MIP Products

2DCC publications are categorized based on four science drivers listed below. For publications that cross multiple science drivers, the publication is listed in the topic area of primary influence and any secondary drivers are listed at the end of its description using the abbreviation for the driver. Publications for each primary science driver are organized into subsections by publications led by ***external user***, ***local user***, and ***in-house***.

Also noted for each publication with a user are instances where co-authors are from non-R1 or minority-serving institutions or government labs. User project ID# is noted which is also cross-referenced in the MIP User Projects document with the DOI. For user projects, sample-only projects are denoted by "S" in the project number, research projects are denoted by "R" in the project number, and RSVP projects are denoted by "V".

In the authors lists, **bold face** is used to designate senior 2DCC faculty (core faculty and research professors that are professional staff in the facility), *italic* is used to designate senior local users not participating in in-house research, and <u>underline</u> is used to designate senior external users.

Physics of 2D Systems (Phys2D)

This science driver focuses on providing enabling materials synthesis, characterization, and modeling capabilities to facilitate fundamental studies of new fundamental physical processes that occur in 2D systems, such as efficient spin-charge conversion and the quantum anomalous Hall effect in topological insulators, valleytronics in transition metal dichalcogenides, and quantum transport in 2D heterostructures.

Epitaxy of 2D Chalcogenides (Epi2DC)

This science driver seeks to understand fundamental mechanisms of 2D film formation in van der Waals bonded systems including the role of the substrate in nucleation and epitaxy, self-limited growth of monolayers, epitaxy in 2D heterostructures, miscibility and alloy formation, intentional doping, and native defects.

Next Generation Devices (NGDev)

This science driver focuses on providing enabling materials synthesis and characterization capabilities to facilitate development of next-generation electronics (2D tunnel transistors, thin film transistors for flexible electronics, etc.) and optoelectronics (2D photodetectors, integrated photonics, single photon sources, etc.).

Advanced Characterization and Modeling (AdvCM)

This science driver focuses on developing techniques and tools to both probe and model 2D chalcogenide films in situ to study the evolution of surface morphology, lateral and vertical domain growth, growth-related defects and grain boundaries, electronic band structure, carrier transport, closely integrated with theory and simulation that targets key kinetic processes during growth, enables new insights on in situ characterization, and accelerates the process of identifying compelling synthetic targets and overcoming experimental obstacles to their synthesis.

Physics of 2D Systems (Phys2D)

External User Publications (Phys2D)

H. Zhang, C.Q. Xu, **S.H. Lee**, **Z. Q. Mao**, X. Ke, "Thermal and Thermoelectric Properties of an Antiferromagnetic Topological Insulator $MnBi_2Te_4$," *Physical Review B* 105, 184411 (2022). 10.1103/PhysRevB.105.184411.
In this paper, electronic, thermal, and thermoelectric transport studies of $MnBi_2Te_4$ are reported The temperature and magnetic field dependence of its resistivity, thermal conductivity, and Seebeck coefficient indicate strong coupling between charge, lattice, and spin degrees of freedom in this system. Furthermore, $MnBi_2Te_4$ exhibits a large anomalous Nernst signal, which is associated with nonzero Berry curvature of the field-induced canted antiferromagnetic state. Bulk Crystal are grown using a 2DCC muffle furnace.

- External User Project S0074

Y. Lin, M. Huber, S. Rajpurohit, Y. Zhu, K.M. Siddiqui, D.H. Eilbott, L. Moreschini, P. Ai, J.D. Denlinger, **Z. Mao**, L.Z. Tan, A. Lanzara, "Evidence of Nested Quasi-one-dimensional Fermi Surface and Decoupled Charge-lattice Orders in Layered $TaTe_2$," *Physical Review Research* 4 (2), L022009 (2022). 10.1103/PhysRevResearch.4.L022009.
Results suggest that TaTe2 manifests intrinsic mixed dimensionality between its electronic and lattice structure and that the CDW-like phase transition is likely governed by multiple mechanisms. This work provides routes for forging unconventional CDW phases and charge-lattice entanglement that would otherwise not be available in materials with fixed dimensionality.

- External User Project S0076 (National Lab)

L.J. Riddiford, A.J. Grutter, T. Pillsbury, M. Stanley, **D.R. Hickey**, P. Li, **N. Alem**, **N. Samarth**, Y. Suzuki, "Understanding Signatures of Emergent Magnetism in Topological Insulator/Ferrite Bilayers," *Physical Review Letters* 128 (12), 126802 (2022). 10.1103/PhysRevLett.128.126802.

Magnetic insulator-topological insulator heterostructures have been studied in search of chiral edge states via proximity induced magnetism in the topological insulator, but these states have been elusive. This study identified $MgAl_{0.5}Fe_{1.5}O_4$=Bi_2Se_3 bilayers for a possible magnetic proximity effect. The results provide a strategy via correlation of microstructure with magnetic data to confirm a magnetic proximity effect. Materials were grown using 2DCC equipment MBE1.

- External User Project S0030

Z. Pan, **S.H. Lee**, K. Wang, **Z. Mao**, D. Li, "Elastic stiffening induces one-dimensional phonons in thin Ta_2Se_3 nanowires," *Applied Physics Letters* 120, 062201 (2022). 10.1063/5.0083980.

Demonstration of Ta_2Se_3 exfoliation into nanowires that indicates strong anisotropy in the bonding strength within the basal place. Systematic thermal property measurements disclose signatures of one-dimensional phonons as the nanowire hydraulic diameter reduces below 19.2 nm with linearly escalating thermal conductivity as temperature increases and size dependence inconsistent with the classical size effect. The nanowires of Ta_2Se_3 used for this study were obtained from microexfoliation of bulk Ta_2Se_3 crystals grown using a chemical vapor transport method at the 2DCC Bulk Growth facility

- External User Project S0061

C. Yan, E. Green, R. Fukumori, N. Protic, **S.H. Lee**, S. Fernandez-Mulligan, R. Raja, R. Erdakos, **Z. Mao**, S. Yang, "An integrated quantum material testbed with multi-resolution photoemission spectroscopy," *Review of Scientific Instruments* 92 (11), 113907 (2021). 10.1063/5.0072979.

Development of a novel quantum material testbed using $FeSe/SrTiO_3$ thin films and bulk $MnBi_4Te_7$ magnetic topological insulators (Tis). The 2DCC grew the magnetic TIs for the user to use in the testbed.

- External User Project S0085

K.M. Siddiqui, D.B. Durham, F. Cropp, C. Ophus, S. Rajpurohit, Y. Zhu, J.D. Carlstrom, C. Stravrakas, **Z. Mao**, A. Raja, P. Musumeci, L.Z. Tan, A.M. Minor, D. Filippetto, R.A. Kaindl, "Ultrafast optical melting of trimer superstructure in layered 1T'-$TaTe_2$," *Communications Physics* 4 (1), 152 (2021). 10.1038/s42005-021-00650-z.

Quasi-two-dimensional transition-metal dichalcogenides are a key platform for exploring emergent nanoscale phenomena arising from complex interactions. Access to the underlying degrees-of-freedom on their natural time scales motivates the use of advanced ultrafast probes sensitive to self-organized atomic-scale patterns. This study reported the ultrafast investigation of $TaTe_2$, which exhibits unique charge and lattice trimer order characterized by a transition upon cooling from stripe-like chains into a (3×3) superstructure of trimer clusters. The work paves the way for further exploration and ultimately rapid optical and electronic manipulation of trimer superstructures. The 2DCC synthesized bulk materials used in this study using non-MIP equipment.

- External User Project S0076 (National Lab)

Y. Tian, Y. Zhu, R. Li, **Z. Mao**, J.H. Ross, "NMR determination of Van Hove singularity and Lifshitz transitions in the nodal-line semimetal ZrSiTe," *Physical Review B* 104 (4), L041105 (2021). PhysRevB.104.L041105.

This study focuses on the Dirac semimetal ZrSiTe. Low-T behavior is dominated by a symmetry-protected nodal line, with NMR providing a sensitive probe of the diamagnetic response of the associate carriers. A Van Hove singularity is identified that is closely connected to this nodal line, and an associated T -induced Lifshitz transition. A disconnect in the NMR shift and linewidth at this temperature indicates the change in electronic behavior associated with this topological change. These features have an orientation-dependent behavior indicating a field-dependent scaling of the associated band energies. The 2DCC synthesized bulk crystals for this study using non-MIP equipment.

- External User Project S0078

C. Yan, S. Fernandez-Mulligan, R. Mei, **S.H. Lee**, N. Protic, R. Fukumori, B. Yan, **C.-X. Liu**, **Z. Mao**, S. Yang, "Origins of electronic bands in the antiferromagnetic topological insulator $MnBi_2Te_4$," *Physical Review B* 104 (4), L041102 (2021). 10.1103/PhysRevB.104.L041102.

Despite the rapid progress in understanding the first intrinsic magnetic topological insulator $MnBi_2Te_4$, its electronic structure remains a topic under debate. This study performs a thorough spectroscopic investigation into the electronic structure of $MnBi_2Te_4$ via laser-based angle-resolved photoemission spectroscopy. The results represent a solid step forward in reconciling the existing controversies in the electronic structure of $MnBi_2Te_4$ and provides an important framework to understand the electronic structures of other relevant topological materials $MnBi_{2n}Te_{3n+1}$.The 2DCC synthesized bulk crystals for this study using MIP equipment. 2DCC theory personnel also contributed to the development of the theoretical model.

- External User Project S0085
- Also science driver AdvCM

A.S. McLeod, A. Wieteska, G. Chiriaco, B. Foutty, Y. Wang, Y. Yuan, F. Xue, **V. Gopalan**, L.-Q. Chen, **Z.Q. Mao**, A.J. Millis, A.N. Pasupathy, D.N. Basov, "Nano-imaging of a strain-tuned stripe textures in a Mott crystal," *npj Quantum Materials* 6, 46 (2021). 10.1038/s41535-021-00339-0.

This work reveals a spontaneous striped texture of coexisting insulating and metallic domains in single crystals of the quasi-2D, bilayer ruthenate $Ca_3(Ti_xRu_{1-x})_2O_7$ across its first-order Mott transition at T≈95K through multi-messenger low-temperature nano-imaging. The sample used in this study was synthesized by the 2DCC researchers using non-MIP equipment.

- External User Project S0059

L. Yang, Y. Tao, Y. Zhu, M. Akter, K. Wang, Z. Pan, Y. Zhao, Q. Zhang, Y.-Q. Xu, R. Chen, T.T. Xu, Y. Chen, **Z. Mao**, D. Li, "Observation of superdiffusive phonon transport in aligned atomic chains," *Nature Nanotechnology* 16, 764-768 (2021). 10.1038/s41565-021-00884-6.

This work reports the experimental observation of divergence of thermal conductivity (κ) at room temperature in ultrathin van der Waals crystal $NbSe_3$ nanowires. The κ of $NbSe_3$ nanowires was also found to follow a 1/3 power law with wire length, consistent with the superdiffusive phonon transport model. These results not only demonstrate the divergent trend of the observed thermal conductivity with sample length in 1-D atomic chain system, but also unveil a possible way of creating novel 1-D van der Waals crystal-based thermal superconductors with exceptionally high κ values. The 2DCC researchers not only synthesized high-quality $NbSe_3$ single crystals using MIP equipment for this work, but also demonstrated $NbSe_3$ nanowires are stable in air using transmission electron microscopy. These combined synthesis and characterization efforts at the 2DCC enable this achievement.

- External User Projects S0049 and S0061

P. Li, J. Ding, S.-L. Zhang, J. Kally, T. Pillsbury, O.G. Heinonen, G. Rimal, C. Bi, A. DeMann, S.B. Field, W. Wang, J. Tang, J.S. Jiang, A. Hoffmann, **N. Samarth**, M. Wu, "Topological Hall Effect in a Topological Insulator Interfaced with a Magnetic Insulator," *Nano Letters* 21 (1), 84-90 (2021). 10.1021/acs.nanolett.0c03195.

In this paper, we used the 2DCC MBE facility to grow thin films of a topological insulator (TI) (Bi_2Se_3) on magnetic insulator (MI) substrates ($BaFe_{12}O_{19}$) provided by the user (Professor Wu, Colorado State). Measurements of the Hall effect made by the user revealed evidence of a genuine topological effect in the temperature range of T = 2–3 K and an anomalous Hall effect at T = 80–300 K. Over T = 3–80 K, the two effects coexist but show opposite temperature dependencies. Control measurements, calculations, and simulations together suggest that the observed topological Hall effect originates from skyrmions formed due to a Dzyaloshinskii–Moriya interaction at the interface. The strength of this interaction based on fitting the data is estimated to be substantially higher than that in the more extensively studied skyrmion systems derived from heavy metal-based systems. The 2DCC synthesized materials for this study using the MIP MBE1 system.

- External User Project S0057

H. Zhang, Y. L. Zhu, Y. Qiu, W. Tian, H. B. Cao, **Z. Q. Mao**, X. Ke, "Field-induced magnetic phase transitions and the resultant giant anomalous Hall effect in the antiferromagentic half-Heusler compound DyPtBi," *Physical Review B* 102 (9), 094424 (2020). 10.1103/PhysRevB.102.094424.

Systematic Neutron scattering and transport studies of a half-Heusler compound DyPtBi were performed using single crystals grown by the 2DCC Bulk Growth facility. This study shows that DyPtBi hosts a delicate balance between two different magnetic ground states, which can be controlled by moderate magnetic fields. One of the magnetic states hosts a giant anomalous Hall effect. These results indicate that DyPtBi is a potential material for realizing the anomalous Hall effect in an antiferromagnet with a face-centered-cubic lattice.

- External User Project S0074

Z. Pan, L. Yang, Y. Tao, Y. Zhu, Y-Q. Xu, **Z. Mao**, D. Li, "Net negative contributions of free electrons to the thermal conductivity of NbSe3 nanowires," *Physical Chemistry Chemical Physics* 22, 21131-21138 (2020). 10.1039/D0CP03484C.

This paper reports comprehensive experimental studies of the thermal transport properties of $NbSe_3$ nanowires, exfoliated from the bulk $NbSe_3$ crystals grown by the 2DCC Bulk Growth facility. This work reveals that the electron-phonon scattering in the $NbSe_3$ nanowire is enhanced as the free electrons are condensed during the charge density wave transition, thus resulting in the decrease of overall thermal conductivity. This result not only reveals a net negative contribution of the free electrons due to the escalated electron-phonon scattering, but also provides insight into the competing roles of free electrons, which could lead to unexpected trends in thermal conductivity.

- External User Project Collaboration between S0061 and S0049

A. Rossi, G. Resta, **S.H. Lee**, **R.D. Redwing**, C. Jozwiak, A. Bostwick, E. Rotenberg, S.Y. Savrasov, I.M. Vishik, "Two phase transitions driven by surface electron doping in WTe_2," *Physical Review B* 102, 121110(R) (2020). 10.1103/PhysRevB.102.121110.
This work identified phase transitions that occur in WTe_2 as a result of electron doping with potassium. A postdoctoral scholar from UC-Davis (external user) received training from 2DCC personnel in the Bulk Growth Facility on CVT synthesis and worked on-site to prepare their samples. The external user also used nano-ARPES equipment at LBNL (national lab) to characterize the surface electronic structure of the samples.

- External User Project R0017

T. Liu, J. Kally, T. Pillsbury, C. Liu, H. Chang, J. Ding, Y. Cheng, M. Hilse, **R. Engel-Herbert**, **A. Richardella**, **N. Samarth**, M. Wu, "Changes of Magnetism in a Magnetic Insulator due to

Proximity to a Topological Insulator," *Physical Review Letters* 125, 017204 (2020). 10.1103/PhysRevLett.125.017204.

Thin films of the topological insulator Bi2Se3 were grown by MBE on a magnetic insulator $Y_3Fe_5O_{12}$ thin film. Ferromagnetic resonance measurements show that the topological surface state in Bi_2Se_3 produces a perpendicular magnetic anisotropy, results in a decrease in the gyromagnetic ratio, and enhances the damping in $Y_3Fe_5O_{12}$. These topological surface state-induced changes become more pronounced as the temperature decreases from 300 to 50 K. These results suggest a completely new approach for control of magnetism in magnetic thin films. Control measurements using $(Bi,In)_2Se_3$, a trivial insulator rule out possible artifacts.

- External User Project S0057

Y. Shao, A.N. Rudenko, J. Hu, Z. Sun, Y. Zhu, S. Moon, A.J. Millis, S. Yuan, A.I. Lichtenstein, D. Smirnov, **Z.Q. Mao**, M.I. Katsnelson, D.N. Basov, "Electronic correlations in nodal-line semimetals," *Nature Physics* 16, 636-641 (2020). 10.1038/s41567-020-0859-z.

Spectroscopic hallmarks of electronic correlations (i.e. strong reduction of the Drude weight and the Fermi velocity) are observed in a topological nodal-line semimetal ZrSiSe. This work establishes the first platform to explore correlation of relativistic fermions in low dimension. Some of the crystals used in this work were grown using the 2DCC Bulk Growth facility.

- External User Project S0082

S.M. Oliver, J.J. Fox, A. Hashemi, A. Singh, R.L. Cavalero, S. Yee, **D.W. Snyder**, R. Jaramillo, H.-P. Komsa, P.M. Vora, "Phonons and excitons in $ZrSe_2$-ZrS_2 alloys," *Journal of Materials Chemistry C* (2020) in press. 10.1039/D0TC00731E.

A comprehensive analysis of photons and excitons in $Z(S,Se)_2$ alloy crystals (synthesized in the 2DCC Bulk Growth facility) was carried out using Raman spectroscopy and spectroscopic ellipsometry. The Raman spectrum was found to be dominated by nominally IR phonons due to the large ionicity of bonding.

- External User Projects R0014 and R0016

A. Gangshettiwar, Y. Zhu, Z. Jiang, J. Peng, Y. Wang, J. He, J. Zhou, **Z. Mao**, K. Lai, "Emergence of a competing stripe phase near the Mott transition in Ti-doped bilayer calcium ruthenates," *Physical Review B*, 101(20), 201106(R) (2020). 10.1103/PhysRevB.101.201106.

This work reveals a new exotic phenomenon of correlated electrons: a competing stripe phase at a Mott transition. The sample used in this study was synthesized using non-MIP equipment.

- External User Project S0060

F. Lupke, D. Waters, S.C. de la Barrera, M. Widom, D.G. Mandrus, J.Q. Yan, R.M. Feenstra, B.M. Hunt, "Proximity-induced superconducting gap in the quantum spin Hall edge state of monolayer WTe_2," *Nature Physics* 16, 526-530 (2020). 10.1038/s41567-020-0816-x.

