



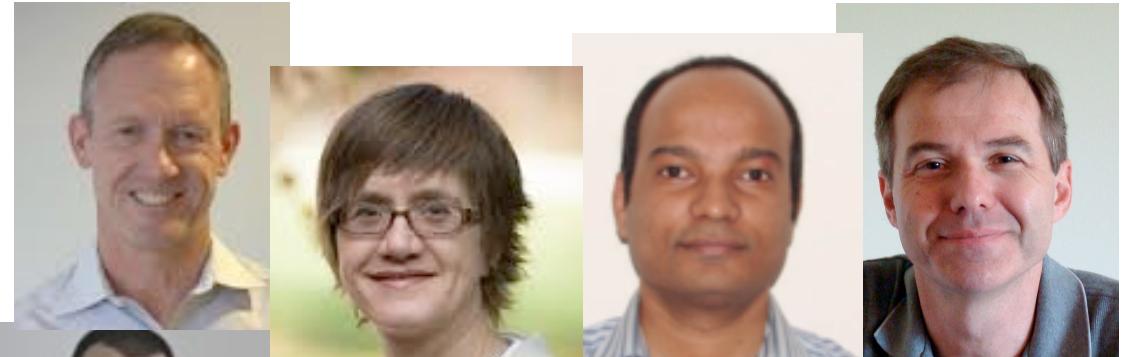
Tom Peterka
Mathematics and Computer Science Division
Argonne National Laboratory

Integrated Imaging Institute Seminar
3/21/16

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

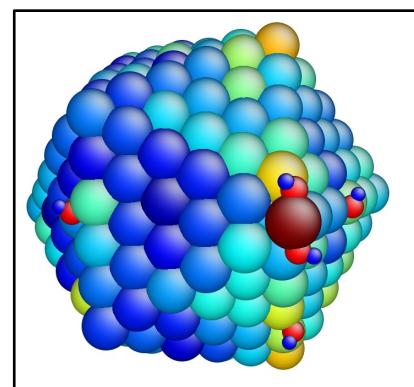
- Ian McNulty (NST)
- Ross Harder (XSD)
- Haidan Wen (XSD)
- Mathew Cherukara (XSD)
- Andrew Ulvestad (MSD)



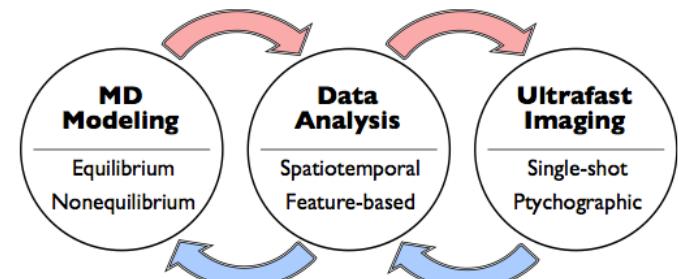
Objectives

technical
collaborative
financial

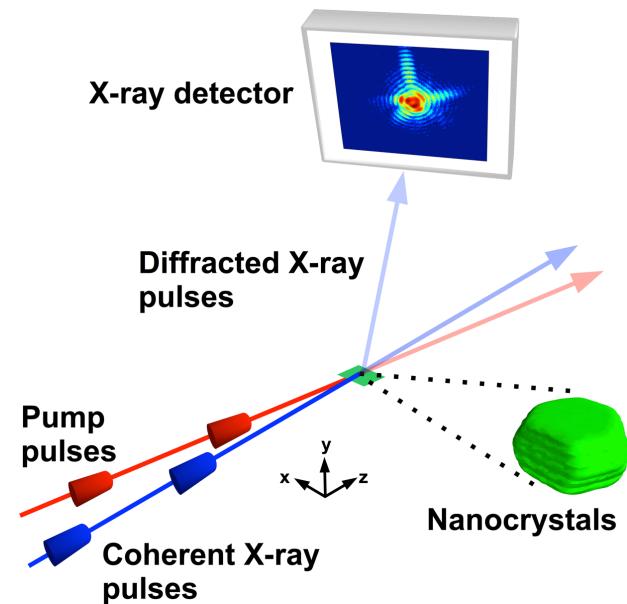
- Pump-probe experiments (Sector 7)
- Reactive MD (LAMMPS Reax)
- Understanding phonon breathing modes
- Understanding reaction-induced strain
- Lattice fitting
- Fast phase reconstruction
- Comparing results in different time/
length scales
- Workflow development
- Attracting new talent
- Learning a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



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



Simulation and experiment are integrated through data analysis.



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



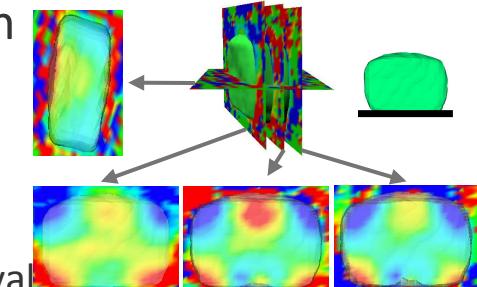
<http://water.org/water-crisis/water-facts/water/>



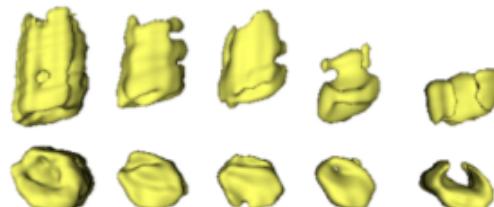
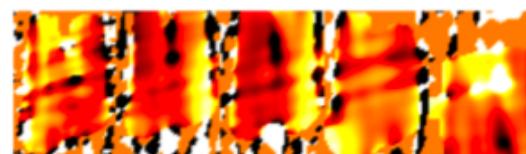
Coherent Diffraction Imaging: Applications

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.

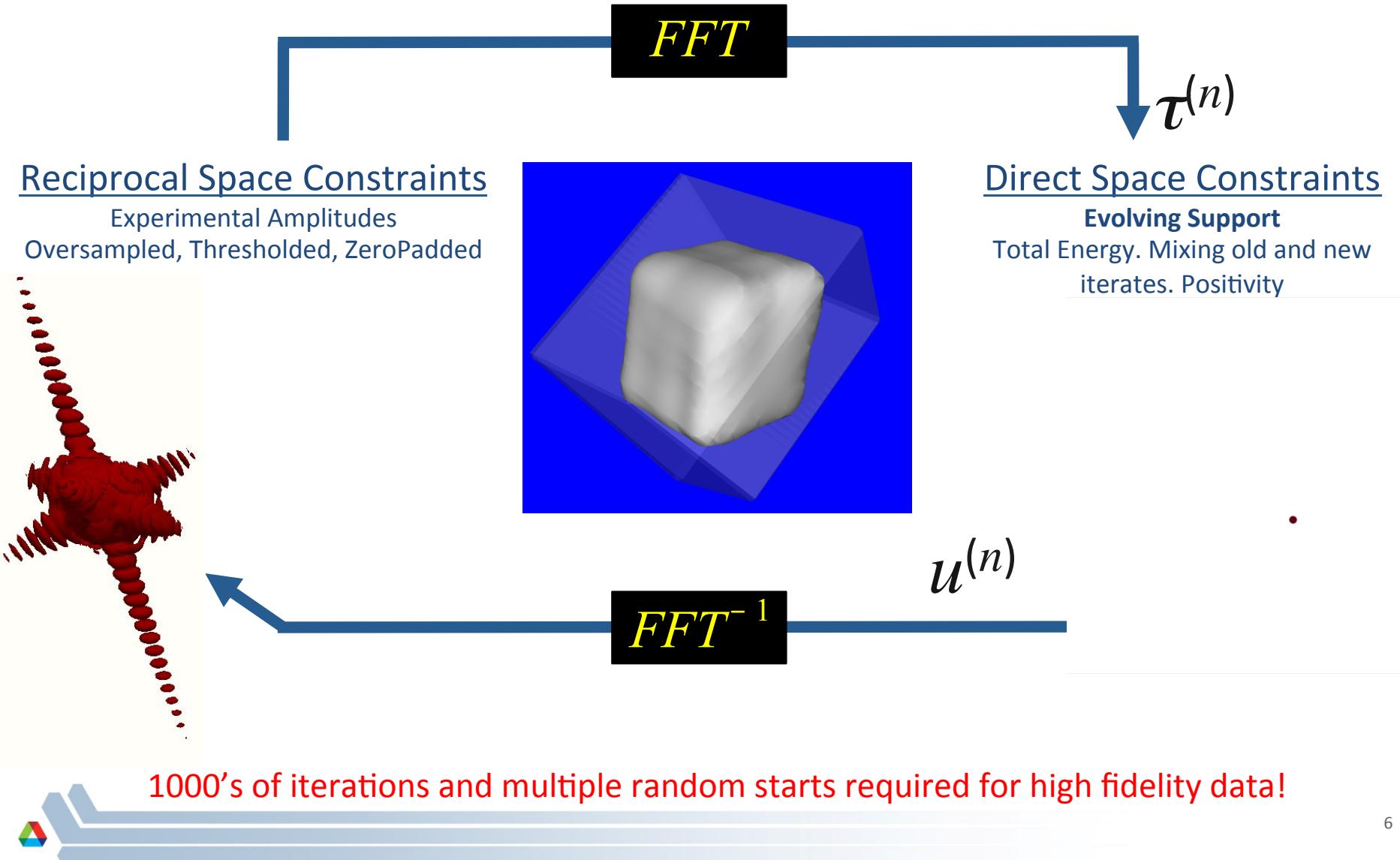
chain thiols, the large stress cannot arise from the van der Waals interactions or other weak forces alone, but at least ionic or covalent rearrangements. Indeed the Au-S bond plays a crucial role in SAM formation: the structure of a 1.6 nm thiolated nanocrystals has its Au-Au spaces disrupted and sulphur intermixed with gold in the outer shell. Our findings support this model and show strong thermal deformations of our 300 nm crystals with strains penetrations of more than 20 nm from the outer surface towards the crystal core. The tight-radius spherical parts of nanocrystals might also undergo strong Au-S interdiffusion. We would indeed be able to provide sufficient stress for reactions involving atomic diffusion of Au at room temperature. This would also be an attractive explanation of the relatively small strain seen in both our X-ray and the cantilever experiments.

