SAND2022-13532C

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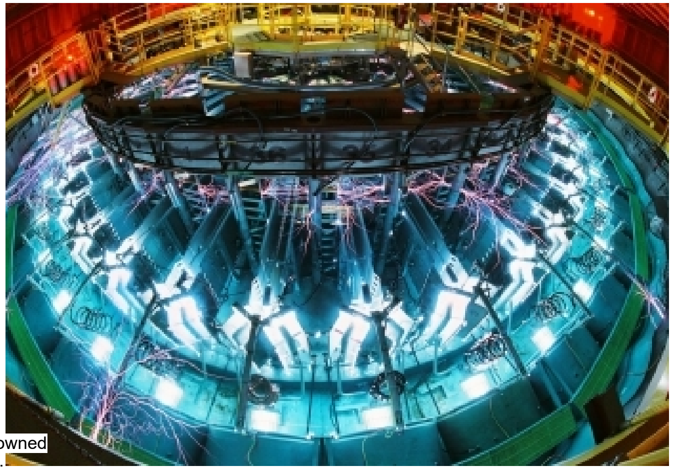
Exploring the behavior of MoNbTaTi refractory CCAs across composition space using a machine-learned interatomic potential

Megan McCarthy[a], Jacob Startt[b], Remi Dingreville[b], Aidan Thompson[a], Mitchell Wood[a]

a Center for Computing Research, Sandia National Laboratories – New Mexico
b Center for Integrated Nanotechnologies, Sandia National Laboratories – New Mexico

Multiscale Materials Modeling (MMM10)

October 6th, 2022



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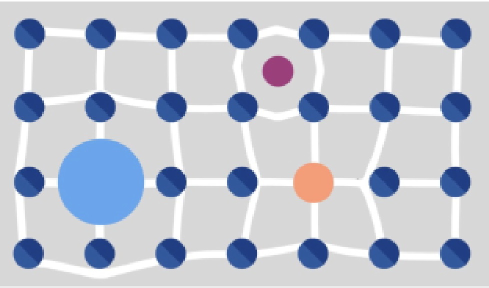
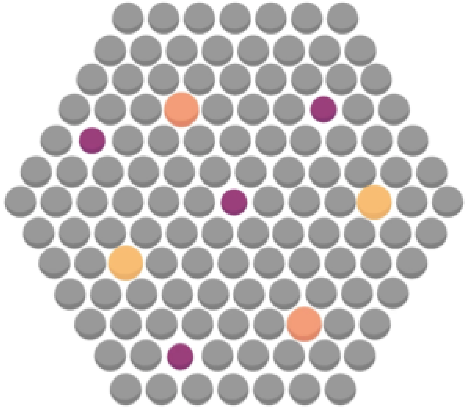
Complex concentrated alloys

Journal of MATERIALS RESEARCH



https://doi.org/10.21203/rs.2.15081/v1



Conventional alloy

High-entropy alloy

CCA: Alloy with high concentrations of 3 or more elements that can coexist without extreme phase separation

AKA: high-entropy alloy (HEA), multi-principal element alloy (MPEA)

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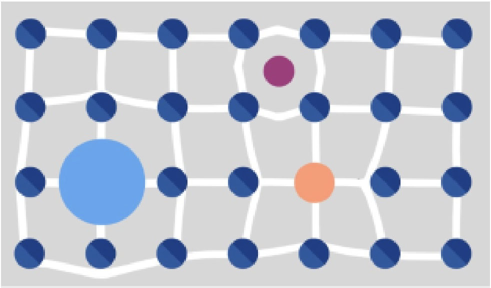

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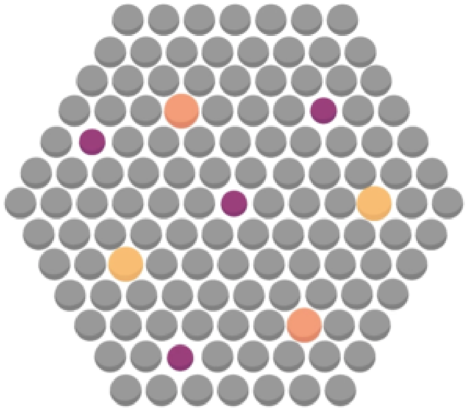

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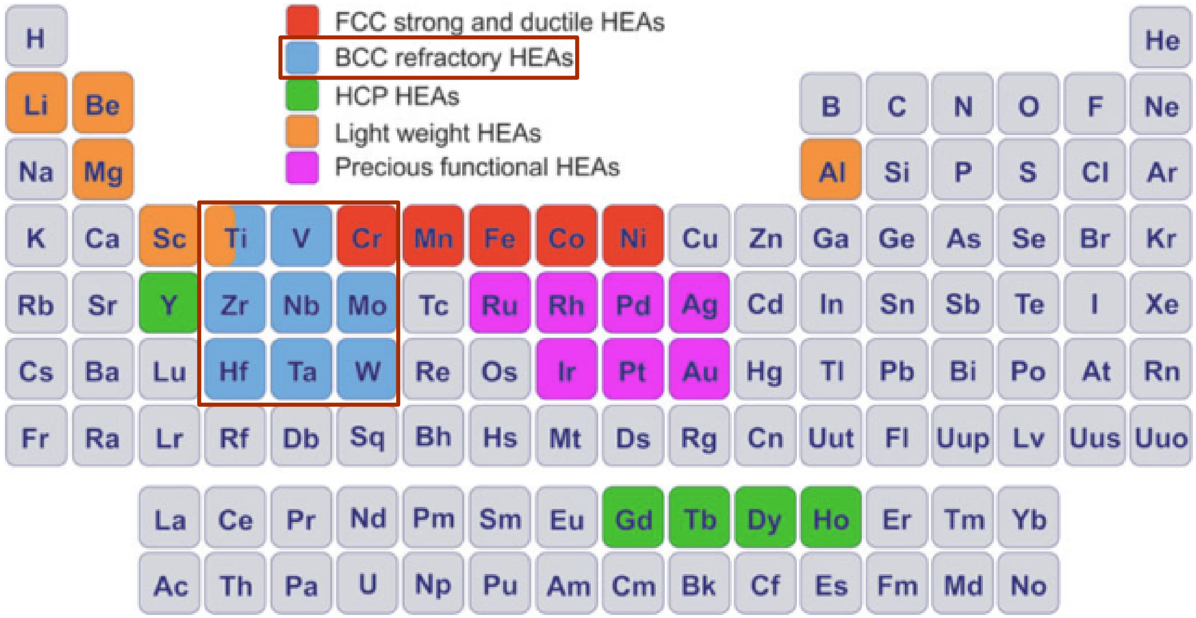

Li et al., J. Mater. Res., Vol. **33**, (2018)

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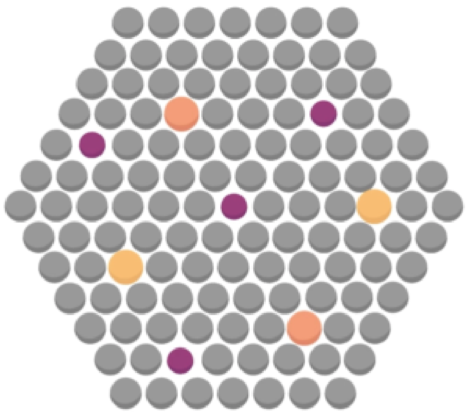

