NAMBE
Room Cummings Ballroom - Session NAMBE2-TuM

Chalcogenides and Topological Materials
Moderator: Stephanie Law, Penn State University

10:45am NAMBE2-TuM-11 Rhombohedral-to-Cubic Phase Transition in $Ge_{1-x}In_xTe$ Thin Films Grown by MBE, *Xinyu Liu*, K. Yoshimura, S. Bey, M. Abdu Karim, J. Wang, L. Riney, M. Zhukovskyi, T. Orlova, B. Assaf, University of Notre Dame

Topological superconductors are the subject of intense research due to their potential to harbor Majorana boundary modes, which hold promising applications in quantum computing. Superconductors that lack inversion symmetry host singlet-triplet mixing and are a precursor of topological superconductivity. GeTe, a non-centrosymmetric semiconductor, has been observed to undergo a semiconductor-superconductor transition above 1K when doped with In at a critical level ($x \approx 0.12$).[1] The transition to superconductivity is accompanied by a structural phase transition to a centrosymmetric cubic phase. This study introduces the first synthesis of $Ge_{1-x}In_xTe$ films via MBE, motivated by recent findings that quantum confinement promotes the stabilization of the non-centrosymmetric structure of IV-VI materials.

Structural analyses of 120nm $Ge_{1-x}In_xTe$ thin films ($0 \leq x \leq 0.4$) on BaF_2 (111) substrates show distinct characteristics not observed in single crystals. The process begins with the deposition of a 40nm GeTe buffer layer to facilitate initial layer nucleation. We then co-deposit Ge, In, and Te from separate elemental sources, maintaining a Ge to Te flux ratio of 0.1. The growth is monitored via in situ RHEED, revealing that only a specific growth temperature range (220–270°C) is suitable for high-quality epitaxial growth.[2] We observe that In atoms tend to migrate into the GeTe buffer layer, confirmed by TEM and EDX mapping.

The samples undergo comprehensive analysis using both SEM and XRD techniques. To evaluate the crystal orientation along the growth axis, specular ω–2θ scans were conducted. To investigate the films' strain relaxation, reciprocal space maps were acquired for both symmetric rhombohedral-GeInTe (0006) and asymmetric rhombohedral-GeInTe (01-18) Bragg peaks. Both SEM and XRD show a rhombohedral-to-cubic phase transition at In level of x=0.19 in MBE grown $Ge_{1-x}In_xTe$ films, higher than what is found in single crystals. We also observe the coexistence of strained and unstrained cubic $Ge_{1-x}In_xTe$ phases for high In content films.

Growing non-centrosymmetric $Ge_{1-x}In_xTe$ films with high In content enables deeper exploration of its unconventional superconductivity through thin-film-specific devices. Upcoming magnetotransport studies will examine the connection between its phase transitions and superconductivity. Additionally, cubic $Ge_{1-x}In_xTe$'s compatibility with topological crystalline insulators positions it as a key material for creating heterostructures that could support topological superconductivity.

11:00am NAMBE2-TuM-12 Coherent strain through quasi van der Waals Epitaxy of magnetic topological insulators Cr: $(Bi_xSb_{1-x})_2Te_3$ on a GaAs (111) substrate and the influence from growth windows, *Yuxing Ren*, K. Pan, Y. Chen, J. Kang, B. Regan, C. Wong, M. Goorsky, K. Wang, University of California at Los Angeles

Quasi van der Waals Epitaxy (qvdWE) has been realized for decades at the interfaces between 3D and 2D materials or van der Waals materials. The growth of magnetic topological insulators (MTI) Cr: (BixSb1-x)2Te3 (CBST) on GaAs (111) substrates for Quantum Anomalous Hall Effect (QAHE) is actually one of the examples of qvdWE, which is not well noticed despite the fact that its advantages have been used in growth of various MTI materials. This is distinguished from the growth of MTIs on other substrates. The specific surface potential and ionicity determines the nature of a quasi van der Waals growth, which has the same advantage of van der Waals epitaxy of getting rid of surface roughness while at the same time maintaining a uniform grain orientation as traditional epitaxy. Despite many reports, there are still no clear conclusions on the root cause of qvdWE and the features of qvdWE have not been well recognized. The typical features are coherent interface and the existence of a strain within a short period of distance. While coherency is much more widely observed for qvdWE, the existence of strain is generally hard to be demonstrated due to the small thickness of strained layers in the van der Waals materials. Our goal in this work is to demonstrate both coherency and the existence of strain at the interface of epitaxial CBST on GaAs (111) substrate.