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

NATURE MATERIALS | VOL 10 | NOVEMBER 2011 | www.nature.com/nmat/
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Phase Retrieval: Challenging and Costly Inverse Problem

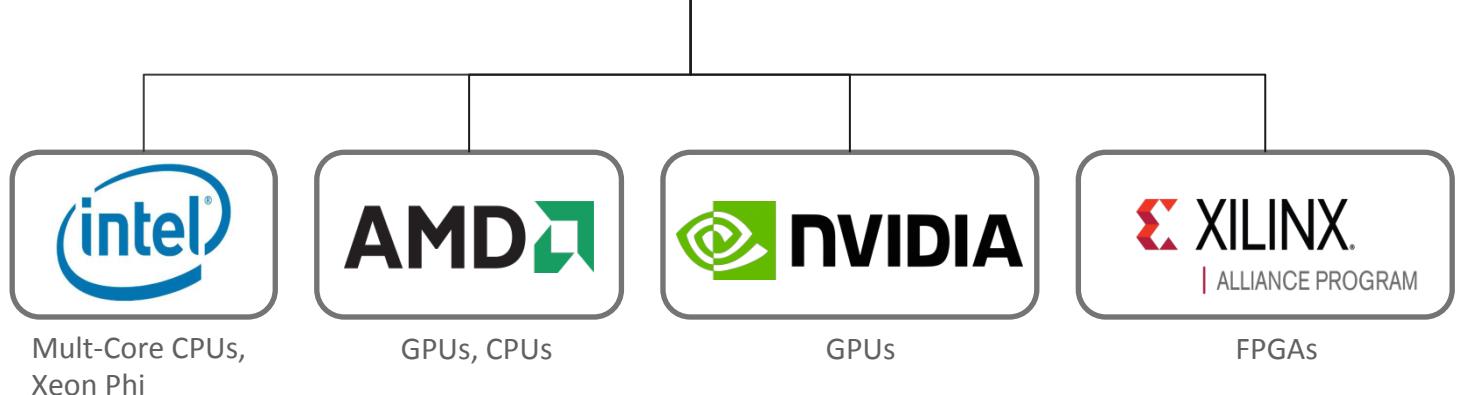


Fast Phasing Library (FPL)

- 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

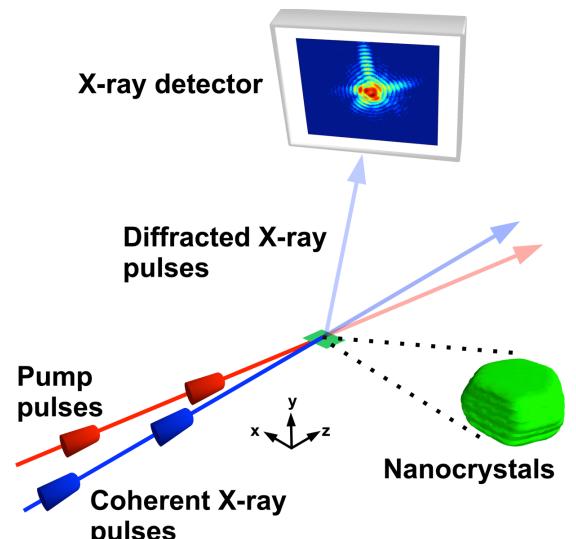


CUDA, Intel MKL, Intel IPP, OpenCL, MAGMA, Eigen,
Armadillo, etc ...

