CONNECTING SCALES: LEARNING DYNAMICS FOR EFFICIENT IONIC CONDUCTIVITY PREDICTIONS WITH GRAPHS

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ABSTRACT

Multiscale approaches are crucial for advancing our understanding of material properties, particularly in the search for novel solid electrolytes essential for solid-state batteries. Estimating ionic conductivity through traditional molecular dynamics (MD) simulations is computationally intensive, requiring significant time to capture macro-scale behavior from micro-scale interatomic interactions. This work addresses the challenge of connecting micro-scale interatomic potentials with macro-scale conductivity measurements. We propose using equivariant graph neural networks to develop a faster mapping between these scales, significantly enhancing the efficiency of ionic diffusion predictions. This proof-of-concept demonstrates the potential to accelerate material discovery for solid electrolytes, addressing a critical need in energy storage technology.

1 INTRODUCTION

Understanding material properties across different scales is fundamental to advancing the development of solid electrolytes for solid-state batteries (Kim et al., 2015). These materials require high ionic conductivity, which is typically estimated through molecular dynamics (MD) simulations for the computational screening of superionic candidates (Kahle et al., 2020). However, the computational demands of MD—often requiring extensive time to capture the necessary macro-scale[1] behavior from micro-scale[2] interactions—pose significant challenges.

MD simulations require the repeated steps for integrating the equations of motion, which can be slow and impractical for large-scale materials screening. The interatomic potentials that drive these equations of motion can be derived from first-principles density functional theory (DFT) methods, which are reliable but also computationally expensive. In contrast, machine-learned and classical potentials offer faster computations but may sacrifice accuracy. Importantly, the mapping from interatomic potentials to diffusivity values remains consistent across these approaches. Thus, by learning this mapping using the more cost-effective methods, we can possibly generalize the solution to DFT calculations without extensive retraining.

To address this, we propose using equivariant graph neural networks to establish an efficient mapping between micro-scale interatomic potentials and macro-scale ionic conductivity predictions. By

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[1] ranging up to hours and centimeters (Takai et al., 2004)

[2] of the order of angstroms and picoseconds

utilizing the inherent symmetry properties of atomic interactions, these networks should be able to capture the relationships that influence ionic diffusion. The proof-of-concept approach outlined in this work illustrates the feasibility of this method and its promise for advancing the discovery of novel solid electrolytes.

2 METHODOLOGY

We use MD trajectories for the 179 lithium-containing materials from the Materials Project database (Jain et al., 2013) simulated by Maevskiy et al. (2024) with the SevenNet universal potential (Park et al., 2024) at a temperature of 1000 K. The slopes of the mean squared displacement (MSD) as a function of time, $\frac{d}{dt}$MSD, served as ground-truth labels for the training process. These slopes determine the ion diffusivity and are related to the ionic conductivity via the Nernst-Einstein equation. Since these values span over several orders of magnitude, we take base-10 logarithm of $\frac{d}{dt}$MSD measured in Å^2 / ps as the prediction target.

To numerically represent the interatomic potential, which, for N atoms, is a $\mathbb{R}^{3\times N} \to \mathbb{R}$ function, we evaluate it at various randomized structural configurations. The optimal randomization strategy remains to be determined; however, in this proof-of-concept, we employ a simple Gaussian smearing of atomic coordinates around their equilibrium positions, introducing a noise power of 0.1 Å. To maintain consistency with the integration of the equations of motion, we make our approach agnostic to atomic types by providing only the accelerations of the atoms as input to our model.

We define a multi-layer message passing architecture using E3 equivariant primitives from the `e3nn` library (Geiger & Smidt, 2022), where E3 refers to the three-dimensional Euclidean group that encompasses transformations such as rotations, translations, and reflections. This design enables the model to respect the inherent symmetries of the physical system. The input graph representation consists of atoms as nodes, with edges established between nodes if their interatomic distance is less than 5 Å. For each material, we sample multiple smeared atomic configurations and predict the MSD slope logarithm independently for each of them. The final slope prediction for each material is taken as the average over these samplings. The model is trained to minimize the mean squared error (MSE) loss. These architectural and training configuration choices were motivated by the need for a straightforward proof-of-concept implementation, acknowledging that this may limit the model's complexity and generalizability.

