Introduction to MD Simulations using Moment Tensor Potential

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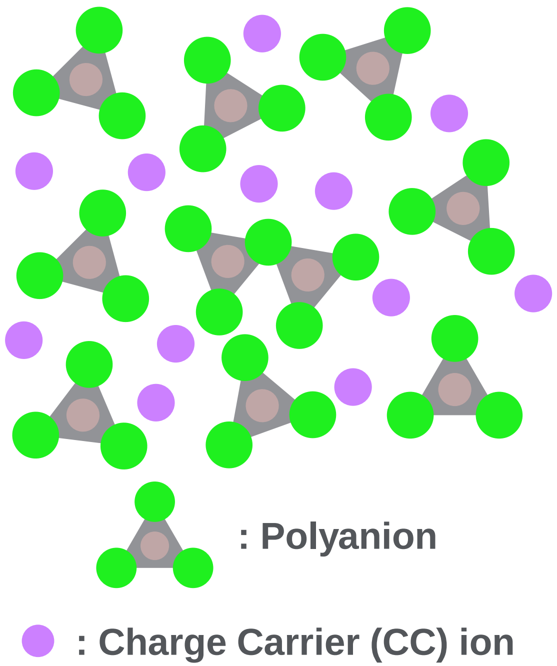
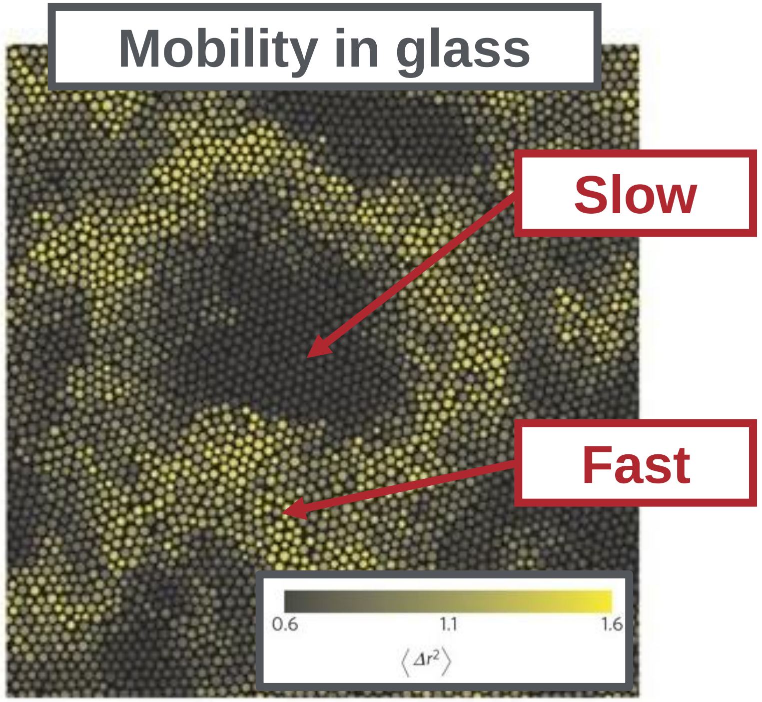
Part I

A brief illustration of our work

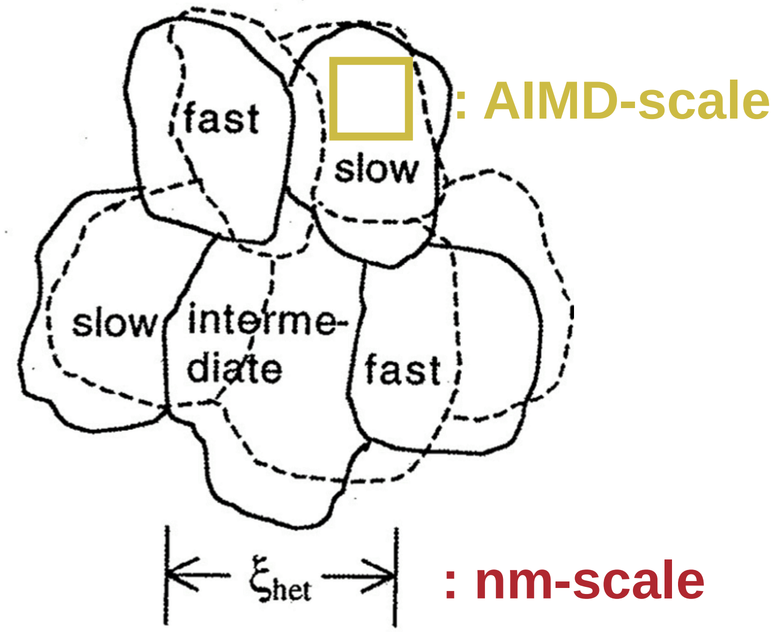
Dynamically heterogeneous glass dynamics

Glass : Low Crystallinity





Nat. Mat. 9, 324-331 (2010)



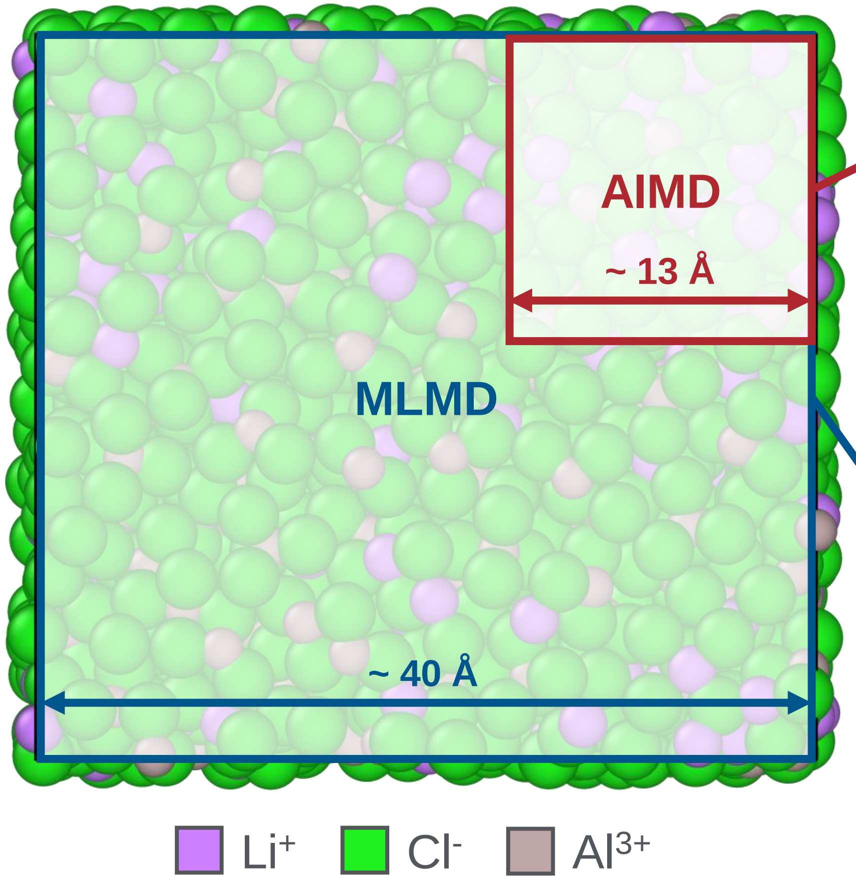
Annu. Rev. Phys. Chem. 51, 99-128 (2000)

