"Novel Materials for Neuromorphic Computing"
2023 CNMS User Meeting Workshop
Monday, August 7th, 2023
Salon A, Crowne Plaza Knoxville, 401 W Summit Hill Dr SW, Knoxville, TN 37902

Organizers: Liangbo Liang, Sabine Neumayer, Saban Hus
Teams link for virtual talks: https://teams.microsoft.com/l/meetup-join/19%3ameeting_ZGY4ZDYyZmMtMDhhOS00YjcwLWIwZWMtZmQwMmE2ODk5ZmMx%40thread.v2/0?context=%7b%22Tid%22%3a%22db3dbd43-4c4b-4544-9f8a-0553f9f5f25e%22%2c%22Oid%22%3a%22755291a1-f59c-4fb2-b30e-57fb3611699a%22%7d

Agenda

8:00-8:10 AM Opening Remarks – Liangbo Liang, Oak Ridge National Laboratory

Session 1 (Chair - Sabine Neumayer)

8:10-8:50 AM Alec Talin, Sandia National Laboratory
Title: "*Dynamically Reconfigurable Electrochemical Random Access Memory (ECRAM)*"

8:50-9:20 AM Santanu Mahapatra, Indian Institute of Science (virtual talk)
Title: "*Generalized theory for nonvolatile resistive switching in 2D memristors*"

9:20-9:50 AM Pavan Nukala, Indian Institute of Science (virtual talk)
Title: "*Materials networks at criticality on 2D hBN platform*"

9:50 AM 15 min Break

Session 2 (Chair - Liangbo Liang)

10:05-10:45 AM Alejandro Strachan, Purdue University
Title: "*Modeling electrochemical processes with large-scale reactive molecular dynamics*"

10:45-11:15 AM Panchapakesan Ganesh, Oak Ridge National Laboratory
Title: "*Harnessing Electron Correlations and Anharmonicity for Energy Efficient Computing*"

11:15-11:45 AM Jerry Bernholc, North Carolina State University
Title: "*Exascale Quantum Transport Simulations*"

11:45 AM-1:00 PM Lunch

Session 3 (Chair - Saban Hus)

1:00-1:40 PM Deji Akinwande, The University of Texas at Austin
Title: "*Atomristors: Single-Atom Memristors and Low-Energy Electronic Systems*"

1:40-2:10 PM Peter Maksymovych, Oak Ridge National Laboratory
Title: "*Multimodal microscopy for intrinsic dynamics of neuromorphic materials*"

2:10-2:40 PM Patrick Collier, Oak Ridge National Laboratory
Title: "*Short-term, long-term, and heterosynaptic plasticity of memristive and memcapacitive lipid bilayer membranes for neuromorphic applications*"

2:40 PM 15 min Break

Session 4 (Chair - Peter Maksymovych)

2:55-3:25 PM Yiyang Li, University of Michigan
Title: "*Why are memristors nonvolatile?*"

3:25-3:55 PM Nikhil Shukla, University of Virginia
Title: "*Insights into the Electrical Stress Response and Endurance of Ferroelectric Hafnium Zirconium Oxide Thin Films*"

3:55-4:15 PM Michael Zachman, Oak Ridge National Laboratory
Title: "*Electron Microscopy Capabilities at the CNMS*"

4:15-4:35 PM Sabine Neumayer, Oak Ridge National Laboratory
Title: "*Functional atomic force microscopy for nanoscale neuromorphic characterization*"

Final Session

4:35 PM Open Discussion

**Each talk includes 2-5 minutes for QA depending on the length of the talk*

See following pages for abstracts.

Abstracts

Dynamically Reconfigurable Electrochemical Random Access Memory (ECRAM)

A. Alec Talin
Sandia National Labs, Livermore, CA

Devices that provide real-time hardware reconfigurability will greatly expand the capabilities of analog computing. In my talk I will discuss our recent results with electrochemical random access memory (ECRAM) devices that are reconfigured by dynamically tuning defect concentration to function as switches for signal routing and demultiplexing, tunable synapses and somas, and variable-frequency oscillators. I will discuss the physical mechanisms behind programming investigated using spectroscopic, and transmission electron microscopy and compositional measurements. Our results suggest that phase transformations and co-existance play an integral role in the switching and state retention characteristics. I will discuss the implications of our findings for emerging neuromorphic computing hardware.