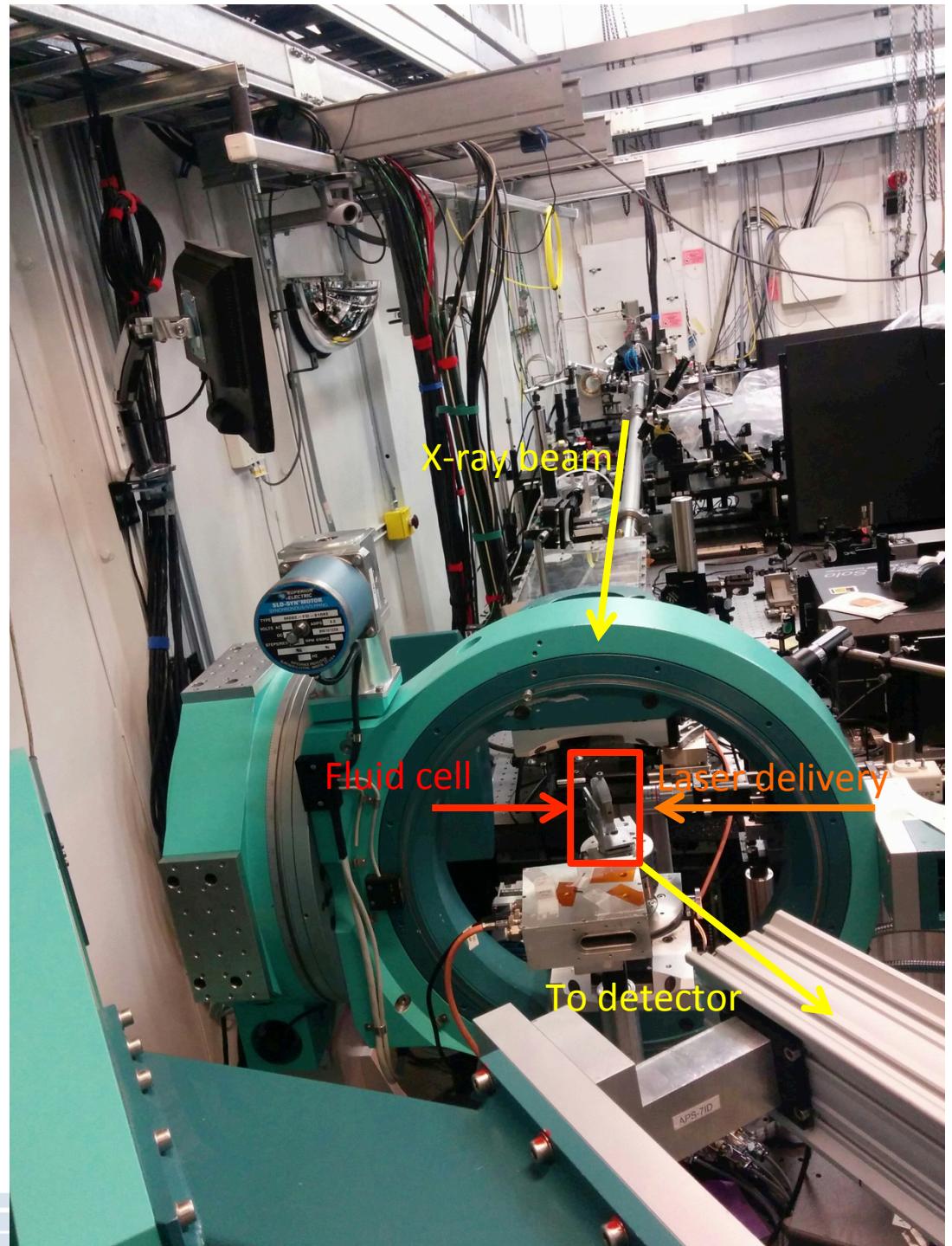


Time-resolved CDI

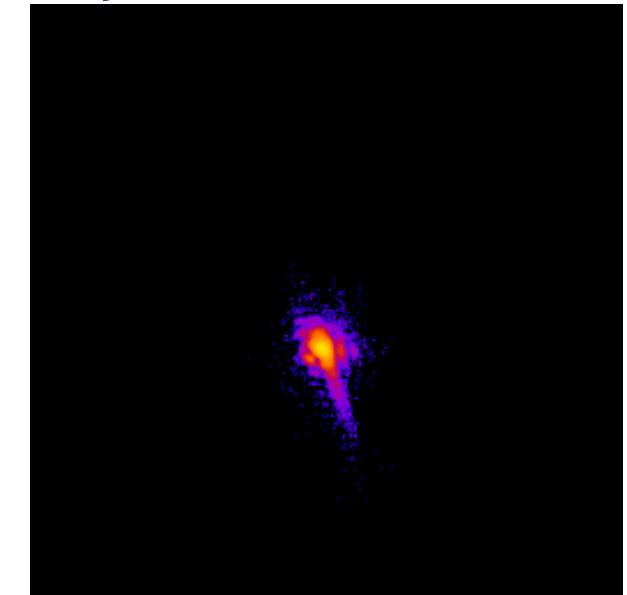
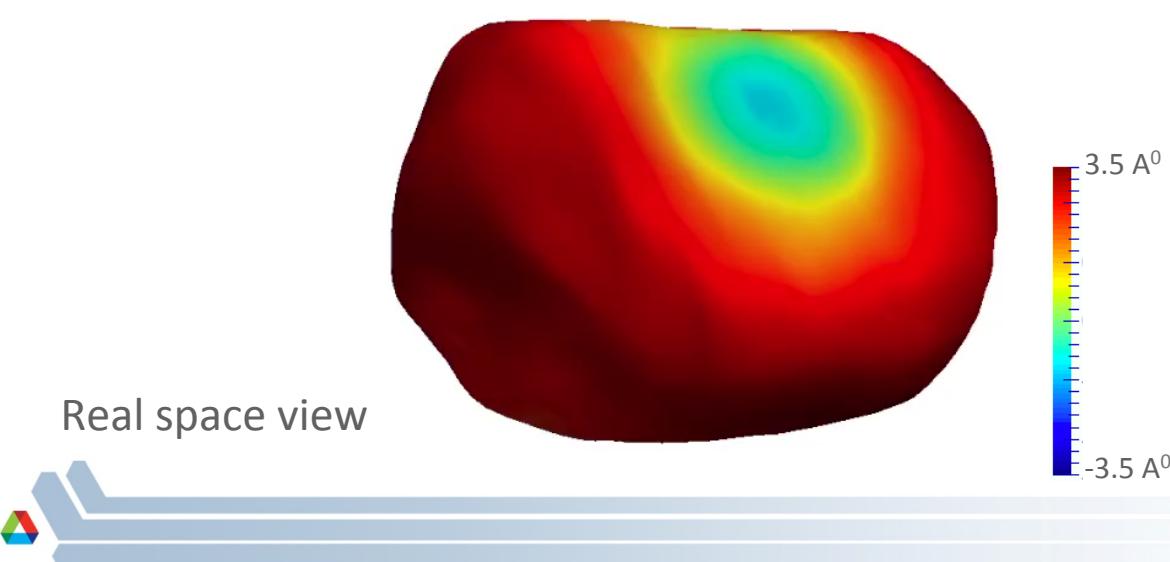
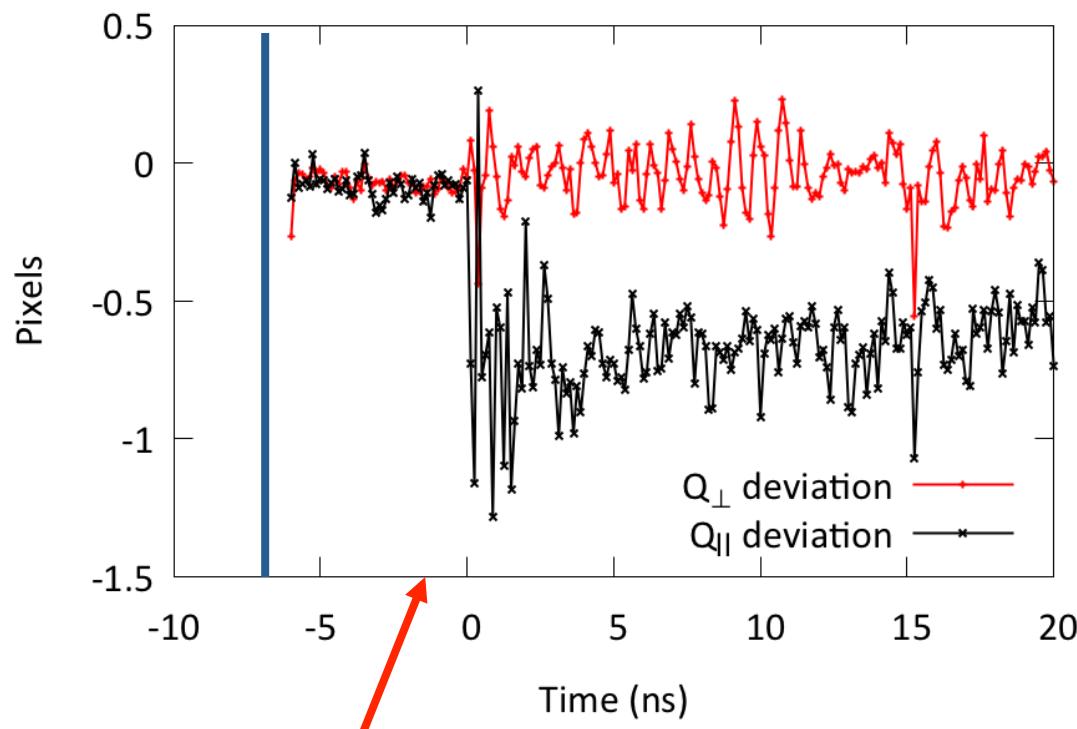
- Laser pump, X-ray probe at Sector 7
- Combine with CDI
- Provide dynamical map of excited modes in sample.
- <10 nm spatial resolution possible with 100 ps temporal resolution.



