

Development of MLIPs for Interfaces in solid state batteries

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Tutorial EN03: Young Scientist Tutorial on Computational Methods for Explicit Interfacial Modeling in All-Solid-State-Batteries



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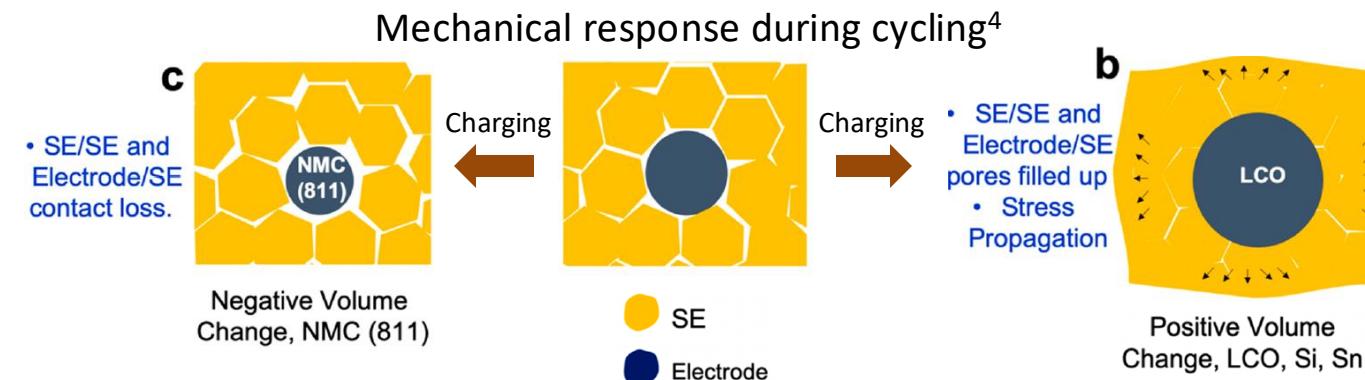
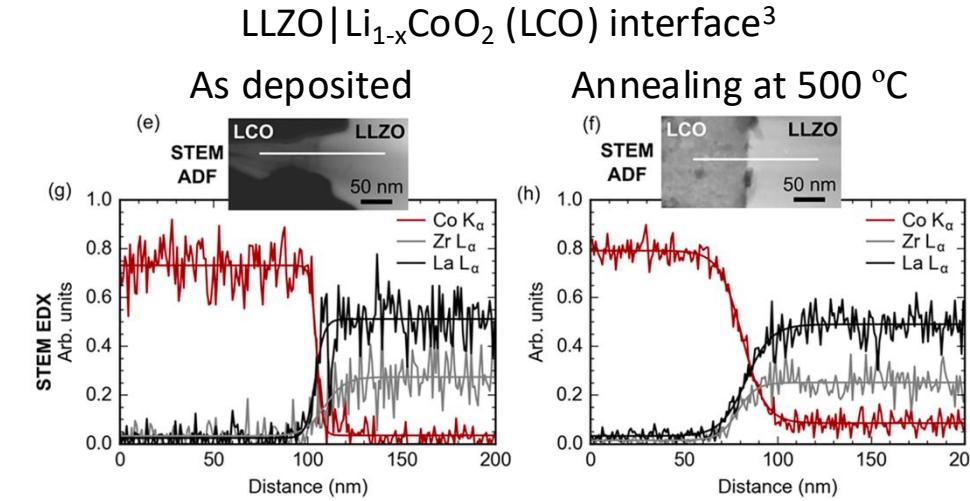
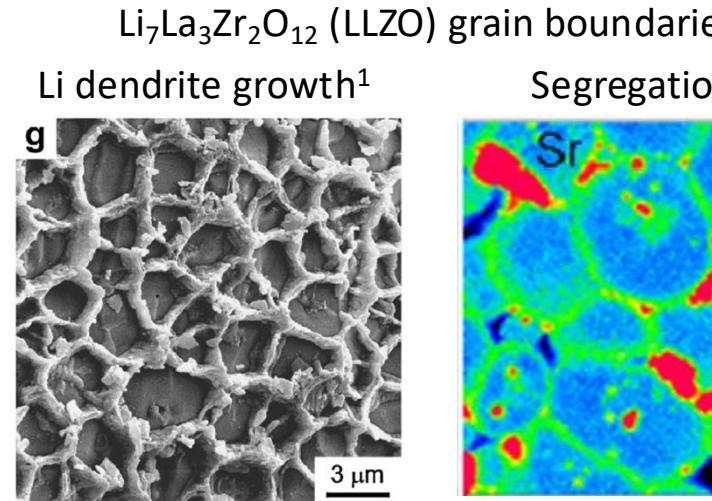
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- Demonstration: application to SSB research

Interface issues in solid-state batteries

- Electro-chemo-mechanical instability and interdiffusion of ions at interfaces degrade the performance of solid-state batteries



[1] E. J. Cheng, A. Sharafi, and J. Sakamoto, *Electrochimica Acta*. **2017**, *223*, 85–91.

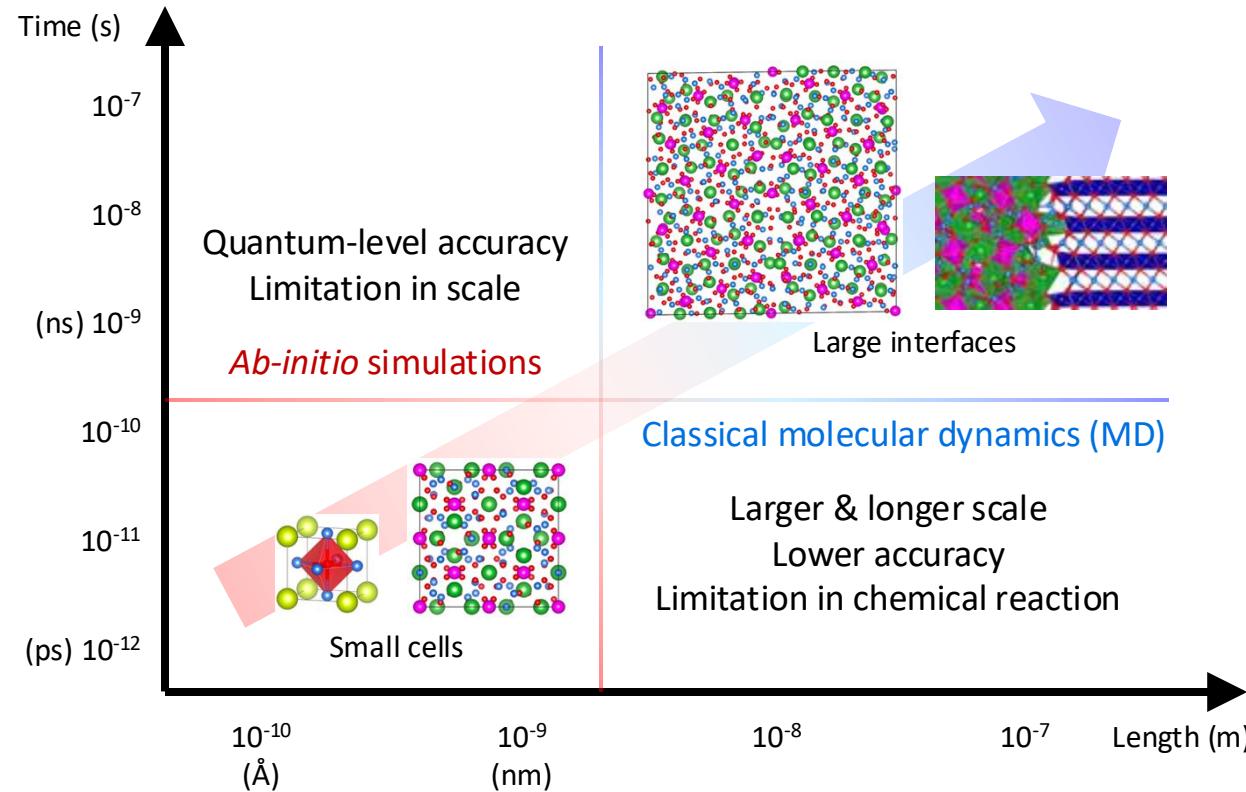
[3] G. Vardar et al., *Chem. Mater.* **2018**, *30*, 6259–6276.

[2] S. Ohta, Y. Kihira, and T. Asaoka, *Front. Energy Res.* **2016**, *4*, 30.

[4] A. Banerjee, X. Wang, C. Fang, E. A. Wu, and Y. S. Meng, *Chem. Rev.* **2020**, *120*, 6878–6933.

