PCSI
Room Ballroom South - Session PCSI-MoM2
2D Materials and Graphene I

Moderator: Kunal Mukherjee, Stanford University

11:00am **PCSI-MoM2-31 Interplay of Valley Polarized Dark Trion and Dark Exciton-Polaron in Monolayer WSe_2**, ***Xiao-Xiao Zhang***, University of Florida **INVITED**

The interactions between charges and excitons involve complex many-body interactions at high densities. The exciton-polaron model has been adopted to understand the Fermi sea screening of charged excitons in monolayer transition metal dichalcogenides (TMD). The results provide good agreement with absorption measurements, which are dominated by dilute bright exciton responses. The Fermi-polaron model treats the quasiparticle responses of a single mobile impurity in a surrounding Fermi sea. In comparison, the exciton density in monolayer TMD can be tuned by laser fluence and be comparable to or exceed the charge density, where the analogy to a single mobile impurity no longer applies. The modification to Fermi sea screening at high exciton densities, however, is still not well understood. Apart from the bright excitons previously studied in reflection contrast measurements, different spin and momentum dark exciton species have been established, which are also expected to have many-body interactions with charges. The coupling between these different species of exciton-polarons has not yet been experimentally investigated.

Here we investigate the Fermi sea dressing of spin-forbidden dark excitons in monolayer WSe_2 . With a Zeeman field, the valley-polarized dark excitons show distinct p-doping dependence in photoluminescence when the carriers reach a critical density (see Fig. 1). This density can be interpreted as the onset of strongly modified Fermi sea interactions and shifts with increasing exciton density. Through valley-selective excitation and dynamics measurements, we also infer an intervalley coupling between the dark trions and exciton-polarons mediated by the many-body interactions. Our results reveal the evolution of Fermi sea screening with increasing exciton density and the impacts of polaron-polaron interactions, which lay the foundation for understanding electronic correlations and many-body interactions in 2D systems.

11:40am **PCSI-MoM2-39 Evidence of Single Photon Emitters from 1L WSe_2 under Electrostatically Induced Strain**, ***Frances Camille Wu***, *S. Wu, B. Fang, X. Li, J. Incorvia, E. Yu,* The University of Texas at Austin

Strain engineering is a powerful tool that strongly influences electronic structure and exciton states in 2D transition metal dichalcogenides (2D TMDs). Among 2D TMDs, monolayer WSe_2 has gained attention as a host for quantum emitters due to its lowest lying dark exciton state that hybridizes with mid-gap defect states under tensile strain, giving rise to bright single photon emitters. The creation of a hybridized state consisting of dark excitons and mid-gap defect states results in the radiative recombination of dark excitons that is otherwise forbidden due to spin and momentum conservation. At cryogenic temperatures, the energetic alignment and coupling of the two abovementioned states results in localized defect emission which possess significant characteristics of single photon emitters. Strain-tunable devices are crucial for investigating the nature of TMD-based single photon emitters which can be beneficial for quantum information processing and secure communications.

In this study, we demonstrate strain modulation of monolayer WSe_2 suspended over a hole-patterned substrate via electrostatic deflection and characterize the resulting photoluminescence. This approach enables the creation of strain-tunable WSe_2 devices that can be operated at cryogenic temperatures, wherein strain fields are generated by applying a bias voltage to the suspended monolayer WSe_2 membrane. We observe a significant monolayer deflection of ~50 nm at 15V gate bias and a ~20 meV redshift of dark exciton peak as the applied is increased to 20V, corresponding to a 0.2% increase in tensile strain of WSe_2. Sharp localized emitters, typically associated with single photon emitters, were observed at 4K which showed less dependence on strain as the applied bias increased. Thus, we attribute these localized emitters to the presence of localized defects which are only weakly influenced by strained lattice environment. We also observe luminescence lifetimes of ~3 ns and saturation of luminescence intensity with incident power, both of which are also characteristic of single photon emitters. These realizations are critical for understanding the origin of single photon emitters based on strained monolayer WSe_2.

11:45am **PCSI-MoM2-40 Comprehensive Study of Interface Chemistry and Electrical Property of Metal Contacts on TMDs**, *S. Kim,* ***Joy Roy***, *X. Wang, R. Wallace,* University of Texas at Dallas

Transition metal dichalcogenides (TMDs) have been introduced due to their exceptional electronic, optical mechanical, and magnetic properties, even in atomically thin thickness, for advanced electronic, optoelectronic, and spintronic devices [1]. However, the limitations in tuning the Schottky barrier height with metal contacts, based on work function greatly hinder efficient carrier injection and electronic performance of TMD-based devices [2]. This study examines contact interfaces and their relationship to electrical contact characteristics. The research encompasses interface chemistry, band alignment, and electronic contact properties of Ni, Ag, Bi, Co and Sn.