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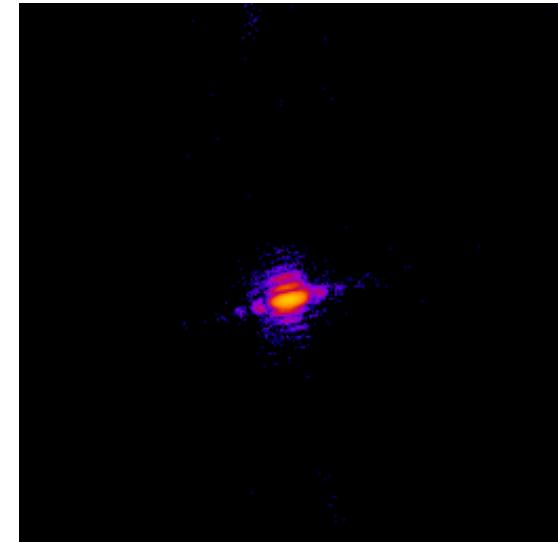
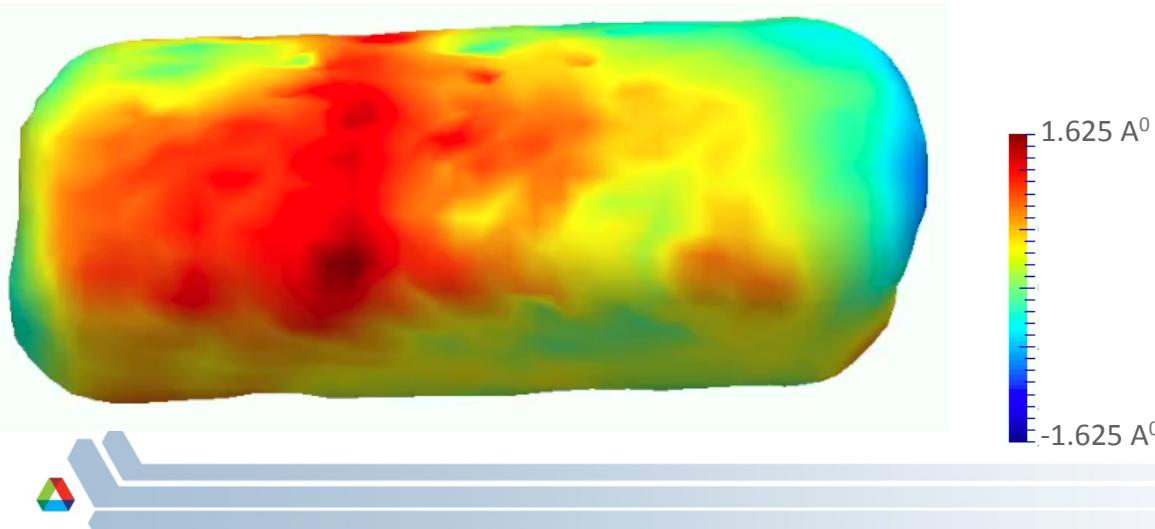
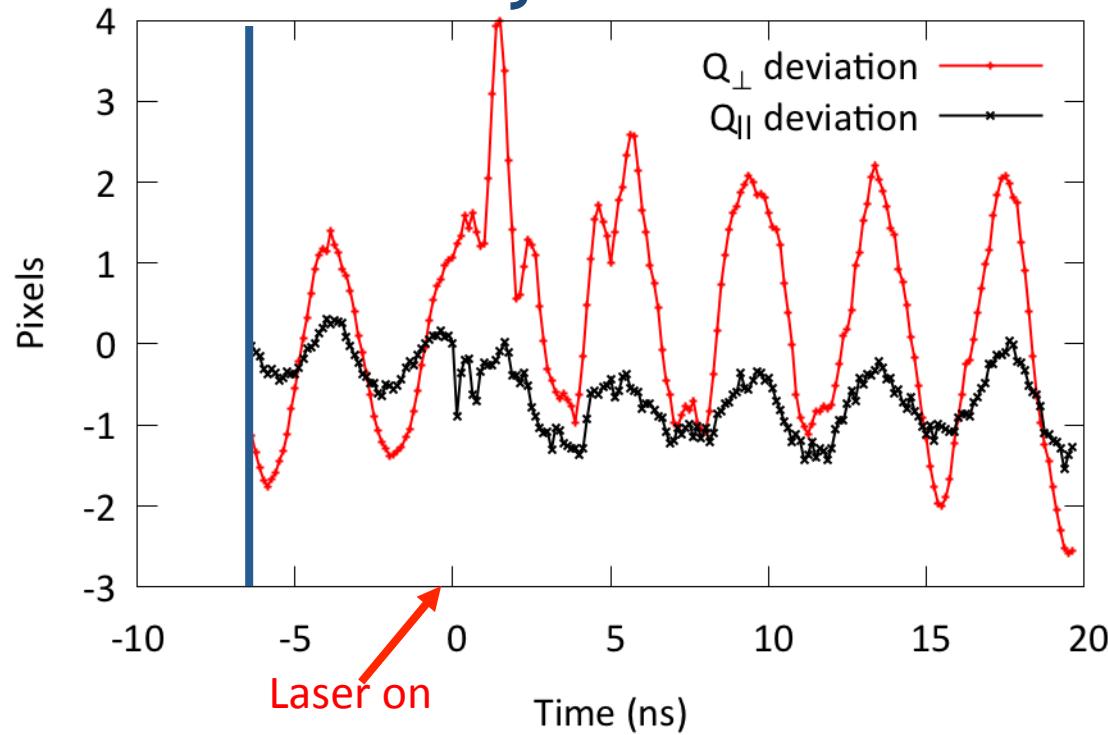


Case Study 1: Dynamic Response of Au Crystals



Reciprocal space view
↓
Phase retrieval
at select times
Need to measure the
complete rocking curve at
those points

