

Science and Technology of MBE

Room Hall of Ideas E-J - Session ST-MoP

Science and Technology of MBE Poster Session

ST-MoP-1 Shadow Mask Molecular Beam Epitaxy, Shagorika Mukherjee, S. Sitaram, X. Wang, University of Delaware; S. Law, Penn State University

Shadow mask molecular beam epitaxy (SMMBE) is a form of selected area epitaxy (SAE) in which one can pattern vacuum-deposited films using a mechanical mask. If the mask is not a contamination source itself and is near the substrate, epitaxial layers may be deposited on the substrate sequentially through the patterned mask. In this way, a patterned film can be produced without etching. A unique feature of SMMBE is the shadowing effect that arises near the mask edges, which causes the elemental fluxes to change as a function of position. The shadowing effect occurs when the mask edges are not perfectly vertical. This results in a gradient of film thickness and/or composition. The gradient can be controlled by changing the mask thickness and/or the angle of the mask edges. In this presentation, we demonstrate the potential of SMMBE to produce in-plane gradient permittivity materials (GPMs) by taking advantage of the shadowing effect.

GPMs are materials in which the permittivity changes horizontally in the plane of the sample. There are a variety of applications of GPMs, we are focused on the excitation of surface plasmon polaritons (SPPs) which can confine light and enhance its electric field. In a GPM, light of different wavelengths is confined at different positions on the sample. If successful, this type of material could be used to develop an on-chip spectrometer. Since we are interested in working in the infrared, our GPMs consist of heavily silicon-doped InAs, which is known to be a good infrared plasmonic material. By varying the silicon doping density in the plane, we can control the Si:InAs permittivity in the lateral direction. We propose to create such a material using SMMBE with reusable silicon shadow masks. The shadow masks are each 200 μm thick and have a dimension of 1 cm \times 1 cm. Each mask has an aperture at its center. The aperture has slanted inner walls, which makes its dimension 0.5 cm \times 0.5 cm at the top and 0.528 cm \times 0.528 cm at the substrate side. To date, we have studied the effect of several growth parameters in controlling the in-plane permittivity in the GPMs. In comparison to non-SMMBE growth recipe, we have found that increased quantities of Bi surfactant (2.5x higher) and As (1.5x higher) in the growth recipe of the GPM can significantly minimize poor growth regions around the edges of GPM where the permittivity gradients are. Characterization by s-SNOM (scattering-type scanning near-field optical microscopy) using mid-IR light source illustrates that the GPMs we have made confine different wavelengths of light at different in-plane positions in the GPMs, setting the stage for the creation of an on-chip spectrometer.

ST-MoP-2 Strain Relaxation of Graded InGaN and AlGaIn, Reem Alhelais, F. Mais de Oliveira, H. Stanchu, M. Sarollahi, Y. Mazur, N. Al-Hosainy, M. Ware, University of Arkansas

III-nitride semiconductor materials have proved to be ideal materials for high-power, high-frequency, and high-temperature applications because of their tunable direct band gaps, high breakdown voltage, high absorption coefficient, resistance to defects, lattice match, and polarization characteristics. These materials form a continuous alloy system with direct bandgaps from 6.2 eV (AlN) through 3.4 eV (GaN) to 0.7 eV (InN). The compositionally graded Group III-nitride alloy enables access to a large range of energies through varying the bandgap. This change in bandgap is achieved by varying the indium and aluminum composition during growth, which yields excellent compatibility for various optoelectronic applications.

The growth and quality of mismatched heteroepitaxial III-Nitrides layers are generally influenced by strain relaxation mechanisms that release the accumulated strain energy. Plastic relaxation is generally started by the formation of misfit dislocations above the critical thickness. This has been well studied and is generally understood for heteroepitaxial films with a fixed composition. Graded composition films have been investigated recently for potential incorporation into semiconductor devices, however, the issue of plastic relaxation for graded III-Nitride semiconductors has not been thoroughly investigated.

