2D Materials
Room A226 - Session 2D+EM+MI+MN+NS+QS-WeM

Novel 2D Materials

Moderator: Phil King, University of St Andrews

8:00am **2D+EM+MI+MN+NS+QS-WeM-1 A Safari Through Thousands of Layered Materials Guided by Data Science Techniques***, **Evan Reed**, G Cheon,* Stanford University **INVITED**

We have utilized data mining approaches to elucidate over 1000 2D materials and several hundred 3D materials consisting of van der Waals bonded 1D subcomponents, or molecular wires. We find that hundreds of these 2D materials have the potential to exhibit observable piezoelectric effects, representing a new class of piezoelectrics. A further class of layered materials consists of naturally occurring vertical hetero structures, i.e. . bulk crystals that consist of stacks of chemically dissimilar van der Waals bonded layers like a 2-D super lattice. We further combine this data set with physics-based machine learning to discover the chemical composition of an additional 1000 materials that are likely to exhibit layered and two-dimensional phases but have yet to be synthesized. This includes two materials our calculations indicate can exist in distinct structures with different band gaps, expanding the short list of two-dimensional phase change materials. We find our model performs five times better than practitioners in the field at identifying layered materials and is comparable or better than professional solid-state chemists. Finally, we find that semi-supervised learning can offer benefits for materials design where labels for some of the materials are unknown.

8:40am **2D+EM+MI+MN+NS+QS-WeM-3 2D Ferroelectric Semiconductor α-In_2Se_3 for Non-Volatile Memory Applications***, M Si, **Peide Ye**,* Purdue University

α-In_2Se_3 is a novel two-dimensional (2D) ferroelectric semiconductor. It has a bandgap of ~1.39 eV, room temperature ferroelectricity, the ability to maintain ferroelectricity down to a few atomic layers and the feasibility for large-area growth. Based on the ferroelectric and semiconducting nature of the material, a ferroelectric semiconductor field-effect transistor (FeS-FET) was proposed and experimentally demonstrated [1]. In the FeS-FET, a ferroelectric semiconductor is employed as the channel material while the gate insulator is the dielectric. The two non-volatile polarization states in FeS-FETs exist in the ferroelectric semiconductor channel. Therefore, a high quality amorphous gate insulator can be used instead of the common polycrystalline ferroelectric insulator for Fe-FETs. The fabricated FeS-FETs exhibit high performance with a large memory window, a high on/off ratio over 10^8, a maximum on-current of 862 µA/µm, low supply voltage with scaled gate insulator and the potential to exceed the existing Fe-FETs for non-volatile memory applications.

[1] M. Si, S. Gao, G. Qiu, J. Qin, Y. Duan, J. Jian, H. Wang, W. Wu, and P. D. Ye, "A Ferroelectric Semiconductor Field-Effect Transistor,", arXiv:1812.02933.

9:00am **2D+EM+MI+MN+NS+QS-WeM-4 *Ab initio* Informed Theory of Axis-dependent Conduction Polarity in Goniopolar Materials***, **Yaxian Wang**, B He, M Arguilla, N Cultrara, M Scudder, J Goldberger, J Heremans, W Windl,* The Ohio State University

$NaSn_2As_2$ has recently been synthesized and was found to be an exfoliatable van der Waals Zintl phase, opening new opportunities for electronic design on the few-atom-thick scale. Although the band structure may suggest a range of metal to semi-metal, it shows strong anisotropy especially in its "polarity", characterized by its dominant carrier type, which strongly affects its electronic and thermal properties. We used DFT calculations to investigate bandstructure and Fermi surface. In addition, we employed BoltzTraP code to calculate the transport behavior in in/cross-plane directions, predicting strongly anisotropic carrier transport and directionally dependent polarity –"goniopolarity" – in this layered material. It is confirmed by experimental thermopower measurements. We show from simulations on a model band structure the Fermi surface geometry origin in a single-band toy model, and we utilize the bandwidth concept from a tight-binding model to give an insight of real space orbital contributions and nature of the bonding states in this layered crystal. Based on that, additional candidate materials for goniopolarity can be proposed, and the design space for goniopolar materials in general will be defined.

