NAMBE
Room Tamaya ABC - Session NAMBE2-WeM
Advanced MBE Techniques

Moderator: Stephanie Law, Penn State University

10:30am **NAMBE2-WeM-11 Artificial Intelligence for on-the-Fly Analysis and Control During Oxide Molecular Beam Epitaxy**, ***Tiffany Kaspar***, *Emily Saldanha, Henry Sprueill, Jenna Pope, Sarah Akers, Derek Hopkins, Ethan King,* Pacific Northwest National Laboratory

Thin film deposition is a fundamental technology for the discovery, optimization, and manufacturing of functional materials. Deposition by molecular beam epitaxy (MBE) typically employs reflection high energy electron diffraction (RHEED) as a real-time in situ probe of the growing film. However, the state of the art for RHEED analysis during deposition requires meticulous human observation and often fails to avoid negative film outcomes. We are working to employ artificial intelligence (AI)-accelerated analysis of in situ data streams for on-the-fly feedback control of the MBE deposition process that will enable successful synthesis of novel materials with desired structure and functional properties.This in situ feedback control is a critical component of autononmous experimentation (AE), which promises orders-of-magnitude acceleration of materials discovery, optimization, and adoption in advanced technologies.Here we present a machine-learning-enabled framework for the analysis of RHEED pattern images in real time (one image per second).We demonstrate this framework using RHEED images collected from the deposition of epitaxial oxide thin films such as anatase TiO_2 on $SrTiO_3$(001).On-the-fly feedback control of the deposition process is shown to improve film outcomes.Both recipe-based interventions and sophisticated AI-enhanced real-time parameter control will be discussed.

10:45am **NAMBE2-WeM-12 A Novel Approach for P-Type Doping in Semiconductor Heterostructures: Interface Fermi-Level Position Engineering**, ***Xiaoyang Liu***, *Xin Qi, Zheng Ju, Nathan Rosenblatt, Razine Hossain, Yong-Hang Zhang,* Arizona State University

Many semiconductors and oxides are difficult to dope p-type. This can be understood by using the hydrogen model, which gives the dopant ionization energy as a function of effective mass and dielectric constant.In most semiconductors, an increase in bandgap leads to a larger effective mass and a smaller dielectric constant. Therefore, many materials like some II-VI and wide-bandgap III-V semiconductors and oxides exhibit high dopant binding energies, limiting the achievable doping concentration. For example, at 300K, CdTe and GaN have high acceptor binding energies of ~73 meV and ~250 meV, respectively. As a result, the hole concentration remains below 8×10^{16} and 4×10^{15} cm^{-3}, respectively, even when the p-type atomic doping concentration reaches 10^{19} cm^{-3}, due to very low acceptor activation.

Here we demonstrate a novel approach to circumvent this fundamental doping limitation by utilizingthe interface statesofITO ona lattice-mismatched heterostructure to achieve “p-type” doping without the use of any dopants. These interfacial states spread across theforbidden gap and have a substantially lower density than statesin the bulk,allowing easier manipulation of the interface Fermi level. We refer to this method as *interface Fermi-level position engineering*.

A model for this approach is developed based on the following assumptions: 1.the interface states arise from dangling bonds at the lattice-mismatched interface; 2.these states are uniformly distributed inthe bandgap; 3. Anderson’s rule determines the initial band offset; and 4.equilibrium is reached as electrons from ITO and the n-typelayer fill the interface states. According to the model, varying the Mg content in the MgCdTe barrier layer modifies the lattice mismatch and the interface density of states so that the Fermi level position at the interface can be tailored.

A series of n-type CdTe/MgCdTe double-heterostructures (DH) with different Mg compositions in the top MgCdTe barrier layers were grown and “p-n junctions” wereformed by simply depositing an n-type ITO layer onto the top MgCdTe barrier layers. The ITO/MgCdTe interface then acts as an effective “p-region” induced by charge transfer. *C-V* measurementsof the samples with different Mg compositionsmatch the theoretical predictions.The sample with 40% Mg shows a V_{bi} over1 V, muchhigher than that (0.2 V) determined by the work function difference between bulk ITO (~4.6 eV) and n-type CdTe (~4.4 eV), confirming the critical role of the interface states in modifying V_{bi}.This method provides a new approach to overcoming p-type doping limitations in wide-bandgap semiconductors and oxides.

