

MBE

Room Max Bell Auditorium - Session MBE-TuM

Bismuth Alloys/Antimonides

Moderators: Richard Mirin, National Institute of Standards and Technology, James Gupta, NRC

8:30am MBE-TuM-1 MBE Young Investigator Award Talk: Tensile-strained Self-assembly of Quantum Dots for Entangled Photon Sources and Band Structure Engineering, *Paul Simmonds*, Boise State University **INVITED**

Since the 1990s, self-assembled quantum dots (QDs) have been the subject of intensive research for technologies ranging from high-stability lasers, to intermediate band solar cells. Driven by compressive strain, semiconductor QDs form spontaneously on the (001) surfaces of III-V and group IV materials. There are however certain applications for which QDs on non-(001) surfaces, or QDs under *tensile* rather than compressive strain, are highly desirable. For example, theory predicts that the low fine-structure splitting of (111) QDs should make them ideal entangled photon sources; tensile strain would induce a dramatic reduction in QD band gap, with implications for infrared optoelectronics and nanoscale band structure engineering. However, until recently it has been extremely challenging to synthesize non-(001) or tensile-strained QDs that are free from crystallographic defects.

Since 2010, I have been working on a robust new MBE-based approach to QD self-assembly that overcomes these difficulties, and enables the controllable growth of defect-free, tensile-strained QDs on (111) and (110) surfaces. By engineering a situation where the kinetic barrier to dislocation nucleation and glide is large, we open a window within which tensile strain is instead relieved elastically by 3D self-assembly. Our model predicts this approach will work for any material with a zinc-blende or diamond-cubic crystal structure.

I will describe some different material systems for which we have explored the use of tensile-strained self-assembly to create nanomaterials with novel functionalities. For example, I will discuss the promising properties of tensile-strained (111) QDs for entangled photon emission. I will also present some early results from our quest to transform Ge from an indirect to a direct band gap semiconductor by producing tensile-strained Ge QDs. The effects of tensile strain on QD band structure could impact a wide range of technologies, from highly tunable infrared devices, to quantum media conversion based on light-hole excitons. I am confident that tensile-strained self-assembly represents a powerful, versatile new tool for heterogeneous materials integration, and nanomaterial development.

[1] P.J. Simmonds et al., ACS Nano, 7, 5017 (2013).

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9:00am MBE-TuM-3 Mechanisms of Compositional Inhomogeneities in Bismide Films, *C Tait, B Carter, V Caro, Joanna Millunchick*, University of Michigan

III-V semiconductor alloys containing Bi have attracted attention due to their novel properties, including a large reduction of bandgap [1], reduced temperature dependence of the bandgap [2], and an increase in spin orbit-coupling [3] with increasing Bi concentration. It has proven difficult to grow high Bi content films, as droplet formation and compositional inhomogeneities arise during growth. These phenomena are crucial to understand, because such fluctuations can cause carrier localization and degradation of device performance. Kinetic Monte Carlo growth simulations (Fig. 1a.) predict that the highest Bi incorporation rates occur when Ga droplets form on the surface [4]. This is because Bi incorporation requires a high availability of Ga [5]. However, growths exhibiting Ga droplets on the surface result in compositional fluctuations (Fig. 1b). We postulate this effect is caused by local variations in Bi incorporation rates due to the nonuniform Ga availability near the droplet. Indeed, high contrast related to high scattering from Bi on the surface near the droplet is seen in Fig. 1b. It was also observed that droplets led to degradation of film crystallinity, verified with X-Ray diffraction [6]. Growing under a higher As/Ga ratio eliminates Ga droplets, but not necessarily compositional inhomogeneities. Figure 1c. shows the formation of a lateral composition modulation. Nanometer-sized clusters of Bi-enriched GaAsBi also form [7]. Raising the growth temperature can mitigate lateral composition modulation and clustering, but results in lower Bi incorporation. In this talk we will also map out the droplet free-growth conditions to maximize the Bi incorporation and mitigate droplet-induced inhomogeneities.

