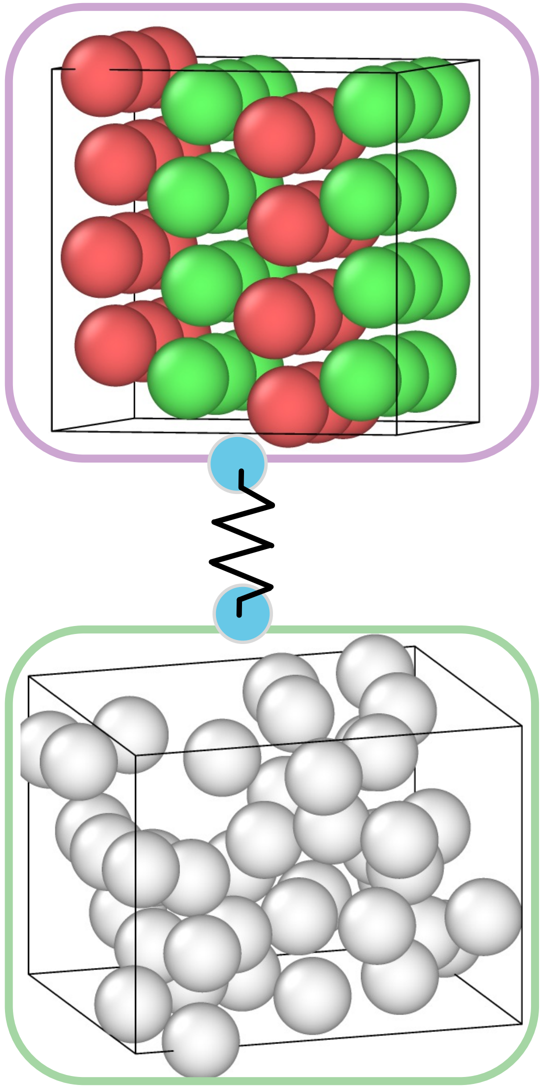




Value Added in MD From Atomic Descriptors

- Data compression easily achieved on large simulations by reducing MD trajectory into descriptor distributions

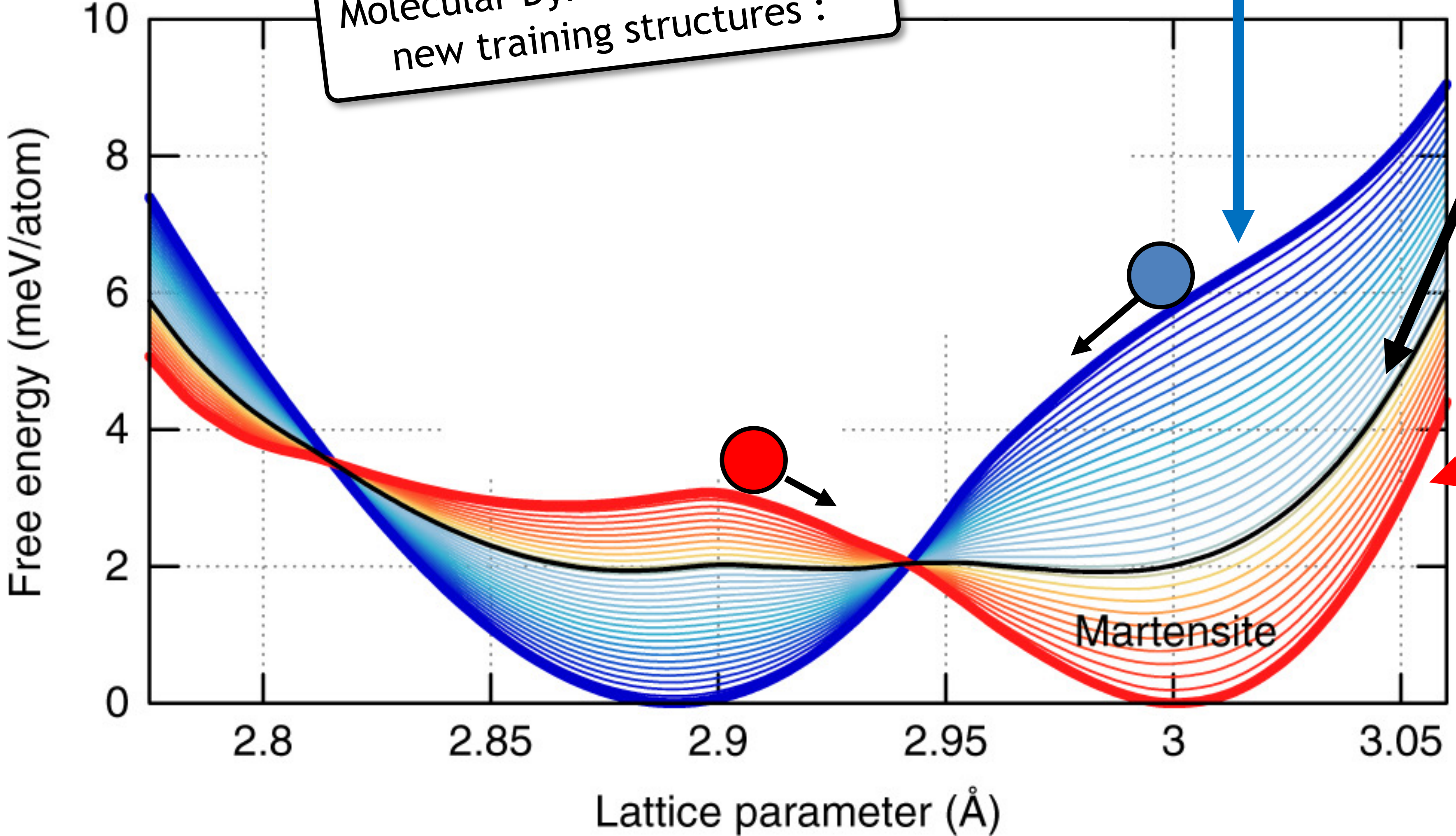




- Descriptors can be used as a scale agnostic collective variable
- Information entropy on the descriptor distribution can be used to apply a force on atoms to move toward/away known structures

Value Added in MD From Atomic Descriptors

Molecular Dynamics to sample new training structures :



Structures preferred by ***energy minimization***

Low ***free energy*** barriers to find new configurations → Easy to achieve in MD!

Structures preferred by ***entropy minimization***

**I have no idea what the entropy surface looks like*

Generalized Representative Structures

"Generalized representative structures for atomistic systems"
OPEN ACCESS
IOP Publishing
Journal of Physics: Condensed Matter
J. Phys.: Condens. Matter **37** (2025) 075901 (13pp) https://doi.org/10.1088/1361-648X/ad9791

- Each atom is represented by a vector of ~100 descriptors, any structure represented by distributions of descriptor values.

Score Target Proposed

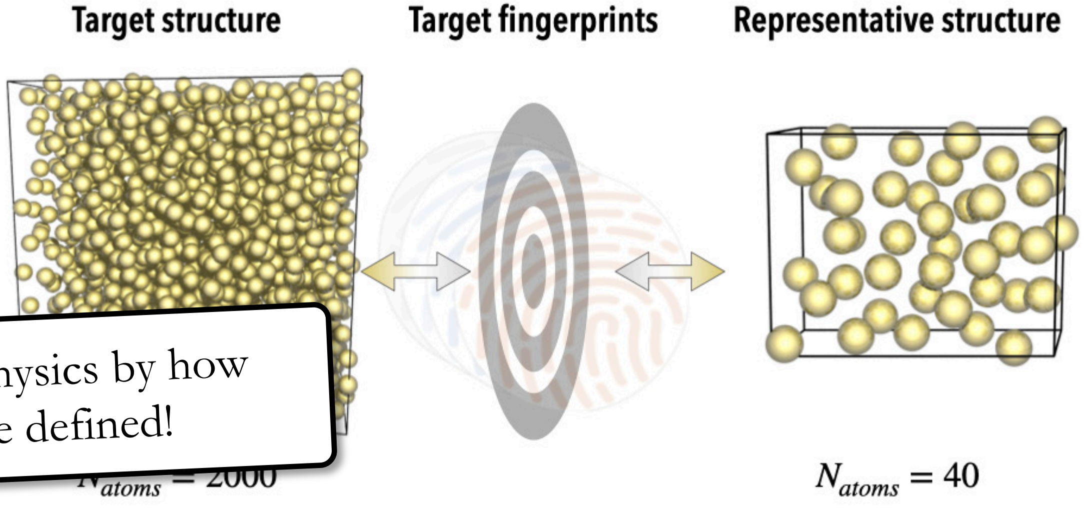
$$Q_{\text{SQS}} = \sum_{\alpha} |\bar{\Phi}_{\alpha}(\boldsymbol{\sigma}) - \bar{\Phi}_{\alpha}^{rand}| \ldots$$

