Nanoscale Science and Technology Division
Room B113 - Session NS1+2D+EM+MN-TuA

Nanofabrication and Characterization of Low-Dimensional Materials

Moderator: Georg Fantner, EPFL

2:20pm **NS1+2D+EM+MN-TuA-1 Atomic-Scale Design and Defect Networks at the 2D/3D Interface***, Kate Reidy,* MIT **INVITED**

'Mixed dimensional' 2D/3D van der Waals heterostructures, where 3D metallic nanostructures are integrated with suspended two-dimensional (2D) van der Waals materials, show unique functionalities including light-matter coupling, charge transfer, and enhanced catalytic activity. To enable such integration, an understanding of how structure and defects at the 2D/3D interface affect heterostructure properties is required. Moreover, 2D/3D heterostructures display fluctuations of opto-electronic properties in nanometer spatial range; and it is advantageous to probe position-dependent properties at the same spatial scales. In this seminar, I will share work exploring the local properties of the 2D/3D interface using a combination of atomic resolution scanning transmission electron microscopy (STEM), in situ ultra-high vacuum (UHV) TEM, and monochromated high-energy resolution electron energy-loss spectroscopy (EELS). We demonstrate epitaxial, single-crystalline metallic nanoisland growth of technologically relevant metals (Au, Ti and Nb) with with ultra-low defect density interfaces and facetted morphologies on several thin suspended 2D materials. We then explore the key parameters of 2D/3D growth, including the role of temperature, defects, moiré, surface chemistry, and thermodynamic equilibrium shapes. Lastly, we fabricate more complex heterostructure stacks with defect densities controlled by the compliance of the 2D material substrate. Through fundamental understanding of the structure-property-performance relationship, we suggest that future electronic, magnetic, and optical nanodevices will utilize versatile fabrication of 2D/3D heterostructures with well-characterized interfaces and morphologies.

3:00pm **NS1+2D+EM+MN-TuA-3 Highly Asymmetric Doping of Epitaxial Bilayer Graphene by Targeted Bonding of the Intercalated Gadolinium***, Marek Kolmer,* Ames National Laboratory*; J. Hall,* Ames National Laboratory and Department of Physics and Astronomy, Iowa State University*; S. Chen, M. Tringides,* Ames National Laboratory, Department of Physics and Astronomy, Iowa State University

Heterostructures consisting of vertically stacked two-dimensional (2D) materials have recently gained large attention due to their highly controllable electronic properties. Particularly, mechanically stacked multilayered systems offer exceptional control over a stacking sequence or interlayer twist angles. On the other hand, epitaxially grown 2D materials express unprecedented quality and stability over wafer-scale lengths. In both cases controlling the interlayer coupling can generate novel electronic and topological phases and its effective implementation is commonly done with a transverse electric field. However, phases generated by high displacement fields are elusive.

Here, we introduce an exceptionally large displacement field by structural modification of a model system: AB-stacked epitaxial bilayer graphene (BLG) on a SiC(0001) surface. We show that upon intercalation of gadolinium with two specific interlayer locations, electronic states in the top two graphene layers exhibit a significant difference in the on-site potential energy (~1 eV), which effectively breaks the interlayer coupling between them. As a result, for energies close to the corresponding Dirac points, the BLG system behaves like two electronically isolated single graphene layers. We prove this fact by a comprehensive multi-technique methodology based on low-temperature scanning tunneling microscopy/spectroscopy (STM/S) and angle-resolved photoelectron spectroscopy, which are corroborated by density functional theory, tight binding, surface diffraction and multiprobe STM transport. The work presents charge transfer from intercalated metal atoms as a promising approach for the synthesis of 2D graphene heterostructures with electronic phases generated by giant displacement fields.

References

[1]M. Kolmer, B. Schrunk, M. Hupalo, J. Hall, S. Chen, J. Zhang, C.-Z. Wang, A. Kaminski, M.C. Tringides, "Highly Asymmetric Graphene Layer Doping and Band Structure Manipulation in Rare Earth–Graphene Heterostructure by Targeted Bonding of the Intercalated Gadolinium", J. Phys. Chem. C. 126, 6863 (2022). https://doi.org/10.1021/acs.jpcc.2c01332.

[2]M. Kolmer, W. Ko, J. Hall, S. Chen, J. Zhang, H. Zhao, L. Ke, C.-Z. Wang, A.-P. Li, M.C. Tringides, "Breaking of Inversion Symmetry and Interlayer Electronic Coupling in Bilayer Graphene Heterostructure by Structural Implementation of High Electric Displacement Fields", J. Phys. Chem. Lett. 13, 11571 (2022). https://doi.org/10.1021/acs.jpclett.2c02407.

4:20pm **NS1+2D+EM+MN-TuA-7 AVS Dorothy M. and Earl S. Hoffman Scholarship Recipient Talk: Exfoliated 2D Nanosheets for Large-Area, Solution-Processed Optoelectronics***, Lidia Kuo*[1]*, S. Rangnekar, V. Sangwan, M. Hersam,* Northwestern University

Two-dimensional (2D) materials exhibit thickness-dependent optoelectronic properties due to their atomically thin nature, unlike their bulk layered crystalline counterparts. In particular, semiconducting MoS_2 undergoes an indirect to direct bandgap transition as the thickness is decreased to the monolayer limit, leading to enhanced optical absorption and emission at the atomically thin scale. Liquid-phase exfoliation (LPE) is a scalable and cost-effective method for obtaining 2D materials from bulk crystals. However, the yield of monolayer sheets by LPE has been impractically low in previous work. The resulting LPE-processed optoelectronic devices have fallen short compared to nanosheets derived from mechanical exfoliation or chemical vapor deposition. Here, we demonstrate that LPE coupled with megasonic exfoliation – i.e., processing at megahertz frequencies compared to the kilohertz frequencies commonly utilized for LPE – yields an unprecedentedly high fraction of monolayer MoS_2. As a result, megasonic exfoliation enables ultrahigh responsivity in printed MoS_2 photodetectors as well as the first demonstration of electroluminescence for large-area, solution-processed MoS_2 films. This work establishes megasonic exfoliation as a scalable and generalizable approach for achieving optoelectronic-grade 2D semiconductors via LPE.

[1] **AVS Dorothy M. and Earl S. Hoffman Scholarship Recipient**

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