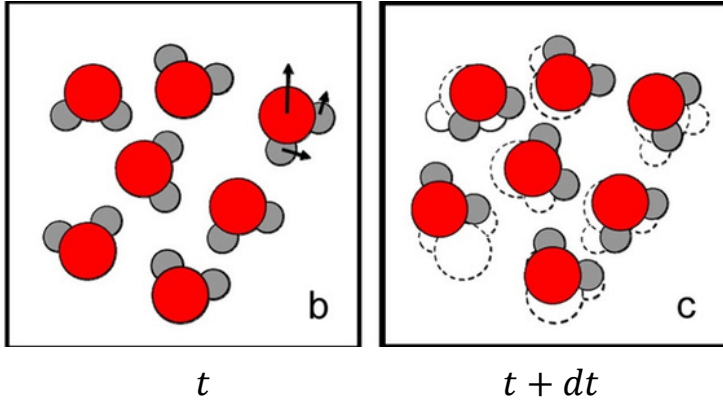


# **Importance of equivariance and message passing in machine-learning interatomic potentials**

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## What is molecular dynamics (MD)?



*J. Manuf. Sci. Eng.* Apr 2014, 136(2): 021015

- Change in atomic positions over time:

$$x = x_0 + vt + \frac{1}{2}at^2$$

- Velocity,  $v$ :

$$\frac{3}{2}kT = \sum \frac{1}{2}mv^2$$

- Acceleration,  $a$ :

$$F = ma \quad -\nabla V = F$$

### Classical interatomic potentials

Noble gases

$$V_{LJ} = 4\epsilon \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^6 \right]$$

Ionic bonding

$$V_{ij}(r) = \frac{Z_i Z_j e^2}{4\pi\epsilon_0 r} + Ae^{-\frac{r}{\rho}} - \frac{C}{r^6}$$

Covalent bonding

$$V_{ij}(r) = V_{\text{repulsive}}(r) + b_{ij} V_{\text{attractive}}(r)$$

- High speed (millions of atoms)
- Limited to specific systems

### Density functional theory (DFT)

$$H\Psi(r_1, r_2, \dots, r_N) = E\Psi(r_1, r_2, \dots, r_N)$$

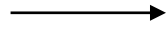


Quantum mechanical calculations

- Accurate & general
- Low speed (<500 atoms)

# Density-functional theory (DFT)

**Input:**  
Structure



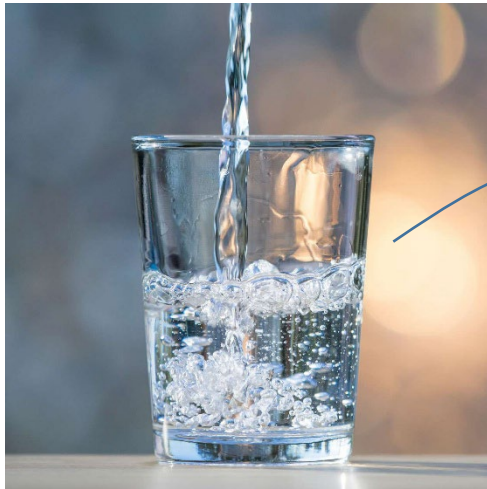
Quantum mechanics



**Output:**  
Energy, wavefunction

$$H\Psi(r_1, r_2, \dots, r_N) = E\Psi(r_1, r_2, \dots, r_N)$$

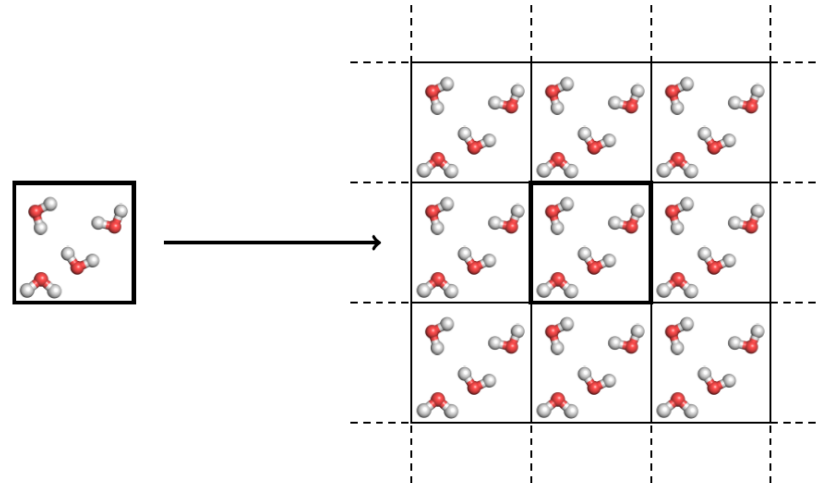
But not exact solution. Using universally applicable approximations.



$0.02 \text{ L} = 6.02 \times 10^{23} \text{ atoms}$

How can we simulate such a large number of atoms?

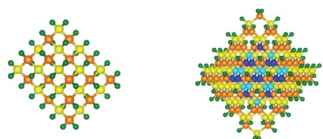
**Periodic boundary condition (PBC)**



Typically, 100–200 atoms are used to perform MD simulations with DFT calculations.

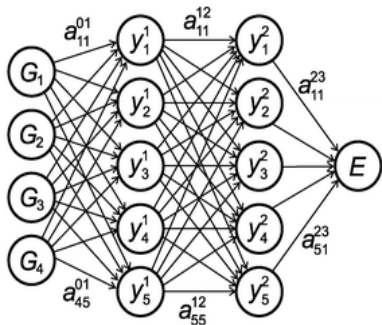
# Machine-learning interatomic potentials (MLIPs)

## Machine-learning interatomic potentials



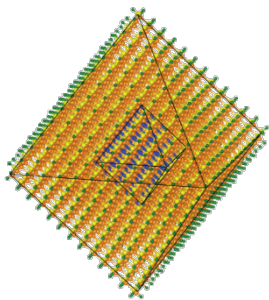
**Small structures**

Training set from  
DFT calculations



**Machine learning  
model**

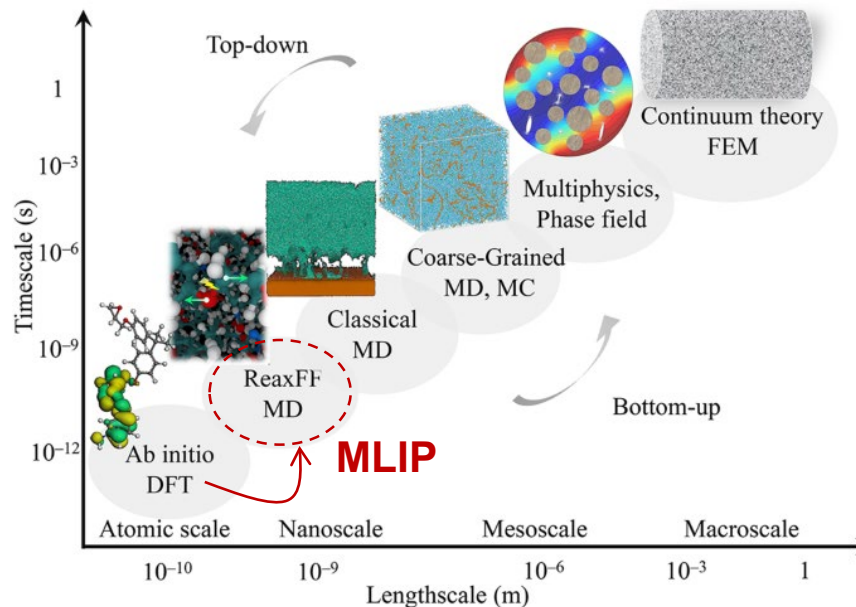
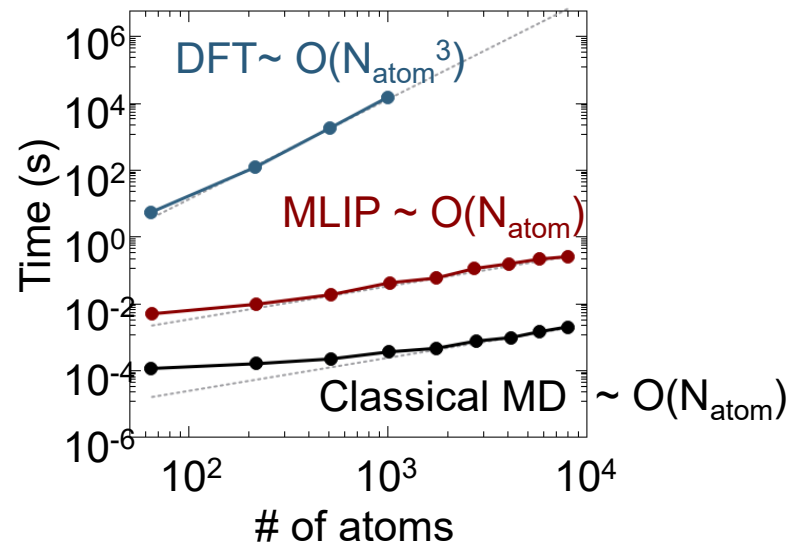
Energy =  $f(\text{structure})$



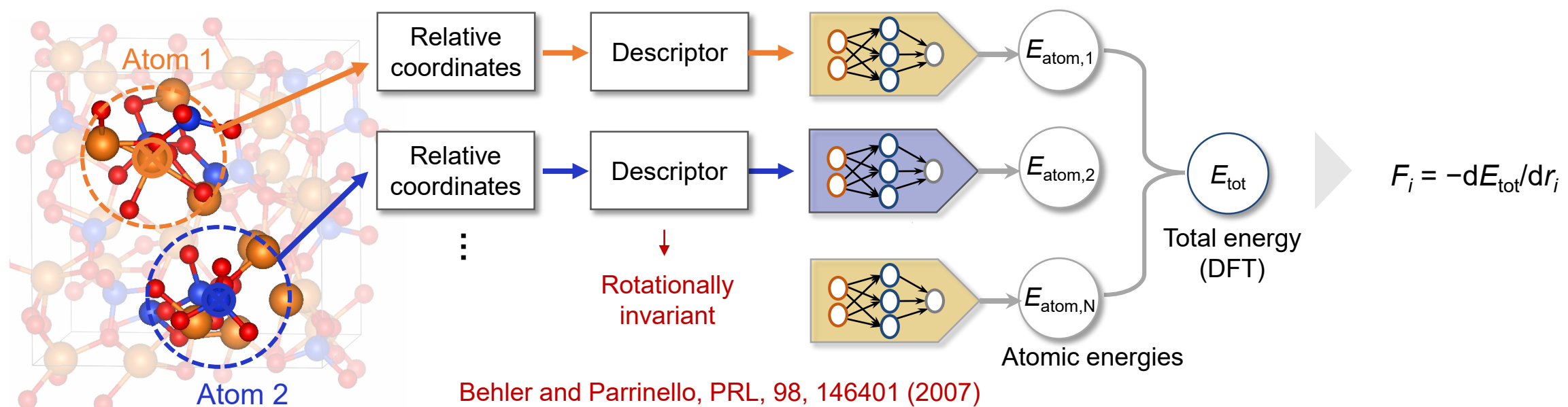
**Big structures**

Target simulation

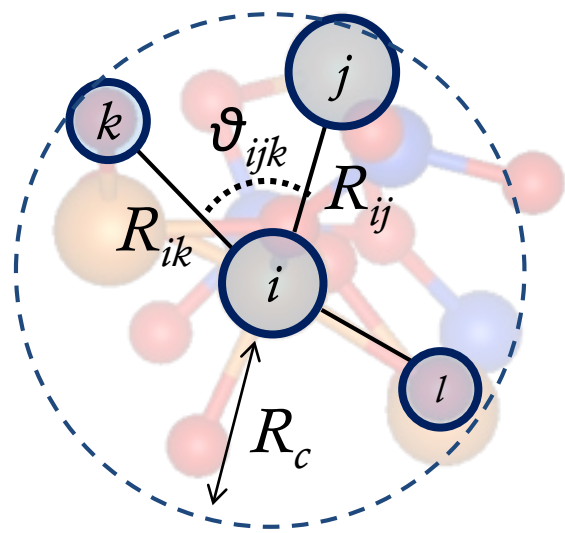
## Computation time for Si



# Architectures of the first-developed MLIP

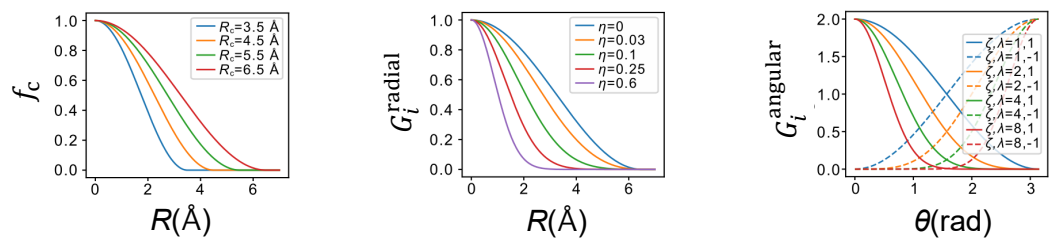


## Example descriptor: symmetry function



2-body  $G_i^{\text{radial}} = \sum_j e^{-\eta R_{ij}^2} f_c(R_{ij})$   $f_c$ : cutoff function

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

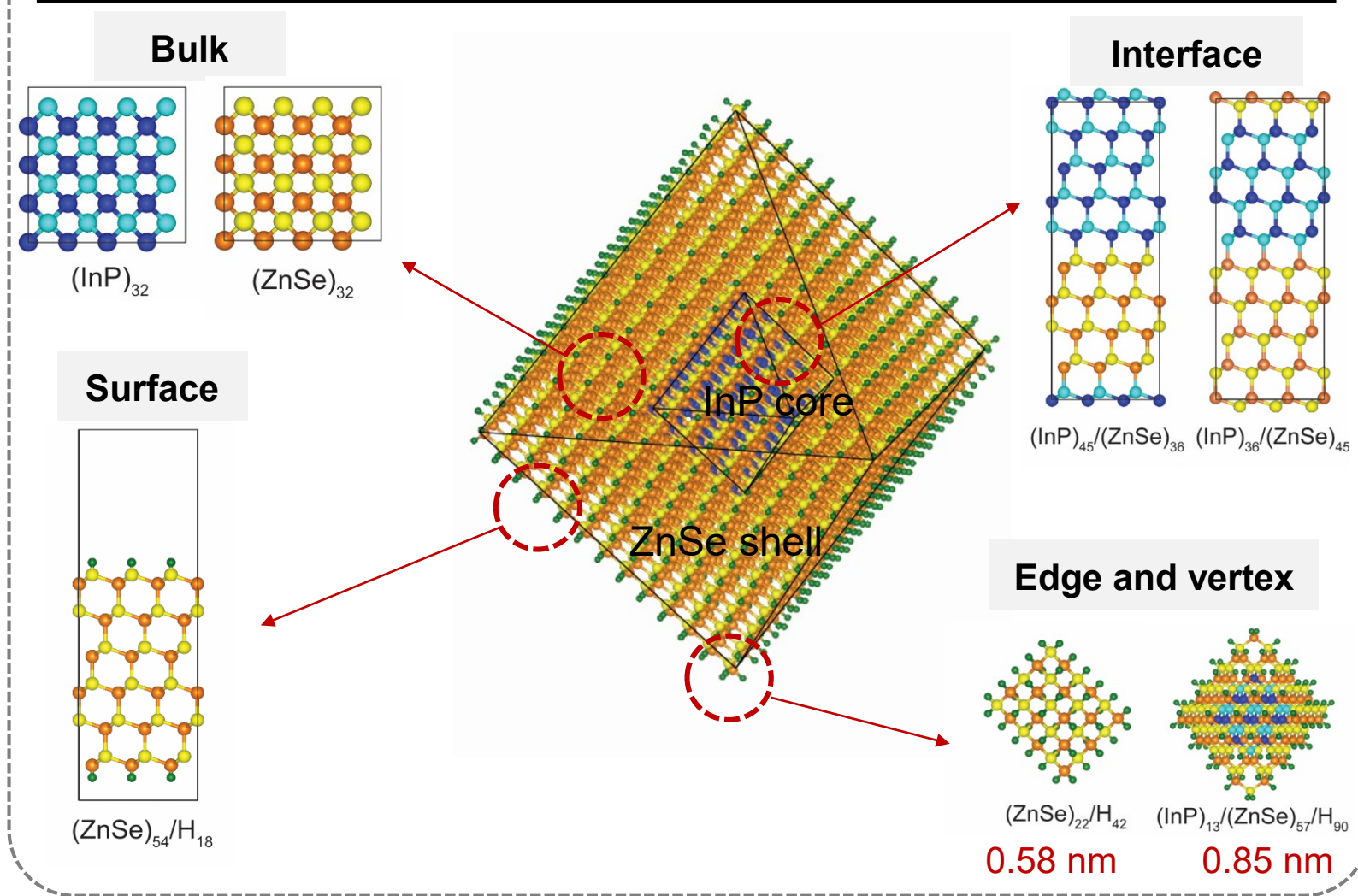


$\mathbf{G}_i = [G_i^{\text{radial},\eta_1}, G_i^{\text{radial},\eta_2}, G_i^{\text{radial},\eta_3}, \dots, G_i^{\text{angular},\zeta_1}, G_i^{\text{angular},\zeta_2}, G_i^{\text{angular},\zeta_3}, \dots]$

