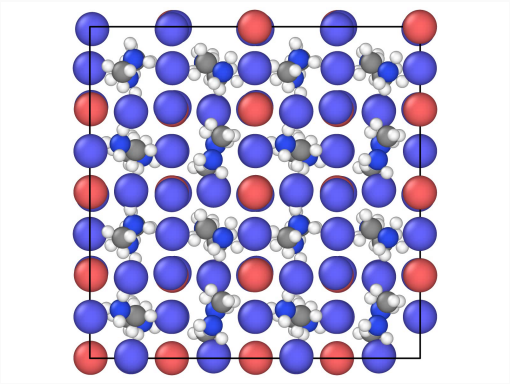
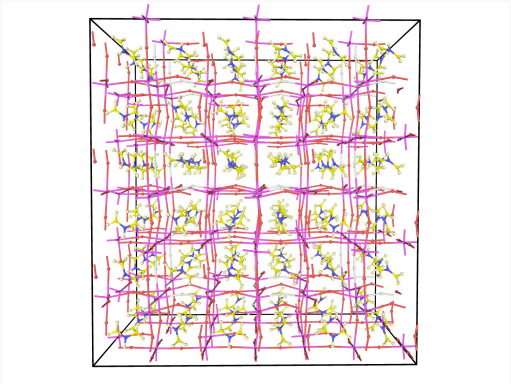




# Neural Network Potential of Complex Perovskite Materials and Their Implementation to LAMMPS

Ping-Han Tang  Sheng-Han Teng  Hsin-An Chen  Guan-Jie Chen  Chun-Wei Pao

August 30, 2019

Research Center for Applied Sciences, Academia Sinica, Taiwan

# Outline

Introduction

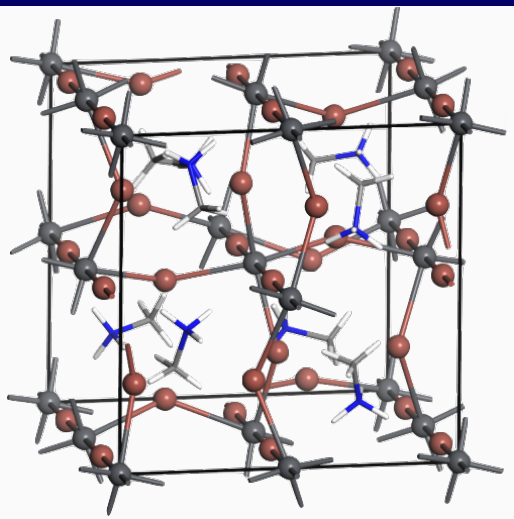
ANN Model for Energy Evaluations

Performance of ANN Potential on $MAPbI_3$ Perovskite Crystals

Implementation of ANN Potential into LAMMPS

Applications to FAMAPbIBr

# Perovskite Solar Cell Materials



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- Mixed covalent, ionic, van der Waals, and hydrogen bond interactions

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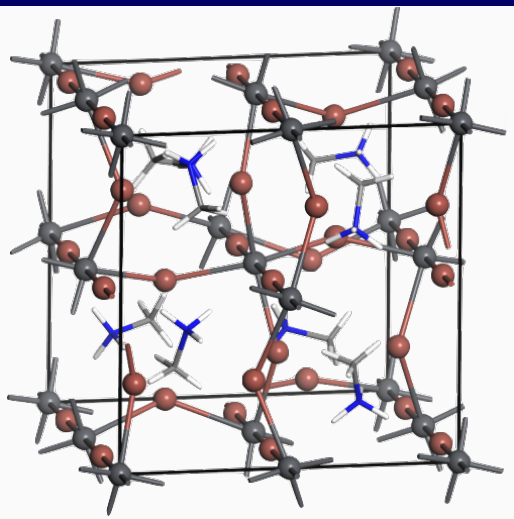
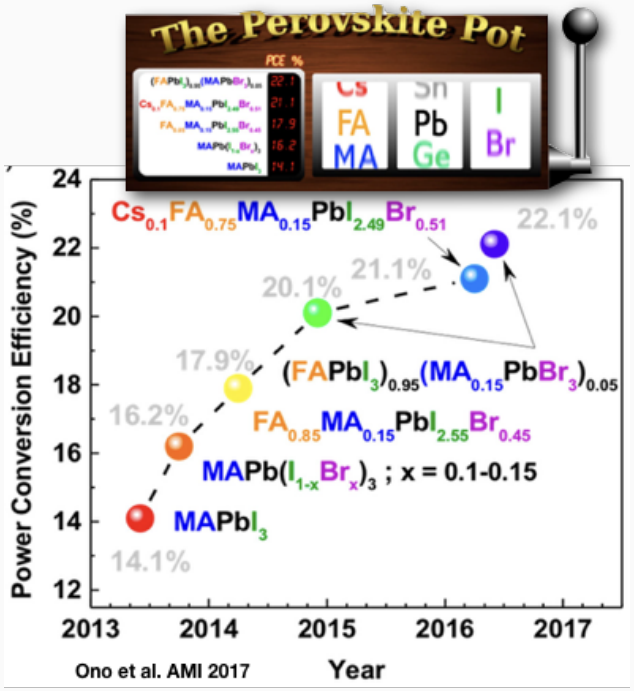


Mixed Perovskite

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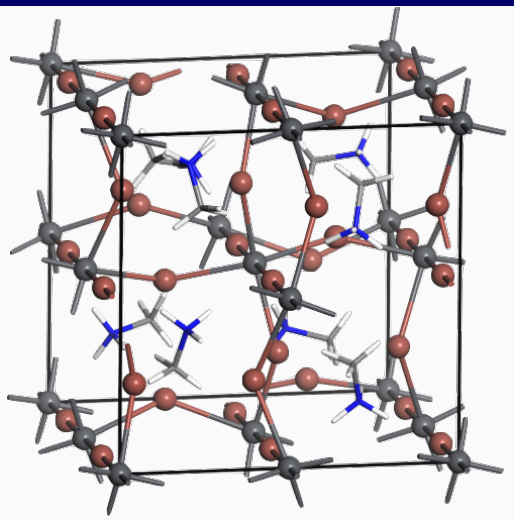
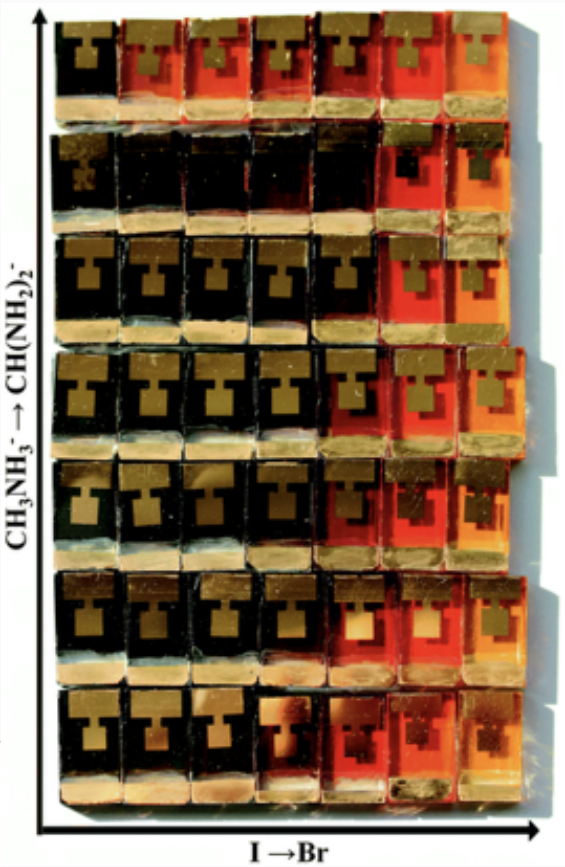
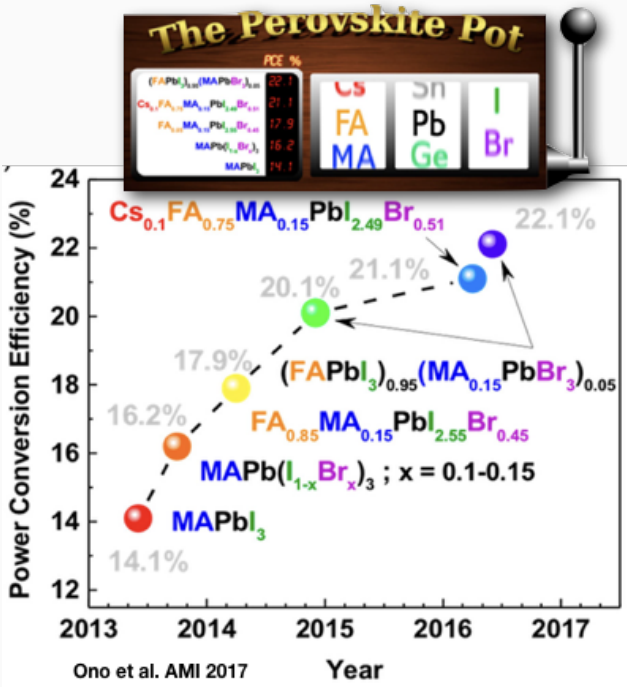
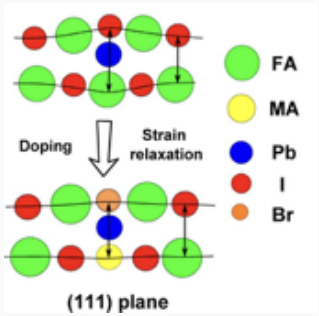





