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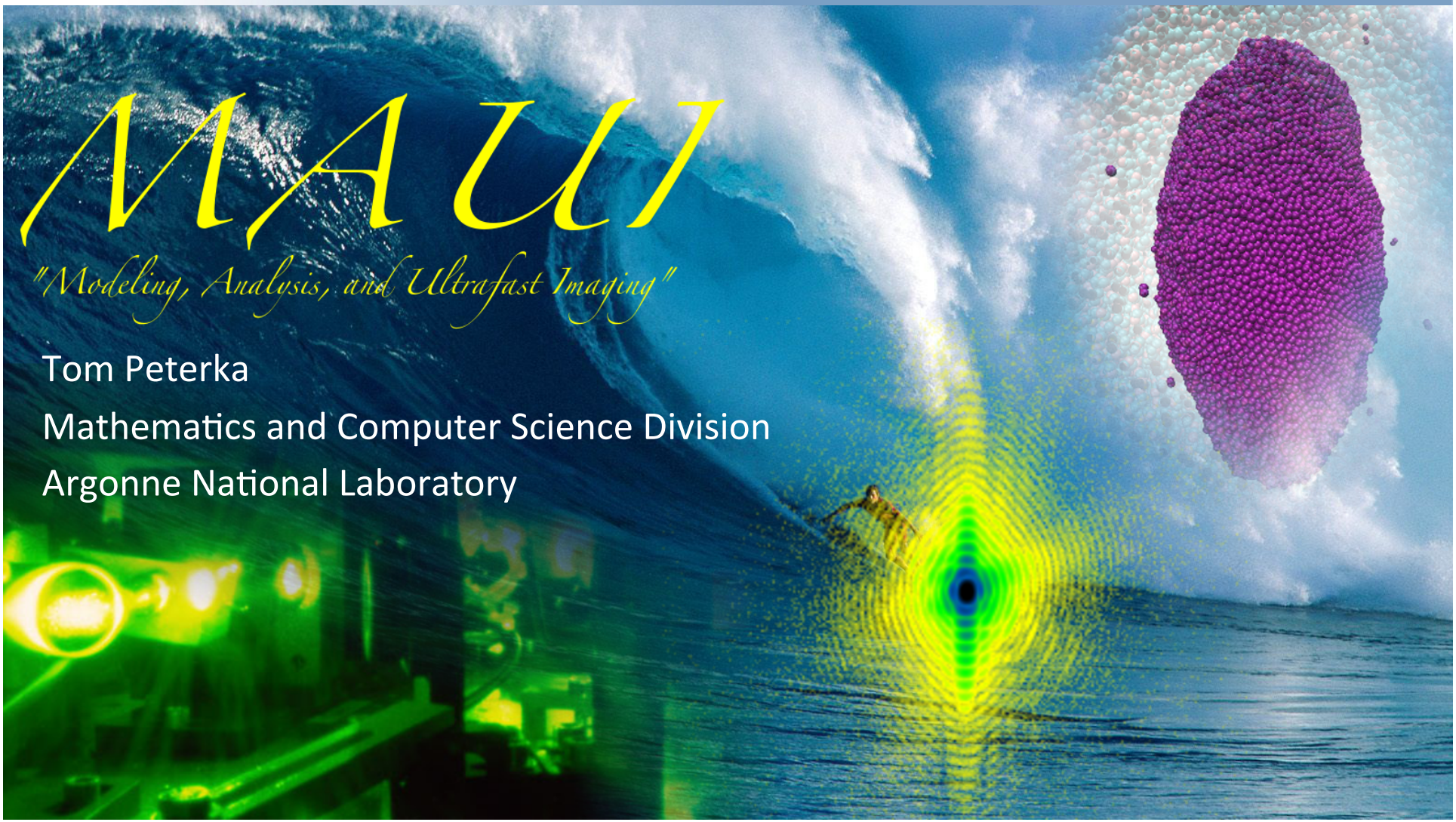


MAUI

"Modeling, Analysis, and Ultrafast Imaging"

Tom Peterka
Mathematics and Computer Science Division
Argonne National Laboratory

Integrated Imaging Institute Seminar
3/21/16

U.S. DEPARTMENT OF ENERGY

Team

Modeling

- Subramanian Sankaranarayanan (CNM)
- Kiran Sasikumar (CNM)

Analysis

- Todd Munson (MCS)
- Sven Leyffer (MCS)
- Nicola Ferrier (MCS)
- Youssef Nashed (MCS)
- Tom Peterka (MCS)

Ultrafast Imaging

- Ian McNulty (NST)
- Ross Harder (XSD)
- Haidan Wen (XSD)
- Mathew Cherukara (XSD)
- Andrew Ulvestad (MSD)

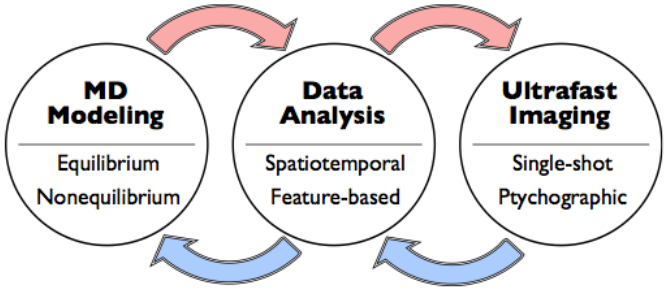
Objectives

technical

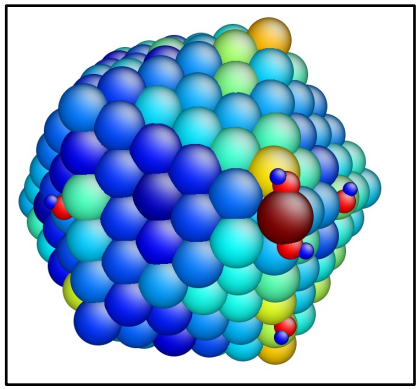
social

financial

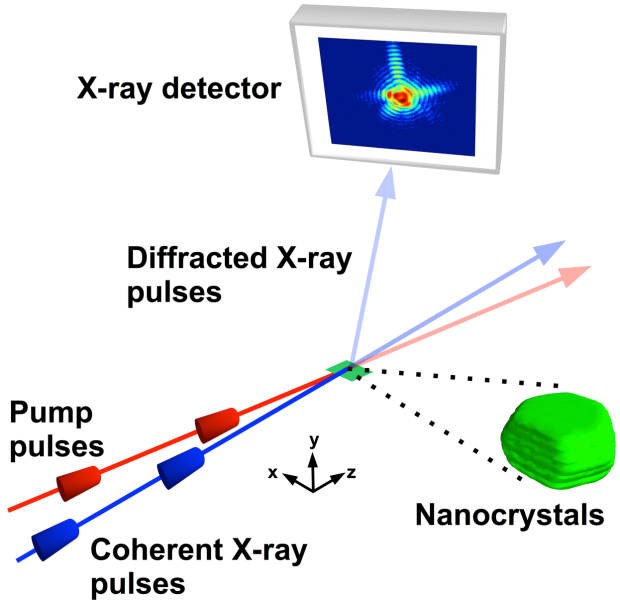
- Pump-probe experiments (Sector 7)
- Reactive MD (LAMMPS Reax)
- Understanding phonon breathing modes
- Understanding reaction-induced strain
- Lattice fitting
- Fast phase reconstruction
- Comparing results in different time/ length scales
- Workflow development
- Attracting new talent
- Learning a new language
- Attracting new funding



Simulation and experiment are integrated through data analysis.



Displacement of Au atoms after 20 ps in 0.55 MM ascorbic acid.



J. N. Clark, et al. *Science* 341 (6141): 56–59.

Phonons: Sound Waves that Carry Heat in Crystal Lattice Vibration

- Heat dissipation of next-generation semiconductors
- Conversion of wasted heat into electricity in thermoelectric materials
- Electrochemical processes across liquid-solid interfaces in water

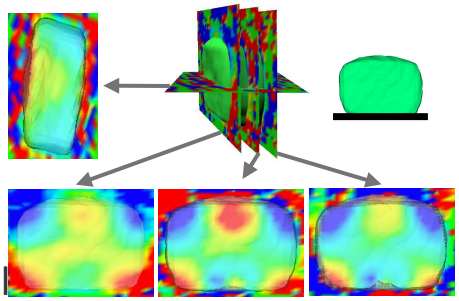
  - phase transitions
  - bond softening/hardening
  - ferroelectricity
  - solid/liquid interfaces
  - heat dissipation
  - phononic local structure
  - phase front propagation
  - spectrometry

http://water.org/water-crisis/water-facts/water/

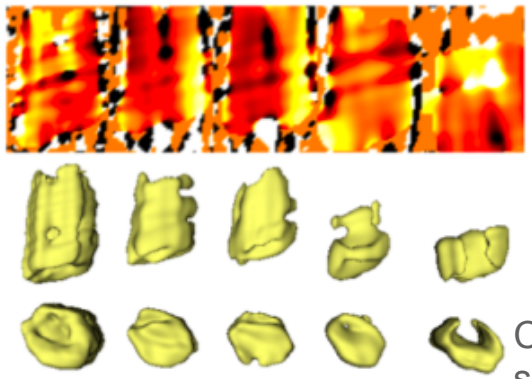
Coherent Diffraction Imaging: Applications

In-situ and operando characterization of materials evolution

- Mechanical response at nanometer length scales
  - Structural response to chemical reactions & in-situ catalysis
  - Decomposition of semiconductors in contact with noble metals
  - Structural changes in crystals due to defect formation and removal
  - Alloying and dealloying
  - High temperature and pressure
- Domain wall (magnetic, orbital, charge) structure in the complex oxides and multi-layer
- Phase transitions vs temperature and magnetic field



Cha, Wonsuk, et. al 2013
Nat Mater 12 (8) 729–34.