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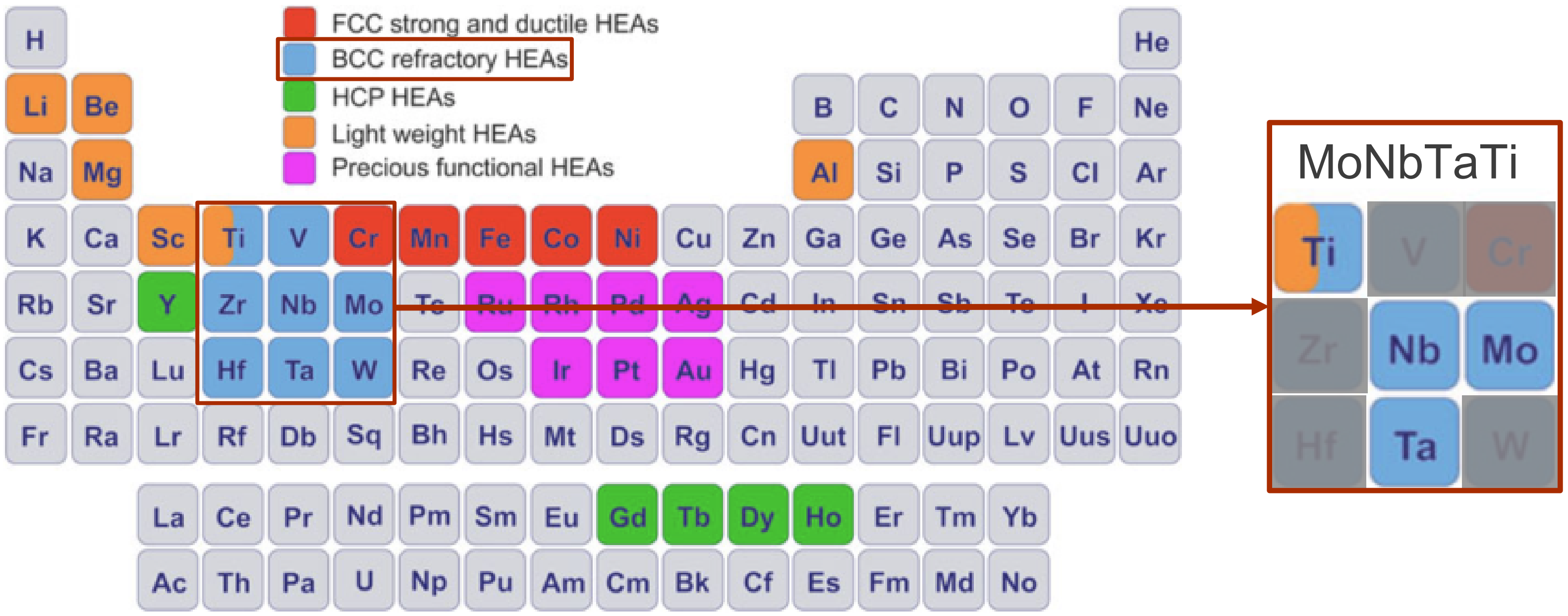

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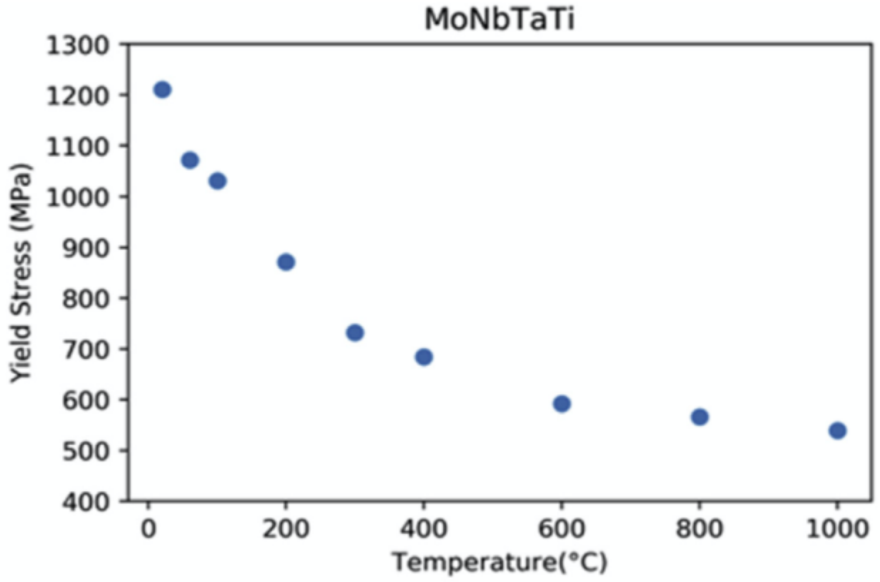

Coury et al., Acta Mat. ***175*** *(2019)*

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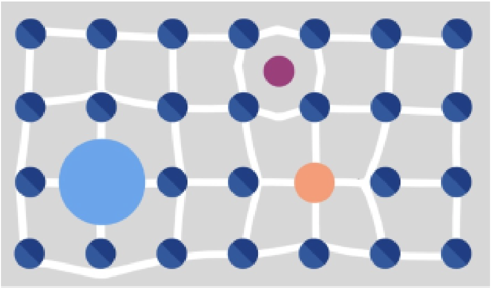

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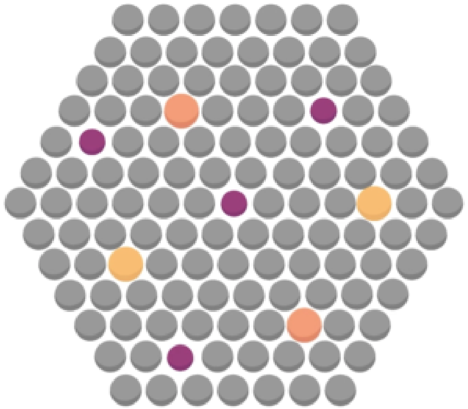

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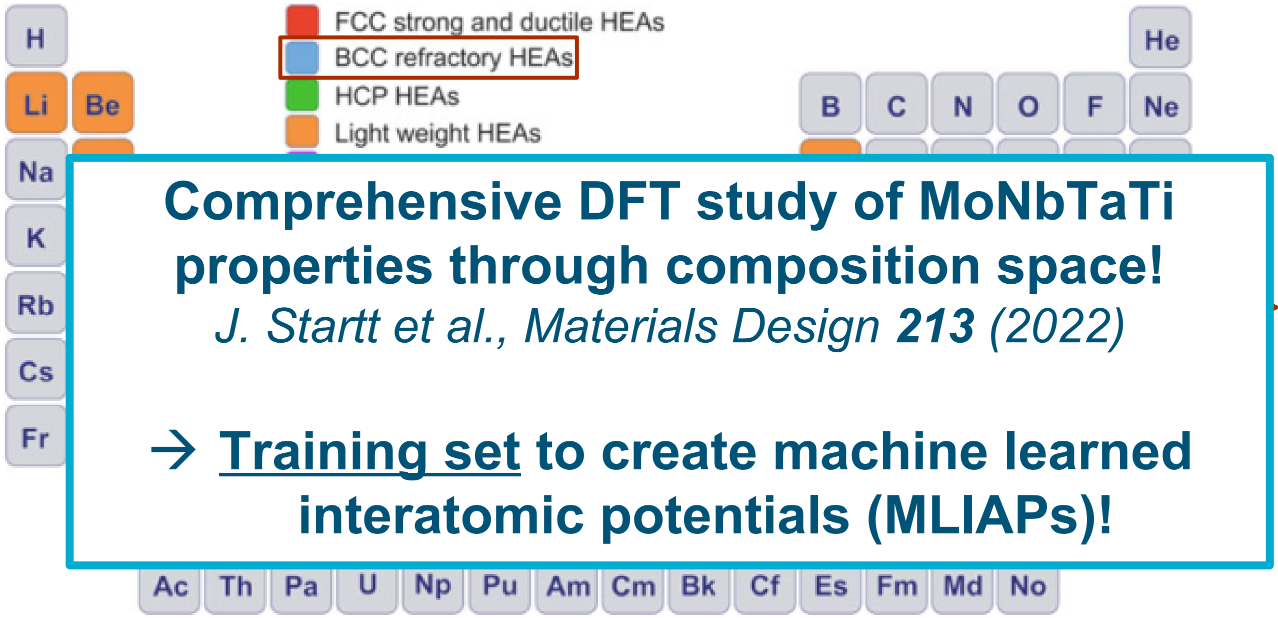
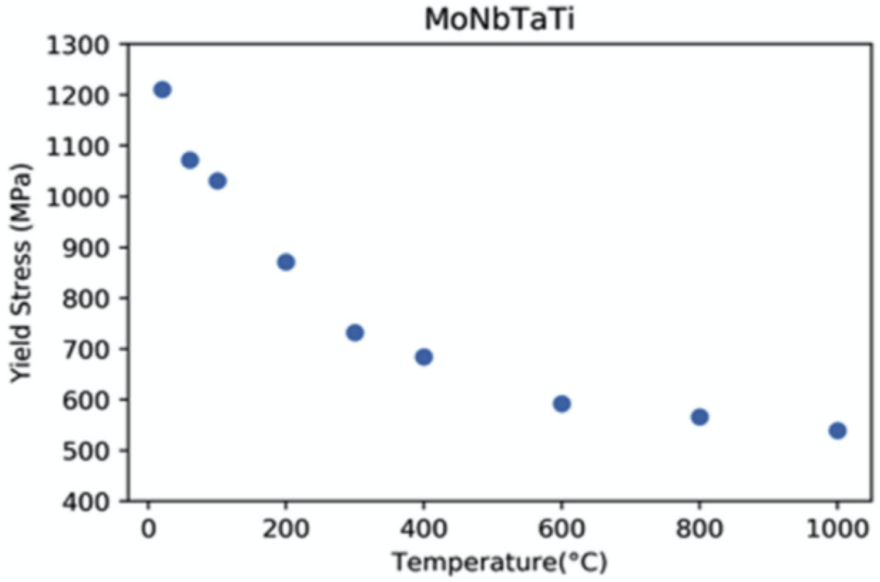


Comprehensive DFT study of MoNbTaTi properties through composition space!

*J. Startt et al., Materials Design **213** (2022)*

→ <u>Training set</u> to create machine learned interatomic potentials (MLIAPs)!