Here in this work, we have for the first time shown the features of both coherent interfaces and the existence of strain originating from qvdWE at the same time. To show the coherently strained interface, we have grown samples with thickness of 1 QL (quintuple layer), 2QL and 6 QL under the same conditions respectively to compare the relaxed layers and strained layers. X-ray pole figures and Reciprocal Space Mapping (RSM), in-situ Reflective High Energy Electron Diffraction (RHEED), STEM (Scanning Transmission Electron Microscope) and EDS (Energy Dispersive X-ray Spectroscopy), and a Raman measurement are used to demonstrate the coherently strained interface.

Growth window of quantization regime is also studied in this work. By controlling source flux and substrate temperatures, we have identified the growth of samples with quantum anomalous Hall effect at the boundary of mass-transport flow and adsorption-control mode on GaAs (111) substrates.

11:15am NAMBE2-TuM-13 Epitaxial Hexagonal $BaZrSe_3$ Thin Films with Strong Birefringence in-Plane, *Ida Sadeghi*, V. Kamboj, MIT; *T. Simonian*, College Green, Ireland; *J. Van Sambeek, M. Xu*, MIT; *V. Nicolosi*, College Green, Ireland; *J. LeBeau, R. Jaramillo*, MIT

Systems of ternary chalcogenides – such as the Ba-Zr-Se system - have a rich diagram of stable and metastable phases, including perovskite, needle-like, and hexagonal structures that vary in the connectivity of octahedra (*e.g.* $ZrSe_6$), and are predicted to vary widely in their opto-electronic properties.[1,2] We previously demonstrated synthesis of $BaZrSe_3$ thin films in the corner-sharing perovskite phase,[3,4] which has a band gap of 1.5 eV and is theoretically predicted to be thermodynamically unstable.[5,1] Others have reported that powder synthesis results in a hexagonal phase with very small band gap and possible defect ordering.[6,7] These results highlight the strong dependence of phase and properties on synthesis conditions for ternary chalcogenides.

Here we demonstrate synthesis of face-sharing hexagonal $BaZrSe_3$ (h-$BaZrSe_3$) thin films by MBE. We can make the hexagonal phase via two routes: (1) by aggressive selenization of a thin film of $BaZrS_3$ in the corner-sharing perovskite phase on $LaAlO_3$ (LAO), or (2) by direct deposition on $YAlO_3$ (YAO) (001) substrates. The kinetics of transformation from perovskite to hexagonal phase depend on the processing temperature and H_2Se gas flux.

The hexagonal structure is highly anisotropic and is expected to feature giant optical birefringence, similar to the iso-structural compound $BaTiS_3$.[8] LAO is pseudo-cubic, and h-$BaZrSe_3$ films form on LAO in two domain types, with the optic axis rotated by 90° in-plane; this renders the birefringence difficult to measure and use. In contrast, YAO is orthorhombic and (001) substrates feature rectangular symmetry. As a result, h-$BaZrSe_3$ films on YAO (001) have a dominant domain orientation with the optic axis in-plane; this enables more direct measurement and use of the optical anisotropy.

To demonstrate the effect of epitaxial control over film phase and orientation, we report measurements of optical transmission versus incident polarization angle, demonstrating a large modulation with a period of ~ 180° at near-infrared wavelengths. We also find using measurements of photoconductivity spectroscopy that the band gap is 1.79 ± 0.1 eV, which is many times larger than theoretically predicted. Our results indicate that epitaxial h-$BaZrSe_3$ may be uniquely useful for free-space modulation of mid-wave and near infrared light.

1. *Nano Lett.* **15**, 581, 2015.

2. *Chem. Mater.* **34**, 6894, 2022.

3. *Adv. Funct. Mater.* **33**, 2304575, 2023.

4. *arXiv:2403.09016*, 2024.

5. *J. Appl. Phys.* **125**, 235702, 2019.

6. *Russ. J. Inorg. Chem.* **9**, 1090, 1964.

7. *J. Am. Chem. Soc.* **120**, 7639, 1998.

8. *Photonics* **12**, 392, 2018.

