東京大学大学院工学系研究科 社会連携・産学協創推進室 主催
ワークショップ：マテリアルズ・インフォマティクスの現状と将来展望
2018年1月29日(月)、東京大学工学部2号館

アモルファス材料中のイオン移動挙動解析のためのニューラルネットワーク原子間ポテンシャルの開発

東大院工 マテリアル工学専攻
NIMS 統合型材料開発・情報基盤部門 情報統合型物質・材料研究拠点
渡邉聡

共同研究者：
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Atom/ion diffusion

Important roles in novel nano-scale devices

Resistive RAM (ReRAM)

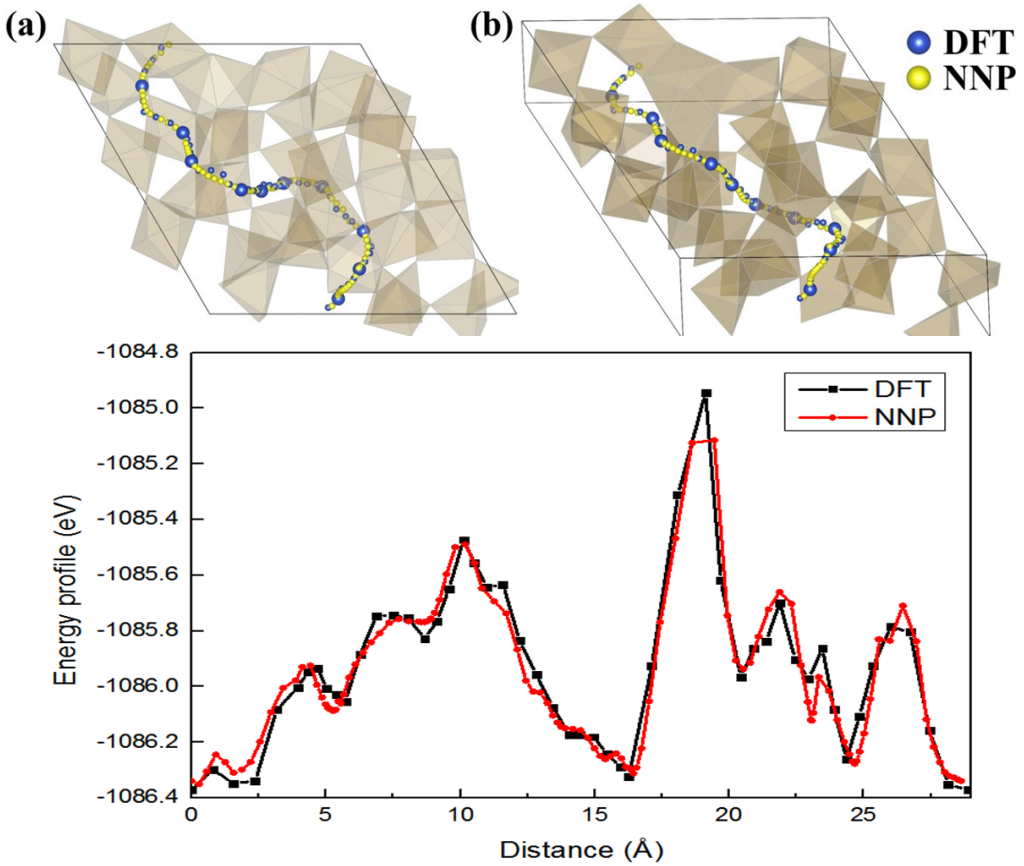
https://phys.org/news/2013-04-battery-memory-device-future-nanoelectronic.html

Solid state batteries

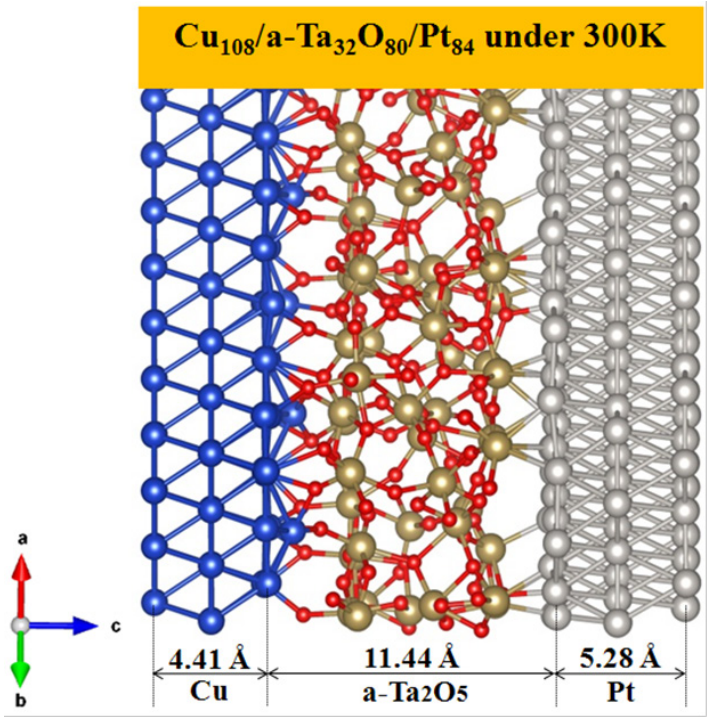
https://www.nanowerk.com/nanotechnology-news/newsid=41813.php

Possible contribution of simulations

- Material ⇔ properties: relation is not straightforward
  - Defects, disorder (**amorphous**), interfaces…
  - Good descriptors: not yet available
  - Experimental data: often insufficient



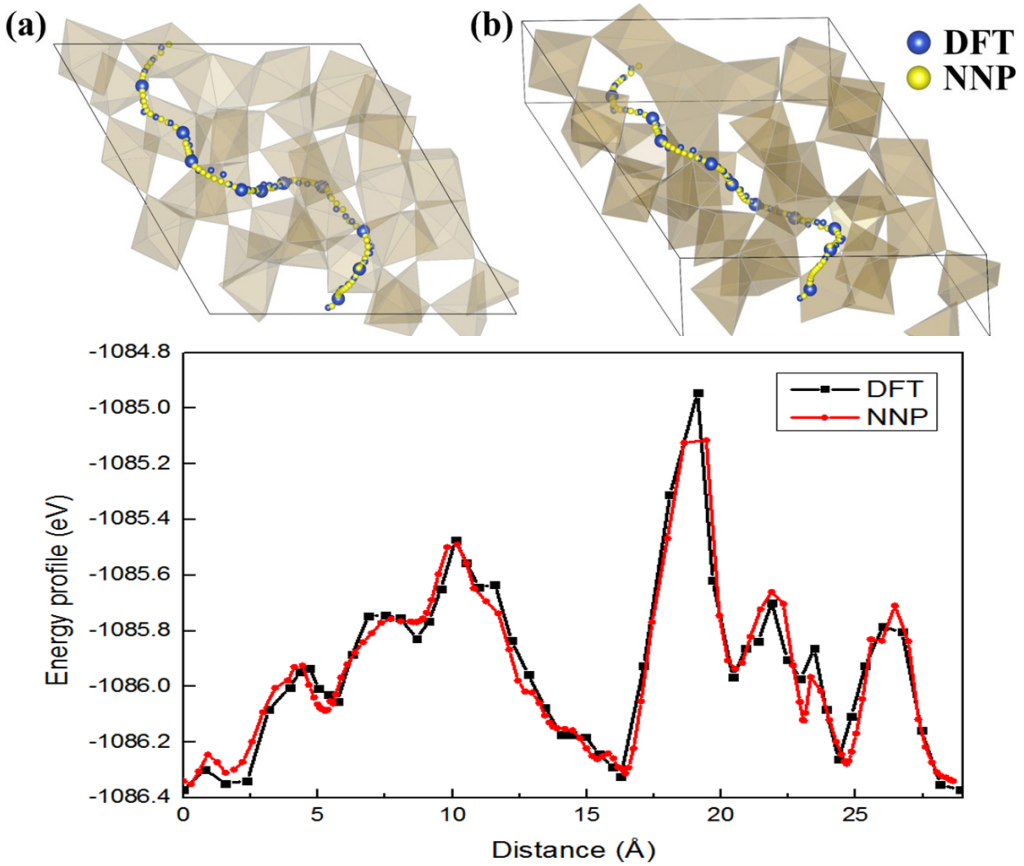
W. Li et al., J. Phys. Soc. Jpn. 86 (2017) 104004