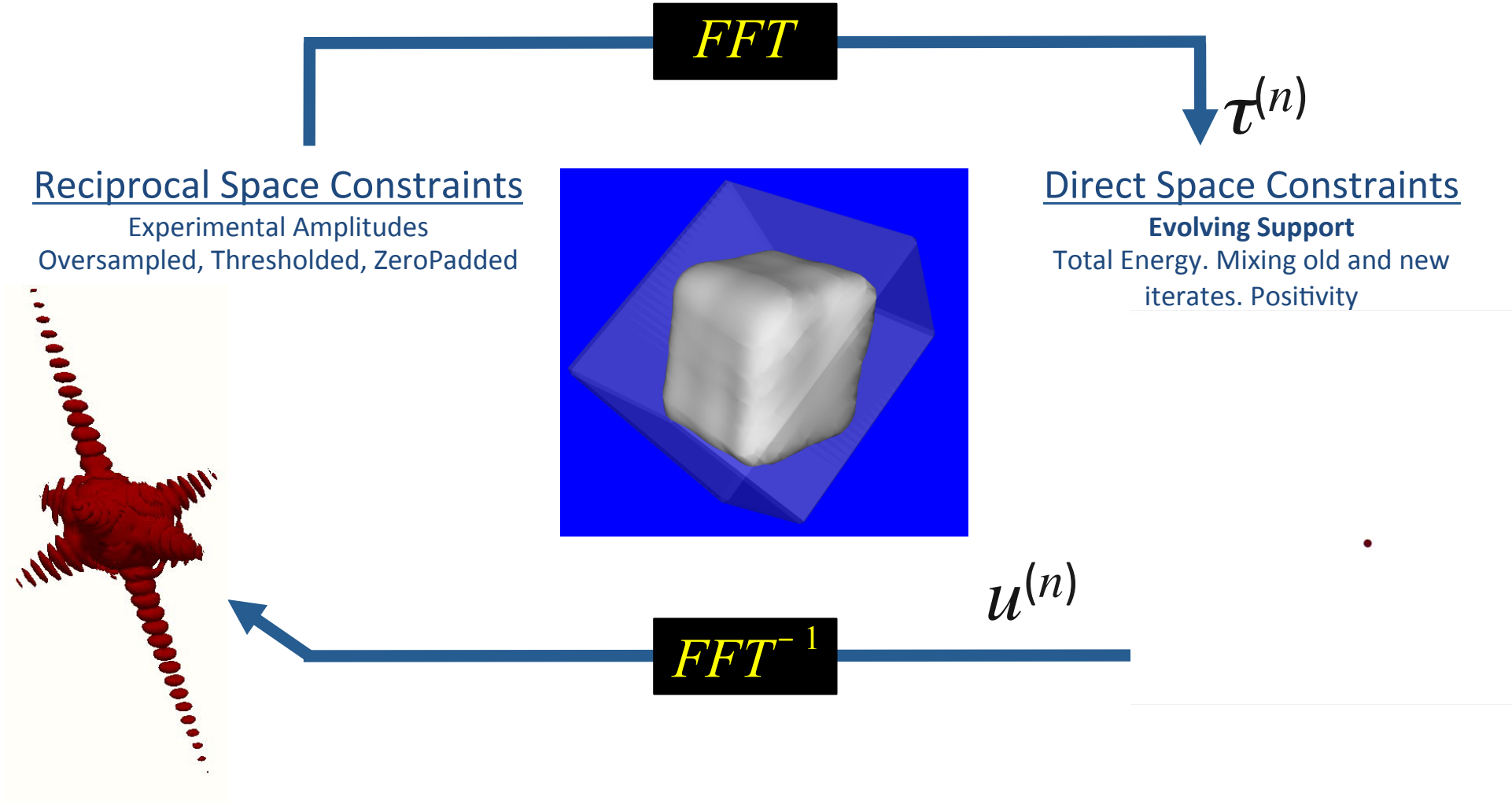
Coherent diffractive imaging of solid state reactions in zinc oxide crystals. Leake, S. J., Harder, R., & Robinson, I. K. (2011) *New Journal of Physics*, *13*(11), 113009

Watari, M., Harder R., et al. (2011). *Nat Mater*, *10*(11), 862–866.

chain thiols, the large stress cannot arise from the va
chain–chain interactions or other weak forces alone, bu
least ionic or covalent rearrangements. Indeed the Au–S
plays a crucial role in SAM formation: the structure o
(1.6 nm) thiolated nanocrystals has its Au–Au spaci
disrupted and sulphur intermixed with gold in the o
Our findings support this model and show strong th
deformations of our 300 nm crystals with strains penet
than 20 nm from the outer surface towards the crystal
strains are absent. The tight-radius spherical parts of
nanocrystals might also undergo strong Au–S inter
would indeed be able to provide sufficient stress.
reactions involving atomic diffusion of Au at room
would also be an attractive explanation of the relatively s
seen in both our X-ray and the cantilever experiments.

Our observation of relative contraction of the
expansion of the curved surface regions of Au r
illustrated in Fig. 3e, leads to the conclusion that the

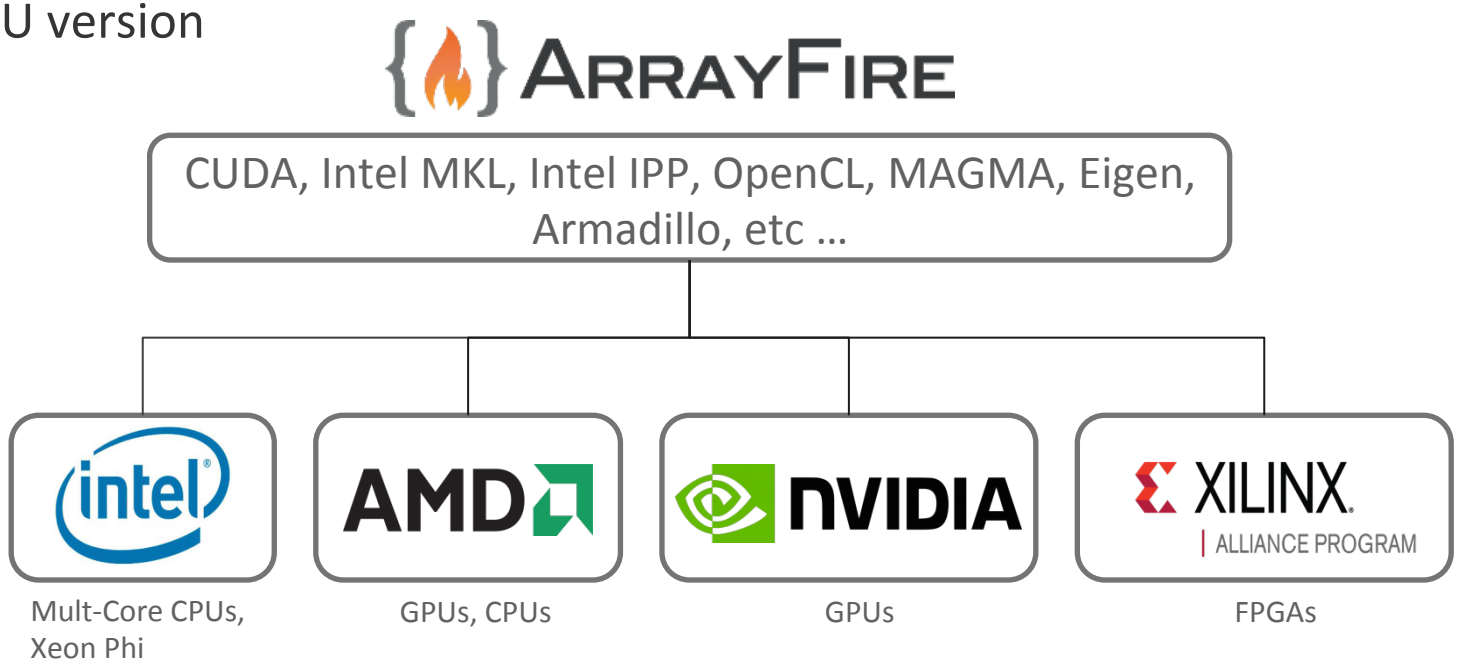
Phase Retrieval: Challenging and Costly Inverse Problem



1000's of iterations and multiple random starts required for high fidelity data!

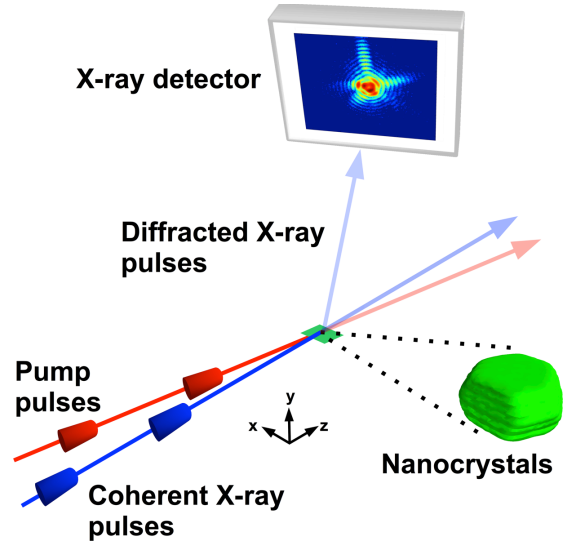
Fast Phasing Library (FPL)

- Group phase retrieval methods and tricks in one library
- Runs on various platforms (from user laptops to HPC systems)
- Hardware-independent code through ArrayFire
- Easy to customize, maintain and extend
- Currently over 100x faster than GPU accelerated MATLAB code, ~500X over the CPU version