Graded InGaIn and AlGaIn can be grown pseudomorphically strained to its substrate until some critical amount of strain energy is built up. This can happen either as a result of reaching a maximum composition or a maximum growth thickness. These two parameters are not independent in terms of their contribution to the buildup of strain energy, and the goal of

this study is to determine both the range over which these alloy layers can be grown without relaxation and the mechanisms by which they exhibit relaxation.

In the present work, we have grown both graded InGaIn and AlGaIn layers with 30 % of In and Al composition of increasing thicknesses for 15 min, 30 min, and 60 min on GaN substrates. We investigated their properties through X-ray diffraction reciprocal space mapping (RSM). With increasing the thickness of these graded layers, the InGaIn or AlGaIn signature in the RSM shifts from a fully strained position. Atomic force microscopy will be also used to characterize the sample surface of interest, including dislocation density, while transmission electron microscopy will be used to understand the nature of relaxing defects that is formed in these layers. How these introduced dislocations impact the electrical and optical properties will be demonstrated through photoluminescence and Raman spectroscopy.

ST-MoP-3 Improved N-Type Doping Activation in AlGaAs:Si Through Digital Alloy Growth, Xizheng Fang, B. Kim, D. Montealegre, Y. Wang, A. Birge, M. Lee, University of Illinois at Urbana Champaign

Despite its maturity, MBE growth of Si-doped Al_xGa_{1-x}As (n-Al_xGa_{1-x}As:Si) with moderately high electron concentration ($n_0 > 1 \times 10^{17} \text{ cm}^{-3}$) remains challenging due to the formation of DX centers and other traps, particularly near the direct to indirect band transition at $x=0.45$.¹ Al_xGa_{1-x}As surfaces are also prone to roughening during growth², which can negatively impact subsequent growth of active regions. In this work, we show that n-Al_{0.4}Ga_{0.6}As:Si grown as a 3 monolayer (ML) GaAs/2 ML AlAs digital alloy at 610°C exhibits ~34x higher activation than in bulk samples, enabling straightforward doping up to $n_0 = 2.5 \times 10^{17} \text{ cm}^{-3}$ with smooth surface morphology.

All samples were grown on semi-insulating GaAs (001) and consisted of a 200 nm GaAs buffer, a 100 nm undoped Al_{0.4}Ga_{0.6}As layer to prevent charge transfer into the GaAs buffer, and a 500-nm-thick n-Al_{0.4}Ga_{0.6}As:Si layer followed by a 9 nm n⁺-GaAs cap to facilitate ohmic contact formation. In all cases, the growth rate was held at 0.65 $\mu\text{m/hr}$ with V/III of 30 and a target Si concentration [Si] = $1 \times 10^{19} \text{ cm}^{-3}$.

We started with the growth of a bulk n-Al_{0.4}Ga_{0.6}As:Si alloy control sample, which gave a root mean square (RMS) roughness of 2.1 nm and $n_0 = 7.3 \times 10^{15} \text{ cm}^{-3}$ (activation = 0.073%) according to Hall effect measurements. In an attempt to increase the activation, we reduced [Si] to $5 \times 10^{18} \text{ cm}^{-3}$ and added 10x Si delta doping spikes³ at 50 nm intervals to bring the integrated Si concentration to $1 \times 10^{19} \text{ cm}^{-3}$. However, the activation only improved by 40%, while the roughness increased by 2x. Next we grew a 6 ML GaAs/4 ML AlAs digital alloy (6/4 DA), with the rationale that Si atoms residing within layers of pure GaAs or AlAs and away from the GaAs/AlAs interfaces may be less prone to DX-center formation. Hall effect measurements showed that activation increased by 2.6x over the control, while the RMS roughness decreased to 1.0 nm; the position of satellite peaks in 004 $\omega/2\theta$ x-ray scans agreed with the 10 ML periodicity. Encouraged by the improvement in surface morphology and activation, we next grew 3/2 DAs at 500°C and 610°C, attaining 14x and 34x improvements in activation, respectively, while maintaining roughness similar to the 6/4 DA; the peak n_0 achieved in this work of $2.5 \times 10^{17} \text{ cm}^{-3}$ is sufficient for use as the n-cladding layer in GaAs/AlGaAs laser diodes.⁴ All samples exhibited photoluminescence at the expected wavelength (~640 nm) and similar reflectance spectra to bulk n-Al_{0.4}Ga_{0.6}As:Si, showing that the DAs mimic the optical properties of random alloys. In future, we will further explore the effects of periodicity and composition on n_0 to elucidate the mechanism for n-doping enhancement in DAs.