✓ IGSSEs consist of polyanions ($AlCl_4^-$) and charge carrier ions (Li^+)

✓ Investigations on dynamic heterogeneity requires large-scale simulations up to nm-scale

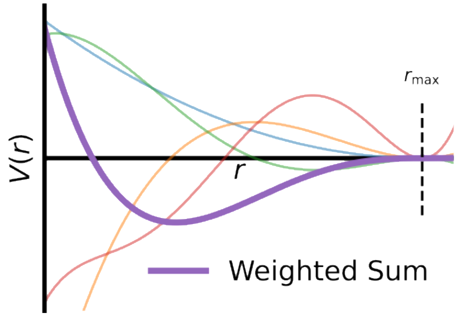
MLP to explore beyond AIMD

Lithium tetracholoroaluminate ($LiAlCl_4$)



- AIMD (PAW_PBE) + DFT (optB88-vdW)
- **96 ions** of $LiAlCl_4$, **1 ps**

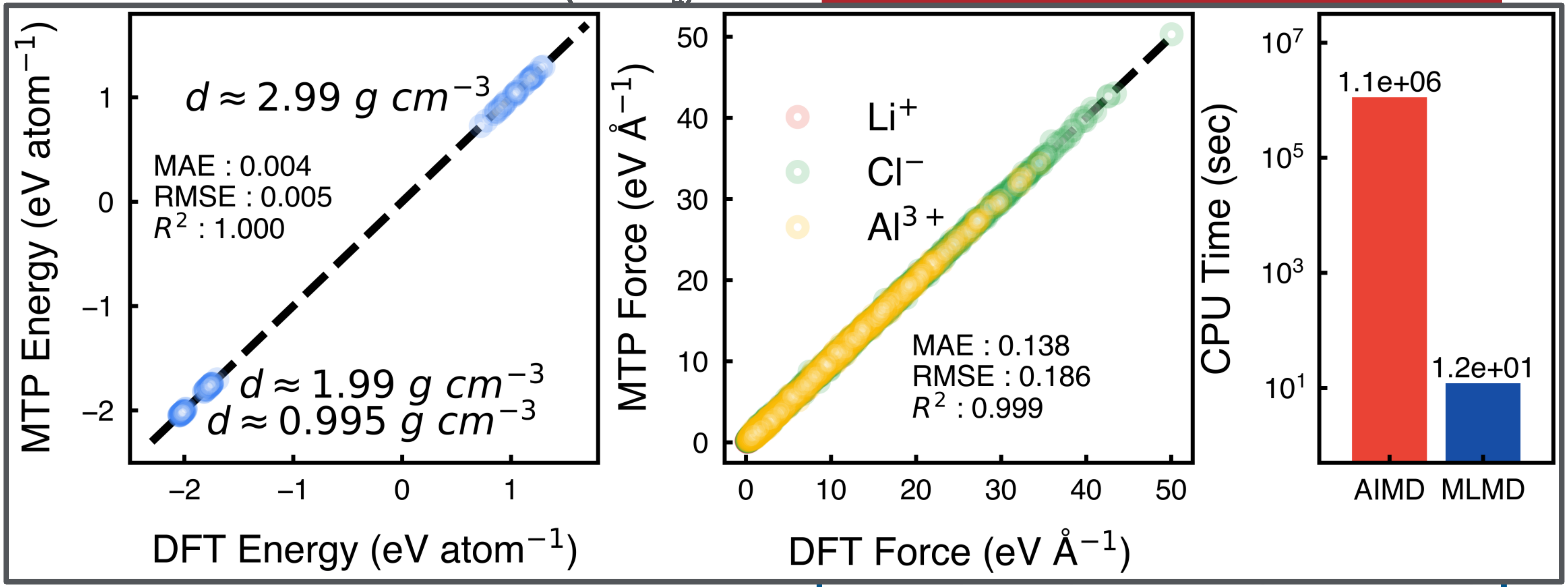
✓ **Moment Tensor Potential** (MTP)



- **MLMD** : Machine-Learning MD
- **2,592 ions** of $LiAlCl_4$, up to **10 ns**
- **Melt** (1000 K) – **Quench** (4.5×10^{11} K s^{-1})

MLP to explore beyond AIMD

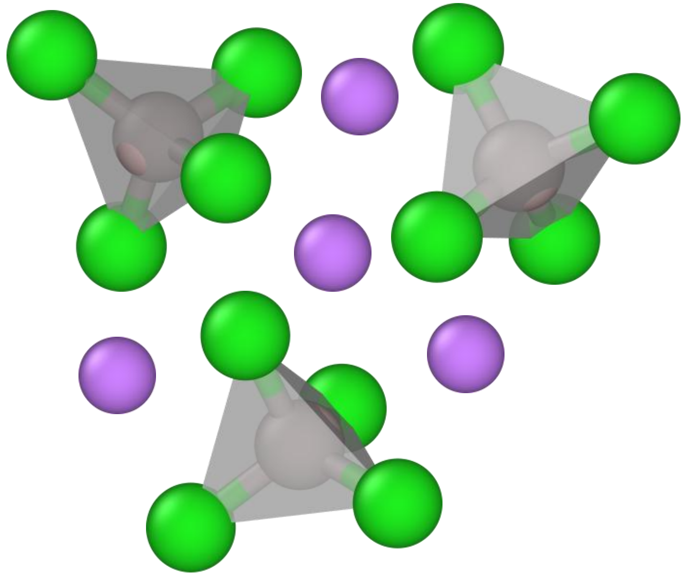
Lithium tetracholoroaluminate ($LiAlCl_4$)