*Li et al., J. Mater. Res., Vol. **33**, (2018)*



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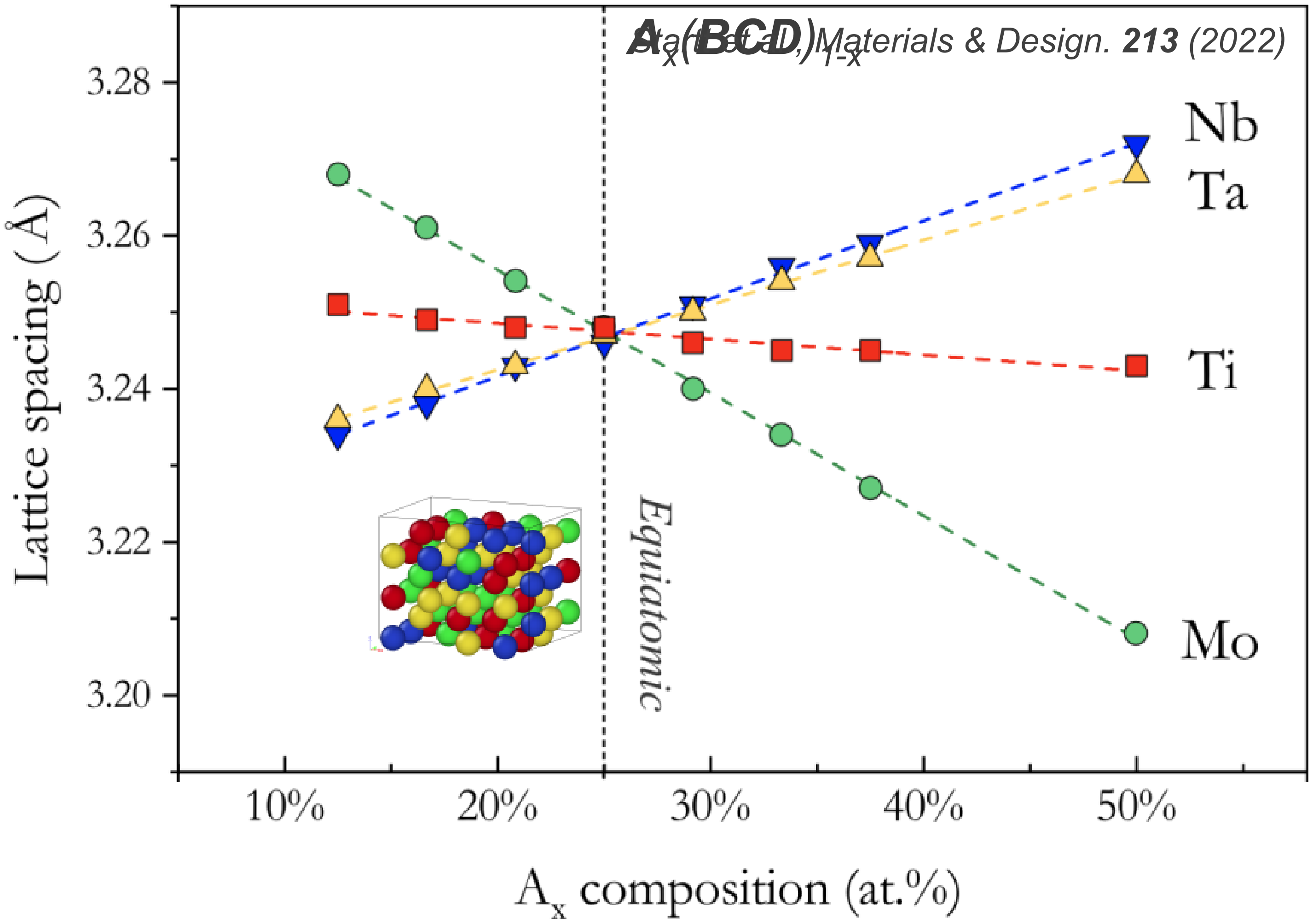
Exploring MoNbTaTi composition space - DFT

*Vary element **A**, hold elements **B, C, and D** constant*

$A_x(BCD)_{1-x}$

*Startt et al., Materials & Design. **213** (2022)*



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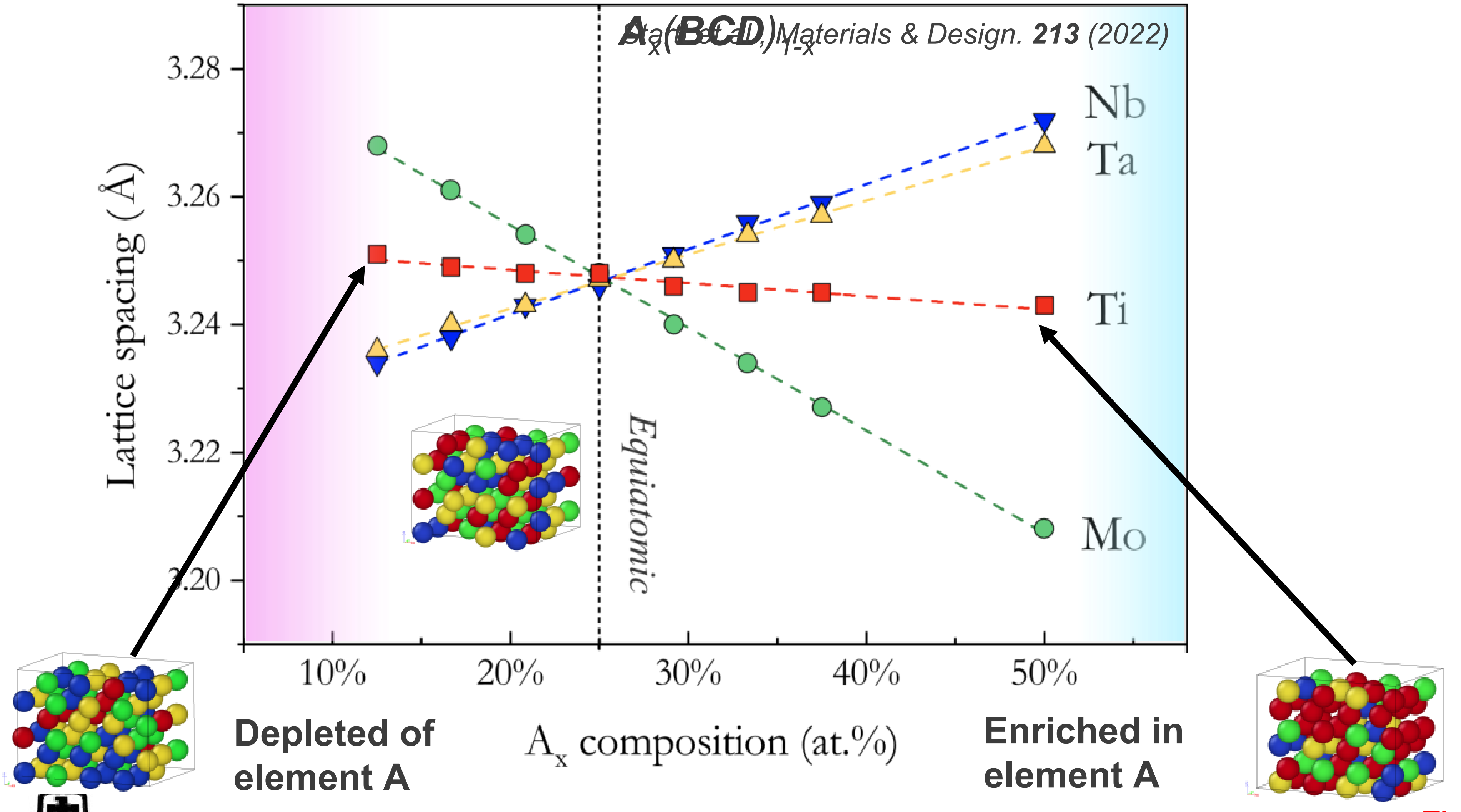
*Stan et al., Materials & Design. **213** (2022)*