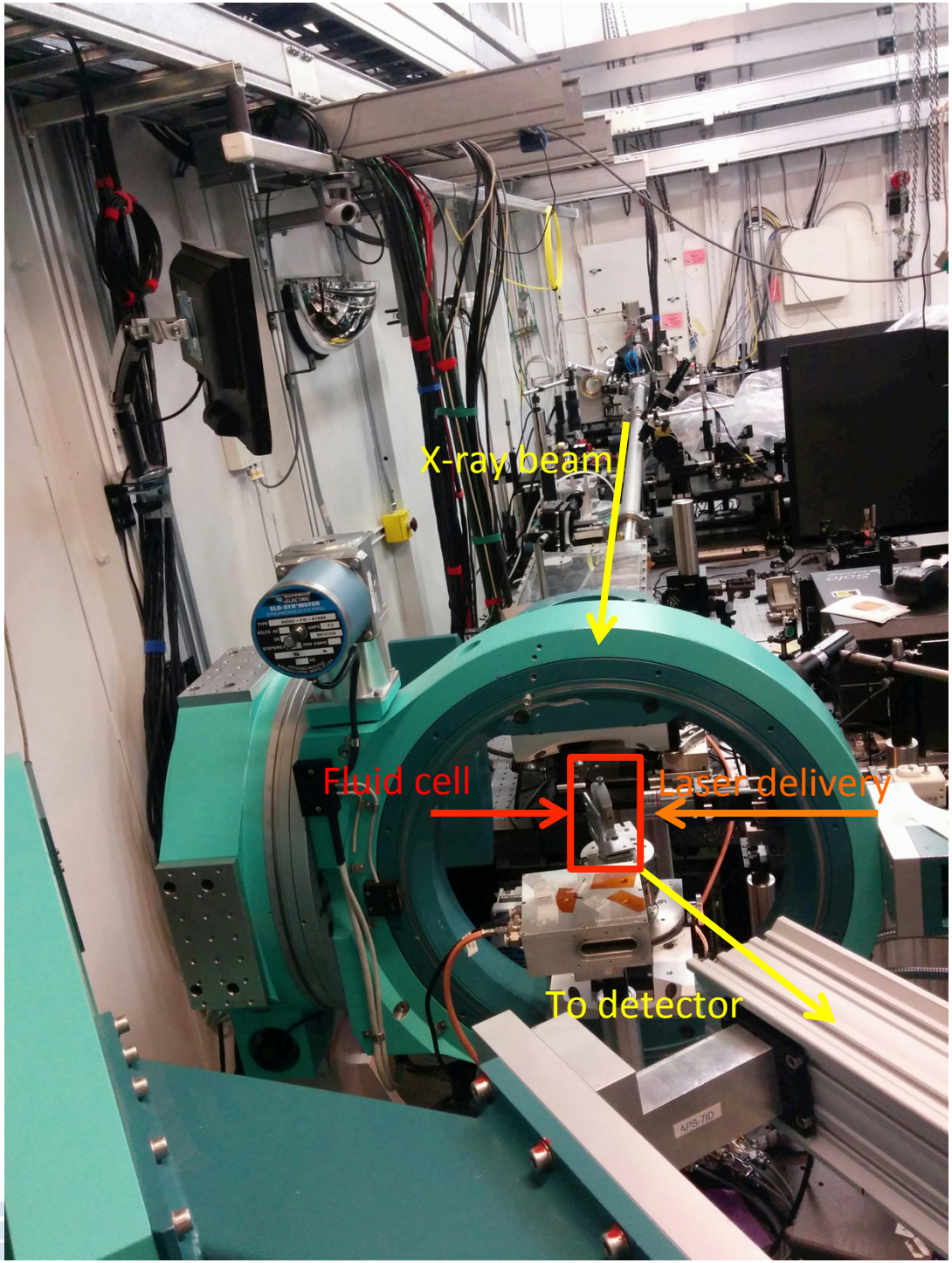


Time-resolved CDI

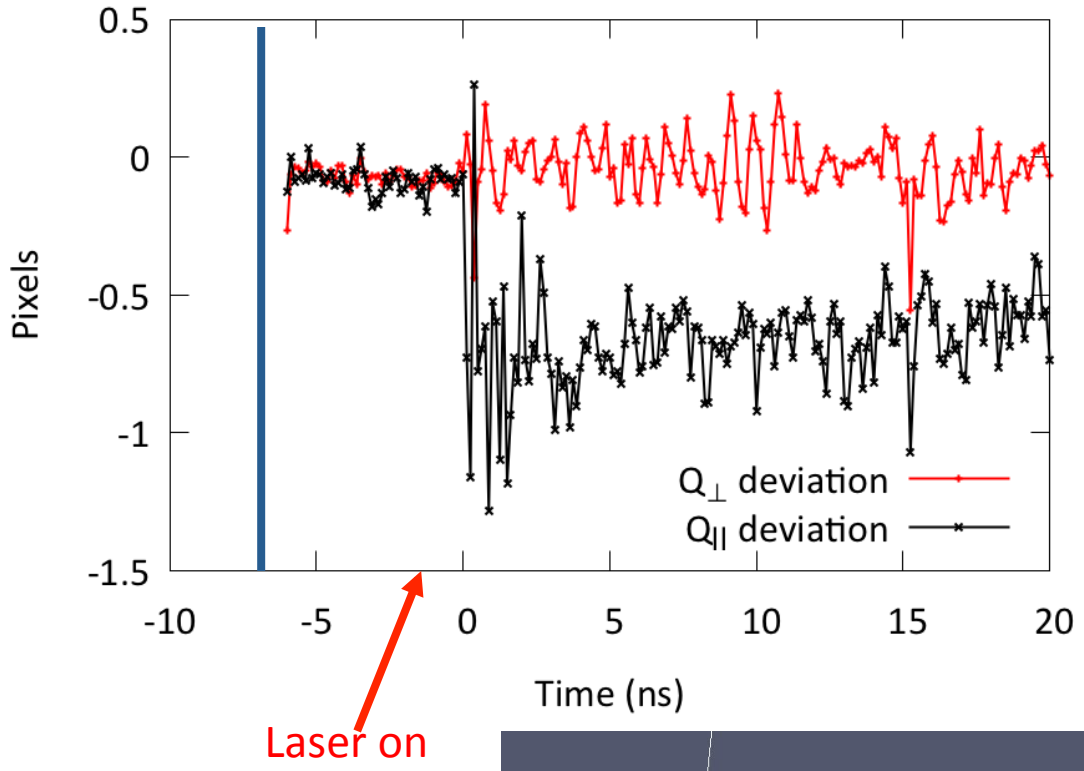
- Laser pump, X-ray probe at Sector 7
- Combine with CDI
- Provide dynamical map of excited modes in sample.
- <10 nm spatial resolution possible with 100 ps temporal resolution.