This study used scanning tunneling spectroscopy of 2DCC-grown WTe_2 monolayer samples in contact with $NbSe_2$ to study proximity-induced superconductivity in the quantum spin Hall phase. This is an important advance toward establishing a 1D topological superconductor and Majorana zero modes in condensed matter.

- Also science driver NGDev
- External User Project S0027

P. Li, J. Kally, S.-L. Zhang, T. Pillsbury, J. Ding, G. Csaba, J. Ding, J.S. Jiang, Y. Liu, R. Sinclair, C. Bi, A. DeMann, G. Rimal, W. Zhang, S.B. Field, J. Tang, W. Wang, O.G. Heinonen, V. Novosad, A. Hoffman, **N. Samarth**, M. Wu, "Magnetization switching using topological surface states," *Science Advances*, 5 (8), eaaw3415 (2019). 10.1126/sciadv.aaw3415

This project used 2DCC MBE-grown topological insulator/ferromagnet insulator bilayers ($Bi_2Se_3/BaFe_{12}O_{19}$) to fabricate spintronic devices that showed current-induced magnetization switching. The pronounced increase in switching efficiency at cryogenic temperatures led to the conclusion that this process is dominated by the spin-momentum locking of topological surface states that have enhanced surface conductivity at low temperatures where bulk conductivity freezes out.

- Also science driver NGDev
- External User Project S0025

Local User Publications (Phys2D)

H. Padmanabhan, M. Poore, P.K. Kim, N.Z. Koocher, **V.A. Stoica**, D. Puggioni, H. Wang, X. Shen, A.H. Reid, M. Gu, M. Wetherington, **S.H. Lee**, R.D. Schaller, **Z. Mao**, A.M. Lindenberg, X. Wang, J.M. Rondinelli, R.D. Averitt, *V. Gopalan*, "Interlayer magnetophononic coupling in $MnBi_2Te_4$," *Nature Communications* 13, 1929 (2022). 10.1038/s41467-022-29545-5.
This study presents evidence for interlayer magnetophononic coupling in the layered magnetic topological insulator $MnBi_2Te_4$. Anomalies in phonon scattering intensities across magnetic field-driven phase transitions are observed, despite the absence of discernible static structural changes. This behavior is a consequence of a magnetophononic wave-mixing process that allows for the excitation of zone-boundary phonons that are otherwise 'forbidden' by momentum conservation. The microscopic model based on density functional theory calculations reveals that this phenomenon can be attributed to phonons modulating the interlayer exchange coupling. In light of the intimate connection between magnetism and topology in $MnBi_2Te_4$, the magnetophononic coupling represents an important step towards coherent on-demand manipulation of magnetic topological phases. Materials grown in this project done with 2DCC muffle furnace equipment.

- Local user project S0062 collaboration with in-house research
- Also science driver AC&M

In-house Research Publications (Phys2D)

J. He, **S.H. Lee**, F. Naccarato, G. Brunin, R. Zu, **Y. Wang**, L. Miao, H. Wang, **N. Alem**, G. Hautier, G.-M. Rignanese, **Z. Mao**, **V. Gopalan**, "SnP_2S_6: A Promising Infrared Nonlinear Optical Crystal with Strong Nonresonant Second Harmonic Generation and Phase-Matchability," *ACS Photonics* 9 (5), 1724-1732 (2022). 10.1021/acsphotonics.2c00131.
High-power infrared laser systems with broad-band tunability are of great importance due to their wide range of applications in spectroscopy and free-space communications. These systems require nonlinear optical (NLO) crystals for wavelength up/down conversion using sum/difference frequency generation, respectively. NLO crystals need to satisfy many competing criteria, including large nonlinear optical susceptibility, large laser-induced damage threshold (LIDT), wide transparency range, and phase-matchability. This study reveals that SnP_2S_6 is an outstanding candidate. Bulk crystals of SnP_2S_6 were grown by 2DCC CVT equipment.

Y. Lv, J. Kally, T. Liu, P. Quarterman, T. Pillsbury, B.J. Kirby, A.J. Grutter, P. Sahu, J.A. Borchers, M. Wu, **N. Samarth**, J.-P. Wang, "Large unidirectional spin Hall and Rashba-Edelstein magnetoresistance in topological insulator/magnetic insulator heterostructures," *Applied Physics Reviews* 9 (1), 011406 (2022). 10.1063/5.0073976.
The unidirectional spin Hall and Rashba−Edelstein magnetoresistance is of great fundamental and practical interest, particularly in the context of reading magnetization states in two-terminal spin–orbit torque memory and logic devices due to its unique symmetry. Here, we report large unidirectional spin Hall and Rashba−Edelstein magnetoresistance in a new material family—magnetic insulator/topological insulator $Y_3Fe_5O_{12}/Bi_2Se_3$ bilayers. Ee demonstrate a prototype memory device based on a magnetic insulator/topological insulator bilayer, using unidirectional spin Hall and Rashba−Edelstein magnetoresistance for electrical readout of current-induced magnetization switching aided by a small Oersted field. The materials in this study were synthesized by the 2DCC MIP MBE1 system.

- In-house collaboration with external user S0057

W. Yanez, **Y. Ou**, R. Xiao, J. Koo, J.Y. Held, S. Ghosh, J. Rable, T. Pillsbury, E. Gonzlaez Delgado, K. Yang, J. Chamorro, A.J. Grutter, P. Quarterman, **A. Richardella**, A. Sengupta, T. McQueen, J.A. Borchers, K.A. Mkhoyan, B. Yan, **N. Samarth**, "Spin and Charge Interconversion in Dirac-Semimetal Thin Films," *Physical Review Applied* 16 (5), 054031 (2021). 10.1103/PhysRevApplied.16.054031.
This study uses spin torque ferromagnetic resonance and ferromagnetic-resonance-driven spin pumping to detect spin-charge interconversion at room temperature in heterostructure devices that interface an archetypal Dirac semimetal, Cd_3As_2, with a metallic ferromagnet, $Ni_{0.80}Fe_{0.20}$ (permalloy). Angle-resolved photoemission directly reveals the Dirac-semimetal nature of the samples prior to device fabrication and high-resolution transmission electron microscopy is used to characterize the crystalline structure and the relevant heterointerfaces. We find that the spin-charge interconversion efficiency in Cd_3As_2/permalloy heterostructures is comparable to that in heavy metals and that it is enhanced by the presence of an interfacial oxide. Spin torque ferromagnetic resonance measurements reveal an in-plane spin polarization regardless of an oxidized or pristine interface. The 2DCC MIP facility was used for ARPES measurements of the Cd_3As_2 thin films via *in vacuo* transfer within the 2DCC highly integrated vacuum environment (HIVE).

S.H. Lee, D. Graf, L. Min, Y. Zhu, H. Yi, S. Ciocys, **Y. Wang**, E.S. Choi, R. Basnet, A. Fereidouni, **V. Gopalan**, H.O.H. Churchill, A Lanzara, **N. Samarth**, **C.-Z. Chang**, J. Hu, **Z. Mao**, "Evidence for a Magnetic-Field-Induced Ideal Type-II Weyl State in Antiferromagnetic Topological Insulator $Mn(Bi_{1-x}Sb_x)_2Te_4$," *Physical Review X* 11 (3), 031032 (2021). 10.1103/PhysRevX.11.031032.
In this article, we report transport evidence for a TRS-breaking type-II WSM observed in the intrinsic antiferromagnetic topological insulator $Mn(Bi_{1-x}Sb_x)_2Te_4$ under magnetic fields. This state is manifested by the electronic structure transition caused by the spin-flop transition. The transition results in an intrinsic anomalous Hall effect and negative c-axis longitudinal magnetoresistance attributable to the chiral anomaly in the ferromagnetic phases of lightly hole-doped samples. Our results establish a promising platform for exploring the underlying physics of the long-sought, ideal TRS-breaking type-II WSM. The 2DCC synthesized bulk crystals for this study using MIP equipment.

- Also science driver AdvCM

J.Y. Liu, J. Yu, J.L. Ning, H.M. Yi, L. Miao, L.J. Min, Y.F. Zhao, W. Ning, K.A. Lopez, Y.L. Zhu, T. Pillsbury, Y.B. Zhang, **Y. Wang**, J. Hu, H.B. Cao, B.C. Chakoumakos, F. Balakirev, F. Weickert, M. Jaime, Y. Lai, L. Yang, J.W. Sun, **N. Alem**, **V. Gopalan**, **C.Z. Chang**, **N. Samarth**, **C.X. Liu**, R.D. McDonald, **Z.Q. Mao**, "Spin-valley locking and bulk quantum Hall effect in a noncentrosymmetric Dirac semimetal $BaMnSb_2$," *Nature Communications* 12 (1), 4062 (2021). 10.1038/s41467-021-24369-1.
Spin-valley locking in monolayer transition metal dichalcogenides has attracted enormous interest, since it offers potential for valleytronic and optoelectronic applications. Such an exotic electronic state has sparsely been seen in bulk materials. Here, we report spin-valley locking in a Dirac semimetal $BaMnSb_2$. This is revealed by comprehensive studies using first principles calculations, tight-binding and effective model analyses, angle-resolved photoemission spectroscopy measurements. The 2DCC synthesized bulk crystals for this study using MIP equipment and contributed first-principles calculations from 2DCC personnel.

- Also science driver AdvCM

N.P. de Leon, K.M. Itoh, D. Kim, K.H. Mehta, T.E. Northup, H. Pak, B.S. Palmer, **N. Samarth**, S. Sangtawesin, D.W. Steuerman, "Materials challenges and opportunities for quantum computing hardware," *Science* 372 (6539), eabb2823 (2021). 10.1126/science.abb2823.

This is a review paper that provides a sweeping overview of the field of quantum computing from the perspective of a materials scientist, describing the key challenges faced by the various principal platforms for building computing hardware (superconducting Josephson junction, semiconductor quantum dots, single spin defects, ion traps, and topological superconductors). Most of the paper discussed materials beyond the scope of 2DCC, but some materials of relevance to 2DCC that were commented on include topological superconductors, graphene, and 2D van der Waals materials.

- Also science driver NGDev

W. Ge, P.M. Sass, J. Yan, **S.H. Lee**, **Z. Mao**, W. Wu, "Direct Evidence of Ferromagnetism in $MnSb_2Te_4$," *Physical Review B* 103 (13), 134403 (2021). 10.1103/PhysRevB.103.134403.

This work reveals direct evidence of ferromagnetism in $MnSb_2Te_4$ using cryogenic magnetic force microscopy. A part of the materials used in this study was synthesized using the 2DCC bulk growth facility. While $MnSb_2Te_4$ was previously reported to be antiferromagnetic, the 2DCC researchers succeeded in growing the ferromagnetic phase which was not predicted in theory through tuning growth conditions. The FM $MnSb_2Te_4$ offers a new platform to explore new exotic quantum states in 2D magnetic materials. The 2DCC synthesized bulk crystals for this study using MIP equipment.

Y. Li, Z. Wang, R. Xiao, Q. Li, K. Wang, **A. Richardella**, J. Wang, **N. Samarth**, "Capping layer influence and isotropic in-plane upper critical field of the superconductivity at the $FeSe/SrTiO_3$ interface," *Physical Review Materials* 5 (3), 034802 (2021). 10.1103/PhysRevMaterials.5.034802.

In this in house project, we used the 2DCC multimodule MBE system to grow and characterize ultrathin FeSe films on $SrTiO_2$. Understanding the superconductivity at the interface of $FeSe/SrTiO_3$ is a problem of great contemporary interest due to the significant increase in critical temperature (Tc) compared to that of bulk FeSe, as well as the possibility of an unconventional pairing mechanism and topological superconductivity. We studied the influence of a capping layer on superconductivity in thin films of FeSe grown on $SrTiO_3$ using molecular beam epitaxy. We used the 2DCC *in vacuo* four-probe electrical resistance measurement facility (LT-Nanoprobe) and *ex situ* magnetotransport measurements to examine the effect of three capping layers that provide distinct charge transfer into FeSe: insulating FeTe, nonmetallic Te, and metallic Zr. Our results show that FeTe provides an optimal cap that barely influences the inherent Tc found in pristine $FeSe/SrTiO_3$, while the transfer of holes from a nonmetallic Te cap completely suppresses superconductivity and leads to insulating behavior. Finally, we used *ex situ* magnetoresistance measurements in FeTe capped FeSe films to extract the angular dependence of the in-plane upper critical magnetic field. Our observations reveal an almost isotropic in-plane upper critical field, providing insight into the symmetry and pairing mechanism of high-temperature superconductivity in FeSe. The 2DCC synthesized materials in this study using the MIP MBE1 and in vacuo STM.

Y. Zhu, J. Hu, D. Graf, X. Gui, W. Xe, **Z. Mao**, "Quasi-two-dimensional relativistic fermions probed by de Haas-van Aplhen quantum oscillations in $LuSn_2$," *Physical Review B* 103 (12), 125109 (2021). 10.1103/PhysRevB.103.125109.

This work revealed the distorted Sn-square net layer in the layered compound $LuSn_2$ generates relativistic fermions. The two-dimensionality of the relativistic band is found to be significantly enhanced due to the suppressed corrugation of the Sn square net layer as compared to the previously reported topological semimetal YSn_2. These results suggest that the dimensionality of the relativistic band in $RESn_2$ (RE=rare earth) can be tuned by the electronegativity of RE atoms. Some samples used in this study were synthesized using MIP equipment in the 2DCC bulk synthesis facility.

F. Wang, X. Wang, Y.-F. Zhao, D. Xiao, L.-J. Zhou, W. Liu, Z. Zhang, W. Zhao, M.H.W. Chan, **N. Samarth**, **C. Liu**, H. Zhang, **C.-Z. Chang**, "Interface-induced sign reversal of the anomalous Hall effect in magnetic topological insulator heterostructures," *Nature Communications* 12, 79 (2021). 10.1038/s41467-020-20349-z.

In this in house research, we used both a faculty MBE chamber (Chang) and the 2DCC MIP equipment (MBE1) to grow quantum anomalous Hall insulator samples derived from V- and Cr-doped Sb_2Te_3 and $(Bi,Sb)_2Te_3$. These samples were then studied using low temperature magnetotransport measurements to understand the intrinsic anomalous Hall effect in terms of non-zero Berry curvature in momentum space. We find that the sign of the anomalous Hall effect in the magnetic chalcogenide topological insulator layer can be changed from being positive to negative by varying the heterostructure details (e.g. layer thickness). First-principles calculations by 2DCC theorists (Liu) show that the built-in electric fields at heterointerfaces influence the band structure of the magnetically doped layers, and thus lead to a reconstruction of the Berry curvature in the heterostructure samples. This enabled the design and demonstration of an artificial "topological Hall effect"-like feature.

- Also science driver AdvCM

W. Ning, **Z. Mao**, "Recent advancements in the study of intrinsic magnetic topological insulators and magnetic Weyl semimetals," *APL Materials* 8 (9), 090701 (2020). 10.1063/5.0015328.

The breaking of time-reversal symmetry in topological materials has been extensively studied as a platform to generate quantum effects, such as the quantum anomalous Hall effect. In this research review, the recent research progress in magnetic topological materials, including intrinsic magnetic topological insulators and magnetic Weyl semimetals, are briefly overviewed.

S.-W. Wang, D. Xiao, Z. Dou, M. Cao, Y.-F. Zhao, **N. Samarth**, **C.-Z. Chang**, M.R. Connolly, C. G. Smith, "Demonstration of Dissipative Quasihelical Edge Transport in Quantum Anomalous Hall Insulators," *Physical Review Letters* 125 (12), 126801 (2020). 10.1103/PhysRevLett.125.126801.

Thin heterostructure films of magnetically-doped topological insulators (TIs), specifically Cr-doped $(Bi,Sb)_2Te_3$, were grown by molecular beam epitaxy on $SrTiO_3$ substrates. Using an electrostatic back gate, the films could be tuned into the quantum anomalous Hall (QAH) insulator state. Collaborators at the University of Cambridge (UK) then studied the temperature- and magnetic-field-dependence of the magnetoresistance of a magnetic TI sandwich heterostructure device. The measurements demonstrated that the predominant dissipation mechanism in thick QAH insulators can switch between nonchiral edge states and residual bulk states in different magnetic-field regimes. The paper provides a way to distinguish between the dissipation arising from the residual bulk states and nonchiral edge states, which is crucial for achieving true dissipationless transport in QAH insulators and for providing deeper insights into QAH-related phenomena.

K. Nisi, S. Subramanian, W. He, K.A. Ulman, H. El-Sherif, F. Sigger, M. Lassauniere, M.T. Wetherington, N. Briggs, J. Gray, A.W. Holleitner, N. Bassim, S.Y. Quek, **J.A. Robinson**, U. Wurstbauer, "Light-Matter Interaction in Quantum Confined 2D Polar Metals," *Advanced Functional Materials*, 2005977 (2020). 10.1002/adfm.202005977.

In this study, we explore the linear optical response of 2D Ga and 2D In. The fundamental light–matter interaction which is described by the complex dielectric functions. We determine the dielectric functions of 2D Ga and 2D In via a combination of spectroscopic ellipsometry (SE) and density functional theory (DFT) in a large spectral range from NIR to UV. The MIP provided the 2D metals for the study.

M. Kopf, J. Ebad-allah, **S.H. Lee**, **Z.Q. Mao**, C.A. Kuntscher, "Influence of magnetic ordering on the optical response of the antiferromagentic topological insulator $MnBi_2Te_4$," *Physical Review B* 102 (16), 165139 (2020). 10.1103/PhysRevB.102.165139.

Comprehensive temperature-dependent optical conductivity studies were performed on $MnBi_2Te_4$ that were grown using the 2DCC Bulk Growth facility. The observations of strong changes in the optical conductivity at Neel temperature confirms the impact of magnetic ordering on the bulk electronic properties of $MnBi_2Te_4$.

M.A. Steves, **Y. Wang**, N. Briggs, T. Zhao, H. Elh-Sherif, B.M. Betrsch, S. Subramanian, C. Dong, T. Bowen, A. De La Fuente Duran, K. Nisi, M. Lassauniere, U. Wurstbauer, N.D. Bassim, J. Fonseca, J.T. Robinson, **V.H. Crespi**, **J.A. Robinson**, K.L. Knappenberger Jr., "Unexpected

Near-Infrared to Visible Nonlinear Optical Properties from 2-D Polar Metals," *Nano Letters* 20 (11), 8312-8318 (2020). 10.1021/acs.nanolett.0c03481.

Near-infrared-to-visible second harmonic generation from air-stable two-dimensional polar gallium and indium metals is described. The photonic properties of 2D metals -including the largest second-order susceptibilities reported for metals (approaching 10nm2/V) –are determined by the atomic-level structure and bonding of two-to-three-atom-thick crystalline films. The MIP played a key role in developing the 2D metals and providing theory on the origin of the optical response.

