FY 2015 LDRD ANNUAL PROJECT REPORT TEMPLATE

PROPOSAL TITLE: Integrated Imaging, Modeling, and Analysis of Ultrafast Energy Transport in Nanomaterials

PROJECT NO.: 2015-149-N0

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PROJECT DESCRIPTION

Integrating ultrafast imaging with molecular dynamics modeling and data analysis and visualization can provide crucial insights for energy research. The temporal behavior of externally stimulated materials beyond equilibrium can lead to breakthroughs, for example, in heat dissipation of next-generation semiconductors, conversion of wasted heat into electricity in thermoelectric materials, and electrochemical processes across liquid-solid interfaces in water purification. These diverse applications all transport energy through phonons (sound waves that carry heat) in a time-evolving crystal lattice. We are researching an integrated approach to predict, image, and analyze phonon dynamics that can be applied to externally stimulated systems. The project consists of three main components: modeling, analysis, and imaging.

MISSION RELEVANCE

Lattice vibrations in individual nanoparticles affect phase transitions, bond softening/hardening, ferroelectricity, solid/liquid interfaces, heat dissipation, phononic local structure, phase front propagation, and spectrometry. Understanding such phenomena can enable energy applications such as photocatalysis, photonics, thermoelectrics, semiconductor design, groundwater photo remediation, and heat transfer in battery interfaces: all critical to DOE's and Argonne's mission to design new materials for energy. The novel integration of model-guided imaging and image-guided modeling allows iterative feedback between the two processes that ultimately enables improved utilization of valuable resources at the APS and ALCF. Modeling information such as the sample temperature and image resolution allows researchers to conduct measurements that are scientifically significant. Likewise, the veracity of simulated results, crucial for the outcome of the experiments, is improved by timely analysis of reconstructed experimental images. In the context of the APS upgrade for transformational sciences, this proposal addresses the need for high-speed, high-volume data processing for novel time-resolved imaging; and it aligns with other Argonne strategic directions in hard X-ray sciences and advanced computing.

RESULTS AND ACCOMPLISHMENTS

We executed MD simulations, analyzed the MD models to compute trajectories from phonons, and we used these MD trajectories to generate strain fields. Such data analysis is the ``glue'' between forward modeling and reverse image reconstruction. We developed advanced GPU methods for rapid, near real-time reconstruction of ptychographic diffraction patterns and for feature detection and tracking in materials. We applied these methods to reconstruct 3D models from CDI images. To complement the imaging experiments and gain further insights into the mechanistic sequence of steps that leads to the onset of strain in a gold-ascorbic acid system, we performed MD simulations using the ReaxFF force-field. These simulations allow for dynamic charge transfer and are suitable for modeling chemically reactive systems. The model test system comprised of an Au slab with 200 ascorbic acid molecules dispersed in 10,000 water molecules. Our preliminary analysis indicated a total of ~90 reactions seen in the adsorption pathway in the short timeframe of ~20 ps. Many of these are fast reversible reactions associated with the rapid dissociation and recombination of ascorbic acid. To identify the atomistic mechanism by which strain is induced, we adapted the Molfract analysis tool in LAMMPS to obtain chemical reaction pathways using gold-ascorbic acid as a representative test system. We developed detailed post-processing of the simulation trajectories in order to understand the correlation between size of the gold nanocrystal and the timescales for fluctuation of ascorbic acid induced strain in the gold lattice (Figure 1). Meanwhile, at beamline 34-ID-C, we conducted experiments of the same reaction, the reduction of ascorbic acid on a nanocrystalline gold catalyst. The coherent imaging (Figure 2) was performed on gold nanocrystals (~200nm) before, during, and after exposure to 1M ascorbic acid. The crystals were observed to exhibit distortions of their lattice near edges of the top and bottom oriented facets and as a function of time in the acid solution, which agreed with our simulations.