Generalized theory for nonvolatile resistive switching in 2D memristors (virtual talk)

Santanu Mahapatra
Nano-Scale Device Research Laboratory, Department of Electronic Systems Engineering, Indian Institute of Science (IISc), Bangalore, Bangalore - 560012, India

Resistive switching (RS) devices have drawn significant interest in the emerging in-memory computing paradigm. While oxide-based memristor technology is nearing commercialization, the recent appearance of diverse two-dimensional (2D) materials has seen a surge of experimental demonstrations in which atomically thin layers are used as insulators. These 2D Metal-Insulator-Metal (MIM) structures can overcome the vertical scaling limitation of oxide-based devices and provide highly dense, rapid, and ultra-low power technology solutions. Despite their technological importance, the underlying physics of ultra-fast resistive switching (RS) in these devices is not fully understood. In this talk we discuss first-principles based atomistic modeling strategy to elucidate the remarkable nonvolatile resistive switching phenomena in 2D materials. Combining reactive molecular dynamics, density functional theory-based calculation and quantum transport modelling techniques we try to assess the movement of atoms in point-defect based monolayer MoS_2 and hBN based, which can explain some of the experimental observations. We also try to come up with a generalized theory which may be applicable for mono and multi-layer based memristive devices.

Materials networks at criticality on 2D hBN platform (virtual talk)

Pavan Nukala
Centre for Nano Science and Engineering, Indian Institute of Science, Bengaluru 560012

Networks and systems which exhibit brain-like behavior can analyze information from intrinsically noisy and unstructured data with very low power consumption. Such characteristics arise due to the critical nature and complex interconnectivity of the brain and its neuronal network. We demonstrate that a system comprising of multilayer hexagonal Boron Nitride (hBN) films contacted with Silver (Ag), that can uniquely host two different self-assembled networks, which are self-organized at criticality (SOC). This system shows bipolar resistive switching between high resistance (HRS) and low resistance states (LRS), each of which is exhibits a neural network of two different universality classes. In the HRS, Ag clusters (nodes) intercalate in the van der Waals gaps of hBN forming a network of tunnel junctions, whereas the LRS contains a network of Ag filaments. The temporal avalanche dynamics in both these states exhibit power-law scaling, long-range temporal correlation, and SOC. These networks can be tuned from one to another with voltage as a control parameter. All in all, I'll speak about two different neuron-like networks that are realized in a single CMOS compatible, 2D materials platform, and I will discuss various efforts (including ours) to start computing with these "black-box" networks.

Modeling electrochemical processes with large-scale reactive molecular dynamics

Alejandro Strachan
School of Materials Engineering and Birck Nanotechnology Center
Purdue University, West Lafayette, Indiana, 47906 USA

First principles-based modeling is playing an increasingly important role in the design and understanding of new materials and devices for a range of applications. I will discuss recent progress by our group on the application of these tools to science and engineering problems and efforts on the open platform nanoHUB aimed at making these tools and the associated data findable, accessible, interoperable, and reusable (FAIR).

Reactive electrochemical simulations. I will discuss reactive molecular dynamics (MD) simulations of the operation of nanoscale resistance switching devices of interest in nanoelectronics. These simulations are enabled by electrochemical dynamics with implicit degrees of freedom (EChemDID), a model to describe the electrochemical driving force in reactive MD simulations. An additional dynamical variable is assigned to each atom to describe the local electrochemical potential in its vicinity, and we use fictitious, but computationally convenient, dynamics to describe its equilibration within connected metallic structures. This local potential is used to dynamically modify the atomic electronegativities used to compute partial atomic changes via charge equilibration. I will demonstrate the use of EChemDID to simulate the operation of two classes of resistance-switching devices: electrochemical metallization and valence change cells. The simulations predict the ultrafast switching observed experimentally and provide new insight into the atomistic mechanisms behind the development and dissolution of stable conducting filaments both in both classes of devices.

