Tuesday Morning, September 20, 2022

Novel Materials Room Swan BC - Session NM-TuM2

Infrared Materials

Moderator: Ida Sadeghi, MIT

10:30am NM-TuM2-11 Strain-Engineered MBE Growth of InAs Quantum Dots Emitting at Telecom Wavelengths, *Bianca Scaparra*, A. Ajay, H. Riedl, G. Koblmueller, J. Finley, K. Mueller, Walter Schottky Institut, Technische Universität München, Germany

In recent years, there has been a great interest in the search of solid-state spin-photon interfaces that couple flying photonic qubits at telecom wavelengths and stationary matter qubits [1,2]. Operating at telecom wavelengths is strongly desired to benefit from the low propagation losses that occur in the commonly employed, industry compatible silica fibers. Deterministically charged semiconductor quantum dots (QDs) are a very promising platform for spin-photon interfaces, due to their fast emission rates and fast optical spin manipulation [1]. However, the most widely studied QDs, InAs QDs embedded in a GaAs matrix, have their transition wavelength mainly centered in the near infra-red region.

In this work, we describe our recent progress on the optimization of two epitaxial growth methods that lead to QDs with emission wavelength in the O and C telecom bands. First, in order to shift the emission wavelengths of the QDs towards the O-band, the study of the growth of a high crystal quality InGaAs strain-reducing layer (SRL) above the QDs is presented [3].

Reciprocal space maps show how, by carefully tailoring the growth conditions of the SRL, the degree of relaxation and alloy composition are affected, thus leading to the expected redshifted QDs photoluminescence emission.

With the goal to further shift the emission wavelength towards the C-band, an In-graded InGaAs metamorphic buffer layer (MBL) is grown beneath the QDs, leading to further strain reduction [3].

With the aim to achieve a deeper understanding of the influence of growth parameters on the material quality of the MBL, reciprocal space maps of In_x Ga_{1-x}As MBLs grown with different grading profiles are presented. In particular, the samples studied consist of linearly graded In_x Ga_{1-x}As buffers with different constant slopes of the In content. These results are combined with compositional EDX mappings for further proving the variation of the In content distribution with different grading profiles.

Finally, in order to understand how the QDs formation arises on the MBL, we discuss how the density and shapes of InAs QDs are correlated to the different In grading profiles and growth conditions.

[1] L. You et al., Nature Comm., 6, 8955 (2015)

[2] Ł. Dusanowski et al., Nature Comm., 13, 748 (2022)

[3] S. L. Portalupi et al., Semicond. Sci. Technol., 34, 053001 (2019)

10:45am NM-TuM2-12 InP-based InAs Quantum Dot/dash Lasers Emitting in the O-band, Sadhvikas Addamane, Center for Integrated Nanotechnologies, Sandia National Laboratories; S. Seth, Center for High Technology Materials, University of New Mexico; S. Hawkins, N. Collins, Sandia National Laboratories; C. Shang, Y. Wan, University of California Santa Barbara; G. Balakrishnan, Center for High Technology Materials, University of New Mexico; J. Klem, Sandia National Laboratories; R. Venables, Intel Corp.; J. Bowers, University of California Santa Barbara

Epitaxially-grown quantum dot (QD) lasers are emerging as the ideal candidate for on-chip sources in fiber-optic communication applications. QDs exhibit 3-dimensional carrier confinement which translates to several advantages over traditional quantum well (QW) lasers: low threshold current density, high temperature-stability, reduced linewidth enhancement factor, and increased tolerance to defects¹. In the O-band (1260-1360nm), most QD-related laser work has been limited to GaAsbased structures and recent advances have demonstrated exceptional device properties including epitaxial integration on Si substrates¹. The case for exploring InP-based QD lasers in the O-band can be made from both material and device perspectives. The lattice mismatch between InAs and InP is lower (3.2%) compared to the InAs/GaAs system (7.2%); this could lead to improved reliability of lasers. Further, strain compensation is more straightforward on InP and could facilitate stacking of multiple QD layers without accumulating strain. On the device side, InP-based QD lasers in the C-band have been shown to yield higher modal gain (per QD layer) values compared to 1.3µm InAs/GaAs lasers. If this advantage extends to the O-

band, higher modal gain would alleviate limitations to device geometry. In this work, we present results from a study focused on realizing $1.3\mu m$ QD lasers based on InP.

