Temperature-transferable coarse-graining of ionic liquids with dual graph convolutional neural networks

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Computer simulations can provide mechanistic insight into ionic liquids (ILs) and predict the properties of experimentally unrealized ion combinations. However, ILs suffer from a particularly large disparity in the time scales of atomistic and ensemble motion. Coarse-grained models are therefore used in place of costly atomistic simulations, allowing simulation of longer time scales and larger systems. Nevertheless, constructing the many-body potential of mean force that defines the structure and dynamics of a coarse-grained system can be complicated and computationally intensive. Machine learning shows great promise for the key coupled challenges of dimensionality reduction and learning the potential of mean force. To improve the coarse-graining of ILs, we present a neural network model trained on all-atom classical molecular dynamics simulations. The potential of mean force is expressed as two jointly-trained neural network interatomic potentials that learn the coupled short-range and the many-body long range molecular interactions. These interatomic potentials treat temperature as an explicit input variable to capture the temperature dependence of the potential of mean force. The model reproduces structural quantities with high fidelity, outperforms the temperature-independent baseline at capturing dynamics, generalizes to unseen temperatures, and incurs low simulation cost.

I. INTRODUCTION

Ionic liquids (ILs) are room temperature liquids composed of a molecular cation and anion. ILs have a wide variety of applications, including ion transport in batteries and catalysts.¹ ² Because of the wide chemical diversity of molecular anions and cations, and their many possible combinations, a vast design space for ILs exists. Molecular dynamics simulations can characterize the structure and dynamics of ILs at the nanoscale, and, to some degree, predict their properties before experimental realization. However, due to the complex intermolecular forces in ILs, classical force fields need to be fine-tuned to recover the correct kinetics and structure.³ ⁴ The properties of ILs are governed by a variety of interactive forces, including weak and isotropic forces such as van der Waals, solvophobic and dispersion forces,⁵ ⁶ strong Coulombic forces, anisotropic hydrogen bonding,⁷ halogen bonding,⁸ and magnetic dipole, electron pair and dipole-dipole interactions.⁹ Further complicating simulation is the sluggish motion of the ensemble. This leads to a large gap in the time scales of atomistic and ensemble motion, limiting the size of the molecular system and the time scales that can be accessed through simulation.

Coarse-graining (CG) can increase accessible simulation sizes by reducing the dimensionality of the full atomistic model to a pseudoparticle representation.¹⁰ ¹¹ A coarse-grained model can be described by a potential energy surface, the many-body potential of mean force (PMF). The PMF can be seen as the configurational free energy in a reduced phase space and is determined by the all-atom potential and the mapping operator from a full to a coarse-grained resolution.¹²

However, constructing an accurate many-body PMF that can faithfully capture the structure, thermodynamics and kinetics of the coarse-grained system is not an easy task. This is because reducing the representation and averaging out the fast atomic motions introduces complex many-body correlations in the PMF. Furthermore, the PMF is conditioned on the choice of mapping from the all-atom to the compressed model and on the temperature of the reference data. Typically, these mappings are chosen based on chemical intuition and trial-and-error, although there exist some methods for automatic and statistical mapping algorithms.¹³ ¹⁴

CG-IL models are particularly challenging because of the different transport properties of the two components, which further complicates the modeling of the many-body PMF.¹⁵ ¹⁶ Many authors have developed CG-IL models, and while each has achieved some success, none has been completely satisfactory. For example, Refs.¹⁷ ¹⁸ studied the 1-butyl-3-methylimidazolium cation with different anions. In one case, idealized models recovered the structural and energetic properties, but their transport properties were unreasonably slow.¹⁹ The model was improved upon by Merlet et al.²⁰, and the transport properties were recovered to a higher degree, but with a loss of structural properties.²¹ Wang et al.²² described an effective force coarse-graining applied to an IL system and recovered structural (RDF) properties over a range of temperatures, but they did not investigate dynamics. Refs.²³ ²⁴ described Newton Inversion and Iterative Boltzmann Inversion methods for coarse-graining ILs, and the thermodynamic properties

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were well recovered. However, the model underperformed with respect to self-diffusivity, both in proximity to experimental data and in temperature scaling. Moreover, ILS exist as liquids over a wide range of temperatures, and many of their desired applications involve temperature swings, but none of the CG approaches mentioned above are temperature transferable. Although energy renormalization has been applied for temperature-transferable CG focused on recovering structure of ILS, work on temperature transferability is lacking.