Making workflows and data FAIR. Finally, I will also describe recent developments in nanoHUB, an open cyberinfrastructure for cloud scientific computing, towards making simulation workflows and their data findable, accessible, interoperable, and reusable (FAIR). We introduce Sim2Ls (pronounced sim tools) and the Sim2L Python library that allows developers to create and share end-to-end computational workflows with well-defined and verified inputs and outputs. The Sim2L library makes Sim2Ls, their requirements, and their services discoverable, verifies inputs and outputs, and automatically stores results in a globally accessible simulation cache and results database.

Harnessing Electron Correlations and Anharmonicity for Energy Efficient Computing

P. Ganesh
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Metal oxide-based Resistive Random-Access Memory (RRAM) exhibits multiple resistance states, arising from the activation/deactivation of a conductive filament (CF) formed by oxygen vacancies inside a switching layer – due an underlying metal-insulator transition (MIT). You can make scalable memory/logic units using the crossbar architecture. So, you have a highly dense information-storage system, that you want to be reliable, and fast switching and utilizing low-power. This can enable emulating 'brain like' neuromorphic computing. Similarly, ferroelectric materials – such as Hafnia – are promising candidates for synaptic weight elements in neural network hardware because of their nonvolatile multilevel memory effects. But conventional RRAM materials require high forming potentials (not viable in crossbars), show high variability (device-to-device or cycle-to-cycle). Similarly, less reliability and voltage-time dilemma are suspected to plague ferroelectric synapses. To address these challenges, we are working to answer these open questions such as: what material characteristics we need when choosing a memristor material? What factors triggers a state-change (e.g. MIT, magnetic-transition, or ferroelectric-switching) in these materials? What determines the dynamics of the switching mechanism?

Using a combination of high-throughput phase-field and machine-learning methods [1] we discovered that harnessing electron-electron correlations in binary oxides can be advantageous for improved performance of RRAM devices. Using a combination of various correlated electronic structure methods, such as Quantum Monte Carlo (QMC) and Dynamical Mean Field Theory (DMFT), we further uncovered the underlying factors that control the MIT in correlated binary oxides – such as VO_2 – when defects such as oxygen vacancies are present [2,3]. We subsequently demonstrated how many of the correlated perovskite metals that undergo MIT are negative charge-transfer metals, with the magnitude of ligand-hole being the key to controlling MIT. As such, the underlying mechanism of MIT is similar in such charge-transfer metals, irrespective of whether the MIT is induced by changes in stoichiometry or chemistry or pressure [4]. This work provides a fundamental understanding to resistive switching in RRAM's. For ferroelectric-based synapses, we explored the recently discovered 2D layered-thiophosphate family of materials [5]. We discovered [6] presence of strong anharmonic coupling in these materials between the polar-mode and a strain-tunable Raman active symmetric-mode, even down to single-layer thickness, which could alleviate the voltage-time dilemma in conventional ferroelectrics. We further show our recent findings of scale-free ferroelectricity in this 2D layered ferroelectric family of materials [7].

References:

[1]" High-throughput phase-field simulations and machine learning of resistive switching in resistive random-access memory", **npj Computational Materials volume 6**, **Article number: 198 (2020)**, Kena Zhang, Jianjun Wang, Yuhui Huang, Long-Qing Chen, **P. Ganesh*** & Ye Cao.