Proposed Future Work

We will continue to model the gold-ascorbic system using ReaxFF and post-process the MD trajectories to derive various structural and dynamical correlation functions (pair correlation functions, structure factors, etc.) to compare/complement the experiments. Since the ascorbic acid reduction is exothermic and might lead to localized heating, we will also carry out non-equilibrium MD simulations to study phonon transport across solid-liquid interfaces such as in the gold-ascorbic acid system. We will subsequently extend the NEMD simulations to study phonon transport across solid interfaces such as in thermoelectric multi-layered materials to complement the imaging experiments. We will carry out coherent imaging of phonon modes excited in a multilayer structure via ultrafast laser pumping. Multilayer structures are particularly promising for thermoelectric (TE) materials as the interfaces between different lattices in the layers act as scattering centers for phonons incident on them leading to phononic band gaps in the materials. This is highly desired in TE materials, as the goal is to transfer maximal energy into the electrons and not into the lattice in the form of phonons. The experiments are proposed for Sector 7 of the APS, as this beamline is equipped with necessary laser and x-ray diffraction instrumentation.

Publications and Presentations

- Deng, J., Vine, D., Nashed, Y., Chen, S., Phillips, N., Peterka, T., Ross, R., Vogt, S. Jacobsen, C.: Fly-Scan Ptychography. Optics Express Journal, 2015.
- Yuelin Li, Zhang Jiang, Xiao-Min Lin, Haidan Wen, Donald A. Walko, Sanket A. Deshmukh, Ram Subbaraman, Subramanian K. R. S. Sankaranarayanan, et al. Femtosecond Laser Pulse Driven Melting in Gold Nanorod Aqueous Colloidal Suspension: Identification of a Transition from Stretched to Exponential Kinetics. Nature Scientific Reports, 5, Article number: 8146 doi:10.1038/srep08146 9 (2015)
- Ulvestad, A., Harder, R., Maxey, E., Clark, J.N., Kim, J.W. Mulvaney, P., Shpyrko, O.G: Nanocatalytic Activity Under Operando Conditions, In preparation.

GRAPHICS (OPTIONAL)

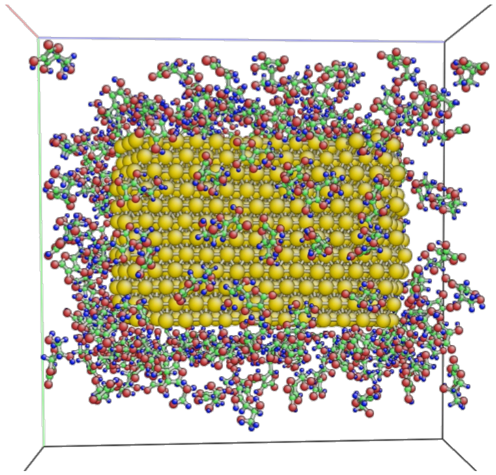

Figure 1. Modeling the atomistic mechanism leading to strain using reactive MD

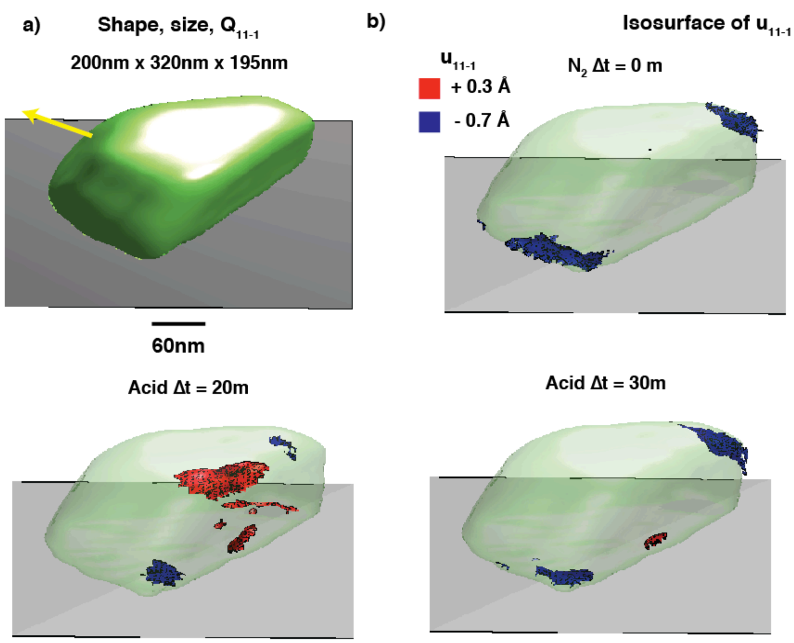

Figure 2. Imaging gold nanoparticles in ascorbic acid using coherent x-rays