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2D Materials
Room On Demand - Session 2D-Invited On Demand

2D Materials Invited On Demand Session

2D-Invited On Demand-7 Tuning Energy Levels and Energy Flow in Nanomaterials Using the External Environment, ***Archana Raja,*** Lawrence Berkeley National Laboratory **INVITED**

The interaction between charge carriers in atomically thin, quasi-two-dimensional (2D) materials is strongly influenced by the local environment. I will discuss how tuning the external dielectric screening can modify the band gap and exciton energies in 2D transition metal dichalcogenides, allowing the formation of lateral heterojunctions without modifying the material itself [1]. Furthermore, using a combination of optical and angle resolved photoemission spectroscopies with microscopic spatial resolution, we show that the band structure rigidly shifts in response to the change in local dielectric screening [2]. This environmental sensitivity can also lead to a new type of disorder that leads to spatially inhomogeneous band gap and exciton energies as a consequence of spatial variations in the external dielectric screening rather than any material imperfections [3]. I will also briefly discuss new experimental approaches to the study of interfacial phenomena in 2D heterostructures and the associated ultrafast dynamics. In addition to the intrinsic scientific interest in understanding materials in this distinctive regime, such control offers a non-invasive approach to engineer material properties by tuning the local environment rather than the material itself, yielding a new paradigm for nanoscale devices.

[1] A. Raja et al. Coulomb engineering of the bandgap and excitons in two-dimensional materials. Nature Communications 8, 15251 (2017)

[2] L. Waldecker*, A. Raja*, M. Rösner* et al. Rigid Band Shifts in Two-Dimensional Semiconductors through External Dielectric Screening. Physical Review Letters, 123(20), 206403 (2019)

[3] A. Raja*, L. Waldecker* et al. Dielectric disorder in two-dimensional materials. Nature Nanotechnology 14 (9), 832 (2019)

2D-Invited On Demand-13 Atomic-Scale Study and Engineering of Low-Dimensional Materials, ***Jani Kotakoski,*** University of Vienna, Austria **INVITED**

Despite the great promise of two-dimensional materials due to their exciting properties, they are not always directly suitable for applications. One way to tune the material properties is to manipulate the atomic structure using particle irradiation. However, as one might expect, this is challenging to do in the case of extremely thin materials, where careful control over the irradiation energy and solid understanding of the underlying atomic-scale phenomena are required. Despite the challenges, electron and ion irradiation have recently evolved into powerful techniques to manipulation the atomic structure of two-dimensional materials. At the same time, the recent advancements in aberration-corrected transmission electron microscopy both provide means to directly image the manipulated structures but also to fine tune them by inducing local structural changes and even to move defects and impurity atoms.In this presentation, I will show that low-dimensional materials change shape due to van der Waals interaction within heterostructures, describe the advances in manipulating low-dimensional materials with electron irradiation, including nanometer-range chemical etching and directed diffusion of impurity atoms, and provide an overview of our latest results in using ion irradiation at a large energy scale to modify 2D materials via introduction of foreign atoms and nanopores. Finally, I will also demonstrate that transmission electron microscopy can be incorporated as an integral part of a ultra-high vacuum setup with different possibilities for experimentation before, during and after atomic-resolution microscopy, including the growth of novel 2D materials using free-standing graphene as the substrate.

2D-Invited On Demand-19 Electronic Structures of Two-Dimensional Topological Materials, ***Sung-Kwan Mo,*** Lawrence Berkeley National Laboratory **INVITED**

Due to the quantum confinement and changes in the symmetry, electrical and topological properties of atomically-thin two-dimensional (2D) materials are often largely different from those of their bulk counterparts. In this talk, I will focus on the changes in topological properties in 2D layers, which is explored by combining bottom-up growth using molecular beam epitaxy (MBE), in situ angle-resolved photoemission (ARPES), scanning tunneling microscopy/spectroscopy (STM/STS), and first principle calculations. We have found that the topological properties of 2D materials can be tuned by thickness, lateral interface, strain, and electric field. The material systems under discussion include 1T'-WTe_2 [1], 1T'-WSe_2 [2], alpha-Sn [3] and Na_3Bi [4].

[1] S. Tang et al., Nat. Phys. **13**, 683 (2017). [2] M. M. Ugeda et al., Nat. Commun. **9**, 3401 (2018). [3] C.-Z. Xu et al., Phys. Rev. Lett. **118**, 146402 (2017); Phys. Rev. B **97**, 035122 (2018). [4] J. L. Collins et al., Nature **564**, 390 (2018).