Depleted of element A

Enriched in element A

$Mo_{29.1}Nb_{29.1}Ta_{29.1}Ti_{12.5}$

$Mo_{16.7}Nb_{16.7}Ta_{16.7}Ti_{50.0}$

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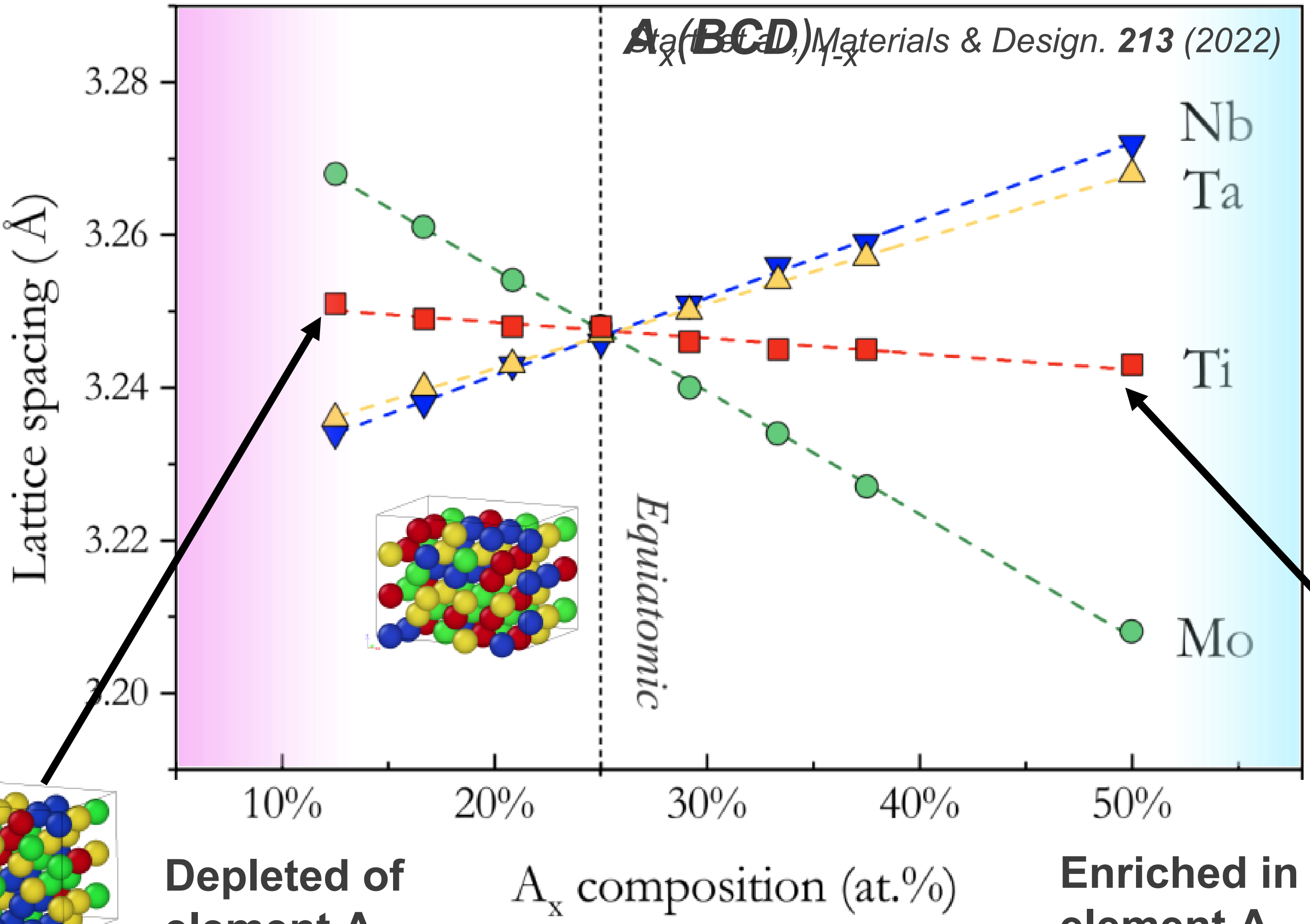
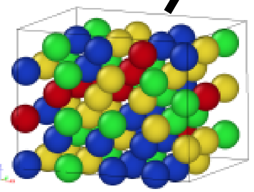


Structures	CCA (per composition)	Pure (per element)
Elastic strain	~ 1.5K - 2K	~ 100
AIMD, solid	~ 2K – 3K	~ 4K
AIMD, liquid	~ 1K – 2K	~ 3K
Volumetric strain	~10 – 20	~ 25
Uniaxial strain	~15 – 20	-
Surfaces	~ 10 – 20	~ 50 – 75



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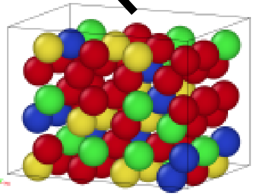

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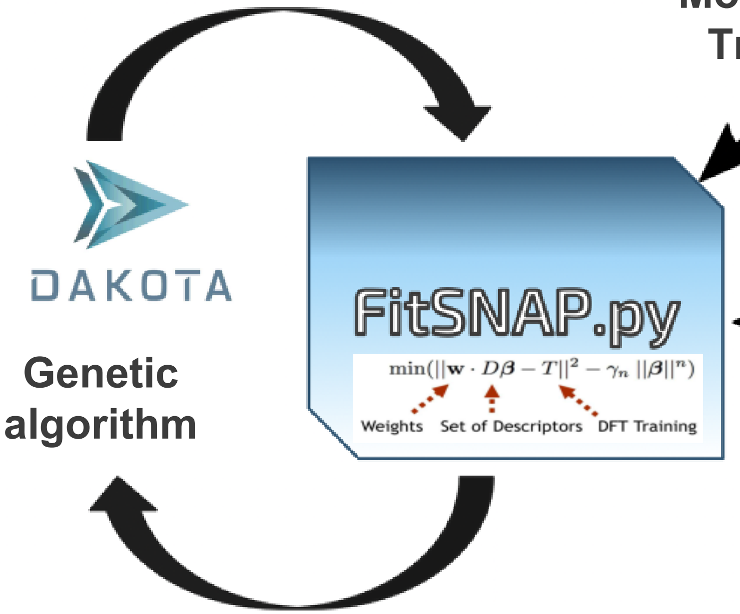
MLIAP training scheme

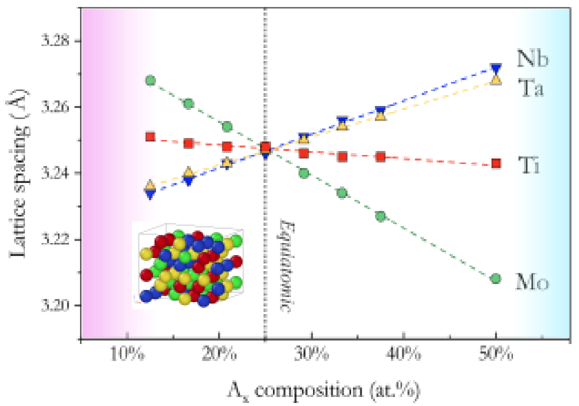
optimize
hyper-parameters



Genetic algorithm

Outputs MLIAP:
Spectral Neighbor Analysis Potential (SNAP)

MoNbTaTi DFT Training set



Calculates **bispectrum** descriptors using **spectral neighbor analysis**

$$E^i_{SNAP} = \beta_0 + \sum_{k=1}^{K} \beta_k (B^i_k - B^i_{k0})$$

MLIAP training scheme

Goal 1: Understand ‘yield’ of rich composition-varied training set
(no additional structures)

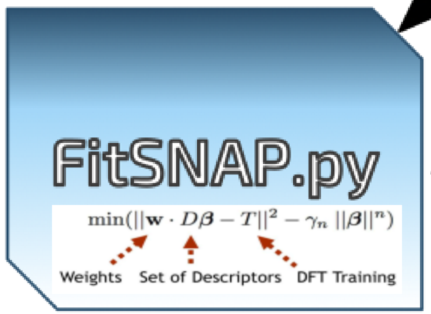
→ MLIAP are trained to match **trends in <u>elastic properties</u> with changes in composition**

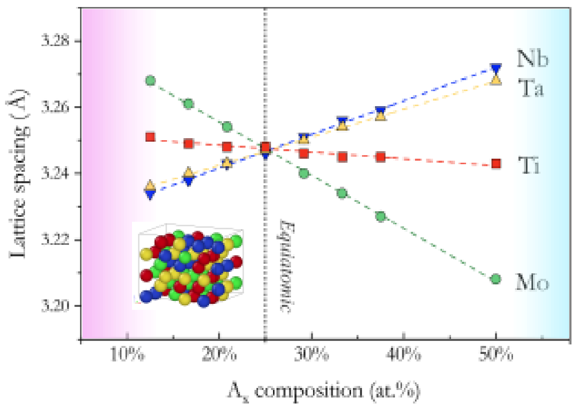
optimize
hyper-parameters

DAKOTA

Genetic algorithm

MoNbTaTi DFT Training set





LAMMPS

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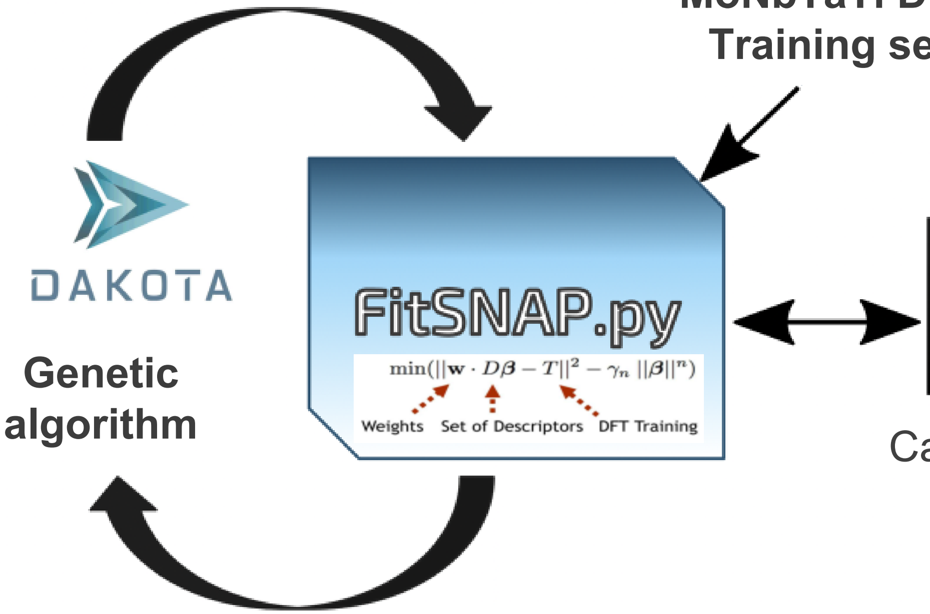

