

PCSI

Room Keauhou II - Session PCSI-MoE

2D Surfaces II/2D Magnetism

Moderators: Jieun Lee, Ajou University, Gunter Luepke, College of William & Mary

7:30pm **PCSI-MoE-1 Towards Strongly Coupled van der Waals Heterostructures Using Layer-by-layer Transfer**, *Emanuel Tutuc, K Kim, G Burg, B Fallahazad, S Larentis, H Movva*, The University of Texas **INVITED**

Layered crystals such as graphite, hexagonal boron nitride (hBN), or transition metal dichalcogenides (TMDs) can be mechanically exfoliated down to a monolayer, thereby providing a large set of two-dimensional (2D) materials with metallic, semiconducting or insulating properties. Combining such 2D materials into layered, van der Waals (vdW) heterostructures using a layer-by-layer transfer approach opens the door to realizing heterostructures with novel functionalities, which may otherwise not be possible using thin film growth techniques.

We review here key techniques and vdW heterostructures realized using layer-by-layer transfer. We demonstrate the realization of vdW heterostructures with high accuracy rotational alignment between different layers [1], which enables the realization of moiré crystals (Fig. 1) in twisted bilayer graphene [2], and gate-tunable resonant tunneling in heterostructures consisting of graphene double layers separated by hBN [1, 3].

A key parameter controlling the functionality of vdW heterostructures is the interlayer coupling in the stacking direction. Measuring such interlayer coupling, and demonstrating heterostructures with strong coupling are key issues relevant to device applications. We discuss recent results in double bilayer graphene heterostructures separated by WSe₂ (Fig. 2) which show gate-tunable resonant tunneling with large current densities [4], comparable or better than values measured in epitaxial heterostructures.

[1] K. Kim, et al., *Nano Lett.* 16, 1989 (2016)

[2] K. Kim, et al., *Proc. Natl. Acad. Sci. USA* 114, 3364 (2017)

[3] B. Fallahazad, et al., *Nano Lett.* 15, 428 (2015)

[4] G. W. Burg, et al., *Nano Lett.* 17, 3919 (2017)

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8:00pm **PCSI-MoE-7 Electronic and Optical Properties of Defects in Transition Metal Dichalcogenide Monolayers**, *B Schuler, S Barja, S Wickenberg, N Borys, E Barnard, A Weber-Bargioni, D. Frank Ogletree*, Molecular Foundry, Lawrence Berkeley Lab

Properties of two-dimensional (2D) transition metal dichalcogenides (TMDs) are highly sensitive to the presence of defects, and a detailed understanding of their structure may lead to tailoring of material properties through 'defect engineering'. Defects in 2D semiconductors are expected to substantially modify material properties. 2D TMD semiconductors are particularly interesting because they exhibit direct bandgaps in the visible range, high charge-carrier mobility, extraordinarily enhanced light-matter interactions, and potential applications in novel optoelectronic devices.

We have investigated the structural and electronic properties of point and 1D defects in MoSe₂ and WSe₂ TMD monolayers through atomically resolved low temperature STM/AFM imaging and spectroscopy [1]. We have also performed scanning near-field photoluminescence hyperspectral imaging of MoS₂ monolayers [2] to image optoelectronic properties at the 40 nm length scale.

The structure and electro-optic properties of mirror-twin domain boundaries and of several types of TMD point defects will be presented and discussed.

[1] Sara Barja, Sebastian Wickenburg, Zhen-Fei Liu *et al.*, *Nature Physics* 12, 751(2016).

[2] Wei Bao, Nicholas J. Borys, Changhyun Ko *et al.*, *Nature Communications* 6, 7993 (2015).

8:05pm **PCSI-MoE-8 Work Function Variations in Twisted Graphene Layers**, *Jeremy Robinson, J Culbertson*, Naval Research Laboratory; *M Berg, T Ohta*, Sandia National Laboratory

Vetting graphene and 2D materials as candidates for advanced electronics requires examination of both intrinsic and extrinsic influences on their material properties, as well as a nuanced characterization of how they respond in different heterogeneous configurations. As the field of 2D crystals expands to include stacked heterosystems, the relative orientation

(or twist angle) between layers becomes important. In the simplest case, stacking two graphene layers to form twisted bilayer graphene (TBG) already leads to measurable differences in variables such as interlayer screening[1], optical absorption[2], chiral charge carriers [3], or chemical reactivity [4].