- Collaboration with External User R0024 (National Lab)

P. Li, J. Koo, W. Ning, J. Li, L. Miao, L. Min, Y. Zhu, Y. Wang, N. Alem, **C.-X. Liu**, **Z. Mao**, B. Yan, "Giant room temperature anomalous Hall effect and tunable topology in a ferromagnetic topological semimetal Co_2MnAl," *Nature Communications* 11 (1), 3476 (2020). 10.1038/s41467-020-17174-9.

This work not only reveals a giant room temperature anomalous Hall effect in a Heusler alloy Co_2MnAl, but also demonstrates its band topology can be tuned by the rotation of magnetization driven by small magnetic fields. These results pay a way for potential applications of 2D thin film of this material in spintronic devices.

X. Wu, D. Xiao, C.-Z. Chen, J. Sun, L. Zhang, M.H.W. Chan, N. Samarth, X.C. Xie, X. Lin, C.-Z. Chang, "Scaling Behavior of the Quantum Phase Transition from a Quantum Anomalous Hall Insulator to an Axion Insulator," *Nature Communications* (2020). 10.1038/s41467-020-18312-z.

Heterostructures of magnetically-doped topological insulators were grown by MBE and used to study the phase transition from the quantum anomalous Hall phase to an axion insulator phase. We find that the transition follows a universal scaling behavior when we analyze the temperature dependence of the derivative of the longitudinal resistance on magnetic field at the transition point. This behavior follows a characteristic power-law that indicates a universal scaling behavior that can be understood by the Chalker-Coddington network model with a critical exponent which agrees with recent high-precision numerical results.

Y. Chen, Y.-W. Chuang, S.H. Lee, Y. Zhu, K. Honz, Y. Guan, Y. Wang, K. Wang, Z. Mao, J. Zhu, C. Heikes, P. Quarterman, P. Zajdel, J.A. Borchers, W. Ratcliff II, "Ferromagnetism in van der Waals compound $MnSb_{1.8}Bi_{0.2}Te_4$," *Phys. Rev. Mater.*, 4, 064411 (2020). 10.1103/PhysRevMaterials.4.064411

A new ferromagnetic phase showing unusual anomalous Hall effect was synthesized through the control of disorders. This material offers opportunity to explore new topological quantum states in 2D. This involves collaboration with a minority researcher at NIST.

Y. Zhu, B. Singh, Y. Wang, C.-Y. Huang, W.-C. Chiu, B. Wang, D. Graf, Y. Zhang, H. Lin, J. Sun, A. Bansil, **Z. Mao**, "Exceptionally large anomalous Hall effect due to anticrossing of spin-split bands in the antiferromagnetic half-Heusler compound TbPtBi," *Physical Review B*, 101, 161105 (2020). 10.1103/PhysRevB.101.161105.

This work reveals a large intrinsic anomalous Hall effect with a record value of the Hall angle in a half Heusler compound. This phenomenon arises from the anticrossing of spin-split bands near the Fermi level. The physics revealed in this work can be extended to 2D systems.

J. Jiang, D. Xiao, F. Wang, J.H. Shin, D. Andreoli, J. Zhang, R. Xiao, Y.-F. Zhao, M. Kayyalha, L. Zhang, K. Wang, J. Zang, **C. Liu**, **N. Samarth**, M.H.W. Chang, **C.Z. Chang**, "Concurrence of quantum anomalous Hall and topological Hall effects in magnetic topological insulator sandwich structures," *Nature Materials* 19, 732-737 (2020). 10.1038/s41563-020-0605-z.

MBE-grown magnetically doped topological insulator heterostructures were used to demonstrate the voltage tuned transition between and concurrence of Berry phase spin texture (characterized by the quantum anomalous Hall effect) and real space spin texture (characterized by the topological Hall effect).

- Also science driver NGDev

M. Kayyalha, D. Xiao, R. Zhang, J. Shin, J. Jiang, F. Wang, Y.-F. Zhao, R. Xiao, L. Zhang, K.M. Fijalkowski, P. Mandal, M. Winnerlein, C. Gould, Q. Li, L.W. Molenkamp, M.H.W. Chan, **N. Samarth**, **C.-Z. Chang**, "Absence of evidence for chiral Majorana modes in quantum anomalous Hall-superconductor devices," *Science*, 367 (6473), 64-67, (2020). 10.1126/science.aax6361

This study used MBE-grown magnetically doped topological insulator heterostructures with highly transparent superconducting contacts to show that the half-quantized conductance of a quantum anomalous Hall insulator channel with proximitized superconductivity is not a signature of chiral Majorana fermions as predicted by theory.

S. Islam, S. Bhattacharya, H. Nhalil, M. Banerjee, **A. Richardella**, A. Kandala, D. Sen, **N. Samarth**, S. Elizabeth, A. Ghosh, "Low-temperature saturation of phase coherence length in topological insulators," *Physical Review B* 99, 245407 (2019). 10.1103/PhysRevB.99.245407

This collaborative paper used 2DCC MBE-grown samples to study the magnetoresistance and conductance fluctuations measurements in topological insulator thin films. The studies indicated the need to identify an alternative source of dephasing that dominates at low temperature in topological insulators, causing saturation in the phase breaking length and time.

S.H. Lee, Y. Zhu, Y. Wang, L. Miao, T. Pillsbury, H. Yi, S. Kempinger, J. Hu, C.A. Heikes, P. Quaterman, W. Ratcliff, J.A. Borchers, H. Zhang, X. Ke, D. Graf, N. Alem, **C-Z. Chang**, **N. Samarth**, and **Z. Mao**, "Spin scattering and noncollinear spin structure-induced intrinsic anomalous Hall effect in antiferromagnetic topological insulator $MnBi_2Te_4$," *Phys. Rev. Res.*, 1, 012011R (2019). 10.1103/PhysRevResearch.1.012011

This study used the 2DCC Bulk Growth facility and the 2DCC ARPES facility to study the antiferromagnetic (AFM) topological insulator $MnBi_2Te_4$. The key findings included the discovery of a magnetic field-driven non-collinear spin structure with an intrinsic anomalous Hall effect and a large intrinsic gap in the surface states created by strong spin fluctuations.

F. Wang, D. Xiao, W. Yuan, J. Jiang, Y.-F. Zhao, L. Zhang, Y. Yao, W. Liu, Z. Zhang, **C. Liu**, J. Shi, W. Han, M. H. W. Chan, **N. Samarth**, and **C.-Z. Chang**, "Observation of Interfacial Antiferromagnetic Coupling between Magnetic Topological Insulator and Antiferromagnetic Insulator," *Nano Letters* 19(5) 2945-2952 (2019). 10.1021/acs.nanolett.9b00027

This study used MBE-grown ferromagnetic topological insulator/antiferromagnetic insulator heterostructures ($(Cr,Sb)_2Te_3/Cr_2O_3$) to demonstration rich temperature-tuned interfacial antiferromagnetic exchange coupling and an exchange-enhanced Curie temperature in the ferromagnetic topological insulator.

L.-H. Hu, **C.-X. Liu**, F.-C. Zhang, "Topological Larkin-Ovchinnikov phase and Majorana zero mode chain in bilayer superconducting topological insulator films," *Commun. Physics*, 2 (1), 1-7 (2019). 10.1038/s42005-019-0126-8

This theoretical paper predicts the emergence of a magnetic field-induced topological Larkin-Ovchinnikov superconducting phase with a finite momentum pairing in bilayer superconducting topological insulator films. The theoretical model can be naturally realized in superconductor/topological insulator sandwich structure or in a Fe(Te, Se) film.

D. Xiao, J. Jiang, J.H. Shin, W. Wang, F. Wang, Y.F. Zhao, **C.X. Liu**, W.D Wu, M. H. W. Chan, **N. Samarth**, and **C.Z. Chang**, "Realization of the Axion Insulator State in Quantum Anomalous Hall Sandwich Heterostructures," *Phys. Rev. Lett.*, 120, 056801 (2018). 10.1103/PhysRevLett.120.056801

This study used 2DCC MBE-grown magnetic topological insulator heterostructures to realize a new quantum state of matter known as the axion insulator, wherein both the longitudinal and Hall conductivity vanish when the opposite surfaces of a topological insulator are oppositely gapped.

- Also science driver NGDev

S. Islam, S. Bhattacharyya, **A. Richardella**, **N. Samarth**, and A. Ghosh, "Bulk-impurity Induced Noise in Large-area Epitaxial Thin Films of Topological Insulators", *Appl. Phys. Lett* **2017**, *111*, 062107. 10.1063/1.4998464

This collaborative paper used 2DCC MBE-grown samples to study the low frequency electrical noise in topological insulator thin films. The studies showed that even in very thin films, defect states within a bulk impurity band are the source of significant electrical noise in surface electrical transport.

Y. Pan, Q-Z. Wang, A. Yeats, T. Pillsbury, T. Flanagan, **A. Richardella**, H. Zhang, D. Awschalom, **C-X. Liu**, **N. Samarth**, "Helicity Dependent Photocurrent in Electrically Gated $(Bi_{1-x}Sb_x)_2Te_3$ Thin Films", *Nature Commun.* **2017**, *8*, 1037; 10.1038/s41467-017-00711-4

This study used 2DCC MBE-grown samples to study the circular photogalvanic effect in topological insulator thin films as a function of chemical potential. The key result shows that even when photocurrents are excited using photon energies well above the bulk band gap, the transitions are still influenced by the spin-momentum correlation present in the Dirac states leading to direction control of photocurrents by the circular polarization of the optical excitation.

- Also science driver NGDev

A. Yeats, P. Mintun, Y. Pan, **A. Richardella**, B. Buckley, **N. Samarth**, and D. Awschalom, "Local Optical Control of Ferromagnetism and Chemical Potential in a Topological Insulator", *PNAS* **2017**, *114* (9), 10379-10383. 10.1073/pnas.1713458114

This collaborative paper demonstrates micron-scale persistent optical patterning of ferromagnetism and chemical potential landscape in magnetically doped topological insulators grown in 2DCC Thin Films facility.

- Also science driver NGDev

W. Dai, **A. Richardella**, R. Du, W. Zhao, X. Liu, C-X. Liu, S-H. Huang, R. Sankar, F. Chou, **N. Samarth**, and Q. Li, "Proximity-effect-induced Superconducting Gap in Topological Surface States - A Point Contact Spectroscopy Study of $NbSe_2/Bi_2Se_3$ Superconductor-Topological Insulator Heterostructures", *Scientific Reports* **2017**, *7*. 10.1038/s41598-017-07990-3

Point-contact study of the proximity-induced superconductivity in a topological insulator/superconductor bilayer ($Bi_2Se_3/NbSe_2$) grown using the 2DCC Thin Films facility.

- Also science driver NGDev

N. Samarth, "Quantum Materials Discovery From a Synthesis Perspective," *Nature Materials* **2017**, *16*, 1068-1076. 10.1038/NMAT5010

Review article on status and opportunities in materials synthesis of quantum materials including those of central interest to the 2DCC Thin Films facility.

- Also science driver NGDev

J. P. Heremans, R.J. Cava, and **N. Samarth**, "Tetradymites as Thermoelectrics and Topological Insulators", *Nat. Rev. Mater.* **2017**, *2*, 17049. 10.1038/natrevmats.2017.49

Review article on the synthesis and properties of chalcogenide crystals (tetradymites) that are of central interest to the 2DCC Thin Films facility.

- Also science driver NGDev

Epitaxy of 2D Chalcogenides (Epi2DC)

External User Publications (Epi2DC)

G. Kim, H.M. Kim, P. Kumar, M. Rahaman, C.E. Stevens, J. Jeon, K. Jo, K.-H. Kim, N. Trainor, H. Zhu, B.-H. Sohn, E.A. Stach, J.R. Hendrickson, N.R. Glavin, J. Suh, **J.M. Redwing**, D. Jariwala, "High-Density, Localized Quantum Emitters in Strained 2D Semiconductors" *ACS Nano* (2022). 10.1021/acsnano.2c02974.
This study demonstrates a bottom-up, scalable, and lithography-free approach for creating large areas of localized emitters with high density (~150 emitters/um^2) in a WSe_2 monolayer. This approach of using a metal nanoparticle array to generate a high density of strained quantum emitters will be applied to scalable, tunable, and versatile quantum light sources. WSe_2 use in this study was grown using the 2DCC MOCVD1 equipment.

- External User Project S0065

A. Basiri, M.Z.E. Rafique, J. Bai, S. Choi, Y. Yao, "Ultrafast Low-pump Fluence All-optical Modulation Based on Graphene-metal Hybrid Metasurfaces," *Light: Science and Applications* 11, 102 (2022). 10.1038/s41377-022-00787-8.
All-optical graphene modulators reported so far require high pump fluence due to the ultrashort photo-carrier lifetime and limited absorption in graphene. This study presents modulator designs based on graphene-metal hybrid plasmonic metasurfaces with highly enhanced light-graphene interaction in the nanoscale hot spots at pump and probe (signal) wavelengths. The proposed designs hold the promise to address the challenges in the realization of ultrafast all-optical modulators for mid-and far-infrared wavelengths. The 2DCC contributed graphene using faculty equipment.

- External User Project S0036

M.H. Alam, S. Chowdhury, A. Roy, X. Wu, R. Ge, M.A. Rodder, J. Chen, Y. Lu, C. Stern, L. Houben, R. Chrostowski, S.R. Burlison, S.J. Yang, M.I. Serna, A. Dodabalapur, F. Mangolini, D. Naveh, J.C. Lee, S.K. Banerjee, J.H. Warner, D. Akinwande, "Wafer-Scalable Single-Layer

Amorphous Molybdenum Trioxide," *ACS Nano* 16 (3), 3756-3767 (2022). 10.1021/acsnano.1c07705.
Molybdenum trioxide (MoO_3), an important transition metal oxide (TMO), has been extensively investigated over the past few decades due to its potential in existing and emerging technologies, including catalysis, energy and data storage, electrochromic devices, and sensors. This study demonstrates a facile route to obtain wafer-scale monolayer amorphous MoO_3 using 2D MoS_2 as a starting material, followed by UV–ozone oxidation at a substrate temperature as low as 120 °C. This simple yet effective process yields smooth, continuous, uniform, and stable monolayer oxide with wafer-scale homogeneity, as confirmed by several characterization techniques, including atomic force microscopy, numerous spectroscopy methods, and scanning transmission electron microscopy. The 2DCC contributed materials in this study from MIP equipment MOCVD1.

- External User Project S0047

O.O Ayodele, S. Pourianejad, A. Trofe, A. Prokofjevs, T. Ignatova, "Application of Soxhlet Extractor for Ultra-clean Graphene Transfer," *ACS Omega* 7 (8), 7297-7303 (2022). 10.1021/acsomega.1c07113.

This study reports a simple, economical, and highly efficient approach for obtaining pristine graphene on a suitable substrate (e.g., SiO_2/Si) by utilizing Soxhlet extraction apparatus for delicate removal of the polymer with a freshly distilled ultrapure solvent (acetone) in a continuous fashion. Excellent structural and morphological qualities of the material thus produced were confirmed using optical microscopy, atomic force microscopy, scanning electron microscopy, and Raman spectroscopy. Compared to the conventional protocol, graphene produced by the current approach has a lower residual polymer content, leading to a root mean square roughness of only 1.26 nm. Graphene materials provided in this study were created using non-MIP faculty equipment.

- External User Project R0062 (Non-R1)

T. Ignatova, S. Pourianejad, X. Li, K. Schmidt, F. Aryeetey, S. Aravamudhan, *S.V. Rotkin*, "Multidimensional Imaging Reveals Mechanisms Controlling Multimodal Label-Free Biosensing in Vertical 2DM-Heterostructures," *ACS Nano* 16 (2), 2598-2607 (2022). 10.1021/acsnano.1c09335.
In this work, a multidimensional optical imaging technique is developed in order to map subdiffractional distributions for doping and strain and understand the role of those for modulation of the electronic properties of the material. As an example, vertical heterostructures comprised of monolayer graphene and single-layer flakes of transition metal dichalcogenide MoS_2 were fabricated and used for biosensing. The 2DCC contributed materials for this study using the MIP equipment MOCVD1.

- External User Project Collaboration between S0016 (Non-R1) and S0034 (Non-R1, MSI/HBCU)

P. Kumar, J. Lynch, B. Song, H. Ling, F. Barrera, K. Kisslinger, H. Zhang, S.B. Anantharaman, J. Digani, H. Zhu, **T.H. Choudhury**, C. McAleese, X. Wang, B.R. Conran, O. Whear, M.J. Motala, M. Snure, C. Muratore, **J.M. Redwing**, N.R. Glavin, E.A. Stach, A.R. Davoyan, D. Jariwala, "Light–matter coupling in large-area van der Waals superlattices," *Nature Nanotechnology* 17, 182-189 (2021). 10.1038/s41565-021-01023-x.

This study presents optical dispersion engineering in a superlattice structure comprising alternating layers of 2D excitonic chalcogenides and dielectric insulators. By carefully designing the unit cell parameters, we demonstrate greater than 90% narrow band absorption in less than 4 nm of active layer excitonic absorber medium at room temperature, concurrently with enhanced photoluminescence in square-centimeter samples. These superlattices show evidence of strong light–matter coupling and exciton–polariton formation with geometry-tuneable coupling constants. The results demonstrate proof of concept structures with engineered optical properties and pave the way for a broad class of scalable, designer optical metamaterials from atomically thin layers. Materials in this study were provided by the 2DCC using MIP equipment MOCVD1.

- External User Project Collaboration among S0065, S0070 and S0077 (Industry)

X. Chen, **B. Huet**, **T.H. Choudhury**, **J.M. Redwing**, T.-M. Lu, G.-C. Wang, "Orientation domain dispersions in wafer scale epitaxial monolayer WSe_2 on sapphire," *Applied Surface Science* 567, 150798 (2021). 10.1016/j.apsusc.2021.150798.
Azimuthal reflection high-energy electron diffraction (ARHEED) is demonstrated to be a powerful technique to measure the symmetry, lattice constants, and in-plane orientation domain dispersion in wafer-scale, continuous monolayer WSe_2 epitaxially grown by metal organic chemical vapor deposition on c-plane sapphire substrate. The constructed 2D reciprocal map from ARHEED reveals few degrees' dispersion in WSe_2 orientation domains due to the step meandering/bunching/mosaic of sapphire substrate. Minor 30° orientation domains are also observed. The methodology can be applied to study other TMDCs epitaxial monolayers, graphene, and confined atomically thin hetero-epitaxial metals. Materials in this study were provided by the 2DCC using MIP equipment MOCVD1.