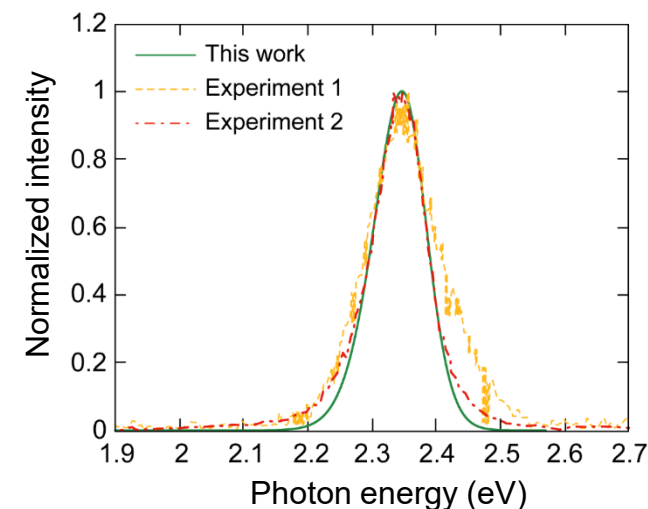
→ Used as input vectors for neural networks predicting atomic energies.



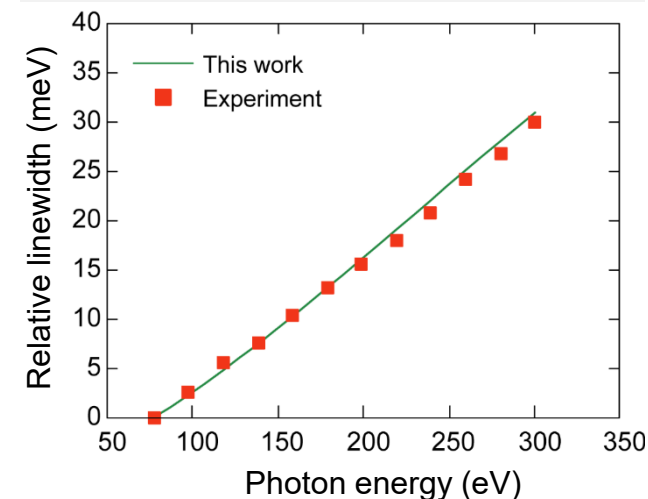
## Training set generation



## Luminescence spectrum



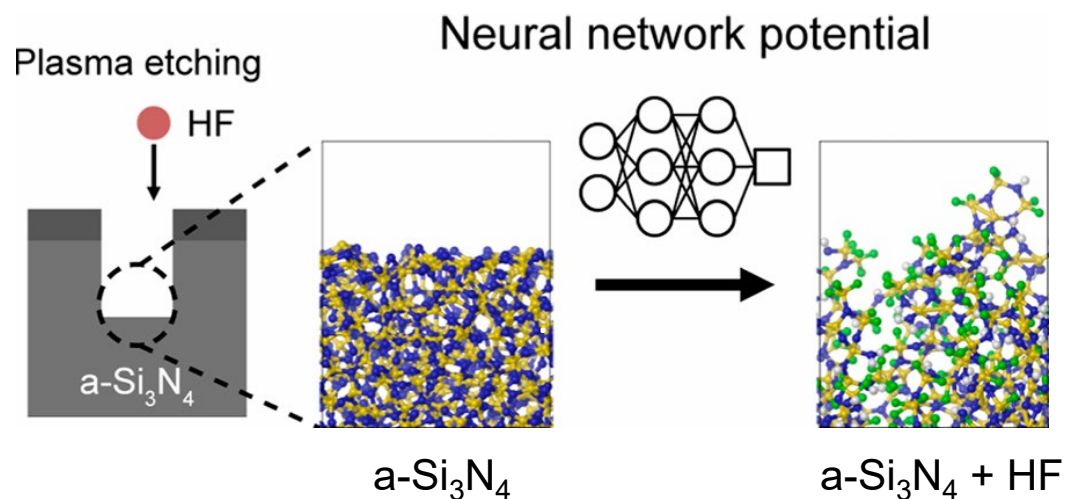
## T-dependence of linewidth



- Sampling a **training set** determines the quality and application range of MLPs.

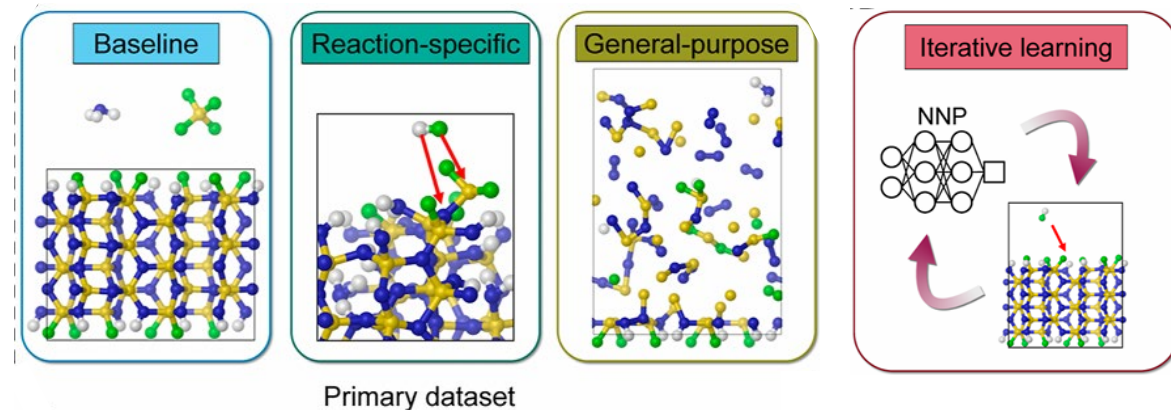
# Example: modeling HF etching process with MLIP

## Simulation target



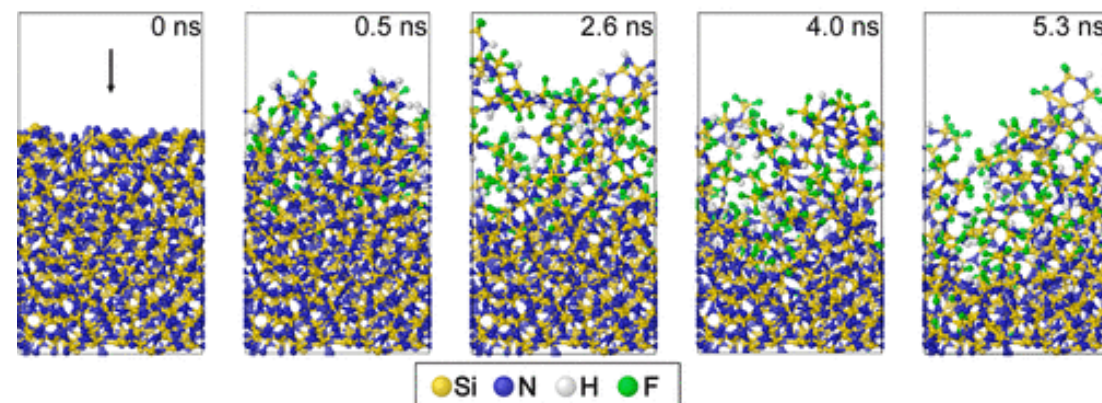
- HF etching of amorphous Si<sub>3</sub>N<sub>4</sub> for semiconductor process

## Training set generation (DFT)



Diverse sampling techniques / Time scale: ps scale

## Simulation (MLIP)

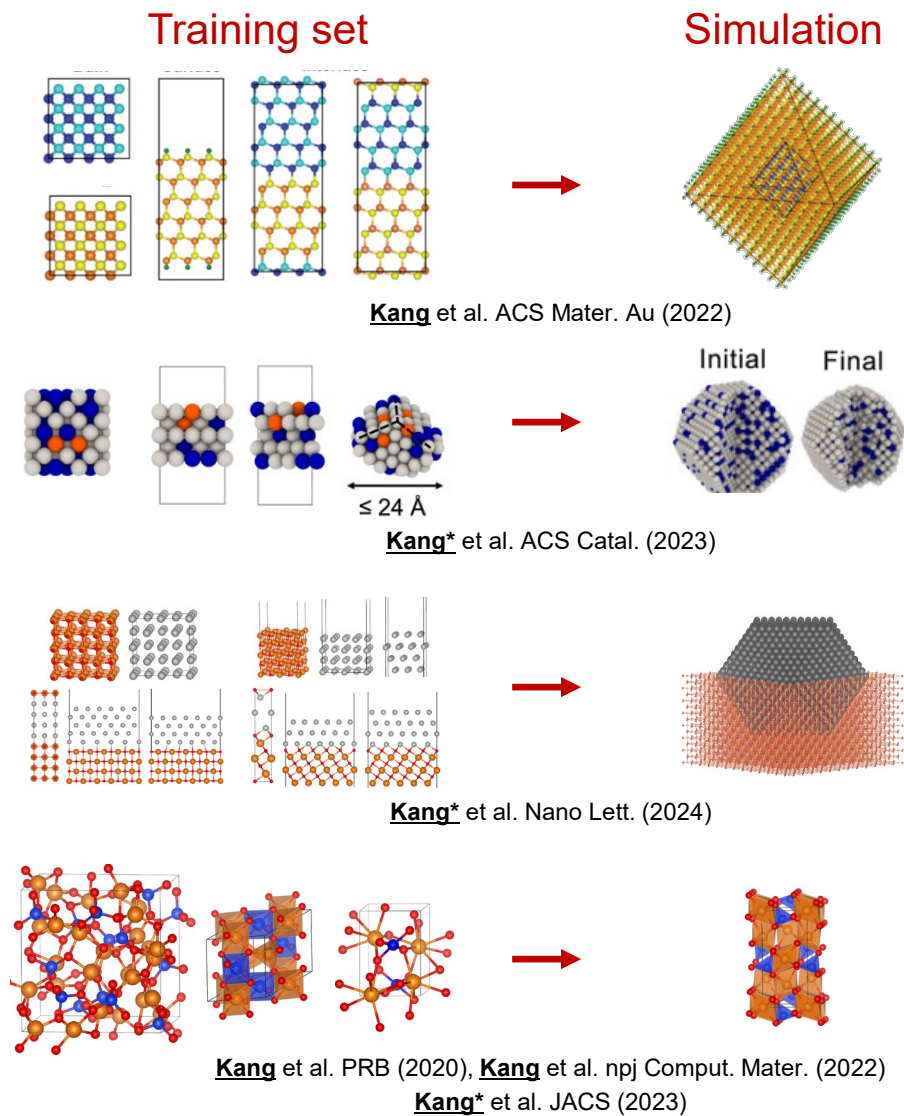


Time scale: ns scale

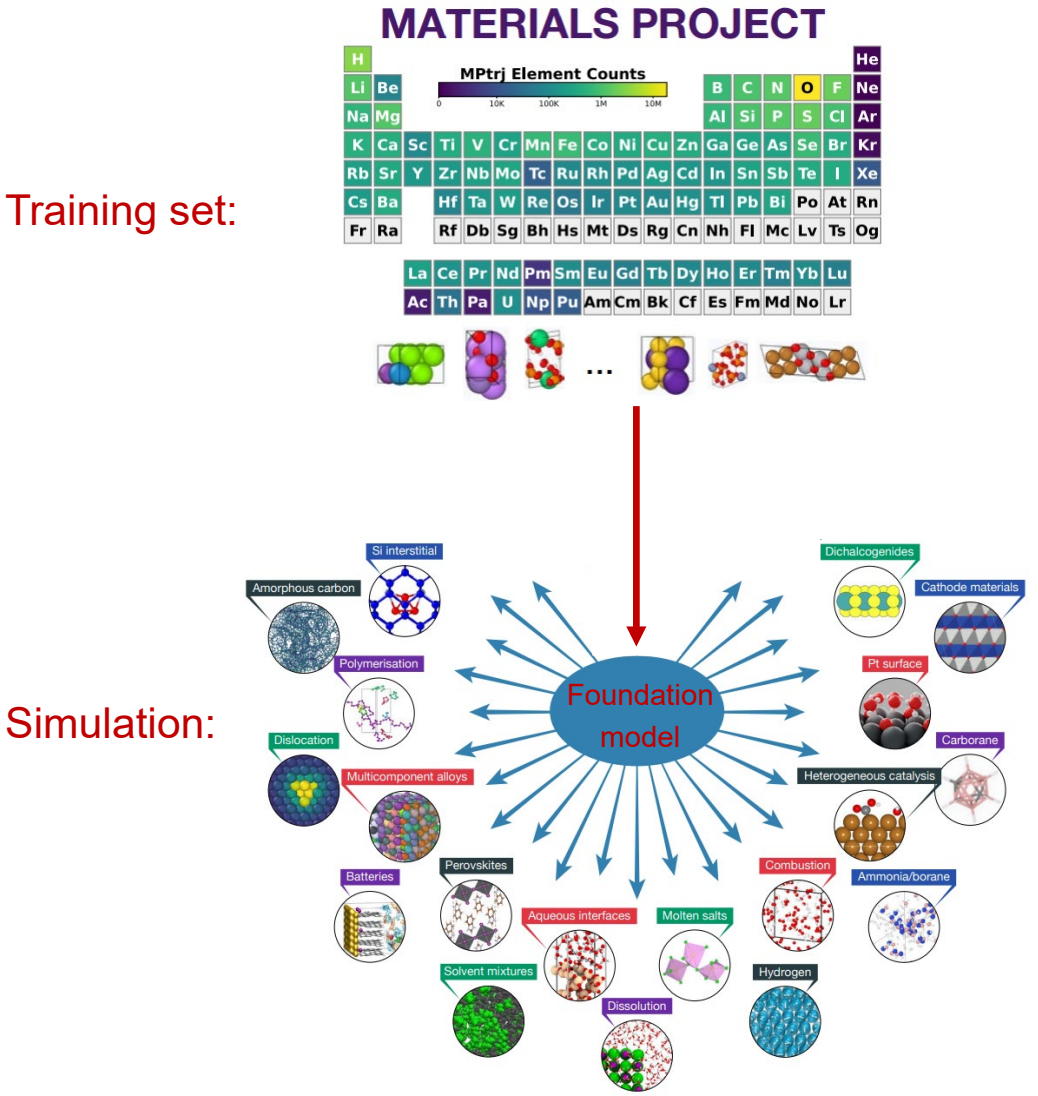


# Foundation model (or universal MLIP)

Conventional approach: MLIPs for individual systems



Recent approach: foundation model (big data)



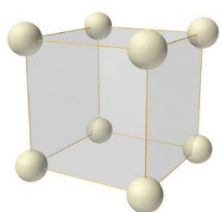


# Extrapolation behavior of universal MLIP

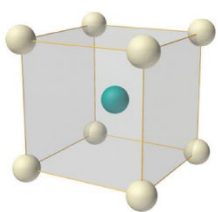
## Training set

- Crystalline material: an ordered solid composed of atoms arranged in a periodic lattice.

