



2015

APS/CNM Users Meeting



GENERAL SESSION

ABSTRACTS



APS

Evolving Rock Structure

Wen-lu Zhu

Department of Geology, University of Maryland, College Park, MD 20782

Correlating physical properties with rock structure at various scales, in three dimensions (3-D), and in four dimensions (4-D, when time evolution is considered), hold the key to unraveling complex geological processes. Synchrotron x-ray imaging, coupled with digital rock physics, is transforming the field of experimental rock deformation by revealing the details of evolving rock microstructures.

3-D images obtained by synchrotron x-ray tomography at nano- and micro-meter scale depict the pore structure of rocks in unprecedented detail. Based on the 3-D microstructure, we determine the bulk electrical conductivity and permeability of partially molten mantle rocks. Numerical experiments of electric currents and fluid flow conducted on the same rock show that the respective pathways for direct current and fluid flow may differ considerably, with more tortuous fluid flow paths in general over the same pore geometry. These results enable better-guided interpretations of geophysical data and constrain melt connectivity and transport at mid-ocean ridges.

Taking advantages of fast developing 4-D x-ray imaging techniques, we investigate the real-time evolution of microscopic pore structure in olivine aggregates reacting with a carbon-rich fluid. The transformation of olivine to magnesium carbonates increases the volume of solids. Our data reveal that after the initial filling of pore space by the growth of new minerals, cracks in polygonal patterns emerge. Subsequently these cracks coalesce into large fractures, which leads to a complete disintegration of the sample. These results provide direct experimental evidence of reaction-induced cracks during mineral carbonation. These data can be used to determine if the reaction between magnesium-iron silicates and carbon-rich fluid is self-sustaining, an important step towards evaluating whether carbon sequestration in ultramafic rocks is feasible.

APS

Operando Investigation of the Hydriding Phase Transformation in Single Palladium Nanocubes

Andrew Ulvestad¹, Ross Harder², Oleg Shpyrko¹, and Paul Mulvaney³¹Department of Physics, University of California-San Diego, La Jolla, CA 92093²Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439³School of Chemistry and Bio21 Institute, University of Melbourne, Parkville, VIC 3010, Australia

Phase transitions in reactive environments are crucially important in energy and information storage, catalysis, and sensors. Nanostructuring materials used in these systems can cause a host of desirable properties, including faster charging/discharging kinetics, increased lifespan, and record activities. However, establishing the causal link between structure and function is challenging for nanoparticles as ensemble measurements convolve intrinsic single particle properties with sample size and shape diversity. Here we study the hydriding phase transformation in individual palladium nanocubes under operando conditions using coherent x-ray diffractive imaging. We directly observe two-phase coexistence in the single particle diffraction data. The phase transformation initiates at the corner of the cube, penetrates further into the particle, and eventually violently rearranges the crystal structure. The strain distributions of the α and β phases are markedly different, indicating more than a simple Wulff geometric construction is required. A phase field model is constructed to interpret the phase transformation. Our results provide a general framework for understanding phase transformations in individual nanocrystals under operating conditions in reactive environments while highlighting the utility and importance of single particle investigations to truly understand important systems.

APS

Connecting Nanoscale Dynamics and Mechanical Properties of Disordered Soft Materials

Robert L. Leheny

Department of Physics and Astronomy, Johns Hopkins University, Baltimore, MD 21218

Complex fluids, such as polymer solutions and colloidal suspensions, are distinguished by their complicated response to mechanical stress. The distinct deformation and flow behaviors of such systems arise from the relaxation of internal structure on the nanometer or micrometer scale; however, the correspondence between this macroscopic mechanical behavior and the microscopic structural dynamics is rarely straightforward. In recent years x-ray photon correlation spectroscopy (XPCS) has emerged as a technique exceptionally suited to characterize the structural dynamics of many complex fluids at the nanometer length scales relevant to their mechanical behavior. This talk will describe a set of related projects employing XPCS on model systems including entangled polymer solutions and colloidal gels that reveal new insight into the microscopic origins of their mechanical properties.

