Materials Synthesis & Characterization

Defect-free Epitaxial Growth of GaAs on Silicon via Nanopatterning

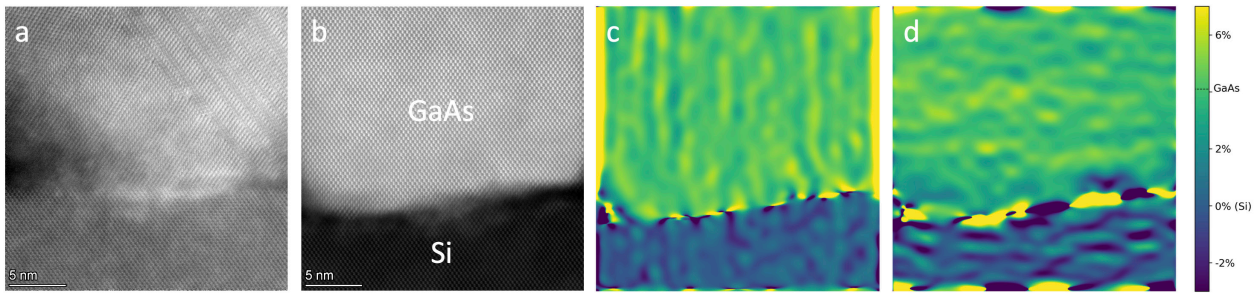
C. Chen, K. Lu, D. Kwon, N. Han, J. Kim, F. M. Ross
Sponsorship: Defense Advanced Research Projects Agency

The advancement of semiconductor technology demands integrating the traditional microelectronic material, Si, with materials that can enhance the performance of devices, e.g., by improving speed. III-V semiconductors like GaAs, with their superior electron mobility, are promising for next-generation transistors. However, when growing GaAs on Si, the difference in atomic spacing between their lattices leads to the formation of interfacial defects that degrade device performance.

To overcome this, we grow GaAs on Si in confined areas, over which the lattice mismatch is small enough that interfacial defects don't form; an array of GaAs islands grows on Si. However, as we continue growing the islands up to coalescence, the contact area between GaAs and Si increases, and interfacial defects form. We thus pattern an interlayer on the Si before GaAs deposition, so that the GaAs and Si only make direct contact through small holes in the interlayer. The GaAs then grows across the interlayer. By optimizing the interlayer, this process of island formation followed by GaAs lateral epitaxial overgrowth lead to defect-free interfaces in a process compatible with standard Si technology.

Measurements of the island and interface structure obtained using scanning transmission electron microscopy (STEM) guide our growth optimization. We use the Thermo Fisher Scientific Themis Z and prepare samples with the FEI Helios 600, both housed in Characterization.nano, to directly visualize atomic arrangements, identify defects such as dislocations and stacking faults (shown in Figure 1a), and map the local strain distribution (Figure 1b-d). Remarkably, geometric phase analysis of high-resolution STEM images reveals that GaAs nuclei can be fully relaxed, despite the lattice mismatch.

By leveraging advanced STEM characterization techniques, we are refining growth strategies to minimize defect formation. These findings open a new path toward integrating III-V materials on Si, so we may realize high-speed, energy-efficient semiconductor devices for future electronics.



▲ Figure 1: STEM images and geometric phase analysis of GaAs on Si. (a) Misaligned region showing polycrystallinity and stacking faults. (b) Defect-free single-crystalline GaAs. (c,d) Strain maps showing full relaxation of the GaAs nucleus in x- and y-directions.

FURTHER READING

- S. H. Bae, K. Lu, Y. Han, S. Kim, K. Qiao, C. Choi, Y. Nie, H. Kim, et al., "Graphene-assisted Spontaneous Relaxation Towards Dislocation-free Heteroepitaxy," *Nature Nanotechnology*, vol. 15, pp. 272–276, 2020. DOI: 10.1038/s41565-020-0633-5
- H. Kim, S. Lee, J. Shin, M. Zhu, M. Akl, K. Lu, N. M. Han, Y. Baek, et al., "Graphene Nanopattern as a Universal Epitaxy Platform for Single-Crystal Membrane Production and Defect Reduction," *Nature Nanotechnology*, vol. 17, pp. 1054–1059, 2022. DOI: 10.1038/s41565-022-01200-6

Defect Complexes in CrSBr Revealed through Electron Microscopy and Deep Learning

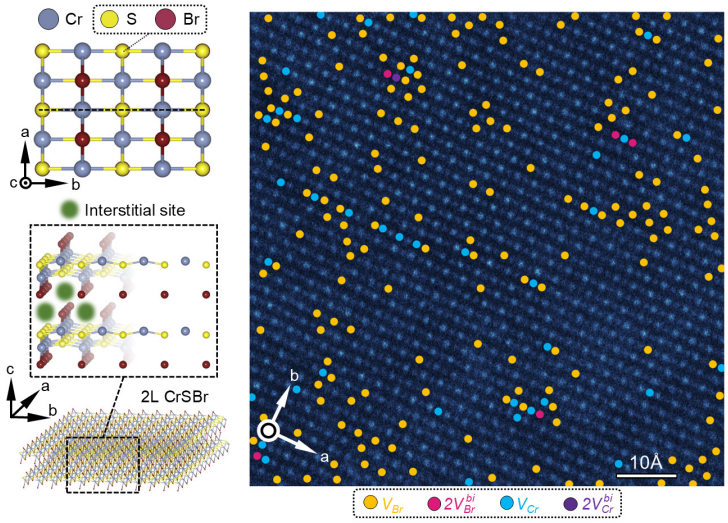
M. Weile, S. Grytsiuk, A. Penn, D. G. Chica, X. Roy, K. Mosina, Z. Sofer, J. Schiøtz, S. Helveg, M. Rösner, F. M. Ross, J. Klein

Atomic-scale defects influence the functional properties of layered materials. In this work, we present a comprehensive defect analysis in bilayer CrSBr, a magnetic quasi-1D van der Waals semiconductor of FeOCl-type symmetry, using a combination of atomic-resolution scanning transmission electron microscopy (STEM), deep learning, and ab-initio calculations.

To overcome beam sensitivity and low signal-to-noise ratios in bilayer CrSBr, we employ a low-dose STEM imaging approach combined with a custom machine learning pipeline for automated detection, classification, and statistical averaging of point defects. This workflow enables the identification of a diverse set of defect types, including single and stacked vacancy defects, interstitial Cr and Br vacancy defect complexes (V_{Cr} + Crint), Cr-Br vacancy complexes (V_{Cr} +V_{Br}), and extended defect lines aligned along the crystallographic a-direction. These features are often difficult to resolve using traditional methods but become visible through ML-assisted averaging across many images.

First-principle simulations support the experimental findings and provide insight into defect formation energies, structural relaxation, and electronic localization. Interstitial Cr defect complexes are shown to introduce highly localized in-gap electronic states with reduced dimensionality. These states are predicted to be optically active and magnetically tunable, offering potential for use in quantum light emitters, defect-based memory elements, or spintronic logic.

