Jerry Bernholc
North Carolina State University

Large-Scale Simulations of Nanomaterials and Nanodevices

We describe large-scale ab initio simulations of nanoscale materials and devices, which complement and explain experimental investigations at CNMS, focusing on graphene nanoribbon (GNR) structures and devices. GNRs have very rich electronic properties that depend on their widths and edge types. We determine the growth mechanism of atomically precise GNRs and also consider incomplete conversion, resulting in an intermediate state consisting of a graphene structure on one edge and a polymer structure on the other. This leads to a novel, experimentally realizable GNR-based negative differential resistance (NDR) device based on GNR/intermediate-structure heterojunctions. We then uncover the general principles governing the design of NDR nanoscale devices. If time permits, we will also discuss exascale simulations of materials and devices. The real-space multigrid code (RMG) has been optimized for exascale supercomputers and can perform high-precision DFT/hybrid-DFT and quantum transport simulations for very large systems. RMG is distributed via www.rmgdft.org, with over 4,000 downloads to date. Due to its computational efficiency, RMG is very suitable for large-scale survey approaches, including Materials-Genome and Machine-Learning projects. In addition, advanced functionalities are provided through interfaces to other codes, including QMCPACK, BerkeleyGW, Phonopy, and ALAMODE.

Amanda B. Marciel
Rice University
Department of Chemical and Biomolecular Engineering

Influence of Charge Fraction and Sequence on Polyelectrolyte Solution and Brush Properties

Polyelectrolytes exhibit unique solution properties compared to neutral polymers due to charge repulsion along the backbone that increases chain size and results in viscoelastic behavior even at low polymer concentration. Consequently, polyelectrolytes are extensively used in industrial applications, including as thickeners and rheology modifiers for aqueous coatings and flocculation agents for colloids and wastewater treatment. They also play a fundamental role in biological processes, including intracellular phase separation and joint lubrication. Polyelectrolytes may also be anchored onto surfaces to create brush architectures that offer flexible design parameters for imparting tailored interfacial functionality at the nanoscale. The influence of charge sequence and fraction on polyelectrolyte solution and brush behavior, however, is lacking. Here, we use solid phase peptide synthesis (SPPS) and surface-initiated copper(0) controlled radical polymerization (SI-CuCRP) to produce polymers with controlled sequence and charge fractions. Systematic studies using small-angle X-ray scattering (SAXS) and 3D single molecule tracking reveal that charge fraction and sequence influence polyelectrolyte solution conformation and phase behavior, as well as brush height and transport properties.

Natalie Holder

Stanford National Accelerator Lab (SLAC)

"*The Top Ten Barriers to Inclusion*"

Despite our best efforts to support DEI and talk about their importance, how are micro-inequities thwarting your best efforts to increase access to your lab and create inclusive cultures? This session will define and explore the most common micro-inequities and discuss strategies to avoid them."

Jon F. Ihlefeld

University of Virginia

Jon F. Ihlefeld,[1] Samantha T. Jaszewski,[1] Shelby S. Fields,[1] Takanori Mimura,[1] Kyle P. Kelley,[2] and Thomas E. Beechem[3]

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Exploring the Mechanisms Leading to Polarization Wake-Up in Ferroelectric Hafnium Oxide Thin Films

The observation of ferroelectricity (switchable permanent dipole) in hafnium oxide just over a decade ago led to great excitement in the materials science and microelectronics communities owing to the promise of this material to serve as a scalable, silicon-compatible non-volatile computer memory component. Such a memory is of great importance as energy consumption for computing is projected to reach 20% of world energy demand at the close of this decade and highly integrated non-volatile memories are one component toward reducing this energy need. While hafnia has great promise, it has been hindered by several issues: an inability to prepare phase-pure materials, a change in polarization magnitude with use, and an inability to unambiguously identify the phases present. In this presentation, we will highlight our team's efforts to identify phases present using characterization tools that are not commonly employed on ferroelectrics. We will show that the variation in polarization magnitude with use can be linked to changes in phase composition of the devices. Specifically, we will show that phase transformations occur between non-ferroelectric metastable phases of the tetragonal $P4_2/nmc$ phase and orthorhombic *Pbca* phase to the ferroelectric metastable orthorhombic phase with space group $Pca2_1$. We will show how oxygen vacancies are a critical variable in phase stability and that control over their concentrations and charge states are necessary to realize reliable and high-performing ferroelectric non-volatile memories using hafnium oxide.

This material is based upon work supported by the Center for 3D Ferroelectric Microelectronics (3DFeM), an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences Energy Frontier Research Centers program under Award Number DE-SC0021118. Scanning probe microscopy research was conducted as part of a user project at the Center for Nanophase Materials Sciences (CNMS), which is a US Department of Energy, Office of Science User Facility at Oak Ridge National Laboratory. This research used resources of the Advanced Light Source, which is a DOE Office of Science User Facility under contract number DE-AC02-05CH11231.

