

Workshop on Quantum Materials Epitaxy

Room Ballroom A - Session QME-SaP

Workshop on Quantum Materials Epitaxy Poster Session

QME-SaP-1 Optimization of Hybridized InAsSb/InGaSb Semiconductor Topological Materials, *Heather Haugan*, Air Force Research Laboratory; *D. Das*, L. Ram-Mohan, Worcester Polytechnic Institute; *J. Corbett*, K. Mahalingam, R. Bedford, K. Eyink, Air Force Research Laboratory

We are in the midst of the second quantum revolution. Research institutes and companies worldwide are working toward harnessing the power of quantum physics for technological applications. Gapless surface states on topological insulators are protected from elastic scattering on nonmagnetic impurities,¹ which makes them promising candidates for low-power electronic applications. In previous years, most research efforts on demonstrating topologically protected edge states were focused on rather exotic topological materials.^{1,2} However it was extremely difficult to generate strong enough edge currents out of these materials, to be practically useful for widespread applications, due to low emission currents. Hence, we are exploring more commonly used infrared materials such as InAsSb/InGaSb quantum wells (QWs) and superlattices (SLs). Both structures can be designed to have an inverted gap by elevating the hole state above the electron state, and a large emission current, in particular in the SL by enhancing wavefunction overlaps. These are critical components on establishing topological states to create the conducting edge and the insulating bulk state. This unique circumstance can create dissipationless transport of electrons in heterostructures, which is particularly important for a variety of sensing applications. Therefore, as an initial test, we examine two hybridized topological structures; one for 6.22 Å atomic lattice constant 82 Å InAs/34 Å In_{0.4}Ga_{0.6}Sb/82 Å InAs symmetric QWs on In_{0.32}Ga_{0.68}Sb substrate³ and the other for 6.10 Å atomic lattice constant 70 Å InAs_{0.9}Sb_{0.1}/35 Å GaSb SLs on GaSb substrate. Both structures are tailored for the same hybridization gap of ~60 meV. By using a combination of theoretical modeling, high-resolution x-ray diffraction, and high-resolution transmission electron microscopy, we optimize the absorber designs and their molecular beam epitaxy process to achieve high-quality materials. Growth parameters in each design are carefully coordinated to mitigate the crystalline defects to produce high-quality materials. Quasiparticle interference mapping through a scanning tunneling microscope is used to investigate the band structure of a SL sample.

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QME-SaP-2 Doping the Undopable: Hybrid Molecular Beam Epitaxy Growth, n-type Doping, and Field-Effect Transistor using CaSnO₃, *Fengdeng Liu*, University of Minnesota, USA; *P. Golani*, University of Minnesota, USA; *T. Truttman*, University of Minnesota, USA; *I. Evangelista*, University of Delaware; *M. Smeaton*, Cornell University; *D. Bugallo*, Drexel University; *J. Wen*, University of Minnesota; *A. Kamath Manjeshwar*, University of Minnesota, USA; *S. May*, Drexel University; *L. Kourkoutis*, Cornell University; *A. Janotti*, University of Delaware; *S. Koester*, University of Minnesota; *B. Jalan*, University of Minnesota, USA

The alkaline earth stannates are touted for their wide band gaps and the highest room-temperature electron mobilities among all the perovskite oxides. CaSnO₃ has the highest measured band gap in this family and is thus a particularly promising ultra-wide band gap semiconductor. However, discouraging results from previous theoretical studies and failed doping attempts had written off this material as "undopable". Here we redeem CaSnO₃ using hybrid molecular beam epitaxy (hMBE), which provides an adsorption-controlled growth for the phase-pure, epitaxial and stoichiometric CaSnO₃ films. By introducing lanthanum (La) as an n-type dopant, we demonstrate the robust and predictable doping of CaSnO₃ with free electron concentrations, n , from $3.3 \times 10^{19} \text{ cm}^{-3}$ to $1.6 \times 10^{20} \text{ cm}^{-3}$. The films exhibit a maximum room-temperature mobility of $42 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at $n = 3.3 \times 10^{19} \text{ cm}^{-3}$. Despite having a smaller radius than the host ion, La expands the lattice parameter. Using density functional calculations, this effect is attributed to the energy gain by lowering the conduction band upon volume expansion. Finally, we exploit the robust doping by fabricating the CaSnO₃-based field-effect transistors. The transistors show promise for CaSnO₃'s high-voltage capabilities by exhibiting low off-state leakage below

20 pA/μm at a drain-source voltage of 100 V and on-off ratios exceeding 10⁶. This work opens the door to future studies on the semiconducting properties of CaSnO₃ and the many devices that could benefit from CaSnO₃'s exceptionally wide band gap.

QME-SaP-3 Solid Source Metal-Organic Molecular Beam Epitaxy for Epitaxial SrRuO₃ Films, *Anusha Kamath Manjeshwar*, *S. Nair*, *A. Rajapitamahuni*, *R. James*, *B. Jalan*, University of Minnesota

The investigation of the electrical and magnetic properties of SrRuO₃ and its associated Sr_{n+1}Ru_nO_{3n+1} Ruddlesden-Popper phases requires a high degree of control over the isolation of the desired phase and its defect density. The growth of ruthenates is fundamentally challenging because ruthenium (Ru) resists scalable evaporation and oxidation. This bottleneck complicates the growth of SrRuO₃ films with low defect densities using inherently low-energy, ultra-high vacuum deposition techniques such as molecular beam epitaxy (MBE). Special modifications to conventional MBE such as electron-beam assisted evaporation and ozone-assisted oxidation of Ru have, so far, enabled the best defect control or the highest residual resistivity ratios (RRR = $\rho_{300\text{K}}/\rho_{2\text{K}}$) in SrRuO₃ films among all physical vapor deposition techniques. However, these modifications are expensive and require additional interlocks to ensure safe operating conditions.