ST-MoP-4 Metal Contact Etch Process Optimization of Metal Contact Etching in 3D Integration Devices, Sung Gyu Pyo, CAU, Republic of Korea

In this paper, we would like to report the metal contact etch, which is different from the existing device contact process, on the film stack side and the supercontact etching characteristics accordingly. General metal contact etch is organically related to physical profile and electrical properties, so evaluating only one item does not have much meaning, but 3D Integration. The physical profile characteristics of metal contact etch and 3D Integrated supercontact were examined. As a result of the 1st step etch evaluation, the etch target in the wafer left area was approximately 2365Å, and the bottom surface was found to be good with a bottom rounded profile.

After the 1st step etch for liner TEOS and nitride removal, the stopping margin was evaluated using 1) metal contact etch conditions where the target was reduced by about 22 sec, 2) CMOS image sensor metal contact etch baseline conditions to which an ILD reduction scheme was

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applied to improve optical characteristics, and 3) the selectivity was improved by increasing the C5F8/O2 ratio and the etch target was reduced. As a result, all three conditions were punch-through of BLC nitride has occurred.

In the 1st and 2nd steps, after proceeding with etch to the appropriate target, in the 3rd step, a good stopping margin was secured as a result of evaluating the 3-step etch recipe that over-etched using high selectivity.

It was confirmed that the stopping margin according to the over etch target split and process window change was also good, and the CD bias also secured good results.

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Room Ballroom A - Session ST-WeM1

Advancement in MBE Growth Approaches

Moderator: Dr. Darrell Schlom, Cornell University

8:00am **ST-WeM1-1 Thermal Laser Epitaxy - the Universal Epitaxy Tool(?)**, **Wolfgang Braun**, D. Kim, F. Hensling, T. Smart, L. Majer, B. Faeth, S. Smink, D. Dereh, Max Planck Institute for Solid State Research, Germany; H. Boschker, Epiray GmbH, Germany; J. Mannhart, Max Planck Institut for Solid State Research, Germany

Thermal laser epitaxy (TLE) uses quasi-CW lasers to heat the substrate and the sources in a deposition geometry similar to MBE. The high energy densities of the laser beams enable the evaporation or sublimation of any element in the periodic table in any combination and substrate temperatures exceeding the melting point of sapphire at 2040 °C. This leads to a dramatic expansion of the parameter space available to MBE-type growth modes such as, e.g., the adsorption-controlled growth of oxides.

In addition, TLE offers new opportunities in terms of the available gas pressures and environments during growth. A TLE growth chamber is conceptually simple, besides the laser entrance windows it basically contains only mechanical mounts for substrates and sources. This means that background gases at any pressure and with any reactivity are possible, as long as these parts do not get damaged, and the laser beam is not absorbed in the gas phase. This is the case for most reactive gases such as oxygen, ozone, plasma-excited nitrogen or ammonia.

As an example, we study the evaporation of Al in a molecular oxygen atmosphere. At high laser power densities, the flux of the source is independent of the oxygen pressure up to 10^{-1} hPa. At lower power densities, the source flux depends on the oxygen pressure and the area of exposed metallic surface on the source. This is consistent with volatile suboxide molecules proportional to the oxygen flux being generated on the exposed surface. For the current maximum laser power, the source finally gets passivated by a stable oxide at pressures above 10^{-1} hPa, and no longer produces a stable flux. The deposition rate on the substrate starts dropping at 10^{-2} hPa for the given working distance of 80 mm, as the mean free path drops below this distance and the transport transitions from the ballistic to the diffusive regime. Irrespective of the reactive flux generation and deposition, the more than five orders of magnitude dynamical range of the deposition flux and high maximum growth rates are maintained.

