Applied Mathematics For Experimental Science

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Simons Chair in Mathematics, Department of Mathematics, UC Berkeley
Want to do two things:

Provide an overview of CAMERA

Discuss detectors, data, mathematics, and models.
The Center for Applied Mathematics for Energy Research Applications (CAMERA)

Mission:  Build the applied mathematics that can accelerate scientific discovery at DOE experimental facilities

Execution:  Coordinated team of applied mathematicians, beam scientists, computational chemists, computer scientists, materials scientists, statisticians, image and signal processors, ...

Initial set of partners:

Advanced Light Source  Molecular Foundry  NCEM

Initial Support:  LBNL LDRD  

Now:  Joint ASCR-BES Pilot Project  
(Steve Lee, P. Lee, B. Harrod, S. Binkley, H. Kung)
Overview of CAMERA

Build the advanced mathematics that can:
- Extract information from murky data, and help interpret experimental results
- Provide on-demand analysis as results are being generated
- Steer experiment and suggest optimal solutions
- Decrease turn-around time/save money: More experiments and more users
- Extend the capabilities of existing and future experimental facilities

To do so, we need to:
- Have experimental scientists/applied mathematicians work together
- Develop common language
- Build new mathematical models, invent algorithms, build prototype codes
- Test on “shop floor”, iterate until codes are solid and useful

Goal: Deliverables users can use (without becoming mathematicians):
- Advanced mathematics embedded in useable software tools
CAMERA: Original/Current Projects

**Pytchography (ALS/MATH)**
Coherent Diffraction with Microscopy

**Automatic Image Analysis (ALS/MATH)**

**New methods for Density Functional Theory (MF/MATH)**

\[ H[\rho] \psi_i(x) = \left( -\frac{1}{2} \Delta + \int dx' \frac{m(x') + \rho(x')}{|x - x'|} + V_{xc}[\rho] \right) \psi_i(x) = \varepsilon_i \psi_i(x) \]

\[ \rho(x) = 2 \sum_{i=1}^{N/2} |\psi_i(x)|^2, \quad \int dx \psi_i^*(x)\psi_j(x) = \delta_{ij}, \quad \varepsilon_1 \leq \varepsilon_2 \leq \ldots \]

**GISAXS (ALS/MATH)**
Grazing incidence small angle x-ray scattering

**Designer Materials (Molecular Foundry/Math)**

**X-Ray nanocrystallography (ALS/MATH)**
**CAMERA: End Goal: Released Software:** (camera.lbl.gov)

**HipGISAXS** ([http://camera.lbl.gov/software/hipgisaxs_software](http://camera.lbl.gov/software/hipgisaxs_software))
- Flexible grazing-incidence small-angle X-ray scattering (GISAXS)
- Distorted wave Born approximation
- Speed: graphics processors and multicore processors.

**PEXSI** ([http://camera.lbl.gov/software/pexsi](http://camera.lbl.gov/software/pexsi))
- Fast method for electronic structure calculation: Kohn-Sham DFT.
- Can regularly handle systems with 10,000 to 100,000 electrons.
- Achieves scalability on more than 10,000 processors.

**SHARP-CAMERA** ([http://camera.lbl.gov/software/sharp_camera_download](http://camera.lbl.gov/software/sharp_camera_download))
- Multi-GPU Accelerated Ptychography Software.
- Combines diffraction + microscopy + high performance GPUS
- Advanced acceleration algorithms for convergence and analysis
- Freely available, open environment for collaboration/customization

**QUANT-CT** ([http://camera.lbl.gov/software/](http://camera.lbl.gov/software/))
- Image enhancement, filtering, segmentation and feature extraction
- Currently on ALS beamline 8.3.2, multi GPU. Open source: FiJi plugin

- Analysis/assembly of crystalline porous materials.
- Geometry-based analysis of structure/topology of material void space
- Current users: EFRC Nanoporous Materials Genome Center, EFRC Materials Project, Bosch, SABIC and Samsung.
Outline of Talk

How did this start?
   History and Motivation

Why mathematics?
   Math/Data/Computing

What is being delivered?
   Four models for delivery

Where is it going?
   A DOE Resource
Background: LBNL/UCB Mathematics:
Long Standing DOE Program: (LBNL+UC Berkeley)

Example: Semiconductor Algorithms: Samsung, Intel, Motorola, Infineon, Synopsis...
Look broadly at mathematical needs of Office of Science facilities, starting with the ALS, Molecular Foundry, NCEM, Joint BioEnergy Institute (JBEI), and future facilities

Question: How can applied mathematics help facilities do More science More efficiently (users, materials, turn-around time...)?
Experimental facilities will be transformed by high-resolution detectors, advanced mathematical analysis techniques, robotics, software automation, and programmable networks.