Methods: The samples described in this study were grown on InP substrates using molecular beam epitaxy (MBE).Short structures consisting of 5x QD layers embedded in InGaAlAs barriers were first grown for photoluminescence (PL) measurements. Each QD layer consists of an asymmetric dot-in-a-well (DWELL) region and the surrounding InGaAlAs QW composition is varied between the different PL samples. Starting from the well-established C-band (1.55µm), increasing the bandgap of the QW slowly moved the emission wavelength towards 1.3µm. Atomic force microscopy (AFM) measurements carried out on uncapped QD layers reveal elongation of the QDs along a preferred crystalline direction (quantum dashes). The optimized active region is integrated into a laser structure with AlInAs cladding and InGaAs (both lattice-matched to InP) p- and n-contact layers. Laser characterization results (I-V, L-I and spectrum) will also be presented.

¹ Norman et al., IEEE J. Quant. Elect. 55(2)–2019

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11:00am NM-TuM2-13 Photonic Crystal Surface Emitting Lasers (PCSELs) grown by Molecular Beam Epitaxy, *Thomas J Rotter*, *S. Seth*, *K. Reilly*, *F. Ince*, University of New Mexico; *A. Kalapala*, *Z. Liu*, *W. Zhou*, University of Texas at Arlington; *G. Balakrishnan*, University of New Mexico

Photonic crystal surface emitting lasers (PCSELs) based on InGaAs quantum wells emitting near 1040nm are fabricated by molecular beam epitaxy (MBE). PCSELs achieve vertical emission with high beam quality while avoiding the complexity associated with DBR growth for VCSELs. The epitaxial structure of a PCSEL is similar to that of an edge emitting laser. Vertical emission is achieved by a photonic crystal, which is realized as a pattern of holes near the waveguide core. PCSELs can be power scaled with area while maintaining coherent emission.

The fabrication process starts with MBE growth of a part of the laser structure. This includes the lower clad and waveguide core with the active region. Next, in the top surface of the waveguide (PC layer) a pattern consisting of periodically spaced voids is fabricated using electron-beam lithography and ICP etch. In the third step, the regrowth, the top clad and contact layers are grown on the patterned surface, which completes the laser structure.

Epitaxial regrowth is challenging due to infilling of the voids and because obtaining a clean surface for regrowth is difficult. Thermal and chemical treatment options to prepare the patterned surface for regrowth are explored.

For the PC design, it is important to match its resonance to the photoluminescence of the active region. Relevant parameters such as void shape and PC fill factor are influenced by the regrowth. SEM characterization of the voids before and after regrowth shows that preregrowth void radius influences the growth dynamics, i.e. growth at the sidewalls versus at the bottom of the void. Regrown PCSEL devices are demonstrated by optical pumping and electrical injection.

In addition to PCSEL fabrication by regrowth, non-regrowth methods are explored. The advantages include significantly reduced fabrication complexity and time. Here the entire laser structure is epitaxially grown and the PC fabricated on the surface of the top contact, which requires etching of very deep voids. Non-regrowth PCSELs are compared to edge-emitters from the same epitaxial structure.

11:15am NM-TuM2-14 Low Growth Temperature Epitaxial PbSe for Heterogeneous Mid-Infrared Emitters, Leland Nordin, J. Meyer, P. Reddy, K. Mukherjee, Stanford University

The mid-infrared (MIR) wavelength range is vital for an abundance of sensing, health/biological monitoring, security and defense, and fundamental science applications. For compact solid-state sources of MIR light, III-V semiconductor heterostructures, such as quantum cascade lasers (QCL), interband cascade lasers (ICL), or type-II superlattice light emitting diodes (SLED), are often used. However, QCL, ICL, and SLED structures all suffer from substantial growth complexity. Recently, thin films of PbSe grown epitaxially on III-V GaAs (001) substrates have shown high internal quantum efficiency (IQE) at room temperature and comparatively low

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growth temperatures of 300 $^\circ\text{C},$ compatible with back end of line processes [1].

In this work, we show that the growth window for high optical-quality PbSe/III-V extends to substantially lower temperatures than previously demonstrated and investigate the recombination dynamics of the low growth temperature films. A high temperature (300 °C) PbSe film [Figure 1(a)] and a lower temperature (170 °C) PbSe film [Fig. 1(b)] were grown on oxide-desorbed and arsenic-capped GaAs (001) substrates in a molecular beam epitaxy (MBE) system using a compound PbSe source. Both growths started with a PbSe dose procedure and nucleation layer [2]. Following the nucleation layer, the growth temperature was lowered to the either 300 °C or 170 °C, and 80 nm of PbSe was grown. Room temperature MIR photoluminescence (PL) was measured on both films and is shown in Fig. 1(c). Notably, the magnitude of PL is nearly identical for both the high and low growth temperature films. To better understand the impact of growth temperature on luminescence efficiency we performed power dependent PL (PDPL) measurements, shown in Figure 2. Remarkably, we do not see a substantial deviation in PL magnitude for lower pump powers, suggesting similar SRH recombination rates