9:15am MBE-TuM-4 In-situ UV Irradiation on the Uniformity and Optical Properties of GaAsBi Epi-layers Grown by MBE, *Daniel Beaton*, ScientaOmicron

The remarkable tunability of the band gap and spin orbit splitting in III-V alloys with only a small amount of bismuth makes the alloy potentially useful material for many applications. However, it is difficult to synthesize GaAsBi epi-layers with sufficiently high optical quality hampering the technological impact of the material. Spectral linewidths are typically exceedingly broad and band edge emission often suppressed at low temperatures by recombination at low energy states. In-situ UV irradiation of semiconductor alloys has been shown to lead to material quality improvements in the past and we apply it here to the growth of GaAsBi. Samples were irradiated by a pulsed 248nm laser focused to a 7x7mm spot, where rotation during growth results in a uniformly illuminated central region and a periphery that is radiated by a fluctuating fluence. With irradiation GaAsBi was shown to be much improved, where luminescence linewidths as low as 14meV were demonstrated and band edge emission observed to low temperatures.

Using the inherent variation of the fluence across the sample we explore the role of the irradiation. In the central uniformly lit region, steady state growth processes are achieved more quickly, yielding more abrupt interfaces, as well as uniform GaAsBi epi-layers. Comparing photoluminescence spectra at low (6K) temperature shows an increasing density of cluster related emission with decreasing fluence. This is observed in the shift in the peak emission energy away from the band edge, where the peak is shifted most dramatically far from the illuminated region. These results indicate a reduction in clustering of incorporated bismuth atoms with the use of the incident UV irradiation, and additionally the density of clusters may be controlled by the degree of irradiation

9:30am MBE-TuM-5 Manipulating Film and Underlayer Strain to Understand Composition Modulation in GaAsBi, *Margaret Stevens, K Grossklaus, J McElearney, T Vandervelde*, Tufts University

GaAs_{1-x}Bi_x is an interesting optoelectronic material that opens up new band gap and lattice constant possibilities for near-, mid-, and far-IR applications. Although thin films of high Bi content GaAs_{1-x}Bi_x (up to x=0.22 at <50nm [1]) have been grown on GaAs in the literature, it is still difficult to grow high Bi content materials that are thick enough to act as active layers in devices. Additionally, we find that materials grown >30nm phase separate into vertically segregated bands and have periodic Bi content. By lowering the compressive strain or adding tensile strain into our 250nm thick GaAsBi layers, we achieve increased Bi incorporation as well as reduced compositional variations, as demonstrated by TEM and atom probe tomography (APT). In this work, we expand our underlayer study, incorporating underlayers of AlGaAs and lower bismuth composition GaAsBi to decouple strain effects from changes in surface reconstruction and surface composition. We hypothesize that moving from high compressive strain to tensile strain in the epilayer provides a more favorable starting surface for both incorporation of Bi as well as for growing homogenous films.

We grew our layers with a Veeco GENxplor MBE using a valved As₄ cracker and a solid source effusion cell for group III elements and Bi. We measured lattice constants and estimated average bismuth content using 004 and 224 HRXRD scans, alongside spectroscopic ellipsometry to measure the room temperature band gap. We characterized the degree of strain relaxation by examining reciprocal space maps around the 224 asymmetric reflection. We additionally used TEM to identify defect centers and APT to measure bismuth variation along the growth direction. We examined structures of GaAsBi/(underlayer)/GaAs with underlayers of InGaAs, GaAsBi, and AlGaAs. We propose that strain engineering may be applied to increase Bi content in GaAsBi films, allowing for the growth of small band gap optoelectronic devices on GaAs substrates.

