S5326
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)
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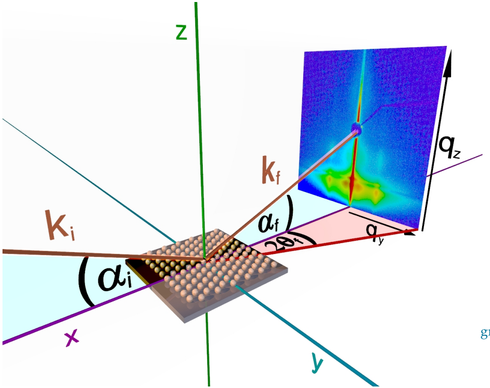
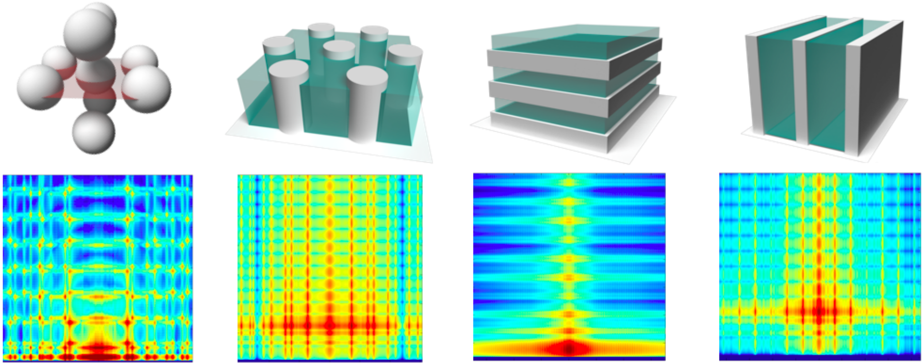
X-ray Scattering at Synchrotrons



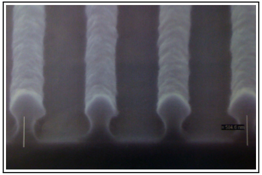
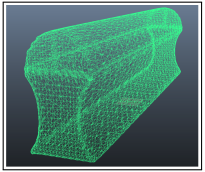
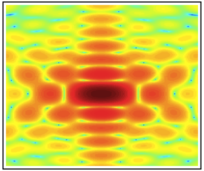
graphic: courtesy of A. Meyer, *www.gisaxs.de*

X-Ray Scattering: Examples

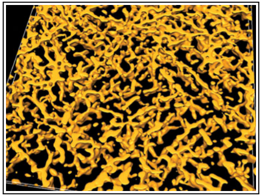
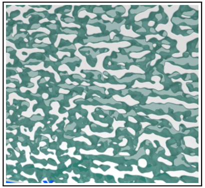
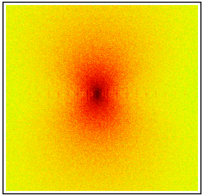


X-Ray Scattering: Complex Examples

Gratings

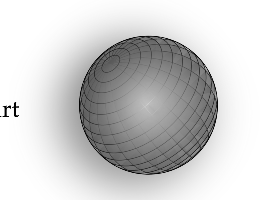
Organic Photovoltaics

Real Sample | Model | Scattering Pattern

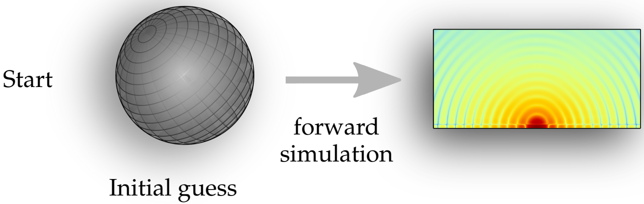
Computational Problems in Structure Recovery: Inverse Modeling

Start



Initial guess

Computational Problems in Structure Recovery: Inverse Modeling



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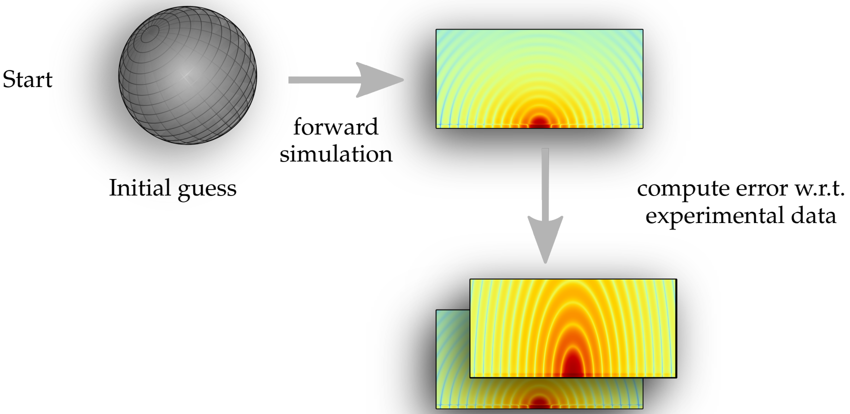

