From Structure to Properties and the Mess Between

Derick Ober

Overview

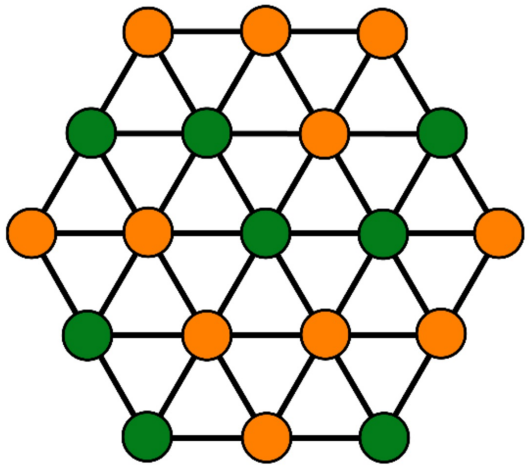
Context
- Structure to Thermodynamic Properties

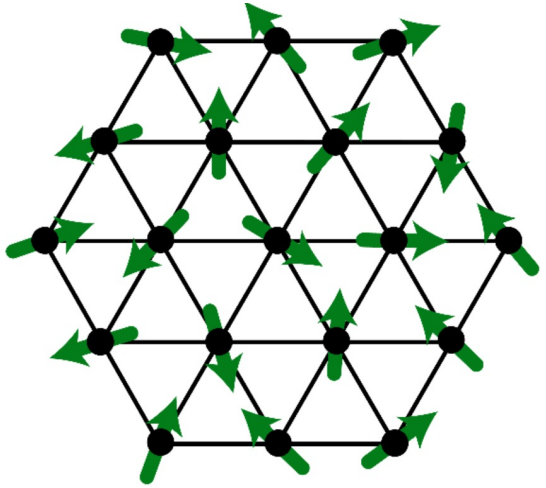
Motivation
- Fidelity and Transparency in Predictions

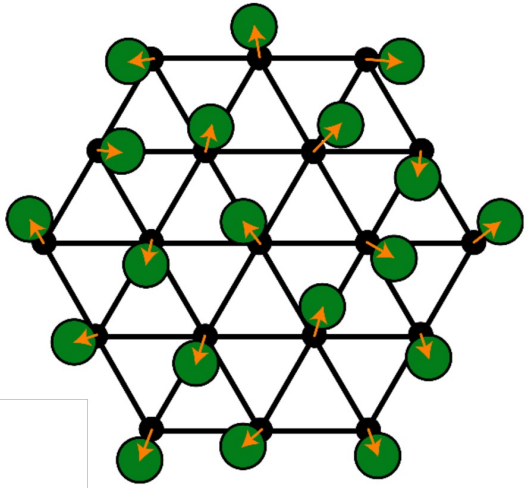
Solutions
- Bayesian Uncertainty Quantification

Structure-Property Relations: Thermo

- Symmetry, elemental, magnetic, vibrational degrees of freedom all impact thermodynamic properties





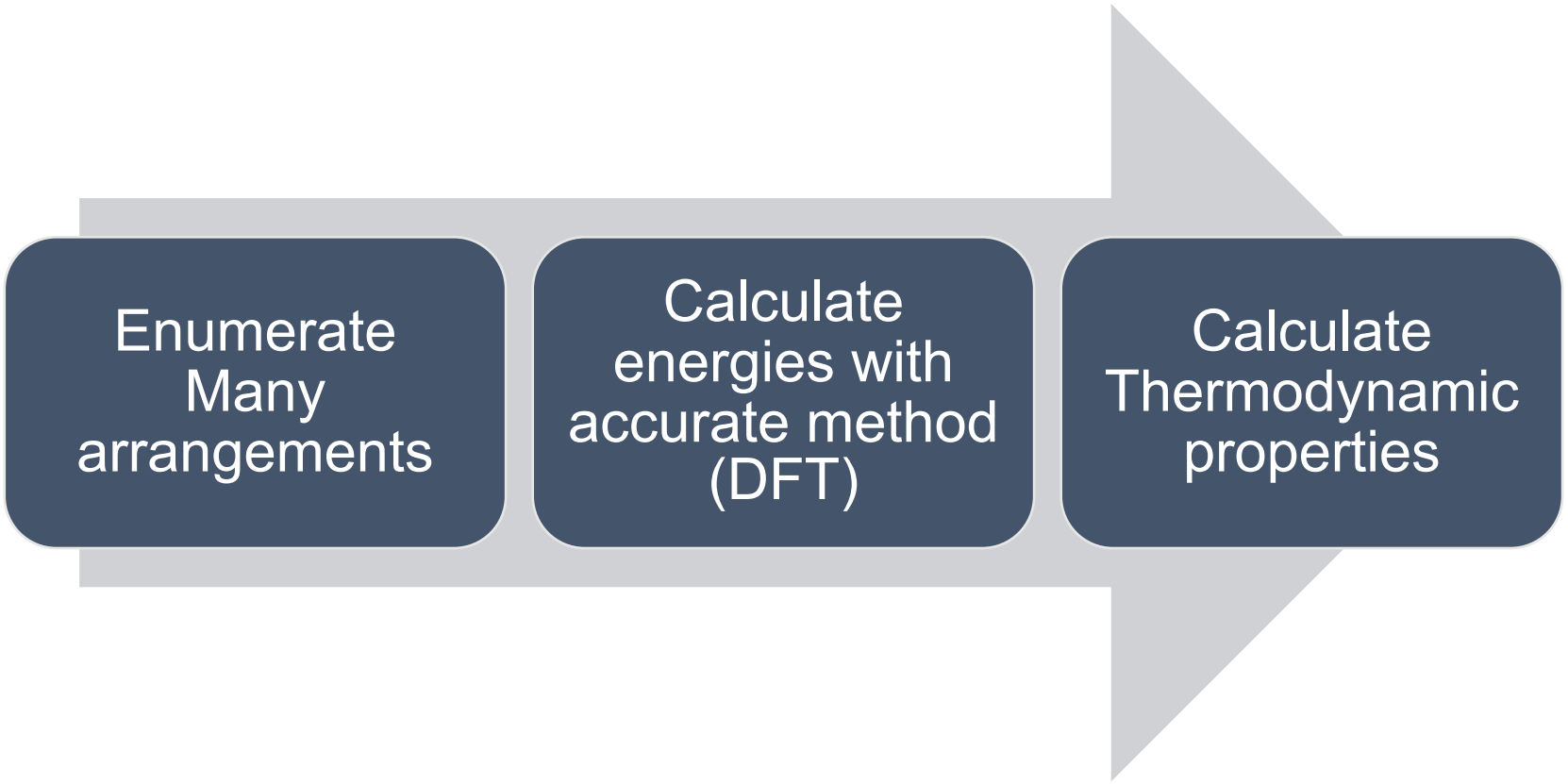


Computing Structure-Property Relations

- If we know energies, we know thermodynamic properties.