- External User Project S0048

H. Kim, K. Lu, Y. Liu, H.S. Kum, K.S. Kim, K. Qiao, S.-H. Bae, S. Lee, Y.J. Ji, K.H. Kim, H. Paik, S. Xie, H. Shin, C. Choi, J.H. Lee, C. Dong, **J.A. Robinson**, J.-H. Lee, J.-H. Ahn, G.Y. Yeom, D.G. Schlom, J. Kim, "Impact of 2D-3D Heterointerface on Remote Epitaxial Interaction through Graphene," *ACS Nano* 15 (6), 10587-10596 (2021). 10.1021/acsnano.1c03296.
This study unveils the respective roles and impacts of the substrate material, graphene, substrate–graphene interface, and epitaxial material for electrostatic coupling of these materials, which governs cohesive ordering and can lead to single-crystal epitaxy in the overlying film. Results demonstrate that simply coating a graphene layer on wafers does not guarantee successful implementation of remote epitaxy, since atomically precise control of the graphene-coated interface is required and provides key considerations for maximizing the remote electrostatic interaction between the substrate and adatoms. This was enabled by exploring various material systems and processing conditions, and we demonstrate that the rules of remote epitaxy vary significantly depending on the ionicity of material systems as well as the graphene–substrate interface and the epitaxy environment. The 2DCC provided graphene through use of faculty equipment.

- External User Project S0097

F. Aryeetey, S. Pourianejad, K. Nowlin, T. Ignatova, S. Aravamudhan, "Bandgap recovery of monolayer MoS_2 using defect engineering and chemical doping," *RSC Advances* 11 (34), 20893-20898 (2021). 10.1039/D1RA02888J.
This paper reports on the successful growth and modification of monolayer MoS_2 (1L MoS_2) by controlling carrier concentration and manipulating bandgap in order to improve the efficiency of

light emission. Atomic size MoS_2 vacancies were created using a Helium Ion Microscope, then the defect sites were doped with 2,3,5,6-tetrafluro7,7,8,8-tetracyanoquinodimethane (F4TCNQ). The carrier concentration in intrinsic (as-grown) and engineered 1L MoS_2 was calculated using Mass Action model. The results are in a good agreement with Raman and photoluminescence spectroscopy as well as Kelvin probe force microscopy characterizations. The 2DCC provided MoS_2 materials in this study synthesized by MIP equipment MOCVD1.

- External User Collaboration S0016 (Non-R1) and S0034 (Non-R1, MSI/HBCU)

M. Hilse, X. Wang, P. Killea, F. Peiris, **R. Engel-Herbert**, "Spectroscopic ellipsometry as an in-situ monitoring tool for Bi2Se3 films grown by molecular beam epitaxy," *Journal of Crystal Growth* 566-567, 126177 (2021). 10.1016/j.jcrysgro.2021.126177.

In-operando spectroscopic ellipsometry used in this study enabled determining the dielectric function of substrate and growing film unobscured by surface or interface reactions. Its sensitivity to sample temperature and film thickness variations allows determining growth temperature, absolute film thickness, and growth rate in real time, rendering it a reliable and universal approach for a direct comparison of growth conditions between different growth campaigns, thus offering the potential to improve reproducibility of the growth conditions for Bi_2Se_3 based films and heterostructures. Materials in this study were synthesized using the 2DCC MIP equipment MBE2 and its onboard spectroscopic ellipsometer.

- External User Project S0013 (non-R1, PUI)

J. Li, J. Wang, X. Zhang, C. Elias, G. Ye, D. Evans, G. Eda, **J.M. Redwing**, G. Cassabois, B. Gil, P. Valvin, R. He, B. Liu, J.H. Edgar, "Hexagonal Boron Nitride Crystal Growth from Iron, a Single Component Flux," *ACS Nano* 15 (4), 7032-7039 (2021). 10.1021/acsnano.1c00115.

This paper reports on the synthesis of hexagonal boron nitride bulk crystal at room temperature using a single component iron flux which is simpler and more cost effective than current multi-component flux processes. A low nucleation density for WSe_2 was observed on the hBN surface indicating a low defect density. The hBN exhibits excellent structural and electrical properties. (WSe_2 growth by MIP equipment MOCVD1 was carried out in the 2DCC facility.)

- External User Project S0087

E. Thompson, E. Manzella, E. Murray, M. Pelletier, J. Stuligross, B.C. Daly, **S.H. Lee**, **R. Redwing**, "Picosecond laser ultrasonic measurements of interlayer elastic properties of 2H-$MoSe_2$ and 2H-WSe_2," *Materials Today Chemistry* 18, 100369 (2020). 10.1016/j.mtchem.2020.100369.

This project explored the use of pump-probe measurements to study the interlayer elastic properties of TMD flakes exfoliated from bulk crystals. Undergraduate students from Vassar College (external user) received training and worked on-site at the 2DCC facility during the summer to synthesize and characterize TMD films which were used during the academic year to carry out the ultrasonic measurements in the lab at Vassar College. Five of the co-authors including the first author are undergraduate students as Vassar.

- Also Science Driver AdvCM
- External User Project R0027 (non-R1, PUI)

G.G. Jernigan, J.J. Fonseca, C.D. Cress, M. Chubarov, **T.H. Choudhury**, J.T. Robinson, "Electronic Changes in Molybdenum Dichalcogenides on Gold Surfaces," *Journal of Physical Chemistry C* 124 (46), 25361-25368 (2020). 10.1021/acs.jpcc.0c07813.

This study employed x-ray and UV photoelectron spectroscopy to study possible charge transfer between TMD monolayer and gold surfaces and revealed that shifts in the XPS spectra were due to interactions of defects in the TMD monolayer with the gold surface. Wafer-scale MoS_2 monolayers from the 2DCC Thin Films facility were supplied to NRL (external user) for this study.

- External User Project R0024 (National Lab)

Y. Xiang, X. Sun, L. Valdman, F. Zhang, **T.H. Choudhury**, M. Chubarov, **J.A. Robinson**, **J.M. Redwing**, **M. Terrones**, Y. Ma, L. Gao, M.A. Washington, T.-M. Liu, G.-C. Wang, "Monolayer MoS_2 on sapphire: an azimuthal reflection high-energy electron diffraction perspective," *2D Materials* 8 (2), 025003 (2020). 10.1088/2053-1583/abce08.

This study employed x-ray and UV photoelectron spectroscopy to study possible charge transfer between TMD monolayer and gold surfaces and revealed that shifts in the XPS spectra were due to interactions of defects in the TMD monolayer with the gold surface. Wafer-scale MoS_2 monolayers from the 2DCC Thin Films facility were supplied to NRL (external user) for this study.

- External User Project S0048 (non-R1)

J. Li, C. Yuan, C. Elias, J. Wang, X. Zhang, G. Ye, C. Huang, M. Kuball, G. Eda, **J.M. Redwing**, R. He, G. Cassabois, B. Gil, P. Valvin, T. Pelini, B. Liu, J.H. Edgar, "Hexagonal Boron Nitride Single Crystal Growth from Solution with a Temperature Gradient," *Chemistry of Materials* 32 (12) 5066-5072 (2020). 10.1021/acs.chemmater.0c00830.

Large hexagonal boron nitride crystals were synthesized by solution growth with a temperature gradient. The crystals have low defect density and narrow Raman peak width, providing an alternative method to high-pressure synthesis. Epitaxial growth of TMDs on hBN was carried out in the 2DCC to assess the defect density.

- External User Project S0087

D.J. Pennachio, C.C. Ornelas-Skarin, N.S. Wilson, S.G. Rosenberg, K.M. Daniels, R.L. Myers-Ward, D.K. Gaskill, C.R. Eddy Jr., C.J. Palmstrom, "Tailoring commensurability of hBN/graphene heterostructures using substrate morphology and epitaxial growth conditions," *Journal of Vacuum Science & Tech*. A, 37, 51503 (2019). 10.1116/1.5110524.

Demonstration of the ability to select different rotational alignments by changing epitaxial growth conditions. This may be used in future wafer-scale growth of hBN/graphene heterostructures to achieve varying degrees of graphene band structure modulation. The project utilized non-MIP equipment to provide epitaxial graphene to external users for hBN growth.

- External user project S0018 (Users from MSI)

K. Zhang, **Y. Wang**, J. Joshi, F. Zhang, S. Subramanian, M. Terrones, P. Vora, **V. Crespi**, and **J.A. Robinson**, "Probing the origin of lateral heterogeneities in synthetic monolayer molybdenum disulfide," *2D Materials* 6 (2) 025008 (2019). 10.1088/2053-1583/aafd9a.

Joint experiment/theory study of the distribution and the origin of inhomogeneities in monolayer MoS_2 of relevance for understanding and optimizing the quality of materials supplied by 2DCC.

- Also science driver AdvCM
- External User Project R0016

X. Zhang, **T.H. Choudhury**, **M. Chubarov**, Y. Xiang, B. Jariwala, F. Zhang, N. Alem, G.C. Wang, **J.A. Robinson**, and **J.M. Redwing**, "Diffusion-Controlled Epitaxy of Large Area Coalesced WSe_2 Monolayers on Sapphire," *Nano Lett*., 18(2), 1049–1056 (2018). 10.1021/acs.nanolett.7b04521.

Development and study of a multi-step process to grow coalesced epitaxial monolayer 2D chalcogenide films on scalable substrates in collaboration with external user.

- External User Project S0014 (User from non-R1).

Local User Publications (Epi2DC)

Q. Qian, W. Wu, L. Peng, **Y. Wang**, A.M.Z. Tan, L. Liang, S.M. Hus, K. Wang, **T.H. Choudhury**, **J.M. Redwing**, A.A. Puretzky, D.B. Geohegan, **R.G. Hennig**, X. Ma, *S. Huang*, "Photoluminescence Induced by Substitutional Nitrogen in Single-Layer Tungsten Disulfide," *ACS Nano* 16 (5), 7428-7437 (2022). 10.1021/acsnano.1c09809.
This study combines experiment and theory in the 2DCC. The electronic and optical properties of two-dimensional materials can be strongly influenced by defects, some of which can find significant implementations, such as controllable doping, prolonged valley lifetime, and single-photon emissions. In this work it is demonstrated that defects created by remote N_2 plasma exposure in single-layer WS_2 can induce a distinct low-energy photoluminescence (PL) peak at 1.59 eV, which is in sharp contrast to that caused by remote Ar plasma. The WS_2 thin films in this study were grown by 2DCC MOCVD equipment.

- Also science driver AdvCM
- Local User Project S0023

A. Dodda, *S. Das*, "Demonstration of Stochastic Resonance, Population Coding, and Population Voting Using Artificial MoS_2 Based Synapses," *ACS Nano* 15 (10), 16172-16182 (2021). 10.1021/acsnano.1c05042.
This study demonstrates how a population of stochastic artificial neurons based on monolayer MoS_2 field effect transistors (FETs) can use an optimum amount of white Gaussian noise and population voting to detect invisible signals at a frugal energy expenditure (~10s of nano-Joules). The findings can aid remote sensing in the emerging era of the Internet of things (IoT) that thrive on energy efficiency. MoS_2 materials in this study were synthesized using MIP equipment MOCVD1.

- Local User Project S0084

R.C. Haislmaier, Y.F. Lu, J. Lapano, H. Zhou, N. Alem, *S.B. Sinnott*, **R. Engel-Herbert**, *V. Gopalan*, "Large Tetragonality and Room Temperature Ferroelectricity in Compressively Strained $CaTiO_3$ Thin Films," *APL Materials*, 7 (5), 051104 (2019). 10.1063/1.5090798.

Demonstration that sizeable epitaxial strain can stabilize a tetragonal distortion and lead to ferroelectric ground state of $CaTiO_3$ at room temperature.

- Local User Project R0002

T.N. Walter, S. Lee, X. Zhang, **M. Chubarov**, **J.M. Redwing**, T.N. Jackson, and *S.E. Mohney*, "Atomic layer deposition of ZnO on MoS_2 and WSe_2," Appl. Surface Sci. 480, 43-51 (2019). 10.1016/j.apsusc.2019.02.182.

Investigation of ALD growth of ZnO on TMD monolayers grown in the 2DCC Thin Films facility.

- Local User Project S0035

In-house Research Publications (Epi2DC)

Y. Lin, R. Torsi, N.A. Simonson, A. Kozhakhmetov, **J.A. Robinson**, "Chapter 6 - Realization of Electronic-grade Two-dimensional Transition Metal Dichalcogenides by Thin-film Deposition Techniques," *Materials Today*, edited by Rafik Addou and Luigi Colombo, Elsevier, 159-193 (2022). 10.1016/B978-0-12-820292-0.00012-4.
This book chapter was contributed based on synthesis techniques used by the 2DCC in-house research team. The chapter discusses challenges in synthesizing high-quality TMDs and provides an overview on the thin-film deposition techniques that show a great potential for making electronic-grade 2D TMD, including powder-based and metal-organic chemical vapor deposition (CVD), as well as molecular beam epitaxy. Second, the chapter discusses several aspects of 2D crystals growth in CVD that would impact the material quality, such as substrates, precursor dissociation dynamics, as well as nucleation and growth kinetics in detail. Lastly, a review of the engineering methods for controlling their heterogeneity through controlling defect type and density, heterostructure formation, and substitutional doping.

S. Lee, X. Zhang, T. McKnight, B. Ramkorun, H. Wang, **V. Gopalan**, **J.M. Redwing**, T.N. Jackson, "Low-temperature Processed Beta-phase In_2Se_3 Ferroelectric Semiconductor Thin Film Transistors," *2D Materials* 9 (2), 025023 (2022). 10.1088/2053-1583/ac5b17.
Ferroelectric semiconductor field effect transistors can be key enablers to improve energy efficiency and overall chip and memory performance. In this work, low-temperature processed, back-end-of-the-line compatible transistors were demonstrated by depositing a layered chalcogenide ferroelectric semiconductor, beta-phase In_2Se_3, at temperature as low as 400 °C. Top gate n-channel In_2Se_3 thin film transistors were fabricated with field-effect mobility ~1 $cm^2\ V^{-1}\ s^{-1}$, and simple polarization switching based memory results are presented. The In_2Se_3 in this study was grown by a high-pressure chemical vapor deposition (HPCVD) system, faculty equipment.

- External user collaborator S0069 (National Lab)

M. Hilse, K. Wang, **R. Engel-Herbert**, "Molecular Beam Epitaxy of $PtSe_2$ Using a Co-deposition Approach," *2D Materials* 9 (2), 025029 (2022). 10.1088/2053-1583/ac606f.
The structural properties of co-deposited ultrathin $PtSe_2$ films grown at low temperatures by molecular beam epitaxy on c-plane Al_2O_3 are studied. Postgrowth anneals under a Se flux was

found to dramatically improve the crystalline quality of the films. Even before the postgrowth anneal in Se, the crystallinity of $PtSe_2$ films grown with the co-deposition method was superior to films realized by thermal assisted conversion. Postgrowth annealed films showed Raman modes with narrower peaks and more than twice the intensity. Transmission electron microscopy investigations revealed that the deposited material transitioned to a two-dimensional layered structure only after the postgrowth anneal. $PtSe_2$ growth was found to start as single layer islands that preferentially nucleated at atomic steps of the substrate and progressed in a layer-by-layer like fashion. Materials in this project were grown by 2DCC equipment MBE2.

A. Bansal, X. Zhang, **J.M. Redwing**, "Gas source chemical vapor deposition of hexagonal boron nitride on C-plane sapphire using B_2H_6 and NH_3," *Journal of Materials Research* 36, 4678-4687 (2021). 10.1557/s43578-021-00446-5.
Chemical vapor deposition (CVD) of hexagonal boron nitride (hBN) using diborane (B_2H_6) and ammonia (NH_3) is reported. The effect of growth conditions on hBN growth rate using continuous vs. flow modulation epitaxy (FME) method is investigated to gain insight into the role of gas-phase chemistry during film deposition. The results provide additional insight into the effects of gas-phase reactions on CVD of hBN. The 2DCC provided materials in this study from non-MIP faculty equipment.

A. Sebastian, S. Das, **S. Das**, "An Annealing Accelerator for Ising Spin Systems Based on In-Memory Complementary 2D FETs," *Advanced Materials* 34, 2107076 (2021). 10.1002/adma.202107076.

In this work, subthreshold Boltzmann transport is exploited in complementary 2D field-effect transistors (p-type WSe_2 and n-type MoS_2) integrated with an analog, nonvolatile, and programmable floating-gate memory stack to develop in-memory computing primitives necessary for energy- and area-efficient hardware acceleration of SA for Ising spin systems. The hardware-realistic numerical simulations further highlight the astounding benefits of SA in accelerating the search for larger spin lattices. MoS_2 and WSe_2 materials in this study were synthesized using MIP equipment MOCVD1.

A. Bansal, **M. Hilse**, **B. Huet**, K. Wang, A. Kozhakhmetov, J.H. Kim, S. Bachu, **N. Alem**, R. Collazo, **J.A. Robinson**, **R. Engel-Herbert**, **J.M. Redwing**, "Substrate Modification during Chemical vapor Deposition of hBN on Sapphire," *ACS Applied Materials & Interfaces* 13 (45), 54516-54526 (2021). 10.1021/acsami.1c14591.
A comparison of hexagonal boron nitride (hBN) layers grown by chemical vapor deposition on *C*-plane (0001) versus *A*-plane ($11\bar{2}0$) sapphire (α-Al_2O_3) substrate is reported. Under the typical growth conditions required for high crystalline quality hBN growth, *A*-plane sapphire provides a more chemically stable substrate. Materials (hBN) in this study were provided by 2DCC faculty non-MIP equipment and RHEED analysis was contributed using MIP equipment MBE2.

M.A. Steves, S. Rajabpour, K. Wang, C. Dong, W. He, S.Y. Quek, **J.A. Robinson**, K.L. Knappenberger, "Atomic-Level Structure Determines Electron-Phonon Scattering rates in 2-D Polar Metal Heterostructures," *ACS Nano* 15 (11), 17780-17789 (2021). 10.1021/acsnano.1c05944.
The electron dynamics of atomically thin 2-D polar metal heterostructures, which consisted of a few crystalline metal atomic layers intercalated between hexagonal silicon carbide and graphene grown from the silicon carbide, were studied using nearly degenerate transient absorption spectroscopy. Optical pumping created charge carriers in both the 2-D metals and graphene

components. Wavelength-dependent probing suggests that graphene-to-metal carrier transfer occurred on a sub-picosecond time scale. These studies provided insights into electronic carrier dynamics in 2-D crystalline elemental metals, including resolving contributions from specific components of a 2-D metal-containing heterojunction. The correlative ultrafast spectroscopy and nonlinear microscopy results suggest that the energy dissipation rates can be tuned through atomic-level structures. Materials provided by the 2DCC in this study were grown on non-MIP faculty equipment.