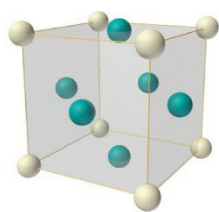
Example:



SC

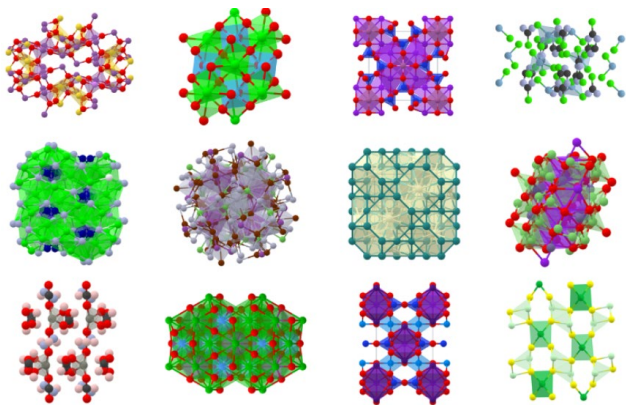


BCC



FCC

- Materials Project DB

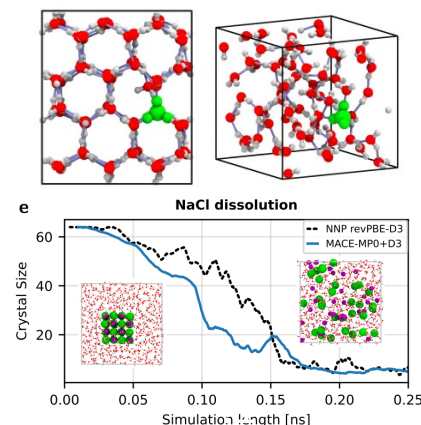


Materials Project is a computational database containing 200,000 **inorganic crystal** structures.

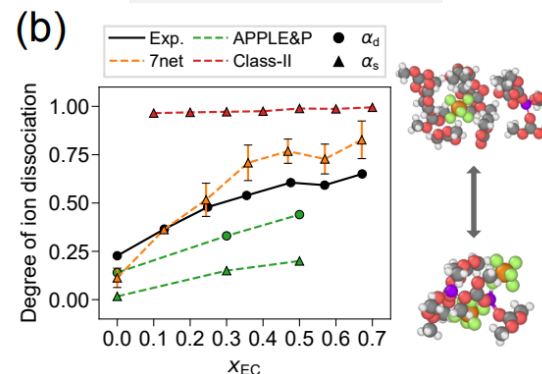
## Prediction results

### Not inorganic

#### Water & ice

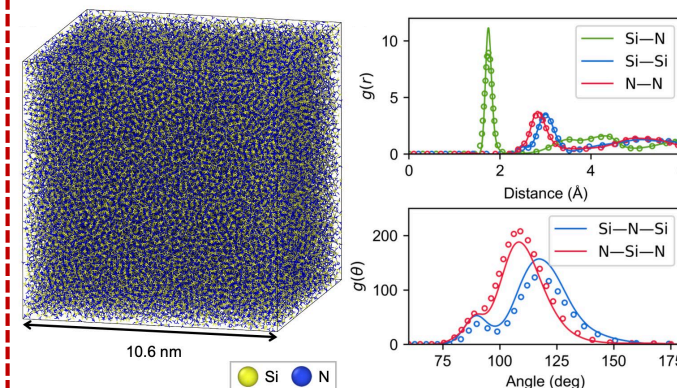


#### Organic liquid

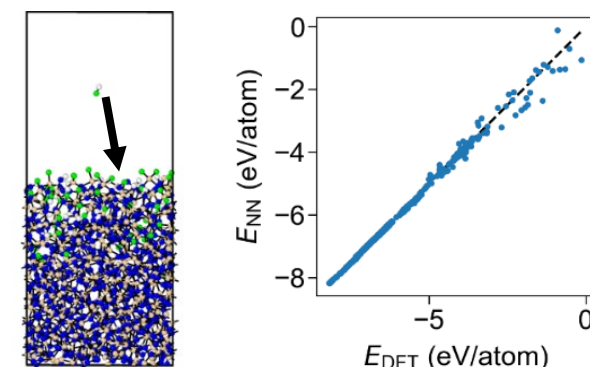


### Not crystal

#### Disordered structure



#### Etching simulation



# Benchmark test of foundation models

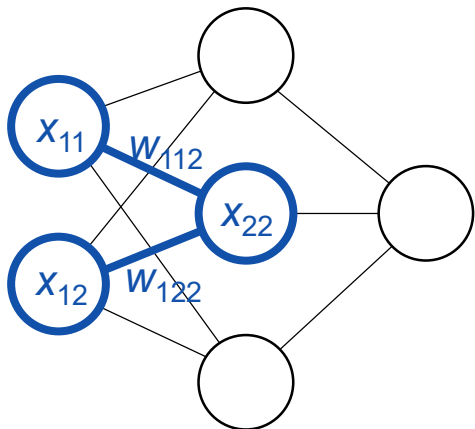
## Matbench Discovery benchmark test

- Energy error: non-listed compositions in Materials Project through substitution
- Thermal conductivity error

	Model	CPS $\uparrow$	Acc $\uparrow$	F1 $\uparrow$	DAF $\uparrow$	Prec $\uparrow$	MAE $\downarrow$	R <sup>2</sup> $\uparrow$	K <sub>SRME</sub> $\downarrow$	RMSD $\downarrow$	Training Set	Params	Targets
META (Facebook)	eSEN-30M-OAM	0.888	0.977	0.925	6.069	0.928	0.018	0.866	0.170	0.061	6.6M (113M) OMat24+MPtrj+sAlex	30.2M	EFS <sub>G</sub>
Orbital Material (start-up)	ORB v3	0.861	0.971	0.905	5.912	0.904	0.024	0.821	0.210	0.075	6.47M (133M) MPtrj+Alex+OMat24	25.5M	EFS <sub>G</sub>
SNU (Prof. Seungwu Han)	SevenNet-MF-ompa	0.845	0.969	0.901	5.825	0.890	0.021	0.867	0.317	0.064	6.6M (113M) OMat24+sAlex+MPtrj	25.7M	EFS <sub>G</sub>
	GRACE-2L-OAM	0.837	0.963	0.880	5.774	0.883	0.023	0.862	0.294	0.067	6.6M (113M) OMat24+sAlex+MPtrj	12.6M	EFS <sub>G</sub>
Ruhr-Universität Bochum	eSEN-30M-MP	0.797	0.946	0.831	5.260	0.804	0.033	0.822	0.340	0.075	146k (1.58M) MPtrj	30.1M	EFS <sub>G</sub>
	MACE-MPA-0	0.795	0.954	0.852	5.582	0.853	0.028	0.842	0.412	0.073	3.37M (12M) MPtrj+sAlex	9.06M	EFS <sub>G</sub>
	MatterSim v1 5M	0.767	0.959	0.862	5.852	0.895	0.024	0.863	0.574	0.073	17M MatterSim	4.55M	EFS <sub>G</sub>
Cambridge	DPA3-v2-OpenLAM	0.762	0.966	0.890	5.747	0.879	0.022	0.869	0.687	0.068	163M OpenLAM	7.02M	EFS <sub>G</sub>
	GRACE-1L-OAM	0.761	0.944	0.824	5.255	0.803	0.031	0.842	0.516	0.072	6.6M (113M) OMat24+sAlex+MPtrj	3.45M	EFS <sub>G</sub>
Microsoft	SevenNet-l3i5	0.714	0.920	0.760	4.629	0.708	0.044	0.776	0.550	0.085	146k (1.58M) MPtrj	1.17M	EFS <sub>G</sub>
	MatRIS v0.5.0 MPtrj	0.681	0.938	0.809	5.049	0.772	0.037	0.803	0.861	0.077	146k (1.58M) MPtrj	5.83M	EFS <sub>G</sub> M
	GRACE-2L-MPtrj	0.681	0.896	0.691	4.163	0.636	0.052	0.741	0.525	0.090	146k (1.58M) MPtrj	15.3M	EFS <sub>G</sub>
DP technology (China)	DPA3-v2-MPtrj	0.646	0.929	0.786	4.822	0.737	0.039	0.804	0.959	0.082	146k (1.58M) MPtrj	4.92M	EFS <sub>G</sub>
	MACE-MP-0	0.644	0.878	0.669	3.777	0.577	0.057	0.697	0.647	0.091	146k (1.58M) MPtrj	4.69M	EFS <sub>G</sub>
	AlphaNet-MPTrj	0.566	0.933	0.799	4.863	0.743	0.041	0.745	1.310	0.107	146k (1.58M) MPtrj	16.2M	EFS <sub>G</sub>
	eqV2 M	0.558	0.975	0.917	6.047	0.924	0.020	0.848	1.771	0.069	3.37M (102M) OMat24+MPtrj	86.6M	EFS <sub>D</sub>
	ORB v2	0.529	0.965	0.880	6.041	0.924	0.028	0.824	1.732	0.097	3.25M (32.1M) MPtrj+Alex	25.2M	EFS <sub>D</sub>
	eqV2 S DeNS	0.522	0.941	0.815	5.042	0.771	0.036	0.788	1.676	0.076	146k (1.58M) MPtrj	31.2M	EFS <sub>D</sub>
	ORB v2 MPtrj	0.470	0.922	0.765	4.702	0.719	0.045	0.756	1.725	0.101	146k (1.58M) MPtrj	25.2M	EFS <sub>D</sub>
	M3GNet	0.428	0.813	0.569	2.882	0.441	0.075	0.585	1.412	0.112	62.8k (188k) MPF	228k	EFS <sub>G</sub>
	CHGNet	0.400	0.851	0.613	3.361	0.514	0.063	0.689	1.717	0.095	146k (1.58M) MPtrj	413k	EFS <sub>G</sub> M
Google DeepMind	GNoME	NaN	0.955	0.829	5.523	0.844	0.035	0.785	n/a	n/a	6M (89M) GNoME	16.2M	EF <sub>G</sub>

# Equivariant graph neural network

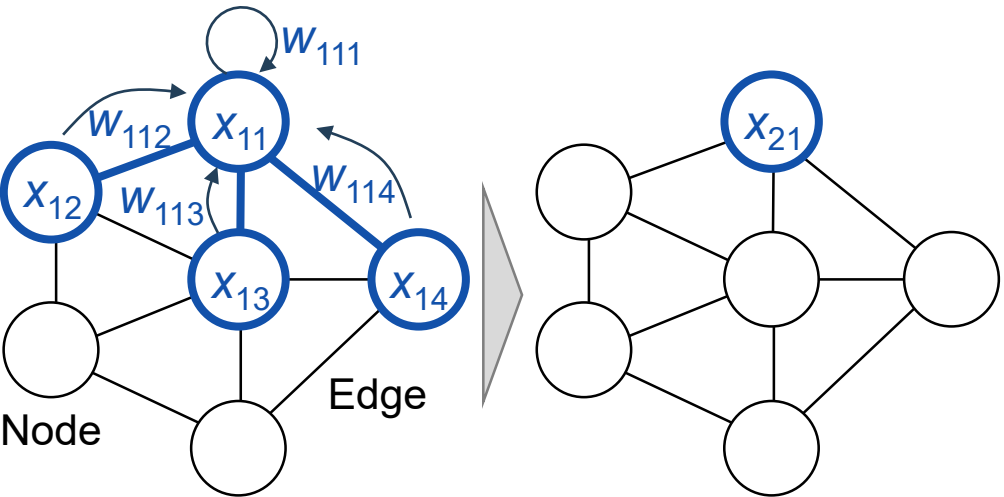
Neural network



Input Hidden Output

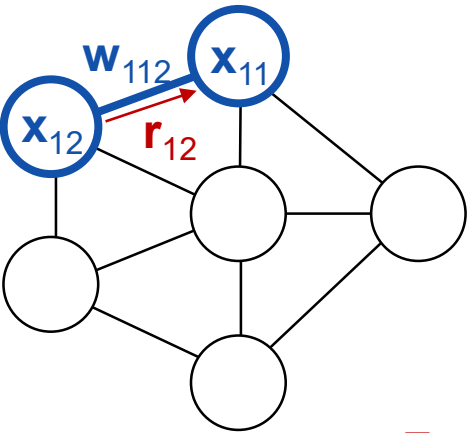
$$x_{22} = \sigma(w_{112}x_{11} + w_{122}x_{12} + b_1)$$

Graph NN (message passing NN)



$$x_{21} = \sigma(w_{112}x_{11} + w_{112}x_{12} + \dots)$$

Equivariant GNN



Tensor

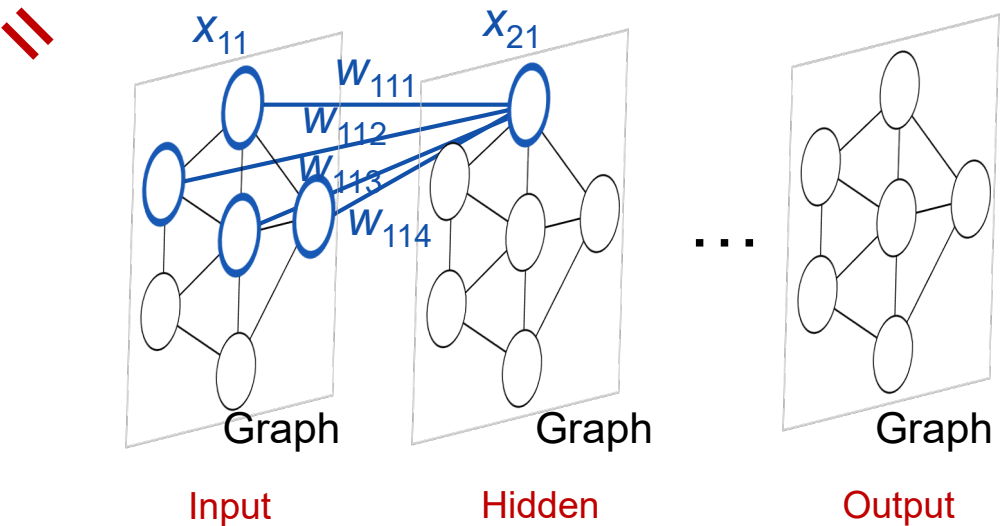
$$\text{Message from 1 to 2} = w_{112} \otimes x_{12}$$

$$\text{Edge tensor, } w_{112,lm} = R(r_{12})Y_{lm}(\hat{r}_{12})$$

Radial term  
(include trainable weights)

Spherical harmonics

For instance, when  $l = 1$   
 $Y_{1-1}(\theta, \varphi) = C \sin\theta \sin\varphi \rightarrow \mathbf{y}$   
 $Y_{10}(\theta, \varphi) = C \cos\theta \rightarrow \mathbf{z}$   
 $Y_{11}(\theta, \varphi) = C \sin\theta \cos\varphi \rightarrow \mathbf{x}$



# E(3)-equivariant neural network

## E(3) group

E(3) = 3D Euclidean group, which comprises translations, rotations, and reflections (parity).

