NAMBE
Room Tamaya ABC - Session NAMBE1-TuA
Oxides and Group IV Materials

Moderators: Ezra Bussman, Sandia National Laboratories, **Sriram Krishnamoorthy**, University of California at Santa Barbara

1:30pm **NAMBE1-TuA-1 Development of Erbium Doped Epitaxial Scheelite Thin Films for Quantum Communication Applications***, **Ignas Masiulionis**,* University of Chicago*; Bonnie Lin,* Massachusetts Institute of Technology*; Gregory D. Grant,* University of Chicago*; Junghwa Kim,* Massachusetts Institute of Technology*; Jiefei Zhang,* Argonne National Laboratory*; James M. LeBeau,* Massachusetts Institute of Technology*; David D. Awschalom, Supratik Guha,* University of Chicago

The Er3+ ion, when embedded in a solid-state dielectric host, is an excellent candidate qubit for quantum communication and networking applications due to its well-shielded 4f-4f telecom c-band transition. CaWO4 has emerged as an intriguing host material for Er, with promising optical and spin properties demonstrated in bulk crystals [1,2]. CaMoO4 is another compound, similar to CaWO4, that is yet to be explored as a host material. Both compounds have a tetragonal (scheelite) structure, with low lattice mismatch (~3.1%) with silicon, presenting a unique opportunity for epitaxial growth on silicon and future on-chip integration for devices.

Precise stoichiometric control is essential for realizing high quality host materials for such applications. This control is best ensured via selection of self-limiting growth regimes in addition to pre-calibrated control of the flux delivery, with pre-growth flux control lacking adequate precision. In this work, we present our results of obtaining epitaxial scheelite thin film growth on oxide substrates under self limiting growth conditions, a first step towards epitaxial growth of these complex oxides on silicon. We utilize Ca, MoO3, WO3, and oxygen (atomic or molecular) for the growths of these scheelites using molecular beam deposition, followed by post-growth anneals in oxidizing environments. We have carried out structural characterization using X-ray diffraction (XRD) and scanning transmission electron microscope (STEM) to confirm epitaxial scheelite growth. These scheelite were doped with natural abundance Er3+ (1 - 100 ppm range).Er3+ 4f-4f emission have been examined via various optical characterization methods, including transient spectral hole burning (TSHB) and time-resolved photoluminescence excitation (PLE). In the annealed Er:CaMoO4 films, we observe TSHB linewidths of 500 MHz, inhomogeneous linewidths of 18 GHz, and T1 optical lifetimes up to 5 ms.

This material is based upon work supported by the Department of Defense/Air Force Office of Scientific Research through the University of Maryland.

[1] Marianne Le Dantec et al.,Twenty-three–millisecond electron spin coherence of erbium ions in a natural-abundance crystal. Sci. Adv. 7, eabj9786 (2021). DOI: 10.1126/sciadv.abj9786

[2] Mahmet T. Uysal et al., Spin-photon entanglement of a single Er3+ ion in the telecom band. arXiv:2406.06515v2

1:45pm **NAMBE1-TuA-2 Rapid Exploration of Oxide Growth Space through *in situ* Growth and Etching***, Stephen Schaefer, Davi Febba,* National Renewable Energy Laboratory*; Michelle Smeaton, Kingsley Egbo, Glenn Teeter, Syed Hasan, William Callahan, Andriy Zakutayev,* national renewable Energy Laboratory*; **M. Brooks Tellekamp**,* National Renewable Energy Laboratory

Beta phase gallium oxide (β-Ga_2O_3) is an emerging ultra-wide bandgap semiconductor that has attracted attention for its potential to outperform existing materials operating at high breakdown voltages and high temperature. Alloying of In and Al in β-Ga_2O_3 provides the ability to individually engineer the bandgap and lattice parameters of the material, providing a useful toolbox for heterostructure engineering. However, the tendency of $(Al,In,Ga)_2O_3$ alloys to form competing phases, along with the complex suboxide chemistry of Ga and In, results in a growth window that is difficult to map and an alloy which is difficult to control.

