NAMBE

Room Tamaya ABC - Session NAMBE1-MoM

Remote and van der Waals Epitaxy

Moderator: Justine Koepke, Sandia National Laboratories

8:00am **NAMBE1-MoM-1 Welcome & Sponsor Thank Yous***,*

8:15am **NAMBE1-MoM-2 Art Gossard MBE Innovator Awardee Talk, INVITED**

8:45am **NAMBE1-MoM-4 Epitaxy and Magnetotransport of GdAuGe on Gallium-Intercalated Graphene/SiC (0001)*, Zachary LaDuca,*** University of Wisconsin - Madison*; Chengye Dong,* Pennsylvania State University*; Nicholas Hagopian, Paul Voyles,* University of Wisconsin - Madison*; Joshua Robinson,* Pennsylvania State University*; Jason Kawasaki,* University of Wisconsin - Madison

Superconducting proximity effects in topological materials and magnetic materials are potential routes to artificially construct topological superconductors and spin-triplet superconductors, respectively. Motivated by recent demonstrations of proximity induced superconductivity in $(Bi,Sb)_2Te_3$ on gallium-intercalated graphene/SiC (*Nat. Mater.* **22**, 570-575 (2023)), here we demonstrate the molecular beam epitaxy growth of the antiferromagnetic topological nodal-line semimetal GdAuGe on gallium-intercalated graphene/SiC(0001), where the intercalated gallium layer is superconducting. This heterostructure provides a platform for investigating proximity-induced superconductivity in GdAuGe.

We compare the growth characteristics of GdAuGe on gallium-intercalated graphene/SiC to those on conventional graphene/SiC. While the in-plane epitaxial relationship remains unchanged—GdAuGe [2 -1 -1 0] || graphene [1 -1 0 0] || SiC [2 -1 -1 0]—the intercalated gallium layer modifies the potential energy landscape beneath the graphene without altering the graphene lattice. This system provides a platform to explore how remote interactions influence film growth in a van der Waals epitaxial system. Ongoing efforts focus on characterizing the structural and electronic properties of the films via x-ray diffraction, atomic force microscopy, and cross-sectional scanning transmission electron microscopy to assess crystallinity and interface quality, while low-temperature magnetotransport measurements will probe superconducting proximity effects from the gallium layer.

9:00am **NAMBE1-MoM-5 Challenges of Remote Epitaxy in Ultra-High Vacuum: Clean Semiconductor-2D Material Stacks*, Manny de Jesus Lopez,*** *Sadhvikas Addamane, Justine Koepke,* Sandia National Laboratories, USA*; Kevin Jones,* University of Florida, Gainesville*; Scott Schmucker,* Sandia National Laboratories, USA

The growth of high-quality heteroepitaxial films is often constrained by lattice mismatch and the availability of suitable substrate materials. Remote epitaxy (RE) offers the potential to overcome these limitations by incorporating a two-dimensional (2D) material, such as graphene, between the substrate and the epitaxial layer. This work explores RE techniques and addresses the challenges associated with traditional remote epitaxy processes. Conventional epitaxy relies on the formation of covalent or ionic bonds between incoming adatoms and the substrate lattice. In contrast, RE leverages a monolayer of graphene to mediate the interaction between the adatoms and the underlying substrate without direct bond formation. This lack of covalent bonding allows the epitaxial layer to relax, reducing the nucleation of dislocation defects and enabling the growth of relaxed epilayers on lattice-mismatched substrates. Despite the straightforward concept of RE, the growth mechanisms differ from conventional models due to the absence of covalent bonds, making it challenging to distinguish RE from processes such as van der Waals epitaxy or pinhole-seeded epitaxy. Additionally, the graphene transfer process can introduce structural defects that promote film nucleation and potentially obscure the RE mechanisms facilitated by weak interfacial forces. Advancing RE requires addressing the cleanliness of the graphene-substrate interface. This work discusses our efforts to improve the fabrication of III-V material stacks via MBE and to understand the competing influences of the remote substrate and graphene defects on through-graphene epitaxial growth. By performing the entire process within an ultra-high vacuum environment and exploring the use of alternative 2D materials, we aim to mitigate contamination and structural defects, providing a clearer understanding of the fundamental growth mechanisms in remote epitaxy. SNL is managed and operated by NTESS under DOE NNSA contract DE-NA0003525

9:15am **NAMBE1-MoM-6 Impact of Graphene-induced Surface Reconstructions on the Mechanisms for Remote and van der Waals Epitaxy of GdAuGe on Graphene/SiC (0001)*, Taehwan Jung,*** *Nicholas Hagopian,* University of Wisconsin - Madison*; Quinn Campbell,* Sandia National Laboratories*; Anshu Sirohi,* University of Wisconsin - Madison*; Chengye Dong,* The Pennsylvania State University*; Sadhvikas Addamane, Justine Koepke,* Sandia National Laboratories*; Joshua Robinson,* The Pennsylvania State University*; Paul Voyles, Jason Kawasaki,* University of Wisconsin - Madison