APS

Utilizing High-energy X-rays for Microstructural Characterization of Nuclear Fuel Materials

Reeju Pokharel and Donald W. Brown

Los Alamos National Laboratory, Los Alamos, NM 87545

Efforts are underway to develop fundamental understanding of microstructural evolution of nuclear fuel materials, which operate at high temperatures for extended periods of time. High-energy synchrotron x-rays can be utilized to non-destructively probe bulk samples, which enables the study of the microstructural evolution and dynamics of grain growth process in nuclear fuels. Near-field high-energy x-ray diffraction microscopy (nf-HEDM) in conjunction with micro-tomography has provided a unique platform for effective and efficient materials characterization, allowing for spatially resolved crystallographic orientation and density maps measurements in three-dimension. To date, these techniques have been used on metals with relatively low Z number (Cu, Zr, Al, Ni), and have been recently extended to measure ceramic fuels with high Z number, materials which require higher incident photon energy x-rays than previously used. The evolution of different microstructural parameters is monitored to understand the effect of fabrication process on fuel performance as well as microstructural changes driven by the thermal gradient that develops during service conditions. The non-destructive nature of this characterization technique allows for the monitoring of various material processes before, during, and after evolution in extreme environments, and will provide unprecedented data for the development and validation of theoretical codes aimed at predicting fuel performance.

APS

Ptychography 2025

D.J. Vine

Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439

Ptychography combined with x-ray fluorescence is a powerful and unique tool for nanoscale imaging. In this presentation, I will speak to how developments over the next decade in source brilliance, scanning microscope design, fast frame rate detectors, and parallel, scalable computational power will revolutionize ptychography. Current and planned projects at the APS have us well placed to take the lead in bringing these advances to bear on exciting scientific problems with national importance.



CNM

New Material Platform for Plasmonics

Vladimir M. Shalaev

Purdue University, West Lafayette, IN 47907

We outline the recent progress in developing new plasmonic materials that will form the basis for future low-loss, CMOS-compatible devices that could enable full-scale development of the metamaterial and nanophotonic technologies.

CNM

Room-temperature Lasing from Nanoparticle Arrays

Teri Odom

Northwestern University, Evanston, IL 60208

This talk will discuss lasing action from band-edge lattice plasmons in arrays of plasmonic nanocavities in a homogeneous dielectric environment. Optically pumped, 2D arrays of plasmonic (Au, Ag) nanoparticles surrounded by an organic gain medium can show directional beam emission (divergence angle $< 1.5^\circ$ and linewidth < 1.3 nm) characteristic of lasing action in the far-field. Lasing in such hybrid systems can be achieved from stimulated energy transfer from the gain to the band-edge lattice plasmons in the deep subwavelength vicinity of individual nanoparticles. Besides our semi-quantum model to explain the lasing action, the idea of a lasing spaser can also be used to describe our system and other array-based cavities. Finally, we will discuss how the emission from lattice-plasmon nanolasers can be dynamically tuned by changing the dielectric environment of the gain media while keeping the nanoparticle-array cavity fixed.

CNM

Ion Transport Controlled by Nanoparticle-functionalized Membranes

Edward Barry¹, Sean P. McBride², Heinrich M. Jaeger^{2,3}, and Xiao-Min Lin¹¹Center for Nanoscale Materials, Argonne National Laboratory, Argonne, IL 60439²James Franck Institute, University of Chicago, Chicago, IL 60637³Department of Physics, University of Chicago, Chicago, IL 60637

From proton exchange membranes in fuel cells to ion channels in biological membranes, the well-specified control of ionic interactions in confined geometries profoundly influences the transport and selectivity of porous materials. Here we outline a versatile new approach to control a membrane's electrostatic interactions with ions by depositing ligand-coated nanoparticles around the pore entrances. Leveraging the flexibility and control by which ligated nanoparticles can be synthesized, we demonstrate how ligand terminal groups such as methyl, carboxyl, and amine can be used to tune the membrane charge density and control ion transport. Further functionality, exploiting the ligands as binding sites, is demonstrated for sulfonate groups resulting in an enhancement of the membrane charge density. We then extend these results to smaller dimensions by systematically varying the underlying pore diameter. As a whole, these results outline a previously unexplored method for the nanoparticle-functionalization of membranes using ligated nanoparticles to control ion transport.

