Learning Interatomic Forces from Experimental Measurements of Fluid Structure

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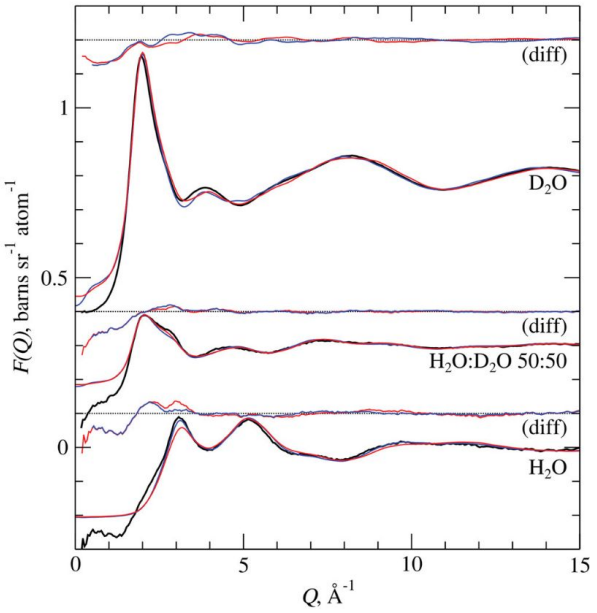
Abdur R. Shazed
Michael P. Hoepfner
University of Utah, Department of Chemical Engineering

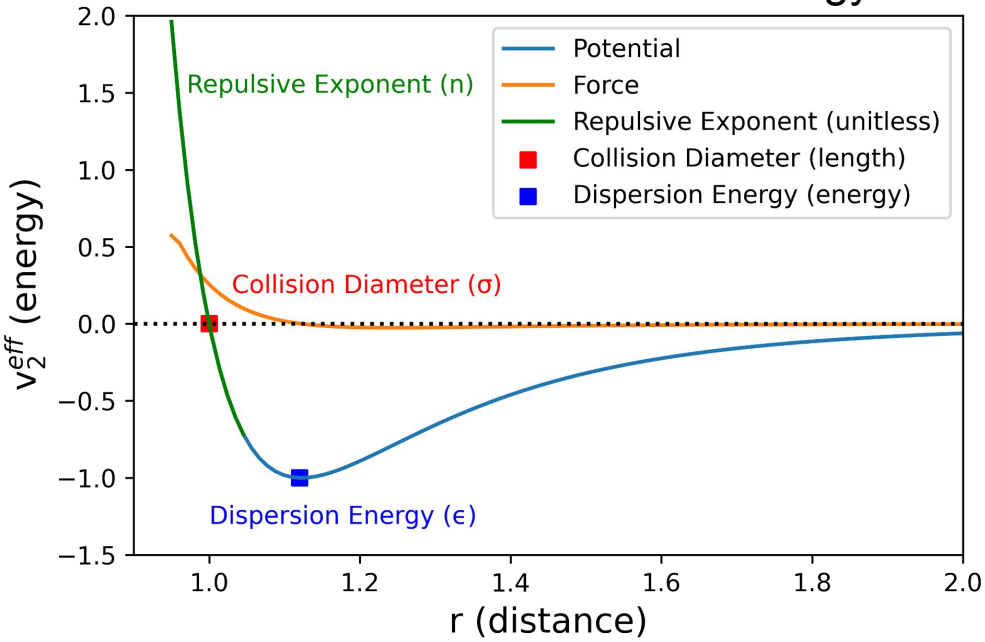
AIChE Machine Learning for Soft and Hard Materials II

Predicting Intermolecular Potentials from Scattering is a Historic Inverse Problem

Neutron/X-ray Scattering



Intermolecular Potential Energy

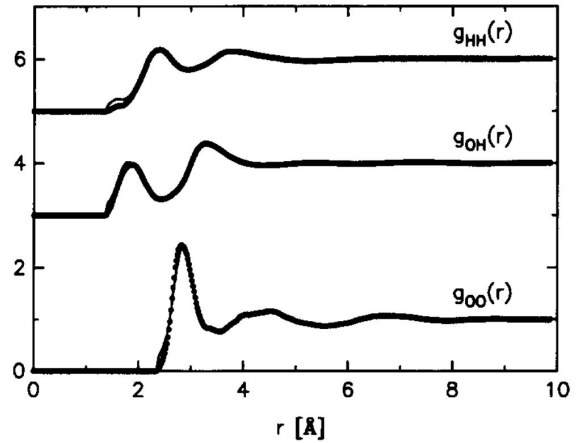


If the structure and potential energy of an ensemble are known, we can (in principle) recover thermodynamic properties of a system through statistical mechanics.

