Encoding locality in an alloy cluster expansion for more efficient atomistic simulations

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Summary

- First-principles simulations of alloys are expensive
- Cluster expansion techniques are effective in circumventing these costs, but do not explicitly encode the locality, recomputing redundant interaction energies during Monte Carlo simulations
- We reformulate the cluster expansion to explicitly encode locality by incorporating the system's topology into feature representation
- This encoding allows us to avoid the computation of redundant interaction energies
- New formulation shows excellent accuracy in the TaW binary system

Introduction

- First-principles simulations are expensive, becoming intractable for systems exceeding a thousand atoms
- Cluster expansion techniques are effective in circumventing these computational costs for solids
- Can very accurately reproduce phase diagrams using Monte Carlo (MC) simulations
- During an MC simulation, only small changes are made to configurations in each step, meaning energy differences should be strictly local
- Current formulations of the cluster expansion do not encode locality of interatomic interactions
- Existing tools, such as ICET [1] and ATAT [2], often recompute redundant interaction energies during Monte Carlo simulations

Standard Cluster Expansion Method

- Calculates enthalpy of mixing as a function of atomic clusters

$$\Delta H_{\text{mix}}(\boldsymbol{\sigma}) = \sum_{\omega} m_{\omega} J_{\omega} \Pi_{\omega}(\boldsymbol{\sigma})$$

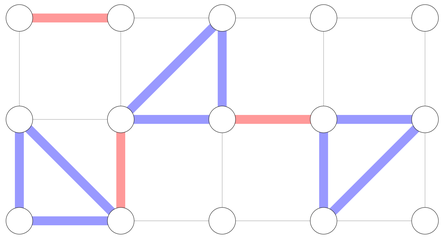

Figure 1:Heuristic visualization of cluster expansion method

Encoding System Topology

$$A_{ij}^{(n)} = \begin{cases} 1 & i \text{ and } j \text{ are } n\text{'th neighbors} \\ 0 & \text{else} \end{cases}$$

$$B_{ijk}^{(n)} = \begin{cases} 1 & i,\ j, \text{ and } k \text{ are } n\text{'th order triplet} \\ 0 & \text{else} \end{cases}$$

$$X_{i\alpha} = \begin{cases} 1 & \text{site } i \text{ occupied by type } \alpha \\ 0 & \text{else} \end{cases}$$

$$\begin{aligned} \mathcal{H}_{\text{eff}}(\mathbf{X}) &= \text{2-body interactions} \\ &\quad + \text{3-body interactions} + \cdots \\ &= \frac{1}{2!}\varepsilon_{\alpha\beta}^{(n)} \underbrace{A_{ij}^{(n)} X_{i\alpha} X_{j\beta}}_{\alpha\text{-}\beta \text{ interaction } N_{\alpha\beta}^{(n)}} \\ &\quad + \frac{1}{3!}\mu_{\alpha\beta\gamma}^{(n)} \underbrace{B_{ijk}^{(n)} X_{i\alpha} X_{j\beta} X_{k\gamma}}_{\alpha\text{-}\beta\text{-}\gamma \text{ interaction } M_{\alpha\beta\gamma}^{(n)}} + \cdots \end{aligned}$$

- Can learn interactions from an atomistic method like DFT
- Linear in interaction coefficients $\varepsilon_{\alpha\beta}^{(n)}$ and $\mu_{\alpha\beta\gamma}^{(n)}$
- Easy linear regression problem
- $\mathbf{A}$ and $\mathbf{B}$ are static and very sparse - only compute once per supercell!

