## PCSI

## Room Ballroom South - Session PCSI-WeM1

## 2D Materials

**Moderator: Ursula Wurstbauer**, University of Münster

8:30am **PCSI-WeM1-1 Flat Bands and Excitons in Transition Metal Dichalcogenide Moiré Patterns***, **Mit Naik**,* University of Texas at Austin **INVITED**

Moiré patterns of 2D van der Waals materials have proven to be an ideal platform to host unusual correlated electronic phases, emerging magnetism, and exciton physics. At small twist-angles, novel moiré exciton states in transition metal dichalcogenide (TMD) heterostructures have been recently discovered through the observation of multiple emergent peaks in the optical spectra, but their atomistic nature has been a mystery. Using first-principles GW-Bethe Salpeter equation calculations we discover a rich diversity of excitonic states in large-area TMD moiré superlattices, particularly a novel exciton with an intralayer charge-transfer character. We uncover a complex interplay between structural reconstruction, the formation of flat bands, and the nature of excitonic states. These studies, which involve thousands of atoms in the reconstructed moiré unit-cell, are made feasible by the development of a new computational approach

While small twist-angles have been widely studied, large twist angle superlattices are often considered electronically layer-decoupled due to misaligned Brillouin zones of the individual layers. Surprisingly, we observe the emergence of flat electronic bands with a distinctive anisotropic dispersion at a large magic twist angle in TMD bilayers. A direct consequence of this flat band is the emergence of phonon-assisted intervalley absorption peaks in reflection contrast spectra measurements. The flat band shows a power-law divergent density of states due to its quasi-one-dimensional character, enhancing the potential for correlated phases.

9:10am **PCSI-WeM1-9 Impact of External Screening on the Valence and Core-Level Photoelectron Spectra of One-Layer $WS_2$***, Alex Boehm, Chris Smyth, Andrew Kim, Don Bethke, Tzu-Ming Lu,* Sandia National Laboratories*; Jose Fonseca Vega,* Naval research Laboratory*; Jeremy Robinson,* naval research Laboratory*; **Taisuke Ohta**,* Sandia National Laboratories

In an effectively-screened environment, transition metal dichalcogenides (TMDs) rearrange their charge carriers to screen the added charges, and reduce the electronic band gap. Consequently, when interfaced with dissimilar materials, a sheet of TMD would change its band gap adapted to its local external screening environment. Similarly, a well-screened environment stabilizes photo-holes or core-holes created in the photoemission process and, in turn, boosts the kinetic energy of photoelectrons resulting in the apparent smaller binding energy. Complication arises when determining the electronic band alignment of TMDs using photoelectron spectroscopy since the screening influences the material property of interest as well as its assessment approach concurrently. Using a sample that contains areas of suspended and gold-supported one-layer $WS_2$, we show how the electronic states of $WS_2$ under the contrasting effective or ineffective external screening environment align at the built-in junction. The photoelectron spectra point to the breakdown of rigid shifts between the valence states and core-levels with the core-levels shifting more than twice as much as the valence states. Additionally, effectively-screened $WS_2$ displays a valence state with a substantially larger photoemission linewidth than ineffectively-screened suspended $WS_2$. Altogether, our result provides key insights into how the local variation of the external screening environment creates essentially a heterojunction within a layer of $WS_2$, and whether commonly accepted photoelectron spectroscopy practices hold when examining the electronic structures of one-layer TMDs.

The work was supported by the Laboratory Directed Research and Development program at Sandia National Laboratories and Base Programs and the Nanoscience Institute at the Naval Research Laboratory via the Office of Naval Research. A.R.K. acknowledges support from the U.S. Department of Energy, Office of Science, Division of Materials Sciences and Engineering (grant BES 20-017574). Samples were fabricated, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the US Department of Energy, Office of Science. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly-owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

9:15am **PCSI-WeM1-10 Disorder-Induced 2D to 3D Dielectric Screening Transition in Single-Layer WS2***, **Christopher Smyth**, Alex Boehm,* Sandia National Laboratories*; Kory Burns,* University of Virginia*; Catalin Spataru, Andrew Kim, Don Bethke, Tzu-Ming Lu, Taisuke Ohta,* Sandia National Laboratories

The quasiparticle band gap ($E_{g,QP}$.) and dielectric permittivity represent fundamental properties of bulk semiconductors as a result of the three-dimensional (3D) character of the dielectric screening. In two-dimensional (2D) materials, the lattice is confined well below the typical dielectric screening length, and the non-local characteristic of the dielectric function emerges, resulting in unusually large exciton radius and binding energy in the weak screening environment. Consequently, the $E_{g,QP}$ and exciton binding energy become highly responsive to perturbations in the dielectric screening with extrinsic origins. Ubiquitous monovacancies in 2D semiconductors host a polarizable dipole, which can impact the screening strength and $E_{g,QP}$ through the internal defect density ($n_v$). Similarly, substrate-induced screening can impact the $E_{g,QP}$ and exciton behavior, which often obscures the combined effects of multiple screening mechanisms.