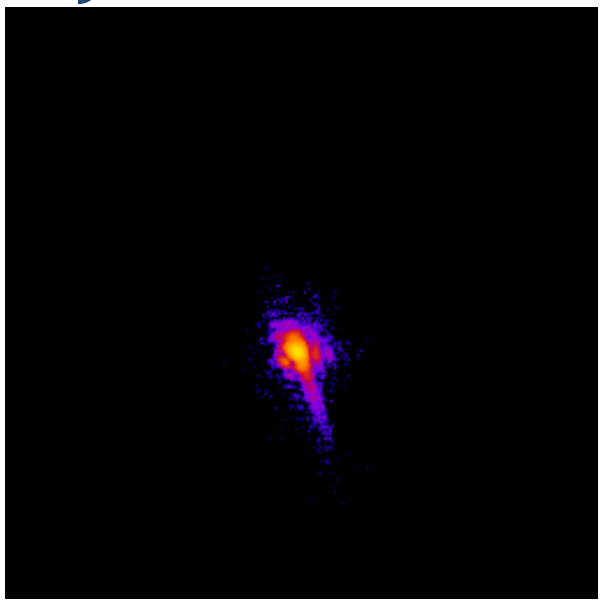


J. N. Clark, et al.
Science 341 (6141): 56–59.



Case Study 1: Dynamic Response of Au Crystals

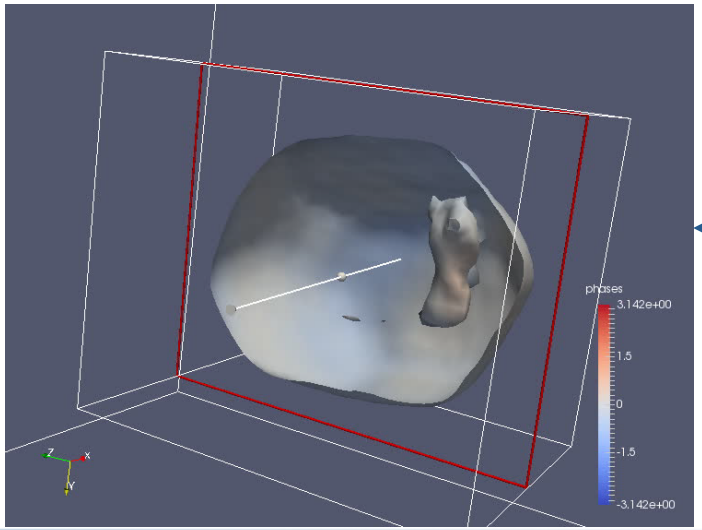




Reciprocal space view

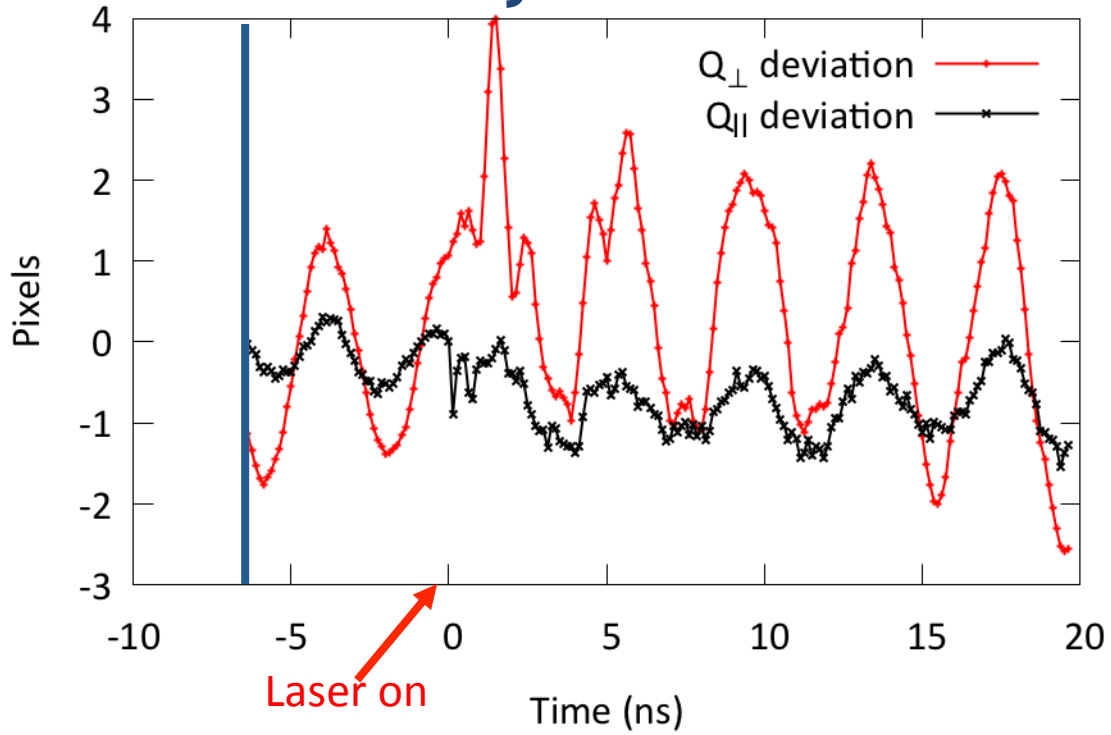
Phase retrieval at select times

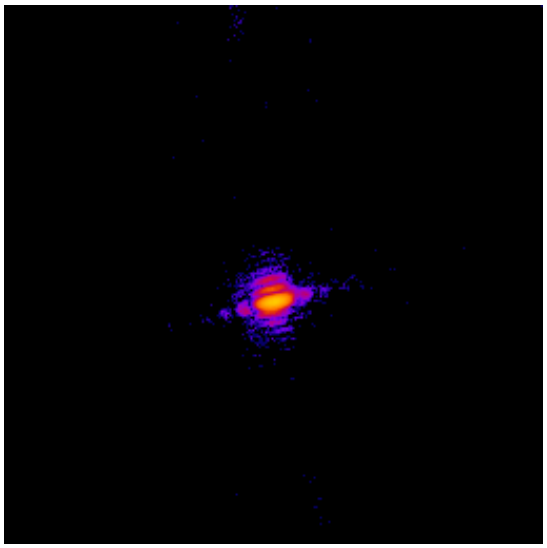
Need to measure the complete rocking curve at those points



Real space view

Case Study 2: Volumetric and Deviatoric Deformation Modes in ZnO Crystal

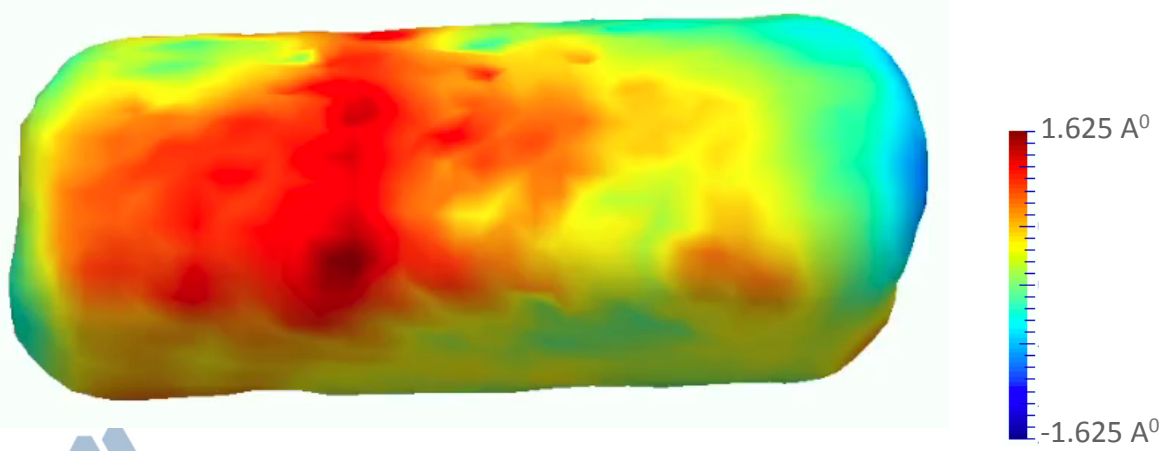




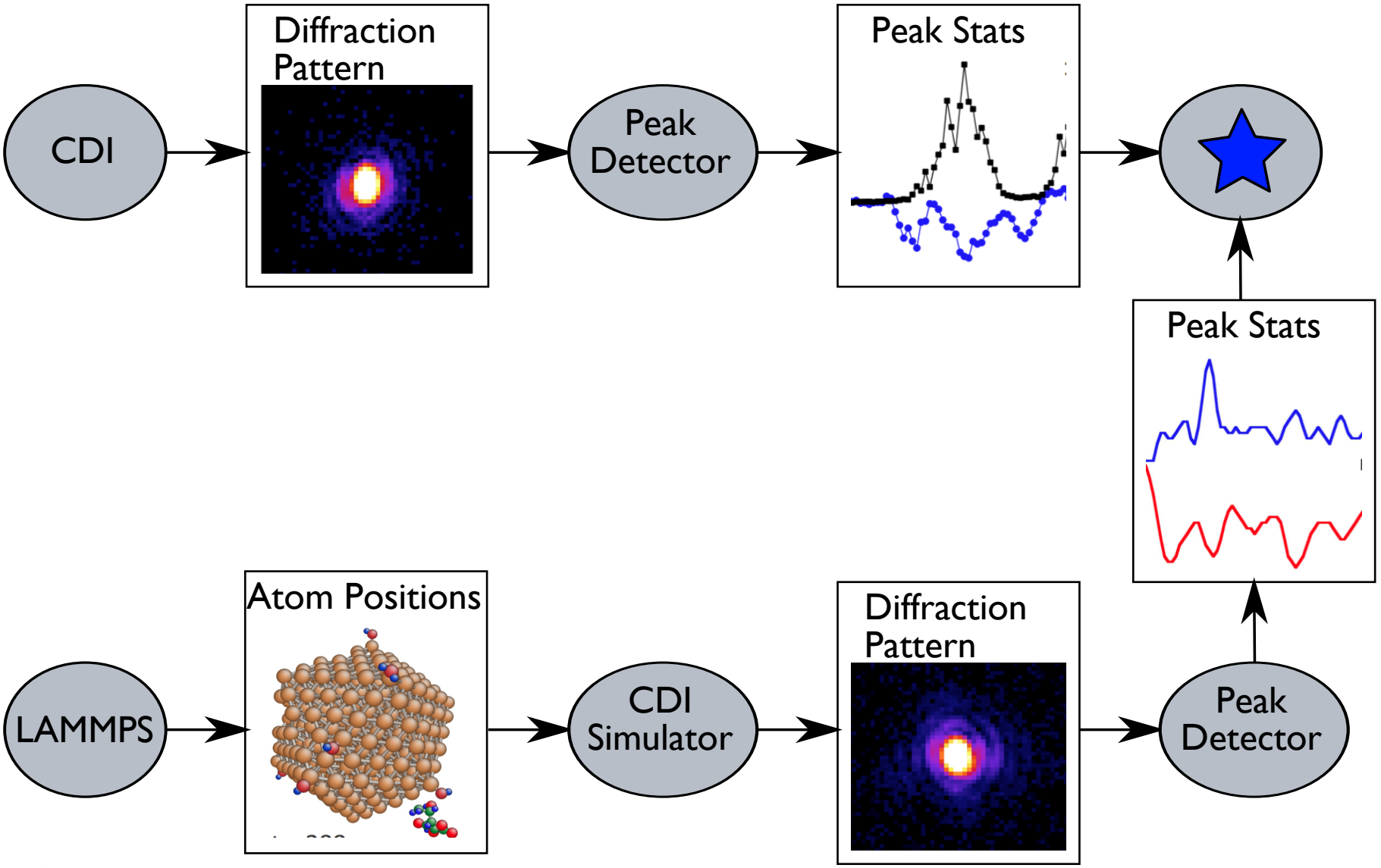
Reciprocal space view

Phase retrieval at select times

Need to measure the complete rocking curve at those points

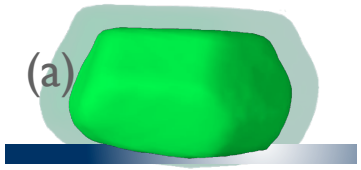


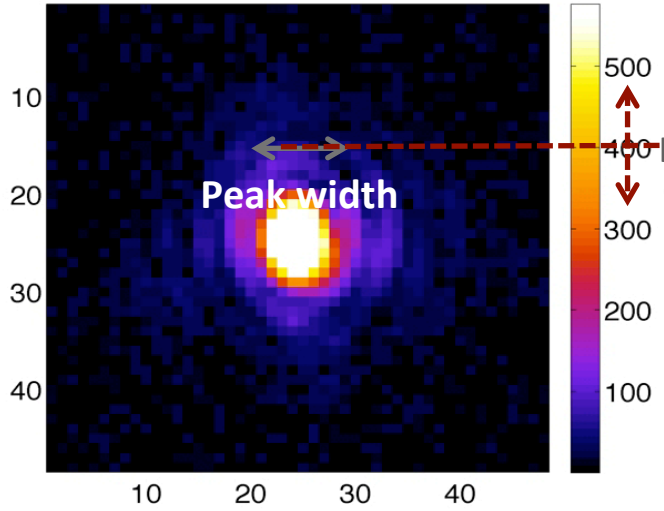
Reverse Problem Workflow

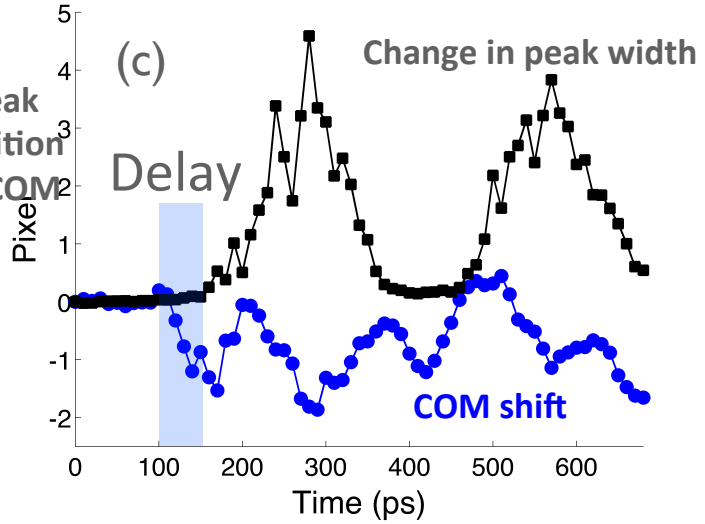


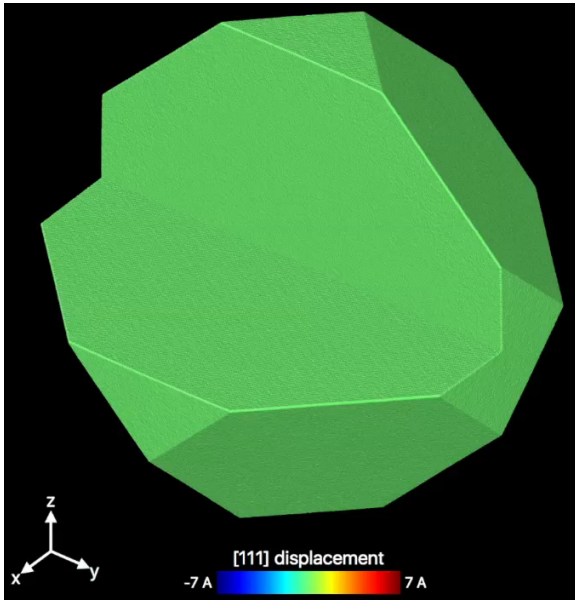
Case Study 3: Induced Shock Waves in Au Coated with Al