CNM

Photoexcited Carrier Dynamics of Cu₂S Thin Films for Photovoltaics

Shannon C. Riha¹, Richard D. Schaller², David J. Gosztola², Gary P. Wiederrecht², and Alex B.F. Martinson¹¹Materials Science Division, Argonne National Laboratory, Argonne, IL 60439²Nanoscience and Technology Division, Argonne National Laboratory, Argonne, IL 60439

Copper sulfide is a simple binary material with promising attributes for low-cost thin film photovoltaics. However, stable Cu₂S-based device efficiencies approaching 10% free from cadmium have yet to be realized. We utilize transient absorption spectroscopy at the CNM to investigate the dynamics of the photoexcited state of isolated Cu₂S thin films

prepared by atomic layer deposition (ALD) or vapor-based cation exchange of ZnS. While a number of variables including film thickness, carrier concentration, surface oxidation, and grain boundary passivation were examined, grain structure alone was found to correlate with longer lifetimes. A map of excited state dynamics is deduced from the spectral evolution from 300 fs to 300 μ s. This study provides insights into why such high device efficiencies may have been achieved in CdS/Cu₂S hetero-structure devices fabricated through topotaxial exchange processes. Revealing the effects of grain morphology on the photophysical properties of Cu₂S is a crucial step toward reaching high efficiencies in operationally stable Cu₂S thin film photovoltaics.

CNM

Looking Out for the Tiniest Lights: Controlling Light with Heterogeneous Interfaces

Jay Foley¹, Hayk Harutyunyan^{1,2}, Daniel Rosenmann¹, Zheng Li¹, Sheng Peng¹, Jeff McMahon³, George Schatz³, Gary Wiederrecht¹, Yugang Sun¹, and Stephen Gray¹

¹Center for Nanoscale Materials, Argonne National Laboratory, Argonne, IL 60439

²Department of Physics, Emory University, Atlanta, GA 30322

³Department of Chemistry, Northwestern University, Evanston, IL 60208

Research in nanophotonics, which seeks to understand and control optical energy and information flow at nanoscale dimensions, has been motivated by a number of applications ranging from energy conversion, imaging, and information storage, to name a few. The ability to confine optical energy beyond the diffraction limit by coupling light into collective electronic motion in metal nanostructures, including localized surface plasmons on metal nanoparticles and surface plasmon polaritons on metal surfaces, has played a key role in many advances in nanophotonics. Consequently, discovering and understanding plasmonics phenomena in new contexts is a considerable focus of nanophotonics research.

This talk will focus on plasmonics phenomena in the context of bimetallic interfaces. I'll describe an entirely new type of plasmon known as an "Inhomogeneous Surface Plasmon Polariton" (ISPP). ISPPs provide new opportunities for controlling the propagation behavior and confinement of light, and can be generated by refraction of ordinary surface plasmon polaritons at the interface between two metal surfaces. I will also describe the enhanced plasmonic properties of interfaced bimetallic nanoparticles, including their ability to support so-called charge-transfer plasmons.

CNM

Origins of Anisotropy in the Magnetic Structure of Artificial Spin Ice Lattices

Vuk Brajuskovic^{1,2}, Charudatta Phatak¹, and Amanda Pefford-Long^{1,2}

¹Materials Science Division, Argonne National Laboratory, Argonne, IL 60439

²Department of Materials Science and Engineering, Northwestern University, Evanston, IL 60208

Artificial spin ice lattices are a two-dimensional analogue to spin ice crystals. The larger length scales of the artificial systems allow for their magnetic frustration behavior to be observed at room temperature. Traditionally, artificial spin ice lattices are patterned in thin films using electron-beam lithography because the elements that make up the lattice need to be small if they are to remain single domain. However, such fabrication is difficult to perform when the thin films have been deposited onto transparent membranes on TEM grids. Therefore, for those lattices whose elements remain single-domain at larger sizes, focused-ion beam (FIB) patterning represents a better alternative. In order to explore the origins of observed anisotropy in the lattices, Lorentz transmission electron microscopy has been used to obtain quantitative maps of the magnetization in square spin ice lattices FIB patterned in Ni₈₀Fe₂₀ (Permalloy) thin films. Our results have shown that although FIB patterning has an effect on the grain size of the Permalloy at the edges of the patterned regions, the anisotropy that is observed is the result of pre-existing anisotropy in the Permalloy film, rather than an effect introduced by the FIB patterning. Our results thus suggest that FIB patterning is an effective method for fabricating larger-scale artificial spin ice lattices.

This work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Material Sciences and Engineering. Use of the Center for Nanoscale Materials was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.