Machine learning accelerated methods to predict interatomic forces from experimental structure measurements

Presenter: **Brennon L. Shanks**
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Abdur R. Shazed
Michael P. Hoepfner

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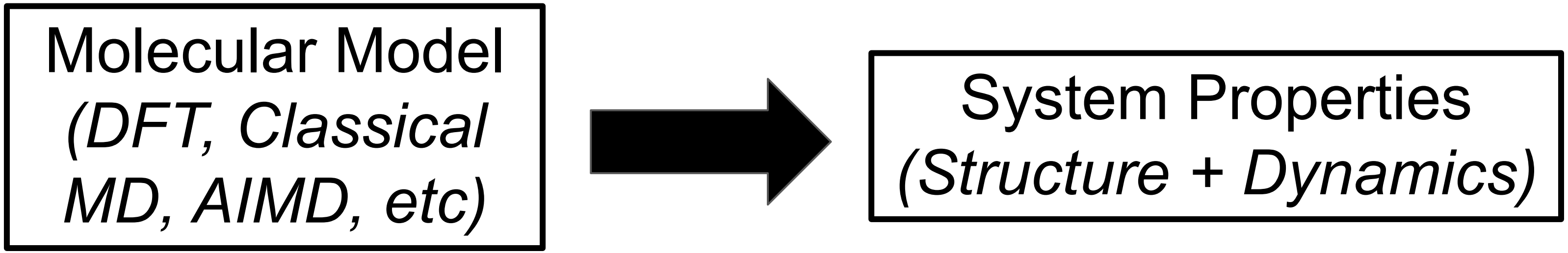
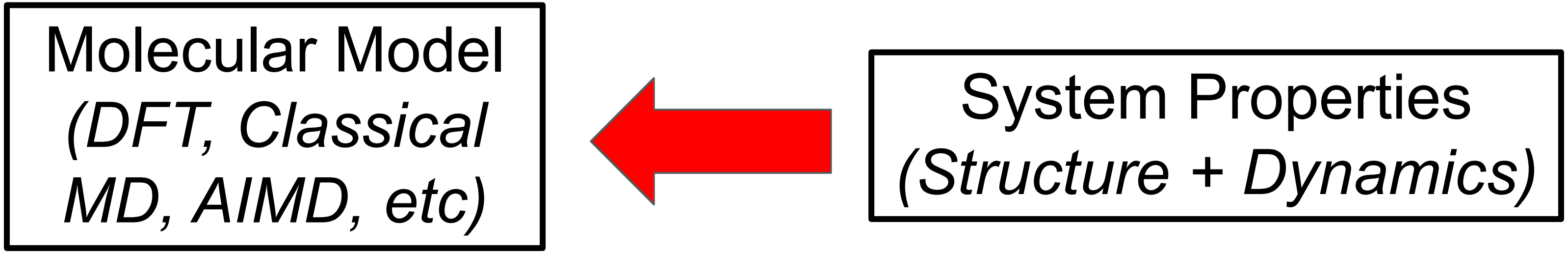
The Standard Paradigm for Atomistic Modeling

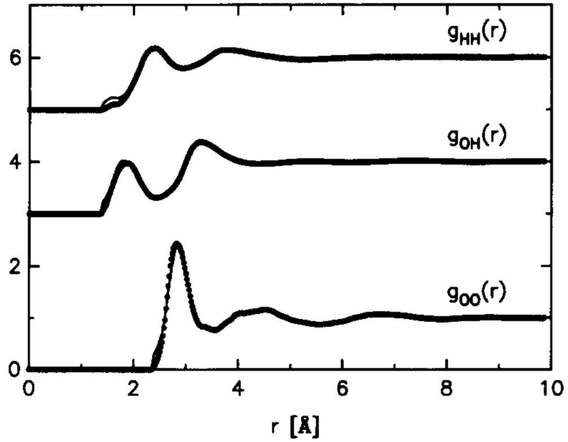


The Inverse Problem for Model Design



Applications of the Inverse Problem for Interesting Chemistry

Scattering Analysis
Alan Soper 1996, *Chem. Phys.*



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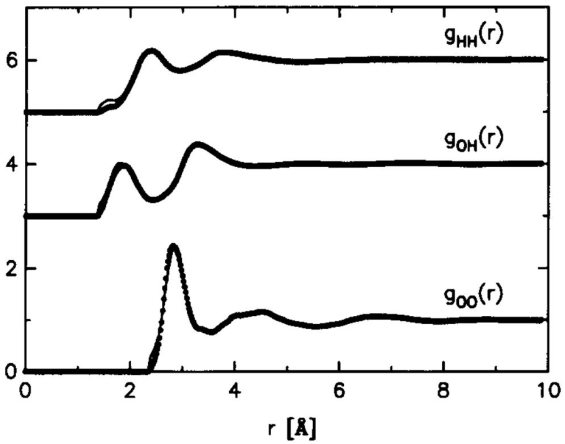

