

## 2D Materials

Room 208 W - Session  
2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM

### 2D Materials: Optoelectronics and Moire Excitons

Moderators: Shengxi Huang, Rice University, Daniel Yimam, Oak Ridge National Laboratory

8:00am **2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM-1 Probing the Ultrafast Charge Dynamics and Exciton Emission from Single Atomic Defects in 2D Semiconductors by Lightwave-Driven STM**, Laric Bobzien, Lysander Huberich, Jonas Allerbeck, Eve Ammerman, Nils Krane, Andres Ortega-Guerrero, Carlo Pignedoli, Oliver Gröning, Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland; Joshua A. Robinson, The Pennsylvania State University; Bruno Schuler, Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland **INVITED**

Two-dimensional (2D) semiconductors provide an exciting platform to engineer atomic quantum systems in a robust, yet tunable solid-state system. This talk explores the intriguing physics of single point defects in transition metal dichalcogenide (TMD) monolayers, investigated through atomically resolved scanning probe microscopy.

We have determined the layer-dependent charge transfer lifetimes of selenium vacancies in  $\text{WSe}_2$  on graphene substrates, spanning picosecond to nanosecond timescales [1]. By leveraging our recently developed lightwave-driven scanning tunneling microscope (THz-STM) [2,3], we could probe the ultrafast charge dynamics on the atomic scale. Time-domain sampling with a THz pump-THz probe scheme enabled capturing atomic-scale snapshots of transient Coulomb blockade, a hallmark of charge transport mediated by quantized defect states [4].

Moreover, the extended charge state lifetimes provided by hBN decoupling layers facilitated the local, electrical stimulation of excitonic emission from pristine  $\text{MoS}_2$  and individual charged defects via STM luminescence (STML).

By combining the structural and electronic properties accessible by conventional scanning probe microscopy with the optical fingerprint from STML and the excited-state dynamics revealed through pump-probe THz-STM, we gain a comprehensive microscopic understanding of localized quantum states in low-dimensional materials.

#### References:

- [1] L. Bobzien et al. Phys. Rev. Lett. (accepted, arxiv: 2407.04508)
- [2] J. Allerbeck et al. ACS Photonics 10, 3888 (2023)
- [3] L. Bobzien et al. APL Mater. 12, 051110 (2024)
- [4] J. Allerbeck et al. arXiv:2412.13718 (2024)
- [5] L. Huberich et al. (in preparation)

8:30am **2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM-3 Many-Body Effects on Excitons, Trions, and Defect-Bound States in 2D Materials**, Kai Xiao, Taegwan Park, Alexander Puzetzy, Oak Ridge National Laboratory, USA; Xufan Li, Honda Research Institute; Kyungnam Kang, Oak Ridge National Laboratory, USA; Austin Houston, University of Tennessee, Knoxville; Christopher Roaleau, David Geohagan, Oak Ridge National Laboratory, USA  
Two-dimensional (2D) materials, particularly transition metal dichalcogenides (TMDs) exhibit strong many-body interactions due to reduced dielectric screening and spatial confinement. These interactions, involving electrons, holes, excitons, phonons, and plasmons, give rise to emergent phenomena distinct from their bulk counterparts. In this talk, I will present our recent investigations into the many-body effects on the optical properties and ultrafast excitonic dynamics of monolayer and bilayer TMDs. Specifically, we synthesized isotopically pure monolayer  $\text{MoS}_2$  and highly defective  $\text{WS}_2$  via nonequilibrium chemical vapor deposition, enabling a controlled study of isotope effects, defects, and background doping on excitonic behavior. Using ultrafast laser spectroscopy and temperature-dependent optical spectroscopy, we observed pronounced many-body interactions, including exciton-phonon and exciton-electron coupling, which significantly influence exciton energy, dynamics, and light-matter interactions in both monolayer and bilayer TMDs. These strong interactions give rise to novel quantum states and make 2D materials promising platforms for next-generation optoelectronics, quantum information technologies, and fundamental condensed matter physics.

Synthesis science was supported by the U.S. Dept. of Energy, Office of Science, Materials Science and Engineering Division. This work was performed at the Center for Nanophase Materials Sciences, which is a DOE Office of Science User Facility.

