Force Field Development for Molecular Simulations with Structure Optimized Potential Refinement

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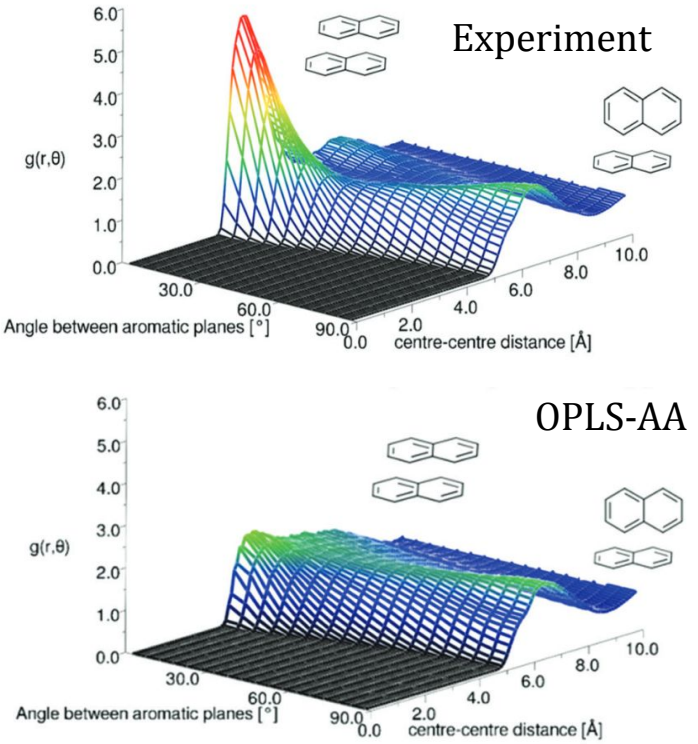
Recent Advances in Molecular Simulation Methods

Force Fields in Classical Molecular Dynamics

- Classical molecular simulation is ubiquitous across chemical physics.
- It is well established that classical methods overfit to the experimental data the force field is trained on.
- From statistical mechanics, we know that we can recover all thermodynamic properties of a system if we know both the structure and potential energy.

Poor Structure Prediction in Napthalene

Headen, 2019 *Phys. Chem. Chem. Phys.*



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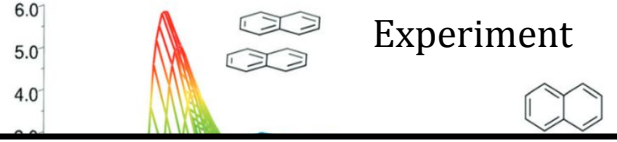
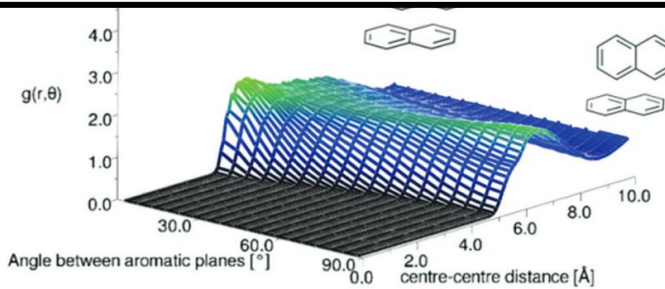
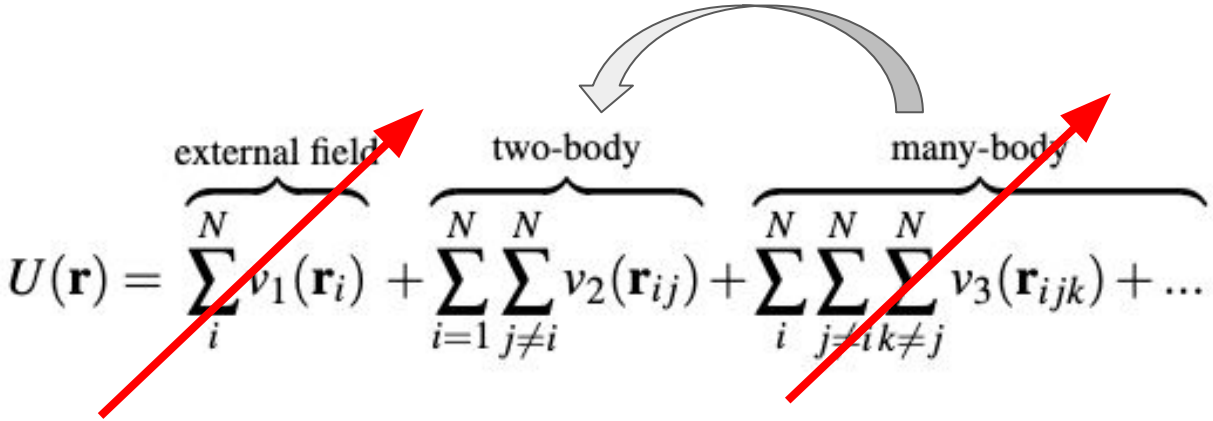


