Science and Technology of MBE
Room Ballroom A - Session ST-WeM2

Fundamentals of MBE Growth

Moderator: Dr. Stephanie Law, University of Delaware

10:00am **ST-WeM2-9 Doping and Surfactant Behavior of Antimony and Gallium in Molecular Beam Epitaxy Grown Germanium-Tin***, **Amanda Lemire**, K. Grossklaus, C. Jamison, V. Vazquez, P. Hennessey, T. Vandervelde,* Tufts University

The bandgap of germanium-tin ($Ge_{1-x}Sn_x$) alloys can vary from ~0.8 eV into the far-IR as the Sn composition increases, and transition from indirect to direct bandgaps between 6 and 25 at% Sn depending on strain. Consequently they are being developed for a range of thermophotovoltaic (TPV), thermal imaging, and photonic data transmission applications. However, at useful Sn contents these alloys are metastable and require low epitaxial growth temperatures. Therefore they can only be grown to limited thickness, otherwise films may experience Sn segregation and defect formation. Growing below ideal temperatures for Ge can also induce defects, reducing electrical conductivity. Growth and thickness limitations complicate PV cell design and optimization. One mitigation strategy for non-optimal growth conditions is to deposit and maintain on the growth surface a thin coverage of an additional atomic species, which acts as a surfactant to modify adatom interactions. Surfactants change the surface energy of the growth front, which controls the mobility of adatoms and affects the rate of positional exchange between buried and surface layers. A surfactant that preferentially moves to the growth surface could increase the incorporation of Sn into deeper layers and reduce defects formed due to growth at low temperatures. Additionally, the filling of partial growth layers by surfactant atoms may reduce islanding and so improve surface roughness.

In this work, antimony was applied as an *n*-type dopant and surfactant during the growth of GeSn alloys to potentially improve the crystallinity of Ge and GeSn films grown by molecular beam epitaxy. Previous work has shown Sb suppresses roughness in SiGe film growth. Gallium was applied as a *p*-type dopant and surfactant, providing a comparison between dopant types and differing surfactant atomic sizes. Pairs of GeSn samples were deposited without and with the codeposition of Sb or Ga. The growth temperature, film thickness, and Sn content were varied to investigate the impact of each surfactant on Sn incorporation and critical thickness. Amount of Sb or Ga incorporation into the growing film depends on growth conditions, so this study also provides data on the retention of Sb and Ga as dopants during low-temperature GeSn growth. Sn content and film quality were characterized by X-ray diffraction, photoluminescence and Raman spectroscopy, spectroscopic ellipsometry, and optical microscopy. Electrical properties were examined by Hall effect measurement. Results will be discussed in terms of how surfactants have modified the kinetics of the growth process and considered with design of a GeSn based photodiodes and TPV cells in mind.

10:15am **ST-WeM2-10 SiO_2 Surface Planarization for Selective Area Regrowth of High Aspect Ratio Microstructures***, **Ashlee Garcia**, A. Skipper, M. Bergthold, S. Bank,* University of Texas at Austin

A molecular beam epitaxy (MBE) approach to selective area epitaxy (SAE) of III-V semiconductors has the potential to advance optoelectronic structures through seamless integration of metals, dielectrics, and high-quality crystalline semiconductors. While SAE by metal organic chemical vapor deposition has been widely successful due to its high material deposition selectivity, an all-MBE method could enable further advances through its high layer precision and access to non-equilibrium growth conditions[1,2].

SAE is difficult to achieve with conventional MBE due to III-V nucleation on the amorphous mask necessitating high growth temperatures and low growth rates to mitigate polycrystal formation[2-4]. While an all-MBE approach has enabled the embedding of features up to 300nm tall, applications requiring high aspect ratio microstructures such as mid-/long-wave infrared high-contrast photonics[5-7] and aspect ratio trapping of dislocations for metamorphic growth[8], are more challenging to access due to increased surface roughness of micron-scale features[7]. Increased roughness and defects on the mask surface lowers the barrier for nucleation further restricting the selective growth regime[8].

In this study, we explore the use of hydrogen silsesquioxane (HSQ) surface planarization to restore the surfaces of micron-scale films for fabrication of high aspect ratio SiO_2 features with surfaces comparable to that of thin films[9,10]. Integrating surface planarization with a 100nm layer of HSQ in the fabrication of 1.5µm tall features demonstrated a significant decrease in film roughness from a root-mean-square roughness of 3.95nm to 0.75nm. Furthermore, selectivity studies on cured HSQ films have shown selectivity equivalent to that of SiO_2 deposited by plasma-enhanced chemical vapor deposition. Experiments are underway to utilize the technique to achieve selective growth of high aspect ratio microstructures.