8:45am **2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM-4 Proximity-Induced "Magic" Raman Bands in TERS Spectra of  $\text{MoS}_2$  /  $\text{WS}_2$  @ 1L h-BN-Capped Gold**, Andrey Krayev, HORIBA Scientific; Pavel Valencia Acuna, PNNL; Ju-Hyun Jung, Pohang University of Science and Technology (POSTECH), Republic of Korea; Cheol-Joo Kim, POSTECH, Republic of Korea; Andrew Mannix, Stanford University; Eleonora Isotta, Max Planck Institute for Sustainable Materials, Germany; Chih-Feng Wang, PNNL

Recently it was proposed to use the monolayer h-BN – capped gold substrates as an ideal platform for the gap mode TERS and TEPL imaging, that on the one hand, should preserve strong gap mode enhancement of Raman signal due to small thickness (0.3 nm) of the dielectric h-BN layer, and on the other hand preserve strong TEPL response due to de-coupling of 2D semiconductors from the metallic substrate. TERS data collected on mono- and a few-layer-thick crystals of  $\text{MoS}_2$  and  $\text{WS}_2$  on 1L-h-BN-capped gold show both the TERS and TEPL response, confirming the validity of the proposed approach.

In addition to the enhancement of both the PL and Raman signal, in the course of assessment of TERS/TEPL response of mono- and a few-layer-thick crystals of  $\text{MoS}_2$  and  $\text{WS}_2$  deposited on 1L h-BN-capped gold we observed in TERS spectra, completely unexpectedly, appearance of Raman bands at about  $796\text{ cm}^{-1}$  and  $76\text{ cm}^{-1}$  which are not normally observed in regular Raman spectra of h-BN or  $\text{WS}_2/\text{MoS}_2$ . We can safely state that these "magic" bands belong to h-BN as they appear at the same spectral position in TERS spectra of both the monolayer  $\text{MoS}_2$  and  $\text{WS}_2$  deposited on the monolayer h-BN capped gold, moreover, the  $796\text{ cm}^{-1}$  band often was the strongest band observed in TERS spectra, even stronger than A' mode from  $\text{WS}_2$  or  $\text{MoS}_2$ . Presence of the transition metal dichalcogenide (TMD) monolayer is mandatory for the appearance of these "magic" bands as they are absent outside of the monolayer TMDs in these samples. Literature search showed that similar (but not identical) phenomenon was observed earlier in h-BN encapsulated  $\text{WSe}_2/\text{MoSe}_2$  and  $\text{WS}_2$ . There have been several significant differences between our data and the earlier reported one: in our case we have not been able to observe the "magic bands" in  $\text{MoSe}_2$  and  $\text{WSe}_2$  @ 1L h-BN@Au, while  $\text{WS}_2$  monolayers deposited on the same substrate as  $\text{WSe}_2$ , showed expected response. More importantly, the excitation laser wavelength dependence in our case was completely different from what was reported earlier: in  $\text{WS}_2$ -based samples we observed strong "magic" bands with excitation at 830 nm, 785nm, 594nm, but not 633nm, the wavelength closest to the A exciton in this material. This excitation profile is remarkably reminiscent of the excitation profile of the monolayer  $\text{WS}_2$  in intimate contact with silver where we observed strong dip of the intensity of main A' mode in TERS spectra at 633nm excitation wavelength.

We will argue that intricate interaction between the tip-substrate gap plasmon, TMD excitons and most probably, normally mid-IR-active phonons in h-BN is responsible for the appearance of observed "magic" bands.

9:00am **2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM-5 Correlated Excitons in TMDC Moiré Superlattice**, Suferi Shi, Carnegie Mellon University **INVITED**

In a strongly correlated electronic system, Coulomb interactions among electrons dominate over kinetic energy. Recently, two-dimensional (2D) moiré superlattices of van der Waals materials have emerged as a promising platform to study correlated physics and exotic quantum phases in 2D. In transition metal dichalcogenides (TMDCs) based moiré superlattices, the combination of large effective mass and strong moiré coupling renders the easier formation of flat bands and stronger electronic correlation, compared with graphene moiré superlattices. Meanwhile, the strong Coulomb interaction in 2D also leads to tightly bound excitons with large binding energy in TMDCs. In this talk, we will discuss how to use optical spectroscopy to investigate excitonic physics and strongly correlated phenomena in TMDC moiré superlattice, along with correlated exciton states arising from strong interactions.