What is preventing classical molecular models from simultaneously predicting structural and thermodynamic properties?



Assumptions in classical force fields limit their accuracy

Pairwise Additive Approximation



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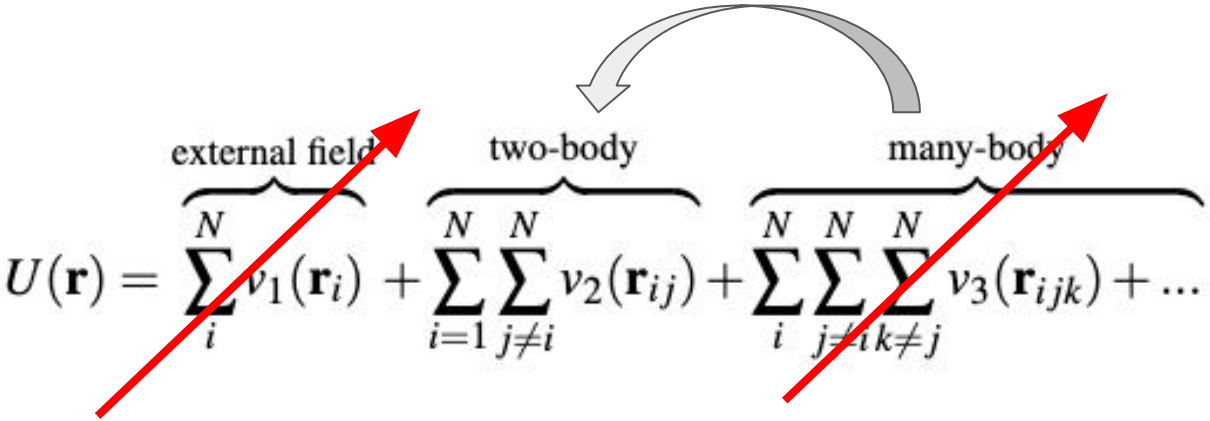

Effective pair potentials are fit to empirical forms

Mie Intermolecular Potential

$$v_2^{\mathrm{Mie}}(r) = \frac{\lambda}{\lambda - 6}\left(\frac{\lambda}{6}\right)^{6/\lambda - 6} \varepsilon \left[\left(\frac{\sigma}{r}\right)^{\lambda} - \left(\frac{\sigma}{r}\right)^{6}\right]$$

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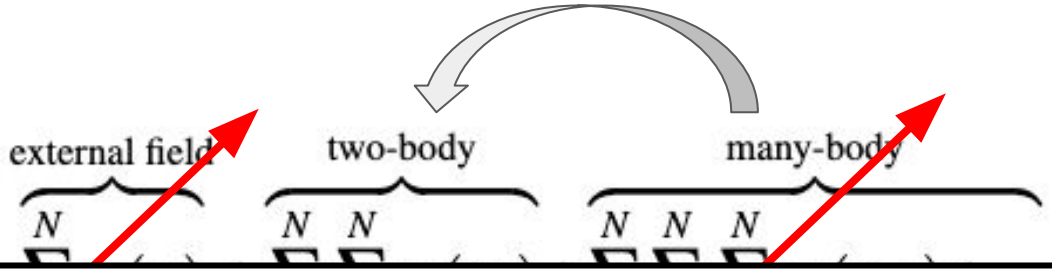

Pairwise Additive

Can we model the potential energy and structure "accurately enough" to recover fluid thermodynamics from a purely classical model?

Effective pair potentials are fit to empirical forms

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The Henderson Inverse Theorem and Iterative Boltzmann Inversion

Henderson's Theorem

If two systems are described by a pairwise additive Hamiltonian and have the same RDF, then their pair interaction term differs at most by a trivial constant.

