

Nanoscale Science and Technology Room 114 - Session NS1+2D+QS-MoA

Functionality in 2D Nanostructures and Devices

Moderator: Dahlia Klein, Weizmann Institute of Science, Israel

1:30pm NS1+2D+QS-MoA-1 Low-Dimensional Neuromorphic Electronic Materials and Applications, Mark Hersam, Northwestern University INVITED

The exponentially improving performance of digital computers has recently slowed due to the speed and power consumption issues resulting from the von Neumann bottleneck. In contrast, neuromorphic computing aims to circumvent these limitations by spatially co-locating logic and memory in a manner analogous to biological neuronal networks [1]. Beyond reducing power consumption, neuromorphic devices provide efficient architectures for image recognition, machine learning, and artificial intelligence [2]. This talk will explore how low-dimensional nanoelectronic materials enable gate-tunable neuromorphic devices [3]. For example, by utilizing self-aligned, atomically thin heterojunctions, dual-gated Gaussian transistors have been realized, which show tunable anti-ambipolarity for artificial neurons, competitive learning, spiking circuits, and mixed-kernel support vector machines [4,5]. In addition, field-driven defect motion in polycrystalline monolayer MoS₂ enables gate-tunable memristive phenomena that serve as the basis of hybrid memristor/transistor devices (i.e., 'memtransistors') that concurrently provide logic and data storage functions [6]. The planar geometry of memtransistors further allows multiple contacts and dual gating that mimic the behavior of biological systems such as heterosynaptic responses [7]. Moreover, control over polycrystalline grain structure enhances the tunability of potentiation and depression, which enables unsupervised continuous learning in spiking neural networks [8]. Finally, the moiré potential in asymmetric twisted bilayer graphene/hexagonal boron nitride heterostructures gives rise to robust electronic ratchet states. The resulting hysteretic, non-volatile injection of charge carriers enables room-temperature operation of moiré synaptic transistors with diverse bio-realistic neuromorphic functionalities and efficient compute-in-memory designs for low-power artificial intelligence and machine learning hardware [9].

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2:00pm NS1+2D+QS-MoA-3 Defect Manipulation in van der Waals Heterostructures and its Applications, Son Le, Laboratory for Physical Sciences; T. Mai, M. Munoz, A. Hight Walker, C. Richter, 100 Bureau Dr.; A. Hanbicki, A. Friedman, 8050 Greenmead Dr. INVITED

Reliable and accurate spatial doping of 2-dimensional (2D) materials is important for future applications using this novel class of materials. Here, we present our work on photo-doping of an h-BN/Graphene/h-BN heterostructure. Natural defect states in bulk h-BN can remotely dope graphene and can be optically activated or deactivated. In this way, we can modify both the carrier density and type in graphene accurately and reversibly by several orders of magnitude. Using a spatially-resolved light source, we can activate photo-dopants in selected areas of the sample, and by laterally modulating the doping, we have created PNP junction (PNPJ) devices. *In-situ* quantum Hall measurements were used to demonstrate the effectiveness of this doping technique and characterize the electrostatic profile of the PNPJ. Doping and undoping the heterostructure in a specific sequence, we were able to introduce and destroy correlation among the dopants. Defect correlation greatly enhances carrier mobility while the destruction of this correlated state significantly degrades the carrier mobility in the graphene, effectively creating a mobility switch. An elegant demonstration of this mobility switch is the observation of spin and valley-resolved Landau levels of the graphene in the quantum Hall regime with high-mobility, dopant correlated states, and spin and valley -degenerate Landau levels in the low-mobility, dopant uncorrelated states. I will discuss

ongoing studies to better understanding the nature of these defects with photo-doping measurements of different hBN thicknesses as well as hBN from different sources. This doping technique opens up the possibility to engineer novel device and expand the applications of 2D heterostructures.

2:30pm NS1+2D+QS-MoA-5 Extraordinary Tunnel Electroresistance in Layer-by-Layer Engineered Van Der Waals Ferroelectric Tunnel Junctions, Qinqin Wang, Department of Electrical and Computer Engineering and Quantum Technology Center, University of Maryland, College Park

The ability to engineer potential profiles of multilayered materials is critical for designing high-performance tunneling devices such as ferroelectric tunnel junctions (FTJs). FTJs comprise asymmetric electrodes and a ferroelectric spacer, promising semiconductor platform-compatible logic and memory devices. However, the traditional FTJs consisting of metal/oxide/metal multilayer heterostructures can only exhibit modest tunneling electroresistance (TER, usually $<10^6$), which is fundamentally undermined by the unavoidable defect states and interfacial trap states. Here, we constructed van der Waals (vdW) FTJs by a layered ferroelectric CuInP₂S₆ (CIPS) and graphene. Owing to the gigantic ferroelectric modulation of the chemical potentials in graphene by as large as ~ 1 eV, we demonstrated a giant TER of 10^9 . While inserting just a monolayer MoS₂ between CIPS/graphene, the off state is further suppressed, leading to $>10^{10}$ TER. Our discovery opens a new solid-state paradigm where potential profiles can be unprecedentedly engineered in a layer-by-layer fashion, fundamentally strengthening the ability to manipulate electrons' tunneling behaviors and design advanced tunneling devices.

