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From First-Principles toward atomistic understanding of mechanical properties in High-Entropy Alloys

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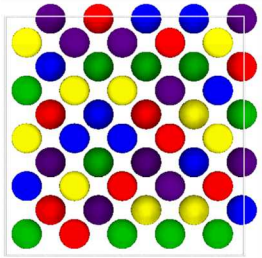
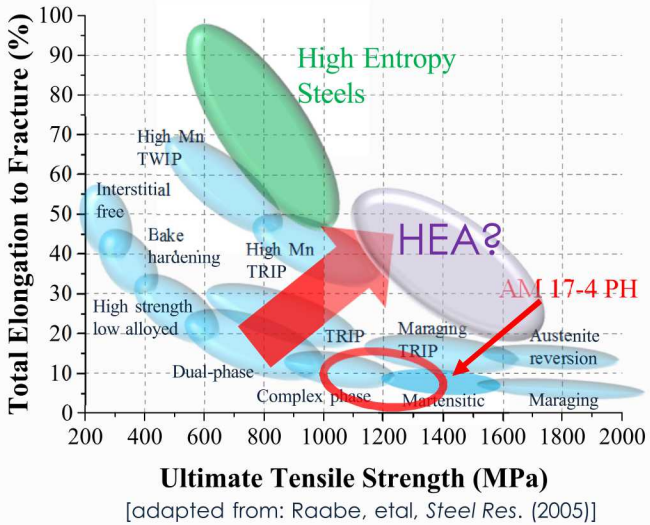
Why High Entropy Alloys - HEA?

High entropy alloys



- Random composition crystalline alloys, four or more principal equiatomic constituents.
- Great potential for structural applications: strong, degradation-resistant.
- Amenable to additive manufacturing-AM?



[adapted from: Raabe, etal, *Steel Res.* (2005)]

Goal:
AM-friendly HEA alloy for high-temperature structural applications: high ductility and fracture resistant

Challenges:
High dimension composition space
Constraints of AM conditions

Need:
Physics-based framework to guide discovery/development of HEA

Focus: **systematic *computational* framework** for atomic-based structure-property relationships.

Anatomy of a multiscale materials model:

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- What do you want? Structure-property relationships
  - structural stability → composition, phases, enthalpies
  - strength → ductility, shear properties (and at high temperature)

- Need:
  1. Accuracy of DFT (or better?) – *too big/complex for DFT*
  2. Dynamical properties with MD – ***I**nter**A**tomic **P**otentials not accurate/available*
  3. Meso-scale microstructure evolution – *lack effective models*

DFT → **IAP** → MD → Meso

MULTISCALE in the face of LARGE COMBINATORIAL COMPLEXITY

Exemplar: refractory **Al-Nb-Ti** — "simple", interesting, less studied

SNAP – Spectral Neighbor Analysis Potentials

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- SNAP describes local environment as atomic (particle) density:

$$\rho_i(\boldsymbol{r}) = \delta(\boldsymbol{r}) + \sum_{r_{ij}<R_{cut}} f_c(r_{ij})\, w_i\, \delta(\boldsymbol{r} - \boldsymbol{r}_{ij})$$