Oana D. Jurchescu
Department of Physics and Center for Functional Materials
Wake Forest University

Organic Transistors for Optoelectronics and Bioelectronics

Our society has become increasingly dependent on electronic devices, which are now integrated into all aspects of our lives, securing our well-being and shaping major fields such as transportation, communication, manufacturing, engineering, medicine, energy, defense, and national security. The emerging Internet of Things (IoT) promises to enhance the quality of our lives even further by providing access to new types of applications and, with that, fundamentally changing the way we interact with electronics. Organic semiconductor devices are prime candidates for such applications due to their low cost, lightweight, versatility, and ease of molding into any shape. Organic thin-film transistors (OTFTs) have been used for decades as both active circuit elements and testbeds for material development. Their performance depends on the structure and the morphology of the organic semiconductor film, the quality of the device interfaces, and the device architecture. In this presentation, I will first discuss device design and processing, and the limitations we currently face in optimizing charge injection and transport. The environmental and operational stability and the major causes of degradation will also be addressed. Finally, I will provide an example of OTFT incorporation in biomedical applications: the RAD-OFET (**RA**diation **D**etector based on **O**rganic **F**ield-**E**ffect **T**ransistor) can validate in real time the radiation dose being delivered to a patient and ensure that for surrounding and/or neighboring organs and tissues an acceptable level of low dose is being received to preserving their bodily functions. Their adoption in clinical settings will facilitate the application of therapeutic radiation with high precision, a process that will increase the effectiveness on treating cancerous tissue and minimize the impact on the surrounding healthy cells. These results uncover new opportunities for organic circuits that will improve the quality of healthcare through better, lower cost in vivo dose monitoring during radiation therapy.

Prof. Sarah B King
Department of Chemistry and James Franck Institute
The University of Chicago

Probing the Ultrafast Electron and Phonon Dynamics of 2D Materials on the Nanoscale

Controlled and predictable energy and phonon transport in 2D materials is critical for their practical applications in nanoscale electronics and thermoelectrics. However, at the 2D limit, abundant morphological variations such as strain, layer thickness, grain boundaries, and edges can modify the optoelectronic and thermoelectric properties. In order to establish a new paradigm of predictably controlled novel functionalities in 2D materials and 2D material heterostructures, it is imperative to determine the interplay of morphological effects and intrinsic optoelectronic and thermoelectric properties in homogeneous and heterogeneous systems. Using polarization-dependent photoemission electron microscopy (PEEM) we have imaged the spatially dependent optical selection rules of black phosphorus,[1] distinguishing edge-specific modes, and antiferroelectric domains of In_2Se_3 with spatial resolution as good as 25 nm. Using ultrafast transmission electron microscopy, we've also been able to probe how the phonon dynamics in few-layer black phosphorus is modified by structural morphology. In the first direct observation of anisotropic phonon propagation on the nanoscale in black phosphorus, we measured the direction-dependent velocity of longitudinal acoustic phonons along the zigzag and armchair crystal directions and showing how phonon dynamics are modified by crystal morphology. Ultimately my group seeks to identify ways to modify the impact of structural heterogeneity in materials and rationally design energy efficient inorganic and organic/inorganic hybrid interfaces on the nanoscale using morphology and molecular interfaces. I'll also provide an introduction to new experiments we've conducted at ORNL using the MAC-STEM on 2D MXenes.

[1] P. P. Joshi, R. Li, J. L. Spellberg, L. Liang, and S. B. King, Nano Lett. (2022) 10.1021/acs.nanolett.1c03849.

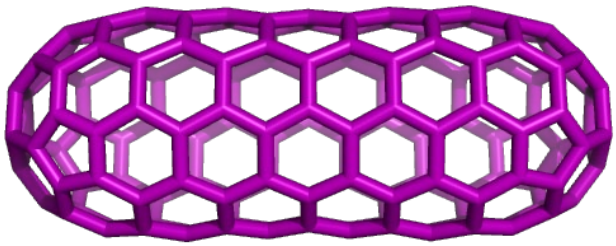
Steven Stevenson

Purdue University Fort Wayne

"Fullertubes: New Molecules and Collaborative Opportunities at ORNL"

Recently in 2020, we experimentally verified and published (https://pubs.acs.org/doi/10.1021/jacs.0c08529) the existence of a new family of carbon molecules of defined molecular weight, reproducible architecture, and synthetically reproducible by others. Formed with either an electric arc synthesis or flame method, these newly isolated fullertube molecules consist of two fullerene hemisphere endcaps with a tubular body of a single layer of rolled graphene resembling a pristine nanotubular region. Since then, in 2022 we isolated an even longer fullertube, C_{120}. See https://pubs.acs.org/doi/10.1021/jacs.2c06951

Now in 2023, we have isolated several new fullertubes, but we need the help of ORNL and the CNMS to collaborate with STM and TEM to cement their tubular structures. In this presentation, I will discuss exciting new findings and request help within the community to, not only imaging assistance, but also seeking new collaborators for help with fundamental science not yet done and ideas on application development. We will conclude this presentation with our recent advances in encapsulating a metal atom(s) inside the cavity of fullertubes (i.e., metallofullertubes). In closing, who among us wouldn't want to work with new fullertube/metallofullertube molecules where everything is seminal and publishable?