The methodology developed here is broadly applicable to other layered systems with limited defect visibility and motivates using STEM for targeted atomic-scale engineering of magnetic semiconductors. The insights gained here can be extended to over 20 related materials in the transition metal oxyhalide and chalcogenide halide families of FeOCl structural type.



▲ Figure 1: Schematic illustration of bilayer CrSBr. High-angle Annular Dark-field-STEM image with overlaid vacancy defects detected by deep convolutional neural networks.

FURTHER READING

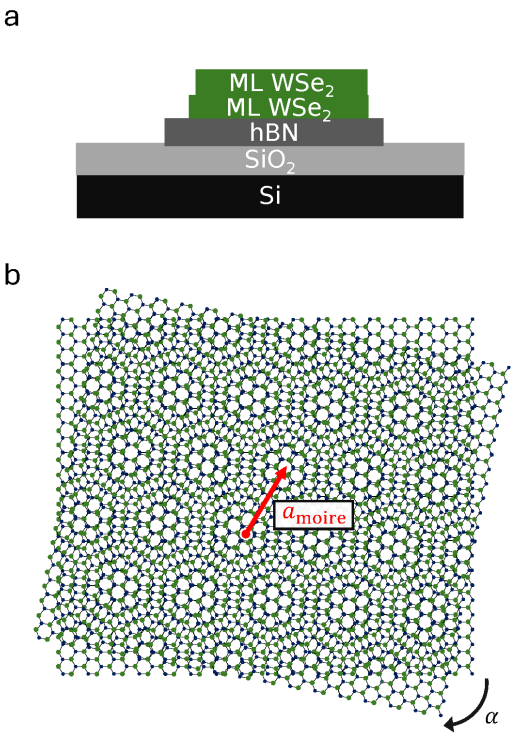
- M. Weile, S. Grytsiuk, A. Penn, D. G. Chica, X. Roy, K. Mosina, Z. Sofer, J. Schiøtz, S. Helveg, M. Rösner, F. M. Ross, and J. Klein, "Defect Complexes in CrSBr Revealed through Electron Microscopy and Deep Learning," *Physical Review X*, vol. 15, p. 021080, 2025.
- J. Klein, T. Pham, J. D. Thomsen, J. B. Curtis, T. Denneulin, M. Lorke, M. Florian, A. Steinhoff, R. A. Wiscons, J. Luxa, Z. Sofer, F. Jahnke, P. Narang, and F. M. Ross, "Control of Structure and Spin Texture in the van der Waals Layered Magnet CrSBr," *Nature Communications*, vol. 13, p. 5420, 2022.
- K. Torres, A. Kuc, L. Maschio, T. Pham, K. Reidy, L. Dekanovsky, Z. Sofer, F. M. Ross, and J. Klein, "Probing Defects and Spin-Phonon Coupling in CrSBr via Resonant Raman Scattering," *Adv. Funct. Mat.*, vol. 33, p. 2211366, 2023.

Twist Disorder in $tWSe_2$: Insights from Lateral Force Microscopy

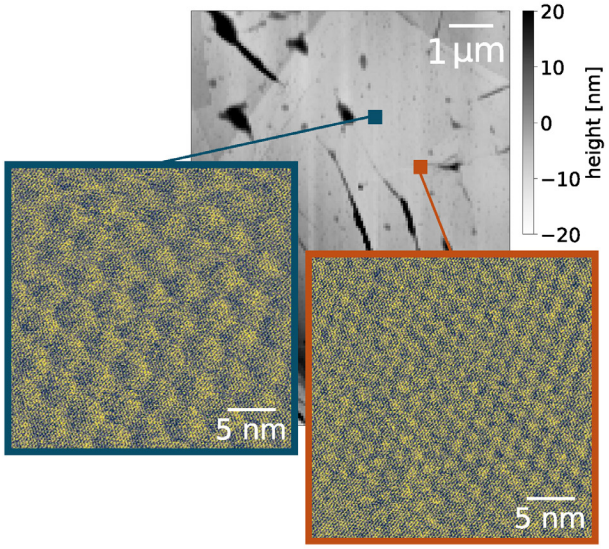
N.-L. Bathen, R. Dana, J. Klein, F. M. Ross, U. Wurstbauer
Sponsorship: NSF

Twisted bilayers of transition metal dichalcogenides (TMDs) form moiré superlattices and host electronic minibands, enabling the exploration of Mott-Hubbard physics and correlated quantum phases. These systems are typically fabricated by stacking van der Waals materials (Figure 1a) with a slight twist mismatch, creating a long-range moiré periodicity determined by the twist angle, as illustrated in Figure 1b. While previous scanning probe microscopy studies have shown that local strain and atomic relaxation can cause real-space variations in the moiré wavelength, these effects have not been systematically analyzed over micrometer-scale distances or across a broad range of twist angles. Such variations are expected to significantly influence optical experiments conducted in our labs.

Here, we use lateral force microscopy (LFM) mode to map the moiré lattice and relocate individual scans within a topographic map of the sample. Thus twist disorder can be visualized, as shown for two positions on the sample in Figure 2. The results reveal twist angle variations of up to several degrees within micrometer-scale distances. This analysis enables identification of homogeneous regions where optical and transport experiments can be reliably performed.



▲Figure 1: Sketch of investigated material system. (a) Stacking order of individual van der Waals materials. (b) Schematic sketch of two static hexagonal lattices with two atomic basis rotated in respect to each other, forming moiré lattice.



▲Figure 2: Height trace of investigated sample with relocated LFM scans, revealing moiré lattice at two individual positions on sample.

FURTHER READING

- S. Shabani, et al., "Deep Moiré Potentials In Twisted Transition Metal Dichalcogenide Bilayers," *Nat. Phys.*, vol. 17, pp. 720–725, 2021.
- N. Saigal, et al., "Collective Charge Excitations between Moiré Minibands in Twisted WSe_2 Bilayers Probed with Resonant Inelastic Light Scattering," *Phys. Rev. Lett.*, vol, 133, p. 046902, 2024.