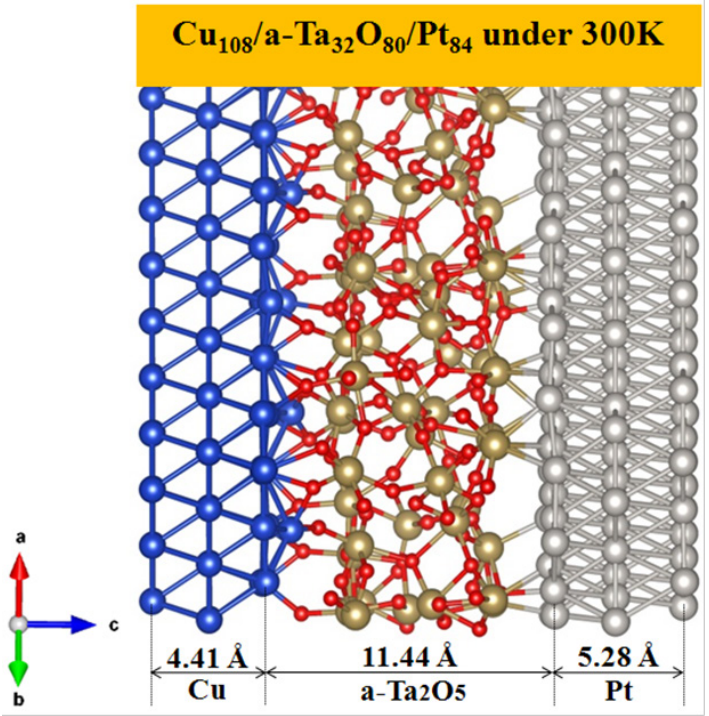
Xiao and Watanabe, *ACS Appl. Mater. Interfaces* 7, 519 (2015).

Conventional approaches of simulations

- First-principles (e.g. density functional theory):
  reliable, but heavy computation (crucial for amorphous)
- Classical interatomic potential:
  light computation but not sufficiently reliable

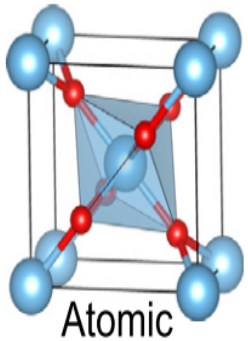


W. Li et al., J. Phys. Soc. Jpn. 86 (2017) 104004



Xiao and Watanabe, *ACS Appl. Mater. Interfaces* 7, 519 (2015).

Another approach: Machine learning interatomic potential



Atomic Coordinates

Black Box

E

$\{\vec{F}_i\}$

Energy and Atomic Forces

Machine learning methods

- Neural network
- Kernel regression
- Linear regression..

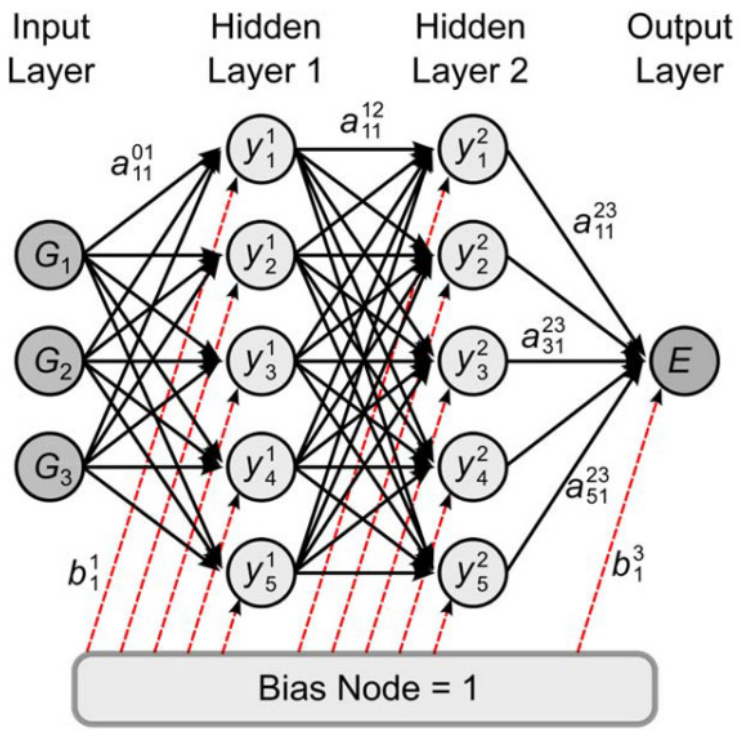
Gaussian approximation potential

High-dimensional Neural Network Potential

Sparse linear regression potential

Kernel regression force field

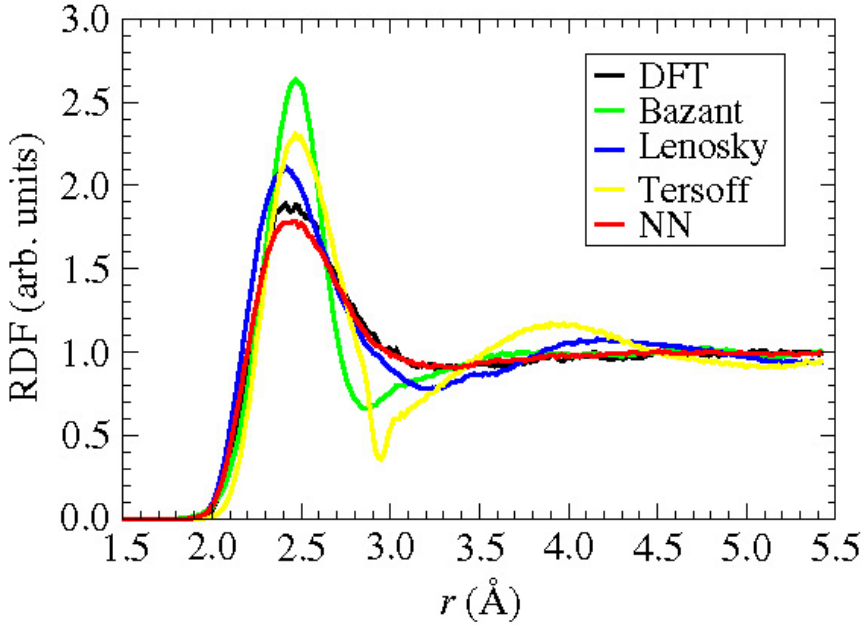
Interatomic potential constructed with neural network



$$y_i^j = f_i^j\left(b_i^j + \sum_{k=1}^{N_{j-1}} a_{k,i}^{j-1,j} \cdot y_k^{j-1}\right)$$

Behler and Parrinello,
Phys. Rev. Lett. 98 (2007) 146401.