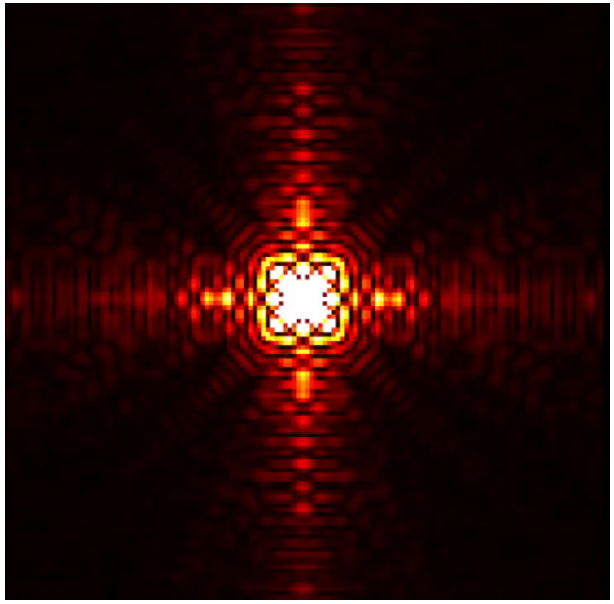
100 nm aluminum shell on 300 nm gold

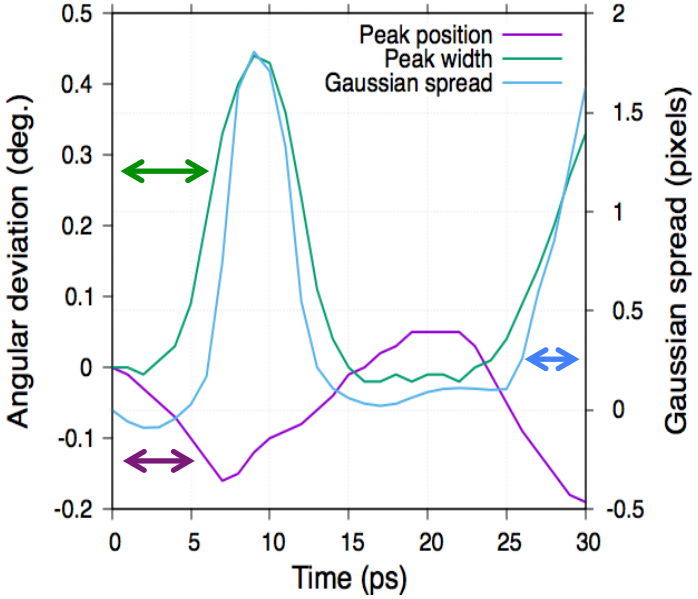






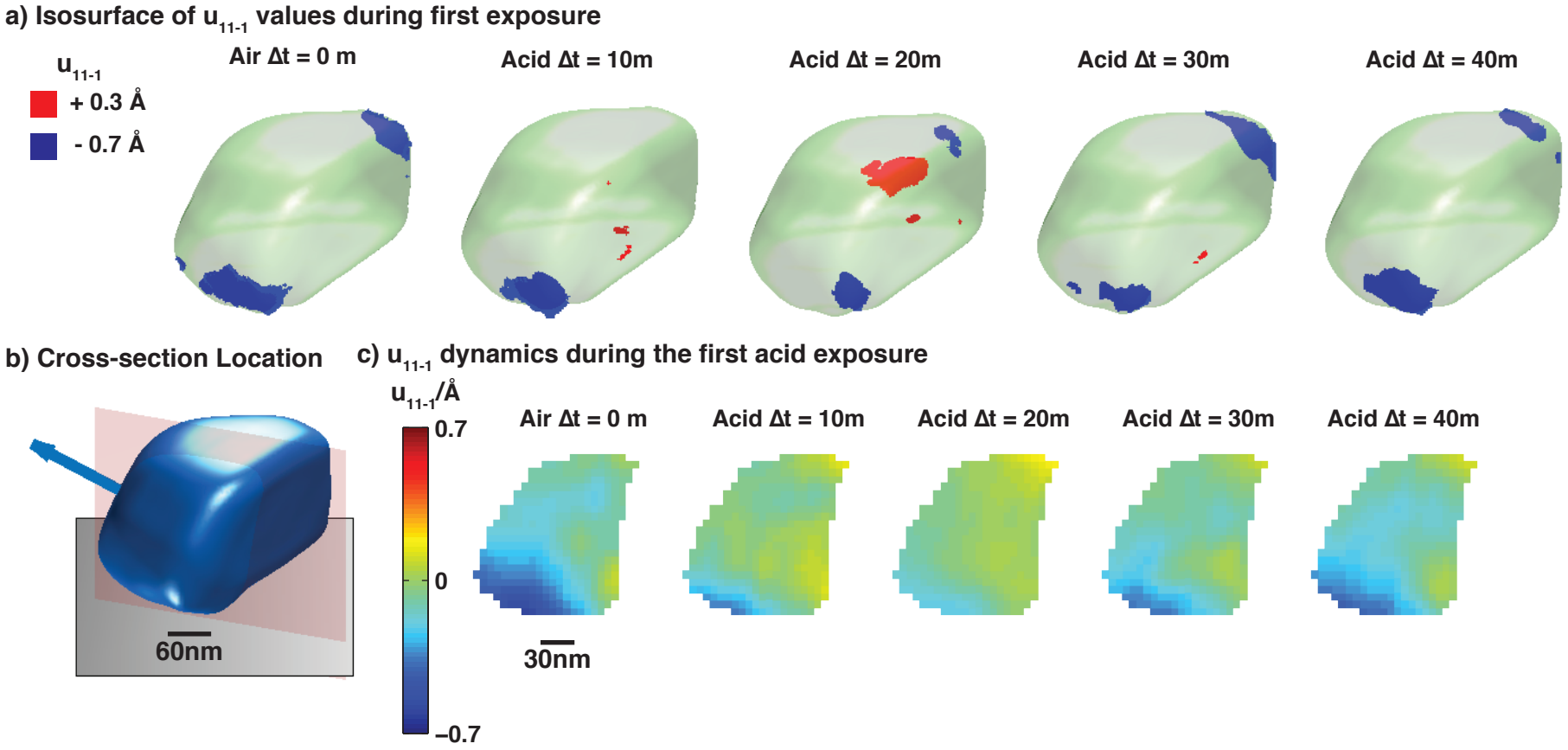






MD simulations → Calculated diffraction pattern

Case Study 4: Ascorbic Acid Decomposition on Gold

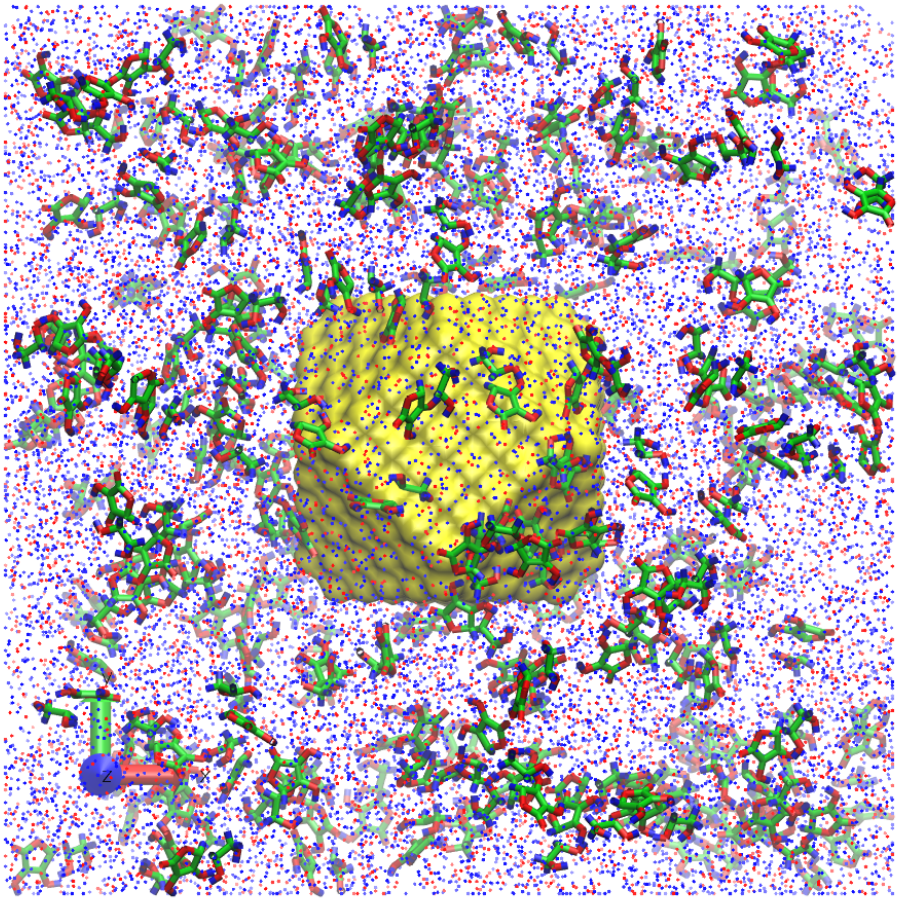


- Lattice change occurs at junctions involving the flattest facet
- Electron injection should create largest electric field at crystallographic discontinuities or apexes, providing “hot spots” for the reaction