Li^+ Cl^- Al^{3+}

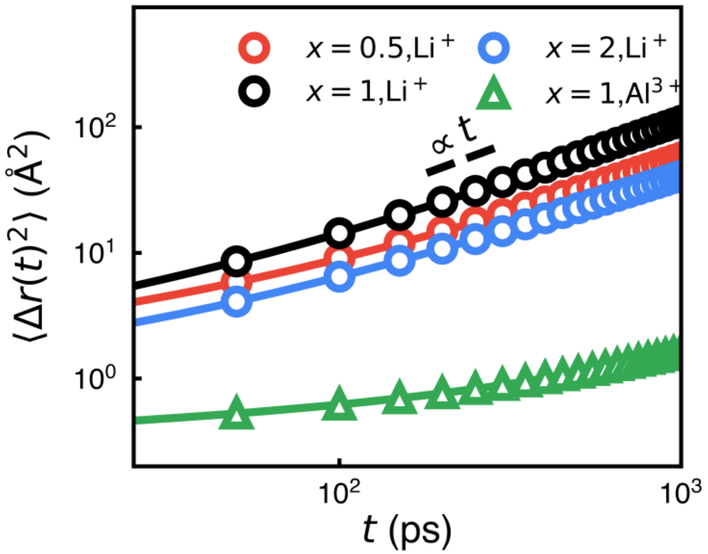
Li^+ exhibits heterogeneous dynamics

Li^+ between $AlCl_4^-$ tetrahedra



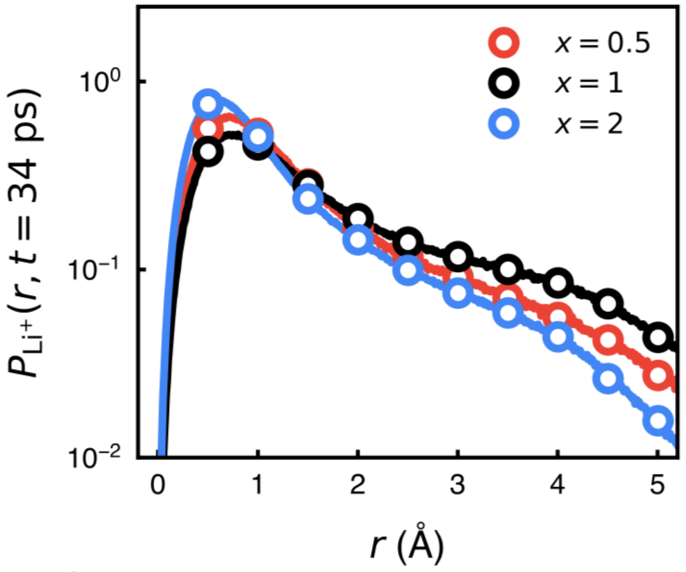
Mean Squared Displacement

$$\langle \Delta r(t)^2 \rangle = \left\langle \left(r(t) - r(0) \right)^2 \right\rangle$$



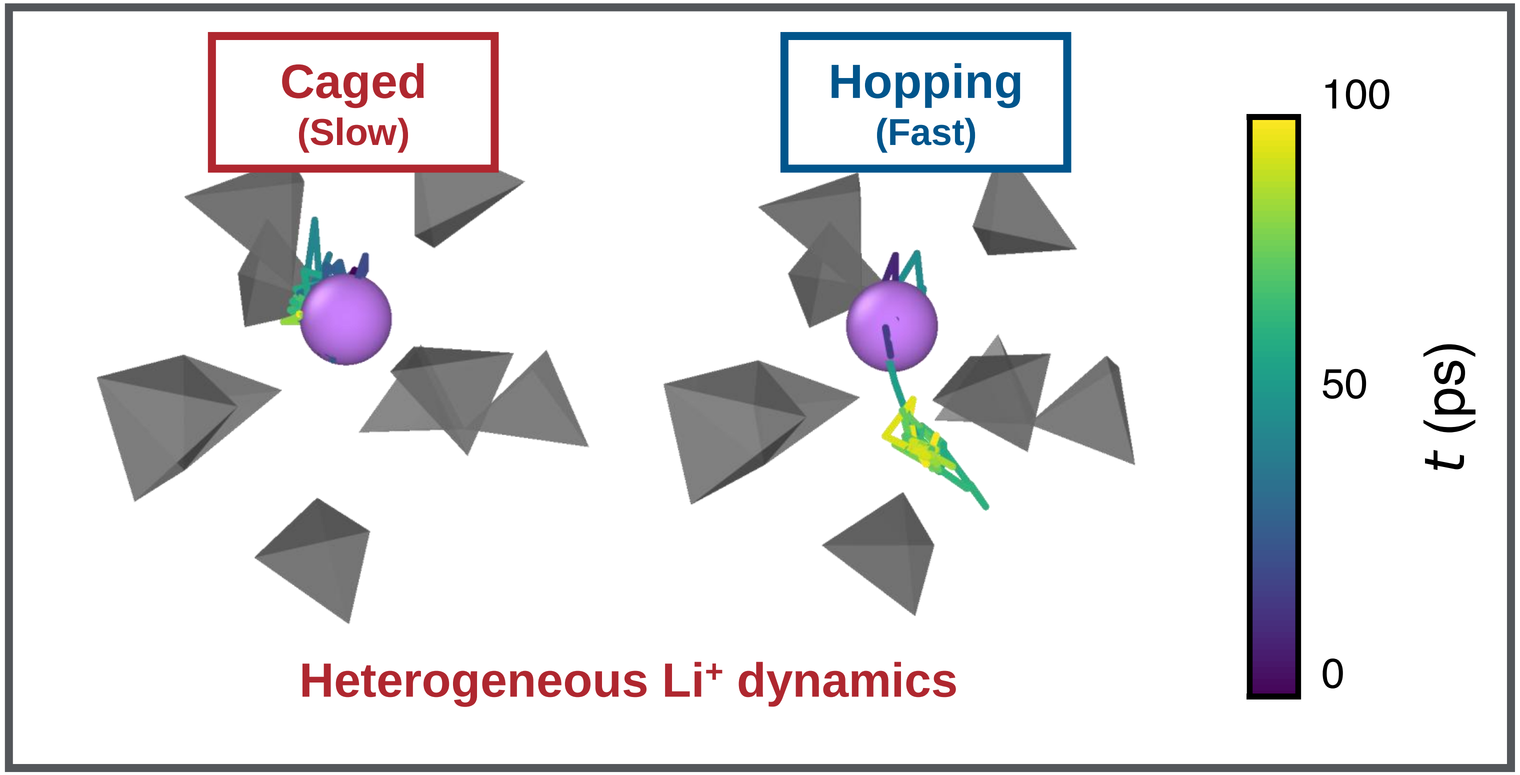
Diffusional van Hove correlation of Li^+

$$P(r,t) = 4\pi r^2 G_s(r,t)$$



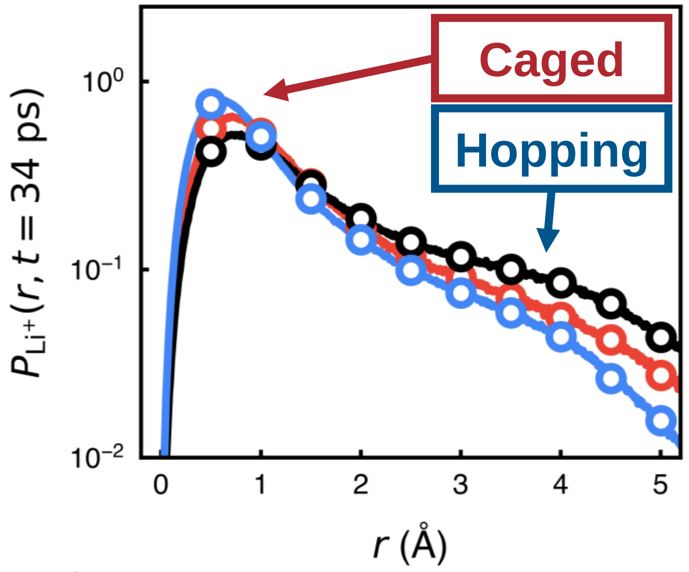
- ✓ Li^+ diffuses between immobile $AlCl_4^-$ tetrahedra, acting as a CC
- ✓ Van Hove correlation, $G_s(r,t)$, represents a distribution of displacement r during time t
- ✓ Li^+ dynamics can be categorized into heterogeneous caged and hopping motions