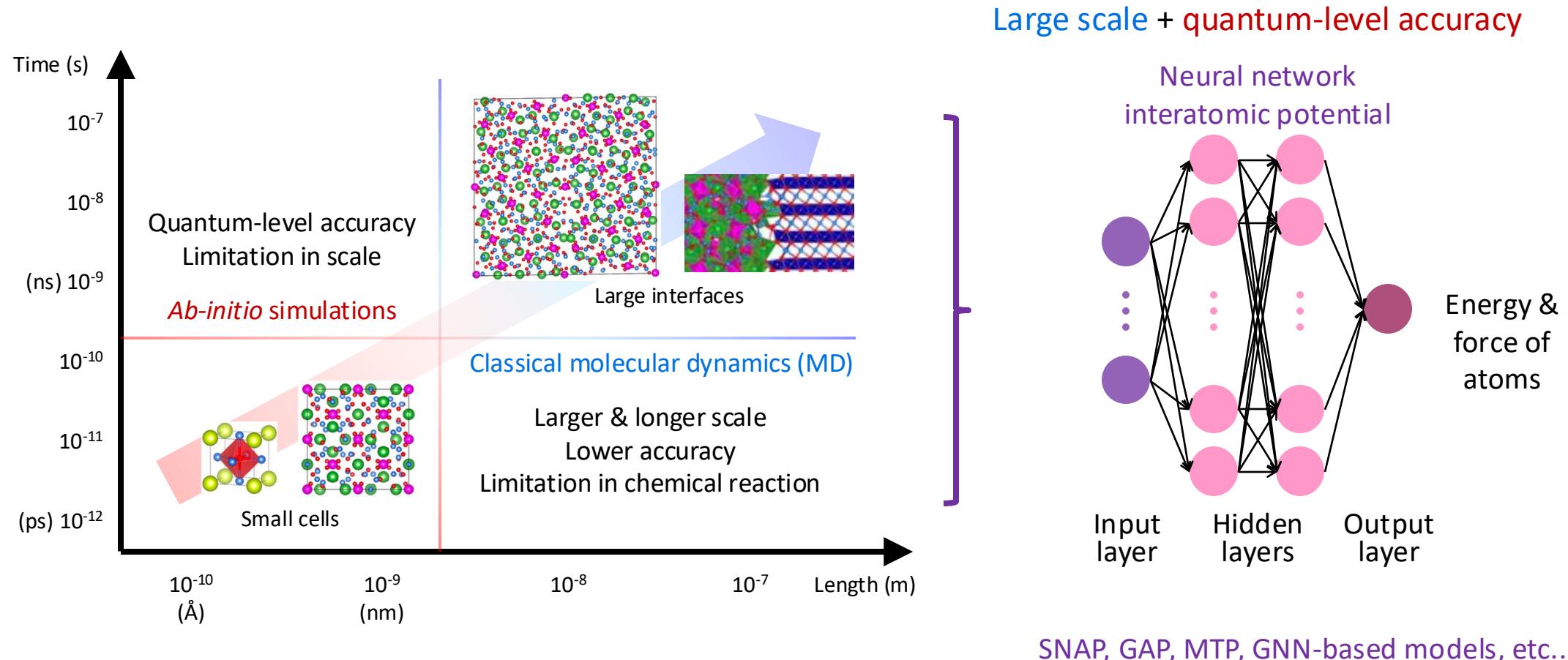
Atomistic modeling for interfaces

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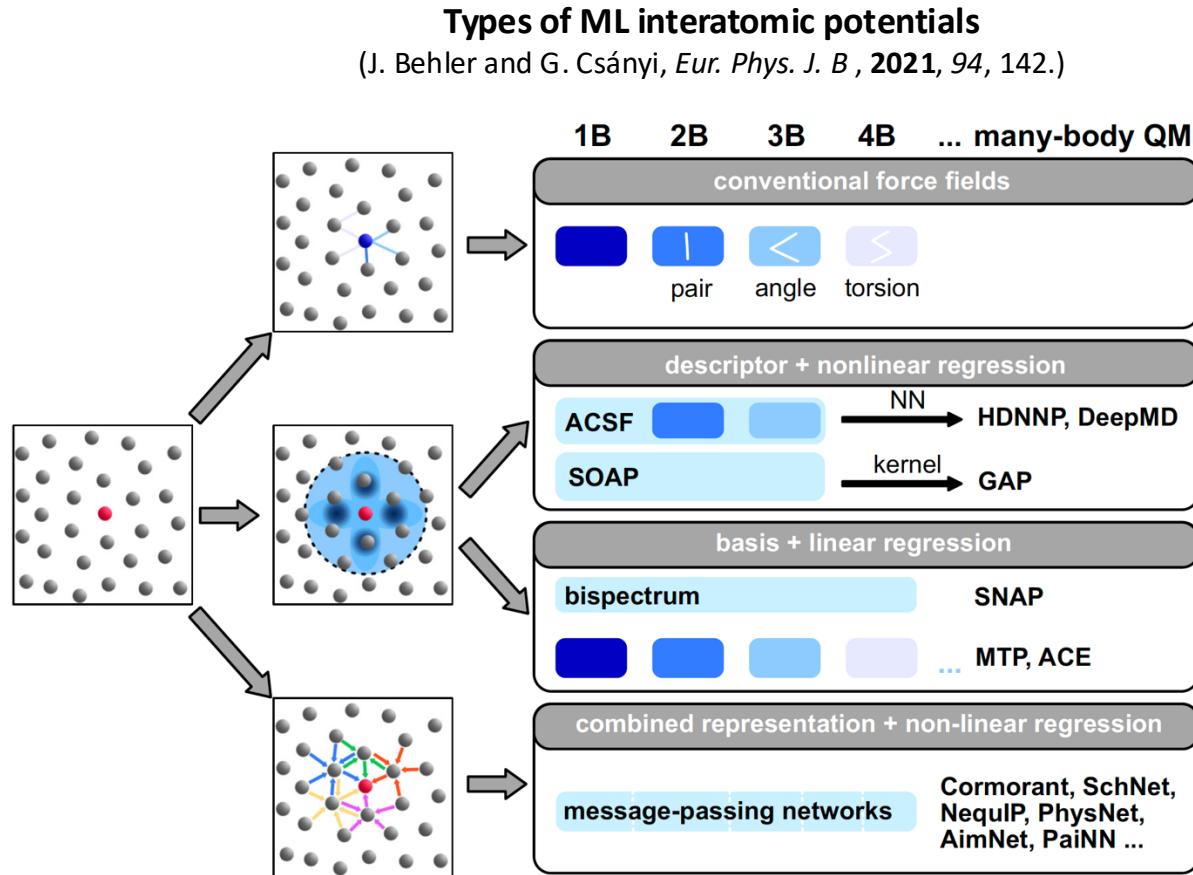
Atomistic modeling for interfaces

- Atomistic modeling can provide fundamental understanding of interfacial phenomena
- Machine-learning interatomic potential (MLIP; or ML force fields) to unbiasedly yet accurately survey the intrinsic properties of interfaces

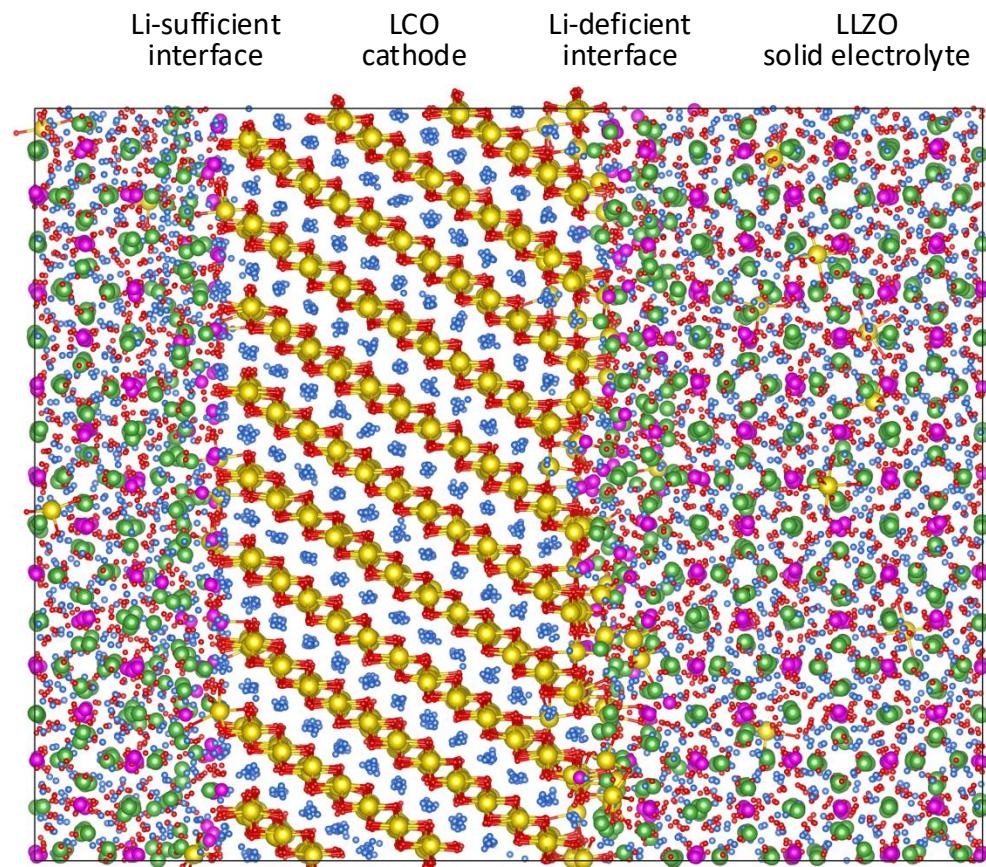


ML models for interatomic potential

- A number of MLIP models are available for interface simulations

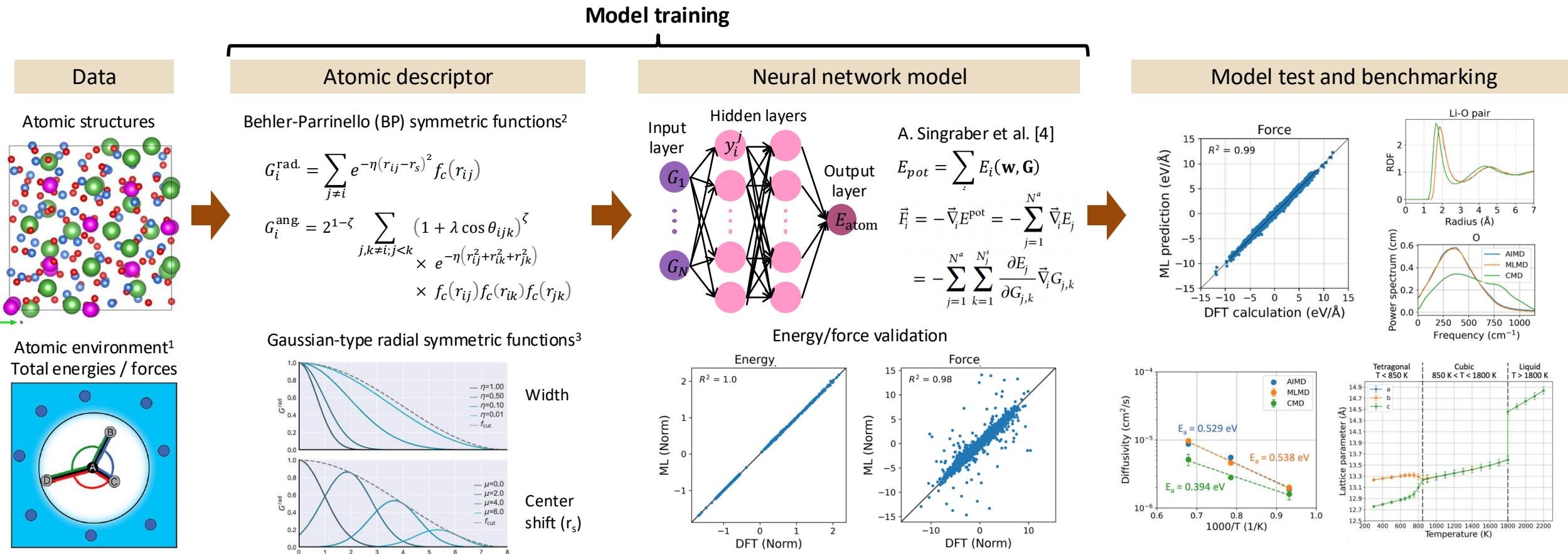


LLZO/LCO interface with 5 elements (n2p2 code)
(K. Kim et al., *Energy Stor. Mater.* **2024**, *73*, 103842.)



MLIP is not magic! Systematic development is essential

- Development process: **data generation, model training, and test**



[1] J. Behler, *Chem. Rev.* **2021**, *121*, 10037–10072.

[3] M. Gastegger et al., *J. Chem. Phys.* **2018**, *148*, 241709.

[2] J. Behler and M. Parrinello, *Phys. Rev. Lett.* **2007**, *98*, 146401.

[4] A. Singraber et al., *J. Chem. Theory Comput.* **2019**, *15*, 1827–1840.

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2023	Li-Si	NNP	a-Li _x Si	$\Delta E/F$ for amorphous & interface models	F. Fu, <i>AFM</i> 2023 , <i>33</i> , 2303936.	No interface but amorphous data
2024	$LiCoO_2/Li_7La_3Zr_2O_{12}$	NNP	Amorphous models	$\Delta E/F$ for interfaces; $\Delta E/F$, RDF, vDOS, diffusivity for amorphous models	K. Kim, <i>Energy Stor. Mater.</i> 2024 , <i>73</i> , 103842.	No interface but amorphous data Dynamic properties validated
2024	Li/Li_6PS_5Cl	MTP	Interfaces	None	G. Chaney, <i>ACS AMI</i> 2024 , <i>16</i> , 24624-24630.	
2024	$Li/\beta\text{-Li}_3PS_4$	NNP	Interfaces	$\Delta E/F$ for interfaces	F. Ren, <i>EES</i> 2024 , <i>17</i> , 2743-2752.	
2024	$Li/Li_7La_3Zr_2O_{12}/Li_xZrO_2$	NNP	a-Li _x La _y Zr _z O & LLZO surface/GB models	None for interfaces	Y. You, <i>npj Comput. Mater.</i> 2024 , <i>10</i> , 57.	
2025	Li/Li_6PS_5Cl	Allegro	interfaces	Unpublished	https://doi.org/10.48550/arXiv.2506.10944	Unpublished

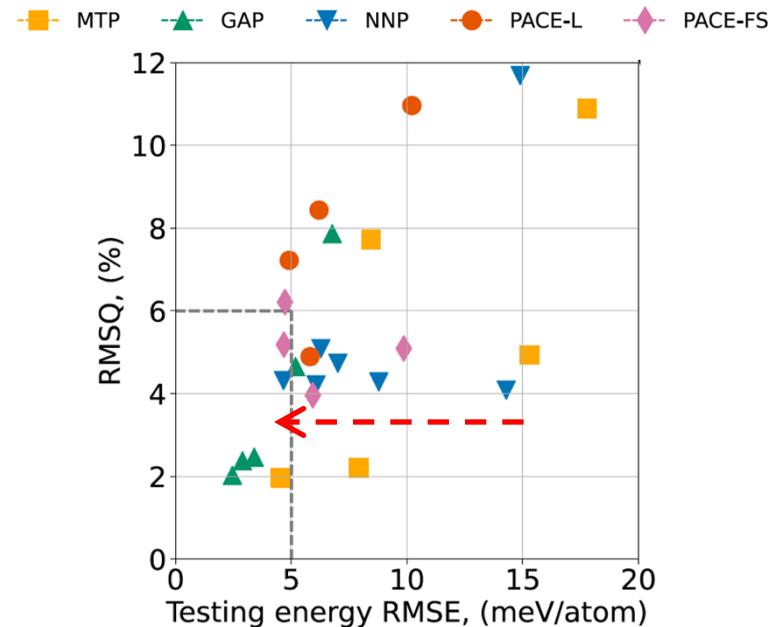
The table may not be complete—please let me know if I missed any papers!

Current status of MLIP application to SSB interfaces

- **Model test:** thorough benchmarking test will be needed
 - Low $\Delta E/F$ errors do not guarantee a good performance for predicting properties accurately
 - Benchmarking tests for properties against AIMD will be necessary

Performance tests for various MLFFs for Fe GBs

(L. Zhang et al., *Acta Materialia* 2024, 270, 119788.)