Applications of Inverse Problems for Interesting Chemistry

Scattering Analysis for Ill-Posed Structure Prediction

A. K. Soper 1996, *Chem.*



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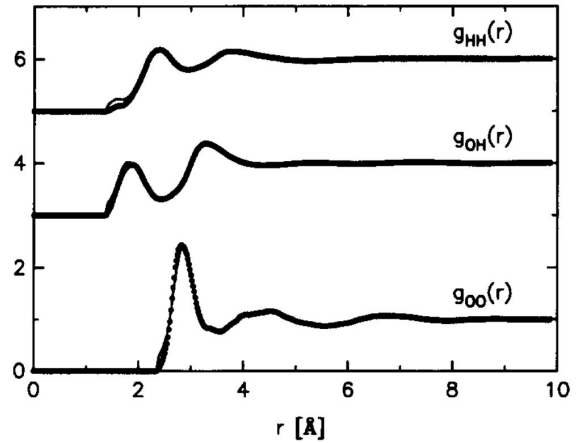

Coarse-Graining

Shell et al. 2008,

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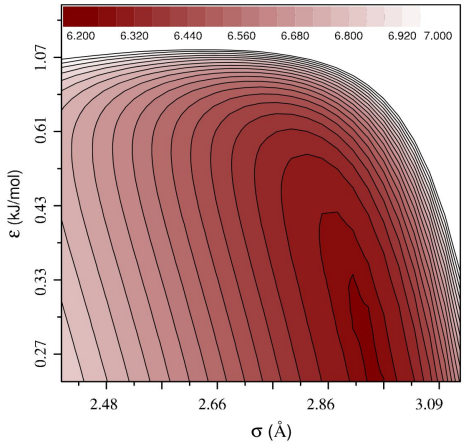

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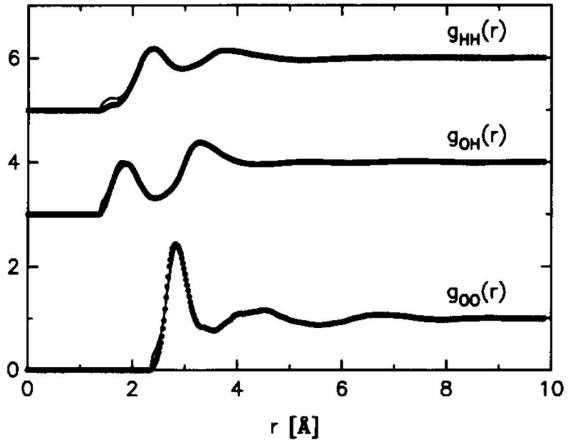

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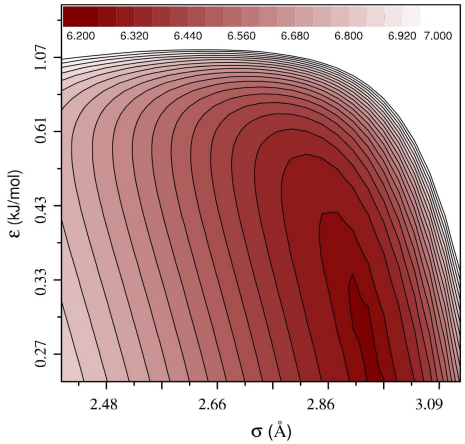
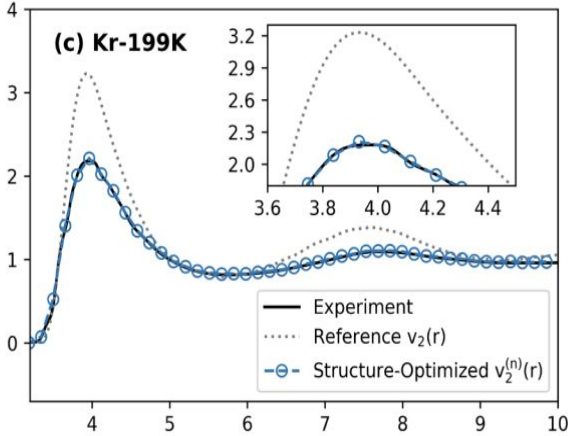


Force Field Development: Structure Optimized Potential Refinement
B. L. Shanks 2022, *J. Phys. Chem. Lett.*



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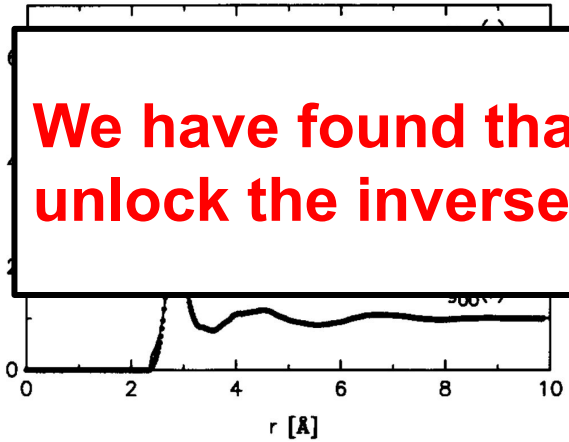

