Improving Quantum Mechanical Calculations Using Machine Learned Electron Densities and Forces

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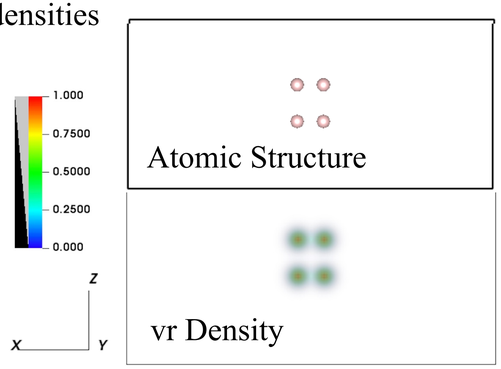
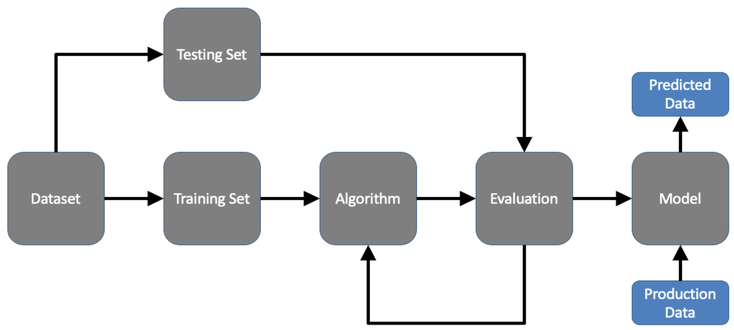
Introduction

Density Functional Theory (DFT) calculations have been used for decades to predict chemical properties of compounds and materials. While DFT calculations are highly chemically accurate, they are computationally costly to perform for systems containing many atoms. On the other hand, classical mechanics based molecular dynamics (MD) simulations can be performed at a larger scale and can model thousands of atoms, albeit with less chemical accuracy. Using machine learning to predict atomic forces and map atomic positions to an electron/energy density will improve the accuracy of molecular dynamics simulations and will allow for faster prediction of material properties.

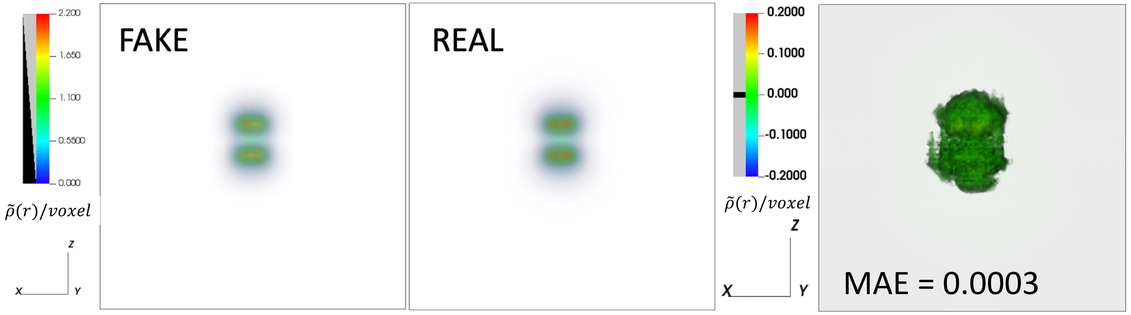
Objectives

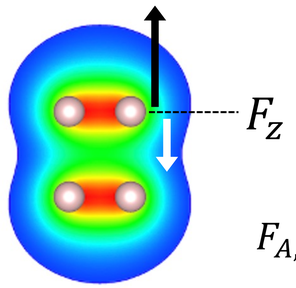
- Develop method of calculating forces from electron density data
- Generate electron density data for hydrogen molecules and train a machine learning algorithm
- Predict forces for machine learned electron densities





Predicted Electron Density for H4





Hellmann-Feynman Force Equation

$$F_{A,x} = Z_A \left(\int \rho(r) \frac{x - x_A}{|r - R_A|^3} dr - \sum_i^m Z_i \frac{x_i - x_A}{|r_i - R_A|^3} \right)$$

Calculated Forces on Real H2 Molecule

Forces in x Direction (Cubic Grid)

Position	Calculated (Eh/bohr)	True (Eh/bohr)	Error
1	0.116597914	0.09805471	0.018543204
2	-0.143777539	-0.09805471	-0.045722829

Forces in x Direction (Radial Grid)

Position	Calculated (Eh/bohr)	True (Eh/bohr)	Error
1	0.125029732	0.09805471	0.026975021
2	-0.125029732	-0.09805471	-0.026975021

Conclusions

- Electron densities for hydrogen molecules are capable of being accurately predicted from vr densities using machine learning
- Integration of the charge density must be improved to decrease the error in calculated forces

Future Work

- Improve force calculations using either better numerical integration or analytical integration of the charge density
- Calculate forces from machine learned electron densities for H2 as opposed to real densities
- Incorporate force calculations into molecular dynamics and geometry optimization algorithms

Acknowledgements

- ASU FURI
- Dr. Christopher Muhich
- Muhich Lab Group

FURI

$\hat{H}|\Psi\rangle = E|\Psi\rangle$

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