Atomistic Simulation of Nanocatalytic Activity on Au Nanosurfaces

Sample schematic of simulated system

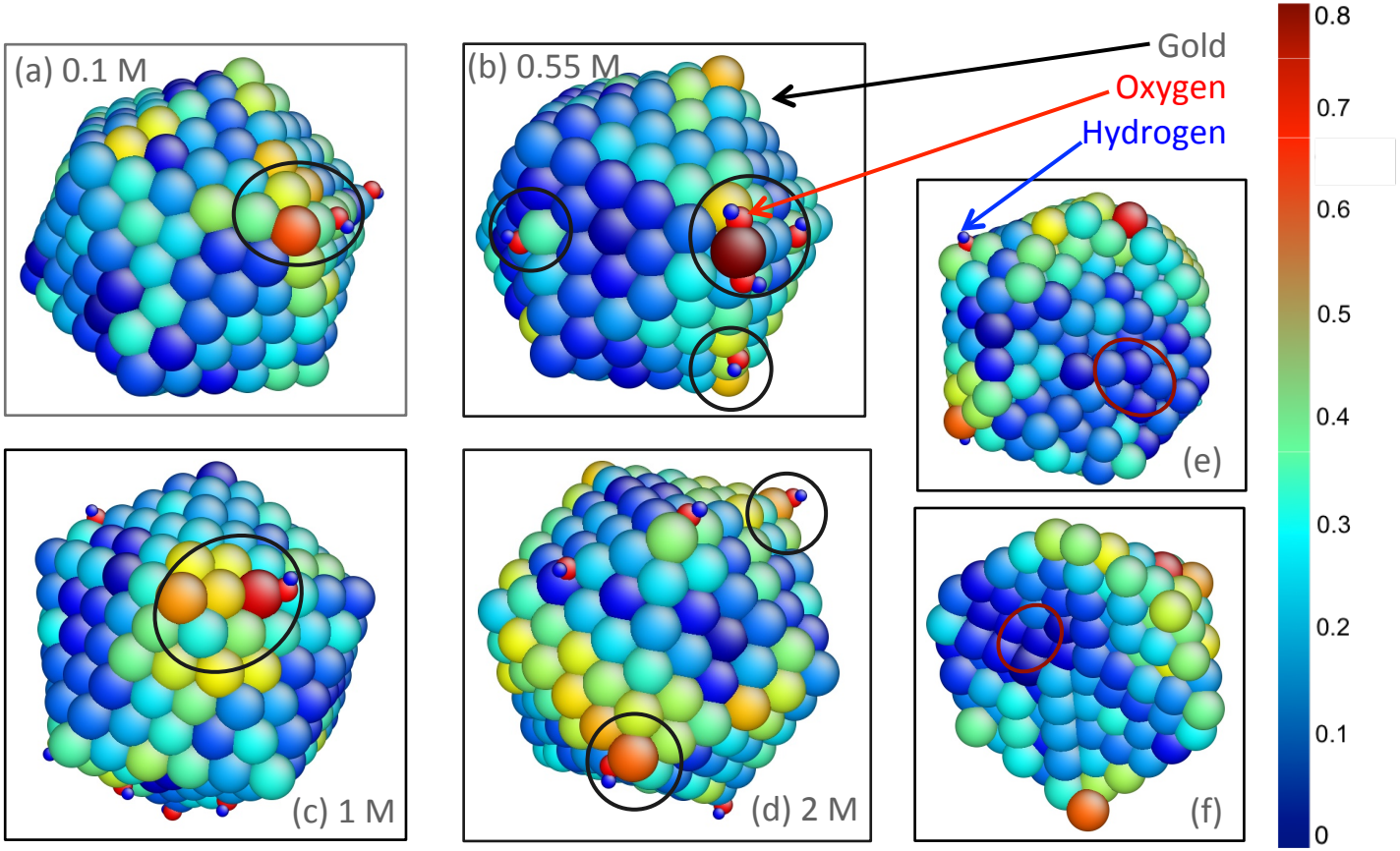
1M acid + water + gold (truncated octahedron)



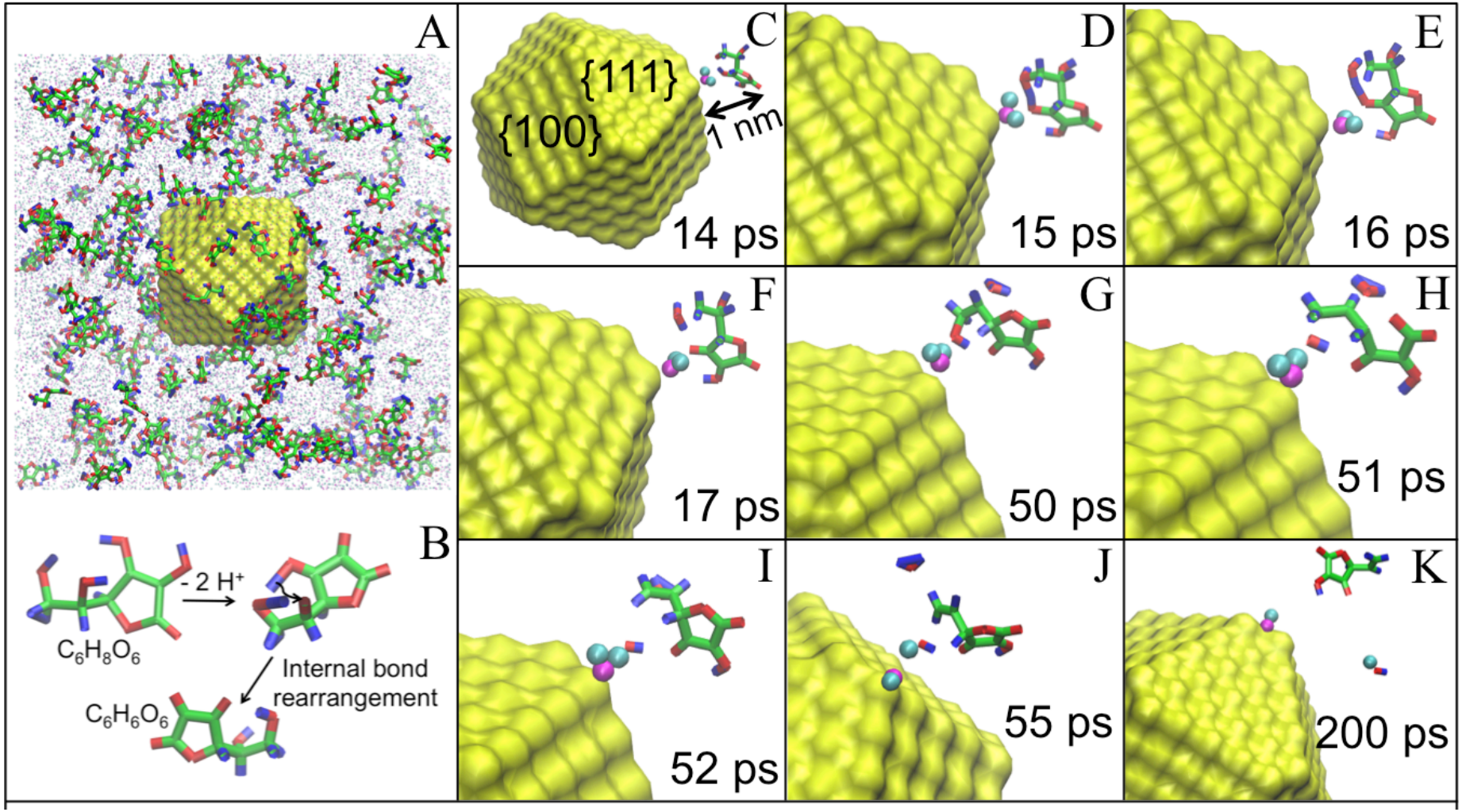
Yellow - Gold
Green - Carbon
Red - Oxygen
Blue - Hydrogen

