2D Materials

Room 304 - Session 2D-TuA

Synthesis and Processing of 2D Materials

Moderator: Tiancong Zhu, Purdue University

2:15pm **2D-TuA-1 Advances in Synthesis, Transfer, and Magnetization Dynamics in van Der Waals Magnets**, ***Roland Kawakami,*** The Ohio State University **INVITED**

In this talk, I will discuss two of our recent results on 2D materials. First is the observation of a new way to excite magnetization dynamics via optical pump pulses in WS_2/$CrGeTe_3$ bilayers [1]. The key result is that absorption of a laser pulse results in charge transfer between 2D semiconductor WS_2 and 2D magnet $CrGeTe_3$, which results in an ultrafast change in the magnetic anisotropy and the subsequent generation of torque on the magnetization of $CrGeTe_3$. This is an optically-excited analog of the magnetoelectric effect, where the optical pulse generates a dynamic interfacial electric field and an ultrafast variation of the carrier density. This ultrafast spin-torque represents a new way to excite magnetization dynamics, beyond typical thermal excitation mechanisms. The second result is the full-film dry transfer of van der Waals films grown by molecular beam epitaxy (MBE). Although there has been much work on the transfer of 2D materials produced by exfoliation or chemical vapor deposition, it turns out that the transfer of MBE-grown films had not been achieved due to a stronger adhesion of MBE films to the growth substrate. We show that by using the polymer PCL and a solid roller with a large radius of curvature to limit the peel-off angle, the MBE film can be released from the growth substrate with minimal cracking and a large area yield (>90%) [2]. Further, the magnetic properties of 2D magnets remained largely preserved during the transfer process. This provides a route for a layer-by-layer fabrication strategy for scalable, CMOS-compatible device arrays and applications.

[1] Wenyi Zhou, Ravi Kumar Bandapelli, Hari Paudyal, Bangzheng Han, I-Hsuan Kao, Ziling Li, Yuqing Zhu, Durga Paudyal, Jyoti Katoch, Simranjeet Singh, Roland K. Kawakami, "Ultrafast Light-Induced Magnetoelectric Effect in van der Waals Magnetic Semiconductor Heterostructures" arXiv:2604.19080

[2] Ziling Li, Wenyi Zhou, Matthew Swann, Vika Vorona, Haley Scott, Roland K. Kawakami, "Full-film dry transfer of MBE-grown van der Waals materials," 2D Materials 12, 035003 (2025).

2:45pm **2D-TuA-3 Approaching Synthesis of Large-Area Monolayer SnSe by Multi-Stage Growth and Bayesian-MBE (BaMBE)**, ***Eric Welp***, *Qihua Zhang, Stephanie Law,* Pennsylvania State University

The outstanding piezoelectric and electronic properties of 2D SnSe has attracted significant interest with a wide range of applications such as efficient solar cells, ferroelectric memory devices, and more. While SnSe nanocrystals and large-area, 3-4 layer-thick deposited crystals have been grown in recent years, the growth of monolayer SnSe at large areas has not been reached due to poor substrate wetting. At the monolayer limit, the centrosymmetry present in multilayer films is removed resulting in strong ferroelectric and piezoelectric properties. Furthermore, SnSe stands out from the currently prominent piezoelectrics which employ lead (PZT, PMN-PT, etc.) in its lack of toxic constituents. This work develops a pathway towards large-area monolayer SnSe by molecular beam epitaxy (MBE) with multi-step growths and reconstruction of the monolayer, guided by recently developed methods in Bayesian optimization for MBE (BaMBE). The reconstruction of a deposited monolayer at high substrate temperature by supplied flux equal to the desorption rate allows small misoriented domains to desorb from the surface and preferentially grow large well-oriented grains that may coalesce into large films. To complete the efficient analysis of the multi-stage growth and reconstruction parameters with supervised machine learning, we implement Bayesian optimization to guide further synthesis by this novel method and approach large-area monolayer films of SnSe for the next generation of ferroelectric and piezoelectric devices.

3:00pm **2D-TuA-4 Plasma Enhanced Atomic Layer Deposition of Layered 2D $ZnIn_2S_4$ Thin Films: Toward Large-Area Synthesis of a Ternary 2D Chalcogenide**, ***Sudipta Mondal,*** *Ryan Weiland, Hamidur Rahman, Ageeth Bol,* University of Michigan, Ann Arbor

