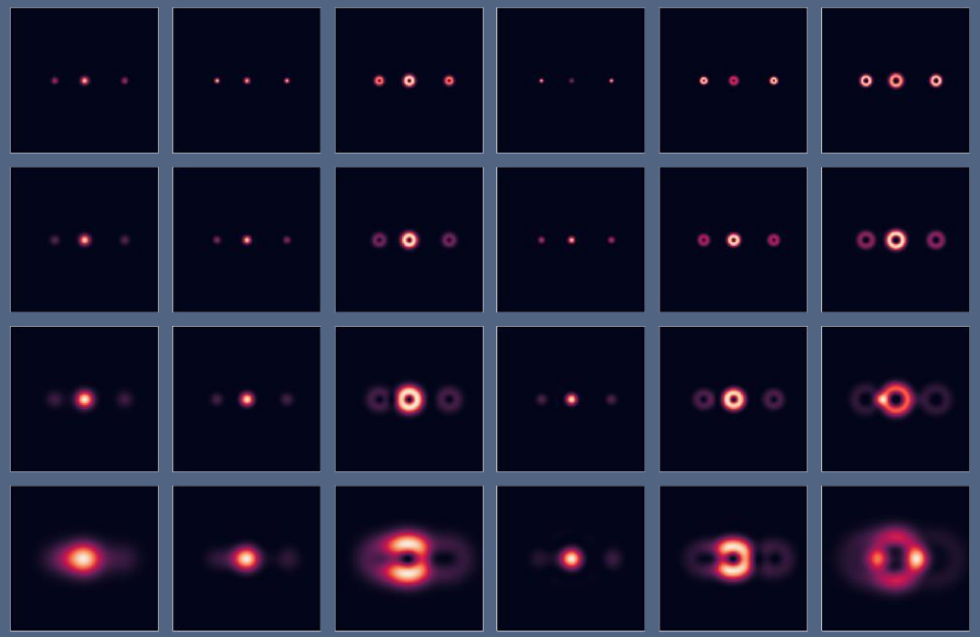
Applying Harmonic Analysis to Predict Quantum Energies and Forces



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ACRES
Advanced Computational Research Experience for Students

MICHIGAN STATE UNIVERSITY

Introduction and Motivation

- A majority of computational chemistry depends on finding Energy and Force within a system
  - Drug development
  - Materials research
  - Battery technology

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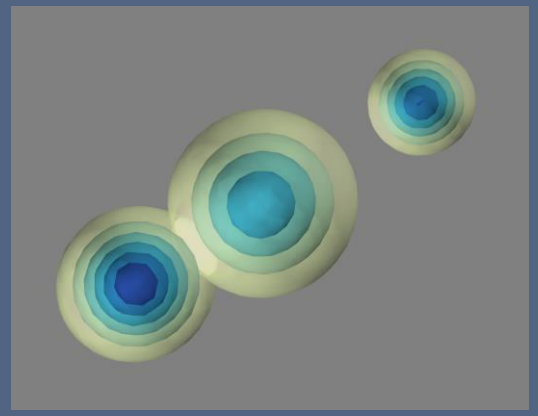
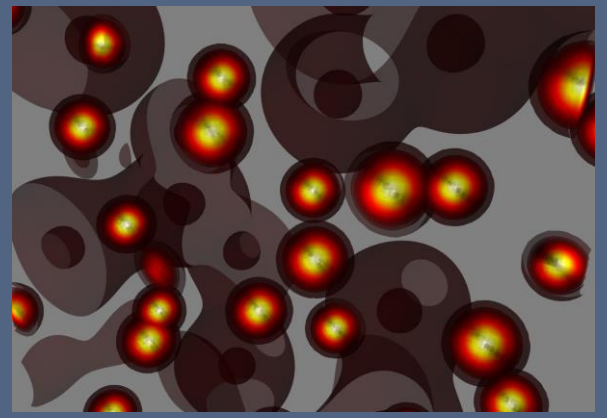
- Simulation methods present a trade-off
- We are building an interpretable learning model that attempts to circumvent the trade-off