[1] R.B. Lewis, M. Masnadi-Shirazi, and T. Tiedje Appl. Phys. Lett.101 082112 (2012)

9:45am MBE-TuM-6 Long-Wavelength InAs-based Interband Cascade Lasers Grown by MBE, *James Gupta, X Wu, G Aers*, National Research Council of Canada, Canada; *Y Li, L Li, W Huang, R Yang*, University of Oklahoma

Interband cascade lasers (ICLs) are becoming a leading semiconductor laser technology for the mid-infrared because of their high efficiency and low power consumption, especially as compared with conventional diode lasers and intersubband quantum cascade lasers (QCLs) in the wavelength range from 3-5 μm . Although a greater effort has been directed towards GaSb-

Tuesday Morning, October 2, 2018

based ICLs in the $\sim 3\text{-}5\mu\text{m}$ range, recent work has highlighted the exciting potential for InAs-based ICLs for reaching longer emission wavelengths.

In this work we report the development of low-threshold InAs-based ICLs with a room-temperature emission wavelength of $6.3\mu\text{m}$. The devices were grown on n+-InAs (100) substrates by solid-source molecular beam epitaxy in a custom V90 system using valved crackers for Sb₂ and As₂. The ICL structures employ an improved waveguide design using intermediate AlAs/AlSb/InAs strain-balanced superlattice cladding layers surrounded by heavily-doped n+-InAs plasmonic claddings. The active region includes 15-stages with AlSb/InAs/In(0.35)Ga(0.65)Sb/InAs/AlSb type-II "W" quantum wells and optimized electron injector doping.

In pulsed mode, broad-area devices lased at 300 K at a lasing wavelength of $6.26\mu\text{m}$ and a threshold current density of 395Acm^{-2} which is the lowest ever reported among semiconductor lasers at similar wavelengths. The broad-area devices lased up to 335K in pulsed mode at a wavelength of $6.45\mu\text{m}$. These results provide strong evidence of the potential for InAs-based ICLs as efficient sources in the mid-IR.

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10:30am MBE-TuM-9 Atomically Smooth InSb Quantum Wells on GaAs Substrates, *Yinqiu Shi, E Bergeron, F Sfigakis, J Baugh, Z Wasilewski*, University of Waterloo, Canada

High-quality InSb quantum wells (QW) are one of the most desirable material systems for the top-down approach in realizing Majorana bound states for topological quantum computing. Such QWs are typically grown with AlInSb metamorphic buffers on GaAs substrates. However, as predicted by the BCF theory [1], high-density pyramid-shaped hillocks form on the surface, which may cause spatial modulation in AlInSb barrier composition as well as variations in InSb QW thickness. Suppression of hillocks is thus essential. Here we report a comparative study on the surface morphology with and without InSb QWs on top of AlInSb metamorphic buffers, as a function of substrate offcut angle.

Modulation-doped InSb/AlInSb QWs were grown on edge-exposed 2" GaAs (001) substrates, using a Veeco Gen10 molecular beam epitaxy (MBE) system (Fig.1(a)). At the wafer centre, a large density of hillocks are formed on the surfaces of both the AlInSb metamorphic buffer and the complete InSb QW structure (Fig.1(b),(d)). Their surface morphologies then transition into smooth regions and eventually become rough again, as the polishing-induced effective offcut increases towards the wafer edge (Fig.1(c),(e)). Formation of hillocks is suppressed for effective substrate offcuts at around $0.4\sim 0.5^\circ$ towards [110] direction on the AlInSb buffer surface, which coincide with the facet angles of the hillock sidewall at the wafer centre, as derived from AFM scans revealing surface atomic steps. With InSb QW overgrown on the buffer, the large hillocks originated from the AlInSb surface are preserved while small hillocks, due to the very thin InSb QW layer, emerge on the large hillock sidewalls at the wafer centre (Fig.1(d)). The steeper sidewalls of these InSb hillocks indicate a larger substrate offcut needed for their complete suppression, as we predicted recently [2]. Indeed, as shown in Fig.1(e), a new morphological transition region is seen, where large AlInSb hillocks are already suppressed while small InSb hillocks persist. With the growth conditions used, an atomically smooth InSb QW surface is found at substrate offcut angles of around $0.5\sim 0.6^\circ$. We propose a model to explain the observed morphological transitions.