11:30am NAMBE2-TuM-14 Quasi-Van Der Waals Epitaxial Growth of Thin γ'-Gase Films, *Mingyu Yu,* University of Delaware*; S. Law,* Pennsylvania State University

GaSe is an advanced 2D layered semiconductor, possessing various appealing properties, such as rare p-type conductivity, nonlinear optical behavior, and high transparency in 650-180000 nm. GaSe also exhibits bandgap behavior opposite to transition metal chalcogenides, as it transitions from an indirect bandgap monolayer to a direct bandgap bulk material, with a reduction in bandgap energy. These features make GaSe rich in potential in many applications, such as quantum photonic devices, field-effect transistors, etc. GaSe has a hexagonal crystal structure composed of Se-Ga-Ga-Se quadruple layers (QLs). Each QL is bonded by weak van der Waals (vdW) interactions, enabling flexible stacking configurations. Therefore, there are multiple polymorphs for GaSe, namely ε-, β-, δ-, and γ-, all having identical non-centrosymmetric QL with a D_{3h} space group. In addition to the 4 extensively explored polymorphs, a new polymorph, γ'-GaSe, was proposed for the first time in 2018. γ'-GaSe is unique for its centrosymmetric D_{3d} QL and the QLs are stacked like γ-GaSe (Fig. S1). On this basis, γ'-GaSe has been predicted to show intriguing performance compared to other polymorphs. However, to date, there are only a few reports showing the observation of γ'-GaSe, and it usually coexists with other polymorphs since the formation energy of γ'-GaSe is less favorable. Moreover, the commercial applications of GaSe are limited by synthesis technology, as common methods such as exfoliation are difficult to achieve wafer-scale, high-quality production of GaSe. Obtaining a single-phase, single-polymorph GaSe is also challenging due to the coexistence of multiple stable phases and polymorphs.

We developed a quasi-vdW epitaxial growth method to obtain high-quality pure γ'-GaSe thin films on GaAs(111)B substrates at a wafer scale. The resulting films exhibit a smooth surface with a root-mean-square roughness as low as 7.2Å (Fig. S2a) and a strong epitaxial relationship with the substrate (Fig. S2b). More interestingly, we observed a pure γ'-type configuration using scanning transmission electron microscopy (Fig. S2c,d) and analyzed its formation mechanism through density-functional theory (Fig. S3). These findings contribute to the exploration of GaSe. We also investigated the unconventional 2D/3D epitaxial growth mechanism and have overcome technical challenges, benefiting the advancement of heterogeneous integration. In the future, we will focus on developing the properties and applications of γ'-GaSe, and delving into the understanding of the epitaxial growth mechanism from a theoretical perspective.

11:45am NAMBE2-TuM-15 Response of Topologically Protected Helical Modes in Monolayer WTe2 to Band-gap Tuning, *Yulia Maximenko,* Colorado State University*; Y. Chang,* Rutgers University*; M. Hirsbrunner, L. Wagner, V. Madhavan, T. Hughes,* University of Illinois at Urbana Champaign

Two-dimensional transition metal dichalcogenide (TMD) materials exhibit a variety of novel quantum phenomena, including topological, superconducting, magnetic, and strongly correlated phases as monolayers and homo-, hetero-, and twisted bilayers. Unlike the exfoliation method, molecular beam epitaxy (MBE) of TMDs produces organics-free atomically pristine layers allowing surface-sensitive measurements. Local atomically resolved studies of such TMD platforms are crucial for unraveling their spatially dependent electronic properties. Here, we report a study of monolayer 1T' WTe$_2$, a quantum-spin-Hall (QSH) insulator and a superconductor, which has been studied extensively using magnetotransport and local-probe techniques previously. We performed an STM study of epitaxially grown monolayer WTe$_2$ imbedded in a device with back-gating capabilities. Using local density of states (LDOS) STM measurements and density functional theory (DFT) simulations, we gained new insights into 1D helical edge modes of WTe$_2$ and report their asymmetric spatial response to out-of-plane displacement fields. Such tunable spatial dependence gives new insights about harvesting spin-momentum-locked 1D modes of WTe$_2$ for energy-efficient devices, spintronics, and topological quantum computing applications. This talk will also address technical challenges of fabricating a device suitable for MBE and gate-tuned STM and epitaxial growth of high-quality monolayer TMDs on commercial CVD graphene.