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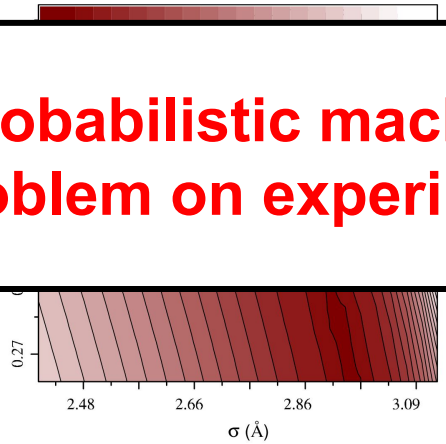

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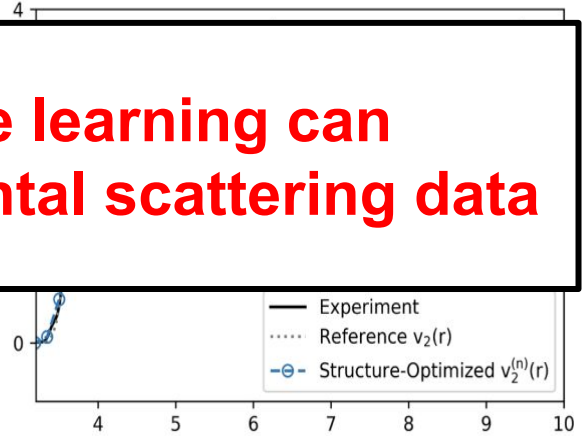
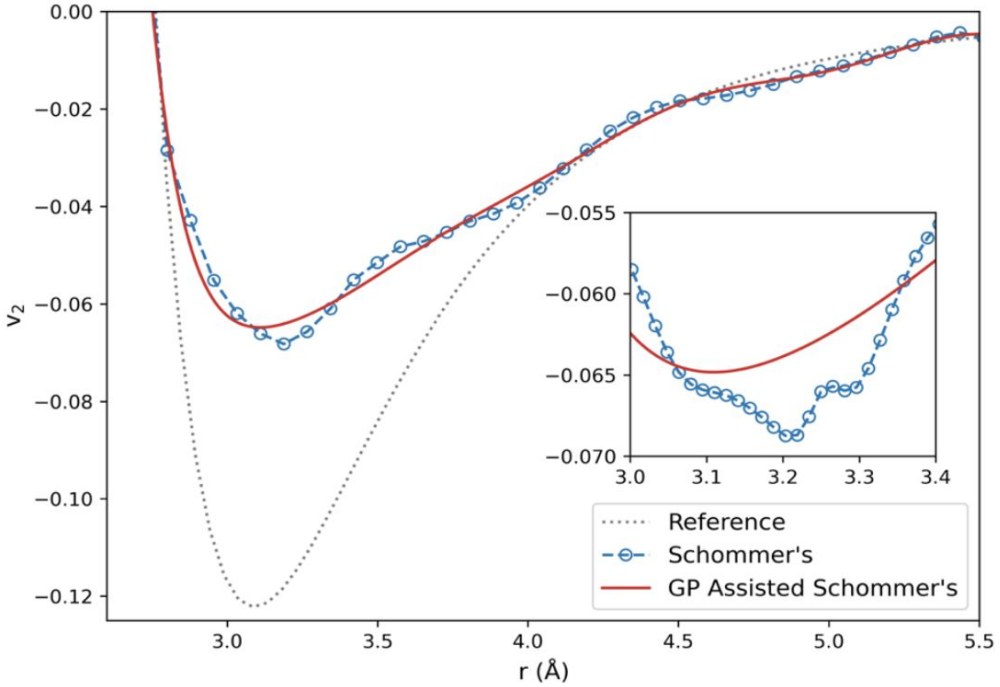


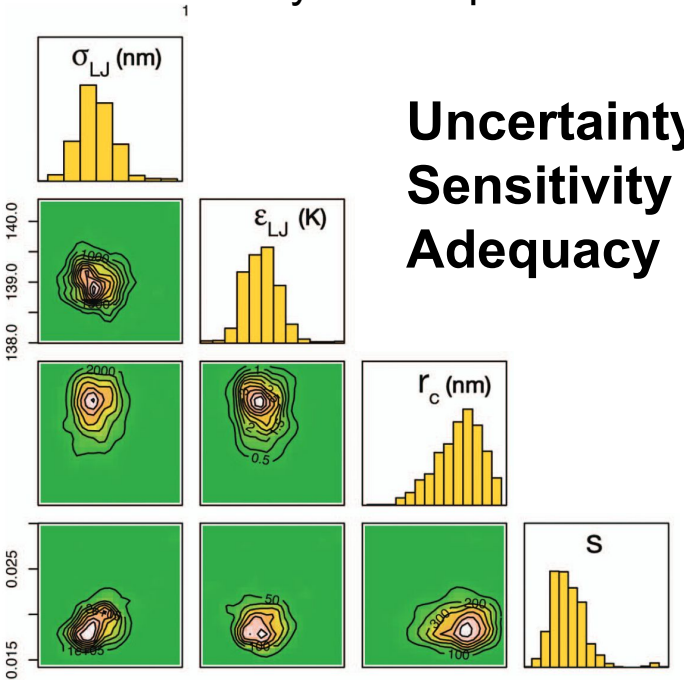
We have found that probabilistic machine learning can unlock the inverse problem on experimental scattering data