Coarse-Graining
Scott Shell 2013,
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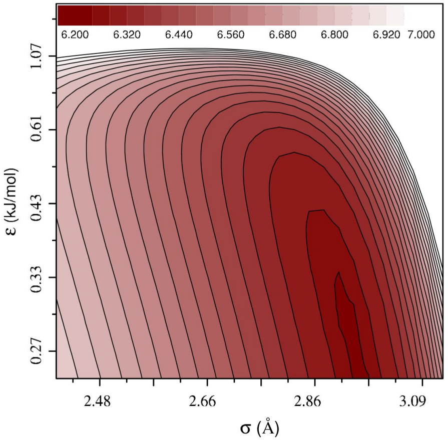

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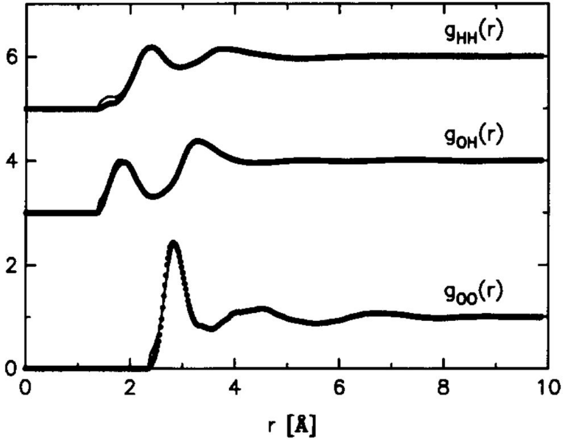

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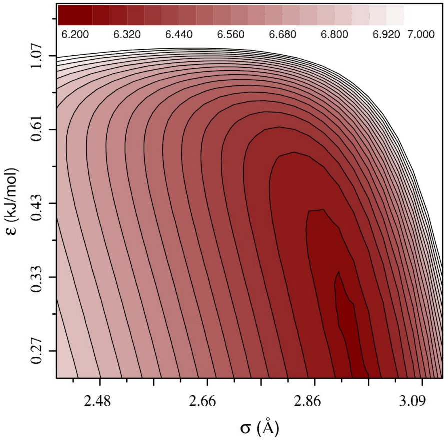
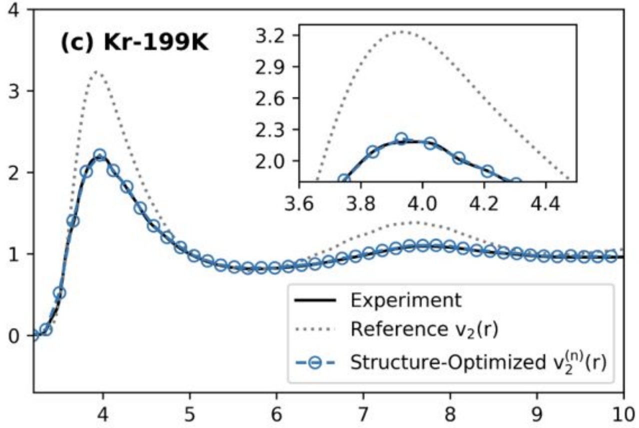


Force Field Development in Simple Liquids
Brennon Shanks 2022, *J. Phys. Chem. Lett.*



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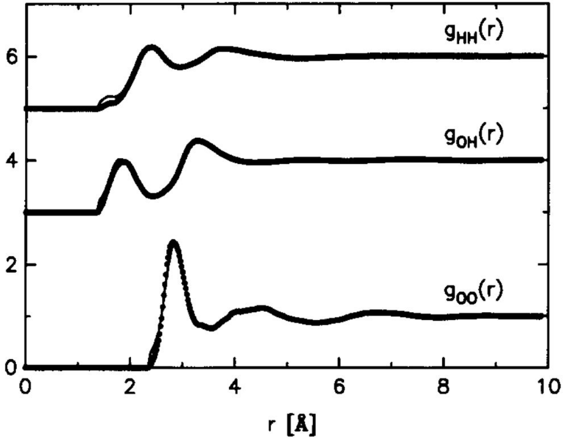

