## Wednesday Morning, September 21, 2022

**Novel Materials** 

**Room Swan BC - Session NM-WeM1** 

#### **Quantum-confined Structures**

Moderator: Joseph Ngai, University of Texas-Arlington

8:00am NM-WeM1-1 Ultra-Strong Light-Matter Coupling in the THz with Continuously Graded Al<sub>\*</sub>Ga<sub>1\*</sub>As Parabolic Quantum Wells, *Chris Deimert*, University of Waterloo (currently at National Research Council Canada), Canada; *P. Goulain, M. Jeannin*, CNRS, Université Paris-Saclay, France; *W. Pasek*, University of Waterloo (currently at University of Campinas), Canada; *A. Bousseksou, R. Colombelli, J. Manceau*, CNRS, Université Paris-Saclay, France; *Z. Wasilewski*, University of Waterloo, Canada

Conventional THz optoelectronic devices operate in the regime where the coupling between light and matter is weak. As an alternative, the strong coupling regime has attracted considerable study in recent years – one can envision the development of coherent light sources based on parametric oscillation and sources of non-classical states of light [1,2]. For such devices, the typical square quantum well struggles above cryogenic temperatures or at frequencies below 3 THz [3], largely due to electronelectron interactions in the well. Parabolic quantum wells (PQWs), on the other hand, are effectively immune to such interactions, providing a strong unified absorption line in situations where square wells cannot be employed [4].

PQWs can be grown with molecular beam epitaxy using digital alloys, but this technique only approximates the parabolic potential and it also generates many interfaces. We instead generate a smooth composition gradient, employing a linear dynamical model of our aluminum effusion cell to smoothly vary the flux at standard growth rates (up to 2.5 Å/s) [5].

Using this technique, we have grown arrays of PQWs in  $AI_xGa_{1-x}As$  with composition in the 2–30% range. THz multipass absorption measurements on samples with different transition frequencies and doping levels reveal clear absorption peaks between 2 and 3 THz. The linewidths observed in these samples are remarkably small for THz intersubband (ISB) transitions – at 2.2 THz, we observe a 69 GHz linewidth, which represents a record-small 3.2% fraction of the centre frequency.

We fabricate several of these samples into metal-insulator-metal microcavities, which are designed to be resonant with the ISB mode of the PQWs. Strong coupling (heralded by the appearance of Rabi splitting) is measured via angle-dependent reflectivity. In one sample, we observe a lower polariton branch centered at 1.8 THz, which is the lowest-frequency ISB polaritonic mode to date. Further, we observe ultrastrong coupling (defined as a Rabi splitting greater than 10% of the central frequency) up to 200 K. Further development of these PQW samples could be of great interest for strong-coupling studies and devices with reduced numbers of electrons or at smaller THz operating frequencies.

S. De Liberato and C. Ciuti, Phys Rev Lett **102**, 136403 (2009).
C. Ciuti, G. Bastard, and I. Carusotto, Phys Rev B **72**, 115303 (2005).
Y. Todorov *et al.*, Phys Rev Lett **105**, 196402 (2010).
C. Deimert *et al.*, Phys Rev Lett **125**, 097403 (2020).
C. Deimert and Z.R. Wasilewski, J Crys Growth, **514**, 103-108 (2019)

# 8:15am NM-WeM1-2 Molecular Beam Epitaxy and Characterization of Bi<sub>2</sub>Se<sub>3</sub> and Sb<sub>2</sub>Te<sub>3</sub> on In<sub>2</sub>Se<sub>3</sub> Layers via Selenium Passivation of InP(111)B Substrates, *Kaushini Wickramasinghe, C. Forrester, I. Levy, M. Tamargo,* City College of New York, City University of New York

Topological Insulators (TI) have attracted a great deal of interest in the past decade due to their non-trivial topology giving rise to metallic surface states protected by time reversal symmetry and an insulating bulk. A wide variety of applications in thermoelectrics, spintronics, twistronics, and quantum computation are being considered. Also, they provide a fundamental platform to explore novel physics. However, there is a large unintentional background doping that conceals their surface channels, and crystal defects such as twin domains are frequently observed. Although these are van der Waals materials, the influence of the substrate on the molecular beam epitaxy (MBE) grown material properties has been shown to be non-negligible. InP(111)B has been used before and reported by some to produce good material properties, although the reports vary and there are few details of the growth conditions. In practice, most materials are grown on sapphire substrates.