We outline a novel technique called solid source metal-organic MBE to supply a solid metal-organic precursor with pre-oxidized ruthenium with an effusion cell at $T < 200 \text{ }^\circ\text{C}$, a drastic decrease from the $\sim 2000 \text{ }^\circ\text{C}$ required to produce comparable fluxes with elemental Ru. With this technique, we demonstrate the growth of phase pure, epitaxial, and stoichiometric SrRuO₃ films with robust ferromagnetism below 150 K on SrTiO₃ (001) substrates. We simplify the route to an adsorption-controlled growth window in SrRuO₃ films, growth conditions where the films can self-regulate their stoichiometry, which is a key ingredient for successful defect control in electron-beam and ozone-assisted MBE-grown SrRuO₃ films. We discuss the intricate relationship between cation stoichiometry, magnetic domains, and RRR in epitaxial SrRuO₃ films and outline new pathways for achieving low defect densities in SrRuO₃. Using these guidelines to optimize stoichiometry and film thickness within a growth window, we achieve a RRR = 87 for a 50 nm-thick SrRuO₃ film, the highest for any SrRuO₃ film on SrTiO₃ (001) substrates. We will also illustrate how solid source metal-organic MBE is a simple and cost-effective method to enhance the capabilities of conventional MBE for the defect-controlled growth of ruthenates.

QME-SaP-4 Growth Mechanism of SrTiO₃ on a Graphene-covered Substrate using Hybrid MBE0, *SooHo Choo*, *H. Yoon*, University of Minnesota, USA, Republic of Korea; *B. Matthews*, Pacific Northwest National Laboratory; *S. Sharma*, University of Minnesota, USA; *S. Spurgeon*, *S. Chambers*, Pacific Northwest National Laboratory; *R. James*, *B. Jalan*, University of Minnesota, USA

Epitaxial films grown on a substrate covered with two-dimensional (2D) materials offer many exciting possibilities: reusability of the substrate; ability to obtain a freestanding membrane; and opportunity to reduce misfit dislocations. Three growth mechanisms are argued to be responsible for epitaxial growth on 2D material-covered substrate: (1) Remote epitaxy; (2) Van der Waals epitaxy; and (3) pinhole-assisted epitaxy. It is, however, still unclear which of these three mechanisms is responsible for epitaxial growth. In this talk, we will first present the successful growth of epitaxial SrTiO₃ nanomembranes on SrTiO₃ (001) substrates covered with bilayer graphene. Titanium tetraisopropoxide (TTIP) was used as a source of titanium and oxygen. No additional oxygen was used to avoid graphene oxidation. By varying Sr/TTIP beam equivalent ratios, we reveal a wide MBE growth window for adsorption-controlled growth of stoichiometric SrTiO₃ membranes [1]. Bulk-like lattice parameter of 3.905 Å was obtained for nanomembranes. By combining heteroepitaxial growth, high-resolution X-ray diffraction, atomic force microscopy, transmission electron microscopy, and Raman spectroscopy, we discuss all three growth mechanisms highlighting the role of graphene thickness, pinholes, and the substrate's ionicity on epitaxial growth.

QME-SaP-5 Growing Clean Crystals from "Dirty" Precursors in MBE, *Rashmi Choudhary*, University of Minnesota, USA; *Z. Liu*, *J. Cai*, *X. Xu*, *J. Chu*, University of Washington; *B. Jalan*, University of Minnesota, USA

Ultra-high purity elemental sources have long been considered a prerequisite for obtaining low impurity concentration in compound semiconductors in the world of molecular beam epitaxy (MBE). Furthermore, to realize intrinsic properties, the material needs to be nearly free of intrinsic and extrinsic defects. For this reason, the use of ultra-high-purity elemental sources has been the historical practice in MBE, perhaps,

for the fear that impurity elements might get incorporated into the film, making it “dirty”.

In this work, we challenge this conventional MBE wisdom by presenting an extension of the hybrid-MBE approach, known as solid-source metal-organic MBE, for growing superconducting Sr_2RuO_4 films using a solid organometallic precursor, ruthenium acetylacetonate, as a source of Ru. We grew 100 nm thick (001) Sr_2RuO_4 films on (001) LSAT substrate at 900°C substrate temperature using co-deposition of Sr, ruthenium acetylacetonate, and oxygen plasma. These films are phase-pure, single-crystalline, fully coherent, and superconducting. The superconducting transition temperature of the film is 0.85 K. In contrast to the conventional MBE, which employs ultra-pure Ru metal evaporated at $\sim 2000^\circ\text{C}$ as a Ru source, along with reactive ozone to obtain $\text{Ru} \rightarrow \text{Ru}^{4+}$ oxidation, the use of ruthenium acetylacetonate precursor requires significantly lower temperature for Ru sublimation (less than 200°C) and eliminates the need for ozone.