An ultrahigh vacuum (UHV) cluster system was employed to investigate the contact properties where in-situ X-ray Photoelectron Spectroscopy (XPS) showed the contact bonding features of metal/TMD interfaces. Ni and Co contacts exhibited stronger bonds with TMD surfaces, resembling covalent-like interfaces, with notable interface reaction products resulting from annealing. However, Bi and Sn showed no robust chemical bonding features under the XPS analysis and van der Waals contact interface was formed due to a weak interaction between metal and TMD. The subsequent ex-situ atomic force microscopy (AFM) measurement supported these contact interface properties. The subsequent electrical characterization using XPS and scanning tunneling microscopy (STM) suggests that the roles of surface defect impact the metal contacts as well. In conclusion, comprehensive research and investigation of metal materials and their contact interface properties with TMDs have shed light on the potential and advantages of metal contact studies.

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[1]Pi, L., Li, L., Liu, K., Zhang, Q., Li, H., and Zhai, T, Adv. Funct. Mater., 29(51), 1904932 (2019).

[2]Guo, Y., Liu, D., and Robertson, J, ACS Appl. Mater. Interface. 7, 25709-25715 (2015)

11:50am **PCSI-MoM2-41 Transport Anisotropy in One-dimensional Graphene Superlattice in the High Kronig-Penney Potential Limit**, ***Tianlin Li***, *H. Chen, K. Wang, Y. Hao, L. Zhang,* University of Nebraska - Lincoln*; K. Watanabe, T. Taniguchi,* National Institute for Materials Science, Japan*; X. Hong,* University of Nebraska - Lincoln

One-dimensional (1D) graphene superlattice (GSL) has drawn considerable research interest as it is promising for realizing the electron lensing effect [1]. Despite the intensive theoretical studies, 1D GSL has only been realized experimentally via periodic dielectric gates in previous studies [2, 3], which yields a moderate Kronig-Penney (KP) potential profile that is not viable to achieve electron supercollimation.

In this work, we demonstrate 1D GSL in the high KP potential limit exploiting nanoscale domains patterning in a ferroelectric bottom gate [4]. We work with 50 nm (001) $PbZr_{0.2}Ti_{0.8}O_3$ (PZT) films deposited on 10 nm $La_{0.67}Sr_{0.33}MnO_3$ buffered $SrTiO_3$ substrates. Monolayer graphene field-effect transistors with top h-BN global gates are fabricated on PZT prepatterned with periodic polarization up (P_{up}) and down (P_{down}) stripe domains, with the period L varying from 200 to 300 nm (Fig. 1a). The polarization shifts the Fermi level of graphene, leading to a KP potential V_0 of about 0.9 eV at 2 K. We fabricate 1D GSL samples in two configurations, with current along the SL vector $\hat{s}$ and perpendicular to $\hat{s}$. For the former samples, additional Dirac points (DP) emerge in the sheet resistance (R_{xx}) *vs.* top-gated induced electron doping δn (Fig. 1b), from which emanates multiple Landau fan branches in the magnetic field (Fig. 1c). This feature is absent in the latter configuration (R_{yy}), which can be attributed to the SL modulated band crossings along $\hat{s}$. The carrier density between consecutive DP positions (Δn_{DP}) scales with the SL period as $\Delta n_{DP} \propto L^{\beta}$, with β = -1.18±0.06 (Fig.

1d), which closely resembles the inversely proportional relation predicted for the high KP potential limit. Figure 1e shows the simulated 1D GSL band structure for our ferroelectric doping scheme, with dimensionless KP potential $u = V_0L/\hbar v_F = 90\pi$, which reveals a highly flattened band that can potentially host electron lensing effect.

[1] C. H. Park, et al., Nano Letters **8**, 2920 (2008)

[2] S. Dubey, et al., Nano Letters **13**, 3990 (2013).

[3] Y. Li, et al., Nature Nanotechnology **16**, 525 (2021).