- Input : information of atomic arrangement
- Output: Energy of the system
- Hidden layers, complex network
  → flexibility of fitting: small error
  (5 meV/atom for Si)



Application to Cu migration in Ta_2O_5

- The total energy of a structure containing amorphous Ta_2O_5 matrix and 1 Cu consists of three parts:

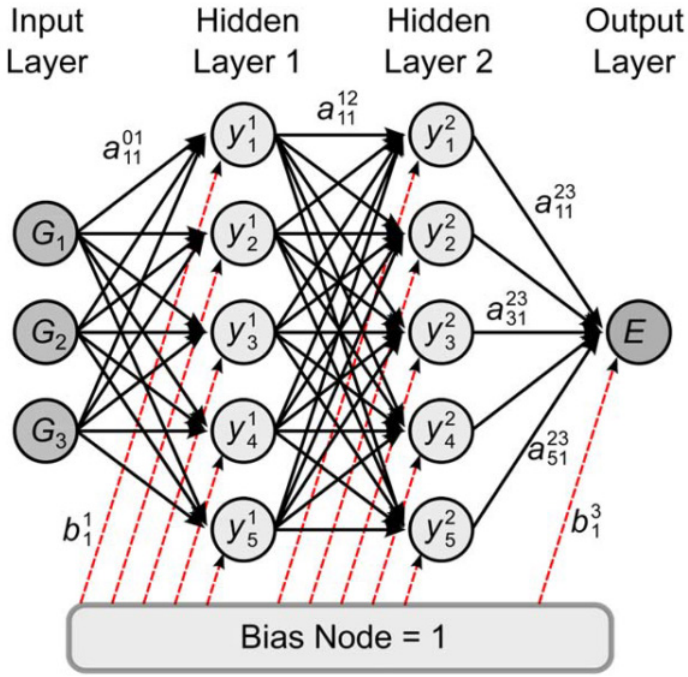
$$E = E_{\text{amorphous}} + E_{\text{Cu}} + E_{\text{relax}}$$

$E_{\text{amorphous}}$: Amorphous Ta_2O_5

E_{Cu}: Cu insertion

E_{relax}: Relaxation after Cu insertion

Represented by NN potential



◎ Ternary system, but only NN potential for Cu is considered

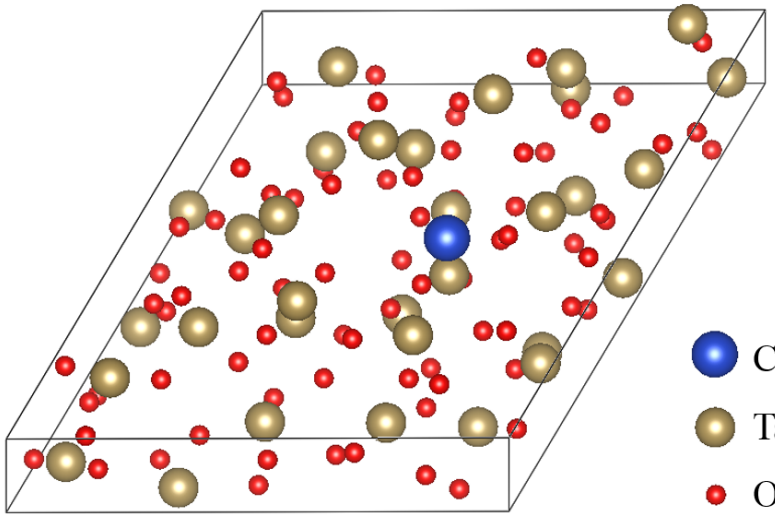
W. Li et al., J. Phys. Soc. Jpn. 86 (2017) 104004 (Editor's Choice)

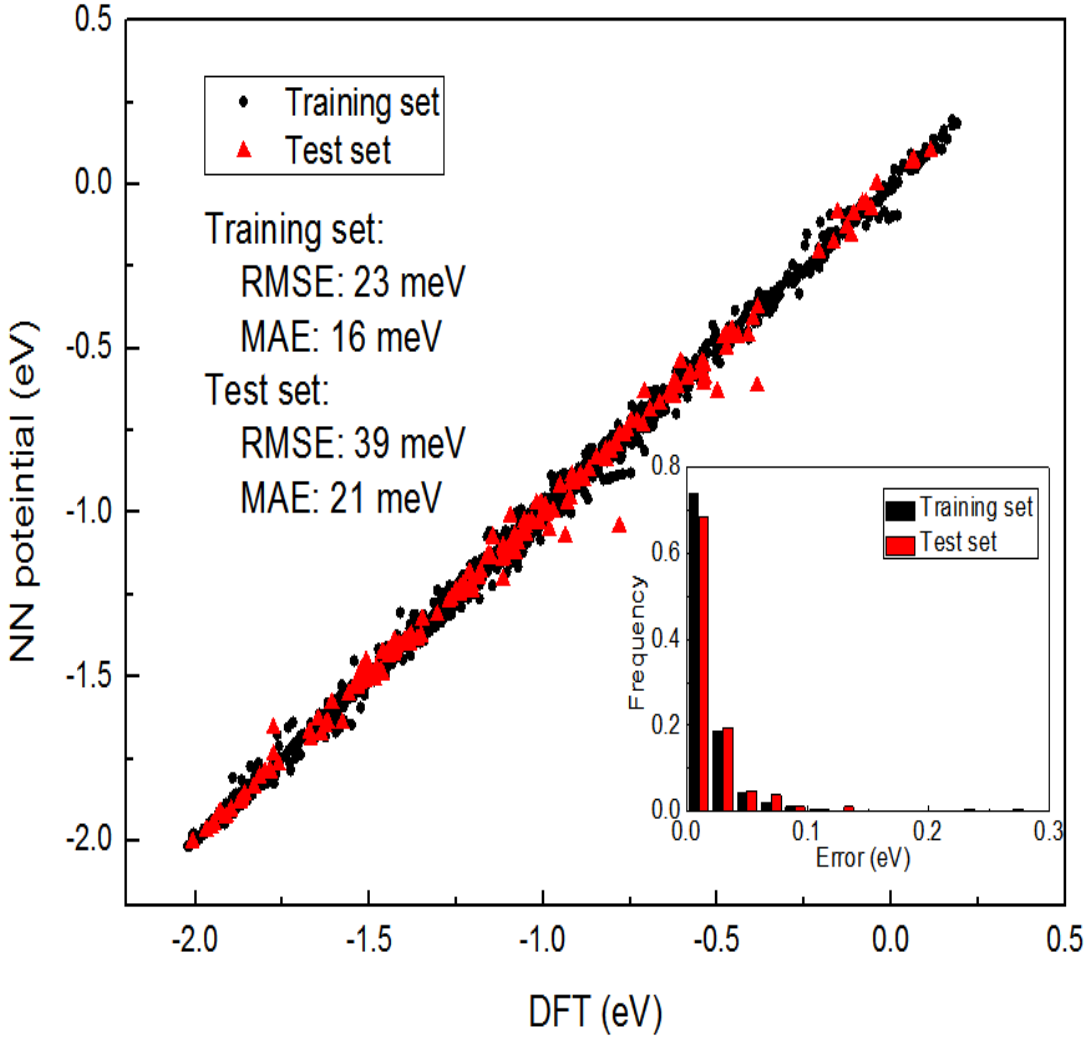
Energy comparison

- Density functional theory (DFT) vs. NN potential
- NN potential:

  trained with 1,800 data

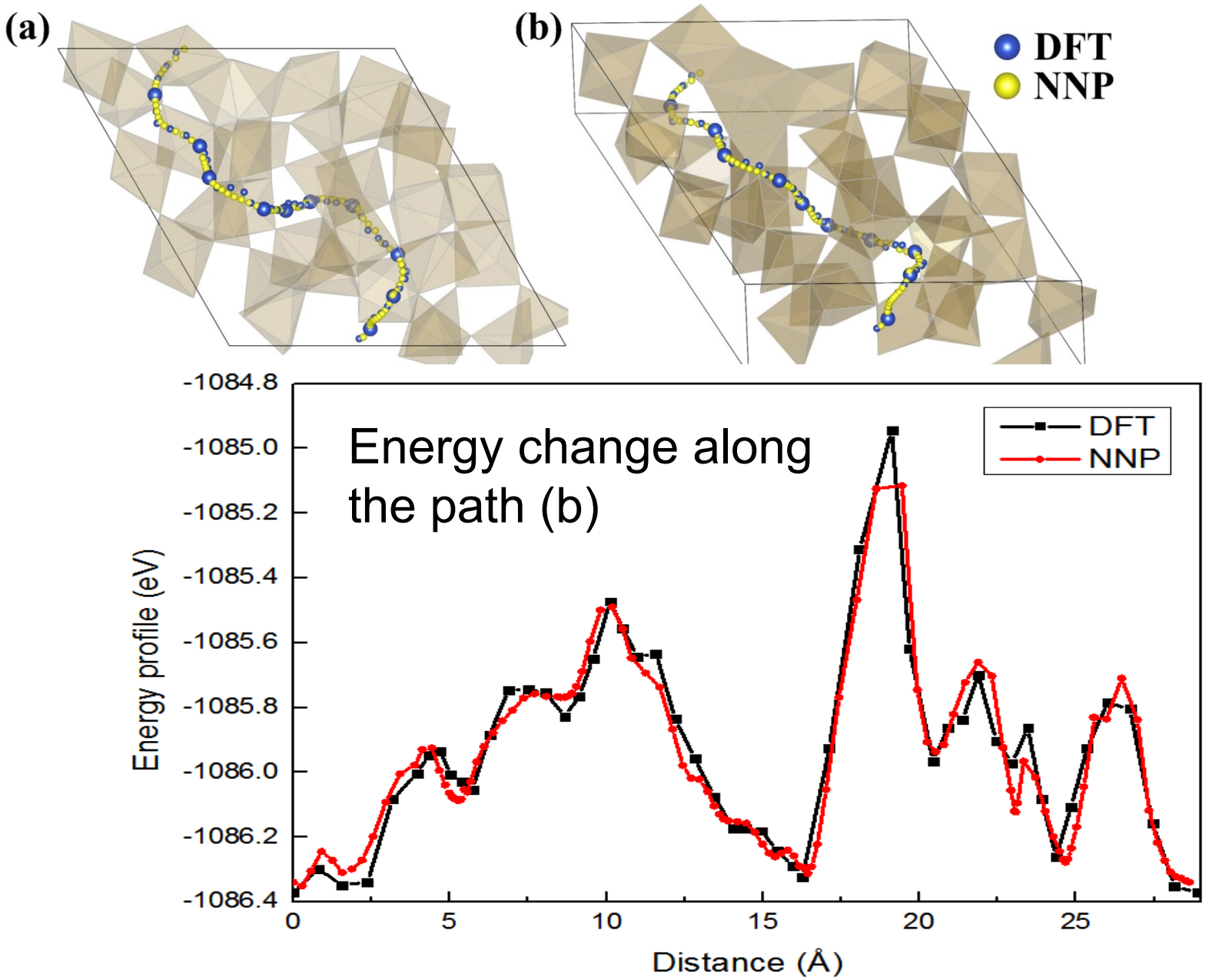
  testing data: 200





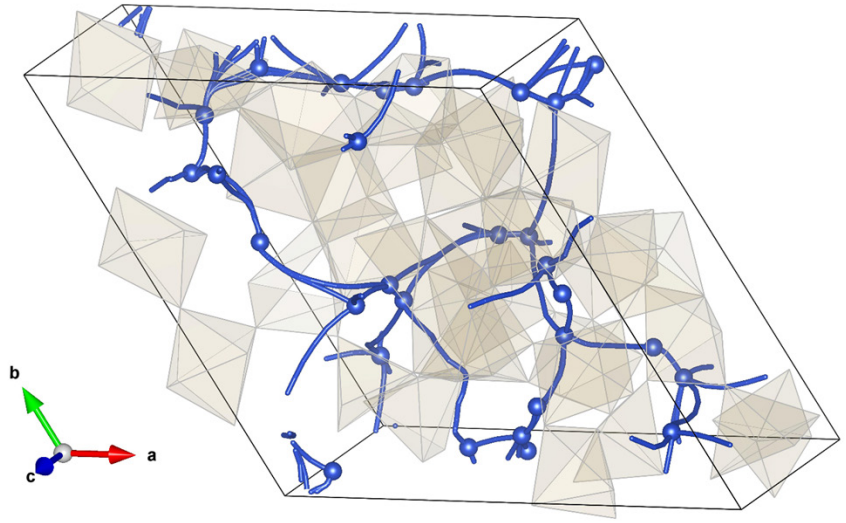
Li et al., *J. Phys. Soc. Jpn.* 86, 104004 (2017)

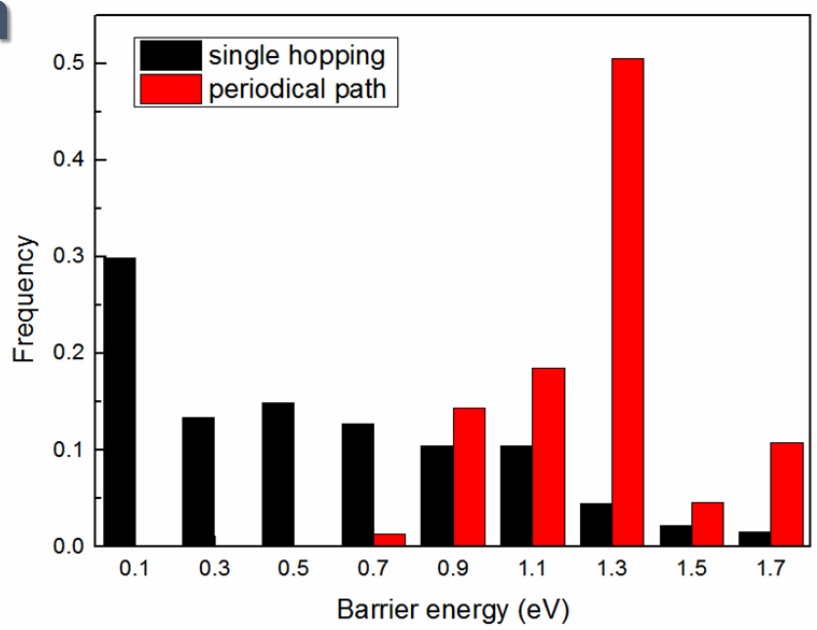
Diffusion path

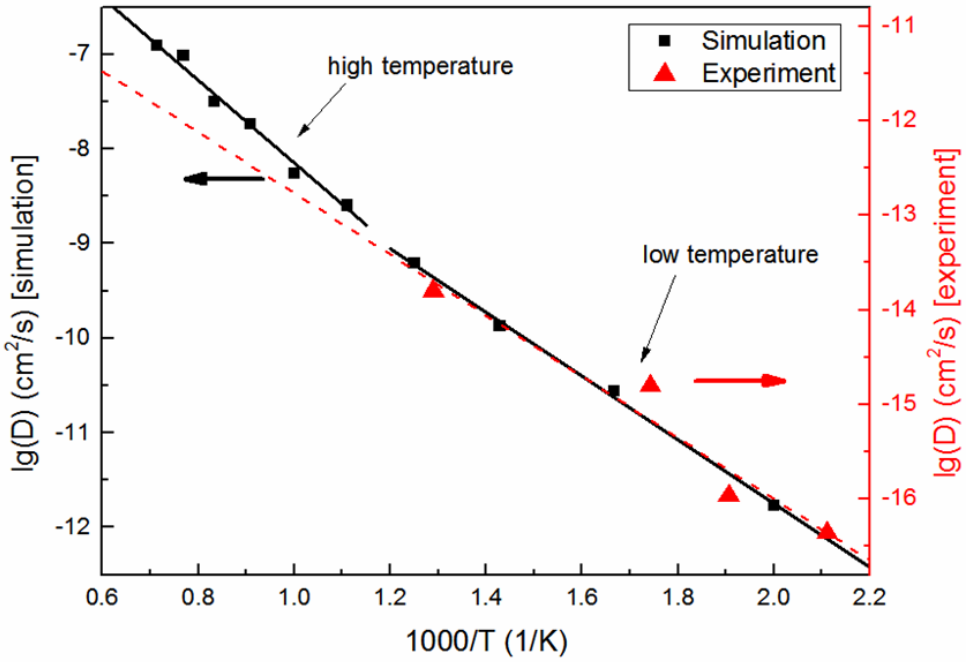


Barrier energy distribution

periodical path = path connecting a metastable site and the equivalent one in the adjacent super cell