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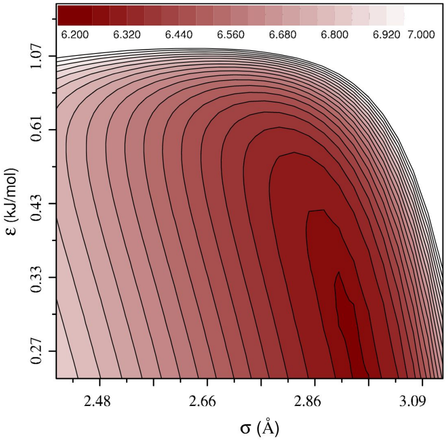

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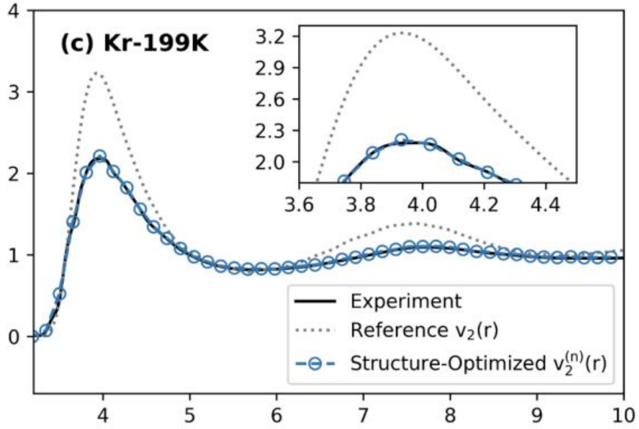
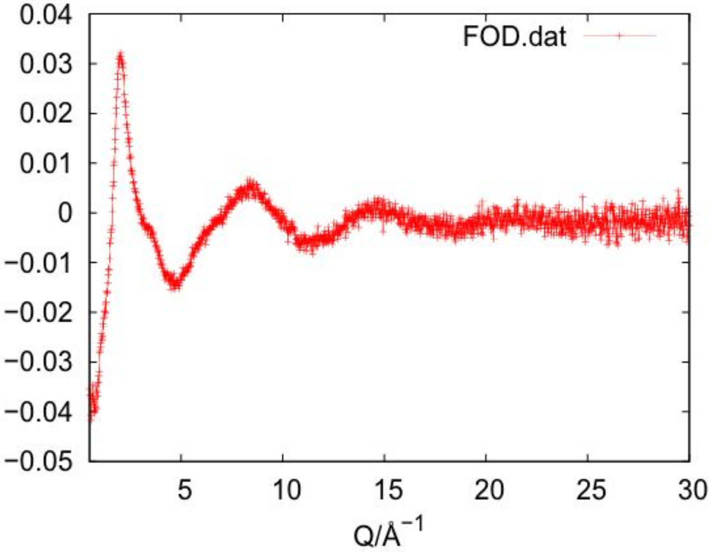


Inverse methods are widely used for scattering analysis and coarse-graining **but** <u>not to build force fields from experimental data</u>… **why?**

Unique Challenges of Inverting Experimental Scattering Data

Measurement Uncertainty



Noise in the Structure Factor of Water
Neuefiend 2012, *Nuc. Inst. Methods.*

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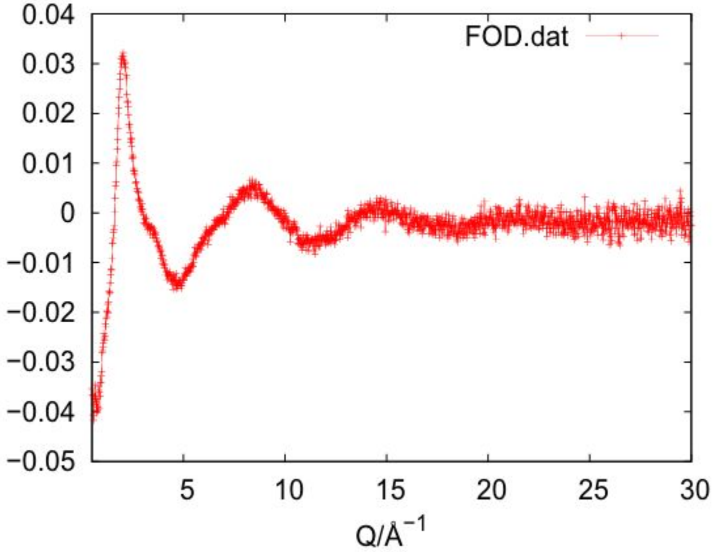