In this study, we propose and explore the quality of epitaxial Bi<sub>2</sub>Se<sub>3</sub>, Sb<sub>2</sub>Te<sub>3</sub> and Sb<sub>2</sub>Te<sub>3</sub> on Bi<sub>2</sub>Se<sub>3</sub> structures on In<sub>2</sub>Se<sub>3</sub> layers grown by a selenium passivation technique during the oxide desorption of the InP(111)B substrates. The crystallinity of the samples is measured using high-

resolution X-ray diffraction (HR-XRD). Surface morphology is explored using atomic force microscopy (AFM). Here we present the process of growth and optimization with detailed analysis of the quality of the samples.

First, the surface is prepared with a self-grown 5 nm layer of  $In_2Se_3$  which is formed during the oxide removal process in the presence of excess selenium. After that, the substrate is cooled down to the required growth temperature. No low temperature seed layer is grown. AFM shows the formation of a smooth  $In_2Se_3$  layer and the HR-XRD shows its high crystallinity, with the presence of satellite peaks from which the thickness could be extracted. Morphology of Bi<sub>2</sub>Se<sub>3</sub> samples grown at different substrate temperatures exhibits smooth surfaces with large terraces.  $\boldsymbol{\Phi}$ scans of the (015) plane of the Bi<sub>2</sub>Se<sub>3</sub> layers show complete suppression of twin domains. AFM of Sb<sub>2</sub>Te<sub>3</sub> layers grown on the In<sub>2</sub>Se<sub>3</sub> layer show planes with steps, with a larger roughness than  $Bi_2Se_3$ , yet the  $\Phi$  scan of the (015) plane of Sb<sub>2</sub>Te<sub>3</sub> also shows complete suppression of twin domains. The surface morphology of Sb2Te3 on Bi2Se3 structure shows three fold symmetry with varying domain sizes. Again, twin suppression is evident in this sample. Thus, our results show that growth of the materials on the In<sub>2</sub>Se<sub>3</sub> interfacial layer results in full suppression of the twin domains compared with samples grown on sapphire, GaAs, or vicinal InP substrates.

8:30am NM-WeM1-3 Structural and Optical Properties of GaNAs Highly Mismatched Alloys Multi-Quantum Well Heterostructures., *Rolando Pinson Ortega*, Universidad Autónoma de San Luis Potosí, Mexico; L. *Espinosa Vega*, E. *Espinoza Figueroa*, A. *Belio Manzano*, P. Olvera Enríquez , M. Villareal Faz, L. Hernandéz Gaytán, F. Perea Parrales, Universidad Autónoma de San Luis Potosí (UASLP), Mexico; C. Yee Rendón, Universidad Autónoma de Sinaloa, Mexico; I. *Cortes Mestizo*, CONACYT-Universidad Autónoma de San Luis Potosí (UASLP), Mexico; V. Méndez García, Universidad Autónoma de San Luis Potosí (UASLP), Mexico