J.A. Robinson, B. Schuler, "Engineering and probing atomic quantum defects in 2D semiconductors: A perspective," *Applied Physics Letters* 119 (14), 140501 (2021). 10.1063/5.0065185.
Semiconducting two-dimensional (2D) transition metal dichalcogenides (TMDs) are considered a key materials class to scale microelectronics to the ultimate atomic level. The robust quantum properties in TMDs also enable new device concepts that promise to push quantum technologies beyond cryogenic environments. In this Perspective, the authors review some recent results on engineering and probing atomic point defects in 2D TMDs. Furthermore, we provide a personal outlook on the next frontiers in this fast evolving field. Materials included in the 2DCC portion of the analysis were synthesized using non-MIP faculty equipment.

M.T. Wetherington, F. Turker, T. Bowen, A. Vera, S. Rajabpour, N. Briggs, S. Subramanian, A. Maloney, **J.A. Robinson**, "2-dimensional polar metals: a low-frequency Raman scattering study," *2D Materials* 8 (4), 041003 (2021). 10.1088/2053-1583/ac2245.
This study discusses newly discovered low-frequency (LF) (<100 cm^{-1}) Raman features due to the formation of unique 2D polar metals (Ag, Cu, Pb, Bi, Ga, In) or metal alloys (In_xGa_{1-x}) intercalated at an epitaxial graphene (EG)/silicon carbide (SiC) interface and demonstrate that 2D-Ag and 2D-Ga can have spatially distinct phases with their own unique Raman responses. Additionally, the study establishes that the 2D-Ga exhibits a structural evolution as a function of temperature, independent of the SiC and EG, that can lead to nucleation of secondary phases. The newly identified LF Raman responses discussed here lay the foundation for rapid, direct, and spatially resolved evaluation of 2D polar metals in ambient. Materials provided by the 2DCC were synthesized using non-MIP faculty equipment.

M. Fromel, R.L. Crisci, C.S. Sankhe, **D.R. Hickey**, T.B. Tighe, E.W. Gomez, C.W. Pester, "User-friendly chemical patterning with digital light projection polymer brush photolithography," *European Polymer Journal* 158, 110652 (2021). 10.1016/j.eurpolymj.2021.110652.
Patterned polymeric coatings are broadly relevant for all areas of bioengineering: anti-biofouling, controlled protein adsorption, guided cell growth, and many more. This contribution describes a robust topographical and chemical patterning platform that combines an LED digital light projector with oxygen-tolerant light-mediated polymerization to design advanced surfaces on the micron scale and in mild ambient conditions. The user-friendly nature of this approach is targeted towards bringing complex chemical patterning abilities based on surface-tethered polymers into the hands of non-experts and enabling both fundamental and applied studies related to patterned surfaces in bioengineering.

S. Rajabpour, A. Vera, W. He, B.N. Katz, R.J. Koch, M. Lassauniere, X. Chen, C. Li, K. Nisi, H. El-Sherif, M.T. Wetherington, C. Dong, A. Bostwick, C. Jozwiak, **A.C.T. van Duin**, N. Bassim, **J. Zhu**, G.-C. Wang, U. Wurstbauer, E. Rotenberg, **V.H. Crespi**, S.Y. Quek, **J.A. Robinson**, "Tunable 2D Group-III Metal Alloys," *Advanced Materials* 33 (44), 2104265 (2021). 10.1002/adma.202104265.

Chemically stable quantum-confined 2D metals are of interest in next-generation nanoscale quantum devices. Bottom-up design and synthesis of such metals could enable the creation of materials with tailored, on-demand, electronic and optical properties for applications that utilize tunable plasmonic coupling, optical nonlinearity, epsilon-near-zero behavior, or wavelength-specific light trapping. In this work, it is demonstrated that the electronic, superconducting, and optical properties of air-stable 2D metals can be controllably tuned by the formation of alloys. Environmentally robust large-area 2D-In_xGa_{1-x} alloys are synthesized by Confinement Heteroepitaxy (CHet). Materials in this study were synthesized using non-MIP faculty equipment. Theoretical analysis was provided by 2DCC personnel.

- Also science driver AdvCM
- In-house Research Collaboration with External User S0048 (Non-R1)

T. Mirabito, **B. Huet**, **J.M. Redwing**, **D.W. Snyder**, "Influence of the Underlying Substrate on the Physical Vapor Deposition of Zn-Phthalocyanine on Graphene," *ACS Omega* 6 (31), 20598-20610 (2021). 10.1021/acsomega.1c02758.

This work investigates the ZnPc physical vapor deposition (PVD) on graphene either as-grown on Cu or as-transferred on various substrates including Si(100), C-plane sapphire, SiO_2/Si, and h-BN. The experiments show that, for identical deposition conditions, ZnPc exhibits various morphologies such as high-aspect-ratio nanowires or a continuous film when changing the substrate supporting graphene. ZnPc morphology is also found to transition from a thin film to a nanowire structure when increasing the number of graphene layers. The observations suggest that substrate-induced changes in graphene affect the adsorption, surface diffusion, and arrangement of ZnPc molecules. This study provides clear guidelines to control MPc crystallinity, morphology, and molecular orientations which drastically influence the (opto)electronic properties. Materials in this study were synthesized using non-MIP faculty equipment.

D.R. Hickey, N. Nayir, M. Chubarov, **T.H. Choudhury**, S. Bachu, L. Miao, **Y. Wang**, C. Qian, **V.H. Crespi**, **J.M. Redwing**, **A.C.T. van Duin**, **N. Alem**, "Illuminating Invisible Grain Boundaries in Coalesced Single-Orientation WS_2 Monolayer Films," *Nano Letters* 21 (15), 6487-6495 (2021). 10.1021/acs.nanolett.1c01517.

Engineering atomic-scale defects is crucial for realizing wafer-scale, single-crystalline transition metal dichalcogenide monolayers for electronic devices. Using electron microscopy and ReaxFF reactive force field-based molecular dynamics simulations, this study provides insights into WS_2 crystal growth mechanisms, providing a direct link between synthetic conditions and microstructure. Imaging and ReaxFF simulations uncover two types of translational mismatch, and we discuss their origin related to relatively fast growth rates. Statistical analysis of >1300 facets demonstrates that microstructural features are constructed from nanometer-scale building blocks, describing the system across sub-Ångstrom to multimicrometer length scales.WS_2 materials in this study were synthesized using MIP equipment MOCVD1 and ReaxFF simulations were conducted by 2DCC personnel.

- Also science driver AdvCM

A. Khozhakhmetov, S. Stolz, A.M.Z. Tan, R. Pendurthi, S. Bachu, F. Turker, **N. Alem**, J. Kachian, **S. Das**, **R.G. Hennig**, O. Groning, B. Schuler, **J.A. Robinson**, "Controllable p-Type Doping of 2D WSe_2 via Vanadium Substitution," *Advanced Functional Materials* 31 (42), 2105252 (2021). 10.1002/adfm.202105252.

Scalable substitutional doping of 2D transition metal dichalcogenides is a prerequisite to developing next-generation logic and memory devices based on 2D materials. In this study scalable growth and vanadium (V) doping of 2D WSe_2 at front-end-of-line and back-end-of-line compatible temperatures of 800 and 400 °C, respectively, is reported. A combination of experimental and theoretical studies confirm that vanadium atoms substitutionally replace tungsten in WSe_2, which results in *p*-type doping via the introduction of discrete defect levels that lie close to the valence band maxima. The *p*-type nature of the V dopants is further verified by constructed field-effect transistors, where hole conduction becomes dominant with increasing vanadium concentration. Therefore, this study presents a method to precisely control the density of intentionally introduced impurities, which is indispensable in the production of electronic-grade wafer-scale extrinsic 2D semiconductors. Materials in this study were synthesized using non-MIP faculty MOCVD equipment for the doping. Theoretical studies were contributed by 2DCC personnel.

- Also science driver AdvCM

D.S.H. Liu, **M. Hilse**, **R. Engel-Herbert**, "Sticking coefficients of selenium and tellurium," *Journal of Vacuum Science and Technology* 39 (2), 023413 (2021). 10.1116/6.0000736.

This work focused on the direct and quantitative determination of sticking coefficients of selenium and tellurium which provides important insights into the kinetics of chalcogenide-based film growth and points toward the need of a precise sample temperature control. Results were obtained from materials grown on the 2DCC MIP equipment MBE2.

M. Chubarov, **T.H. Choudhury**, **D.R. Hickey**, S. Bachu, T. Zhang, A. Sebastian, A. Bansal, H. Zhu, N. Trainor, S. Das, **M. Terrones**, **N. Alem**, **J.M. Redwing**, "Wafer-Scale Epitaxial Growth of Unidirectional WS_2 Monolayers on Sapphire," *ACS Nano* 15 (2), 2532-2541 (2021). 10.1021/acsnano.0c06750.

This paper demonstrated that steps on the sapphire substrate surface can be used to achieve a preferential alignment of WS_2 monolayer domains grown by MIP equipment MOCVD1 in the 2DCC facility resulting in a dramatic reduction of anti-phase boundaries coalesced wafer-scale WS_2 films. Translational boundaries which result from coalescence of WS_2 domains with the same crystallographic direction, but sub-unit cell offsets were observed to be the predominant line defect in the films. The optical and transport properties of the MOCVD-grown TMD monolayers were comparable to that reported for exfoliated flakes.

A. Kozhakhmetov, R. Torsi, C.Y. Chen, **J.A. Robinson**, "Scalable Low-Temperature Synthesis of Two-Dimensional Materials Beyond Graphene," *Journal of Physical Materials*, 4 (1), 012001 (2020). 10.1088/2515-7639/abbdb1.

This review article summarizes recent breakthroughs in low-temperature synthesis for TMDs for semiconductor applications. It is the first review of its kind.

A. Kozhakhmetov, B. Schuler, A.M.Z. Tan, K.A. Chochrane, J.R. Nasr, H. El-Sherif, A. Bansal, A. Vera, V. Bojan, **J.M. Redwing**, N. Bassim, *S. Das*, **R.G. Hennig**, A. Weber-Bargioni, **J.A. Robinson**, "Scalable Substitutional Re-Doping and Its Impact on the Optical and Electronic Properties of Tungsten Diselenide," *Advanced Materials* 32 (50), 2005159 (2020). 10.1002/adma.202005159.

The study demonstrates a viable approach to introducing dopant-level impurities with high precision, specifically focusing on Re as the dopant atom, which can be scaled up to batch production for applications beyond digital electronics. The MIP provided support for both theory and experiment.

- Also Science Driver AdvCM

D.R. Hickey, D.E. Yilmaz, M. Chubarov, S. Bachu, **T.H. Choudhury**, L. Miao, C. Qian, **J.M. Redwing**, **A.C.T. van Duin**, **N. Alem**, "Formation of metal vacancy arrays in coalesced WS_2 monolayer films," *2D Materials* 8 (1), 011003 (2020). 10.1088/2053-1583/abc905.

This work identified a new type of structural defect – metal vacancy arrays – which form in epitaxial WS2 monolayers grown by MOCVD due to coalescence of domains that are slightly offset from one another. This study was a collaboration between 2DCC personnel in the Thin Films and In Situ Characterization Facility and the Theory/Simulation working on epitaxial growth of TMDs, advanced electron microscopy characterization and theory and simulation.

- Also Science Driver AdvCM

T.H. Choudhury, X. Zhang, Z.Y. Al Balushi, M. Chubarov, **J.M. Redwing**, "Epitaxial growth of two-dimensional layered transition metal dichalcogenides," *Annual Review of Materials Research*, 50 (2020). 10.1146/annurev-matsci-090519-113456.

Review article highlighting fundamental issues associated with vapor phase growth and epitaxy of TMDs

- Included external user from Government Lab on user project S0069 and external user from project S0067

Y.C. Lin, B. Jariwala, B. Bersch, K. Xu, Yi.F. Nie, B.M. Wang, **S.M. Eichfeld**, X. Zhang, **T.H. Choudhury**, Y. Pan, R. Addou, C. Smith, J. Li, K. Zhang, M. Aman Haque, S. Folsch, R. Feenstra, R.M. Wallace, K.J. Cho, S. Fullerton-Shirey, **J.M. Redwing**, and **J.A. Robinson**, "Realizing Large-Scale, Electronic-Grade Two-Dimensional Semiconductors," *ACS Nano* 12(2), 965–975 (2018). 10.1021/acsnano.7b07059.

Demonstration of MOCVD growth and properties of WSe_2 epitaxial films grown on sapphire in collaboration with external user. The project utilized non-MIP equipment as part of the Thin Film facility to create the 2D films.

- Included external user from project R0011

F. Zhang, K. Momeni, M. AlSaud, A. Azizi, M. Hainey, **J. Redwing**, *L-Q. Chen*, and N. Alem. "Controlled Synthesis of 2D Transition Metal Dichalcogenides: from vertical to planar MoS_2", *2D Materials* (2017), *4*, (2), 025029. 10.1088/2053-1583/aa5b01

Combined experimental and computational modeling study of powder vapor transport of MoS_2 films carried out in collaboration with local and external users.

- Included external user from project R0001 (User from Non-R1)

N. Briggs, B. Bersch, **Y. Wang**, J. Jiang, R.J. Koch, N. Nayir, K. Wang, M. Kolmer, W. Ko, A. De La Fuente Duran, S. Subramanian, C. Dong, J. Shallenberger, M. Fu, Q. Zou, Y.-W. Chuang, Z. Gai, A.-P. Li, A. Bostwick, C. Jozwiak , **C.-Z. Chang**, E. Rotenberg, J. Zhu, **A.C.T. van Duin**, **V. Crespi**, **J.A. Robinson**, "Atomically thin half-van der Waals metals enabled by confinement heteroepitaxy," *Nature Materials* 19, 637-643 (2020). 10.1038/s41563-020-0631-x.

This work introduces a new class of 2D materials – air-stable atomically thin metals encapsulated in graphene – whose discovery and development followed from a close coupling of experiment with predictive in-house first-principles calculations of formation energies, electronic properties, electron-phonon coupling, and superconductivity. The project utilized non-MIP equipment as part of the Thin Films facility to create the 2D films, and 2DCC theory. Sample provision of this emerging family of novel 2D metals will be a core future offering of the 2DCC.

- Also science driver AdvCM

X. Zhang, S. Lee, A. Bansal, F. Zhang, **M. Terrones**, T.N. Jackson, **J.M. Redwing**, "Epitaxial growth of few-layer beta-In_2Se_3 thin films by metalorganic chemical vapor deposition," *Journal of Crystal Growth*, 533 (1), 125471 (2020). 10.1016/j.jcrysgro.2019.125471.

First demonstration of epitaxial growth of beta-In_2Se_3 by MOCVD.

B. Huet, J.-P. Raski, **D.W. Snyder**, **J.M. Redwing**, "Fundamental limitations in transferred CVD graphene caused by Cu catalyst surface morphology," Carbon, 163, 95-104 (2020). 10.1016/j.carbon.2020.02.074.

Demonstration of effects of surface roughness and processing conditions on the transfer process for CVD graphene. Samples were grown using individual faculty equipment in the 2DCC.

T. Mirabito, **B. Huet**, A.L. Briseno, **D.W. Snyder**, "Physical vapor deposition of zinc phthalocyanine nanostructures on oxidized silicon and graphene substrates," *Journal of Crystal Growth,* 533 (1), 125484 (2020). 10.1016/j.jcrysgro.2020.125484.

CVD graphene layers (synthesized using individual faculty equipment) were used as substrates for the growth of zinc phthalocyanine nanostructures.

J. Fox, X. Zhang, Z. Al Balushi, M. Chubarov, A. Kozhakhmetov, **J. Redwing**, "Van der Waals epitaxy and composition control of layered SnS_xSe_{2-x} alloy thin films," *Journal of Materials Research*, 1-11 (2020). 10.1557/jmr.2020.19.

Layered $Sn(S,Se)_2$ alloys are of interest for the top absorber layer in tandem Si photovoltaics. This study explored the epitaxial growth and properties of these alloys across the entire composition range.

B. Huet, X. Zhang, **J.M. Redwing**, **D.W. Snyder**, J.-P. Raskin, "Multi-wafer batch synthesis of graphene on Cu films by quasi-static flow chemical vapor deposition," *2D Materials*, 6, 45032 (2019). 10.1088/2053-1583/ab33ae

Demonstration of high-throughput synthesis of uniform single-layer highly crystalline graphene on 3-inch wafers.

N. Briggs, Z.M. Gebeyehu, A. Vera, T. Zhao, K. Wang, A. Duan, B. Bersch, T. Bowen K.L. Knappenberger Jr., **J.A. Robinson**, "Epitaxial graphene/silicon carbide intercalation: a minireview on graphene modulation and unique 2D materials," *Nanoscale*, 11, 15440-15447 (2019). 10.1039/C9NR03721G

In-house research minireview on processes of epitaxial-graphene/SiC intercalation that underpin Platform advances in developing the new family of atomically, air-stable two-dimensional metals described in 10.1007/s11664-020-08087-w. The project also utilized faculty owned equipment as part of the Thin Film facility to create the 2D films.

X. Zhang, F. Zhang, **Y. Wang**, D. S. Schulman, T. Zhang, A. Bansal, N. Alem, S. Das, **V. H. Crespi**, **M. Terrones**, **J. M. Redwing**, "Defect-controlled nucleation and orientation of WSe_2 on hBN – a route to single crystal epitaxial monolayers", *ACS Nano*, 13 (3), 3341, (2019). 10.1021/acsnano.8b09230

A collaborative follow-up to initial work with MoS_2 that provides evidence for the generality of the proposed mechanism of defect-assisted epitaxial growth with orientation control, here demonstrated for WSe_2 grown on hBN with extensive characterization. Uses 2DCC facilities closely coupled to 2DCC-supported theory and informs growth of materials supplied to Platform users.

M. Chubarov, **T.H. Choudhury**, X. Zhang and **J.M. Redwing**, "In-plane x-ray diffraction for characterization of monolayer and few-layer transition metal dichalcogenide films," *Nanotechnol.* 29, 055706 (2018). 10.1088/1361-6528/aaa1bd

Demonstration of in-plane x-ray diffraction for characterization of epitaxial TMD monolayers grown at 2DCC.

A. Kozhakhmetov, **T.H. Choudhury**, Z.Y. Al Balushi, **M. Chubarov**, and **J.M. Redwing**, "Effect of substrate on the growth and properties of thin 3R NbS_2 films grown by chemical vapor deposition," *J. Crystal Growth* 486, 137-141 (2018). 10.1016/j.jcrysgro.2018.01.031

Study of epitaxial growth and properties of NbS_2 thin films.