Ternary 2D layered chalcogenides have emerged as a promising class of n-type semiconductors for next-generation optoelectronic and energy conversion technologies due to their tunable electronic structure and strong light-matter interaction. Among them, $ZnIn_2S_4$ (ZIS), a wide-band-gap ternary chalcogenide, has attracted significant interest for photoelectrochemical (PEC) conversion, photocatalysis, and photodetector applications. Yet, scalable large-area synthesis routes, especially thin-film deposition techniques for the ZIS system, remain largely unexplored. In this work, we report the first atomic layer deposition (ALD) growth of ternary 2D $ZnIn_2S_4$ thin films. $ZnIn_xS_y$ thin films were deposited by plasma-enhanced atomic layer deposition (PEALD) using supercycles composed of ZnS and In_2S_3 subcycles, with growth monitored by in-situ ellipsometry. β-diketonate indium and zinc precursors, chosen for their thermal stability over a wide temperature window, were combined with H_2S plasma to develop the individual ZnS and In_2S_3 processes and, subsequently, the ternary ZIS process. Crystalline films were obtained at 500 °C, and the supercycle ratio was carefully tuned to approach the target $ZnIn_2S_4$ stoichiometry. Among the conditions investigated, a 2 ZnS:8 In_2S_3 (2-8) supercycle led to the formation of $ZnIn_{2.29}S_{3.48}$ as determined by XPS, closest to the desired stoichiometry. GIXRD analysis revealed the presence of a $(00l_1)$ reflection, characteristic of a layered ZIS phase. The observation of only basal-plane ZIS reflections, with no non-basal peaks, indicates highly preferred c-axis-oriented growth, with the basal planes parallel to the substrate. An increase in the number of indium subcycles in the ALD supercycle structure, such as a 2-10 recipe, led to the formation of a mixed ZIS and tetragonal β-In_2S_3 phase with a stoichiometry of $ZnIn_{3.03}S_{4.27}$, highlighting the importance of cycle ratio control to achieve a pure ZIS phase. AFM and SEM analyses showed the presence of triangular and hexagonal layered 2D-platelet-like morphology, confirming the GIXRD results and consistent with layered 2D growth. UV-Vis measurements gave an optical band gap of 2.6 eV for the 2-8 ZIS film, in good agreement with reported values. Moreover, the 2-8 sample was annealed in a tube furnace at 700 °C under H_2S atmosphere, leading to a stoichiometric $ZnIn_{2.10}S_{4.02}$ sample with an additional low-angle $(00l_2)$ reflection appearing, indicating a more crystalline and layered ZIS film formation. This study establishes a thin film deposition technique for layered $ZnIn_2S_4$ and provides insights into phase and defect engineering in ternary layered chalcogenide semiconductors.

3:15pm **2D-TuA-5 Two-Step Synthesis of MoSe2 from MoOx Thin Films by Low Temperature Thermal Atomic Layer Deposition**, *Icelene Leong,* Boise State University*; Brian Everhart, Drake Austin,* AFRL*; Steven M. Hues,* Boise State University*; Nicholas R. Glavin,* AFRL*;* ***Elton Graugnard***, Boise State University

Two-dimensional (2D) semiconducting materials, including transition metal dichalcogenides (TMDs) and transition metal oxides, are of increasing interest for applications in microelectronics, photonics, catalysis, and energy technologies. Atomic layer deposition (ALD) provides a scalable route for synthesis of molybdenum oxide (MoO_x) thin films; however, the structural and functional properties of the resulting materials depend strongly on precursor chemistry, oxidation state, and post-deposition processing conditions. A two-step conversion approach offers a promising pathway to TMD formation from ALD metal oxides. Here, we report a laser-assisted two-step processing method for low-temperature ALD-grown MoO_x films for the conversion to $MoSe_2$. Laser annealing was employed to locally modify surface composition, crystallinity, and oxidation state under controlled environments, enabling systematic tuning of the intermediate oxide prior to selenization. High-throughput processing produced many distinct material conditions on a single sample by varying laser intensity and exposure time. Complementary high-throughput characterization using Raman spectroscopy, x-ray photoelectron spectroscopy (XPS), and x-ray diffraction, enabled rapid identification of processing conditions associated with high-quality $MoSe_2$ films. These results demonstrate new routes for processing ALD oxides to TMD that can be used in numerous applications.

4:00pm **2D-TuA-8 Epitaxy Growth of P-Type 2d Semiconductors**, ***Vincent Tung,*** The University of Tokyo, Japan **INVITED**

The rise of two-dimensional (2D) semiconductors represents the next wave of innovation in semiconductor technology, offering new opportunities for scaling channel materials and rethinking device architectures beyond the limits of bulk silicon CMOS. However, despite considerable progress in n-type 2D materials, such as MoS_2 and WS_2, the development of high-performance p-type counterparts remains significantly underdeveloped. This imbalance presents a critical bottleneck, as the lack of robust p-type 2D semiconductors hinders the realization of complementary logic circuits and limits the overall performance gain relative to silicon CMOS. A key challenge in identifying suitable p-type 2D semiconductors lies in the inherent difficulty of achieving stable valence band alignment, sufficient hole mobility, and reliable contact formation without introducing Fermi-level pinning or unintentional doping. Moreover, the performance of 2D devices is often compromised by degraded semiconductor/dielectric and semiconductor/electrode interfaces, especially when harsh fabrication

steps or transfer processes are involved. These interfaces, though only a few atoms thick, play a decisive role in governing charge transport, threshold voltage stability, and scalability. In this talk, I will present our recent efforts in addressing these challenges by focusing on the synthesis and integration of p-type 2D semiconductors, particularly under low-temperature conditions. Emphasis will be placed on strategies for the direct growth of these materials on insulating substrates, thereby preserving electronically clean and atomically well-defined interfaces. This approach offers a practical pathway toward realizing high-performance complementary 2D logic while maintaining compatibility with back-end-of-line (BEOL) process constraints.