In this talk, we describe the characterization (e.g. imaging, Raman, kelvin probe force microscopy (KPFM), and photoemission electron microscopy (PEEM)) of rotationally faulted graphene. The variable inter-layer interactions in twisted graphene layers leads to a wide range of variable properties, including optical absorption and surface potential (Φ). We find that F can vary up to $\Delta\Phi=39\text{mV}$ between 'small' ($q_s \gg 0^\circ$) and 'large' ($q_l > 16^\circ$) twist angles, and between $\Delta\Phi=36\text{-}129\text{mV}$ for different layer thickness ($N=1\text{-}4$). The PEEM measured work function of 4.4eV for graphene is consistent with doping levels on the order of 10^{12}cm^{-2} , and we find that F scales linearly with Raman G-peak wavenumber shift (slope = 22.2 mV/cm^{-1}) for all layers and twist angles, which is consistent with doping-dependent changes to graphene's Fermi energy in the 'high' doping limit[5]. The results discussed here emphasize that layer orientation is equally important as layer thickness when designing multilayer 2D systems.

[1] Sanchez-Yamagishi, et al., *PRL* 108, 076601 (2012)

[2] Robinson et al., *ACS Nano* 7, 637 (2013)

[3] Ding et al., *Chem. Mater.*, 28, 1034 (2016)

[4] Kim et al., *Nat Nano* 11, 520 (2016)

[5] Robinson et al., submitted (2017)

8:10pm **PCSI-MoE-9 Quantum Hall Effect Observed for Covalently and non-Covalently Functionalized Epitaxial Graphene**, *Evgeniya Lock, J Prestigiacomo*, Naval Research Laboratory; *P Dev*, Howard University; *A Nath*, George Mason University; *R Myers-Ward, M Osofsky, T Reinecke, K Gaskill*, Naval Research Laboratory

We demonstrate no deterioration of electrical properties for epitaxial graphene, formed on SiC substrates via Si sublimation at elevated temperatures, functionalized using both covalent and non-covalent chemical approaches. In particular, we show that these functionalized samples exhibit the Quantum Hall Effect and possess Shubnikov-de Haas oscillations, which are typical for high quality exfoliated graphene flakes. We did not observe these phenomena on the initial pristine epitaxial graphene films. From first principle calculations, these results are likely due to reduced sheet charge density after functionalization, derived from charge transfer from the chemical molecules to the epitaxial graphene. This mechanism is supported by experimental data. This means that using our approach, and through the appropriate choice of functionalization moieties, the Fermi level of graphene can be controlled without detriment to electrical properties.

8:15pm **PCSI-MoE-10 2D Magnets and Heterostructures**, *Xiaodong Xu*, University of Washington **INVITED**

Since the discovery of graphene, the family of two-dimensional (2D) materials has grown to encompass a broad range of electronic properties. However, until recently 2D crystals with intrinsic magnetism were still lacking. Such crystals would enable new ways to study 2D magnetism by harnessing the unique features of atomically-thin materials, such as electrical control for magnetoelectronics and van der Waals engineering for novel interface phenomena. In this talk, I will describe our recent magneto-optical spectroscopy experiments on van der Waals magnets, chromium(III) iodide CrI₃. I will first demonstrate the existence of isolated monolayer semiconductor with intrinsic Ising ferromagnetism. I will then show the layer number-dependent magnetic phases. The magnetic ground state evolves from being ferromagnetic in a monolayer, to antiferromagnetic in a bilayer, and back to ferromagnetic behavior in a trilayer. Lastly, I will discuss the emerging spin phenomena in monolayer WSe₂/CrI₃ ferromagnetic semiconductor heterostructures, including ferromagnetic control of valley pseudospin in WSe₂ via large magnetic exchange field, and optical analog of giant magnetoresistance effect.