Stress, Strain, and Temperature Tunability of 2D Metal Thiophosphate Materials Probed via Defect-induced Photoluminescence, X-ray, and Raman Spectroscopies

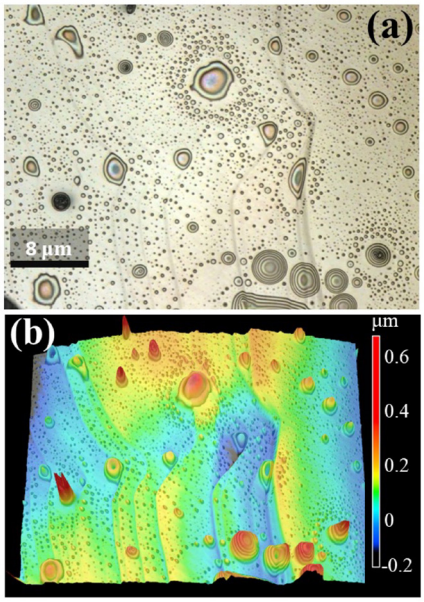
A. Mukherjee, S. V. Boriskina
Sponsorship: ARO-Multidisciplinary University Research Initiatives program, MIT International Science and Technology Initiatives-Poland Seed Fund, MIT-Monterrey Technology Program

Metal thiophosphates (MTPs) are a new family of intermediate-bandgap (1.3–3.5 eV) two-dimensional (2D) materials that exhibit diverse electronic, magnetic, and nonlinear optical properties and show promise for applications in energy harvesting, storage, and photo-detection. These properties are strongly influenced by the transition metal element within the MPTs.

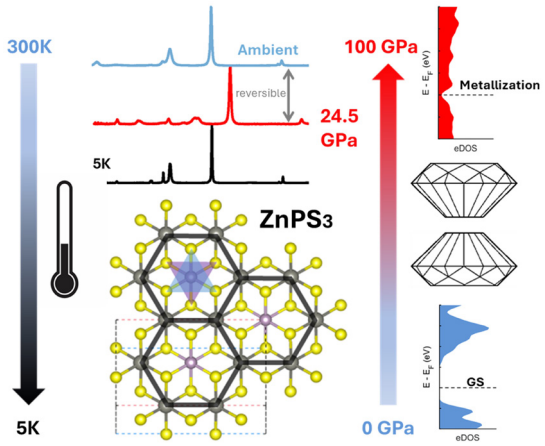
Sulfur vacancies and other defects in MTPs allow defect-state-to-valence-band transitions leading to visible light emission at sub-band gap energies. Photoluminescence (PL) measurements under a variety of external stimuli can shed light on the structural and electronic properties evolution in these materials and reveal material candidates to achieve high tunability or high stability under extreme conditions.

In collaboration with the Institute of Physics in Warsaw, the US Air Force Research Lab, and Technology de Monterrey, we showed experimentally that defect-mediated PL in $AgScP_2S_6$ can be enhanced and spectrally shaped by structural defects in the material (Figure 1). These defects form during material growth, originating from dislocations buried under surface planes, and exhibit varying thickness and inhomogeneous localized strain distribution. Our data also show that PL can be further enhanced and tuned via thermal annealing, which increases the density of sulfur vacancies, and by temperature-induced strain gradients.

In turn, zinc phosphorus trisulfide ($ZnPS_3$), demonstrated remarkable structural stability under extreme pressures and cryogenic temperatures (Figure 2). PL measurements and Raman spectroscopy revealed a fully reversible pressure-induced phase transition starting at ~7 GPa, after which $ZnPS_3$ demonstrates stability up to 24.5 GPa. Ab-initio density functional theory (DFT) calculations support these observations and predict a semiconductor-to-semimetal transition at 100 GPa. Cryogenic X-ray diffraction measurements revealed that $ZnPS_3$ has a high mean thermal expansion coefficient of about 4.4×10^{-5} K^{-1}, among the highest reported for 2D materials. This unique combination of tunable electronic properties under low pressure and high thermal sensitivity makes $ZnPS_3$ a strong candidate for sensing applications in extreme environments.



▲ Figure 1: (a) Morphology of an $AgScP_2S_6$ crystallite under optical microscope, which reveals multiple structural defects. (b) 3D height profile reconstruction using laser scanning confocal microscopy.



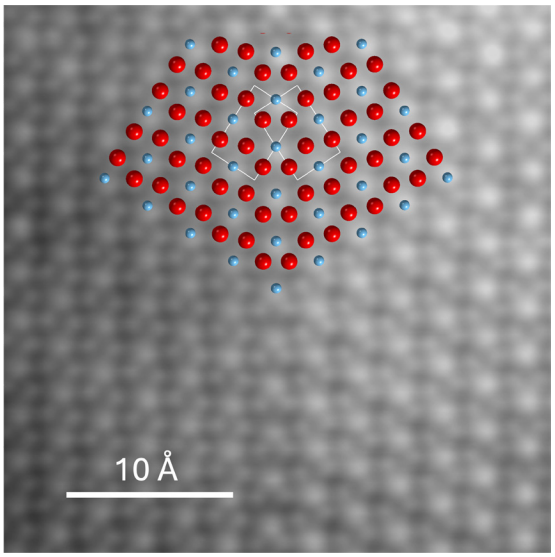
▲ Figure 2: $ZnPS_3$ undergoes reversible pressure-induced phase transition, followed by semiconductor-to-semimetal transition at 100 GPa. It exhibits defect-assisted emission, large Grüneisen parameter at cryogenic temperatures, and high thermal expansion coefficient of approximately 4.4×10^{-5} K^{-1}.

Electron Microscopy Studies of the Mechanisms of Epitaxial Titanium Oxide Growth on Graphene

P. Knight, F. M. Ross

The pressing need to reduce the energy used in computation is driving the creation of new designs for transistors and other computing architectures with reduced energy consumption. One promising concept is to use two-dimensional (2D) van der Waals materials as channel materials in transistor designs, which can be scaled to the atomically thin limit while avoiding short channel effects, thus minimizing static power consumption. Energy savings will also come from innovations in dielectric materials: for example, ferroelectric gate stacks that contain crystalline HfO_2 and ZrO_2 have recently been shown to exhibit "negative capacitance" effects that allow lower operating voltages by enabling a steeper switching transition between "on" and "off" states than is possible with conventional microelectronic materials. However, integrating any new material into advanced device designs faces the constraints of materials science and engineering. Here we need to create stacks of materials that include 2D layers with well-defined crystal structures and crystal orientations, perfect interfaces, and minimal defects. These problems are particularly challenging in the context of incorporating 2D materials into devices because of the known difficulties in nucleating 3D materials on van der Waals surfaces.

We quantify oxidation transformations on 2D materials of Ti-group metal oxides, all of which are of interest as next-generation dielectric materials. We aim for the self-assembly of an epitaxial metal layer, followed by oxidation of the metal via a series of epitaxial suboxides to form a fully oxidized phase. We use the Themis Z-STEM in MIT.nano to study the strain and defects within these oxide phases to better understand the effect of those defects on the dielectric properties of the material. Figure 1 shows an example visualizing the arrangement of Ti atoms (bright) and O atoms (less bright) in rutile TiO_2 at a twin boundary. We anticipate that polarization at these kinds of defects could enable use of Ti oxides as dielectric layers in devices. Our ongoing studies aim to extend understanding of nucleation and growth of epitaxial Ti oxides, which defects result, and what influence these defects will have on electronic device performance.



◀ Figure 1: (a) Morphology of an $AgScP_2S_6$ crystallite under optical microscope, which reveals multiple structural defects. (b) 3D height profile reconstruction using laser scanning confocal microscopy.

