## MBE

## Room Max Bell Auditorium - Session MBE-MoA

## Novel Materials and Oxides/2D Materials and Characterization

**Moderators:** Joshua Zide, University of Delaware, Geoffrey Gardner, Microsoft Research

1:30pm **MBE-MoA-1 Epitaxial Stabilization of Monoclinic $Fe_2O_3$ on β-$Ga_2O_3$, *John Jamison,*** *B May, R Myers,* The Ohio State University

There is a surge in interest in β-$Ga_2O_3$ because of its thermodynamic stability, wide bandgap, and excellent figures of merit for high power devices. Additionally, β-$Ga_2O_3$ is quite similar to structures found in magnetic 3d transition metal oxides, which also consist of networks of tetrahedra and octahedra. Specifically, there are several naturally occurring $Fe_2O_3$ phases, and $Fe^{3+}$ and $Ga^{3+}$ cations exhibit similar ionic radii. However, there are no $Fe_2O_3$ phases the same monoclinic structure as β-$Ga_2O_3$. Here, we investigate the possibility of using epitaxial strain to stabilize a new form of monoclinic $Fe_2O_3$ (m-$Fe_2O_3$) on β-$Ga_2O_3$. Molecular beam epitaxy was used to grow a sample on a (010) β-$Ga_2O_3$ substrate, consisting of multiple Fe depositions of increasing amounts separated by 10 nm β-$Ga_2O_3$ spacers (Fig. 1(a)). Reflection high energy electron diffraction (RHEED) shows the preservation of the β-$Ga_2O_3$ overgrowth quality even for quite high m-$Fe_2O_3$ thicknesses. High resolution X-ray diffraction of the structure shows distinct thickness fringes and superlattice peaks. High resolution scanning transmission electron microscopy confirms that the overgrown β-$Ga_2O_3$ remains high quality after multiple Fe containing layers. The high Fe regions also show the same crystal structure as β-$Ga_2O_3$, i.e. m-$Fe_2O_3$. The optical and magnetic properties of this new form of $Fe_2O_3$ will also be discussed.

1:45pm **MBE-MoA-2 Homo- and Hetero-epitaxial Growth of β -$Ga_2O_3$ Thin Films by Molecular Beam Epitaxy, *Neeraj Nepal,*** *D Katzer, B Downey, V Wheeler, M Hardy, D Storm, D Meyer,* U.S. Naval Research Laboratory

β-$Ga_2O_3$ is emerging as a next generation ultra-wide bandgap semiconductor (UWBGS) material with a bandgap of 4.5-4.9 eV with applications in high-power/temperature electronics devices [1-3]. A distinct advantage of β-$Ga_2O_3$ over other UWBGS materials is availability of inexpensive large area bulk substrates synthesized by melt growth techniques at atmospheric pressure [2]. Homoepitaxial growth on bulk substrates offers the potential of low defect density films for vertical power devices. Despite the crystalline quality advantages of homoepitaxy, future device performance is anticipated to be limited by the low thermal conductivity of β-$Ga_2O_3$, so one approach to improve thermal performance is through hetero-epitaxy of β-$Ga_2O_3$ on a high thermal conductivity substrate such as SiC. For these reasons, both homo- and hetero-epitaxial growth of $Ga_2O_3$ films are of general interest to be investigated.

Figure 1. X-ray diffraction measurements of epitaxial β-$Ga_2O_3$ on on-axis 4H-SiC (blue, 86 nm thick), c-sapphire (black, 126 nm) and (010) β-$Ga_2O_3$ (purple, ~200 nm).