Each of the methods above achieved success in many, but not all, aspects of coarse graining. Moreover, each method required user expertise to create CG models that were not transferable to other ionic liquids. An alternative approach is to use machine learning (ML). ML approaches can achieve dimensionality reduction in a variety of domains and learn complex functions such as the CG many-body PMF. Neural coarse graining has been shown to recover ensemble quantities of liquid-phase alkanes with high accuracy, and without expert user input. Here we expand on this bottom-up approach to learn the encoding function and the associated PMF, with two key extensions. First, we condition the PMF on the temperature, achieving temperature-transferable dynamics at temperatures between two extremes. Second, in order to capture the complex many-body intermolecular forces acting on CG ILS, we utilize two separate neural network models to learn the coupled intra- and inter-molecular PMF. These two key features allow coarse-graining more complex condensed phase systems like ILS and offer a reliable, automated alternative to expert-based IL coarse graining.

FIG. 1. Model framework of the coarse-grained auto-encoder. a - The encoder-decoder architecture is optimized by minimizing the sum of the reconstruction and force regularization losses (Equation 1). b - To achieve discrete mapping, the computational graph is optimized using the Gumbel-softmax reparametrization, with weights randomly initiated as atom-wise vectors. c - Example of the training scheme with the convergence to an atom-wise mapping, the x-axis represents each heavy atom in the cation and the y-axis the contribution to each bead.

II. METHODS

A. Auto-encoder coarse-graining

The general schematic of neural network coarse graining is shown in Fig. 1. The model is an autoencoder, consisting of an encoder that maps the atomistic data to a coarse-grained latent representation, and a decoder maps the latent representation back to the atomistic representation. To train the model we minimize the autoencoder loss, $L_{AE}$, given by

$$L_{AE} = \frac{1}{N} \mathbb{E}_{x \sim P(x)}[(D(E(x, g, \tau)) - x)^2 + \rho F_{inst}(E(x))^2].$$

The first term in Eq. 1 is a reconstruction loss, where $D$ is the decoder mapping operator from the coarse-grained variables to the all atom representation, $E$ is the encoder mapping operator from the all atom representation to the coarse-grained beads, $x$ is the
atomistic ground truth coordinates, \( g \) is a Gumbel distribution sampled variable for reparametrizing the coarse-grained atom-wise weights and \( \tau \) is a temperature-like variable that decreases along training to achieve discrete encoding in the coarse-grained mapping \( E \). The second term is an instantaneous force regularization, obtained by applying the encoding function to the all-atom forces. The reconstruction loss statistically minimizes the loss of information that occurs when going through the latent space, and the force regularization is used to minimize the fluctuations in the mean force of the encoded space. This way a smoother energy landscape is achieved and more reasonable geometries can be obtained. A weight parameter \( \rho \) is introduced to adjust the balance between the two terms.

B. Temperature-dependent graph neural network potentials

Each mapping was used to fit a matching PMF through a graph convolutional neural network based on the SchNet model. In the SchNet model the geometry is transformed into a set of atomic fingerprints (the “convolution” component), and a fully connected network transforms the fingerprints into an output (the “readout” component). The convolution blocks consist of a message step and update step to systematically gather information from neighboring atoms. A neighbor is defined as an atom within a fixed distance cutoff (discussed further below). Defining \( v \) as index for each atom and its neighbors as \( N(v) \), the \( t \)th graph convolution updates the atomic feature vectors \( h_v \) by aggregating “messages” from their connected atoms \( v \) and their edge features \( e_{uv} \), through:

\[
h_v^t = h_v^{t-1} + \sum_{u \in N(v)} \text{message}^t(h_u, e_{uv})
\]

The initial feature vector \( h^0 \) is generated from a learnable embedding of atomic numbers. By performing the convolution operations several times, a many body correlation function can be parametrized to represent the potential energy surface of the ionic liquid complex. In the case of SchNet, the update function is simply a sum over the atomic embeddings. The message function is parameterized by

\[
\text{message}^t(e_{uv}, h_v) = MLP_3(MLP_1(e_{uv}) \circ MLP_1(h_v)),
\]

where MLP denotes a multi-layer perceptron and \( \circ \) is the concatenation operation. The readout consists of a sum over MLPs acting on each of the \( N \) atomic embedding at the final \( (T^{39}) \) convolution:

\[
\text{Energy} = \sum_v N \cdot MLP(h_v^T).
\]

Further details of the original model can be found in Ref. [3].