w_i = weights defining atomic species
f_c = radial switching functions

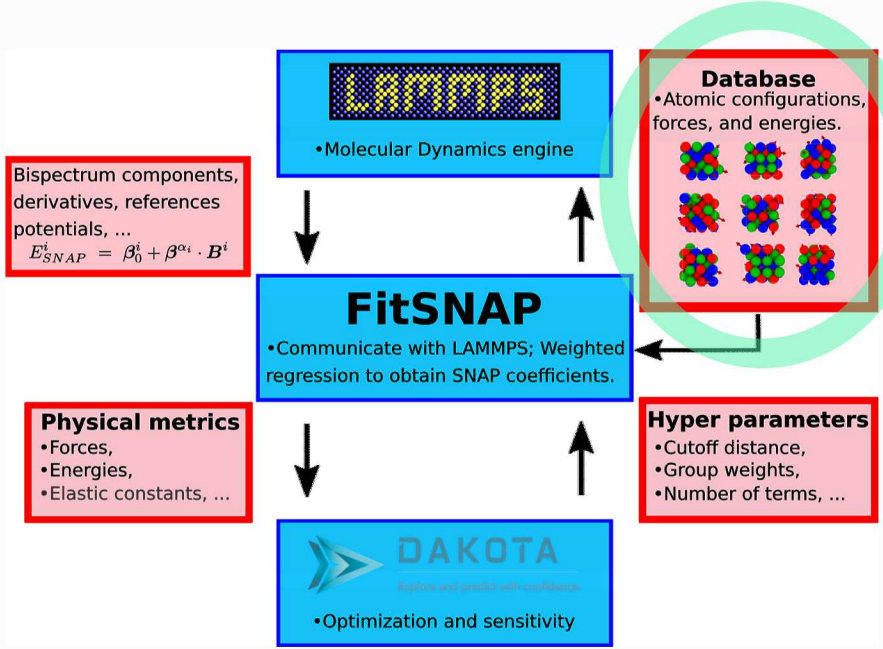
- Density expressed in terms of bispectrum components. Those bispectrum components express energy and forces on each atom:

$$\begin{cases} E^i_{SNAP} = \beta^i_0 + \boldsymbol{\beta}^{\alpha_i} \cdot \boldsymbol{B}^i \\ \boldsymbol{F}^j_{SNAP} = -\boldsymbol{\beta} \cdot \sum_{i=1}^{N} \dfrac{\partial \boldsymbol{B}_i}{\partial \boldsymbol{r}_j} \end{cases}$$

- Fit bispectrum components and weights to reproduce potential energy surface of DFT.

Data/ML-driven interatomic potential

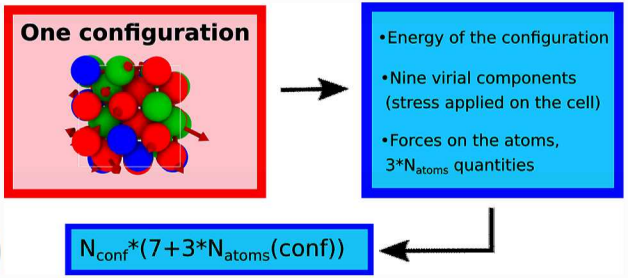
Thompson, A. P., *et al*.. (2015). J. Comp. Phys., 285, 316-330.



… Only as good as its data

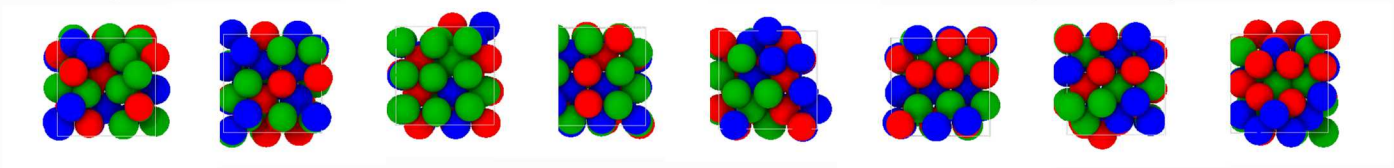
DFT – HEA training data design

Goals with DFT:
- accurate structure/composition energies
- accurate elastic properties
- discern crucial materials physics of HEA
- computationally practical (combinatorial complexity)



Representative "**SQS**" model ***impractical*** and ***insufficient*** for HEA alloys

Strategic sampling of random 54-atom cubic (3x3x3) cells:



- atoms isolated (near-sightedness), "cheap", equi-composition (18-18-18)

