## 2D Materials
## Room 122 - Session 2D+EM+MI+QS-WeM
## 2D Materials: Heterostructures, Twistronics, and Proximity Effects

**Moderators: Aaron Bostwick**, Advanced Light Source, Lawrence Berkeley National Laboratory, **Tiancong Zhu**, Purdue University

8:00am **2D+EM+MI+QS-WeM-1 Van der Waals Semiconductors: From Stacking-Controlled Crystals to Unconventional Heterostructures***, **Peter Sutter**, E. Sutter,* University of Nebraska - Lincoln **INVITED**

2D materials have attracted broad interest due to novel properties that arise in atomically thin crystals. As interesting scientifically and important technologically, but much less explored are van der Waals (vdW) crystals that, assembled from 2D building blocks, lie between a monolayer and the bulk. In this regime, phenomena such as phase separation, transformations between crystal polymorphs, and competition between different stacking registries provide unprecedented opportunities for controlling morphology, interface formation, and novel degrees of freedom such as interlayer twist. But going beyond a single layer also poses significant challenges, both due to the diversity of the possible few-layer structures and the difficulty of probing functionality such as optoelectronics and ferroics at the relevant length scales.

Here, we discuss our recent research that addresses these challenges focusing on group IVA chalcogenides, an emerging class of anisotropic layered semiconductors promising for energy conversion, optoelectronics, and information processing. Advanced *in-situ* microscopy provides insights into the growth process, interlayer twisting,and emerging functionality such as stacking-controlled ferroelectricity. Nanometer-scale electron excited spectroscopy identifies photonic light-matter hybrid states and reveals anisotropic and valley-selective charge carrier flows across interfaces in heterostructures. Our results highlight the rich sets of materials architectures and functionalities that can be realized in van der Waals crystals and heterostructures beyond the 2D limit.

8:30am **2D+EM+MI+QS-WeM-3 Deterministic Assembly, Transfer, and Flipping of 2D Materials Using Tunable Polymer Films***, **Jeffrey J. Schwartz**, S. Le,* University of Maryland, College Park*; K. Grutter, A. Hanbicki, A. Friedman,* Laboratory for Physical Sciences

Assembly of two-dimensional (2D) materials into van der Waals heterostructures is a crucial step in creating precisely engineered nanoscale and quantum devices for use in a wide variety of spintronic, electronic, and other applications. Numerous strategies exist to pick-up, stack, transfer, and even flip over these atomically thin structures. One popular strategy leverages the ability to tune the adhesion between a polymer stamp and 2D sheets to pick-up, stack, and release structures at different temperatures. Although relatively easy to implement, this technique is tedious to perform and has a low throughput. Here, we demonstrate a significant improvement to a deterministic, all-dry, polymer-assisted transfer technique using polyvinyl chloride (PVC) thin films to manipulate 2D materials and to fabricate devices. We construct stamps from pairs of commercially available PVC films that controllably pick-up and release 2D sheets within known, overlapping temperature ranges. These mechanically durable stamps can be produced quickly and without the time-consuming preparation and annealing steps required by most other commonly used polymers. Importantly, these stamps not only facilitate deterministic transfer of 2D materials, but they also enable polymer-to-polymer transfer (e.g., between separate stamps) and flipping of material stacks to create inverted heterostructures that are important for many applications, including scanning tunneling microscopy measurements. We characterize the thermal transition properties of the PVC films employed here as well as assay the cleanliness and performance of devices produced using this technique. These improvements enable rapid production of 2D devices with fewer interactions required by the operator, which is especially significant when working in controlled environments (e.g., glovebox) or in remote or autonomously controlled contexts.

8:45am **2D+EM+MI+QS-WeM-4 Cleaning of Low-Dimensionality Materials: Challenge and Solutions***, **Jean-Francois de Marneffe**, P. Wyndaele, M. Timmermans, C. Cunha,* IMEC, Belgium*; B. Canto, Z. Wang,* AMO GmbH, Aachen, Germany*; R. Slaets, G. He, I. Asselberghs, C. J. Lockhart de la Rosa, G. Sankar Kar, C. Merckling, S. De Gendt,* IMEC, Belgium

Over the last few years, significant efforts have been made in exploring low-dimensionality materials such as single layer Graphene (SLG), transition metal dichalcogenides (TMDCs) and carbon nanotubes (CNTs), for a wide range of applications covering beyond CMOS logic, EUV pellicles, photonics, and sensing (amongst others). Due to their intrinsic 2D or 1D nature, these materials are highly sensitive to processing damage leading to stoichiometric changes or crystalline defects. Among the many manufacturing steps required for building devices, the cleaning of these systems is an absolute requirement and a bottleneck. Typically, during processing, residual polymers or carbon of ambient origin, do contaminate the surface leading to nanometric deposits that change the intrinsic transport/optical properties of the materials, and cause parasitic dielectric drift or high contact resistance. Wet cleaning, using organic solvents, is a mainstream approach, which proves to be inefficient for irreversibly physically adsorbed polymer residues. In this paper, we explore dry cleaning approaches, based on plasma treatment and UV cure. Plasma-based cleaning proves to be very efficient but leads to material damage, which can be minimized by tuning the average ion energy, the processing temperature, the plasma chemistry or adding a post-cleaning restoration step. For TMDCs, damage consist essentially in the creation of chalcogen vacancies, which lead to metal oxidation upon ambient exposure. For Graphene and CNTs, damage consist in carbon vacancies, causing lattice distortions, oxidation and ultimately a dramatic change of the material's transport properties. Part of this presentation will explore the use of UV cure, which is a known method for cleaning polymers from semiconductor surfaces.