Li^+ exhibits heterogeneous dynamics



Diffusional van Hove correlation of Li^+

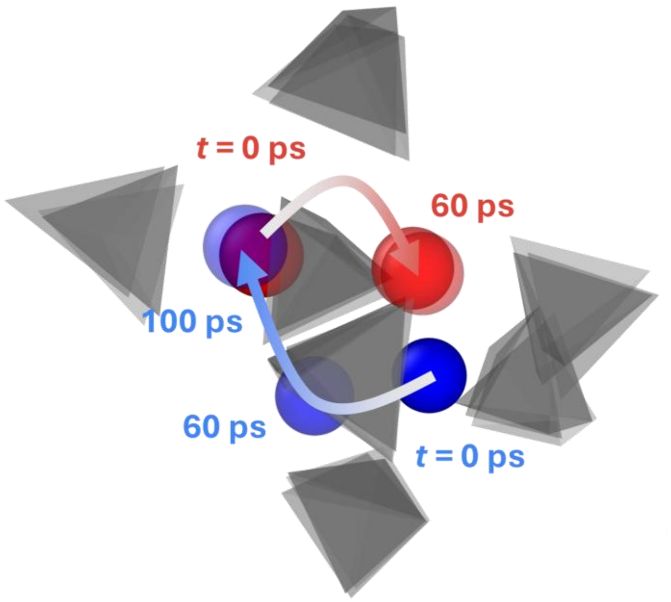
$P(r,t) = 4\pi r^2 G_s(r,t)$

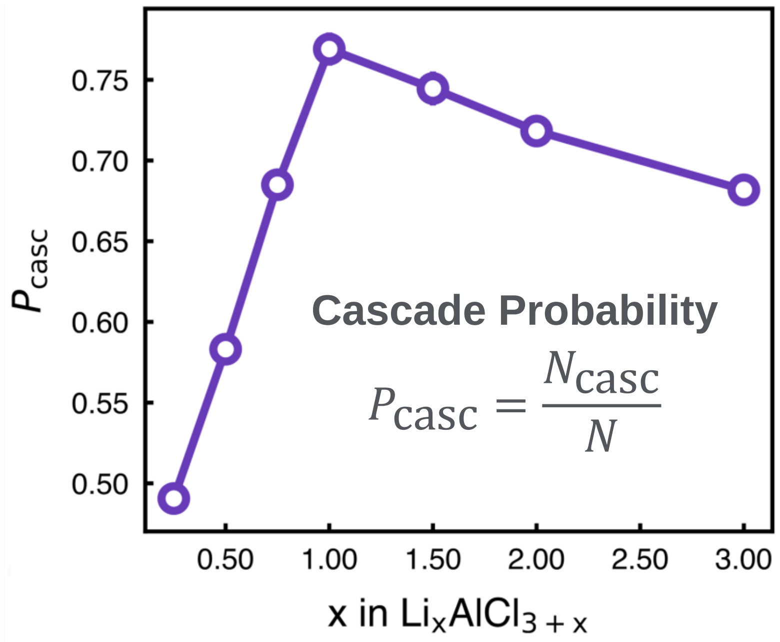


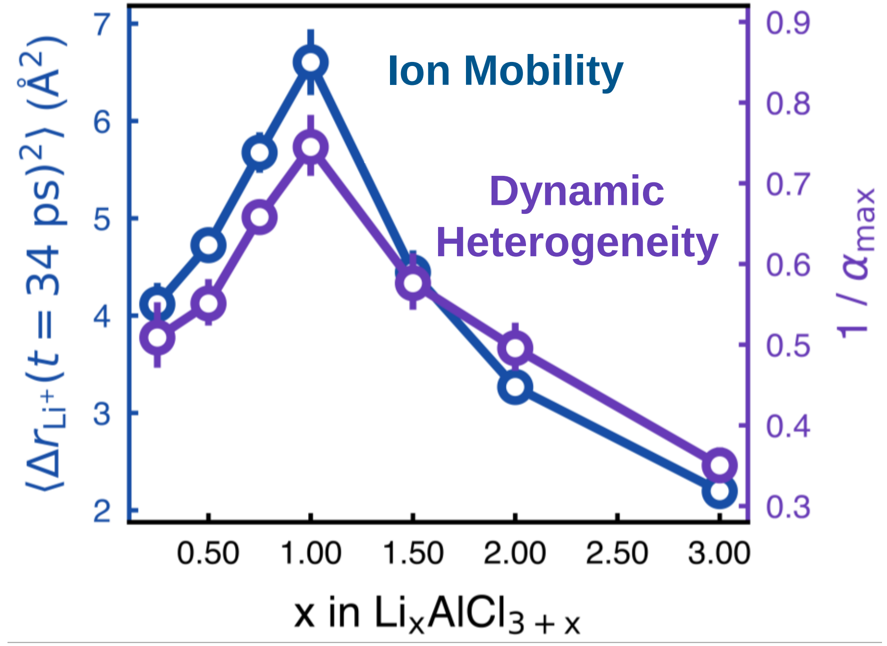
- ✓ Li^+ diffuses between immobile $AlCl_4^-$ tetrahedra, acting as a CC
- ✓ Van Hove correlation, $G_s(r,t)$, represents a distribution of displacement *r* during time *t*
- ✓ Li^+ dynamics can be categorized into heterogeneous caged and hopping motions

Dynamic heterogeneity affects ion mobility

The *cascade* hopping event







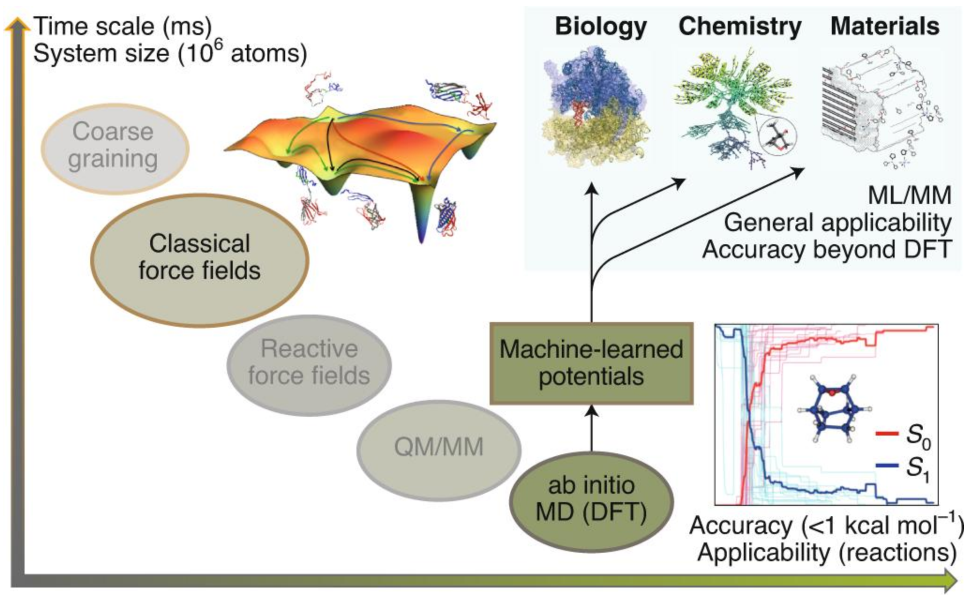
✓ Cascade hopping of Li^+ can be identified