Odd parity (p = -1)      Even parity (p = 1)

l = 0



Pseudo scalar (0o)



Scalar (0e)

l = 1



Vector (1o)



Pseudo vector (1e)

l = 2

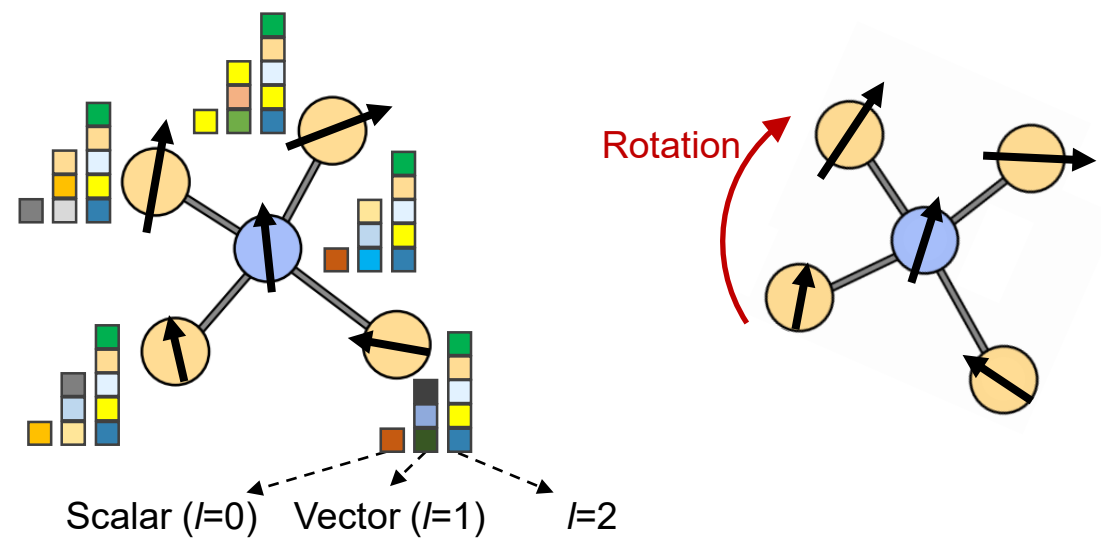


2o



2e

## Network architecture



Message  
from node *b* to *a*

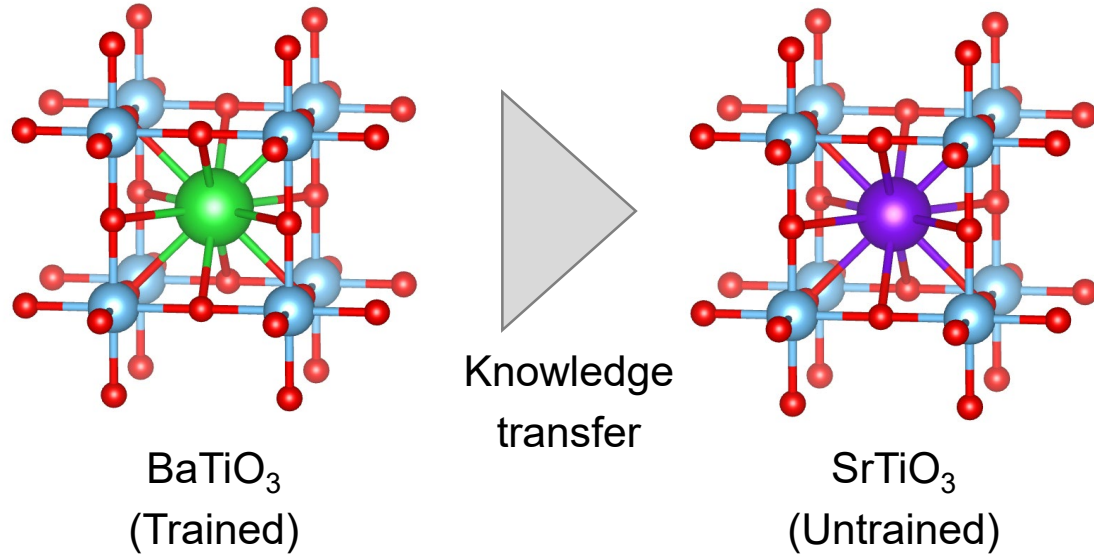
$$\mathcal{L}_{acm_o}^{l_o, p_o, l_f, p_f, l_i, p_i}(\vec{r}_a, V_{acm_i}^{l_i, p_i}) = \text{Edge tensor} \otimes \text{Node tensor}$$
$$= \sum_{m_f, m_i} \underbrace{C_{l_i, m_i, l_f, m_f}^{l_o, m_o}}_{\text{Clebsch-Gordon coeff.}} \sum_{b \in S} \underbrace{(R(r_{ab}))_{c, l_o, p_o, l_f, p_f, l_i, p_i}}_{\text{Radial part}} \underbrace{Y_{m_f}^{l_f}(\hat{r}_{ab})}_{\text{Spherical harmonics}} \underbrace{V_{bcm_i}^{l_i, p_i}}_{\text{Node feature}}$$

Feature vectors consist of tensors, in addition of scalars.

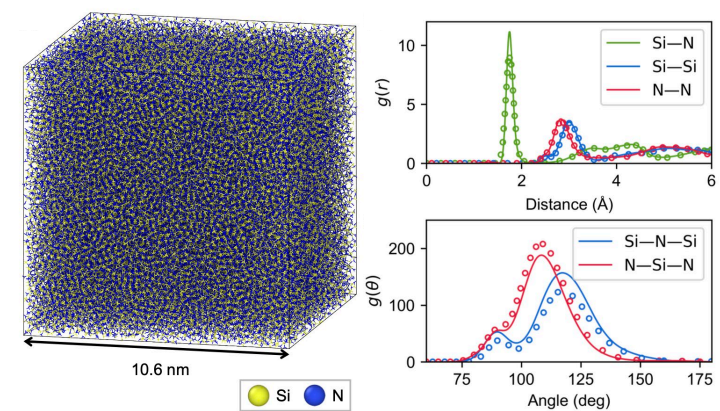
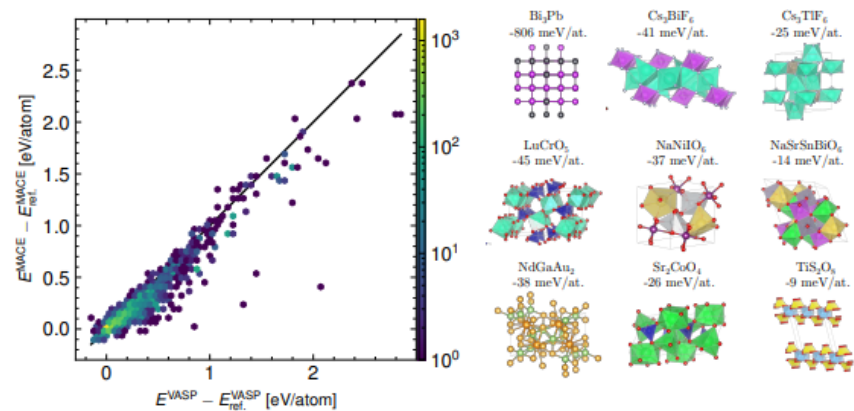
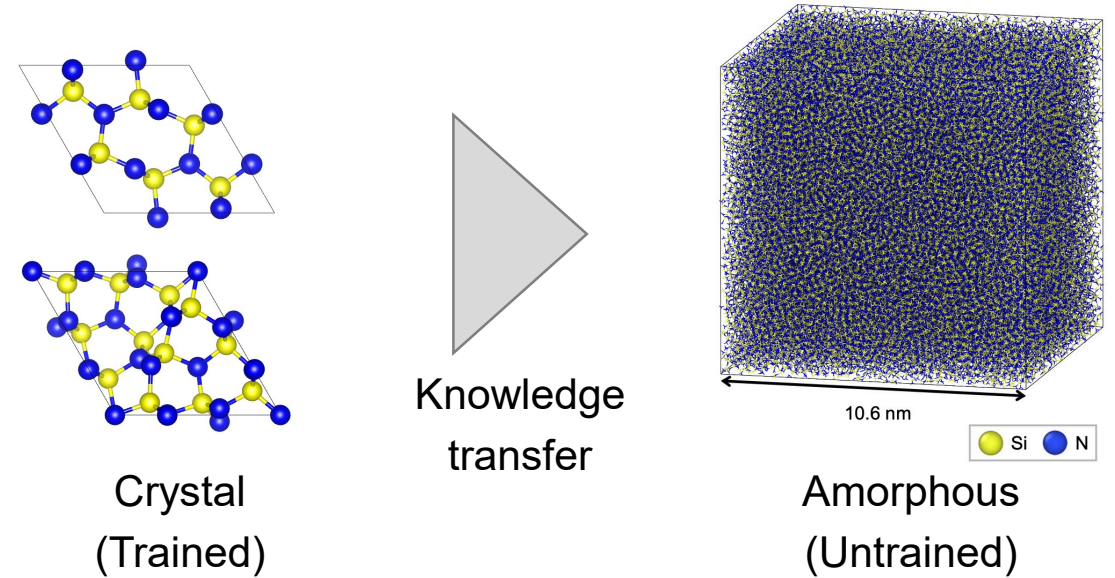


# Types of extrapolation made by graph neural network interatomic potentials (GNN-IPs)

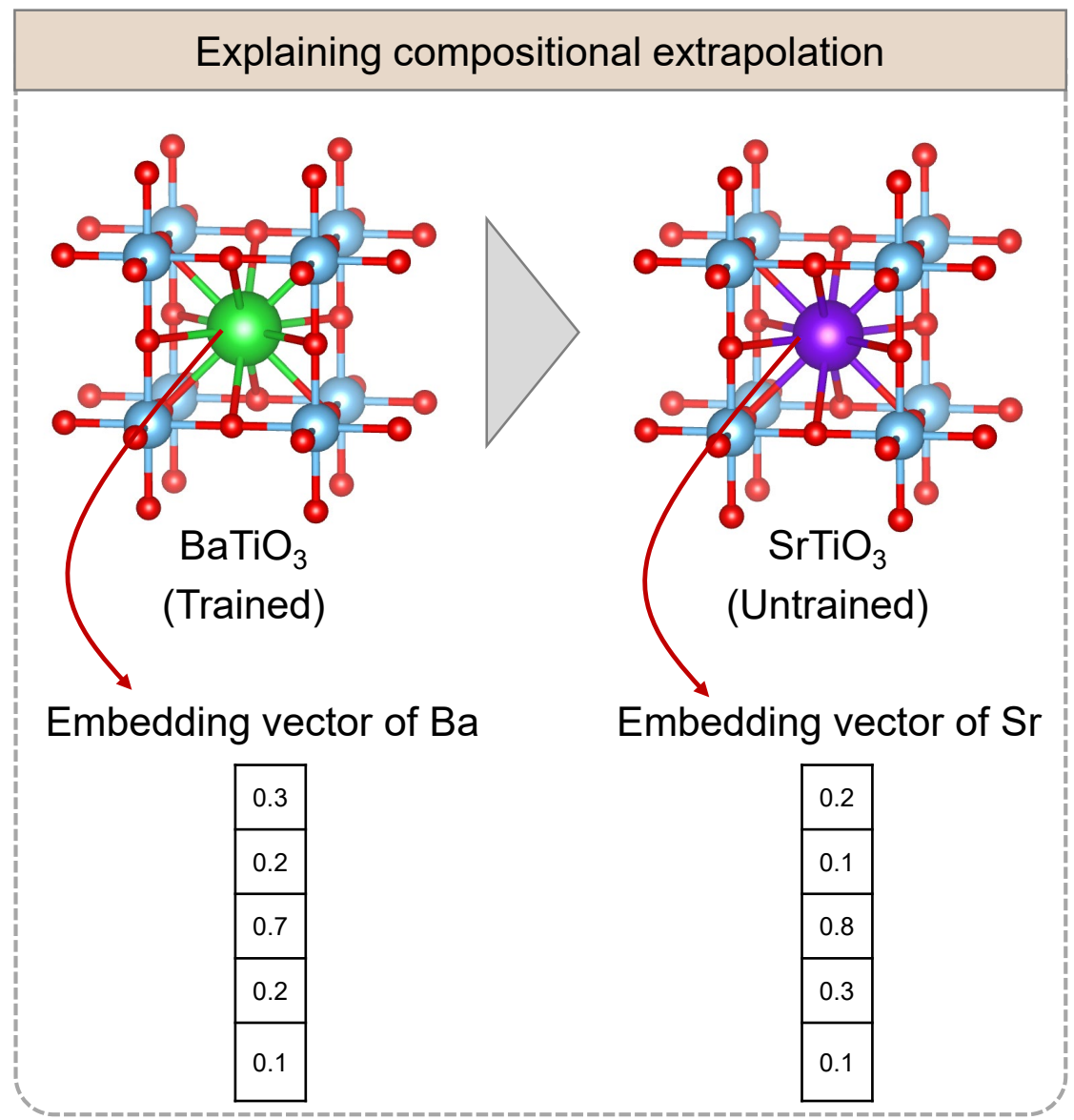
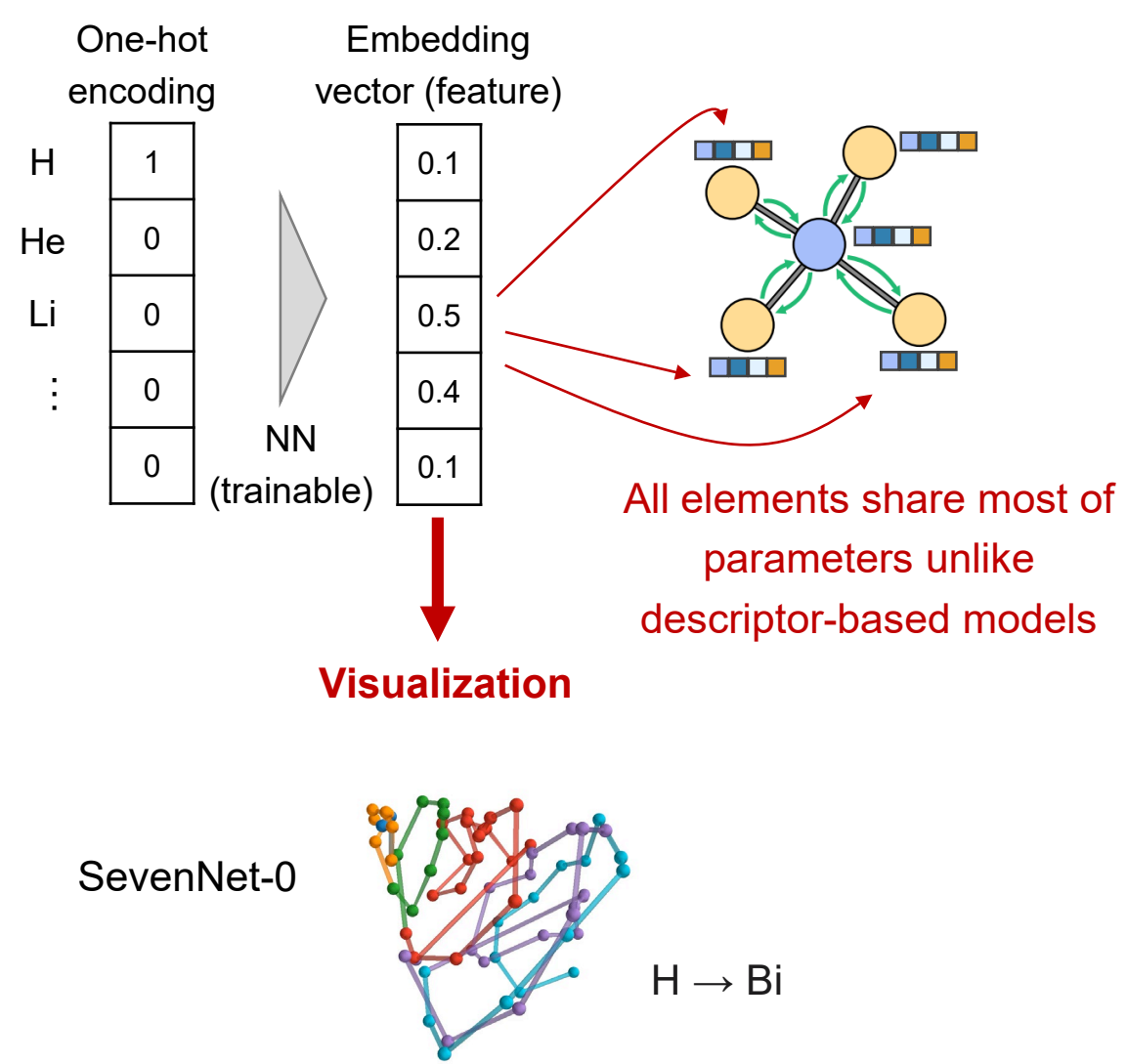
## Compositional extrapolation