Jacobsson et al. EES 2016

**Mixed Perovskite**




Zheng et al. ACS Energy Lett. 2016

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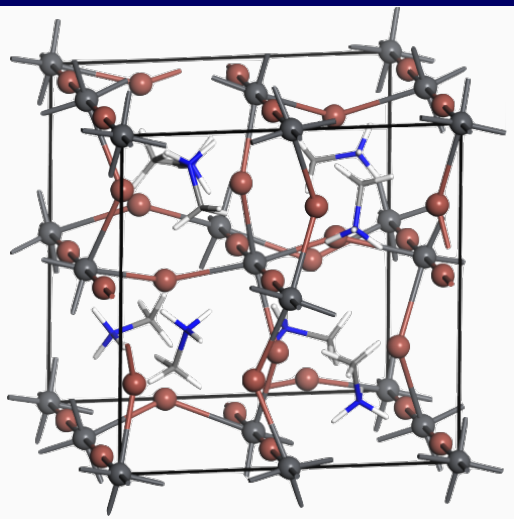
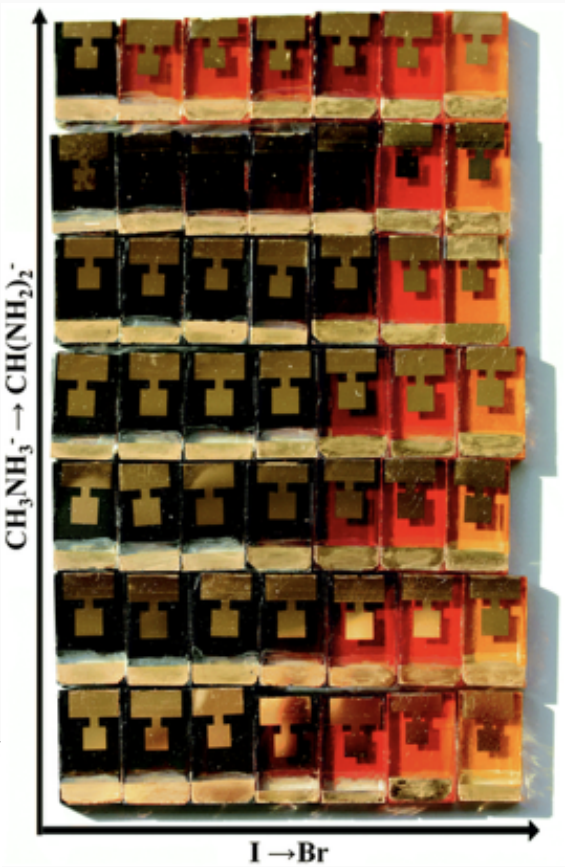
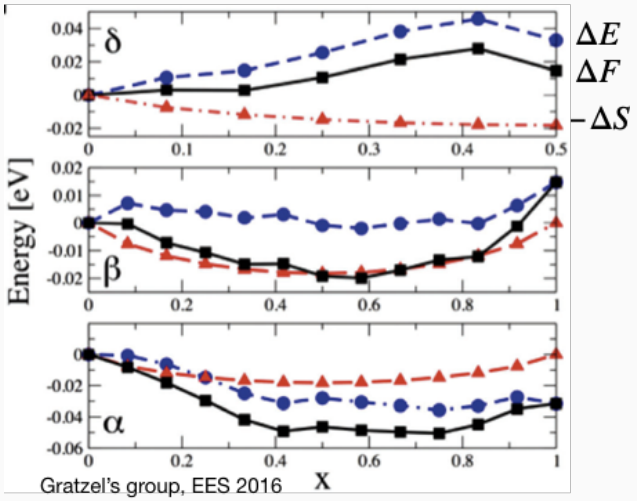





Jacobsson et al. EES 2016

**DFT calculation of $FA_xCs_{1-x}PbI_3$**




Gratzel's group, EES 2016

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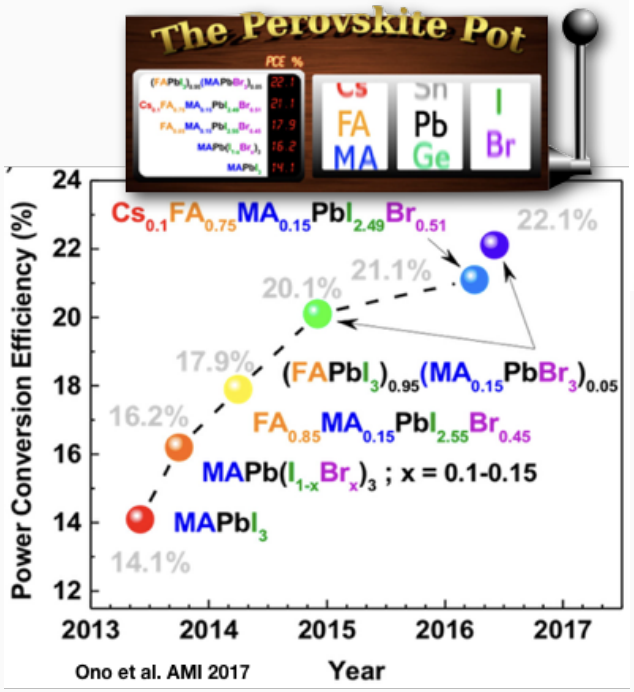
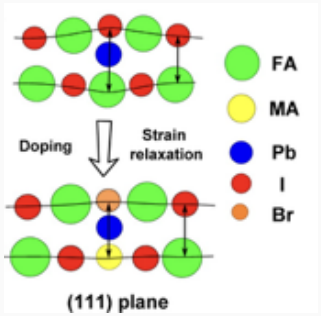


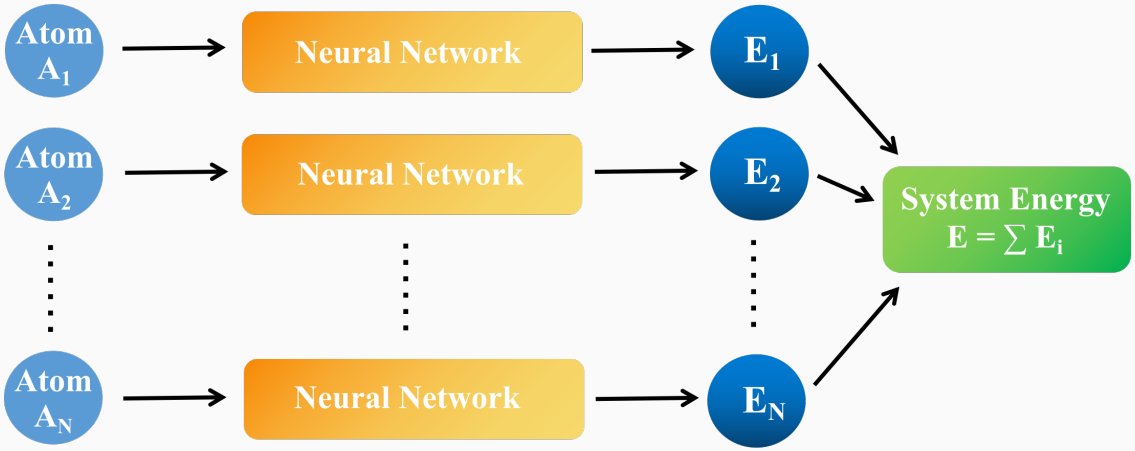

Ono et al. AMI 2017




Zheng et al. ACS Energy Lett. 2016

- Hybrid perovskite are promising photovoltaic materials
- Mixed covalent, ionic, van der Waals, and hydrogen bond interactions
- Boost in PCE and lifetime by mixing cations and halogen ions
- Morphology dependent on compositions
- DFT calculations are computationally intensive to explore the configurational space
- We develop an artificial neural network potential model to predict energy/forces with efficacy