9:00am **2D+EM+MI+QS-WeM-5 Spin-Valley Physics in Mixed-Dimensional Van Der Waals Heterostructures***, **Vikram Deshpande**,* University of Utah **INVITED**

Spin-valley physics has become ubiquitous in 2D materials-based van der Waals (vdW) heterostructures, particularly those hosting flat bands, wherein various ground states with spin-valley character including magnetic, insulating and superconducting states have been observed. On the other hand, mixed dimensional vdW heterostructures, such as those between 2D and 1D materials have been less explored for intricate spin-valley physics. The reduced phase space for scattering in 1D in particular might lead to qualitatively different phenomena. We are guided by our studies of ultraclean carbon nanotube quantum dots wherein we have observed subtle effects from the degeneracy lifting between the speeds of right- and left-moving electrons within a given Dirac cone or valley. Bound states can be purely fast-moving or purely slow-moving, giving rise to incommensurate energy level spacings and a vernier spectrum. Using quantum interferometry [1] and Coulomb blockade spectroscopy [2] of such ultraclean carbon nanotube quantum dots, we have found evidence for this vernier spectrum. The addition-energy spectrum of the quantum dots reveals an energy-level structure that oscillates between aligned and misaligned energy levels. Our data find that the fast- and slow-moving bound states hybridize at certain gate voltages. We extend existing theory to show that our experiment probes the degree of isospin polarization/hybridization of the various quantum states probed in our system. As a result, gate-voltage tuning can select states with varying degrees of hybridization, suggesting numerous applications based on accessing this isospin degree of freedom in conjunction with 2D materials in the form of mixed dimensional vdW heterostructures. We have fabricated prototypical mixed dimensional vdW heterostructures between carbon nanotubes and 2D materials and extended our measurements to these structures. I will discuss our recent and ongoing work studying spin-valley physics in such systems.

References:

Lotfizadeh, N.; Senger, M. J.; McCulley, D. R.; Minot, E. D.; Deshpande, V. V. Quantum Interferences in Ultraclean Carbon Nanotubes. Phys. Rev. Lett.2021, 126 (21), 216802. https://doi.org/10.1103/PhysRevLett.126.216802.

Berg, J.; Lotfizadeh, N.; Nichols, D.; Senger, M. J.; De Gottardi, W.; Minot, E. D.; Deshpande, V. V. Vernier Spectrum and Isospin State Control in Carbon Nanotube Quantum Dots. arXiv November 20, 2023. http://arxiv.org/abs/2311.12332.

9:30am **2D+EM+MI+QS-WeM-7 Exploring Incommensurate Lattice Modulations in BSCCO van der Waals Heterostructures: Implications for Q-Bit Development***, **Patryk Wasik**,* Brookhaven National Laboratory*; S. Zhao,* Harvard University*; R. Jangid,* Brookhaven National Laboratory*; A. Cui,* Harvard University*; J. Sinsheimer,* Brookhaven National Laboratory*; P. Kim,* Harvard University*; N. Poccia,* IFW Dresden, Germany*; C. Mazzoli,* Brookhaven National Laboratory

Quantum computers (QC) are poised to revolutionise computational capabilities by naturally encoding complex quantum computations, thereby significantly improving computation time compared to silicon-based technologies. Currently, Q-bits, the essential components of QCs, are made from conventional superconductors that operate efficiently only near absolute zero temperatures. To address this limitation, two-dimensional van der Waals (vdW) encapsulated high-temperature superconductor (HTSC) stacks have been proposed as future Q-bit candidates, driven by recent advancements in nanofabrication techniques. However, a detailed understanding of their structural and electronic properties is crucial.

We present low-temperature resonant soft X-ray investigations on ultrathin $Bi_2Sr_2CaCu_2O_{8+y}$ (BSCCO) vdW heterostructures, promising candidates for large-scale Q-bit applications. BSCCO crystals exhibit two incommensurate lattice modulations (ILMs), providing an excellent opportunity to explore the relationship between structure and electronic behaviour in low dimensions. We report ILMs (Cu $L_3$) and structural peak (off resonance) maps obtained across the superconducting transition temperature ($T_c \approx 60$ K). These signals, under external gating, present significant potential for further exploration offering new insights into the electronic interactions in vdW HTSC systems.