Probabilistic ML Approaches to the Structure Inverse Problem

Non-Parametric Approaches



Parametric Bayesian Optimization



Uncertainty
Sensitivity
Adequacy

Marginal Posteriors on LJ Parameters
Koumoutsakos 2012, *J. Chem. Phys.*

Accelerated Bayesian Inference with Gaussian Process Surrogates

Evaluating the Bayesian probability distributions is easy! Just run 100,000+ molecular simulations to populate the model parameter space and you're done!

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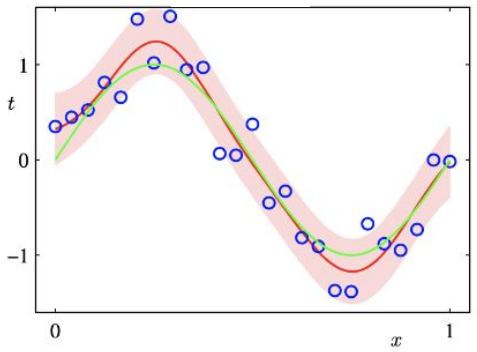
Instead, we train a GP surrogate model on properties from MD trajectories.

~ O(eta^3) scaling

$$GP(\boldsymbol{\theta}^*)$$



$$(N\eta \text{ x } \dim(\boldsymbol{\theta}) + 1)$$

$$\hat{\mathbf{X}} = \begin{bmatrix} \theta_{1,1} & \theta_{2,1} & \dots & r_1 \\ \theta_{1,1} & \theta_{2,1} & \dots & r_2 \\ \vdots & \vdots & \vdots & \vdots \\ \theta_{1,1} & \theta_{2,1} & \dots & r_\eta \\ \theta_{1,2} & \theta_{2,2} & \dots & r_1 \\ \vdots & \vdots & \vdots & \vdots \\ \theta_{1,N} & \theta_{2,N} & \dots & r_\eta \end{bmatrix}$$

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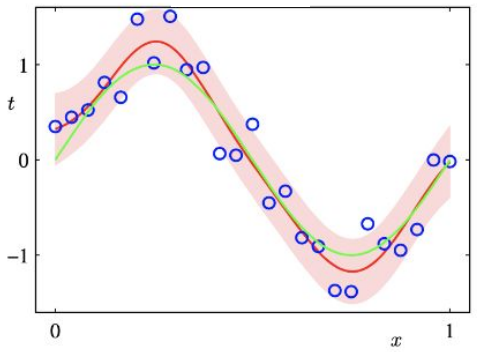

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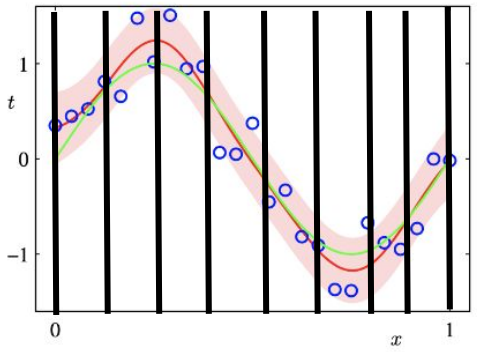
Local GPs are a **greedy**, low dimensional representation of a GP

~ O(eta) scaling

$GP_k(\boldsymbol{\theta}^*)$



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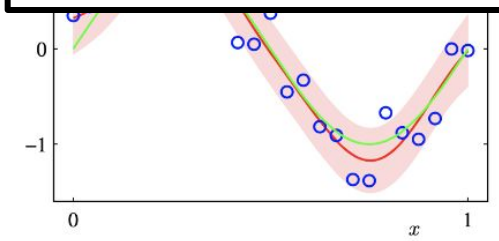
Instead, we train a GP surrogate model on … **Local GPs** are a **greedy** … low …

By assuming that points along a function are independent, **the computational time to evaluate the surrogate model reduces from cubic to linear** in the number of independent variables.



