Multiscale Simulations of Ionic Liquids With Deep Neural Networks

Jurgis Ruza[1,2]

Supervised by Prof. Rafael Gómez-Bombarelli[1] and Prof. Michele Ceriotti[2]

1 – Masachusetts Institute of Technology
2 - École polytechnique fédérale de Lausanne

Motivation

- Coarse-grained models provide the capability of performing longer simulations for a fraction of the cost
- Determining the best pseudo-particle mapping remains an unsolved and important issue
- Constructing the many body potential of mean force (PMF) is time consuming and complicated

Solution

- Using an auto-encoder structure [1] to learn the statistically most viable coarse-grained atom mapping of 1-Butyl-3-methylimidazolium tetrafluoroborate – a common ionic liquid (Figure 1)
- Learning the many body PMF with a novel double graph convolutional neural network – neural force field (NFF) with data from molecular dynamics simulations

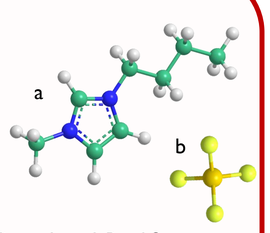

Figure 1, a - 1-Butyl-3-methylimidazolium (cation)
b - tetrafluoroborate (anion)

Coarse grained mapping

- Variational auto-encoder based generative model for constructing a coarse-grained mapping (Figure 2) [1]
- Trained to optimize a reconstruction loss from the CG mapping to the ground truth

$$min_{D,\phi} L_{AE} = min_{D,\phi} \mathbb{E}_{x\sim P(x)} \mathbb{E}_{g\sim Gumbel(0,1)} (D(E(x,g,\tau)) - x)^2$$

- As well as the instantaneous mean force is minimized to regularize the coarse-grained space