- If we could compute as fast as we wanted, and store as much data as we liked… how would we do this?

Computing Structure-Property Relations

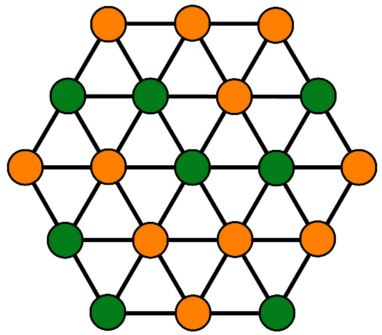


Computing Structure-Property Relations

Enumerate Many arrangements

Calculate energies with accurate method (DFT)

Calculate Thermodynamic properties



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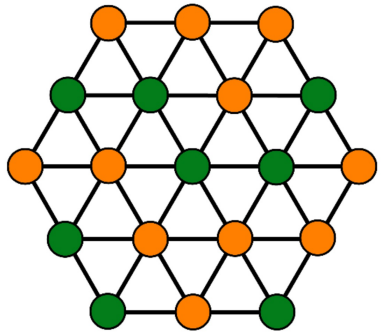

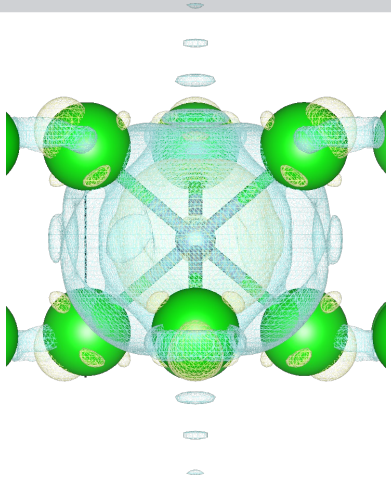

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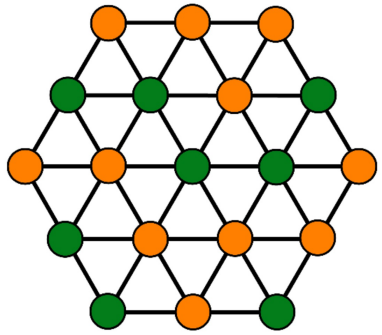

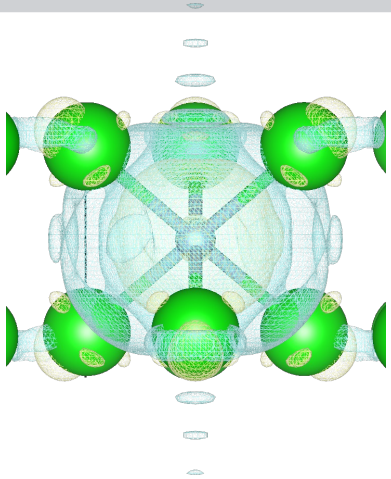

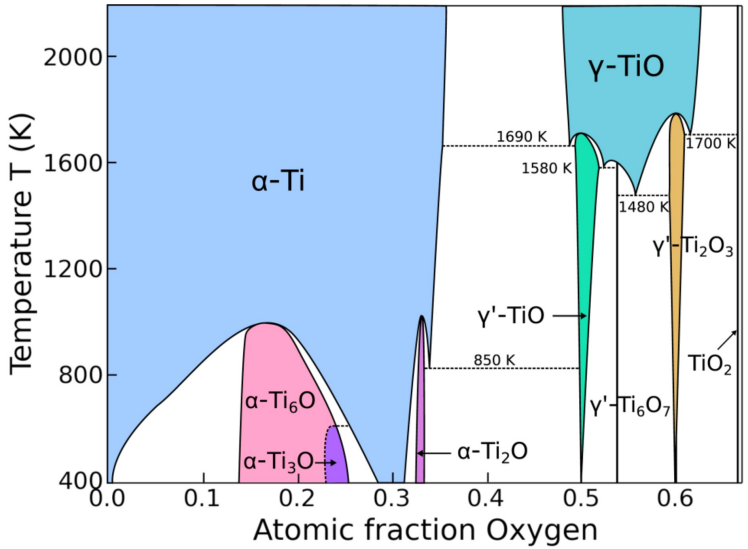

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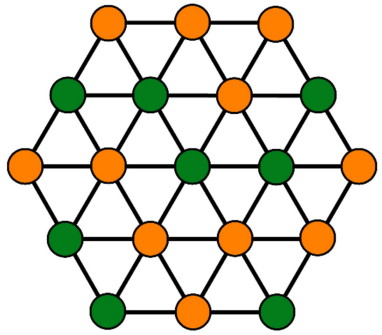
Computing Structure-Property Relations

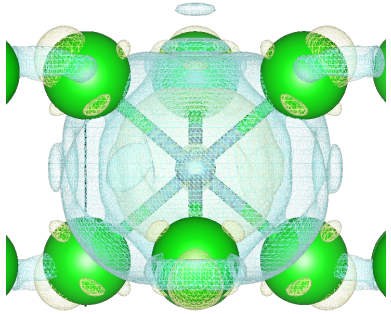
Millions of energy calculations?

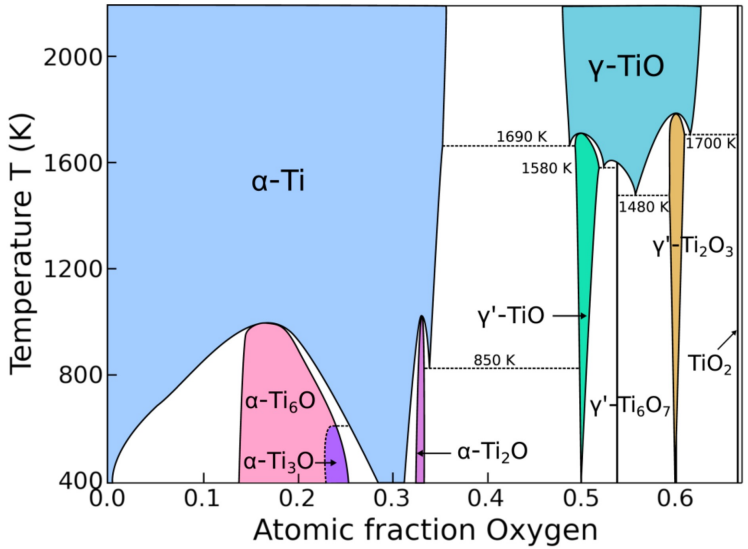
Enumerate Many arrangements

Calculate energies with accurate method (DFT)