11:00am **2D+EM+MI+QS-WeM-13 Atomic Layer Deposition of Transition Metal Dichalcogenides: Precursors, Processes, and Applications Perspectives***, **Thong Ngo**, A. Azcatl, N. Vu, C. Cheng, M. Miller, C. Chen, R. Kanjolia, M. Moinpour, M. Clark,* EMD Electronics, USA **INVITED**

Transition metal dichalcogenides (TMD) are three-atom-thick layer materials that possess a wide array of properties, such as insulating, semiconducting, conducting, and superconducting. While there has been a significant amount of research on TMD for various applications including energy storage, photovoltaics, biomedicals, catalysis, hydrogen production processes, and healthcare, the usage of TMD for electronic applications has been researched most in the past two decades.

Each layer of TMD is a 2D sheet with the thickness of ~6-7Å. These layers are bonded by Van der Waals force. The ultra-thin structure enables TMD for semiconductor industry applications, which continuously requires both size-scaling of materials layers and electrical performance improvement of devices. The semiconductor-range band gap and the high electron/hole mobility of several TMD, such as $MoS_2$, $WS_2$, $WSe_2$, and $MoSe_2$allow their usage for high mobility ultra-thin channel transistor. In addition, the relatively high conductivity of some other TMD, such as $TaS_2$ and $NbS_2$ make them promising candidates for interconnect barrier/liner as a replacement of TaN/Ta bilayer.

TMD materials need to pass certain quality requirements to provide desirable property/performance; therefore, method of synthesizing TMD plays an important role for high quality materials. Solution-based deposition, non-vacuum electrodeposition, polymer-assisted deposition, physical vapor deposition (PVD), chemical vapor deposition (CVD), and atomic layer deposition (ALD) have been used to deposit ultra-thin TMD. Among these techniques, CVD is the most popular deposition method, and to date, the highest quality of TMD for semiconductor applications is CVD-TMD. However, the need of lower thermal budget, better layer controllability, uniformity, and conformality requires the semiconductor community to explore ALD methods. In this presentation, we will review multiple ALD processes for TMD including precursors, process requirements for high mobility channel and barrier/liner applications. We will highlight challenges of TMD applications for logic, memory, and interconnects. The presentation will also feature our recent work on ALD $MoS_2$ for high-mobility channel transistor with >5 decades On/Off ratio and >1 μA/μm Ion. Our achievement of 300mm ALD $MoS_2$ deposition brings ultra-thin TMD materials closer to a manufacturable fab process for semiconductor industry.

11:30am **2D+EM+MI+QS-WeM-15 Writing and Detecting Topological Spin Textures in Exfoliated $Fe_5$-xGete$_2$***, **Luis Balicas**,* Florida State University - National High Magnetic Field Lab - FSU Quantum Initiative

$Fe_{5-x}GeTe_2$ is a centrosymmetric, layered van der Waals (vdW) ferromagnet that displays Curie temperatures $T_c$ (270-330 K) that are within the useful range for spintronic applications. Little is known about the interplay between its topological spin textures (e.g., merons, skyrmions) with technologically relevant transport properties such as the topological Hall effect (THE), or topological thermal transport. We found via high-resolution Lorentz transmission electron microscopy that merons and anti-meron pairs coexist with Néel skyrmions in $Fe_{5-x}GeTe_2$ over a wide range of temperatures and probe their effects on thermal and electrical transport [1]. It turns out that we detect a THE, even at room *T*, that senses merons at higher *T*'s as well as their coexistence with skyrmions as *T* is lowered, indicating an on-demand thermally driven formation of either type of spin texture.Remarkably, we also observe an unconventional THE, i.e., in absence of Lorentz force, and attribute it to the interaction between charge carriers and magnetic field-induced chiral spin textures. We find that both the anomalous Hall effect (AHE) and THE can be amplified considerably by just adjusting the thickness of exfoliated $Fe_{5-x}GeTe_2$, with the THE becoming observable even under zero magnetic field due to a field-induced unbalance in topological charges [2]. Using a complementary suite of techniques, including electronic transport, Lorentz transmission electron microscopy, and micromagnetic simulations, we reveal the emergence of substantial coercive fields upon exfoliation, which are absent in the bulk, implying thickness-dependent magnetic interactions that affect the topological spin textures (TSTs). We detected a 'magic' thickness of *t* ~30 nm where the formation of TSTs is maximized, inducing large magnitudes for the topological charge density, and the concomitant AHE and THE resistivities at *T* ~ 120 K. Their values are observed to be higher than those found in magnetic topological insulators and, so far, the largest reported for 2D magnets. The hitherto unobserved THE under zero magnetic field could provide a platform for the writing and electrical detection of TSTs aiming at energy-efficient devices based on vdW ferromagnets.

[1] B. W. Casas *et al*., Adv. Mater. **35**, 202212087 (2023).

[2] A. Moon *et al.*, ACS Nano **18**, 4216-4228 (2024).

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