In this paper, we report homo- and hetero-epitaxial growth 100-200 nm thick β-$Ga_2O_3$ thin films on sapphire, (010) β-$Ga_2O_3$ and 4H-SiC substrates by molecular beam epitaxy (MBE) at 650 °C and compare the impact of substrate. The growth parameter space including thermocouple-measured growth temperature, relative Ga flux, and oxygen plasma flow were varied to grow β-$Ga_2O_3$ films on c-plane sapphire substrates. Figure 1 shows about 86-130nm thick single phase MBE-grownβ-$Ga_2O_3$ films that are insulating with relatively low surface roughness. The heteroepitaxial films have rocking curve full-width-at-half-maximum of 256 and 720 arc-sec on sapphire and SiC, respectively. In this paper we will discuss MBE growth parameter space optimization of β-$Ga_2O_3$ on sapphire and the structural, morphological, and electrical properties of MBE grown β-$Ga_2O_3$ thin films on (010) $Ga_2O_3$ and SiC.

[1] H.H. Tippins, Physical Review **140**, A316 (1965).

[2] K. Akito et al., Jpn. J. Appl. Phys. **55**, 1202A2 (2016).

[3] J.Y. Tsao, Adv. Electron. Mater. **4**, 1600501 (2018).

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2:00pm **MBE-MoA-3 Epitaxial Growth and Electronic Structure of Semiconducting Half-Heusler FeVSb, *Estiaque Haidar Shourov,*** *P Strohbeen, D Du,* University of Wisconsin Madison*; J McChesney,* Argonne National Laboratory*; A Janotti,* University of Delaware*; J Kawasaki,* University of Wisconsin Madison

Although FeVSb is experimentally known as a high figure of merit thermoelectric material [1], challenges associated with fabricating high quality single crystalline samples have hampered a fundamental understanding of its electronic structure [2]. For example, while recent first-principles calculations show that the DFT band gap is highly sensitive to the choice of exchange and correlation functional (LDA predicts 0.36 eV and HSE predicts 1.45 eV [3,4]), its experimental bandgap is not known. Here, we demonstrate the epitaxial growth of FeVSb on MgO (001) by solid source molecular beam epitaxy. The single crystalline phase and epitaxial alignment were confirmed by reflection high-energy electron diffraction (RHEED) and X-ray diffraction (Fig. 1). By tuning the growth temperature and relative Sb flux, we find that FeVSb can be grown in a self-limiting, Sb adsorption-controlled window. Further tuning of the Fe:V flux ratio (by QCM and RBS measurements) then allows us to grow stoichiometric FeVSb. Our angle-resolved photoemission spectroscopy (ARPES) reveals that the band gap of FeVSb is at least 0.6 eV (Fig. 2), much larger than the 0.36 eV band gap predicted by LDA calculations, and the measured valence band width is smaller than the LDA width by nearly a factor of two. We present further calculations and experimental results to decipher this discrepancy.

2:15pm **MBE-MoA-4 Growth of Candidate Polar Metal Hexagonal Half Heuslers, *Dongxue Du,*** *J Kawasaki,* University of Wisconsin Madison

Hexagonal half Heuslers (space group P63mc, LiGaGe-type structure) have recently been proposed as a new hyper-ferroelectric materials system. In these ABC intermetallic compounds, layers of B and C atoms form a buckled honeycomb lattice, resulting in a net polarization along the c axis that is robust against the depolarizing field [1]. Moreover, many of these compounds exhibit large Rashba coefficients and magnetic order, making them a promising system for finding multiferroics [2]. However, demonstration of these properties and understanding the mechanism for hyper-ferroelectricity require high quality epitaxial films, which haven't yet been demonstrated.

Here we demonstrate the first epitaxial growth of LaPtSb and LaAuGe. These compounds are grown on *c*-plane Al2O3 by solid source MBE, using an Sb adsorption controlled window for LaPtSb, and by flux matching for LaAuGe. Symmetric 2theta-omega (Fig. 1) and in-plane rotation (phi scans Fig. S1) x-ray diffraction measurements confirm that the films are epitaxial and single crystalline, with the desired LiGaGe-type buckled hexagonal structure. RHEED patterns confirm well-ordered surfaces with surface reconstructions. Through a combined analysis of cross sectional TEM, second harmonic generation (SHG), and angle-resolved photoemission spectroscopy (ARPES) measurements, we are exploring the coupling of polar distortions to electronic structure and magnetism in these materials.

