NAMBE

Room Cummings Ballroom - Session NAMBE1-TuA

Oxides I

Moderator: Matthew Brahlek, Oak Ridge National Laboratory

2:00pm **NAMBE1-TuA-1 Plasma Assisted Molecular Beam Epitaxial Growth of β-ga2o3 (100) Thin Films on Mgo(100) Substrates**, ***Seth Hibbert***, *R. Reeves, M. Allen,* University of Canterbury, New Zealand

Gallium oxide (Ga2O3) is a transparent conducting oxide undergoing active research for its many potential uses in power electronics. It has a larger breakdown field (~8MV/cm) and a larger bandgap (4.6 – 4.9 eV) than silicon carbide, a popular material for power electronics. Currently five phases of gallium oxide have been identified, these have been labeled the α, β, γ, δ, and ε phases. Of these phases, β-Ga2O3 is the most stable and thus the most commonly grown by epitaxial methods. β-Ga2O3 has a monoclinic structure (figure 1a in supplemental document) and can be either conducting or insulating depending on the conditions of the crystal growth. Due to the monoclinic structure of β-Ga2O3, the asymmetric unit cell leads to anisotropies in the structural, electrical and optical properties. The (-201) orientation of β-Ga2O3 can be easily grown on a variety of materials with c-plane sapphire being a common substrate for heteroepitaxy. The (100) orientation, however, has seen little research due to a relatively slow growth rate when grown heteroepitaxially via molecular beam epitaxy. In the work presented here, β-Ga2O3 thin films of ~90nm thickness, were grown on MgO(100) substrates using plasma-assisted molecular beam epitaxy. The films exhibited very smooth, epitaxial surfaces as measured by in-situ RHEED and atomic force microscopy after growth (figure 2). X-ray diffraction showed crystalline thin films having the (100) orientation and figure 3 shows a comparison with a (-201) oriented film grown under similar conditions on a c-plane sapphire substrate. It is apparent the (100) oriented film has excellent crystal quality with FWHM of the (400) peak measured as 0.280 degrees compared to 0.397 degrees for the (-402) peak. The films showed high transmission in the optical region with indications the (100) oriented film has a slightly lower bandgap energy (figure 4). While it is known epitaxial strain can change bandgap values, the excellent crystal quality suggests anisotropy of lattice directions is the probable cause.

2:15pm **NAMBE1-TuA-2 Progresses Towards Production-Worthy Epitaxy of $BaTiO_3$ and $SrTiO_3$ Perovskites on Si(001) Substrates**, ***Mark O'Steen***, Veeco Instruments Inc.*; M. Baryshnikova, G. Croes,* IMEC, Belgium*; Y. Wang, S. Farrell, G. Sundaram,* Veeco Instruments Inc.*; C. Merckling,* IMEC, Belgium

Barium titanate ($BaTiO_3$ or BTO) exhibits a large, linear electro-optic effect (or Pockels effect), making it potentially useful for a variety of applications including datacom/telecom, LIDAR, and quantum computing. To be practical for such applications, it is advantageous to integrate BTO to a substrate material such as Si, which is readily available in large wafer sizes at low cost. Since BTO is chemically unstable in contact with Si, this integration requires the use of an intermediate strontium titanite ($SrTiO_3$ or STO) template.

BTO (using an STO buffer) can be grown on Si by several techniques; however, so far, only conventional, solid-source MBE (SS-MBE) produces material exhibiting a high Pockels coefficient due to higher crystalline quality. SS-MBE has only a small process window to produce high quality material due to the challenges in maintaining cation stoichiometry in STO and BTO. Alternatively, hybrid MBE (*h*MBE), in which the precursor Ti-tetraisopropoxide (TTIP) is used as the Ti source, has a much larger process window to achieve stoichiometric STO and BTO perovskites due to the presence of self-regulating growth regimes.

In this study, we investigate the epitaxy and characteristics of STO and BTO thin films on Silicon by *h*MBE approach. A novel precursor delivery system, designed and optimized for use in *h*MBE, enabled optimizing the growth processes and methodologies for consistent deposition results. Epitaxial materials are characterized using several techniques including TEM, XRD, XRF, RBS, AFM, and electro-optical measurements. Finally, SS-MBE and *h*MBE growth of BTO will be compared and the growth characteristics and tradeoffs of the two techniques will be discussed. In particular, the potential and challenges for scaling to large diameter substrates and to develop the robust processes needed for manufacturing will be highlighted.

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2:30pm **NAMBE1-TuA-3 Epitaxial Growth of Si-doped $(Al, Ga)_2O_3$ Films by Hybrid MBE**, ***Zhuoqun Wen***, *E. Ahmadi,* University of Michigan

The oxidation of Al metal source in the conventional oxide MBE requires frequent opening of the system to reload materials. A regular crucible filled with Al element can be depleteing within 10 growth cycles. Additionally, oxidation of silicon source in these systems makes controlled and uniform Si-doping of Ga_2O_3 challenging. To overcome these challenges, we are using a hybrid MBE which uses conventional effusion cells for Ga and Ge, and gas sources to supply Al and Si. In this talk, we will give an overview of our custom-built gas delivery system with dilluted disilane as Si source and TTBAl as Al source, and present our latest results on achieving AlGaO films with different Al compositions (1%-25%) by controlling TTBAl flow and Ga_2O_3 films with Si doping concentrations ranging from 4×10^{16} cm^{-3} to 2×10^{19} cm^{-3} concentrations with different disilane flows. Fig.1 shows the β-$(Al_xGa_{1-x})_2O_3$ grown by TTBAl at different flows at 525 ℃, Ga beam-equivalent pressure (BEP) at 10^{-8} Torr, and O plasma 410W, 2 sccm. By increasing the TTBAl beam-equivalent pressure (BEP) from 1.1×10^{-7} Torr to 4.3×10^{-7} Torr, the Al incorporation increases from 1.8% to 14.9%. The Al composition can be further increased up to 25% by controlling the TTBAl flow and increasing the growth temperature. The electron properties of Si doped β-$(Al_xGa_{1-x})_2O_3$ grown by disilane and TTBAl will be studied.