8:45pm **PCSI-MoE-16 Antiferromagnetic Ordering in Atomically Thin 2-dimensional Materials Studied by Raman Spectroscopy**, *J Lee, K Kim, S Lim*, Sogang University, Republic of Korea; *S Lee, J Ryoo, S Kang, T Kim, P Kim, C Park, J Park*, Seoul National University; *Hyeonsik Cheong*, Sogang University, Republic of Korea

Magnetism in low dimensional systems is attracting much interest not only for the fundamental scientific interest but also as a promising candidate for numerous applications in sensors and data storage. However, most experimental studies on magnetism in 2-dimension so far have been

Monday Evening, January 15, 2018

limited to the magnetism arising from defects, vacancies, edges or chemical dopants which are all extrinsic effects. Recent discovery of ferromagnetism in atomically thin materials [1,2] ignited much interest in magnetism in 2 dimension in general. Antiferromagnetic ordering, on the other hand, is much more difficult to detect as the net magnetism is zero. Neutron scattering, which is a powerful tool to detect antiferromagnetic order in bulk materials, cannot be used for atomically thin samples. Raman spectroscopy has proven a powerful tool to detect ferromagnetic ordering by monitoring the zone-folding due to the antiferromagnetic order [3,4] or the signal from two magnon scattering. We report on the observation of *intrinsic* antiferromagnetic ordering in the two-dimensional limit. We demonstrate that FePS₃ exhibits an Ising-type antiferromagnetic ordering down to the monolayer limit, in good agreement with the Onsager solution for two-dimensional order-disorder transition. The transition temperature remains almost independent of the thickness from bulk to the monolayer limit with $T_N \sim 118$ K, indicating that the weak interlayer interaction has little effect on the antiferromagnetic ordering. [4] For an XXZ-type antiferromagnet NiPS₃, a signal due to two-magnon scattering and the low-energy scattering signal due to spin fluctuations are monitored to find antiferromagnetic ordering down to the monolayer limit. [5]

[1] C. Gong, *et al.*, Nature **546**, 265 (2017).

[2] B. Huang, *et al.*, Nature **546**, 270 (2017).

[3] X. Wang, *et al.*, 2D Materials **3**, 031009 (2016).

[4] J.-U. Lee, H. Cheong, *et al.*, Nano Letters **16**, 7433 (2016).

[5] K. Kim, H. Cheong, *et al.*, *in preparation*.

Author Index

Bold page numbers indicate presenter

— B —

Barja, S: PCSI-MoE-7, **1**
Barnard, E: PCSI-MoE-7, **1**
Berg, M: PCSI-MoE-8, **1**
Borys, N: PCSI-MoE-7, **1**
Burg, G: PCSI-MoE-1, **1**
— C —
Cheong, H: PCSI-MoE-16, **1**
Culbertson, J: PCSI-MoE-8, **1**
— D —
Dev, P: PCSI-MoE-9, **1**
— F —
Fallahazad, B: PCSI-MoE-1, **1**
— G —
Gaskill, K: PCSI-MoE-9, **1**
— K —
Kang, S: PCSI-MoE-16, **1**
Kim, K: PCSI-MoE-1, **1**; PCSI-MoE-16, **1**

Kim, P: PCSI-MoE-16, **1**
Kim, T: PCSI-MoE-16, **1**
— L —
Larentis, S: PCSI-MoE-1, **1**
Lee, J: PCSI-MoE-16, **1**
Lee, S: PCSI-MoE-16, **1**
Lim, S: PCSI-MoE-16, **1**
Lock, E: PCSI-MoE-9, **1**
— M —
Movva, H: PCSI-MoE-1, **1**
Myers-Ward, R: PCSI-MoE-9, **1**
— N —
Nath, A: PCSI-MoE-9, **1**
— O —
Ogletree, D: PCSI-MoE-7, **1**
Ohta, T: PCSI-MoE-8, **1**
Osofsky, M: PCSI-MoE-9, **1**

— P —

Park, C: PCSI-MoE-16, **1**
Park, J: PCSI-MoE-16, **1**
Prestigiacomo, J: PCSI-MoE-9, **1**
— R —
Reinecke, T: PCSI-MoE-9, **1**
Robinson, J: PCSI-MoE-8, **1**
Ryoo, J: PCSI-MoE-16, **1**
— S —
Schuler, B: PCSI-MoE-7, **1**
— T —
Tutuc, E: PCSI-MoE-1, **1**
— W —
Weber-Bargioni, A: PCSI-MoE-7, **1**
Wickenberg, S: PCSI-MoE-7, **1**
— X —
Xu, X: PCSI-MoE-10, **1**