Quality factor

$$Q = \sqrt{\frac{1}{N_p} \sum_{i=1}^{N_p} \left(\frac{f_i^{ML} - f_i^{DFT}}{f_i^{DFT}} \right)^2},$$

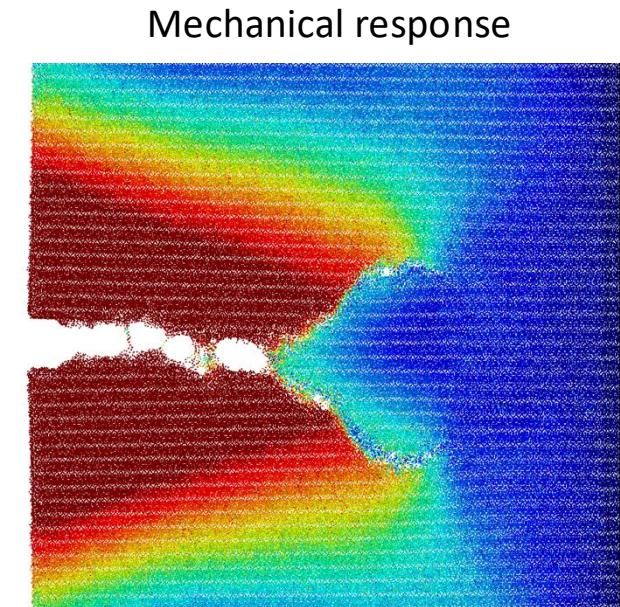
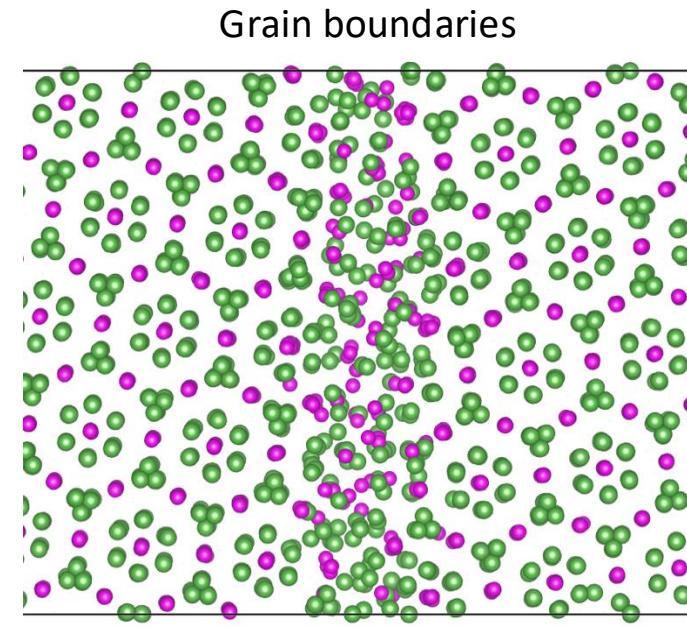
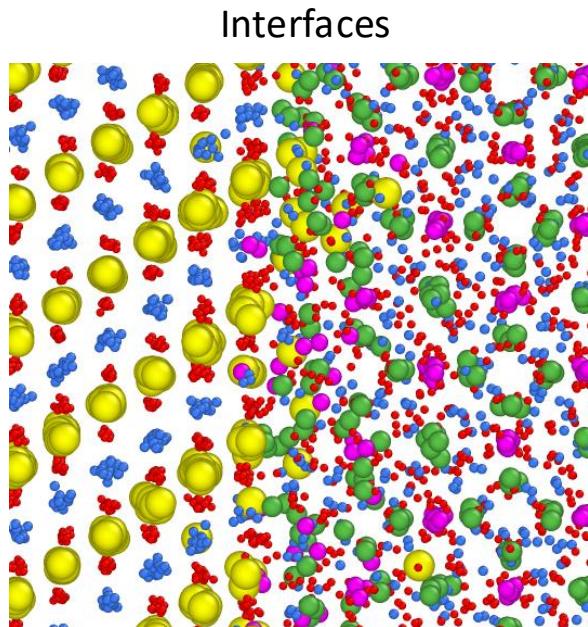
where f_i is the value of the property (a_0 , C_{ij} , E_v , γ_s , and γ_{us})

Tutorial for HDNNP with $\text{LiCoO}_2/\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ example

- Data generation
- Descriptors
- Training (n2p2 code)
- Model test/benchmarking

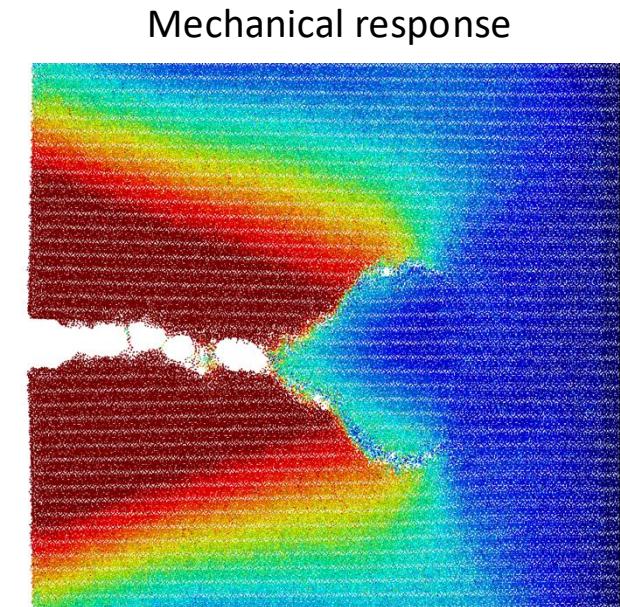
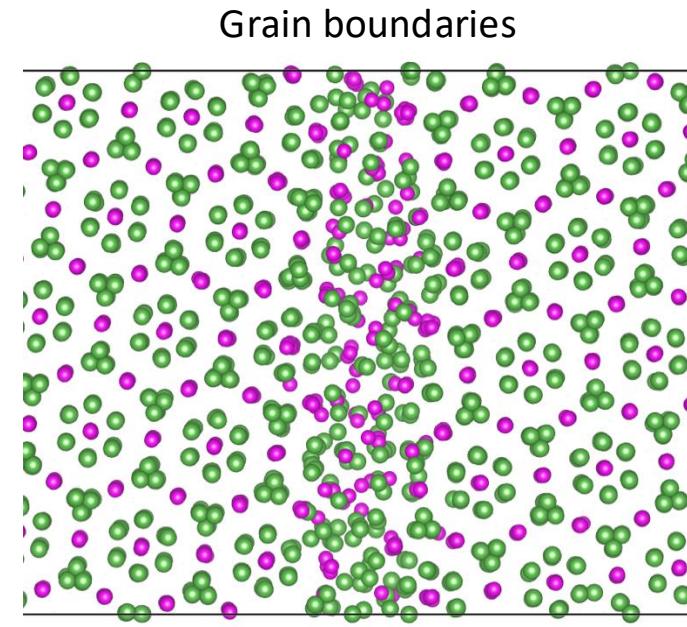
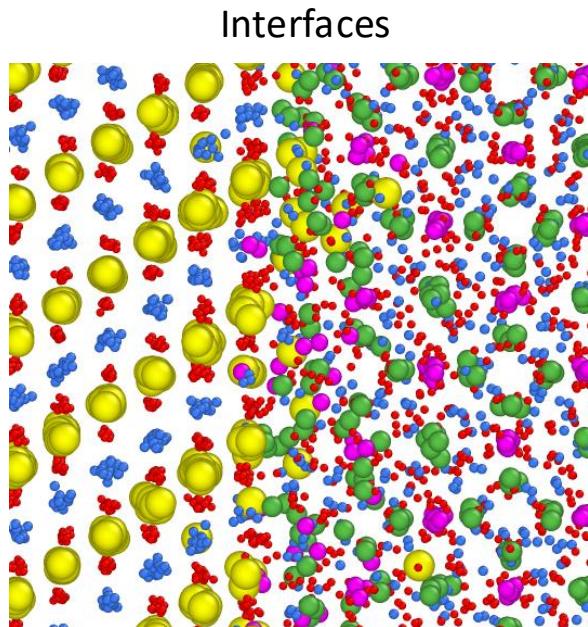
Dataset generation

- Structure data should provide information of local atomic environments at interfaces
 - Disordered/amorphous geometries with high compositional and configurational complexities increase difficulties for data generation



Dataset generation

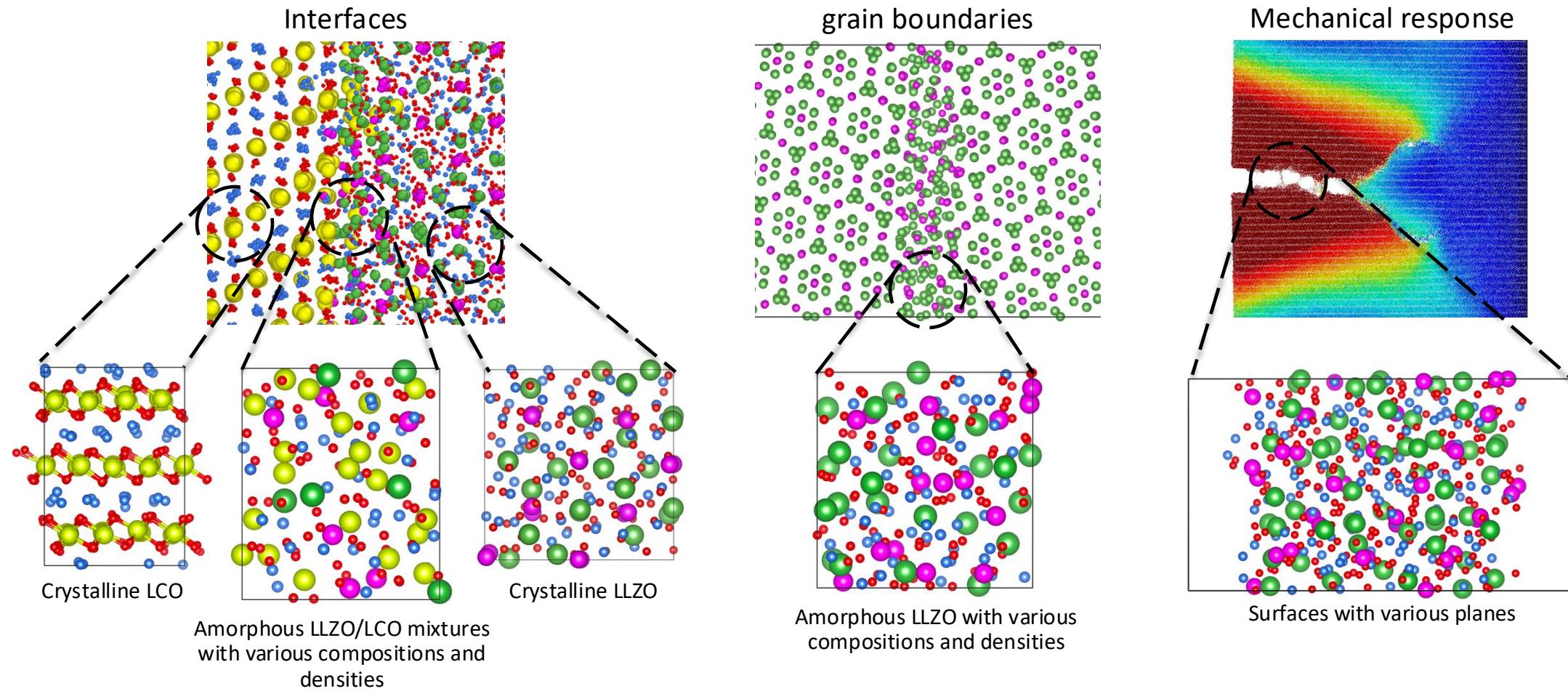
- Structure data should provide information of local atomic environments at interfaces
 - Disordered/amorphous geometries with high compositional and configurational complexities increase difficulties for data generation
 - DFT/AIMD cannot simulate actual disordered geometries over thousands of atoms due to limitation in length scale
→ size issue



Dataset generation

- Structure data should provide information of local atomic environments at interfaces

Strategy: collect various “local” configurations that could be observed at disordered geometries



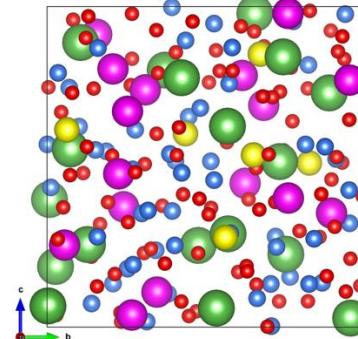
Dataset generation

- Example for data generation for LLZO/LCO interface [1]
 - Structures at high temperatures are necessary

Table S2. Data type and number of structures in the entire dataset. The total number of data is 12,651. The information of Models #1-5 of LLZO-LCO mixtures can be found in Table S1. For Co-doped LLZO, $n_{Co_x=Li, La, or Zr}$ indicates species x replaced by n Co atom(s) in a unit cell.