T.H. Choudhury, H. Simchi, R. Boichot, **M. Chubarov**, *S.E. Mohney* and **J.M. Redwing**, "Chalcogen precursor effect on cold-wall gas-source chemical vapor deposition growth of WS_2," *Cryst. Growth Des.* 8, 4357-4364 (2018). 10.1021/acs.cgd.8b00306

Investigation of the effect of precursor chemistry on the growth and properties of WS_2 thin films grown by MOCVD in the 2DCC Thin Films facility.

N. Briggs, M.I Preciado, Y.F. Lu, K. Wang, J. Leach, X.F. Li, K. Xiao, S. Subramanian, B.M. Wang, A. Haque, **S. Sinnott** and **J.A. Robinson**, "Transformation of 2D group III-selenides to ultra-thin nitrides: enabling epitaxy on amorphous substrates," *Nanotechnol.* 29, 47 (2018). 10.1088/1361-6528/aae0bb

Development of chemical transformation routes towards expanding the suite of 2D systems that are synthetically accessible starting from a chalcogenide initial state. The project utilized faculty owned equipment as part of the Thin Films facility to create the 2D films.

- Also science driver AdvCM

Next Generation Devices (NGDev)

External User Publications (NGDev)

T. Wang, Y. Zhu, **Z. Mao**, Y.-Q. Xu, "Tunneling Effects in Crossed $Ta_2Pt_3Se_8$-$Ta_2Pd_3Se_8$ Nanowire Junctions: Implications for Anisotropic Photodetectors," *ACS Applied Nano Materials* 4 (2), 1817-1824 (2021). 10.1021/acsanm.0c03223.
This work demonstrated nanoscale crossed p-n junctions formed by nanowires of two quasi-1D van der Waals (vdW) materials, i.e. p-type $Ta_2Pd_3Se_8$ (TPdS) and n-type $Ta_2Pt_3Se_8$ (TPtS). Such p-n junctions exhibit asymmetric nonlinear output behaviors, inelastic tunneling effects, and

isotropic photocurrent signals. This study not only offers a way to build nanoscale junctions but also provides fundamental understandings of the electronic and optoelectronic properties of vdW nanowires and their heterojunctions. TPdS and TPtS single crystals used in this were synthesized using non-MIP CVT faculty equipment.

- External User S0049

S.S. Jo, A. Singh, L. Yang, S.C. Tiwari, S. Hong, A. Krishnamoorthy, M.G. Sales, S.M. Oliver, J. Fox, R.L. Cavalero, **D.W. Snyder**, P.M. Vora, S.J. McDonnell, P. Vashishta, R.K. Kalia, A. Nakano, R. Jaramillo, “Growth Kinetics and Atomistic Mechanisms of Native Oxidation of ZrS_xSe_{2-x} and MoS_2 Crystals,” *Nano Letters* 20 (12), 8592-8599 (2020). 10.1021/acs.nanolett.0c03263.

Quantifying and understanding the oxidation mechanisms in the 2DCC-grown ZrS_xSe_{2-x} alloy series is particularly useful for processing electronic devices from Zr-based TMD. In this study, we provide insight and quantitative guidance for designing and processing semiconductor devices.

- External User Project Collaboration between R0014 and R0016

K. Xiong, X. Zhang, L. Li, F. Zhang, B. Davis, A. Madjar, A. Göritz, M. Wiestruck, M. Kaynak, N.C. Strandwitz, **M. Terrones**, **J.M. Redwing**, J.C.M. Hwang, "Temperature-dependent RF Characteristics of Al_2O_3-passivated WSe_2 MOSFETs," *IEEE Electron Device Letters* (2020) in press. 10.1109/LED.2020.2999906.

High-frequency characteristics of WSe_2 MOSFETs were studied as a function of temperature to assess device performance. WSe_2 samples provided by 2DCC were used in this study

- External User Project S0009 (non-R1)

J.J. Fox, S. Bachu, R.L. Cavalero, R.M. Lavelle, S.M. Oliver, S. Yee, P.M. Vora, **N. Alem**, **D.W. Snyder**, “Chemical Vapor Transport Synthesis, Characterization and Compositional Tuning of ZrS_xSe_{2-x} for Optoelectronic Applications,” *Journal of Crystal Growth*, 542, 125609 (2020). 10.1016/j.jcrysgro.2020.125609.

The high anisotropy of the 1T phase of $ZrSe_2$ and ZrS_2 gives rise to a high absorption coefficient which is of interest for photovoltaics and photodetectors. This study explored the CVT synthesis and optical properties of the $Zr(S,Se)_2$ alloy bulk crystals, synthesized in the 2DCC Bulk Growth facility, over the entire composition range.

- External User Project R0016

K. Xiong, M. Hilse, L. Li, A. Göritz, M. Lisker, M. Wietstruck, M. Kaynak, **R. Engel-Herbert**, A. Madjar, J.C.M. Hwang, “Large-Scale Fabrication of Submicrometer-Gate-Length MOSFETs With a Trilayer $PtSe_2$ Channel Grown by Molecular Beam Epitaxy,” *IEEE Transactions on Electron Devices*, 67 (3), 796-801 (2020). 10.1109/TED.2020.2966434

Successful integration of $PtSe_2$ (synthesized in the 2DCC Thin Films facility) as a new channel material into field effect transistor geometry and analysis of device characteristics.

- External User Project R0026 (non-R1)

J. Zhang, M. Boora, T. Kaminski, C. Kendrick, Y.K. Yapa, J.Y. Suh, “Fano resonances from plasmon-exciton coupling in hetero-bilayer WSe_2-WS_2 on Au nanorod arrays,” *Photonics and*

Nanostructures – Fundamentals and Applications, 100783 (2020). 10.1016/j.photonics.2020.100783.

Plasmon-exciton coupling was studied in WSe_2/WS_2 bilayers (synthesized in the 2DCC Thin Films facility) that were integrated with patterned Au nanorod arrays.

- External User Project S0040 (User from Non-R1).

J.J. Fonseca, A.L. Yeats, B. Blue, M.K. Zalalutdinov, T. Brintlinger, B.S. Simpkins, D.C. Ratchford, J.C. Culbertson, J.Q. Grim, S.G. Carter, M. Ishigami, R.M. Stroud, C.D. Cress, J.T. Robinson, "Enabling remote quantum emission in 2D semiconductors via porous metallic networks," *Nature Communications*, 11, 5, (2020). 10.1038/s41467-019-13857-0.

Demonstration of how two-dimensional crystal overlayers influence the recrystallization of relatively thick metal films and the subsequent synergetic benefits this provides for coupling surface plasmon-polaritons (SPPs) to photon emission in 2D semiconductors. TMD samples were grown in the 2DCC facility.

- External User Project R0024 (Government Lab User)

Z. Wu, J. Li, X. Zhang, **J.M. Redwing**, Y. Zheng "Room-Temperature Active Modulation of Valley Dynamics in a Monolayer Semiconductor through Chiral Purcell Effects," *Advanced Materials*, 1904132, (2019). 10.1002/adma.201904132

Demonstration of tunable and active modulation of valley dynamics in a monolayer WSe_2 (synthesized in 2DCC Thin Films facility) at room temperature through controllable chiral Purcell effects in plasmonic chiral metamaterials.

- External User Project S0064

D.R. Hickey, J.G. Azadani, **A.R. Richardella**, J.C. Kally, J.S. Lee, H.C. Chang, T. Liu, M.Z. Wu, **N. Samarth,** T. Low, K. A. Mkhoyan, "Structure and basal twinning of the topological insulator Bi_2Se_3 grown by MBE onto crystalline $Y_3Fe_5O_{12}$," *Physical Review Materials* [Rapid Communications] 3, 061201(R) (2019). 10.1103/PhysRevMaterials.3.061201

Detailed microscopy study of types of disorder present in topological insulator films grown on YIG using atomic force microscopy and scanning transmission electron microscopy, revealing the presence of an amorphous metal oxide layer between the substrate and the film, which appears to smooth out the nanometer-scale undulations in a YIG surface. Using density functional theory, the study explores the impact of observed basal twins on the electronic structure of TI films.

- External User Project S0025

W. Wu, C.K. Dass, J.R. Hendrickson, R.D. Montano, X. Zhang, **T.H. Choudhury**, **J.M. Redwing** and M.T. Pettes, "Locally defined quantum emission from epitaxial few-layer WSe_2," *Appl. Phys. Lett*. (2019). 10.1063/1.5091779

Demonstration of quantum emission from strain-localized WSe_2 epitaxial films that were grown in the 2DCC Thin Films facility.

- External User Project S0007

X. Ge, M. Minkov, **T. Choudhury**, **M. Chubarov**, S. Fan, **J. Redwing**, X. Li, W. Zhou, "Room Temperature Photonic Crystal Surface Emitting Laser with Synthesized Monolayer Tungsten Disulfide," *IEEE International Semiconductor Laser Conference*, 167-168 (2018). 10.1109/ISLC.2018.8516219

Demonstration of lasing with a narrow linewidth from WS_2 epitaxial monolayers grown in 2DCC Thin Films facility and integrated into a silicon nitride photonic crystal cavity.

- External User Project S0010 (User from MSI).

J. Han, **A. Richardella**, S. S. Siddiqui, J. Finley, **N. Samarth**, and L. Liu, "Room Temperature Spin-orbit Torque Switching Induced by a Topological Insulator," Phys. Rev. Lett., 119, 077702 (2017). 10.1103/PhysRevLett.119.077702

This project used Bi_2Se_3 and $(Bi,Sb)_2Te_3$ grown in the 2DCC Thin Films facility to carry out the first room temperature demonstration of energy efficient current driven spin-orbit torque switching in topological insulator-ferrimagnet heterostructure spintronic devices.

- External User Project S0003

Local User Publications (NGDev)

A. Sebastian, R. Pendurthi, **T.H. Choudhury**, **J.M. Redwing**, *S. Das*, "Benchmarking monolayer MoS_2 and WS_2 field-effect transistors," *Nature Communications* 12, 693 (2021). 10.1038/s41467-020-20732-w.

This paper benchmarks device-to-device variation in field-effect transistors (FETs) based on wafer-scale monolayer MoS_2 and WS_2 grown by MIP equipment MOCVD1 in the 2DCC facility. Statistical measures were used to evaluate key FET performance indicators for several hundred 2D FETs and were compared against existing literature as well as ultra-thin body Si FETs. Our results show consistent performance of the 2D FETs owing to high quality uniform layers and clean transfer onto device substrates. We demonstrate record high carrier mobility of 33 cm^2/Vs was measured in WS_2 FETs, which is a 1.5X improvement compared to the best literature report. Our results confirm the technological viability of 2D FETs in future integrated circuits.

- Local User project S0084
- Also science driver Epi2DC

A. Dodda, A. Oberoi, A. Sebastian, **T.H. Choudhury**, **J.M. Redwing**, *S. Das*, "Stochastic resonance in MoS_2 photodetector," *Nature Communications* 11, 4406 (2020). 10.1038/s41467-020-18195-0.

An ultra-low-power sensor based on stochastic resonance phenomena was demonstrated in photodetectors fabricated using large-area MoS_2 monolayers synthesized in the 2DCC facility. Stochastic resonance enables the detection of weak signals within the noise limit of the system and mimics the sensory information processing abilities of animals adapted to extreme and resource limited environments.

- Local User project S0084

D. Jayachandran, A. Oberoi, A. Sebastian, **T.H. Choudhury**, B. Shankar, **J.M. Redwing**, *S. Das*, "A low-power biomimetic collision detector based on an in-memory molybdenum disulfide photodetector," *Nature Electronics* (2020). 10.1038/s41928-020-00466-9.

A compact, low power nanoscale collision detector is demonstrated that mimics the lobula giant movement detector (LGMD) neuron in locusts which can detect an approaching object and prevent collisions within a swarm of millions of locusts. The biomimetic collision detector is comprised of molybdenum disulfide photodetectors stacked on top of a non-volatile and programmable floating-gate memory architecture. Large area MoS_2 monolayers synthesized in the 2DCC facility were used for photodetector fabrication.

- Local user project S0084

J.R. Rodriguez, W. Murray, K. Fujisawa, **S.H. Lee**, A.L. Kotrick, Y. Chen, N. McKee, S. Lee, **M. Terrones**, S. Trolier-McKinstry, T.J. Jackson, **Z. Mao**, Z. Liu and *Y. Liu*, "Electric field induced metallic behavior in thin crystals of ferroelectric alpha-In_2Se_3," *App. Phys. Lett.* 117 (5), 052901 (2020). 10.1063/5.0014945.

Field-effect transistors (FET), which use exfoliated nano flakes of ferroelectric semiconductor α-In_2Se_3 grown by the 2DCC bulk growth facility as the channel material were fabricated and tested. The transport measurements on these devices reveal evidence for the reorientation of electrical polarization and an electric field-induced metallic state in α-In_2Se_3. These results suggest the α-In_2Se_3 based FET devices can serve as a platform for the fundamental study of ferroelectric metals as well as the exploration of potential applications of semiconducting ferroelectrics.

- Local User Project S0039

A.D. Agyapong, K.A. Cooley, *S.E. Mohney*, "Reactivity of contact metals on monolayer WS_2," *Journal of Applied Physics* 128 (5), 055306 (2020). 10.1063/5.0014005.

A rapid non-destructive method based on Raman spectroscopy was developed to analyze the reactivity of contact metals with WS2 monolayers prepared in the 2DCC Thin Films facility. The metal/WS_2 reactivity observed in this study is in excellent agreement with predictions from bulk thermodynamics, which can provide good guidance for studies of other metal/TMD systems.

- Local User Project S0035

Q. Qian, L. Peng, N.P. Lopez, K. Fujisawa, K. Zhang, X. Zhang, **T.H. Choudhury**, **J. Redwing**, **M. Terrones**, X. Ma, *S. Huang*, "Defect creation in WSe_2 with microsecond photoluminescence lifetime by focused ion beam irradiation," *Nanoscale*, 12, 2047-2056 (2020). 10.1039/C9NR08390A

Focused ion beam was used to create defects in WSe_2 (bulk crystals and MOCVD monolayers synthesized in 2DCC Thin Films facility). Long photoluminescence lifetime was measured for defect-related emission peaks which is valuable for valleytronics, quantum emitters and other applications.

- Local User Project S0023

L. Ding, M.S. Ukhtary, **M. Chubarov**, **T.H. Choudhury**, F. Zhang, R. Yang, A. Zhang, J.A. Fan, **M. Terrones**, **J.M. Redwing**, T. Yang, M.D. Li, R. Saito, and *S.X. Huang*, "Understanding interlayer coupling in TMD-hBN heterostructures by Raman spectroscopy*," IEEE Trans. Electron. Dev*. 64(10), 4059-4067 (2018). 10.1109/TED.2018.2847230

Investigation and interpretation of interlayer interactions in 2D heterostructures grown in the 2DCC Thin Films facility by Raman spectroscopy.

- Local User Project S0023

In-house Research Publications (NGDev)

S. Novakov, B. Jariwala, N.M. Vu, A. Kozhakhmetov, **J.A. Robinson**, J.T. Heron, "Interface Transparency and Rashba Spin Torque Enhancement in WSe_2 Heterostructures," *ACS Applied Materials & Interfaces* 13 (11), 13744-13750 (2021). 10.1021/acsami.0c19266.

In this paper, enhanced spin transfer torques from the Rashba spin current in heterostructures of permalloy (Py) and WSe_2 is reported. The study shows that insertion of up to two monolayers of WSe_2 enhances the spin transfer torques in a Rashba system by up to 3×, without changing the fieldlike Rashba spin–orbit torque (SOT), a measure of interface polarization. The results indicate that low layer count TMD films can be used as an interfacial "scattering promoter" in heterostructure interfaces without quenching the original polarization. Materials in this study were provided by the 2DCC using non-MIP MOCVD faculty equipment.

- Also science driver Epi2DC

A. Woeppel, K. Xu, A. Kozhakhmetov, S. Wate, **J.A. Robinson**, S.K. Fullerton-Shirey, "Single- versus Dual-Ion Conductors for Electric Double Layer Gating: Finite Element Modeling and Hall-Effect Measurements," *ACS Applied Materials & Interfaces* (2020). 10.1021/acsami.0c08653.

Demonstration of how TMD transport can be electrostatically controlled using advanced polymer electrolytes. The project utilized non-MIP equipment as part of the Thin Film facility to create the 2D films, with contributions from in-house researchers.

- Also science driver AdvCM

M. Hilse, K. Wang, **R. Engel-Herbert**, "Growth of ultrathin Pt layers and selenization into $PtSe_2$ by molecular beam epitaxy," *2D Materials* 7 (4), 045013 (2020). 10.1088/2053-1583/ab9f91.

2D transition metal dichalcogenide system $PtSe_2$ was grown by MBE using in-situ post-deposition selenization to study layer crystallinity of this material system to be used as high mobility transistor channel materials for ultra-thin-body electronics.

N. Briggs, S. Subramanian, Z. Lin, X. Li, X. Zhang, K. Zhang, K. Xiao, D. Geohegan, R. Wallace, *L.-Q. Chen*, **M. Terrones**, *A. Ebrahimi*, S. Das, **J. Redwing**, C. Hinkle, K. Momeni, **A. van Duin**, **V. Crespi**, S. Kar, and **J.A. Robinson**, "A roadmap for electronic grade 2D materials," *2D Materials* 6 (2), 022001 (2019). 10.1088/2053-1583/aaf836

Review article highlighting applications, current status and future directions for the synthesis, processing and characterization of 2D layered chalcogenides with contributions from in-house researchers, local users and external users of 2DCC.

- Included external users from projects R0037 (User from Non-R1) and R0011

S. Subramanian, K. Xu, **Y. Wang**, S. Moser, N.A. Simonson, D. Deng, **V.H. Crespi**, S.K. Fullerton-Shirey, **J.A. Robinson**, "Tuning transport across MoS_2/graphene interfaces via as-grown lateral heterostructures," *npj 2D Materials and Applications*, 4, 9 (2020). 10.1038/s41699-020-0144-0.

Close coupling of theory and experiment here helps to accelerate the development of device applications for 2D materials through advancing the understanding of interfaces in lateral heterostructures that include transition metal dichalcogenides. The project utilized non-MIP equipment as part of the Thin Film facility to create the 2D films.

- Also science driver AdvCM

Z. Islam, A. Kozhakhmetov, **J. Robinson**, A. Haque, "Enhancement of WSe_2 FET Performance Using Low-Temperature Annealing," *Journal of Electronic Materials* (2020). 10.1007/s11664-020-08087-w.

