Ab-Initio Accuracy at Large Length and Time Scales for Lithium Metal Simulations

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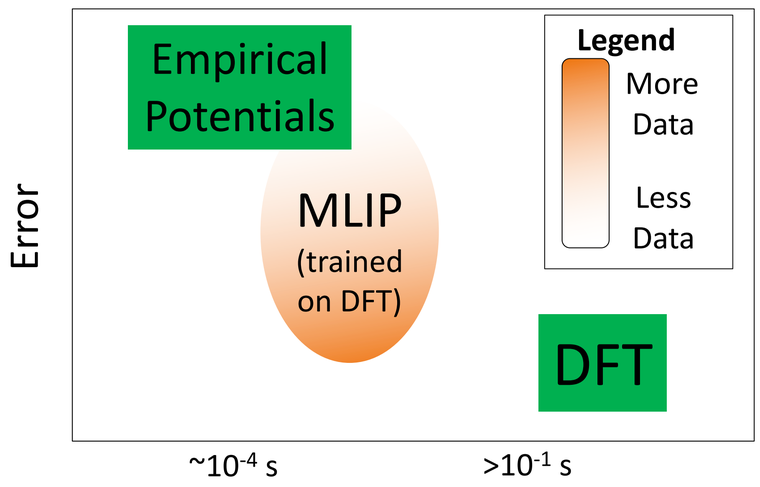
MOTIVATION

Material properties such as those of lithium metal are key parameters engineering applications. Often, they are difficult to probe experimentally due to the contamination, complex microstructures, lack of characterization methods etc. Computationally, there is a lack of quantitatively accurate empirical potentials while more accurate Density Functional Theory (DFT) calculations are too costly. Machine Learning Interaction Potentials trained on DFT can achieve both high accuracy and computational efficiency. In this work, we generate data and train three MLIPs for lithium metal with DFT data and find that equivariant MLIPs are the most accurate. We then test the ability of Allegro, a MLIP implementation that can scale over multiple GPUs on Google Cloud to push the boundaries of simulation accuracy, size and timescale in Molecular Dynamics (MD) simulations at Ab-Initio accuracy.

METHODS

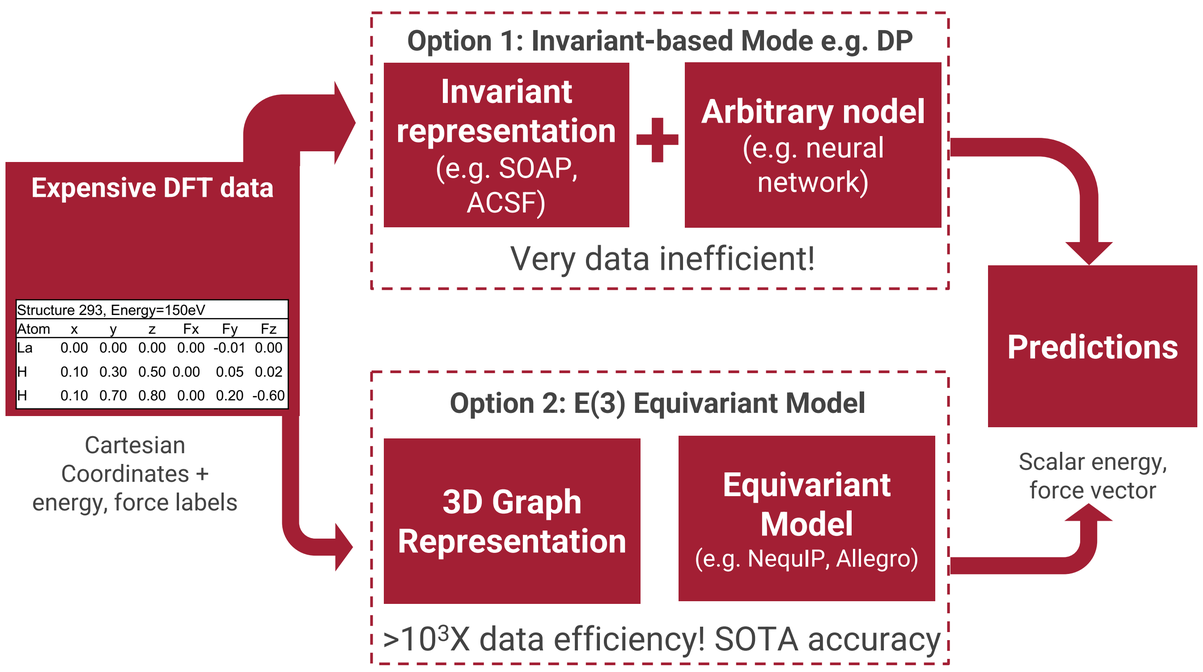
Machine Learning Interaction Potentials (MLIPs)

MLIPs map atomic structure to a potential energy surface for Molecular dynamics (MD) simulations used to predict material properties



Empirical potentials are inaccurate, DFT is limited to ~400atoms and <0.015ns/day, MLIPs give both speed and accuracy

There are two classes of MLIPs based on how they represent geometric data



NequIP and Allegro are E(3) equivariant MLIP matching SOTA accuracy on multiple materials benchmarks and Allegro is parallelizable across GPUs

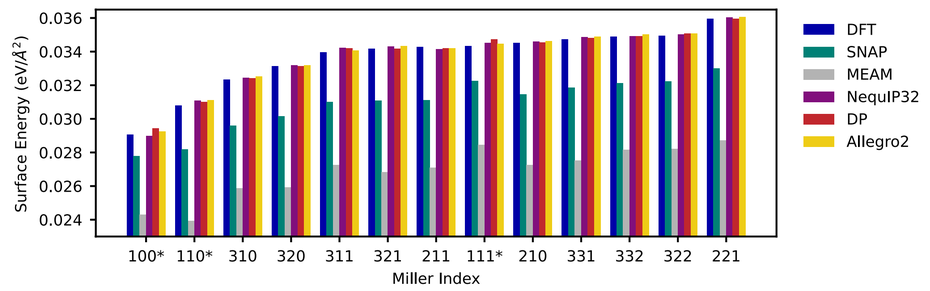
Allegro Implementation [2]

- E(3) Equivariant model
- Matches SOTA data efficiency and generalization of NequIP
- Fully local and parallelizable
- Integrated with LAMMPS for Molecular Dynamics

TRAINING AND SIMULATION RESULTS

We trained 3 MLIPs. NequIP has best accuracy, Allegro models perform similarly well. Deep Potential (Invariant) is also very good

Model	Energy RMSE (meV/atom)	Force RMSE (meV/\AAA˚)	Stress RMSE (meV/\AA^3A˚3)
Deep Potential	3.4	20.2	1.4
NequIP32	1.2	12.1	0.4
Allegro2	4.3	25.0	0.8



Equivariant MLIPs have much higher data efficiency and accuracy but are slower and more memory intensive than invariant MLIPs hence the need for parallelization on GPUs

Num. of A100 GPUs	Max num. of atoms	ns/day	Timesteps/s
1	27,648	0.222	1.286
2	59,582	0.207	1.201
4	118,638	0.207	1.200
8	182,250	0.219	1.268
16	476,656	0.201	1.161

MD simulations previously impossible to do with DFT can now be done at DFT accuracy at much larger length and time scales using NequIP and Allegro

Simulations we have done:
~476,000 atoms on 16 A100 GPUs

Future simulation:

ACKNOWLEDGEMENTS

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