9:30am **2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM-7 Sub-Stoichiometric Phases in 2D  $\text{MoTe}_2$** , Onyedikachi Alanwoko, Nirasha Rajapakse, Matthias Batzill, University of South Florida

Atom vacancy formation in crystalline materials is energetically expensive. To lower the energy cost for non-stoichiometry, point defects can condense into energetically more favorable extended defects. Studies on Mo-dichalcogenides have shown that excess Mo is condensed into closed, triangular Mirror Twin Boundary (MTB) loops. These MTBs can form in high densities where the triangular loops connect and form a cross-hatched network of MTBs. Here we show through Scanning Tunneling Microscopy (STM) that periodically ordered MTB networks can obtain a homologous series of sub-stoichiometric  $\text{MoTe}_{2-x}$  phases. We systematically investigate

the preparation conditions (which include a variation of the growth temperature, Te-desorption by post-growth annealing, and vapor-deposited Mo), enabling the controlled synthesis of these new phases. The different phases require different synthesis procedures, and once formed, these phases appear thermally stable in vacuum. The ability to control and create these different phases of MoTe<sub>2</sub> and other two-dimensional (2D) materials is a promising way of realizing new electronic and chemical properties of 2D materials. Particularly promising is the observation that we can react MoTe<sub>2</sub> with dissimilar transition metals to create new doped or alloyed 2D materials with potentially desirable properties.

**9:45am 2D+AQs+EM+MI+MN+NS+QS+SS+TF-ThM-8 Quantum Confining Excitons with Electrostatic Moiré Superlattice, Liuxin Gu, Lifu Zhang, Sam Felsenfeld, University of Maryland, College Park; Rundong Ma, University of Maryland College Park; Suji Park, Houk Jang, Brookhaven National Laboratory; Takashi Taniguchi, Kenji Watanabe, National Institute for Materials Science, Japan; You Zhou, University of Maryland, College Park**  
Quantum confining excitons has been a persistent challenge in the pursuit of strong exciton interactions and quantum light generation. Unlike electrons, which can be readily controlled via electric fields, imposing strong nanoscale potentials on excitons to enable quantum confinement has proven challenging. In this study, we utilize piezoelectric force microscopy to image the domain structures of twisted hexagonal boron nitride (hBN), revealing evidence of strong in-plane electric fields at the domain boundaries. By placing a monolayer MoSe<sub>2</sub> only one to two nanometers away from the twisted hBN interface, we observe energy splitting of neutral excitons and Fermi polarons by several millielectronvolts at the moiré domain boundaries. By directly correlating local structural and optical properties, we attribute such observations to excitons confined in a nanoscale one-dimensional electrostatic potential created by the strong in-plane electric fields at the moiré domain boundaries. Intriguingly, this 1D quantum confinement results in pronounced polarization anisotropy in the excitons' reflection and emission, persistent to temperatures as high as ~80 Kelvins. These findings open new avenues for exploring and controlling strongly interacting excitons for classical and quantum optoelectronics.

**11:00am 2D+AQs+EM+MI+MN+NS+QS+SS+TF-ThM-13 Microwave Imaging of Excitonic States and Fractional Chern Insulators in 2D Transition Metal Dichalcogenides, Zhurun Ji, SLAC National Accelerator Laboratory/ MIT**  
**INVITED**

Nanoscale electrodynamics offers a unique perspective on states with bulk-edge correspondence or spatially dependent excitations. I will introduce our latest advancements in optically coupled microwave impedance microscopy, a technique that enhances our capability to explore electrodynamics at the nanometer scale. I will discuss our recent studies utilizing this technology to extract spectroscopic information on exciton excitations within transition metal dichalcogenide systems. Additionally, I will share our recent findings on probing topological and correlated electronic states, specifically the fractional Chern insulator states in twisted TMD bilayers.

**11:30am 2D+AQs+EM+MI+MN+NS+QS+SS+TF-ThM-15 Control and Properties of Single Dislocations in Van Der Waals Nanowires, Peter Sutter, Eli Sutter, University of Nebraska - Lincoln**

Line defects (dislocations) not only govern the mechanical properties of crystalline solids but they can also produce distinct electronic, thermal, and topological effects. Identifying and accessing this functionality requires control over the placement and geometry of single dislocations embedded in a small host volume to maximize emerging effects. We have identified a synthetic route that enables the rational placement and tuning of dislocation in van der Waals nanowires, where the 2D/layered crystal structure limits the possible defect configurations and the nanowire architecture puts single dislocations in close proximity to the entire host volume.<sup>1</sup> While homogeneous layered nanowires carry individual screw dislocations, the synthesis of radial (core-shell) nanowire heterostructures transforms the defect into a mixed (helical) dislocation whose edge-to-screw ratio is continuously tunable via the core-shell lattice mismatch.

Such deterministic control over defects now enables the probing of functionality arising with single dislocations. For example, germanium sulfide van der Waals nanowires carrying single screw dislocations incorporate Eshelby twist and thus adopt a chiral twisted structure,<sup>2</sup> which for the first time allowed the identification of chirality effects in the photonic properties of a single nanostructure.<sup>3</sup> Using cathodoluminescence spectroscopy, whispering gallery modes could be excited and probed to directly compare the photonics of chiral and achiral segments in single nanowires. The data show systematic shifts in energy, which with the help

of simulations are assigned to chiral whispering gallery modes in wires hosting a single dislocation.