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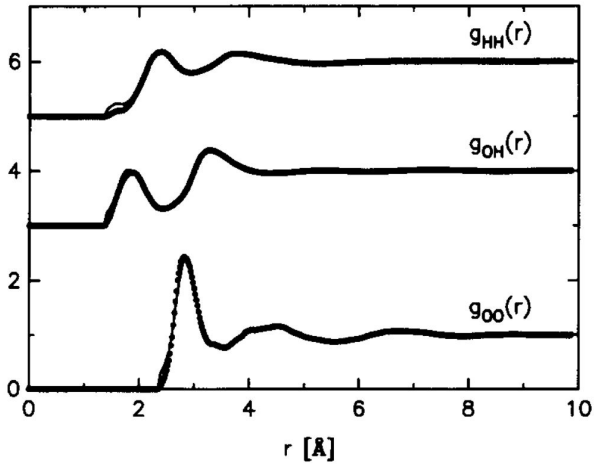
Neutron Scattering Analysis

Soper, 1996 *Chem. Phys.*



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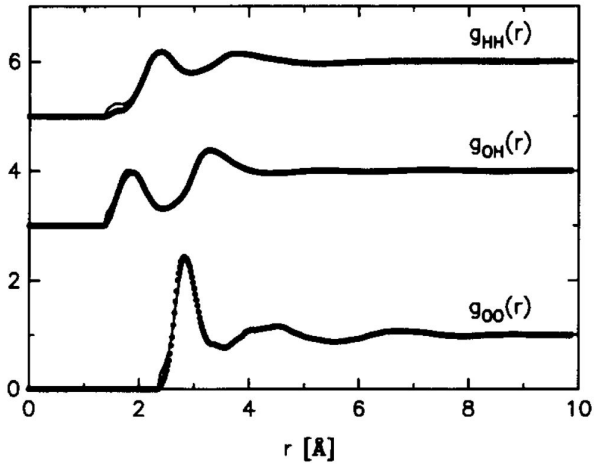
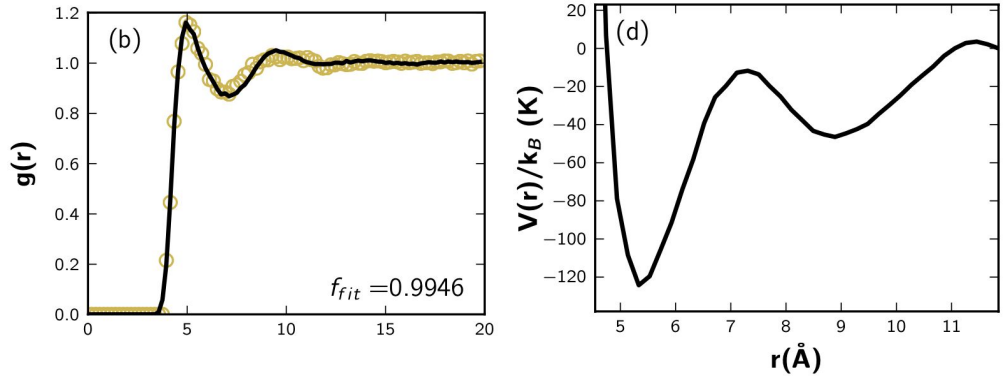


