## PCSI
## Room Keahou I - Session PCSI-WeA4
## 2D Materials and Graphene II

**Moderator: Scott Crooker**, Los Alamos National Laboratory

4:55pm **PCSI-WeA4-42 UPGRADED: Topotaxy in 2D Materials: Towards Synthesis of Novel 2D Materials by Surface Reactions*, Matthias Batzill,*** University of South Florida

Topotaxy is a surface reaction of deposited elements with a substrate, during which the substrate retains some structural characteristics. Such newly formed materials thus have a crystallographic relationship with the original substrate. For 2D materials, surface reactions with single molecular layers may enable their transformation into new 2D crystals. Here the potential for making new 2D materials by topotactical reactions of transition metal dichalcogenides (TMDs) with transition metals are investigated. Three distinct examples are discussed: (i) the transformation of $PtTe_2$ into $Pt_2Te_2$ by reaction with Pt atoms; (ii) the reaction of Cr or Mn with bilayer $VSe_2$ to form $VSe_2$/Mn(Cr)/$VSe_2$, and (iii) reaction of $MoTe_2$ with Mo to create mirror twin grain boundaries that may self-organize in periodic lattice networks (Figure (c)). The common concept in these surface reactions is that the reacted metals occupy ad- or ab-sorption sites which maintain a low energy van der Waals termination and thus enables the creation of new (meta) stable 2D materials. The three examples discussed here, illustrate the diversity of possible reaction products and the potential for synthesizing novel 2D materials by topotaxy.

5:15pm **PCSI-WeA4-46 Thickness Calculation of HBN and Graphene Using RGB Colors*, Gabriel Ruiz,*** New Mexico State University*; Ben Xie,* University of California Santa Barbara

The main objective of our research is primarily due to the required scientific exploration in the properties of a two-dimensional material called graphene. In order to achieve our goals, we want to induce a flat band graphene in order to maintain a nice platform that allows study of correlating physics. When graphene is combined with other materials in van der Waals heterostructures, we can electronically tune its band flatness. When achieved, electron kinetic energy decreases. This allows us to observe and study various correlation phenomena. Using Van der Waals heterostructures as a methodology for the measurement and alteration of graphene requires atomically homogeneous material to build it. The homogeneity of these materials plays an important role when using them to build our heterostructures.

We normally obtain them through mechanical exfoliation then search for them under a microscope. However, it is complicated to characterize the exact thickness of these materials optically. With this problem we looked for a solution by creating a program code. We seek to indicate through saturation comparisons between the different layers of hexagonal boron nitride (HBN) which serves as a dielectric material required for the composition of the heterostructure and Graphene. The code has been polished and altered to generate more efficiency towards the search for homogenous 2D materials. This project will significantly improve the efficiency for us to search for better flakes. Eventually leading to a higher device quality and potential observation of novel physics phenomena.

5:20pm **PCSI-WeA4-47 Optoelectronic Properties of MoS2/Graphene Heterostructures Prepared by Dry Transfer Method for Light-induced Energy Harvesting Applications*, Sanju Gupta,*** Penn State University and Gdansk University of technology

Optoelectronic properties of atomic thin van der Waals heterostructures (vdWHs) comprising transition metal dichalcogenides that harvest light energy are of paramount interest. In this work, the effects of underlying single and bi-layer graphene (Gr) layers on structural and physical properties of $MoS_2$/Gr vertical heterostructures *i.e.,* (1-2L) $MoS_2$/(1-2L) Gr, besides additional interfaces including $MoS_2$ folds/edges [$MoS_2$(1L+1L))/Gr(1L)] and $MoS_2$(1-2L)/Au, are investigated to unravel the excitonic properties. By employing correlative scanning probe microscopy combined with micro-spectroscopy, we observed multiple effects related to excitons (*i.e.,* redshifted neutral exciton, ratio of charged exciton or trion to neutral exciton population, and long-tailed trions) and surface electronic properties (*i.e.,* reduced work function suggesting electron transfer) in addition to significantly enhanced near-field Raman spectra, apparent n-p type current rectification behavior and increase in photo-generated carriers. These experimental findings are attributed to interlayer electronic interactions while minimizing Fermi level pinning at $MoS_2$/Au interface, commonly observed in 2D semiconductor-3D metal junction, and corroborated with theoretical DFT calculations, which deepened our understanding of dissimilar 2D materials junctions. Integrating $MoS_2$ with optimal number of graphene layers as 'nanospacer' signified substrate engineering that are versatile for key optoelectronic and photovoltaic applications [1, 2, 3, 4].

[1] Gupta S., Johnston A., Khondaker, A., Optoelectronic properties of $MoS_2$/graphene heterostructures prepared by dry transfer method for light-induced energy applications, *J. Electron. Mater.* 51 (2022) 4257.

[2] Gupta, S., Johnston, A., Khondaker, S., Correlated KPFM and TERS imaging to elucidate defect-induced inhomogeneities in oxygen plasma treated 2D $MoS_2$ nanosheets, *J. Appl. Phys.* 131 (2022) 164303.