The Data

- 625 LiOLi molecules
- 80,000 amorphous periodic Li-Si structures at various concentrations
- Truth values calculated via VASP



Build Model on Simple Data



Eventually Generalize to Complex Data

Why Use Harmonic Analysis

Our data is well behaved, Energy and Forces are:

1. Invariant under isometries
2. Invariant under re-indexing of atoms
3. Continuous under small deformations

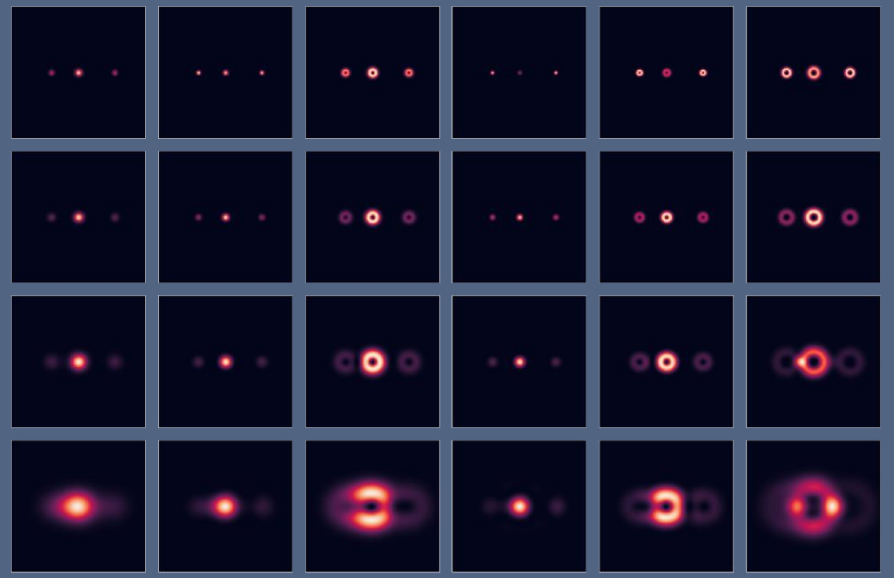
We should take advantage of these facts

Construct features $\vec{\phi}$ which respect physics

A Closer Look at $\vec{\phi}$

Given a state, each feature is based on:

1. Atom positions and types
2. A specific spherical harmonic
3. Scaling parameters (range of interaction)



Example features prior to integration

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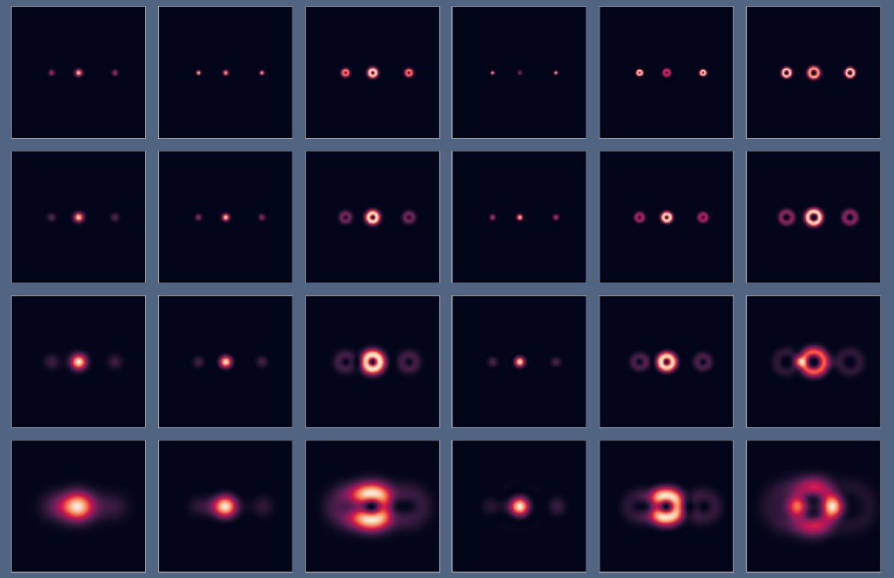