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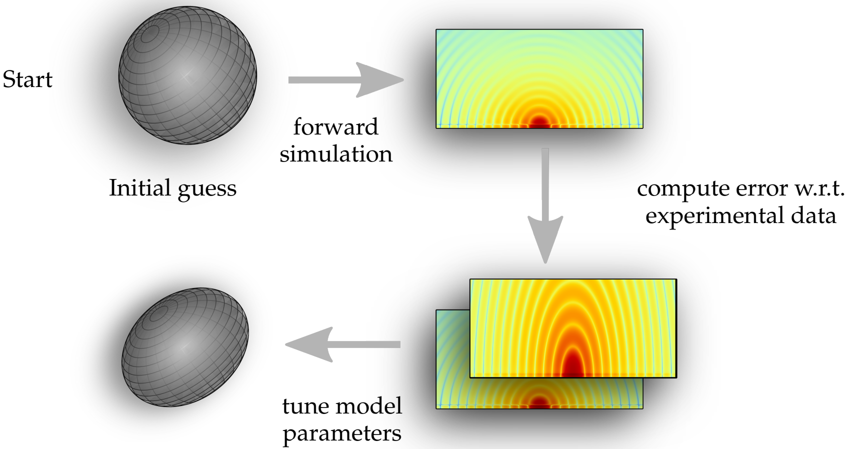

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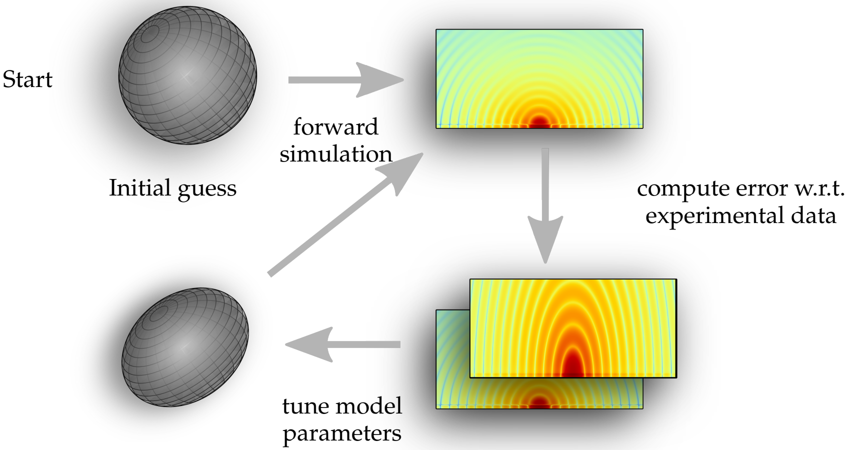

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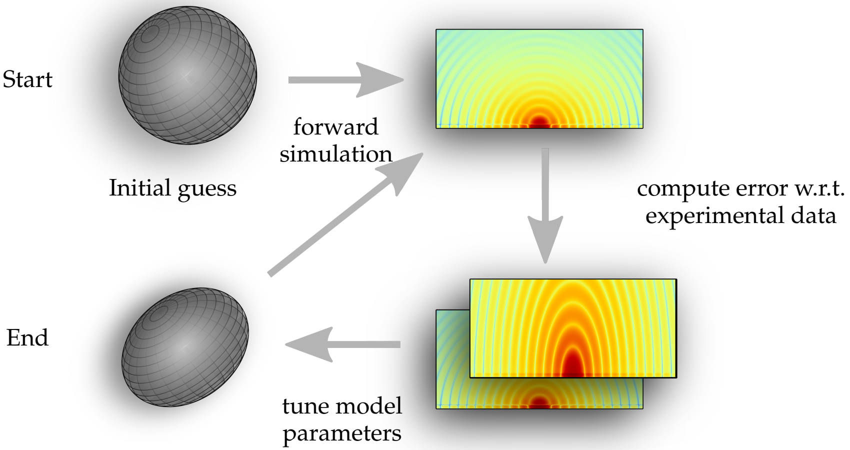


Need for High-Performance Computing

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:

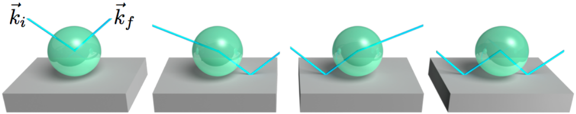
- Beam-line scientists lack access to high-performance algorithms and codes.
- In-house developed codes limited in compute capabilities and performance.
- Also, they are extremely slow – wait for days and weeks to obtain basic results.

