From Structure to Properties and the Mess Between

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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

Enumerate Many arrangements

Calculate energies with accurate method (DFT)

Calculate Thermodynamic properties
Computing Structure-Property Relations

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Surrogate Model: Cluster Expansion

Calculate Thermodynamic properties

Surrogate Model: Cluster Expansion

- Fast

- Like an Ising model with more interactions

- Gives physical intuition

- Exact when using infinite terms (Think Fourier Series)
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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• **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
  • Phase Diagrams
  • 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

How to find $\vec{V}$?

- Ordinary Least Squares
- Ridge
- LASSO
Surrogate Model: Cluster Expansion

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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

\[
P(q|d_0) = \frac{P(d_0|q) \times P(q)}{\text{Normalization Constant}}
\]

Measuring what you don’t know…??


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- Confidence is automatically included
- Allows surgical use of DFT
Measuring what you don’t know…??
Questions?