- Can move between scales by simplifying th descriptors, $d^s(\{\Phi_\alpha\})$ and $d^t(\{\Phi_\alpha\})$ with their moments

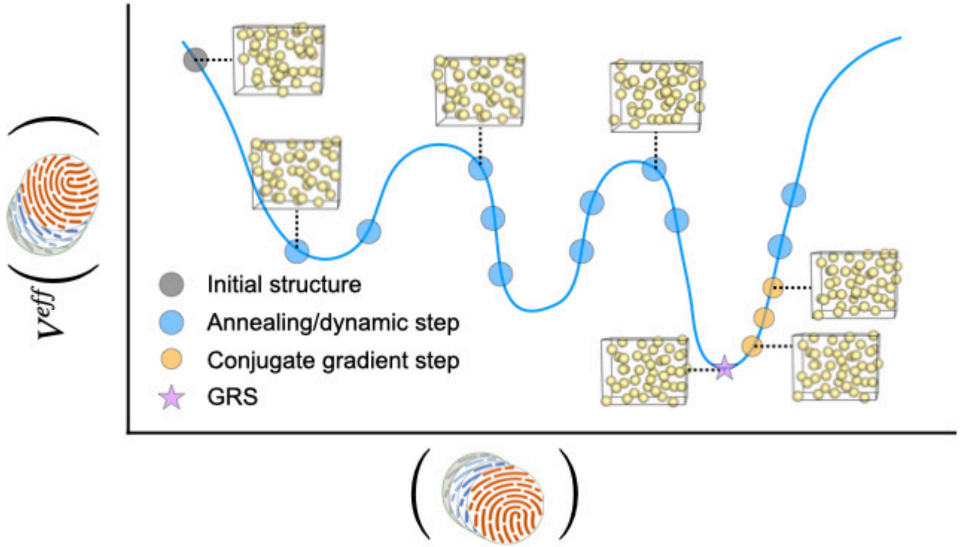
$$Q_{\text{GRS}}^{\text{intra}} = \alpha_1 \Delta\mathcal{M}_1[d^S, d^t] + \alpha_2 \Delta\mathcal{M}_2[d^S, d^t] + \cdots.$$

- Now construct an effective potential that is the difference between target and current structure, sampling is challenging.

$$V^{\text{eff}}(\{\boldsymbol{R}\}_S, \{\boldsymbol{\sigma}\}_S) = V_{\text{core}}(\boldsymbol{R}, \boldsymbol{\sigma}) + w_1 Q_{\text{GRS}}(\boldsymbol{R}_s, \boldsymbol{\sigma}_s) + w_2 Q_{\text{GRS}}^{\text{intra}}(\{\boldsymbol{R}\}_S, \{\boldsymbol{\sigma}\}_S).$$



Access to new physics by how potentials are defined!



Dynamic Interatomic Potentials

Approximate Electronic Excitations

- Even the most accurate ML-IAP fail to capture large changes in temperature due to electronic occupation changes.
- Born-Oppenheimer → Sommerfeld Potentials

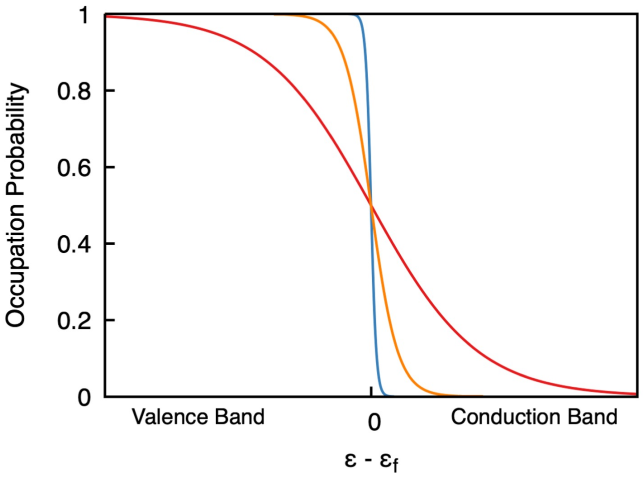
$$E = \int f_{FD}(\epsilon, T_e) D(\epsilon, \{R\}) d\epsilon \quad ; \quad f_{FD} = \frac{1}{e^{\epsilon / k_b T_e} + 1}$$

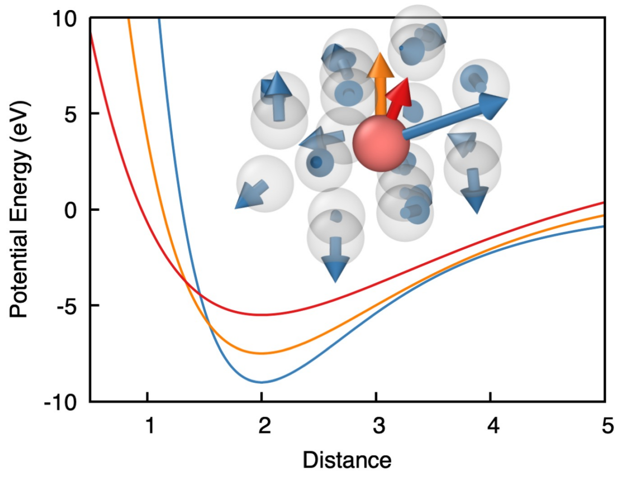
$$E = \int_{-\infty}^{\epsilon_f} \mathcal{H}(\epsilon) d\epsilon + \sum_n^{\infty} (k_b T)^{2n} a_n \frac{d^{2n-1}}{d\epsilon^{2n-1}} \mathcal{H}(\epsilon_f)$$

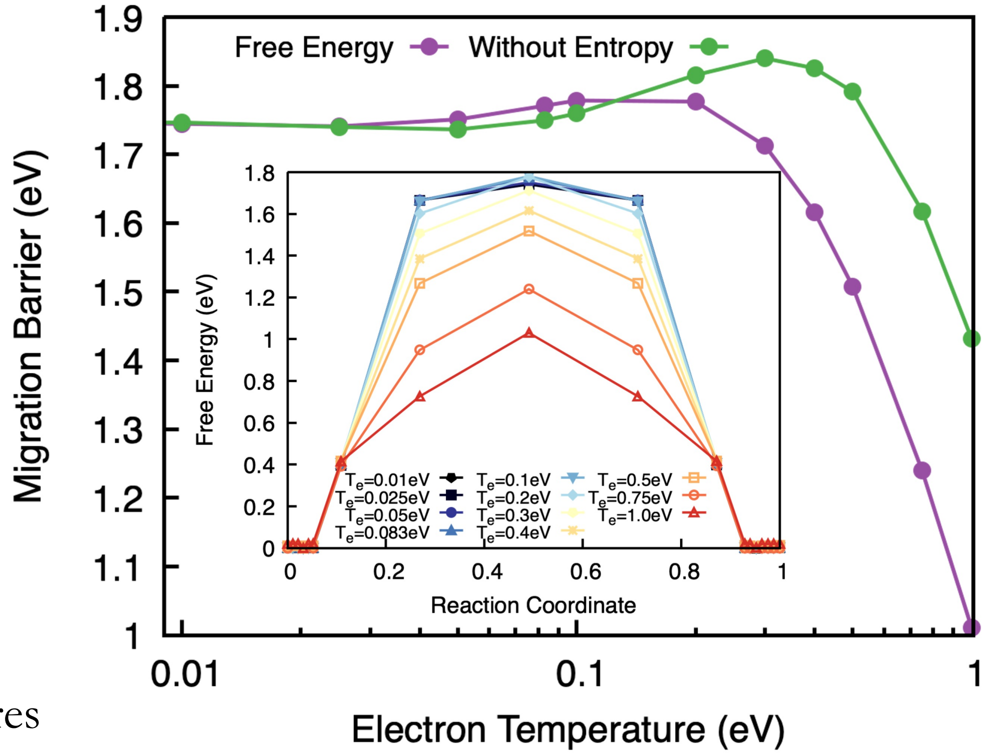
$$\mathcal{U}_{Som}(R_i) = \sum_j \mathcal{U}_{BO}(R_j, R_i) + T_e^2 \sum_j g_{\epsilon_f}(|R_i - R_j|)$$

