INDUCTIVE BIAS GRAPH NETWORK FOR ROBUST MOLECULAR DYNAMICS

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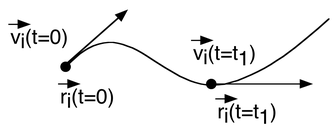
Abstract

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 $GeSe_2$ 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 $GeSe_2$ 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 $O(N^3)$ 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.**



Solve Newton's equation of motion

$$\vec{F}(t) = m\frac{d^2\vec{r}_i}{dt^2} = -\frac{d}{d\vec{r}_i}V(\vec{r}_i \dots \vec{r}_N)$$

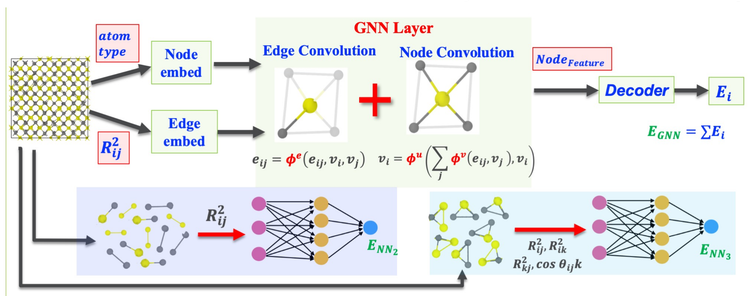
V = potential function, non-trivial and system specific

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

$$E = E_{GNN}(R_1, R_2, \dots, R_N) + \sum_{i=1}^{N}\sum_{j\in NN_{R_c}} E_{NN_2}(R_{ij}^2) + \sum_{i=1}^{N}\sum_{j\neq k; j,k\in NN_{R_c}} E_{NN_3}(R_{ij}^2, R_{ij}^2, R_{jk}^2, \cos\theta_{ijk})$$

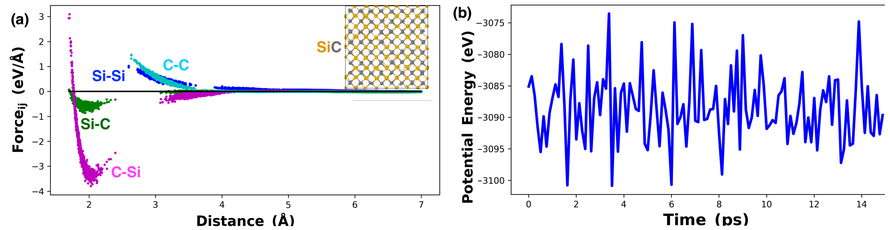


$$Loss = p_e\frac{1}{M}\sum_{i=1}^{M}\frac{1}{N_i}(E_{AIMD} - E)^2 + p_f\frac{1}{M}\sum_{i=1}^{M}\sum_{j=1}^{N}\frac{1}{N_i}\sum_{k}^{3}(F_{ijk,AIMD} - F_{ijk})^2$$

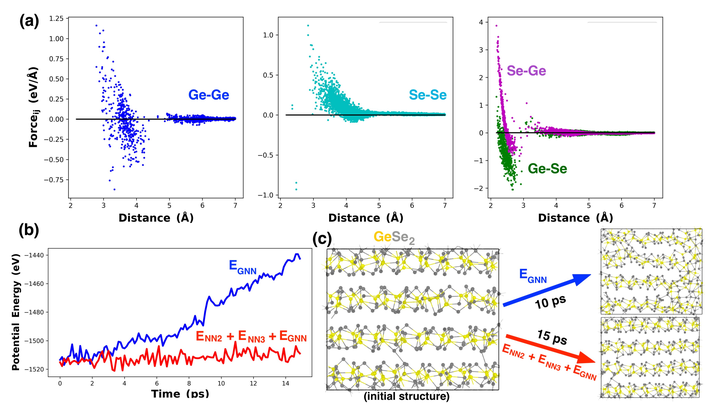
- E_{NN_2} and E_{NN_3} uses cutoff distance of $R_c = 3$Å for neighbor list (NN_{R_c}), whereas cutoff of $R_{GNN} = 7$Å is used in GNN layers.
- Hierarchical training strategy is used for E_{NN_2}, E_{NN_3} and E_{GNN}.
- MD simulation is performed with velocity-Verlet algorithm in NVT ensemble.
- Atomic coordinates are updated as $R_{ik}(t+\delta t) = R_{ijk}(t) + v_{ik}\delta t$. Here, v_{ik} is updated using $a_{ik} = F_{ik}/mass_i$

Results

- **For simple system such as Silicon Carbide (SiC), GNN model without any inductive bias is sufficient to learn robust potential function for MD simulation.**
  - **MD stability is dependent upon learned local interaction $F_{ij}(r_{ij})$ between atoms during training such that total force $F_i = \sum_j F_{ij}(r_{ij})$**



  - **Learned $F_{ij}(r_{ij})$ and potential energy (PE) of 512 atom SiC in MD after training.**
  - **RMSE energy on energy was 2.5 meV/atom and 2.3 meV/atom respectively, on training and test data.**
- **Germanium Selenide ($GeSe_2$) is a complex phase-change material that has multiple crystalline configuration and several non-equivalent atomic positions and oxidation state for anions.**



Energy (meV/atom)

Model	Training	Test
E_{GNN}	2.16	2.48
$E_{GNN} + E_{NN_2} + E_{NN_3}$	1.06	1.64

- **Learned $F_{ij}(r_{ij})$ and PE for 384 atom $GeSe_2$ in MD simulation using GNN along and with energy decomposition $(E_{GNN} + E_{NN_2} + E_{NN_3})$**
- **Se-Se interaction is attractive between 2 – 3Å. The attractive interaction happens between inter-layer Se atoms. Without energy decomposition, system shows Se-Se clustering in MD.**
- **Decomposing energy into 2-body and 3-body not only decreases RMSE errors but also make MD simulation stable.**

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 $GeSe_2$ with multiple metastable configurations 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.**