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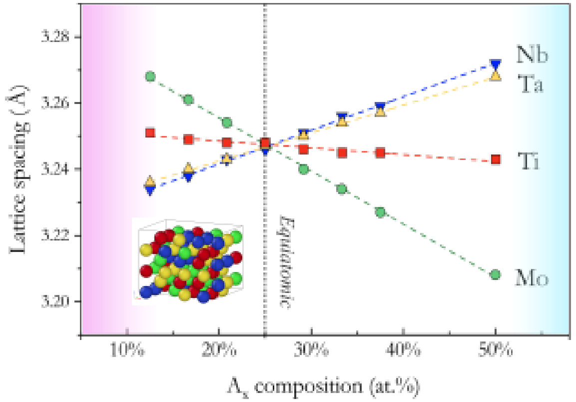

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$$E^i_{SNAP} = \beta_0 + \sum_{k=1}^{K} \beta_k (B^i_k - B^i_{k0})$$

$$E = \frac{9GB}{3B+G}$$

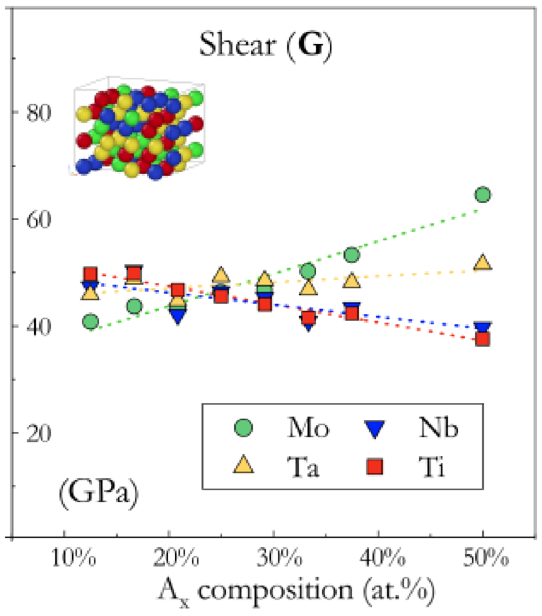
$$B_0 = \frac{\mathbb{C}_{1111} + 2\mathbb{C}_{1122}}{3}$$

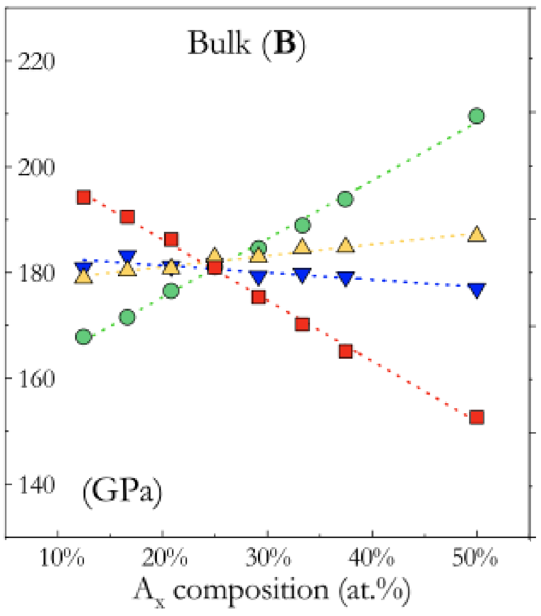
$$G = \frac{1}{2}\left[\frac{\mathbb{C}_{1111} - \mathbb{C}_{1122} + 3\mathbb{C}_{1212}}{5} + \frac{5\mathbb{C}_{1212}(\mathbb{C}_{1111} - \mathbb{C}_{1122})}{4\mathbb{C}_{1212} + 3(\mathbb{C}_{1111} - \mathbb{C}_{1122})}\right]$$

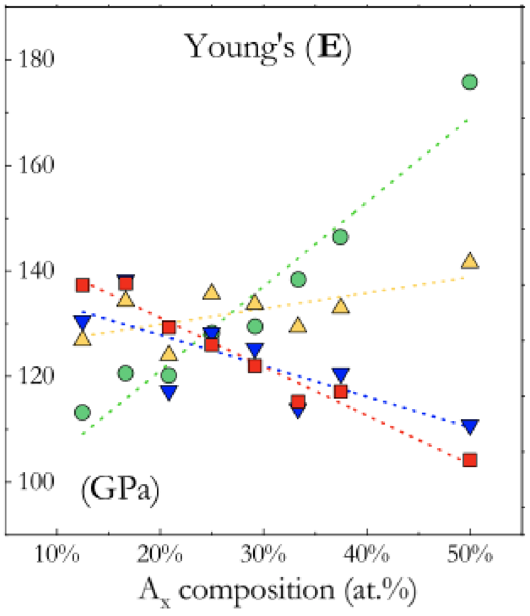
Example: Young's modulus E → Genetic algorithm favors MLIAP with low errors on cubic C11, C12, C44

Fits to higher-order elastic properties

DFT







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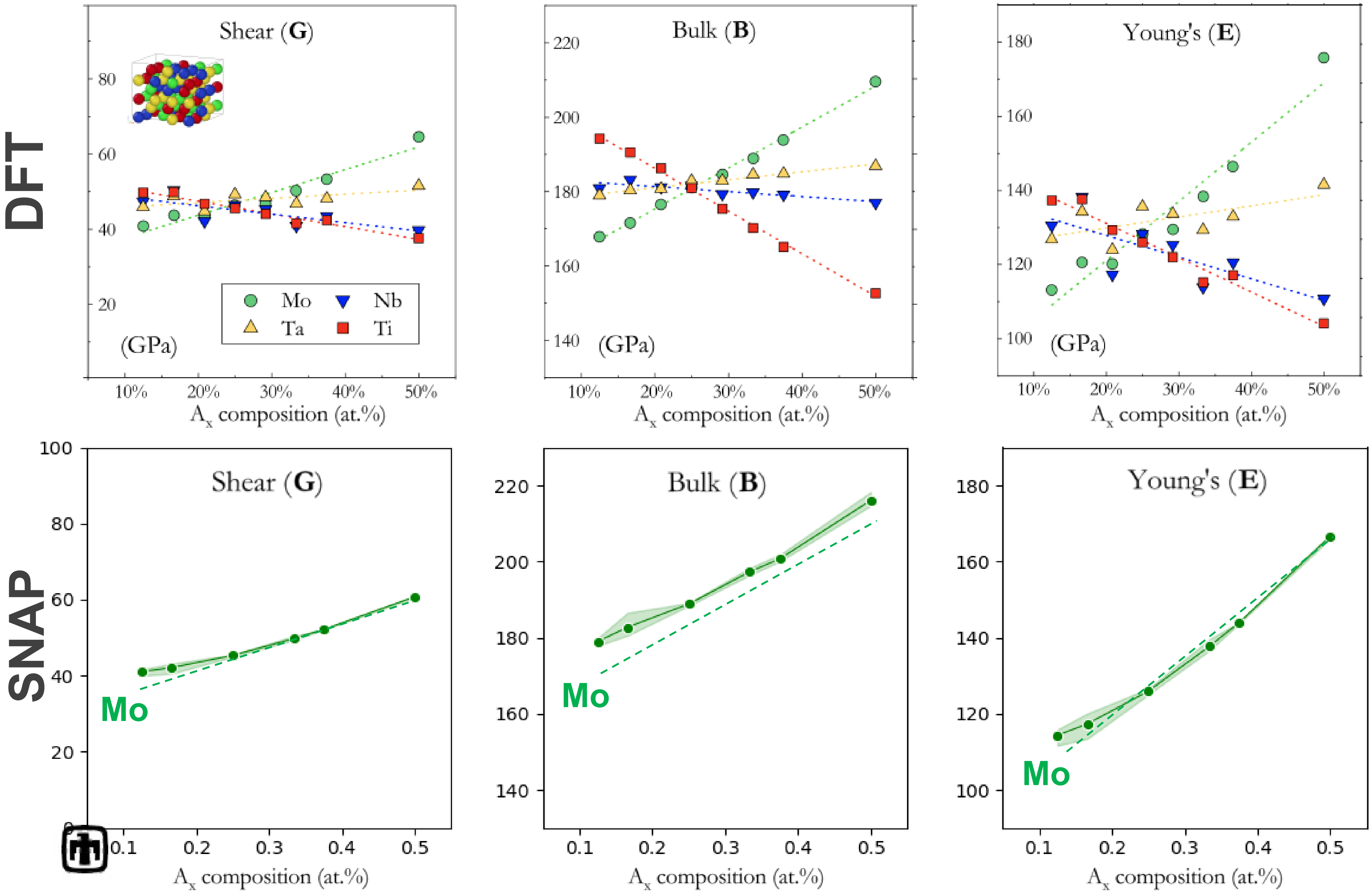