Moment expansion for electron entropy can be captured with simple adaptations of ML-IAP

Pictorial PES Distortion by T_e:





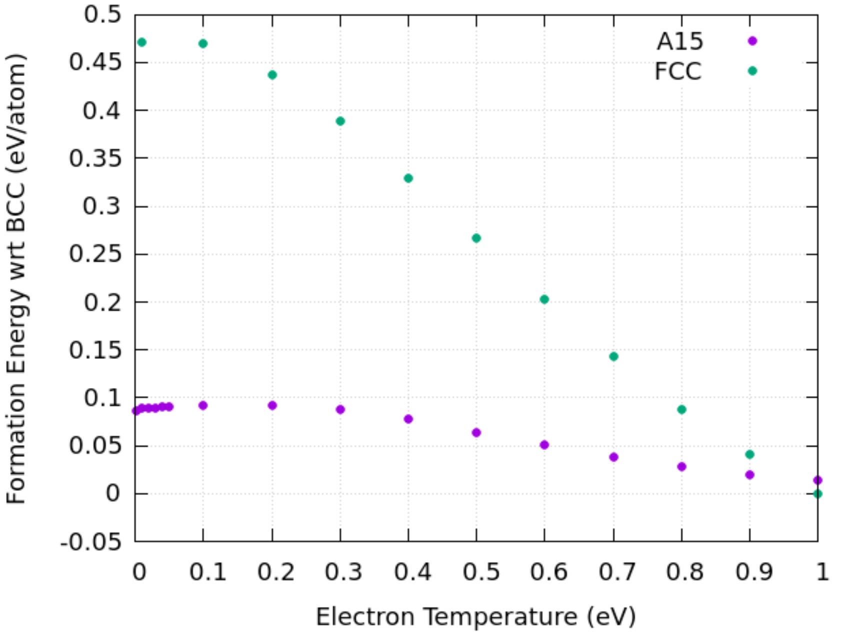


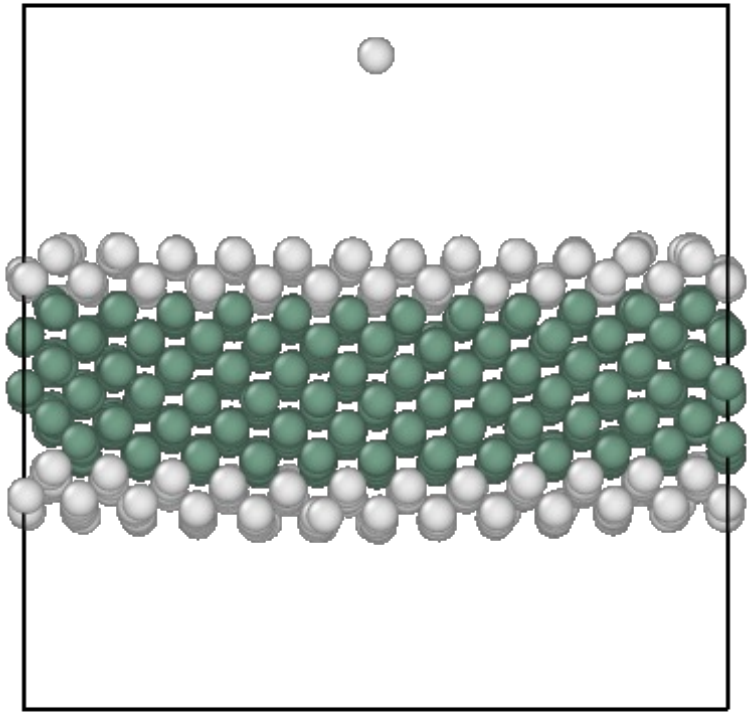
(Right) Vacancy migration barrier in Tungsten at elevated Temperatures

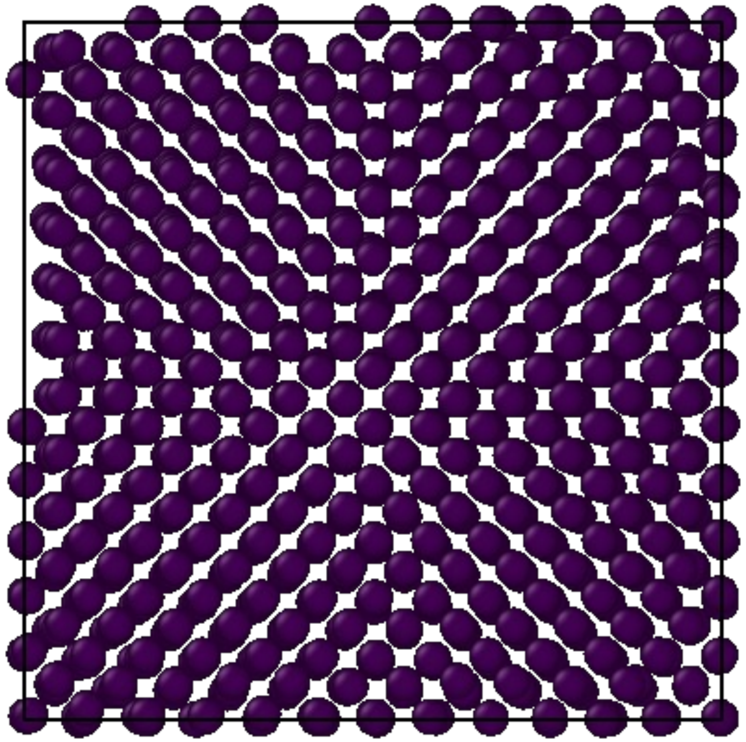
In Practice Using LAMMPS

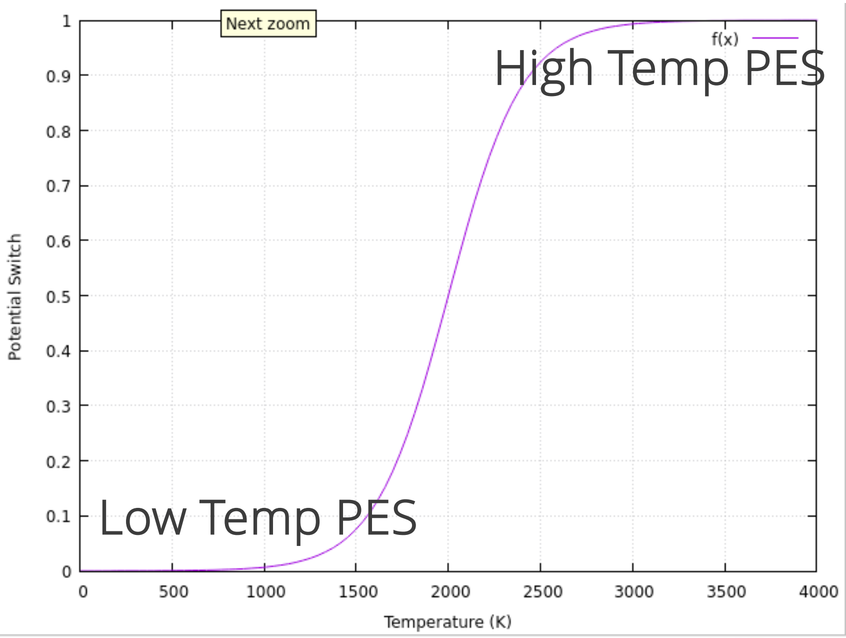
- Partition your training into two bins: High and Low Temperature

```
compute      lavg all ave/sphere/atom cutoff ${ravg}
variable        psratio1 atom 1/(1+exp(-(c_lavg[2]-v_tdamp)/v_twidth)) #High T surface
variable        psratio2 atom 1-v_psratio1                             #Low T surface
pair_style       hybrid/scaled v_psratio2 eam/fs v_psratio1 pace product
pair_coeff       * * eam/fs ../2013Marinica_W_EAM2.fs W
pair_coeff       * * pace W_pot.yace W
```