2:45pm **NAMBE1-TuA-4 Correlated Phase Diagram Tunable by Structural Layering in Square-Planar Nickelates**, ***Grace Pan***, *D. Ferenc Segedin, S. TenHuisen,* Harvard University*; L. Bhatt,* Cornell University*; H. LaBollita,* Arizona State University*; A. Jiang,* Harvard University*; Q. Song,* Cornell University*; A. Turkiewicz,* Harvard University*; H. Paik,* University of Oklahoma*; C. Brooks, M. Mitrano,* Harvard University*; B. Goodge,* Max Planck Institute for Chemical Physics of Solids*; A. Botana,* Arizona State University*; J. Mundy,* Harvard University

Since the discovery of superconductivity in Sr-doped $NdNiO_2$[1], the identification of independent tuning parameters has further expanded the materials and correlated phases in this family of nickel-based compounds. One such knob has been the structural layering, which has been used to realize a superconducting phase in the five-layer square-planar nickelate $Nd_6Ni_5O_{12}$[2]. In the square-planar $Nd_{n+1}Ni_nO_{2n+2}$ compounds the primary role of layering is to provide a nominal doping level of $1/n$ electrons per nickel site. Nonetheless, one might expect substantial modification of the electronic structure, correlation effects, or even nominal doping levels from intentional restructuring of the atomic lattice. Here, we present the correlated phase diagram of the layered $Nd_{n+1}Ni_nO_{2n+2}$ compounds. We observe a superconducting dome of clear resemblance to the chemically doped infinite-layer nickelates, indicating that structural layering can generally tune the electronic doping levels. This furthermore supports the potential universality of doping-dependent superconducting phases[3–6]. We discuss the role of layering beyond providing rigid doping shifts, including effects on correlations, spin fluctuations, and disorder.

[1]Li, D. *et al.* Superconductivity in an infinite-layer nickelate. *Nature* **572**, 624–627 (2019). [2]Pan, G. A. *et al.* Superconductivity in a quintuple-layer square-planar nickelate. *Nat. Mater.* **21**, 160–164 (2022). [3]Lee, K. *et al.* Linear-in-temperature resistivity for optimally superconducting (Nd,Sr)NiO_2. *Nature* **619**, 288–292 (2023). [4]Keimer, B. *et al*. From quantum matter to high-temperature superconductivity in copper oxides. *Nature* **518**, 179–186 (2015). [5]Johnston, D. C. The puzzle of high temperature superconductivity in layered iron pnictides and chalcogenides. *Adv. Phys.* **59**, 803–1061 (2010). [6]Cao, Y. *et al.* Unconventional superconductivity in magic-angle graphene superlattices. *Nature* **556**, 43–50 (2018).

3:00pm **NAMBE1-TuA-5 Synthesis of Layered Square-planar Lanthanum Nickelate Thin Films, $La_{n+1}Ni_nO_{2n+2}$**, ***Dan Ferenc Segedin***, *G. Pan, A. Turkiewicz, A. Jiang, C. Brooks, J. Mundy,* Harvard University

The layered square-planar nickelates, $R_{n+1}Ni_nO_{2n+2}$ (R = trivalent rare-earth cation and n > 1), are an exciting platform to tune the properties of superconducting square-planar nickelates via dimensionality [1-2]. To date, superconducting $Nd_{n+1}Ni_nO_{2n+2}$ films have been achieved only on $NdGaO_3$ due to the competing requirements for the synthesis of the precursor Ruddlesden-Popper, $Nd_{n+1}Ni_nO_{3n+1}$, and reduction to the square-planar phase [3]. On $SrTiO_3$, the high tensile strain in the precursor phase, $Nd_{n+1}Ni_nO_{3n+1}$, leads to a high density of stacking faults [3]. Improved crystalline quality and an enhanced superconducting transition temperature have been achieved in (Nd,Sr)NiO_2 on LSAT by lowering the tensile strain in the parent perovskite phase [4]. Here, we investigate the role of epitaxial strain in the synthesis of Lanthanum-based layered nickelates on $NdGaO_3$. We demonstrate a substantially higher crystalline quality in $La_{n+1}Ni_nO_{3n+1}$ on $NdGaO_3$ than $Nd_{n+1}Ni_nO_{3n+1}$, due to the lower

tensile strain. Finally, we discuss the reduction to the square-planar phase on $NdGaO_3$.

[1] Li, D. *et al.* Superconductivity in an infinite-layer nickelate. *Nature* 572, 624–627 (2019).

[2] Pan, G. A. *et al.* Superconductivity in a quintuple-layer square-planar nickelate. *Nat Mater* 1–5 (2021).

[3] Ferenc Segedin, D. *et al.* Limits to the strain engineering of layered square-planar nickelate thin films. *Nat. Commun.* 14, 1468 (2023).

[4] Lee, K. *et al.* Linear-in-temperature resistivity for optimally superconducting (Nd,Sr)NiO2. *Nature* 619, 288–292 (2023).

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