11:00am **NAMBE2-WeM-13 GaP Planar Coalescence Over Embedded Dielectric Gratings by Molecular Beam Epitaxy**, ***Ashlee Garcia***, *Will Doyle,* University of Texas at Austin*; Yiteng Wang, Corey White,* University of Illinois at Urbana-Champaign*; Byron Aguilar,* University of Texas at Austin*; Minjoo Lee,* University of Illinois at Urbana-Champaign*; Daniel Wasserman, Seth Bank,* University of Texas at Austin

Selective area growth (SAG) of III-V semiconductors by molecular beam epitaxy (MBE) enables the seamless integration of dielectrics, metals and high-quality crystalline semiconductors, which presents exciting pathways for the advancement of GaP-based applications such as nonlinear integrated photonics,[1] interfacing with cold atoms,[2] metasurfaces,[3] high-contrast photonics,[4] and site-controlled quantum-confined structures.[5-7]

While GaP has long been a valuable material in photonics, there has been a recent growing interest in realizing it as a nanophotonic platform due to its high transparency, high refractive index, large $\chi^{(2)}$ nonlinearity and lattice constant near that of silicon.[1] However, etch-induced roughness and growth quality due to substrate requirements currently limit the performance of GaP nanophotonics.[8] Leveraging all-MBE regrowth of GaP could enable deterministic growth of quantum-confined structures and patterned thin films boasting epitaxially smooth surfaces,[5-8] and realize advanced photonic design capabilities via the incorporation of amorphous materials into single-crystalline GaP. Here we demonstrate all-MBE regrowth of (100) GaP over SiO_2 gratings using a two-stage approach pioneered by Ironside,[9] which combines periodic supply epitaxy (PSE) for lateral epitaxial overgrowth with planar coalescence.

Selective GaP PSE was performed over SiO_2 gratings, aligned to [0-11], [010] and [011], on a GaP wafer. Enhanced lateral growth was observed for [010]-aligned gratings forming {101} facets. By then continuously growing at 0.35 μm/hr, restoration of the planar (100) surface was achieved above [010]- and [011]-aligned gratings. Atomic force microscopy showed high quality restored surfaces with root-mean-square (RMS) roughness values ranging from 0.9 to 3 nm, lower than that of optimized GaAs regrowth[5]. Cross-sectional scanning electron microscopy showed that, like GaAs, the [010]-aligned gratings were completely encapsulated by GaP, whereas, gratings along [011], saw the formation of high aspect ratio voids due to limited {0-11} growth, demonstrating a wide range of embedded GaP geometries achievable under a restored planar surface. This work was supported by NASA via the Quantum Pathways Institute (Award 80NSSC22K0287).

References: [1] D. J. Wilson et al. Nat. Photon, 2020. [2] A. González-Tudela et al. Nat. Photon, 2015. [3] M. Melli et al. Sci. Rep, 2020. [4] C. J. Chang-Hasnain et al. Adv. Opt. Photon, 2012. [5] P. Aseev et al. Nano Lett, 2019. [6] S. Birudavolu et al. Appl. Phys. Lett, 2004. [7] T. Schumann et al. Nanotechnol, 2011. [8] V. Fedorov et al. ACS Appl. Nano Mater., 2022. [9] D. J. Ironside et al. Crys. Growth Des, 2019.

11:15am **NAMBE2-WeM-14 High-Efficiency Entangled Photon Sources Using (111)-Oriented Quantum Optical Metasurfaces**, ***Trevor Blaikie***, University of Waterloo, Canada*; Simon Stich,* Walter Schottky Institut, Technische Universität München, Germany*; Vitaliy Sultanov,* Max-Planck Institute for the Science of Light, Germany*; Maria Chekhova,* Max Planck Institute for the Science of Light, Germany*; Mikhail Belkin,* Walter Schottky Institut, Technische Universität München, Germany*; Zbig Wasilewski,* University of Waterloo, Canada

High-efficiency, sub-wavelength-thickness sources of entangled photons for quantum optical applications enable the miniaturization of photonic quantum processing units and advancements in nanoscale photonic quantum state engineering, with promising applications in quantum imaging, computing, sensing, metrology, and communication.

Spontaneous parametric downconversion (SPDC), a second-order nonlinear optical process, is widely used for generating entangled photon pairs. The SPDC conversion rate increases with the second-order electrical susceptibility ($\chi^{(2)}$) of the nonlinear medium.

