## Novel Materials
## Room Swan BC - Session NM-TuA2

### Heterogenous Integration
**Moderator: Vladimir Vladimirovich Protasenko**, Cornell University

3:45pm **NM-TuA2-10 sub-Monolayer Surface Termination Control of Charge Transfer and Band Alignment Across a Semiconductor-Crystalline Oxide Heterojunction**, *M. Chrysler,* University of Texas-Arlington*; T. Lee, J. Gabel,* Diamond Light Source, UK*; Z. Zhu, P. Sushko, S. Chambers,* Pacific Northwest National Lab*; **Joseph Ngai,*** University of Texas-Arlington

Semiconducting heterojunctions exhibiting built-in electric fields that stem from charge transfer are the building-blocks for virtually all device technologies. While such heterojunctions are typically formed between covalent semiconductors, the epitaxial growth of SrTiO3 on Si(100) using MBE enables charge transfer and the formation of built-in fields to be studied in hybrid heterojunctions in which ionic and covalent properties are coupled. We find that the surface of SrTiO3 drives the transfer of electrons from Si to SrTiO3, and that electron transfer can be tuned by altering the composition of the terminating layer, as revealed by electrical transport and hard x-ray photoelectron spectroscopy measurements. The transferred electrons in turn create space charge across the interface which modifies the interfacial dipole that stems from epitaxial bonding. The modulation of the dipole leads to a change in band alignment from type-II to type-III, which promotes additional electron transfer. The surface driven transfer of charge across the interface is discussed within the context of surface depletion, surface adsorbates, as well as physical structure. The strong sensitivity of interfacial charge transfer to surface termination as well as the tunability of band alignment via the interfacial dipole, opens degrees of freedom by which functional behavior can be engineered in hybrid heterojunctions.

4:00pm **NM-TuA2-11 Heteroepitaxial Growth of (111)-oriented $Sr_{1-x}Ca_xTiO_3$ Thin Films on III-Nitride Semiconductors**, ***Eric Jin,*** *B. Downey, V. Gokhale, J. Roussos, M. Hardy, N. Nepal, D. Katzer, J. Calame, V. Wheeler, D. Meyer,* U.S. Naval Research Laboratory

Heterogeneous integration of epitaxial functional oxides with semiconductors has attracted significant interest in recent decades with the goal to couple the novel functionalities observed in these oxides—including high-temperature superconductivity, multiferroicity, tunable electronic phases, and high dielectric constants (κ)—with technologically-relevant semiconductor platforms. Advances in epitaxial growth techniques have enabled the monolithic integration of perovskite oxides with Si, Ge, GaAs, and more recently, GaN. Development of electronics based on wide and ultra-wide bandgap (UWBG) semiconductors such as AlN, AlGaN, and ScAlN is especially attractive for high frequency and power electronics applications, due to the larger breakdown electric field of these materials when compared to conventional semiconductors.

High quality epitaxial growth of perovskite oxides on wurtzite-phase III-nitrides is challenging due to both the crystal and chemical mismatches. For example, the lattice mismatch between $SrTiO_3$ (STO) and GaN is 13.3%, which leads to polycrystalline films when STO is directly deposited onto GaN. However, a thin rutile $TiO_2$ buffer layer can significantly reduce the lattice mismatch [1]. In this work, we demonstrate the epitaxial growth and characterization of (111)-oriented STO and $Sr_{1-x}Ca_xTiO_3$ (SCTO) thin films on AlGaN/GaN and ScAlN/GaN high-electron-mobility transistor (HEMT) heterostructures by RF-plasma-assisted oxide molecular beam epitaxy. A 1-nm-thick $TiO_2$ buffer layer is used to orient the SCTO film and greatly improves crystal quality [2].

As an application, we extract the dielectric constant of the oxide/III-nitride heterostructures with capacitance-voltage measurements and find a κ value as high as 290 and a fixed positive interface charge density of $2.38 \times 10^{13}$ $cm^{-2}$ at the SCTO/AlGaN interface [3]. RF characterization of interdigitated capacitors fabricated on the SCTO films show that the high κ values are maintained at 2 GHz. These results demonstrate the epitaxial integration of an “extreme κ” functional oxide with GaN that can potentially improve electric field management in RF HEMTs [4]. Moreover, the epitaxial connection between a perovskite and wurtzite crystal structure can pave the way for perovskite oxide-UWBG semiconductor integration to realize next-generation multifunctional oxide/nitride hybrid electronics.

This work is supported by the Office of Naval Research.