9:20am **2D+EM+MI+MN+NS+QS-WeM-5 In-Plane Mechanical Properties and Strain Engineering of 2D Hybrid Organic-Inorganic Perovskites***, **Qing Tu**, I Spanopoulos, S Hao, C Wolverton, M Kanatzidis, G Shekhawat, V Dravid,* Northwestern University

Two-dimensional (2D) hybrid organic-inorganic perovskites (HOIPs) are new members of the 2D materials family with wide tunability, highly dynamic structural features and excellent physical properties. Mechanical strain is inevitable in 2D-HOIP-based applications due to materials processing, thermal expansion and substrate deformation. Understanding the mechanical properties and strain engineering of such functional materials are both fundamentally and practically important to achieve high performance and mechanically stable (flexible) devices. Here the in-plane mechanical properties and the impact of in-plane uniaxial tensile strain on the electronic properties of 2D lead iodide perovskites with a general formula $(CH_3(CH_2)_3NH_3)_2(CH_3\text{-}NH_3)_{n\text{-}1}Pb_nI_{3n+1}$ were reported for the first time. The in-plane Young's modulus and breaking strength of ultrathin 2D HOIP flakes were measured by AFM-based nanoindentation of suspended 2D HOIP membranes.[1] The in-plane Young's moduli of 2D HOIPs are smaller than that of conventional covalently bonded 2D materials like graphene and MoS_2 due to the much more deformable $[PbI_6]^{4-}$ octahedra structure. Both the Young's modulus and breaking strength first decrease and then plateau as the thickness of 2D HOIP flake increases from monolayer to 4 layers, which is attributed to interlayer slippage during deformation. Ultrathin 2D HOIPs exhibit outstanding breaking strength/Young's Modulus ratio compared to many other widely used engineering materials and polymeric flexible substrates, which renders them suitable for application into flexible electronic devices. Furthermore, the uniaxial tensile strain was found to increase the band gap of 2D HOIPs.[2] Such strain effect on the band gap of 2D HOIPs is fully reversible and depends on the structural unit of the materials. For2D HOIP with n = 5, the strain response of the band gap can be as high as 13.3 meV/%. First-principles simulations show that the strain response of the band gap arises from the rotation of the inorganic $[PbI_6]^{4-}$ octahedra and the consequential Pb-I bond stretching and increase of Pb-I-Pb angle. The observed band gap–strain relationship can be harnessed to map the local mechanical strain in 2D HOIP-based devices and allow 2D HOIPs for sensing applications.

References

[1]. Tu Q, Spanopoulos I, Yasaei P, Stoumpos CC, Kanatzidis MG, Shekhawat GS, et al. Stretching and Breaking of Ultrathin 2D Hybrid Organic–Inorganic Perovskites. ACS Nano. 2018;12(10):10347-54.

[2]. Tu Q, Spanopoulos I, Hao S, Wolverton C, Kanatzidis MG, Shekhawat GS, et al. Probing Strain-Induced Band Gap Modulation in 2D Hybrid Organic–Inorganic Perovskites. ACS Energy Letters. 2019;4(3):796-802.

9:40am **2D+EM+MI+MN+NS+QS-WeM-6 Collective Electronic States of Epitaxial Monolayer 1T-$NbSe_2$***, **Zhuozhi Ge**,* University of Wisconsin*; H Zhang, L Liu, C Yan,* West Virginia University*; M Weinert,* University of Wisconsin*; L Li,* West Virginia University

At the single layer limit, transition metal dichalcogenides (TMDs) can adopt two different structural variants depending on the anionic environment around the metal ions: the anions are arranged in trigonal prismatic fashion in the 1H polymorph, whereas in 1T the arrangement is octahedral. While bulk 1T $NbSe_2$ doesn't exist in nature, here we show that single layer 1T $NbSe_2$ polymorph can be grown by molecular beam epitaxy on epitaxial graphene/SiC(0001) substrates. A (Ö13xÖ13) Star-of-David charge density waves is observed by *in situ* scanning tunnelling microscopy, which persists above room temperature. A gap of 0.50 eV is further observed by tunnelling spectroscopy and angle resolved photoemission spectroscopy, indicating that this monolayer 1T phase of $NbSe_2$ is also a Mott insulator, similar to that of bulk 1T TaS_2. Our findings indicate that the presence of epitaxial constraints can generate structural configurations that are prohibited in fully-bonded TMD crystals. These findings and their implication on the collective electronic states of single layer 1T-$NbSe_2$ will be discussed at the meeting.