[1] D.J. Ironside et al., J. Cryst. Growth (2019). [2] A.M. Skipper et al., 2019 MRS EMC. [3] F.E. Allegretti et al., J. Cryst. Growth (1995). [4] S.C. Lee et al. J. of Appl. Phys. (2002). [5] Jun Wang et al. 2017 Laser Phys. Lett. 14 125801. [6] J.Z. Li et al. Appl. Phys. Lett. 91 (2) (2007). [7] M. R. Amirzada et al. Appl Nanosci 6, 215–222 (2016). [8] M. Ohring, The Material Science of Thin Films, Academic Press (1992). [9] F. Salmassi et al, Applied Optics, Vol. 45, No. 11 (2006). [10] C.-C. Yang and W.-C. Chen, J. Mater. Chem., 2002,12, 1138-1141.

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10:30am **ST-WeM2-11 InP Lateral Epitaxial Overgrowth by Solid-Source Molecular Beam Epitaxy***, **Yiteng Wang**, R. Hool, W. North, S. Pandey, E. Raftery, K. Choquette, M. Lee,* University of Illinois at Urbana-Champaign

Lateral epitaxial overgrowth (LEO) of InP on buried dielectrics (typically SiO2 and SiN) has been investigated for optical telecommunication devices [1],[2]. LEO on dielectric patterns usually requires coalescence from multiple crystal fronts, which can introduce structural defects including threading dislocations (TDs) and stacking faults (SFs) [3]. In this work, we demonstrate selective InP nucleation and coalescence on patterned InP substrates with designed feature sizes of 100-1110 nm by solid-source molecular beam epitaxy. Structural defects formed due to lateral growth and coalescence for LEO on all features, including triangles, polygons, and stripes. We found that LEO on features < 300 nm with sides aligned away from [0 -1 1] and [0 1 1] directions gave the lowest defect density.

The dielectric patterns investigated here included arrays of 40 nm thick SiN isosceles-right triangles with side lengths of 465-1110 nm. The sides of the triangles were aligned along [0 -1 1] and [0 1 1], and the hypotenuses were aligned along [0 1 0] crystallographic directions. 20 and 350 nm layers of Be-doped InP were grown at a substrate temperature (Tsub) of 500°C and a growth rate of 0.2 µm/hr to maintain selectivity [4]. Thicker samples of 1300 nm p-InP were initiated using the same conditions for the first 500 nm, followed by a second growth step at more typical InP MBE growth conditions of 440°C, 1 µm/hr.

InP growths of 20-350 nm at T_{sub} = 500°C show selective growth was maintained, as no polycrystalline nucleation was found on dielectrics by secondary electron imaging (SEI). Next, after 1300 nm growth, the InP surface was fully coalesced for all feature sizes, with small voids enclosed above the dielectrics. Electron channeling contrast imaging (ECCI) reveals that LEO on 465 and 1110 nm triangles possess high threading dislocation densities TDD > 10^7 cm^{-2}. Further study of InP LEO on SiO2 lines (120 nm thick/230 nm wide) oriented along the three low-index in-plane directions shows high defect density along [0 1 1] and [0 -1 1] and lower defect density along [0 1 0].

To reduce TDD, we performed overgrowth on an array of 40 nm thick SiN polygons with side lengths of 266 nm and most of the sides oriented away from [0 1 1] and [0 -1 1] directions. Taking advantage of the side orientation and reduced feature sizes < 300 nm, InP LEO on polygon patterns shows an order of magnitude reduction in TDD (5.0×10^6 cm^{-2}) and fewer SFs compared to the 465 nm and 1110 nm triangles. Future work will focus on better understanding the effect of dielectric thickness, size, and orientation on the formation of extended defects during InP LEO.