[4] T. Li, et al, arXiv: 2309.04931 (2023)

11:55am **PCSI-MoM2-42 Terahertz Emission Spectroscopy Revealing Nanoscale Vectorial Photocurrents in Symmetry-Broken Optoelectronic Metasurfaces***, J. Pettine, P. Padmanabhan,* Los Alamos National Laboratory*; L. Gingras, R. Holzwarth,* Menlo Systems, Germany*; R. Prasankumar, A. Taylor, S. Lin,* ***Hou-Tong Chen****,* Los Alamos National Laboratory

Terahertz (THz) emission spectroscopy has emerged in the past several decades as a versatile method for directly tracking the ultrafast evolution of physical properties, quasiparticle distributions, and order parameters within bulk materials and nanoscale interfaces. Ultrafast optically-induced THz radiation is widely utilized to analyze nonlinear polarization, magnetization, and various transient free charge currents, where the underlying broken symmetries (surface and bulk) enable THz emission by defining a system directionality in space and/or time [1]. The broken spatial or temporal symmetries responsible for second-order nonlinearities and bias-free photocurrent generation in materials are typically either intrinsic to the lattice and thus constrained to specific light–matter interaction geometries, or otherwise dependent upon applied static fields that are difficult to texture on small length scales.

In this talk we show that asymmetric gold nanoantennas on graphene exhibit strong light-driven directional responses [2]. The local photocurrent directionality is determined by the orientation of individual nanoantennas, which can be patterned into an arbitrary direction profile for global spatially-varying and optically-controlled photocurrents. We use THz emission spectroscopy, complementary to the direct photocurrent readout, to validate such a concept. Our experimental results clearly demonstrate that these vectorial optoelectronic metasurfaces serve as efficient and versatile sources of ultrafast THz radiation, including broadband THz vector beams. Electrostatic gating and multiphysics modeling reveal a local photothermoelectric driving mechanism and elucidate previously unexplored dynamics occurring at the intersection of femtosecond excitation and nanoscale localization.

[1] J. Pettine *et al.*, arXiv.2307.11928 (2023).

[2] J. Pettine *et al.*, *Light Sci. Appl.* **12**, 133 (2023).

12:00pm **PCSI-MoM2-43 Excitons, Electrons, and Holes in Monolayer Semiconductors: Insights from Spectroscopy in (Really) High Magnetic Fields***, **Scott Crooker,*** National High Magnetic Field Lab

Historically, magnetic fields have played an essential role in revealing the properties of semiconductors, and the many-body physics that can emerge when they are doped with mobile carriers. However, for atomically-thin 'transition metal dichalcogenide' (TMD) semiconductors such as MoS_2 and WSe_2, the relevant field scale is substantial (of order 100 tesla!) due to heavy carrier masses, huge exciton binding energies, and typically large Fermi energies.Fortunately, modern pulsed magnets can achieve this scale. This talk will discuss a few recent optical studies that probe the physics of -- and many-body correlations between -- excitons, electrons, and holes in TMD monolayers. These experiments used dual-gated TMD monolayers, assembled via van der Waals stacking directly atop single-mode optical fibers to enable polarized absorption spectroscopy at low temperatures in 60-100T fields.

*In charge-neutral monolayers, spectroscopy up to ~90T reveals the diamagnetic shifts of the neutral exciton's *1s* ground state *and* its excited *2s, 3s,… ns* Rydberg states, revealing exciton masses, radii, binding energies, dielectric properties, and free-particle bandgaps – essential ingredients for the rational design of optoelectronic van der Waals structures. [1]

*In hole-doped monolayer WSe_2, high-field spectroscopy of both neutral and charged exciton transitions revealed the (often-hypothesized) spontaneous valley polarization of mobile holes, due to exchange interactions, occurring at ~40T. [2]

*In electron-doped WSe_2 monolayers, the ordering of the conduction bands in the *K* and *K'* valleys allows studies of not only neutral excitons (*X0*) and charged excitons (*X*- trions) at low carrier density, but also many-body states that can emerge at higher doping. We investigate the so-called *X-'* resonance that emerges at high electron density, known since 2013 but never understood. The data suggest that *X-'* is, in fact, a six-particle "hexciton" state that arises when the photoexcited electron-hole pair couples simultaneously to two Fermi seas having quantum-mechanically distinguishable spin/valley quantum numbers. This state also appears in WS_2 and may appear in MoS_2, and appears in $MoSe_2$ at the B-exciton resonance [3,4]

[1] *Nat. Comm.***10**, 4172 (2019); [2] *PRL***125**, 147602 (2020);[3] *Nano Letters***22**, 426 (2022);[4] *PRL***129**, 076801 (2022).

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