NAMBE

Room Tamaya ABC - Session NAMBE1-WeM

Heteroepitaxy

Moderator: Seth Bank, University of Texas at Austin

8:00am **NAMBE1-WeM-1 Heteroepitaxial Growth of Highly Anisotropic Sb_2Se_3 Films on GaAs**, ***Kelly Xiao,*** Stanford University*; Virat Tara,* University of Washington*; Pooja Reddy, Jarod Meyer,* Stanford University*; Alec Skipper,* University of California Santa Barbara*; Rui Chen,* University of Washington*; Leland Nordin,* University of Central Florida*; Arka Majumdar,* University of Washington*; Kunal Mukherjee,* Stanford University

Antimony selenide (Sb_2Se_3) is a versatile semiconductor with applications in photovoltaics, optoelectronics, and most recently as a phase change material (PCM) for reconfigurable photonic integrated circuits. Sb_2Se_3 is one of few binary PCMs that simultaneously exhibits both large amorphous-crystalline refractive index contrast and low loss in the near-infrared.[1] Notably, crystalline Sb_2Se_3 has a highly anisotropic "quasi-1D" orthorhombic structure due to covalent bonding along one axis and van der Waals bonding along the other two axes. Harnessing this unique anisotropy has the potential to mitigate inconsistent performance in current polycrystalline PCM films associated with individual randomly oriented grains exhibiting different optical responses,[2] as well as to unlock light polarization control or detection functionalities in birefringent and dichroic single crystalline films. Heteroepitaxial integration and solid-phase epitaxy of Sb_2Se_3 on single crystal substrates are therefore important avenues to exploit anisotropic waveguide-integrated Sb_2Se_3 for photonics.

The quasi-1D structure has prominent consequences on growth. In this work, we demonstrate a synthesis route towards textured-epitaxial Sb_2Se_3 films directly on arsenic-capped GaAs(001) substrates via molecular beam epitaxy (MBE). We use a large relative Se/Sb flux ratio of 20 to achieve coalesced films, in contrast with previous MBE work showing sparse Sb_2Se_3 nanostripe formation on GaAs.[3] Not only do we find a ribbon-like surface morphology, the 1D axis unexpectedly remains aligned to the (2x1) reconstructed GaAs template to largely suppress 90° rotational domains, which otherwise commonly form on cubic substrates. X-ray diffraction indicates that for growth temperatures of 230–265°C, neighboring in-plane aligned domains deviate slightly in their out-of-plane orientations to constitute an in-plane "rotated" fiber texture. More importantly, we identify a narrow epitaxial growth window at a lower range of 180–200 °C. We show remarkable optical anisotropy along all three primary directions in epitaxial films, with giant out-of-plane birefringence ($\Delta n > 1$) at telecom bands.

Furthermore, we find that below 150 °C, MBE conditions can produce amorphous Sb_2Se_3 films, opening opportunities for heteroepitaxy. We will present initial results on laser-crystallized Sb_2Se_3, using the as-grown crystalline Sb_2Se_3 model system to inform our understanding of activating anisotropic crystallization in initially disordered phases.

[1]M. Delaney et al., *Adv. Func. Mat.* **30**(36), 2002447 (2020).

[2]C. Laprais et al., *Adv. Opt. Mat.* **12**(28), 2401214 (2024).

[3]P. Wojnar et al., *Nanoscale* **16**(41), 19477 (2024).

8:15am **NAMBE1-WeM-2 Evaluating Dopant Candidates for N-Type SnTe Films Grown by Molecular Beam Epitaxy**, ***Qihua Zhang,*** *Mary Kathleen Caucci, Maria Hilse, Susan Sinnot, Stephanie Law,* The Pennsylvania State University

SnTe is a desirable narrow bandgap semiconductor owing to its mid-infrared plasmonic capabilities, in-plane ferroelectricity, and topologically non-trivial band structure. However, due to the negative formation energy of Sn vacancies, the synthesized SnTe thin films are predominantly *p*-type even without extrinsic dopants, which limits their potential in thermoelectric and spintronic applications. While attempts have been made to develop *n*-type SnTe bulk crystals, these efforts have not extended to thin films.