- Detectors capable of generating terabit data streams.
- Computational tools for analysis, data reduction & feature extraction *in situ*, using advanced algorithms and special-purpose hardware.
- Increase scientific throughput from robotics and automation software.
- Post-processing: reconstruction, inter-comparison, simulation, visualization.
- Data management and sharing, with federated identity management and flexible access control.
- Integration of experimental and computational facilities in real time, using programmable networks.

DOE Facilities in 2025: More Data, More Users, More Discovery
Mathematics for accelerating the analysis of experimental data

**Now**

Computational tools for analysis, data reduction & feature extraction *in situ*, using advanced algorithms and special-purpose hardware.

**Later**

Post-processing: reconstruction, inter-comparison, simulation, visualization.

**Mathematics for each can be quite different:**

- What is the minimum/fastest computational model/algorithm that gives (at least some) useful information?
- Can you quickly determine if data is useful, not useful, or in between?
- Can you quickly do analysis and steer experiment to more optimal configurations or output?
- What is the maximal amount of information you can get out of the data?
- Can data be measured, processed, organized and displayed to help understand/suggest further experiment?
- Can data be transformed to initialize computational models, and output framed to complement experiment?
(a) Problems have not yet been “mathemeticized”.
(b) No “equations of motion”
(c) Deep connections between the science and math

To tackle these problems requires new mathematics that bridges across mathematical disciplines.

Fortunately, Applied Mathematics is Undergoing a Profound Transformation

Traditional walls between continuous math, discrete math, analysis, probability and statistics, topology, algebra, geometry .... are all breaking down.
Mathematics is what changes data into information
Challenges are growing:

More data, more resolution
More complexity
Less obvious relational linking
More noise
More false signals

... 

Mathematics is what changes data into information

Going to need mathematics more than ever...
**Goal:** Build the applied mathematics that helps *transform experimental data into understanding*

**Today:**  
Facilities data is time-consuming

**Tomorrow:**  
More data.  
More quickly.  
High resolution.

**Critical need:**  
 algorithms and analysis for understanding

**LBNL approach:**  
Focused teams of mathematicians/domain scientists

**New math to:**  
Guide and optimize experiments

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**Pilot Partners**  
ALS  
Foundry & NCEM

**Key:** Leverage state-of-the-art mathematics

- Spectral clustering
- Clique analysis
- Maximum likelihood estimators
- Hamilton-Jacobi solvers
- Computational harmonic analysis
- PDE-based image segmentation
- Voronoi methods
- Graph theory
- Representation theory
- Machine learning
- Discrete Galerkin methods
- Statistical sampling
- Bayesian analysis
- Optimization methods
- Discrete/continuous shape descriptors
- Mori-Zwanzig theory
CAMERA: Personnel
Who is working on this?

Advanced Light Source (ALS):
  A. Hexemer  (Beam Scientist/GISAXS)
  S. Marchesini (Ptychography)
  D. Parkinson  (Beamline Scientist, Hard X-ray tomography)
  D. Shapiro (Beamline scientist)

Molecular Foundry
  D. Britt  (Organic and Macromolecular Synthesis)
  J. Neaton (Electronic Structure)
  W. Queen (Inorganic Nanostructures)

National Center for Electron Microscopy (NCEM)
  P. Ercius (Scanning transmission electron microscope)

Computational Research Division (CRD)
  M. Haranczyk (Materials Design)  T. Perciano (Image Analysis)
  X. Li  (GISAXS/)  H. Krishnan (Image Analysis/HPC)
  L. Lin  (Electronic Structure)
  R. Martin (Materials Design)  CRD Mathematics Department:
  C. Yang (Electronic Structure)  J. Donatelli (X-Ray Nanocrystallography)
  D. Ushizima  (Image Analysis)  C. Rycroft  (Optimal Chemical Design)
  J.A. Sethian (Director)

• Opportunity: Steady stream of new Berkeley faculty/postdocs/grad students
What Does CAMERA Deliver?

(1) Codes that run locally on computers embedded at facilities.

(2) Remote browsers executing code locally running at facilities.

(3) Codes remotely run on data downloaded from facilities to supercomputer centers.