[2]" Doping a bad metal: Origin of suppression of the metal-insulator transition in nonstoichiometric VO2", **Phys. Rev. B 101, 155129, (2020)**, **P. Ganesh***, Frank Lechermann, Ilkka Kylänpää, Jaron T. Krogel, Paul R. C. Kent, and Olle Heinonen

[3]" Metal–insulator transition tuned by oxygen vacancy migration across -TiO2/VO2 interface", **Scientific Reports**, **10**, **1854 (2020)**, Qiyang Lu, Changhee Sohn, Guoxiang Hu, XiangGao, Matthew F. Chisholm,

Ilkka Kylänpää, JaronT. Krogel, Paul R. C. Kent, Olle Heinonen, **P.Ganesh** and Ho Nyung Lee
[4]"Origin of Metal-Insulator Transitions in Correlated Perovskite Metals", **Phys. Rev. Research 4**, **L022005 (2022)**, M. Chandler Bennett, Guoxiang Hu, Guangming Wang, Olle Heinonen, Paul R. C. Kent, Jaron T. Krogel, **P. Ganesh***.
[5]"Tunable quadruple-well ferroelectric van der Waals crystals", **Nature Materials**, **19**, **43 (2020)**, John A. Brehm, Sabine M. Neumayer, Lei Tao, Andrew O'Hara, Marius Chyasnavichus, Michael A. Susner, Michael A. McGuire, Sergei V. Kalinin, Stephen Jesse, **P. Ganesh**, Sokrates T. Pantelides, Petro Maksymovych and Nina Balke
[6]"Origin and stabilization of ferrielectricity in CuInP2Se6", **PHYSICAL REVIEW RESEARCH 4**, **013094 (2022)**, Nikhil Sivadas, Peter Doak, **P. Ganesh***
[7]"Scale Free Ferroelectric Polarization and Responses in 2D Ferroelectrics", Nikhil Sivadas, Bobby G. Sumpter and **P. Ganesh***, (under review)

Exascale Quantum Transport Simulations

Jerry Bernholc
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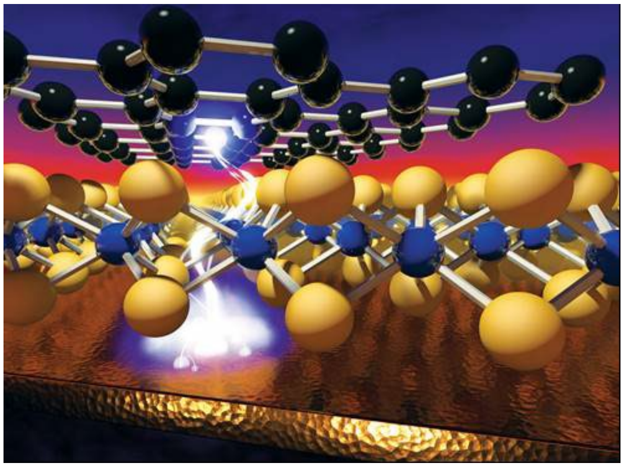
Neuromorphic computing relies on novel materials and device structures that must be discovered and evaluated. Large-scale DFT calculations can predict, prototype, and evaluate candidate materials and devices before experimental exploration and/or help interpret experimental findings and suggest future directions. We describe large-scale DFT calculations modeling paradigmatic device structures, starting from small-molecule sensors and negative differential resistance devices to a nano circuit that could potentially enable the electrical sequencing of DNA. The calculations use the open-source real-space multigrid (RMG) suite of codes, which contains a non-equilibrium Green's function module capable of handling tens of thousands of atoms with full DFT precision. RMG performs very well on DOE's exascale Frontier and pre-exascale Summit, Perlmutter, and Polaris supercomputers. It is downloadable from github.com/RMGDFT.

Atomristors: Single-Atom Memristors and Low-Energy Electronic Systems

Deji Akinwande
University of Texas – Austin



This presentation focuses on the discovery of memory effect in 2D atomically-thin nanomaterials towards greater scientific understanding and advanced engineering applications. Non-volatile memory devices based on 2D materials are an application of defects and is a rapidly advancing field with rich physics that can be attributed to vacancies combined with metal adsorption. In particular the talk will highlight our pioneering work on monolayer memory (atomristors) that has expanded to over a dozen 2D sheets and can enable various applications including zero-power devices, non-volatile RF switches, and memristors for neuromorphic computing. These memory devices offer high-energy efficiency and fast switching that may benefit mobile systems, cloud computing and data centers towards reduced energy consumption worldwide. Much of these research achievements have been published in nature, advanced materials, IEEE, and ACS journals.