[1] W. Tian et al., *Appl. Phys. Lett.* (2007)
[2] E. N. Jin et al., *J. Appl. Phys.* (2020)
[3] E. N. Jin et al., *APL Mater.* (2021)
[4] N. K. Kalarickal et al., *IEEE Trans. Electron Devices* (2021)

4:15pm **NM-TuA2-12 MBE Growth and Electronic Properties of Epitaxial $SrNiO_3$-based Heterostructures**, ***Le Wang,*** *P. Sushko, S. Spurgeon, Y. Du, S. Chambers,* Pacific Northwest National Laboratory

The rich phase diagrams of nickelates $RNiO_3$ ($R$ denotes the lanthanide) are of critical importance for both material physics and oxide electronics. The recent discovery of superconductivity in infinite-layered $R_{1-x}Sr_xNiO_2$ further motivates the synthesis of other related nickelates in searching for high-$T_c$ materials and should provide new insights into the origin of high-temperature superconductivity. However, Sr substitution in $RNiO_3$ solid solutions has been shown to induce local defects and structural instabilities, presumably because bulk $SrNiO_3$ exhibits a hexagonal phase and is not stable. Thus, it is important to understand how to stabilize perovskite structured $SrNiO_3$ and how the local charges are distributed in the lattice.

One of our recent efforts to stabilize $SrNiO_3$ thin films on perovskite substrates by oxide molecular beam epitaxy (MBE) led to spontaneous phase segregation. Two co-existing oxygen-deficient Ruddlesden-Popper (RP) phases, $Sr_2NiO_3$ and $SrNi_2O_3$, are formed to balance the stoichiometry and stabilize the energetically preferred $Ni^{2+}$ cation.[1] We envision that this kind of spontaneous phase segregation and RP faults may occur during the synthesis of Sr doped $RNiO_3$ when the Sr doping level is high.On the other hand, by fabricating epitaxial $(SrNiO_3)_m/(ABO_3)_n$ superlattices, we show that normally hexagonal-structured $SrNiO_3$ can be stabilized as a perovskite at the single unit cell level ($m$ = 1).[2,3] Spectroscopy reveals that the Ni valence can exceed $Ni^{3+}$ and can be also tuned by controlling the magnitude of the $BO_6$ octahedral rotations. In-plane transport data are consistent with variable-range hole hopping conductivity for these superlattices, whereas hole hopping along the $c$-axis direction can be tuned by changing $n$. Our results offer exciting opportunities to generate novel ground-states unobtainable in bulk crystals and will inspire the epitaxial synthesis of $(SrNiO_3)_1/(RNiO_3)_n$ superlattices. Measurements of the associated properties may open new avenues for studying superconductivity in nickel-based oxides.

[1] L. Wang *et al.* Sci. Adv. 7, eabe2866 (2021).

[2] L. Wang *et al.* Adv. Mater. 32 (45), 2005003 (2020).

[3] L. Wang *et al.* Phys. Rev. Mater. submitted (2022)

4:30pm **NM-TuA2-13 Controlling Dislocation Formation and Dynamics in GaAs-Based Films on Silicon via Indium Alloying**, ***Eamonn Hughes,*** *M. Dumont, J. Selvidge, J. Norman,* University of California, Santa Barbara*; Y. Hu,* Hewlett-Packard Labs*; C. Shang, D. Jung, A. Taylor, M. Kennedy,* University of California, Santa Barbara*; R. Herrick,* Intel Corporation*; D. Liang, R. Beausoleil,* Hewlett-Packard Labs*; J. Bowers,* University of California, Santa Barbara*; K. Mukherjee,* Stanford University

Much dislocation research sensibly focuses on growth strategies to reduce dislocation densities and thereby improve device performance and reliability, but one often overlooked stage of growth is the cooldown phase. In many III-As films on silicon, this phase is especially important because dislocations continue to evolve due to (1) thermal stress generated by thermal expansion mismatch between film and substrate and (2) the lack of obstacles to glide particularly in (Al)GaAs. For a GaAs-based film grown on silicon at a typical molecular beam epitaxy temperature of 580 °C, cooling to room temperature leaves about 0.15% residual thermal tensile strain. We have recently shown that alloying such a film with just 5% indium can effectively halt dislocation glide under these low stress conditions. Here, we detail two cases where we leverage this indium alloy-hardening effect to mitigate certain undesirable dislocation formations.