The ability to design nanomaterials containing individual dislocations with controlled geometry paves the way for identifying a broad range of functional properties of dislocations, with the potential to herald a paradigm shift from the traditional strategy of suppressing dislocations to embracing and harnessing them as core elements of new technologies.

1. P. Sutter, R.R. Unocic, and E. Sutter, *Journal of the American Chemical Society* 145, 20503 (2023); DOI: 10.1021/jacs.3c06469
2. P. Sutter, S. Wimer, and E. Sutter, *Nature* 570, 354 (2019); DOI: 10.1038/s41586-019-1147-x
3. P. Sutter, L. Khosravi-Khorashad, C.V. Ciobanu, and E. Sutter, *Materials Horizons* 10, 3830 (2023); DOI: 10.1039/D3MH00693J

**11:45am 2D+AQs+EM+MI+MN+NS+QS+SS+TF-ThM-16 Electrical Manipulation of Valley Polarized Charged Excitons in 2d Transition Metal Dichalcogenides, Kuan Eng Johnson Goh, Agency for Science Technology and Research (A\*STAR), 2 Fusionopolis Way, Innovis #08-03, Singapore 138634, Singapore**

The control of excitons in 2-dimensional (2D) Transition Metal Dichalcogenide (TMD) semiconductors is a key enabler for their use in optoelectronic, valleytronic and quantum applications. Reproducible electrical control of excitons remains elusive as excitons are intrinsically charge neutral quasiparticles. Here, we demonstrate that charge defects present in 2D TMDs like single-layer H-phase WS<sub>2</sub> [1,2], could be advantageous for electrical control through the coherent coupling of the exciton or biexciton with intrinsic charges in the single-layer WS<sub>2</sub>, thus enabling a simple and robust method for electrical manipulation of the degree of valley polarization from <10% to >60% [3]. Such robust electrical tunability of the spectral resonance of the charged states indicates resonant control of valley polarization by exploiting the intricate interplay between the charged and neutral exciton/biexciton states, representing a key advance towards using the valley degree of freedom as an alternate information carrier.[4].

## References

- [1] Bussolotti, F., et al., ACS Nano 15 (2021) 2686
- [2] Bussolotti, F., et al., ACS Nano 18 (2024) 8706
- [3] Das, S., et al., ACS Nano 18 (2024) 30805
- [4] Goh, K. E. J., et al., Advanced Quantum Technologies 3 (2020) 1900123

**12:00pm 2D+AQs+EM+MI+MN+NS+QS+SS+TF-ThM-17 Thickness Dependent Band Gap and Electrical Anisotropy of 2DSnSe, Marshall Frye, Jonathan Chin, Joshua Wahl, Jeremy Knight, Georgia Institute of Technology; Walter Smith, Purdue University; Dilara Sen, Samuel Kovach, Kenyon University; Frank Peiris, Kenyon College; Charles Paillard, University of Arkansas; Thomas Beechem, Purdue University; Anna Osterholm, Lauren Garten, Georgia Institute of Technology**

2D SnSe presents unique opportunities for optoelectronics, and scalable microelectronics, but it is first critical to understand how the electrical and optical response change upon downscaling. Tailoring the band gap and electrical anisotropy of 2D monochalcogenides, like SnSe, has previously been shown but the mechanisms that drive the changes in band gap are still not understood. This study reveals how changes in bond length and structure drive the thickness dependences of band gap, carrier mobility and lifetime of SnSe thin films. Molecular beam epitaxy is used to deposit (2h00) oriented SnSe thin films with thicknesses ranging from 4 nm to 80 nm. The direct band gap increases from 1.4 eV at 80 nm to 1.9 eV at 4 nm, underscoring the potential of SnSe as a tunable and direct band gap material for thin film optoelectronics. Raman spectroscopy shows different simultaneously changes in the crystal structure and bonding occurring parallel versus perpendicular to the 2D plane with decreasing film thickness. TEM further supports the hypothesis that the increase in the band gap with reduced thickness is due to changes in crystal structure resulting in a contraction of the out-of-plane SnSe covalent bonds, while the in-plane bond length increases. In addition to the reduction in band gap, tracking the time dependent photoluminescence shows an increase in carrier lifetime with decreasing film thickness, while Hall measurements show a change in the carrier mobility with decreasing thickness. Overall, this work provides the critical missing insight needed to design these optically and electronically relevant 2D materials for scalability.

**Bold page numbers indicate presenter**

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Alanwoko, Onyedikachi:  
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Allerbeck, Jonas:  
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