Calculate Thermodynamic properties







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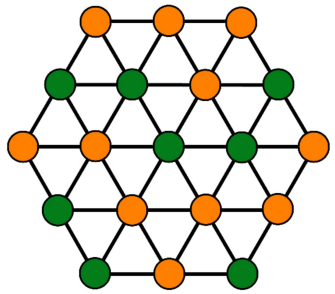
Structure-Property Relations at Scale

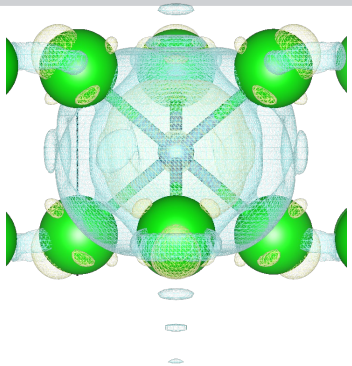
Enumerate Many arrangements

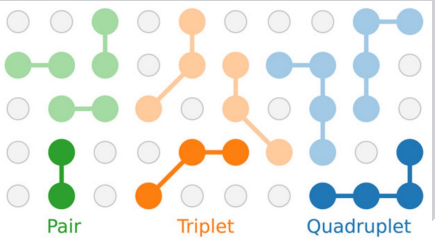
Calculate energies with accurate method (DFT)

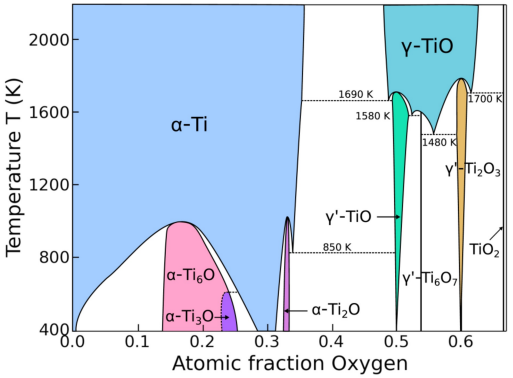
Surrogate Model: Cluster Expansion

Calculate Thermodynamic properties









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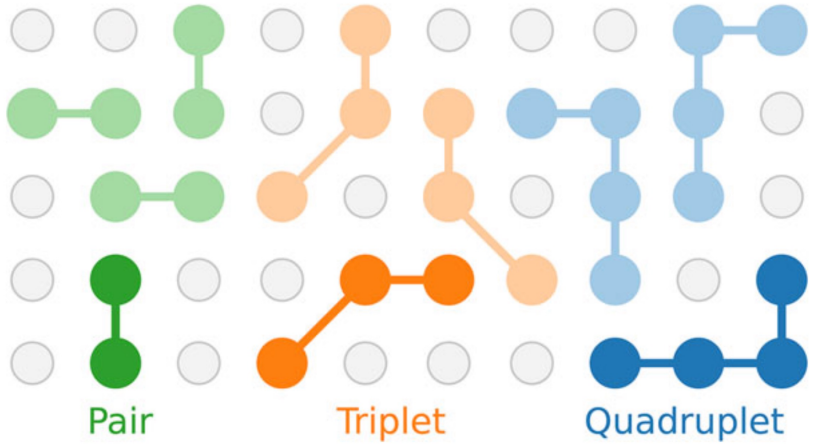
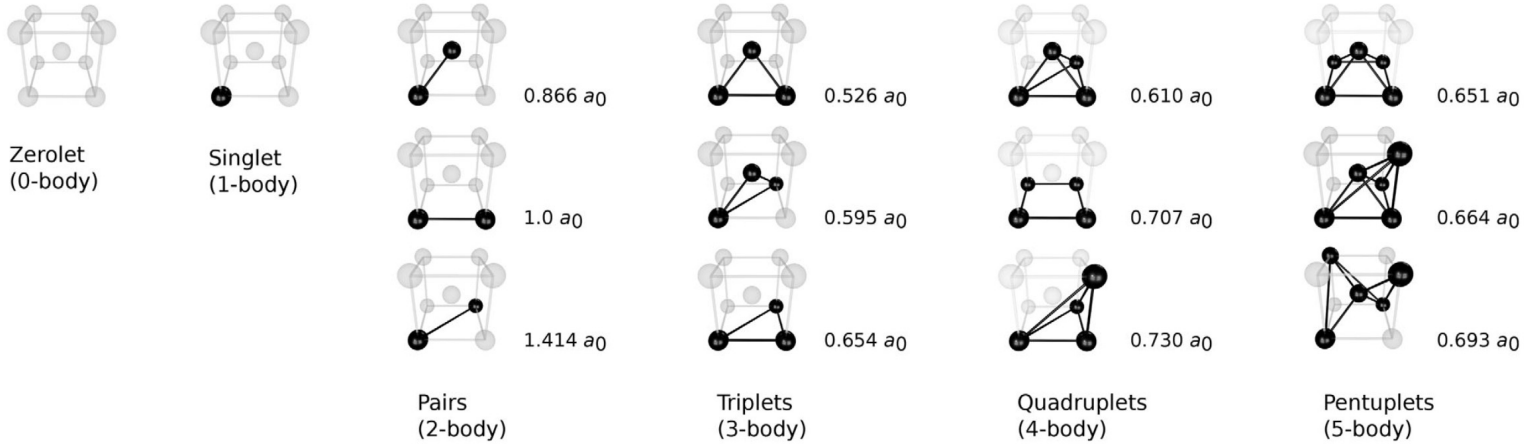
Surrogate Model: Cluster Expansion

- Fast
- Like an Ising model with more interactions
- Gives physical intuition
- Exact when using infinite terms (Think Fourier Series)





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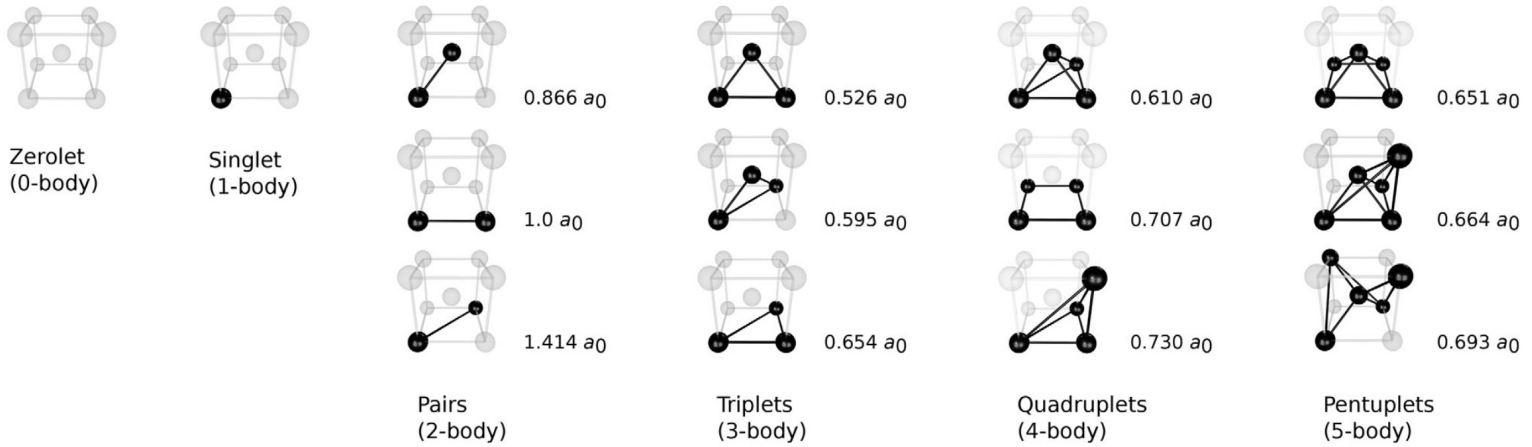
Surrogate Model: Cluster Expansion