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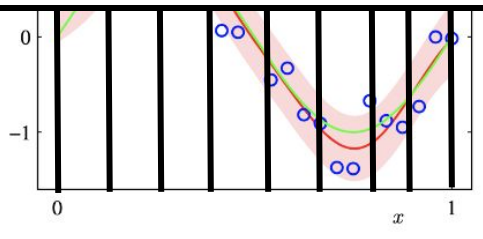

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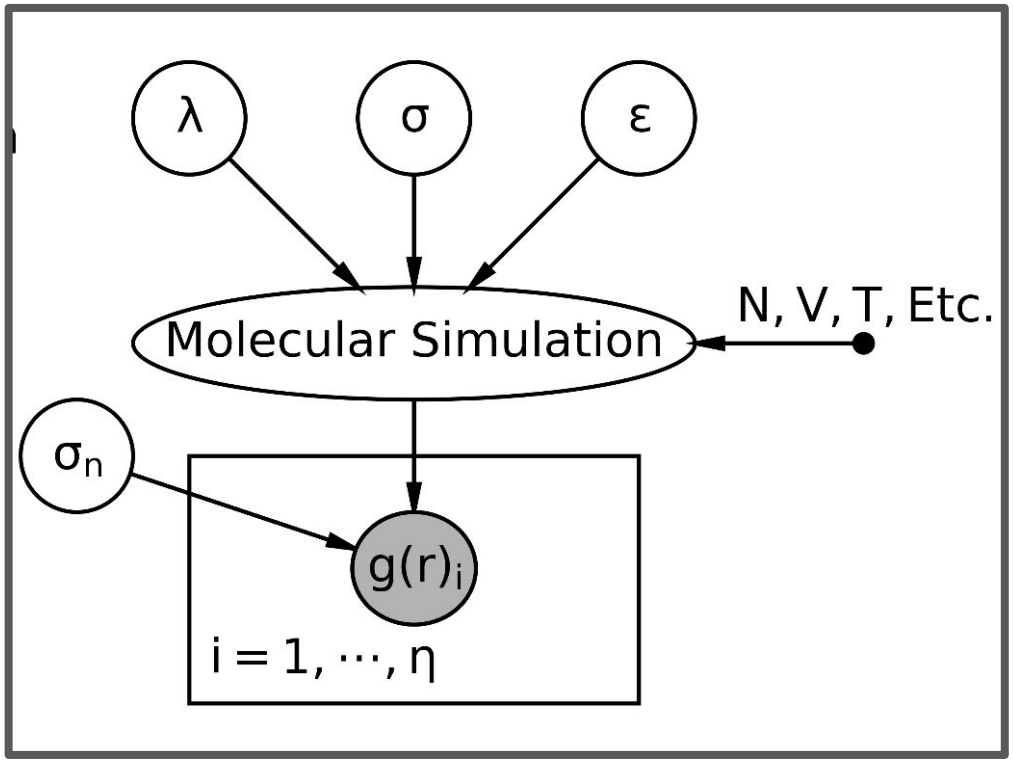
Example: Building a LGP Surrogate Model for the RDF of Liquid Ne



Mie Potential w/ 3 Parameters

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

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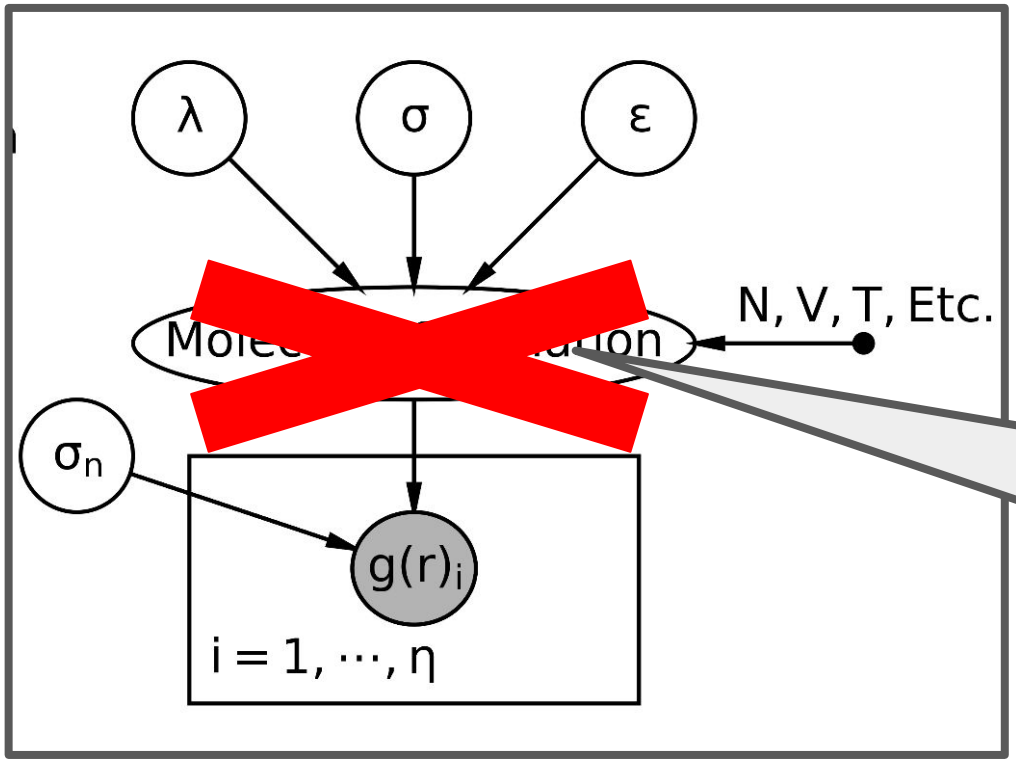