# Energy Partitioning and Atomic Neural Network Function




The system energy is partitioned into contributions from individual atoms, namely

$$E_{sys} = \sum_i E_i$$

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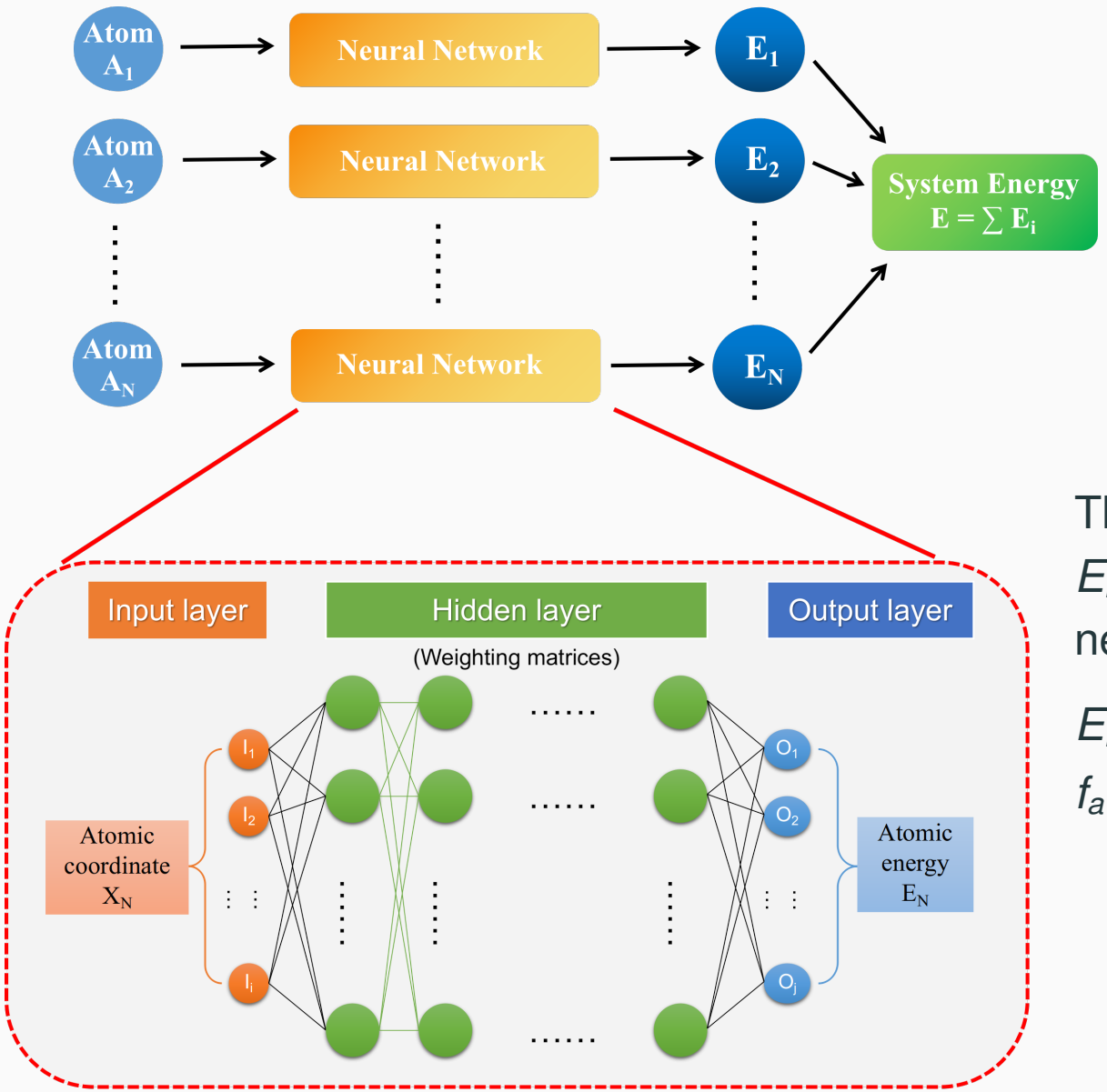


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The contribution from each atoms $E_i$ can be computed by a neural network function

$$E_i = \mathcal{N}(\mathbf{I_i}, \{\mathbf{W}\}) = f_a\Big(\mathbf{W_M} f_a\big(\mathbf{W_{M-1}} f_a(\cdots f_a(\mathbf{W_1 I})\cdots)\big)\Big)$$

- $\mathbf{I_i}$: feature vector of atom $i$
- $\{\mathbf{W}\}$: weighting parameters connecting nodes

# Feature Vectors

Feature vector $\mathbf{l_i}$ of atom *i* is comprised of a series of descriptor functions converting cartesian coordinates into fingerprints representing chemical environments. We employed the Gaussian type descriptors developed by Behler and Parrinello.

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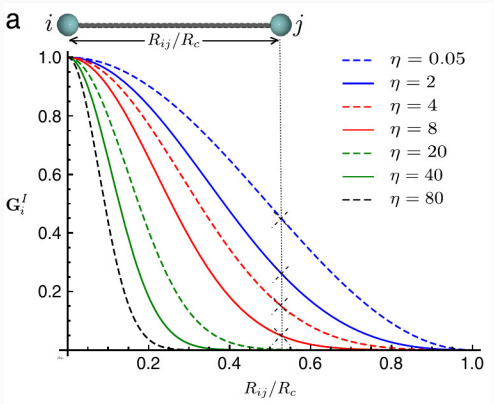
## Two-Body Descriptor

$$G_i^{II} = \sum_{j \neq i} e^{-\eta \frac{(R_{ij} - R_s)^2}{R_c^2}} f_c(R_{ij})$$




Khorshidi and Peterson, 2016

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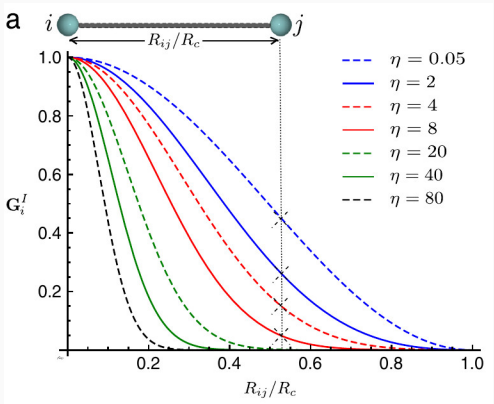
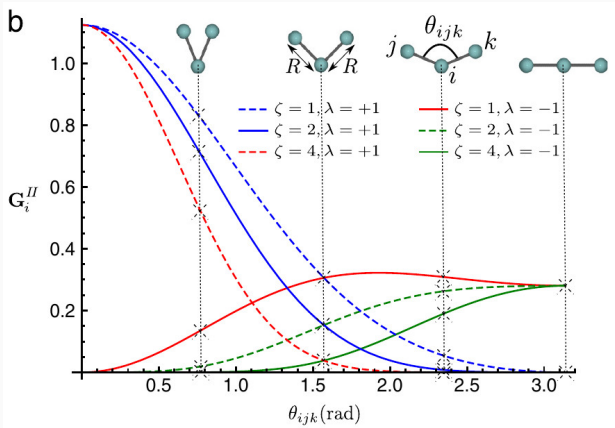



Khorshidi and Peterson, 2016

## Three-Body Descriptor

$$G_i^{III} = 2^{1-\zeta} \sum_{j,k \neq i} (1 + \lambda cos\theta_{ijk})^{\zeta} e^{-\eta \frac{R_{ij}^2 + R_{ik}^2 + R_{jk}^2}{R_c^2}} f_c(R_{ij}) f_c(R_{ik}) f_c(R_{jk})$$

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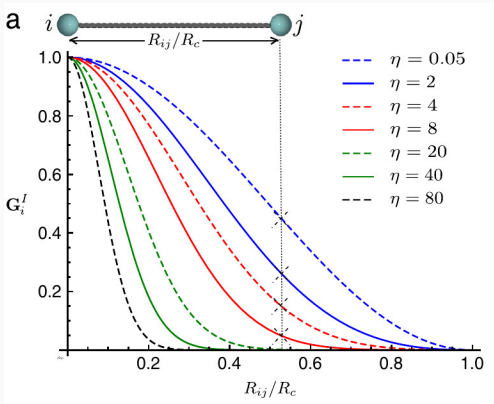


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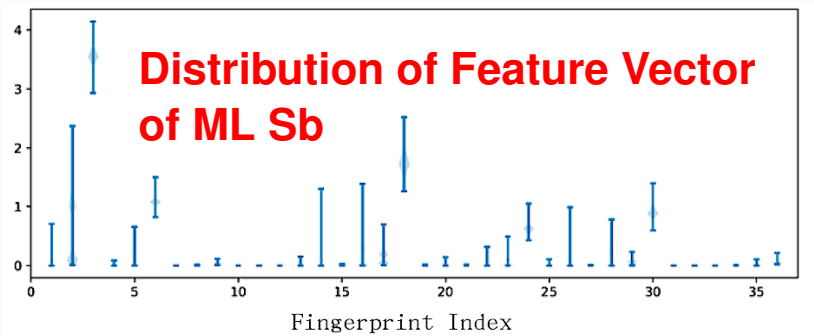


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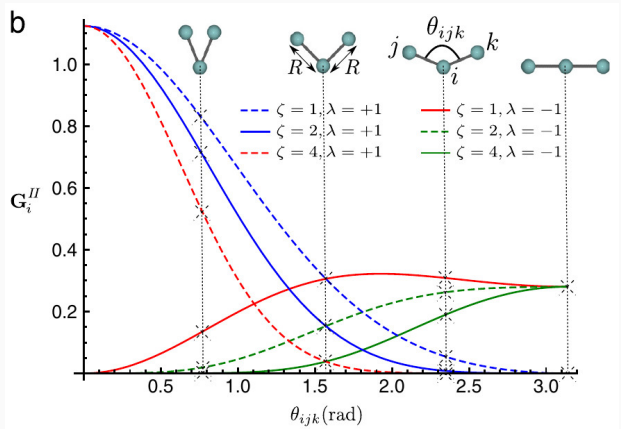