This implies that TLE is able to cover the entire range of growth conditions traditionally employed in MBE, GS-MBE, MOMBE, MOCVD, CBE and CVD. TLE thereby enables the evaporation or sublimation of practically any combination of elements, together with reactive gas sources, at low and also very high growth rates. CVD growth modes, e.g. for thick buffer layers, may be combined with kinetically limited growth, e.g. for active layers at low temperatures, in a single growth to produce devices with very dissimilar layers and growth conditions in a single growth run.

Results of TLE with nitrogen and ammonia will be presented and discussed.

8:15am **ST-WeM1-2 Routes Towards Making BaZrSe₃ Thin Films in the Perovskite Structure by MBE**, **Ida Sadeghi**, K. Ye, J. Van Sambeek, R. Jaramillo, MIT

Chalcogenide (sulfide and selenide) perovskite semiconductors are anticipated to have favorable structural, optical and electronic characteristics for solar energy conversion. The most studied compound is BaZrS₃, with a band gap of 1.9 eV. Alloying on the anion or cation sites has been explored to lower the band gap into a range suitable for single-junction solar cells. The pure selenide perovskite BaZrSe₃ is predicted to have band gap 0.5 eV lower than the sulfide.¹ However, BaZrSe₃ may form in different polymorphs, theory predicts that the needle-like (non-perovskite) phase with band gap below 1 eV is the most stable, and solid-state synthesis attempts have resulted in a semi-metallic hexagonal ordered defect phase.^{1,2,3,4}

We previously reported the first epitaxial synthesis of chalcogenide perovskite thin films by gas-source MBE: BaZrS₃ film on LaAlO₃ substrate.⁵ The films grow on a self-assembled interface layer that relieves the epitaxial strain. Here we demonstrate alloying BaZrS₃ with Se, including the first report of a pure selenide perovskite, and we confirm the tunability of the direct band gap of this alloy system.⁶ We demonstrate two processing routes to make BaZrS_(3-y)Se_y alloys in the perovskite structure. (i) We deposit BaZrS_(3-y)Se_y on a BaZrS₃ template layer on LaAlO₃. The template stabilizes

the perovskite structure for BaZrS_(3-y)Se_y, which otherwise does not grow as a perovskite directly on LaAlO₃. Scanning transmission electron microscopy analysis shows that there is anion intermixing between the sulfide template and the alloy during growth, such that the final film has nearly uniform composition. (ii) We deposit BaZrS₃ on LaAlO₃, followed by post-growth selenization. The RHEED pattern during selenization remains unchanged, indicating that the crystal structure is static even as the sulfur and selenium anions exchange. Both processing routes can produce selenide perovskite films with band gap of 1.4 eV.

This work sets the stage for developing chalcogenide perovskites as a family of semiconductor alloys with properties that can be tuned with composition in high-quality epitaxial thin films, as has been long-established for other semiconductor materials. The band gap of high-selenium-content BaZrS_(3-y)Se_y suggests applications in thin-film solar cells.

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8:30am **ST-WeM1-3 Adsorption Controlled Homoepitaxial Growth of c-Plane Sapphire by Thermal Laser Epitaxy**, **Felix Hensling**, L. Majer, S. Smink, J. Mannhart, W. Braun, Max Planck Institute for Solid State Research, Germany

The interest in sapphire as an electronic material is due to its low cost, superior properties over silicon, high quality wafer availability, and the possible integration with silicon. Indeed, its rapidly growing market share suggests sapphire to be the substrate material of the future.[1] A cornerstone for further establishing sapphire in a wide range of (electronic) applications is the ability to grow high quality homoepitaxial sapphire films. Key applications that can benefit from this are, e.g. diodes and high-power electronics based on nitrides [1] and (ultra) wide band gap semiconductors, respectively [2,3]. However, epitaxial films of c-plane oriented sapphire, one of the most common cuts of sapphire, have so far been out of reach due to a preferred formation of the Al₂O₃-phase.[3]

I will present how thermal laser epitaxy (TLE) overcomes this issue thanks to the availability of a parameter space that far exceeds the reach of other deposition methods.[4] The substrate laser heating system allows to easily heat sapphire substrates even beyond their melting point. The high accessible temperatures enable a precise and smooth sapphire substrate preparation – the first step for successful homoepitaxy.[5] I further present how the crystal quality and surface smoothness of homoepitaxial sapphire increase with increasing substrate temperature in case the growth is performed in the adsorption-controlled mode. Even at the respective high temperatures growth rates far exceeding 1 μm/h can be realized. Films were investigated by scanning transmission electron microscopy, atomic force microscopy, and x-ray diffraction. At a growth temperature of 1600 °C, the films were found to be practically undistinguishable from the underlying substrate.