10:45am MBE-TuM-10 Narrow Bandgap InAsSb Detector on Digital Alloy AlInSb Metamorphic Buffer, *Vinita Dahiya, A Kazemi*, The Ohio State University; *E Fraser*, Intelligent Epitaxy Technology, Inc.; *J Deitz, J Boyer, S Lee*, The Ohio State University; *P Pinsukanjana*, Intelligent Epitaxy Technology, Inc.; *T Grassman, S Krishna*, The Ohio State University

Recently, the InAs_{1-y}Sb_y alloy system has emerged as a promising material in the long wave infrared (LWIR, 8-14 μm). However, to target LWIR wavelengths, Sb composition of more than 40% is required, which is relatively difficult to grow due to the non-availability of lattice-matched commercial substrates. This requires the growth of virtual substrates to target the desired lattice constant using, for example, step- or linear-graded metamorphic buffers. However, these structures typically suffer from various complexities, such as long growth interruptions and intricate cell temperature ramp profiles. In our previous work, we demonstrated the application of digital alloy (DA) defined compositions to produce step-graded Al_{1-x}In_xSb metamorphic buffers, which helps eliminate the issues mentioned above. The buffer layers grown via the DA technique exhibited relaxation behavior similar to conventional bulk, random alloy compositions. In the present work, this effort was further extended to grow subsequent lattice-matched InAsSb absorber layers and nBn detectors.

Tuesday Morning, October 2, 2018

To enable probing of the material and optical properties, 1.5 μm thick InAs_{0.55}Sb_{0.45} absorber material was grown lattice-matched to an Al_{0.68}In_{0.32}Sb terminal composition Al_{1-x}In_xSb DA (1.85 nm period thickness) metamorphic buffer, as described previously [2]. Based on high resolution X-ray diffraction reciprocal space mapping measurements, the absorber layer InAs_{0.55}Sb_{0.45} was found have residual strain of $\sim 0.1\%$. Electron channeling contrast imaging characterization indicated a low threading dislocation density on the order of $5\times 10^6\text{cm}^{-2}$ in the target InAs_{0.55}Sb_{0.45} layer. A photoluminescence peak was observed at $\sim 9\mu\text{m}$ at 77K, close to the expected value based on the XRD-derived composition [1].

nBn detector structures were then grown with an undoped 1.5 μm thick InAs_{0.55}Sb_{0.45} absorber and 100 nm thick Al_{0.68}In_{0.32}Sb barrier (Be doped; $1\times 10^{16}\text{cm}^{-3}$). Single pixel detectors of different sizes were fabricated via standard lithographic techniques. Preliminary spectral response of front side illuminated pixels revealed a 50% cutoff wavelength of $9\mu\text{m}$ at 150K. Radiometric characterization of the devices including dark current and quantum efficiency are being undertaken and results will be presented later.

[1] W. L. Sarney et al. , J. Appl. Phys. **122**, 025705, (2017).

[2] V. Dahiya et al., J. Vac. Sci. Technol., **B36**, 02D111(2018).

11:00am MBE-TuM-11 Molecular Beam Epitaxy of Wide-Bandgap InAlAsSb, *Stephanie Tomasulo*, U.S. Naval Research Laboratory; *M Gonzalez*, Sotera Defense Solutions; *M Lumb*, The George Washington University; *M Twigg, I Vurgafman, J Meyer, R Walters, M Yakes*, U.S. Naval Research Laboratory

Triple-junction solar cells, lattice-matched to InP, have recently gained interest as an alternative to traditional GaAs-based devices. To maximize efficiency, this design requires a top subcell with bandgap (E_g) of 1.74 eV, thus motivating the development of the widest direct-gap material lattice-matched to InP, In_xAl_{1-x}As_ySb_{1-y}. Both the immaturity and mixed group-V nature of this alloy pose significant challenges, requiring in depth investigation. Initial attempts at molecular beam epitaxy (MBE) of In_xAl_{1-x}As_ySb_{1-y} resulted in anomalously low photoluminescence (PL) emission energies, compared with energies extracted from variable angle spectroscopic ellipsometry (referred to as ellipsometry herein). To further investigate the cause of this discrepancy, in this work we have performed a systematic study of the substrate temperature (T_{sub}) and V/III of In_{0.26}Al_{0.24}As_{0.28}Sb_{0.22} (expected $E_g=1.64\text{eV}$).