In this work, we investigate on the effect of incorporating electron dopants in SnTe thin films by molecular beam epitaxy. We first demonstrate the molecular beam epitaxy (MBE) growths of SnTe layers on InP substrate, which has a 7.8% lattice mismatch to SnTe. Using interfacial misfit array, high quality SnTe layers with a full-width-at-half-maximum (FWHM) of 0.09° in XRD rocking curves and root-mean-square (RMS) roughness of 0.2 nm has been obtained. We next study the effects of group V elements, Sb and Bi, as dopants in SnTe films. We found that Sb is an unsuitable electron dopant and has a detrimental effect on the SnTe surface morphology. However, by doping Bi into SnTe films, a 2.5 times reduction in free hole concentrations is observed while the smooth surface can be retained. However, the addition of Bi dopants also induces twin domains in the films. Using first-principles calculations with density functional theory, we show that the preferred substitutional site of the Sb and Bi dopants depends on growth conditions: positively charged Sb_{Sn}^{+1} impurity is only found in heavily Sn-poor conditions while the Bi_{Sn}^{+1} is favored in all but the Sn-rich growth conditions. Moreover, for both Sb and Bi dopants, the substitutional sites form complexes with Sn vacancies which act as shallow acceptors and limit the reduction of free hole concentrations. We further evaluate alloying SnTe with In and Pb. Surprisingly, by alloying SnTe with up to 30% In content, up to 1.7×10^{16} cm^{-2} in electron sheet concentrations is obtained, while alloyed InSnTe film retained the rock-salt cubic structure and excellent crystal quality with a FWHM of 0.11° in the InSnTe (222) diffraction peak. The surface band structure of both SnTe and In-doped SnTe samples are studied using angle-resolved photoemission spectroscopy. DFT results regarding to the In substitutional sites in SnTe will also be discussed. This study presents a rare demonstration of n-type SnTe films grown by MBE, and serves as a critical milestone towards realizing n-type SnTe epitaxial layers.

8:30am **NAMBE1-WeM-3 Tunable Electrical Conductivity in Ferromagnetic Semiconductor Samarium Nitride**, ***Kevin Vallejo,*** Idaho National Laboratory

Rare-earth nitrides (RENs) are an exciting family of materials with a wide variety of properties desirable in the field of spintronics, infrared detectors, intrinsically ferromagnetic-based tunnel junctions, and as strongly correlated electron materials. The electronic configuration of elements containing 4*f* orbitals is a source of interesting new physics: as an example, samarium nitride (SmN) has been reported to support the coexistence of semiconductor behavior, ferromagnetic states, and superconductivity. Motivated by these properties and exciting opportunities, there has been an increased interest in the synthesis and study of high-quality rare-earth nitride materials. In this study we present an analysis of the synthesis of SmN thin films on MgO(001) using molecular beam epitaxy with varying growth conditions to create different levels of N vacancies. We report on the structure of different samples grown under different regimes of N availability and substrate temperature (T_{sub}), and measure their transport properties as a function of carrier concentration. We find that T_{sub} impacts the availability of carriers by a factor of 13x, compared to N availability that only increases carriers by a factor of 2x.

Structural characterization of these films indicate a uniform rocksalt crystal structure, with no appreciable difference in lattice constant or crystal quality beyond the difference in full width half-maximum of the SmN(200) peak of >0.14º. These promising results indicate a path forward in the epitaxy of versatile materials able to provide monolithic integration of different electronic behaviors without the associated strain brought about by heteroepitaxial integration of dissimilar materials.