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[5] S. Chen, M. R. Mahmoodi, … D. Akinwande, D. B. Strukov, and M. Lanza, "Wafer-scale integration of two-dimensional materials in high-density memristive crossbar arrays for artificial neural networks," *Nature Electronics*, 10/2020.

Multimodal microscopy for intrinsic dynamics of neuromorphic materials

Petro Maksymovych
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The connection between device function and fundamental physical processes is a long-standing challenge. In the case of resistive and ferroelectric switching at the heart of emerging computing devices, the challenge is further exacerbated by non-equilibrium nature of the underlying dynamics. Scanning probe microscopy is a very promising technique, that can bridge the nanoscale gap between idealized atomic models and the functional scale of information devices. Here I will present our recent work on multimodal switching microscopy, that enabled robust inference of polarization dynamics in multiwell ferroelectrics and extension of SPM toward switching dynamics for advanced microelectronic and neuromorphic materials. Specifically, we recently succeeded to probe of the characteristic multiwell shape of the polarization potential in in oxygen- and lead-free thiophosphate ferroelectrics [2-4]. Here I will discuss the application of topological data analysis to statistically robust identification of the signatures of the multiwells from the switching hysteresis loops. Abstracting from the raw coordinates into dynamic embedding representations enables effective comparison across the spectrum of measurements, as well as comparison to numerical models on dynamical grounds. These analyses also identify the signatures of the specific dynamics that leads to still rare memcapacitive behavior [2,4], that, somewhat ironically, is prohibited in ideal double-well ferroelectrics. Finally, I will discuss the possibility to bridge multimodal microscopy to intrinsic resistive switching in oxides, where the connection to underlying dynamics remains elusive.
Research funded in part by the DOE Office of Science Research Program for Microelectronics Codesign (sponsored by ASCR, BES, HEP, NP, and FES) through the Abisko Project. Scanning probe experiments were carried out at the Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, which is a DOE sponsored User facility.

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Short-term, long-term, and heterosynaptic plasticity of memristive and memcapacitive lipid bilayer membranes for neuromorphic applications

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Synaptic plasticity refers to synapses between neurons increasing in strength. It is usually associated with homosynaptic plasticity, which refers to a synaptic junction that is controlled by interactions with a specific neuron. Generally, it is associated with specific chemical interactions of a single species or type of neurotransmitters and associated membrane receptors and ion channels across a synaptic cleft. Heterosynaptic plasticity, on the other hand, lacks this specificity. It involves much larger populations of synapses and neurons and can be associated with changes in synaptic strength due to nonlocal changes in the ambient electrochemical environment. In biology, heterosynaptic plasticity regulates homeostasis in synaptic inputs during associative learning and memory. In this talk, specific examples will be given that show how the background electrochemical environment of lipid membranes can be varied to affect the nonlinear dynamical behaviors of memristive and memcapacitive systems in droplet interface bilayers (DIBs). Examples include:

1. Memristance in DIB membrane systems that was strongly affected by crowded environments, manifested as downward shifts in threshold voltages for alamethicin pore formation as a function of aqueous macromolecular crowding in the two droplets. This was due to an increased chemical potential of alamethicin monomers at the membrane in the presence of macromolecular crowders and increased osmotic stress in the bilayers due to excluded volumes.

2. The use of pH as a modulatory “interneuron” that changes the voltage-dependent memristance of alamethicin ion channels in lipid bilayers by changing the structure and dynamical properties of the bilayer. Barriers to conduction from membrane-bound ion channels can be lowered by reducing solution pH, resulting in higher currents, and enhanced short-term learning behavior due to “force from lipid” interactions related to pH-dependent, peptide-lipid hydrophobic mismatch.

