Recovering Structural Information about Nanoparticle Systems

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Nanoparticle Systems

- Materials (natural or artificial) made up of nanoparticles.
- Sizes ranging from 1 nanometer to 1000s nanometers.
- Wide variety of applications in optical, electronic and biomedical fields. E.g.:
  - Inorganic nanomaterials in optoelectronics.
  - Organic material based nano-devices such as Organic Photovoltaics (OPVs), OLEDs.
  - Chemical catalysts, drug design and discovery, biological process dynamics.

Importance of structural information:

- Nanomaterials exhibit shape and size-dependent properties, unlike bulk materials which have constant physical properties regardless of size.
- Nanoparticle characterization is necessary to establish understanding and control of material synthesis and applications.
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Measuring Structural Information at Nano-scale

- Electron microscopy (TEM, SEM),
- atomic force microscopy (AFM),
- X-ray photoelectron spectroscopy (XPS),
- X-ray diffraction (XRD),
- X-ray scattering,
- and more.

**X-ray scattering:**
- Determine the size distribution profile of nanoparticles in suspension or polymers in solution.
- Probe the behavior of complex fluids such as polymer solutions.
- Probe structures of non-crystalline thin-film materials.

**Examples:**
- Small-Angle X-ray Scattering (SAXS)
- Grazing-Incidence SAXS (GISAXS)
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X-ray Scattering at Synchrotrons

X-Ray Scattering: Examples
X-Ray Scattering: Complex Examples

Gratings

Organic Photovoltaics

Real Sample

Model

Scattering Pattern
Computational Problems in Structure Recovery: Inverse Modeling

Start

Initial guess
Computational Problems in Structure Recovery: Inverse Modeling

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forward simulation
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compute error w.r.t. experimental data
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End
Need for High-Performance Computing

Data generation and analysis gap:

- High measurement rates of current state-of-the-art light beam detectors.
- Wait for days for analyzing data with previous softwares.
- Extremely inefficient utilization of facilities due to mismatch.
- *Example:* 100 MB raw data per second. Up to 12 TB per week.
Need for High-Performance Computing

High computational and accuracy requirements:

• Errors are proportional to the resolutions of various computational discretization.

• Higher resolutions require higher computational power.

• Example:
  • $O(10^7)$ to $O(10^{15})$ kernel computations for one simulation.
  • $O(10^2)$ experiments per material sample.
  • $O(10)$ to $O(10^3)$ forward simulations for inverse modeling per scattering pattern.
Need for High-Performance Computing

Science Gap:

- Beam-line scientists lack access to high-performance algorithms and codes.
- In-house developed codes limited in compute capabilities and performance.
- Also, they are extremely slow – wait for days and weeks to obtain basic results.
Forward Simulations: Computing Scattered Light Intensities

Given:
1. a sample structure model, and
2. experimental configuration,
simulate scattering patterns.

Based on *Distorted Wave Born Approximation* (DWBA) theory.
Inverse Modeling

Forward simulation kernel: computing the scattered light intensities. E.g.

- FFT computations (SAXS)
- Complex form factor and structure factor computations (GISAXS)

Various inverse modeling algorithms:

- Reverse Monte-Carlo simulations for SAXS.
- Sophisticated optimization algorithms for GISAXS.
  - Gradient based: LMVM (Limited-Memory Variable-Metric.)
  - Derivative-free trust region-based: POUNDerS.
  - Stochastic: Particle Swarm Optimization.
Inverse Modeling

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Reverse Monte Carlo Simulations

Initial model (random) → FFT → Experimental Structure Factor

Accept or reject move

Compute χ²-error

Move particle at random

FFT
Reverse Monte Carlo Simulations: Validation

Actual Models
Reverse Monte Carlo Simulations: Validation

Actual Models

Initial Models
Reverse Monte Carlo Simulations: Validation

Actual Models

Initial Models

Recovered Models
Reverse Monte Carlo Simulations: Strong and Weak Scaling

Titan

Hopper

asarje@lbl.gov Lawrence Berkeley National Laboratory
Limited-Memory Variable-Metric and POUNDerS

- Methods from the optimization package TAO.
- LMVM is a gradient-based method.
- POUNDerS is a derivative-free trust-region-based method.
Limited-Memory Variable-Metric and POUNDerS: Two Parameter Case

A Single Cylindrical Nanoparticle
Limited-Memory Variable-Metric and POUNDerS: Two Parameter Case

A Single Cylindrical Nanoparticle

X-Ray Scattering Pattern
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Objective Function Map
Limited-Memory Variable-Metric and POUNDerS: Two Parameter Case

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Objective Function Map

LMVM Convergence Map

POUNDerS Convergence Map
Limited-Memory Variable-Metric and POUNDerS: Six Parameter Case

Pyramidal Nanoparticles forming a Lattice
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Objective Function Map

LMVM does not converge

POUNDerS Convergence Map
Particle Swarm Optimization

- Stochastic method.
- Multiple agents, “particle swarm”, search for optimal points in the parameter space.
- Agent velocities influenced by history of traveled paths.
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\[
\vec{v}_i \leftarrow \omega \vec{v}_i + (\vec{b}_i - \vec{x}_i) r_1 \phi_1 + (\vec{b}_g - \vec{x}_i) r_2 \phi_2
\]
Particle Swarm Optimization: Fitting X-Ray Scattering Data

Fitting 2 Parameters

Fitting 6 Parameters
Particle Swarm Optimization: Performance

Convergence w.r.t. Search Space Volume

Strong Scaling on Titan
Particle Swarm Optimization: Agents vs. Generations

![Graph showing the relationship between Relative Error and Generation for different numbers of agents.](image)
An Ongoing Work

We saw that:

- Derivative-based methods converge only for simple cases.
- Trust-region-based methods are very sensitive to initial guess.
- PSO is robust, nearly always converging, but expensive.
- Need better optimization algorithms.
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Near future:

- GPUs have brought data analysis time from days and weeks to just minutes and seconds.
- Opening gates to much more sophisticated analyses.
- We are applying Deep Learning for feature and structural classification to generate initial models to fit.
- Our codes are already being used at various synchrotrons world-wide.
Our Current Team

- **Alexander Hexemer**, *Advanced Light Source, Berkeley Lab*.
- **Dinesh Kumar**, *Advanced Light Source, Berkeley Lab*.
- **Xiaoye S. Li**, *Computational Research Division, Berkeley Lab*.
- **Abhinav Sarje**, *Computational Research Division, Berkeley Lab*.
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And we are open for collaborations ...
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Code: http://portal.nersc.gov/project/als/hipgisaxs