This is the first-time realization of superconducting Sr_2RuO_4 films using ozone-free MBE. By combining our results with the recent developments in hybrid-MBE, we argue that leveraging precursor chemistry will be necessary to realize next-generation breakthroughs in the synthesis of atomically precise quantum materials. Our results establish hybrid-MBE as a viable method for growing highest quality crystals and put this technique at the forefront of vacuum deposition technologies despite the use of a “dirty” chemical precursor.

QME-SaP-6 Growth of EuIn_2As_2 Thin Films by MBE: Towards Investigating the Topological Properties of a Candidate Axion Insulator, *Muhsin Abdul Karim*, University of Notre Dame, Ghana; *J. Wang*, University of Notre Dame, China; *K. Yoshimura*, University of Notre Dame, Japan; *S. Bac*, University of Notre Dame, Korea (Democratic People's Republic of); *X. Liu*, University of Notre Dame, China; *B. Assaf*, University of Notre Dame

EuIn_2As_2 is a promising topological material with an antiferromagnetic ground state predicted to support axion electrodynamics[1]. To observe these predicted properties and effects, thin films of the material must be synthesized. However, thin films of EuIn_2As_2 are not easy to make due to its nature of crystallization and the fact that it competes with thermodynamically stable zincblende phases of III-V materials. In this work, we present the thin film growth of EuIn_2As_2 on sapphire substrates by molecular beam epitaxy where we show that a high substrate temperature stabilizes the crystal structure of this compound. We have successfully grown thin films of thickness ranging between 50 – 120 nm. Magnetometry studies on the thin films agree very well with the result on bulk crystals[2]. The films exhibit the expected (ab)-plane magnetic easy axis and a Neel temperature close to 16 K. All our films are p-type and have charge carrier densities varying between $(2.8 - 7.4) \times 10^{14} \text{ cm}^{-2}$. We also find that the sample resistivity, ranging from $1.8 \times 10^{-5} - 2.8 \times 10^{-4} \Omega\text{m}$, is mainly determined by changing morphology of the films under different growth conditions. Our work provides a developed growth scheme for EuIn_2As_2 thin films, a crucial gateway towards realizing the predicted topological properties in this candidate axion insulator material.

QME-SaP-7 Synthesis of Free-Standing Membranes Using a Sacrificial Layer Method Grown by Hybrid MBE, *Shivasheesh Varshney*, *S. Choo*, *Z. Yang*, *J. Wen*, *S. Koester*, *B. Jalan*, University of Minnesota, USA

Free-standing membranes have broad applications in the creation of symmetry-mismatched, non-equilibrium, and artificial heterostructures. We use sacrificial layer method to synthesize phase-pure epitaxial SrTiO_3 membranes. In this study, we will discuss the growth of strain-engineered SrTiO_3 films using different sacrificial layer(s) grown by hybrid MBE. We characterize the as-grown films using x-ray diffraction (XRD) and atomic force microscopy (AFM). We show exfoliation and transfer of films onto dissimilar substrates, followed by their structural characterization. Finally, we use impedance spectroscopy to characterize the dielectric properties and show a bulk-like dielectric constant of ≈ 300 for SrTiO_3 membranes transferred on Au coated Si substrate.

QME-SaP-8 Epitaxial Growth of Precursor Phases of Novel Cuprate Superconductors Using Oxide MBE, *Jinkwon Kim*, *C. Kim*, *D. Schlom*, Cornell University

Since the discovery of high- T_c cuprates,^[1] a plethora of research has been conducted to understand their superconducting origin. They usually have layered perovskite structures and CuO_2 planes are considered as crucial ingredients to host Cooper pairs and d -wave superconductivity. The CuO_6 coordination octahedron is elongated along the c -axis, the $3d_{x^2-y^2}$ orbital dominantly contributes to the electronic structure at the Fermi level.^[2] In

2019, a totally different type of cuprate superconductor was discovered: $\text{Ba}_2\text{CuO}_{4-\delta}$ (bulk $T_c \sim 73 \text{ K}$ for $\delta = 0.8$).^[3] The octahedron of $\text{Ba}_2\text{CuO}_{4-\delta}$ was compressed along the c -axis, hence it becomes a multi-band system composed of $3d_{x^2-y^2}$ and $3d_{3z^2-r^2}$ orbitals. Moreover, oxygen vacancies even exist on the CuO_2 plane. These unique characteristics of $\text{Ba}_2\text{CuO}_{4-\delta}$ strongly suggest that the Cooper pair behavior is different from previously reported cuprate superconductors. Hence the study of $\text{Ba}_2\text{CuO}_{4-\delta}$ is expected to contribute to unveiling clues about the superconducting mechanism of high- T_c cuprates.