FURTHER READING

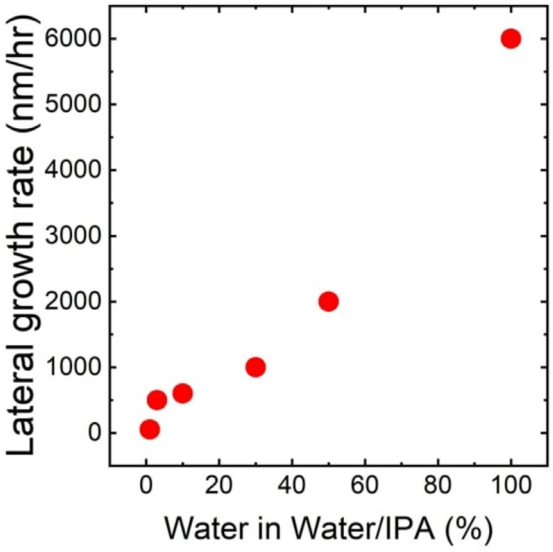
- P. J. Knight, K. Reidy, A. Penn, A. Foucher, and F. M. Ross, "In-situ Phase Transformations of 3D Nanoislands on 2D Materials in the Ti-Graphene System," presented at *Materials Research Society Fall Meeting*, Boston, MA, 2024.

Seeding Promoter Effect on Metal Organic Chemical Vapor Deposition Synthesized Molybdenum Disulfide

Y. Jiao, J. Zhu, T. Palacios
Sponsorship: Center for Heterogeneous Integration of Micro Electronic Systems (JUMP 2.0)

Two-dimensional (2D) transition metal dichalcogenide (TMD) materials are promising for the next generation electronics thanks to their excellent electronic and photonic properties. Metal organic chemical vapor deposition (MOCVD) can provide excellent uniformity and quality. Seeding promoters, such as sodium chloride (NaCl), have been reported to have significant improvement in the MOCVD growth rate and quality of molybdenum disulfide (MoS_2). Understanding the effect of seeding promoters can benefit overcoming limitations of the large-scale fabrication of 2D electronics.

Here, we quantitatively investigate the concentration dependence of seeding density, and flake quality. We also demonstrate material uniformity across the 200 mm platform and a boost in the average mobility to above 75 $cm^2/(V \cdot s)$. This effort has provided insight into the fundamental growth mechanism and heterogeneous integration of TMD material with complementary-metal-oxide-semiconductor (CMOS) technology.



▲ Figure 1: Lateral growth rate change based on NaCl concentration in the saturated water:IPA solution.

Metal-Organic Chemical Vapor Deposition-based Synthesis of P-type Tungsten Diselenide

S. Chakravarthi, H. W. Lee, Y. Jiao, X. Zheng, J. Zhu, J. Kong, T. Palacios
Sponsorship: Intel

Two-dimensional(2D) materials with their high mobility, excellent gate electrostatic control, and atomically thin nature have emerged as promising channel materials for highly-scaled, energy-efficient electronics. However, most of the previous research focuses on n-type semiconductors, e.g., molybdenum disulfide (MoS_2). P-type 2D materials, e.g., tungsten diselenide (WSe_2), possess equal importance in constituting the complementary electronics circuits, but both the material synthesis and electrical characterizations are less developed.

In this work, using metal-organic chemical vapor deposition (MOCVD), the impact of precursor concentration, seeding promoter, carrier gas and forming gas (H_2) flow on WSe_2 synthesis are systematically discussed, providing insights for obtaining higher-quality materials. Comprehensive material and device characterizations are carried out to evaluate the quality of as-grown WSe_2. This paves the way for our future research on scaling up the material synthesis system to 8" wafer-scale and enabling direct integration of 2D WSe_2 on silicon complementary-metal-oxide-semiconductor (CMOS) circuits.

FURTHER READING

- J. Zhu, J. Park, S. A. Vitale, et al., "Low Thermal Budget Synthesis of Monolayer Molybdenum Disulfide for Silicon Back-end-of-line Integration on 200 mm Platform," *Nat. Nanotechnol.* 18, 456-463 (2023).

Fabrication of Å-Scale Graphene Pores for Efficient Isomer and Ionic Separation

D. R. Chen, T. Zhang, S. M. He, Z Wang, C. Cheng, J. Wang, J. Kong
Sponsorship: NSTC Graduate Students Study Abroad Program (113-2917-I-002 -050)

Å-scale pores in monolayer graphene provide spatial confinement for small particles of various sizes, showing great potential in separation science, such as isomer and ionic separation. Conventional top-down methods for fabricating Å-scale pores with high density and uniformity often lack atomistic precision, resulting in large, non-selective pores that cause leakage and reduced selectivity. We previously demonstrated a Cu-ion-sputtering cascade compression approach in Nature that addresses the problem that minimizes the tail-end of the pore size distribution while enhancing pore density. Despite this, it remains challenging to repeatably achieve precise, uniform nanopores for designed separation applications due to the uncontrollable number of Cu ions. Here, we utilize Ar, which delivery is better controlled through a mass flow controller, as an ion source to enhance the reliability of in-situ pore fabrication. By applying voltage within the CVD chamber, Ar gas is ionized into Ar ions through a gas-phase electrochemical reaction and sputters carbon atoms out to create Å-scale pores. As a result, a repeatable production of sub-nanometer pores is implemented by such Ar-cascaded compression, verified by Raman spectroscopy and Conducting-AFM. The resulting Å-scale porous graphene film exhibits remarkable ion-ion selectivity along with high permeance. We plan to collect more data to further verify the repeatability for as-synthesized samples as separation films.

We hope the findings pave the way for Å-scale porous graphene, leading to future sustainable separation technologies.

Mechanocaloric Polymer Discovery and Multi-Scale Engineering for Green HVAC Technologies

D. Xu, H. Gold, T. Yu, S. V. Boriskina
Sponsorship: SUSTech and ONR Global, MathWorks MechE Graduate Fellowship, NDSEG and PEM Graduate Fellowships

Residential thermal management (heating and refrigeration) is one of the most energy-intensive technology sectors, with space cooling alone accounting for nearly 16% of global electricity consumption in the buildings sector in 2021. The direct CO2 emissions from space and water heating reached a record high of 2.5 G tons in the same year. Solid-state cooling technologies offer a sustainable alternative to conventional HVAC systems, providing opportunities for enhanced energy efficiency and reduced environmental impact.