11:00am **2D+EM+MI+MN+NS+QS-WeM-10 Magnetic Interfaces of MnSe2 Monolayer***, **Tomas Rojas**, S Ulloa,* Ohio University

Until recently, 2D magnetism was thought to occur together with defects or doping on different substrates. This situation changed drastically, as intrinsic Cr-based ferromagnetic monolayer materials were discovered, namely CrI3 and Cr2Ge2Te6. A different material, MnSe2, was predicted as stable ferromagnetic monolayer by first-principles calculations, and it has been successfully grown on several substrates. In this study, the authors confirm the intrinsic ferromagnetism of the monolayer, while for thicker

samples they report an interface of the MnSe2 monolayer with bulk α-MnSe(111). This phase of the material is non-magnetic, and yet the observed magnetic moments are of up to twice the value of those in the monolayer alone. In this work, we present a detailed analysis of the interactions at this interface between the two phases, using the Heyd-Scuseria-Ernzerhof hybrid functional. We have studied the effects on the electronic and magnetic structure of both phases of the material, and the dependence on the sample thickness. We study the role that strain plays at the interface, and how it affects the magnetic moments of the structure.

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11:20am **2D+EM+MI+MN+NS+QS-WeM-11 Orbital Design of Topological Insulators from Two-dimensional Semiconductors***, **Shixuan Du,*** Institute of Physics, Chinese Academy of Sciences, China

Two-dimensional (2D) materials have attracted much recent attention because they exhibit various distinct intrinsic properties/functionalities, which are, however, usually not interchangeable. Interestingly, here we propose a generic approach to convert 2D semiconductors, which are amply abundant, to 2D topological insulators (TIs), which are less available, via selective atomic adsorption and strain engineering. The approach is underlined by an orbital design principle that involves introducing an extrinsic -orbital state into the intrinsic -bands of a 2D semiconductor, so as to induce - band inversion for a TI phase, as demonstrated by tight-binding model analyses. Remarkably, based on first-principles calculations, we apply this approach to convert the semiconducting monolayer CuS and CuTe into a TI by adsorbing Na and K respectively with a proper -level energy, and CuSe into a TI by adsorbing a mixture of Na and K with a tuned -level energy or by adsorbing either Na or K on a strained CuSe with a tuned -level valence band edge. Our findings open a new door to the discovery of TIs by a predictive materials design, beyond finding a preexisting 2D TI.

This work is in collaboration with Lei Gao and Jia-Tao Sun from IoP-CAS, Gurjyot Sethi and Feng Liu from University of Utah, Yu-Yang Zhang from UCAS.

11:40am **2D+EM+MI+MN+NS+QS-WeM-12 Rotationally Controlled van der Waals Heterostructures of 2D Materials***, **Emanuel Tutuc**, K Kim, G Burg, H Movva,* The University of Texas at Austin **INVITED**

Heterostructures of atomic layers such as graphene, hexagonal boron-nitride, and transition metal dichalcogenides (TMDs) can serve as testbed for novel quantum phenomena in two-dimensions, and potential device applications. A key ingredient that can add a new dimension to the atomic layer heterostructures palette is the rotational control, and alignment of different two-dimensional (2D) layers. We review here an experimental technique that enables rotationally controlled heterostructures with accurate alignment of the individual layer crystal axes [1]. We illustrate the applicability of this technique to the rotationally aligned double layers of graphene [2], or TMDs [3] separated by a tunnel barrier which display resonant, energy- and momentum-conserving tunneling in vertical transport, consistent with theoretical expectations. When two 2D layers are overlaid with a relative twist, the resulting heterostructure shows a new type of periodicity associated with the moiré superlattice, which are only beginning to be systematically investigated as platform for strongly correlated electron physics. We discuss the electron transport in tunable moiré patterns realized in twisted bilayer [4], and double bilayer graphene heterostructures.

Work done in collaboration with S. K. Banerjee, L. F, Register, B. J. LeRoy, A. H. MacDonald, T. Taniguchi, and K. Watanabe.

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

[2] G. W. Burg *et al*., *Nano Lett.* **17**, 3919 (2017); G. W. Burg *et al*., *Phys. Rev. Lett.* **120**, 177702 (2018).

[3] K. Kim *et al*., *Nano Lett.* **18**, 5967 (2018).

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

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