In recent years, novel materials, and new designs of solar cells (SCs) have been proposed with the aim of contributing to the universal attempt to retrieve solar energy more efficiently. Up to now, the highest conversion efficiencies have been achieved with tandem or multi-junction SCs, a technology that comprise the simultaneous absorption of photons of different wavelengths. This mechanism allows to capture a wider region of the solar spectrum, as compared to simple pn junctions. An alternative approach to achieve the multi-wavelength photon absorption is throughout the utilization of highly mismatched alloys (HMA) where the decoupling in electronegativity and size of the atomic radii of the constituting elements, generates a splitting of the host material conduction band (CB). The lower and higher energy CBs after splitting are called as Eand E+, respectively. Thus, in a single layer three different transitions are allowed: i) valence band - E-, ii) E- - E+ and iii) VB -E+. Presumably, a larger number of transitions can be obtained if quantum confinement effects are added like in any other quantum well system. In this work, the physical and optical properties of multi-quantum well heterostructures of GaNAs are investigated. The GaNAs alloy, with nitrogen mole fraction  $x_N$  of 1.1%, was inserted between AlGaAs barriers with x<sub>Al</sub>from 0 to1 to selectively achieve the confinement of E+ and/or E-. Theoretically both GaNAs' bands may contribute to the localization of carriers, which certainly depends on  $x_{AI}$ . There is not clear evidence of quantum confined states related with E+, suggesting that E- is the sole energy band that contributes to the electron confinement. According to HRXRD measurements the Al concentration in the alloys partially relieves the strain of the multi-quantum well heterostructures. The Raman LO phonon can be related to the residual strain in films. In spite that this technique gets information only from those layers laying within the laser excitation source penetration length and that the heterostructures are capped with GaAs, the LO peak shifts to higher vibrating frequency with respect to the natural LO frequency of homoepitaxial GaAs (291.9cm<sup>-1</sup>), which is indicative of a reduction of tensile strain in the heterostructures.

#### 8:45am NM-WeM1-4 Strained Ge Quantum Wells by Molecular Beam Epitaxy for Superconducting Quantum Circuits, *Patrick Strohbeen*, *M. Hatefipour, W. Strickland, I. Levy, J. Shabani*, New York University

Superconducting quantum circuits utilizing superconductor-semiconductor (S-Sm) Josephson Junctions (JJs) enable rapid gate-tunability of qubit frequency and are an extremely promising avenue for future quantum computing devices [1,2]. However, these systems utilizing InAs 2D electron systems (2DESs) suffer from strong dielectric losses in the substrate they are grown on, resulting in significantly reduced qubit lifetimes [1,3]. To this end, it is highly advantageous to develop a new materials platform for gate-tunable superconducting qubits that are not limited by substrate dielectric loss.

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In this talk, I will discuss our work on the molecular beam epitaxy (MBE) growth of a 2D hole system (2DHS) in shallow strained Ge quantum wells as an alternative structure to InAs 2DESs. Samples are grown in an MBE system via electron-beam evaporation to enable rapid deposition of group IV species. Shallow Ge quantum wells are proximitized via in-situ deposition of a superconducting metal enabling higher quality S-Sm interfaces than can be achieved via ex-situ metal deposition. Use of the 2DHSs in strained Ge quantum wells for superconducting quantum circuitry will also be discussed.

[1] L. Casparis, M. R. Connolly, M. Kjaergaard, et. al., Nat. Nanotechnol.13, 915-919 (2018).

[2] M. C. Dartiailh, W. Mayer, J. Yuan, et. al., Phys. Rev. Lett. 126, 036802 (2021).

[3] J. O'Connell Yuan, K. S. Wickramasinghe, W. M. Strickland, et. al., J. Vac. Sci. Technol. A 39, 033407 (2021).

9:00am NM-WeM1-5 Vertical Transport in Bulk InAsSb and InAs/InAsSb and InGaAs/InAsSb Superlattices Grown on GaSb is Investigated using Photoluminescence Spectroscopy and Compared to Magnetoresistance Measurements, *Marko Milosavljevic*, Arizona State University; *R. Carracso*, *A. Newell*, Air Force Research Laboratory, USA; *J. Love*, New Mexico State University; *S. Zollner*, University of New Mexico; *C. Morath*, *P. Webster*, Air Force Research Laboratory, USA; *S. Johnson*, Arizona State University

Due to strong carrier confinement in type-II superlattices, vertical transport is much smaller than lateral transport. This asymmetry in carrier mobility affects the performance of type-II superlattice photodetector devices, is complicated to evaluate, and has been determined using temperature and magnetic field dependent magnetoresistance measurements on Van der Pauw structures [1].On the other hand, photoluminescence spectroscopy provides a noninvasive method to investigate carrier transport in bulk and superlattice materials. In which case, photoexcited carriers generated in a mid-wave absorber diffuse into a long-wave well. The ratio of the long to mid wave photoluminescence intensity strongly depends on the diffusion rate of photoexcited carriers into the long-wave well, providing carrier mobility for the mid-wave region. In this work, temperature dependent steady state photoluminescence measurements are performed on samples containing 800 to 2100 nm thick mid-wave absorber regions that are bulk InAsSb or superlattice InAs/InAsSb or InGaAs/InAsSb on top of 130 nm thick long-wave InAs/InAsSb superlattice wells.