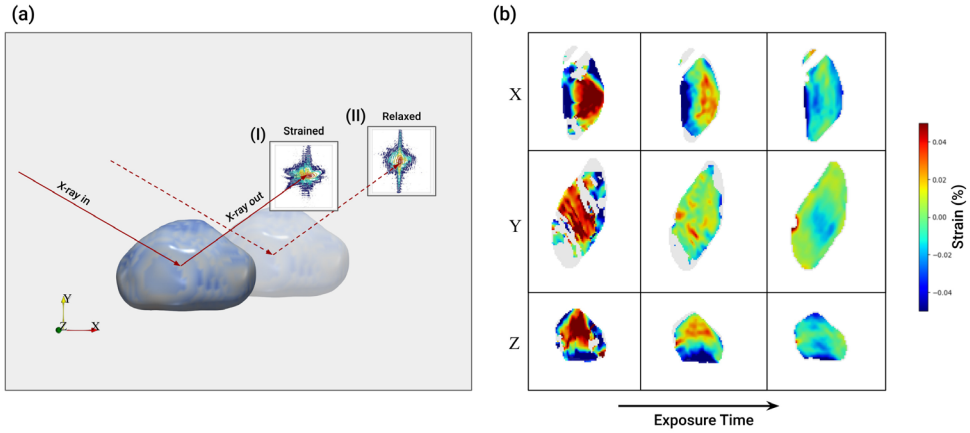
This work focuses on the AI-driven discovery, testing, and optimization of mechanocaloric (mC) polymers. We use a combination of ab-initio density functional theory and machine-learning surrogate model development to identify most promising polymers and co-polymers for mC applications, a molecular dynamics simulations to evaluate realistic material performance under mechanical deformations, followed by experimental verification and optimization of the most promising materials. Our experimental results show that some of the new materials identified through our discovery pipeline exhibit strong performance, achieving temperature changes exceeding 5 ˚C under uniaxial strain and up to 20 ˚C under twisting deformation. Additionally, they exhibit a high coefficient of performance, low actuation stress, and a competitive cost-to-performance ratio, showing promise for scalable applications. Finally, we use macroscopic finite-element mechanical modeling to study the impact of larger-scale structural features, such as fiber plying, braiding and knotting on improving the material performance and fatigue resistance.

Through this multi-scale optimization and testing pipeline, we aim to accelerate the development of durable, high-performance mC materials for scalable and sustainable solid-state cooling and heating technologies.

X-ray Induced Relaxation of Nickel and Nickel-based Nanoparticles on Silicon

R. Hultquist, D. Simonne, E. Jossou
Sponsorship: MathWorks Fellowship, Startup

Nickel and Nickel-Cobalt single crystals are model systems for studying the effects of radiation and corrosion in Light Water Reactor cooling components. While bulk alloys are used in full-scale systems, nanoparticle analogues can accelerate the research and development of bulk materials for use in extreme environments. Their morphologies, formed by dewetting, distinguish them from their bulk forms and influence local mechanical and chemical properties. Utilizing coherent X-rays to resolve strain and defects, Bragg Coherent Diffraction Imaging (BCDI) enables deep insight into single crystal models. In this work, we demonstrate that coherent X-rays can be used to relax highly strained Nickel alloy single crystals on Silicon. Furthermore, we use atomistic density functional theory calculations to show how photo-assisted modulation of charge density at the interface leads to the observed relaxation. Understanding and controlling heteroepitaxy with coherent X-rays will provide deeper insight into the physical chemistry of heterointerfaces while also promoting the use of BCDI to characterize complex models of materials in extreme environments.



▲ Figure 1: (a): Schematic of crystal in initial state (I) and final state (II) with detected diffraction pattern showing changes from highly strained to relaxed state. (b): Crystal cross sections along X, Y and Z directions showing time dependent strain field relaxation under the influence of X-rays.

Material Characterization of Niobium Nitride Sputtered Thin Films for SNSPDs

F. Incalza, D. J. Paul, M. Castellani, O. Medeiros, E. Batson, K. K. Berggren
Sponsorship: Darpa Program

Niobium Nitride (NbN) superconducting thin films have garnered significant interest for applications in superconducting electronics, quantum computation, and superconducting nanowire single-photon detectors (SNSPDs). SNSPD performance is highly dependent on the superconducting properties of NbN films, which are primarily determined by the film's microstructure and the substrate on which it is grown. A detailed understanding of the crystallographic structure and chemistry of NbN is essential for evaluating and optimizing device performance. Variations in the phononic spectrum, affected by whether the films are crystalline or polycrystalline, can lead to significant differences in SNSPD characteristics, including thermal conductance, detection efficiency, switching current, depairing current, critical temperature, and diffusion coefficients. This insight enables a direct relationship between crystal structure and device performance, guiding the optimization of NbN-based devices.

In this work, NbN thin films were deposited using sputtering techniques on sapphire, thermally oxidized silicon, and magnesium oxide substrates. The deposition process was analyzed and optimized based on the substrate type. Material and superconducting properties were examined using various techniques, including XRD, XPS, EBSD, ellipsometry, MPMS, and TEM. The critical temperature, crystal structure and orientation, grain dimensions, and optical properties were analyzed and compared across films, with each set of properties related to the achievable device characteristics. Using high-temperature (800 ˚C) sputtering, we achieved high-quality epitaxial NbN thin films on sapphire, with a critical temperature reaching the bulk value of 16 K, and compared their properties with those of films deposited on other substrates.

Uncovering Origins of Poor Photoluminescence Quantum Yield in 2D Hybrid Metal Organic Chalcogenolates (MOCs)

N. J. Samulewicz, W. S. Lee, W. A. Tisdale
Sponsorship: NSF Graduate Research Fellowship, U.S Army Research Office Award

Innovations in blue emitting semiconductors and widespread adoption of light emitting diodes (LEDs) in recent years have drastically reduced global energy consumption associated with lighting. While this reduction is monumental, production and other energy intensive processes associated with LED lighting still contribute significant yearly CO_2 emissions. To reach future sustainability targets, advancements in light emission are essential, driving research interest towards novel materials – particularly ones with more sustainable synthesis pathways.

Our lab investigates metal organic chalcogenolates (MOCs) – novel 2D van der Waals stacking hybrid semiconductors with strong exciton binding energies, in-plane anisotropy, and direct bandgap. Amongst others, these properties make MOCs suitable candidates for applications as LEDs, excitonic switches, and other optoelectronics. Importantly, high-quality millimeter scale single crystals are synthesizable through a simple, ambient single-phase reaction. Covalent intralayer bonding in MOCs bolsters strong environmentally stability and enables property tuning through various functionalizations. Among them, mithrene, $[AgSePh]_\infty$, is of particular interest due to its narrow blue luminescence. However, nonradiative recombination dominates excited state dynamics, limiting its potential to revolutionize future optoelectronics. This work details efforts to uncover the origins of the low photoluminescence quantum yield in mithrene through spectroscopic analysis of defects, physical structure, and brighter MOC variants. Emission is highly impacted by defects, yet impervious to simpler defect-mitigation strategies, forcing exploration of more novel passivation techniques. Despite interest in mithrene's sharp blue emission, future viability of this material class to promote sustainable energy consumption is most dependent on the efficiency of light emission, and is necessary to pursue improvements in both performance and understanding.