3 RESULTS

The architecture and training procedure described in Section 2 yield a model, referred to as the *baseline model*, that achieves the coefficient of determination value, R^2, of approximately 0.5 for the validation sample. While this R^2 value is relatively modest, it indicates that the model possesses predictive power. We present the learning curves and sample predictions for this model in Fig. 1.

One of the challenges that we face is that there is no guarantee that the model extracts the relevant information from the force field rather than from the atomic environment. In fact, we observe that the same architecture and training procedure lead to a comparable level of model performance when substituting the true atomic accelerations in the node features with random vectors. This can be addressed by restricting the model, which comes at the cost of reduced prediction quality. Namely, reducing the hidden representation size by a factor of $3/5$ and increasing the number of samplings per single prediction by a factor of 5 allows us to obtain a positive R^2 value of approximately 0.25 with SevenNet accelerations, while that value with randomized accelerations is just 0.06. We demonstrate the learning curves and predictions for these restricted models in Fig. 2.

We observe in our exploration, that sampling strategy plays an important role in achieving better performance. A sampling technique that we try, as an alternative to the Gaussian smearing described in Section 2, is to take the structural configurations from an actual MD trajectory. Obviously, if we were able to sample the same set of structural configurations that appear during an MD simulation, we could make an ideal prediction by simply calculating MSD analytically. We note, however, that using force-field evaluations from even a few frames of an actual MD trajectory results in better performance. For our best such model, referred to as the *trajectory-based model*, we achieve a validation R^2 value of 0.72.

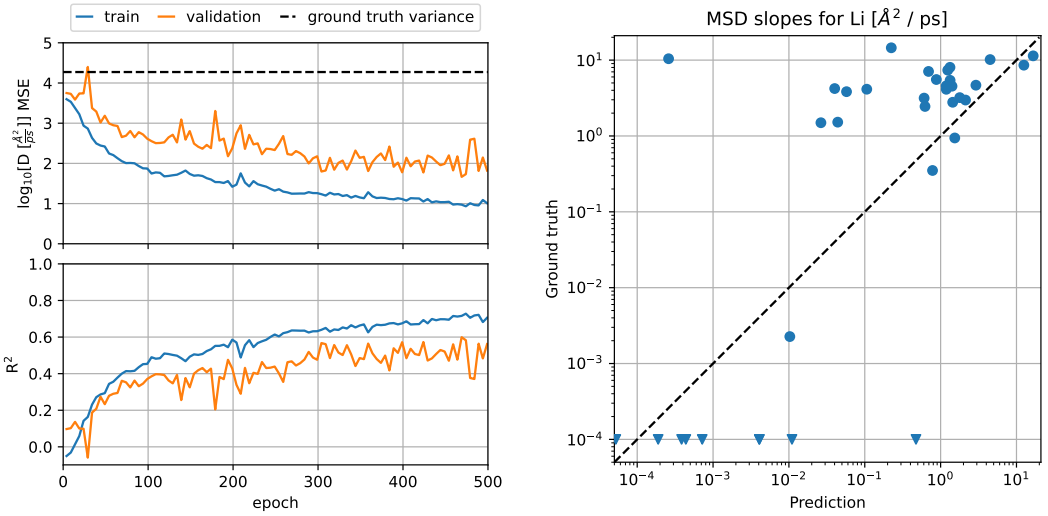
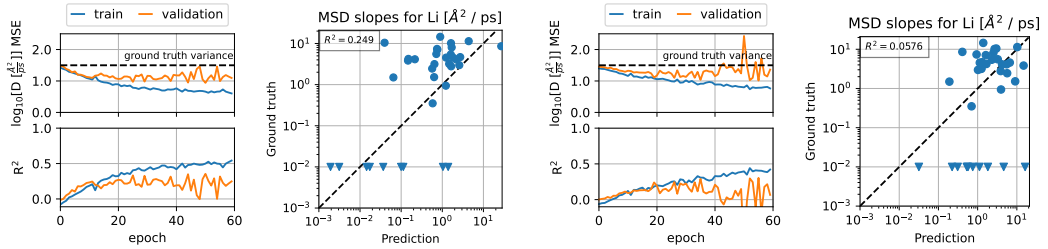