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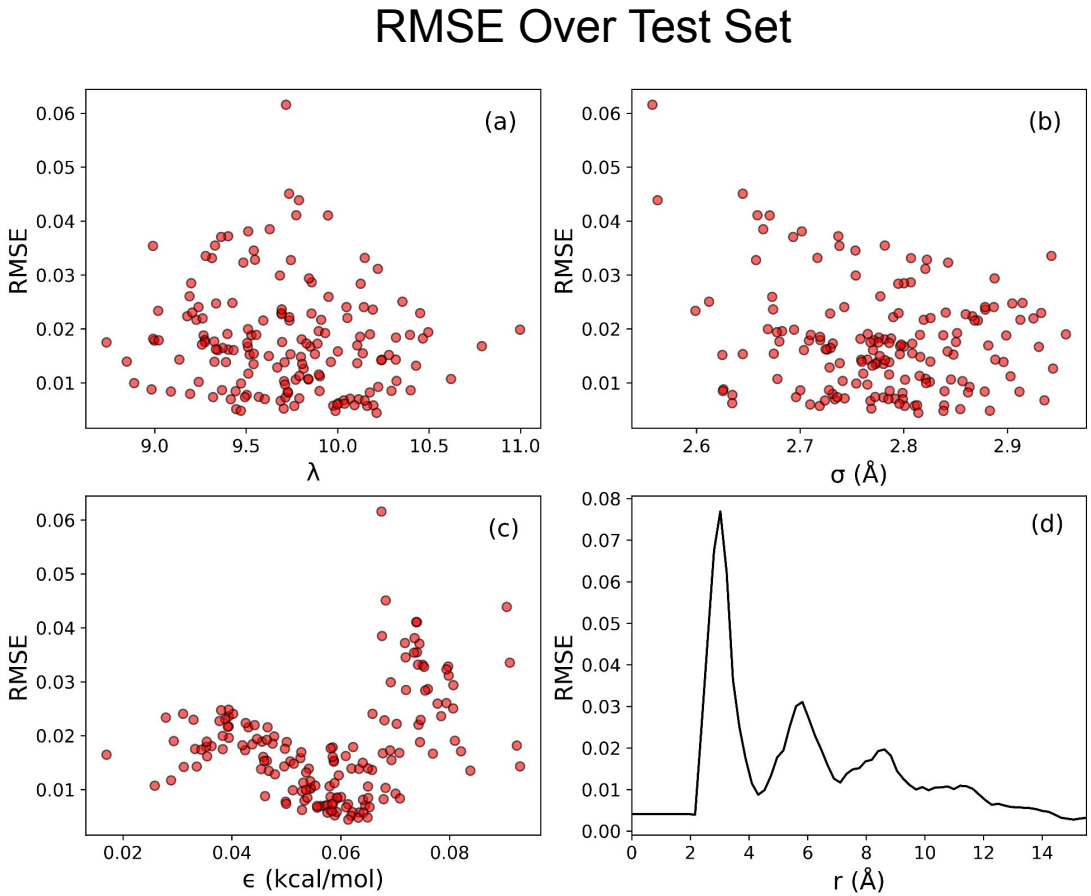
Replace with Machine Learning (Local GP) Surrogate Model!