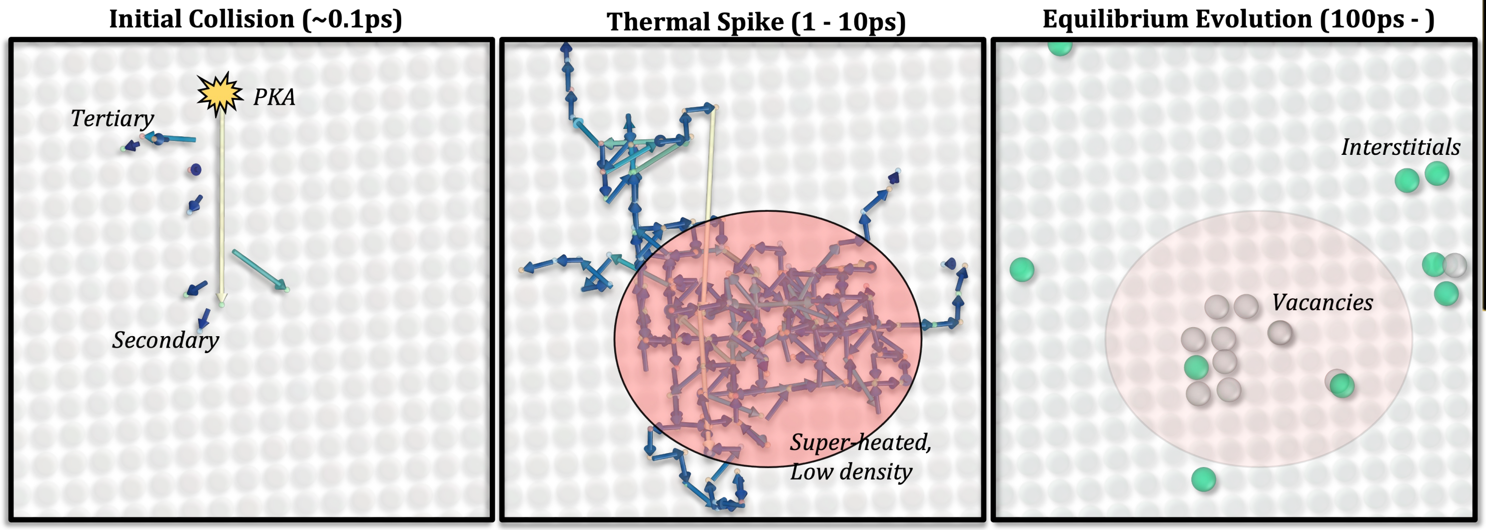


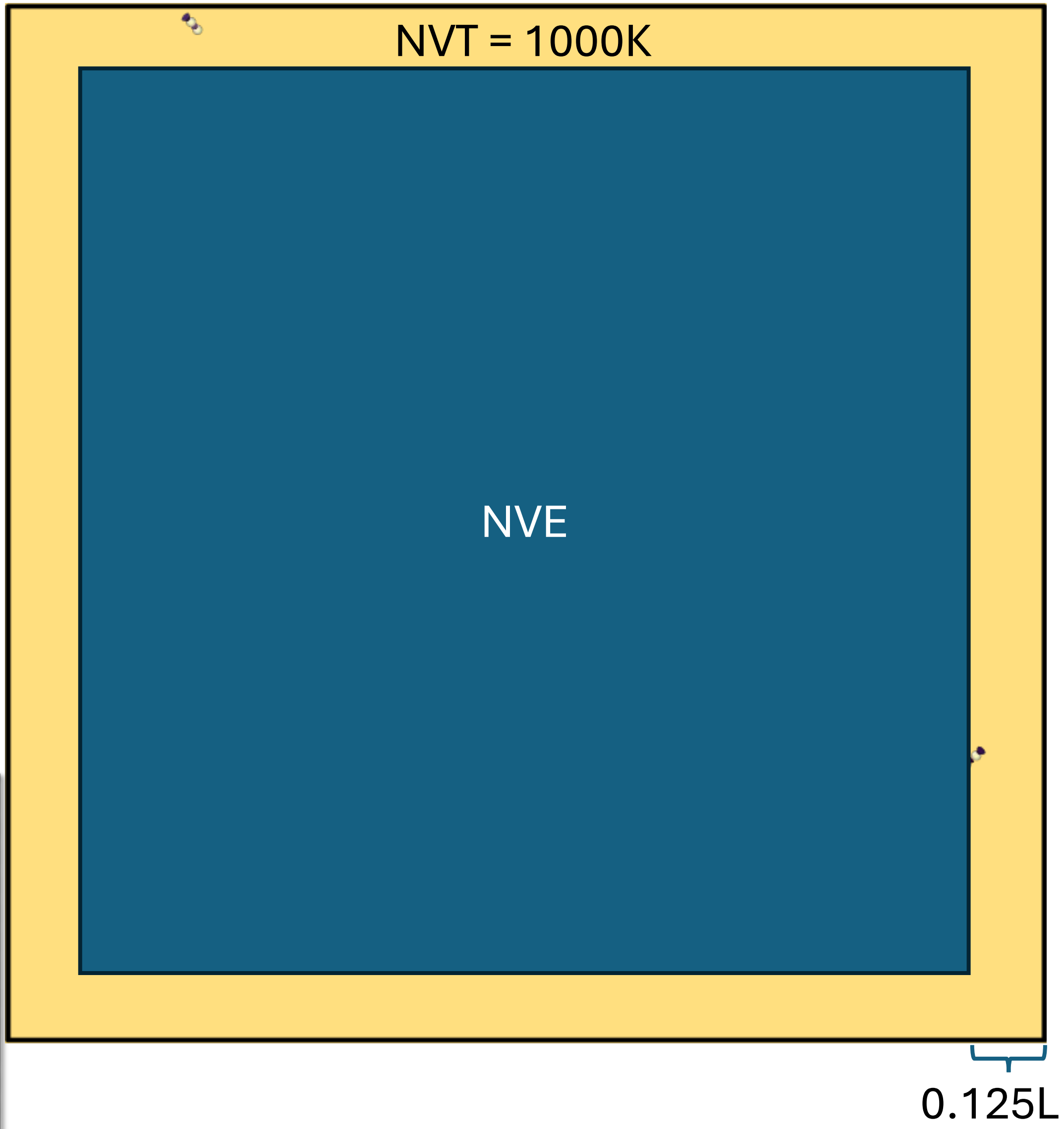




Use Case for Radiation Damage

- During a neutron collision with the lattice, very high energy density states are generated
- Local temperature/density of atoms will determine which PES we sample from
- PKA = Primary Knock-on Atom