$$E = \vec{X} \cdot \vec{V}$$

- $\vec{X}$: A fingerprint describing an arrangement of atoms on a lattice. (KNOWN)
- $\vec{V}$: Effective Cluster Interactions (ECIs) describe how each type of interaction contributes to the energy (UNKNOWN)



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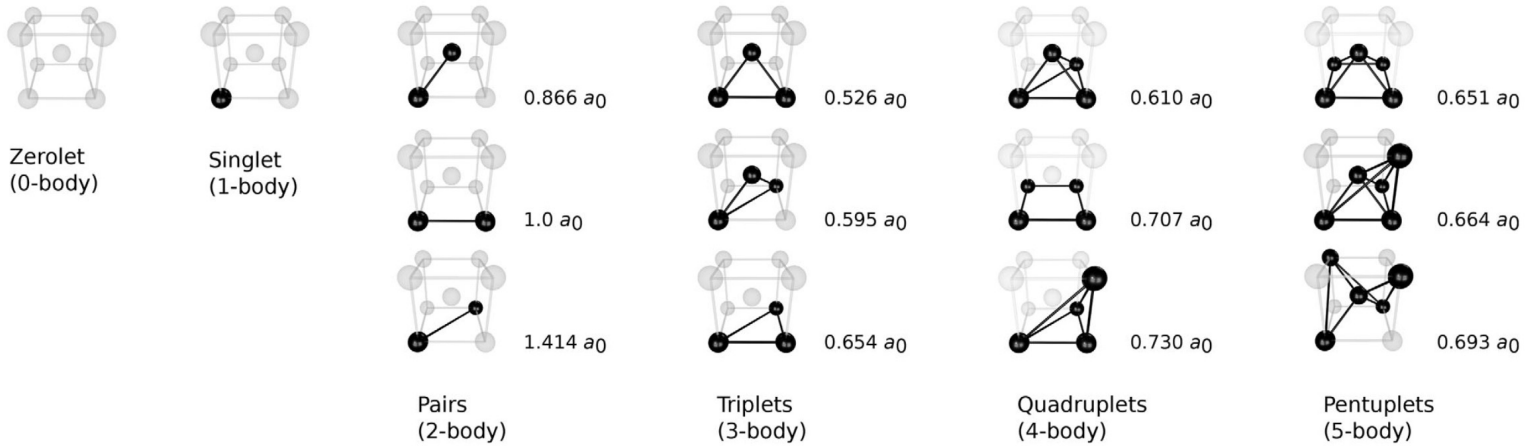

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Motivation: Current Work

- **Big** composition spaces:
  - HCP, FCC
  - Ti, Zr, Hf, Nb, O, N

- Refractory Material uses:
  - Nuclear cladding
  - High temperature barriers
  - Battery anodes
  - Ferroelectrics

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- Want thermodynamic properties
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  - Voltage curves