Local Gaussian Process Surrogate Models are Fast and Accurate

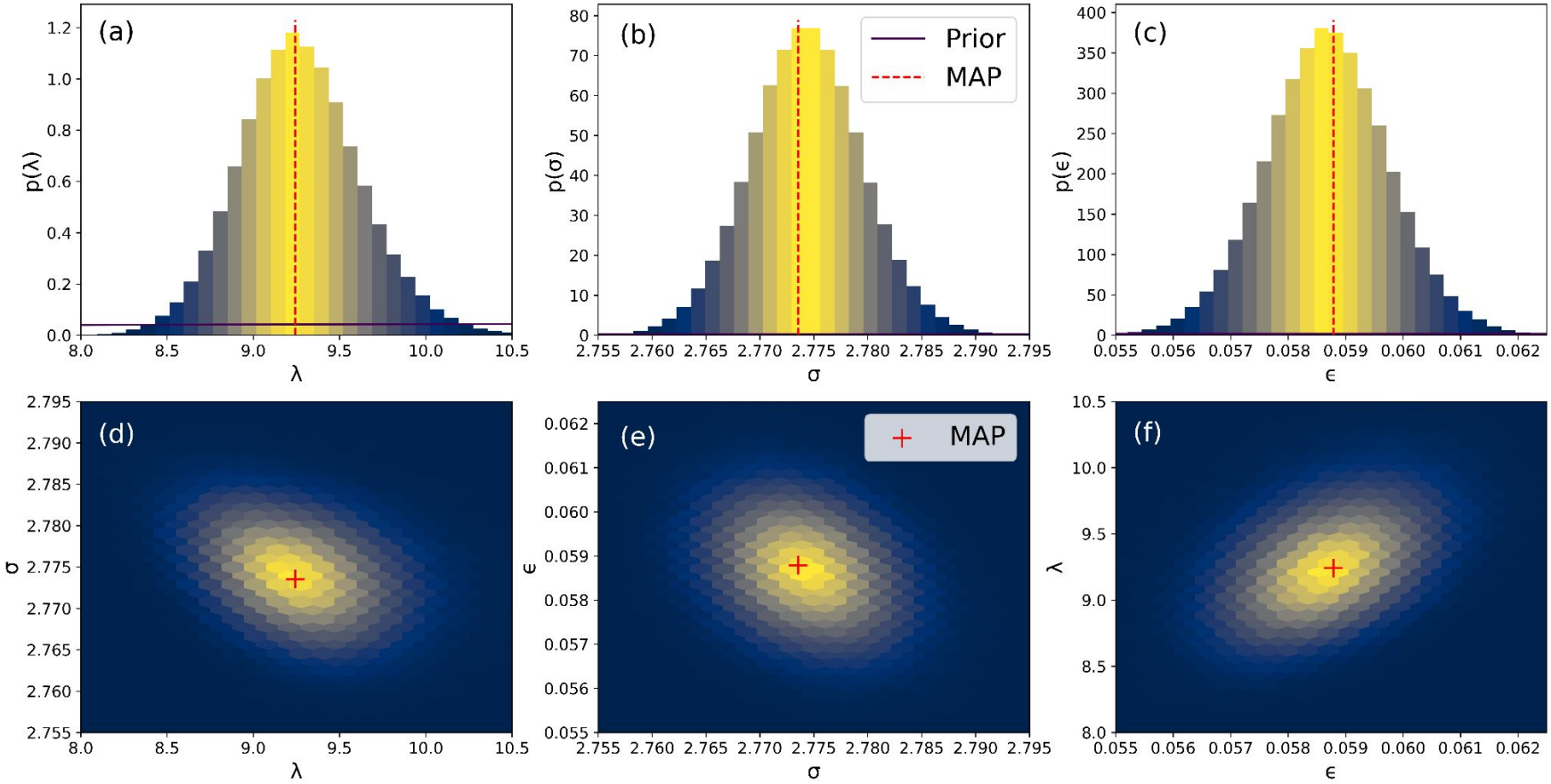
A local GP can predict the RDF 288,000x faster than MD

