MAUI

"Modeling, Analysis, and Ultrafast Imaging"

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Integrated Imaging Institute Seminar
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Team

Modeling
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Analysis
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Ultrafast Imaging
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- Mathew Cherukara (XSD)
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Displacement of Au atoms after 20 ps in 0.55 MM ascorbic acid.
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

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


Phase Retrieval: Challenging and Costly Inverse Problem

Reciprocal Space Constraints
Experimental Amplitudes
Oversampled, Thresholded, ZeroPadded

Direct Space Constraints
Evolving Support
Total Energy. Mixing old and new iterates. Positivity

$\text{FFT}^{-1}$

$\tau^{(n)}$

$u^{(n)}$

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.
- \(<10 \text{ nm spatial resolution possible with } 100 \text{ ps temporal resolution.}\)

Need to measure the complete rocking curve at those points.
Case Study 2: Volumetric and Deviatoric Deformation Modes in ZnO Crystal

Need to measure the complete rocking curve at those points.

Phase retrieval at select times.

Reciprocal space view.
Reverse Problem Workflow

- **CDI** → **Diffraction Pattern** → **Peak Detector** → **Peak Stats**
- **LAMMPS** → **Atom Positions** → **CDI Simulator** → **Diffraction Pattern** → **Peak Detector**
Case Study 3: Induced Shock Waves in Au Coated with Al

100 nm aluminum shell on 300 nm gold

(c) Delay

Change in peak width

COM shift

MD simulations → Calculated diffraction pattern
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  Red - Oxygen
Green - Carbon  Blue - Hydrogen
Effect of Acid Molarity

Displacement of Au atoms after 200 ps

(a) 0.1 M  (b) 0.55 M  (c) 1 M  (d) 2 M  (e)  

Gold
Oxygen
Hydrogen

15
## Adsorption Pathway

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- **A**: Adsorption of molecules onto the surface.
- **B**: Representation of chemical reactions and internal bond rearrangement.
- **C-D**: Sequential stages of adsorption and chemical interaction.
- **E**: Transition to the next stage.
- **F-G**: Further progression in the adsorption pathway.
- **H**: Final state or product.

Timing: 14 ps, 15 ps, 16 ps, 17 ps, 50 ps, 51 ps, 52 ps, 55 ps, 200 ps.
Forward Problem Workflow

CDI → Diffraction Pattern

Reconstruction

Amplitude & Phase

Max

Strain Stats

Thresholding

Density Isovolume

Lattice Fitting

Atom Positions

CDI

Atom Positions

Atom Displacements

Max

LAMMPS

Strain Stats

(d) 2 M
Lattice Fitting
Modeling Ascorbic Acid Decomposition on Gold

**CXDI Experiment**
- Area Detector
- Gold Nanoparticles
- Scattered Coherent X-rays
- Incident Coherent X-rays
- Equation: \( \text{C}_6\text{H}_8\text{O}_6 + 0.5 \text{O}_2 \rightarrow \text{H}_2\text{O} + \text{C}_6\text{H}_4\text{O}_6 \)
- Acid \( \Delta = 20 \text{m} \)
- Central Slice: Projected displacement along \( u_{111}(\text{Å}) \)

**FEM (COMSOL)**
- Minimum element size: 1 nm
- Maximum element size: 20 nm
- 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.

**Experiment vs. Theory**
- Experimental reconstruction as input to FEM
- Force from MD as input to FEM
- FEM Result

- Experiment vs. Theory
- Central Slice: Projected displacement along \( u_{111}(\text{Å}) \)
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)
References

- Maui website (public)
  http://tpeterka.github.io/maui-project/
- Maui project site (private)
  https://bitbucket.org/tpeterka1/maui
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