Abstract

Machine learning (ML) force field models for molecular dynamics (MD) simulations often suffer from poor system stability with instabilities such as atom clustering that must be corrected by active learning approaches. However, the correlation between the structural and chemical complexity of a multi-component systems and the robustness of long-time ML-based MD dynamics has not been studied in detail. We develop graph neural network (GNN) model for SiC and GeSe₂ systems to perform classical MD simulations with quantum mechanical accuracy. A GNN model is sufficient to ensure robust long-time dynamics in a ‘simple’ system like SiC. However, we need additional inductive bias, in the form of energy decomposition into 2-body and 3-body terms to generate stable MD trajectories for complex GeSe₂ systems, which can exist in multiple metastable atomic configurations.

Molecular Dynamics Simulation

- Ab-initio molecular dynamics (AIMD), which computes interatomic forces quantum mechanically is commonly used to study materials properties.
- However, AIMD is expensive and scales as $O(N^2)$ with the number of electron
- Classical molecular dynamics simulation (MD) is commonly used to simulate large systems, which scales linearly with number of atoms and models each atom as point mass with point charge.

**Goal**

- Design a Graph Neural Network (GNN) based potential function for MD simulation at AIMD level accuracy
- Incorporate inductive bias - two-body and three-body interaction - into GNN model for robust MD dynamics

Inductive Bias Graph Network for Classical MD Simulation

$$E = E_{GNN}(R_1, R_2, \ldots, R_N) + \sum_{i=1}^{N} \sum_{j=k \in \mathcal{N}_R} E_{NN}(R_i, R_j) + \sum_{i=1}^{N} \sum_{j=k \in \mathcal{N}_{GNN}} E_{NN}(R_i, R_k, R_j, \cos \theta_{ik})$$

- $E_{NN}$ and $E_{NN}$ uses cutoff distance of $R_c = 3\sigma$ for neighbor list ($N_{NN}$), whereas cutoff of $E_{GNN}$ is $7\sigma$ in GNN layers.
- Hierarchical training strategy is used for $E_{NN}$ and $E_{GNN}$.
- MD simulation is performed with velocity-Verlet algorithm in NVT ensemble.
- Atomic coordinates are updated as $R_k(t + \Delta t) = R_k(t) + v_{ik}\Delta t$. Here, $v_{ik}$ is updated using $a_{ik} = F_{ik}/m_{ik}$. 

**Conclusions**

- Developed a GNN model to perform classical MD simulations with quantum mechanical accuracy.
- For a ‘simple’ system like SiC, a GNN model is sufficient to have robust long time MD dynamics.
- For complex systems such as GeSe₂ with multiple metastable points in its potential energy surface, decomposition of total energy into 2-body, 3-body and many-body (GNN) terms is needed to create models that generate stable MD trajectories.