Data type		Number of structures (Temperature)
LLZO	Crystalline (cubic)	500 (1273 K); 500 (1473 K)
	Amorphous	500 (4000 K)
LCO	Crystalline (layered)	312 (1000 K); 692 (2000 K)
	Amorphous	502 (4000 K)
LLZO-LCO mixtures	Model #1	355 (2000 K); 508 (3000 K); 683 (4000 K)
	Model #2	366 (2000 K); 535 (3000 K); 710 (4000 K)
	Model #3	330 (2000 K); 534 (3000 K); 686 (4000 K)
	Model #4	339 (2000 K); 533 (3000 K); 712 (4000 K)
	Model #5	324 (2000 K); 527 (3000 K); 682 (4000 K)
	1Co _{Li}	47 (3000 K); 48 (4000 K)
	1Co _{La}	51 (3000 K); 48 (4000 K)
	1Co _{Zr}	52 (3000 K); 48 (4000 K)
	2Co _{Li}	47 (3000 K); 46 (4000 K)
	2Co _{La}	49 (3000 K); 52 (4000 K)
Co-doped amorphous LLZO	2Co _{Zr}	51 (3000 K); 48 (4000 K)
	3Co _{Li}	48 (3000 K); 47 (4000 K)
	3Co _{La}	47 (3000 K); 54 (4000 K)
	3Co _{Zr}	52 (3000 K); 52 (4000 K)
	4Co _{Li}	54 (3000 K); 49 (4000 K)
	4Co _{La}	52 (3000 K); 47 (4000 K)
	4Co _{Zr}	56 (3000 K); 48 (4000 K)
	5Co _{Li}	54 (3000 K); 49 (4000 K)
	5Co _{La}	56 (3000 K); 49 (4000 K)
	5Co _{Zr}	55 (3000 K); 54 (4000 K)
	6Co _{Li}	56 (3000 K); 47 (4000 K)
	6Co _{La}	52 (3000 K); 52 (4000 K)
	6Co _{Zr}	53 (3000 K); 51 (4000 K)

Bulk/amorphous
(Co-aLLZO)



LLZO-LCO mixture models
(local environment at interfaces)

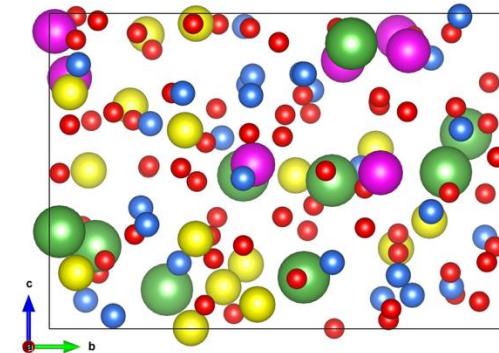
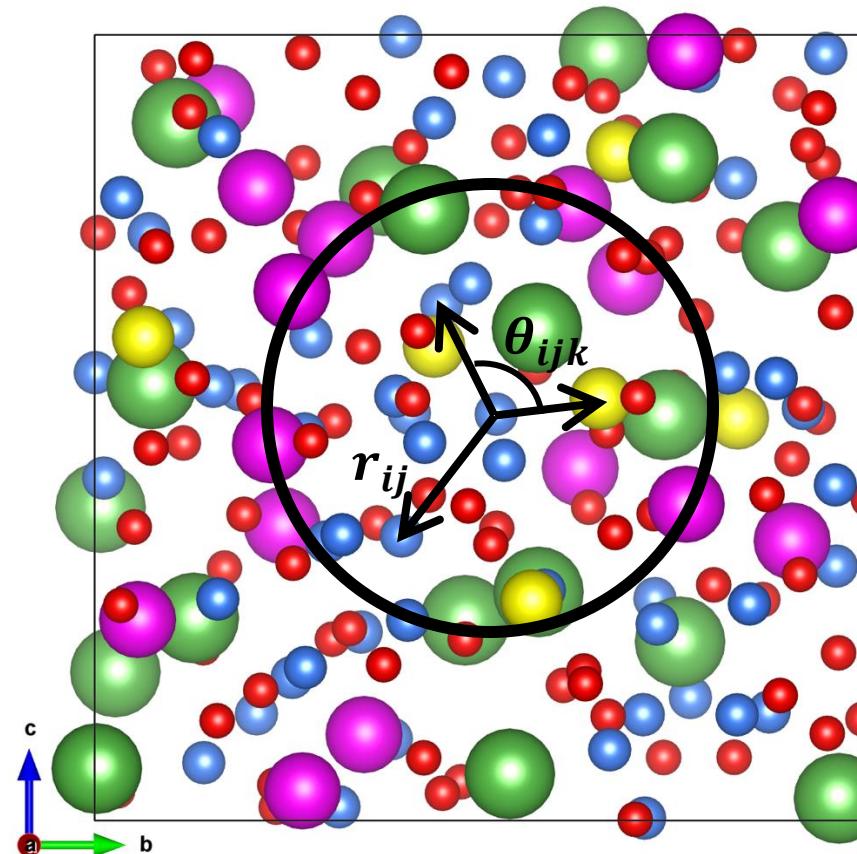


Table S3. Formula and the number of atoms in LLZO-LCO mixture models which were not used for training but only for validation. The number of Li in LLZO is counted assuming fully-lithiated LCO, i.e., $n_{Li-LLZO} = n_{Li-cell} - n_{Co}$, where $n_{Li-cell}$ and n_{Co} are the number of Li and Co atoms in the entire cell, respectively. Values in parenthesis are stoichiometries of Li, La, and Zr in LLZO assuming that the stoichiometry of O is 12, i.e., $s_{cation-LLZO} = n_{cation-LLZO} / (n_{O-LLZO}/12)$, where $n_{cation-LLZO}$ and n_{O-LLZO} are the number of cation and O atoms in LLZO, respectively.

	Formula	Li ₇ La ₃ Zr ₂ O ₁₂				LiCoO ₂			Total
		Li	La	Zr	O	Li	Co	O	
Model #1	Li ₄₂ La ₁₂ Zr ₄ O ₇₄ Co ₁₈	24 (7.58)	12 (3.79)	4 (1.26)	38	18	18	36	150
Model #2	Li ₄₃ La ₁₃ Zr ₆ O ₈₆ Co ₂₂	21 (6.00)	13 (3.71)	6 (1.71)	42	22	22	44	170
Model #3	Li ₂₉ La ₁₀ Zr ₅ O ₆₄ Co ₁₄	15 (5.00)	10 (3.33)	5 (1.67)	36	14	14	28	122
Model #4	Li ₃₉ La ₁₂ Zr ₈ O ₉₈ Co ₂₆	13 (3.39)	12 (3.13)	8 (2.09)	46	26	26	52	183
Model #5	Li ₃₂ La ₁₃ Zr ₆ O ₈₆ Co ₂₂	10 (2.86)	13 (3.71)	6 (1.71)	42	22	22	44	159

Behler-Parrinello (BP) symmetry functions for descriptors

- Local atomic environments need to be described quantitatively for inputs in ML model
 - 2B (**radial**) and 3B (**angular**) interactions
 - Satisfy rotational, translational, and permutational invariance



Behler-Parrinello (BP) symmetry functions for descriptors

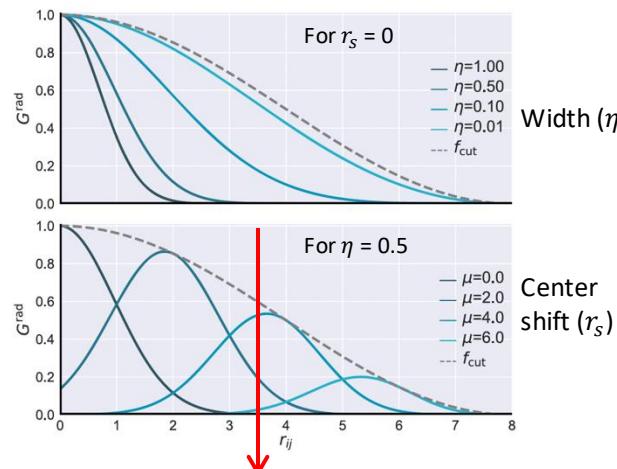
- Atom-centered symm. Functions (ACSFs) describe local atomic environments, which are inputs for neural network

Behler-Parrinello (BP) symmetric functions^{1,2}

Gaussian-type radial symm. functions

$$G_i^{\text{rad.}} = \sum_{j \neq i} e^{-\eta(r_{ij}-r_s)^2} f_c(r_{ij})$$

Effects of parameters on the shape of $G_i^{\text{rad.}}$ [4]

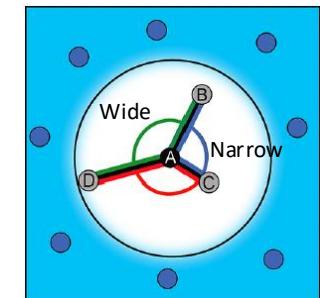


For a certain ij pair,
the data in vector form = $[G_{\text{rad}1} \dots G_{\text{rad}4}, \dots] = [0, 0.18, 0.53, 0.04, \dots]$
Each component should be summed over all neighbor atoms j

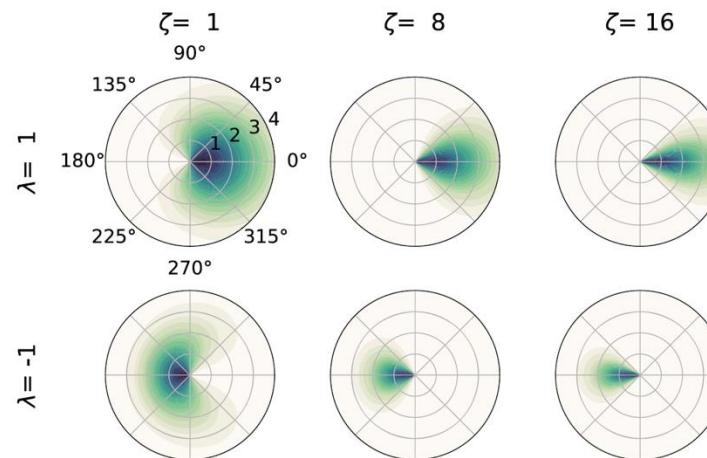
Narrow and wide angular symm. functions

$$\begin{aligned} G_i^{\text{ang.narrow}} &= 2^{1-\zeta} \sum_{j,k \neq i, j < k} (1 + \lambda \cos \theta_{ijk})^\zeta \times e^{-\eta(r_{ij}^2 + r_{ik}^2 + r_{jk}^2)} \times f_c(r_{ij}) f_c(r_{ik}) f_c(r_{jk}) \\ G_i^{\text{ang.wide}} &= 2^{1-\zeta} \sum_{j,k \neq i, j < k} (1 + \lambda \cos \theta_{ijk})^\zeta \times e^{-\eta(r_{ij}^2 + r_{ik}^2)} \times f_c(r_{ij}) f_c(r_{ik}) \end{aligned}$$

Atomic environment [3]

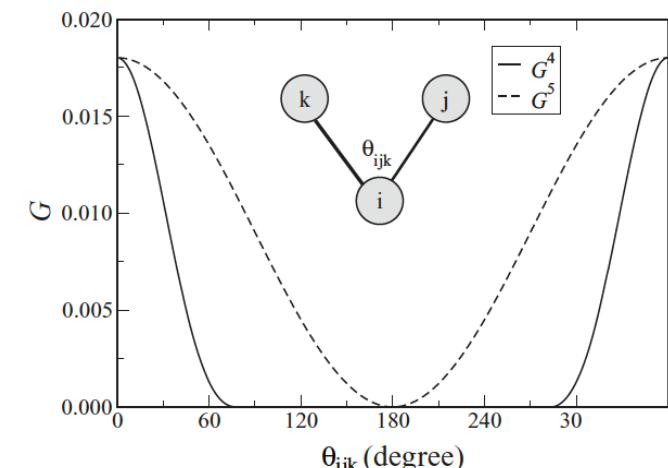


Effects of parameters on the shape of $G_i^{\text{rad.}}$ [4]



Detection range and direction are changed with ζ and λ , respectively.
 r_s is not shown but also can be included.
Wide version can be optional.