✓ The non-monotonic behavior of P_{casc} on [CC] follows the trend of ion mobility and heterogeneity

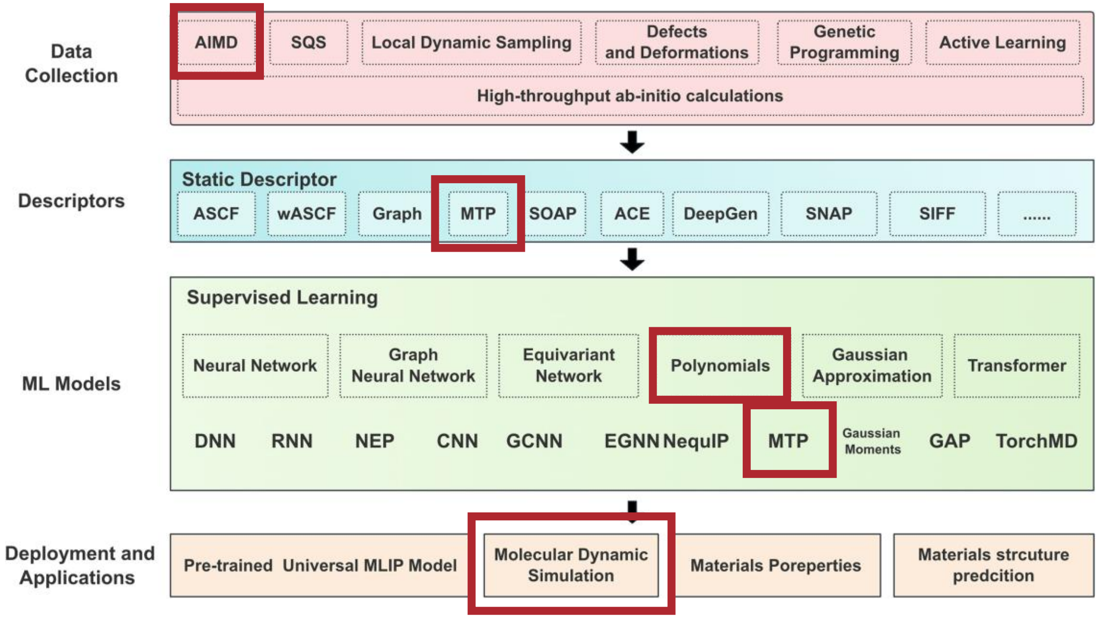
Part II

About Moment Tensor Potentials

Machine Learning Potentials (MLPs)



Nat. Mat. 20, 750-761 (2021)

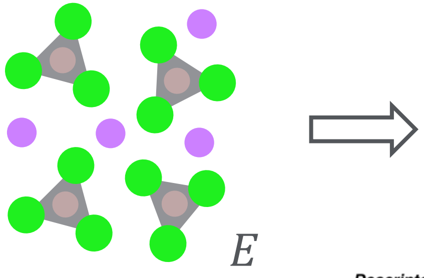


iScience 27 (5), 109673 (2024)

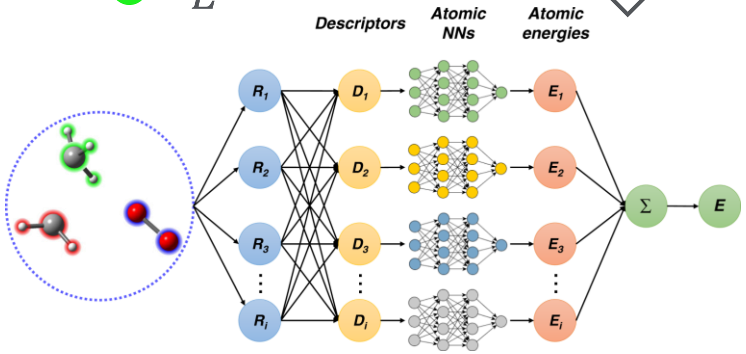
- ✓ Machine learning potential, trained from DFT results, can acquire high accuracy with large size
- ✓ Several types of MLP exists, sharing similar workflows (data-descriptor-model-application)

What's special about MTP?

DeePMD-Kit (Descriptor-based)

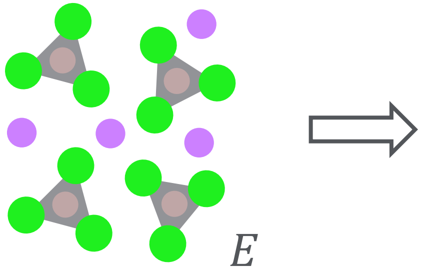


$$\boldsymbol{R}^i = \{\boldsymbol{r}_{1i}^T, \dots, \boldsymbol{r}_{ji}^T, \dots, \boldsymbol{r}_{N_i i}^T\}$$
$$\boldsymbol{r}_{ij} = (x_{ji}, y_{ji}, z_{ji})$$



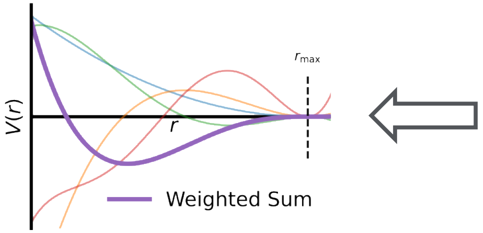
Nat. Comm. 11, 5713 (2020)

Moment Tensor Potential (MTP)



Pair	Types	r (Å)
1-2	0-1	5.388
1-3	0-2	3.997
	…	

$$f_\mu(|r_{ij}|, z_i, z_j) = \sum_{\beta=1}^{N_Q} c_{\mu, z_i, z_j}^{(\beta)} Q^{(\beta)}(|r_{ij}|)$$



- ✓ Descriptor-based models calculates the energy of the local environment through neural network
- ✓ MTP describes interatomic potential through linear combination of polynomials

What's special about MTP?