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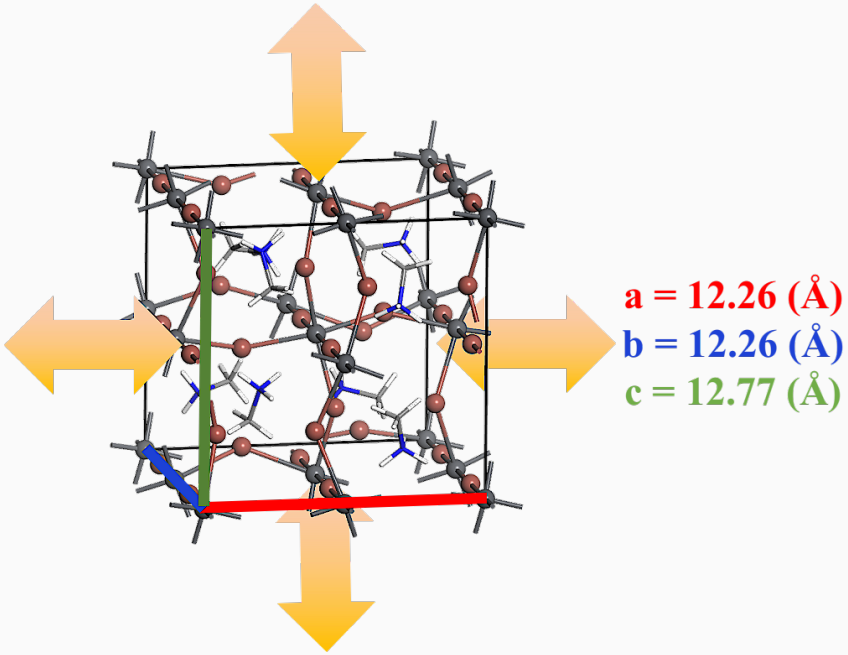
# ANN Model for Perovskite Materials




## Training Set Generation

- Tetragonal perovskite crystal subjected to hydrostatic strains $-5\%$ $+5\%$.
- AIMD using VASP, $E_{cutoff} = 400eV$, PAW-PBE with vdW correction (DFT-D3).
- Langevin NVT at 300K.
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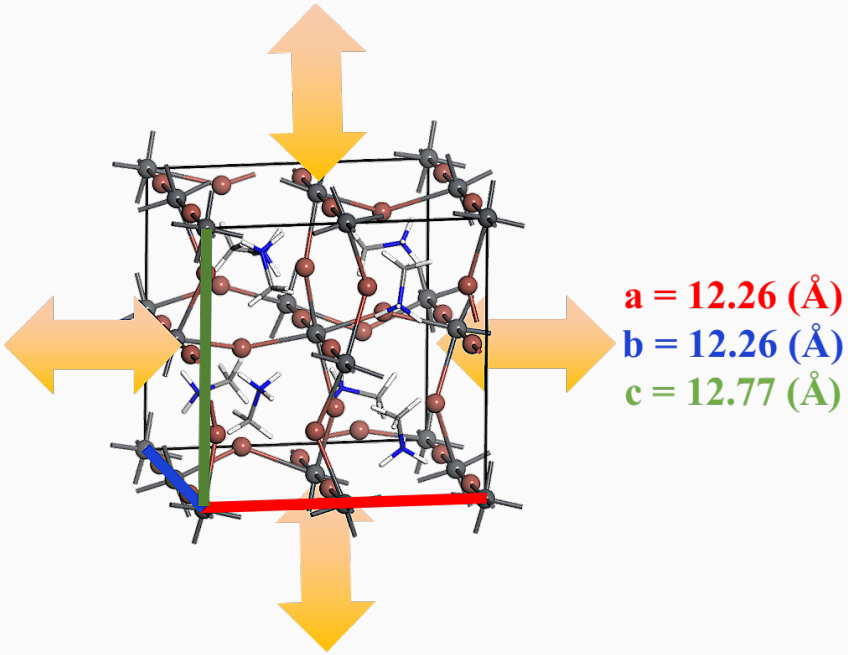


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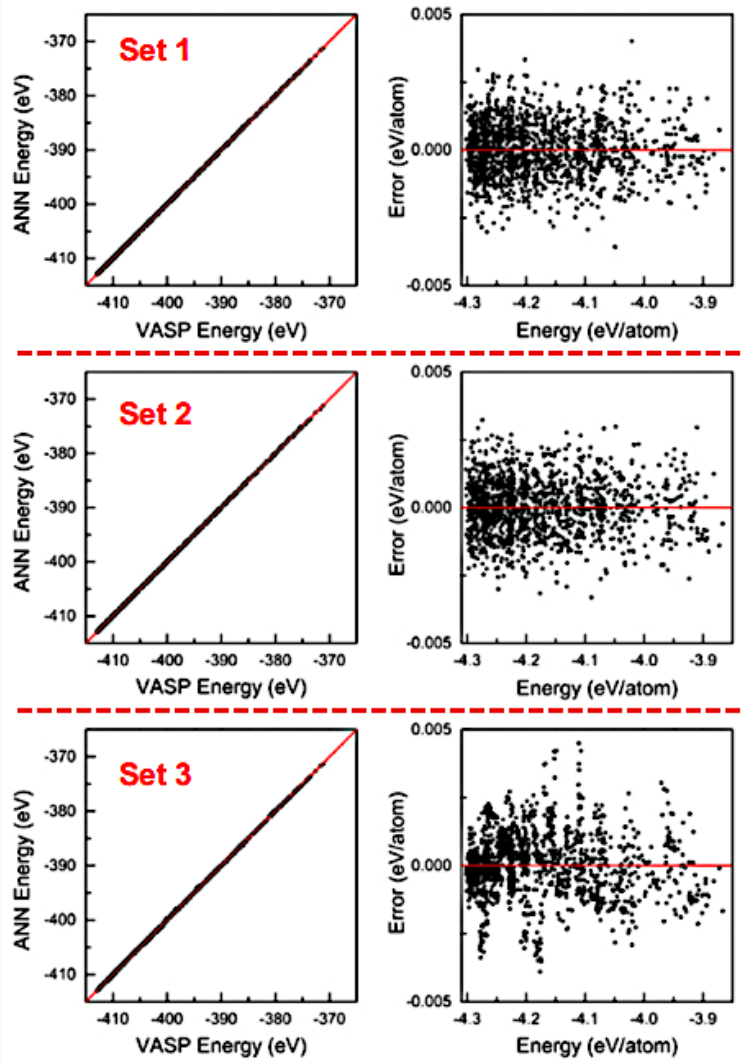
## Training Setup

| | Pb, I architecture | C, N, H architecture | Energy | Force |
|---|---|---|---|---|
| Setting 1 | $\{10, 5, 5\}$ | $\{5, 5, 5\}$ | ◯ | X |
| Setting 2 | $\{10, 5, 5\}$ | $\{5, 5\}$ | ◯ | X |
| Setting 3 | $\{10, 5, 5\}$ | $\{5, 5\}$ | ◯ | ◯ |

Force training using Tensorflow implemented in *Atomistic Machine-Learning Package*.

# Perovskite ANN Model Training Results

## Parity of Trained Model



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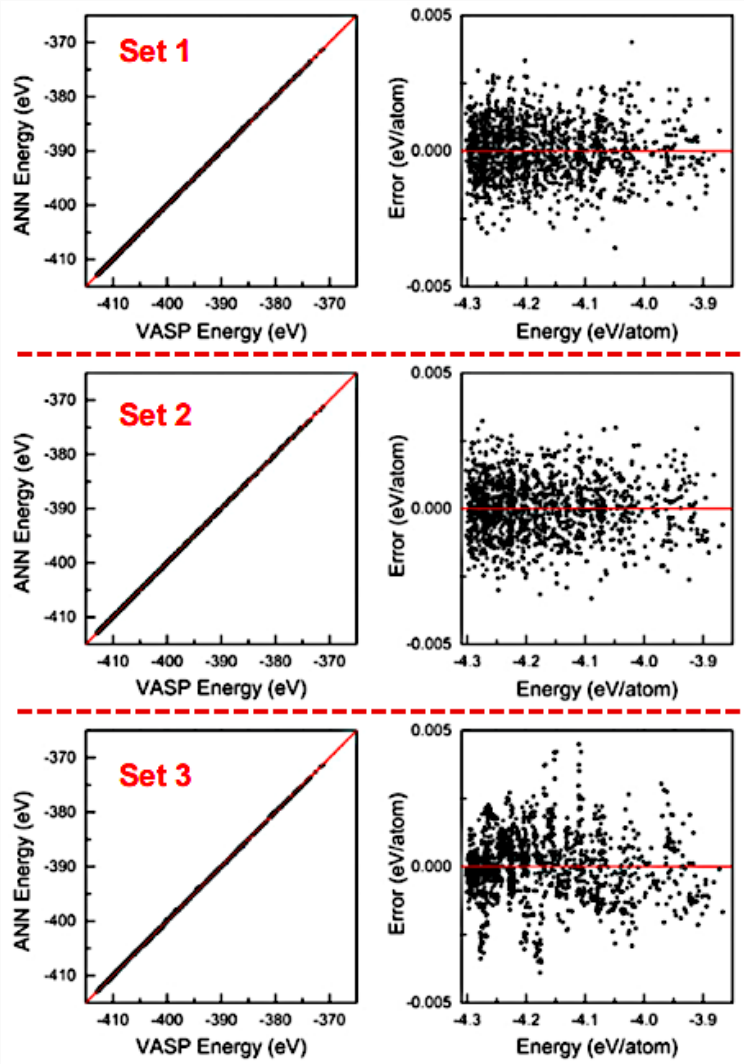
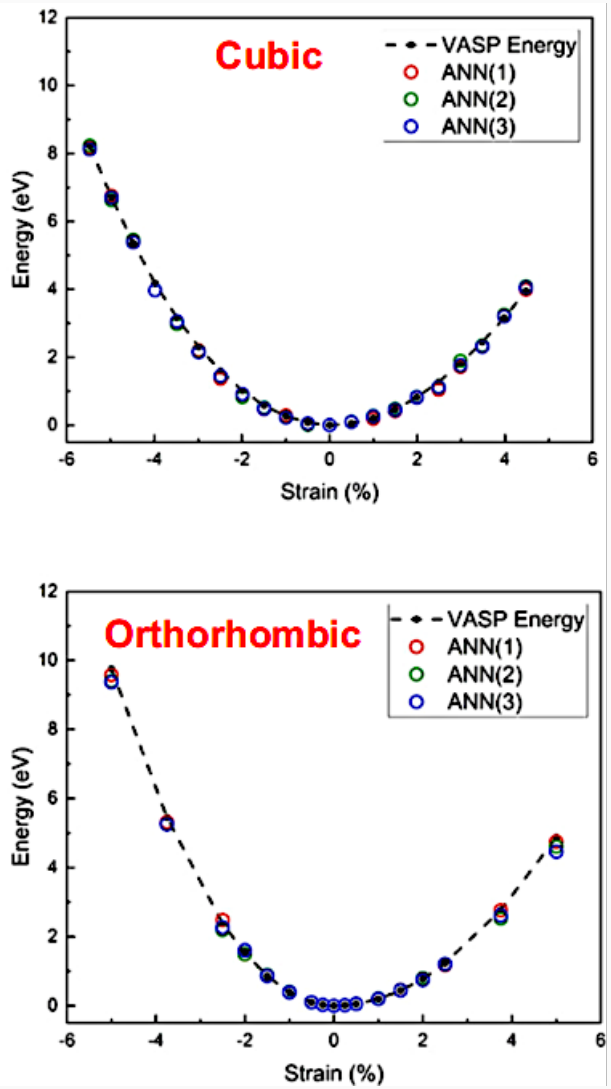