Unfortunately, the synthesis of $\text{Ba}_2\text{CuO}_{4-\delta}$ poses significant challenges. Ba-Cu-O compounds energetically prefer forming the Ba_2CuO_3 phase (a 1D CuO chain structure), hence strong oxidation is required to achieve the desired $\text{Ba}_2\text{CuO}_{3.2}$ phase. Li *et al.* achieved superconducting $\text{Ba}_2\text{CuO}_{4-\delta}$ specimens by high-pressure synthesis methods.^[3] But the chemical instability and polycrystalline structure limited deeper understanding of its electronic structure and superconductivity. Epitaxial thin film growth can be an alternative approach since it provides a strong oxidation environment with a large surface-to-volume ratio, a low reaction temperature, a pseudomorphic constraint from the substrate, and the ability to reveal the electronic structure with angle-resolved photoemission spectroscopy. In this study, using oxide MBE, we grew epitaxial thin films of the precursor phase of the $\text{Ba}_2\text{CuO}_{4-\delta}$ superconductor, Ba_2CuO_3 , and its homolog, Sr_2CuO_3 . After growth, the Ba_2CuO_3 and Sr_2CuO_3 films were oxidized by various methods such as post-growth ozone exposing or topotactic oxidation. We characterized the cuprate films by reflection high-energy electron diffraction (RHEED), x-ray diffraction (XRD), and atomic force microscopy (AFM). Our work on a thin-film approach toward single-crystalline $\text{A}_2\text{CuO}_{4-\delta}$ (A: Ba, Sr) superconductors will be presented.

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QME-SaP-9 MBE of $\text{Ba}_2\text{BiTaO}_6$, a Candidate p -type Oxide Semiconductor, *Anna Park*, *Y. Birkhölzer*, *M. Barone*, *D. Schlom*, Cornell University

Complementary metal-oxide-semiconductor (CMOS) technology is an important part of today's integrated circuit technology. CMOS replaced n -channel metal-oxide-semiconductor (NMOS) in the 1980s and with the 100 to 1000x power savings advantage it provides, enabled integrated circuits to grow from tens of thousands of NMOS transistors on a chip to tens of billions of CMOS transistors today. Today we stand at similar cross-roads for transistors made from oxide semiconductors. Only high-performance n -channel oxide transistors (and thus NMOS) exist for oxide transistors. If high performance p -channel oxide transistors could also be made, low-power CMOS would be possible in oxide systems and enable low-power transparent electronics. Although many p -type semiconducting oxides have been predicted based on their theorized electronic properties, few have been realized in experiment and those that have been achieved have much lower mobility than established n -type oxides like indium-gallium-zinc-oxide or In_2O_3 . Unfortunately, the realization of p -type oxides is particularly difficult due to the localization of the oxygen $2p$ orbitals. One design criterion to realize p -type oxides is to create oxides with Sn^{2+} , Pb^{2+} , or Bi^{3+} cations that have dispersive filled s -orbitals that will hybridize with oxygen $2p$ orbitals to delocalize the hole states and enhance mobility. We show that suboxide MBE can be used to grow $\text{Ba}_2\text{BiTaO}_6$, a candidate transparent p -type oxide.

Suboxide MBE utilizes molecular beams of suboxides, where the incoming cation precursors are already in the desired oxidation states. Many oxides crystallize with a perovskite structure and offer flexibility in accommodating a variety of cations, resulting in a wide range of properties. Double perovskites, of which $\text{Ba}_2\text{BiTaO}_6$ is an example, offer additional flexibility and another degree of freedom to explore in the interplay of structure and properties. For $\text{Ba}_2\text{BiTaO}_6$, we take advantage of suboxide MBE and avoid using an electron beam to evaporate Ta by supplying a molecular beam of TaO_2 from a Ta_2O_5 source. Additionally, with the volatility of Bi at our growth temperatures, we grow in an adsorption-controlled regime and fine tune the flux of Ba and Ta cations to create the desired phase.

QME-SaP-10 Flexomagnetism and Strain Induced Superconductivity in Rippled GdAuGe Heusler Membranes, Tamalika Samanta, Z. LaDuca, D. Du, T. Jung, S. Manzo, K. Su, M. Arnold, J. Kawasaki, University of Wisconsin - Madison

Rare earth-based Heuslers are prospective materials platforms for magnonics, topological spin texture, superconductivity, THz spintronics, etc. [1, 2]. The magneto-mechanical coupling in these materials allows for better control and manipulation of the primary order parameter and magnetic flexibility [3]. Here, we demonstrate novel flexomagnetic responses i.e., the coupling between strain gradient and magnetism, and strain-induced superconductivity, in GdAuGe Heusler membranes. The thin films of GdAuGe Heusler composition have been grown on monolayer Graphene/ Ge (111) by molecular beam epitaxy (MBE). GdAuGe films are then mechanically exfoliated to form free-standing rippled membranes.

GdAuGe shows an antiferromagnetic ordering below ~ 17 K, which is sustained when a homogeneous strain is applied. However, the application of strain gradient dramatically alters the magnetic ground state of GdAuGe in the rippled membranes. A phase diagram of the rippled GdAuGe membranes is shown in Fig. 1(a). Notably, a moderate strain gradient of a few tenths of a percentage transforms the ground state from antiferromagnetic to unconventional ferrimagnetic phases. These ferrimagnetic ground states in the rippled membranes offer the possibility of discovering spin reorientation and other unique magnetic phenomena; the most exciting observation is the emergence of superconductivity in GdAuGe membranes when a very large strain gradient is applied, with superconducting transitions occurring at low temperatures below ~ 3.5 K. Figure 1(b) shows the magnetic characterization of a superconducting GdAuGe rippled membrane.

At present, the microscopic origin of flexomagnetism and its effects on the thermodynamics of spin reorientation and phase transitions in these membranes remain unclear. Advanced spectroscopic measurements and magneto-transport experiments, combined with theoretical modeling, are planned to further investigate the phenomena in these rippled membranes.