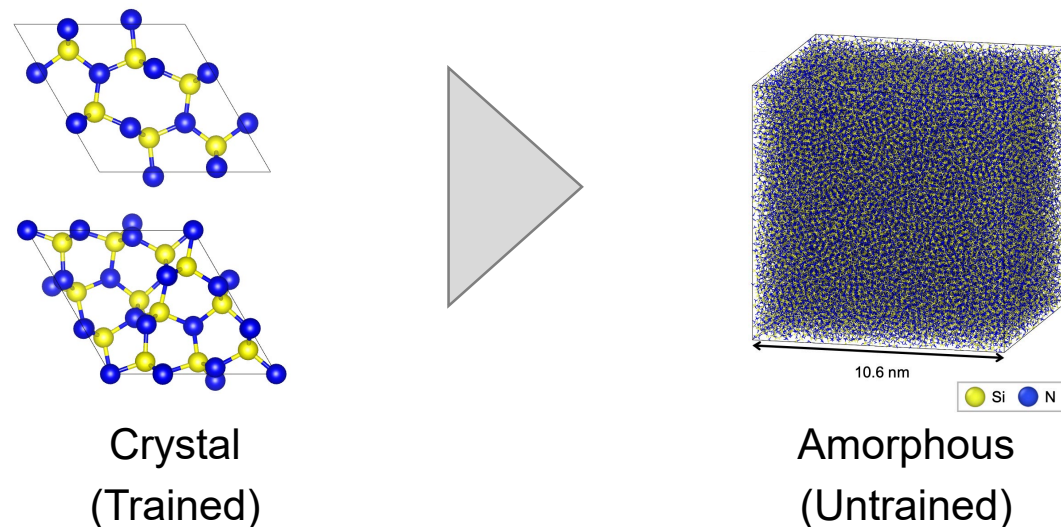
## Configurational extrapolation



# Compositional extrapolation is well described by the embedding characteristics of GNN-IPs



# How to explain configurational extrapolation?



Physical laws should be explicitly learned by MLIP to extrapolate in configurational space.

Then, which physical interaction?

Kinetic energy:

$$E_{\text{kin},i} = -\frac{1}{2} \int \nabla_{\mathbf{r}}^2 \rho_{\text{at},i}(\mathbf{r}, \mathbf{r}')|_{\mathbf{r}=\mathbf{r}'} d\mathbf{r}'$$

XC energy:

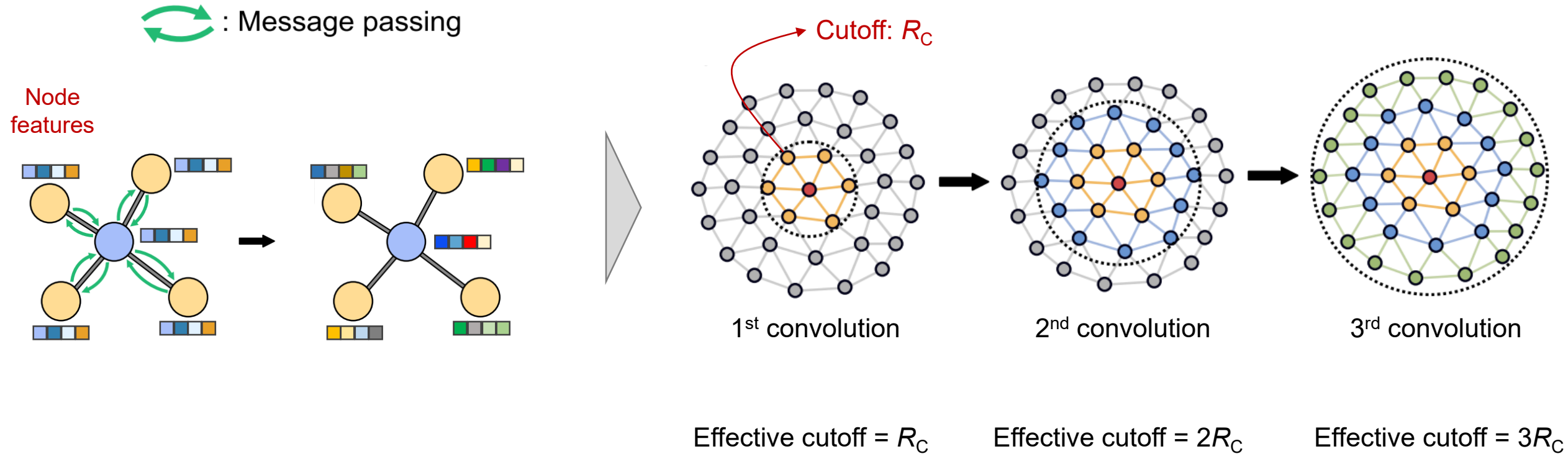
$$E_{\text{XC},i} = \int \rho_i(\mathbf{r}) \varepsilon_{\text{XC}}(\rho_i(\mathbf{r}), \nabla \rho_i(\mathbf{r})) d\mathbf{r}$$

Coulomb energy:

$$E_{\text{Coul},i} = \frac{1}{2} \sum_{j \neq i} \frac{\rho_{\text{tot},i}(\mathbf{r}) \rho_{\text{tot},j}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}' + \frac{1}{2} \int \frac{\rho_i(\mathbf{r}) \rho_i(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}' - \frac{1}{2} \int \frac{q_i \rho_i(\mathbf{r})}{|\mathbf{r} - \mathbf{r}_i|} d\mathbf{r}$$

Approximated  
to  $q_1 q_2 / r_{12}$

# Role of the message-passing algorithm



## Descriptor MLIPs

- Typically,  $R_C = 5\sim 6$  Å

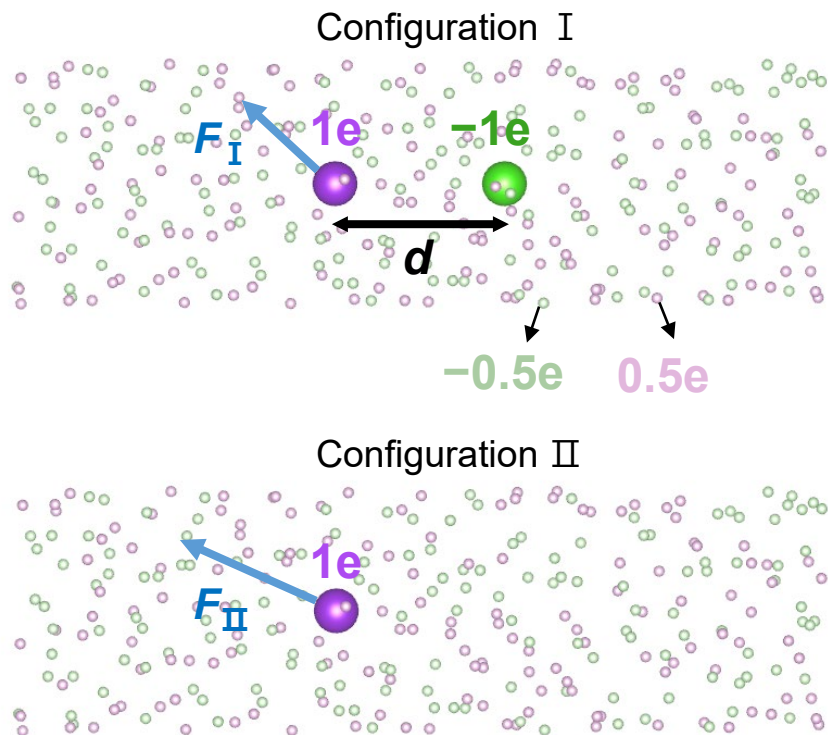
## Graph MLIPs

- Typically,  $R_C = 5\sim 6$  Å, No. of layers = 2~5
- Effective cutoff is 15 ~ 25 Å



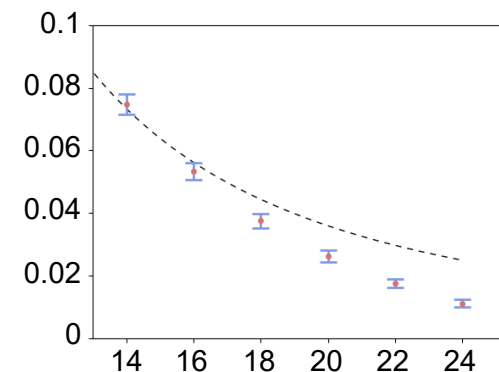
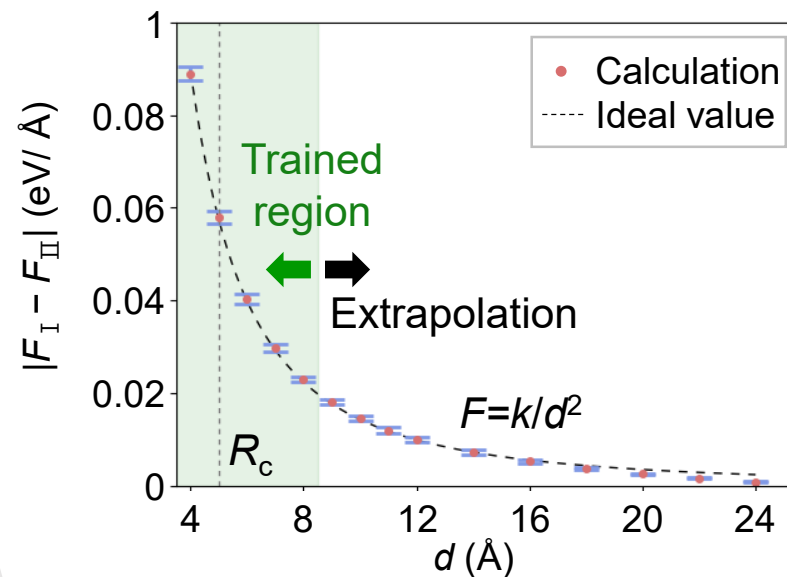
# Extrapolating Coulomb interactions using GNN-IPs

## Toy model

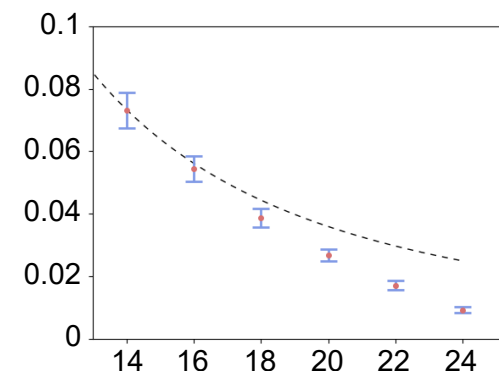
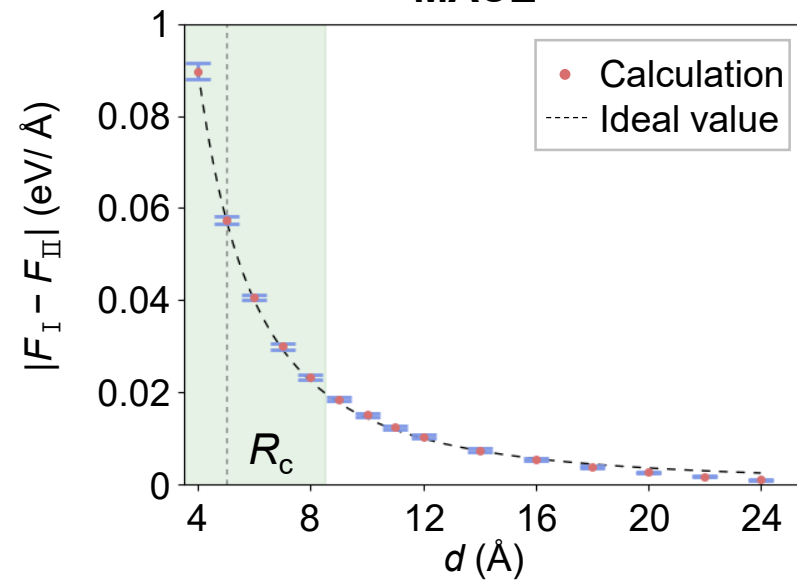


- 0.5 and -0.5 particles are placed randomly.
- Only Coulomb interactions are applied to the system non-periodically.
- **Training set:**  $d = 4, 5, 6, 7, 8 \text{ \AA}$
- **Cutoff radius ( $R_c$ )** =  $5 \text{ \AA}$ , 5 convolution layers