Computational methods

- **General purpose DFT code SeqQuest** (http://dft.sandia.gov/Quest)
  - well-converged (Gaussian-based) local orbital basis
  - both LDA and PBE functionals
  - converged norm-conserving pseudopotentials
  - full force-relaxed atoms
  - stress-relaxed cells (accurate stress-strain)
  - carefully verified against converged FP-LAPW results

- **HEA supercells**
  - 54-atom cubic supercells (3x3x3 conventional 2-atom bcc cubic cell)
  - real-space grids: 96^3
  - offset k-grids
  - Constrained to cubic

- **And then also Quantum Espresso**

DFT – validation/accuracy

			a_0 (Å)	c_0 (Å)	c/a	B(GPa)	Energy
Al	expt.	fcc	4.04			72.2	
	PBE	fcc	4.053			75	0
		bcc	**3.246**			**67**	**+0.093 eV/Al**
Nb	expt.	bcc	3.30			170.2	
	PBE	**bcc**	**3.316**			**171**	**0**
Ti	expt.	hcp	2.95	4.68	1.586	105.1	
	PBE	hcp	2.931	4.662	1.590	114	
		bcc	**3.352**			**108**	**+0.104/Ti**

SeqQuest/PBE: lattice constants within < 1%
bulk moduli within 8%

DFT– two-component B2/bcc alloys

a_0 (Å)	Al	Nb	Ti
Al	3.246		
Nb	3.236	3.316	
Ti	3.188	3.270	3.352

Expect a_0 ~ 3.2-3.3 Å for HEA

B (GPa)	Al	Nb	Ti
Al	67		
Nb	141	171	
Ti	113	135	108

Expect *B* ~ 110-140 GPa for HEA

H_f (eV/fu)	Al	Nb	Ti
Al	+0.186		
Nb	-0.050	0	
Ti	-0.537	+0.136	+0.206

Ti – Al very happy
Nb – Ti less happy

H_f^{bcc} (eV/fu)	Al	Nb	Ti
Al	0		
Nb	-0.143	0	
Ti	-0.733	+0.035	0

Binding dominated by Ti – Al?

Need to be concerned about intermetallics?

DFT Equation-of-State for HEA

7 random colorings X 6 element mappings
42 Al_{18}-Nb_{18}-Ti_{18} cells

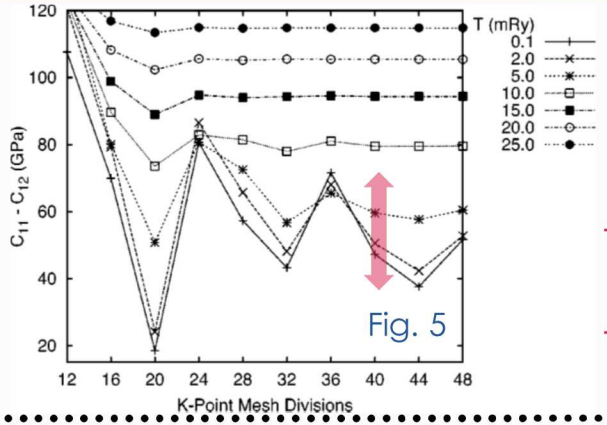
	a_0 (Å)	*B* (GPa)	H_f (meV/at)
18-18-18	**3.246 (2)**	**125 (2)**	**176 (14)**
Al-Nb-Ti			
20-19-15	3.245	125	190
19-21-14	3.249	129	175
17-15-22	3.242	124	170
21-14-19	3.240	114	186
20-16-18	3.242	122	184

Consistent between 42 random cells:
Random-54 cell ~ "SQS"

Deviation from equi-atomic:
Not sensitive

Vindicates sufficiency of 54-site cells.