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QME-SaP-11 Engineering Metal Oxidation Towards Epitaxial Growth of Complex Iridates using Molecular Beam Epitaxy, Sreejith Nair, Z. Yang, D. Lee, S. Guo, University of Minnesota, USA; J. Sadowski, Brookhaven National Laboratory; S. Johnson, Auburn University; A. Saboor, University of Delaware; Y. Li, H. Zhou, Argonne National Laboratory, USA; R. Comes, W. Jin, Auburn University; K. Mkhoyan, University of Minnesota, USA; A. Janotti, University of Delaware; B. Jalan, University of Minnesota, USA

The platinum group metals like Ir and Ru have captured significant interest in the condensed matter physics and materials science community due to the exotic electronic and magnetic properties that they exhibit when combined with oxygen. The oxides of these metals provide a unique platform to study and leverage the delicate interplay between electron correlations, crystal field and spin-orbit coupling energies. High quality thin films of complex platinum group metal oxides are hence, critical to realizing new phenomena such as the predicted unconventional superconductivity in Sr_2IrO_4 . However, the platinum group metals have extremely low vapor pressures and low oxidation potentials. These factors make it challenging to synthesize their oxide thin films using an ultra-high vacuum (UHV) technique like Molecular Beam Epitaxy (MBE). Here, we have addressed these challenges using a novel solid-source metal-organic MBE approach [1,2]. We demonstrate atomically precise synthesis of binary IrO_2 using $\text{Ir}(\text{acac})_3$ as the metal-organic Ir source at substrate temperatures as low as 250°C . The use of the metal-organic precursor allows Ir supply at source temperatures less than 200°C and enables easy oxidation due to the +3 Ir oxidation state in the precursor. Further, by combining epitaxially strained IrO_2 thin film growth on different substrates, x-ray diffraction, electron microscopy, spectroscopy techniques, and DFT calculations, we demonstrate a vital role of epitaxial strain in Ir oxidation. Thus, epitaxial strain can be an additional tuning knob to engineer metal oxidation which can aid the conventional thermodynamic and kinetic driving forces [3].

However, the true test of metal oxidation in UHV occurs at high growth temperatures where oxidation becomes increasingly thermodynamically

unfavorable. Hence, in order to examine the efficacy of the solid-source metal-organic MBE approach and to realize the elusive unconventional superconducting state, we study the synthesis of Sr_2IrO_4 thin films, which is favored at growth temperatures greater than $600\text{--}700^\circ\text{C}$. We will present a detailed growth study, structural characterization, electrical and magneto-transport in epitaxial Sr_2IrO_4 films, along with alternative ways to tackle the Ir oxidation challenge in UHV synthesis.

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QME-SaP-12 Improved Epitaxy of Unconventional Metals for Quantum Applications, Stefania Isceri, M. Giparakis, R. Svagera, M. Waas, Technische Universität Wien, Austria; V. Butera, E. Kolibalova, O. Man, Central European Institute of Technology, Czechia; L. Fischer, H. Detz, W. Schrenk, G. Strasser, S. Buehler-Paschen, A. Andrews, Technische Universität Wien, Austria

Strange metal thin films have attracted attention due to their promising applications in quantum devices. Strong correlations in YbRh_2Si_2 lead to intriguing phenomena, including linear-in-temperature strange metal behavior, phase transition from Landau-Fermi liquid to antiferromagnetic at the quantum critical point, electron delocalization transition [1], unconventional superconductivity [2], and suppression of shot noise [3]. In the Weyl-Kondo semimetal $\text{Ce}_3\text{Bi}_4\text{Pd}_3$ films, non-trivial surface states are present [4]. In this study, we demonstrate the improvement of epitaxial YbRh_2Si_2 films on $\text{Ge}(001)$ and the achievement of epitaxial growth of $\text{Ce}_3\text{Bi}_4\text{Pd}_3$ on sapphire.

We use an MBE chamber equipped with Knudsen cells for Bi, Ce, Pd, Yb, and e-beam evaporation sources for Rh and Si. First, we investigate the conditions in terms of growth temperature, and Yb flux to obtain smooth and stoichiometric samples without any surface treatment. Then, we performed density functional theory (DFT) calculations to analyze the most favorable adsorbed atoms in the first layer of YbRh_2Si_2 on the Ge surface. This study indicates that the adsorption of Rh on Ge (binding energy $E_b = -5.4$ eV) is favored over Si and Yb ($E_b = -4.4$ eV and -2.9 eV, respectively), so strongly that Rh atoms tend to kick out the Ge atoms. On the other hand, Si atoms diffuse on the Ge surface. We analyzed the improvement of the samples' surface by soaking the substrate with 1-2 ML of Yb before the deposition of YbRh_2Si_2 in the temperature range between 400°C and 475°C , measured by a pyrometer. The thickness of the samples spans 10 to 60 nm. The results show that with increasing Yb soaking time, a transition of the RHEED pattern from spotty to streaky, as well as the reduction of the surface roughness and defects occur.