3500x speed up compared to standard GP

We also find that the RMSE is within the RDF uncertainty



Learning Parameter Relationships from the Posterior Distribution



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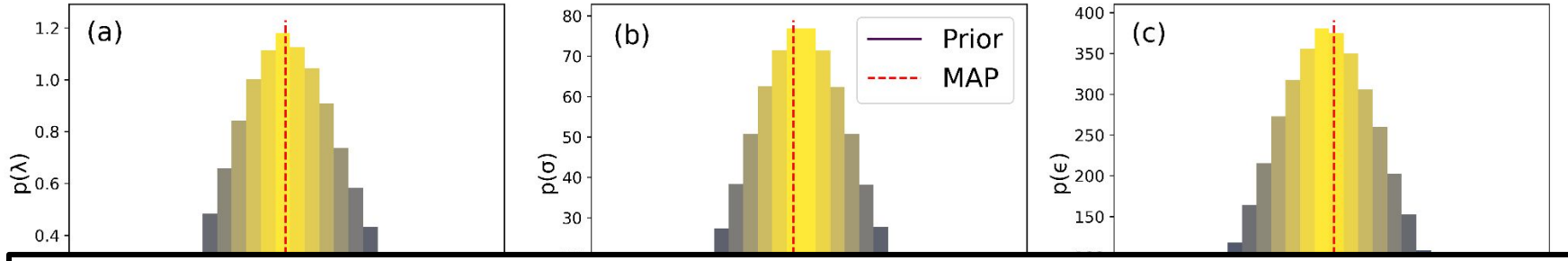
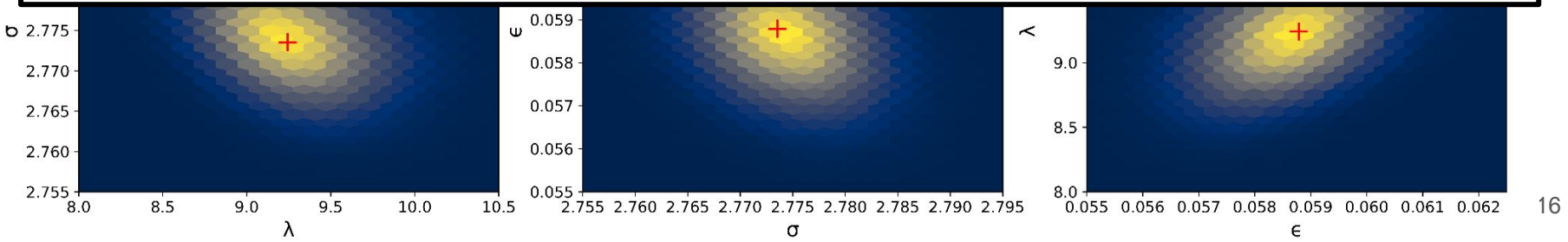
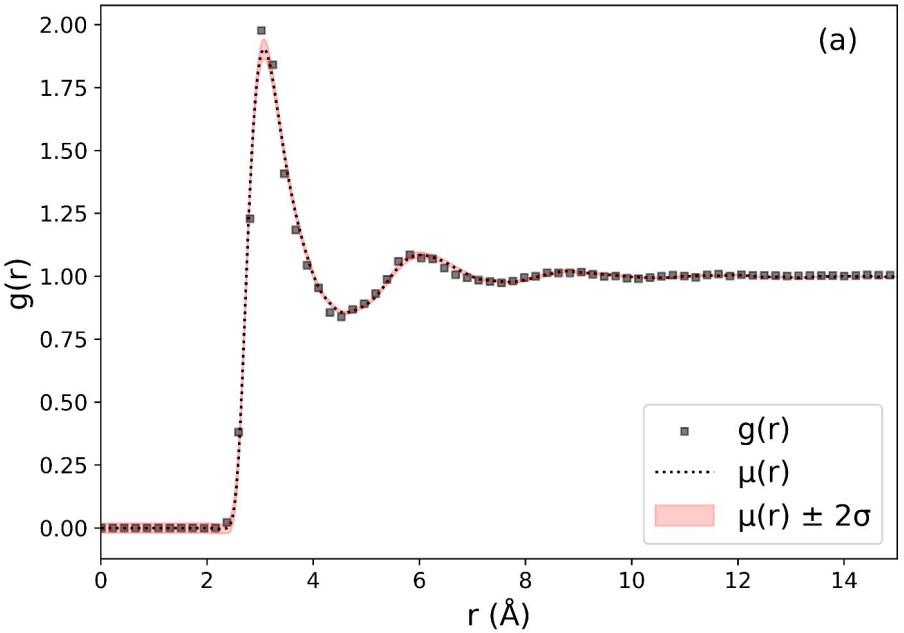


Local GP surrogates reduce the calculation of the Bayesian posterior from ~22 days with a standard GP to under 9 minutes on our local cluster!

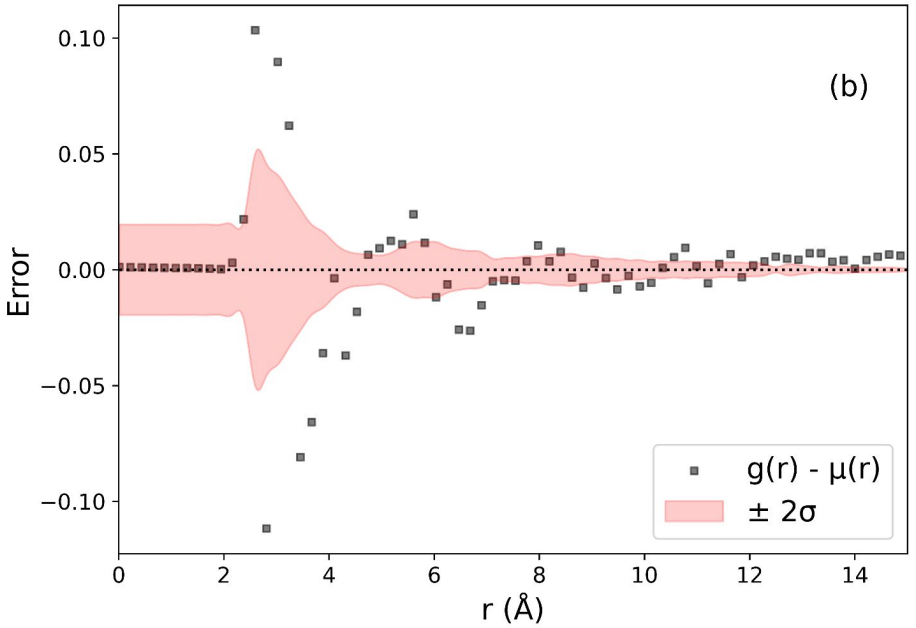


Learning from the Posterior Predictive Distribution

Radial Distribution Function Predictive



Experiment and Posterior Residuals



Summary and Key Takeaways

- A robust and accurate method that can predict intermolecular potentials from scattering data has numerous fundamental and practical applications.
- Probabilistic approaches to scattering analysis are necessary to extract meaningful information on intermolecular potentials.
- **Local Gaussian processes can facilitate Bayesian inference by acting as fast and reliable surrogate models for complex data with built-in UQ.**