Figure 2 shows the neural network architecture for fitting a coarse-grained potential. The model is based on the SchNet architecture, but uses graph convolutions performed at two scales, those of the intra-molecular and inter-molecular graphs. We implemented graph convolution architectures for both scales with separate networks (as defined in Eq. (2)) to fit the energy and coarse-grained forces. The message passing step was performed to create two fingerprints for each CG bead. The fingerprint is summed and is then fed into a multi-layer perceptron for atomic energy parametrization. The purpose of this restrictive design was to differentiate the atomic energy after coarse-graining: the convolution procedure can differentiate its CG bead neighbor by their edge features \( e_{uv} \), through:

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h_v^t = h_v^{t-1} + \sum_{u \in N(v)} \text{message}^t(h_u, e_{uv})
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FIG. 2. Overview of the neural network architecture, which is based on the SchNet model\textsuperscript{28}. Instead of having a single interaction block, two are used to model the inter- and intramolecular interactions of the coarse-grained atoms. Temperature learning and classical priors in the forms of excluded volume and bond length are also used.

C. Stochastic force matching

The force-matching approach for parameterizing force fields was proposed in Ref.\textsuperscript{10} to reproduce structural correlation functions. Given an atomistic potential energy function $V(x)$ with the partition function $Z$, the probabilistic distribution of atomistic configurations is:

$$P(x) = \frac{1}{Z} e^{-\beta V(x)}.$$  \hfill (7)

The distribution function of coarse-grained variables $P_{CG}(z)$ and the corresponding many-body potential of mean force $A(z)$ is the log-likelihood of the distribution of coarse-grained variables. It is a free energy obtained from marginalizing over micro-states that are mapped to the coarse-grained coordinates,

$$A(z) = -\frac{1}{\beta} \ln P_{CG}(z).$$  \hfill (8)

The mean force of the coarse-grained variables is the average of instantaneous forces conditioned on the mapping $b_x = z$:\textsuperscript{30,31}

$$-\frac{dA}{dz} = F(z) = \langle -b \nabla V(x) \rangle_{E(x)=z}$$  \hfill (9)

where $b$ corresponds to the mapping assignment from the all-atom to the coarse-grained representation. Force matching requires the minimization of the loss function:

$$\min_{\theta} L = \min_{\theta} \mathbb{E}[(F(z) + \nabla_z V_{CG}(z))^2]$$  \hfill (10)

where $\theta$ are the parameters in $V_{CG}$ and $\nabla V_{CG}$ represents the “coarse-grained forces” which can be obtained from automatic differentiation. As proposed in Zhang et al., if $\mathbb{E}_z[F_{inst}] = F(z)$ is assumed, the loss function can be reformulated as the following minimization target:

$$\min_{\theta} L_{inst} = \min_{\theta} \mathbb{E}[F_{inst}(z) + \nabla V_{CG}(z))^2].$$  \hfill (11)

This minimization target has also been used in several works with stochastic gradient descent\textsuperscript{15,26} instead of matching mean forces that need to be obtained from constrained dynamics, the model minimizes $L_{inst}$ with respect to $V_{CG}(z)$ and $E(x)$. This target provides a data-driven target for optimizing coarse-grained force fields.
III. RESULTS

The model was trained on molecular dynamics simulations performed with the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) engine\textsuperscript{32}, with a publicly available force field for ionic liquids\textsuperscript{3,4}. The simulation box was created with a size of 300 cation and anion combinations with PACKMOL\textsuperscript{33}. A 1 ns NPT ensemble with a Nosé-Hoover thermostat was run to equilibrate the box followed by a 10 ns simulation performed at different temperatures, each repeated 3 times with different randomly initiated starting velocities to obtain larger space sampling. A subsample of the dataset was used for training: out of the 30,000 timesteps, 6,000 were evenly sampled and used further. The model was then trained on data from 300, 350, 450 and 500 K simulations. New CG molecular dynamics simulations with the trained models were run at 300-500 K in 50 K increments, thus including experiments at 400 K, outside of the training data. The structure and dynamics of CG simulations compared with the all-atom ground truth.

In keeping with the convention in the field, we utilized 1 bead to represent the anion. The choice of number and mapping of beads to represent the cation is less settled. Here, we tested two representations of 1-butyl-3-methylimidazolium through either 2 or 4 pseudoparticles (CG3 and CG5 respectively, when counting anion bead).

