2D Materials

Room 304 - Session 2D+QS-TuM

Novel Quantum 2D Materials

Moderators: Fei Yao, University at Buffalo, **Roland Kawakami**, The Ohio State University

11:00am **2D+QS-TuM-13 Watching 2D Materials Form: Real-Time Atomic-Scale Visualization of TMD Growth and Crystallization Pathways**, ***Raymond Unocic***, *Prajakta Prabhune, Aidan Cotton,* North Carolina State University*; Caitlin Obrero,* Stanford University*; Suman Jaiswal, Masoud Mahjouri-Samani,* Auburn University*; Kyungnam Kang, Kai Xiao,* Oak Ridge National Laboratory **INVITED**

Achieving control over 2D material functional properties requires direct insight into the dynamic pathways that drive nucleation, crystallization, and growth. Aberration-corrected scanning transmission electron microscopy (STEM), coupled with in situ capabilities, has emerged as a powerful platform that can be used to directly observe these processes with atomic-scale spatial resolution and under a range of synthesis conditions. In this work, we investigate thermolysis-driven growth and pulsed laser deposition (PLD)-derived amorphous-to-crystalline transformations for 2D transition metal dichalcogenides (TMDs). Using MEMS based heating microchips, controlled heating experiments were performed to directly visualize the evolution of amorphous precursor phases into crystalline layered structures, revealing the atomic-scale pathways governing growth and phase evolution. By systematically varying thermal conditions and electron dose, we aimed to establish correlations between processing parameters and the resulting kinetic and energetic landscapes that dictate crystallization behavior. To further uncover hidden relationships within the transformation dynamics, we employ data analytics and machine learning approaches to quantify atomic trajectories and to identify transient states. The direct observation of these transformations provides unprecedented insight into the role of local structural rearrangements and transient intermediate states in governing the evolution of final material architectures. These findings establish a foundation for predictive synthesis strategies and enable atomic-scale control over TMD growth pathways.

11:30am **2D+QS-TuM-15 Airgap-Mediated Remote Epitaxial Control of Bandgap Opening in Bilayer Silicene on Semiconductor Substrates**, ***Kumar Vishal,*** Wright University

Bilayer silicene (BLSi) is a promising silicon-compatible 2D material for future nanoelectronic channels, but practical bandgap engineering requires simultaneous strain retention, substrate interaction control, and release from growth templates. We use first-principles density functional theory to model epitaxial and remote-epitaxial BLSi on lattice-mismatched semiconductor substrates, with emphasis on Ga- and Sb-terminated GaSb surfaces. Substrate-induced biaxial tensile strain is used to flatten and electronically activate BLSi, while the BLSi/substrate separation is varied to emulate an airgap interlayer that suppresses direct chemical bonding.The calculations reveal three interaction regimes governed by optimized interface spacing, surface termination, electron transfer, and residual buckling. For Ga-terminated GaSb, direct epitaxy relaxes to a strongly bonded interface at Δd0 = 0.817 Å, producing a 112 meV bandgap but limiting transferability. Increasing the spacing creates a metastable remote-epitaxial regime at Δd0 ≈ 2.6–2.9 Å, where electron density at the interface drops below 0.075 Å^-3 and the bandgap becomes tunable in the 20–30 meV range. At larger spacing, interfacial charge transfer becomes negligible and BLSi approaches the strained free-standing limit near 57–59 meV. For Sb-terminated GaSb, the weaker Si-Sb interaction enables a wider practical tuning window: an airgap of approximately 3.0–4.3 Å gradually opens the BLSi bandgap from near zero to 57 meV while maintaining a weakly coupled, releasable interface.These results show that an atomically controlled airgap can function as a passive remote-epitaxial layer, preserving lattice-mismatch strain while preventing irreversible covalent bonding. The approach offers a low-complexity route for tuning bilayer silicene electronic structure and for integrating strain-engineered 2D silicon materials with scalable semiconductor device processing.The technical details above are grounded in your uploaded paper, including the DFT modeling, GaSb_Ga/GaSb_Sb comparison, three interface-spacing regimes, electron-density reduction, and tunable bandgap results.

11:45am **2D+QS-TuM-16 Synthesis and Characterization of Two-Dimensional Indium Gallium Nitride (2d $In_xGa_{1-x}N$) via Graphene Encapsulation**, ***Krishnan Mekkanamkulam Ananthanarayanan***, *Soukyoon Kim, Md Farhan, Nutifafa Y. Doumon, Adri van Duin, Joshua Robinson,* The Pennsylvania State University

Group-III nitrides are widely explored semiconducting material for optoelectronics including light-emitting diodes (LEDs), photodetectors, solar cells and lasers; however, quantum applications such as single photon emitters, tunnel junctions and quantum sensors remain unrealized. Atomically thin two-dimensional (2D) nitrides are predicted to have large tunable bandgap than the bulk counterpart via quantum confinement with electronic and optical properties for wide spectrum of applications such as deep UV light emitters, single photon sources and other optoelectronic devices. First-principles calculations predict 2D $In_xGa_{1-x}N$ to possess a tunable bandgap ranging from 1.7 to 4.1 eV depending on the composition. However, experimental realization of large area 2D nitrides growth other than hBN remains elusive due to 3D growth induced by surface energy constraints. Graphene encapsulation has paved the way for realization of MOCVD growth of 2D GaN_x (E_g≈5eV), InN_x (E_g≈2eV) and AlN_x (E_g≈9eV). The significant limitation in the MOCVD growth of 2D nitrides via graphene encapsulation is the 3D island formation on top of graphene and small domain growth. In this study, we have synthesized large-area 2D $In_xGa_{1-x}N$ by intercalation of 2D In_xGa_{1-x} alloy at the interface of EG/SiC using confinement heteroepitaxy (CHet) and followed by ammonolysis to intercalate nitrogen to counter the 3D island formation. The high energy interface between epitaxial graphene (EG) and silicon carbide (SiC) efficiently stabilizes the metal and other compounds into confined 2D crystalline form. The intercalation of tunable 2D In_xGa_{1-x} metal alloy composition by changing the relative elemental composition of the precursor using CHet technique have been previously demonstrated. XPS, cross sectional transmission electron microscopy (TEM), energy dispersive X-ray spectroscopy (EDS) and AES mapping provides evidence of ~100 μm^2 2D $In_xGa_{1-x}N$ at the EG/SiC interface and determine the III-alloy nitride composition. The electron energy loss spectroscopy (EELS) and UV-vis spectroscopy extract the energy bandgap of 2D $In_xGa_{1-x}N$ with varying alloy compositions. The vertical transport measurements of the heterostructure are carried out using conductive atomic force microscopy (C-AFM) using a conductive SiC substrate. Scanning electron microscopy (SEM) and AFM validate the reduced 3D island formation. The surface morphology of 2D III-nitride consists of graphene wrinkles, tears and nitrogen trapped bubbles formed underneath the EG. We combine the study with ReaxFF reactive molecular dynamics simulations to elucidate the mechanism of graphene bubbles formation and the role of EG, In and Ga.

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