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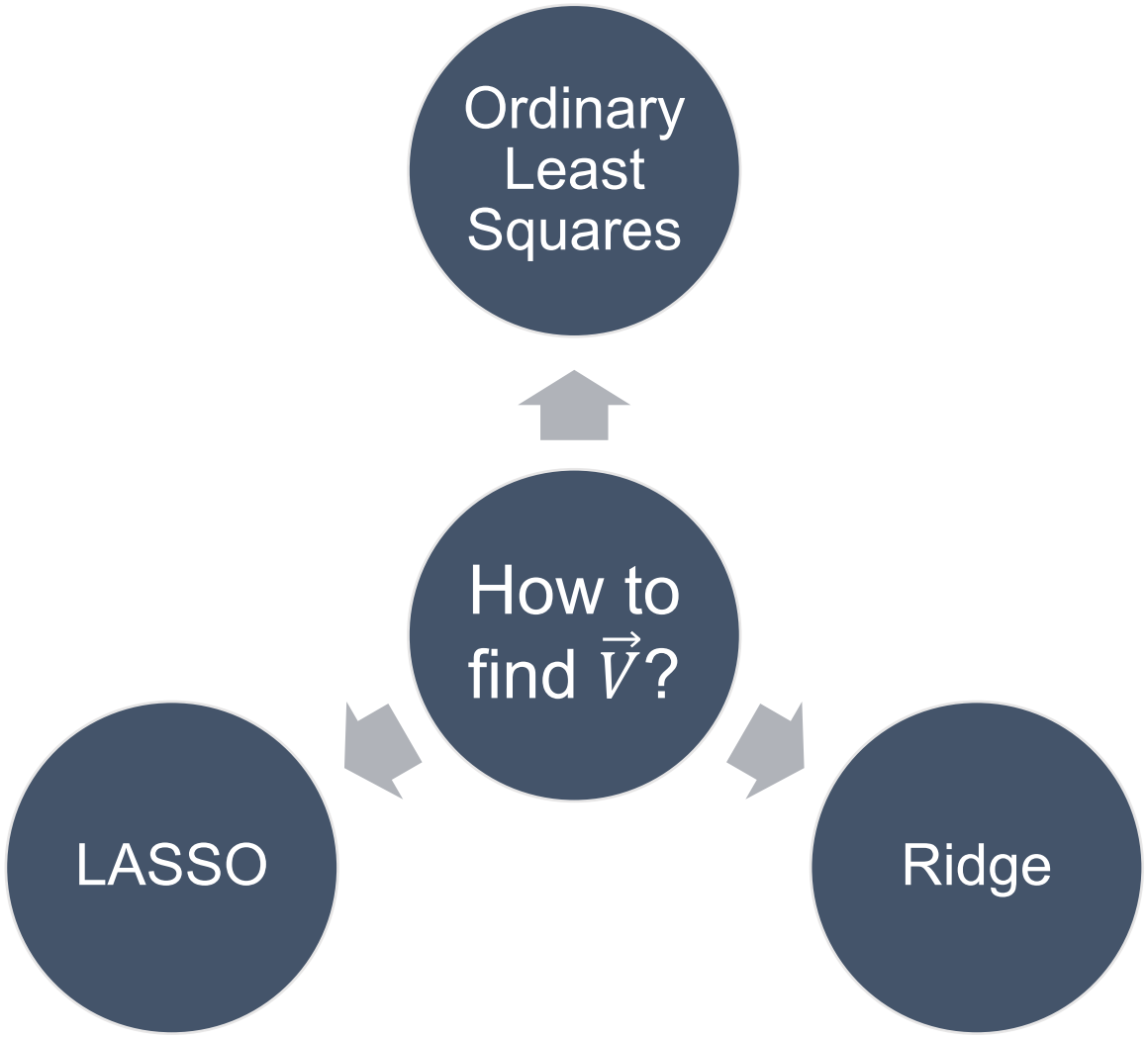
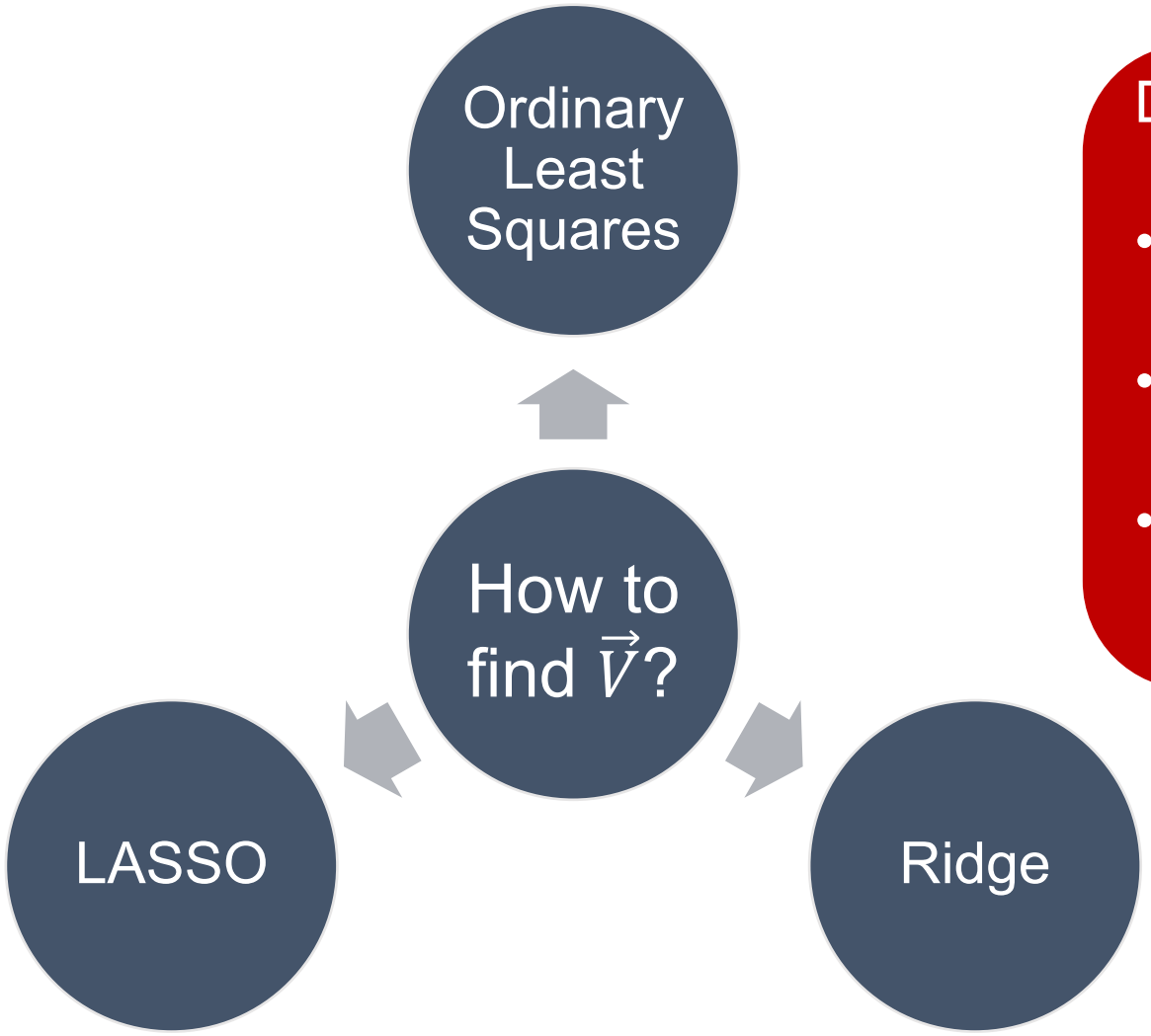
We want our predictions to be CONFIDENT!

Surrogate Model: Cluster Expansion



Let's play a game: pick your favorite linear regression method

Surrogate Model: Cluster Expansion



Depending on your choice:

- Thermodynamic results may not match
- Subjective
- Doesn't report confidence

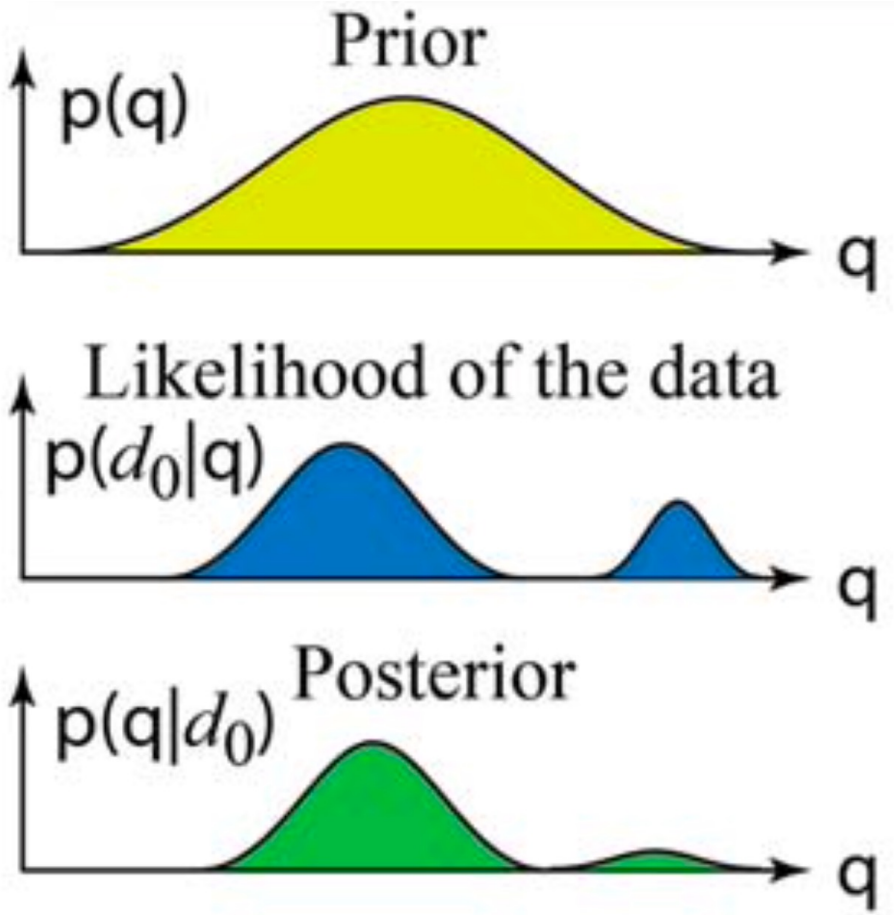
Surrogate Model: Cluster Expansion

How to find $\vec{V}$?

Bayesian Cluster Expansion

- Distribution of Thermodynamic results
- Combines Intuition and observed data
- Reports confidence

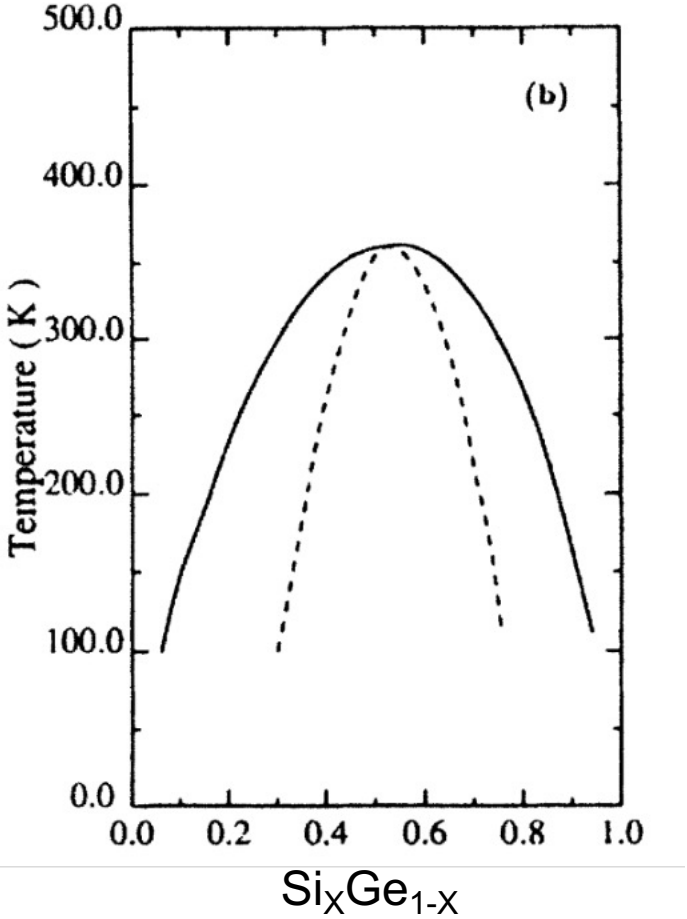
Bayes Theorem: Graphically

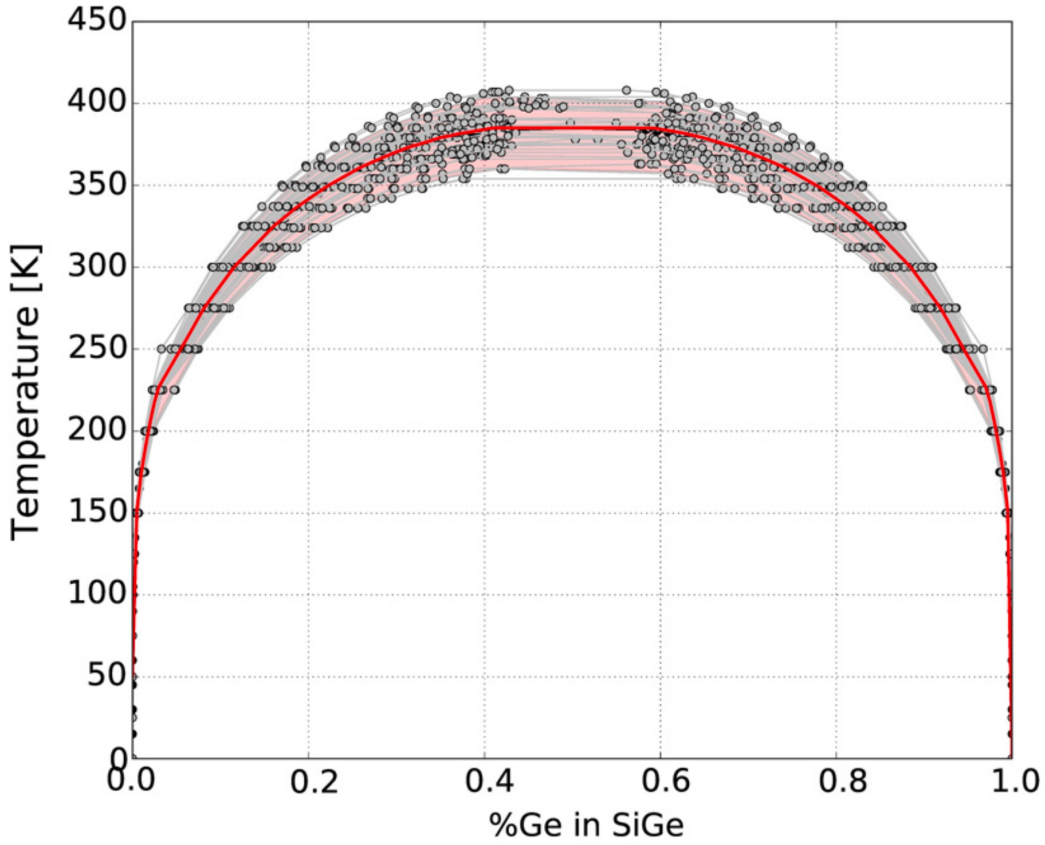


$$\text{Posterior } P(q|d_0) = \frac{\text{Likelihood } P(d_0|q) \times \text{Prior } P(q)}{\text{Normalization Constant}}$$

P. Honarmandi, N. H. Paulson, R. Arróyave, and M. Stan, "Uncertainty quantification and propagation in CALPHAD modeling," *Modelling and Simulation in Materials Science and Engineering*, vol. 27, no. 3, Mar. 2019, doi: 10.1088/1361-651X/ab08c3.