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

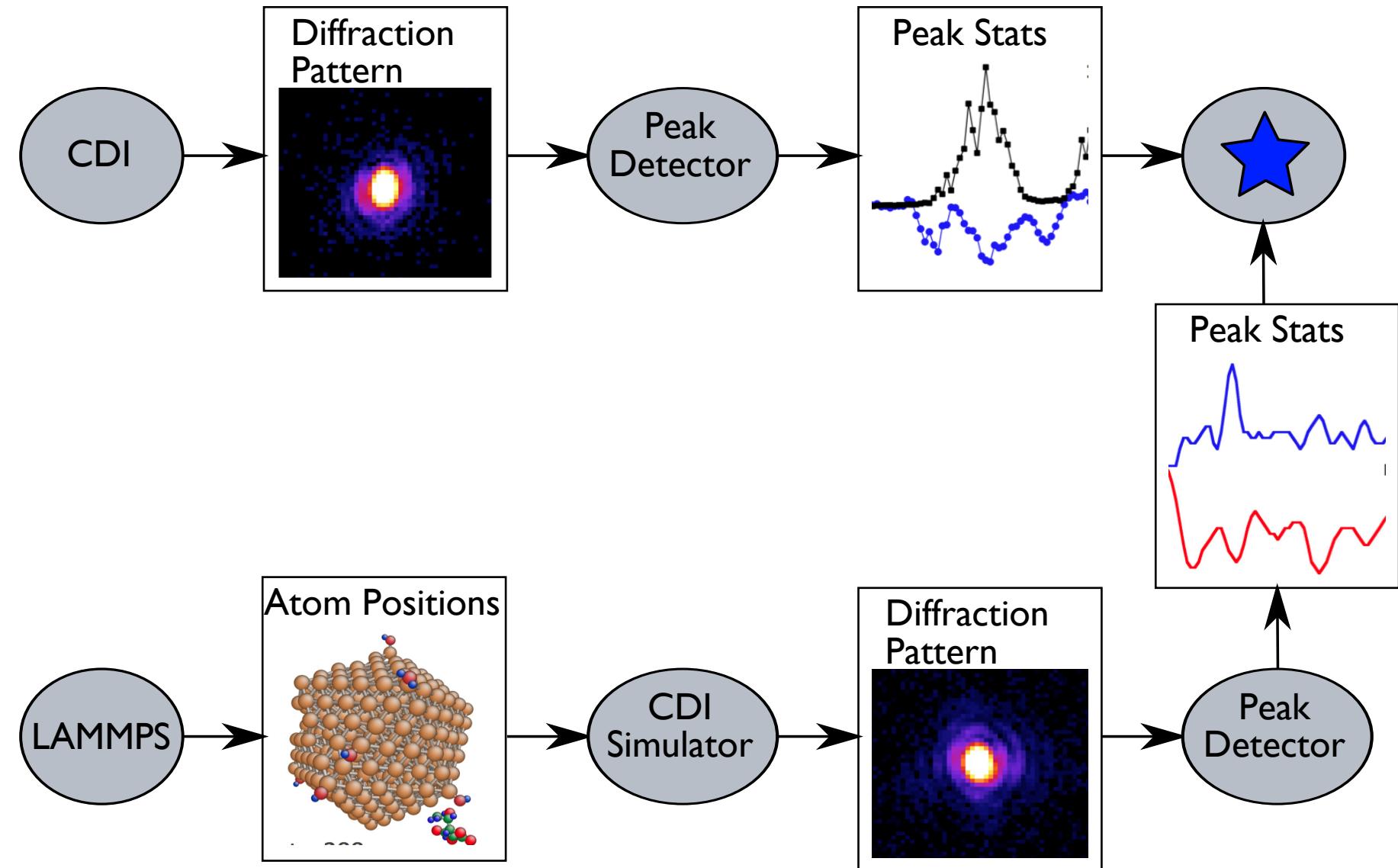


Reciprocal space view

Phase retrieval
at select times

Need to measure the
complete rocking curve at
those points

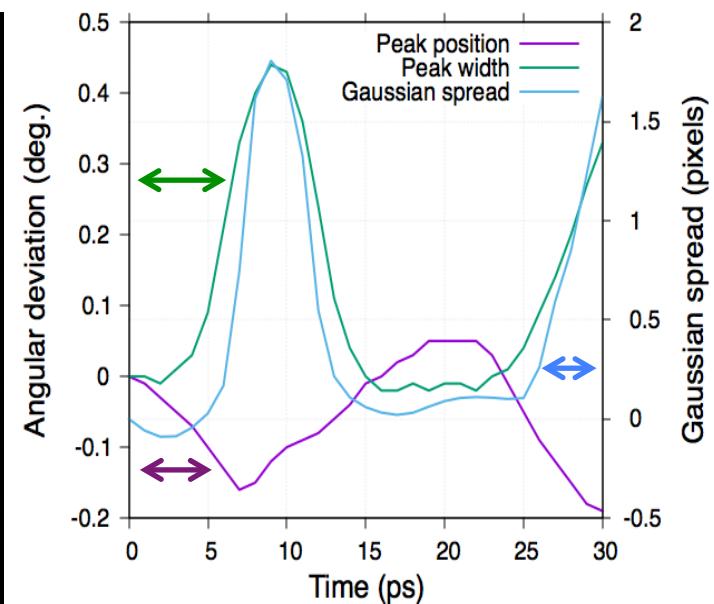
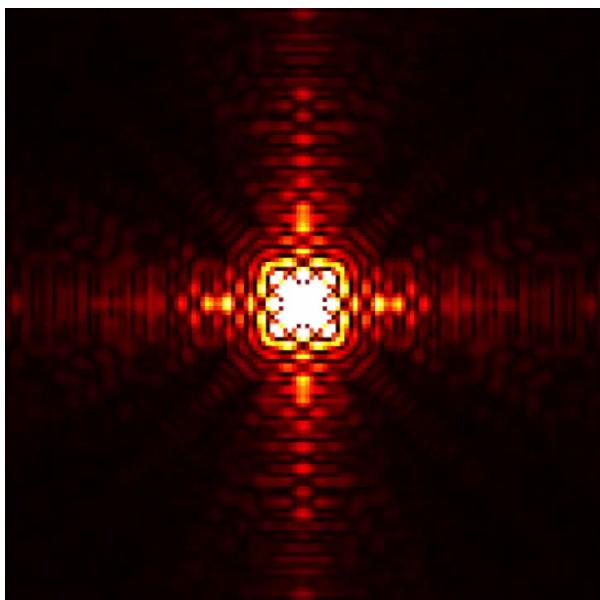
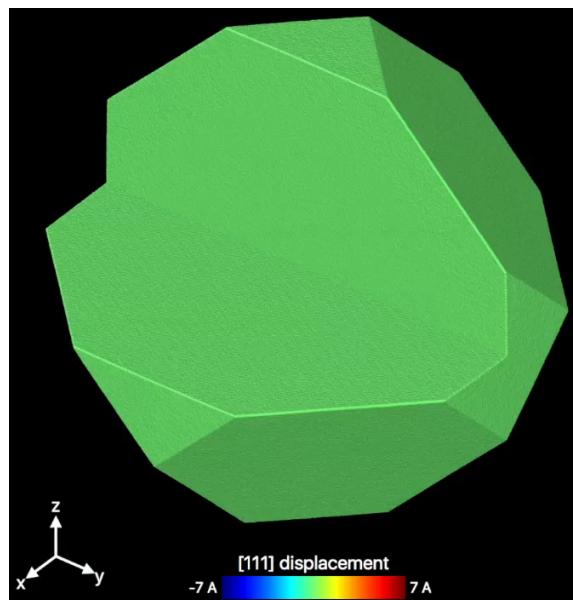
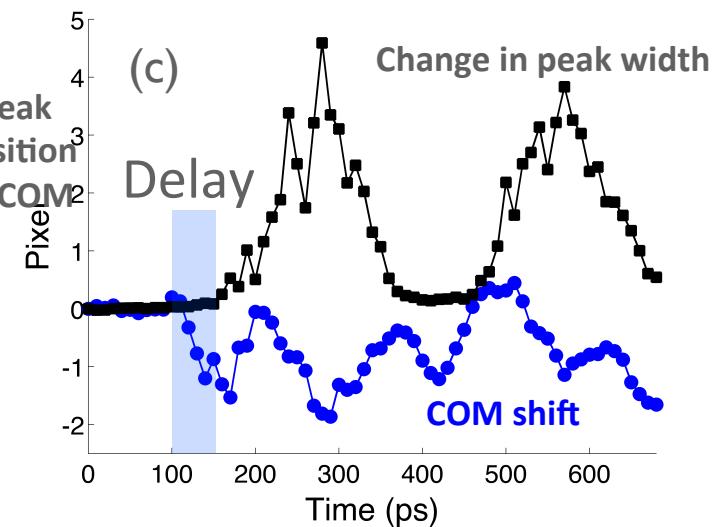
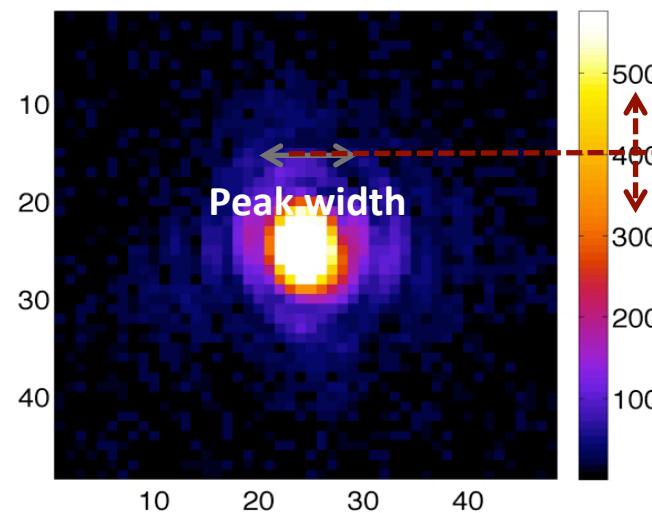
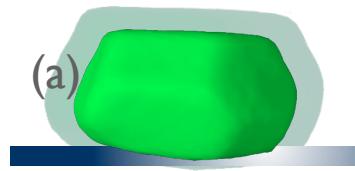
Reverse Problem Workflow



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

100 nm aluminum shell on 300 nm gold

(a)