DeePMD-Kit (Descriptor-based)

- Complex & Heavy
- Slow (GPU required)
- Unfixed functional form (model learns itself)

Moment Tensor Potential (MTP)

- Simple & Lightweight
- Fast (CPU available)
- Fixed functional form (limited to polynomials)

✓ Descriptor-based models calculates the energy of the local environment through neural network

✓ MTP describes interatomic potential through linear combination of polynomials

Mathematical Details (1)

- ✓ The energy is the sum of contributions of atomic neighborhoods

$$E^{\mathrm{MTP}} = \sum_{i=1}^{N} V^{\mathrm{MTP}}(\boldsymbol{n}_i), \qquad \boldsymbol{n}_i = (\{r_{i1}, z_i, z_1\}, \dots, \{r_{iN_{\mathrm{nbh}}}, z_i, z_{N_{\mathrm{nbh}}}\})$$

- ✓ Each contribution can be expanded through a set of basis functions

$$V^{\mathrm{MTP}}(\boldsymbol{n}_i) = \sum_{\alpha=1}^{N_{\mathrm{lin}}} \xi_\alpha B_\alpha(\boldsymbol{n}_i)$$

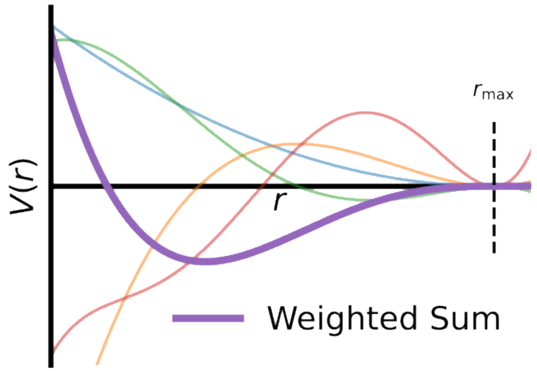
- ✓ MTP basis functions can be defined from *moment tensor descriptors*

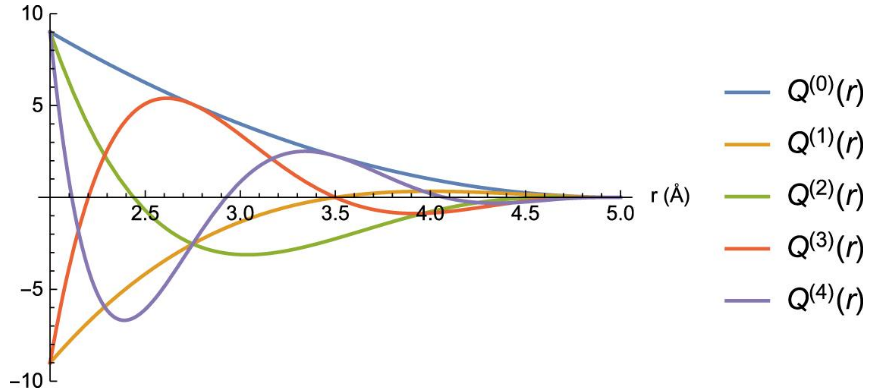
$$M_{\mu,\nu}(\boldsymbol{n}_i) = \sum_{j=1}^{N_{\mathrm{nbh}}} f_\mu(|r_{ij}|, z_i, z_j) r_{ij}^{\otimes \nu}$$

Mathematical Details (2)

- ✓ The radial part has the following form

$$f_\mu(|r_{ij}|, z_i, z_j) = \sum_{\beta=1}^{N_{\text{polyn}}} c^{(\beta)}_{\mu,z_i,z_j} T^{(\beta)}(|r_{ij}|)(R_{cut} - |r_{ij}|)^2$$





Mach. Learn.:Sci. Technol. 2, 025002 (2020)

- ✓ The MTP basis function, B_α, is a contraction of one or more moment tensor descriptors

e.g. $B_4 = M_{0,1} \cdot M_{0,1}, \quad B_8 = M_{0,0}(M_{0,1} \cdot M_{0,1})$

Loss function of MTP

✓ If we denote a MTP parameter set as $\boldsymbol{\theta} = (\xi_\alpha, c^{(\beta)}_{\mu,z_i,z_j})$,

✓ The loss function is defined as below

$$\sum_{k=1}^{K}\left[\omega_e\left(E_k^{\mathrm{MTP}}(\boldsymbol{\theta}) - E_k^{\mathrm{QM}}\right)^2 + \omega_f \sum_{i=1}^{N^{(k)}}\left|\boldsymbol{f}_{i,k}^{\mathrm{MTP}}(\boldsymbol{\theta}) - \boldsymbol{f}_{i,k}^{\mathrm{QM}}\right|^2 + \omega_s \sum_{a,b=1}^{3}\left(\sigma_{ab,k}^{\mathrm{MTP}}(\boldsymbol{\theta}) - \sigma_{ab,k}^{\mathrm{QM}}\right)^2\right] \to min.$$

✓ The loss function contains errors from energy (required), force, and stress (optional)

More details on MTP

PAPER • OPEN ACCESS

The MLIP package: moment tensor potentials with MPI and active learning

Ivan S Novikov, Konstantin Gubaev, Evgeny V Podryabinkin and Alexander V Shapeev

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https://iopscience.iop.org/article/10.1088/2632-2153/abc9fe

RESEARCH ARTICLE | AUGUST 28 2023

MLIP-3: Active learning on atomic environments with moment tensor potentials

Special Collection: Software for Atomistic Machine Learning

Evgeny Podryabinkin; Kamil Garifullin; Alexander Shapeev; Ivan Novikov

Check for updates

+ Author & Article Information

J. Chem. Phys. 159, 084112 (2023)

https://doi.org/10.1063/5.0155887 **Article history**

https://pubs.aip.org/aip/jcp/article/159/8/084112/2908187/MLIP-3-Active-learning-on-atomic-environments-with

Skoltech MLIP

Меню

Machine Learning Interatomic Potentials (MLIP for short) is a software package being developed by the group of Alexander Shapeev (Skoltech). The currently supported is the second version supporting multicomponent MTP, see the Download and Documentation sections.

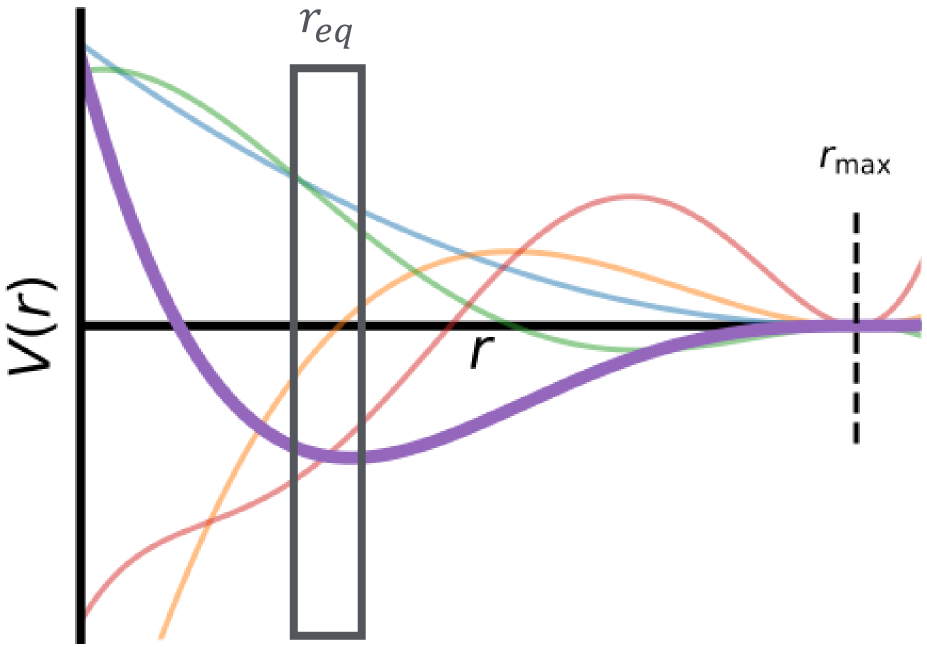
This project has started as a part of the Skoltech NGP Program No. 2016-7/NGP (a Skoltech-MIT joint project) and is now funded by the Russian Science Foundation project RSF 18-13-00479.