- Can reliably get stable MLIAPs that match DFT trends well across composition space for single elements
- Tested successfully at high T, P, and strain rates

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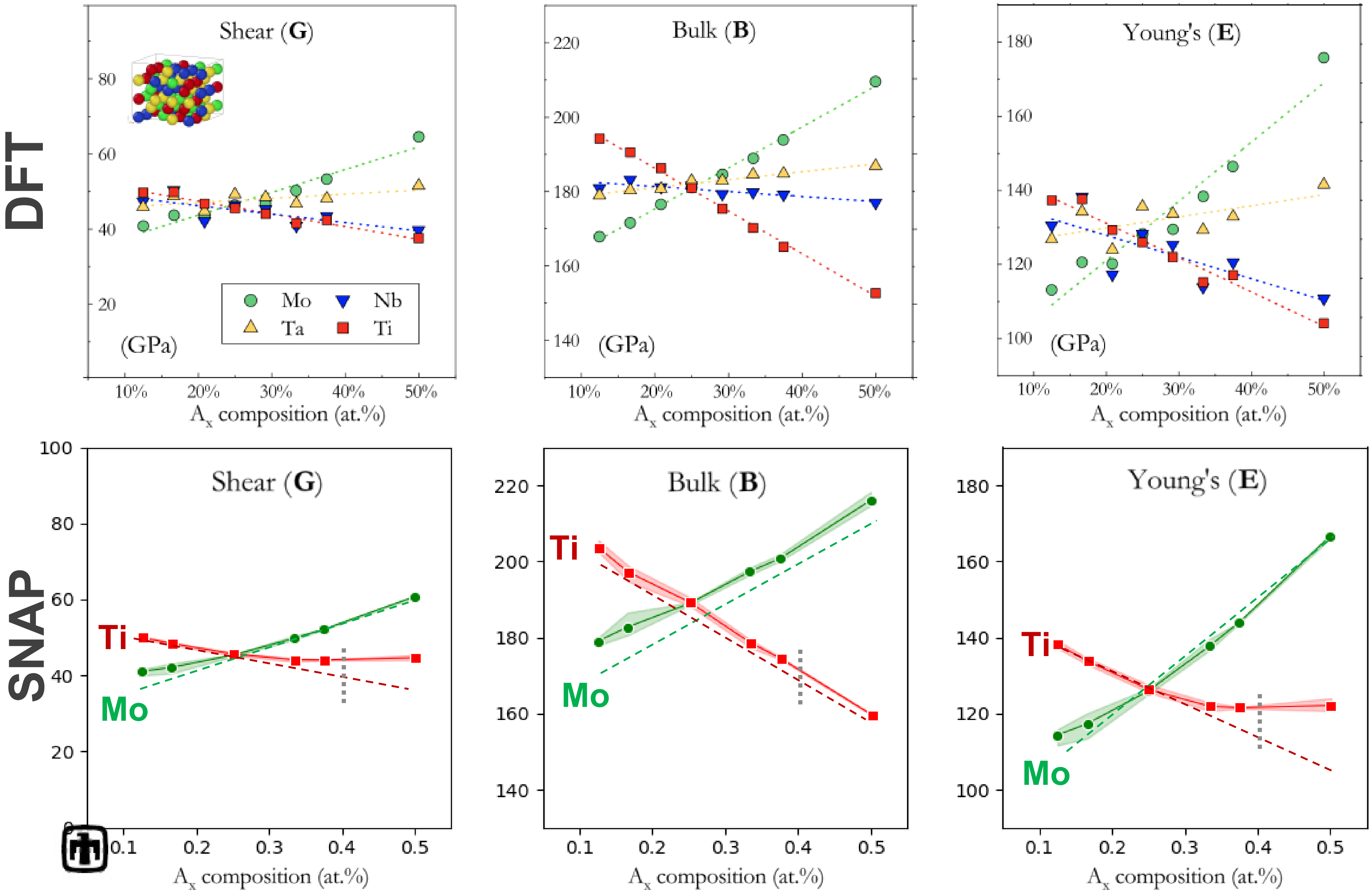

- Can reliably get stable MLIAPs that match DFT trends well across composition space for single elements
- Tested successfully at high T, P, and strain rates
- Now beginning to get stable MLIAP that can match **two elements → can extrapolate** to new compositions! *(here: most Mo at-%, up to Ti ~35 %)*

Extrapolation from training set

Untrained composition (vary 2 elements)

$Mo_{18.1}Nb_{23.6}Ta_{23.6}Ti_{34.7}$

Property	SNAP 2-element MLIAP	New DFT (not in training)	|SNAP - DFT|
C11 (GPa)	239.6	237.6	**0.8 %**
C12 (GPa)	143.6	129.7	**10.7 %**
C44 (GPa)	39.5	37.8	**4.5 %**
B (GPa)	175.6	165.7	**6.0 %**
G (GPa)	42.7	43.6	**2.1 %**
E (GPa)	118.5	120.1	**1.3 %**

Startt et al., Materials & Design. ***213*** *(2022)*

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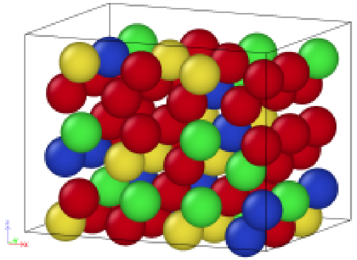
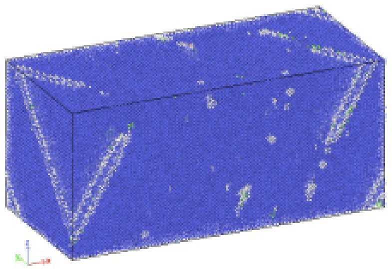
Increase system size



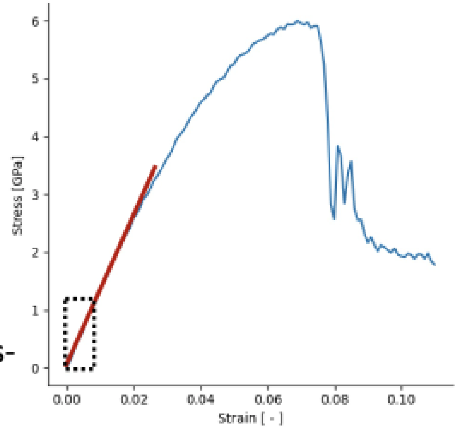
- SNAP | DFT

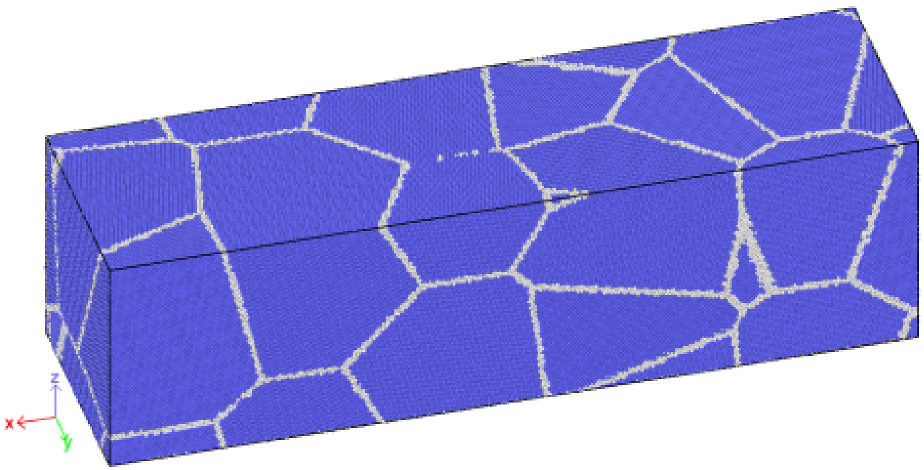
72-atom cell

ε = 6.25%



Young's Modulus from stress-strain curve: ~120 GPa





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FitSNAP →
Active learning (AL) and uncertainty quantification (UQ) modules in development!