David J. Masiello
University of Washington

Nanometer-Scale Spatial and Spectral Mapping of Exciton Polaritons in Structured Plasmonic Cavities

Exciton polaritons (EPs) are ubiquitous light-matter excitations under intense investigation as test beds of fundamental physics and as components for all-optical computing. Owing to their unique attributes and facile experimental tunability, EPs potentially enable strong nonlinearities, condensation, and superfluidity at room temperature. However, the diffraction limit of light and broad momentum content of fast electron probes preclude the characterization of EPs in nanoscale structured cavities exhibiting energy-momentum dispersion. Through an innovative combination of electron beam and optical probes, I will show theoretically that these limitations can be overcome to measure EPs in periodic nanophotonic cavities at their natural energy, momentum, and length scales via lattice electron energy gain spectroscopy. With the combined high momentum resolution of light and nanoscale spatial resolution of free-space focused electron beams, lattice electron energy gain spectroscopy can expose deeply subwavelength EP features using currently available monochromated, aberration-corrected scanning transmission electron microscopes (STEMs). Time permitting, I will also discuss the addition of polarization structure to the STEM electron probe and demonstrate theoretically how this degree of freedom can be harnessed to characterize the 3D polarization-resolved response field and local chirality of an optically excited target with nanoscale spatial resolution.

B. Jill Venton
University of Virginia

B. Jill Venton, Nickolay Lavrik, Zijun Shao, He Zhao

Nanoscale Carbon Electrodes for Neurotransmitter Detection

Microelectrodes are the standard method for measuring neurotransmitters *in vivo*, but their design has been based on carbon fibers, which limits the geometries possible. However, improvements in sensitivity and decreases in size are necessary to improve performance and allow measurements in small brain regions. Here, we introduce a novel, implantable and freestanding microsensor fabrication method using two-photon nanolithography followed by pyrolysis. This 3D printing method allows the fabrication of free-standing carbon microelectrodes with customizable geometry and electroactive carbon surface, which is suitable for neurotransmitter detection. We have optimized the shape and size of these electrodes to allow custom electrochemistry and to promote trapping effects. We have also developed methods to make 3D printed carbon nanoelectrodes, as pyrolysis carbonizes and shrinks the photoresist, so the final sensor feature size can be smaller than the resolution of the printer. Electrodes can be made with nanosized features, such as spikes, or as nanoelectrode disks. Recently, we developed a pulling based method to create long nanoneedle electrodes using 3D printing. This technology is increasing the functional geometries that can be designed using 3D printing and expanding our capabilities for making long electrodes that are useful at synapses or in small animals. We have demonstrated the use of these electrodes *in vivo* and in *Drosophila* for measurements of neurotransmitters. We are also using the Nanoscribe laser to produce laser-induced graphene and to continue to develop new electrodes for neurochemistry. Overall, 3D printing is a useful process for making precision nanoelectrodes for neurotransmitter detection.

Liping Yu
University of Maine

Descriptor-Enabled High-Entropy Materials Design and Discovery Over Vast Chemical Spaces

Advanced energy and electronic technologies rely heavily on materials that face significant challenges in development and discovery. As the development of new materials composed of two or three elements approaches the limit of feasible combinations, a promising field in materials science has emerged: High-Entropy Materials (HEMs), which consist of five or more principal elements. HEMs possess many unique properties that are beneficial for a wide range of energy and electronic applications, including catalysts, solid fuel cells, batteries, solar cells, high-temperature transistors, coatings, and sensors. However, despite their potential, only a few hundred HEMs have been experimentally reported to date, which is negligible compared to the millions of known inorganic solid materials. In this talk, I will present (i) the critical knowledge gaps in the design and discovery of new HEMs, (ii) our recently developed Mixed Enthalpy-Entropy Descriptor (MEED), which enables robust high-throughput first-principles prediction of new HEMs over large chemical spaces, and (iii) MEED predictions of new high-entropy metal carbides and new high-entropy layered 2D transition metal chalcogenides, along with their experimental validation.

Dr. Feng-Yuan Zhang
University of Tennessee-Knoxville

Engineering Catalysts and Electrodes for High-Efficiency Hydrogen Production

Water electrolysis is becoming's a more promising technology for green hydrogen/oxygen production with renewable energy sources and playing more crucial roles for decarbonization and net-zero economy. High-efficiency and low-cost electrodes with ultrahigh catalyst utilizations are strongly urgently desired. By taking advantage of the novel liquid/gas diffusion layers (LGDLs) coupled with the development of a transparent PEMEC and a high-speed micro visualization system, the rapid electrochemical reactions and multiphase transport inside PEMECs are revealed to occur only on the catalyst layer adjacent to good electrical conductors. Based on these findings, an engineered catalyst and innovative electrode design strategy of catalyst-coated LGDLs is proposed to tune electrocatalysts with abundant exposed edges and nanopores, and to promote electron/proton transport nanohighways for scalable, low-cost, and robust water electrolysis. Various electrocatalysts with well-controlled morphologies/nanostructures, tunable crystal structures (e.g., amorphous vs. crystalline) and precisely regulated chemical compositions will be are discussed to enhance oxygen evolution reactions (OERs) and hydrogen evolution reactions (HERs).