8:45am **NAMBE1-WeM-4 Formation of [111]-Ge Domains in Layered α-FeGe2 by MBE and Solid Phase Epitaxy**, ***Moritz Hansemann,*** *Michael Hanke, Achim Trampert, Jens Herfort,* Paul-Drude-Institut für Festkörperelektronik Leibniz-Institut im Forschungsverbund Berlin, Germany

α-$FeGe_2$ is a promising new candidate in the emerging field of magnetic 2D-materials [1]. The α-phase of $FeGe_2$ fits neatly into this category with a predicted ferromagnetic to antiferromagnetic phase transition [2][3] and its self-organized layered structure [4]. This makes it also an interesting candidate for spintronics applications and the study of 2D-magnetism. We grow α-$FeGe_2$ on GaAs (001) by MBE utilizing solid phase epitaxy. In a two step process we first grow a layer of Fe_3Si that is subsequently overgrown by amorphous Germanium. This approach limits interdiffusion of Fe into the GaAs and allows for a sharp interface. The solid phase epitaxy is subsequently fulfilled by in situ annealing and forming the α-$FeGe_2$. This method allows us to grow α-$FeGe_2$ with high crystalline quality and long range order with thicknesses ranging between 4 nm and 25 nm. We present comprehensive analysis by XRR, XRD, AFM and TEM. From AFM we report very smooth surfaces with RMS= 0.5 nm, reproducing the GaAs surface. Additionally we find agglomerates on the surface, that we can link to the Germanium content in the layer. In the TEM micrographs we see layers with very few crystalline defects and homogeneity over long distances. The layers are occasionally interrupted by small Germanium domains, in which the [111] crystal direction is aligned with the [001] growth direction. These grow in a competing manor to the α-$FeGe_2$ in the solid phase epitaxy and are a known problem of the solid phase epitaxy process [5]. Further we

present GID measurements performed at the synchrotron PETRA III at DESY, that confirm the presence of the [111]-domains. These domains are also likely nucleation sites for the previously mentioned Ge-agglomerates visible in AFM.

References

[1] S. O. Valenzuela et al. “The phase diagram of 2D antiferromagnets”. In: Nature Nanotechnology 14.12 (Dec. 2019), pp. 1088–1089. doi: 10.1038/s41565-019-0592-x.

[2] D. Czubak et al. “Electronic and magnetic properties of α − FeGe2 films embedded in vertical spin valve devices”. doi: 10.1103/PhysRevMaterials.4.104415.

[3] D. Czubak et al. “Supplemental Material: Electronic and magnetic properties of α − FeGe2 films embedded in vertical spin-valve devices”. doi: 10.1103/PhysRevMaterials.4.104415.

[4] B. Jenichen et al. “Ordered structure of FeGe2 formed during solid-phase epitaxy”. doi: 10.1103/PhysRevMaterials. 2.051402.

[5] P. G. Evans et al. “Crystallization of amorphous complex oxides: New geometries and new compositions via solid phase epitaxy”. doi: https://doi.org/10.1016/j.cossms.2018.09.001

9:00am **NAMBE1-WeM-5 Twin-free $(MnSb_2Te_4)_x(Sb_2Te_3)_{1-x}$ Growth on In_2Se_3/InP(111):B Substrates by Molecular Beam Epitaxy**, ***Jisoo Moon**, Candice R. Forrester,* City College of New York*; Sina Mohammadi,* City College of New York, City University of New York*; Lia Krusin-Elbaum, Maria C. Tamargo,* City College of New York

$MnSb_2Te_4$ (MST) is a magnetic topological material in which Mn-Sb antisite defects play a critical role in determining the magnetic ground state. Due to the similar ionic radii of Mn and Sb, site-mixing is energetically preferred, driving the system easily to the ferromagnetic state. Antiferromagnetic MST crystals have been achieved by tuning growth parameters, such as the amount of Mn during synthesis, and through a post-growth treatment that introduces H^+ ions into the system. However, the effects of other types of crystalline defects on the site-mixing and, consequently, on the magnetic properties of this material system have not been considered. Here, we utilize our recent twin-free In_2Se_3-InP(111):B substrate for molecular beam epitaxy growth of $(MnSb_2Te_4)_x(Sb_2Te_3)_{1-x,}$ aimed at suppressing interfacial defects between the substrate and MST layers, thus towards twin-free MST. Un-twinned thin In_2Se_3 layers are formed via a selenium passivation technique during the oxide desorption of InP(111):B substrates. To prevent MST intermixing with In_2Se_3, thin topological insulator (TI) insertion layers are employed to separate the MST from In_2Se_3. ϕ-scan result of X-ray diffraction (XRD) shows the MST grown on the un-twinned In_2Se_3 is indeed twin-free, too. Low-temperature magnetic susceptibility measurements indicate a weakened ferromagnetic ground state, which is also consistent with electronic transport results. The concentration of the magnetic septuple layer (SL), van der Waals unit layer of MST, is investigated using high-resolution XRD, energy-dispersive X-ray spectroscopy, and transmission electron microscopy, which exhibit significantly lower concentrations of the SLs than in the ones grown on sapphire substrates. Considering the latter is expected to be twinned and, therefore, more defective, the SL concentration and Mn distribution under similar growth conditions appear to be closely related to crystalline defects. We will present more findings from XRD and atomic force microscopy for different growth parameters. The successful growth of twin-defect-suppressed MST will provide a new opportunity to better understand the relationship between material properties and defect types and levels in this material system.