Group-IV-based Nanostructure Materials for Quantum Computing and Photonics

S. Ben-David, S. P. Ramanandan, N. Amador-Mendez, A. Rudra, A. Fontcuberta i Morral
Sponsorship: NCCR SPIN, SNSF SUGAR, Fulbright/Swiss Government Open Research Award

Group-IV semiconductors (Si, Ge, Sn) are key materials for nanoelectronics and show promise for quantum devices. While Si remains the most widely used semiconductor, Ge and GeSn draw interest due to their inherently higher electron and hole mobility. This work explores methods to realize Ge- and GeSn-based nanostructures.

Ge nanowires (NWs) offer a versatile platform for exploring quantum transport phenomena as the 1D charge confinement enables spin-based qubit logic. Despite these convenient properties, NW scalability remains a major challenge. Selective area epitaxy (SAE) offers a scalable solution for achieving horizontal NWs on device substrates. In this work, we advance the SAE of Ge NWs by growing the Ge in a Si V-groove nanomembrane (NM) instead of holes on a planar substrate. This approach confines the NWs fully within the Si V-groove, eliminating the direct interface between the Ge and SiO_2 dielectric mask that hinders NW spin-qubit functionality. We present the fabrication of the Si V-groove NM and the temporal evolution of the Ge NW growth using SEM, AFM, and cross-section TEM.

The incorporation of Si V-groove NMs enables precise confinement of Ge nanowires for scalable quantum devices while addressing limitations in spin-qubit functionality. Nonetheless, the indirect bandgap of Ge restricts use in optoelectronics, which can be overcome by leveraging GeSn alloys with direct bandgap properties. GeSn can be monolithically integrated into Si photonics to act as a NIR emitter. We present a novel approach to realize GeSn nanostructures with Sn contents beyond the solubility limit (1%at) via flash annealing. We show the top-down fabrication of GeSn nanostructures and the material properties evaluated using XRD, AFM, Raman spectroscopy, and TEM. Future work focuses on integrating Ge and GeSn nanostructures into optoelectronic platforms to enhance device functionality and scalability.

Electric Field Tunable Chern Insulators in Tetralayer Rhombohedral Graphene

S. Ye, T. Han, Z. Lu, Y. Yao, J. Yang, J. Seo, L. Ju
Sponsorship: NSF Grant

Rhombohedral graphene, characterized by its unique stacking order and flat electronic bands, has emerged as a versatile platform for exploring correlated and topological phenomena. The rhombohedral tetralayer graphene/hBN moiré system has been proposed to host novel superconductivity and topological phases, including the chiral superconductivity and quantum anomalous Hall effect. In this work, we present DC electrical transport measurements in a rhombohedral tetralayer graphene/hBN moiré superlattice. We observed electric-field-induced topological phase transitions of Chern insulators, with the Chern number tunable by displacement field from C=3 to C=4 at the first hole filling of the superlattice. Magnetic hysteresis measurements revealed that these states emerge at extremely low magnetic fields (B=0.1) and exhibit perfect quantization. While the C=4 state aligns with previous prediction and observation of a quantum anomalous Hall state, the C=3 state observed at lower displacement fields is unexpected. Additionally, we identified another C=3 state at filling nu = -2. These findings enrich the topological landscape of this intriguing system. Combined with its unique superconducting properties, rhombohedral multilayer graphene is a promising platform for studying and engineering novel quantum phases of matter.

Investigation of Novel Atomic Layer Deposited Dielectric Films for High Frequency, High Temperature Gallium Nitride Electronics

J. Kang, J. Niroula, T. Palacios
Sponsorship: AFOSR, SRC Jump 2.0 SUPREME Center

High temperature rated electronics are used in various ways such as hypersonic aircrafts, deep well oil drilling, and exploring Venus. Unfortunately, traditional silicon (Si) devices cannot operate above 250˚C due to fundamental limits on its intrinsic carrier concentration. Therefore, semiconductors like silicon carbide (SiC) and gallium nitride (GaN) are more suitable to use for high temperatures because of their wide-band gap and negligible carrier thermal generation at temperatures around 500˚C. Currently, plasma enhanced chemical vapor deposition (PECVD) silicon nitride (SiN) is the current state of the art in GaN passivation, but for high frequency devices, thin (<25 nm) passivation layers are needed to minimize parasitic capacitances. Thus, for next generation passivation layers, using Atomic Layer Deposition (ALD) is the preferred deposition method on these devices as ALD allows for atomically precise control of very thin, high quality dielectric layers.

In this project, we investigate the electrical properties of ALD deposited Ga_2O_3, AlN, and Al_2O_3 as future passivation layers for next generation GaN devices. This is done through high temperature (25-300˚C) electrical characterization of metal-insulator-metal (MIM) capacitors as well as through current collapse measurements of these films on GaN high electron mobility transistors (HEMTs). ALD grown films provide excellent passivation for silicon surfaces as they exhibit high breakdown voltage, low leakage current, and better thermal conductivity.

Hybrid Deposition Process of Perovskite Photovoltaics

T. Kadosh Zhitomirsky, K. Yang, S. Srinivasan, W.J. Hsu, E. Pettit, R. J. Holmes, H. L. Tuller, V. Bulović
Sponsorship: U.S. Department of Energy

Halide perovskites have demonstrated remarkably high solar to electrical energy conversion efficiencies and are therefore of great interest for rapid commercialization. Currently, popular fabrication processes involve the hazardous solvent DMF, which is required to dissolve the lead based precursors, such as PbI2. These fabrication routes cannot be readily up-scaled as required. Vapor Transport Deposition (VTD) is an alternative, low-cost manufacturing technique that has been proven before for other solar cell materials.

VTD base process bypasses some solvent related challenges, namely uniform coverage of large areas, chemical compatibility, and toxicity. As a low-cost alternative to thermal evaporation, VTD has the potential to deposit organic and inorganic perovskite precursor materials either sequentially or via co-deposition. Furthermore, VTD potentially offers higher tunability of deposition parameters, to enable film growth with improved composition and microstructure control. In this work, we demonstrate harnessing VTD to deposit the inorganic precursor lead iodide. By that we can eliminate the use of the toxic solvent DMF, and replace it by isopropanol which is considered 40 times less toxic.

We are currently working with a custom-made VTD system, with which we achieved champion FAPI and MAPI cells of 12% power conversion efficiency. We report our progress in investigating the influence of underlying layer, substrate and sublimation temperatures, chamber pressure and flow rate ratios on the morphology and stoichiometry of the forming perovskite film, and in turn, its photo-active and electronic properties.