[3] Gupta S., Dimakis N., First-Principles Calculations Integrated with Experimental Optical and Electronic Properties for $MoS_2$/Au and $MoS_2$/Graphene/Au Heterostructures, Appl. Surf. Sci. 623 (2023) 156948.

[2] Gupta *et al.,* Appl. Surf. Sci. **623**, 156948 (2023).

5:25pm **PCSI-WeA4-48 The Case of the Missing Sulfur*, Mirette Fawzy,*** Dept. of Physics, Simon Fraser University, Canada*; Mohammad Reza Mohammadzadehb, Amin Abnavi, Thushani de Silva, Ribwar Ahmadi, Hamireza Ghanbari, Fahmid Kabir, Amirhossein Hasani, Michael M. Adachi,* School of Engineering Science, Simon Fraser University, Canada*; **Karen Kavanagh**,* Dept. of Physics, Simon Fraser University, Canada

M. Fawzy[a], M. Reza Mohammadzadeh[b], A. Abnavi[b], T. de Silva[b], R. Ahmadi,[b], H. Ghanbari,[b], F. Kabir[b], A. Hasani[b], M. M. Adachi[b] and K. L. Kavanagh[a]

[a]*Dept. of Physics, Simon Fraser University, Burnaby, BC V5A 1S6*

[b]*School of Engineering Science, Simon Fraser University, Burnaby, BC V5A 1S6*

A common strategy for obtaining an ohmic contact to any semiconductor is to form a tunnel junction using a heavily doped surface layer. Independent of the magnitude of the interfacial barrier, the narrow width of the depletion layer allows for efficient tunneling and a linear current-voltage transport. This has worked well for classical bulk devices where transport is through a thick crystal. The low resistance ohmic contact can be a large area, placed for example at the bottom of a wafer. For two-dimensional semiconductors, such as $MoS_2$, the transport of interest is typically parallel to the surface meaning lateral contacts are required. However, if the semiconductor is an exfoliated triangular flake, parallel contacts become naturally asymmetric in area once the metal is evaporated and patterned on top. It was also soon noticed by a few groups that different area contacts resulted in rectification ($10^5$ decades), even when the same metal was used, with the *smaller* area contact having the lower resistance ohmic transport. In particular, the application of different area reactive Cr/Au contacts on (20-60) nm thick exfoliated $MoS_2$ flakes has been an effective method to fabricate a two terminal diode that has been applied towards optoelectronics and biosensing applications [1-2].

The latest example from our collaboration is the sensing of volatile gases using a UV optically powered asymmetric $MoS_2$ diode [3]. Shown in the figure is a schematic diagram of the device (55 nm thick $MoS_2$) with a set of I-V characteristics as a function of UV power. This presentation will discuss the likely mechanisms as a function of metal thickness, contact edge lengths, and $MoS_2$ source and thickness. Topics such as Fermi level pinning, sulfur reaction and diffusion, and buried depletion regions might be discussed.

Corresponding author: kavanagh@sfu.ca

[1] Flexible High-Performance Photovoltaic Devices based on 2D $MoS_2$ Diodes with Geometrically Asymmetric Contact Areas, Amin Abnavi, Michael M. Adachi, et al. *Adv. Funct. Mater*. **33** (2022) 2210619.

[2] Ultrasensitive rapid cytokine sensors based on asymmetric geometry two-dimensional $MoS_2$ diodes, Thushani de Silva, Mirette Fawzy, et al., *Nature Comm.* **13**, 7593 (2022).

[3] A Photovoltaic Self-Powered Volatile Organic Compounds Sensor Based on Asymmetric Geometry 2D $MoS_2$ Diodes, Mirette Fawzy, MR Mohammadzadeh, et al. *ECS Sensors Plus,* in press (2024).

5:35pm **PCSI-WeA4-50 Investigating Modulation of Coulomb Interaction in Graphene on a High-k Dielectric**, ***Rubi Km,*** Los Alamos National Laboratory*; Junxiong Hu,* National University of Singapore*; Maurice Bal,* Radboud University Nijmegen, Netherlands*; Mun Chan,* Los Alamos National Laboratory*; Ariando Ariando,* National University of Singapore*; Uli Zeitler,* Radboud University Nijmegen, Netherlands*; Neil Harrison,* Los Alamos National Laboratory

Graphene on $SrTiO_3$ (STO) exhibits interesting quantum phenomena, such as quantum Hall ferromagnetism [1] and charge-density-wave order [2]. These effects are believed to stem from the large dielectric permittivity of STO [1,2,3,4], which is expected to significantly screen Coulomb interactions in graphene. However, angle-resolved photoemission spectroscopy (ARPES) measurements reveal that the Fermi velocity of carriers in graphene on STO is comparable to that of graphene on conventional substrates $SiO_2$ and hBN [5], suggesting minimal screening of Coulomb interactions.

To further investigate the electronic band properties and resolve the question of interaction screening in graphene on STO, we conducted electrical transport measurements in high magnetic fields up to 60 T, across a broad temperature range of 1.5–300 K. In this talk, we will present findings inferred from the quantum Hall effect and quantum oscillations results on graphene/STO devices (Fig 1). Our detailed analysis of the back-gate and temperature dependence of these phenomena indicates a strong effect of the STO substrate on the Fermi energy of graphene, but not on its Fermi velocity.