For the second semimetal $\text{Ce}_3\text{Bi}_4\text{Pd}_3$, the sapphire substrates are cleaned with solvents and then annealed in the MBE machine to remove hydrocarbons. Then 50-nm-thick $\text{Ce}_3\text{Bi}_4\text{Pd}_3$ films are grown at 60°C (heater temperature) and a 10-nm-thick Si capping layer is deposited to prevent oxidation of the samples. In the early development of this research, x-ray diffraction shows that the epitaxy of polycrystalline films of this material is possible, whilst energy dispersive x-ray spectroscopy (EDX) and inductively coupled plasma-optical emission spectroscopy (ICP-OES) techniques are used to adjust the stoichiometric composition. Future investigations are planned on diamond (lattice mismatch=0.4%).

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Saturday Evening, September 16, 2023

QME-SaP-13 Growth and Angle-Resolved Photoemission of Strain-and Thickness-Tuned α -Sn Films, Aaron Engel, C. Dempsey, H. Inbar, S. Nishihaya, Y. Chang, University of California, Santa Barbara; A. Fedorov, Advanced Light Source, Lawrence Berkeley National Laboratory; M. Hashimoto, D. Lu, SLAC National Accelerator Laboratory; P. Taylor, P. Folkes, US Army Research Laboratory; C. Palmström, University of California, Santa Barbara

α -Sn, the diamond structure allotrope of Sn, is a zero-gap semiconductor with band inversion. Calculations suggest that epitaxial tensile strain induces a transformation to a topological insulator (TI) phase, while epitaxial compressive strain induces a transformation to a Dirac semimetal (DSM) phase [1,2]. When this DSM phase is confined, it is suggested to form a quasi-3D TI phase [3]. There is little consensus, however, on exactly how or if these transitions occur. The α -Sn based system is expected to have less alloy disorder and anti-site defects compared to the typical $(\text{Bi,Sb})_2(\text{Se,Te})_3$ TI system. Bulk α -Sn is also only stable at low temperatures, transforming into the topologically trivial β -Sn above 13.2 °C. This transformation temperature is raised above 100 °C by epitaxial stabilization of α -Sn ($a=6.489$ Å) on a closely lattice matched substrate like InSb ($a=6.479$ Å) [4]. Even at the low growth temperatures (<30 °C) necessary due to the phase transformation, incorporation of indium from the substrate as a p -type dopant in the epi-layer is difficult to prevent [5].

We first explore the essential role that surface preparation of the InSb(001) substrate has on both the quality and dopant density of the α -Sn films. Through magnetotransport and ultraviolet photoelectron spectroscopy measurements, we find that growth on the Sb-terminated $c(4\times 4)$ surface reconstruction results in higher mobility films with significantly reduced p -type doping. Using spin- and angle-resolved photoemission spectroscopy (ARPES), we study compressively strained α -Sn films on InSb(001) at a range of film thicknesses. These measurements provide essential clarification to the band structure of α -Sn: we observe the presence of a 3D TI-like phase in 13 bilayer films. Potential causes of this contradiction to the literature will be discussed.

With the previous behavior benchmarked, we then alloy the α -Sn films with Ge to tune from low (-0.15%) compressive strain on InSb to multiple tensile strains (+0.5%, +0.8%, +1.3%) at the same film thicknesses. Morphology changes as a function of Ge alloying were studied with *in-situ* scanning tunneling microscopy, and strain was confirmed through XRD. Finally, the presence of topological phase transitions induced by tensile strain is studied via ARPES. Our results pave the way for a better understanding of the effect of strain and confinement on α -Sn's band structure.

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QME-SaP-14 Electrostatic Gating of SrSnO₃ Thin Films with Improved Mobilities, Zhifei Yang, F. Liu, T. Truttmann, B. Jalan, University of Minnesota, USA

Ultra-wide-bandgap (UWBG) semiconducting oxides are becoming more crucial in sustainable technologies due to their promising use in applications including transparent electronics and power switching. Among them, alkaline earth stannates such as SrSnO₃ with the perovskite crystal structure have gained much interest in recent years. However, the room-temperature mobility of SrSnO₃ thin films has been shown to be limited by defective surface scattering. By using a 4 nm undoped SrSnO₃ capping layer on 19 nm La-doped SrSnO₃ thin film, the measured room-temperature mobility has been shown to improve. In this structure, charge spill over from the doped layer to the undoped layer is expected to happen as Fermi levels equilibrate. Here, we demonstrate a reversible and electrostatic doping of SrSnO₃ films grown by Hybrid molecular beam epitaxy with tunable carrier densities using electric-double-layer transistor configurations with ion gels. Using modeling and a discrete two-channel model, we show that the modulation due to gating is confined within 4 nm at the top capping layer and the modulation leads to an increase of mobility in SrSnO₃ up to 130 cm²V⁻¹s⁻¹ at 250 K. A detailed growth study combined with temperature-dependent Hall effect measurements and transport analysis will be presented.

QME-SaP-15 Epitaxial Growth and Transport Properties of Square-Net Rare Earth Telluride Thin Films, Adrian Llanos, J. Falson, California Institute of Technology

The square-net family of materials constitute a set of crystal structures which host a wide array of quantum phenomena including charge-density wave order, magnetism, superconductivity and topological band structures. The rare-earth ditellurides (RTe₂) (Space group P4/nmm) are an especially exciting subclass of these materials whose structure consists of square-planar, conducting Te sheets interspersed with insulating R-Te corrugated layers. Due to their unique crystal structures and chemical tunability, the RTe₂ compounds offer opportunities to study the effects of topological phenomena in the context of broken-symmetry ground states. This research seeks to use molecular beam epitaxy to open new avenues for control of the low-temperature properties of these materials.