Effect of acid molarity

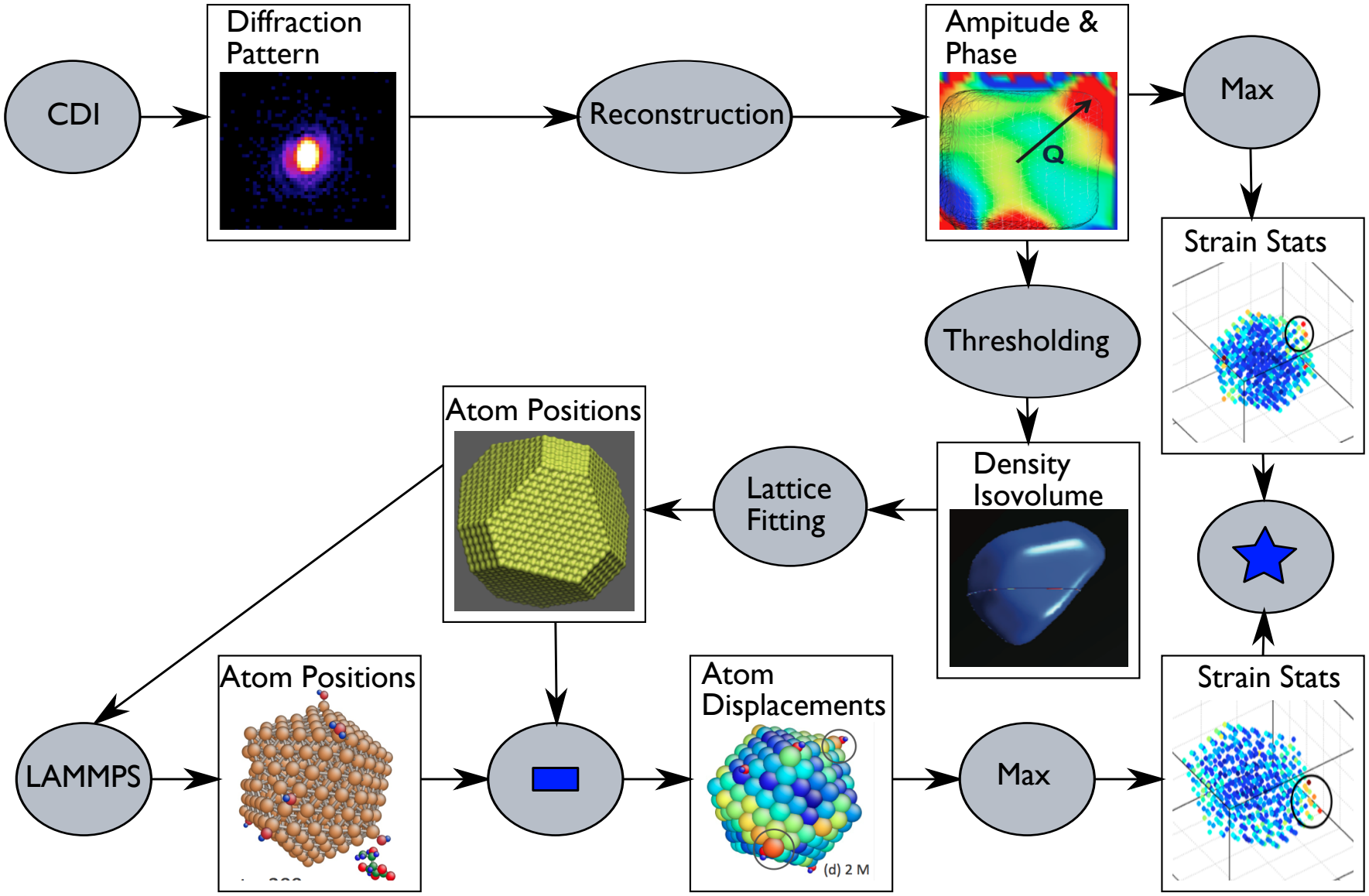
Displacement of Au atoms after 200 ps



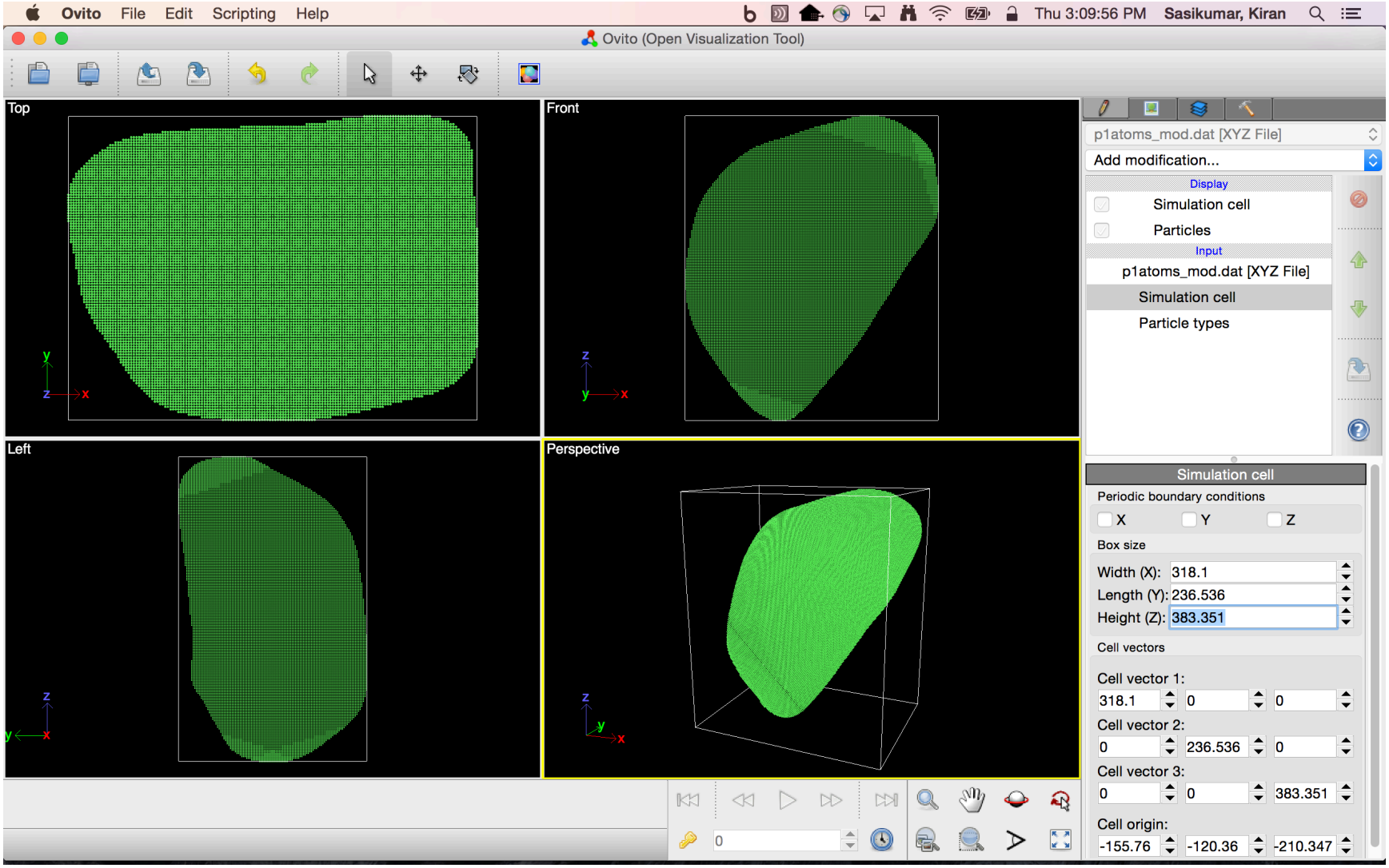
Adsorption pathway



Forward Problem Workflow

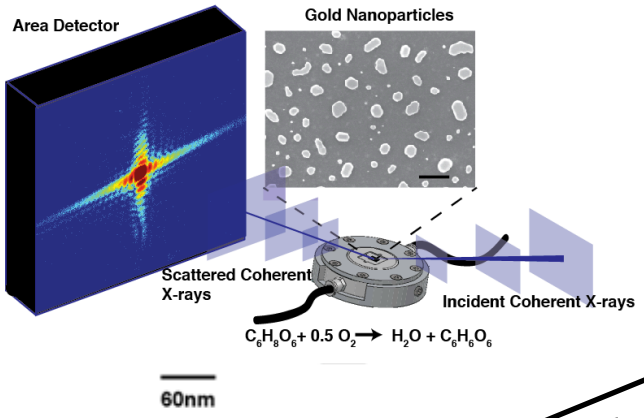


Lattice Fitting

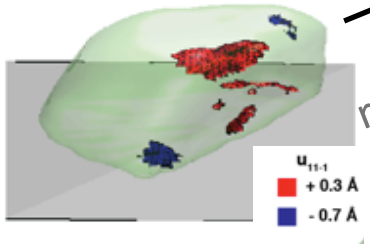


Modeling Ascorbic Acid Decomposition on Gold

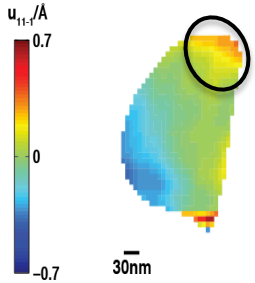
CXDI Experiment



Acid Δt = 20m

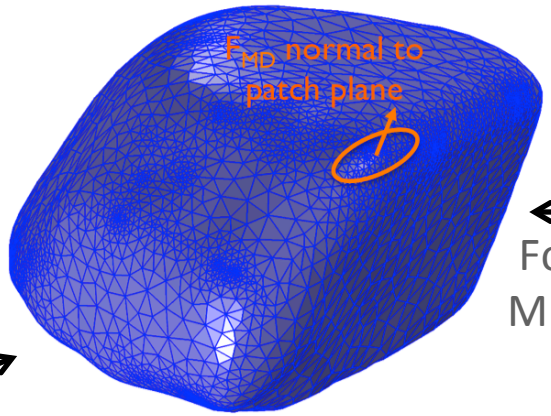


Experimental reconstruction as input to FEM



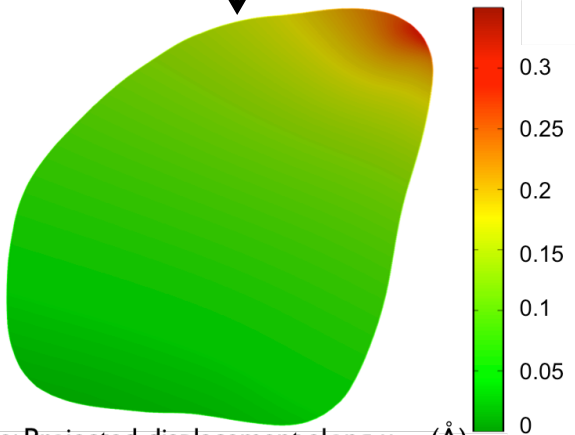
Experiment vs. Theory

FEM (COMSOL)



Minimum element size: 1 nm
Maximum element size: 20 nm

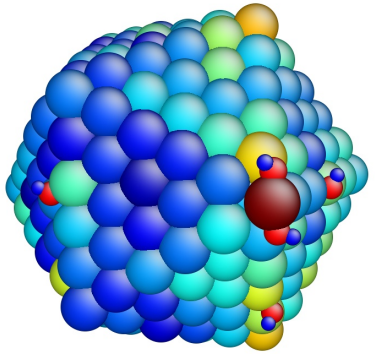
FEM Result



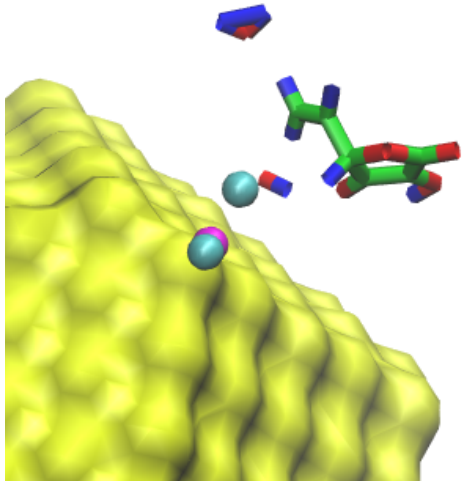
Central Slice: Projected displacement along u_{11-1}(Å)

Force from MD as input to FEM

MD (ReaxFF – LAMMPS)



Snapshot of Au and chemisorbed OH. Au colored by displacement from reference lattice.