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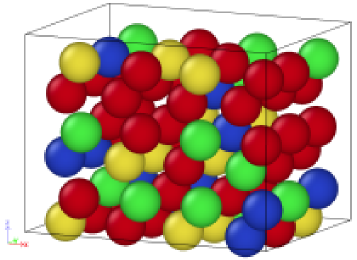

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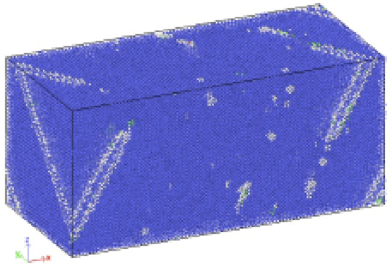

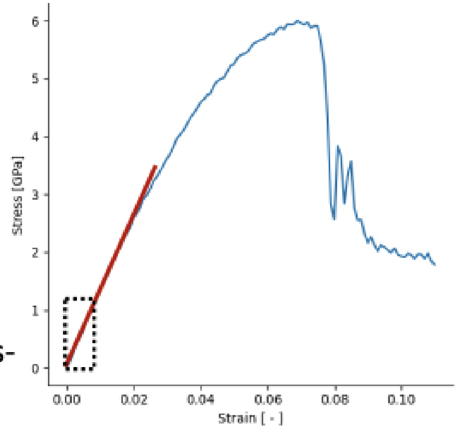

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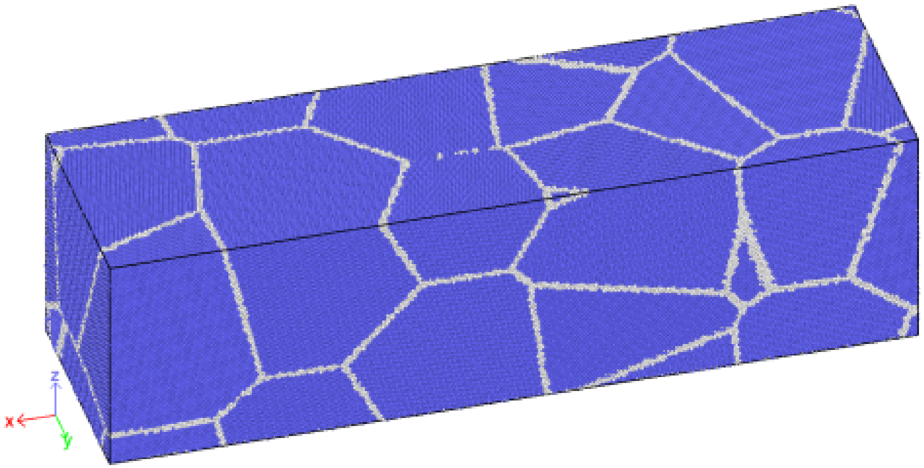

Sample new MD environments, add DFT data

Building the training set beyond elasticity

- Multiple compositions – explosion in number of training structures to include in DFT (expensive!)
  - Current MoNbTaTi comprehensive DFT set: **no point defect structures** or **derivative (binary, ternary) alloys**
- Use AL/UQ studies of **point defect behavior** and **alloy chemistry** to expand training set *where necessary*

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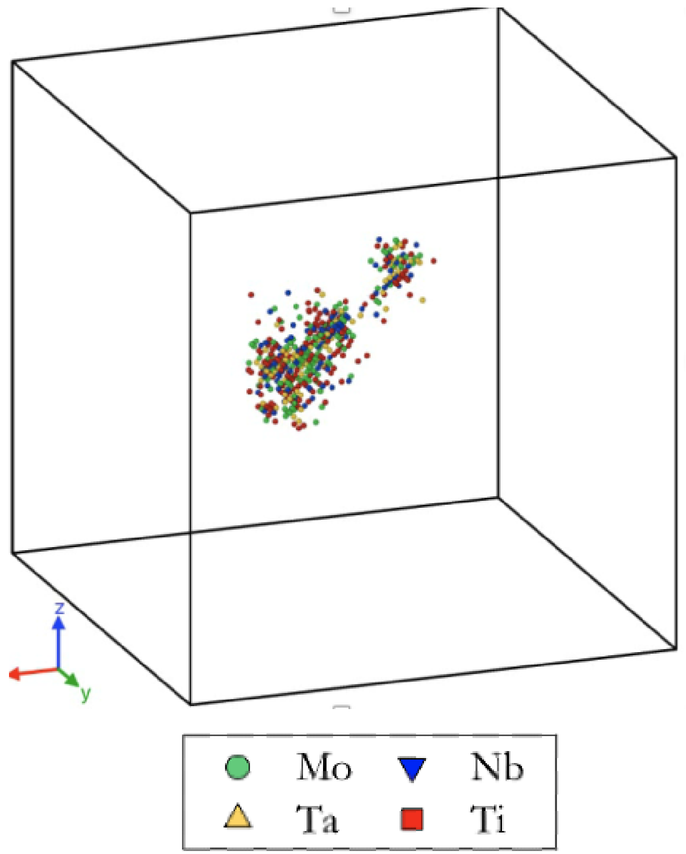
Point defects: Radiation damage

Primary knock-on atom (PKA) simulations → Expose SNAP to per-atom energy levels far exceeding range seen in training set

Training: order of ~10 eV
PKA:
5 keV, 10 keV, 20 keV



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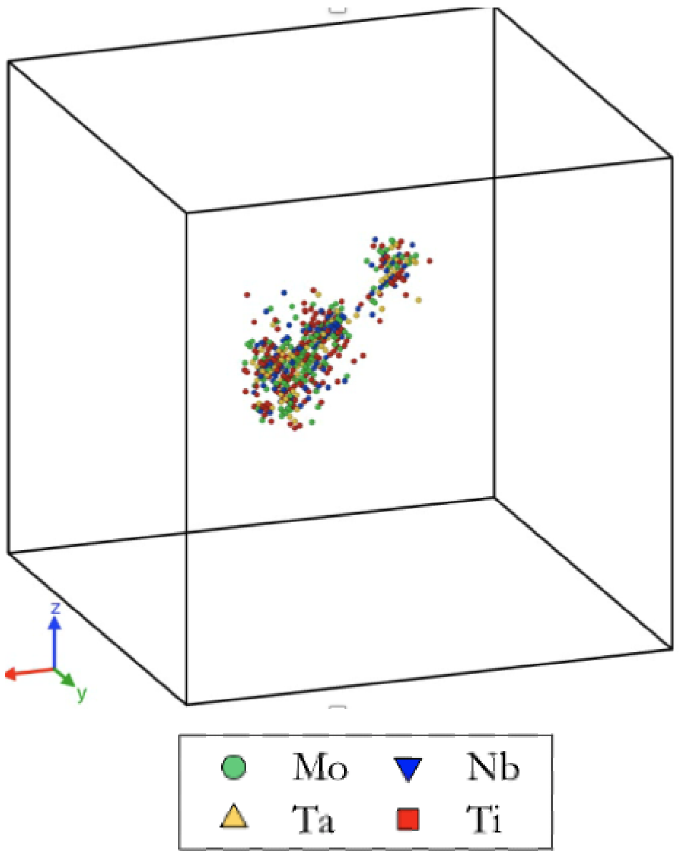
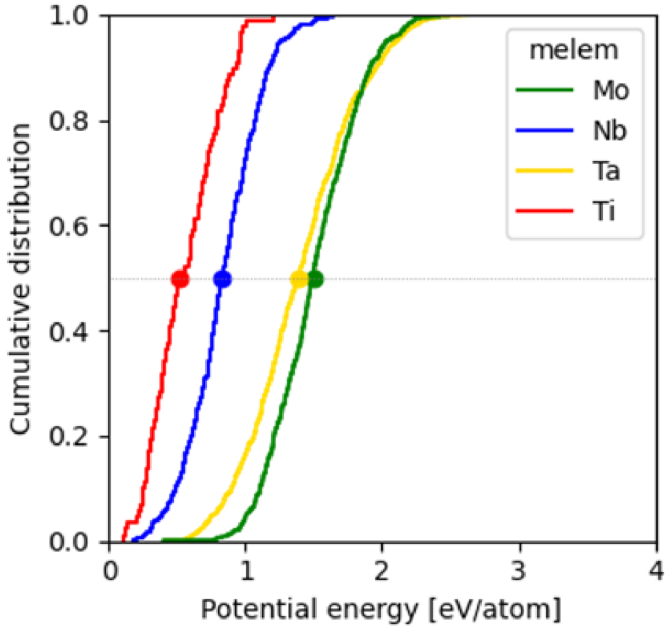