References

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- [2] H. Okumura, "Sn and Si doping of α-Al₂O₃ (10-10) layers grown by plasma-assisted molecular beam epitaxy," *JJAP*, 61, 125505, 2022.
- [3] R. Jinno *et al.*, "Crystal orientation dictated epitaxy of ultrawide-bandgap 5.4- to 8.6-eV α-(AlGa)₂O₃ on m-plane sapphire", *Sci. Adv.*, 7, eabd5891, 2021
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8:45am **ST-WeM1-4 Prediction of MBE-grown Oxide Film Composition Using Neural Networks and Big Data Analytics**, **Patrick Gemperline**, R. Paudel, S. Thapa, S. Provence, S. Battles, R. Markland, Auburn University; R. Vasudevan, Oak Ridge National Laboratory; R. Comes, Auburn University
Reflected high energy electron diffraction (RHEED) is a highly common form of real time analysis used in growth systems such as molecular beam

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epitaxy (MBE) and pulsed laser deposition (PLD). Traditional RHEED analysis focuses only on the intensity and shape of the diffraction pattern for a few still images taken during growth or on the intensity of a single diffraction peak in real time. While this information can be quite insightful, there is far more information that can be gleaned from RHEED, which is often qualitative and learned through many repeated trials by the film grower. In order to obtain greater insight from RHEED videos, component analysis (PCA) and k-means clustering were applied to the recordings of the RHEED taken during the MBE growth of epitaxial thin film perovskite oxides, including SrTiO₃, LaFeO₃ and SrHfO₃. To further enhance the utilization of RHEED, a generative adversarial neural network was trained to predict X-ray photoelectron spectroscopy data from RHEED images, thus allowing for real-time prediction of film stoichiometry. These methods yield more quantitative results from the RHEED with minimal time requirements and open the door for future development of real-time computer control of film growth for optimal growth conditions.

9:00am ST-WeM1-5 Autonomous Synthesis in the MBE Using Real-Time Artificial Intelligence, Tiffany Kaspar, L. Wang, J. Christudasjustus, M. Sassi, B. Helfrecht, J. Pope, A. Harilal, S. Akers, S. Spurgeon, Pacific Northwest National Laboratory

Materials are the key components of nearly all advanced technologies, including quantum information systems, microelectronics, catalysis, and energy conversion and storage. Modern synthesis methods enable the fabrication of an ever-expanding array of novel, non-equilibrium, and/or metastable materials and composites that may possess unique and desirable functionality. Thin film deposition by molecular beam epitaxy (MBE) can produce atomically precise (or nearly so) materials with a wide range of functional electronic, magnetic, ferroelectric/multiferroic, optical, and/or ion-conducting properties. The current state of the art in precision design of functional materials is to manually explore the "growth phase space" of the deposition technique to optimize the film properties of interest. Limitations of time and resources often result in incomplete exploration of the growth phase space and resulting properties. Faced with this lack of complete information, materials design and synthesis decisions are made based in part on intuition and luck, slowing both materials optimization and materials discovery. This current synthesis paradigm can be disrupted by employing artificial intelligence (AI)-accelerated analysis of in situ and ex situ data streams that will enable targeted synthesis of novel materials with desired structure, chemical stability, and functional properties. Here we present a preliminary implementation of such an AI-controlled MBE. We are integrating the control of key synthesis parameters (temperatures, gas flow rates, shutters) with AI-guided computer control. Guidance will be based on near-real-time analysis of reflection high energy electron diffraction (RHEED) patterns using sparse data analytics, with low-latency feedback to the control software. As an initial demonstration, we will control the morphology and phase purity of epitaxial anatase TiO₂ thin films.