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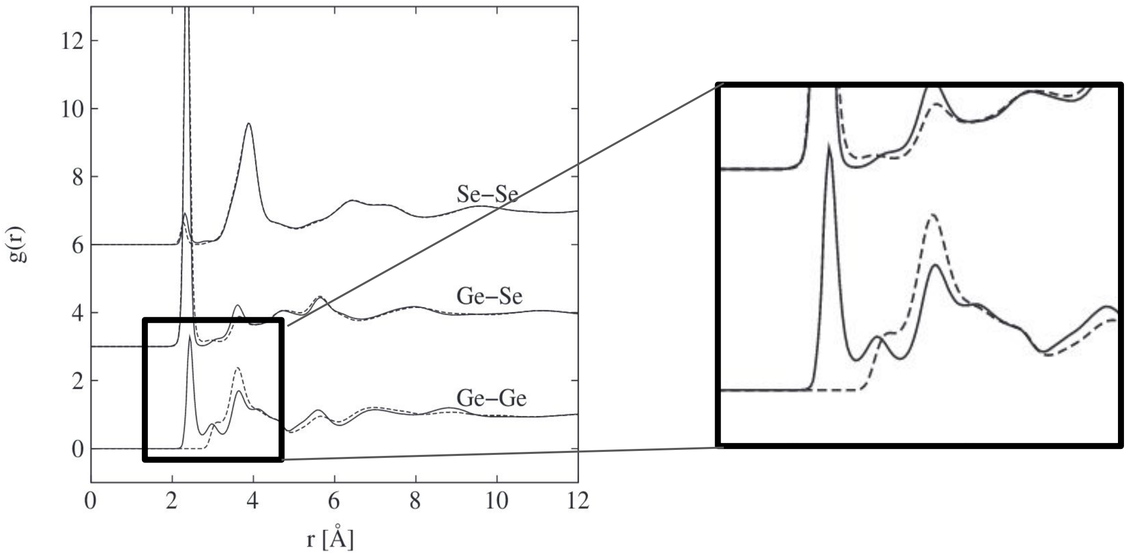
"Experimental" Model Uncertainty



Real-space structure is non-unique
Alan Soper 2007, *Condens. Matt.*

Bayesian Methods Can Quantify Parameter Uncertainty Given Uncertain Experimental Data

The Basic Outline of Bayesian Approaches

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(1) Define **'prior'** distributions

$$p(\theta)$$

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(2) Define and evaluate a **'likelihood'** function

$$p(\mathscr{Y}|\theta)p(\theta)$$

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The Basic Outline of Bayesian Approaches

(1) Define **'prior'** distributions

(2) Define and evaluate a **'likelihood'** function

(3) Solve for the **'posterior'** distribution

$$p(\theta|\mathscr{Y}) = \frac{p(\mathscr{Y}|\theta)p(\theta)}{p(\mathscr{Y})}$$

The posterior is a direct quantification of parameter uncertainty based on your experimental data, Y.

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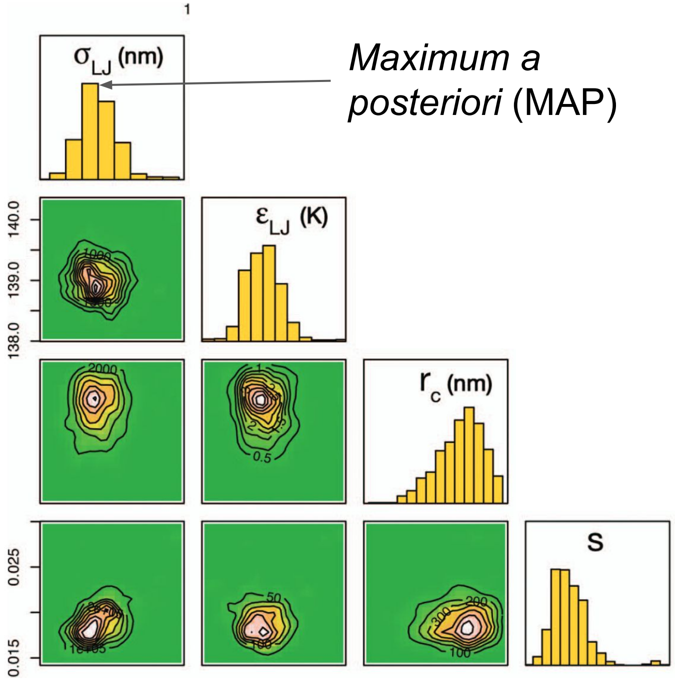

Marginal Posteriors on LJ Parameters
Koumoutsakos 2015, *J. Chem. Phys.*

Accelerated Bayesian Inference with Gaussian Process Surrogates

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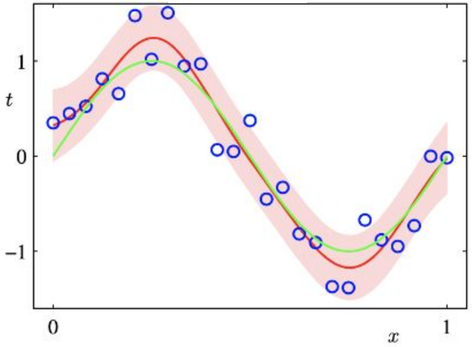
Instead, we train a GP on N ~ 1000 simulations
For data containing η independent variables.