MD simulations

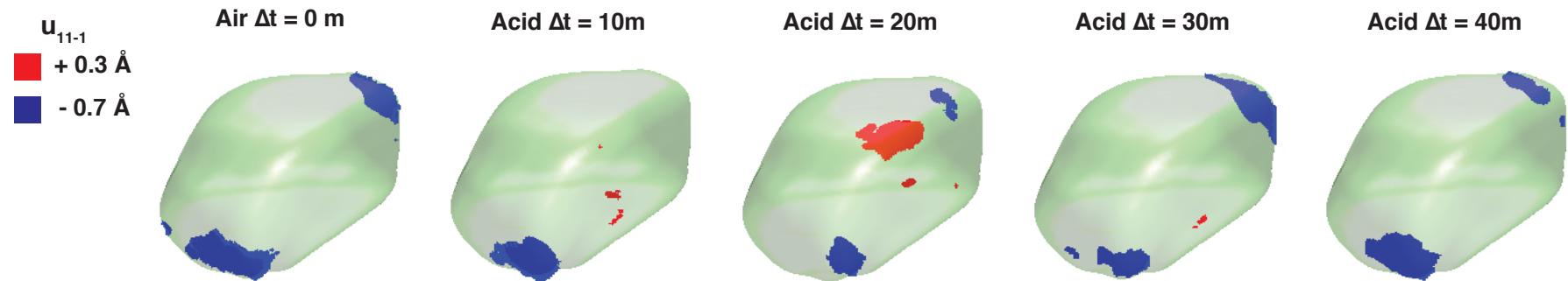


Calculated diffraction pattern

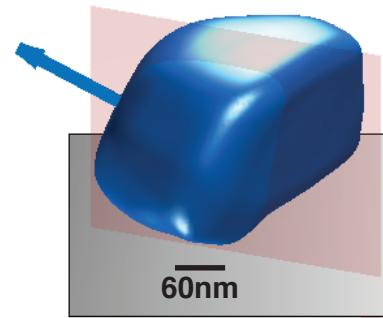


Case Study 4: Ascorbic Acid Decomposition on Gold

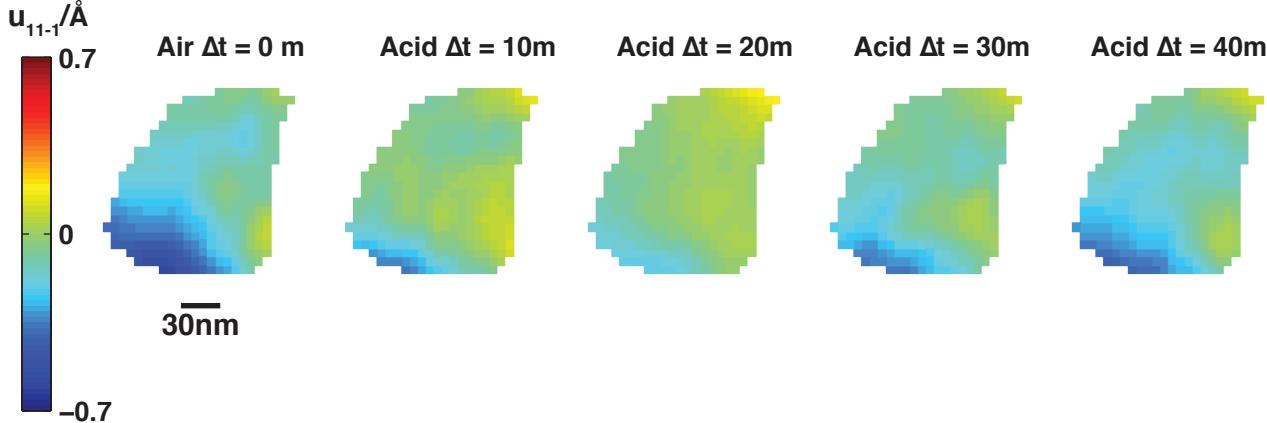
a) Isosurface of u_{11-1} values during first exposure



b) Cross-section Location



c) u_{11-1} dynamics during the first acid exposure

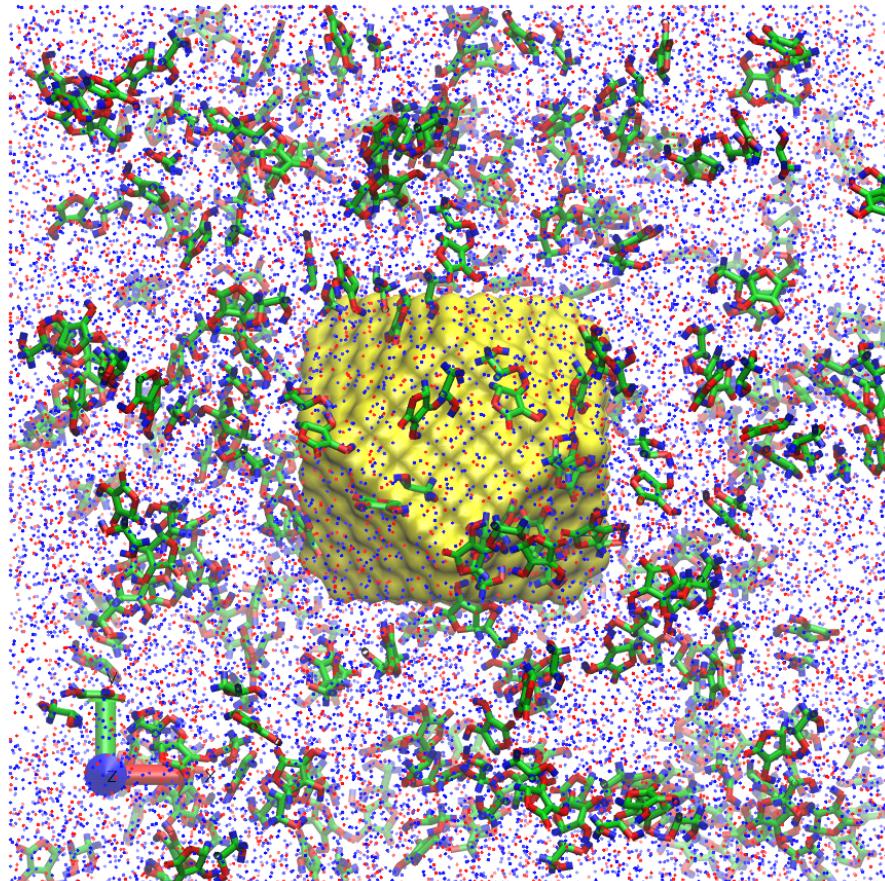


- Lattice change occurs at junctions involving the flattest facet
- Electron injection should create largest electric field at crystallographic discontinuities or apexes, providing “hot spots” for the reaction



Atomistic Simulation of Nanocatalytic Activity on Au Nanosurfaces

Sample schematic of simulated system
1M acid + water + gold (truncated octahedron)