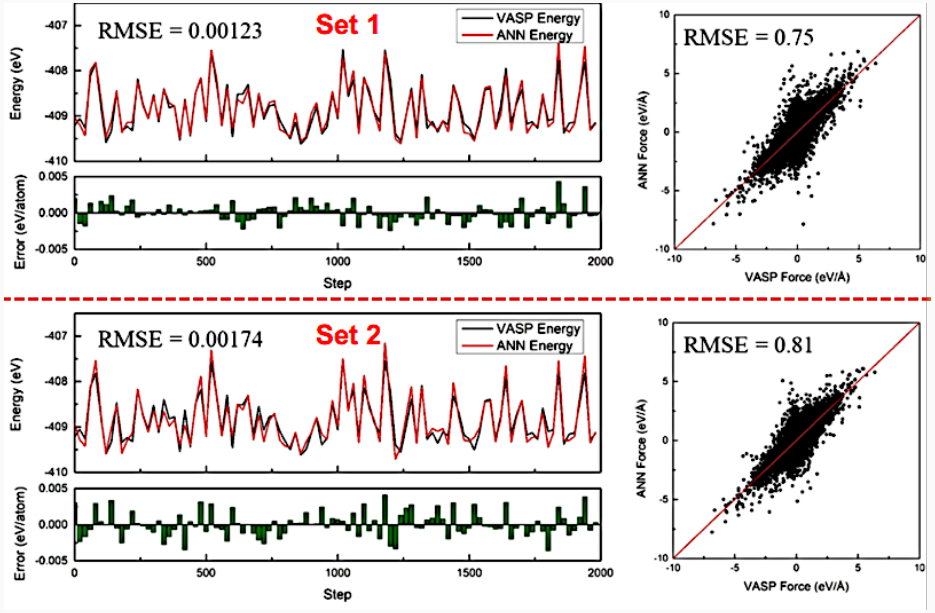

## Crystal Structure Tests



# Validation: Comparing AIMD Trajectory of CUBIC Perovskite

- To further validate the ANN models, coordinates from separate VASP AIMD trajectories of CUBIC perovskite were fed to ANN models and energies/atomic forces were evaluated.

## ANN Models w.o. Force Training

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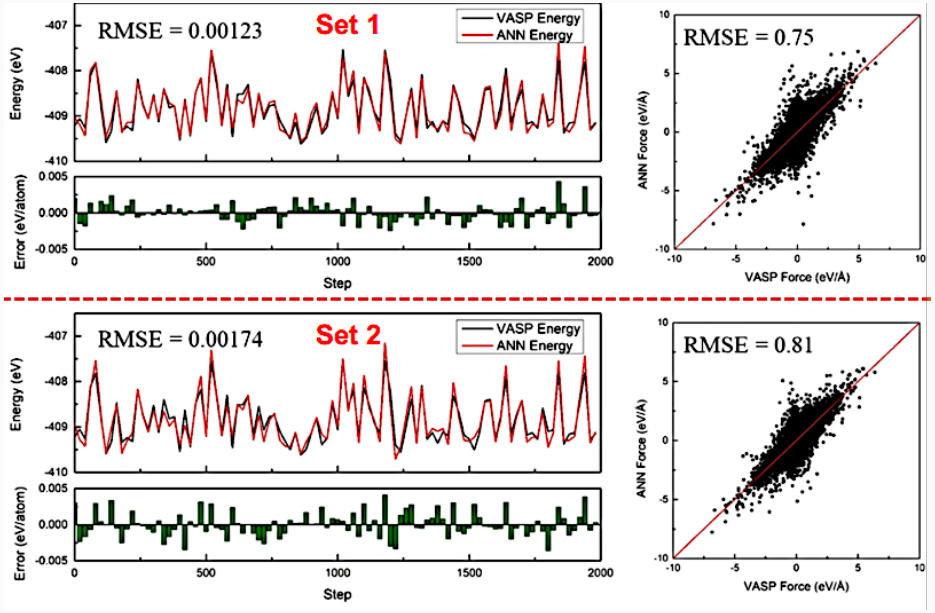