In the analysis, the relative luminescence intensity from the long-wave well depends on its carrier lifetime and radiative coefficient, and most significantly on the diffusion rate of the carriers from the mid-wave absorber into the long-wave well. The ratio of the long to mid wave luminescence is proportional to the ratio of the respective photon extraction factors (~1), radiative coefficients (~0.1), and integrated carrier densities squared. The carrier density term has a strong power law dependence on the carrier diffusion length in the mid-wave region, providing the desired sensitivity to vertical transport. The optical constants of the mid and long wave materials are measured using spectroscopic ellipsometry to accurately determine the radiative coefficients.The extracted vertical hole mobility for the mid-wave InAs/InAsSb superlattices are consistent with those reported for magnetoresistance measurements [1]. There are methods of analysis that do not require knowledge of the material optical constants, as they assume that the external quantum efficiency ratio, and hence radiative lifetime ratio, are approximately unity.For the samples investigated here, this approximation underestimates the hole mobility by two orders of magnitude and is not utilized

[1] L.K. Casias, C.P. Morath, E.H. Steenbergen, G.A. Umana-Membreno, P.T. Webster, J.V. Logan, J.K. Kim, G. Balakrishnan, L. Faraone, S. Krishna, Vertical carrier transport in strain-balanced InAs/InAsSb type-II superlattice material, Appl. Phys. Lett. **116**, 182109 (2020).

9:15am NM-WeM1-6 Tensile-Strained InGaAs Quantum Dots With Interband Emission in the Mid-Infrarred, *Kevin Vallejo*, Idaho National Laboratory; *T. Garrett*, Boise State University; *C. Cabrerara-Perdomo*, Autonomous University of the State of Morelos, Mexico; *M. Drake*, Boise State University; *B. Liang*, University of California, Los Angeles; *K. Grossklaus*, Tufts University; *P. Siimonds*, Boise State University

We determined a robust set of growth conditions for the self-assembly of tensile-strained  $In_{1x}Ga_xAs$  quantum dot (QD) nanostructures on GaSb(111)A surfaces. During molecular beam epitaxy (MBE),  $In_{1x}Ga_xAs$  QDs form spontaneously on GaSb(111)A seemingly with less than 1 ML deposited, indicating a Volmer-Weber growth mode. We characterized

these nanostructures using atomic force microscopy, transmission electron microscopy and energy-dispersive X-ray spectroscopy to understand InGaAs/GaSb(111)A QD structure as a function of the MBE conditions. A combination of photoluminescence spectroscopy and computational modeling shows that residual tensile strain in the QDs reduces the InGaAs band gap energy to produce band-to-band emission at 3.5-3.9 µm. When coupled with quantum size effects, the use of tensile strain to red-shift QD emission offers an attractive way to create highly tunable mid-IR light sources.

9:30am NM-WeM1-7 High Quality Quantum Dot Formation on 300 Mm Si Photonic Wafers for Monolithic on-Chip Light Source, *Chen Shang, E. Hughes,* UC Santa Barbara; *A. Clark,* IQE Inc.; *R. Koscica, K. Feng,* UC Santa Barbara; *G. Leake, D. Harame,* SUNY Polytechnic Institute; *J. Bowers,* UC Santa Barbara

Monolithic integration via direct epitaxial growth III-V gain material onto Si substrates is the ultimate solution for laser integration with silicon photonics. High reliability has been achieved in InAs quantum dot (QD) lasers grown on (001) blanket Si substrate. However, the required thick buffer layers for the defect reduction makes the evanescent light coupling to the underneath Si waveguides difficult. Growing the III-V gain materials in pockets in a butt-coupled configuration is a more promising approach. Transferring the QD active region from the blanket Si substrate into the pockets on the patterned Si template has proven to be nontrivial. In this work, we have successfully realized high quality QD nucleation in pockets after identifying the growth challenge introduced by the template architecture. Pockets are aligned to either the [1 1 0] or the [1 -1 0] crystal orientation.