~200 fold speed up

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



99% LOOCV Score

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

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

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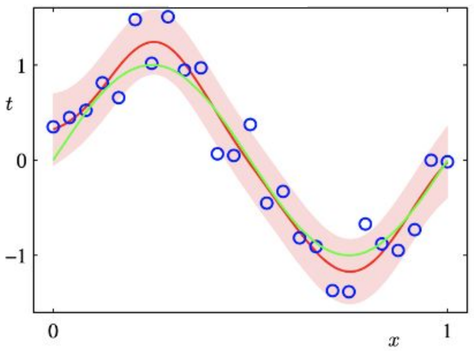

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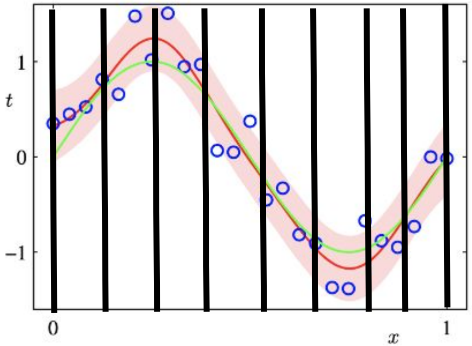
Local GPs reduce matrix size and are about 600 fold faster than full GPs

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

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



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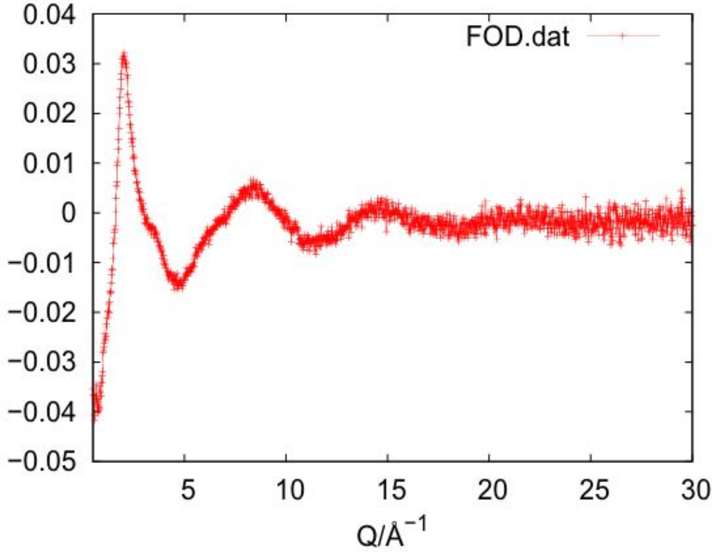
Application: Understanding Experimental Uncertainty Under a Known Model

Measurement Uncertainty



Noise in the Structure Factor of Water

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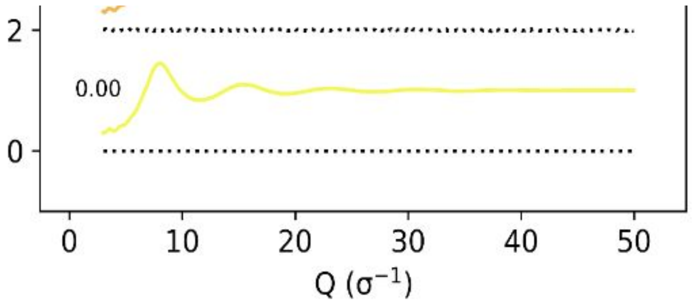
Investigating the impact of measurement uncertainty in Mie fluids