10:45am **ST-WeM2-12 Effect of Molecular Beam Epitaxy (Mbe) Growth Conditions on the Structural and Magnetic Properties of High Curie Temperature $(MnSb_2Te_4)_x(Sb_2Te_3)_{1-x}$ Magnetic Topological Insulators***, **Candice Forrester,*** The City college of New York, Lehman College*; C. Testelin,* Institute des NanoSciences de Paris, France*; K. Wickramasinghe,* The City College of New York*; I. Levy,* New York University*; X. Ding, L. Krusin-Elbaum,* The City College of New York*; G. Lopez,* Lehman College*; M. Tamargo,* The City College of New York

Recently it has been shown that structural disorder in 3D Topological Insulators (TIs) has considerable effects on the properties of the materials. The addition of magnetic ions like Mn breaks time reversal symmetry and opens a gap in the Dirac point.[1] This addition also changes the crystal structure from the typical quintuple layer (QLs) structure of non-magnetic TIs to a septuple layer (SLs) structure.[2] Furthermore, addition of a Mn flux during MBE growth results in self-assembled structures of mixed QLs and SLs.[2]

Previously we reported the MBE growth of self-assembled structures of $(MnSb_2Te_4)_x(Sb_2Te_3)_{1-x}$ magnetic topological materials, and showed that their Curie temperature (T_C) is dependent on the composition x (or %SL). Samples with $0.7 < x < 0.85$ exhibit very high T_C values. Additionally, it was observed that as the Mn beam equivalent pressure (BEP) ratio used was increased, there was a corresponding increase in T_C.Decreasing the growth rate further increased T_C to >100 K, the highest values reported for this material system. An understanding of how the changes in growth conditions lead to the T_C enhancement is not well-established.

Here we investigate the structural properties of the materials as they relate to the growth conditions, specifically growth rate (GR). Samples grown at slow GRs (0.4 – 0.6 nm/min) were compared to samples grown at fast GRs (0.8 – 1.0 nm/min). Samples were investigated by X-ray diffraction, Energy Dispersion X-Ray Spectroscopy (EDS), Hall effect and scanning transmission electron microscopy (STEM).

We found that for the same Mn BEP ratio, low GR yields similar composition x as fast GRs. On the other hand, EDS showed that for $x > 0.7$ there was increased intermixing between Sb and Mn in both the fast and the slow GR samples. However, the samples grown with slow GR showed much greater Mn and Sb intermixing, as well as Mn and Te intermixing, suggesting increased Mn incorporation at the slow GR. Cross sectional EDS studies reveal a high Mn content in the QLs, consistent with $(Sb_{1-y}Mn_y)_2Te_3$ alloy formation. Hall effect measurements show that GR does not significantly affect the electrical doping in $(MnSb_2Te_4)_x(Sb_2Te_3)_{1-x}$ supporting the proposal that a super-exchange magnetic mechanism is likely at play. Other techniques, such as magnetic force microscopy (MFM) are being explored to better understand the magnetic mechanisms leading to high T_C values. Our results provide insight as to how to achieve on-demand magnetic TIs with enhanced properties.

[1] Y. Tokura et al, *Nature Reviews Physics* **1**, 126 (2019)

[2] J.A. Hagmann et al, *New Journal of Physics* **19**, 085002 (2017)

[3] I. Levy et al, *Crystal Growth & Design* **22**, 3007 (2022)

11:00am **ST-WeM2-13 Evolution of Antiferromagnetic Spin Texture in MBE-Grown Epitaxial Multiferroic BiFeO3***, **Maya Ramesh,*** 105 Worth Street*; P. Meisenheimer,* University of California, Berkeley*; S. Zhou,* Brown University*; P. Stevenson,* Northeastern University*; L. Caretta,* Brown University*; R. Ramesh,* Rice University*; D. Schlom,* Cornell University

Bismuth ferrite (BiFeO3) is a lead-free magnetoelectric multiferroic showing antiferromagnetic order and a large spontaneous polarization at room temperature. This antiferromagnetic order in BiFeO3 is complex, where, as a consequence of the Dzyaloshinskii-Moriya Interaction (DMI), a small canting of the antiferromagnetic order forms a chiral spin cycloid in bulk samples. Understanding the interplay between the ferroelectric polarization and the spin cycloid, as well as its electric field manipulation, is of significant interest for antiferromagnetic spintronics and next generation computation. There is still much to learn about BiFeO3's intrinsic antiferromagnetic structure in thin films, where epitaxial strain imposed by an underlying substrate can perturb and even destroy the spin cycloid. MBE provides an ideal pathway to create BiFeO3 with unparalleled structural perfection. As a model system, we have synthesized epitaxial thin films (2-100 nm) of BiFeO3 on (110) TbScO3 substrates via oxide molecular-beam epitaxy. In this work, we explore how epitaxial strain affects the formation and orientation of the spin texture in BiFeO3 using nitrogen-vacancy magnetometry (NV magnetometry). NV magnetometry uses a nitrogen vacancy implanted at the tip of a diamond cantilever which acts as a single-spin magnetometer to sensitively map nanoscale surface stray fields. By correlating the spin texture to ferroelectric information measured using piezoresponse force microscopy, we are able to explore coupling between the two order parameters. Electric-field-dependent measurements show that the spin cycloid can be manipulated by switching the ferroelectric polarization, which is of great interest in low dissipation magnonics and spintronics.