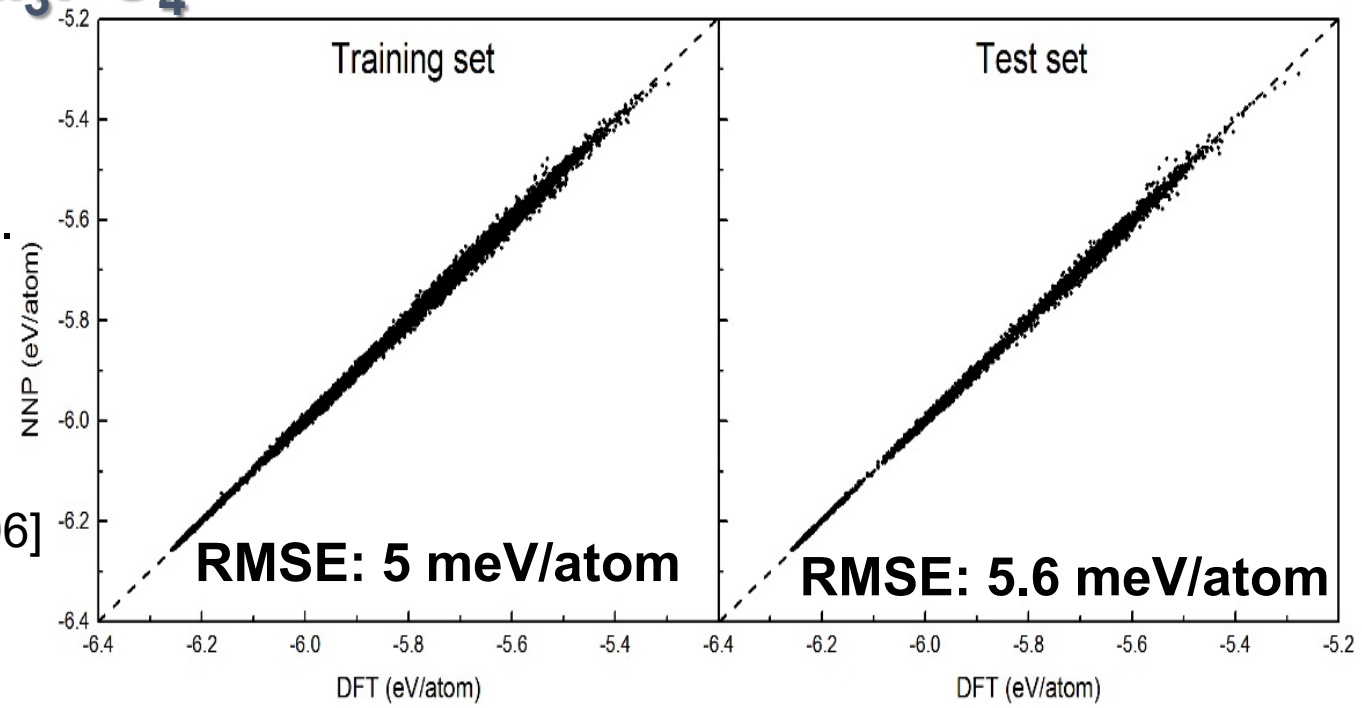


Kinetic Monte Carlo simulation
Activation energy at low T:
0.67eV (calc.)
cf. 0.64eV (exp.)

Example (2): Li_3PO_4

- Li, P & O are explicitly considered.
- Training set: 30874
  Testing set: 7718

[W. Li et al., J. Chem. Phys. 147 (2017) 214106]