Mie Fluid Interaction Potential

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

Run Model Simulation



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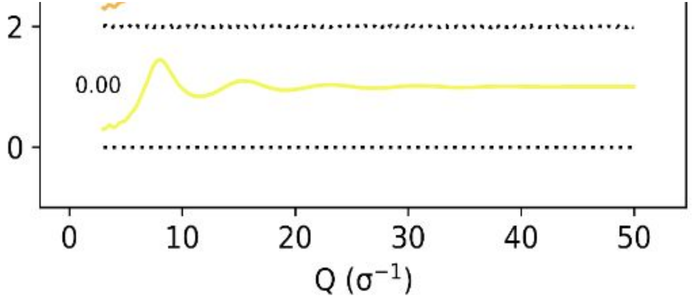
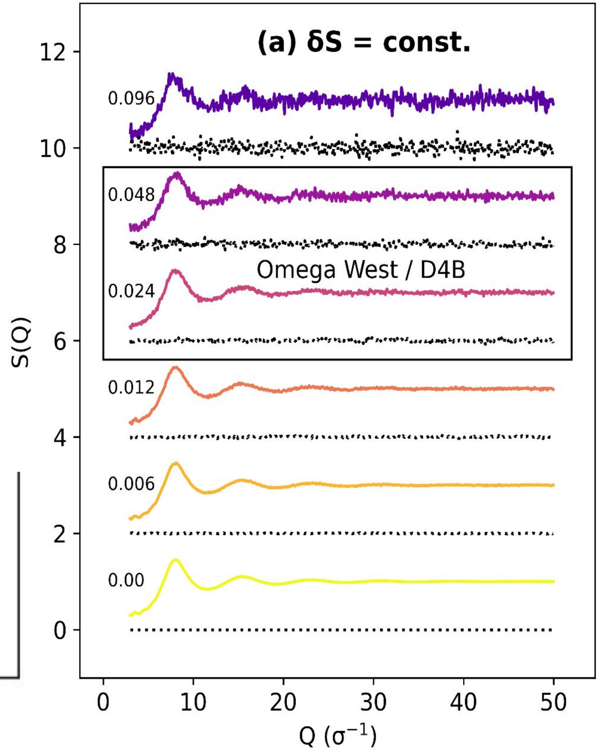
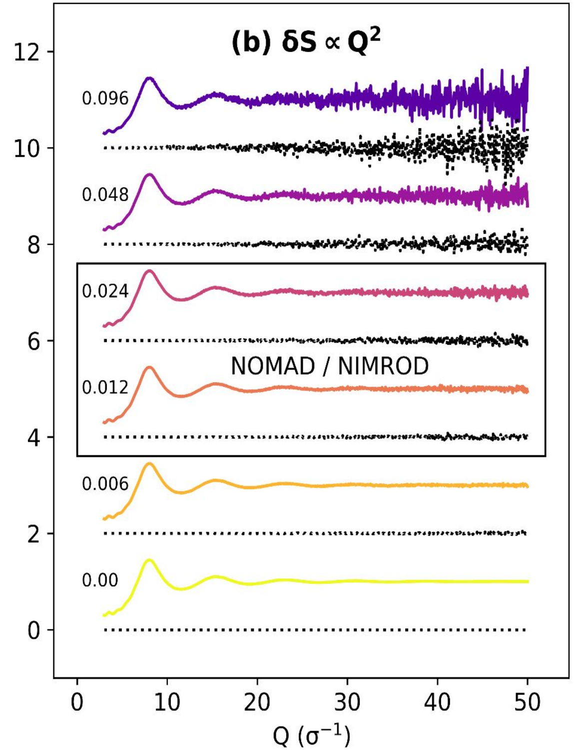