12:00pm NAMBE2-TuM-16 Phase-selective Growth of the Topological Insulators Bi$_2$Te$_3$ and Bi$_4$Te$_3$ for Integration with the Superconductor Fe(Te,Se), *Matthew Brahlek, J. Chen, J. Lu,* Oak Ridge National Laboratory*; R. Moore,* Oak Ridge Natinal Laboratory

Realizing exotic forms of superconductivity by epitaxially integrating high-transition-temperature (T_C) superconductors with topological insulators can open new paths for quantum-based applications. In this talk, we will discuss how molecular beam epitaxy can be used to controllably synthesize the topological insulators Bi$_2$Te$_3$ and Bi$_4$Te$_3$ by carefully controlling the flux ratio during growth. Alternatively, high-quality Bi$_4$Te$_3$ can readily be achieved by reducing Bi$_2$Te$_3$ post-growth. It is found that both phases can be integrated them with high T_C superconductor Fe(Te,Se) with sharp interfaces. In the low Se-doping regime, measurements of the electronic and crystalline structure reveal that a large electron transfer, epitaxial strain, and novel chemical reduction processes likely affect superconductivity at the interface of Fe(Te,Se) with Bi$_4$Te$_3$ compared to Bi$_2$Te$_3$. This novel route for epitaxial phase control at topological/superconducting interfaces provides new insight into the nature of unconventional superconductivity while being a new platform for identifying and utilizing new electronic phases.

12:15pm NAMBE2-TuM-17 Origin of the high Curie Temperature in (Sb$_2$Te$_3$)$_{1-x}$(MnSb$_2$Te$_4$)$_x$ structures grown by molecular beam epitaxy, *Candice Forrester,* The Graduate Center (CUNY)*; C. Testelin,* CNRS, France*; K. Wickramasinghe,* City College of New York, City University of New York*; S. Mohammadi,* The Graduate Center (CUNY)*; M. Tamargo,* City College of New York, City University of New York

Previously we reported the growth of (Sb$_2$Te$_3$)$_{1-x}$(MnSb$_2$Te$_4$)$_x$ magnetic topological materials with Curie temperatures (T_C) as high as 100K, much higher than reported values for these materials. They are formed by combining Mn, Sb and Te in molecular beam epitaxy (MBE) via self-assembly, where MnSb$_2$Te$_4$ septuple layers (SLs) and Sb$_2$Te$_3$ quintuple layers (QLs) form compositions (x) that depend on the growth parameters used. By reducing the growth rate to ~ 0.5 nm/min, from the previously used 0.9 nm/min, the highest T_C values were obtained for structures with $0.7 \leq x \leq 0.85$.[1] Derivative curves of temperature dependent Hall resistance revealed the presence of two T_C components, T_{C1} about 20-25K and T_{C2} as high as 100K. Energy dispersive X-ray spectroscopy (EDS) data as a function of x suggests that excess Mn incorporation in our samples is responsible for the high T_C values. We have proposed that the lower T_{C1} originates from the MnSb$_2$Te$_4$ SLs of the structure, while the higher T_{C2} is associated with the formation of (Mn$_y$Sb$_{1-y}$)$_2$Te$_3$ QLs alloys.[2] However, direct evidence for this proposal is difficult to obtain due to the complex nature of the materials.

Although transition metal (TM) ternary alloys like (TM$_y$Sb$_{1-y}$)$_2$Te$_3$, where TM = Cr or V, have been demonstrated and shown to exhibit high T_C values, Mn alloys of Sb$_2$Te$_3$, with y > 0.03, have not been experimentally realized. However, DFT simulations have predicted (Mn$_y$Sb$_{1-y}$)$_2$Te$_3$ to have very high T_C as the Mn content increases.[3] An alloy formation equation was derived y = 1/2 [(5+2x) χ_{Mn}-x], where χ_{Mn} is the Mn fraction in the samples obtained from the EDS data. This equation assumes that excess Mn incorporates into QLs and SLs equally and all the excess Mn is substituting for Sb. The equation was employed to fit our experimental data to the predicted data for samples with $0.7 \leq x \leq 0.85$. Although a good fit was observed for samples with low Mn content, samples with high Mn content deviated significantly from the simulated data. Here we explore modifications of the alloy formation equation to better describe the observed results. Specifically, a better fit to all the data is obtained when Mn substitution is not limited to only Sb sites, as seen when χ_{Sb} is used instead of χ_{Mn}. Other scenarios, such as Mn/Te substitution or Mn incorporation in the vdW gaps are also considered. Our results are consistent with the formation of (Mn$_y$Sb$_{1-y}$)$_2$Te$_3$ with y > 0.35, much higher than ever obtained before, and bring us closer to achieving magnetic topological materials with T_C values for practical applications.

1. arXiv:2311.10891 (2023)

2. Sci Rep 13, 7381 (2023)

3. Phys. Rev. B 89, 165202 (2014)

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