GaAs has one of the highest $\chi^{(2)}$ values (400–500 pm/V) among traditional materials, but its use is limited by SPDC momentum conservation requirements. These constraints relax at sub-wavelength thicknesses of the medium, allowing SPDC over a broader wavelength range. However, GaAs’s $\chi^{(2)}$ tensor symmetry requires electric field components in all three crystal basis dimensions for SPDC to occur. With the pump beam incident normally on a (001) surface, achieving the necessary vertical electric field component is challenging, leading to low conversion efficiency.

To address this, we switch the optical axis to the [111] direction, where photons traveling normally to the surface naturally have electric field components in all three crystal basis dimensions. This approach enables SPDC without the need for additional layer patterning.

Towards this goal, we have grown GaAs/AlGaAs layers on GaAs (111)B substrates. Initial measurements show that unpatterned AlGaAs (111)B layers generate photon pairs at rates two orders of magnitude higher than thin films of GaP, a commonly used nonlinear material. Additional patterning of these layers enabled still further efficiency improvement.

Epitaxial growth on GaAs (111) substrates has been largely abandoned since the late 90s due to significant difficulties in growing defect-free homoepitaxial and heteroepitaxial layers with smooth surfaces and interfaces. We will discuss the latest SPDC results and present the strategies developed to optimize the growth of smooth GaAs/AlGaAs layers on GaAs (111)B substrates.

11:30am **NAMBE2-WeM-15 Crystalline Direction and Shadowing Effect on Overgrowth of Patterned Features on GaAs (001)**, ***Xizheng Fang***, *Yiteng Wang, Adrian Birge, Minjoo Lee,* University of Illinois Urbana-Champaign

Epitaxial regrowth on patterned substrates has been widely explored for photonics applications[1], including embedded voids for photonic crystal surface emitting lasers[2] and Bragg mirror on patterned lasers[3]. Regrowth on etched heterostructures also enables the formation of quantum-confined wires with novel charge transport properties[4]. Regrowth morphology depends on many factors, including mass redistribution during deoxidation, shadowing, adatom diffusion, and different incorporation rates on different facets. In this work, we show that growth on etched trenches can be switched from planar to faceted based on trench orientation and that growing without substrate rotation can lead to nonconformal growth and the formation of lateral heterostructures.

The starting materials in this study were either epi-ready GaAs or 125 nm GaAs/125 nm AlAs templates, both with (001) surface orientation. 3 μm-wide trenches were dry etched to a depth of 0.25-0.70 μm over a wide range of in-plane directions. GaAs/AlAs superlattices (SLs) with periods of 60-100 nm were grown at 0.5-1.0 μm/hr with V/III ratios of 15-30. Substrate temperatures (T_{sub}) of 400-610°C were used to analyze the effects of adatom diffusion on regrowth.

The morphology of GaAs/AlAs SLs grown on trenches depends strongly on their in-plane direction. On [010]-oriented trenches, SL growth is highly conformal and became increasingly planar with time, even at T_{sub}=400°C. In contrast, overgrowth on [110]-oriented trenches led to the formation of prominent {111}B and {113}B facets at the end of SL growth with decreased conformality and thickness uniformity; layers grown on the {111}B facet were ~40% as thick as those grown on the (001) surface.

We next investigated shadowing effects by performing SL growth over trenches without substrate rotation. Thermal oxide desorption at 650°C without rotation causes GaAs etching near sidewalls where the As_2 flux is shadowed; ~60% of the Ga evaporates, and the rest redeposits epitaxially against the opposite sidewall with line-of-sight to the As_2 cell. SL growth at 610°C without rotation on trenches etched into 125 nm GaAs/125 nm AlAs templates enables novel vertical+lateral heterostructures (e.g., nanosheets) where thin GaAs layers are surrounded by AlAs barriers on three sides. In summary, crystallographic direction and shadowing effects strongly influences the conformality of MBE regrowth over trenches, enabling the formation of lateral heterostructures with potential (opto)electronics applications.

[1] Koshiba, S., et. al., *JAP 76*, (1994).

[2] McKenzie et. al., *APL 118*, (2021).

[3] Gebretsadik, et. al., *APL 71*, (1997).

[4] Chinni, et. al., *IEEE J-EDS 5*, (2017).