We gratefully acknowledge support from the ARO YIP (W911NF-17-1-0254, Dr. Chakrapani Varanasi)

[1] Kevin F, Phys. Rev. Lett. **112**,127061 (2014).

[2] Awadhesh Narava, Phys. Rev. B. 92.220101® (2015).

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2:30pm **MBE-MoA-5 Optimizing Cesium Antimonide Photocathode Performance Using Real-time *In-situ* Monitoring of Photoemissive Properties, *Mark Hoffbauer,*** Los Alamos National Laboratory*; S Celestin,* Northeastern University*; V Pavlenko, F Liu, N Moody,* Los Alamos National Laboratory

Alkali antimonide semiconductor photocathodes like $Cs_3Sb$ or $K_2CsSb$ are promising electron sources for use in next-generation light sources such as advance Free Electron Lasers (FEL) due to their high quantum efficiency in the visible spectrum, short response time, good lifetime, and the ability to produce high-brightness beams with a relatively low emittance. Traditional methods of alkali antimonide photocathode growth, sequential deposition, dates back to 1960s when quantum efficiency (QE, number of electrons emitted per incident photon) was prioritized over other parameters. Sequential deposition allows fabrication of acceptable photocathodes, but the crystalline quality is always low and surface roughness is high. Photocathodes for next-generation light sources must be smooth and have high crystalline quality in order to generate "colder" electron beams (low emittance). Recently, a co-deposition alkali antimonide growth technique was introduced that mimics MBE but, lacking meaningful feedback, fails to achieve acceptable control over the growth parameters. Understanding the

correlation between growth conditions (substrate temperature, fluxes of alkali metals and Sb), film characteristics (stoichiometry, crystal structure, roughness), and cathode metrics (QE, emittance, and response time) is necessary for developing reliable growth procedures for alkali antimonide photocathodes.

We have performed detailed studies on the growth of $Cs_3Sb$ photocathodes using a new MBE growth capability at LANL with better control of the growth kinetics for optimizing photoemissive properties. The growth capability utilizes a Sb effusion cell and a custom-built Cs evaporator assembly for precise control of their fluxes. A multiple wavelength laser assembly is used to illuminate the surface of the growing cathode and measure the spectral response (QE vs. wavelength) in real-time. These *in situ* measurements were used to tune the growth parameters (fluxes, temperature, etc.) and attain the spectral response indicative of a stoichiometric $Cs_3Sb$ film. Our results demonstrate the ability to fine tune the Sb and Cs fluxes in a co-deposition film growth mode and improve the overall spectral response. Improved photoemissive properties can be correlated with initiating the film growth under conditions for forming $Cs_3Sb$ at the earliest stages and maintaining the film stoichiometry throughout the growth. These films can be grown over a range of substrate temperatures and show excellent long term photoemission stability. The relationship between the optimized growth conditions and the photocathode emittance properties will also be discussed.

2:45pm **MBE-MoA-6 Optically Triggered Semiconductor Hyperbolic Metamaterial for Controlled Single Photon Emission**, ***Kurt Eyink***, *H Haugan, V Pustovit, A Urbas,* Air Force Research Laboratory

Quantum photonics opens doors for applications in sensing, data transfer, quantum computing. A key technological hurdle is a system for controlled single photon emission. Hyperbolic metamaterials, composed of metallic building blocks embedded in dielectric media control emission lifetime by modifying the photon density of states. However, limited previous efforts have explored the transient modification of metamaterials to control emission. Antimony-based semiconductor hyperbolic metamaterials (SHMMs) offer a route to modulation of these resonances at the mid-infrared (IR) wavelength range, which would modulate emission. In this work we propose to demonstrate SHMMs such as InAsSb alloys, and InAs/InAsSb stacks embedded with dielectric GaSb media in which a transient carrier concentration will be generated through optical pumping. Modelling of these films show that optical concentration of $10^{19}$-$10^{20}$ e-h/cm$^3$ would generate responses in the IR range. This transient excitation of the SHMM would enable triggered single photon emission as well as optical gating and modulation. Calculations show 2-3 orders of magnitude change in the photon density of states predicting dramatic changes in the emission rate. If successful, this study would establish a new platform for deterministic single photon emission that would be integrable into opto-electronic platforms and dramatically advance optical quantum technologies. This initial study will serve as an ideal test bed for next-generation plasmonic architectures, where optically engineered metals can be integrated with a loss-less dielectrics to explore the ultimate limits of plasmonic devices.