https://mlip.skoltech.ru/

Part III

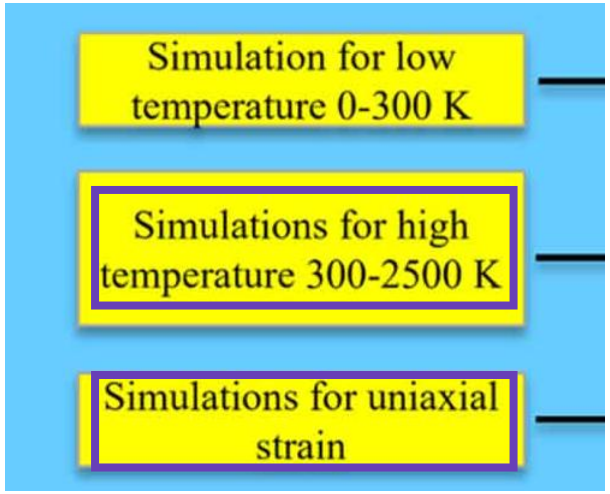
MTP Training Strategies

Strategy: Sampling various r

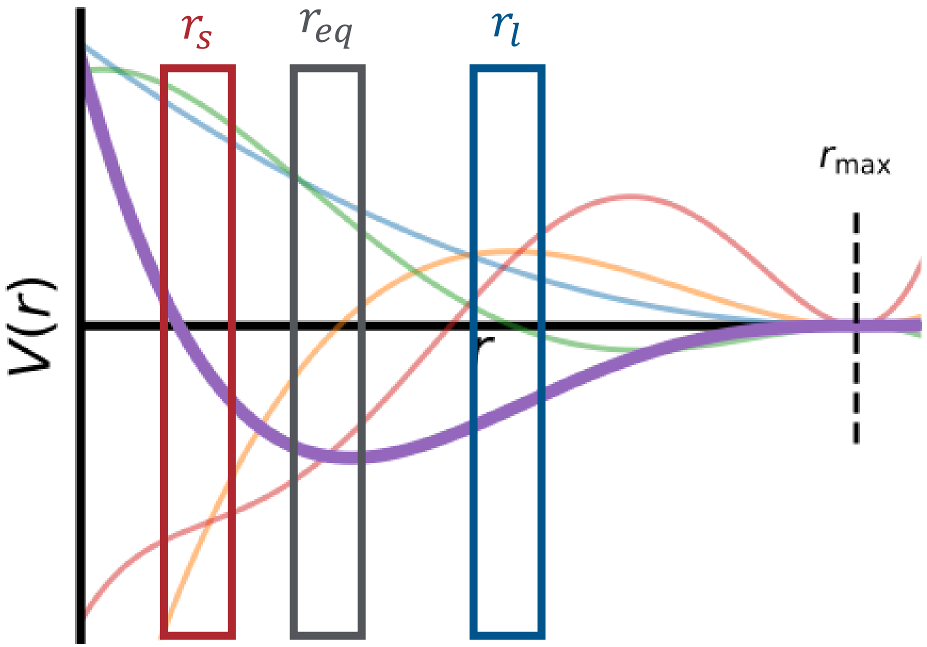


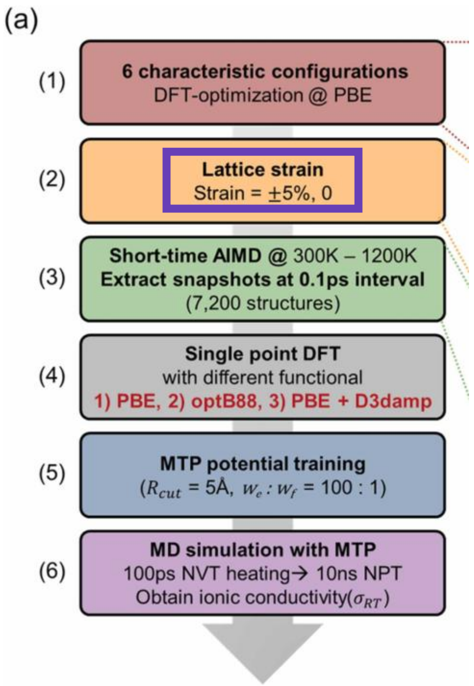
- ✓ A special tip: MTP trained with only r_{eq} fails molecular dynamics simulations
- ✓ A method for acquiring samples of various r is critical

Strategy: Sampling various r



J. Phys. Mater. 8, 015003 (2025)

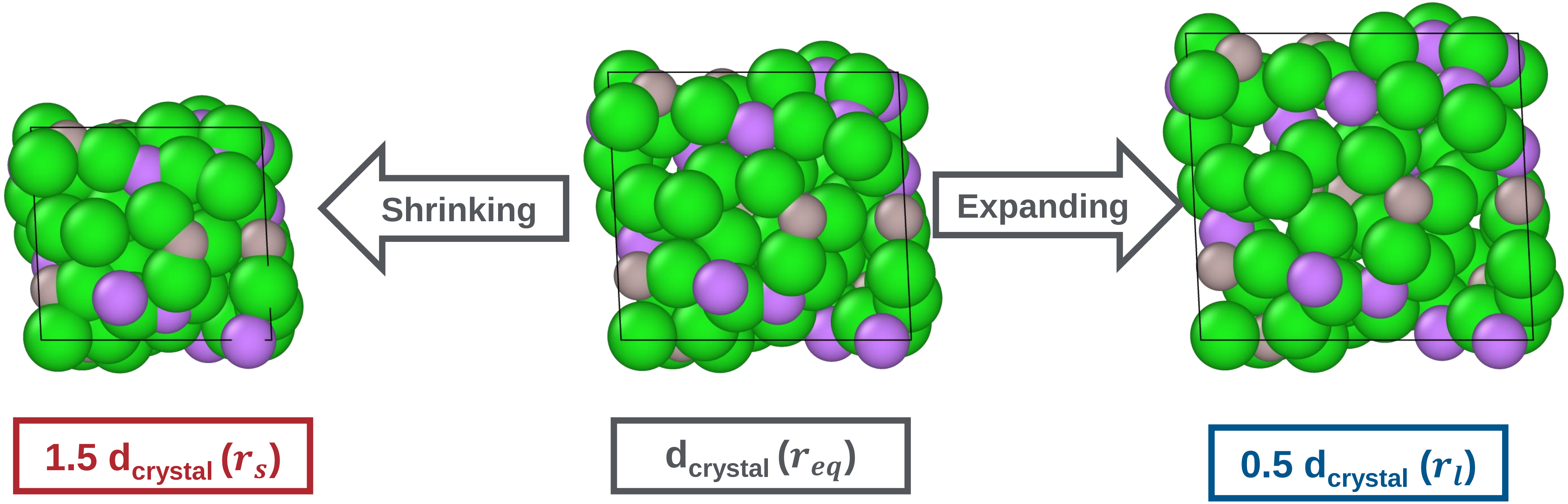




Nano Energy 1, 109436 (2024)