Figure 1: (Left) Learning curves for the baseline model displaying the MSE loss (top panel) and R^2 metric (bottom panel) as functions of epoch number, with training and validation values indicated. (Right) Scatter plot of predicted versus ground truth MSD slope values for the baseline model, where ground truth values clipped from below at 10^{-4} Å^2 / ps are represented by triangles. The dashed line indicates the line of equality ($X = Y$) for reference.



(a) SevenNet accelerations

(b) Random accelerations

Figure 2: Learning curves and prediction versus ground truth label scatter plots for the restricted models (see main text) with (a) SevenNet accelerations and (b) random accelerations used as node features. For each subplot, left panels contain the MSE loss (top) and R^2 metric (bottom) as a function of epoch number, while right panels show the scatter plots of predicted versus ground truth MSD slope values. The ground truth MSD slopes are clipped from below at 10^{-2} Å^2 / ps in these experiments.

4 Discussion and Summary

Our model achieves a coefficient of determination value, R^2, of approximately 0.5 for the validation sample, indicating its predictive power despite the modest performance. We hypothesize that improving this result involves further tuning of the architecture. However, a major challenge in improving this result and achieving generalizability to arbitrary potentials lies in ensuring that the model extracts relevant information from the force field rather than relying solely on atomic environment and structural information. We observe that our best architecture is capable of achieving a comparable level of performance even when random vectors are assigned to the graph nodes instead of true atomic accelerations. We can mitigate this by restricting the model, but it comes at the cost of reduced prediction quality. Overall, this indicates that, to improve the model's robustness, a larger dataset is required and, possibly, a more suitable inductive bias for the architecture needs to be identified.

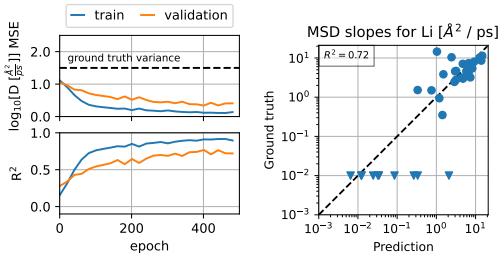


Figure 3: (Left) Learning curves for the trajectory-based model displaying the MSE loss (top panel) and R^2 metric (bottom panel) as functions of epoch number, with training and validation values indicated. (Right) Scatter plot of predicted versus ground truth MSD slope values for the trajectory-based model, where ground truth values clipped from below at 10^{-2} Å^2 / ps are represented by triangles. The dashed line indicates the line of equality ($X = Y$) for reference.

In our exploration, we also find that using structural configurations from actual MD trajectories, rather than Gaussian smearing, improves performance, achieving a validation R^2 value of 0.72. This suggests that a more sophisticated sampling strategy could further enhance predictive power. Alternatively, a hybrid setting could be implemented where a short true MD trajectory is used to make the diffusivity prediction.

Building on these findings, our results demonstrate the feasibility of connecting micro-scale interatomic potential description to macro-scale material properties like diffusivity. Once fully developed, this approach can facilitate a more efficient search for novel solid-state battery materials by providing insights that help identify and optimize materials with improved ionic conductivity.

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(Will be added after unblinding.)

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