We grew seven samples of In_xAl_{1-x}As_ySb_{1-y} ($x\approx 0.26$, $y\approx 0.28$) on InP by solid source MBE with valved crackers supplying cracked As and Sb. We investigated T_{sub} (measured via bandedge thermometry) ranging from 325 to 455 °C and V/III ratios (beam equivalent pressure) of 16 and 30. Given that the alloy composition varies with T_{sub} , we re-calibrated the group-V fluxes for each growth, using energy extracted from ellipsometry ($E=1.69\pm 0.05\text{eV}$) and lattice matching (to within 0.1% mismatch) via x-ray diffractometry as our compositional guide. Room temperature PL yielded emission from only four of the seven samples and we again found that it underestimates the energies extracted from ellipsometry. Low-temperature PL will be performed to inform the remaining three samples. Furthermore, we hypothesize that phase separation and clustering is responsible for this behavior and will probe this via power- and temperature-dependent PL measurements and transmission electron microscopy. By quantifying phase separation in this way, we can relate degree of phase separation to growth conditions, guiding us toward the appropriate conditions for In_xAl_{1-x}As_ySb_{1-y} yielding the least phase separation and widest E_g .

11:15am MBE-TuM-12 Minority Carrier Lifetime and Recombination Dynamics in Strain-Balanced GaInAs/InAsSb Superlattices, *Preston T. Webster, E Steenbergen, G Ariyawansa, C Reyner*, Air Force Research Laboratory; *J Kim*, Sandia National Laboratories

Strain-balanced InAs/InAsSb superlattices are rapidly emerging as a contending mid-infrared sensing technology as decreasing dark currents lead to ever more sensitive detectors. Dark current can be minimized by increasing the absorption coefficient and utilizing a thinner absorber region, thereby reducing the volume over which dark current is generated. While the InAs/InAsSb superlattice design may be optimized for maximum absorption, there remains great room for improvement by establishing a more favorable strain-balance condition. Specifically, replacing the lightly-tensile InAs layers with more-tensile GaInAs leads to a more symmetric wavefunction overlap profile and correspondingly stronger absorption for the same energy cutoff. The absorption coefficient in GaInAs/InAsSb superlattices improves substantially with Ga content up to 20%.

Tuesday Morning, October 2, 2018

In this work, two strain-balanced GaInAs/InAsSb superlattices (0% and 20% Ga) designed for maximum absorption at 5 μm wavelength are examined using temperature- and excitation-dependent photoluminescence spectroscopy and time-resolved microwave reflectance. The superlattices are 1 μm thick and doped $4 \times 10^{15} \text{ cm}^{-3} n$ -type in order to examine the optimal doping density \times lifetime product in a potential diffusion-limited detector. The 77 K time-resolved microwave reflectance decays of the 0% and 20% Ga designs are 1.2 μs and 2.1 μs respectively. The photoluminescence is evaluated using a recombination rate model to extract the Shockley-Read-Hall, radiative, and Auger rate constants as a function of temperature, to compare to the temperature-dependent minority carrier lifetimes determined by the time-resolved photoconductivity decay.