Example features prior to integration

$$\phi_{j,n,l,q}(R) = \int_{\mathbb{R}^3} \left(\sum_{m=-l}^{l} |\rho_R * \Psi_{j,n,l}^m|^2 \right)^{q/2}$$

Energy and Forces

Regressing upon these features leads to a good energy model [1]

$$R \rightsquigarrow \vec{\phi} \rightsquigarrow \tilde{E} = \sum_{\lambda} w_{\lambda} \phi_{\lambda}$$

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Note: $F = -\nabla E$

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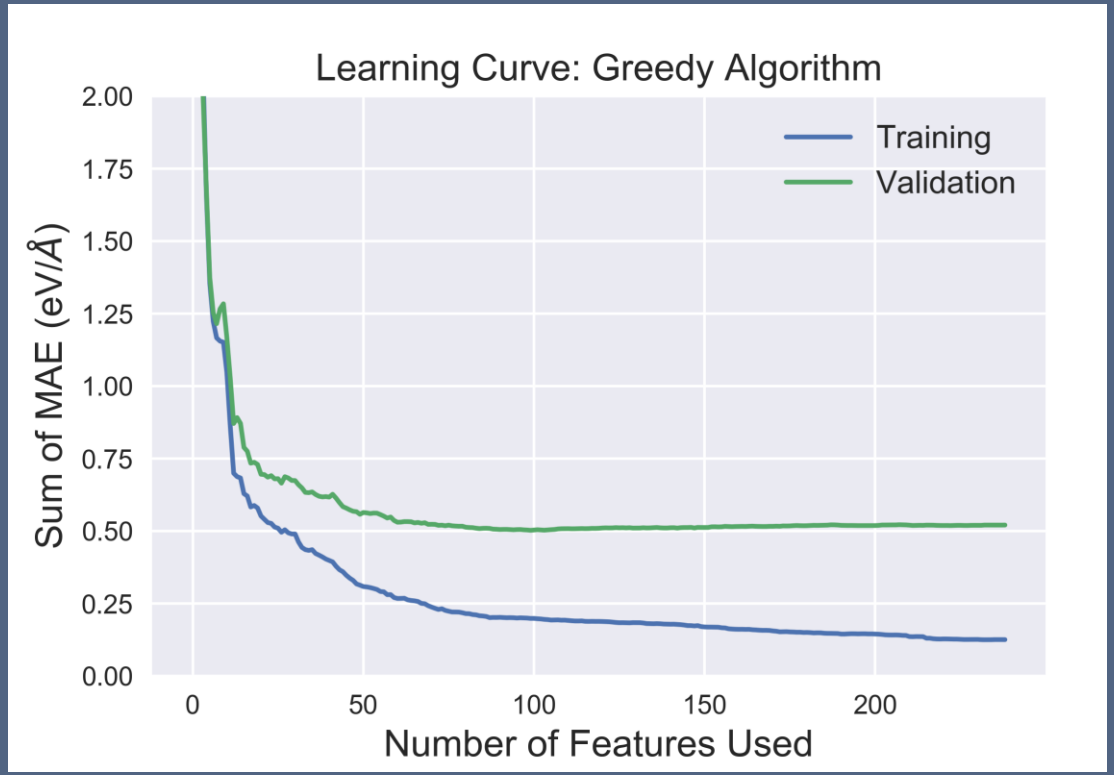
So we hope: $\tilde{F} = -\nabla \tilde{E} = -\sum_{\lambda} w_\lambda \nabla \phi_\lambda$

Results on LiOLi Data

LiOLi Data:

- 625 molecules
- Each force vector has 9 components with units eV/Å

We use a greedy algorithm to select the most influential features first



Future Plans

- Hope to tune model to reduce the error
  - Goal is less than 0.02eV/Angstrom/Force Direction
- Calculate energy and force concurrently
- Move towards Li-Si data set

References

[1] M. Hirn, S. Mallat, and N. Poilvert, Multiscale Model. Simul. **15**, 827 (2017).

[2] M. Eickenberg, G. Exarchakis, M. Hirn, S. Mallat, NIPS **30**, 6543 (2017).