In one application involving regrowing GaAs-based films on a thin GaAs template bonded to silicon for integrated photonics, we explore the potential for thin indium-alloyed layers to reduce threading dislocation (TD) densities, which nucleate due to thermal expansion mismatch. We find that the initial compressive stress is relaxed efficiently by a small density of dislocations nucleated at the bond interface, but during cooldown when the stress state reverses, the dislocation density rises sharply. We block the propagation of these latter dislocations by inserting strained indium-alloyed ‘trapping’ layers and reduce dislocation densities in the remainder of the film by 40×.

In a separate application, misfit dislocations (MDs) are seen to form during cooldown in InAs QD lasers directly above and below the active region due to TD pinning and thermal stress. These MDs cannot pass through the active region because, in addition to pinning, they experience a strong repulsive force due to the tensile-to-compressive stress reversal at the interface. To address this, we displace the network of MDs away from the active region by inserting a thin indium-alloyed trapping layer a short distance on either side of the active region to extend the region of TD pinning. By placing the trapping layers outside the depletion region, the MDs are starved of minority carriers, rendering them largely benign during device operation and yielding up to two orders of magnitude improvement in reliability.

These studies showcase a method to engineer around cooldown-generated dislocations rather than eliminate them entirely, particularly useful when doing so is exceedingly difficult or comes with unwanted side effects such as cracking.

4:45pm **NM-TuA2-14 Grafted Si/GaN, AlN/Si, and GaAs/GeSn PN Junctions with Epitaxy-Like Interface Qualities***, **Jie Zhou,*** University of Wisconsin - Madison*; P. Wang, D. Wang,* University of Michigan, Ann Arbor*; T. Ng,* King Abdullah University of Science and Technology, Saudi Arabia*; H. Wang, S. Xu,* National University of Singapore*; S. Ojo,* University of Arkansas*; Z. Mi,* University of Michigan, Ann Arbor*; B. Ooi,* King Abdullah University of Science and Technology, Saudi Arabia*; X. Gong,* National University of Singapore*; S. Yu,* University of Arkansas*; T. Grotjohn,* Michigan State University*; Z. Ma,* University of Wisconsin - Madison

Semiconductor heterojunction-based devices have revolutionized modern human society. Epitaxy growth employing MBE, MOCVD, MOVPE, etc. has been the only approach to form abrupt semiconductor heterojunctions. The essential requirement to form abrupt heterojunctions using the epitaxy techniques is lattice match (or a close match for pseudomorphic growth) between two different semiconductors. Lattice-mismatched heterojunctions can open much broader applications than the lattice-matched ones, however, cannot be realized easily with heteroepitaxy growth. Metamorphically grown interface of such heterojunctions retains a large quantity of density of interface states (Dit). While wafer bonding/fusion techniques have offered limited applications in tunnel junction devices, they cannot be generally used to fabricate abrupt interface with low defect densities [1][2].

Here, we present a novel semiconductor grafting technology as a strategy to overcome the lattice-mismatched constraints faced by epitaxy growth communities. By grafting single-crystalline epitaxially grown semiconductors with a quantum-tunneling gluing layer in between any two semiconductors, high-quality semiconductor heterojunctions, exhibiting significantly suppressed Dit ($10^{10}$-$10^{11}$ /cm$^3$.eV) at their interfaces, have been formed. The interface quality reflected from the diode ideality factor rivals the lattice-matched epitaxy interfaces [3]. Specifically, we will report Si/GaN, AlN/Si, and GaAs/GeSn PN junctions formed via the grafting approach. The approach can be applied to any semiconductor heterojunctions that have arbitrary lattice structures and lattice constants.

[1] M. N. Hasan et al (2022). Influences of Native Oxide on the Properties of Ultrathin Al2O3-Interfaced Si/GaAs Heterojunctions. Advanced Materials Interfaces, 2101531.

[2] Y. Ohno et al (2020). Chemical bonding at room temperature via surface activation to fabricate low-resistance GaAs/Si heterointerfaces. Applied Surface Science, 525, 146610.

[3] D. Liu, et al (2018). Lattice-mismatched semiconductor heterostructures. arXiv preprint arXiv:1812.10225.