Coarse-Graining with Iterative Boltzmann Inversion
Moore, 2014 *J. Chem. Phys.*



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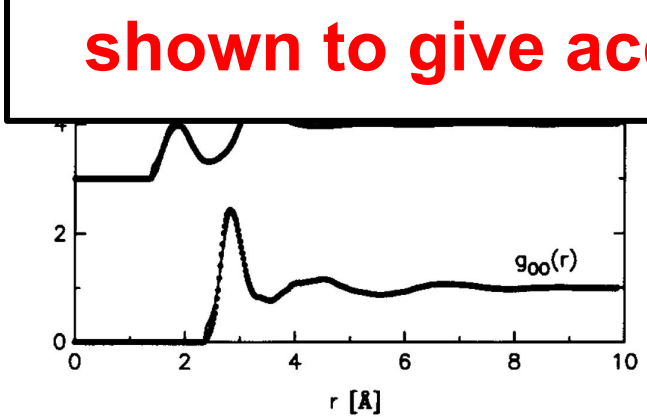
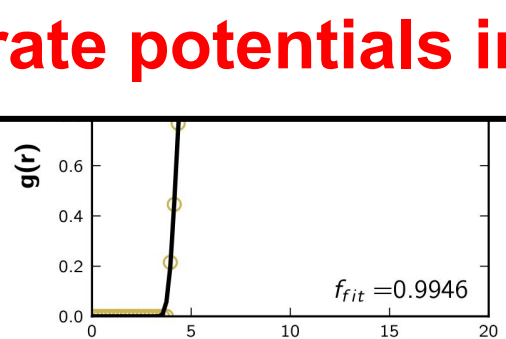
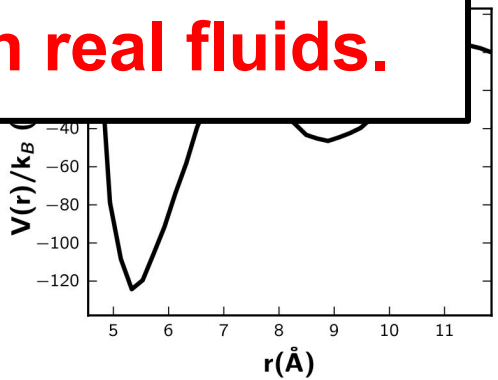
Neutron scattering analysis methods (EPSR) and Iterative Boltzmann Inversion have not been shown to give accurate potentials in real fluids.

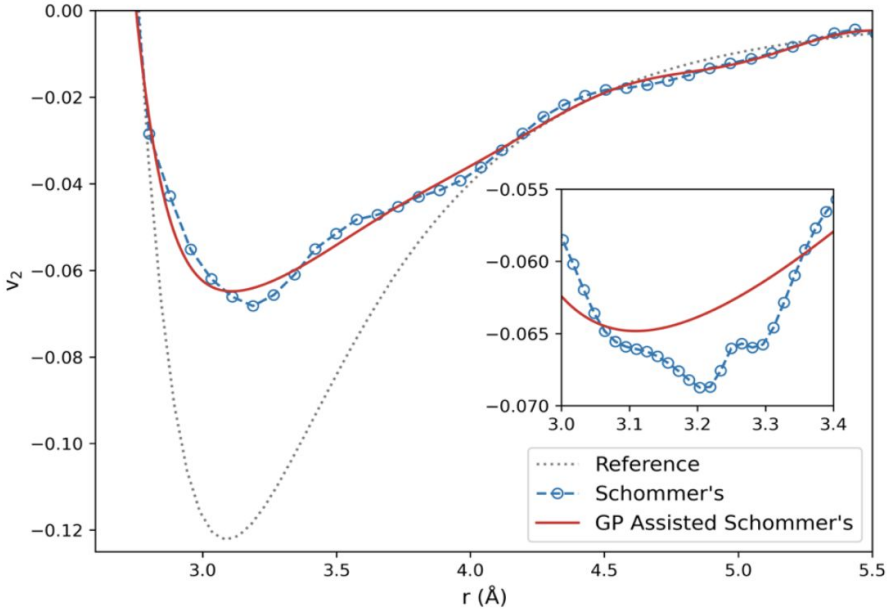






Structure Optimized Potential Refinement - A Physics-Guided, Probabilistic Iterative Boltzmann Inversion

Probabilistic Regression w/ GPs



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B. L. Shanks 2022, *J. Phys. Chem. Lett.*