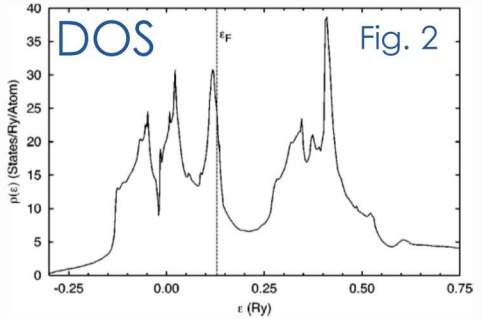
Shear – k-point catastrophe?



Mike Mehl, PRB **61**, 1654 (2000)
Nb – Tight Binding

- Shear moduli do not converge

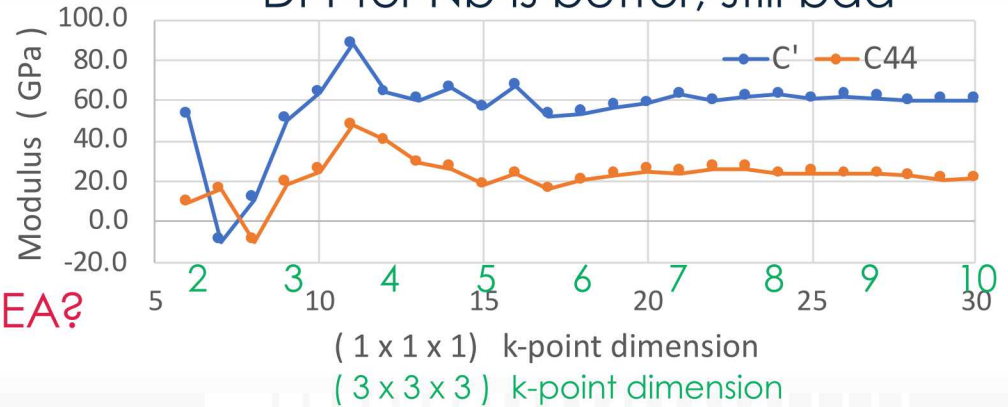
- Structure interacts with DOS at ε_f



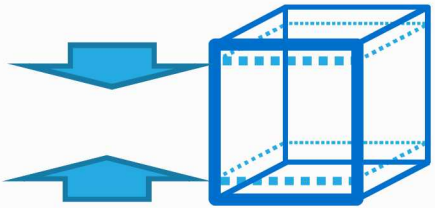
General issue for metals –
- Ta is worse (MSMSE 13, R1 2009)
- Al is worse
- NbTi (B2) is worse

Need expensive >5^3 k-points for HEA?