5:00pm **NM-TuA2-15 Integrating GaSb-Based Infrared Detectors with Si Substrates via Interfacial Misfit Arrays***, **Trent Garrett**, M. Drake* , Boise State University*; P. Reddy,* Stanford University*; K. Mukherjee,* Stanford University*; K. Grossklaus,* Tufts University*; S. Maimon,* Netz Vision*; P. Simmonds,* Boise State University

With applications from night vision and aerial target acquisition, to space telescope operation, infrared (IR) detectors are of great interest to the defense and scientific communities alike. The functionality of these detectors hinges on achieving a high signal-to-noise ratio so that weak signals can still be resolved. Of the many IR detector designs, the nBn device has emerged as a leading choice. As the name suggests, nBn detectors comprise an electron-blocking barrier between *n*-type absorber and contact layers [1].

nBn-based IR detectors are typically grown on GaSb substrates, which represents perhaps their biggest disadvantage. GaSb substrates are expensive and only widely available up to 4" diameter, placing a limit on high volume manufacturing. Successfully integrating III-V based nBn detectors with Si substrates would overcome these wafer cost and size constraints. However, this approach comes with its own set of challenges, primarily due to the large lattice mismatch between GaSb and Si. GaSb deposited directly onto Si typically forms large 3D islands with a high density of threading dislocations that help relieve the strain [2].

We therefore adopt the use of interfacial misfit (IMF) arrays grown by molecular beam epitaxy (MBE) to manage strain at the III-Sb/Si heterointerface. IMFs consist of the spontaneous formation of a 2D array of 90º dislocations that lie in the plane of the heterointerface. Previous studies show that thin initiation AlSb layers between the GaSb and Si are critical. Compared with GaSb grown directly on Si, GaSb deposited onto an AlSb/Si IMF heterostructure has dramatically improved material quality and lower threading dislocation density (TDD) [3]. To ensure the growth of GaSb-on-Si virtual substrates with the highest possible quality for subsequent nBn growth, perfecting the growth of this AlSb/Si IMF layer is fundamentally important.

We will discuss how choices regarding AlSb growth initiation, substrate temperature, annealing, AlSb thickness, and AlSb growth rate affect the quality of GaSb overlayers. By optimizing these MBE growth parameters, initial results suggest that we can grow GaSb layers with quality comparable to the current state-of-the-art, giving us a benchmark against which to measure further improvements. We will also discuss the use of dislocation filtering superlattices above the IMF to further reduce TDD.

This work is supported by the Office of Naval Research through grant ##N00014-21-1-2445.

[1] S. Maimon and G.W. Wicks, Applied Physics Letters 89, (2006).

[2] S.H. Vajargah, S. Ghanad-Tavakoli, J.S. Preston, R.N. Kleiman, and G.A. Botton, Journal of Applied Physics 114, 113101 (2013).

[3] K. Akahane, N. Yamamoto, S.-ichiro Gozu, and N. Ohtani, Journal of Crystal Growth 264, 21 (2004).

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5:15pm **NM-TuA2-16 Epitaxial Growth of Highly Mismatched Antimonide-Based Alloys Using Imf and Defect Filter Layers***, **Fatih Ince**, T. Rotter, A. Mansoori,* University of New Mexico*; S. Addamane,* Sandia National Laboratories*; D. Shima, G. Balakrishnan,* University of New Mexico

Antimonide-based alloys have the capability to detect medium and long-wavelength infrared ranges of the EM spectrum. However, realizing fully relaxed lattice constants between 6.09 Å and 6.47 Å remains a challenge due to the lack of substrates. This limitation can be overcome by using interfacial misfit dislocation (IMF) array growth mode and it has been shown that fully relaxed GaSb growth on GaAs with dislocation density of ~108 dislocations/cm2 is possible [1]. In this presentation, it will be shown that further reduction of the dislocation density to ~107 dislocations/cm2 is possible using AlSb defect filter layers that bend dislocations due to the strain at the AlSb/GaSb interface.

The IMF growth of GaSb on GaAs involves the exchange of the group V element. The same principle is applied to the growth of In0.53Ga0.47Sb on In0.53Ga0.47As in order to further extend the lattice constant to ~6.3 Å. The latter can be grown on an InP substrate, since it is lattice matched. TEM images show the formation of the interfacial misfit dislocation array at the interface similarly to GaSb on GaAs. Defect filter layers can be realized in this case by varying the composition of InGaSb or using InAlSb. However, XRD indicates low crystalline quality of In0.53Ga0.47Sb. This might be due to phase segregation in In0.53Ga0.47Sb.

To reduce the complexity associated with ternaries, the growth of InSb on InAs substrates is investigated. XRD results indicate that InSb can be grown fully relaxed on InAs as in the case of GaSb on GaAs. Studying the direct growth of InSb on InAs helps to identify parameters to improve the growth of the ternary In0.53Ga0.47Sb on In0.53Ga0.47As/InP. In this presentation, our current efforts to optimize the growth quality of InSb and InGaSb alloys as well as the use of antimonide-based defect filter layers to reduce dislocation density will be presented.