We have grown PbSe films on GaAs substrates at 300 °C and 170 °C and investigated their luminescence properties. PL is not dramatically impacted by the lower growth temperature, corroborated by both the magnitude of spectral dependent PL and power dependent PL. Most importantly, the low growth temperatures investigated are compatible with back end of line processes, likely extend the accessible crack-free film thicknesses, and possess bright PL. Additionally, these low growth temperatures enable the investigation of more esoteric ternaries, such as PbGeSe, which require extremely low growth temperatures.

[1] J. Meyer et al. ... K. Mukherjee, APL Mater., 9,111112 (2021).

[2] B. B. Haidet et al. ... K. Mukherjee, Phys. Rev. Mater., 4, 033402(2020).

11:30am NM-TuM2-15 Structural Properties of MBE-grown PbSnSe on GaAs (001) Films for Mid-infrared Optoelectronics Investigated by X-ray Diffraction, Jarod Meyer, Stanford University; E. Hughes, University of California at Santa Barbara; L. Nordin, K. Mukherjee, Stanford University

The narrow gap IV-VI semiconductor PbSe is an intriguing material for heterogeneously integrated mid-infrared light emitters in the 3-5 um wavelength range. The combination of an anomalously low Auger recombination coefficient and high tolerance of minority carriers to crystal defects suggests the potential for mid-infrared light emission with high quantum efficiency. Recently, we showed that bare, epitaxial thin films of PbSe on (001)-oriented GaAs emitted brightly despite an 8% lattice mismatch, indicating a high internal quantum efficiency of luminescence at room-temperature.

Moving beyond binary PbSe, the bandgap may be tuned across the midand far-IR via alloying with Sn in the ternary Pb_{1:x}Sn_xSe.² This motivates us to explore how alloying Sn alters the microstructure and luminescence properties of PbSe films. Strain-induced structural distortions, stemming from lattice and thermal-expansion mismatches with GaAs, are also important for understanding the optoelectronic properties of these thin films.

We grew Pb_{1*}Sn_xSe epitaxial films on (001)-oriented GaAs substrates by molecular beam epitaxy at 280–330 °C using PbSe and SnSe compound source effusion cells, following a 400 °C surface pretreatment with PbSe flux that we find essential for good photoluminescence. High resolution x-ray diffraction was used to extract alloy compositions and structural properties. Across a Sn composition range of x = 0–0.31, full-width-at-half-maximums of (004) and (224) rocking curves were in the range of 370 – 510 and 700 – 840 arcseconds, respectively, indicating only minor differences in structural quality. For the x = 0.41 sample, however, structural quality was significantly degraded, likely due to the presence of both rock-salt and orthorhombic phases in the film. In-plane residual tensile strains of 0.25–0.50% at room-temperature were also found for the Pb_{1*}Sn_xSe films, in agreement with the predicted thermal mismatch strain between PbSe and GaAs upon cooldown from growth temperature.^{1,3}

Preliminary room-temperature photoluminescence of dilute Pb_{1-x}Sn_xSe ternaries shows redshifted peak wavelengths out to 4.6 μ m for x = 0.03, but with significantly decreased intensity compared to PbSe. We will present results investigating and comparing loss mechanisms in PbSe and Pb_{1-x}Sn_xSe to understand whether these are fundamental or may be overcome by engineering.

[1] J. Meyer, A. J. Muhowski, L. J. Nordin, E. T. Hughes, B. B. Haidet, D. Wasserman, K. Mukherjee *APL Materials* **9** 111112 (2021).

[2]H. Preier, Appl. Phys. 20 189 - 206 (1979).

[3] C. P. Li, P. J. McCann, X. M. Fang, J. Cryst. Growth. 208 423 – 430 (2000).

11:45am NM-TuM2-16 MBE Growth and Characterization of an InAs/AIAs_{0.16}Sb_{0.84} Quantum Cascade Detector at 2.7 μm, *M. Giparakis*, *H. Knötig, S. Isceri, M. Beiser, H. Detz, W. Schrenk, B. Schwarz, G. Strasser, Aaron Maxwell Andrews*, Technische Universität Wien, Austria

We present the molecular beam epitaxy (MBE) growth, design, and characterization of an InAs/AIAs_{0.16}Sb_{0.84} quantum cascade detector (QCD) grown lattice-matched to an InAs substrate detecting at 2.7 μ m (0.459 eV), which is above the band gap energy of the InAs substrate of 0.345 eV (3.5 μ m) [1] and in the center of a CO₂ absorption line.