Narrow (G^4) and wide (G^5) ang. symm. Functions [2]



Behler-Parrinello (BP) symmetry functions for descriptors

- Atom-centered symm. Functions (ACSFs) describe local atomic environments, which are inputs for neural network

Behler-Parrinello (BP) symmetric functions^{1,2}

Gaussian-type radial symm. functions

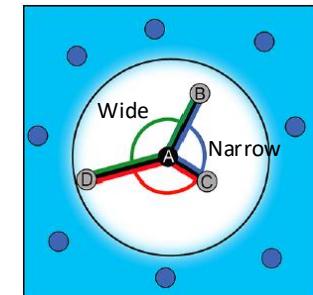
$$G_i^{\text{rad.}} = \sum_{j \neq i} e^{-\eta(r_{ij}-r_s)^2} f_c(r_{ij})$$

Narrow and wide angular symm. functions

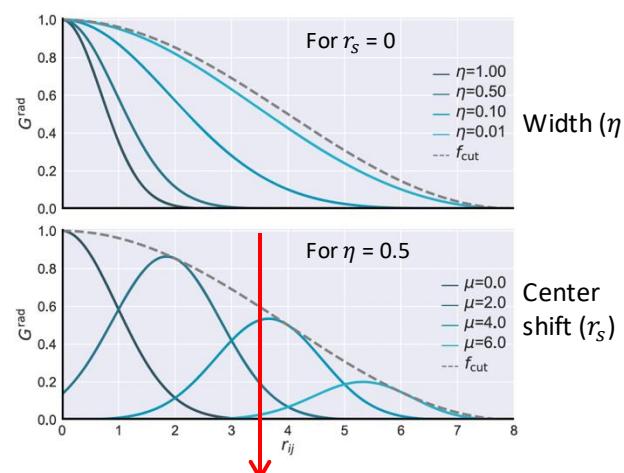
$$G_i^{\text{ang.narrow}} = 2^{1-\zeta} \sum_{j,k \neq i; j < k} (1 + \lambda \cos \theta_{ijk})^\zeta \times e^{-\eta(r_{ij}^2+r_{ik}^2+r_{jk}^2)} \times f_c(r_{ij}) f_c(r_{ik}) f_c(r_{jk})$$

$$G_i^{\text{ang.wide}} = 2^{1-\zeta} \sum_{j,k \neq i; j < k} (1 + \lambda \cos \theta_{ijk})^\zeta \times e^{-\eta(r_{ij}^2+r_{ik}^2)} \times f_c(r_{ij}) f_c(r_{ik})$$

Atomic environment [3]

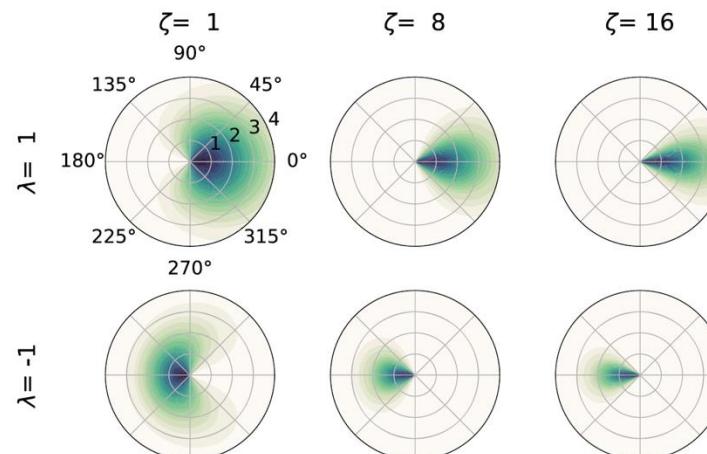


Effects of parameters on the shape of $G_i^{\text{rad.}}$ [4]



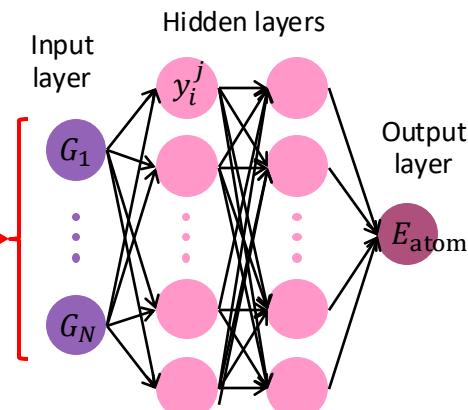
For a certain ij pair,
the data in vector form = $[G_{\text{rad}1} \dots G_{\text{rad}4}, \dots] = [0, 0.18, 0.53, 0.04, \dots]$
Each component should be summed over all neighbor atoms j

Effects of parameters on the shape of $G_i^{\text{rad.}}$ [4]



Detection range and direction are changed with ζ and λ , respectively.
 r_s is not shown but also can be included.
Wide version can be optional.

Neural-network architecture (per element)



A list of symmetry function values to the nodes in the input layer

Behler-Parrinello (BP) symmetry functions for descriptors

- There are typical ways to determine parameters in the symmetry functions
- The number of symmetry functions increases significantly with the number of elements

Radial symmetry functions

```
#####
# Radial symmetry function set, for elements ['Li', 'La', 'Zr', 'O', 'Co']
#####
# r_cutoff      = 6.0
# The following settings were used for generating sets
# of values for the radial parameters r_shift and eta:
# rule          = imbalzano2018
# mode          = shift
# nb_param_pairs = 9
# Sets of values for parameters:
# r_shift_grid  = [0.6667 0.851  1.0863 1.3867 1.7702 2.2597 2.8845 3.6821 4.7003]
# eta_grid      = [29.4263 18.0585 11.0822 6.801   4.1737 2.5613 1.5719 0.9646 0.592 ]

symfunction_short Li 2 Li 2.943E+01 6.667E-01 6.000E+00
symfunction_short Li 2 Li 1.806E+01 8.510E-01 6.000E+00
symfunction_short Li 2 Li 1.108E+01 1.086E+00 6.000E+00
symfunction_short Li 2 Li 6.801E+00 1.387E+00 6.000E+00
symfunction_short Li 2 Li 4.174E+00 1.770E+00 6.000E+00
symfunction_short Li 2 Li 2.561E+00 2.260E+00 6.000E+00
symfunction_short Li 2 Li 1.572E+00 2.884E+00 6.000E+00
symfunction_short Li 2 Li 9.646E-01 3.682E+00 6.000E+00
symfunction_short Li 2 Li 5.920E-01 4.700E+00 6.000E+00
```

$$G_i^{\text{rad.}} = \sum_{j \neq i} e^{-\eta(r_{ij} - r_s)^2} f_c(r_{ij})$$

$$\begin{aligned} r_{s,m} &= r_c / n^{m/n} \\ \eta_m &= 1 / (r_{s,m} - r_{s,m-1})^2 \\ m &= \{1, 2, \dots, n\}; n = 9 \end{aligned}$$

G. Imbalzano et al., *J. Chem. Phys.* **2018**, *148*, 241730.

Python code for generating symmetry function set is available (by n2p2 developer) [1,2]

Narrow and wide angular symmetry functions

```
#####
# Narrow angular symmetry function set, for elements ['Li', 'La', 'Zr', 'O', 'Co']
#####
# r_cutoff      = 6.0
# The following settings were used for generating sets
# of values for the radial parameters r_shift and eta:
# rule          = gastegger2018
# mode          = center
# nb_param_pairs = 3
# r_lower       = 1.5
# r_upper       = 6.0
# Sets of values for parameters:
# r_shift_grid  = [0. 0. 0.]
# eta_grid      = [0.2222 0.0356 0.0139]
# lambdas       = [-1. 1.]
# zetas         = [1. 6.]

symfunction_short Li 3 Li Li 2.222E-01 -1 1.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 2.222E-01 1 1.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 2.222E-01 -1 6.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 2.222E-01 1 6.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 3.556E-02 -1 1.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 3.556E-02 1 1.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 3.556E-02 -1 6.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 3.556E-02 1 6.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 1.389E-02 -1 1.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 1.389E-02 1 1.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 1.389E-02 -1 6.000E+00 6.000E+00 0.000E+00
symfunction_short Li 3 Li Li 1.389E-02 1 6.000E+00 6.000E+00 0.000E+00
```

12 symm functions × 15 blocks for each element
= 180 symm. functions

Twice if using both types

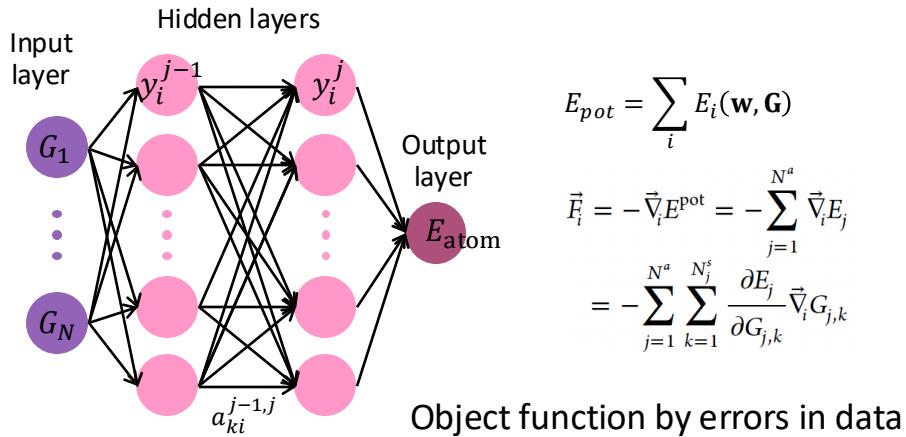
$$\begin{aligned} G_i^{\text{ang.narrow}} &= 2^{1-\zeta} \sum_{j,k \neq i; j < k} (1 + \lambda \cos \theta_{ijk})^\zeta \times e^{-\eta(r_{ij}^2 + r_{ik}^2 + r_{jk}^2)} \times f_c(r_{ij}) f_c(r_{ik}) f_c(r_{jk}) \\ \eta_m &= 1/(2r_m^2) \text{ with } N \text{ equally spaced points } (N=3); \\ r_m &\text{ ranging from } r_0 = 1.5 \text{ \AA} \text{ and } r_{N-1} = r_c \\ \zeta &= \{1, 6\} \\ \lambda &= \{-1, 1\} \end{aligned}$$

M. Gastegger et al., *J. Chem. Phys.* **2018**, *148*, 241709.

Training ML model

- A number of model/training parameters, a few important to test/determine
 - Important: # layers and nodes, force weights, ratio, and selection mode; use default settings for other parameters

Neural-network architecture



$$y_i^j = f_a^j \left(b_i^j + \sum_k a_{ki}^{j-1,j} \cdot y_k^{j-1} \right)$$

Activation function: on/off function by tanh, logistic, softplus, linear, etc.
They work well similarly.[1]

Important parameters (n2p2)

global_hidden_layers_short	2
global_nodes_short	20 20

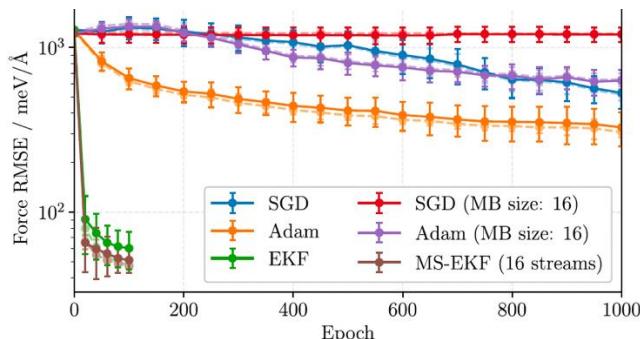
test_fraction	0.1
force_weight	10.0

force_energy_ratio	10.0
selection_mode (0 = Random, 1 = Sort, 2 = Threshold)	

Sample force data for training every epoch for effective/efficient training

Training algorithm (n2p2)

Multi-stream extended Karman filter outperforms other methods [1]



$(\eta_0, \eta_{\max}, \tau_{\eta}^{\text{epoch}}), (q_0, q_{\min}, \tau_q^{\text{epoch}})$

kalman_epsilon	1.0E-2
kalman_q0	0.01
kalman_qtau	2.302
kalman_qmin	1.0E-6
kalman_eta	0.01
kalman_etatau	2.302
kalman_eta_max	1.0