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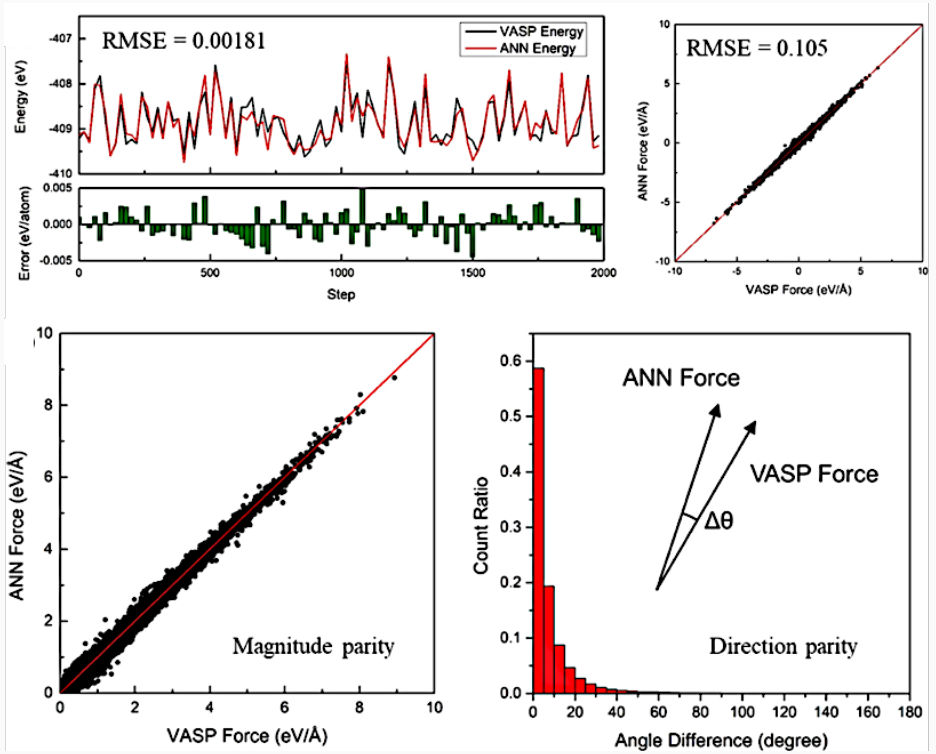
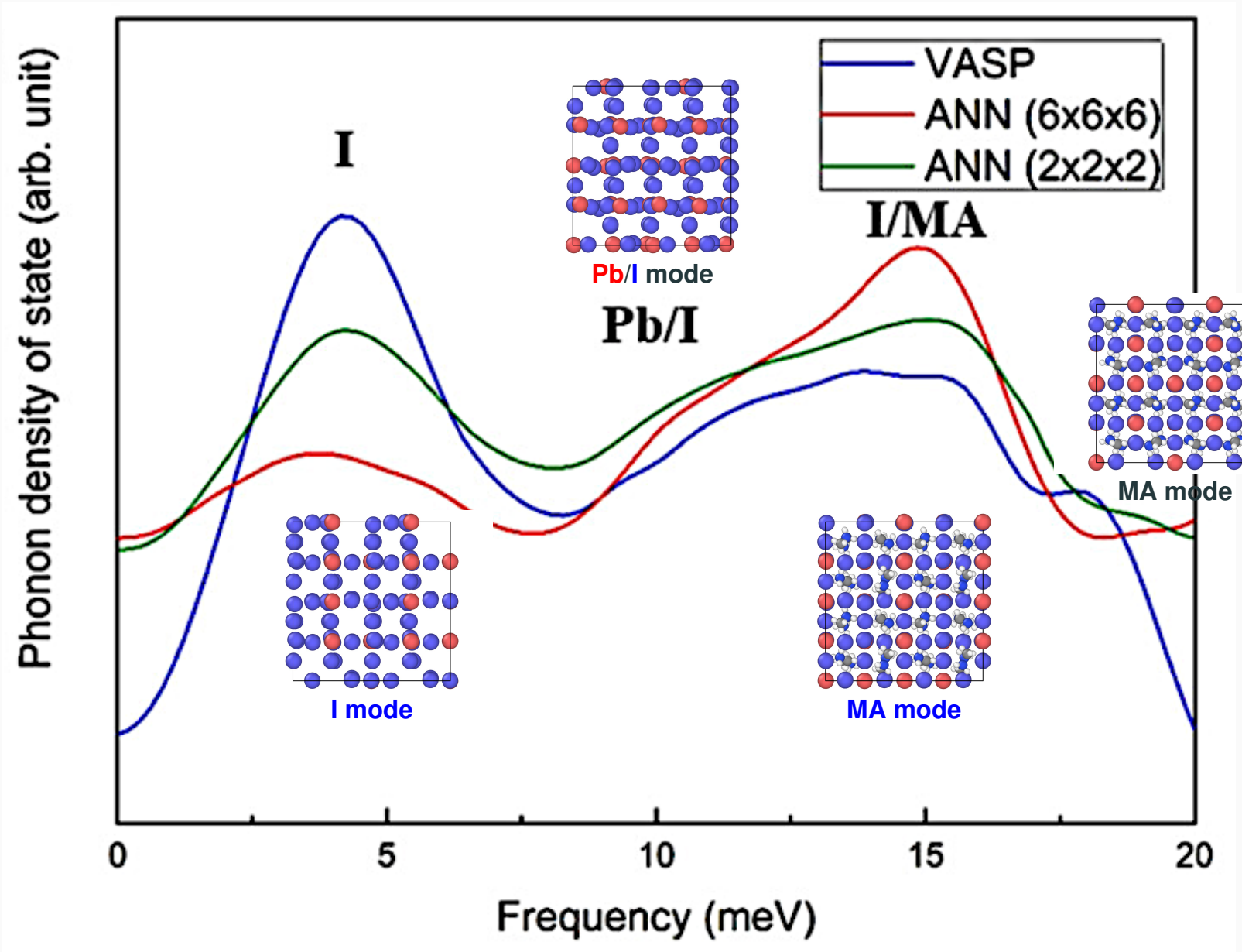
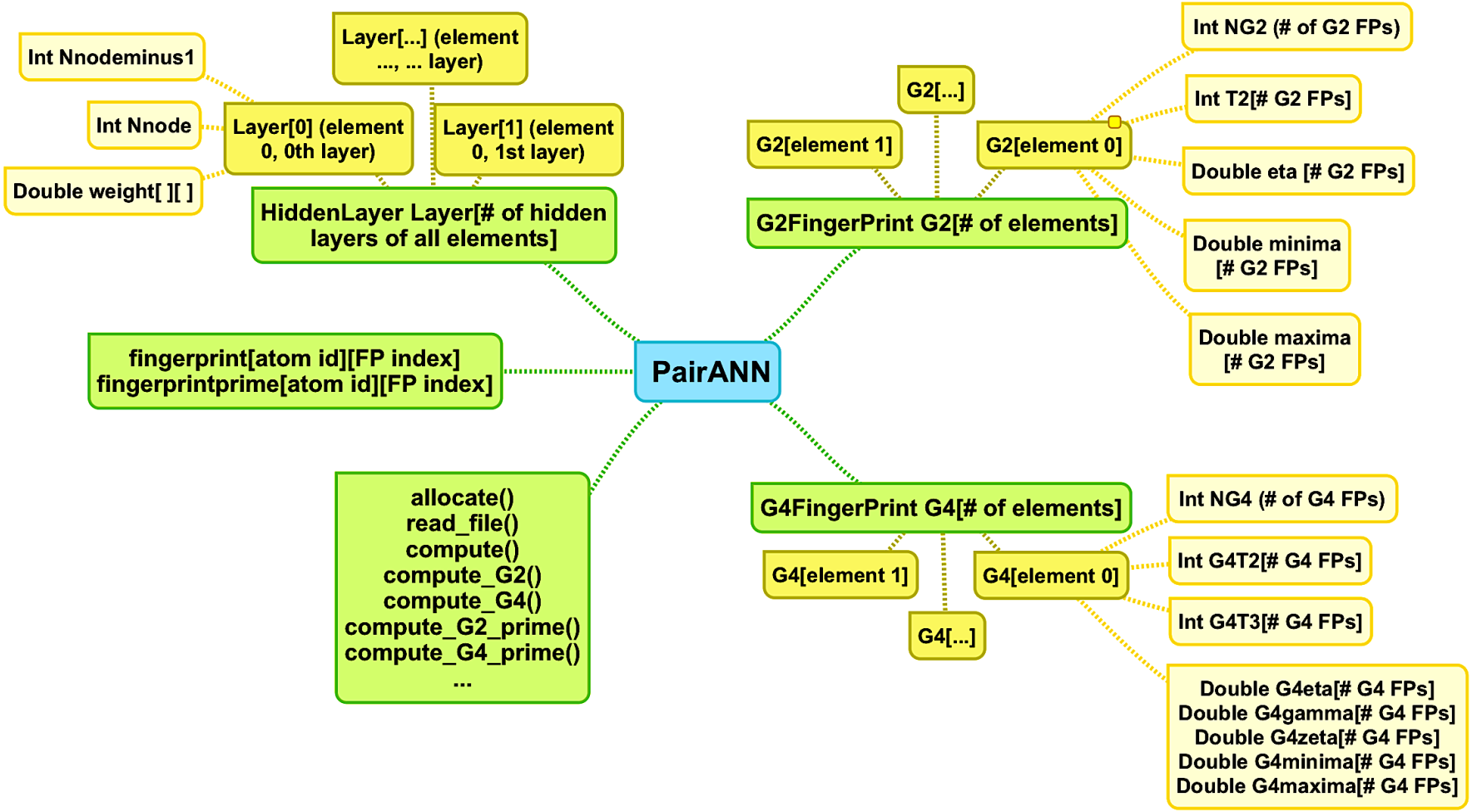


# Phonon Density of State Calculations



# Structure of *pair ann*



# Usage of *pair ann*

1. Train the potential using the *Atomic Machine Learning Package* (AMP)
2. Covert the potential file into *pair ann* format
3. Potential file format of *pair ann*:

```
# potential for Ni-Co-Ti-Zr-Hf
# Pb potential
G2 50
Pb 0.05 0.0383631306 6.82461076
I ......
.....
G4 60
Pb I ....
.....
Nlayer 3
Layer 40
.......
Layer 20
.......
# I potential
```

# Usage of pair ann and benchmark

*pair ann* can be invoked by the following commands in LAMMPS input script:

```
.............
pair_style ann 6.5
.............
pair_coeff * * MAPbI3.ann Pb I C N H
.............
```

Benchmark of the *pair ann*:

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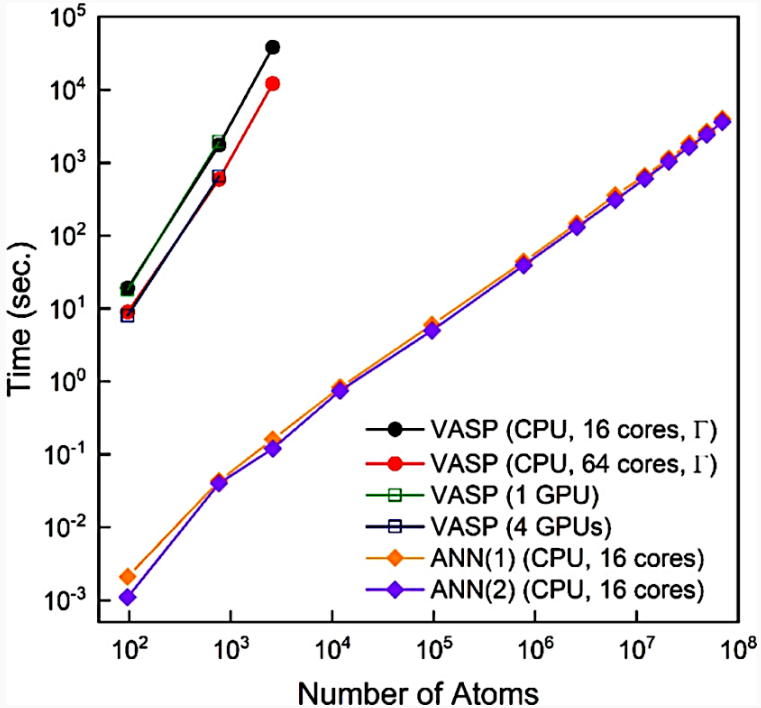