9:15am **NAMBE1-WeM-6 Epitaxial Growth of SnGeSe Ternary Alloys on GaAs Substrates**, ***Kira Martin**, Pooja D. Reddy, Jarod E. Meyer, Kelly Xiao, Tri Nguyen, Kunal Mukherjee,* Stanford University

The heteroepitaxy of low-symmetry, non-cubic semiconductors on cubic substrates is a key challenge for integrating anisotropic electronic and optical properties on to technologically relevant device platforms. Tin selenide (SnSe) and Germanium selenide (GeSe) are layered orthorhombic (a≠b≠c, α=β=γ=90°)IV-VI semiconductors with bandgaps in the near-infrared and useful electrical, optical, and thermoelectric properties.[1,2] SnSe and GeSe comprise of bilayer structures with strong in-plane covalent bonding and weak van der Walls (vdW) bonding out-of-plane.[3] Alloying across the completely miscible SnSe-GeSe system helps tune the anisotropy, as the structure of GeSe is more anisotropic than SnSe. Developing heteroepitaxial growth of SnGeSe alloys on cubic III-V and Si/Ge substrates is a critical step towards utilizing the tunable anisotropic optical and electronic properties of orthorhombic structures for applications such as polarization sensitive detectors.

Despite the structural similarities between SnSe and GeSe, we find that thin film alloy synthesis is complicated by the low sticking coefficient and glass-forming character of GeSe. We deposit IV-VI alloys on arsenic-capped GaAs(001) substrates using molecular beam epitaxy (MBE). The substate was prepared by thermally desorbing the arsenic cap and then dosing the substrate surface with a PbSe flux at 420˚C. A 50 nm buffer layer of SnSe was then grown at 300˚C before growing the SnGeSe alloy film at 160˚C, based on seeing minimal Ge sticking in related IV-VI materials above 230˚C.[4] The relative beam equivalent pressures (BEP) of SnSe and GeSe compound effusion cells were changed to systematically sweep composition from crystalline SnSe to amorphous GeSe. Reflection high energy electron diffraction indicates Sn-rich growths are crystalline, but increasing GeSe BEP ratio eventually results in amorphous growth.

Alloy composition and structural information for the crystalline phases was determined using high resolution X-ray diffraction. The vdW-bonded ‘a-axis’ is in the out-of-plane direction, and reciprocal space maps show an in-plane epitaxial relationship with two 90˚ rotated domains—the ‘b’ and ‘c’ axes of the IV-VI alloys aligned to the in-plane <110> axes of GaAs. We will explore how the structural quality and optical properties of SnGeSe change as a function of Ge composition and study the potential for post-growth annealing to crystallize Ge-rich amorphous SnGeSe alloys into the orthorhombic phase.

[1]Z. Chen et al., *Prog. Mater. Sci.* **97**, 283 (2018).

[2]Y. Kim et al., *J. Korean Phys. Soc*. **72**(2), 238 (2018).

[3]S. Yang et al., *Nano Res.* **11**(1), 554 (2018).

[4]K. Xiao, *arXiv.* 2411.15464(2024).