Tested by
developers [1]

$$\text{Learning rate } \eta(t) = \min\left(\eta_0 e^{t/u\tau_{\eta}^{\text{epoch}}}, \eta_{\max}\right)$$

$$\text{Artificial process noise } q(t) = \max\left(q_0 e^{t/u\tau_q^{\text{epoch}}}, q_{\min}\right)$$

Training ML model

- At least 40-50 steps are needed, but at most 100 steps
 - Training curve and validation parity plots for energies and forces

	ep	E_count	E_train	E_test	E_pt
force	ep	F_count	F_train	F_test	F_pt
timing	ep	count	train error	other	epoch total
ENERGY	0	0	5.15840E-01	5.18112E-01	0.0
FORCE	0	0	2.44126E+00	2.42716E+00	0.0
TIMING	0	0.0	90.6	9.4	32.92 32.92

ENERGY	1	316	1.20730E-02	1.26898E-02	7.8
FORCE	1	3202	6.36616E-01	6.52999E-01	92.2
TIMING	1	3518	99.8	0.1	0.0 10943.74 10976.66

ENERGY	2	316	1.59176E-02	1.61653E-02	7.7
FORCE	2	3202	6.43364E-01	5.88598E-01	92.3
TIMING	2	3518	99.8	0.2	0.0 10553.65 21530.32

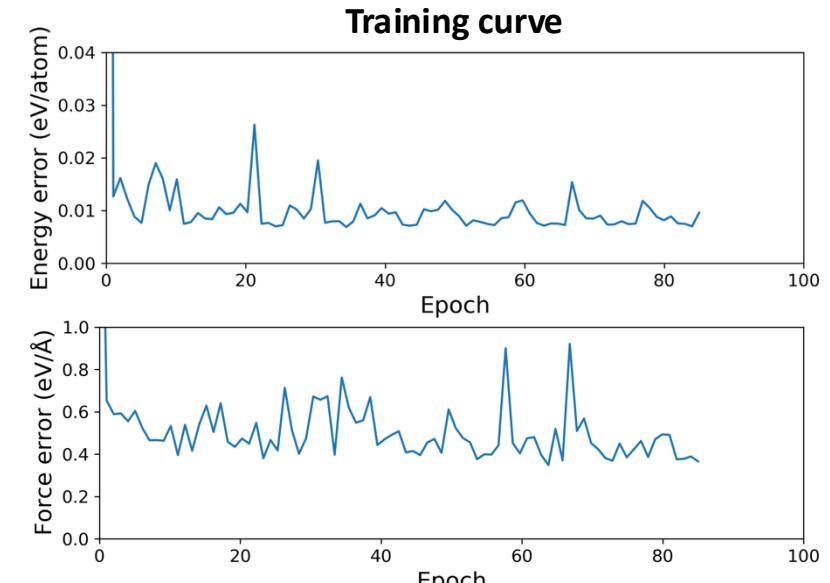
ENERGY	3	316	1.22059E-02	1.21437E-02	7.7
FORCE	3	3202	6.20207E-01	5.92178E-01	92.3
TIMING	3	3518	99.8	0.1	0.0 10558.98 32089.29

ENERGY	4	316	8.40317E-03	8.82657E-03	7.8
FORCE	4	3202	4.77805E-01	5.55042E-01	92.2
TIMING	4	3518	99.8	0.1	0.0 10769.56 42858.85

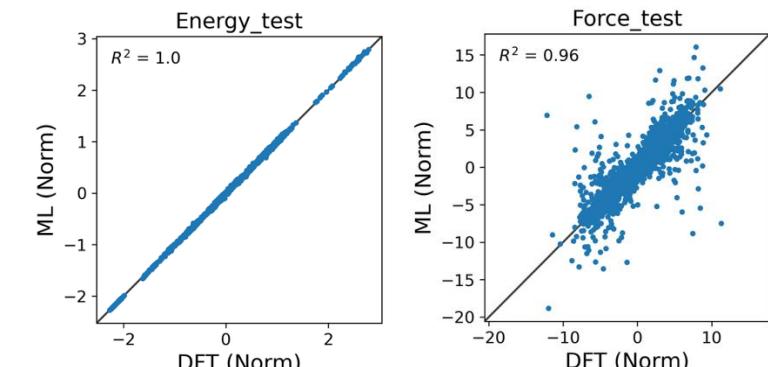
ENERGY	5	316	7.38112E-03	7.67552E-03	7.7
FORCE	5	3202	5.14072E-01	6.03842E-01	92.3
TIMING	5	3518	99.8	0.1	0.0 10924.30 53783.15

```

INFO  Switching selection mode for property "energy" to SM_SORT (1).
INFO  Switching selection mode for property "force" to SM_SORT (1).
ENERGY 6   316  1.43133E-02  1.48987E-02    7.9
FORCE   6   3202 4.76752E-01  5.24212E-01  92.1
TIMING  6   3518 99.9  0.1    0.0  10090.00  63873.15
-----
ENERGY 7   316  1.94100E-02  1.89989E-02    7.9
FORCE   7   3202 4.76752E-01  4.65406E-01  92.1
TIMING  7   3518 99.8  0.1    0.0  10293.32  74166.47
-----
ENERGY 8   316  1.65912E-02  1.60942E-02    7.9
FORCE   8   3202 1.54716E+00  4.66020E-01  92.1
TIMING  8   3518 99.8  0.1    0.0  10662.03  84828.50
-----
ENERGY 9   316  9.93164E-03  1.00522E-02    7.8
FORCE   9   3202 6.32645E-01  4.63204E-01  92.2
TIMING  9   3518 99.8  0.1    0.0  10180.90  95009.40
-----
ENERGY 10  316  1.58756E-02  1.59296E-02    7.9
FORCE   10  3202 6.02403E-01  5.32629E-01  92.1
TIMING 10  3518 99.8  0.1    0.0  10232.01  105241.41
-----
INFO  Switching selection mode for property "energy" to SM_THRESHOLD (2).
INFO  Switching selection mode for property "force" to SM_THRESHOLD (2).
ENERGY 81  316  7.17798E-03  7.55830E-03    7.5
FORCE   81  3202 3.67075E-01  3.75701E-01  92.5
TIMING 81  3518 99.8  0.1    0.0  10001.37  823972.97
-----
ENERGY 82  316  6.94839E-03  7.47458E-03    7.7
FORCE   82  3202 3.73480E-01  3.77231E-01  92.3
TIMING 82  3518 99.8  0.1    0.0  10562.59  834535.56
-----
ENERGY 83  316  6.59720E-03  7.01826E-03    7.6
FORCE   83  3202 4.09406E-01  3.88988E-01  92.4
TIMING 83  3518 99.8  0.1    0.0  10289.92  844825.48
-----
ENERGY 84  316  9.53091E-03  9.60208E-03    7.4
FORCE   84  3202 4.40382E-01  3.65881E-01  92.6
TIMING 84  3518 99.8  0.1    0.0  9506.21  854331.69
-----
```



→ **Energy/force validation (test set)**

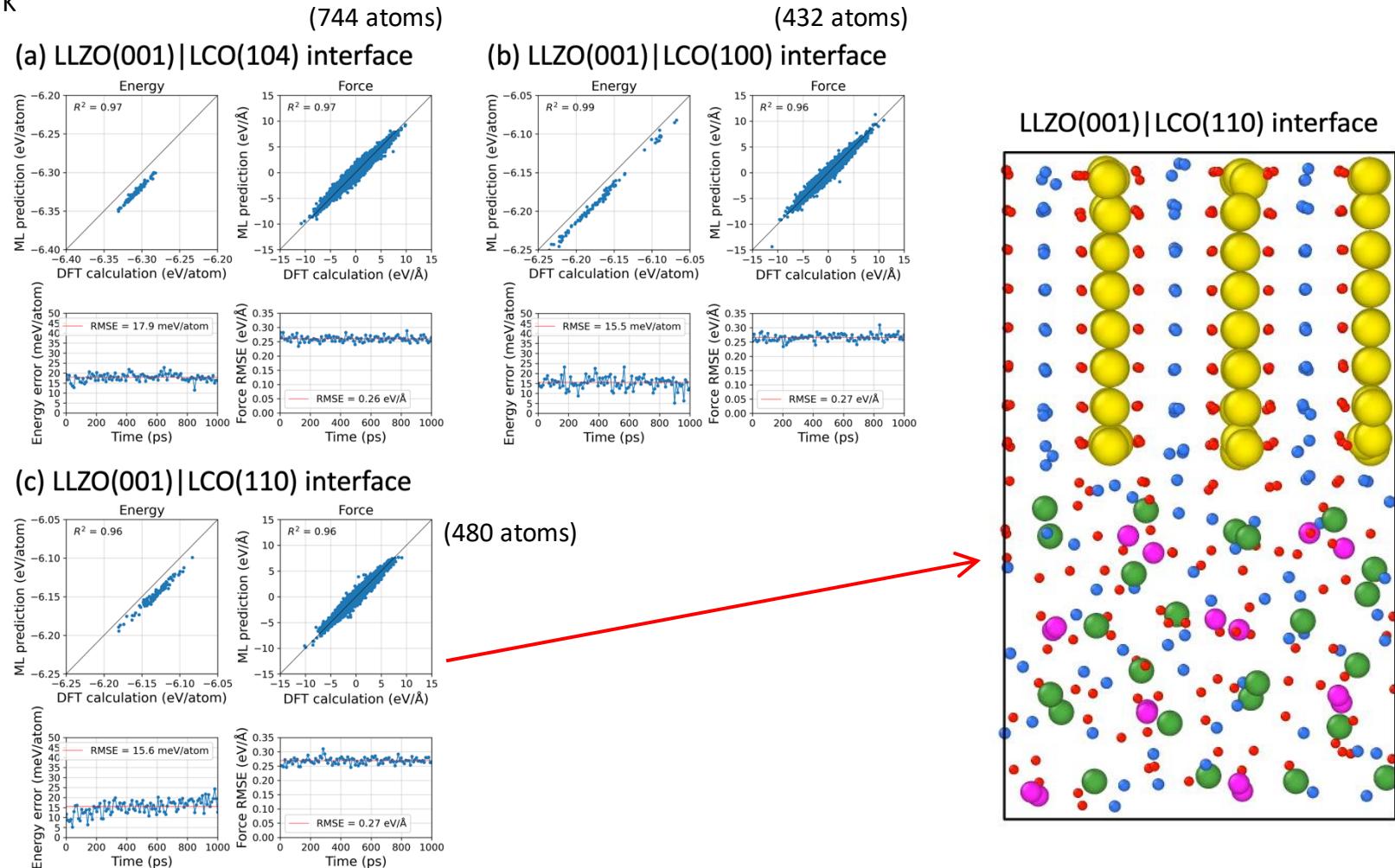


Model benchmark tests

- Test errors for energies and forces
 - Low energy and force errors during 1 ns with large changes in the atomic structures