The InAs/AlAs_{0.16}Sb_{0.84} material system was chosen because it combines two important characteristics for improved optical absorption strength, low noise, high detectivity, and a broad designable wavelength range – one of the lowest effective electron masses m^*_e in III-V semiconductors of $m^*_e = 0.023 m_0$ and second the highest cubic conduction band offsets of 2.1 eV [2].

The growth of this material system is challenging due to the mixed group V compound in the barrier. The As and Sb fluxes must be precisely controlled, especially when growing in lattice-matched conditions. Additionally, QCDs designed for short wavelength absorption, like the one presented here, need thin wells and barriers – which were ranging from 1.38 to 3.85 nm for this QCD. The precise layer thickness control necessary for this structure was achieved by implementing special shutter sequences to reduce intermixing at the interfaces and growth rate variations caused by shutter operations were taken into consideration. Lattice matching well and barrier materials to the substrate improves the growth quality and more degrees of freedom exist, because strain balancing does not have to be considered.

The QCD was fabricated into mesas with a diffraction grating on top that allows top-side illumination, which is normally forbidden by the selection rule of intersubband devices, such as QCDs. It also allows for a detection wavelength above the band gap energy of the InAs substrate. To find the optimal parameters for the grating-period, grating-duty, and grating-depth simulations were performed using COMSOL.

The absorption spectrum was measured using a Fourier-transform infrared spectrometer together with a Globar and a longpass filter with a cut-on wavelength of 2.4 μm . The designed absorption wavelength of 2.7 μm was confirmed. The QCD shows a peak responsivity at room temperature of 5.63 mA/W and a specific detectivity of 1.14×10⁸ Jones.

[1] M. Giparakis et al., Appl. Phys. Lett. 120, 071104 (2022).

[2] P. Reininger et al., Appl. Phys. Lett. 107, 081107 (2015).

12:00pm NM-TuM2-17 Substrate Preparation and MBE Growth of High Quality α -Sn Topological Insulator Thin Films on InSb(001) Surfaces, Aaron Engel, C. Dempsey, University of California, Santa Barbara; S. Nishihaya, Y. Chang, University of California, Santa Barabara; M. Hashimoto, D. Lu, Stanford Synchrotron Radiation Lightsource; C. Palmstrøm, University of California, Santa Barabara

 α -Sn is one of the only elemental materials that shows multiple topological phases. Of high interest is the conversion from a Dirac semimetal state under compressive strain to a topological insulator state under both compressive strain and confinement. When in this topological insulator state, α -Sn has been calculated to have a large band gap and measured to have Dirac surface states with high Fermi velocity, making it attractive for spintronics applications [1]. α -Sn does not suffer from stoichiometry, uniformity, and disorder issues to the degree seen in compound topological materials. Unfortunately, interactions with the substrate have made clean studies into this system difficult. Particularly, the typical sputter cleaning and annealing preparation of InSb results in poor crystal quality at the interface and significant hole doping of the α -Sn films [2]. Here we present the combination of in-situ atomic hydrogen cleaning and Sb-termination of the InSb(001) substrates in an interconnected growth and characterization system to 1) prepare high quality α -Sn(001) by MBE and 2) limit the effects of hole doping. These methods, along with a UHV vacuum suitcase, allowed the α -Sn electronic bands to be studied at high resolution using synchrotron-based angle-resolved photoelectron spectroscopy (ARPES). We confirm that the 3D Dirac semimetal state transitions to a topological insulator under confinement. A two-dimensional electron gas is found to coexist with the Dirac surface state after electron doping the Sn surface. Magnetotransport measurements will be performed and compared with the ARPES spectra. Our work paves the way for deeper studies into pristine α-Sn prepared on a larger scale.

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[1] Y. Ohtsubo, P. le Fèvre, F. Bertran, and A. Taleb-Ibrahimi, Dirac Cone with Helical Spin Polarization in Ultrathin-Sn(001) Films, Physical Review Letters 111 (2013).
[2] I. Madarevic et al., Structural and Electronic Properties of the Pure and Stable Elemental 3D Topological Dirac Semimetal α-Sn, APL Materials 8, 031114 (2020).

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