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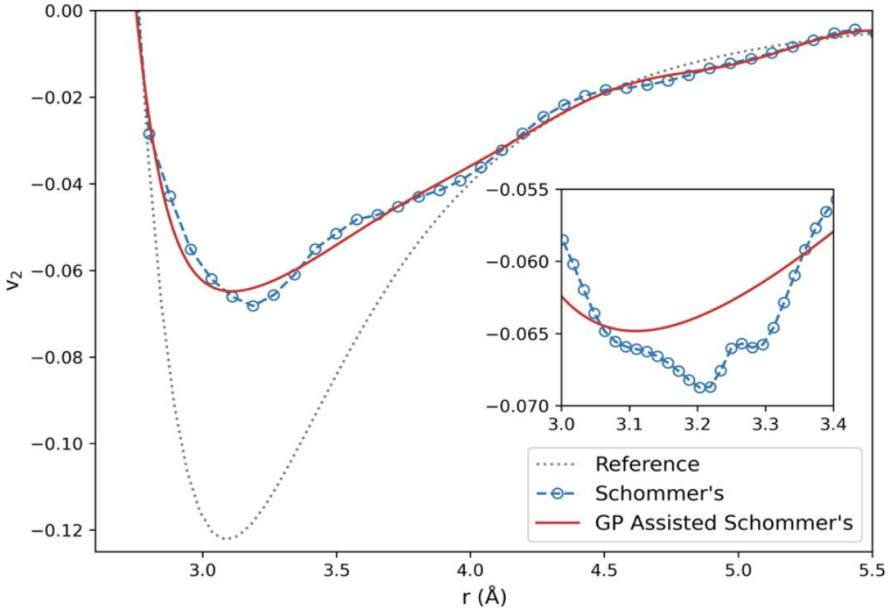
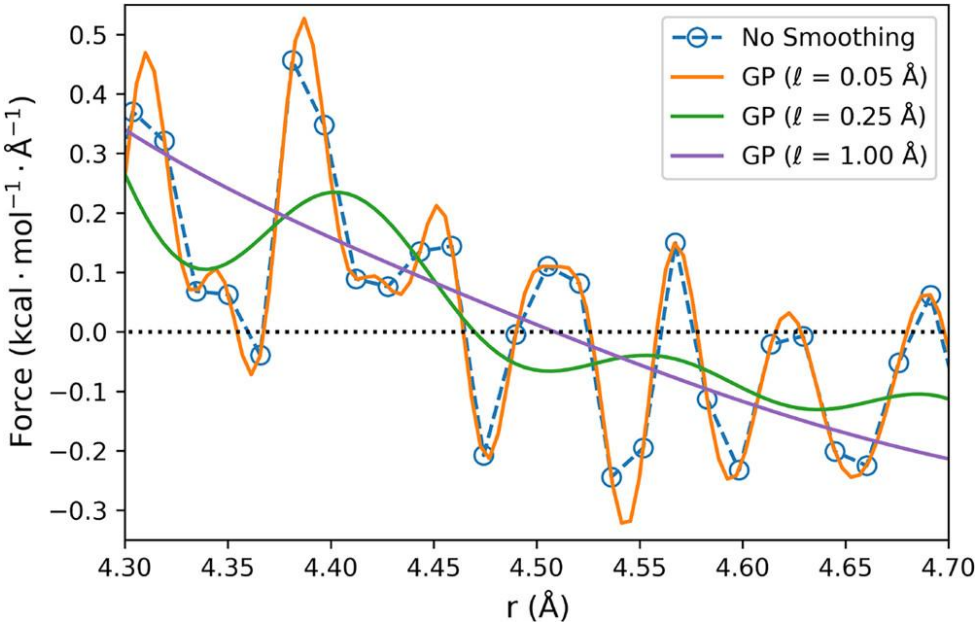


GPs promote functional stability



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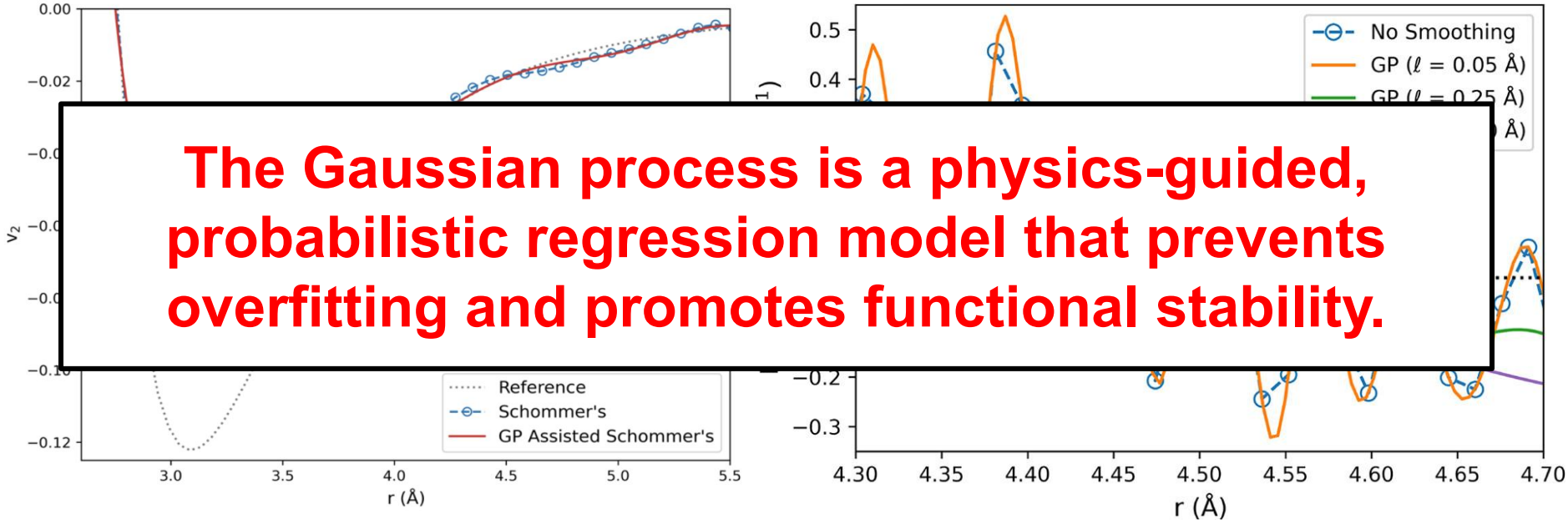
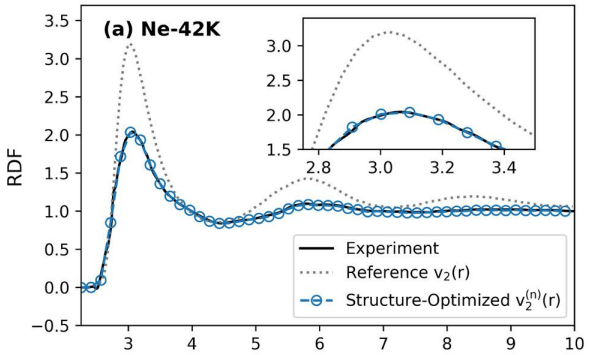