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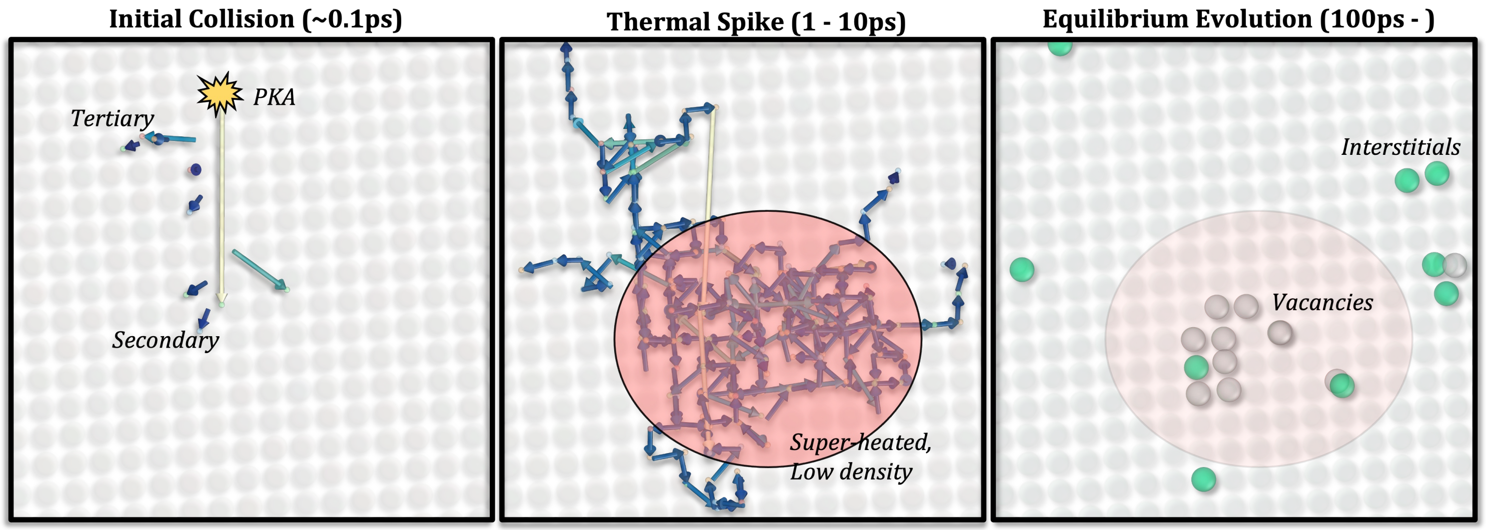
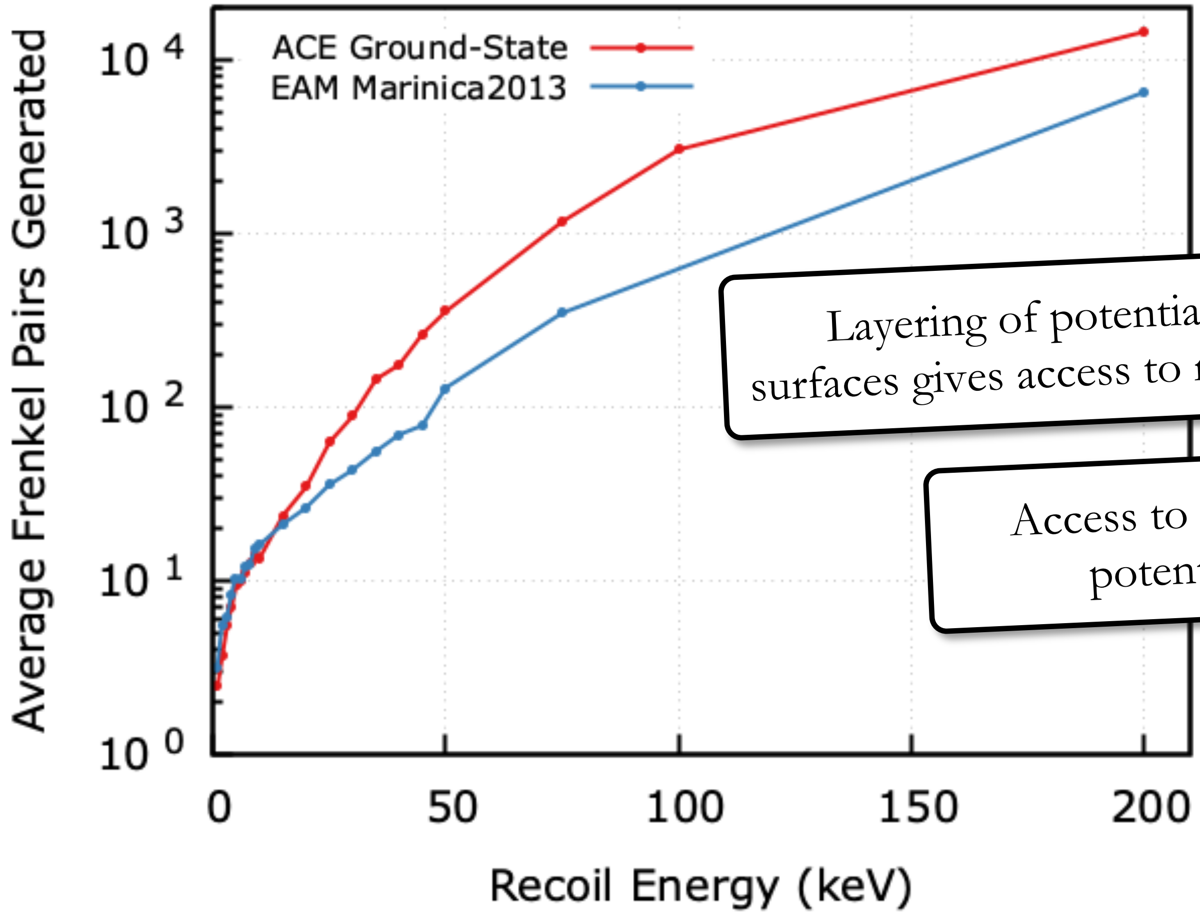


Use Case for Radiation Damage



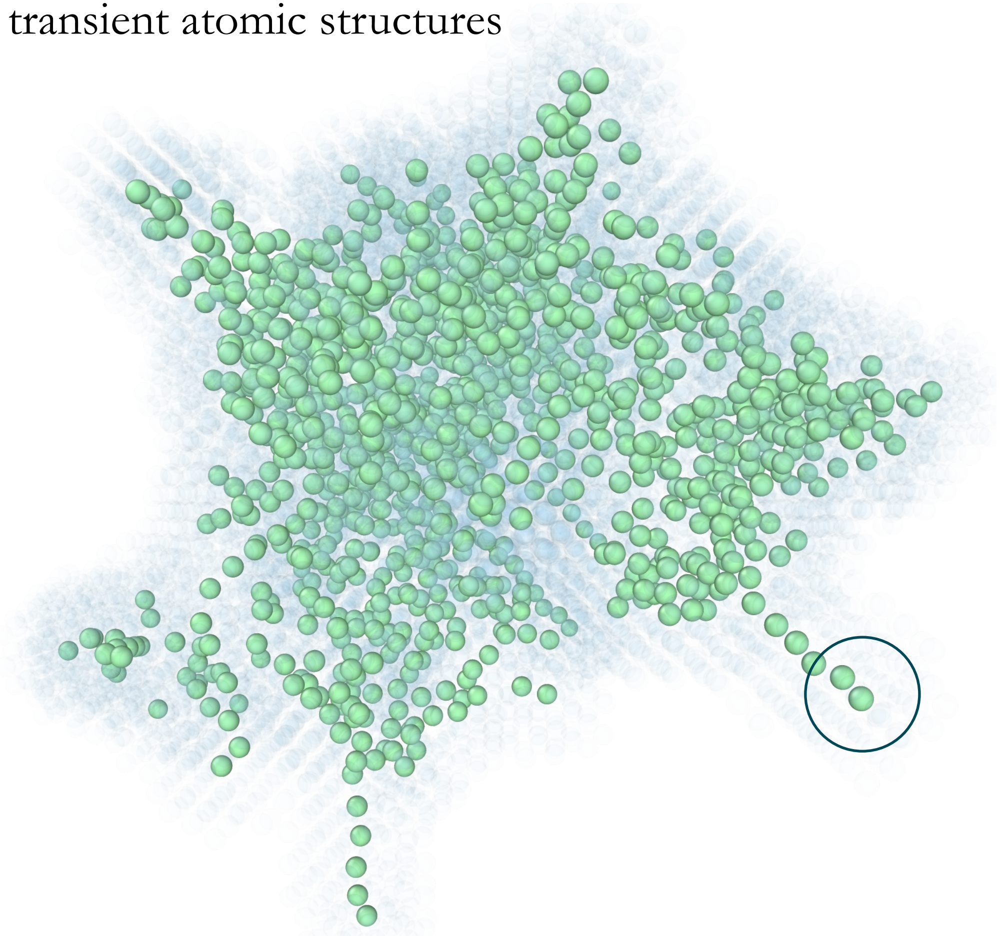
Layering of potential energy surfaces gives access to new physics!