Vacancy migration energy distributions (from NEB)

Mo-25% Nb-25% Ta-45% Ti-05%
(Ta enriched, Ti depleted)



Mo-25% Nb-25% Ta-05% Ti-45%
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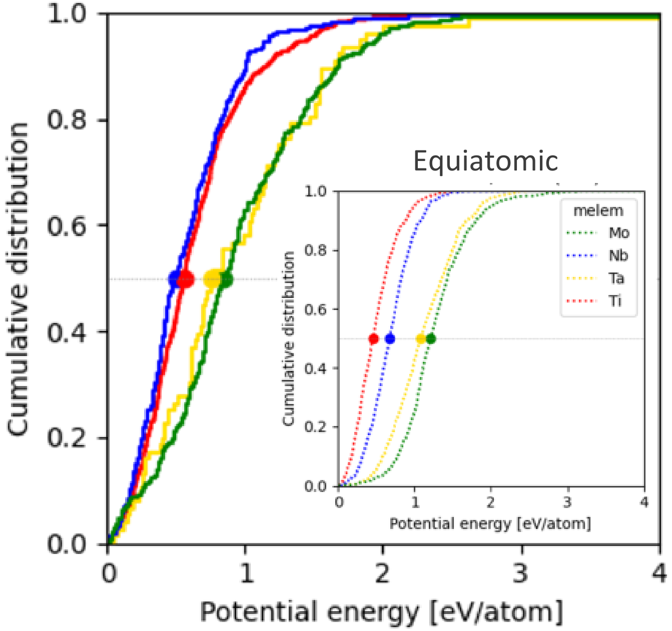

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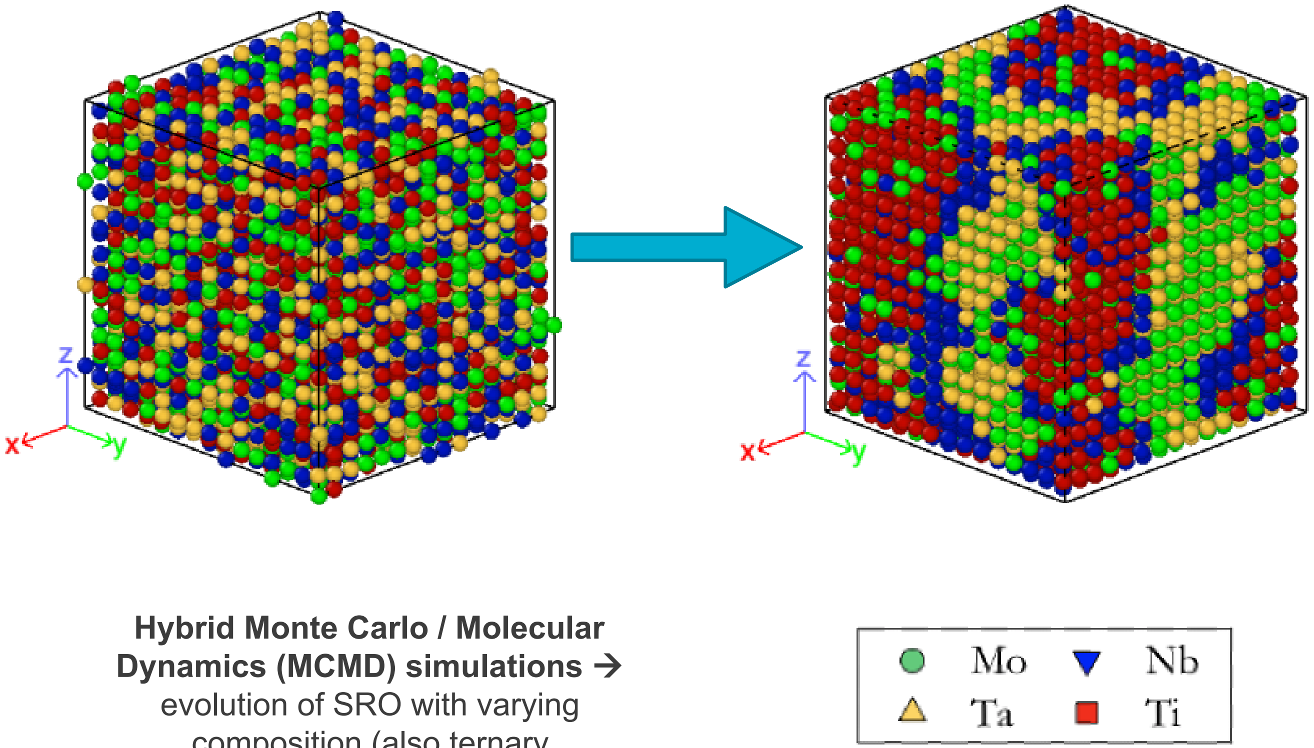
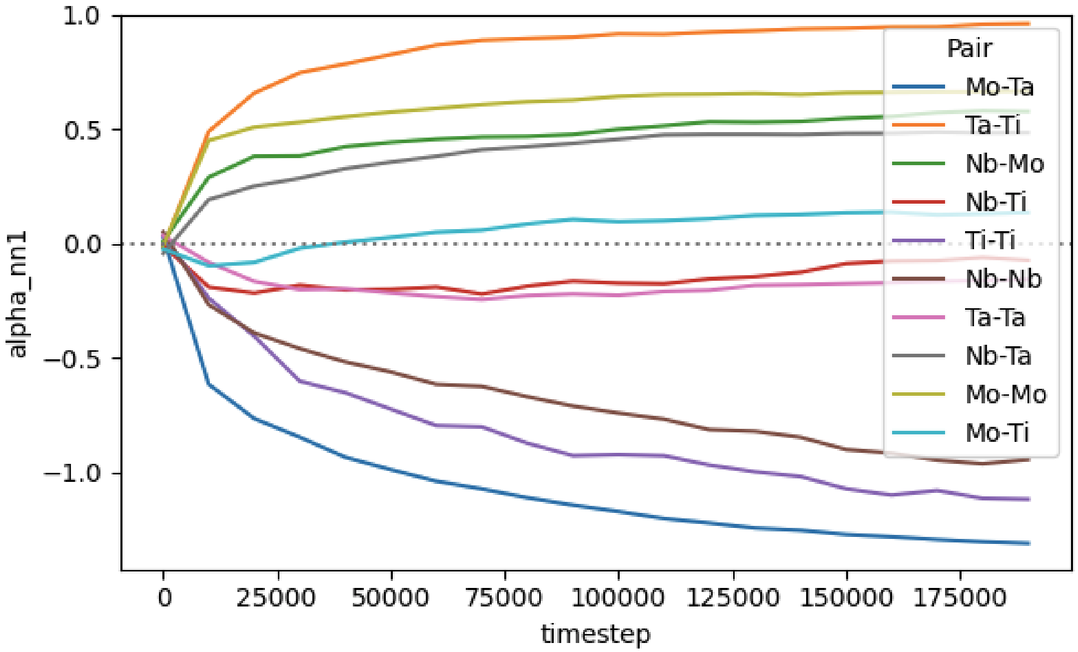
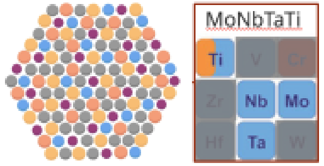
Alloy chemistry: short-range order studies



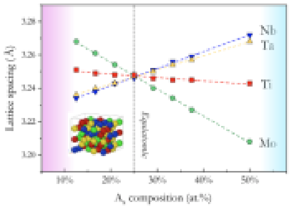


Hybrid Monte Carlo / Molecular Dynamics (MCMD) simulations → evolution of SRO with varying composition (also ternary

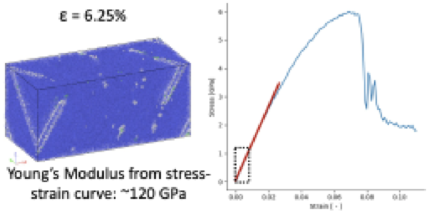
Summary



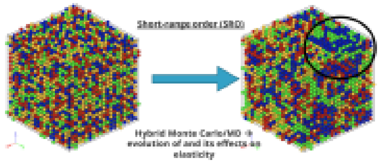
<u>MoNbTaTi</u> is a promising member of the refractory complex concentrated alloys



A comprehensive DFT study was undertaken by Startt et al. to understand how <u>non-equiatomic composition affects material properties</u> such as elasticity



Using FitSNAP and a genetic algorithm on the as-received DFT training set, we are successfully generating <u>stable MLIAPs</u> that can <u>match elastic property trends across multiple compositions</u> and <u>extrapolate successfully to new compositions</u>



Active learning and uncertainty quantification will be used to understand and expand scope of current MoNbTaTi training set

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

Sandia National Laboratories

Contact:
megmcca@sandia.gov