Reactor Source

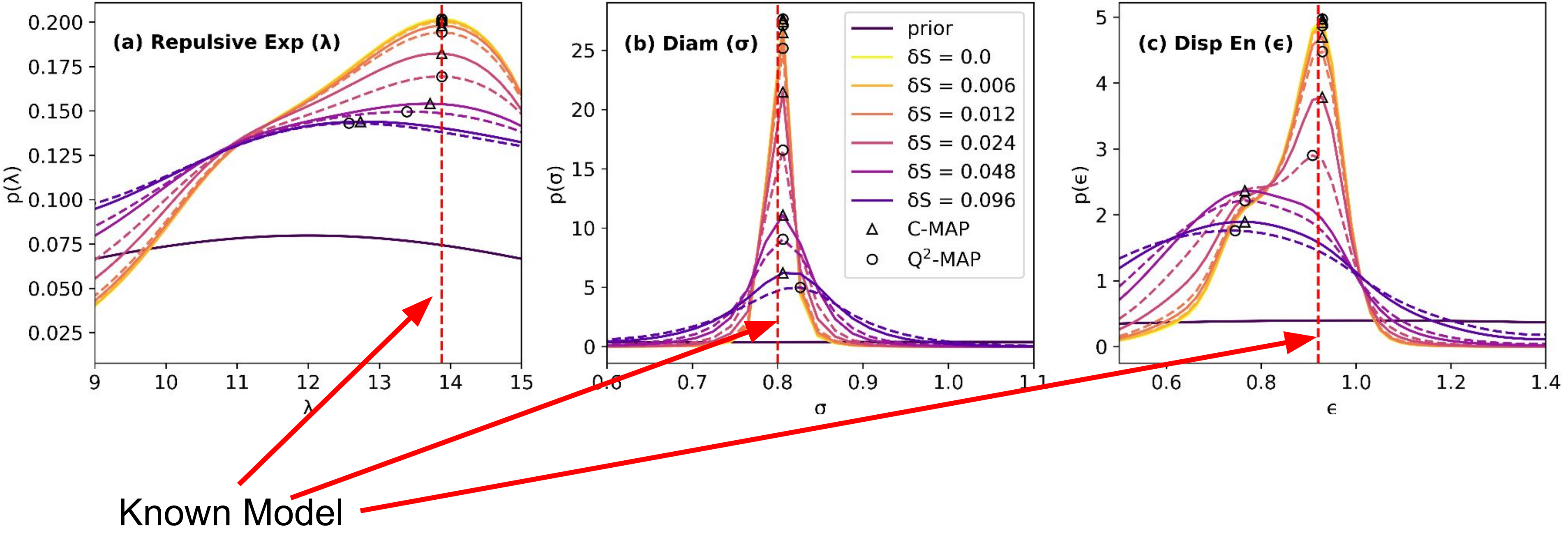


Spallation Source



Can we recover our original model from the structure?

Bayesian Marginal Probability Distribution on Model Parameters



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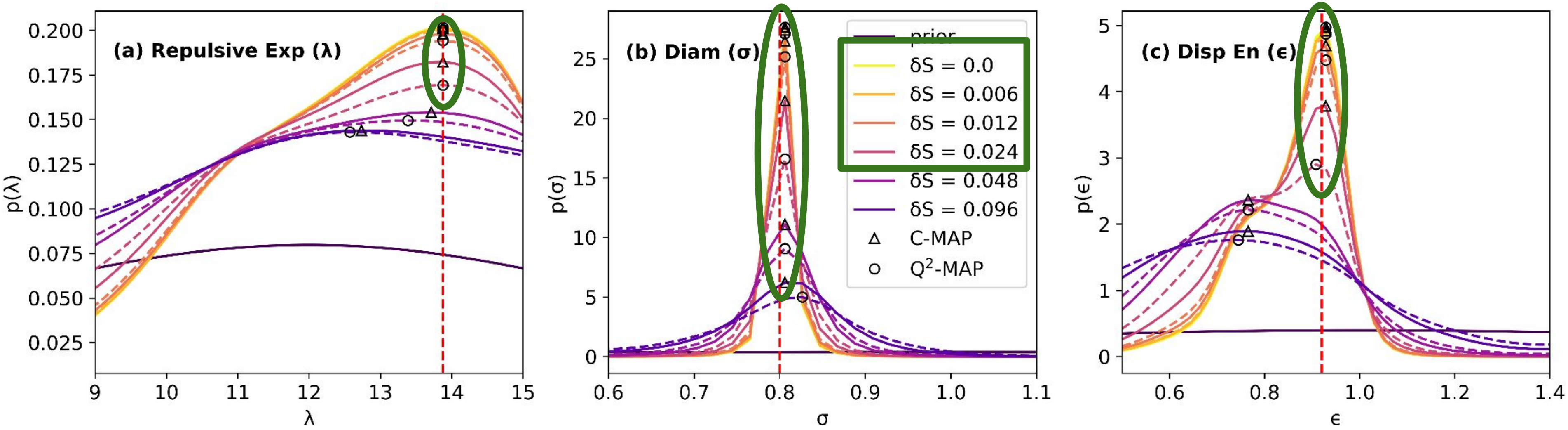

Bayesian optimization recovers force field parameters with high-accuracy for low uncertainty structure factor measurements.