[1]S. Huang, G. Balakrishnan, and D. L. Huffaker, “Interfacial misfit array formation for GaSb growth on GaAs,” Journal of Applied Physics, vol. 105, no. 10. p. 103104, 2009. doi: [http://paperpile.com/b/GBqBar/qFoQ]10.1063/1.3129562. [http://dx.doi.org/10.1063/1.3129562.]

5:30pm **NM-TuA2-17 Controlling the Balance between Remote, Pinhole, and van der Waals Epitaxy of Heusler Films on Graphene/Sapphire**, *D. Du, S. Manzo, T. Jung, X. Zheng, M. Arnold,* ***Jason Kawasaki,*** University of Wisconsin - Madison

Remote epitaxy on monolayer graphene is promising for synthesis of highly lattice mismatched materials, exfoliation of free-standing membranes, and re-use of expensive substrates. However, clear experimental evidence of a remote mechanism remains elusive. In many cases, due to contaminants at the transferred graphene/substrate interface, alternative mechanisms such as pinhole-seeded lateral epitaxy or van der Waals epitaxy can also explain the resulting exfoliatable single-crystalline films. Here, using Heusler film growth on clean graphene on sapphire substrates, we observe a 30 degree rotated epitaxial superstructure that cannot be explained by pinhole or van der Waals epitaxy. With decreasing growth temperature the volume fraction of this 30 degree domain increases compared to the direct epitaxial 0 degree domain. We further show that careful graphene/substrate annealing (T ~ 700 C) and consideration of the film/substrate vs film/graphene lattice mismatch are required to obtain epitaxy to the underlying substrate. The 30 degree rotated superstructure provides the first experimental fingerprint of remote epitaxy that cannot be explained by the other mechanisms.

5:45pm **NM-TuA2-18 Improved-Quality of 3D Semiconductors at Low Temperature Using Intermediate 2D Materials**, ***Guanyu Zhou,*** *R. Younas, T. Sun, G. Harden, Y. Li, A. Hoffman, C. Hinkle,* University of Notre Dame

Low-temperature growth of materials with high-quality is of great importance as it will enable advanced technologies such as monolithic three-dimensional (3D) integration and flexible electronics. However, low growth temperature prohibits sufficient atomic diffusion and directly leads to poor growth quality, as described by the exponential decay of atomic diffusion $D$ with growth temperature $T$ in classic nucleation theory[1] $D \propto exp(-V_s/k_BT)$, where $V_s$ is the potential-energy barrier for adatom diffusion. To enhance atomic diffusion at low $T$, $V_s$ must be reduced. Here we show, using a thin layer of 2D materials to separate the adatom and the substrate spatially, $V_s$ is significantly reduced based on a Lennard-Jones potential diagram. Simultaneously, the 2D materials behave like a “transparent layer”, that permit the adatom-substrate interactions such that the 3D materials can retain epitaxial growth, which have been demonstrated by the novel growth method “remote epitaxy”.[2] The maintained, but reduced, potential field through the 2D layer, coupled with the inert surface of the 2D materials, enables long-distance atomic diffusion and enhanced growth quality at lower growth temperatures. As model systems, GaN and ZnSe using $WSe_2$ and graphene as intermediate layers, exhibit larger grains, preferred orientation, reduced strain, and improved carrier mobility, all at temperatures lower by >200 °C compared to direct growth as characterized by diffraction, X-ray photoelectron spectroscopy (XPS), Raman, and Hall measurements. Moreover, the impact of thickness and grain-size of the $WSe_2$ on the 3D materials' growth quality was also studied taking advantage of molecular beam epitaxy that can realize excellent uniformity and thickness control. We further demonstrate an improved method to obtain band alignment of the $ZnSe/WSe_2$ heterostructure using XPS, taking advantage of the easy exfoliation of the grown 2D/3D heterostructure, eliminating quantum confinement issues caused by needing to use thin overlayers to remain photoelectron transparent in conventional methods.

Our results reveal the benefits of a reduced potential field through atomically thin 2D layers, showing promise for reducing the growth temperature of 3D semiconductors and other materials as a solution to severe thermal budget constraints. Moreover, the 2D/3D heterostructures could also enable promising new heterostructures for novel device designs.

This work was supported in part by NEWLIMITS, a center in nCORE, an SRC program sponsored by NIST through award number 70NANB17H041.

1. Z. Zhang, *et al.* Science 276, 377 (1997).

2. Y. Kim, *et al.* Nature 544, 340 (2017).

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