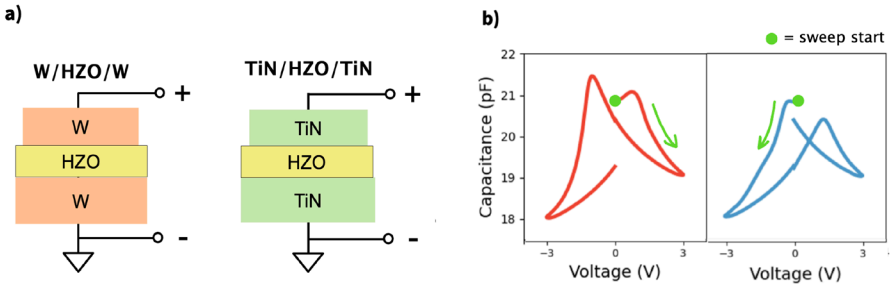
Characterization of Interface-Driven Wake-Up and Fatigue of Ferroelectric $Hf_{0.5}Zr_{0.5}O_2$ for Non-Volatile Memory Applications

T. E. Espedal, Y. Shao, E. R. Borujeny, J. A. del Alamo
Sponsorship: MIT UROP, Intel Corporation, Semiconductor Research Corporation

Ferroelectric (FE) memory based on CMOS-compatible $Hf_{0.5}Zr_{0.5}O_2$ (HZO) has emerged as an NVM technology may provide low-voltage operation, fast switching, long data retention, and high device endurance. Plasma-Enhanced ALD has shown enhanced HZO film quality, potentially improving memory. However, certain metal-FE-metal (MFM) capacitive structures show premature breakdown. Endurance characterization shows asymmetric wake-up, indicating unique physics at either metal-HZO interface.

We identify the asymmetric electrical characteristics of W/HZO and TiN/HZO devices that emerge with device cycling. Consistent symmetry patterns across pulsed I-V, DC I-V and C-V measurements indicate interfacial phenomena that eventually lead to device breakdown.

In this work, we show that interface physics are a critical aspect of MFM device endurance: specifically, process conditions and electrode metal are important in realizing high-endurance HZO films and thereby potential NVM applications.

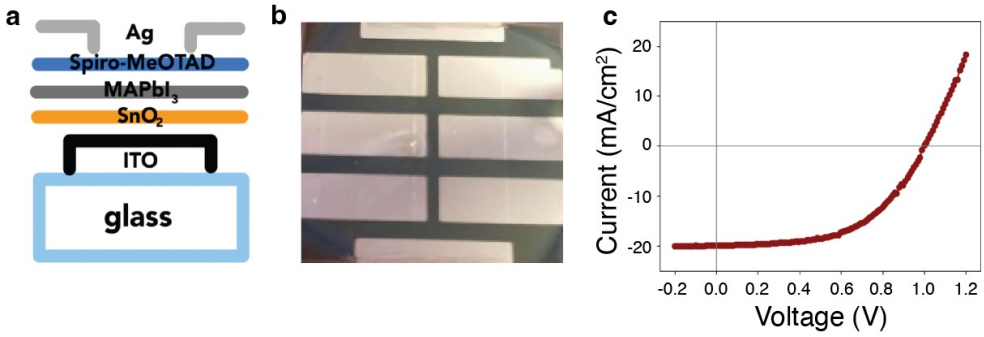


▲ Figure 1: a) Schematic of MFM structures. b) The observed W/HZO C-V characteristic is invariant to sweep direction reversal, indicating device asymmetry.

Semi-transparent Perovskite Solar Cells

E. Delarosa, K. Jander, V. Sandrapaty, F. Shangguan, R. Zhang, T. K. Zhitomirsky
Sponsorship: MIT EECS, 6.2540 class

With growing global energy demands, renewable energy sources like solar are more critical than ever. Designing solar cells with a formfactor that allows their ubiquitous integration into the buildings, including windows as energy generating surfaces, can expand access to solar energy. Perovskite solar cells are especially promising due to their high efficiency and thin-film design which can facilitate their manufacturing, enable semi-transparency, and light weight. In this work, we designed and fabricated perovskite solar cells with efficiency as high as 10.7%, and investigated the transparency-efficiency tradeoff in these devices.



▲ Figure 1: a) Schematic illustration of the designed perovskite solar cell. b) Top-view photo of an example fabricated solar cell. c) Example I-V characteristic of a fabricated solar cell with 10.7% efficiency.

Synthesis of Mixed Dimensional Perovskite Heterostructures

M. Chattoraj, S. Saris, E. K. Price, W. A. Tisdale

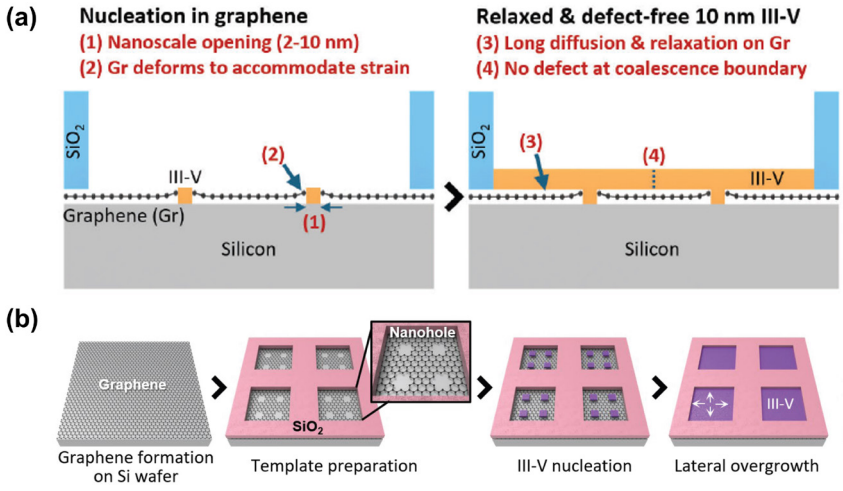
Continued advancements in the synthesis and application of nanomaterials dictate the need for well-developed analysis frameworks that fully capture the fundamentals behind their interactions. Förster theory accurately describes the transfer of excitons between molecules. However, this framework has repeatedly failed to quantitatively predict energy transfer rates between semiconducting nanomaterials, particularly in heterostructure systems that combine nanomaterials of different composition and dimensionality. The shortcomings of Förster theory are exacerbated in the case of lead halide perovskites, offering an optimal platform for pushing past the limits of current energy transfer models. Here, we synthesize mixed dimensional perovskite heterostructures and set the stage for studying energy transfer in these systems using time resolved photoluminescence spectroscopy. We focus on heterostructures combining spheroidal $CsPbBr_3$ quantum dots and two dimensional hybrid organic inorganic perovskite crystals, and aim to contribute to a deeper, quantitative understanding of energy transfer mechanisms in novel nanomaterials, which will inform the design of optoelectronic devices with nanomaterial interfaces.

Selective Area Epitaxy of Defect-free III-V Layers on Silicon via Graphene Nanoholes

N. M. Han, K. Lu, D. A. Kwon, J. Kim
Sponsorship: DARPA M-STUDIO (HR001124S0019--FP-006)

The integration of III-V semiconductors on silicon is a boon for electronic and optoelectronic devices due to their high electron mobility and direct bandgaps. However, their large lattice mismatch often leads to high dislocation densities, severely impacting performance. Standard techniques like graded-buffer layers, epitaxial lateral overgrowth, and aspect ratio trapping have shown limited success, and pseudomorphic growth, while dislocation-free, can result in interface roughening and mobility loss in thin channels.