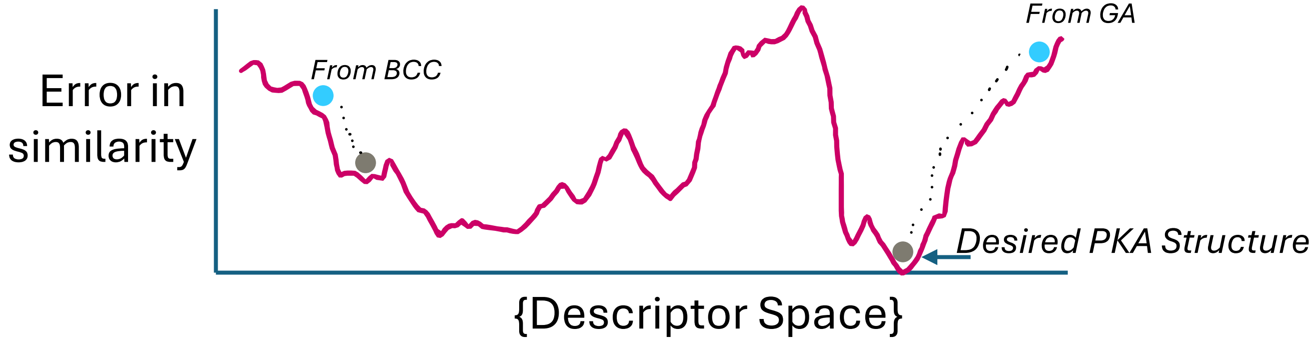
Access to new physics by how potentials are defined!

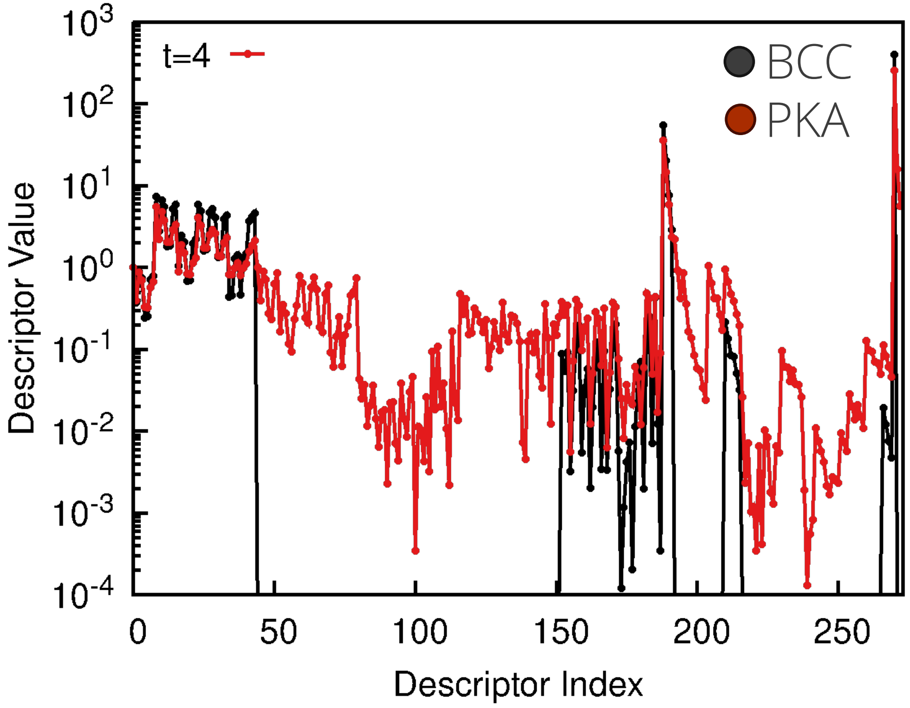
ACE PES T=0.0eV,100keV PKA
7.3E-4 DPA before relax
0.52ps Elapsed, adaptive timestep interval

Application to Radiation Damage in Materials

- ML-IAP are being refined toward accurate displacement damage by curating training to transient atomic structures

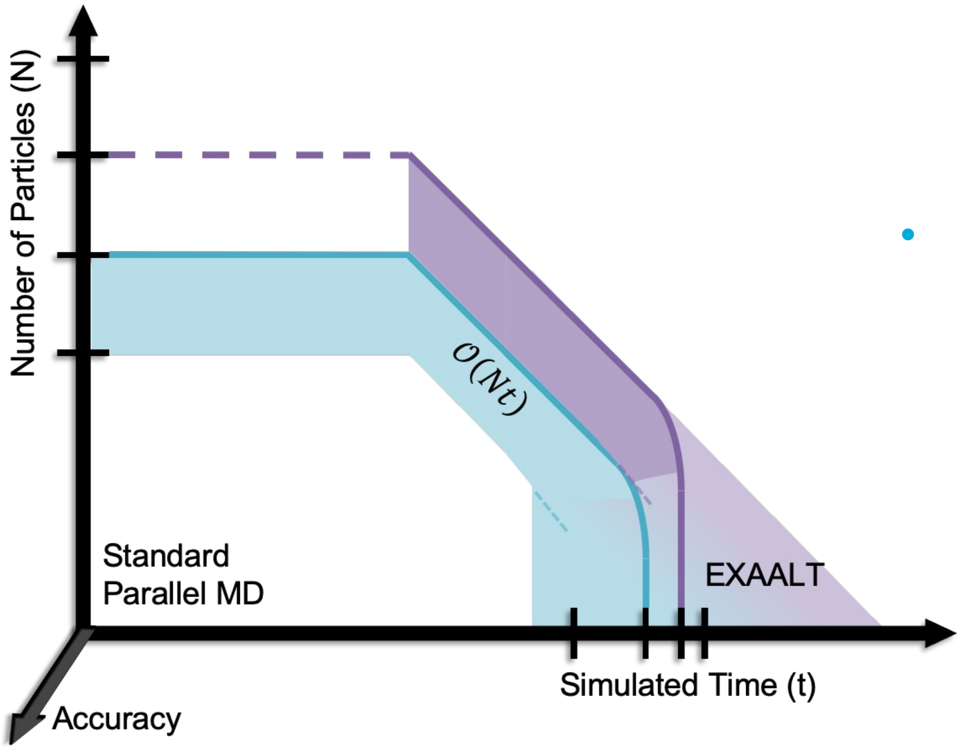






Conclusion and Path Forward

- Data-driven interatomic potentials allow for MD predictions of challenging material problems.
- While harder to quantify, the fidelity of our MD simulations needs to be a key consideration



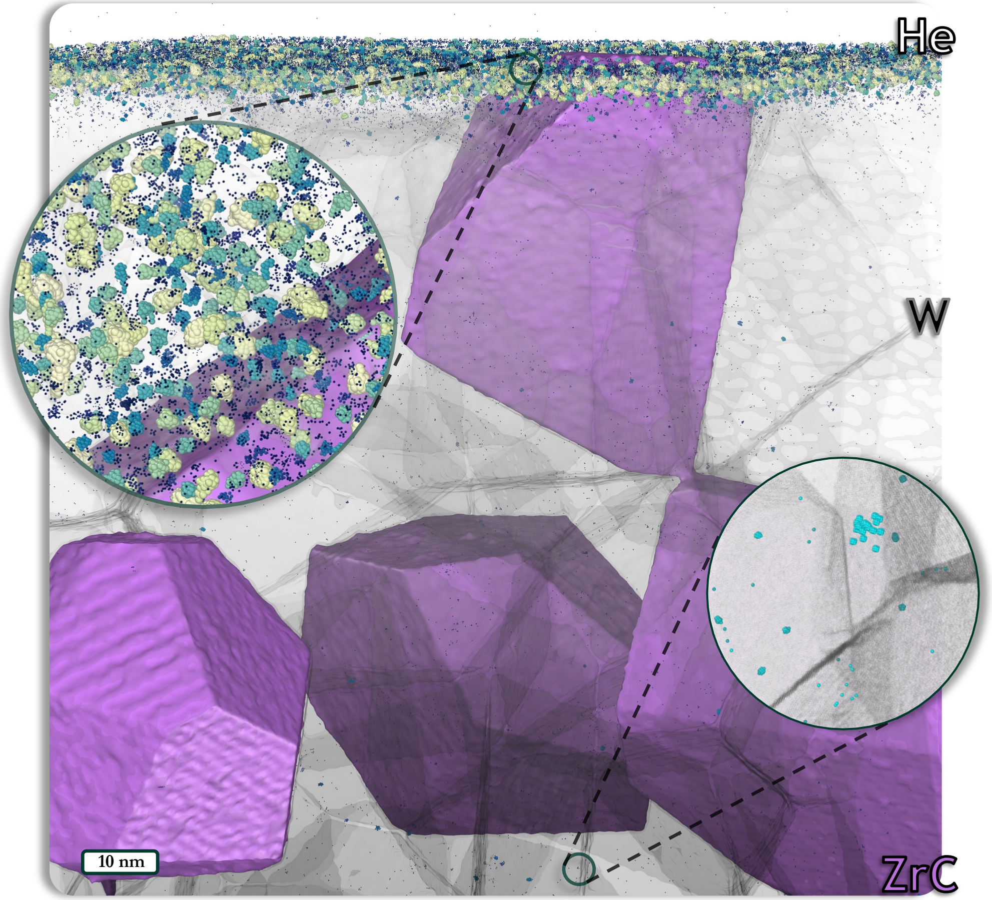
- Thank you to all my collaborators: Aidan Thompson, Mary Alice Cusentino, Krupa Ramasesha, Svetoslav Nikolov, Drew Rohskopf, Charlie Sievers, David Montes do Oca Zapian, Danny Perez, Nick Lubbers, Julien Tranchida, Steve Plimpton, Ivan Oleynik, Jon Willman, Ember Sikorski, Megan McCarthy, James Goff, James Stewart, Carlos Pereyra, Nat Trask, Michael Sakano, and many others!