In this presentation, we describe our recent work on epitaxial growth of rare-earth telluride thin films LaTe₂ and DyTe₂ grown on MgO substrates. The good lattice match of DyTe₂ (+~1.7%) has enabled the growth of epitaxially strained films in the ultra-thin limit (<3 unit cells (uc)). Observation of RHEED oscillations along with measured surface roughness on the order of ~1uc indicates layer-by-layer growth. Out-of-plane X-ray diffraction shows intense peaks with prominent Laue fringes and rocking curve full width at half maximum of ~0.02°. Thin films of LaTe₂ have also been produced with comparably high structural quality yet are relaxed within 1uc due to the +7% mismatch with MgO.

Using grazing incidence X-ray diffraction, a modulated superlattice in the (hk0) plane has been observed in DyTe₂ and shows a $\sqrt{5}\times\sqrt{5}$ modulation expected for Te-deficient DyTe₂. Additionally, incommensurate modulations that cannot be indexed according to previously reported modulated structures have also been observed. The role of Te deficiency in producing these modulated structures and in relaxing epitaxial strain will be discussed.

Magnetotransport studies on LaTe₂ have revealed previously unobserved, non-saturated, negative magnetoresistance that persists to room temperature. The temperature dependence of resistivity also shows a strong dependence on growth temperature and charge density, likely resulting from variations in Te deficiency. We will discuss our current understanding of these phenomena informed by the unique features in the band structure as well as the complex defect chemistry found in these materials.

QME-SaP-16 Improving MBE (Bi,Sb)₂(Te,Se)₃ Topological Materials Via Resonant and Magnetic Dopants, Patrick Taylor, Army Research Laboratory

The (Bi,Sb)₂(Se,Te)₃ topological insulator (TI) system hosts robust Dirac-like topological surface states, but suffers from parasitic conduction in the bulk. This parasitic conduction channel limits the technology transition of these emergent materials into useful device technologies. In this work, we present results of the MBE growth, *in-situ* ARPES measurements, DFT calculations, and magneto-transport investigations of select (Bi,Sb)₂(Se,Te)₃ alloys doped with Sn, a resonant dopant, [1,2] as well as Mn and Eu magnetic dopants whose function is intended to reduce bulk parasitic conduction.

The addition of tin is predicted by DFT to shift the Dirac point upwards in energy, and reduce the energy of the Fermi level out of the conduction band, and closer to the mid-gap Dirac states. Those DFT predictions were tested by *in-situ* ARPES measurements of MBE-grown films and are found to be consistent as shown in Figure 1.

Interestingly, as EuS is added during MBE growth as an independent flux, the carrier concentration drops and the mobility increases as the general transport behavior is consistent with significantly lower bulk conduction. *In-situ* ARPES measurements show the near complete absence of conduction band states. Figure 2 summarizes the magneto-transport and *in-situ* ARPES results from EuS doping. The significance of this work is that it highlights the potential for significant reduction in parasitic bulk conduction toward a loffe-Regel metal-insulator transition.

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QME-SaP-17 Growth of Cd₃As₂ on GaAs(001), GaAs(110), and Si(001) Substrates, Anthony Rice, I. Leahy, A. Norman, K. Alberi, National Renewable Energy Laboratory

The three-dimensional Dirac semimetal Cd₃As₂ has been shown to exhibit a variety of novel physics, providing a promising platform for their study. Thin film synthesis is enabling for scientific study as well as the realization of new devices, and growth has already been carried out on GaAs, GaSb, CdTe, SrTiO₃ and mica substrates. Due to its low energy (112) surface, however, the majority of thin film synthesis routes result in this orientation, while single crystals are limited by this cleave plane when performing studies requiring pristine surfaces. By expanding compatible substrate orientations, and ultimately the Cd₃As₂ orientation, much more of the band structure may be probed via photoemission, and a broader range of device structures may be integrated with it.

Here, we present the design of II-VI buffer layers to template high quality Cd₃As₂ in the (001) and (110) orientations on GaAs substrates. Lattice-matched Zn_xCd_{1-x}Te buffers are known to reduce defects in Cd₃As₂ epilayers grown on GaAs and improve their electron mobility [1]. We find that the addition of a ZnTe nucleation layers is critical for stabilizing Cd₃As₂ (001) growth on GaAs (001) substrates, while Zn₃As₂ nucleation layers are required to remove tilt in the Zn_xCd_{1-x}Te buffer when growing on GaAs (110). These films have a much different morphology due to the higher surface energy, and also a much different dependence on arsenic incorporation compared to Cd₃As₂ (112). However, we show that the Cd₃As₂ epilayers exhibit electron mobilities greater than 10,000 cm²/V-s. Finally, we present a methodology for growing Cd₃As₂ (112) epilayers on GaAs (001) substrates using CdTe to switch between orientations [2], also allowing for integration with Si (001) [3]. Such schemes will allow for the design of Cd₃As₂ orientation for specific measurement and application needs.

This work was performed by the National Renewable Energy Laboratory, operated by Alliance for Sustainable Energy, LLC, for the U.S. Department of Energy (DOE) under Contract No. DE-AC36-08GO28308. Funding was provided by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Division of Materials Sciences and Engineering, Physical Behavior of Materials Program under the Disorder in Topological Semimetals project.