We describe a “high-throughput” molecular beam epitaxy (MBE) technique to screen the growth conditions for complex oxide growth space where suboxide desorption plays a key role. As a model system, we apply the method to the Ga_2O_3-In_2O_3 alloy system to determine appropriate synthesis conditions for monoclinic $(Al_xGa_{1-x-y}In_y)_2O_3$ alloys. By leveraging the sub-oxide chemistry of Ga_2O_3 and *in-situ* monitoring by reflection high-energy electron diffraction (RHEED), a cyclical growth and etch-back method is developed to rapidly characterize the $(In_yGa_{1-y})_2O_3$ growth space. This cyclical method provides approximately 10x increase in experimental throughput and 46x improvement in Ga_2O_3 substrate utilization. Growth conditions for monoclinic $(In_yGa_{1-y})_2O_3$ are identified and targeted growths are characterized *ex-situ* to confirm improved In incorporation. These conditions are then used to grow quaternary $(Al_xGa_{1-x-y}In_y)_2O_3$ with Al cation composition *x* ranging from 1% – 24% and In cation composition *y* ranging from 3% to 16%. The structural, chemical and optical properties of the alloys are investigated. An $(Al_{0.17}Ga_{0.76}In_{0.07})_2O_3$ alloy lattice-matched to Ga_2O_3 is examined by high resolution microscopy, highlighting the correlation between surface facets and composition. Such lattice-matched material can be grown arbitrarily thick without elastic strain and relaxation, making it suitable for high voltage diodes, transistor barriers, and epitaxial dielectrics.

2:00pm **NAMBE1-TuA-3 Metastable Iron-Oxide Phases by Epitaxial Matching to 4H-SiC (0001)***, **Alexandra Fonseca Montenegro**, Faisal Kimbugwe Kimbugwe, Marzieh Baan, Sevim Polat Genlik, Maryam Ghazisaeidi, Tyler Grassman, Roberto Myers,* The Ohio State University

Wustite Fe1-xO is the non-stoichiometric cubic rock salt phase of iron oxide which is stable at ambient pressure above 500C, but decomposes into Fe and Fe3O4 at room temperature. We aim to use epitaxial lattice matching to stabilize stoichiometric FeO. Lattice matching of iron oxide to miscut (4-deg) 4H-SiC (0001) substrates is explored via gas source molecular beam epitaxy (GSMBE) using an Fe-effusion cell and either O-plasma or molecular O2. Si-face 4H-SiC provides close lattice matching to stoichiometric FeO, however the heterovalent interface poses strong challenges, e.g. careful control of the interface bond ordering to avoid formation of SiOx silicate ring structures at the interface. Density functional theory (DFT) calculations predict the most energetically favorable interface structure. HRXRD simulation and measurement is used to identify the present phases, such as Fe1-xO, Fe, Fe3O4, and Fe2O3. We will discuss observations in RHEED, AFM, and XPS. Additionally, defect characterization is extensively explored via transmission electron microscopy (TEM) and electron channeling contrast imaging (ECCI), enabling a statistical quantification of misfit dislocations along the interface. Superconducting quantum interference device (SQUID) magnetometry, with its high sensitivity to weak magnetic fields, is employed to measure the magnetic responses of films with varying misfit dislocation densities. The relationship between dislocation density, phase stability, and magnetic response provides valuable insight into the fundamental properties of Fe-O thin films and their potential for spintronic and magnetoelectric applications.