Contact Information:

mitwood@sandia.gov

Conclusion and Path Forward

- While harder to quantify, the fidelity of our ML-IAP training sets needs to be a key consideration



"Generalized representative structures for atomistic systems"

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Journal of Physics: Condensed Matter
J. Phys.: Condens. Matter **37** (2025) 075901 (13pp) https://doi.org/10.1088/1361-648X/ad9791

"Permutation-adapted complete and independent basis for atomic cluster expansion descriptors"

Journal of Computational Physics
Volume 510, 1 August 2024, 113073

FitSNAP / **structure-generation**

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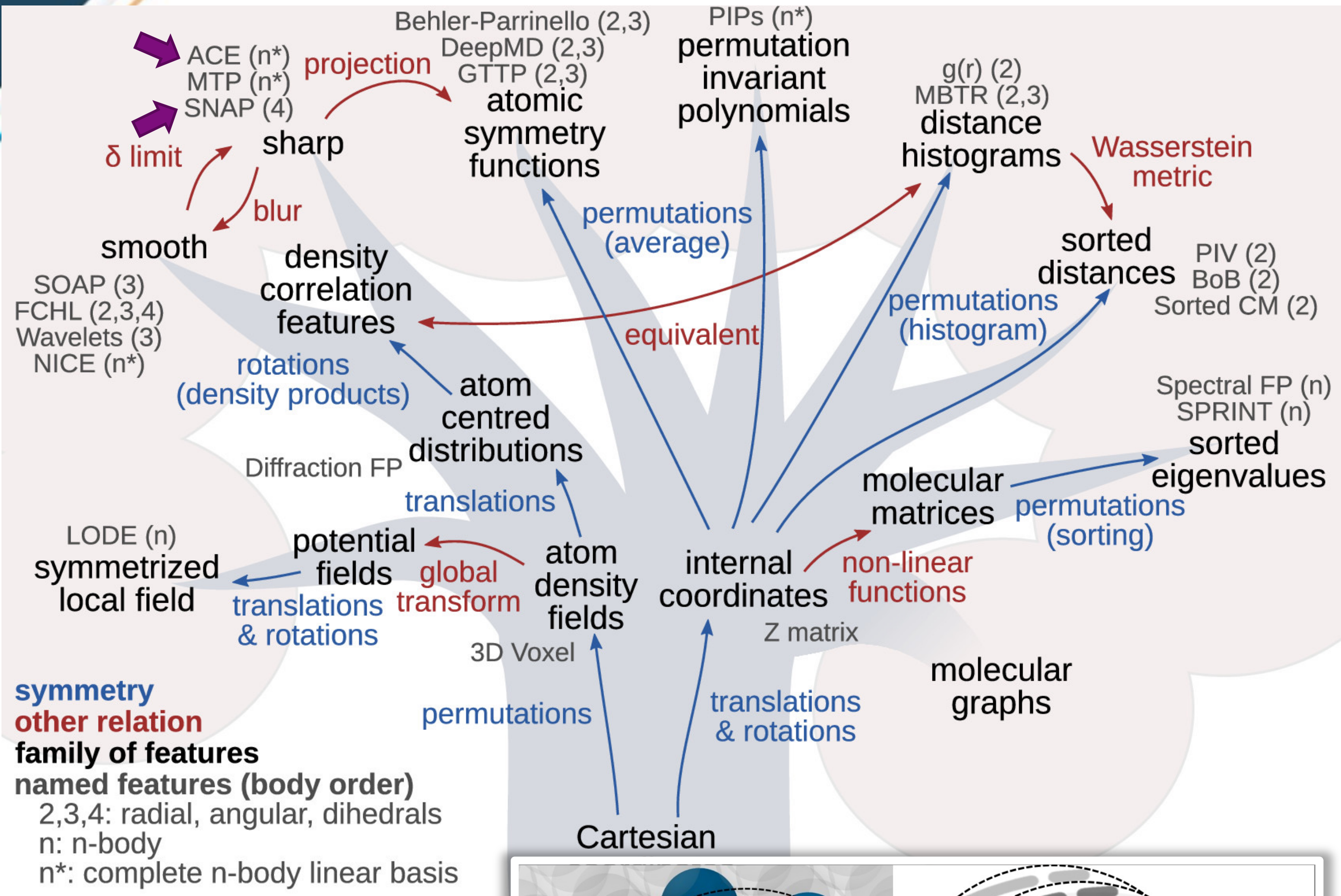
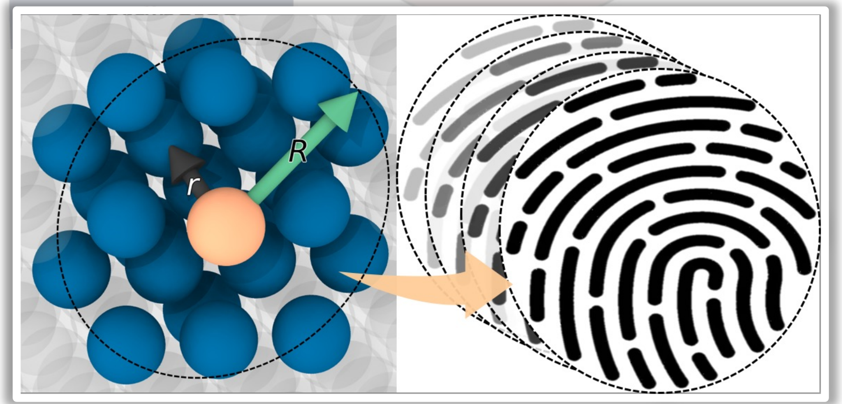
U.S. DEPARTMENT OF ENERGY | Office of Science

Descriptor Sets

"Permutation-adapted complete and independent basis for atomic cluster expansion descriptors"
Journal of Computational Physics
Volume 510, 1 August 2024, 113073



