Advanced Characterization and Modeling (AdvCM)

External User Publications (AdvCM)

K. Momeni, Y. Ji, N. Nayir, N. Sakib, H. Zhu, S. Paul, **T.H. Choudhury**, S. Neshani, **A.C.T van Duin**, **J.M. Redwing**, L.-Q. Chen, "A computational framework for guiding the MOCVD-growth of wafer-scale 2D materials," *npj Computational Materials* 8 (1), 240 (2022). 10.1038/s41524-022-00936-y.

A multiscale model for WSe_2 MOCVD was developed using ReaxFF, CFD and Phase field methods. At the atomic scale, using reactive MD simulations based on DFT calculations, it provides the relationship between the ratio of precursor concentrations and stable morphology of 2D materials; At the macroscale, it connects experimentally controllable parameters with concentration of precursor and its gradient over the substrate; It combines the information from both nanoscale and macroscale simulations to predict the coverage and uniformity of as-grown 2D materials. This computational framework provides a unique alternative to exhaustive trial-and-error experimentations and a powerful tool to develop and optimize the synthesis of new 2D materials. It can further serve as an observer for controllers of the growth process, providing the feedback loop capability, thus, precise control over the growth process, which opens new routes to design and fabricate the next generation of nanodevices for application in quantum computing and artificial intelligence. The 2DCC Theory/Simulation/Data facility provided the atomic-scale (DFT, ReaxFF) expertise to provide parameters for CFD simulations carried out by the external user. The wafer-scale growth of WSe_2 films was carried out by MOCVD1 in the 2DCC Thin Films facility.

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

N. Sakib, S. Paul, N. Nayir, **A.C.T. van Duin**, S. Neshani, K. Momeni, "Role of tilt grain boundaries on the structural integrity of WSe_2 monolayers," *Physical Chemistry Chemical Physics* 24, 27241-27249 (2022). 10.1039/d2cp03492a.

The role of grain boundary defects with varying misorientation angles on the mechanical properties of WSe_2 at different temperatures was investigated using MD simulations. The results presented here provide a fundamental understanding of the role of grain boundaries in bi-atomic monolayers, such as TMDCs, on their final mechanical performance. Thus, it paves the way to design next-generation optoelectronic devices. The 2DCC computational allocation on ICDS was used for ReaxFF simulations.

- External User Project R0037 (Non-R1)

Q. Mao, Y. Zhang, **M. Kowalik**, N. Nayir, M. Chandross, **A.C.T. van Duin**, "Oxidation and hydrogenation of monolayer MoS_2 with compositing agent under environmental exposure: The ReaxFF Mo/Ti/Au/O/S/H force field development and applications," *Frontiers in Nanotechnology* 4 (2022). 10.3389/fnano.2022.1034795.

This study developed a new ReaxFF reactive force field for Mo, Ti, Au, O, S, and H (ReaxFF Mo/Ti/Au/O/S/H-2022) to investigate the role of Ti clusters in preventing the oxidation and hydrogenation of a monolayer MoS_2 surface in O_2- and H_2O-rich environments. The ReaxFF molecular dynamics (MD) simulations reveal the oxidation and hydrogenation mechanisms for

the MoS_2 surfaces exposed to O_2 and H_2O environments with and without the presence of Ti clusters. The 2DCC's allocations on the PSU's Institute for Computational and Data Sciences - Advanced Cyber Infrastructure (ICDS-ACI) cluster enabled the ReaxFF force field training and the series of MD simulations for this work.

- External User Project R0051 (National Lab)

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)

Y. Wang, **V.H. Crespi**, M.L. Cohen, A. Nourhani, "Nonstoichiometric Salt Intercalation as a Means to Stabliize Alkali Doping of 2D Materials," *Physical Review Letters* 129, 266401 (2022). 10.1103/PhysRevLett.129.266401.
This study demonstrates a means to dope layered materials that stabilizes the doped layers against oxidation (making them more useful for applications) and also decoupling the individual layers from each other by means of the insulating salt slabs intercalated between them, allowing one to recover monolayer-like behavior in bulk materials. Many intriguing properties of 2D materials have limited application due to the thinness of the samples. If such properties (such as a direct bandgaps or 2D superconductivity) can be achieved in bulk materials by decoupling the layers, it may open the scope of possible applications for 2D and layered materials. This publication features a combination of in-house and external user expertise in computational materials design and use of 2DCC's allocations on the ICDS cluster.

- In-house collaboration with external user R0076 (R1)

B.N. Katz, L. Krainov, **V. Crespi**, "Shape Entropy of a Reconfigurable Ising Surface," *Physical Review Letters* 129, 096102 (2022). 10.1103/PhysRevLett.129.096102.
This theoretical study addresses disclinations in a 2D sheet of graphene. The study demonstrated regions of Gaussian curvature whose inversion produces a reconfigurable surface with many distinct metastable shapes, as shown by molecular dynamics of a disclinated graphene monolayer. This material has a near-Gaussian "density of shapes" and an effectively antiferromagnetic interaction between adjacent cones. As this approach is purely geometrical, it should apply to any atomically thin sheet, which could be polar (h-BN), hydrophilic (graphene oxide), or stiffer than graphene. This publication includes use of the 2DCC's allocation on the ICDS cluster.

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.