(4) Downloaded and run remotely.
“Long-distance Delivery”: SPOT-Suite
Led by C. Tull, LBNL

Towards an End-to-End Solution for Light Source Data, Analysis, & Simulation

• **Combining...**
  – scalable software systems.
  – HPC/HTC/network resources.
  – advanced algorithms & analysis.
  – advanced simulation.
  – realtime feedback.

• **Multi-division team:** CRD, ALS, Math, CAMERA, ESNet, MSD, & NERSC.

• **Extending to include** SAXS, μDiff, and Ptychography beamlines. Focus on in-situ, time-resolved experiments, new algorithms, data sharing & collaboration.

• **SC14 Demos include** LCLS, APS, NSLS datasets.

Research into generalizable real time workflows and metadata already yielding valuable insight for photon scientists.
## Who is using CAMERA’s deliverables?

<table>
<thead>
<tr>
<th>Software</th>
<th>Users and Details</th>
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<tbody>
<tr>
<td><strong>PTychography</strong></td>
<td>CXRO/SEMTEC, LLNL/NASA, UI Chicago, UC San Diego, UC Davis, UCB, McMaster, Stanford. ALS, BNL, F. Maia, Uppsala, BYU, multiple workshops, tutorials</td>
</tr>
<tr>
<td><strong>QuantCT</strong></td>
<td>Advanced Light Source Users, available on “shop floor”. Downloadable Fiji Plugin (world-wide user base).</td>
</tr>
<tr>
<td><strong>PEXSI</strong></td>
<td>Accelerated Kohn-Sham Density Functional Algorithms Embedded in SIESTA (<em>Spanish Initiative for Electronic Simulations with Thousands of Atoms</em>): Next: CP2K</td>
</tr>
<tr>
<td><strong>Zeo++</strong></td>
<td>Open-source package - <a href="http://www.zeoplusplus.org">www.zeoplusplus.org</a>. Zeo++: a default tool for two BES Materials Genome Centers (Nanoporous Materials Genome Center (Minnesota) and Center for Functional Electronic Materials (LBNL)) and LBNL EFRC for Gas Separations Roughly 200 registered users world-wide in both academia/industry (e.g. Bosch, Samsung)</td>
</tr>
<tr>
<td><strong>HipGISAXS</strong></td>
<td>ORNL, ANL, ALS, Molecular Foundry, numerous universities, …</td>
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AN OVERVIEW OF SOME OF THE WORK UNDERWAY

Application

Mathematical issues
New mathematics we needed to build and exploit

Software for Users

(Describe problem, emphasize new mathematics, describe deliverables)
SHARP
(Scalable Heterogeneous Adaptive Robust Ptychography)

Fast scalable methods for ptychographic reconstructions

S. Marchesini, D. Shapiro (Advanced Light Source)
H. Krishnan (LBNL Computing Sciences)
F. Maia (LBL/Uppsala)
H-T Wu (LBNL/Stanford, now Toronto)
Fundamental idea: combine:
- High precision scanning microscope with
- High resolution diffraction measurements.
- Replace single detector with 2D CCD array.
- Measure intensity distribution at many scattering angles

Each recorded diffraction pattern:
- contains short-spatial Fourier frequency information
- only intensity is measured: need phase for reconstruction.
- phase retrieval comes from recording multiple diffraction patterns from same region of object.

Pytchography:
- uses a small step size relative to illumination geometry to scan sample.
- diffraction measurements from neighboring regions related through this geometry
- Thus, phase-less information is replaced with a redundant set of measurements.

Lots of ptychographic equipment/codes throughout DOE, universities, world-wide
When does it (not) work?
(no convergence proof yet available for method)

Existing algorithms may have trouble converging on large data sets:
(iterative methods intrinsically operate by interchanging information between nearest neighbor frames (diffraction patterns) at each step, so it might take many iterations for frames far apart to communicate.)

Effects of noise and physical uncertainties:
(how do reconstruction algorithms perform with uncertainties in photon statistics, lens perturbations, illumination positions, incoherent measurements, detector response and discretization, time fluctuations, etc.)

What is the best lens and illumination scheme for arbitrary specimens?
(given a detector, with a limited rate, dynamic range and response function, what is the best scheme to encode and extract more information per detector channel?)
Phase retrieval in high dimensional space

Tackling Efficiency: Challenges with basic alternating projection algorithm:

**Poor scaling:**
- Long range interactions among frames decay exponentially with distance.

**Poor initial guess:**
- Can significantly delay convergence.

Ultimately, an overdetermined problem in high dimensional space.

