Novel Materials
Room Swan BC - Session NM-TuM1

Novel Materials
Moderator: Debdeep Jena, Cornell University

8:00am **NM-TuM1-1 Navigating MBE Growth of Atomically Precise Complex Oxides of Stubborn Metals Using Source Chemistry**, ***Bharat Jalan,*** University of Minnesota, USA **INVITED**

Molecular-beam epitaxy (MBE) has come to the forefront for the thin film synthesis of materials with an exceptionally high structural quality, and with the best figures of merits whether it is related to their electrical or optical properties. From its beginnings as a successful method for III–V semiconductor growth to today for the growth of many contenders for next-generation electronics and spintronics devices, several new synthesis challenges have however emerged. For instance, it has been notoriously difficult to grow metal oxides, in an atomically precise manner, of metals having ultra-low vapor pressures and difficulty of oxidation.

In this talk, we will review these issues and will present our group's effort to address these challenges using a novel solid-source metal-organic MBE approach. We show, for the first time, controlled synthesis of metal and metal oxides of these "stubborn" elements with the ***same ease and control*** as afforded by III-V MBE. We will present detailed growth study utilizing chemistry of source materials as a controlling knob to navigate synthesis. With the goal to understand and control electronic ground states in defect-managed complex oxide films and nano-membranes, we will discuss how chemistry of source materials can be used to navigate synthesis on-demand.

8:30am **NM-TuM1-3 Pinhole-Seeded Lateral Epitaxy and Exfoliation on Graphene-Terminated Surfaces**, ***Sebastian Manzo,*** *P. Strohbeen,* University of Wisconsin - Madison*; Z. Lim,* University of Wisconsin - Madison, Malaysia*; V. Saraswat,* University of Wisconsin - Madison, India*; D. Du, S. Xu,* University of Wisconsin - Madison, China*; N. Pokharel,* University of Wisconsin - Madison, Nepal*; K. Su, L. Mawst, M. Arnold , J. Kawasaki,* University of Wisconsin - Madison

Remote epitaxy is a promising approach for synthesizing exfoliatable crystalline membranes and enabling epitaxy of materials with large lattice mismatch. However, the atomic scale mechanisms for remote epitaxy remain unclear. Here we experimentally demonstrate that GaSb films grow on graphene-terminated GaSb (001) via a seeded lateral epitaxy mechanism, in which pinhole defects in the graphene serve as selective nucleation sites, followed by lateral epitaxy and coalescence into a continuous film. Remote interactions are not necessary in order to explain the growth. Importantly, the small size of the pinholes permits exfoliation of continuous, free-standing GaSb membranes. Due to the chemical similarity between GaSb and other III-V materials, we anticipate this mechanism to apply more generally to other materials. By combining molecular beam epitaxy with in-situ electron diffraction and photoemission, plus ex-situ atomic force microscopy and Raman spectroscopy, we track the graphene defect generation and GaSb growth evolution a few monolayers at a time. Our results show that the controlled introduction of nanoscale openings in graphene provides a powerful route towards tuning the growth and properties of 3D epitaxial films and membranes on 2D material masks.

8:45am **NM-TuM1-4 Molecular Beam Epitaxial Growth of Cr-Sn Thin Films on Al2O3**, ***Tyler Erickson,*** *S. Upadhyay, A. Abbas, D. Ingram, A. Smith,* Ohio University

The suggestion of a spin liquid state in spin-1/2 Kagome antiferromagnetic materials poses interesting possibilities for investigating Kagome materials[1]. The confirmation of spin liquid states in the $ZnCu_3(OD)_6Cl_2$ spin-1/2 Kagome lattice antiferromagnet[2] and recent demonstration of a large anomalous Hall effect present in Mn_3Sn fims[3] demonstrate interesting magnetic properties valuable for future developments in spintronics and quantum materials. With a recent theoretical investigation into the Cr-Sn system that predicts an antiferromagnetic alloy[4], we now pursue the growth and characterization of this system. We investigate the growth of the Cr-Sn system on Al_2O_3 using molecular beam epitaxy with precise control over the flux ratio of Cr to Sn (from 1.7:1 to 5.1:1) and substrate growth temperature (650 °C - 850 °C). Growths for 1.7:1 and 5.1:1 Cr:Sn ratios were performed at 750 °C, and growths for 3.4:1 ratios have been performed at 650 ≥°C, 750 ≥°C, and 850 ≈°C. Reflection high energy electron diffraction patterns give *in-situ* characterization of the *in-plane* lattice, while X-ray diffraction characterizes the *out-of-plane* lattice. Preliminary analysis of RHEED patterns for Cr-Sn flux ratios of 3.4:1 and 5.1:1 give two distinct ranges of *in-plane* lattice constants. These ranges are 3.11 Å - 3.29Å and 3.98 Å - 4.06 Å. The second range gives good agreement with the *in-plane* lattice constant a = 4.054 Å calculated by Senthur Pandi Rajasabai *et al*[4]. We are currently in the process of further characterizing our Cr-Sn samples using Rutherford backscattering and XRD. This research was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. DE-FG02-06ER46317.