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QME-SaP-18 Molecular Beam Epitaxy of Superconducting ZrN Thin Films on GaN Substrates, Kevin D. Vallejo, D. Hurley, K. Gofryk, B. May, Idaho National Laboratory

Group III-Nitride materials have found applications in optoelectronics and photonic devices due to the large variation in direct bandgap spanning from the infrared to the deep ultraviolet. Recent

research has pursued the integration of this well-established material system with transition-metal nitrides to create complex heterostructures with additional magnetic or superconducting functionality. ZrN is a well-known refractory conductor with high oxidation resistance, high hardness, and has been shown to be a superconductor at temperatures <10K. The estimated lattice mismatch of ZrN with InN, GaN, and AlN is 8.5%, -1.5%, and -4.2%, respectively, suggesting strain free integration with In-based ternaries. This work focuses on the epitaxial growth of ZrN on c-plane GaN substrates via molecular beam epitaxy. An electron beam source was used to evaporate Zr, and an RF-plasma source supplied the active nitrogen. Reflection high energy electron diffraction (RHEED) and X-ray diffraction (XRD) did not reveal any crystallographic texture of ZrN deposited on fused silica at temperatures >700°C. However, growth of ZrN on c-plane GaN substrates at similar temperatures was epitaxial. RHEED maintained similar hexagonal symmetry during the entirety of ZrN deposition and post-growth examination via XRD showed (111) oriented ZrN thin films. A physical properties measurement system (PPMS) was used to measure electrical transport as a function of temperature and magnetic field. Using a 4-point probe in a PPMS, initial results reveal that the epitaxial ZrN film is superconducting with a critical temperature of ~ 4 K and a critical field of 2 T. Because the critical temperature is lower than expected, the presence of off stoichiometry or structural disorder is suspected. These results pave the way for integration of superconductors and quantum phenomena in the III-

N photonic systems

QME-SaP-19 Quasi Van Der Waals Epitaxy of Magnetic Topological Insulators on a GaAs (111) Substrate, Yuxing Ren, University of California at Los Angeles; L. Tai, 404 Westwood Plaza, Engineering VI, 310; K. Pan, S. Srivastava, Y. Xie, M. Goorsky, K. Wang, University of California at Los Angeles

Magnetic topological insulator could achieve quantum anomalous Hall (QAH) effect and spin-orbit torque (SOT) switching in the same structure. This is promising for its future applications in memory or switching with its robust surface properties by topological protection. Considering the van der Waals nature of the epitaxial layers, it has very weak van der Waals bonding with the substrate. This gives rise to a novel quasi Van der Waals epitaxial growth mode at the interface of GaAs(111) substrates and the epitaxial layers, which has the advantages of both good crystallinity from substrate confinement, and a less influence from defects and roughness on the substrate surfaces. This is very crucial for achieving the quantization regime.

In this work we have done hetero-epitaxy of Cr:(Bi_xSb_{1-x})₂Te₃ and other magnetic topological insulators on GaAs(111) substrates by MBE (Molecular Beam Epitaxy). Unlike the pure Van der Waals epitaxy which has more freedom at the interfaces epitaxial layer and substrates, we found out that in this quasi Van der Waals growth mode, strain exist and relaxes quickly within the 1st epitaxial layer. While the surface defects quickly get screened within the 1st layer, the surface confinement also gives the epitaxial layer a uniform in-plane orientation which is important for achieving a single crystalline structure. Growth mechanism and the influence on its transport properties are also discussed.

QME-SaP-20 Transforming Rotating RHEED Data for Post-Growth Characterization Using Automated Machine Learning, Chris Price, Atomic Data Sciences

Reflection high-energy electron diffraction (RHEED) data is a characterization technique used to monitor the real-time surface structure and material morphology during epitaxial growth. Analyzing RHEED data over the growth duration can reveal a wealth of information about the relationship between the structure of the growing material and the growth procedure. In practice, this information is difficult to access and frequently neglected because transforming RHEED data into physically interpretable information is challenging and time-consuming. This is especially true if the growth stage is rotating, an important step to synthesize materials at device-relevant length scales with uniform growth across the substrate. Existing strategies to deconvolute the rotational and intensity oscillation frequencies must be calibrated to the materials system and measurement conditions and are brittle to structural changes during growth. We present an automated, material system-agnostic, and parameter-free approach to analyze rotating RHEED data. Using an entire unlabeled RHEED video as input, we extract the rotational frequency as a function of time in the video and create a complementary dataset averaged over the rotational period. The averaged data is used as input to a series of unsupervised dimensional reduction and clustering algorithms to identify transition points in the growth independent of rotation. Within each growth segment identified between these transition points, we extract original RHEED patterns at high symmetry scattering angles and quantify them using image segmentation models. Transitions are validated using small, labeled datasets of expert-identified growth transitions. The metrics automatically extracted at these angles are compared with the equivalent angles throughout the growth to label and quantify the evolution of the materials system. We align these quantified pattern metrics with in-situ environmental metrology data, such as quenching temperature, to build correlations between synthesized material structure and process variables. Fusing domain knowledge with machine learning, we reduce the time and effort barriers to accessing all the physical information collected with RHEED, producing physically interpretable datasets on materials structure over the course of a rotating growth.

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