11:45am **NAMBE2-WeM-16 Thermal Laser Epitaxy System for Synthesis of Thin Films Containing Refractory Elements**, ***David Catherall***, *Yifei Yan, Fin Donachie, Austin Minnich,* California Institute of Technology

Molecular beam epitaxy (MBE) has well-known challenges in producing flux from highly-refractory elements such as W owing to high temperatures ~3000 C required to achieve adequate vapor pressure. Thermal laser epitaxy (TLE) has been proposed as a new technique to overcome this challenge by relying on laser heating of elemental sources. Here, we introduce a custom TLE system at Caltech which employs laser substrate heating and fiber laser heating of source materials. Our system is comprised of a UHV chamber with 1070 nm fiber lasers for source material heating, 10.6 um CO_2 laser for substrate heating, various in-situ diagnostics, and connection with a transfer line into into a nitrogen glovebox. In this talk, we will describe the design and preliminary results from this system.

12:00pm **NAMBE2-WeM-17 Rolled-Up Metamaterials (RUMMS) for Infrared Imaging**, ***Gokul Nanda Gopakumar***, *Stephanie Law,* Pennsylvania State University

Subwavelength information about an object is carried by waves with large wavevectors. The diffraction limit is caused by the rapid evanescent decay of these large wavevector modes at the surface of a material. Hyperbolic materials allow light with large wave vectors to propagate within the material without decaying exponentially close to the surface. These materials have a negative real part of the permittivity tensor along at least one direction and a positive permittivity along at least one other direction, leading to an open isofrequency surface, in contrast to the closed isofrequency surface of normal materials. In a flat hyperbolic material, the sub-diffractional information will still exponentially decay once it leaves the hyperbolic medium. However, in a rolled-up hyperbolic material, the wavevector of the light decreases as it propagates radially, and the image is magnified, enabling propagation beyond the surface.

In this work, we present rolled up semiconductor-based infrared hyperbolic metamaterials. We fabricate these structures by using a strained bilayer that can be released from the substrate. The strained bilayer is grown using molecular beam epitaxy and comprises of a compressively strained $Ga_{1-x}In_xSb$ bottom layer and tensile strained GaSb top layer grown on top of an AlSb sacrificial layer. A layer of Si:InAs layer is grown on top of the bilayer because heavily doped III-V semiconductors can act as an optical metal in the IR. Fabrication of rectangular mesas is done using standard lithographic and wet etching techniques. Finally, a wet etch that selectively removes the sacrificial layer is used to gradually release the strained bilayer, causing it to roll up. By changing the alloy composition, we tune the stress in the bilayers to change the diameter of the rolled-up tube. The number of turns in the rolled-up tube can also be increased by increasing the etching time. The result is a RUMM that has alternating layers of dielectric ($Ga_{1-x}In_xSb$ and GaSb) and metal (Si:InAs) in the radial direction.

The growth of the strained bilayer and determination of the strain are evaluated using High resolution X-ray diffraction. Scanning electron microscopy is used to image the rolled-up tubes and correlate their diameter to the bilayer strain. Finally, infrared spectroscopy will be used to measure the optical properties of the RUMMs. This is the first step in creating a fully semiconductor-based curved hyperbolic metamaterial that can be used in subdiffractional imaging in the IR wavelength range.

12:15pm **NAMBE2-WeM-18 Spectroscopic Ellipsometry as an in Situ Technique to Control MBE Growth**, *Jackson Niedel, Owen Peterson, Hatim Saeed,* Kenyon College*; Qihua Zhang, Stephanie Law, Maria Hilse,* Penn State University*;* ***Frank Peiris***, Kenyon College

It is imperative to develop *in-situ* tools that will yield instant feedback in growing novel and challenging structures, such as topological insulators and transition metal dichalcogenides. Using several examples, we will show that by incorporating a spectroscopic ellipsometer into a molecular-beam epitaxy growth chamber, the growth parameters of binary and ternary films can be obtained layer-by-layer, during their entire growth cycle. Obtaining continuous ellipsometry spectra during the growth of a ~25 nm thick $PtSe_2$ film, we determined the thickness dependent dielectric functions of $PtSe_2$. In addition, we map how the sample composition (i.e., $PtSe_2$ and void) changes during the growth of the film. We have also explored $(Bi_xIn_{1-x})_2Se_3$ and have obtained the composition-dependent dielectric functions of this ternary system. By using the calibrated dielectric functions, the composition and thickness of any $(Bi_xIn_{1-x})_2Se_3$ film can be obtained immediately at any stage of the growth cycle. We tested the model for universality among MBE growth systems and found that the model was transferable between systems. Furthermore, the generalized model allowed us to monitor sticking and desorption coefficients as well as temperature-induced composition changes in the $(Bi_xIn_{1-x})_2Se_3$ thin films.

12:30pm **NAMBE2-WeM-19 Closing Remarks & Thank Yous**,

Author Index

Bold page numbers indicate presenter