Energy Difference Shortcut

$$\begin{aligned} \Delta\tilde{N}_{\alpha\beta}^{(n)} &= \sum_{d\in\mathcal{D}} A_{dj}^{(n)} X'_{d\alpha} X'_{j\beta} - \sum_{d\in\mathcal{D}} A_{dj} X_{d\alpha} X_{j\beta} \\ \Delta N_{\alpha\beta}^{(n)} &= 2\Delta\tilde{N}_{(\alpha\beta)}^{(n)} \\ \Delta\tilde{M}_{\alpha\beta\gamma}^{(n)} &= \sum_{d\in\mathcal{D}} B_{djk}^{(n)} X'_{d\alpha} X'_{j\beta} X'_{k\gamma} - \sum_{d\in\mathcal{D}} B_{djk}^{(n)} X_{d\alpha} X_{j\beta} X_{k\gamma} \\ \Delta M_{\alpha\beta\gamma}^{(n)} &= 3\Delta\tilde{M}_{(\alpha\beta\gamma)}^{(n)} \end{aligned}$$

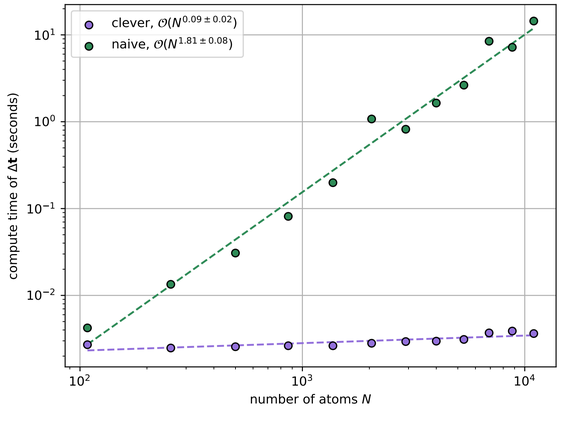

Figure 2:Feature vector difference calculation with and without shortcut

Case study: TaW

- Fit standard cluster expansion using DFT data
- Run Monte Carlo (MC) on traditional cluster expansion with ATAT
- Calculate DFT energies for configurations from MC run
- Use DFT energies in our cluster expansion

Results

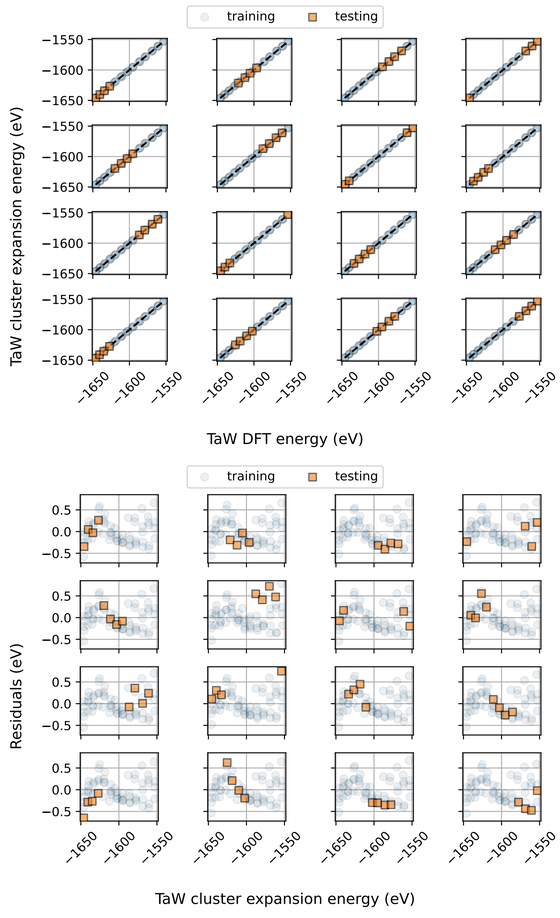

Figure 3:Cross-validation of our cluster expansion model for TaW. Parity plots showing DFT vs. cluster expansion energies (top) and residual plots showing model error (bottom) for various training and testing splits

Conclusion

- The cluster expansion technique is very effective for bypassing expensive DFT calculations. Current formulations do not explicitly encode system topology in the effective Hamiltonian. As a result, popular implementations of the technique recompute redundant energies when calculating energy differences.
- We have reformulated the cluster expansion technique to encode system topology, allowing us to avoid redundant energy computations, yielding a quadratic speedup in computation time. Preliminary cross-validation computations on the TaW system showcase that the model is very accurate.

Future Work

- More alloy systems
- 4-body terms
- Saddle points for kinetics! (very difficult)

References

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