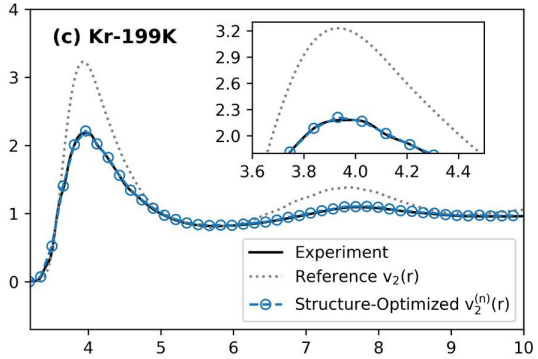
The Gaussian process is a physics-guided, probabilistic regression model that prevents overfitting and promotes functional stability.

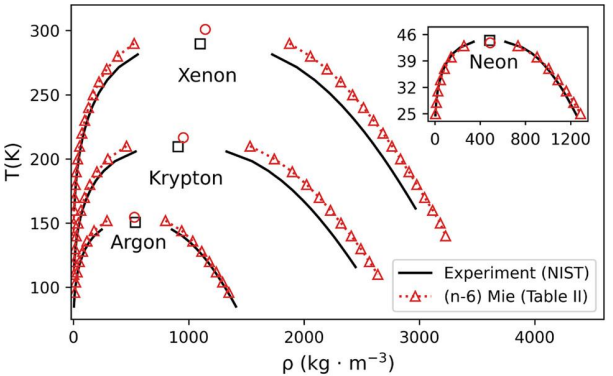
Structure Optimized Potential Refinement
B. L. Shanks 2022, *J. Phys. Chem. Lett.*

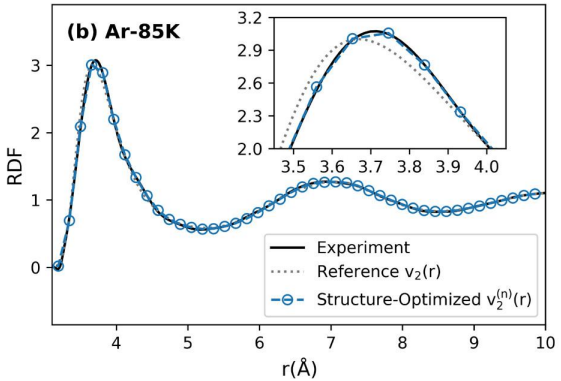
Noble Gas Force Fields from Scattering Data