Keywords: 2d materials, ferroelectric tunnel junctions, tunneling electroresistance

2:45pm NS1+2D+QS-MoA-6 Scanning Tunneling Microscopy Studies of Twisted Transition Metal Dichalcogenides, Adina Luican-Mayer, STEM 150 Louis Pasteur Private, Canada

Material systems, devices, and circuits, based on the manipulation of individual charges, spins, and photons in solid-state platforms are key for quantum technologies. Two-dimensional (2D) materials present an emerging opportunity for the development of novel quantum technologies, while also pushing the boundaries of fundamental understanding of materials. Our laboratory aims to create quantum functionality in 2D systems by combining fabrication and assembly techniques of 2D layers with atomically precise microscopy.

In this talk, I will focus on experimental observations of novel phenomena in moiré structures created by twisting 2D layers using scanning tunnelling microscopy and spectroscopy. I will discuss the demonstration of reversible local response of domain wall networks using scanning tunneling microscopy in ferroelectric interfaces of marginally twisted WS₂ bilayers. Moreover, in the case of twisted WS₂ bilayers close to 60°, we observe signatures of flat bands and study the influence of atomic relaxation on their band structure.

3:00pm NS1+2D+QS-MoA-7 Hybrid Molecule/Quantum Material Van Der Waals Heterostructures, Emanuele Orgiu, Institut National de la Recherche Scientifique (INRS), Canada

The rise of graphene and related 2D materials makes it possible to form heterostructures held together by weak interplanar van der Waals (vdW) interactions. The interactions of such 2D layers with adventitious contaminants is able to exert a strong effect on its major electronic characteristics [1]. However, the controlled incorporation of ordered organic molecules within these systems holds an immense potential. Whilst nature offers a finite number of 2D materials, an almost unlimited variety of molecules can be designed and synthesized with predictable functionalities [2-3]. The possibilities offered by systems in which continuous molecular layers are interfaced with inorganic 2D materials to form hybrid organic/inorganic van der Waals heterostructures are emphasized. Similar to their inorganic counterpart, the hybrid structures have been exploited to put forward novel device architectures. Moreover, specific molecular groups can be employed to modify intrinsic properties and confer new capabilities to 2D materials. This work intends to provide the audience with a brief overview of how molecular self-assembly at the surface of 2D materials can be mastered to achieve precise control over position and density of (molecular) functional groups, paving the way for a new class of hybrid functional. In particular, within such vdW heterostructures, currently assembled by mechanical superposition of different layers, periodic potentials naturally occur at the interface between the 2D materials. These potentials significantly modify the electronic structure of the individual 2D

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components within the stack and their alignment, thus offering the possibility to build up hybrid and novel materials with unique properties.

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3:15pm **NS1+2D+QS-MoA-8 Revealing Quantum Functionality of Thin Films by in situ Characterization with Materials Cluster System, Wonhee Ko**, University of Tennessee Knoxville

Achieving unique quantum properties from the nanostructures is a key to realize novel electronic and quantum devices. Thin films of quantum materials are a promising candidate, but the quantum states in these films are highly fragile to the ambient condition and require in situ growth and characterization techniques. With collaboration between several groups in Oak Ridge National Laboratory and University of Tennessee, Knoxville, we built materials cluster system that combines in situ epitaxial film growth and characterization instruments, such as molecular beam epitaxy (MBE), pulsed laser deposition (PLD), angle-resolved photoemission spectroscopy (ARPES), and scanning tunneling microscopy (STM). We used the materials cluster system to grow thin films of topological insulators with MBE and observed lattice and electronic structures in atomic scale with STM and ARPES. Interestingly, we found that the step edges possess Rashba edge states with unique spin texture, which interacts with topological surface states depending on the film thickness. The results demonstrate the potential of materials cluster system to exhibit the unique quantum functionality of thin film materials, which will become a foundation for realizing future quantum devices.

3:30pm **NS1+2D+QS-MoA-9 Functionalized 2D Materials for Sustainable Energy Nanomaterials, Nozomi Shirato**, Argonne National Lab

Advancements in carbon dioxide capturing and reduction (CDR) technology are essential to mitigate greenhouse gas-induced climate changes. Currently, the main challenges of CDRs are economical unviability and poor scalability. The need for next-generation high-performance and cost-effective CDR nanomaterials is paramount. Here, we explore functionalized nanomaterials synthesized on a platinum single crystal for various thermocatalytic functionalities. We aim to understand the roles of single monolayer thick 2D materials for thermocatalytic performance by harnessing atomic-scale surface imaging capability. Utilizing a low-temperature scanning tunneling microscope (LT-STM) to characterize surfaces at atomic resolution, the fully automated temperature-programmed desorption (TPD) measurements evaluate catalytic performance and repeatability under various conditions. The first-principles-based study supports the results, providing additional fundamental insights into CDR technologies.

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