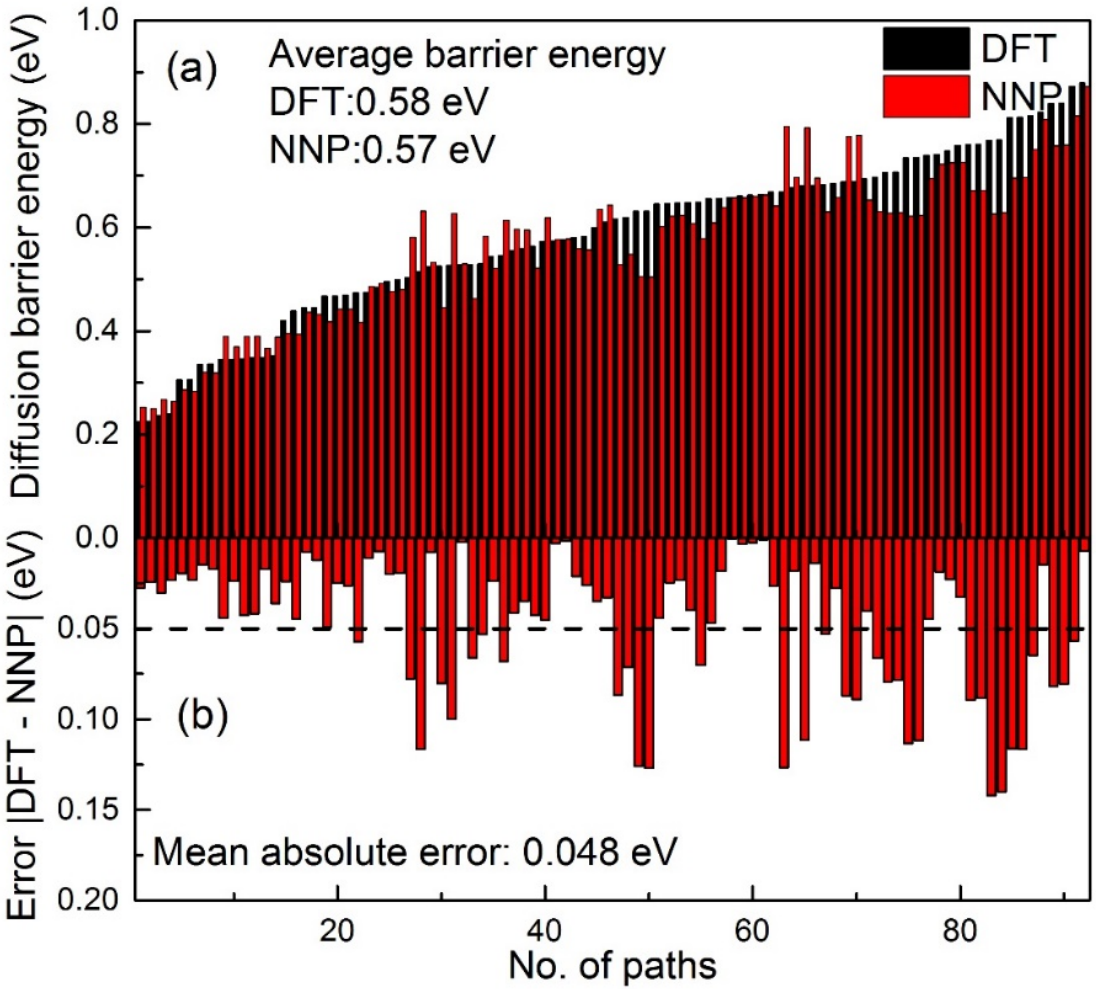
Properties of γ and β crystal Li_3PO_4

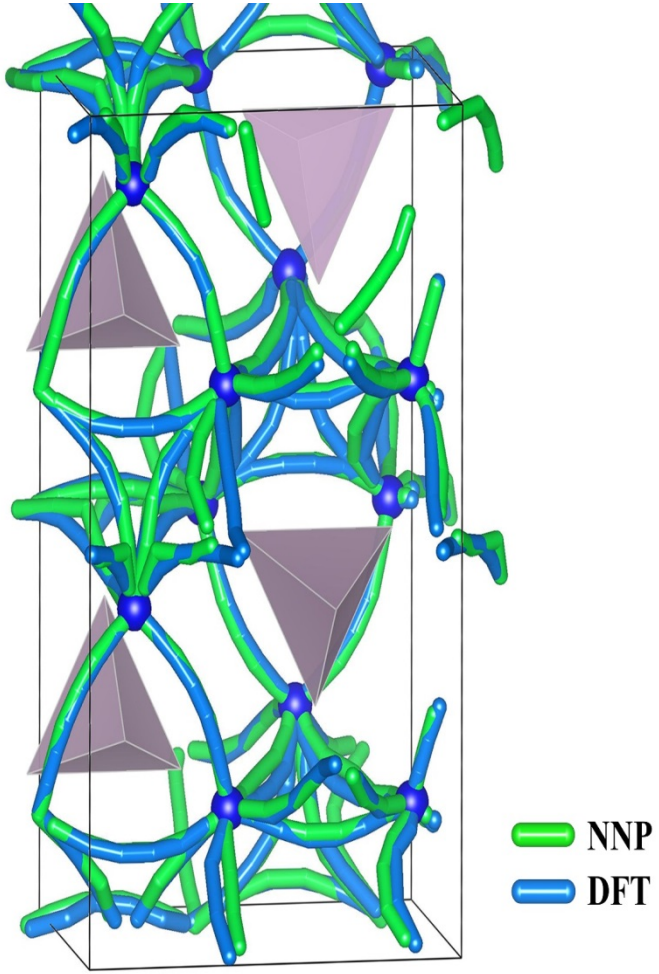
	NNP		DFT		Expt.
	Energy (eV)	Lattice constant (Å)	Energy (eV)	Lattice constant (Å)	Lattice constant (Å)
γ- Li_3PO_4	-200.96	a = 5.003 b = 6.108 c = 10.502	-200.94	a = 5.001 b = 6.177 c = 10.615	a = 4.927 b = 6.120 c = 10.490
β- Li_3PO_4	-100.49	a = 4.914 b = 5.287 c = 6.164	-100.48	a = 4.923 b = 5.298 c = 6.177	a = 4.856 b = 5.240 c = 6.115

* O. V. Yakubovich and V. S. Urusov, Crystallogr. Rep. 42, 261 (1997).

Diffusion paths and barrier energies

- Diffusion paths in *a*-Li_3PO_4





46 diffusion paths in the amorphous Li_3PO_4 structure

Transferability to Larger models

From experiment:

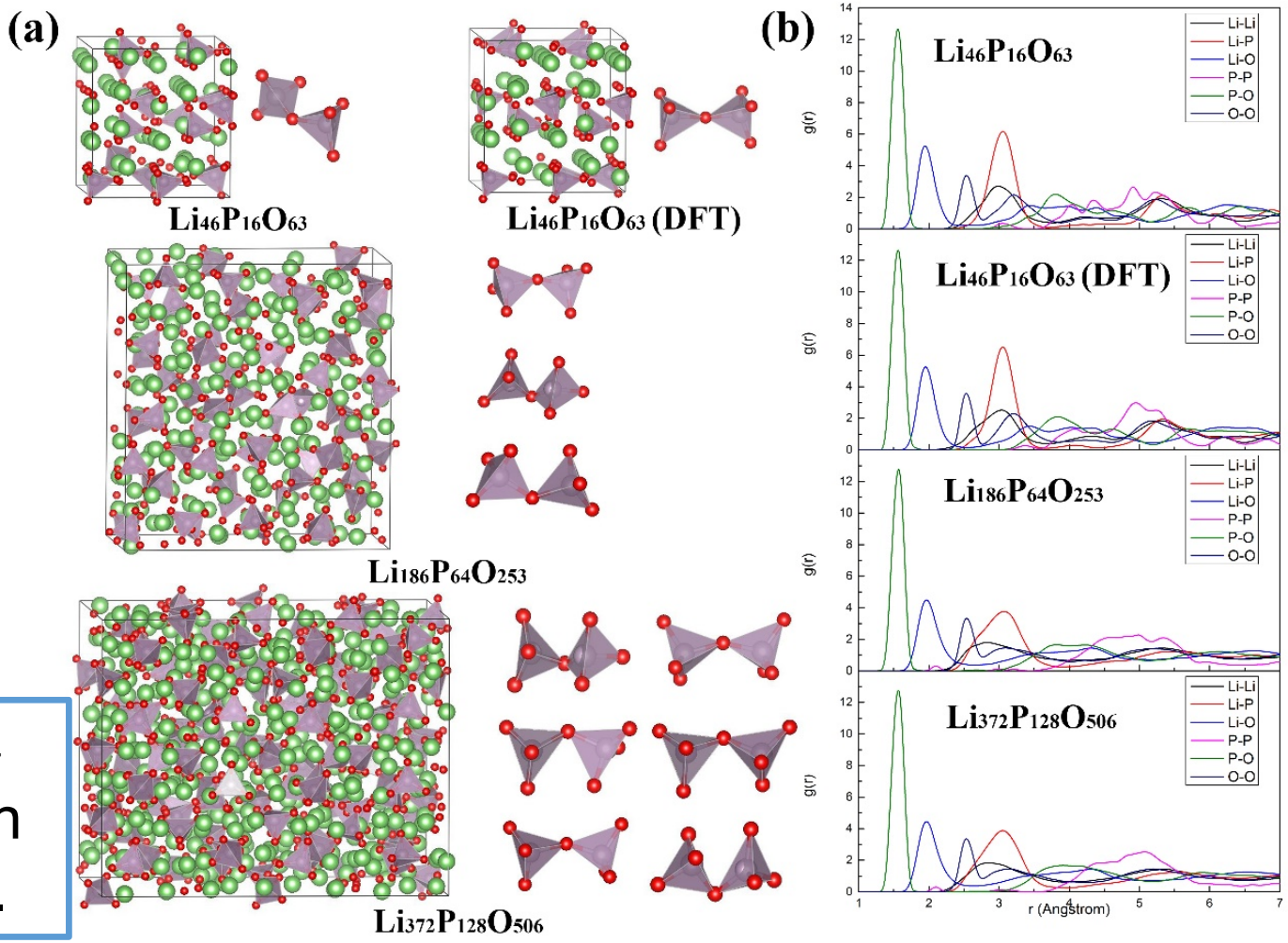
[W. Li et al., J. Chem. Phys. 147 (2017) 214106]