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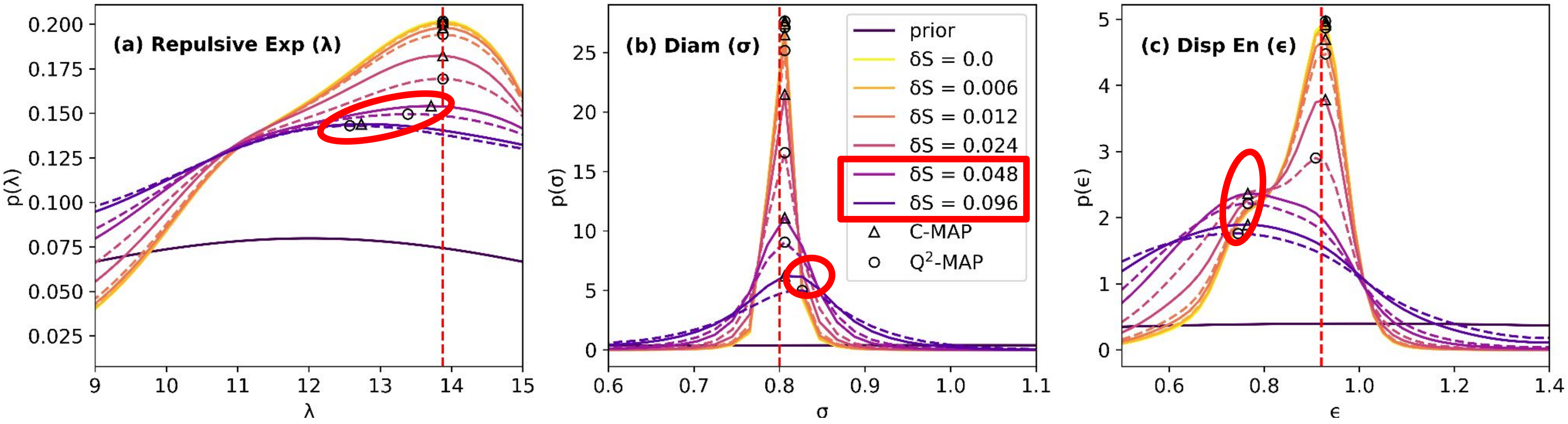

Uncertainty increases and accuracy declines rapidly below a 0.024 variance.

This data quality is representative of the 1960s-1980s neutron sources.

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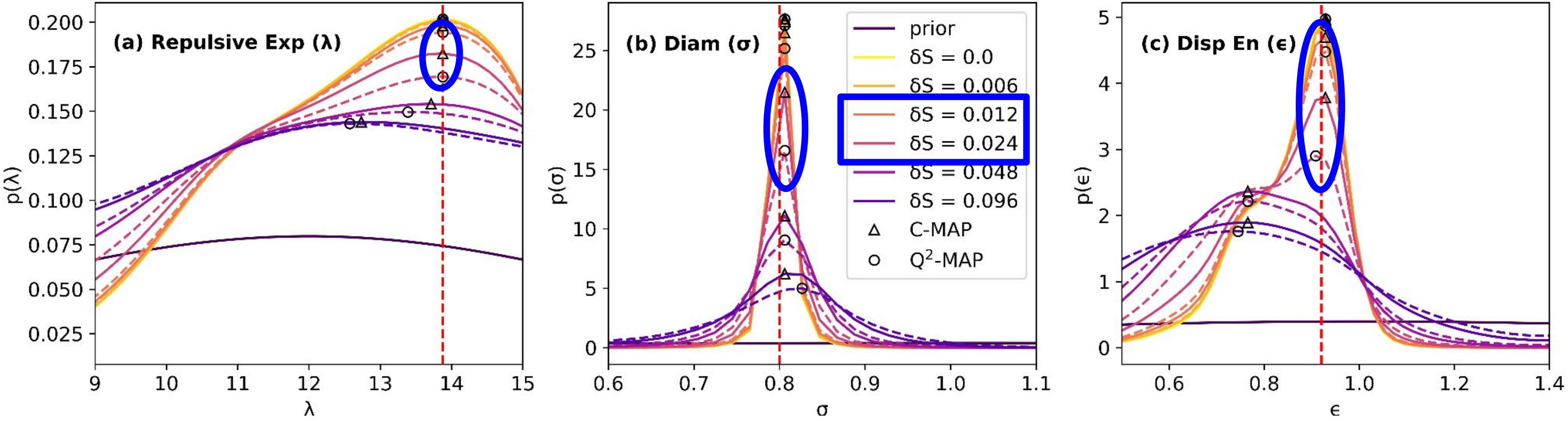

Existing instruments (NOMAD/NIMROD) can provide measurements below the precision threshold.

Summary and Key Takeaways

- Inverse problems are useful for interesting chemistry, including scattering analysis, coarse-graining, and atomistic force field development.
- Bayesian uncertainty quantification is a rigorous framework to understand our confidence in our models.
- We need ML accelerated approaches to populate Bayesian likelihood distributions.
- Bayesian UQ can help answer fundamental questions around interatomic forces, enable active learning approaches using decision theory, and rigorously incorporate experimental data into atomistic models.