- ✓ Previous studies added strained configuration or configurations at high temperature
  - ✓ Additional dataset significantly increases the computational cost of DFT

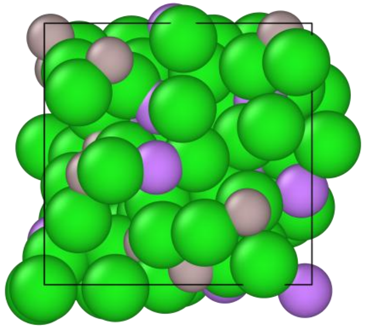
Strategy: Multi-Density Training Set



✓ Additional datasets can be made from manually shrinking / expanding melt configurations

Workflow of MTP training (1)

Initial Configuration
Random distribution of
16 Li^{+}, 64 Cl^{-}, 16 Al^{3+}

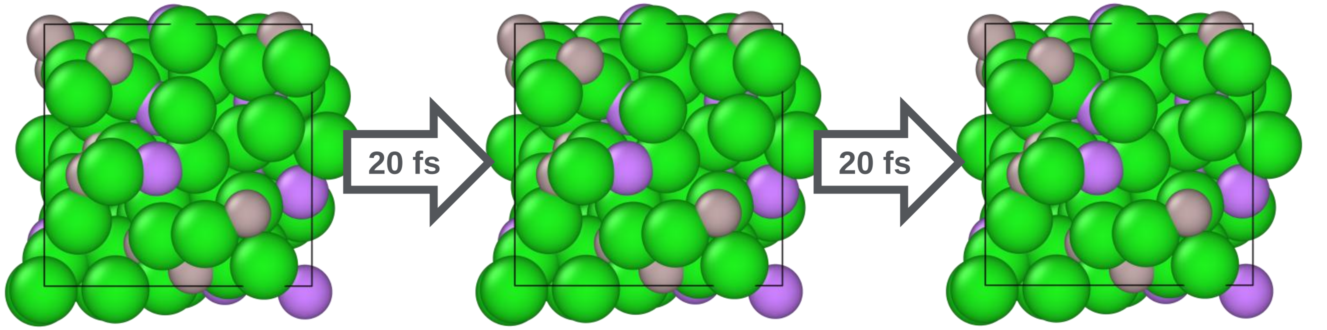


Ab initio MD
VASP – Vienna Ab initio
Simulation Package

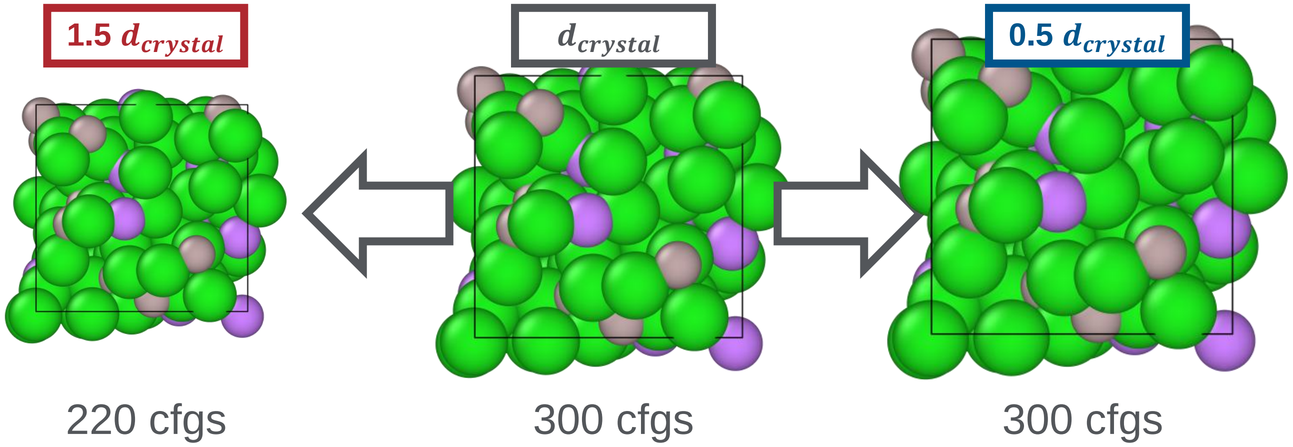
- Nosé-Hoover, 1000K
- 2 fs timestep
- Perdew-Burke-Ernzerhof (PBE)
- Projector Augmented Wave Potentials (PAW)
- Wave energy cutoff of 520 eV
- Γ-centered 1 x 1 x 1 *k*-point mesh

Workflow of MTP training (2)

Configuration Select
Sample configurations from a AIMD trajectory w/ 20 fs gap



Manual Modification
Expand / shrink equilibrium configuration to each density



Workflow of MTP training (2)

Single Point Calculation

- OptB88-vdW functional
- Projector Augmented Wave Potentials (PAW)
- Wave energy cutoff of 520 eV
- Γ-centered 1 x 1 x 1 *k*-point mesh

MTP Training

lev_{max} = 16
r_{cut} = 5 Å
ω_e = 1
ω_f = 0.01
ω_s = 0.001

1.5 $d_{crystal}$ | **$d_{crystal}$** | **0.5 $d_{crystal}$**

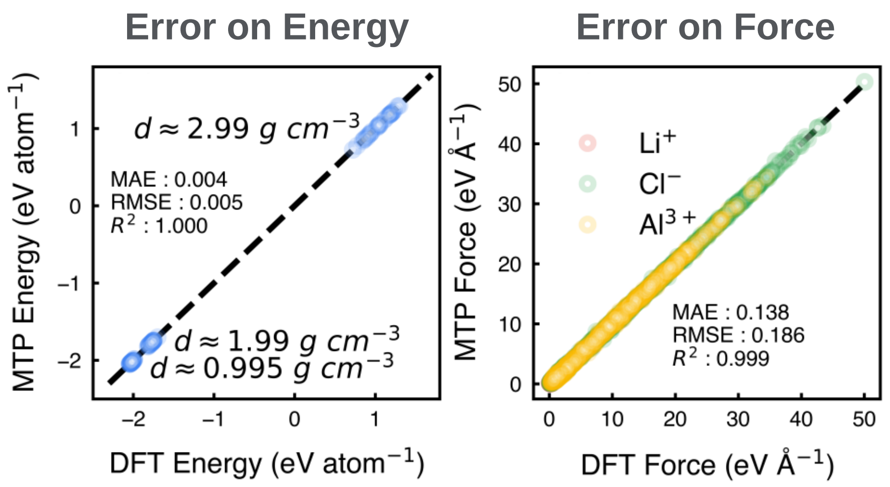
Total Dataset

Training | **Test**

Workflow of MTP training (3)

Error Analysis

MAE:
Mean Squared Error
RMSE:
Root Mean Squared Error
R^2:
Coefficient of Determination



MD Simulation

Part IV

The Hands-on Guidebook of MLIP Package

- ✓ https://github.com/SinsuSquid/MLIP_Guidebook