9:15am ST-WeM1-6 The Role of Optical Excitation on Misfit Dislocations in Epitaxial ZnS on GaP, Alexandra Fonseca Montenegro, M. Baan, A. Blackston, R. Myers, T. Grassman, The Ohio State University

Light decreases dislocation mobility in bulk ZnS, as was recently shown through photo-indentation measurements. Here we investigate the impact of above band gap optical excitation on ZnS epilayers grown by molecular beam epitaxy (MBE) on (001) GaP. For example, optical excitation during MBE could potentially increase the critical thickness for misfit dislocation (MD) nucleation at the ZnS/GaP heterovalent interface. GaP (001) homoepitaxial buffer layers are prepared on GaP wafers, As-capped, and transferred to the chalcogenide system where ZnS is grown by compound source MBE at 150C on the As-desorbed GaP surface. Using electron channeling contrast imaging (ECCI), the MD ensembles are measured at various film thicknesses to determine the onset of MD formation. Although HRXRD shows negligible strain-relaxation in films up to 50nm, ECCI reveals that the MD nucleation processes begins at ZnS thicknesses of between 15 and 20nm, far lower than previously reported critical thicknesses. Additionally, high densities of dipole-like features appear at large densities, which appear to be dislocation loops nucleated at the surface. Image analysis is used to quantify the MD density, length, as well as the density of dipole features as a function of film thickness. We will discuss changes in the MD content with and without above band gap excitation. Additionally, post-growth strain biasing is used to increase the MD content taking advantage of the thermal mismatch via temperature cycling. Photoluminescence spectra are acquired revealing the emergence of a sub-band gap peak at 3.1 eV, which increases in intensity within ZnS epilayers upon repeated temperature cycles. We will discuss changes in the MD

content and PL spectra for samples thermally cycled with and without optical excitation.

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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. Grossklauss, 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 SiO₂ 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 formation²⁻⁴. 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 photonics⁵⁻⁷ and aspect ratio trapping of dislocations for metamorphic growth⁸, are more challenging to access due to increased surface roughness of micron-scale features⁷. Increased roughness and defects on the mask surface lowers the barrier for nucleation further restricting the selective growth regime⁹.

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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 SiO₂ 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 SiO₂ deposited by plasma-enhanced chemical vapor deposition. Experiments are underway to utilize the technique to achieve selective growth of high aspect ratio microstructures.

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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 (T_{sub}) 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 > 10⁷ 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 1] and [0 -1 1].

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 × 10⁶ 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 (MnSb₂Te₄)_x(Sb₂Te₃)_{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 (MnSb₂Te₄)_x(Sb₂Te₃)_{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)₂Te₃ alloy formation. Hall effect measurements show that GR does not significantly affect the electrical doping in (MnSb₂Te₄)_x(Sb₂Te₃)_{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.

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

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

³I. 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

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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 SrTiO₃ by Oxide MBE**, *Dylan Sotir, M. Barone, D. Schlom*, Cornell University

Historically, the growth of stoichiometric SrTiO₃ 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 SrTiO₃ was self-regulating. One demonstrated way to achieve such automatic control of stoichiometry in SrTiO₃ is by supplying an excess of the volatile organometallic precursor titanium isopropoxide to grow SrTiO₃ by metalorganic MBE (MOMBE). Another means, and the one investigated here, is to stick with elemental sources but increase the substrate temperature. When SrTiO₃ 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 SrTiO₃ 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 SrTiO₃ 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 Ba₂BiTaO₆, 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 In₂O₃. 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 Sn²⁺, Pb²⁺, or Bi³⁺ 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 Ba₂BiTaO₆, 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 Ba₂BiTaO₆ is an example, offer additional flexibility and another degree of freedom to explore in the interplay of structure and properties. For Ba₂BiTaO₆, we take advantage of suboxide MBE and avoid using an electron beam to evaporate Ta by supplying a molecular beam of TaO₂ from a Ta₂O₅ 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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