In this study, we investigate a non-thermal annealing process for two-dimensional materials. Instead of high temperature, we exploit the electron wind force at near-room temperature conditions. The process is demonstrated on back-gated WSe_2 transistors. To explain the atomistic mechanisms behind the room-temperature annealing, we perform molecular dynamics simulation. The project utilized non-MIP equipment as part of the Thin Films facility to create the 2D films.

- Also science driver AdvCM

A. Kozhakhmetov, J.R. Nasr, F. Zhang, K. Xu, N.C. Briggs, R. Addou, R. Wallace, S. Sullerton-Shirey, **M. Terrones**, S. Das, **J.A. Robinson**, "Scalable BEOL compatible 2D tungsten diselenide," *2D Materials*, 7 (1), 15029, (2019). 10.1088/2053-1583/ab5ad1

Benchmark of carbon and alkali salt-free synthesis of fully coalesced, stoichiometric 2D WSe_2 films on amorphous SiO_2/Si substrates at BEOL- compatible temperatures (475 °C) via gas-source metal-organic chemical deposition. This work highlights the necessity of a Se-rich environment in a kinetically limited growth regime for successful integration of low-temperature 2D WSe_2. The project utilized non-MIP equipment as part of the Thin Films facility to create the 2D films.

Advanced Characterization and Modeling (AdvCM)

External User Publications (AdvCM)

S. Paul, R. Torsi, **J.A. Robinson**, K. Momeni, "Effect of the Substrate on MoS_2 Monolayer Morphology, An Integrated Computational and Experimental Study," *ACS Applied Materials & Interfaces* 14 (6), 18835-18844 (2022). 10.1021/acsami.2c03471.
This study combines external user theory and in-house experiment. The study reveals the role of the substrate's energy landscape on the orientation of as-grown TMDs, where the presence of monolayer–substrate energy barriers perpendicular to the streamlines hinder the detachment of precursor nuclei from the substrate. MoS_2 monolayers with controlled orientations could not be grown on the SiO_2 substrate and revealed that amorphization of the substrate changes the intensity and equilibrium distance of monolayer–substrate interactions. Simulations indicate that 0° rotated MoS_2 is the most favorable configuration on a sapphire substrate, consistent with experimental results. The experimentally validated computational results and insight presented in this study pave the way for the high-quality synthesis of TMDs for high-performance electronic and optoelectronic devices. Materials were grown using a faculty CVD machine.

- Also science driver Epi2DC
- External User Project R0037 (Non-R1)

K. Momeni, Y. Ji, L.-Q. Chen, "Computational synthesis of 2D materials grown by chemical vapor deposition," *Journal of Materials Research* 37 (1), 114-123 (2021). 10.1557/s43578-021-00384-2.
This study introduces a multiscale/multiphysics model based on coupling continuum fluid mechanics and phase-field models for CVD growth of 2D materials. It connects the macroscale

experimentally controllable parameters, such as inlet velocity and temperature, and mesoscale growth parameters such as surface diffusion and deposition rates, to morphology of the as-grown 2D materials. The model can guide the CVD growth of monolayer materials and paves the way to their synthesis-by-design. Data from MIP equipment MOCVD1 was used in this study. Simulations were conducted using the computational resources of the non-MIP ICDS facility at Penn State.

- External User Project R0037 (non-R1)

Y. Ji, K. Momeni, L.-Q. Chen, "A multiscale insight into the growth of h-BN: effect of the enclosure," *2D Materials* 8 (3), 035033 (2021). 10.1088/2053-1583/abfcaa.

This study sought to understand fundamental growth mechanisms governing 2D materials synthesized by CVD and their correlation with experimentally specified parameters. A multiscale computational framework was developed and deployed to correlate the macroscale heat and mass flow with the mesoscale morphology of the as-grown 2D materials by solving the coupled system of heat/mass transfer and phase-field equations. Hexagonal boron nitride (h-BN) was used as the model material and investigated the effect of substrate enclosure on its growth kinetics and final morphology. Results included observation of lower concentration with a more uniform distribution on the substrate in an enclosed-growth than open-growth. Simulations were conducted using the computational resources of the non-MIP ICDS facility at Penn State.

- External User Project R0037 (non-R1)

S. Paul, D. Schwen, M.P. Short, K. Momeni, "Effect of Irradiation on Ni-Inconel/Incoloy Heterostructures in Multimetallic Layered Composites," *Journal of Nuclear Materials* 547, 152778 (2021). 10.1016/j.jnucmat.2021.152778.

This molecular dynamics study used interface analysis techniques developed under user project R0037 as applied to heterostructures to delve into relationships of interface thickness and formation of vacancies and interstitials. This relationship is important to the 2DCC in understanding the broader behavior of heterostructures.

- External User Project R0037 (non-R1)

R.A. Rowe, A. Tajyar, M. Munther, K.E. Joahanna, P.G. Allison, K. Momeni, K. Davami, "Nanoscale serration characteristics of additively manufactured superalloys," *Journal of Alloys and Compounds* (2020). 10.1016/j.jallcom.2020.156723.

A study on surface effects and their role in the strength and mechanical properties of materials. The knowledge and expertise developed will apply to materials, where surface effects dominate, including 2D materials. Particularly, the indentation experiments can be used to make new 2D materials such as diamane.

- External User Project R0037 (non-R1)

S. Paul, K. Momeni, V.I. Levitas, "Shear-induced diamondization of multilayer graphene structures: A computational study," *Carbon* 167, 140-147 (2020). 10.1016/j.carbon.2020.05.038.

Computational study with reactive force fields of the role of shear in the generation of interlayer bonding in a 2D material, multilayer graphene, providing potential insights into the generation of interlayer bonds in other 2D multilayers.

- External User Project R0037 (non-R1)

F. Aryeetey, T. Ignatova, S. Aravamudhan, "Quantification of Defects Engineered in Single Layer MoS_2," *RSC Advances* (2020) in press. 10.1039/d0ra03372c

This work used a helium ion beam to create defects in MoS_2. The defect structure was correlated to the appearance of an acoustic phonon mode in the Raman spectra which introduces a new method for quantifying defects in 2D materials. The study used samples provided by 2DCC.

- External User Projects S0016 (User from Non-R1) and S0034 (User from MSI/HBCU)

X. Wang, C.R. Cormier, A. Khosravi, C.M. Smyth, *J.R. Shallenberger*, R. Addou, R.M. Wallace, "In situ exfoliated 2D molybdenum disulfide analyzed by XPS," *Surface Science Spectra*, 27, 014019 (2020). 10.1116/6.0000153.

Quantitative analysis of MoS_2, providing direct evidence that bulk crystals exhibit a sulfur deficient surface composition of $MoS_{1.8}$, and impurities below the XPS detection limit.

- External User Project R0011

K. Momeni, Y. Ji, **Y. Wang**, S. Paul, S. Neshani, D.E. Yilmaz, Y.K. Shin, D. Zhang, J.-W. Jiang, H.S. Park, **S. Sinnott**, **A. van Duin**, **V. Crespi**, *L.-Q. Chen*, "Multiscale computational understanding and growth of 2D materials: a review," *npj Computational Materials*, 6, 22 (2020). 10.1038/s41524-020-0280-2.

This comprehensive review of computational and data-centric approaches to materials growth and discovery (led by a 2DCC user) spans from atomistic to mesoscopic, macroscopic and materials genomic methods and thus embodies the core theory/data mission of the 2DCC in materials discovery and development. Resources reviewed here, such as advanced reactive force fields, are provided to the community.

- External User Project R0037 (non-R1).

W. Wang, C.M. Smyth, A. Khosravi, C.R. Cormier, *J.R. Shallenberger*, R. Addou, R.M. Wallace, "2D Topological Insulator Bismuth Selenide Analyzed by in situ XPS," *Surface Science Spectra*, 26 (2), 024014, (2020). 10.1116/1.5130891

Development of XPS protocols for the analysis of 2D TIs, in concert with and in support of external users.

- External User Project R0011

J.R. Shallenberger, C.M. Smyth, R. Addou, R.M. Wallace, "2D Bismuth Telluride analyzed by XPS," *Surface Science Spectra*, 26, 024011, (2019). 10.1116/1.5120015

Development of XPS protocols for the analysis of 2D TIs, in concert with and in support of external users.

- External user Project R0011

S. Paul and K. Momeni, "Mechanochemistry of Stable Diamane and Atomically Thin Diamond Films Synthesis from Bi-and Multilayer Graphene: A Computational Study," *Journal of Phys. Chem. C*, 123, 15751 (2019). 10.1021/acs.jpcc.9b02149

Demonstration of the possibility to synthesize diamond films from multilayer graphene using the molecular dynamics approach with reactive force fields provided by the 2DCC.

- External User Project R0001 (User from Non-R1)

K. Momeni, Y. Ji, K. Zhang, **J.A. Robinson**, and *L-Q. Chen*. "Multiscale framework for simulation-guided growth of 2D materials," npj 2D Materials and Applications 2, no. 1 (2018): 27. 10.1038/s41699-018-0072-4

Development of computational tools to simulate CVD growth of 2D materials in conditions relevant to 2DCC.

- External User Project R0001 (User from Non-R1).

Local User Publications (AdvCM)

Y. Lu and *S.B. Sinnott*, “Density functional theory study of epitaxially strained monolayer transition metal dichalcogenides for piezoelectricity generation,” *ACS Applied Nano Materials*, 3 (1), 384-390 (2020). 10.1021/acsanm.9b02021

A high-throughput computational analysis of the elastic and piezoelectric response of fifty-six 2D chalcogenide materials that identifies synthetic targets of potential interest to the Platform and broader materials community based on predicted extreme piezoelectric response.

- Local User Project R0002

J.R. Shallenberger, “2D tungsten diselenide analyzed by XPS,” *Surf. Sci. Spectra* 25, 014001 (2018). 10.1116/1.5016189
Development of XPS protocols for the analysis of 2D transition metal dichalcogenides, in concert with and in support of external users.

- Local user participating on external user project R0011

In-house Research Publications (AdvCM)

A.M.Z. Tan, M.A. Garcia, **R.G. Hennig**, “Giant Stokes Shift for Charged Vacancies in Monolayer SnS,” *Physical Review Materials* 6 (4), 044003 (2022). 10.1103/PhysRevMaterials.6.044003.
First-principles density-functional theory study to determine the equilibrium defect structures, formation energies, charge transition levels, and electronic structures of Sn and S vacancies in monolayer SnS. Both Sn and S vacancies exhibit multiple charge transition levels and in-gap defect states, indicating that they may be stable in different charge states depending on the Fermi level in the system.

N. Nayir, M.Y. Sengul, A.L. Costine, P. Reinke, S. Rajabpour, A. Bansal, A. Kozhakhmetov, **J.A. Robinson**, **J.M. Redwing**, **A.C.T. van Duin**, “Atomic-scale probing of defect-assisted Ga intercalation through graphene using ReaxFF molecular dynamics simulations,” *Carbon* 190, 276-290 (2022). 10.1016/j.carbon.2022.01.005.
This work provides an in-depth atomic scale understanding into the complex interplay between defects and precursors, thus providing an effective way to design defects for 2D metal fabrication. It is a joint theory and experimental investigation on the defect-mediated surface interactions of gallium (Ga) metals and trimethyl-gallium (TMGa) molecules with graphene. Experimental results are connected to ReaxFF simulations, which further confirm that the Ga and TMGa

adsorption on graphene is strongly impacted by the presence and size of defects. Non-MIP equipment was used for synthesis and ReaxFF parameters developed by MIP were used.

A. Lele, P. Krstic, **A.C.T. van Duin**, “ReaxFF Force Field Development for Gas-Phase hBN Nanostructure Synthesis,” *Journal of Physical Chemistry A* 126 (4), 568-582 (2022). 10.1021/acs.jpca.1c09648.
Two-dimensional (2D) hexagonal boron nitride materials are isomorphs of carbon nanomaterials and hold promise for electronics applications owing to their unique properties. Understanding the growth mechanism of BN nanostructures through modeling and experiments is key to improving its widespread production. This work presents the development of a ReaxFF-based force field capable of modeling the gas-phase chemistry important for the chemical vapor deposition (CVD) synthesis process.

B. Rijal, A.M.Z. Tan, C. Freysoldt, **R.G. Hennig**, “Charged vacancy defects in monolayer phosphorene,” *Physical Review Materials* 5 (12), 124004 (2021). 10.1103/PhysRevMaterials.5.124004.
Two-dimensional semiconductor phosphorene has attracted extensive research interests for potential applications in optoelectronics, spintronics, catalysis, sensors, and energy conversion. To harness phosphorene's potential requires a better understanding of how intrinsic defects control carrier concentration, character, and mobility. Using density functional theory and a charge correction scheme to account for the appropriate boundary conditions, this comprehensive study elucidates the effect of structure on the formation energy, electronic structure, and charge transition level of the charged vacancy point defects in phosphorene.

D. Akbarian, N. Nayir, **A.C.T. van Duin**, “Understanding physical chemistry of $Ba_xSr_{1-x}TiO_3$ using ReaxFF molecular dynamics simulations,” *Physical Chemistry Chemical Physics* 23 (44), 25056-25062 (2021). 10.1039/d1cp03353k.
This study developed a ReaxFF reactive force field verified against quantum mechanical data to investigate the temperature and composition dependency of BSTO in non-ferroelectric/ferroelectric phases. This potential was also explicitly designed to capture the surface energetics of STO with SrO and TiO_2 terminations. This is an important study for the 2DCC in understanding substrate materials in a number of semiconductor applications.

N. Nayir, **Y. Wang**, Y. Ji, **T.H. Choudhury**, **J.M. Redwing**, L.Q. Chen, **V.H. Crespi** and **A.C.T. van Duin**, “Theoretical modeling of edge-controlled growth kinetics and structural engineering of 2D-$MoSe_2$,” *Materials Science and Engineering: B* 271, 115263 (2021). 10.1016/j.mseb.2021.115263.
This study reports the first reactive force field (ReaxFF) for Mo/Se/H interactions, which enables large-scale molecular dynamics simulations of the synthesis, processing, and characterization of 2D-$MoSe_2$ and whose parameters are trained primarily on first-principles energetics data including both periodic and non-periodic calculations. This new potential elucidates the structural transition from metallic to semiconducting phases, the energetics of various defects, and the Se-vacancy migration barrier. $MoSe_2$ materials used in the analysis were synthesized by the MIP equipment MOCVD1.

- Also science driver Epi2DC

A. Verma, W. Zhang, **A.C.T. van Duin**, “ReaxFF reactive molecular dynamics simulations to study the interfacial dynamics between defective h-BN nanosheets and water nanodroplets,” *Physical Chemistry Chemical Physics* 23, 10822-10834 (2021). 10.1039/d1cp00546d.

This paper describes the development of a reactive force field (ReaxFF) description for hexagonal boron nitride (h-BN) and the effect of water molecules on the interfacial interactions with vacancy defective hexagonal boron nitride (h-BN) nanosheets by introducing parameters suitable for the B/N/O/H chemistry. This study provides important information for the use of h-BN nanosheets in nanodevices for water desalination and underwater applications, as these h-BN nanosheets possess the desired adsorption capability and structural stability.

S. Rajabpour, Q. Mao, N. Nayir, **J.A. Robinson**, **A.C.T. van Duin**, "Development and Applications of ReaxFF Reactive Force Fields for Group-III Gas-Phase Precursors and Surface Reactions with Graphene in Metal-Organic Chemical Vapor Deposition Synthesis," *Journal of Physical Chemistry C* (125 (19), 10747-10758 (2021). 10.1021/acs.jpcc.1c01965.

In this paper, two ReaxFF reactive force fields are reported, GaCH-2020 and InCH- 2020, which were developed to investigate the , metal−organic chemical vapor deposition (MOCVD) gas-phase reactions of Ga and In film growth from trimethylgallium (TMGa) and trimethylindium (TMIn) precursors, respectively, and the surface interactions of TMGa and TMIn with graphene. The newly developed force fields were applied to determine the optimal conditions for the thermal decomposition of TMGa/TMIn to achieve Ga/In nanoclusters with low impurities. Additionally, the cluster formation of Ga/In on a graphene substrate with different vacancies and edges was studied with ReaxFF, providing targets for future experimental work. Data from non-MIP MOCVD equipment was used in analysis.

- Also science Driver Epi2DC

M.Y. Sengul, Y. Song, N. Nayir, Y. Gao, Y. Hung, T. Dasgupta, **A.C.T. van Duin**, "INDEEDopt: a deep learning-based ReaxFF parameterization framework," *npj Computational Materials* 7, 68 (2021). 10.1038/s41524-021-00534-4.

Complex empirical interatomic potentials, like ReaxFF, require optimization of many force field parameters to tune interatomic interactions to mimic ones obtained by quantum chemistry-based methods. Here, we report an INitial-DEsign Enhanced Deep learning-based OPTimization (INDEEDopt) framework to accelerate and improve the quality of the ReaxFF parameterization. The procedure starts with a Latin Hypercube Design (LHD) algorithm that is used to explore the parameter landscape extensively. The LHD passes the information about explored regions to a deep learning model, which finds the minimum discrepancy regions, eliminates unfeasible regions and constructs a more comprehensive understanding of physically meaningful parameter space. We demonstrate the procedure here for the parameterization of a nickel–chromium binary force field and a tungsten–sulfide–carbon–oxygen–hydrogen quinary force field. We show that INDEEDopt produces improved accuracies in shorter development time compared to the conventional ReaxFF optimization methods.

M. Kowalik, M. J. Hossain, A. Lele, W. Zhu, R. Banerjee, T. Granzier-Nakajima, **M. Terrones**, **E.W. Hudson**, **A.C.T van Duin**, "Atomistic-Scale Simulations on Graphene Bending Near a Copper Surface," *Catalysts* 11 (2), 208 (2021). 10.3390/catal11020208.

This paper validates the capability of ReaxFF to reproduce complex graphite bending patterns near metal support surfaces. Using ReaxFF reactive molecular simulations, we have investigated the possible bending of graphene in vacuum and near copper surfaces. We describe the energy cost for graphene bending and the binding energy with hydrogen and copper with two different ReaxFF parameter sets, demonstrating the relevance of using the more recently developed ReaxFF parameter sets for graphene properties. Moreover, the draping angle at copper step edges obtained from our atomistic simulations is in good agreement with the draping angle determined from experimental measurements, thus validating the ReaxFF results.

- Also science Driver Epi2DC

W. Zhang, **A.C.T. van Duin**, "Atomistic-Scale Simulations of the Graphene Growth on a Silicon Carbide Substrate Using Thermal Decomposition and Chemical Vapor Deposition," *Chemistry of Materials* 32 (19), 8306-8317 (2020). 10.1021/acs.chemmater.0c02121.