To address this, I developed a graphene-assisted epitaxy method to achieve dislocation-free III-V growth on silicon. Here, graphene is grown on the silicon surface with nanoholes that expose nucleation sites for III-V growth, while lateral overgrowth over the graphene-covered surface facilitates strain relaxation. This scalable, CMOS-compatible method enables high-quality III-V material integration on silicon for next-generation electronic and optoelectronic devices.



▲Figure 1: Schematic of the (a) mechanisms and (b) process of graphene-assisted epitaxy to achieve defect-free III-V films on Silicon.

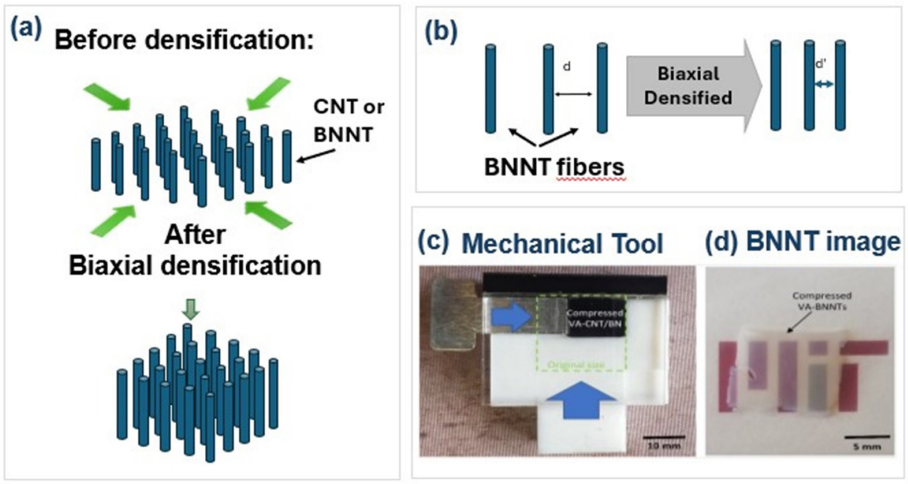
Densification of Vertically Aligned Boron Nitride Nanotubes via Biaxial Mechanical Compression

S. Sharma, A. R. C. Neto, M. Rogers, L. Acauan, B. L. Wardle
Sponsorship: NECST consortium

Boron nitride nanotubes (BNNTs) have garnered significant interest due to their one-dimensional structure, piezoelectric properties, superior chemical, and thermal stabilities. All these attributes make BNNTs highly versatile, supporting diverse applications ranging from structural reinforcement to energy storage [1]. However, the synthesized vertically aligned BNNTs have a low volume fraction of around ~1% [2], which can limit their use in applications like piezoelectric sensor/actuators [3]. In this work, we demonstrate a biaxial densification technique, previously applied to VA-CNTs [4], that increases the volume fraction of VA-BNNTs by a factor of four.

In this study, the BNNTs were synthesized by coating VA-CNTs with hexagonal boron nitride (VA-CNT/BN), followed by thermal oxidation to remove the CNT scaffold [2]. The work compares three different routes for densifying VA-BNNTs: (1) biaxially densifying the VA-BNNTs directly; (2) biaxially densifying the VA-CNT/BN and posteriorly removing the VA-CNT scaffold; and (3) coating the pre-densified VA-CNTs with BN and subsequently removing the VA-CNT scaffold.

Our results show that method (3) produces a poor BN coating on the CNTs, while method (1) leads to a nonuniformly densified VA-BNNT structure that breaks easily when the densifying forces are released. In contrast, method (2) achieves a uniformly densified VA-BNNT structure after removal of the CNT scaffold. These findings suggest that further densification is feasible with this approach, enabling applications that require a high-volume fraction of BNNTs.

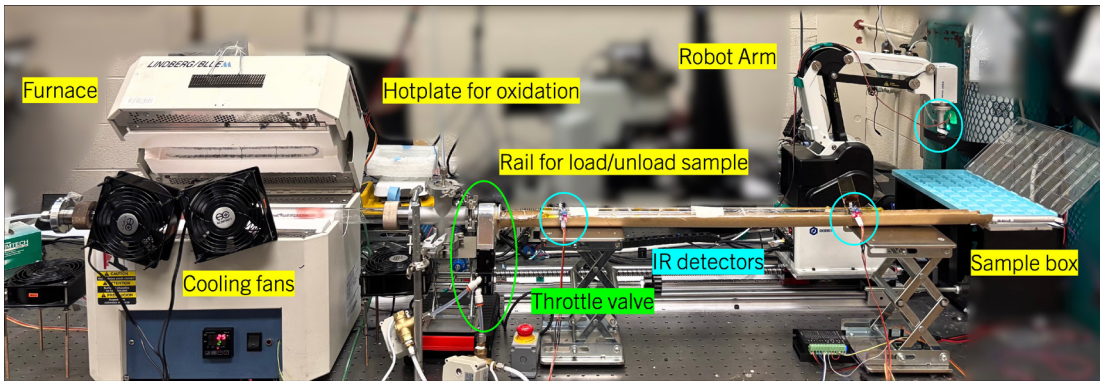


▲ Figure 1: (a,b) Schematic of the biaxial mechanical densification process, (c) Mechanical tool used for compressing VA-CNT compressed, and (d) image of bi-axially compressed VA-BNNT after CNT removal.

Artificial Intelligence-Assisted Synthesis and Integration Optimization of 2D Materials

Z. Wang, S. He, A. Lu, J. Wang, J. Kong
Sponsorship: The Semiconductor Research Corporation Center 7 in JUMP 2.0

Chemical vapor deposition (CVD) has developed to be the most efficient and scalable synthesis method for various two-dimensional materials, which hold great promise for advancing the semiconductor technology. However, manual process execution in the CVD synthesis is both time-consuming and labor-intensive. Here, we propose an autonomous CVD system that integrates AI and automation for increased efficiency and high-throughput. To demonstrate this idea, we have built an autonomous graphene CVD system, incorporating stationary robots and automated synthesis. This system can control all the growth parameters including temperature, gas flow, pressure, through input parameters, and handle samples with a robot arm. This autonomous CVD system has the capacity to process up to 50 sets of different growth parameters and synthesize continuously without any manual operation. We have tested the stability of this system with 50 graphene growths within a time period of two weeks, which shows high reproducibility and stability. Enabled by this fully automated system, we plan to incorporate AI into recipe optimization and explore different parameters space of graphene synthesis.



▲ Figure 1: Photograph of the autonomous CVD system for graphene, consists of automated furnace (on the left) and robotic arm for sample handling (on the right).