For snapshots from 100 ps traj. by nvt MD with MLFF at 2000 K

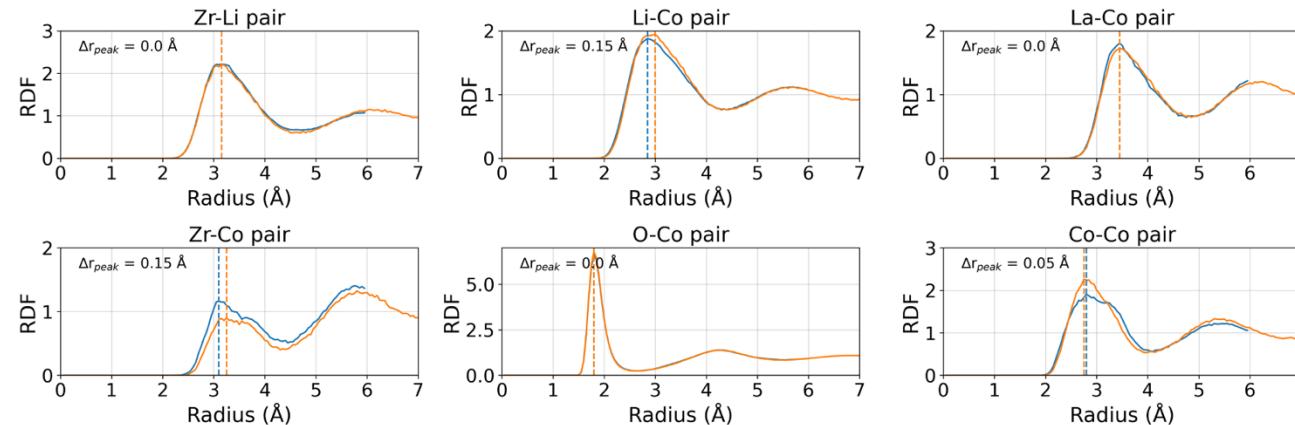
System type		Energy	Force
LLZO	Cubic	4.3 (0.99)	0.14 (0.99)
	Amorphous	11.0 (0.97)	0.19 (0.98)
LCO	Layered	8.0 (0.99)	0.19 (0.99)
	Model #1	21.9 (0.93)	0.26 (0.96)
LLZO-LCO mixtures	Model #2	22.4 (0.90)	0.28 (0.96)
	Model #3	24.9 (0.91)	0.27 (0.96)
	Model #4	28.6 (0.96)	0.28 (0.96)
	Model #5	23.9 (0.93)	0.29 (0.96)
	6Co _{Li}	27.6 (0.89)	0.28 (0.96)
Co-doped amorphous LLZO	6Co _{La}	15.6 (0.96)	0.22 (0.97)
	6Co _{Zr}	20.3 (0.95)	0.24 (0.97)
	Model #1	15.8 (0.93)	0.27 (0.96)
LLZO-LCO mixtures (unseen)	Model #2	20.9 (0.92)	0.26 (0.96)
	Model #3	26.2 (0.95)	0.26 (0.96)
	Model #4	25.8 (0.94)	0.27 (0.96)
	Model #5	22.6 (0.96)	0.28 (0.96)
	LLZO(001)/LCO(104)	17.9 (0.97)	0.26 (0.97)
LLZO-LCO interfaces	LLZO(001)/LCO(100)	15.5 (0.99)	0.27 (0.96)
	LLZO(001)/LCO(110)	15.6 (0.96)	0.27 (0.96)



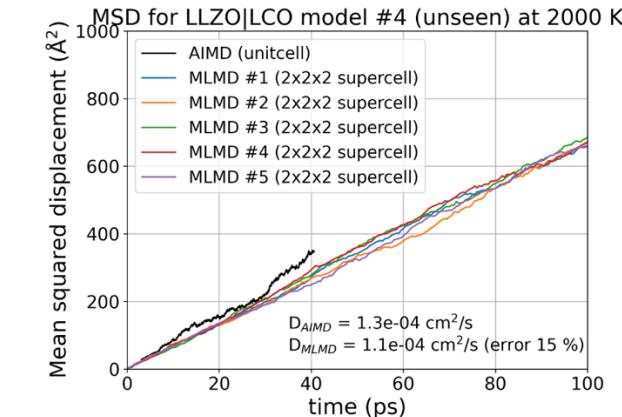
Model benchmark tests

- Benchmarking tests for structural, vibrational, and dynamical properties against DFT/AIMD

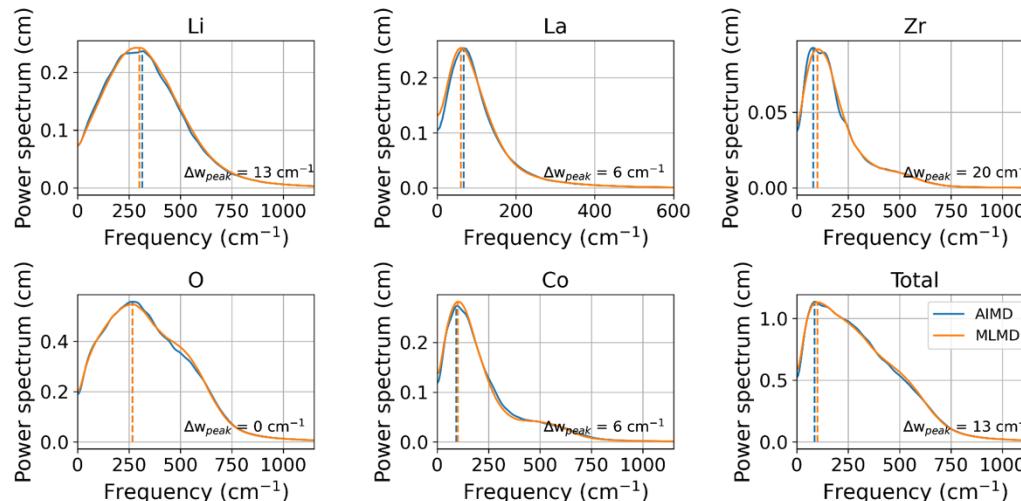
Radial distribution functions



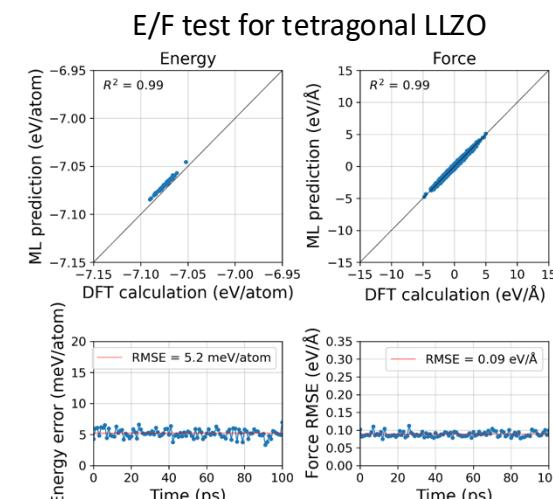
Li diffusivity



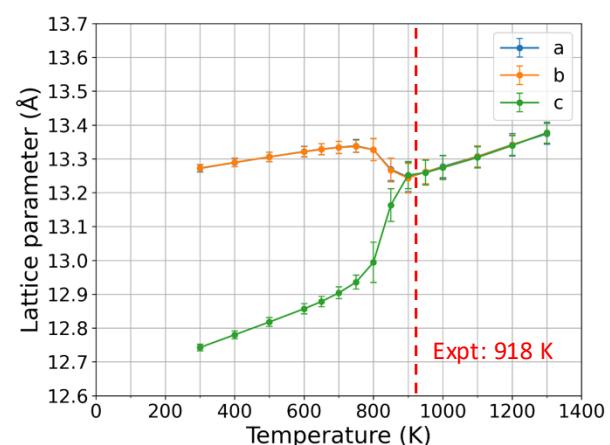
Vibrational density of states



LLZO phase transformation



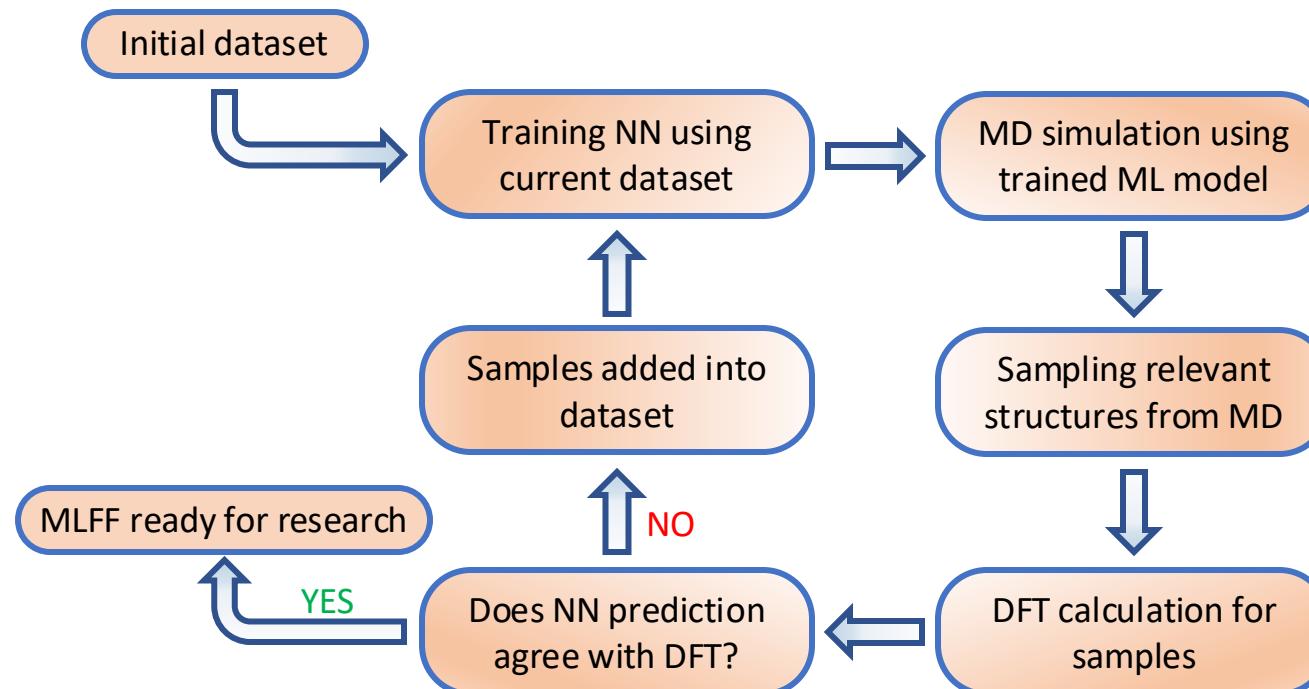
Tetragonal-to-cubic phase transformation



Active learning

- Iterative training to satisfy desired performance
 - DFT/AIMD cannot simulate all possible configurations due to limitation in time scale → **sampling issue**
 - The initial dataset is not likely to be complete to provide enough information for local atomic environment

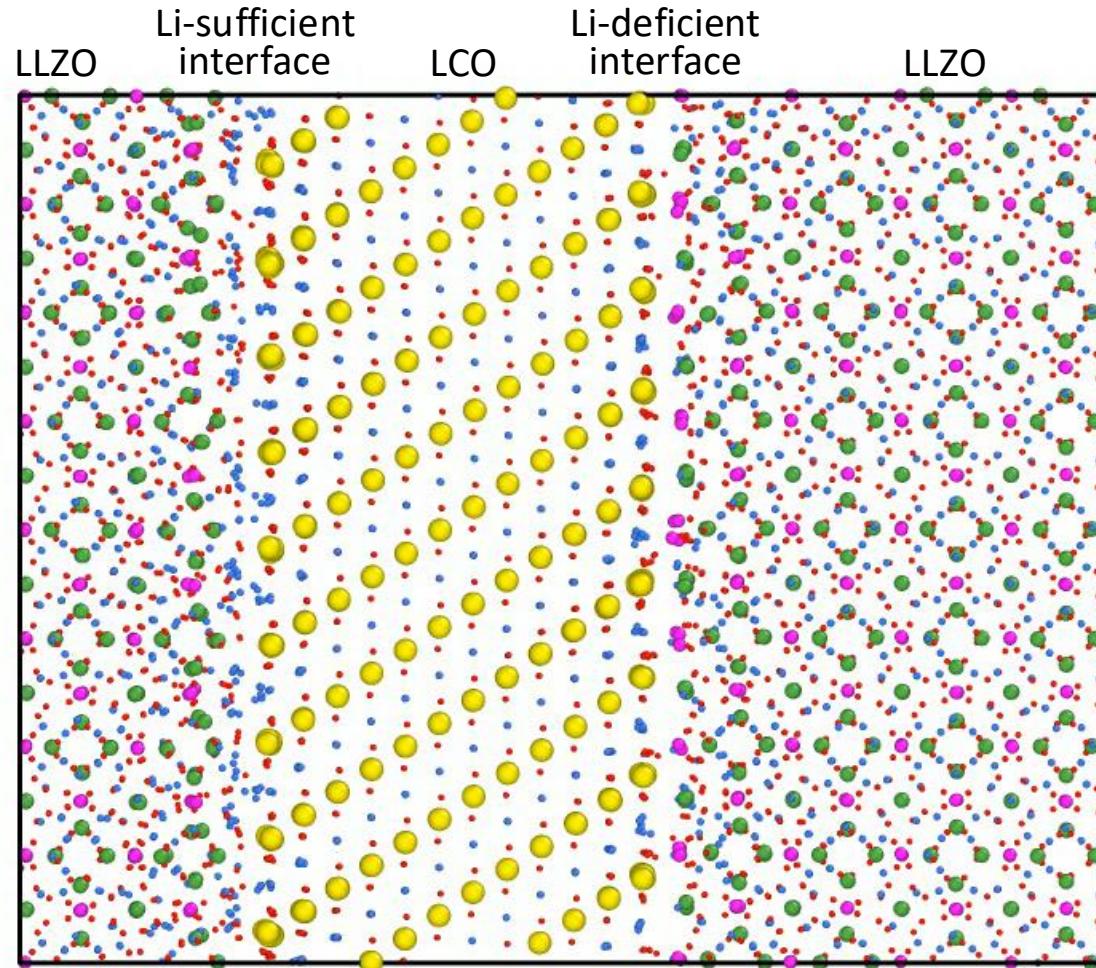
Active learning to generate data and train MLFF iteratively