2D-Invited On Demand-25 2020 AVS Medard W. Welch Award Lecture: Chemically Tailoring Interfaces in Two-Dimensional Heterostructures, ***Mark Hersam***[1]***,*** Northwestern University **INVITED**

As a result of their unique electronic, optical, and physical properties, two-dimensional (2D) materials are actively being explored for applications in next-generation computing [1], quantum information science [2], and energy technologies [3]. With exceptionally high surface-to-volume ratios, 2D materials are highly sensitive to their environment, resulting in a strong dependence of their properties on substrate effects, extrinsic adsorbates, and interfacial defects. Furthermore, the integration of 2D materials into heterostructure devices introduces further demands for controlling interfaces with atomic precision. With this motivation, this talk will explore emerging efforts to understand and utilize interfacial chemical functionalization to influence the properties of 2D heterostructures. For example, organic adlayers can tailor chemical reactivity to enable conformal atomic layer deposition of pinhole-free encapsulation layers that mitigate the deleterious effects of ambient exposure, particularly for ambient-unstable 2D materials such as black phosphorus and monochalcogenides [4]. The integration of organic self-assembled monolayers with 2D semiconductors also allows for tailoring of electronic and optical properties such as photoinduced charge separation in fullerene/InSe heterojunctions [5] and mixed-dimensional excitonic states in phthalocyanine/MoS_2 heterojunctions [6]. By exploiting spatially inhomogeneous surface chemistry, seamless lateral 2D heterostructures can also be realized including perylene/borophene [7] and graphene/borophene [8] heterostructures, each of which show atomically sharp electronic interfaces as confirmed by ultrahigh vacuum scanning tunneling microscopy and spectroscopy. Overall, by providing substantial tailoring of interfaces, chemical functionalization presents opportunities for improved functionality in 2D heterostructure devices.

[1] V. K. Sangwan, *et al.*, *Nature Nanotechnology*, DOI: 10.1038/s41565-020-0647-z (2020).

[2] X. Liu, *et al.*, *Nature Reviews Materials*, **4**, 669 (2019).

[3] S. Padgaonkar, *et al.*, *Accounts of Chemical Research*, **53**, 763 (2020).

[4] S. A. Wells, *et al.*, *Nano Letters*, **18**, 7876 (2018).

[5] S. Li, *et al.*, *ACS Nano*, **14**, 3509 (2020).

[6] S. H. Amsterdam, *et al.*, *ACS Nano*, **13**, 4183 (2019).

[7] X. Liu, *et al.*, *Science Advances*, **3**, e1602356 (2017).

[8] X. Liu, *et al.*, *Science Advances*, **5**, eaax6444 (2019).

2D-Invited On Demand-31 First-Principles Calculations of 2D Materials for Gas Sensing Applications, ***Udo Schwingenschlogl,*** KAUST, Saudi Arabia **INVITED**

We study the potential of material simulations based on first-principles methods to predict gas sensing properties of 2D materials. This emerging class of materials is of particular interest to gas sensing applications due to high surface-to-volume ratios and chemical stability. We discuss in detail results of electron transport calculations within the Landauer-Büttiker formalism and compare the conclusions to analyses in terms of adsorption energies, charge transfers, and work functions. Specific examples include the effects of the interlayer interaction in bilayer MoS_2 and WS_2 on the gas sensing performance and the consequence of the presence of reactive Si in Si_2BN. We also address the properties of C_3N and para/meta-C_3Si. Potential of very sensitive gas sensing is demonstrated for para-C_3Si and is explained by the susceptibility of Dirac states to symmetry breaking distortions rather than by a mechanism based on charge transfer. Finally, the enhanced gas sensing performance of monovacant C_6BN is studied and it is shown that the work function changes of both pristine and monovacant C_6BN during

[1] **AVS 2020 Medard W. Welch Awardee**

gas adsorption do not correlate with the changes observed in the I-V characteristics.

2D-Invited On Demand-37 Modeling the Growth of 2D Crystals: Analytical, Phase-Field and Machine Learning Methods*, Vivek Shenoy,* University of Pennsylvania **INVITED**

Vertical stacking of monolayers *via* van der Waals (vdW) interaction opens promising routes toward engineering physical properties of two-dimensional (2D) materials and designing atomically thin devices. Increasingly, the bottleneck in this field is the controlled synthesis of these materials through methods such as chemical etching and chemical vapor deposition (CVD). In this talk, I will present insights into synthesis and growth of 2D materials developed from analytical, phase-field, and machine learning models. First, we adapt the state-of-the-art positive and unlabeled (PU) machine learning framework to predict which theoretically proposed 2D materials in the MXene family have the highest likelihood of being successfully synthesized. By considering both the MXenes and their precursors, we identify 18 MXene compounds that are highly promising candidates for synthesis. Next, we develop a general multiscale model for scalable CVD growth of layered materials and predict the necessary growth conditions for vertical (initial + subsequent layers) *versus* in-plane lateral (monolayer) growth. An analytic thermodynamic criterion is established for multilayer growth that depends on the sizes of both layers, the vdW interaction energies, and the edge energy of 2D layers. We connect the model to experimental controls and find that temperature and adatom flux from vapor are the primary criteria affecting the self-assembled growth. This model agrees with experimental observations of various monolayer and bilayer transition metal dichalcogenides grown by CVD. Finally, we consider CVD synthesizable transition metal dichalcogenide heterostructures as a robust platform for engineering quantum confinement of Dirac fermions using a multiscale model for electronic properties.

Author Index

Bold page numbers indicate presenter