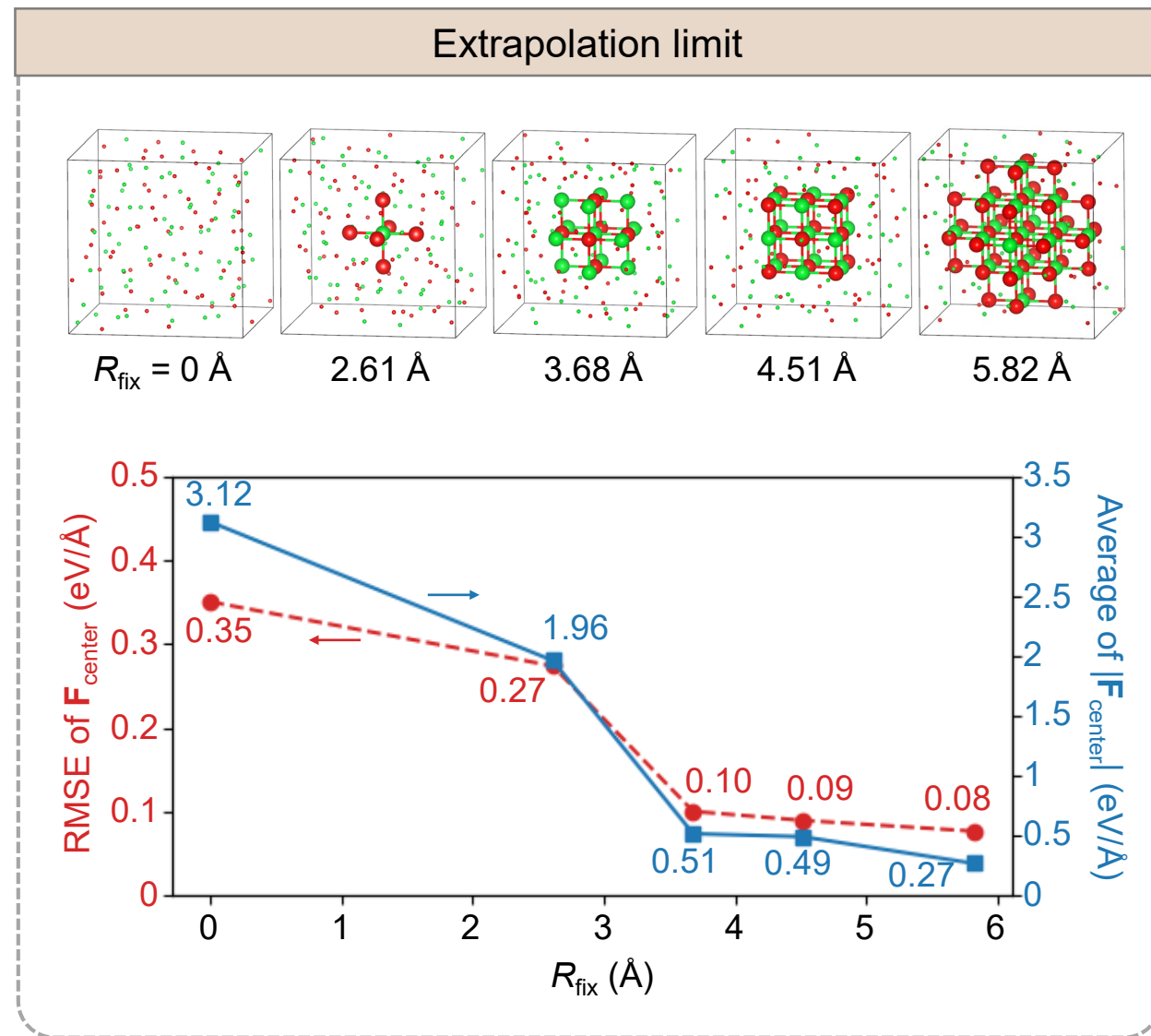
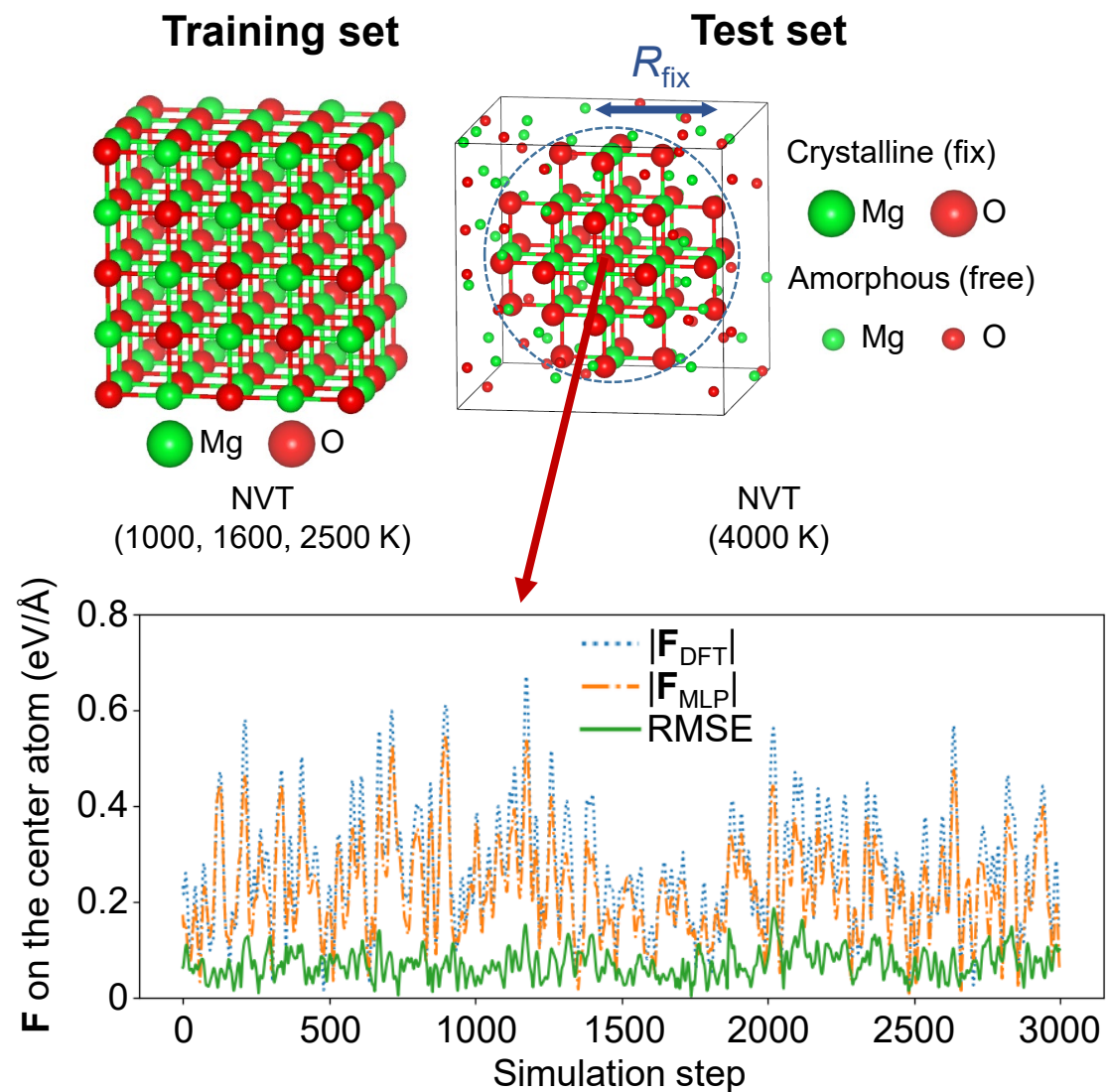
## <SevenNet>



## <MACE>



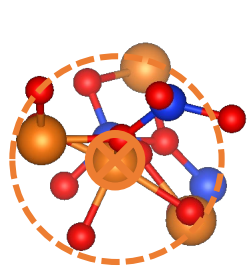
# Learning long-range forces from crystal structures



Electrostatic interactions in disordered structures are predicted by the potential trained from the crystal structure.

# Role of equivariance

Descriptor model = Invariant model

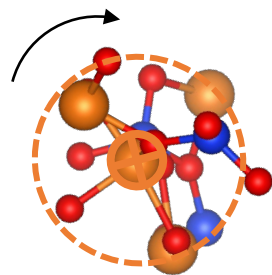


$\mathbf{x}$



$f(\mathbf{x})$

$=$



$R\mathbf{x}$



$f(R\mathbf{x})$

Descriptor  
(input)



$g(f(\mathbf{x}))$

$=$



$g(f(R\mathbf{x}))$

Hidden  
layers



Energy

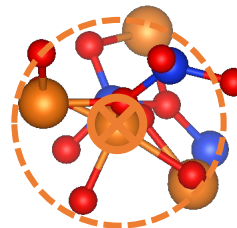
$=$



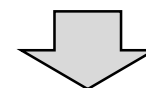
Energy

Output

Equivariant model

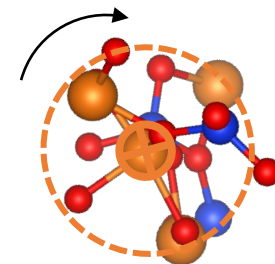


$\mathbf{x}$

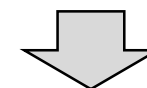


$f(\mathbf{x})$

$\neq$

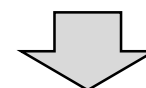
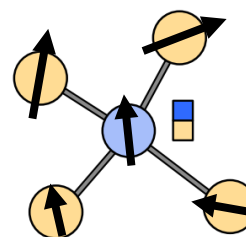


$R\mathbf{x}$



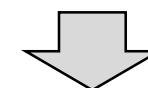
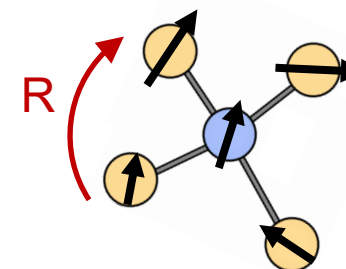
$f(R\mathbf{x}) = Rf(\mathbf{x})$

Convolution  
layers



Energy

$=$

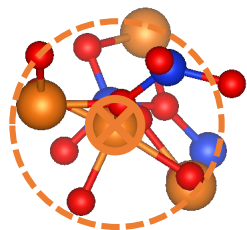


Energy

Output

# Role of equivariance

## Invariant model



$\mathbf{x}$



Descriptor  
(input)

$f(\mathbf{x})$

Structural  
representation



Hidden  
layers

$g(f(\mathbf{x}))$

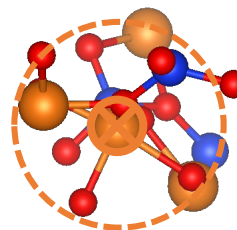
Energy  
regression



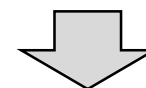
Output

Energy

## Equivariant model

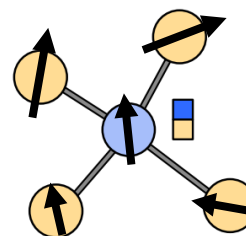


$\mathbf{x}$

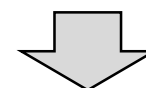


$f(\mathbf{x})$

Convolution  
layers



Structural representation +  
energy regression  
at the same time



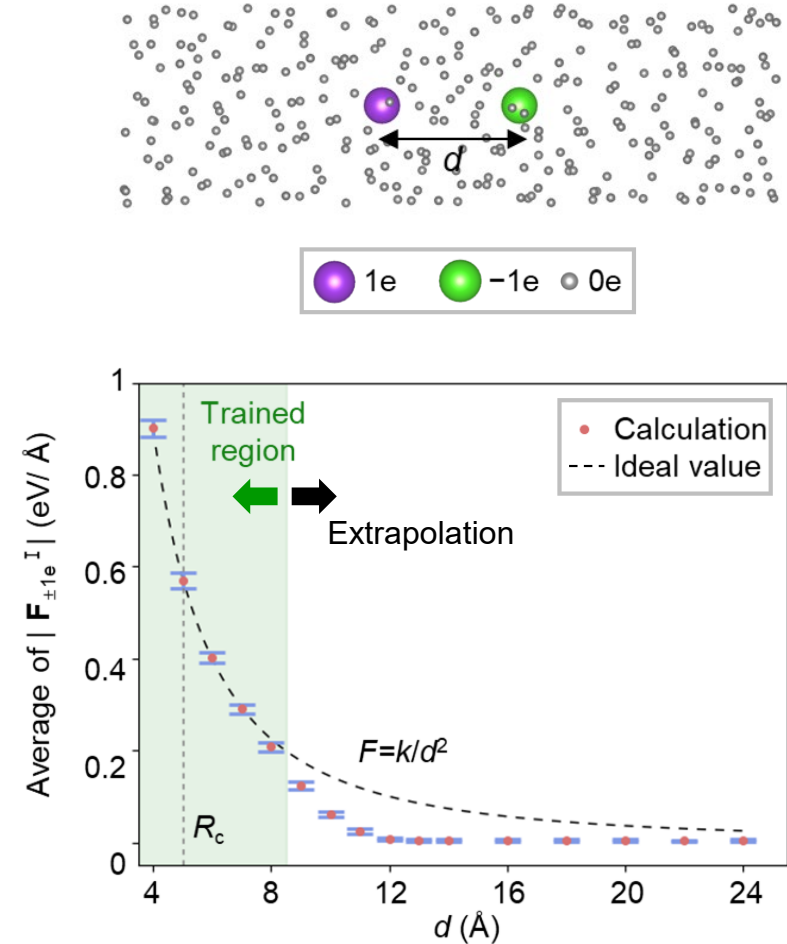
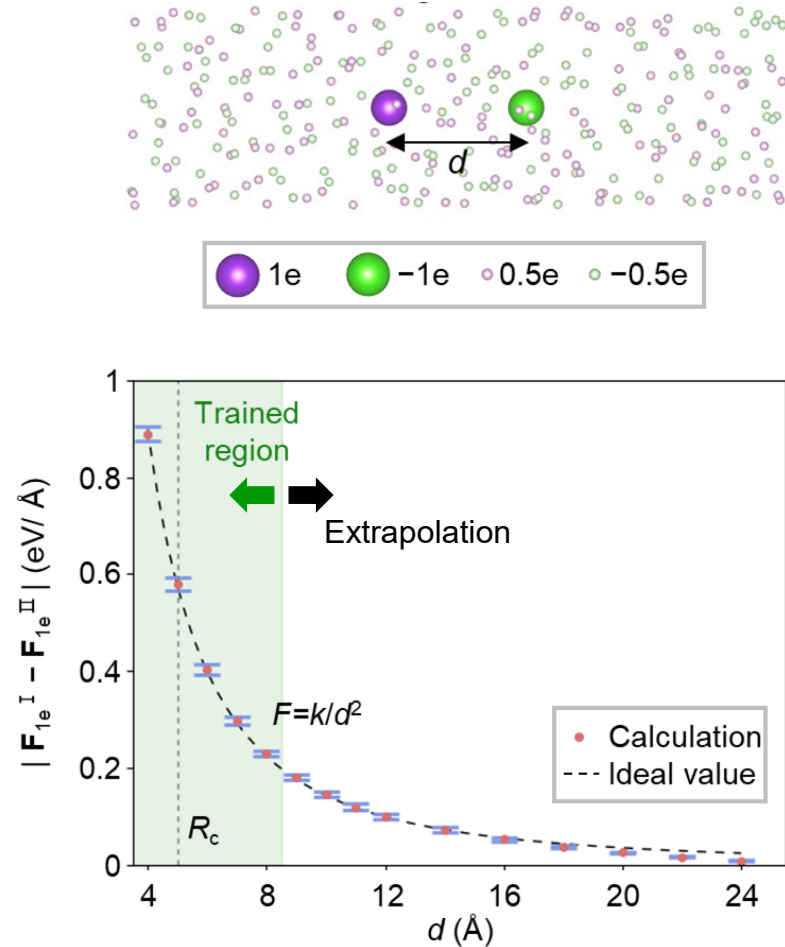
Output