This work addresses the key first step in CHet synthesis of novel 2D systems through the development and application of reactive force fields adapted to the specific physical processes active during the growth of the graphene layer that acts as a "superstrate" to the subsequent growth of CHet materials, combined with treatment of the SiC substrate and the interaction between the two. Optimizations in reactive potentials are available to the community.

- Also science driver Epi2DC

A.M.Z. Tan, C. Freysoldt, **R.G. Hennig**, "First-principles investigation of charged dopants and dopant-vacancy defect complexes in monolayer MoS_2," *Physical Review Materials* 4 (11), 114002 (2020). 10.1103/PhysRevMaterials.4.114002.

This work advances understanding of defects and dopants in 2D chalcogenides through first-principles simulation, which correlated strongly with experimental characterization of these systems and optimization of growth to control defect and dopant properties. Defect properties are an important component of developing data resources.

- Also science driver Epi2DC

F. Zhang, B. Zheng, A. Sebastian, D.H. Olson, M. Liu, K. Fujisawa, Y.T.H. Pham, V.O. Jimenez, V. Kalappattil, L. Miao, T. Zhang, R. Pendurthi, Y. Lei, A.L. Elias, **Y. Wang**, **N. Alem**, P.E. Hopkins, *S. Das*, **V.H. Crespi**, M.-H. Phan, **M. Terrones**, "Monolayer Vanadium-Doped Tungsten Disulfide: A Room-Temperature Dilute Magnetic Semiconductor," *Advanced Science* 7 (24), 200174 (2020). 10.1002/advs.202001174.

This closely coupled experimental and theoretical work employs and investigates the role of dopants and defects in inducing ferromagnetism in 2D semiconductors, with careful consideration of the role of defect-defect coupling. This fundamental study initiates a pathway towards possible 2D magnetic semiconducting devices.

- Also science drivers Epi2DC and NGDev

N. Nayir, **Y. Wang**, S. Shabnam, **D.R. Hickey**, L. Miao, X. Zhang, S. Bachu, **N. Alem**, **J. Redwing**, **V.H. Crespi**, **A.C.T. van Duin**, "Modeling for Structural Engineering and Synthesis of Two-Dimensional WSe_2 Using a Newly Developed ReaxFF Reactive Force Field," *Journal of Physical Chemistry C* 124 (51), 28285-28297 (2020). 10.1021/acs.jpcc.0c09155.

This work combines experimental validation with computational development of new reactive force fields for chalcogenide systems that can interrogate questions of synthesis and post-synthesis annealing, structural modification, and environmental interactions. These new reactive potentials add to the suite currently available to the community.

K. Burns, A.M.Z. Tan, H. Gordon, T.Y. Wang, A. Gabriel, L. Shao, **R.G. Hennig**, A. Aitkaliyeva, "Strain modulation using defects in two-dimensional MoS_2," *Physical Review B*, 102 (8), 085421 (2020). 10.1103/PhysRevB.102.085421.

The interaction of tensile and compressive strain and ion irradiation on the formation and evolution of defects in 2D transition metal dichalcogenides, including the crystalline-to-amorphous transition, is elucidated through first-principles calculations to establish insights on new ways to modify the properties of 2D materials.

A.M.Z. Tan, C. Freysoldt, **R.G. Hennig**, "Stability of charged sulfur vacancies in 2D and bulk MoS_2 from plane-wave density functional theory with electrostatic corrections", *Phys. Rev. Mater.* 4, 064004 (2020). 10.1103/PhysRevMaterials.4.064004
Computational investigation of the formation energies and symmetry-lowering relaxations of charged chalcogenide vacancies in transition metal dichalcogenides, a major target in optimization of materials quality.

K. Burns, A.M.Z. Tan, A. Gabriel, L. Shao, **R.G. Hennig**, A. Aitkaliyeva, "Controlling neutral and charged excitons in MoS_2 with defects," *Journal of Materials Research*, 35 (8), 949-957 (2020). 10.1557/jmr.2019.404.

The platform-supported component of this joint experimental/computational work comprises first-principles calculations of defect properties in 2D transition metal dichalcogenides to elucidate formation energies, charge state, and influence on optical response.

Y. Sun, **Y. Wang**, J.Y.C. Chen, K. Fujisawa, C.F. Holder, J.T. Miller, **V.H. Crespi**, **M. Terrones**, R.E. Schaak, "Interface-mediated noble metal deposition on transition metal dichalcogenide nanostructures," *Nature Chemistry*, 12, 284-293 (2020). 10.1038/s41557-020-0418-3

The platform contributed in-house first-principles computations on the energetics and kinetics of metal deposition onto two-dimensional transition metal dichalcogenides in a close theory/experiment collaboration that advances Platform goals in accelerating the development of device applications, here through understanding the metal/2D interface, which is crucial for contact formation.

- Also science driver NGDev

B.R. Carvalho, **Y. Wang**, K. Fujisawa, T. Zhang, E. Kahn, I. Bilgin, P.M. Ajayan, A.M. de Paula, M.A. Pimenta, S. Kar, **V.H. Crespi**, **M. Terrones**, L.M. Malard, "Nonlinear dark-field imaging of one-dimensional defects in monolayer dichalcogenides," *Nano Letters*, 20 (1), 284-291 (2020). 10.1021/acs.nanolett.9b03795.

In-house first-principles calculations here closely couple to experimental work to demonstrate a means to optically image atomic-scale defects in two-dimensional transition metal dichalcogenides; this work relates to Platform goals in understanding and controlling defects in

2D materials with special focus on high-throughput optical methods whose application is enabled or facilitated by supporting first-principles computations.

Y. Xuan, A. Jain, S. Zafar, R. Lotfi, N. Nayir, **Y. Wang**, **T.H. Choudhury**, S. Wright, J. Feraca, L. Rosenbaum, **J.M. Redwing**, **V. Crespi**, **A. van Duin** "Multi-scale modeling of gas-phase reactions in metal-organic chemical vapor deposition growth of WSe_2," *Science Direct*, 527, 125247, (2019). 10.1016/j.jcrysgro.2019.125247

This comprehensive multi-disciplinary computational framework helps to advance the understanding of gas-phase kinetics in MOCVD synthesis of TMDs by combining first-principles methods, empirical atomistic reactive molecular dynamics, and computational fluid dynamics to efficiently model gas-phase physiochemical processes leading to WSe_2 growth in a cold-wall chamber whose geometry is designed to model the 2DCC tool used to provide many MOCVD-based user samples.

L.T. Alameda, R.W. Lord, J.A. Barr, P. Moradifar, Z.P. Metzger, B.C Steimle, C.F. Holder, N. Alem, **S.B. Sinnott**, R.E. Schaak, "Multi-Step Topochemical Pathway to Metastable Mo_2AlB_2 and Related Two-Dimensional Nanosheet Heterostructures," *J. Amer. Chem. Soc*., 141 (27), 10852-10861 (2019). 10.1021/jacs.9b04726

Study demonstrating that the combination of chemical destabilization, size-selective precipitation, and low-temperature annealing provides a potentially generalizable kinetic pathway to metastable variants of refractory compounds, including bulk Mo_2AlB_2 and Mo_2AlB_2–AlO_x nanosheet heterostructures, and opens the door to other previously elusive 2-D materials.

P. Zhao, **Y. Wang**, B. Katz, E. Mockensturm, **V.H. Crespi**, S. Zhang, "Geometry and chiral symmetry breaking of ripple junctions in 2D materials," *Journal of the Mechanics and Physics of Solids*, 131, 337-343 (2019). 10.1016/j.jmps.2019.07.007

Atomistic simulations of the mechanical response of deformed 2D materials with particular focus on distinct 2D morphologies such as ripples, whose formation (or suppression thereof) can play a key role in device fabrication from 2D materials, making use of intermolecular potentials developed by 2DCC personnel and provided to the community.

R. Rao, V. Carozo, **Y. Wang**, A.E. Islam, N. Perea-Lopez, K. Fujisawa, **V.H. Crespi**, **M. Terrones**, B. Maruyama, "Dynamics of cleaning, passivating and doping monolayer MoS_2 by controlled laser irradiation," *2D Materials*, 6, 45031 (2019). 10.1088/2053-1583/ab33ab

In situ study that elucidates the passivation mechanism in TMDs upon laser irradiation and demonstrates a way to controllably n-dope CVD-grown monolayer MoS_2 on SiO_2 substrates, with in-house 2DCC theory/computation work in close concert with experiment.

F. Zhang, **Y. Wang**, C. Erb, K. Wang, P. Moradifar, **V. H. Crespi**, N. Alem, "Full orientation control of epitaxial MoS_2 on hBN assisted by substrate defects", *Phys. Rev. B*, (2019), 99, 155430. 10.1103/PhysRevB.99.155430

Joint experiment/theory discovery of a defect-complex mechanism that results in a preferred orientation for transition metal dichalcogenides grown epitaxially on hexagonal boron nitride, providing insights towards achieving single-crystal monolayers of materials relevant to 2DCC mission, performed using 2DCC Theory/Simulation facility. Insights deriving from these results inform MOCVD synthesis efforts on samples for 2DCC users.

Y. Yuan, Y. Lu, G. Stone, K. Wang, C.M. Brooks, D.G. Schlom, **S.B. Sinnott**, H. Zhou, V. Gopalan, "Three-dimensional atomic scale electron density reconstruction of octahedral tilt epitaxy in functional perovskites," *Nature Comm*. 9, 5220 (2018). 10.1038/s41467-018-07665-1

Combined experimental and theoretical study of octahedral tilts and polar distortions at perovskite interfaces including collaborators from 2DCC and PARADIM.

Z. Zhang, **Y. Wang**, X.X. Leng, **V. H. Crespi**, F. Kang, and R. Lv, "Controllable Edge Exposure of MoS_2 for Efficient Hydrogen Evolution with High Current Density," ACS Appl. Energy Mater. 1(3), 1268–1275 (2018). 10.1021/acsaem.8b00010

Joint experimental/computational effort on the catalytic properties of the edges of 2D transition metal dichalcogenides, of relevance for both application and understanding and controlling edge exposure and edge properties in these systems, using 2DCC Theory/Simulation facility.

Y.J. Tang, C.I. Chia, and **V. H. Crespi**, "Dual-Sided Adsorption: Devil's Staircase of Coverage Fractions," *Phys. Rev. Lett*. 120, 056101 (2018). 10.1103/PhysRevLett.120.056101

Theoretical and computational proposal for a novel 2D system formed from adsorption onto a suspended 2D monolayer, with a general scheme that could apply to any sufficiently thin semiconducting or insulator 2D layer, performed using 2DCC Theory/Simulation facility.

M. Hasanian, B. Mortazavi, A. Ostadhossein, T. Rabczuk, and **A.C.T. van Duin**, "Hydrogenation and defect formation control the strength and ductility of MoS_2 nanosheets: Reactive molecular dynamics simulation," *Extreme Mech. Lett*. 22, 1570164 (2018). 10.1016/j.eml.2018.05.008

Investigation of defects and functionalization of 2D transition metal dichalcogenide thin films through reactive force field simulation performed in part by the 2DCC Theory/Simulation facility and using reactive force fields in the class developed under Platform support.

D.E. Yilmaz, **R. Lotfi**, C. Ashraf, S.W. Hong and **A.C.T. van Duin**, "Defect design of two-dimensional MoS_2 structures by using a graphene layer and potato stamp concept," J. *Phys. Chem. C*, 122(22), 11911-11917 (2018). 10.1021/acs.jpcc.8b02991

Computational development of a new controlled defect induction concept utilizing adhesion of 2D chalcogenide monolayers through reactive force field simulation carried out using the 2DCC Theory/Simulation facility. This work advances general understanding of defect properties in 2D materials and their description at an empirical potential level.

F.A. Soria, W.W. Zhang, P.A. Paredes-Olivera, **A.C.T. van Duin** and E.M. Patrito, "Si/C/H ReaxFF reactive potential for silicon surfaces grafted with organic molecules," *J. Phys. Chem. C.* 122 (41), 23515-23527 (2018). 10.1021/acs.jpcc.8b07075

Development of reactive force fields to handle silicon, carbon, and hydrogen of relevance to platform efforts on confinement heteroepitaxy, a novel means of growing new types of 2D materials.

Y. Wang, B.R. Carvalho, **V.H. Crespi**, "Strong exciton regulation of Raman scattering in monolayer MoS_2," *Phys. Rev. B*. 98 (16), 161405 (2018). 10.1103/PhysRevB.98.161405

Development of new theoretical/computational tools to understand and interpret optical response of 2D systems, in close concert with experiment, to enhance capabilities of interpretation of in situ and ex situ platform optical probes, performed using 2DCC Theory/Simulation facility and of particular interest to applications and fundamental phenomena exploiting the excitonic optical response of 2D TMDs.

A. Ostadhossein, A. Rahnamoun, **Y. Wang**, P. Zhao, S. Zhang, **V.H. Crespi**, and **A.C.T. van Duin**, "ReaxFF Reactive Force-Field Study of Molybdenum Disulfide (MoS_2)", *Journal of Physical Chemistry Letters* **2017**, *8*, 631–640. 10.1021/acs.jpclett.6b02902

The first reactive potential to describe TMD systems, of broad general utility in simulations of kinetic processes e.g. (growth) and also structural distortions of TMDs, with initial application to ripple deformations; this potential is available to users through the 2DCC website, with extensions to other metals, chalcogens and also substrate interactions completed or underway in the Platform.

C-X. Liu, "Unconventional Superconductivity in Bilayer Transition Metal Dichalcogenides", *Phys Rev. Lett.* **2017**, *118*, 087001. 10.1103/PhysRevLett.118.087001

Theoretical study predicting superconducting phases in bilayer transition metal dichalcogenides.

- Also science driver Phys2D

Y. Wang and **V. H. Crespi**, "Theory of Finite-Length Grain Boundaries of Controlled Misfit Angle in Two-Dimensional Materials", *Nano Letters* **2017**, *17*, 5297. 10.1021/acs.nanolett.7b01641

Theory-driven proposal for a general mechanism of grain boundary engineering in a 2D material, which could provide a way to place grain boundaries of desired misfit angles at desired locations, performed using 2DCC Theory/Simulation facility. We are currently extending this theory as a possible route to growing multilayer magic angles, encouraged by preliminary experimental results that suggest certain 2D materials may support growth modes that are conducive to this mechanism.

Y. Wang and **V. Crespi**, "NanoVelcro: Theory of Guided Folding in Atomically Thin Sheets with Regions of Complementary Doping", *Nano Letters* **2017**, *17* (11), 6708-6714. 10.1021/acs.nanolett.7b02773

Theory-driven methodology to program a folding structure into an arbitrary 2D semimetallic or semiconducting system by applying key concepts from origami to complementary p and n type doping, using 2DCC Theory/Simulation facility.

A. McCreary, J. Simpson, **Y. Wang**, D. Rhodes, K. Fujisawa, L. Balicas, M. Dubey, **V. Crespi**, **M. Terrones**, and A. Hight Walker, "Intricate Resonant Raman Response in Anisotropic ReS_2", *Nano Lett.* **2017**, *17*, 5897−5907. 10.1021/acs.nanolett.7b01463

The first calculation of resonant Raman response in a Rhenium-based TMD in close collaboration with experiment, identifying the origins of a complex assembly of Raman modes in this low-symmetry 2D chalcogenide. This work extends the suite of 2D chalcogenides for which we are able to interpret optical probes and uses the 2DCC Theory/Simulation facility.

V. Carozo, **Y. Wang**, K. Fujisawa, B. R. Carvalho, A. McCreary, S. Feng, Z. Lin, C. Zhou, N. Perea-López, A. L. Elías, B. Kabius, **V. H. Crespi**, and **M. Terrones**, "Optical identification of sulfur vacancies: Bound excitons at the edges of monolayer tungsten disulfide", *Sci. Adv.* **2017**, *3*, e1602813. 10.1126/sciadv.1602813

A methodology to identify important defects in TMDs through rapid optical spectroscopic characterization, and elucidation of the mechanisms of exciton/defect binding, using 2DCC Theory/Simulation facility and supportive of optical characterization of thin films produced by the Platform.

B. R. Carvalho, **Y. Wang**, S. Mignuzzi, D. Roy, **M. Terrones**, C. Fantini, **V. H. Crespi**, L. M. Malard, and M. A. Pimenta, "Intervalley scattering by acoustic phonons in two-dimensional MoS_2 revealed by double-resonance Raman spectroscopy", *Nature. Commun.* **2017**, *8*, 14670. 10.1038/ncomms14670

Elucidation of the correct resonant intervalley origin for key Raman modes in TMD MoS_2 through close theory/experiment collaboration, an effort that provides guidance for the interpretation of optical characterization of samples produced by 2DCC and in the community at large, using 2DCC Theory/Simulation facility.

A. Azizi, **Y. Wang**, G. Stone, A. L. Elias, Z. Lin, **M. Terrones**, **V. H. Crespi**, and N. Alem, "Defect Coupling and Sub-Angstrom Structural Distortions in $W_{1-x}Mo_xS_2$Monolayers", *Nano Lett.* **2017**, *17*, 2802. 10.1021/acs.nanolett.6b05045

Experimental and theoretical study demonstrating coupling of vacancies and metal atoms in transition metal dichalcogenide alloys carried out by 2DCC Thin Films and Theory/Simulation facilities. This work advances the understanding of defects in 2D materials, which is important for optimizing growth and understanding properties of thin films produced by the platform and the community at large.

A. Azizi, **Y. Wang**, Z. Lin, K. Wang, A.L. Elias, **M. Terrones**, **V.H. Crespi**, and N. Alem, "Spontaneous Formation of Atomically Thin Stripes in Transition Metal Dichalcogenide Monolayers," *Nano Lett.* **2016**, 16 (11), 6982−6987. 10.1021/acs.nanolett.6b03075

Experimental and theoretical study of atomic scale ordering in 2D transition metal dichalcogenide alloys carried out by collaborators in 2DCC Thin Films and Theory/Simulation facilities; this work has guided later exploratory synthetic efforts to exploit the phenomena therein discerned to potentially create sharp, thin lateral heterostructures.

- Also science driver Epi2DC

S-L. Shang, G. Lindwall, **Y. Wang**, **J.M. Redwing**, T. Anderson, and Z-K. Liu, "Lateral Versus Vertical Growth of Two-Dimensional Layered Transition-Metal Dichalcogenides: Thermodynamic Insight into MoS_2," *Nano Lett.* **2016**, 16 (9), 5742-5750. 10.1021/acs.nanolett.6b02443

Thermodynamic investigation into the effects of processing conditions on the growth mode of transition metal dichalcogenide films carried out in collaboration with 2DCC Thin Films facility.