4:30pm **2D-TuA-10 Repeatability and Reproducibility in the Chemical Vapor Deposition of 2D Films: A Physics-Driven Exploration of the Reactor Black Box***, **Shahana Chatterjee**,* Make Materials, Canada*; Thomas Abadie,* University of Birmingham, UK*; Meihui Wang,* IBS Center for Multidimensional Carbon Materials, Republic of Korea*; Omar Matar,* Imperial College London, UK*; Rodney Ruoff,* IBS Center for Multidimensional Carbon Materials, Republic of Korea

Chemical vapor deposition (CVD) on metal substrates is the method of choice for growing single-crystalline high-quality 2D films for various opto-electronic applications. Though one and a half decades have passed since the CVD synthesis of graphene was first reported (and followed by that of other 2D films), multiple problems remain, stemming from the fact that the CVD reactors are still virtual black-boxes with poorly understood reaction environments. For example, repeatability issues, such as an optimized growth condition changing over time in the same reactor, or reproducibility issues, such as the failure to reproduce the growth process in a different reactor (even in the same research laboratory), are well known. It is indeed quite difficult to (1) study process kinetics, (2) correlate experimental results to atomic level simulations, (3) build up from previous publications or test a new hypothesis, and, (4) scale-up and commercialize lab-scale processes. This presentation will explore what may be done to circumvent this complicated problem, as monitoring or measuring the reactor environment directly is challenging or even impossible. We will discuss how relevant external process and reactor parameters may first be identified with the help of the Computational Fluid Dynamics (CFD) toolbox OpenFOAM and then utilized to recreate the reaction environment. Finally, the critical importance of the reactor environment on such processes, and, a protocol for experimentalists to use while monitoring and reporting them will be discussed.

References

1. Chatterjee, S.; Abadie, T.; Wang, M.; Matar, O. K.; Ruoff, R. S. *Repeatability and Reproducibility in the Chemical Vapor Deposition of 2D Films: A Physics-Driven Exploration of the Reactor Black Box*. Chemistry of Materials 2024, 36, 1290-1298.

2. Wang, M.; Kim, Y. C.; Meng, Y.; Chatterjee, S.; Bakharev, P.; Luo, D.; Gong, Y.; Abadie, T.; Kim, M. H.; Sitek, J.; Seong, W. K.; Lee, G.; Ruoff, R. S. *Growth Kinetics of Graphene on Cu(111) Foils from Methane, Ethyne, Ethylene, and Ethane*. Angewandte Chemie International Edition 2024, 63, e202412131.

4:45pm **2D-TuA-11 Topotactic Formation of 2D Ternary Single Crystals***, Nitin Shinde, Shavit Ben David, **Ariel Ismach**,* Tel Aviv University, Israel

The growth of 2D materials has advanced significantly in recent decades, enabling the synthesis of high-quality, epitaxial graphene, hexagonal boron nitride (h-BN), and transition metal dichalcogenides (TMDs). Beyond these mono- and binary systems, a vast number of 2D ternary compounds offer compelling physical and chemical properties. However, the growth of such ternary thin films and crystals *via* methods like chemical vapor deposition (CVD) remains largely unexplored.

In this talk, I will present our approach for synthesizing oriented ternary compounds of the type $MePS_3$, where Me represents a transition metal (*e.g.*, Ni, Fe, *etc.*). We propose a two-step methodology in which it begins with the growth of an epitaxial transition metal thin film, which is subsequently transformed into a ternary compound via a topotactic reaction. Because topotaxy is a solid-state reaction that maintains structural correlation with the initial film, oriented thin crystals form on the substrate, a methodology well-suited for large-scale growth and integration. I will detail the underlying growth mechanism and discuss the resulting crystal structure and magnetic properties. This work introduces a viable pathway for the scalable synthesis of complex 2D ternary compounds with tailored chemical compositions.

5:00pm **2D-TuA-12 Spatial Doping of CVD-Grown WSe2 for Lateral Junction and Optoelectronic Applications***, **Utsab Kafley**, Anupama Kaul,* University of North Texas

Two-dimensional (2D) transition metal dichalcogenides (TMDCs), particularly tungsten diselenide (WSe_2), have emerged as promising materials for next-generation electronic and optoelectronic applications due to their tunable electrical and optical properties. Here, we investigate localized chemical modification of chemical vapor deposition (CVD)-grown WSe_2 using a methylammonium based dopant to achieve selective carrier modulation and lateral junction engineering. The influence of selective chemical doping on the electronic, structural, and optical response of WSe_2 is systematically explored to understand charge-transfer-induced modulation in atomically thin semiconductors. The localized doping process leads to measurable variations in carrier transport, field-effect mobility, surface morphology, vibrational response, excitonic emission, rectification characteristics, and light sensitivity, indicating effective tuning of material properties through spatial carrier engineering. The formation of laterally modulated regions further enables investigation of junction-like transport behavior and optoelectronic response within single CVD-grown WSe_2 membranes.

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