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2:15pm **NAMBE1-TuA-4 SiGe/SnGe Superlattices Grown Using Molecular Beam Epitaxy***, **Allison McMinn**, Tyler McCarthy,* Arizona State University*; Yicheng Wang,* Dartmouth*; Xiaoyang Liu, Razine Hossain, Xin Qi, Zheng Ju,* Arizona State University*; David Jaeger,* University of North Texas*; Jifeng Liu,* Dartmouth*; David Smith, Yong-Hang Zhang,* Arizona State University

The Group-IV material system, including Si, Ge, and their alloys, has been one of the semiconductor industry’s most utilized and researched material families for over half a century. The expansion of this material family by the addition of α-Sn, the diamond cubic semiconducting form of Sn, in the last couple of decades has fostered new research into low-cost IR materials, novel IR detector concepts based on momentum(*k*)-space charge separation (*k*-SCS) effect, and short-range ordering (SRO) effects in semiconductors. SRO is referred to as the preferential local arrangements of constituent atoms over a short distance deviating from a completely random distribution. Superlattices (SLs) made of these Group-IV elements and alloys offer a platform with additional degrees of freedom regarding band structure design, alloy composition profile control, and strain balancing for these device applications and materials physics studies.

Structures such as $Si_xGe_{1-x}/Sn_yGe_{1-y}$ SL were designed and grown via MBE at substrate temperatures between 200 to 250°C to study these effects. To the best of our knowledge, this is the first report on SiGe/SnGe SLs. The targeted compositions chosen were x=15% and y=17.5% with period thicknesses of 2.38 and 2.35 nm, respectively, for a SL of 50 periods. HRXRD shows multiple satellite peaks and Pendellösung fringes; some features are due to shutter operation deviating from the designed SL.

APT measurements confirm expected interdiffusion at the interfaces in the SL, creating an equivalent tertiary $Si_aGe_{1-a-b}Sn_b/Si_cGe_{1-c-d}Sn_d$ SL instead of the designed binary $Si_{0.15}Ge_{0.85}/Sn_{0.175}Ge_{0.825}$ SL. The layers exhibit periodical composition variations between average values of a~ 2% and b~ 9% and c~ 4% and d~ 6%. The actual Si and Sn compositions in the corresponding layers were lower than the designed 15% and 17.5% values, respectively, with maximum incorporation values being ~ 6% Si in a SiGe and ~ 11% Sn in a SnGe layer. Dark and bright field TEM images reveal structure features introduced by the actual shutter operations, which have some irregularities. The individual SL period thicknesses estimated from TEM are 4.5 nm for the $Si_{0.02}Ge_{0.89}Sn_{0.09}$ layers and 3.2 nm for the $Si_{0.04}Ge_{0.9}Sn_{0.06}$ layers. Higher magnifications verify the interdiffusion seen in APT between adjacent SL layers, and the interfaces are not perfectly abrupt. Additionally, the chemical SRO in the SL was characterized as showing a significant difference compared to CVD-grown SiGeSn alloys. The study of SRO may lead toward new band engineering techniques beyond composition and strain, as well as the emergence of novel phase-change materials for Si electronics/photonics.

2:30pm **NAMBE1-TuA-5 Intervalence Band Transitions of α-Sn Films on InSb Substrates with Different Surface Reconstructions***, **Jaden Love**, Jan Hrabovsky, Carlos A. Armenta,* New Mexico State University*; Aaron N. Engel, Chris Palmstrom,* University of California at Santa Barbara*; Stefan Zollner,* New Mexico State University

Gray-tin, α-Sn, is a single crystalline phase of tin that crystallizes in the FCC diamond-like cubic structure with a known lattice constant of 6.4892 Å at room temperature. The unit cell belongs to space group O_h^7 (Fd3m) and contains 8 atoms located at the Wyckoff positions (0,0,0) and (1/4, 1/4, 1/4) [5]. α-Sn is a gapless semiconductor that has an inverted Γ_7^- band positioned between the Γ_8^{+v} (heavy hole) and the Γ_7^+ (split off) bands [1]. The $\bar{E}_0$ peak of α-Sn is attributed to allowed interband transitions from the Γ_7^- band to Γ_8^{+v} band or Γ_8^{+c} (light hole) band [1]. The interest in α-Sn stems from the ability to tune its band structure by changing the amount of doping, strain, and film thickness causing the material to behave like a topological insulator or semi-metal [3,4,6].