Figure 1\textsuperscript{a} shows the mapping and average reconstruction of 1-butyl-3-methylimidazolium. As expected, the 4-bead model led to good reconstruction, since it captures the orientation of the alkyl chain. The assignment learned by the auto-encoder structure agrees with prior intuition: one bead for the side methyl group and two ring atoms, one bead for the two following carbon atoms in the ring, one bead for the other imidazolium nitrogen and two connected linear carbons (Fig. 6), and a final bead for the terminal ethyl group. Because the model only learns to reconstruct average positions, rotationally equivalent atoms were reconstructed near their center of gyration.

Figure 3 shows the radial distribution functions (RDF) of the CG5 model (4-bead cation plus tetrafluoroborate anion). The ground truth data is the result of applying the CG mapping to held out all-atom frames. The RDFs are in close agreement with the ground truth data. Both the cation-anion and anion-anion interactions have been reproduced to a high degree. Additionally, the cation-cation interactions and the positions of all peaks have been accurately recovered, though their intensities show higher deviations from the ground truth. The difference between the second and third cation-cation peaks further decrease at higher temperatures.

The RDF at 400 K shown in Fig. 3b shows that the model produces stable dynamics at temperatures outside the training set, and can recover structural properties of systems with similar accuracy as at reference temperatures. Bond distance distribution comparisons can be found in the Supplementary Information.

Transport properties such as self-diffusivity are very relevant to the use of ILs as ionic conductors, and effective CG simulations should recover dynamic properties to the best degree possible. Experiments have shown that the 1-butyl-3-methylimidazolium...
FIG. 4. Transport properties of the coarse-grained ionic liquid. a) Mean squared displacement (MSD) over 1 ns long range at 300 K. b) Diffusivity across a temperature range from 300 K to 500 K.

| Molecule | GT    | T-NFF | NFF   | CG3   |
|----------|-------|-------|-------|-------|
| Cation   | 20.7±1.5 | 14.3±1.6 | 10.4±0.6 | 9.4±0.7 |
| Anion    | 21.6±2.9 | 16.7±1.8 | 12.2±0.5 | 8.7±0.6 |

TABLE I. Activation energy for diffusivity of the ground truth and CG neural force field for the tested cases. T-NFF and NFF are with the 5 pseudoparticle representation, and CG3 is trained on the T-NFF model with 3 pseudoparticle representation.

The tetrafluoroborate ionic liquid exhibits the Arrhenius relationship\cite{17,18}. Figure 4a shows the mean-squared displacement calculated by the Einstein relation at 300 K:

\[
D_{a,\text{self}} = \frac{1}{2} \lim_{\tau \to \infty} \langle [r_{i,a}(t+\tau) - r_{i,a}(t)]^2 \rangle / \tau.
\]  

The evolution of CG T-NFF displacement is about an order of magnitude faster than the ground truth all-atom system. This is because in the CG model the small, fast movements of hydrogen atoms are averaged out, thus reducing the overall friction of the system and speeding up the dynamics. Fig. 4b shows the self-diffusivity across a temperature range from 300 K to 500 K, including out-of-sample simulations at 400 K. Both the ground truth and the T-NFF show similar relations between the cation and the anion. At lower temperatures, the cation has strictly faster dynamics than the anion, but at higher temperatures this difference shrinks. The T-NFF shows the same relation at lower temperatures, but at higher temperatures the difference shrinks faster than for the ground truth.

Figure 5b shows the Arrhenius-type relation of the coarse-grained anion and cation. The Pearson correlation coefficient between for the linear fit is -0.981 and -0.982 for cations and anions respectively. The cation and anion curves intersect in the high temperature range as in the ground truth simulation (Table I), although the intersection occurs at a higher temperature.

Figure 5 weighs the impact of the temperature embedding in the model by comparing the outcome of CG simulations from a temperature-independent model (NFF) with the temperature-dependent T-NFF results. Both models were trained on the same data from all-atom simulations at 300, 350, 450 and 500 K and have the same hyperparameters. Figure 5 shows the RDFs and Arrhenius relations between the ground truth and the models. The two models reproduce the structural properties to a similar degree and the Arrhenius relation of the system is similarly linear. However, as seen from the slopes in Fig. 5b and activation energies in Table I, the temperature-averaged NFF has not learned the temperature effect on dynamics as well as its counterpart and produces a much lower activation energy in addition to accelerated dynamics. It also underestimates the temperature at which cation and anion diffusivities cross.