Remote epitaxy has emerged as a promising technique for synthesizing single-crystalline membranes, enabling flexible electronics and the exploration of novel properties under extreme strain. This method relies on the premise that a film can grow on a graphene-covered substrate through remote interactions that penetrate the graphene, allowing the film to grow as if the graphene were semi-transparent. However, in several cases, epitaxial graphene is known to induce long-range reconstructions in the underlying substrate, which implies strong graphene-substrate interactions. Prime examples include the (6x2) reconstruction of graphene/Ge (110) and the $(6\sqrt{3} \times 6\sqrt{3})R30°$ reconstruction of Buffer-graphene on SiC (0001). The impact of these surface reconstructions on the mechanisms for remote and van der Waals epitaxy is typically overlooked. In this study, we investigate the transparency of graphene using two types on a 6H-SiC (0001) substrate: Epitaxial-graphene, which is more decoupled from the substrate, and Buffer-graphene, which remains partially covalently bonded to the underlying SiC. We find that GdAuGe films exhibit a distinct in-plane rotation of 30 degrees when grown on Buffer-graphene/SiC compared to Epitaxial-graphene/SiC. Additionally, scanning transmission electron microscopy (STEM) reveals interfacial reconstructions and semi-disordered layers in the GdAuGe at the graphene/GdAuGe interface. To elucidate the origin of these findings, Density Functional Theory (DFT) calculations show that Buffer-graphene induces stronger substrate potential fluctuations than Epitaxial-graphene, while X-ray Photoelectron Spectroscopy (XPS) is expected to reveal differences in bond strength between GdAuGe and the two graphene types, further supporting our results. These findings provide new insights into the role of graphene-substrate interactions in remote epitaxy and suggest possible strategies for controlling film orientation and interface structure.

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9:30am **NAMBE1-MoM-7 Selective Area Epitaxy of van der Waals Materials*, Ryan Trice,*** *Stephanie Law,* Penn State University

Two-dimensional (2D) van der Waals (vdW) materials are interesting for a variety of applications ranging from optoelectronics and photocatalysis to energy storage and topological devices. However, vdW materials synthesized using common techniques like chemical or physical vapor deposition often have a high density of growth-related defects ranging from grain boundaries and twin defects to pyramidal growth and spiral defects. While pyramidal growth can be minimized through higher growth temperatures, grain boundaries, twin defects, and spiral defects are much harder to overcome.For many applications, especially in electronics and optics, these defects lead to non-radiative recombination, electron scattering, and other undesirable effects. Furthermore, the fabrication of 2D materials into quantum dots (QDs) through bottom-up methods faces problems with precise location placement and polydispersity in the QDs diameters. This makes the QDs difficult to characterize and is not ideal for most quantum computing and optical setups. Top-down nanofabrication approaches fix this issue but often cause significant damage to the surfaces or edges of the materials. To address these issues, we used selective area epitaxy (SAE) to grow Bi_2Se_3 thin films. SAE is a technique in which thin films nucleate and grow in defined areas on a wafer. This is done through use of a patterned mask where growth conditions are selected such that the film will only nucleate on the substrate.

In this talk, we will describe SAE growth of Bi_2Se_3 on Al_2O_3 (001) and Si (111) substrates using a SiO_2 mask. The mask was deposited onto a 10x10mm substrate by atomic layer deposition. Etching of the SiO_2 mask was done with standard photolithography techniques and a direct write laser beam lithography system, and the SiO_2 was removed from selected areas using a wet chemical etch, resulting in micron-scale holes of various shapes and sizes. The processed substrates were then loaded into a molecular beam epitaxy chamber for growth of the Bi_2Se_3 film. First, we will discuss the effects of different substrate temperatures on the selective growth of the Bi_2Se_3 thin films. Second, we will discuss the geometric influence of variously shaped patterns on the crystal quality of the selectively grown

films. Third, we look at the effect and viability of nano-scale patterns for selective growth of vdW materials. Further studies will focus on using different materials for the substrate and mask. This approach could allow us to grow wafer-scale, defect-free 2D vdWs QDs at specified areas on the wafer, thereby increasing the scalability and applicability of these materials to real-world challenges.

9:45am **NAMBE1-MoM-8 Impact of Point and Extended Defects on the Mechanism for Remote Epitaxy of GaAs on Graphene/Ge**, ***Anshu Sirohi***, *Patrick Strohbeen, Sebastian Manzo, Katherine Su, Vivek Saraswat, Nicholas Hagopian, Paul Voyles, Michael Arnold, Jason Kawasaki,* University of Wisconsin - Madison

Remote epitaxy is an advanced approach that enables the growth of single crystalline thin films on graphene-terminated substrates without direct chemical bonding between the films and the substrates[1, 2]. In the simplest picture, the grown films imitate the crystallographic structure of the underlying substrate as if the graphene layer were transparent. This approach facilitates lattice-matched epitaxy while empowering film exfoliation, wafer recycling and flexible device integration[3]. However, the microscopic mechanisms for remote epitaxy remain unclear, in particular, the role of point and extended defects as nucleation sites. The potential fluctuation through graphene on atomically flat GaAs is calculated by DFT to be ~15 meV, which is small compared to kT at growth temperatures. This opens the question of whether 15 meV is enough to serve as a template for epitaxial alignment. On the other hand, the potential fluctuation at a graphene covered atomic step edge should be much larger compared to flat regions, potentially enhancing remote interaction. Therefore, investigating early-stage nucleation on graphene-terminated substrates and the effect of the density of atomic steps are crucial to comprehend the possible intrinsic mechanism of epitaxy.

Here, we focus on most commonly used system i.e. III-V semiconductor GaAs. The films of GaAs were synthesized on graphene terminated germanium substrates using molecular beam epitaxy. The structural and chemical properties were characterized using RHEED, XRD, and XPS, evidencing the formation of crystalline GaAs films. Detailed surface morphological information before and after film growth were analyzed using AFM and in-situ STM. Preliminary results indicate that the nucleation of the GaAs film is influenced by the atomic steps on Ge111. Additionally, on Ge110, the nucleation appears to be strongly affected by the surface reconstructions of Gr/Ge110.

All these insights will allow me to shed light on the early stage nucleation in the monolayer regime in GaAs/Gr/Ge system.

References:

[1] Kim Y et al. Nature 544 340 (2017).

[2] Lee M L. Nature 544 301–2 (2017).

[3] Guo, Y. et al. Nano Lett. 20, 33–42 (2019).

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