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

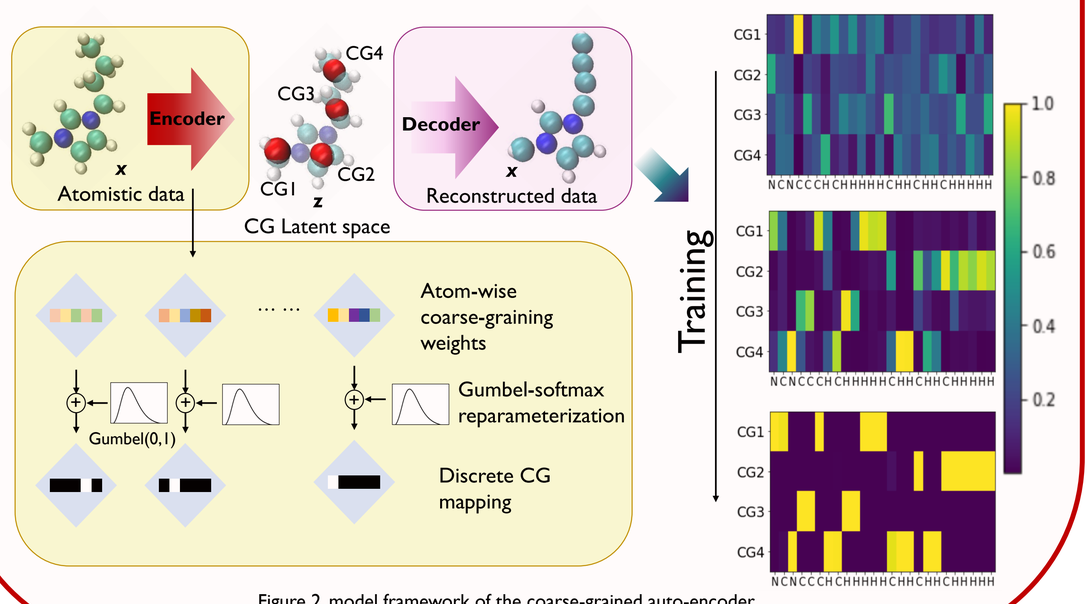

Figure 2, model framework of the coarse-grained auto-encoder

Graph Convolutional Neural Network - Hybridgraph

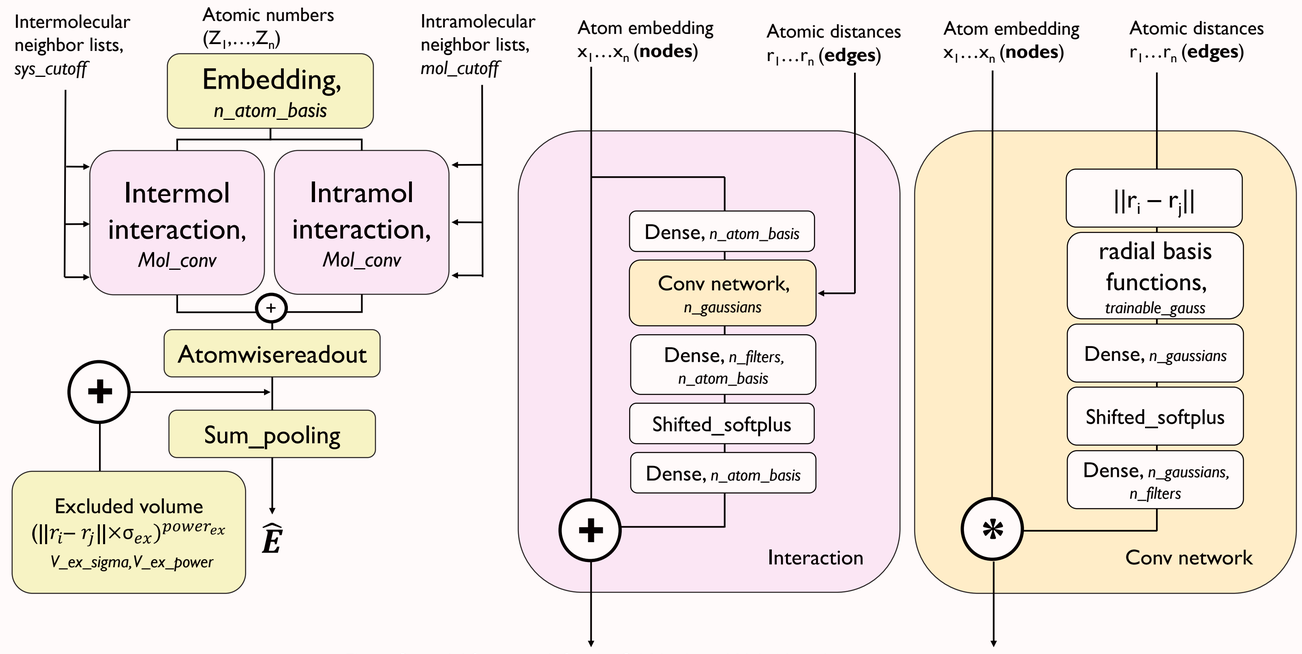

Figure 3, model framework for the SchNet based Hybridgraph

- Based on the continuous-filter convolutional neural network model SchNet [2]
- Coarse-grained hidden many body correlations are harder to learn
- **Two interaction** blocks are introduced to model the many body PMF (Figure 3):
  - **Intermolecular** and **intramolecular** interactions learned separately
- The neural network is trained by minimizing the MSE to the instantaneous mean forces with intermolecular and intramolecular distances as training data
- Classical prior in the form of excluded volume is introduced for better results

Results

- Training the variational autoencoder on a 4 pseudoparticle representation provided the mapping in Figure 4
- After a hyperparameter optimization, molecular dynamics simulation was run and characteristic properties were compared with the ground truth
- Radial distribution functions of different interactions showed well learned structural properties (Figure 5)
- Self-diffusivity across a temperature range (Figure 6) used as rate constant *k*, to plot the Arrhenius relation