11:15am **ST-WeM2-14 Adsorption-Controlled Growth of $SrTiO_3$ by Oxide MBE***, **Dylan Sotir**, M. Barone, D. Schlom,* Cornell University

Historically, the growth of stoichiometric $SrTiO_3$ by conventional molecular-beam epitaxy (MBE), utilizing elemental sources, has been challenging due to the precise calibration required. It would be useful if there were a thermodynamic regime where the composition of $SrTiO_3$ was self-regulating. One demonstrated way to achieve such automatic control of stoichiometry in $SrTiO_3$ is by supplying an excess of the volatile organometallic precursor titanium isopropoxide to grow $SrTiO_3$ by metalorganic MBE (MOMBE). Another means, and the one investigated here, is to stick with elemental sources but increase the substrate temperature. When $SrTiO_3$ is heated to high temperature, it does not evaporate congruently; rather, it loses more strontium than titanium. This difference in the vapor pressures of the volatile species suggests that adsorption-controlled growth of $SrTiO_3$ may be possible by conventional MBE. We report a thermodynamic window in which SrTiO3 can be grown via adsorption control by conventional MBE. A new high-temperature laser substrate heater installed in Cornell University's PARADIM Thin Film Facility allows substrate temperatures to reach up to 2000 °C.

We show that the growth window for our highest quality adsorption-controlled $SrTiO_3$ is from approximately 1450-1475°C, with a Sr:Ti ratio of 5:1. All films were grown on LaAlO3 (100) substrates. Figure 1 shows XRD data for samples grown at several temperatures including the adsorption-control window.

11:30am **ST-WeM2-15 MBE of Ba_2BiTaO_6,a Candidate *p*-type Oxide Semiconductor***, **Anna Park**, Y. Birkhölzer, M. Barone, D. Schlom,* Cornell University

Complementary metal-oxide-semiconductor (CMOS) technology is an important part of today's integrated circuit technology. CMOS replaced *n*-channel metal-oxide-semiconductor (NMOS) in the 1980s and with the 100 to 1000x power savings advantage it provides, enabled integrated circuits to grow from tens of thousands of NMOS transistors on a chip to tens of billions of CMOS transistors today. Today we stand at similar cross-roads for transistors made from oxide semiconductors. Only high-performance n-channel oxide transistors (and thus NMOS) exist for oxide transistors. If high performance *p*-channel oxide transistors could also be made, low-power CMOS would be possible in oxide systems and enable low-power transparent electronics. Although many *p*-type semiconducting oxides have been predicted based on their theorized electronic properties, few have been realized in experiment and those that have been achieved have much lower mobility than established n-type oxides like indium-gallium-zinc-oxide or In_2O_3. Unfortunately, the realization of *p*-type oxides is particularly difficult due to the localization of the oxygen 2*p* orbitals. One design criterion to realize *p*-type oxides is to create oxides with Sn^{2+}, Pb^{2+}, or Bi^{3+} cations that have dispersive filled *s*-orbitals that will hybridize with oxygen 2*p* orbitals to delocalize the hole states and enhance mobility. We show that suboxide MBE can be used to grow Ba_2BiTaO_6, a candidate transparent *p*-type oxide.

Suboxide MBE utilizes molecular beams of suboxides, where the incoming cation precursors are already in the desired oxidation states. Many oxides crystallize with a perovskite structure and offer flexibility in accommodating a variety of cations, resulting in a wide range of properties. Double perovskites, of which Ba_2BiTaO_6 is an example, offer additional flexibility and another degree of freedom to explore in the interplay of structure and properties. For Ba_2BiTaO_6, we take advantage of suboxide MBE and avoid using an electron beam to evaporate Ta by supplying a molecular beam of TaO_2 from a Ta_2O_5 source. Additionally, with the volatility of Bi at our growth temperatures, we grow in an adsorption-controlled regime and fine tune the flux of Ba and Ta cations to create the desired phase.

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