3. The discovery of long-term potentiation (LTP) in lipids-only, DIBs-based memcapacitive systems after applying electrical stimulation protocols (i.e., tetani) that induced progressive asymmetric charging of the membrane as a result of the system being driven into nonequilibrium steady states.

Tuning environmental variables such as these provide additional training and learning algorithms that can be used to elicit complex functionality within artificial neural networks comprised of lipid bilayer membranes.

Why are memristors nonvolatile?

Yiyang Li
Materials Science and Engineering, University of Michigan

Redox-based resistive memory devices, also known as memristors or ReRAM, are highly promising elements for embedded nonvolatile memory, in-memory computing, and neuromorphic computing. Such devices switch resistance states through the electrochemical migration of oxygen vacancies in transition metal oxides. We present our recent research on the materials thermodynamics of ionic motion in these oxygen-based memristors. Using a combination of device measurements, materials characterization, and multiscale physical modeling, we find that oxygen vacancies do not obey Fick's First Law of diffusion as conventionally believed, but instead undergo composition phase separation, which enables diffusion against the concentration gradient. This phase separation is critical to the ability of memristors to retain information for long periods of time. Finally, we utilize this understanding of phase separation in transition metal oxides to engineer retention time in a new class of three-terminal electrochemical memory.

Insights into the Electrical Stress Response and Endurance of Ferroelectric Hafnium Zirconium Oxide Thin Films

Nikhil Shukla
Electrical and Computer Engineering, University of Virginia

Despite its scalability and CMOS process compatibility, the limited endurance and sub-optimal stress response of ferroelectric Zr-substituted hafnia [(Hf,Zr)O2] have been one of the key impediments toward its integration into practical device and technology applications. In this talk, we investigate the underlying mechanisms behind this behavior using detailed electrical measurements complemented by photoluminescence spectroscopy. Analyzing the evolution of leakage current with stress cycles and the spectroscopic response of the stress-induced leakage current, we attribute the behavior to defect levels, which lie at 0.6 eV from the conduction band edge of the ferroelectric. Photoluminescence spectroscopy, in turn, further corroborates the defect level's position within the bandgap while enabling its attribution to the presence of oxygen vacancies. Our work helps to identify oxygen vacancies as the key factor responsible for the degraded endurance and stress response in (Hf,Zr)O2 and subsequently motivates the exploration of methods to reduce the oxygen vacancy concentrations without destabilizing the ferroelectric orthorhombic phase. Our work advances the understanding of the reliability and failure mechanisms in ferroelectric hafnia. The specific defect levels revealed through our work also provides clear directions towards improving the reliability and endurance of this materials to meet technological needs.

Electron Microscopy Capabilities at the CNMS

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The Center for Nanophase Materials Sciences (CNMS) is a US Department of Energy, Office of Science User Facility and houses a variety of high-end electron microscopes, from dedicated scanning transmission electron microscopes (STEMs) and cryogenic transmission electron microscopes (cryo-TEMs) to versatile analytical (S)TEM instruments. In this talk, I will outline the instruments available through the CNMS user program and the capabilities that they provide, as well as the areas of expertise of the microscopy staff at the CNMS.

Functional atomic force microscopy for nanoscale neuromorphic characterization

Sabine M. Neumayer
Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA

The increasing importance of big data and artificial intelligence demands computing paradigms that combine high performance with high energy efficiency. Neuromorphic computing integrated with neuromorphic hardware architectures provides a solution by emulating the highly parallel and energy efficient way the brain can process and store information in biological building blocks called synapses and neurons. To achieve neuromorphic hardware elements, the functional properties of materials and material systems are crucial. In this talk, I will show how atomic force microscopy tools that are available at the Center for Nanophase Materials Sciences can be used to locally characterize neuromorphic material characteristics, including adaptable signal transfer, memory functionality and multi-state behavior. The micro- and nanoscale information gained from atomic force microscopy bridges the gap between macroscopic devices performance and atomic scale experimental and theoretical data.

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