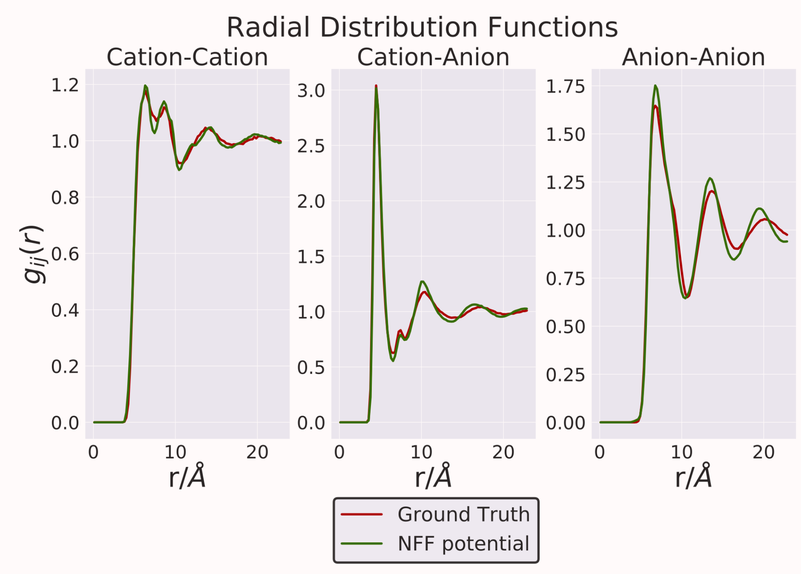

Figure 5, RDF of the coarse-grained model propagated with the hybdridgraph neural force field, compared with the ground truth,

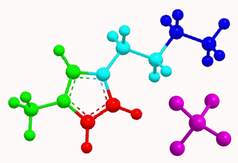

Figure 4, auto-encoder determined mapping of the cation and anion molecule

$$\ln k = \ln \frac{-E_a}{R}\left(\frac{1}{T}\right) + \ln A$$

$$E_a = slope\ of\ curve * (-R)$$

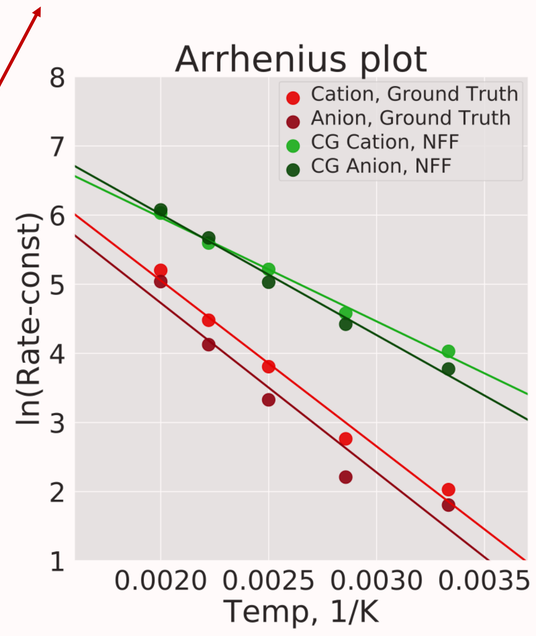

Figure 6, Arrhenius plot of the ground truth and neural force field across a temperature range, with the rate constant as self-diffusivity

- In the temperature range both relationships are linear – NFF has learned the Arrhenius relation
- Dynamics are faster as the all atom friction is hidden
- Activation energy for the NFF is considerably lower – dynamics slower at higher temperatures

	E_a, kJ mol^{-1}
Ground truth, Cation	**19.95**
NFF, Cation	**12.49**

Table 1, Activation energy of self-diffusivity

Conclusions

- Successfully learned an alternative and innovative coarse grained mapping to a common ionic liquid [3]
- Using a novel approach for modeling inter and intramolecular interactions the model reproduced ground truth structural properties to a high degree
- As expected dynamic properties were faster than ground truth, which is taken as a welcome view
  - Gives the possibility of larger phase space sampling in less time

Outlooks

- Developing a temperature transferable model, that can recover structural and dynamic properties across a temperature range
- Integrating the coarse-grained reconstruction, to fully recover the dynamic properties at long intervals with more thoroughly sampled phase space

References

[1] WujieWang and Rafael Gómez-Bombarelli. Coarse-graining auto-encoders for molecular dynamics. npj ComputationalMaterials, 5(1):1–9, 12 2019.
[2] K. T. Schütt, H. E. Sauceda, P. J. Kindermans, A. Tkatchenko, and K. R. Müller. SchNet - A deep learning architecture for molecules and materials. Journal of Chemical Physics, 148(24):241722, 6 2018.
[3] Durba Roy, Nikhil Patel, Sean Conte, and Mark Maroncelli. Dynamics in an idealized ionic liquid model. Journal of Physical Chemistry B, 114(25):8410–8424, 7 2010.