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9:00am **NM-TuM1-5 Growth of Mn_3Sn on Sapphire Using Molecular Beam Epitaxy**, ***Sneha Upadhyay,*** Ohio University*; T. Erickson , D. Ingram ,* Ohio University*; K. Sun,* University of Michigan, Ann Arbor*; A. Smith,* Ohio University

The kagome antiferromagnet Mn3Sn has become fascinating in the current times because it's one of the rare antiferromagnets that exhibits large anomalous Hall and Nernst effects. This opens a new area of research using functional antiferromagnets[1], but for future device applications, it requires fabricating high-quality thin films. There are reports of the controlled growth of Mn_3Sn on substrates like m-plane sapphire,[2] Pt/Al2O3 (0001)[3] and others using sputtering growth, but this often can result in polycrystalline films. In this work, the goal is to grow a crystalline high quality Mn_3Sn film using molecular beam epitaxy. Effusion cells are used for Mn and Sn sources which are calibrated using a quartz crystal thickness monitor. The growth is monitored in-situ using reflection high energy electron diffraction (RHEED) and measured ex-situ using X-ray diffraction, Rutherford backscattering and cross-sectional STEM. The samples are grown at 500 ± 9°C and 416 ± 9 °C with Mn: Sn atomic flux ratio of 3.2: 1 on c-plane sapphire for 60 mins. We observed that, for both temperatures, the RHEED patterns are streaky; however, the resulting orientations of the films are different. Additional results pertaining to surface morphologies, film orientations, chemical compositions, as well as empirical models will be discussed in detail.

Acknowledgement:

The authors acknowledge support from the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. DE-FG02-06ER46317. The authors would like to thank Dr. Stinaff and his students for back coating the sapphire substrates, as well as Ashok Shrestha for useful discussions and help.

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9:15am **NM-TuM1-6 Relaxed Epitaxial Constraints for Semi-freestanding Shape Memory Alloy Ni_2MnGa Films Grown on Graphene/MgO**, ***Zachary LaDuca,*** *S. Manzo, D. Du, K. Su, M. Arnold, J. Kawasaki,* University of Wisconsin - Madison

Ferromagnetic shape memory alloys such as Ni_2MnGa have great potential as microelectronic actuators due to their ability to reversibly transform between multiple crystalline phases. However, epitaxial growth of shape memory alloys can prevent their ability to undergo structural phase transformations through substrate clamping effects, especially in nanometer scale films below their relaxation thickness. Here, we demonstrate interfacial decoupling between the ferromagnetic shape memory alloy Ni_2MnGa and MgO via the introduction of a graphene interlayer. X-ray diffraction experiments of Ni_2MnGa films grown by molecular beam epitaxy on bare and graphene terminated MgO(001) show that the constraints of epitaxy are relaxed on graphene terminated surfaces, allowing for multiple epitaxial relationships between substrate and film. This decoupling from graphene allows the Ni_2MnGa to be exfoliated, providing an etch-free method to produce free-standing membranes. Finally, we will discuss the similarities and differences between the three sample types based on magnetization, transport, and structural measurements.