We present an experimental investigation of the isolated and combined impacts of structural disorder and external dielectric screening on the $E_{g,QP}$ and exciton behavior in single-layer $WS_2$. Introducing dilute structural disorders, primarily sulfur monovacancies, to weakly screened $WS_2$ results in a significant 190 meV renormalization of the $E_{g,QP}$, while the optical bandgap remains effectively unchanged at around 2.0 eV, as confirmed by photoemission spectroscopy (PES) and electron energy loss spectroscopy (EELS). When the $n_v$ reaches a threshold of $5 \times 10^{12}$ $cm^{-2}$, the interactions between isolated excitons and defects in poorly screened $WS_2$ transition to correlated interactions. The exciton-defect interaction changes because the screening radius and interdefect distance converge at ~4 nm. The emergence of correlated exciton-defect interactions is attributed to the dielectric function transitioning from a 2D to 3D character. In contrast, the $E_{g,QP}$ and exciton binding energy of $WS_2$ remain unaffected by vacancies when the screening environment is dominated by a strongly screening Au substrate. Therefore, the sensitivity of the electronic band structure and exciton stability in 2D materials to the screening environment hinges on their polarizability.

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9:20am **PCSI-WeM1-11 Tuning Phonon and Exciton Dynamics Through Alloying in 2D Transition Metal Dichalcogenides***, **Tinsae Alem**, Hayden Barry, Stephen McDonnell,* University of Virginia*; Jordan Hachtel,* Oak Ridge National Laboratory*; Chris Smyth,* Sandia National Laboratories*; Kory Burns,* University of Virginia

Quasiparticles play a key role in thermal transport and electronic behavior in 2D materials, where dipole interactions and lattice dynamics can be tuned via compositional alloying. However, controlling momentum transitions of phonons, coupling strength of excitons, and their locality based on intrinsic stresses in the lattice remains a challenge. This is attributed to a limitation in bulk characterization techniques; they give global information that averages thousands of unit cells, making it impossible to correlate which bonds result in individual displacement modes or optical transitions. Hereby, we investigate how alloying in $W_xMo_{1-x}S_2$ modulates local dipole moments and phonon/exciton populations, with implications for thermoelectric performance. By varying stoichiometry across five compositions, we examine how vibrational properties evolve with atomic-scale disorder and dipole perturbation. Bulk vibrational modes were measured using Raman and Infrared (IR) spectroscopy, with Raman providing superior energy resolution and enabling the collection of low-energy optical phonons. The optical spectrum was interpreted with photoluminescence (PL) to measure band transitions and splitting in the valence band. To access vibrational and optical behavior beyond the diffraction limit set by the wavelength of photons, we used monochromated electron energy loss spectroscopy (EELS) inside an aberration-corrected scanning transmission electron microscope (STEM),

enabling atomic-scale mapping of phonon and exciton populations. X-ray photoelectron spectroscopy (XPS) and energy-dispersive X-ray spectroscopy (EDX) confirmed chemical uniformity over micron and nanometer length scales. This combined approach reveals how alloying influences spin-orbit coupling and lattice vibrations, offering insight into thermal transport and tunability of exciton-phonon coupling in low-dimensional semiconducting platforms.

9:25am **PCSI-WeM1-12 UPGRADED: Quantum Emitters from Electrostatically Strained WSe2 Monolayers Suspended Over Nanocavities**, ***Frances Camille Wu***, *Jadon Zheng, Shang-Hsuan Wu, Bin Fang, Edward Yu,* The University of Texas at Austin

Tungsten diselenide ($WSe_2$) monolayers are promising hosts for quantum emitters due to their intrinsic dark exciton state, which hybridizes with defect states under tensile strain, resulting in localized defect emission. To date, strain engineering has focused primarily on creating static strain via techniques such as draping $WSe_2$ monolayers on patterned substrates or indentation with an atomic force microscope, which suffers from poor control over strain. In this study, we employ an electrostatic straining approach demonstrated for monolayer $WSe_2$ suspended over micron-scale cavities in a patterned substrate. In this approach, a back-gate voltage applied to the silicon substrate below the cavity induces a downward deflection of the suspended $WSe_2$ monolayer due to the capacitive interaction between the monolayer and substrate.[1]

Here, we investigate scaling of the cavity to nanoscale dimensions, allowing the distance between emitters to be smaller than the wavelength of light and enabling the study of cooperative emission behaviors from coupled emitters such as those leading to superradiant emission. This collective emission from coupled emitters has been previously observed in laterally arranged quantum dot ensembles, but not in highly scalable $WSe_2$-based quantum emitters. By performing electrostatic straining on a $WSe_2$ monolayer suspended over 100-nm diameter cavities, monolayer deflections of 2 nm ($V_g$ = 10 V) and 3.5 nm ($V_g$ = 27.5 V) were observed, corresponding to 0.2 and 0.4% increase in tensile strain, respectively. Initial studies of photoluminescence from a $WSe_2$ monolayer suspended over 10 cavities showed a linear (rather than saturating) behavior of localized emitter intensity as a function of increasing excitation power, a distinct characteristic of cooperative emission. Second-order photon correlation measurements showed an emitter antibunching behavior of $g^{(2)}(0)$ = 0.24±0.03, indicative of high-purity emitters, which can be attributed to the effective exciton funneling on nanoscale cavities. Current studies are focused on the electrostatic biasing of the suspended $WSe_2$ monolayer, which could enable precise strain modulation and enhanced coupling of emitters for possible observation of additional cooperative emission phenomena. This can be beneficial for the development of highly scalable and tunable coupled quantum emitters from strained $WSe_2$ monolayers for potential applications in quantum information processing, quantum laser technology, and quantum computing.

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