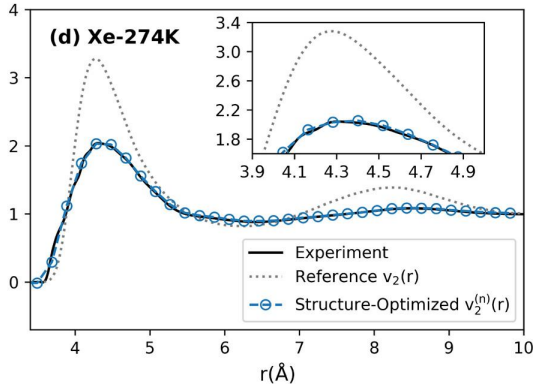
Radial Distribution Functions and Vapor Liquid Equilibrium Match with Excellent Agreement

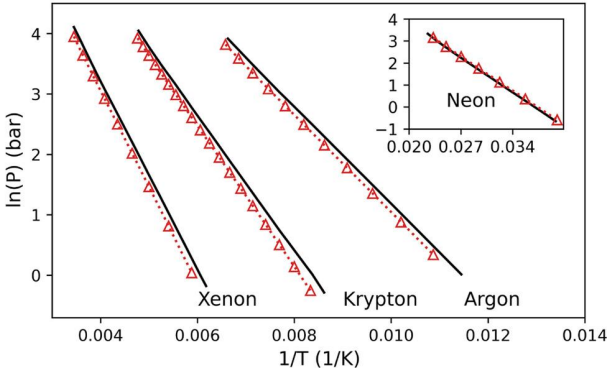












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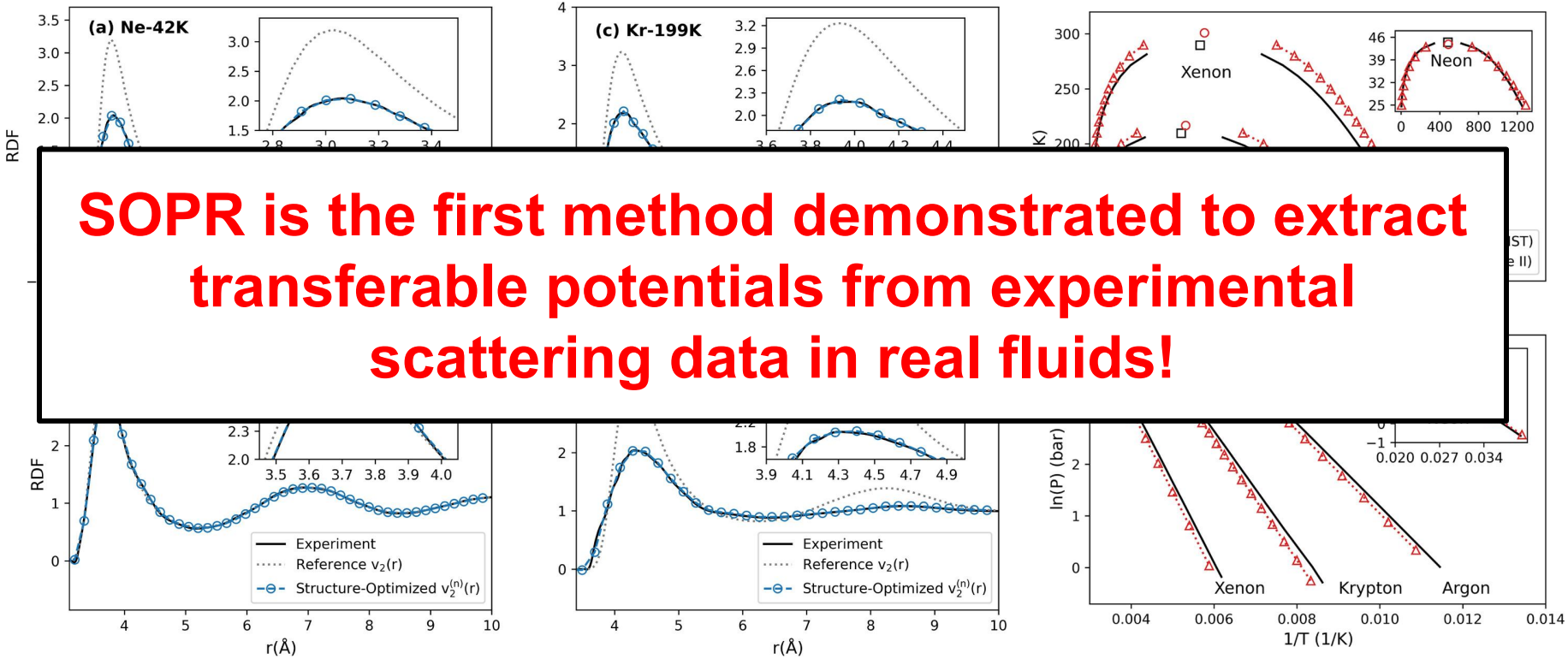
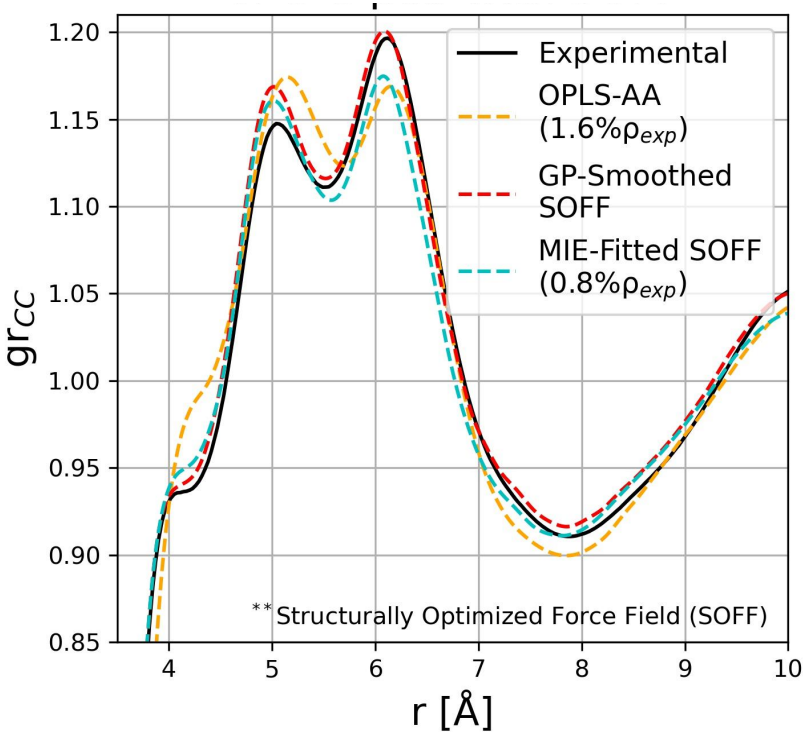
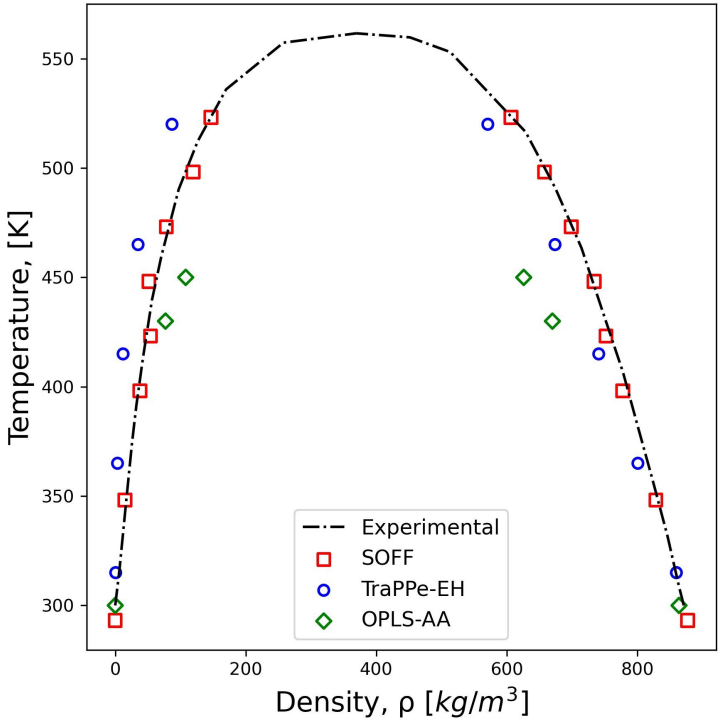


SOPR is the first method demonstrated to extract transferable potentials from experimental scattering data in real fluids!

Current and Future Work - Extending SOPR Beyond Simple Liquids

Excellent RDF + VLE Agreement for Water, Benzene and Methane





Abdur Shazed

Harry Sullivan

Summary and Key Takeaways

- SOPR is the first validated method to determine transferable potentials from experimental scattering data.
- Excellent performance on structure and vapor-equilibria in single atom and molecular liquids.
- Probabilistic machine learning techniques continue to shine for complex and indeterministic physical modeling.