Monday Morning, January 16, 2023

PCSI
Room Redondo - Session PCSI-MoM1
2D and VdW Materials

Moderator: Anthony Rice, National Renewable Energy Laboratory

8:30am **PCSI-MoM1-1 Step-Edge Nucleation and Domain Orientation Control in Epitaxy of Transition Metal Dichalcogenides on Sapphire***, H. Zhu,* The Pennsylvania State University*; T. Choudhury,* The Pennsylvania State University, India*; N. Nayir,* The Pennsylvania State University, Turkey*; T. Mc Knight, N. Trainor, A. van Duin, **Joan Redwing**,* The Pennsylvania State University **INVITED**

Wafer-scale synthesis of semiconducting transition metal dichalcogenide (TMDs) monolayers is of significant interest for device applications to circumvent size limitations associated with the use of exfoliated flakes. Promising results have been demonstrated for epitaxial films deposited by vapor phase techniques such as CVD and MOCVD. However, the three-fold symmetry of TMDs such as MoS_2 and WSe_2, results in two energetically equivalent domain alignments, often referred to as 0°and 60° domains, when grown on flat high symmetry substrates such as c-plane sapphire. The oppositely oriented domains give rise to inversion domain boundaries (IDBs) upon coalescence which exhibit a metallic character and are generally undesirable. In this study, we demonstrate the epitaxial growth of unidirectional TMD monolayers on 2" diameter c-plane sapphire substrates with a significantly reduced density of inversion domains. Steps on the sapphire surface are shown to play a key role in TMD nucleation and impart a preferred orientation to the domains depending on the step edge structure and chemistry.

Metalorganic chemical vapor deposition (MOCVD) was used for the epitaxial growth of WSe_2 monolayers on c-plane sapphire, miscut ~0.2^o toward the m-axis, in a cold-wall horizontal quartz-tube reactor. A three-step nucleation-ripening-lateral growth process, carried out at temperatures ranging from 850^{o}C to 1000^{o}C, was used to achieve epitaxial films using $W(CO)_6$ and H_2Se as precursors in a H_2 carrier gas. Density functional theory calculations demonstrate that the extent of Se passivation of the sapphire surface and the presence of oxygen remnants near the step edge are critical factors in determining the location of TMD nucleation on the step edge and the subsequent domain orientation relative to the underlying sapphire. By changing the reactor pressure, which modifies the sapphire surface via changes in the chemical potential of H and Se, the site of WSe_2 nucleation can be modified which switches the preferred orientation of the domains. Fully coalesced TMD monolayers are obtained with a reduced density of inversion domain boundaries (<15% areal coverage).The results demonstrate the important role of step structure and chemistry in nucleation and epitaxial growth of TMD monolayers.

9:10am **PCSI-MoM1-9 Effects of Strain and Local Topography on Electromechanical Coupling in Monolayer Transition Metal Dichalcogenides***, **Claire Ganski**, A. De Palma, E. Yu,* The University of Texas at Austin

Two-dimensional transition metal dichalcogenides (2D TMDs) have been extensively studied in recent years due to their remarkable electronic, optical, and mechanical properties. Their ability to withstand high levels of strain without fracture makes 2D TMDs attractive candidate materials for devices such as quantum emitters and energy harvesters. However, the high sensitivity of 2D material properties to nanoscale morphology and strain can just as easily interfere with as aid in optimal device performance if not fully understood and accounted for. In this work, we use piezoresponse force microscopy (PFM) to demonstrate that nanobubbles present in exfoliated MoS_2 monolayers significantly alter the electromechanical behavior of these samples and exhibit strong out-of-plane PFM responses due to flexoelectricity. Small bubbles with diameters under 100 nm consistently exhibit enhanced piezoresponse compared to flat regions. Without filtering images for these ubiquitous features, effective piezoelectric coefficients based on PFM of exfoliated monolayer MoS_2 will be systematically overestimated. Large bubbles with diameters on the order of hundreds of nanometers present an even more remarkable pattern. Rather than increasing monotonically from perimeter to apex, the piezoresponse amplitude of large bubbles reaches its peak at the perimeter and subsequently decreases toward a local minimum at the bubble apex. This profile can be correlated with the curvature of the bubble topography, which is related to the local strain gradient. These correlations suggest a link between local electromechanical properties and strain gradients present in these regions. We argue that the large strain gradients at the bubble edges induce a local reduction in spatial symmetry, which produces out-of-plane polarizations via the flexoelectric effect. On subsequent characterization using PFM, these regions effectively behave not as a 2H MoS_2 monolayer, but as a low-symmetry material exhibiting out-of-plane piezoelectricity.