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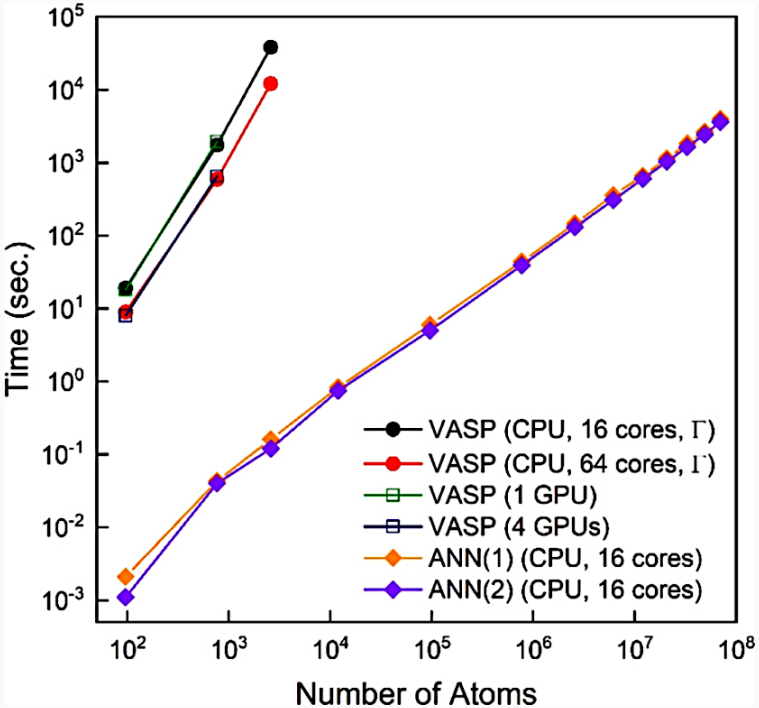
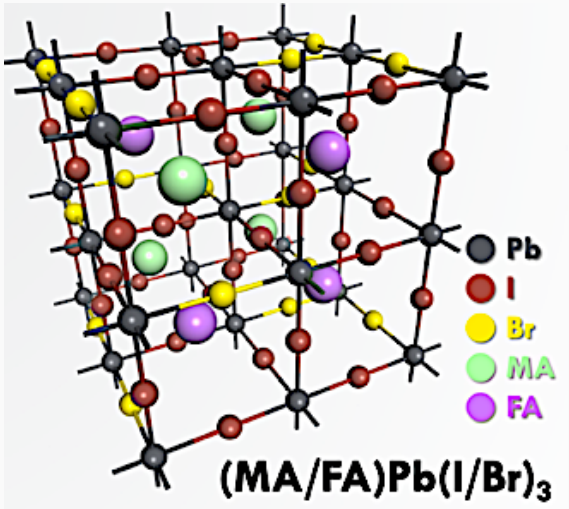
Benchmark of the *pair ann*:

An up to $10^5$ computational speedup relative to VASP can be achieved, thereby allowing exhaustive sampling of perovskite materials



# Training of ANN Potential for FAMAPbIBr



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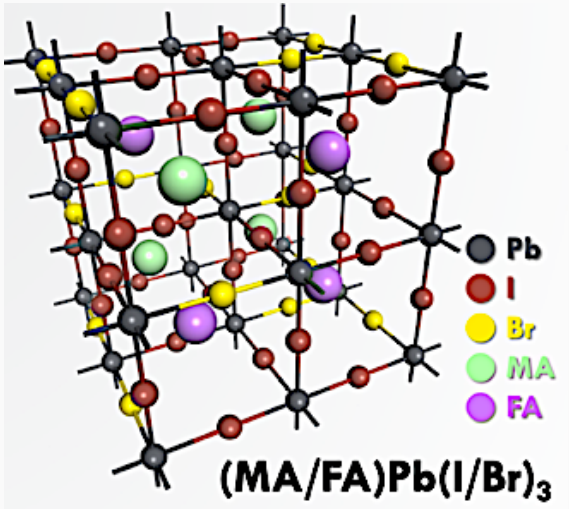
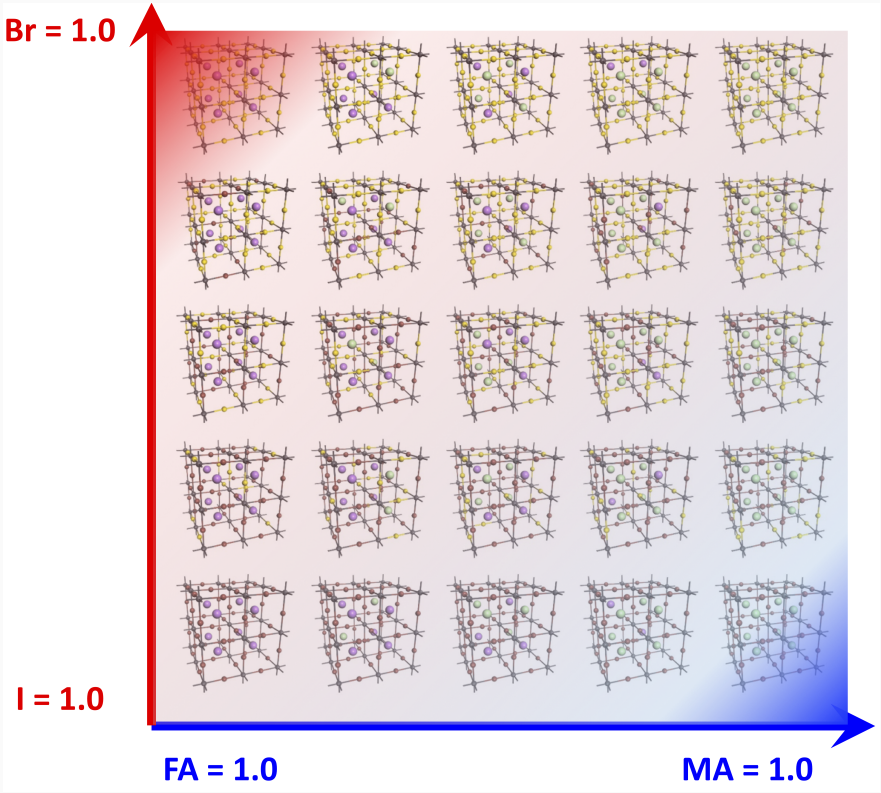
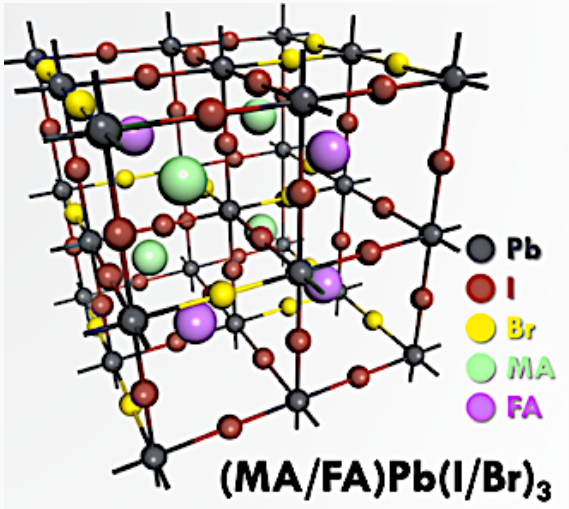