DFT for Nb is better, still bad

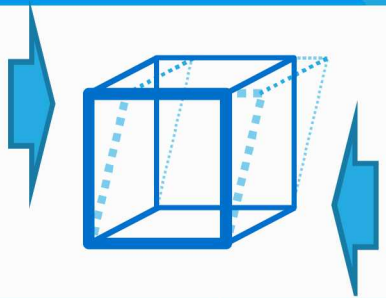


HEA elastic constants – strain-stress Hooke's Law



C_{11}, C_{12}

$$C' = \frac{C11 - C12}{2}$$



C_{44}

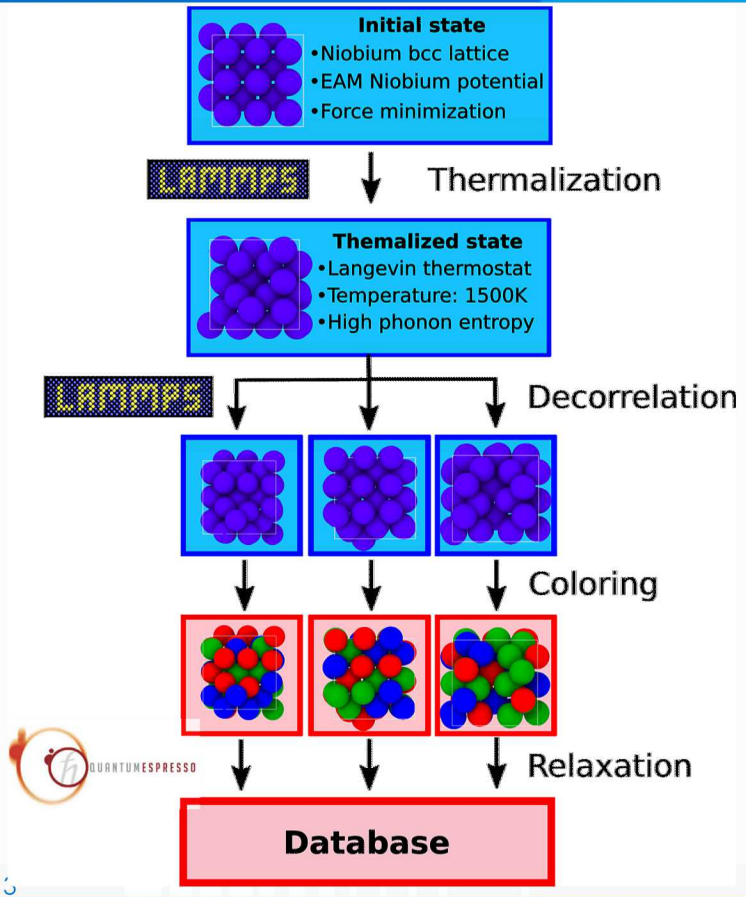
k(1x1x1)	k(54-cell)	C' (GPa)	C_{44} (GPa)	B (GPa)
6 x 6 x 6	2 x 2 x 2	15	69	125
9 x 9 x 9	3 x 3 x 3	17	70	125
12 x 12 x 12	4 x 4 x 4	18	71	126

Very fast convergence

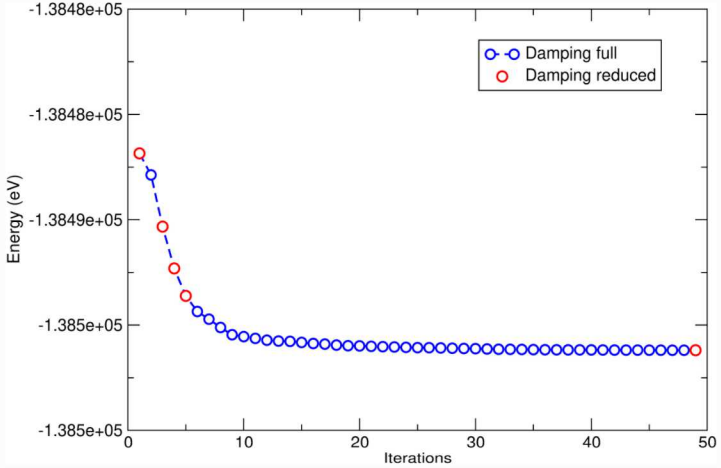
Compositional randomness → localizes electrons, damps electronic effects

DFT → IAP: sampling of small (54-atom), fast (2x2x2 k-pts) cells suffices

Framework for generating (decorrelated) DFT data



- 600 decorrelated MD configurations sampled
- DFT relaxation ~40 steps → ~24000 configurations
- Use 5 energy-distinct points → 3000 configurations



- **Future**:
  Use ML to filter this copious data set.
  Use ML to identify and augment deficiencies

Summary and Prospects

- Developed physics-based protocol for DFT training data
  - Sampling small 54-site cells, random-composition sufficient
  - Small k-points suffice – atomic structure ←→ electronic structure
  - Huge training data possible → ideal for ML

- Path to combinatorial 4-5-6-7 component HEA
  - Insensitivity to most variations in local environment
  - SNAP-ML potentials & training data protocols accommodate large data

- Future challenges:
  - Will these potentials really work? - *see you next year*
  - Can DFT and MD-SNAP give good enough data to inform meso?

paschul@sandia.gov http://dft.sandia.gov/Quest