9:30am **NAMBE1-WeM-7 Controlling Antiphase Twins in Bi_2Se_3 via Step-Terminated Al_2O_3 Substrates**, ***Matthew Brahlek**, Jane Chen,* Oak Ridge National Laboratory*; Rob Moore,* Oak Ridge Nation*; Alessandro R. Mazza,* Oak Ridge National Laboratory

The epitaxial synthesis of high-quality 2D layered materials is an essential driver of both fundamental physics studies as well as being central to bridge to technological applications. Bi_2Se_3, a prototypical 2D layered topological insulator, suffers from myriad defects imparted during the growth, either thermodynamically or due to the interaction with substrates. In this study, we demonstrate that step-terminated Al_2O_3 substrates with a high miscut angle (3°) can effectively suppress a particular hard-to-mitigate defect, the antiphase twin. Systematic investigations across a range of growth temperatures and substrate miscut angles confirm that atomic step edges act as preferential nucleation sites, stabilizing a single twin domain. First-principles calculations suggest a significant energy barrier for twin boundary formation at step edges, supporting the experimental observations. Detailed structural characterization indicates that this twin-selectivity is lost through the mechanism of the 2D layers overgrowing the step edges, leading to higher twin density as the thickness increases. These findings highlight the complex energetic landscape unique to 2D materials that is driven by interplay between substrate topology, nucleation dynamics, and defect formation, which is critical to optimizing growth strategies to improve material quality for quantum and electronic applications.

9:45am **NAMBE1-WeM-8 “Kinetic Roughening” in Low-temperature MBE Growth of III-As Heterostructures on InP(111)B**, ***Esperanza Luna**,* Paul-Drude-Institut für Festkörperelektronik Leibniz-Institut im Forschungsverbund Berlin, Germany*; Seyed Ali Hosseini Farahabadi, Milad Entezami, Man Chun Alan Tam, Zbigniew Roman Wasilewski,* University of Waterloo, Canada

MBE growth of III-V semiconductors on (111)-oriented substrates has gained renewed interest in recent years. For example, the use of InGaAs/InAlAs short-period superlattices SL grown on InP(111)B substrates have been proposed as an alternative antenna material for time-domain spectroscopy (TDS) systems using 1.55 μm femtosecond laser pumps. Furthermore, low temperature (LT) growth leading to the incorporation of excess As via arsenic antisites should enable more efficient TDS systems due to a faster recombination of photocarriers through As antisites in the structure. The first prerequisite however is the growth of high-quality LT InGaAs/InAlAs SPSL on InP(111)B, an effort which combines the challenges of InGaAs/InAlAs growth on InP(111)B with those of LT MBE.

To explore the impact of the LT substrate temperature (Ts) on the SL structure, 50 periods of lattice-matched InGaAs/InAlAs on InP(111)B were grown changing Ts every 10 periods of SL from 450°C to 250°C while maintaining other parameters unchanged. The sample microstructure was investigated using a combination of scanning transmission electron

microscopy (S)TEM techniques.

We find that whereas the first 10 periods of InGaAs/InAlAs SL (Ts = 450°C) grow in a regular fashion, the structure progressively deteriorates as Ts is reduced, with a dramatic degradation for the SL grown at the lowest Ts of 250 °C (Fig. 1). We observe a noticeable change in the microstructure after reducing Ts from 450° to 400°C, the resulting periods are characterized by a high density of in-plane microtwins and staking-faults (Fig. 2). The situation rapidly evolves towards even more defective layers as Ts is further reduced (Fig. 1). In fact, the SL grown at Ts = 250°C is highly defective, its microstructure resembles the initial stages of breakdown of epitaxy.

We note that the observed microstructure and its dependence on Ts can be explained in terms of the critical role of step-flow growth and of Ehrlich-Schwoebel (ES) barriers when growing on the (111)B-oriented substrate. Despite the use of InP(111)B substrates with a 2° miscut to promote step-flow growth, it is obvious that reduction of Ts below 400° C without further change of i.e. the V/III ratio, is not enough to maintain the regular step-flow regime, leading to a “kinetic roughening”. We suggest that a readjustment of the growth conditions for each individual Ts might help to preserve the step-flow growth mode and to overcome the impact of ES barriers, leading to higher quality layers. We believe that these results are of general validity to other material systems and contribute to the understanding of the always challenging growth on (111)-oriented substrates.

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