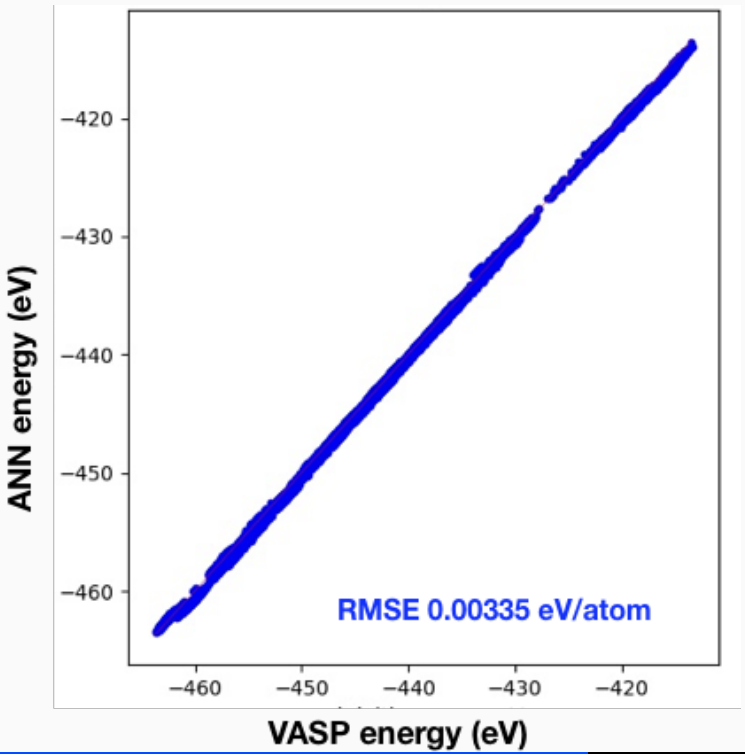



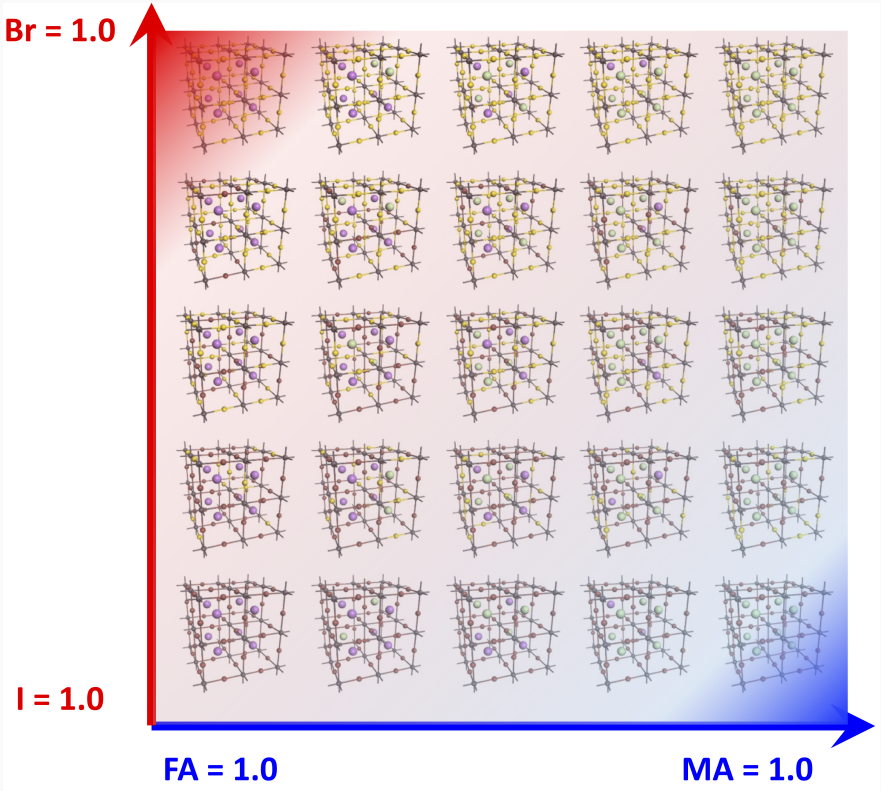

## Training Setup

1. MA, FA, I, Br of a variety of concentrations as well as permutations
2. (10,10) architecture for Pb, I, Br, C, N, and H atoms
3. Force training enabled

# Training of ANN Potential for FAMAPbIBr

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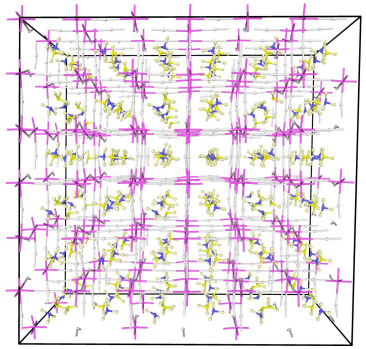
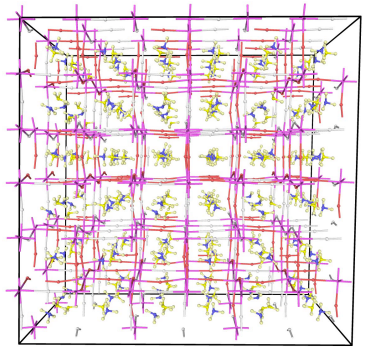
# Atomistic Simulation of FAMAPbIBr

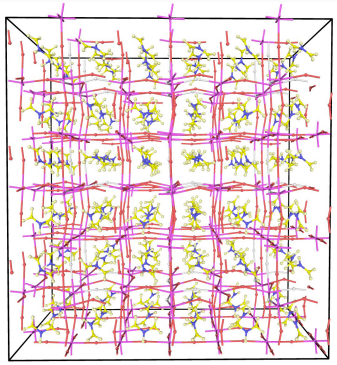
$MAPbI_3$



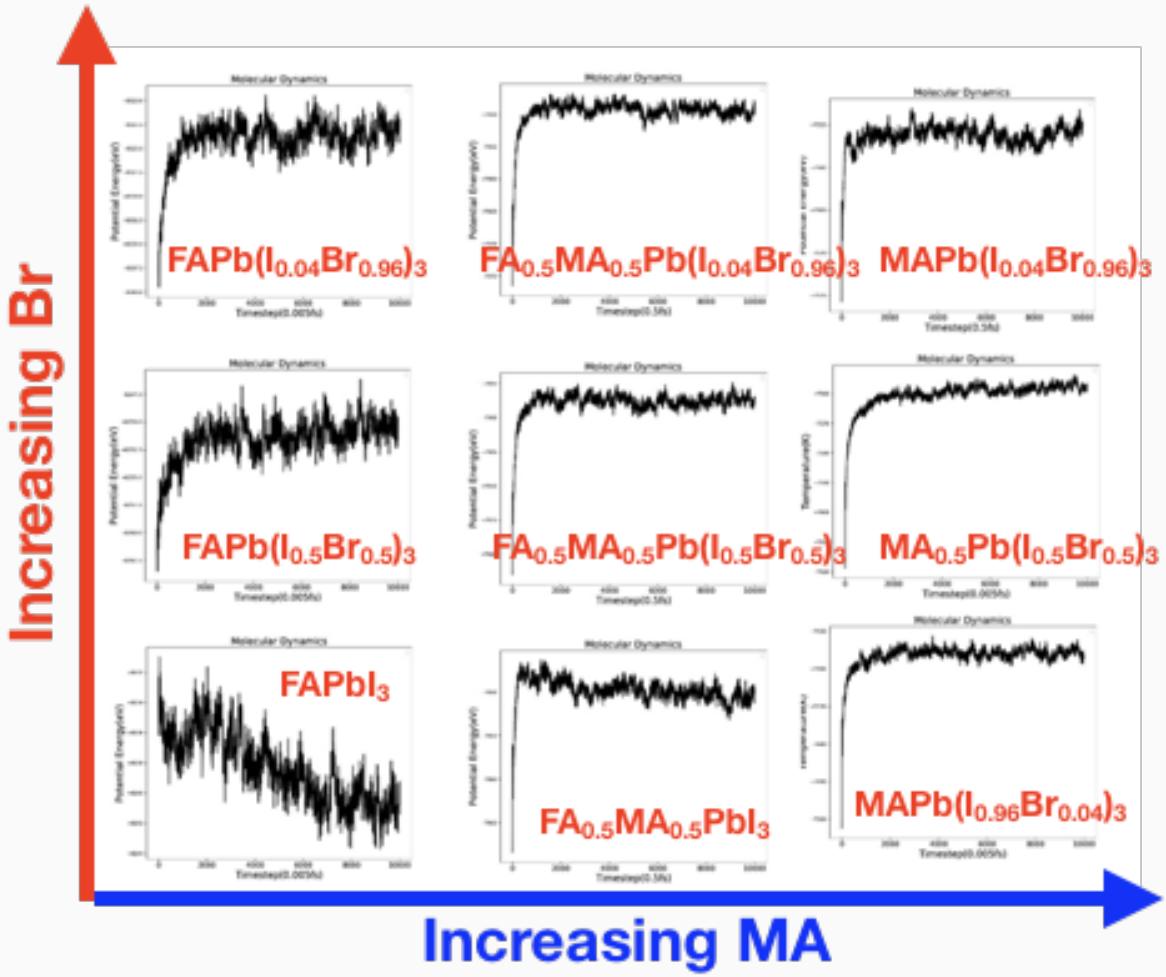
$MAPbI_{1.5}Br_{1.5}$



$FAPbIBr_3$



Potential energy evolution of MD trajectories of different FAMAPbIBr compositions using trained ANN potential



# Summary

## Pro of Neural Network Model

- Can successfully evaluate system energies/forces of chemically complex systems with high fidelity to respective *ab initio* calculations.
  - e.g. 2D perovskite, and CoCrFeNi and NiCoTiHfZr HEAs (ongoing work)
- Computational speedup allows exhaustive sampling of chemical configurations

## Challenges Ahead

- Accuracy of neural network model critically depends on the span of training sets.
  - A self-learning ANN to resolve the issue (ongoing).

## References


- Hsin-An Chen and Chun-Wei Pao* (2019), "Fast and Accurate Artificial Neural Network Potential Model for MAPbI3 Perovskite Materials", *ACS Omega* **4**, 10950
- Yu-Xuan Wang, Hsin-An Chen, Chun-Wei Pao*, Chien-Cheng Chang (2019), "Artificial Neural Network Model for Atomistic Simulations of Sb/MoS2 van der Waals Heterostructures", *Multiscale Science and Engineering* **1**, 119.

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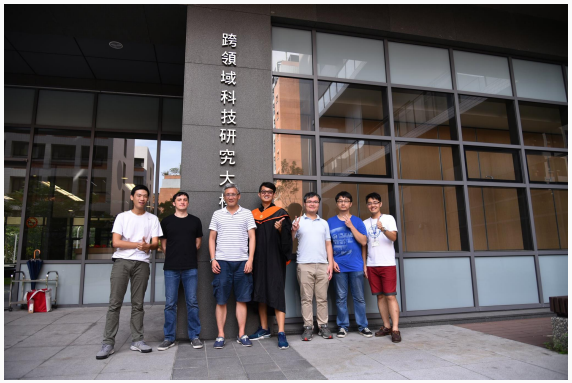

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