Energy

→ MLIP learns effective  
structural representation  
way as well



# Charged medium vs Neutral medium



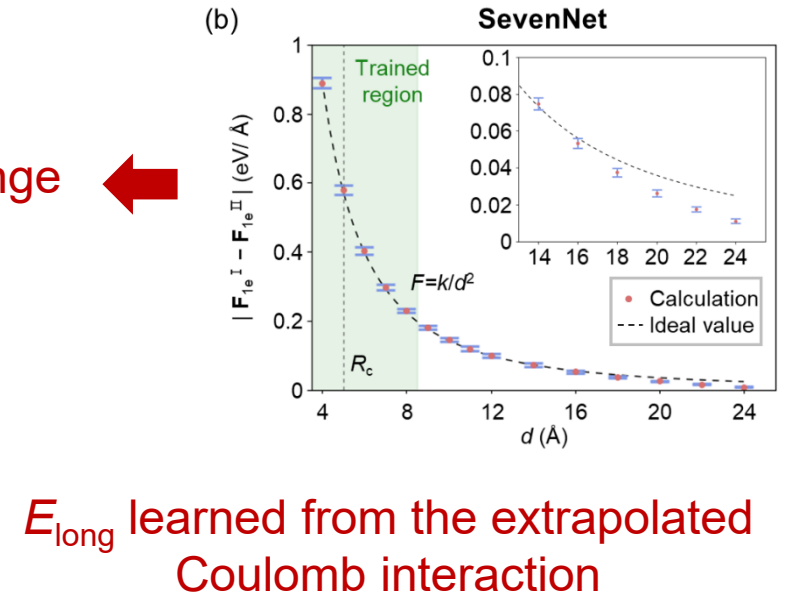
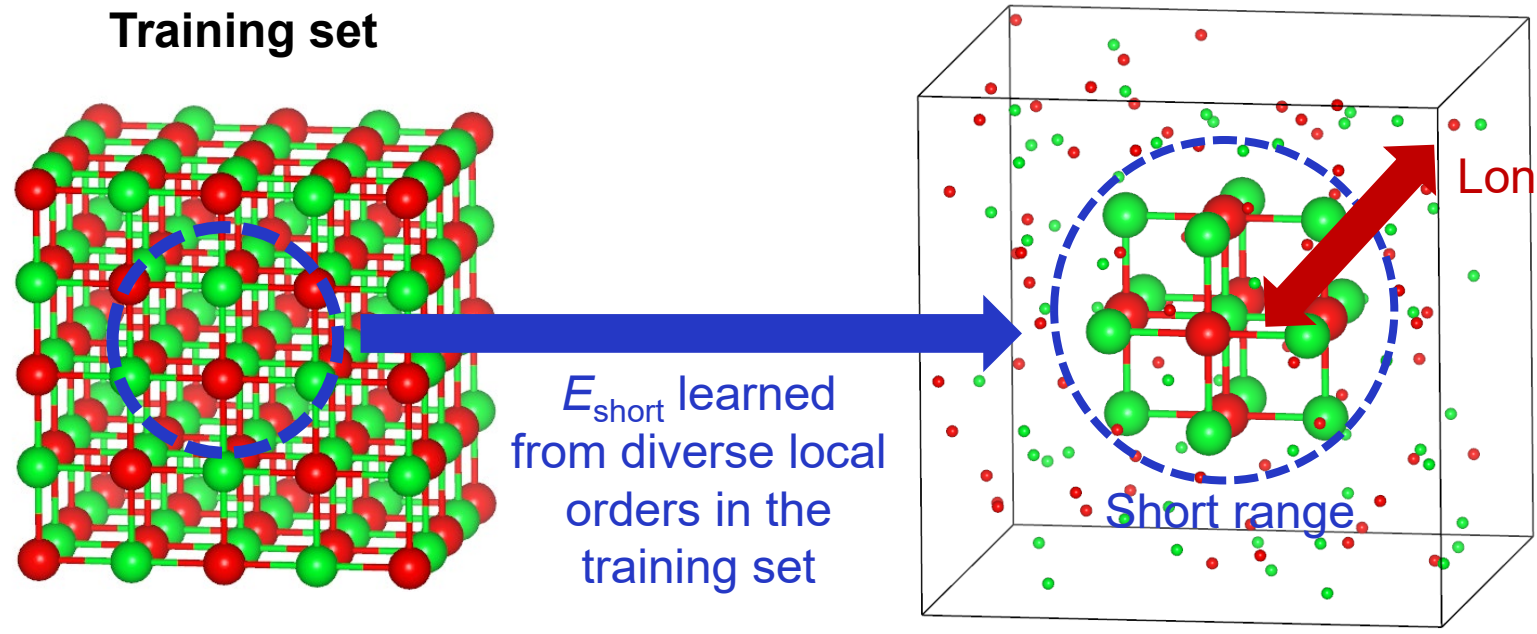
- GNN-IP well predicts electrostatic interaction in the charged medium, but fails in the neutral medium.
- It can be related to the fact that the structural representation and energetics is separated in the neutral medium case.

# How graph neural networks extrapolate?

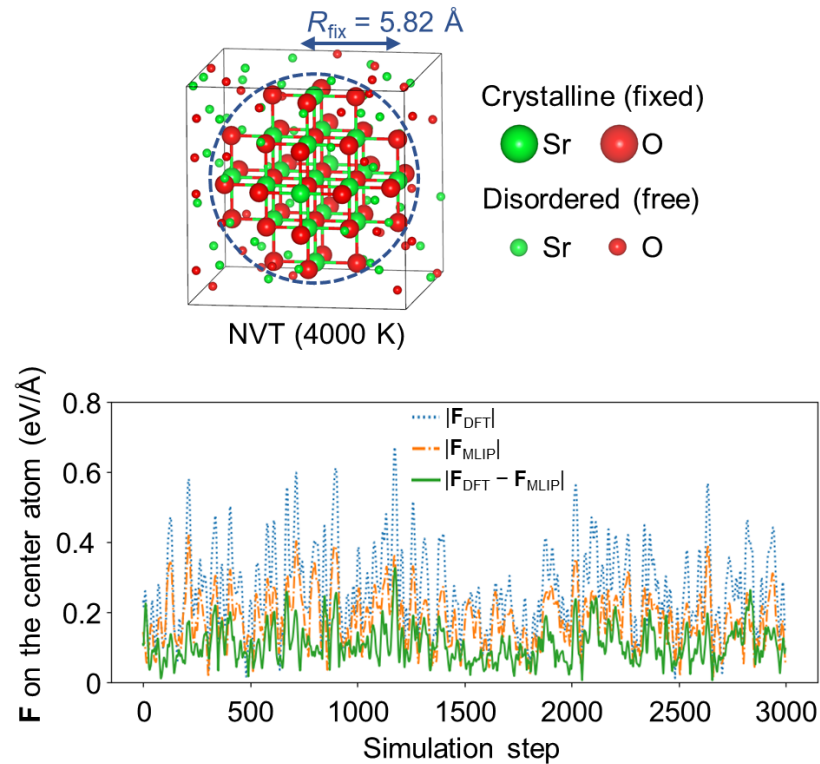
$$E_{\text{at},i} = \underbrace{E_{\text{kin},i}}_{\text{Short range}} + \underbrace{E_{\text{XC},i}}_{\text{Long range}} + \underbrace{E_{\text{Coul},i}}_{\text{Long range}}$$

↑  
Element embedding,  
Equivariance

↑  
Message-passing

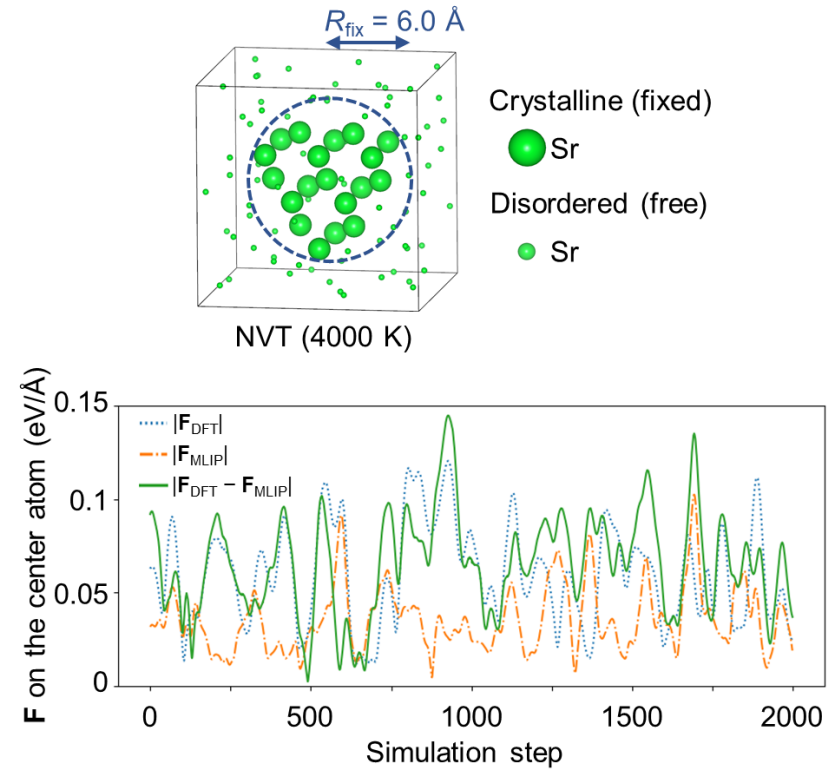


## Ionic system



- Coulomb term: extrapolate

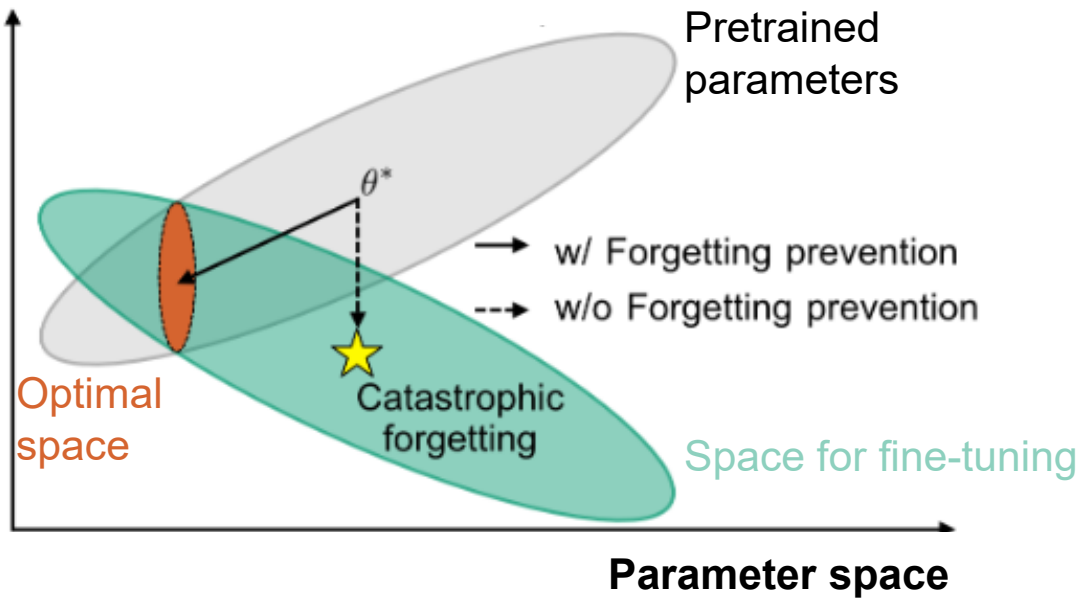
## Unary metallic system



- Kinetic term: do not extrapolate

# Forgetting-aware fine-tuning of universal MLIPs

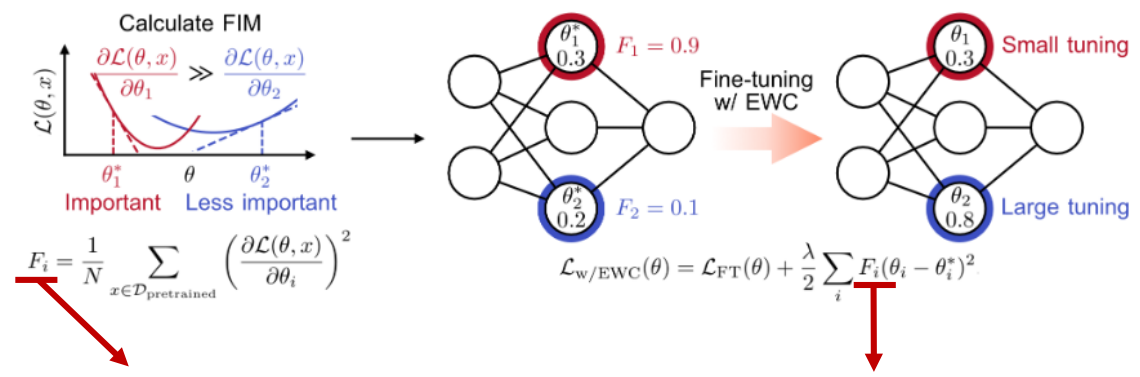
## Catastrophic forgetting



Catastrophic forgetting occurs if we fine-tune ML model with no forgetting-prevention methods.

## Forgetting-aware fine-tuning methods

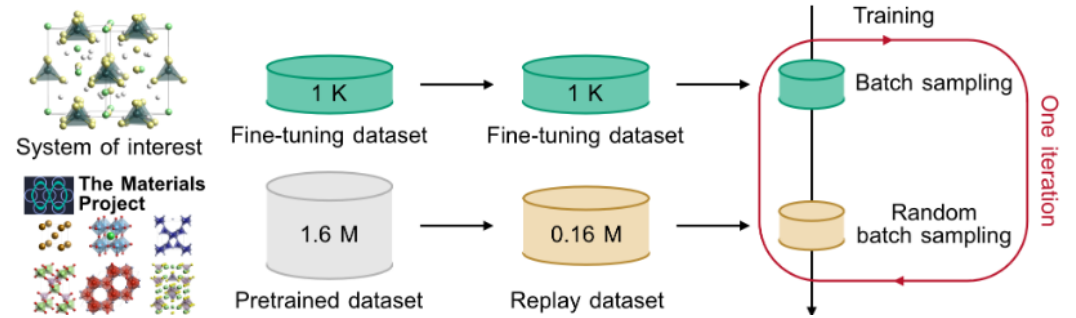
### Elastic weight consolidation (EWC)



Fisher information matrix: importance of parameters to certain training set

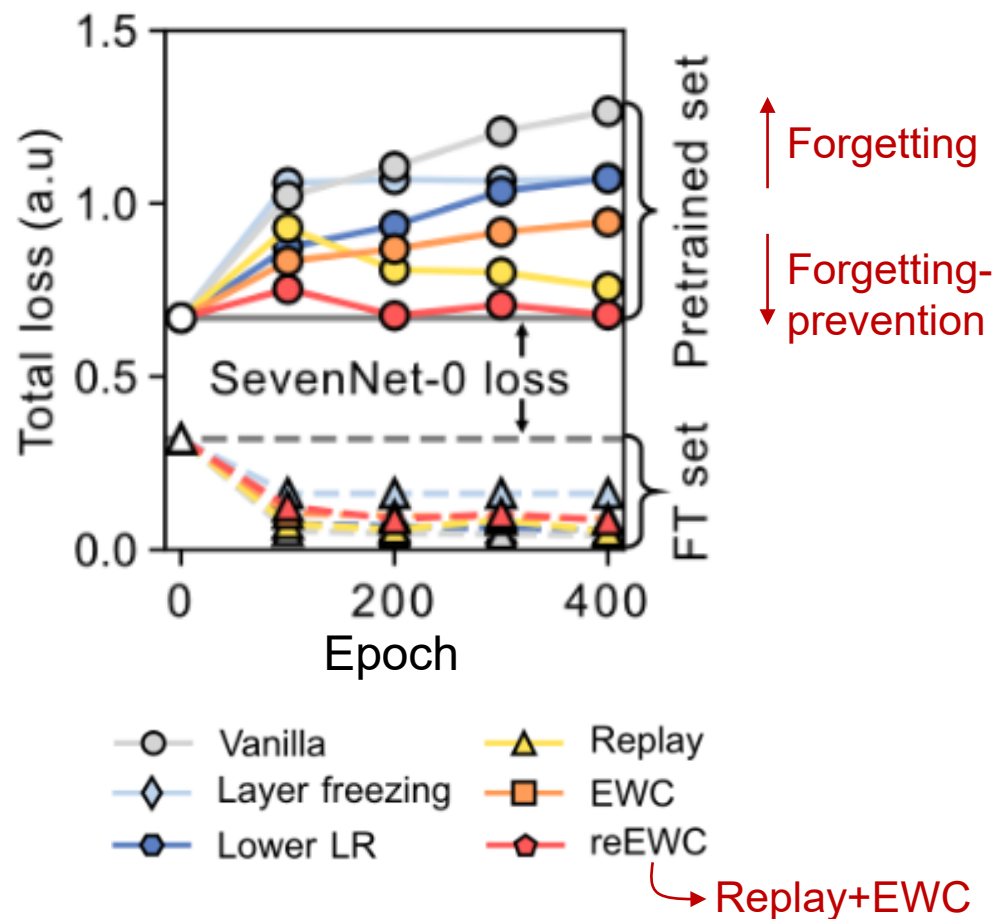
Fisher information matrix are used as weights for parameters during training