Short-time Fourier Transform

How can we speed this up?

Large dimensional data

Low dimensional space
Building a better starting guess:

(1) View every pixel of every frame as a dimension. Each data point lives on a torus (complex plane)

(2) Build “relationship network RN: a graph (V,E) that relates each frame to its neighbors.

(3) Construct Graph Laplacian of RN: defined as difference between the degree matrix D and the adjacency matrix A: GL = D - A

(4) The largest eigenvector of the Connection graph provides the most aligned phases encoding the (approximate) data topology.

This provides a strong starting guess.
Code: Scalable Ptychography Solver

**Code: Open source, downloadable package**
- Release
- Prototype
- Under way/testing

- Scalable code, (source package, remote interface, web interface, API).
- Real time feedback by reducing latency
- 80x speedup with algorithms
- 30x speedup with GPUs
- >16x speedup with distributed GPU

- Optimal Network fabric design for throughput
- Optimal lens design for SNR

- Iterative tomography (network/bandwidth optimized)
- Chemical mapping (robust PCA/SVD)
- Dynamics

**“Compute design”**

SHARP real time specs:
- 3D torus p2p fabric
- CCD/RDMA streaming
- Instrument calibration

**Intercalation Battery Research:**
Mechanisms in Lithium Ion Phosphate
ALS BL 5.3.2 (Nat. Phot. /in press)

**Partners:**
CXRO/SEMETEC, LLNL/NASA, UI Chicago, UC San Diego, UC Davis, UCB, McMaster, Stanford. ALS, BNL, F. Maia, Uppsala, BYU

**Software presentations:** Ptycho 2013, FIO/LS, SIAM IM14, MSPPR, XRM, Coherence 14

**Software tutorials:** Coming: SSRL/CAMERA xx/2014, CAMERA/ALS/BNL AUG 2014
CAMERA/ALS/APS Sep 10/14, COHERENCE, XRM, SIAMIM, FIO/LS, RACIR summer school, ALS Users workshop
Toward real-time feedback

Currently
the user interface starts processing at the end
of a full scan. (1 minute each)

In the future
low Latency (<5 ms) feedback by streaming
detector frames on distributed direct memory
access fabric.

Real time enables smart self-calibrating, auto-
tuning feedback of the microscope control
system.

Two Weeks Ago: Finished a prototype “Real
Time” version-code directly off of CCD
QuantCT

Automatic image analysis tools for micro-CT

D. Ushizima, D. Morozov, H. Krishnan, T. Perciano (LBNL Computing Sciences)
D. Parkinson (Advanced Light Source)
Goal: Develop algorithms for 3D/4D quantitative analysis of experiments, addressing challenges posed by noise, artifacts, sheer size, and heterogeneous materials.

Analyze structure: porosity, pathways, interior voids, ...

- Application: High-resolution synchrotron-based X-ray absorption microtomography.
- Suitability of materials and biomineralization processes for carbon sequestration.
- Acquire projection views at equi-spaced angles: produce 2D cross-sections.
- Gray level value of image voxels reflects x-ray attenuation and density.
- Compute pathways through materials:

Imaging Pipeline Requires:

- Filtering: remove noise, sharpen contrasts (bi-lateral and non-linear filters)
- Segmentation to isolate, and extract shapes from images (PDE-VIIM methods)
- Feature detection/analysis (Reeb graphs, topological analysis, channel detection)
QuantCT: Timeline of Mathematics/Algorithm Development

2011

Filtering of microCT
- Gaussian
- Median
- Bilateral
- Anisotropic diffusion
- Non-linear tensor PDE

2014

Segmentation of (near) homogeneous regions
- Thresholding (local/global)
- Variational Level Set Methods
- Fast Marching Methods
- Statistical Region Merging
- Voronoi Implicit Interface

Analysis of microstructures
- Porosity
- Intensity descriptors
- Topological descriptors
  - Pore network
  - Max Flow curves
  - Slope of max flow
  - Persistent pockets

Get rid of artifacts
Get structure from clutter
Get parameters from grains
(1) **Mumford-Shah functional** for image segmentation of two phases  
(index i indicates separate phases, Find interface G to minimize E)

\[ E(I, I_1, I_2) = \int_A (I(x,y) - I_1)^2 \, dx + \int_B (I(x,y) - I_2)^2 \, dx + mg \int G(s) \, ds \]