1) Partial condensation reaction

$2Li_3PO_4 \rightarrow Li_4P_2O_7 + Li_2O$

2) Off-stoichiometric composition

Li : P = 2.9

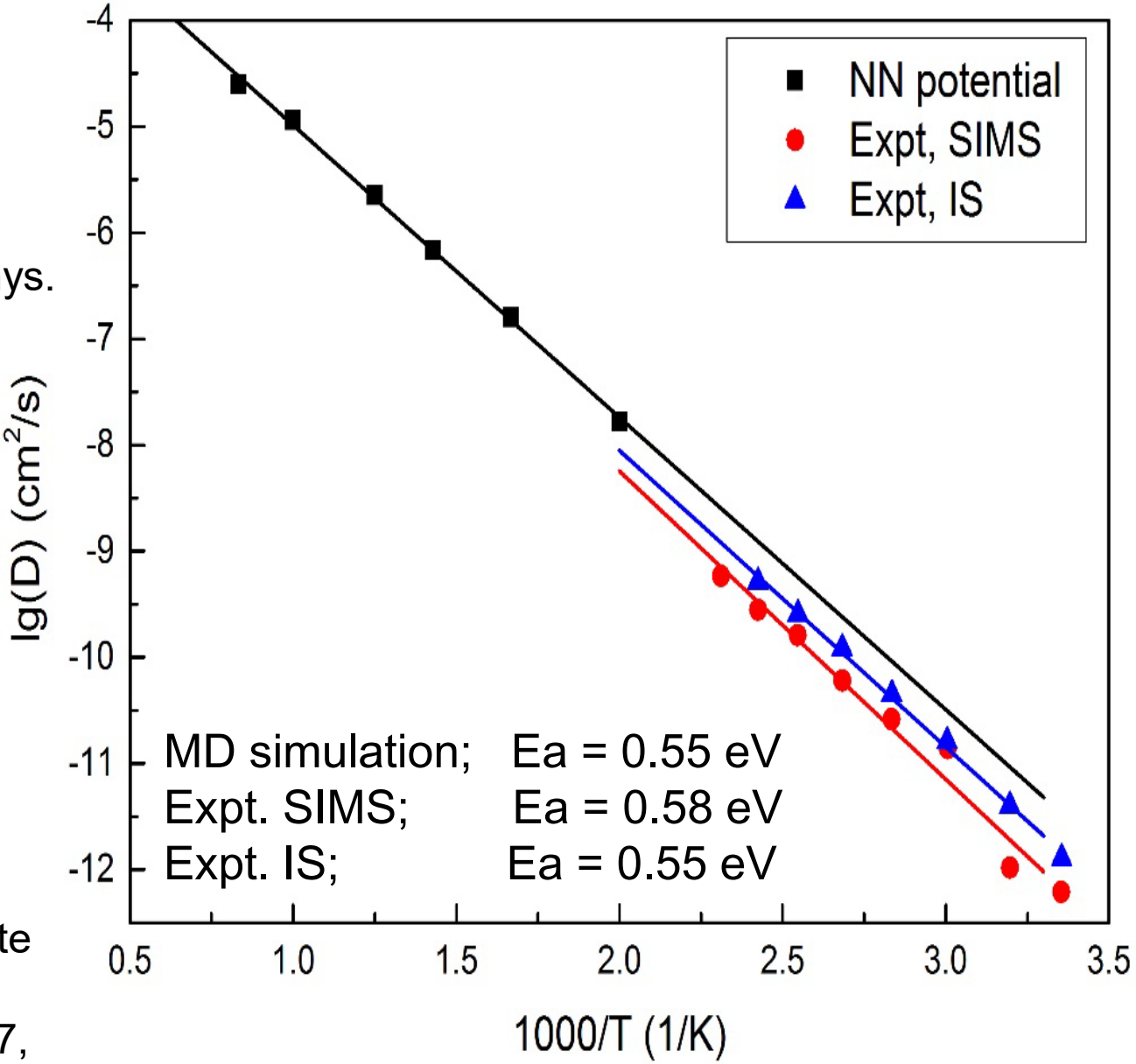


Existence of P_2O_7 dimmer agree with NMR observation.

Diffusion coefficients

Calculated with the largest model

[W. Li et al., J. Chem. Phys. 147 (2017) 214106]



Exp:
Kuwata, et. al, Solid State Ionics 294, 59 (2016);
J. Electrochem. Soc. 157, A521 (2010).

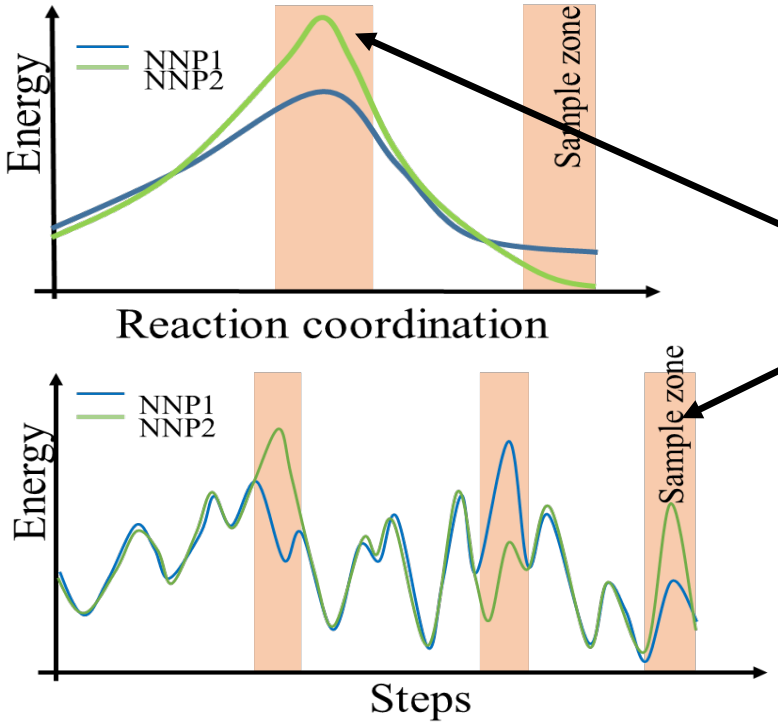
Example (3): Cu in Al_2O_3

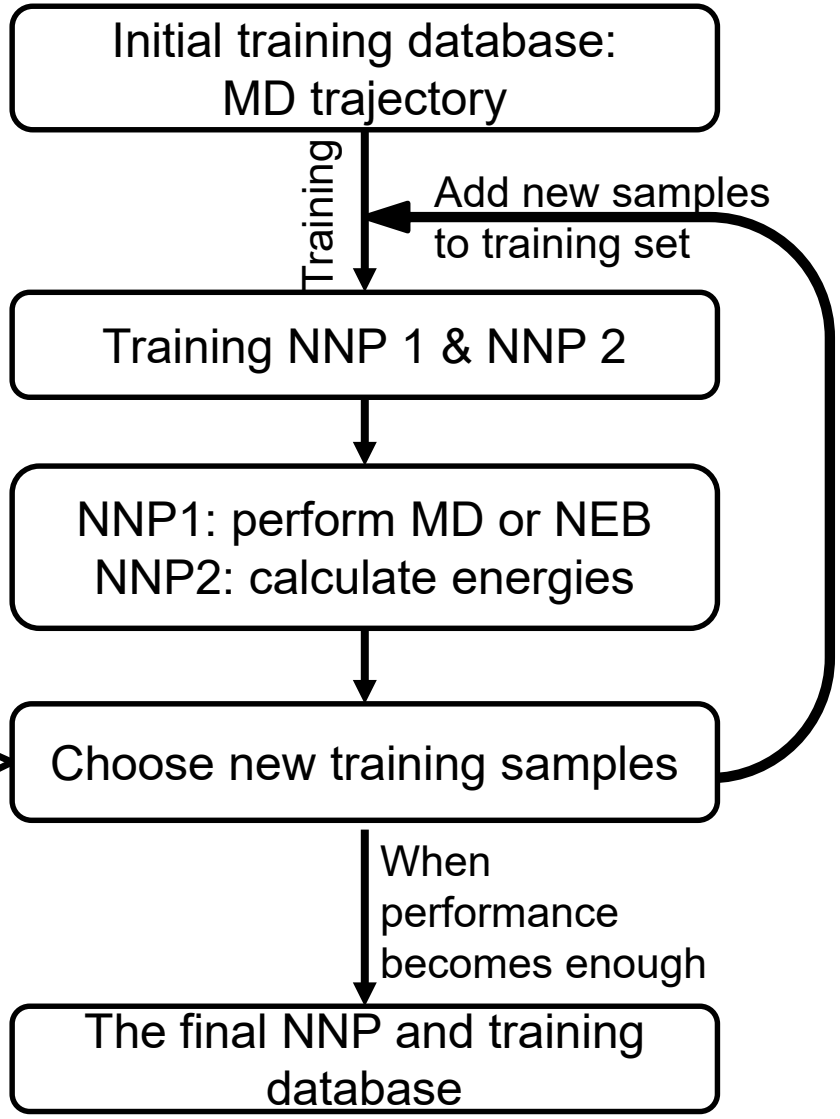
[W. Li, Doctoral thesis]