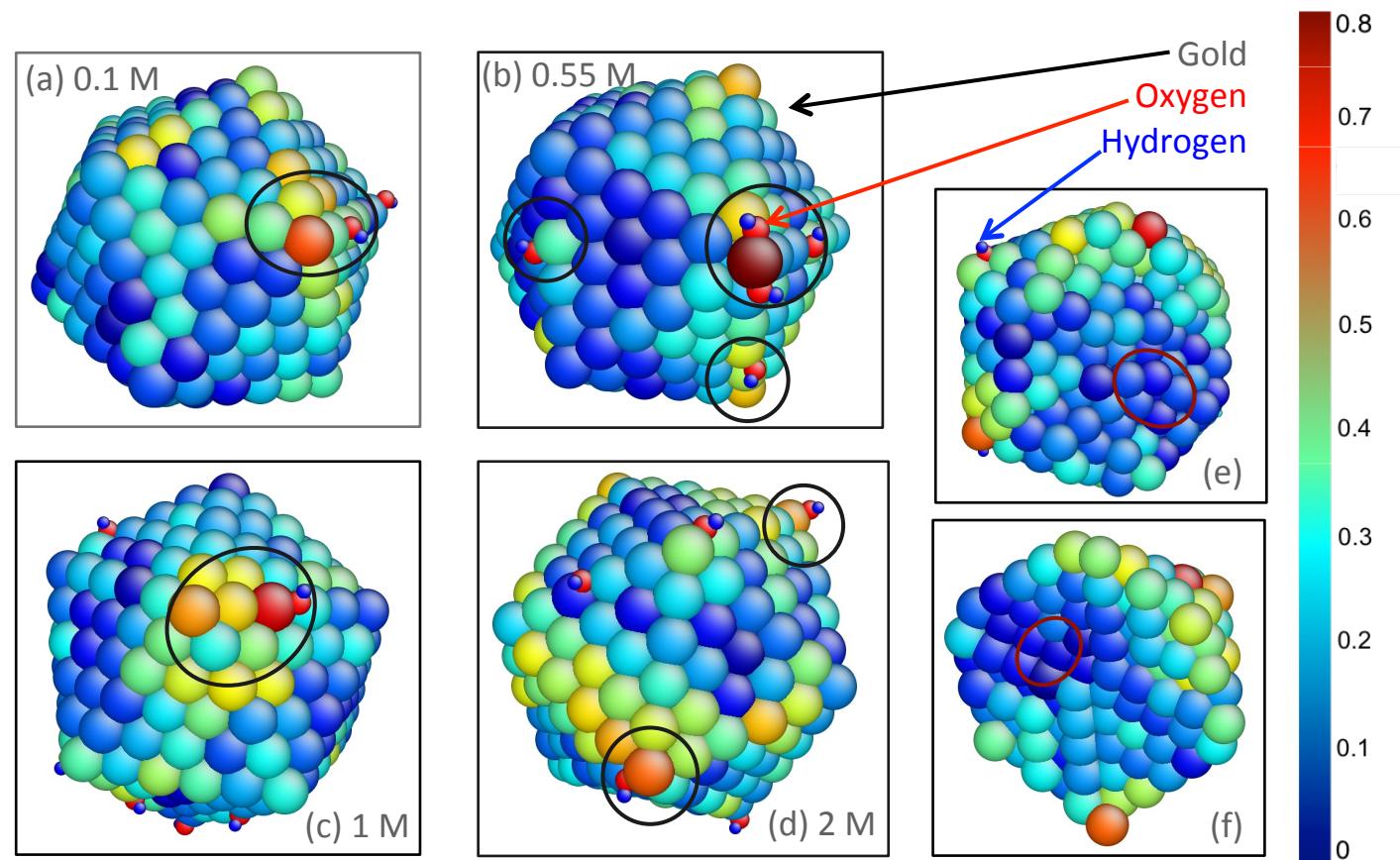
Yellow - Gold
Green - Carbon

Red - Oxygen
Blue - Hydrogen

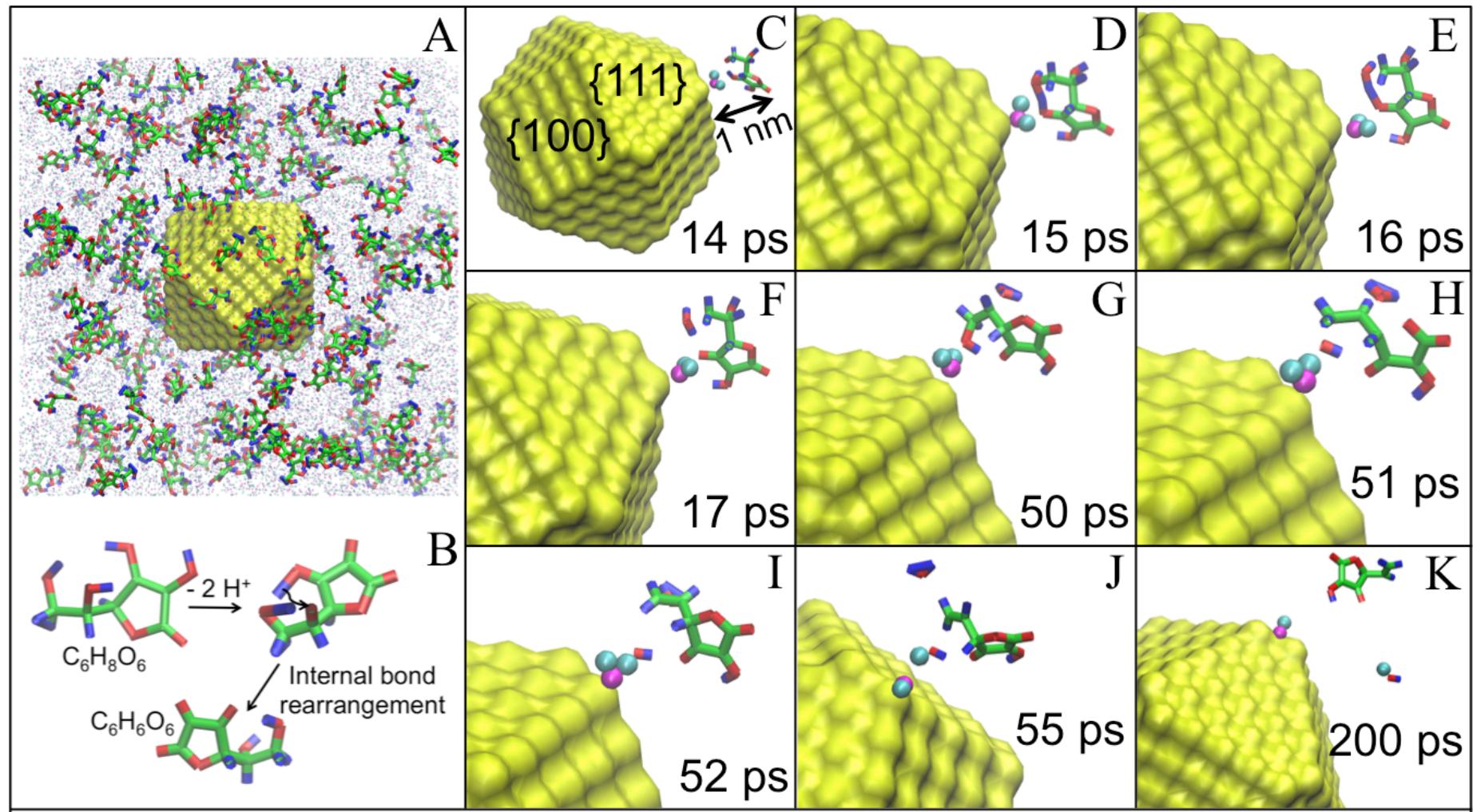


Effect of Acid Molarity

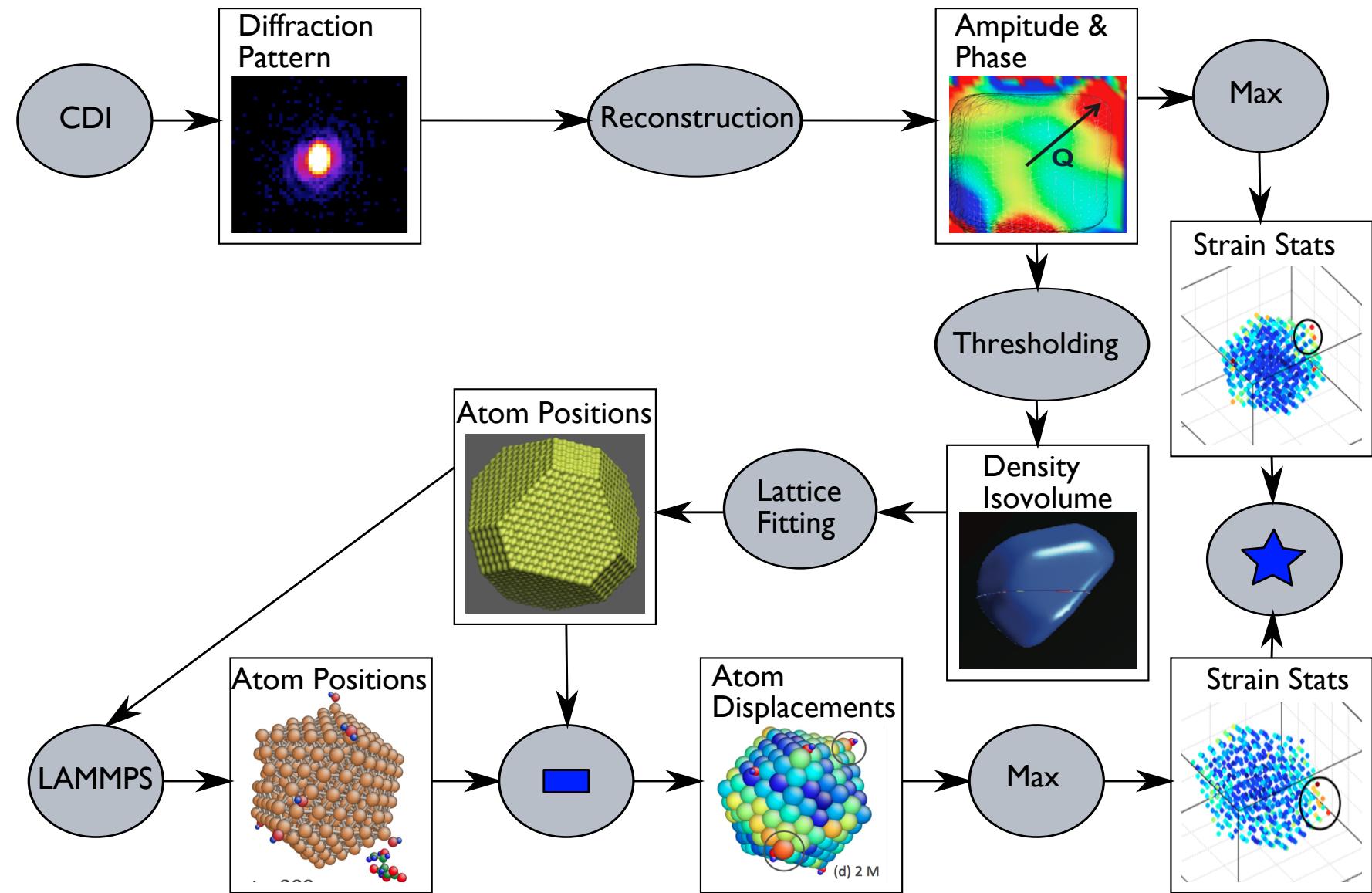
Displacement of Au atoms after 200 ps



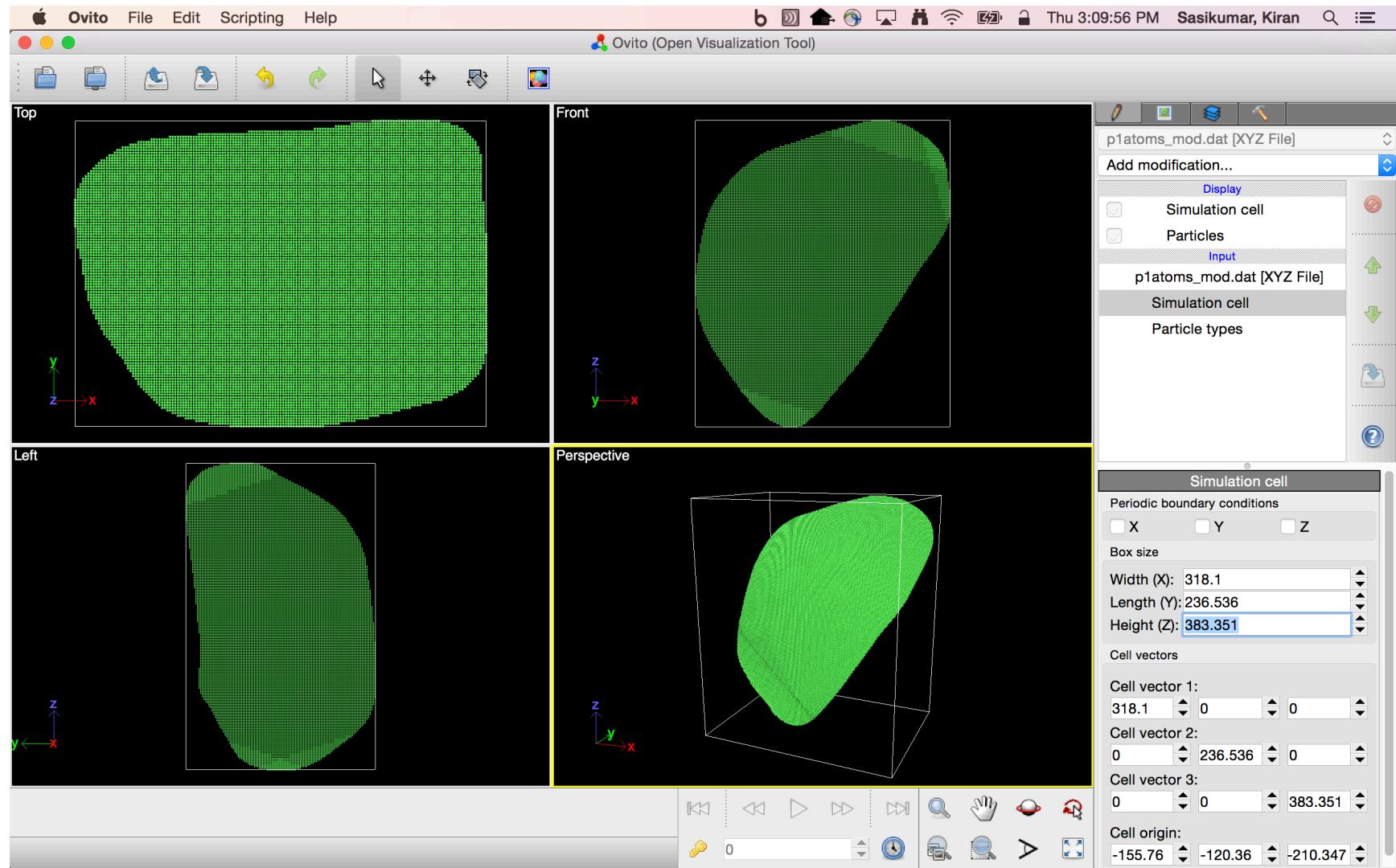
Adsorption Pathway



Forward Problem Workflow

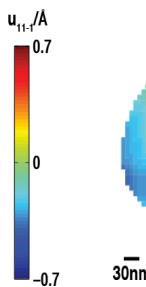
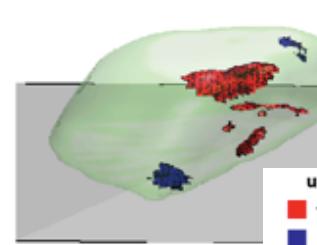
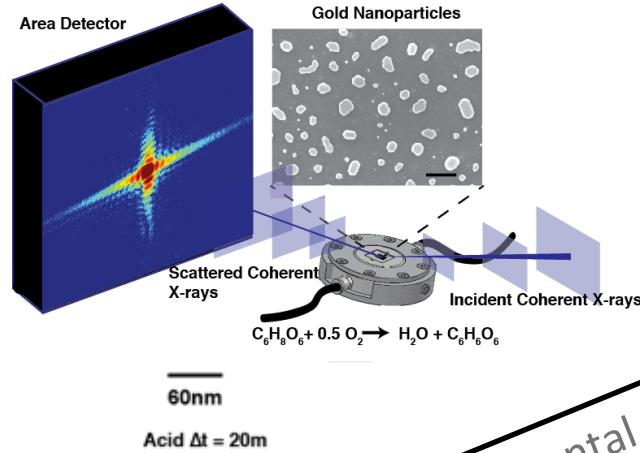


Lattice Fitting



Modeling Ascorbic Acid Decomposition on Gold

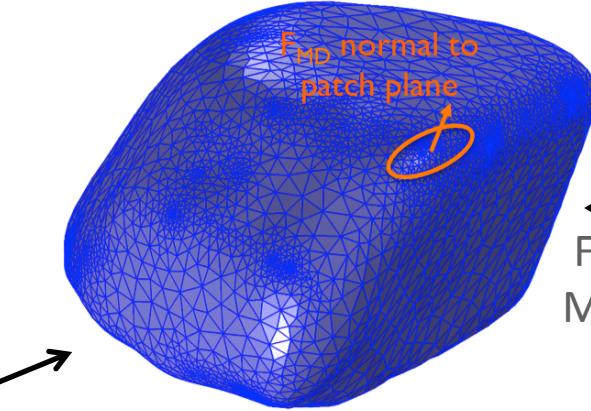
CXDI Experiment



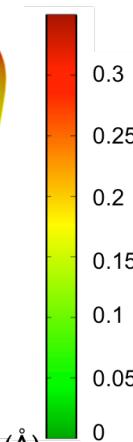
Experimental reconstruction as input to FEM

Experiment vs.
Theory

FEM (COMSOL)

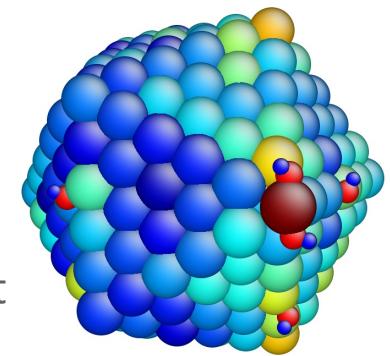


FEM Result



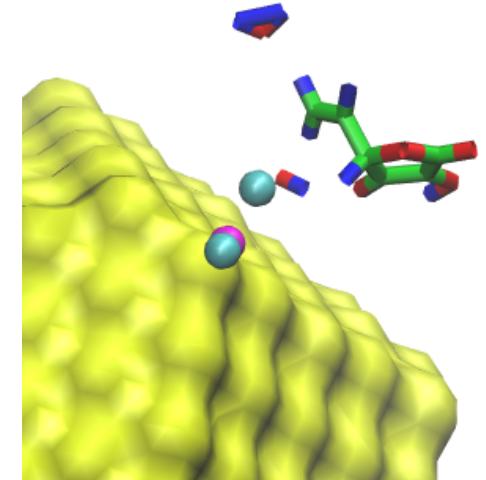
Central Slice: Projected displacement along u_{11-1} (Å)

MD (ReaxFF – LAMMPS)



Force from MD as input to FEM

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
 - 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)



[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 enable science applications to better manage their end-to-end data flow. The following...

The image shows the front cover of a report titled "CHALLENGES AT THE FRONTIERS OF MATTER AND ENERGY: Transformative Opportunities for Discovery Science". The cover has a yellow and orange textured background. The title is at the top, followed by a subtitle "Transformative Opportunities for Discovery Science". A vertical strip on the right edge contains the text "A Report from the Basic Energy Sciences Advisory Committee".

[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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