Measuring what you don’t know…??





A. Qteish, R. Resta “Thermodynamic Properties of Si-Ge alloys” *Phys.Rev. B* 1987

M. Aldegunde , N. Zabaras and J. Kristensen , "Quantifying uncertainties in first-principles alloy thermodynamics using cluster expansions," Journal of Computational Physics, vol. 323, pp. 17-44, 2016.

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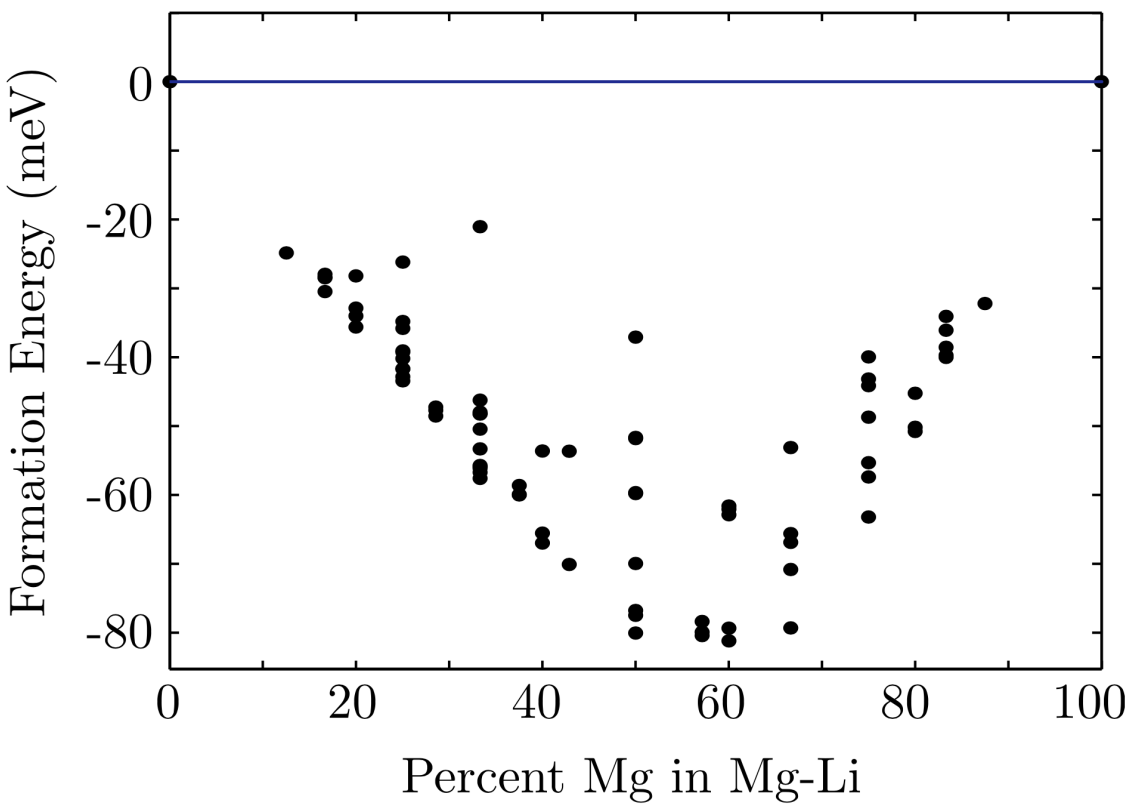