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 Optmization.

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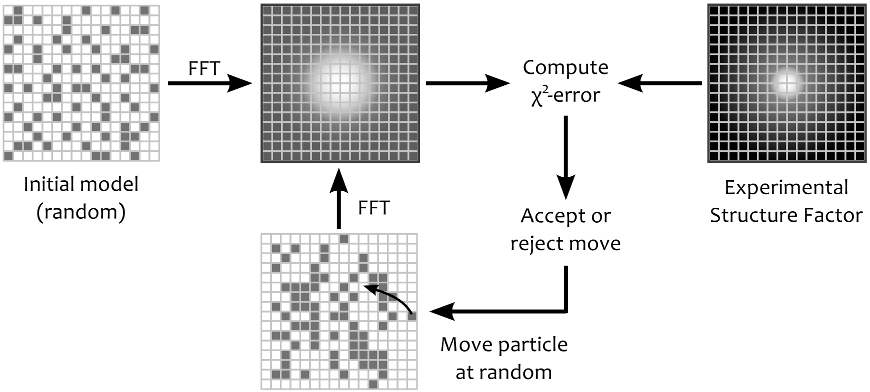
Reverse Monte Carlo Simulations



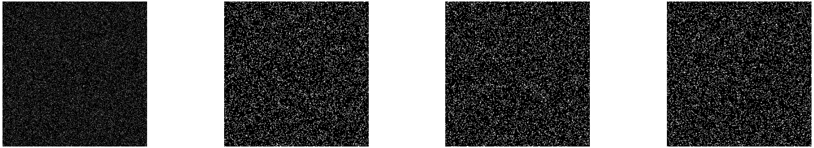
Reverse Monte Carlo Simulations: Validation

Actual Models

Reverse Monte Carlo Simulations: Validation

Actual Models

Initial Models



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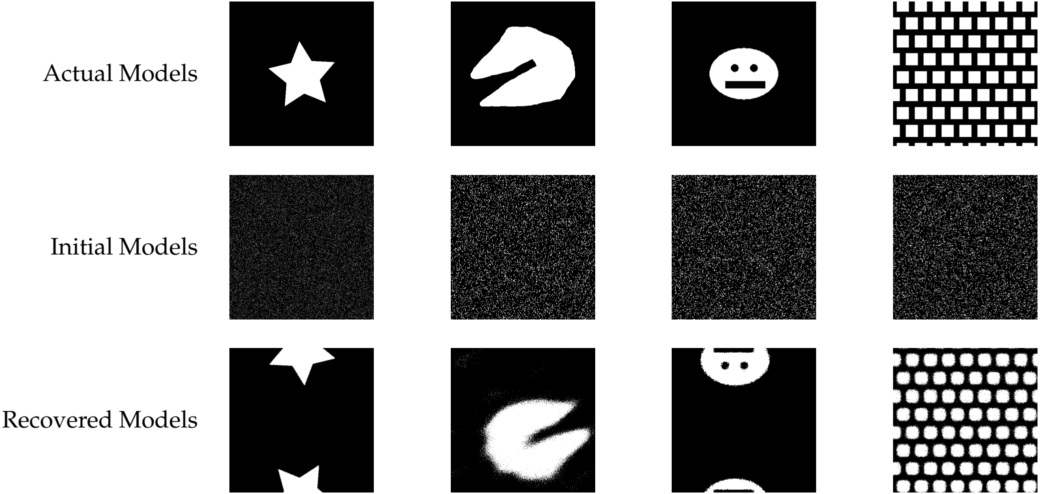
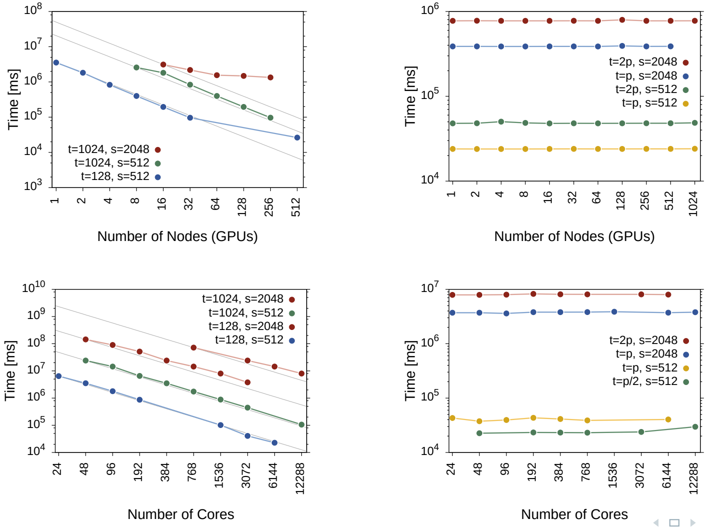