9:15am **PCSI-MoM1-10 Probing Edge State Conductance in Ultra-Thin Topological Insulator Films***, A. Leis, **Jonathan Karl Hofmann**, M. Schleenvoigt, K. Moors, H. Soltner, V. Cherepanov, P. Schüffelgen, G. Mussler, D. Grützmacher, B. Voigtländer, F. Lüpke, F. Tautz,* Forschungszentrum Juelich GmbH, Germany

Quantum spin Hall (QSH) insulators have unique electronic properties, comprising a band gap in their 2D interior and 1D spin-polarized edge states in which current flows ballistically. In scanning tunneling microscopy (STM), the edge states manifest themselves as an enhanced local density of states (LDOS). However, there is a significant research gap between the observation of edge states in nanoscale spectroscopy, and the detection of ballistic transport in edge channels which typically relies on transport experiments with microscale lithographic contacts. Here, few-layer films of the 3D topological insulator (BixSb1−x)2Te3 are studied [1], for which a topological transition to a 2D topological QSH insulator phase has been proposed. The samples were grown on silicon-on-insulator substrates by MBE [1]. After the growth, a vacuum transfer into the room temperature 4-tip STM was carried out. Indeed, an edge state in the LDOS is observed within the band gap. Yet, in nanoscale transport experiments with a four-tip STM, two-quintuple-layer (QL) films do not exhibit a ballistic conductance in the edge channels, as the measured 4-point resistance is fully in accord with finite element calculations using the measured terrace conductivities [2]. Thus, no QSH edge states are present at the 2 QL films. This demonstrates, that the detection of edge states in spectroscopy can be misleading with regard to the identification of a QSH phase. In contrast, nanoscale multi-tip transport experiments are a robust method for effectively pinpointing ballistic edge channels, as opposed to trivial edge states, in quantum materials.

[1] A. Leis et al., Adv. Quantum Technol. 4, 2100083 (2021)
[2] A. Leis et al., Adv. Quantum Technol. 5, 2200043 (2022)

9:20am **PCSI-MoM1-11 MBE Growth of Transition-Metal Dichalcogenides***, **Wojciech Pacuski**,* University of Warsaw, Poland **INVITED**

Monolayer transition-metal dichalcogenides (TMDs) are two-dimensional materials with exceptional optical properties such as high oscillator strength, valley-related excitonic physics, efficient photoluminescence, and several narrow excitonic resonances. Long time the above effects have been explored for structures produced by techniques involving mechanical exfoliation or post-growth transfer, and more recently, also encapsulation in hBN, inevitably inducing considerable large-scale inhomogeneity. On the other hand, techniques which are essentially free from this disadvantage, such as molecular beam epitaxy (MBE), have yielded only structures characterized by considerable spectral broadening, which hinders most of the interesting optical effects. In this talk, I will present the MBE-grown TMD ($MoSe_2$) exhibiting narrow and fully resolved spectral lines of neutral and charged exciton [1]. Moreover, our monolayers exhibit unprecedented high spatial homogeneity of optical properties, with variation of the exciton energy as small as 0.16 meV over a distance of tens of micrometers. Importantly, good optical properties are achieved for as-grown samples, without any post-growth exfoliation and encapsulation in hBN. Our best recipe for MBE growth includes an extremely slow growth rate, the annealing at very high temperatures, and the use of atomically flat hBN substrate in the form of flakes exfoliated from bulk. Moreover, comparable results we also obtained using an hBN buffer that we grow by MOCVD on 2" Al_2O_3 wafers [2]. Our optical characterization includes Raman scattering, second-harmonic generation, and low-temperature photoluminescencere. Thanks to sharp and intense exciton lines, we were able to perform also magneto-spectroscopy and time resolved spectroscopy [3], which reveal subtle differences between 2D epilayers and mechanically exfoliated materials. We compare structural and optical properties of $MoSe_2$ grown on hBN to properties of various TMDs ($MoTe_2$ [4,5], $NiTe_2$ [6], WSe_2, VSe_2) grown on various substrates (2D, 3D, polycrystalline). This reveals particularly high diffusion parameters of transition metals on hBN [5], the role of distribution of orientation of TMD grain domains, the tendency to merge grains, form bilayers and 1D or 3D structures.

[1] W. Pacuski et al., Nano Letters 20, 3058 (2020).

[2] K. Ludwiczak et al., ACS Appl. Mater. Interfaces 13, 47904 (2021).

[3] K. Oreszczuk et al., in preparation (2022) .

[4] Z. Ogorzałek et al., Nanoscale 12, 16535 (2020).

[5] B. Seredyński et al., J. Cryst. Growth 596, 126806 (2022).

[6] B. Seredyński et al., Cryst. Growth Des. 21, 5773 (2021).

Author Index

Bold page numbers indicate presenter