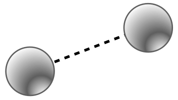


Musil *et. al.* Chem. Rev. (2021) 121, 9759–9815

Complete, generalizable single-bond basis

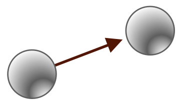
Spatial

$$\phi_{inlm} = R_n(r_{ij})Y_l^m(\widehat{r_{ij}})$$



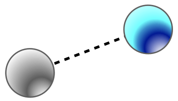
Charge (transfer)

$$T_k(q_{ij})Y_l'^m(\widehat{\delta q_{ij}})$$



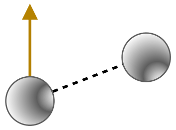
Chemical (bonds)

$$T_i(\mu_{ij})$$

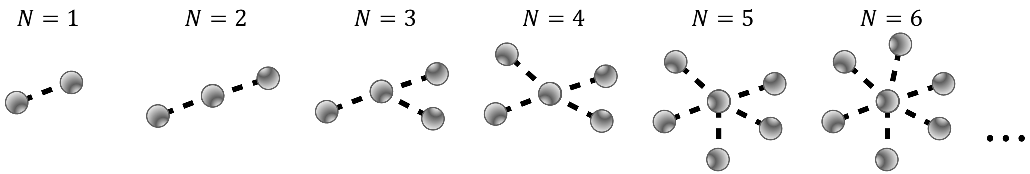


Magnetism

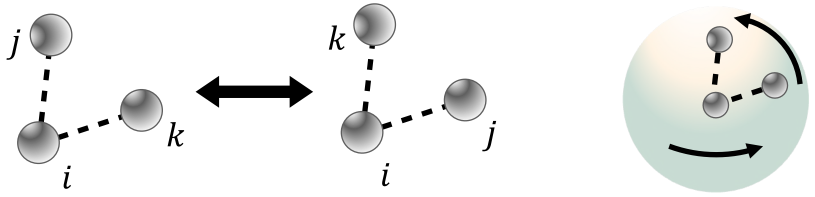
$$T_k(M_i)Y_l''^m(\widehat{M_i})$$



Form a complete N-bond basis

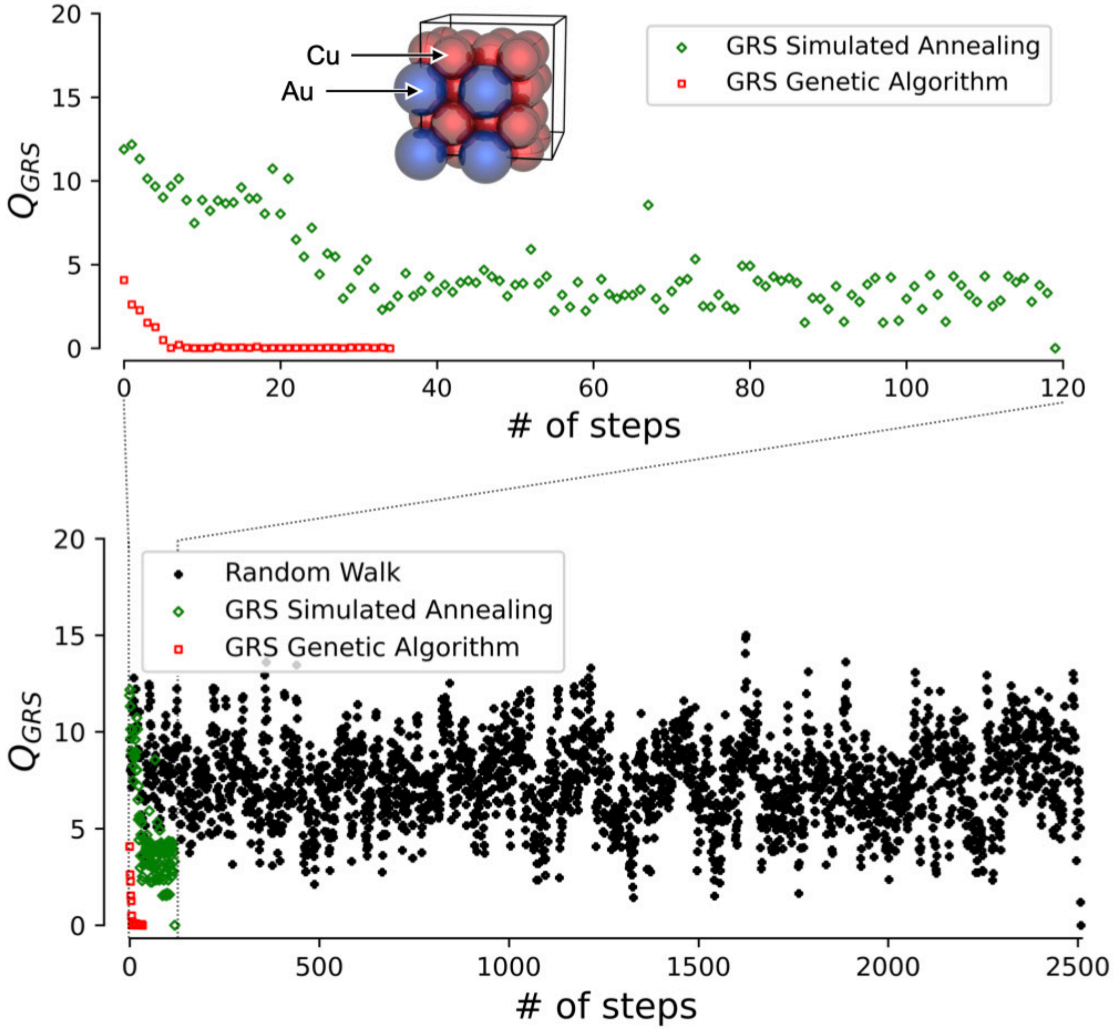


Impose invariance w.r.t. rotations and permutations

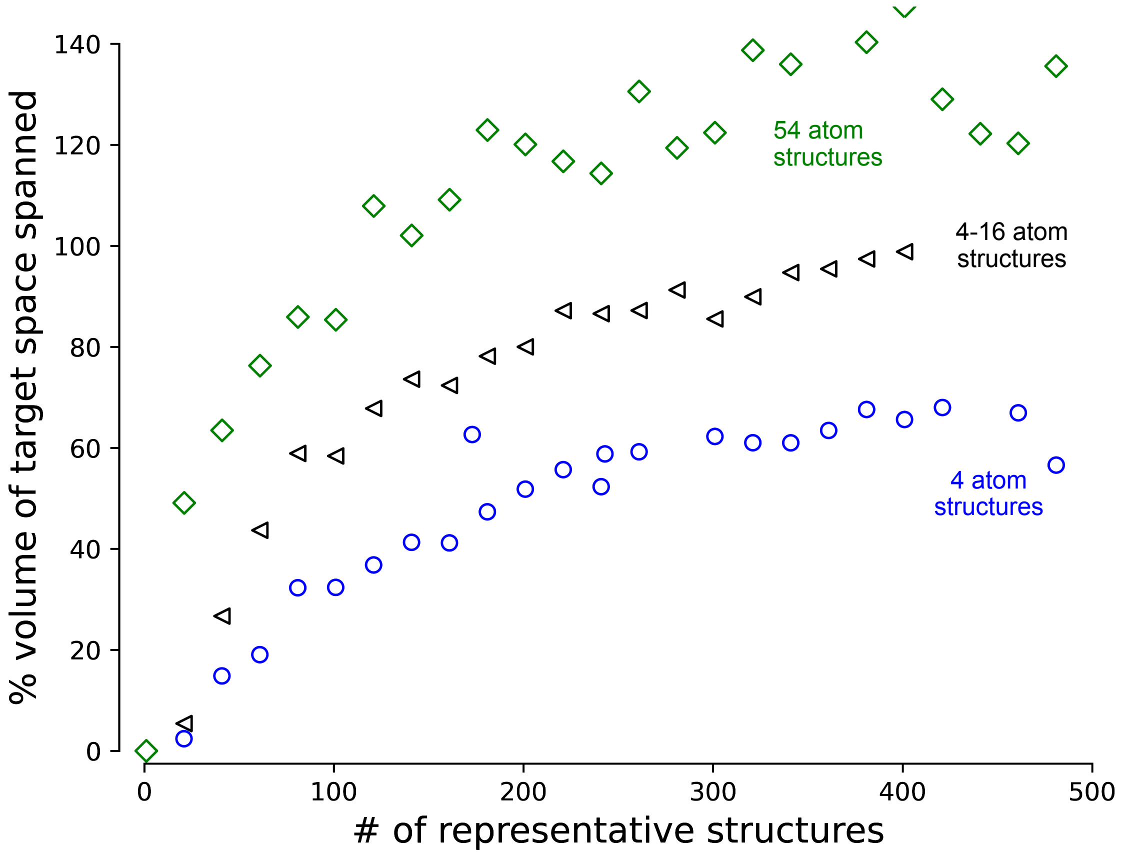


Sampling the Loss Function Potential

"Generalized representative structures for atomistic systems"
OPEN ACCESS
IOP Publishing
Journal of Physics: Condensed Matter
J. Phys.: Condens. Matter 37 (2025) 075901 (13pp) https://doi.org/10.1088/1361-648X/ad9791



- More representation in larger structures, batches of structures.



Representing Complex, Heterogeneous Structures

- Descriptor basis is chemically informed, making complex chemistry and structural disorder capable of miniaturizing.