11:30am **MBE-TuM-13 Inhibited Hot-Carrier Cooling in InAs/AlAs_{1-x}Sb_x Quantum Wells**, *H Esmailpour*, *V Whiteside*, University of Oklahoma; *H Piyathilaka*, West Virginia University; *S Vijayaragunathan*, *B Wang*, University of Oklahoma; *E Adcock-Smith*, *K Roberts*, University of Tulsa; *T Mishima*, **Michael Santos**, University of Oklahoma; *A Bristow*, West Virginia University; *I Sellers*, University of Oklahoma

Semiconductor quantum wells (QWs) have been shown to exhibit decreased hot-carrier thermalization relative to bulk systems. Recently, we proposed that type-II QWs have the potential to further inhibit hot-carrier relaxation via the decoupling of the phonon channels through the spatial separation of photogenerated carriers. The spatial separation increases the radiative lifetime for the hot electrons, and leads to the formation of a robust phonon bottleneck at elevated temperatures. A decoupling of the thermalization coefficient was observed when the system transitioned from efficient type-I radiative recombination at low temperature to less efficient type-II recombination at elevated temperatures.

The multiple-QW structure consists of a 10 nm AlAs_{0.14}Sb_{0.86} barrier followed by 30 repetitions of a 2.4 nm InAs QW and a 10 nm AlAs_{0.14}Sb_{0.86} barrier grown by molecular beam epitaxy on a GaAs (001) substrate. Photoluminescence (PL) measurements at several lattice temperatures were modeled via a generalized Planck radiation law using the carrier temperature as a fitting parameter. The effective temperature of the carriers above the lattice temperature is weakly dependent on the excitation power, but becomes significantly hotter with increasing lattice temperature. When the lattice is at room temperature, the carrier temperature is 490K. Carrier lifetimes were determined by THz time-domain spectroscopy, which measures the AC photocurrent as a function of delay time between a near-infrared pump pulse and a THz probe pulse. Inspection of the three decay contributions reveals that the mechanism for the fast component (~ 0.01 ns) is different than for the intermediate and slow components. The fast decay is attributed to direct recombination within the QW, due to transitions from the electron ground state to localized hole states arising from alloy fluctuations. The intermediate (~ 0.3 ns) and slow (~ 2 ns) decay times, which dominate at high temperatures, are due to a two-step decay process between the same sets of initial and final states. These competing faster and slower components are attributed to the redistribution of photogenerated holes and the degeneracy of the valence band at elevated temperatures. This leads to competition and the convolution of the PL from multiple confined hole states and the ground state electrons. Confirmation of the long-lived nature of the photogenerated electrons in the InAs QWs supports the notion that the carriers in the QW facilitate a phonon bottleneck.

11:45am **MBE-TuM-14 Observation of Interface Electronic States from InAs/GaSb Multi Quantum Wells Grown by Molecular Beam Epitaxy**, *S Alyamani*, **Jong Su Kim**, *J Shin*, Yeungnam University, Korea; *S Lee*, *J Kim*, Korea Research Institute of Standards and Science, Korea; *S Lee*, *V Dahiya*, *S Krishna*, The Ohio State University

We have investigated optical transitions in the InAs/GaSb multiple quantum wells (MQWs) by photoreflectance (PR) and photoluminescence (PL) spectroscopy with various temperatures and excitation intensities. PR measurements were performed using a 405 nm laser diode as an excitation source. The probe beam obtained from a tungsten-halogen lamp dispersed through a monochromator. The reflected beam was collected by using a Si (400 \sim 1100 nm: high energy region) and InGaAs (1200 \sim 2400 nm: low energy region) photodiodes. The PR was employed to investigate the inter-band transitions such band-to-band (E_{GaSb}), spin-orbit split off (Δ_0), E_1 and Δ_1 of GaSb [1] as well as their interface quantum states (IQS).

