Novel Materials
Room Ballroom A - Session NM-TuM2

Thin Film Membranes

Moderator: Dr. Roman Engel-Herbert, Paul Drude Institute

10:30am **NM-TuM2-12 Doping the Undopable: Hybrid Molecular Beam Epitaxy Growth, n-type Doping, and Field-Effect Transistor using $CaSnO_3$**, ***Fengdeng Liu****, P. Golani, T. Truttmann,* University of Minnesota, USA*; I. Evangelista,* University of Delaware*; M. Smeaton,* Cornell University*; D. Bugallo,* Drexel University*; J. Wen, A. Kamath Manjeshwar,* University of Minnesota*; S. May,* Drexel University*; L. Kourkoutis,* Cornell University*; A. Janotti,* University of Delaware*; S. Koester,* University of Minnesota*; B. Jalan,* University of Minnesota, USA

ABSTRACT: The alkaline earth stannates are touted for their wide band gaps and the highest room-temperature electron mobilities among all the perovskite oxides. $CaSnO_3$ 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 $CaSnO_3$ using hybrid molecular beam epitaxy (hMBE), which provides an adsorption-controlled growth for the phase-pure, epitaxial and stoichiometric $CaSnO_3$ films. By introducing lanthanum (La) as an n-type dopant, we demonstrate the robust and predictable doping of $CaSnO_3$ with free electron concentrations, n, from 3.3×10^{19} cm^{-3} to 1.6×10^{20} cm^{-3}. The films exhibit a maximum room-temperature mobility of 42 cm^2 V^{-1}s^{-1} at n = 3.3×10^{19} 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 $CaSnO_3$ -based field-effect transistors. The transistors show promise for $CaSnO_3$’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 10^6. This work opens the door to future studies on the semiconducting properties of $CaSnO_3$ and the many devices that could benefit from $CaSnO_3$’s exceptionally wide band gap.

10:45am **NM-TuM2-13 Controlling the Balance between Remote, Pinhole, and van der Waals Epitaxy of Heusler Films on Graphene/Sapphire**, ***Taehwan Jung****, Z. LaDuca, D. Du, S. Manzo, K. Su, X. Zheng, V. Saraswat,* University of Wisconsin - Madison*; J. McChesney,* Argonne National Lab*; M. Arnold, J. Kawasaki,* University of Wisconsin - Madison

Remote epitaxy is promising for the synthesis of lattice-mismatched materials, exfoliation of membranes, and reuse of expensive substrates. However, clear experimental evidence of a remote mechanism remains elusive. Alternative mechanisms such as pinhole-seeded epitaxy or van der Waals epitaxy can often explain the resulting films. Here, we show that growth of the Heusler compound GdPtSb on clean graphene/sapphire produces a 30° rotated (R30) superstructure that cannot be explained by pinhole epitaxy [1]. With decreasing temperature, the fraction of this R30 domain increases, compared to the direct epitaxial R0 domain, which can be explained by a competition between remote versus pinhole epitaxy. Careful graphene/substrate annealing and consideration of the relative lattice mismatches are required to obtain epitaxy to the underlying substrate across a series of other Heusler films, including LaPtSb and GdAuGe. The R30 superstructure provides a possible experimental fingerprint of remote epitaxy, since it is inconsistent with the leading alternative mechanisms.

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[1] D. Du, et. al. *Nano Lett.*2022, 22, 21, 8647–8653

11:00am **NM-TuM2-14 Synthesis of Flexomagnetic GdAuGe Membranes via Van Der Waals Epitaxy on Graphene Terminated Germanium**, ***Zachary LaDuca****, S. Manzo, T. Jung, T. Samanta, K. Su, M. Arnold, J. Kawasaki,* University of Wisconsin - Madison

The ability to synthesize freestanding membranes of crystalline materials is critical for advancing the understanding of strain and strain gradient effects on materials properties. A common approach for the synthesis of freestanding membranes is epitaxial growth on graphene terminated substrates, where direct bonding between the film and substrate is prevented, allowing for exfoliation of the film from the substrate. However, challenges with wetting on the low surface energy graphene frequently prevents the growth of smooth epitaxial films. Here we demonstrate high quality growth of the flexomagnetic Heusler compound GdAuGe on graphene terminated germanium substrates using a cold-seeded growth approach. Scanning electron microscopy, atomic force microscopy, and x-ray diffraction experiments illustrate the tradeoffs resulting from growth at high and low temperatures and confirm that multistep growth approach comprised of a few-nanometer, amorphous deposition at room temperature followed by an anneal and subsequent growth at elevated temperature result in smooth, highly ordered films. This improved morphology and crystallinity enhances the ability to control the strain state in rippled GdAuGe membranes.

11:15am **NM-TuM2-15 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.

References

1. Graf, Tanja, et al. "Simple rules for the understanding of Heusler compounds." Progress in solid state chemistry 39.1 (2011): 1-50.

2. Kawasaki, Jason K. "Heusler interfaces—Opportunities beyond spintronics?." APL Materials 7.8 (2019): 080907

3. Du, Dongxue, et al. "Epitaxy, exfoliation, and strain-induced magnetism in rippled Heusler membranes." Nature Communications 12.1 (2021): 1-7

11:30am **NM-TuM2-16 Growth Mechanism of $SrTiO_3$ on a Graphene-covered Substrate Using Hybrid MBE**, ***Sooho Choo****,* University of Minnesota, Republic of Korea*; 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 $SrTiO_3$ nanomembranes on $SrTiO_3$ (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 $SrTiO_3$ 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.

11:45am **NM-TuM2-17 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.

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