Reverse Monte Carlo Simulations: Strong and Weak Scaling

Titan

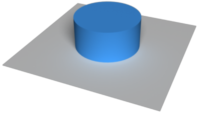
Hopper



Limited-Memory Variable-Metric and POUNDerS

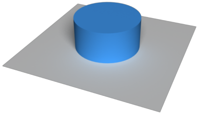
- Methods from the optimization package TAO.
- LMVM is a gradient-based method.
- POUNDerS is a derivative-free trust-region-based method.

Limited-Memory Variable-Metric and POUNDerS: Two Parameter Case



A Single Cylindrical Nanoparticle

Limited-Memory Variable-Metric and POUNDerS: Two Parameter Case



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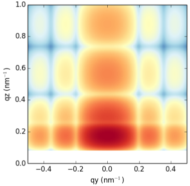

X-Ray Scattering Pattern

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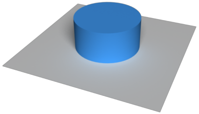

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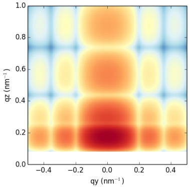

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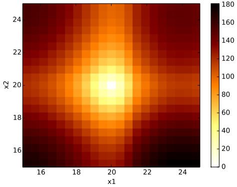
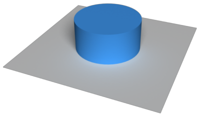


Objective Function Map

Limited-Memory Variable-Metric and POUNDerS: Two Parameter Case



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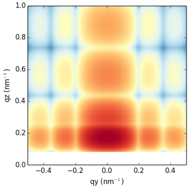

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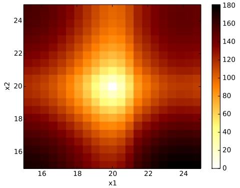

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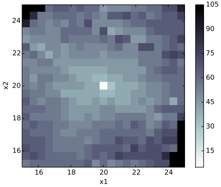
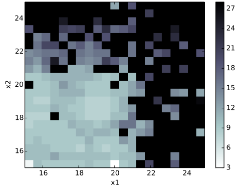
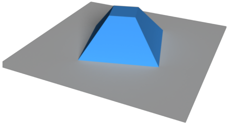


LMVM Convergence Map



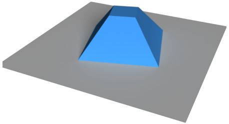
POUNDerS Convergence Map

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



Pyramidal Nanoparticles
forming a Lattice

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



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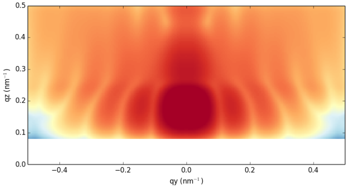

X-Ray Scattering Pattern

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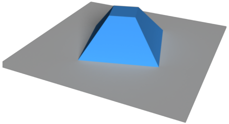

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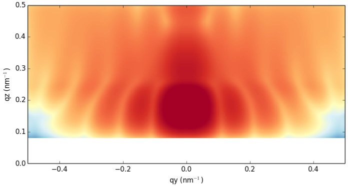

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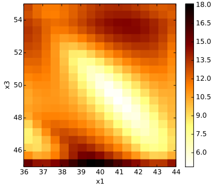

Objective Function Map

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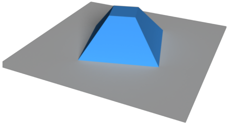

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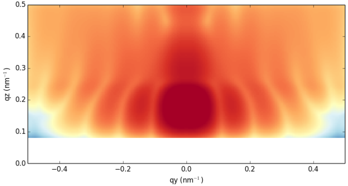