Since most of the template is covered with the oxide mask, the use of RHEED as the *in-situ* monitor for QD nucleation is completely forbidden. We observed that the sample with the oxide mask gives a lower pyrometer reading compared to that measured on blanket GaAs/GaP/Si. Once the oxide mask is covered with polycrystalline III-V from the non-selective growth, the pyrometer reading is higher than that measured on blanket GaAs/GaP/Si, depending on the thickness of the polycrystal. The consequence of the temperature uncertainty results in either an over- or under-estimated actual growth temperature. Sample heater power calibrated on a blanket GaAs/GaP/Si template was then used for the pocket growth instead of using the pyrometer, resulting in a blanket-substratelevel low defect III-V film with no QD contrast observed. This is attributed to the much lower heat conductivity of the oxide mask than the III-V material in the pockets. Thus, even though the sample was heated to the same temperature as if it were a blanket GaAs/GaP/Si piece, less heat is radiated from the oxide mask, resulting in a hotter-surface that evaporates the InAs dots. Arbitrary lowering of the heater power was then carried out at 5 °C intervals and QD nucleation was realized after an approximately 30 C° decrease with respect to the temperature profile obtained on blanket GaAs/GaP/Si template. Room temperature photoluminescent and crosssection TEMs were obtained from materials grown in pockets aligned to either [1 1 0] or the [1 -1 0] crystal orientation. Clear differences were observed, potentially attributed to the asymmetric surface diffusion. Further investigation is required to understand the underlying cause of the material asymmetry and to provide guidance to future template and growth designs.

9:45am NM-WeM1-8 Manipulating Surface Diffusion for InAs Quantum Emitters at Telecommunication Wavelengths by Droplet Epitaxy, Margaret Stevens, NRC Postdoctoral Fellow residing at the Naval Research Laboratory; W. McKenzie, G. Baumgartner, Laboratory for Telecommunication Sciences; J. Grim, S. Carter, A. Bracker, Naval Research Laboratory

Droplet epitaxy (DE) is a useful growth technique to achieve light emitters that span the near-infrared wavelengths, including ranges important for telecommunication applications. The two-step growth process, including nucleation of group-III droplets in a group-V depleted environment, followed by crystallization under group-V overpressure, enables tuning of the quantum dot size, density, and morphology without being constrained by strain. DE can also yield quantum dots with very low fine structure splitting [1], an important quality to achieve entangled photon pairs. InAs DE quantum emitters are typically grown by (1) metal organic chemical vapor deposition on (001) surfaces or (2) molecular beam epitaxy on (111)A or (311)A surfaces. InAs DE structures grown on (001) surfaces often yield "ring" shapes, unless the droplet deposition temperature is near room temperature. These ring shapes form due to the high surface diffusion of indium adatoms. While these remaining rings, often made up of smaller

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clusters of quantum dots, have interesting optical properties, quantum dots for telecommunication sciences require symmetric, isolated dots with low areal density.

In this study, we explored methods of manipulating indium surface diffusion to achieve symmetric, hemispherical quantum dots on (001) InP surfaces grown by MBE. We explored deposition temperature and group-III coverage, the impact of strained underlayers, and a two-step group-V flux technique, previously demonstrated for InAs droplets on GaAs [2], to discourage indium from diffusing away from the center of the droplet faster than it can crystallize. We used atomic force microscopy to characterize the droplet morphology, and photoluminescence spectroscopy of capped samples to characterize the optical properties. Ultimately, we showed that controlling indium surface diffusion is critical to achieving quantum dots that are suitable for telecommunication sciences experiments.

[1] J. Skiba-Szymanska et al., Phys. Rev. Appl, 8, 014013, 2017

[2] S. V. Balakirev et al., Appl. Surf. Sci. 578, 152023, 2022

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