Figure 6 compares the performance the CG3 model (2-bead cation) to the CG5 model. In Fig. 6a, it can be seen that both the 3 and 5 bead models recover structural properties at a temperature that has not been included in the training data. Nevertheless, significant differences are seen in the dynamical properties derived from the Arrhenius relation. Both the models have recovered the linear relation, including the 400K out-of-sample trajectory, but the CG3 model grossly underestimates the activation energy (Table I).
IV. DISCUSSION

We have introduced a novel extension of graph convolutional neural networks to learn many body PMFs by separating short-range intramolecular and longer-range many-body intermolecular terms, and incorporated a temperature transferable embedding to learn the temperature dependence. The model has been trained on all-atom simulations of an ionic liquid system, 1-butyl-3-methylimidazolium tetrafluoroborate, whose PMF is known to be difficult to construct. The reconstruction of the structural properties has been achieved to a high degree. Furthermore, the Arrhenius relation for self-diffusivity has been obtained with the neural force field, though with faster dynamics. The fast dynamics arise due to the averaging of fast motions in the system, which in turn potentially decrease the overall friction of the system. If dynamics cannot be reconstructed exactly, it is preferable that the activation energy for diffusion remain constant between all-atom to CG simulations, and a simple scaling of the dynamics is observed. Other strategies based on spectral matching or differentiable simulations could be utilized to further condition the encoding function and PMF to reproduce system dynamics.

Results from a single graph convolutional neural network with a single interaction block for both inter- and intramolecular interaction have not been included in this work for comparison with the novel dual graph convolutional neural network. This is because no stable simulations could be performed with the simpler model. No choice of hyperparameters led to a model that could run coarse-grained IL molecular dynamics simulations without the system collapsing on itself. This is likely because in a coarse-grained model the distances between the beads of a single molecule are on a much larger scale than those of atoms in the same molecule. These intramolecular distances are then closer to intermolecular distances, and as such it becomes harder for a single interaction layer to separate them and learn the structural and dynamics properties of the system. This highlights the key contribution of the dual graph convolutions introduced in this work.

A comparison in performance was made between the temperature dependent neural force field (T-NFF) and the neural force field without the temperature learning embedding layer (NFF) (see Fig. 5). Both models overestimated the dynamics at lower temperatures equally, but at higher temperatures NFF was significantly slower. The T-NFF showed acceleration of dynamics across the temperature range and as such reproduced the Arrhenius-like temperature dependence much better, largely scaling the overall kinetics by a constant factor at all temperatures. Interestingly, the T-NFF model faithfully captured the loss of linearity in the Arrhenius plot at the lowest temperature that is present in the all-atom simulation. Previous works on coarse-grained ILs,
such as Roy et al\textsuperscript{19} and Moradzadeh et al\textsuperscript{33}, heavily underestimated dynamics. The model by Roy et al. was improved to attain an agreement of diffusivity\textsuperscript{21} though at the cost of accuracy of structural properties.

Different cation representations were also compared. Both 3- and 5-bead models recovered the structural properties to a similar degree, though the 5 bead model led to better learning of the dynamic temperature relation, despite the overestimation of the speed of the self-diffusion in both cases. This reflects the fact that coarser representations lead to more averaging over small atomic movements, and therefore to faster dynamics.

The model described in this work can accurately learn the many-body PMF of a CG model for a complicated system, is accurate over a range of temperatures, requires little user input and can to be applied to other condensed-phase systems, leading to easier, more accurate, and more generalizable CG simulations of complex systems.

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**DATA AND CODE AVAILABILITY**

The data that support the findings of this study are available from the corresponding author upon reasonable request. The computer code use to train the models is available at https://github.com/learningmatter-mit

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SUPPLEMENTARY INFORMATION

Figure 7 shows the learned bond distributions of the temperature transferable graph convolutional neural network at 300 K. The model has properly learned the complex multivariate distributions of the interatomic distances to a high degree. Figure 8 shows the RDFs across a temperature range for the 2 pseudoparticle representation of the cation as well as the recovered bond distribution. It can be seen from Fig. 8 that the model has performed reasonably well in recovering the structural properties at 400 K even when this temperature has not been included in the training data.
FIG. 8. Radial distribution functions and bond distributions of the ionic liquid system with 300 cation and anion molecules, where the cation is represented with 2 pseudoparticles. a) At 300 K, b) at 400 K, and c) at 500 K. d) Recovered bond distribution at 300 K of the 2 bead cation representation.