5:40pm **PCSI-WeA4-51 MBE Growth of Transition Metal Dichalcogenides**, ***Matthew Swann,*** *Ziling Li,* The Ohio State University*; Corbin Helton,* Columbus State Community College*; Roland Kawakami,* The Ohio State University

3D materials such as silicon have been the workhorse of the semiconductor industry for decades. However, as transistor technology approaches nanoscale, the performance of these materials is seriously impacted by short-channel effects. In contrast, 2D van der Waals materials hold several distinct advantages, including relative immunity from short-channel effects and a lack of dangling bonds. Monolayer transition metal dichalcogenides (TMDs) have been shown to exhibit modest and direct bandgaps, making them ideal semiconductors. Field effect transistors (FETs) fabricated utilizing exfoliated TMDs have already exhibited high On/Off ratio, small hysteresis and small subthreshold swing, and high mobilities. Exfoliated materials are typically of high quality but aren't scalable. While methods like chemical vapor deposition (CVD) can grow these materials to scale more economically, molecular beam epitaxy (MBE) can deposit large-area films with atomically precise thickness, as well as precisely control the composition of deposited films, making it ideal for studying the transport properties of TMDs. While the growth of TMDs on c-sapphire is common in CVD, its use in MBE growth is uncommon due to the large lattice mismatch between TMDs and c-sapphire. Growth on c-sapphire requires temperatures 900°C and higher in ultra-high vacuum in order to make oriented films [1], without which, a randomly oriented polycrystalline film is obtained. [2,3] Our films are grown with precise thickness control, are highly crystalline, and uniform. The aggressive heating that is required to obtain oriented films causes chalcogenide vacancies to accumulate in the film, which has been demonstrated with annealing temperatures as low as 600°C. The accumulation of these vacancies lead to increased scattering of charge carriers and shorter exciton lifetimes. We will discuss the optical and transport properties of the films.

[1] M. Nakano, Y. Wang, Y. Kashiwabara, H. Matsuoka, and Y. Iwasa, Layer-by-Layer Epitaxial Growth of Scalable WSe2 on Sapphire by Molecular Beam Epitaxy, Nano Lett. **17**, 5595 (2017).

[2] M. T. Dau et al., Millimeter-scale layered MoSe2 grown on sapphire and evidence for negative magnetoresistance, Applied Physics Letters **110**, 011909 (2017).

[3] A. Roy, H. C. P. Movva, B. Satpati, K. Kim, R. Dey, A. Rai, T. Pramanik, S. Guchhait, E. Tutuc, and S. K. Banerjee, Structural and Electrical Properties of MoTe2 and MoSe2 Grown by Molecular Beam Epitaxy, ACS Appl. Mater. Interfaces **8**, 7396 (2016).

5:45pm **PCSI-WeA4-52 Improvement of $HfO_2$ on TMDCs using Thermal Expansion Coefficient difference with Substrate**, ***Sukhyeon Eom,*** *Jinhong Park,* Sungkyunkwan University (SKKU), Republic of Korea

Recently, two-dimensional Transition Metal Dichalcogenide (TMDCs), such as $MoS_2$, have gained attention as next-generation semiconductor materials. However, due to the nature of these 2D materials, which lack dangling bonds that form interlayer bonds, it is challenging to form gate oxide materials like high-k materials. To address this, methods such as plasma or functional group treatment for surface modification of 2D materials and the use of interlayer materials like h-BN have been attempted. However, surface treatment methods can cause damage to the $MoS_2$ surface, leading to performance degradation, and interlayer materials like h-BN are mostly low-k, requiring very thin EOT formation, which introduces other side effects.

In order to overcome these challenges, methods for directly forming high-k materials via Atomic Layer Deposition (ALD) have been explored. The representative method is the CVD-ALD Mode approach using physical adsorption as a seed due to the low binding energy of 2D materials. While materials like HZO and $Al_2O_3$ have been successfully deposited using this method, bulkier materials like $HfO_2$ tend to form islands and pinholes, resulting in non-uniform growth.

To achieve uniform $HfO_2$ on $MoS_2$, PMMA is used as substrate material which induce strain by thermal expansion coefficient differences. The thermal expansion coefficients of $MoS_2$ and $SiO_2$ are generally known to be 7.0 x 10^-6 /K and 0.5 x 10^-6 /K, respectively. The difference in the thermal expansion coefficients between these two materials is 6.5 x 10^-6 /K. In contrast, the thermal expansion coefficient of PMMA is around 7.5 x 10^-5 /K, indicating a difference of 6.8 x 10^-5 /K with $MoS_2$, which is more than 10 times higher. Therefore, it is expected that the strain induced by the difference in the thermal expansion coefficient with PMMA will be higher compared to that with a Si substrate. As we expected, uniform $HfO_2$ is formed on $MoS_2$ (Fig 1). This method is expected to be utilized in next-generation semiconductor devices structure as it does not damage the channel.

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