MD also reveals underlying mechanism.

Food for Thought

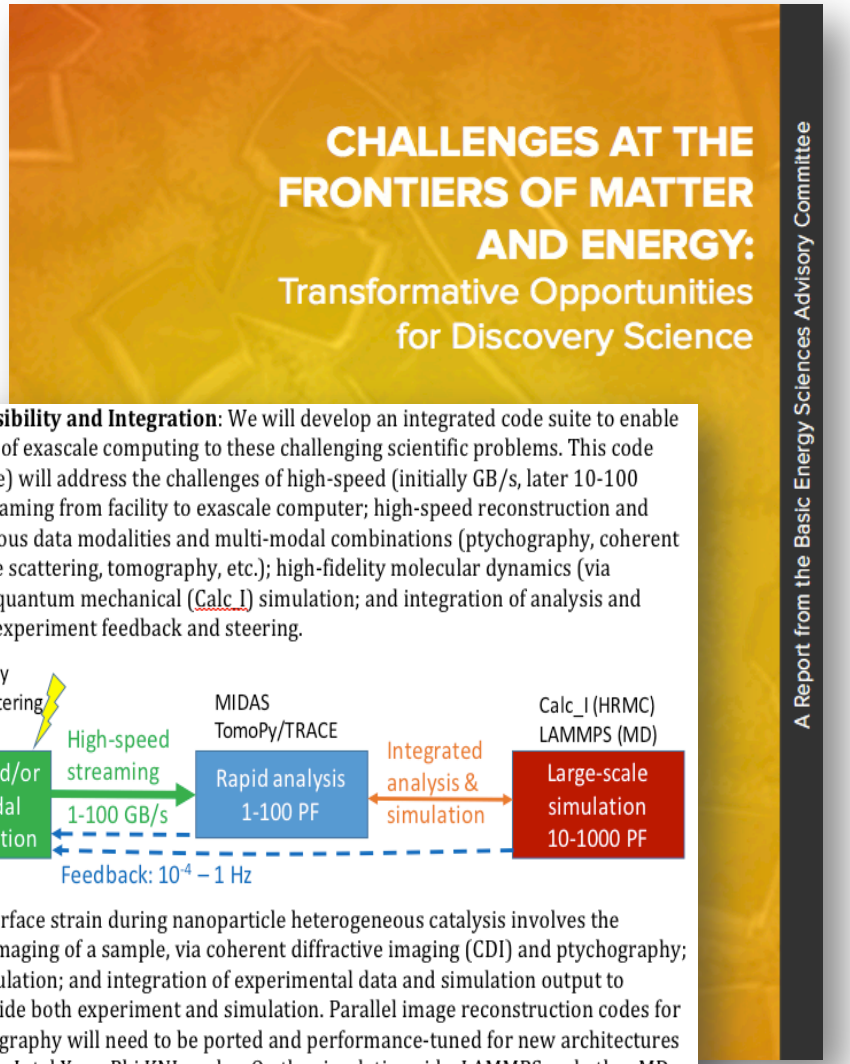
- Science challenges
  - Time and space scales
    - Experiment
      - 10-20 nm resolution, particle size > 100 nm
    - time resolution 100 ps at the APS, free electron laser at SLAC 1 ps, 100 fs laser pulse
    - Modeling
      - 1B atoms, 100 nm particle, 1 atom spatial resolution
    - time step 1 fs, 10s of ns duration
  - Comparing experiment with model
    - Measure different quantities or the same quantity differently
    - We can measure strain on one lattice vector; how do we do that in 3d? In atomic scale?
    - Various techniques are appropriate for different scale models (eg FEM instead of MD)
- CS challenges
  - Language barriers
  - Time and space scales
  - Workflows: automation, reproducibility

Food for Thought

- Next steps
  - Draft paper for submission to a journal
  - Complete analysis of systems we measured ZnO and AuAl, MD model those systems
  - Corrosion experiment, submitted proposal for beam time at 34 and compute time Cu in salt water
  - In situ reconstruction and analysis
  - Machine learning to fit measured diffraction pattern to database of diffraction patterns (AICDI LDRD)

Funding Opportunities

- Workshops: Workflows workshop, Experimental and Observational Data workshop, Rumored upcoming experiment + simulation workshop
- ECP
- BESAC future directions
  - "Beyond Ideal Materials and Systems: Understanding the Critical Roles of Heterogeneity, Interfaces, and Disorder" (e.g., corrosion)



Technical Feasibility and Integration: We will develop an integrated code suite to enable the application of exascale computing to these challenging scientific problems. This code suite (see figure) will address the challenges of high-speed (initially GB/s, later 10-100 GB/s) data streaming from facility to exascale computer; high-speed reconstruction and analysis of various data modalities and multi-modal combinations (ptychography, coherent imaging, diffuse scattering, tomography, etc.); high-fidelity molecular dynamics (via LAMMPS) and quantum mechanical (Calc_I) simulation; and integration of analysis and simulation for experiment feedback and steering.

Our work on surface strain during nanoparticle heterogeneous catalysis involves the simultaneous imaging of a sample, via coherent diffractive imaging (CDI) and ptychography; a large MD simulation; and integration of experimental data and simulation output to validate and guide both experiment and simulation. Parallel image reconstruction codes for CDI and ptychography will need to be ported and performance-tuned for new architectures such as Aurora's Intel Xeon Phi KNL nodes. On the simulation side, LAMMPS and other MD codes will need to include multiple levels of parallelism: MPI, OpenMP, and vectorization in order to fully utilize exascale architecture. Similarly, data analytics to compare simulation and experimental results will need to be done in situ with LAMMPS in order to process data as quickly as it is being produced. Software such as DIY and Decaf can be used to manage data movement within and between LAMMPS and analytics codes to compute lattice displacement and strain. Overall workflow orchestration can be accomplished with Swift.

Workshop on the Future of Scientific Workflows

MissionThe mission of this workshop is to develop requirements for workflow methods and tools in a combined HPC and distributed work environment to ena

to-end

Data Management, Visualization, and Analysis of Experimental and Observational Data (EOD) Workshop

The Data Management, Visualization, and Analysis of Experimental and Observational Data (EOD) Workshop is aimed at helping ASCR and the research community better understand data management, analysis and visualization needs related to user...

References

- Maui website (public)

  http://tpeterka.github.io/maui-project/

- Maui project site (private)

  https://bitbucket.org/tpeterka1/maui

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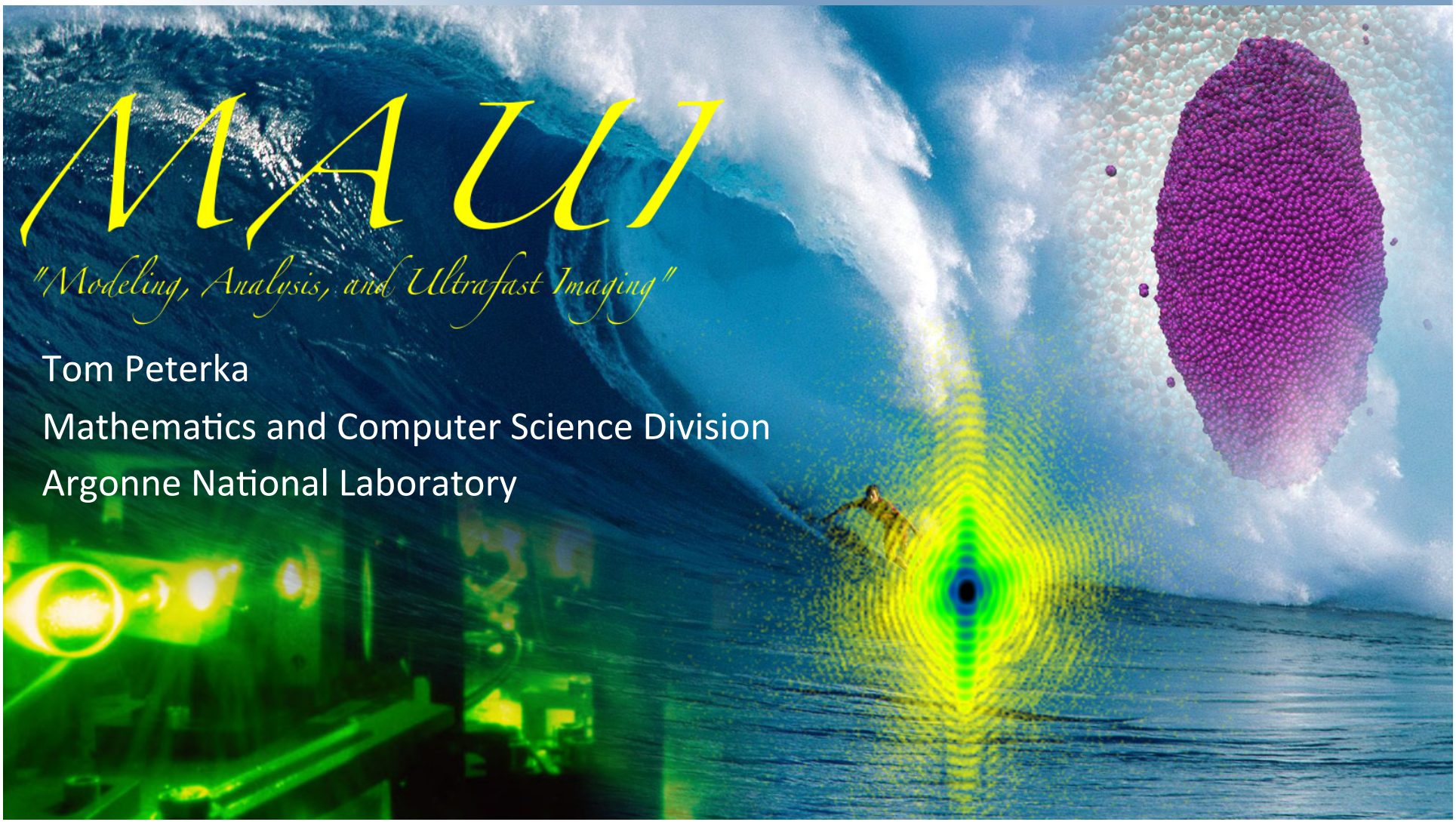

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