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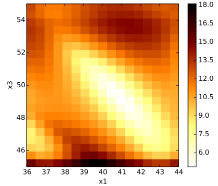
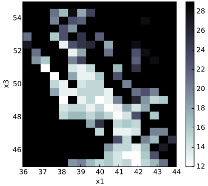


Objective Function Map

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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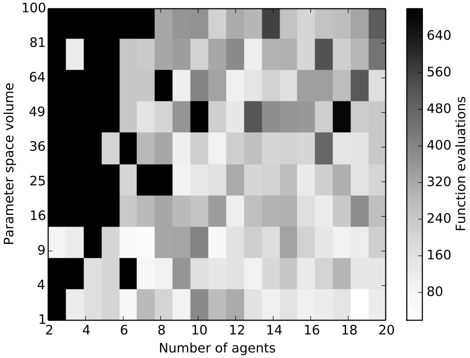
inertia coefficient force coefficients

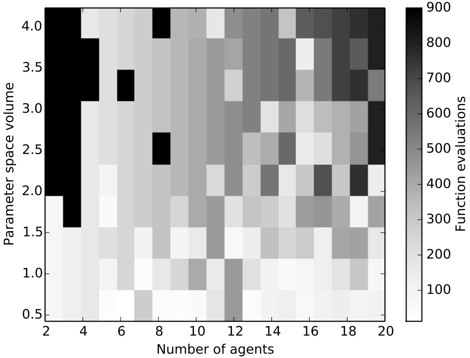
$$\vec{v_i} \leftarrow \omega\vec{v_i} + (\vec{b_i} - \vec{x_i})\vec{r_1}\phi_1 + (\vec{b_g} - \vec{x_i})\vec{r_2}\phi_2$$

local best position global best position

Particle Swarm Optimization: Fitting X-Ray Scattering Data

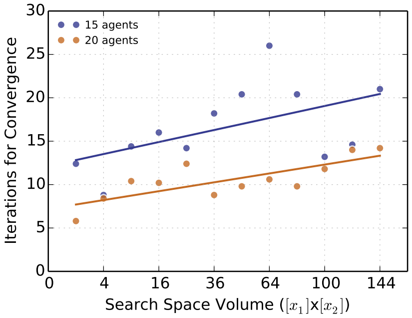


Fitting 2 Parameters

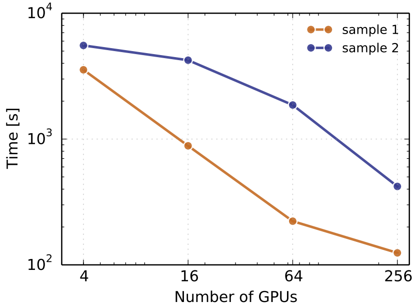


Fitting 6 Parameters

Particle Swarm Optimization: Performance

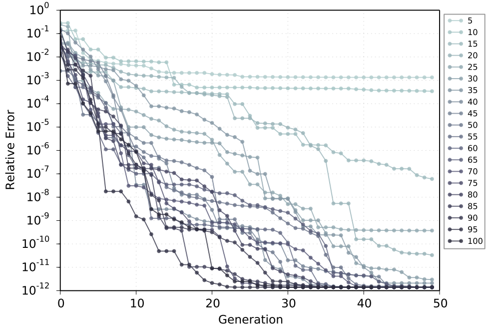


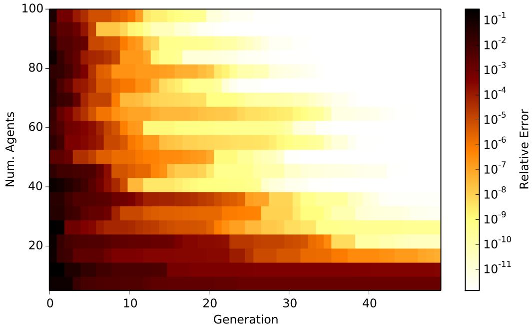
Convergence w.r.t. Search Space Volume



Strong Scaling on Titan

Particle Swarm Optimization: Agents vs. Generations





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*.
- **Singanallur Venkatakrishnan**, *Advanced Light Source, Berkeley Lab*.

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- **Singanallur Venkatakrishnan**, *Advanced Light Source, Berkeley Lab*.

And we are open for collaborations ...

Acknowledgments

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- Used resources of the Oak Ridge Leadership Computing Facility at the Oak Ridge National Laboratory, which is supported by the Office of Science of the U.S. Department of Energy under Contract No. DE-AC05-00OR22725.

Thank you!

Contact: asarje@lbl.gov

Code: http://portal.nersc.gov/project/als/hipgisaxs