### Replay



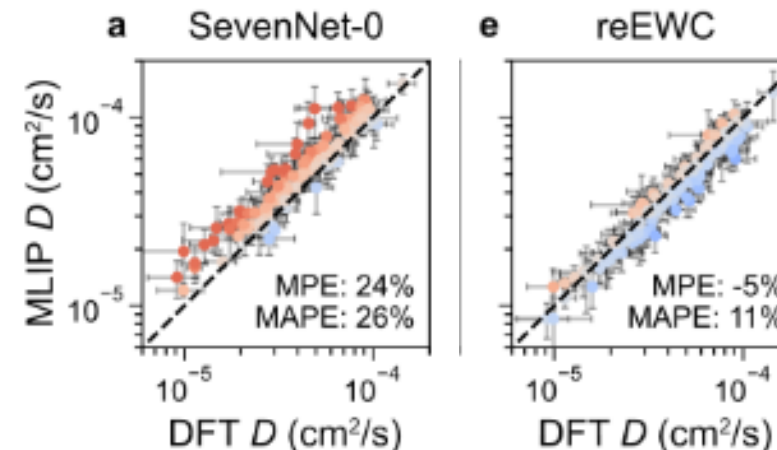


## Preventing forgetting for pretrained set

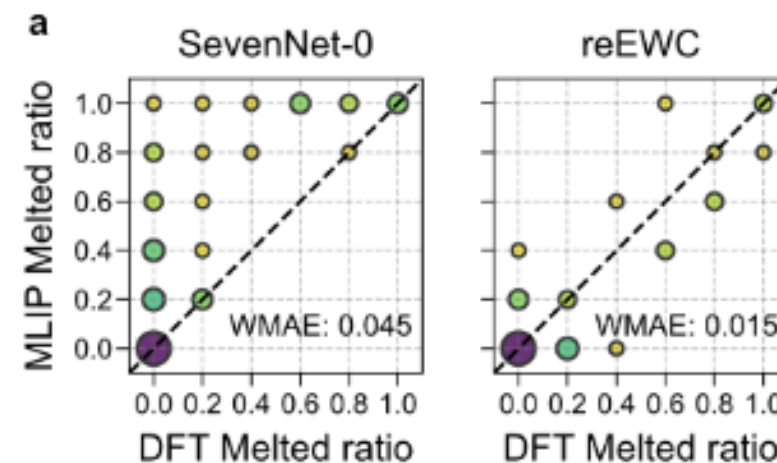


- Fine-tuning (FT) set: one MD trajectory of LPSC

## Li diffusivity

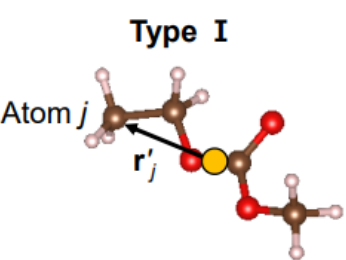
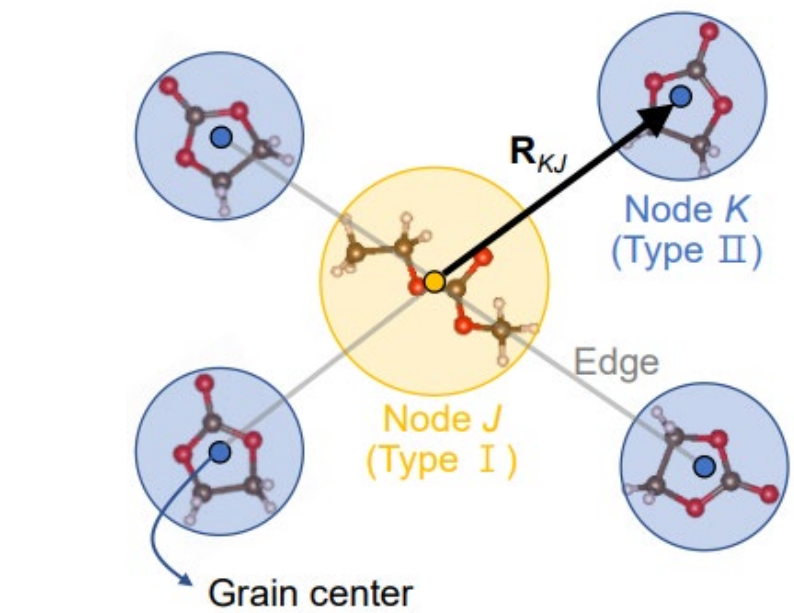


## MD stability



# Coarse-grained all-atom force field (CGAA-FF) formalism

## Grain embedding



**Grain embedding**

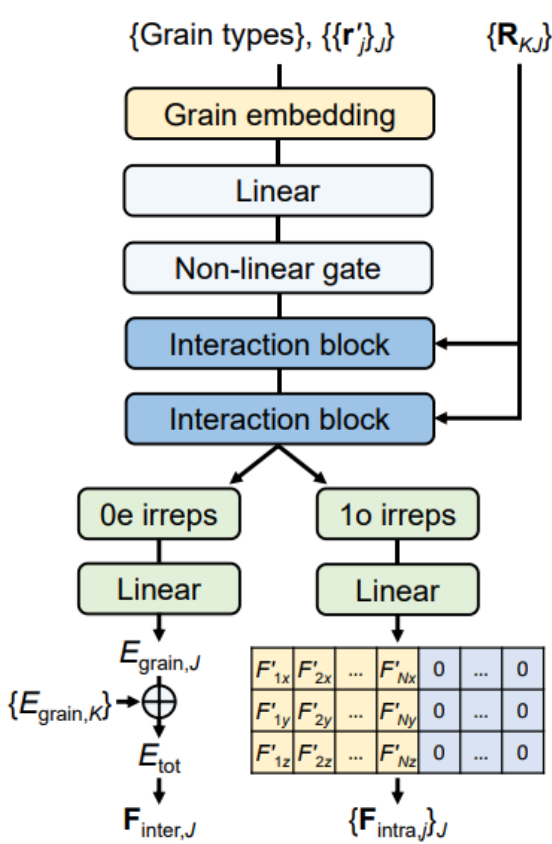
1o irreps	$r'_{1x}$	$r'_{2x}$	...	$r'_{Nx}$	0	...	0
	$r'_{1y}$	$r'_{2y}$	...	$r'_{Ny}$	0	...	0
	$r'_{1z}$	$r'_{2z}$	...	$r'_{Nz}$	0	...	0
0e irreps	1	1	...	1	0	...	0



**Type II**

1o irreps	0	0	...	0	$r'_{1x}$	...	$r'_{Mx}$
	0	0	...	0	$r'_{1y}$	...	$r'_{My}$
	0	0	...	0	$r'_{1z}$	...	$r'_{Mz}$
0e irreps	0	0	...	0	1	...	1

## Network architecture



$$F_{j\alpha} = -\frac{\partial E_{\text{tot}}}{\partial r_{j\alpha}} = -\sum_{k \in \{n_J\}} \frac{\partial E_{\text{tot}}}{\partial r'_{k\alpha}} \frac{\partial r'_{k\alpha}}{\partial r_{j\alpha}} - \frac{\partial E_{\text{tot}}}{\partial R_{J\alpha}} \frac{\partial R_{J\alpha}}{\partial r_{j\alpha}}$$

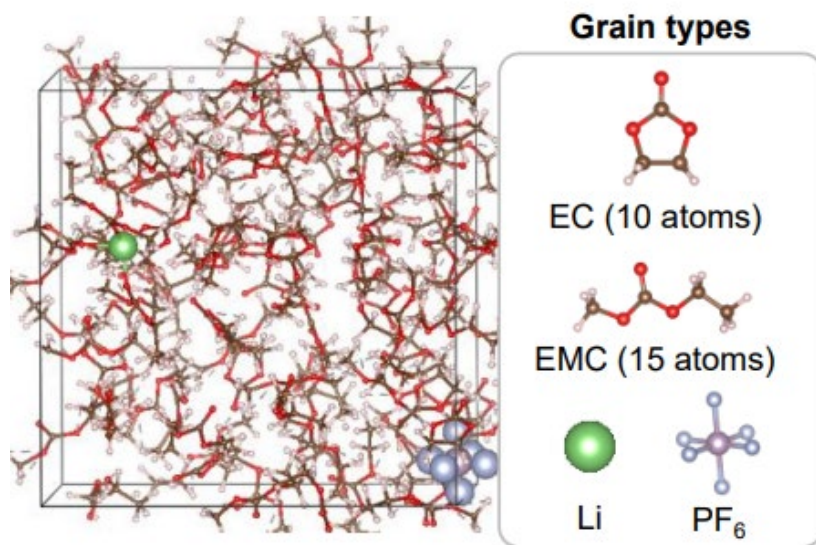
$$= -\frac{\partial E_{\text{tot}}}{\partial r'_{j\alpha}} + \frac{1}{N_J} \sum_{k \in \{n_J\}} \frac{\partial E_{\text{tot}}}{\partial r'_{k\alpha}} - \frac{1}{N_J} \frac{\partial E_{\text{tot}}}{\partial R_{J\alpha}}$$

Inter-grain force ( $F_{\text{inter}}$ )

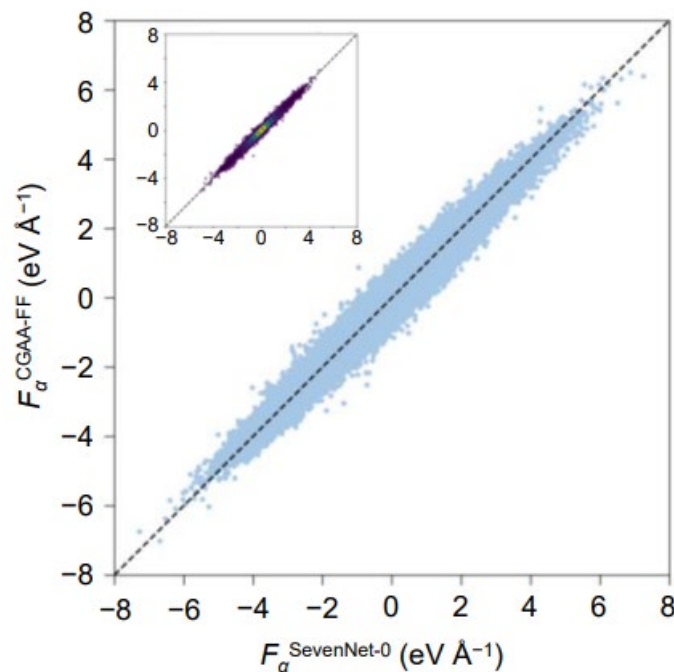
Intra-grain force ( $F_{\text{intra}}$ )

→ New formalism that enables all-atom force predictions in coarse-grained energy model

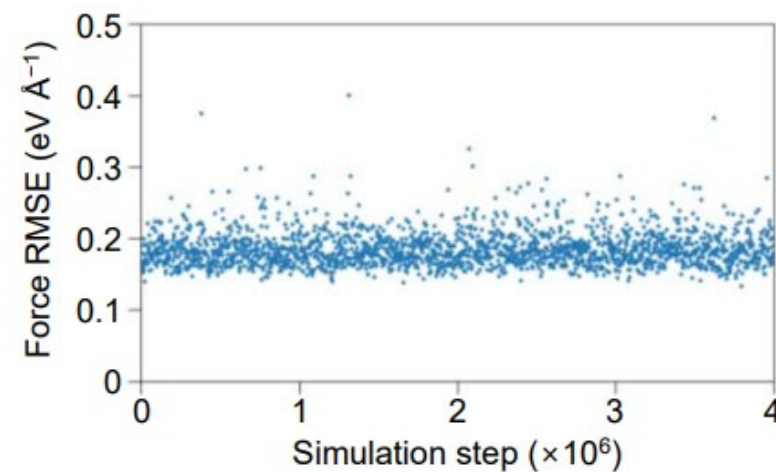
## Test systems: Li-ion battery electrolyte



## Training performance



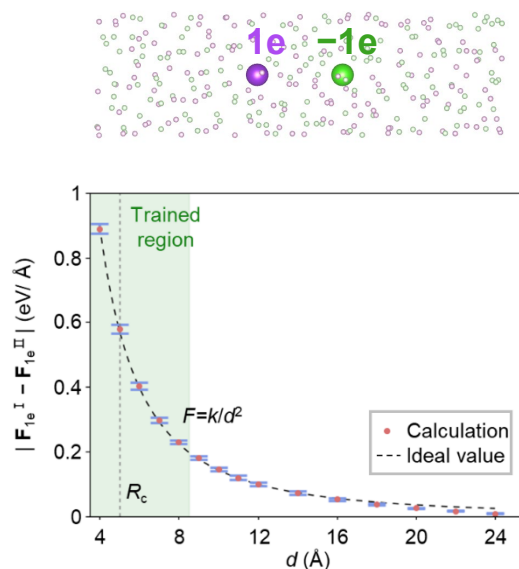
## Test simulation



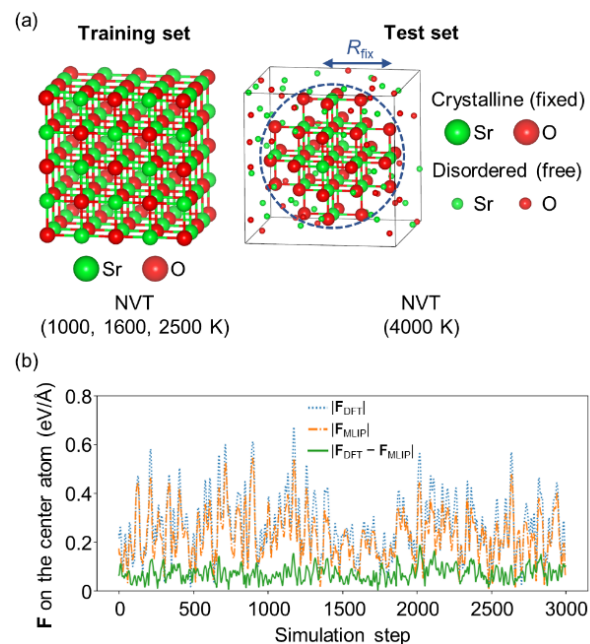
- Energy RMSE = 4.96 meV atom<sup>-1</sup>
- Force RMSE = 0.201 eV Å<sup>-1</sup>
- Speed: 22 times improved
- Memory: 14 times improved

## Extrapolation of Coulomb interactions via GNN-IPs

### Toy model



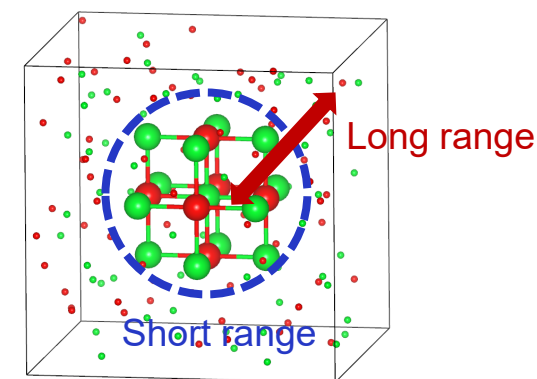
### DFT



$$E_{at,i} = E_{kin,i} + E_{XC,i} + E_{Coul,i}$$

Short range Long range

Element embedding Message-passing, Equivariance



- Universal interatomic potentials based on GNN-IP models well extrapolate to untrained domain and configurations.
- Extrapolation capability of GNN-IPs originate from the extrapolation capability in Coulomb interactions, and enhanced representation from the equivariant features.