MBE was used to grow two 30 nm α-Sn layers on InSb (100) with c(8x2) and c(4x4) surface reconstructions. The c(8x2) substrate surface was prepared using an atomic hydrogen clean and light annealing to smooth the surface. The c(4x4) substrate surface was prepared using an atomic hydrogen clean and an anneal at higher temperatures under Sb_4 flux. These differences in MBE surface preparations lead to an Sb-terminated c(4x4) substrate surface and an In rich c(8x2) substrate surface [2].Termination of the substrate surface with Sb reduces unintentional hole doping by In therefore limiting the allowed transitions from the Γ_7^- band to the Γ_8^{+v} band, especially at low temperatures.

Here we discuss the presence of a strong $\bar{E}_0$ peak appearing in the extinction coefficient at 0.45 eV in room temperature infrared spectroscopic ellipsometry measurements. Temperature-dependent IR ellipsometry spectra were taken from 300 K - 10 K and show that the $\bar{E}_0$ peak amplitude is larger at higher temperatures for α-Sn films grown on InSb (100) c(4x4) substrates. Using the integrated $\bar{E}_0$ peak intensity of the dielectric function, we will calculate the carrier concentration as a function of temperature. Additionally high-resolution x-ray diffraction will be used to determine the strain of the α-Sn films.

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[2] A. N. Engel, et al., Phys. Rev. Mater. 8, 044202 (2024)

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[4] S. Kufner, et al., Phys. Rev. B 87, 235307 (2013).

[5] Landolt and R. Bornstein, Zahlenwerte und Funktionen aus Physik, Chemie, Astronomie, Geophysik und Technik, 4 (1955).

[6] H. Song, et al., Adv. Eng. Mater. 21, 1900410, (2019).

2:45pm **NAMBE1-TuA-6 Epitaxial Growth and Optical Properties of GeSn Alloys on Ge (100) and Si (100) via Molecular Beam Epitaxy***, **Nirosh Meckamalil Eldose**, Diandian Zhang, Dinesh Baral, Hryhorii Stanchu, Fernando Maia de Oliveira, Sudip Acharya, Wei Du, Fisher Yu, Gregory J Salamo,* University of Arkansas

Group IV alloys of Ge and Sn have attracted significant interest for electronic and optoelectronic applications on a Si platform. [1] The incorporation of α-Sn into Ge with concentrations as low as 6% induces an indirect-to-direct bandgap transition, offering enhanced optical properties compared to pure Ge. Higher Sn content enables mid- and long-wavelength infrared emission and detection, making GeSn a promising material for photonic integration. [2,3] However, the low (~1%) solid solubility of Sn in Ge and the substantial (~14%) lattice mismatch between α-Sn and Ge present major challenges in achieving Sn-rich $Ge_{1-x}Sn_x$ structures.

We report on the synthesis of high-quality strained and relaxed GeSn layers with significant Sn content grown on Ge (100) and Si (100) substrates via molecular beam epitaxy (MBE). A critical challenge in MBE growth is incorporating high Sn concentrations while suppressing Sn surface segregation. Growth temperatures ranging from 100°C to 200°C were investigated to optimize the crystalline quality of GeSn. High crystalline quality was demonstrated using high-resolution X-ray diffraction (HRXRD), while atomic force microscopy (AFM) provided insights into the surface morphology of the films. In this study, we report on the roles of strain relaxation and defect density on the GeSn optical properties. To accomplish this, we use photoluminescence (PL) from the defect transitions to measure the change in defect density with changes in GeSn film thickness and relaxation.

In addition, we report the first observation of direct bandgap PL emission from MBE-grown GeSn on Si (100) substrates without post annealing as shown in supplementary section Fig.1 (e). Secondary ion mass spectrometry (SIMS) depth profile of GeSn, confirming a Sn content of 11.4%, consistent with XRD-RSM measurements shown in supplementary section Fig. 1 (b and d). The GeSn layer, approximately 500 nm thick, exhibits a uniform Sn distribution, indicating homogeneous incorporation and ensuring compositional consistency and structural integrity, marking a significant step toward using MBE for integrating GeSn-based optoelectronic devices with Si technology.

References:

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