9:30am **NM-TuM1-7 Ferroelectricity at 900 °C in a 1 Unit-Cell-Thick Film***,* ***Yilin Evan Li,*** *R. Steinhardt, M. Holtz,* Cornell University*; P. Silva,* University of California, Berkeley*; Z. Xiao,* Lawrence Berkeley National Laboratory*; R. Ozgur,* University of California, Berkeley*; C. Brooks,* Cornell University*; D. Tenne,* Boise State University*; D. Muller,* Cornell University*; P. Shafer, E. Arenholz,* Lawrence Berkeley National Laboratory*; J. Mundy,* Cornell University*; R. Ramesh,* University of California, Berkeley, Lawrence Berkeley National Laboratory*; D. Schlom,* Cornell University, USA, Leibniz-Institut für Kristallzüchtung, Berlin, Germany

Ferroelectric and multiferroic materials are of great importance for functional devices including low-power non-volatile memories and logic. Proper ferroelectrics, in which spontaneous polarization drives the ferroelectric phase transition, have attracted broad interest in the past few decades. Both theoretical and experimental studies indicated that critical thicknesses exist for proper ferroelectrics. For example, a synchrotron X-ray study revealed that, at room temperature, $PbTiO_3$ films can be structurally ferroelectric with thicknesses down to 3-unit cells [1], which agrees well with theoretical predictions [2]. Unfortunately, far less is known about improper ferroelectrics, where an ordering other than polarization (e.g., structural, charge or spin ordering) drives a phase transition that results in ferroelectricity.

To explore the intrinsic properties of improper ferroelectrics, we have investigated a model system, hexagonal $LuFeO_3$. Phase-pure films of this improper ferroelectric are grown on epitaxial iridium bottom electrodes on YSZ (111) substrates by MBE. Atomic force microscopy (AFM) shows that *h*-$LuFeO_3$ films grow smoothly on the iridium bottom electrode in a layer-by-layer fashion with an RMS roughness of 0.55 nm. X-ray ω-rocking curves are measured to assess the structural quality. The full width at half maximum (FWHM) of 111 Ir peak is 29 arc sec (0.008°) and of the 002 *h*-$LuFeO_3$ peak is 0.12°, narrower than any prior reports [3,4] by factors of about 60 and 2, respectively. Our results show that *h*-$LuFeO_3$ films as thin as 1-unit cell are still ferroelectric at ~900 °C.

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9:45am **NM-TuM1-8 Strange Metal $Ybrh_2Si_2$ Grown by Molecular Beam Epitaxy***,* ***Stefania Isceri,*** Institute of Solid State Electronics, Technische Universität Wien, Austria*; M. Giparakis,* Institute of Solid-State Electronics, Technische Universität Wien, Austria*; E. Bakali,* Insitute of Solid-State Physics, Technische Universität Wien, Austria*; R. Svagera,* Institute of Solid-State Physics, Technische Universität Wien, Austria*; M. Waas,* Institute of Solid State Physics, Technische Universität Wien, Austria*; D. Nguyen,* Institute of Solid-State Physics, Technische Universität Wien, Austria*; H. Detz, W. Schrenk,* Institute of Solid State Electronics, Technische Universität Wien, Austria*; S. Buehler-Paschen,* Insitute of Solid-State Physics, Technische Universität Wien, Austria*; G. Strasser, A. Andrews,* Institute of Solid-State Electronics, Technische Universität Wien, Austria

$YbRh_2Si_2$ is a heavy fermion material [1, 2] crystallizing in the tetragonal $ThCr_2Si_2$-type structure (space group I4/mmm). It has a well-defined quantum critical point [2, 3], a linear-in-temperature strange metal behavior resulting from a dynamical electron localization-delocalization transition [1] and shows superconductivity below 10 mK [4]. $YbRh_2Si_2$ crystal thin films have been recognized as promising for novel applications, such as THz transmission spectroscopy and shot noise detection.

In this work, we study the growth of $YbRh_2Si_2$ on Ge(001) substrates by molecular beam epitaxy (MBE). The substrate chosen is Ge (fcc, a=5.658 Å [5]), due to the low lattice mismatch between its nearest neighbor distance $a'=(a\sqrt{2})/2=$ 4.001 Å and the a lattice parameter of $YbRh_2Si_2$ (a= 4.007 Å, c= 9.860 Å [6]). The thickness of the deposited films is around 60 nm.

Complementary characterization techniques, *in situ* reflection high-energy electron diffraction (RHEED), *ex situ* atomic force microscopy (AFM), x-ray diffraction (XRD), and energy-dispersive x-ray (EDX) spectroscopy, demonstrate that epitaxial crystalline films are obtained.

The crystal structure analysis highlights that small changes in the out-of-plane lattice parameter correlate with the Rh content. In particular, larger values are directly related to higher Rh concentration. XRD φ-scans show that the films have a four-fold symmetry and are rotated by 45° with respect to the substrate. Resistivity measurements show that the 60 nm thin film $YbRh_2Si_2$ exhibit similar characteristics to bulk single crystals.

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