(2) **Becomes PDE transport method using level set methodology:**

\[ t + F \cdot \nabla |\nabla| = 0, \text{ where } F = \left[ ((I - I_1)^2 + (I - I_2)^2) \cdot \nabla \cdot \left( \frac{g \nabla}{|\nabla|} \right) \right] \]

(3) **New approach: Extend the Mumford-Shah energy functional to multi-phase multi-interface**

**Voronoi Implicit Interface Method (VIIM)**

\[ F_i = \left[ ((I - I_i)^2) \cdot \nabla \cdot \left( \frac{g \nabla}{|\nabla|} \right) \right] \]

(combination implicit embedding plus dual Eikonal Voronoi reconstruction)

(4) **Allows simultaneous extraction of multiple structures in 3D.**

Calcite precipitation: "pore clogging"
Augmented Topological Descriptors: Max Flow Graphs and Persistence Diagrams

Max-Flow:
- Reeb graph: Evolution of level sets of function on manifold.
- Use to detect pathways for particle of size $\alpha$
- Edge capacities = Intersection area between slices
- Flow between source/sink without exceeding capacities
- Family of graphs: Vary $\alpha$

Persistence Diagram:
- Track components in superlevel set of distance function
- When component merge: "younger" component merges into "older" component

Ford Fulkerson

Pocket distribution from persistence diagram
QuantCT: Results

Pore network through porous material

software for microCT analysis (0.33 images/s)

Automatic detection of 3D fibers and matrix cracking from assembled 2D slices
QuantCT: Workflow

Figure 1: Flow diagram of Quant-CT segmentation workflow: yellow indicates user-interaction event and blue indicates a program action.

QuantCT: Delivery Mechanisms

Delivery mechanisms:

Current:
(1) Browser/computer at ALS
(2) Available as FiJi plugin
(3) Prototype source downloadable.

Code Specifics:

- Implemented in Java.
- Part of Fiji framework.
- Implemented in OpenCL.
- Called from Java code through JOCL.
- Dedicated thread assigned to each OpenCL device to handle multiple accelerators on any given node.
- Each thread requests unprocessed slices up to the maximum allowed by the hardware.

Fluctuation X-Ray Scattering

Mathematics for structure reconstruction

J. Donatelli, J.A. Sethian (LBNL Computing Sciences)
P. Zwart (LBNL Advanced Light Source and Physical Biosciences)
Fluctuation X-Ray Scattering  
(Brand-new work)

(1) Fluctuation X-Ray Scattering:  
- Extension of Small- and Wide- angle X-Ray Scattering  
- X-Ray snapshots taken below rotational diffusion times  
- Significantly more experimental information than traditional techniques  
- Powerful technique for modern synchrotrons and free electron lasers (FEL)

(2) Going from real space structure to fluctuation scattering data is straightforward.

(3) But the reverse “inverse” problem is tough.

(4) A joint CAMERA collaboration between LBNL Physical Biosciences, ALS, and Computing Sciences has produced a new technique: “M-TIP”, which exploits multi-tiered iterative phasing and solves this inverse problem.

(5) The new method figures out structure of objects that cannot be crystallized, at a far higher resolution than previously available (to appear: PNAS in few weeks)

(6) Example: Using M-TIP, was able to reconstruct 3D profile of pentameric ligand-gated ion channel (pLGIC), from Protein Data Bank entry 4NPP
CAMERA: Where is this going?
Reaching out:

Positive response from the community

BESAC Committee Presentation (June 2014)
Oak Ridge/SNS Joint NSRC Workshop (June 2015)
  “Big, Deep, Smart Data Analytics in Materials Imaging”
Brookhaven and NSLS (May 2015)
  Invited Seminar Talk
Argonne/APS Joint Workshop (March 2015)
  “Frontiers in Data, Modeling, and Simulation
LBNL/Advanced Light Source (Oct. 2014)
  “CAMERA Workshop on Real-Time Robust Ptychography”

Many software requests:
Download, use CAMERA codes
Requests for CAMERA to house, curate, and host algorithms and software across the light sources

New Joint Projects starting up:
GISAXS and extensions to neutron sources (ORNL/SNS)
  Ptychography (BNL, ANL)
Fluctuation scattering (LCLS)
Knowing **what to build, how to build it, and how to use it** requires close-knit, coordinated teams with many different skills.

With careful attention to mathematics and algorithms, we can build codes and software tools that can transform data into the information that users really want.

camera.lbl.gov