Fig. 1(a) and (b) show the room temperature PR spectra at near band transition and above band transition for InAs/GaSb (5 ML/50 nm) MQW, respectively. PR spectra of the InAs/GaSb MQWs showed the E_{GaSb} , the

Franz-Keldysh oscillation (FKO) and IQS. We confirmed the transition energies from 0.72 eV, 1.52 eV, 2.07 eV and 2.53 eV corresponding to the E_{GaSb} , $E_{\text{GaSb}} + \Delta_0$, E_1 and $E_1 + \Delta_1$, respectively. Moreover, at room temperature PR spectrum, we observed sharp transition features due to the IQSs from the interface of InAs/GaSb. At near 1.2 eV region, we found unidentified transitions (UIS) and which were investigated by excitation intensity and temperature dependent PR. At low temperature PR and PL results, we found the QS transition between confined electrons states in InAs QW and GaSb valence band at energy of 0.506 eV as shown in Fig. 2

Author Index

Bold page numbers indicate presenter

— A —

Adcock-Smith, E: MBE-TuM-13, **3**

Aers, G: MBE-TuM-6, **1**

Alyamani, S: MBE-TuM-14, **3**

Ariyawansa, G: MBE-TuM-12, **2**

— B —

Baugh, J: MBE-TuM-9, **2**

Beaton, D: MBE-TuM-4, **1**

Bergeron, E: MBE-TuM-9, **2**

Boyer, J: MBE-TuM-10, **2**

Bristow, A: MBE-TuM-13, **3**

— C —

Caro, V: MBE-TuM-3, **1**

Carter, B: MBE-TuM-3, **1**

— D —

Dahiya, V: MBE-TuM-10, **2**; MBE-TuM-14, **3**

Deitz, J: MBE-TuM-10, **2**

— E —

Esmailpour, H: MBE-TuM-13, **3**

— F —

Fraser, E: MBE-TuM-10, **2**

— G —

Gonzalez, M: MBE-TuM-11, **2**

Grassman, T: MBE-TuM-10, **2**

Grossklaus, K: MBE-TuM-5, **1**

Gupta, J: MBE-TuM-6, **1**

— H —

Huang, W: MBE-TuM-6, **1**

— K —

Kazemi, A: MBE-TuM-10, **2**

Kim, J: MBE-TuM-12, **2**; MBE-TuM-14, **3**

Krishna, S: MBE-TuM-10, **2**; MBE-TuM-14, **3**

— L —

Lee, S: MBE-TuM-10, **2**; MBE-TuM-14, **3**

Li, L: MBE-TuM-6, **1**

Li, Y: MBE-TuM-6, **1**

Lumb, M: MBE-TuM-11, **2**

— M —

McElearney, J: MBE-TuM-5, **1**

Meyer, J: MBE-TuM-11, **2**

Millunchick, J: MBE-TuM-3, **1**

Mishima, T: MBE-TuM-13, **3**

— P —

Pinsukanjana, P: MBE-TuM-10, **2**

Piyathilaka, H: MBE-TuM-13, **3**

— R —

Reyner, C: MBE-TuM-12, **2**

Roberts, K: MBE-TuM-13, **3**

— S —

Santos, M: MBE-TuM-13, **3**

Sellers, I: MBE-TuM-13, **3**

Sfigakis, F: MBE-TuM-9, **2**

Shi, Y: MBE-TuM-9, **2**

Shin, J: MBE-TuM-14, **3**

Simmonds, P: MBE-TuM-1, **1**

Steenbergen, E: MBE-TuM-12, **2**

Stevens, M: MBE-TuM-5, **1**

— T —

Tait, C: MBE-TuM-3, **1**

Tomasulo, S: MBE-TuM-11, **2**

Twigg, M: MBE-TuM-11, **2**

— V —

Vandervelde, T: MBE-TuM-5, **1**

Vijayaragunathan, S: MBE-TuM-13, **3**

Vurgaftman, I: MBE-TuM-11, **2**

— W —

Walters, R: MBE-TuM-11, **2**

Wang, B: MBE-TuM-13, **3**

Wasilewski, Z: MBE-TuM-9, **2**

Webster, P: MBE-TuM-12, **2**

Whiteside, V: MBE-TuM-13, **3**

Wu, X: MBE-TuM-6, **1**

— Y —

Yakes, M: MBE-TuM-11, **2**

Yang, R: MBE-TuM-6, **1**