Demonstration: application to SSB research

Application: understanding interfacial evolution and interdiffusion

- Direct observation of interfacial evolution with ML potential
 - Different surface conditions at two interfaces: Li sufficiency and deficiency



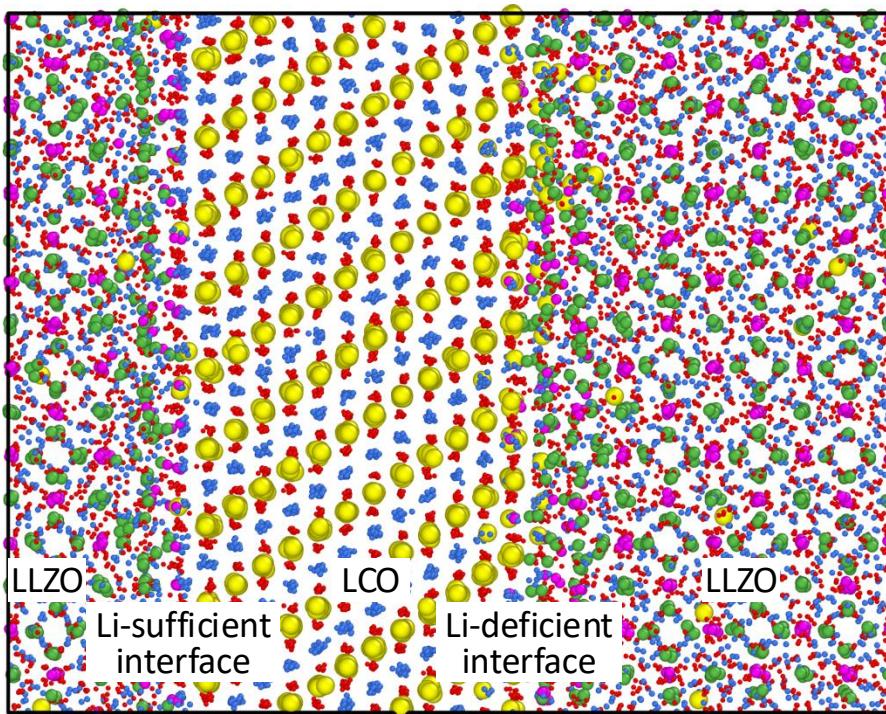
LLZO(001)/LCO(104) at 1500 K
during 1.8 ns

Blue: Li, Green: La, Magenta: Zr,
Red: O, Yellow: Co
(total 17k atoms)

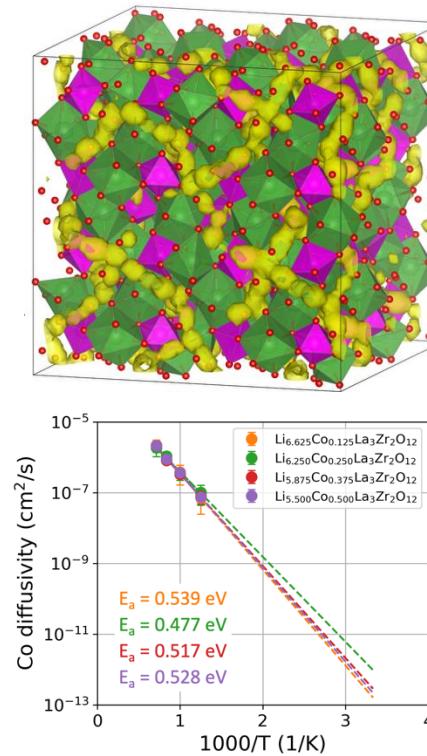
LLZO|LCO interface evolution / formation of Co-rich phase at GBs

- Different LLZO surface chemistries result in different propensities of interfacial evolution
- Fast Co diffusion via Li channel in cubic LLZO at high temperature
- Co ions show segregation, trapping, and clustering at the GB

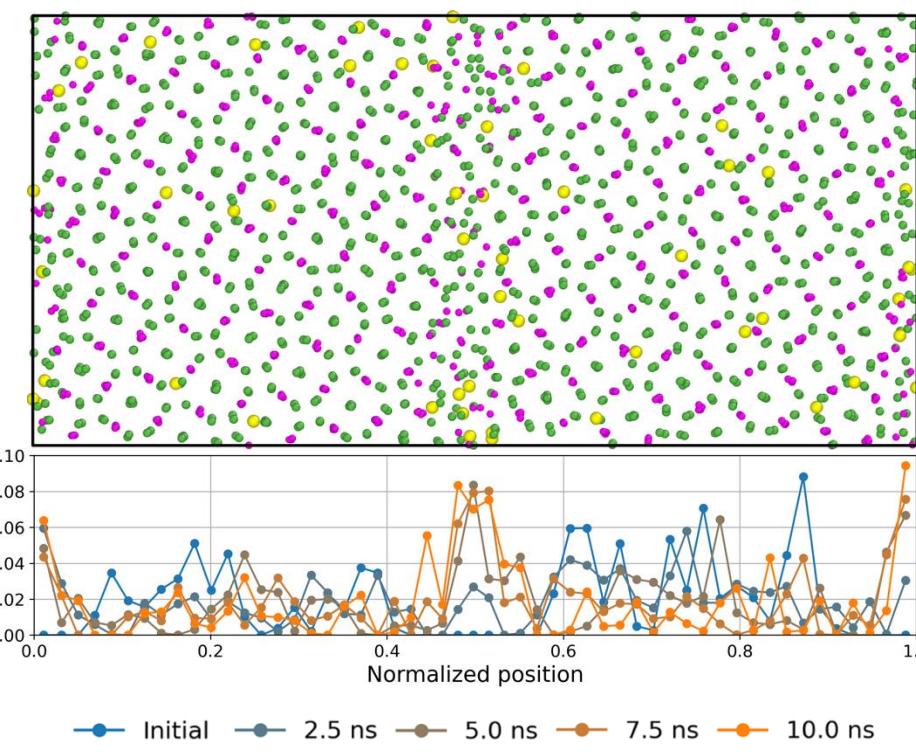
LLZO(001)/LCO(104) at 1500 K after 10 ns



Co diffusion in Li channel



$\Sigma 13(230)/[001]$ symmetric tilt GB at 1300 K after 10 ns



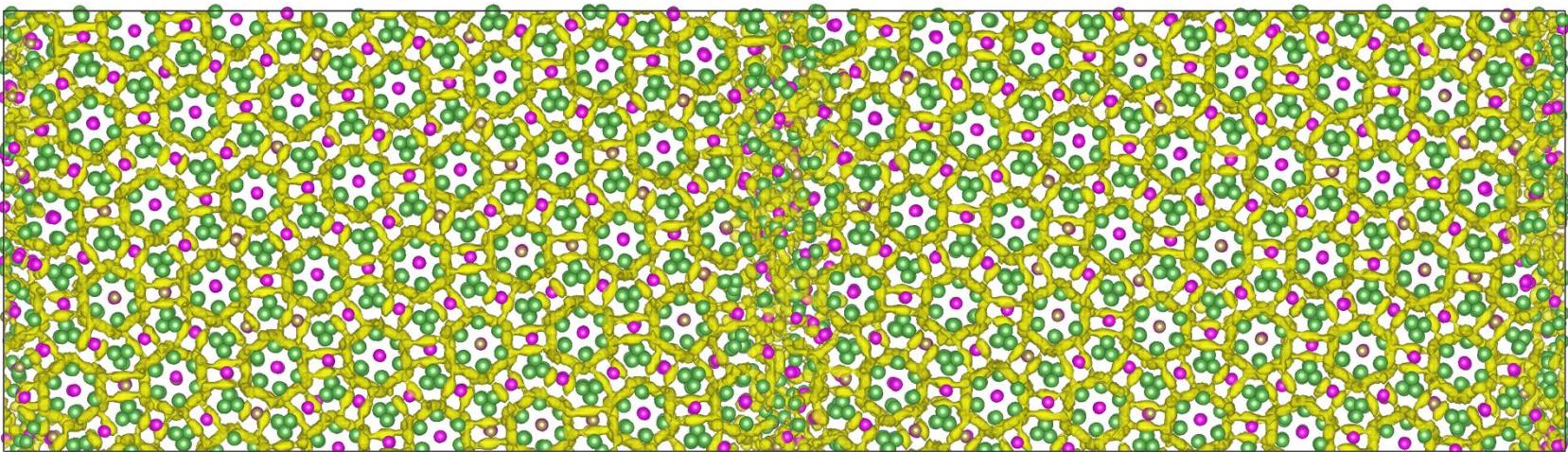
Application: understanding dopant segregation at LLZO GBs

- Mechanisms of dopant segregation at GBs and their effects on the properties of LLZO
 - Thermodynamics and kinetics determine the degree of segregation
 - Detrimental effects of segregations on ionic and electronic transports

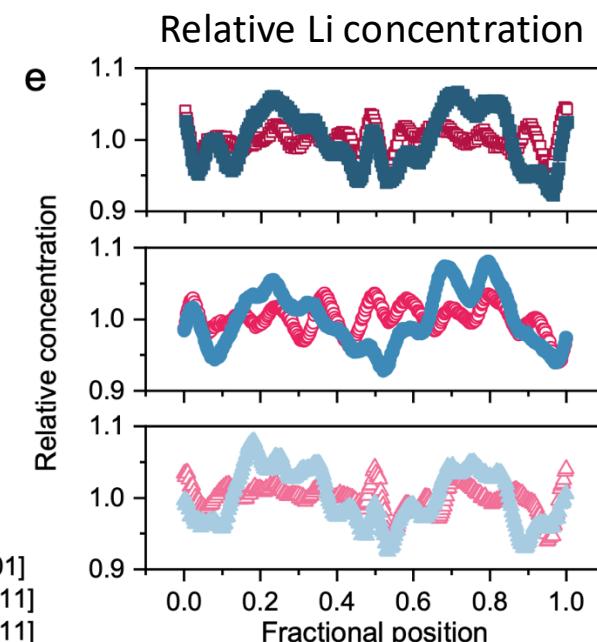
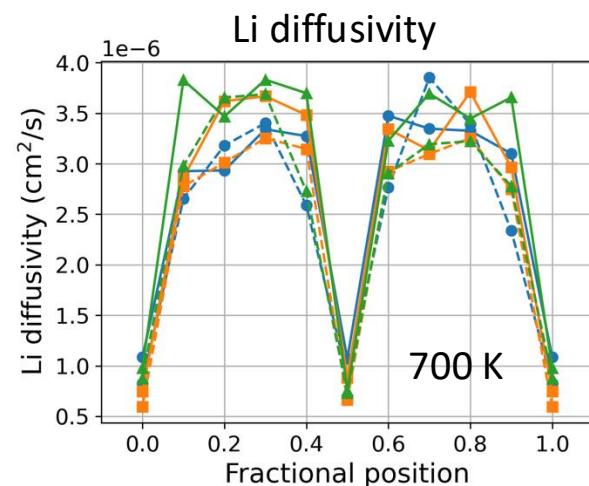
Upcoming talk:

EN03.06.02 - Interfacial Degradation in All-Solid-State Batteries
in Atomic Scale Using Machine Learning Force Fields
Wednesday 8:15 am; Hynes, Level 3, Room 304

Li positional probability density plot in $\Sigma 19(32-5)/[111]$ GB at 1100 K for 1 ns



- | | |
|----------------------------|---------------------------------|
| □ $\Sigma 17 (530) [001]$ | ■ Seg. $\Sigma 17 (530) [001]$ |
| ○ $\Sigma 33 (52-2) [011]$ | ● Seg. $\Sigma 33 (52-2) [011]$ |
| △ $\Sigma 19 (32-5) [111]$ | △ Seg. $\Sigma 19 (32-5) [111]$ |



Summary

- Introduction to MLIP
- Current status of MLIP application to SSB interfaces
- Tutorial for HDNNP with $\text{LiCoO}_2/\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ example
- Demonstration: application to SSB research
- Perspectives

K. Kim et al., **Machine-learning Interatomic Potentials for Interfaces in All-solid-state Batteries: Perspectives on Training Data, Model Selection, and Validation**, Under review.



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