- NN potential applicable to composition/density variation

Total number: 41495

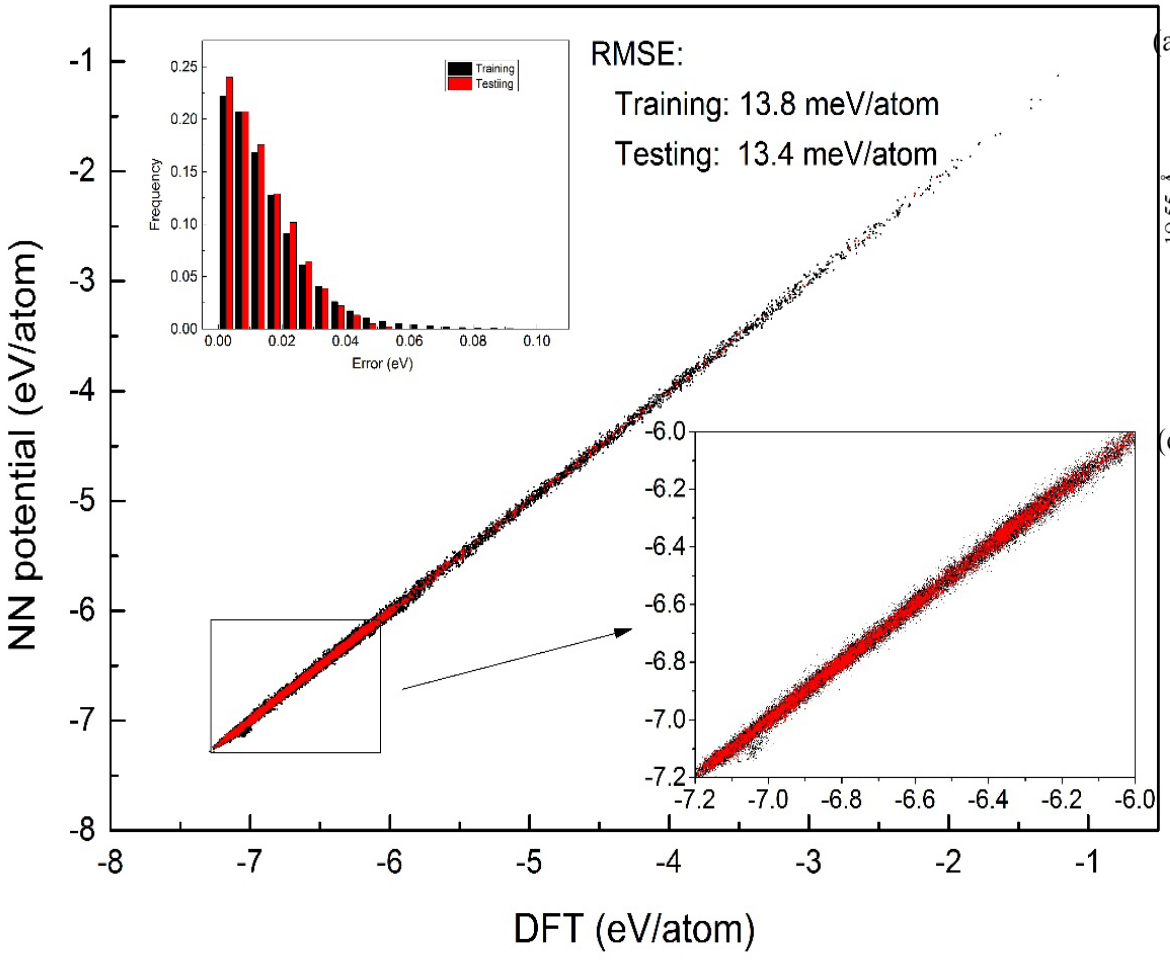
- Number of atoms: < 161 atoms
- Density: 2.9 ~ 3.2 g/cm^3
- Al/O ratio: 1 ~ 1.5



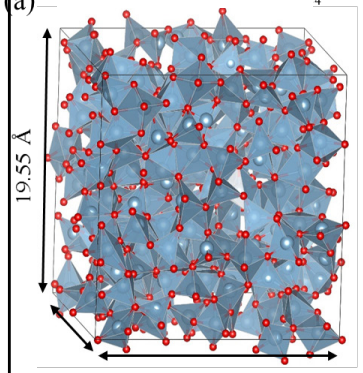


Energy prediction

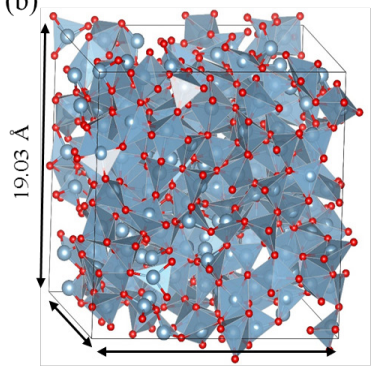
[W. Li, Doctoral thesis]

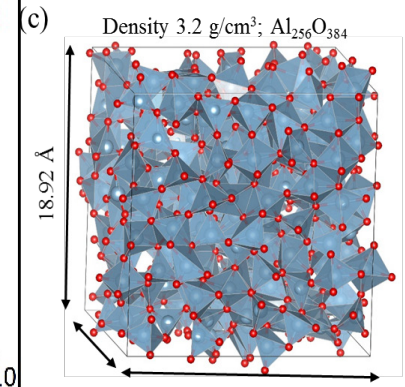


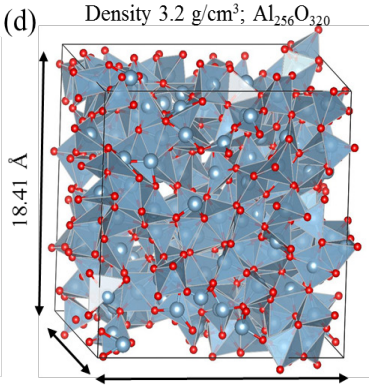
2.9g/cm^3, $Al_{256}O_{384}$



2.9g/cm^3, $Al_{256}O_{320}$







3.2g/cm^3, $Al_{256}O_{384}$

3.2g/cm^3, $Al_{256}O_{320}$

Diffusion coefficients & activation energies: dependence on composition & density

Stoichiometric case

Density =2.9 g/cm^3

Summary

Interatomic potentials constructed using neural network and first-principles simulation data to examine atomic diffusion behaviors in amorphous materials

- Cu in Ta_2O_5: simplified method (considering only Cu explicitly)
- Li in Li_3PO_4: transferability to much larger models than those used in training
- Cu in Al_2O_3: considering composition and density variation

Having both reliability and computational efficiency: promising!

Future plans

- ✓ Improvements (under electric fields, data sampling method, large-scale MD)
- ✓ Defects (in GaN etc.) and interfaces (Au/Li_3PO_4 etc.)
- ✓ Phonon & thermal transport
- ✓ Ferroelectric materials (long-range Coulomb interaction)