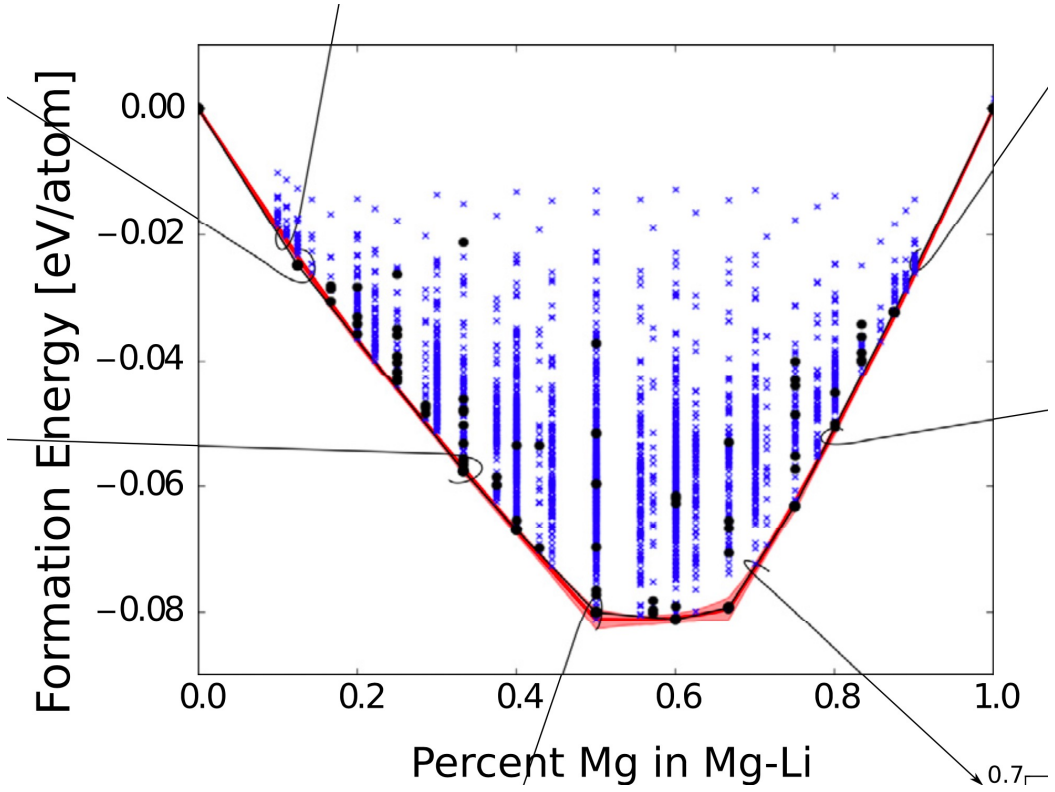

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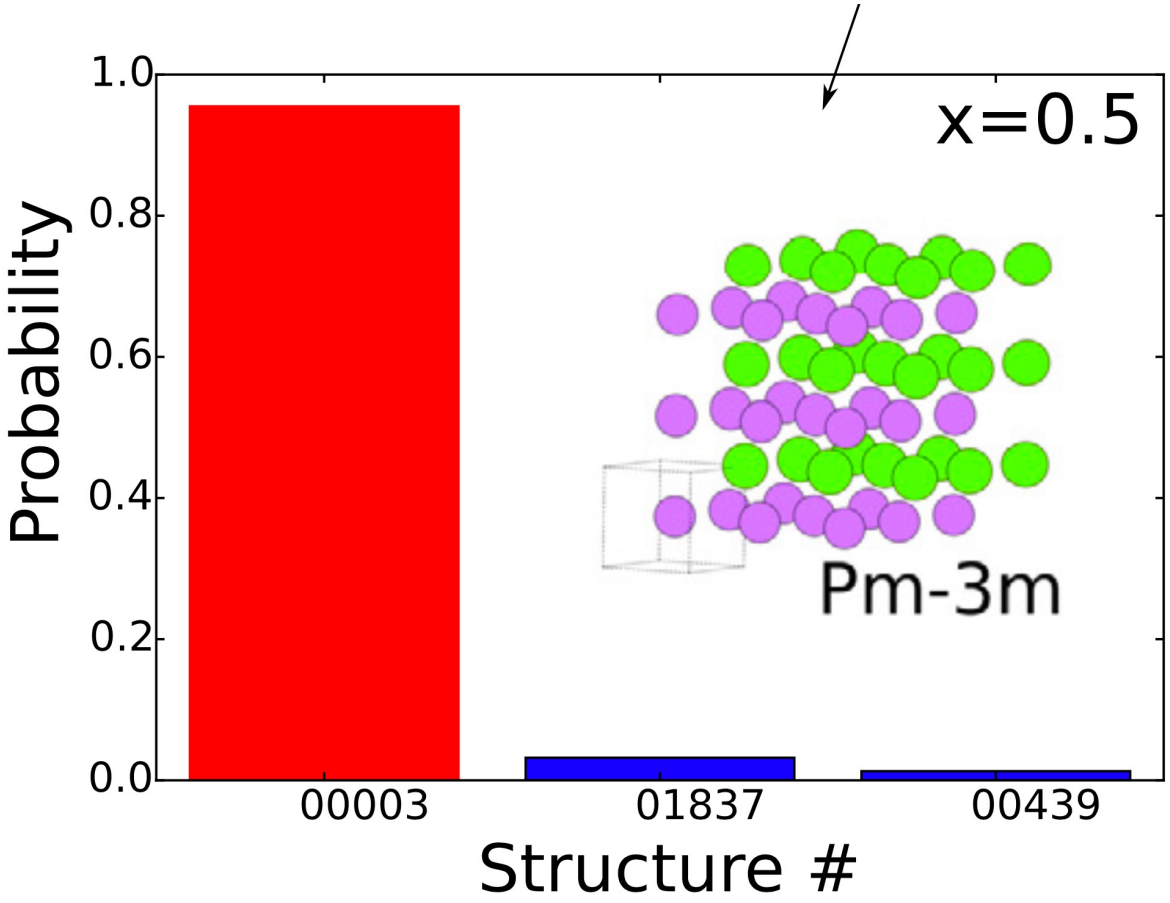

- Confidence is automatically included
- Allows surgical use of DFT

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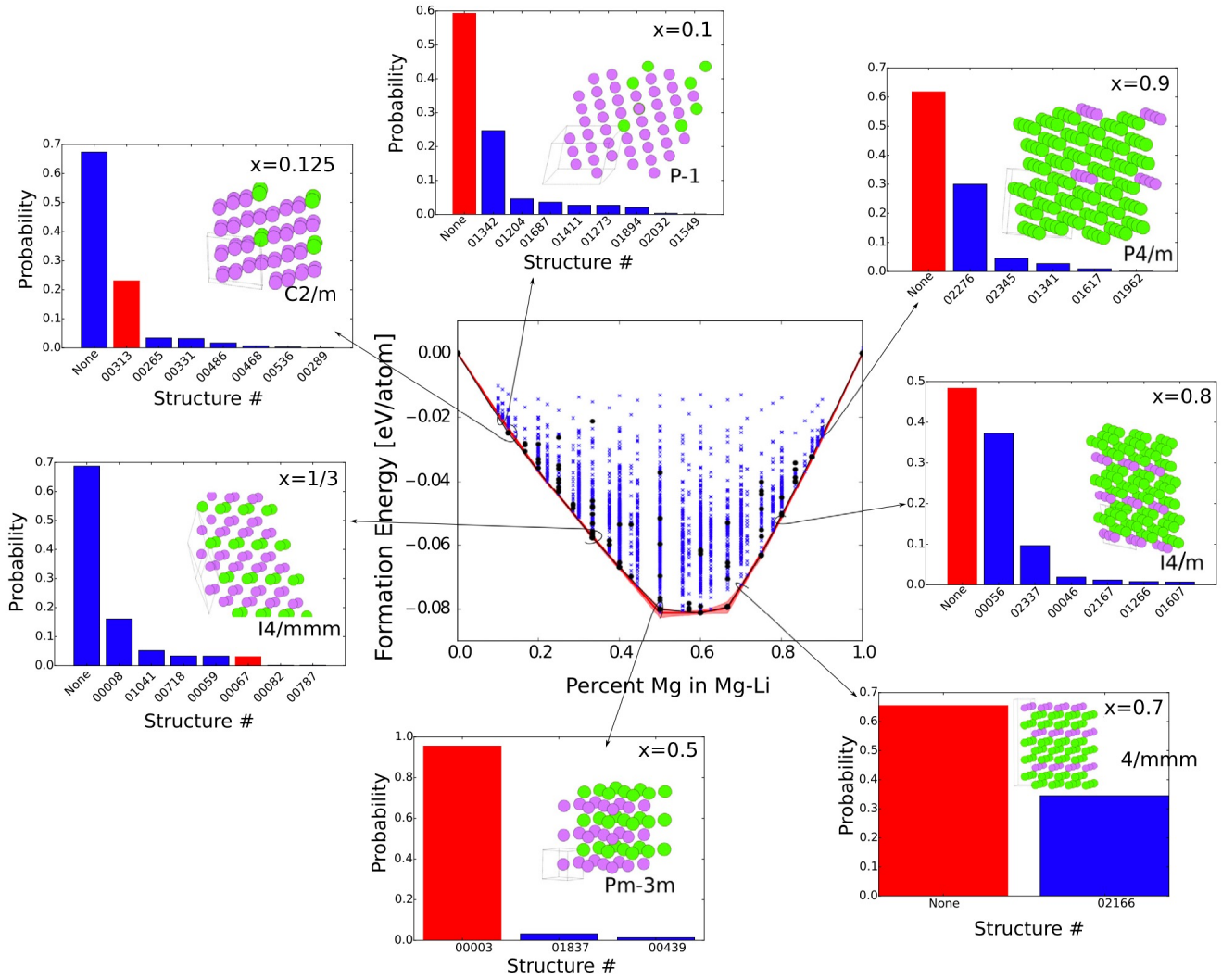

Questions?