3:30pm **MBE-MoA-9 Epitaxy of *M*/graphene/Ge (*M* = Fe, Sb) Heterostructures: Testing the Limits of Remote Heteroepitaxy**, ***Patrick J. Strohbeen***, *E Shourov, V Saraswat, D Du, M Arnold, J Kawasaki,* University of Wisconsin Madison

It was recently demonstrated through the creation of GaAs/graphene/GaAs (001) heterostructures that monolayer graphene may act as a general platform for epitaxy through an atomic barrier[1]. However, the underlying mechanisms of "remote epitaxy" and its generalization to other material systems, e.g. transition metal compounds or oxides, remains unclear. Here, using *M*/graphene/Ge (*M* = transition metal or Sb) as a model system we (1) explore the limits of the remote epitaxy mechanism and (2) demonstrate that single layer graphene is also an excellent solid state diffusion barrier.

In systems containing more volatile species (*M* = Sb) we have found that carefully controlling growth kinetics both via substrate temperature and the cracked Sb species enables growth of nearly single oriented Sb/graphene/Ge (111) heterostructures. The resultant films are readily exfoliated using scotch tape (Fig. 1). In contrast, we find that when *M* = Fe, the films grown on graphene are polycrystalline regardless of substrate temperature and Ge orientation. Though we still show that the polycrystalline films are easily exfoliated. These results suggest that volatile adatom species may be a required ingredient for "remote epitaxy". With *M* = Fe we also show that graphene behaves as an excellent solid state diffusion barrier as supported by our in-situ x-ray photoemission spectroscopy (XPS) measurements as a function of annealing temperature. Our work suggests highly flux dependent growth mechanisms due to both the difficulty in wetting the graphene monolayer as well as the high in-plane diffusivity on graphene. The effects of growth conditions as well as the effectiveness of graphene as a solid state diffusion barrier will be discussed.

[1] Y. Kim *et al.*, Nature, **544**, 7650, Apr. 2017.

3:45pm **MBE-MoA-10 Molecular Beam Epitaxy of $MoSe_2$ Directly on Si**, ***Elline Hettiaratchy***, *B May, R Myers,* The Ohio State University

Van der Waals bonding relaxes the constraints of lattice matching, making two-dimensional (2D) transition metal dichalcogenides attractive in the field of epitaxy. Recently, molecular beam epitaxy (MBE) of $MoSe_2$ has been demonstrated on AlN and GaAs [1,2] but, to our knowledge, the direct growth of $MoSe_2$ on Si by MBE has not yet been reported. Here we investigate the early stages of 2D nucleation of $MoSe_2$ grown on Si by MBE in order to pursue tunable grain size. In principle, large area $MoSe_2$ (0001) will grow on Si (111) with two domain orientations. After removing the oxide by a Piranha etch, Mo and Se are codeposited on Si (111). At constant flux ratios the 2D nucleation rate is controllable with substrate temperature, as confirmed using x-ray diffraction and atomic resolution force microscopy (AFM). Film morphology and structural quality in the high temperature, Mo-limited, regime of $MoSe_2$ growth using high Se vapor overpressures will be discussed.

4:00pm **MBE-MoA-11 Atomic Scale Characterization Showing Kinetic Compositional Instability and Phase Separation in MBE-grown InGaAs**, ***Michael Yakes***, *M Twigg, N Kotulak, N Mahadik, S Tomasulo,* U.S. Naval Research Laboratory

Phase separation in III-V semiconductor alloys remains a problem that limits the performance and quality of electronic materials. As the first stage in a comprehensive program addressing this issue, we have begun investigating an alloy system in which only the group III elements differ: InGaAs. Lattice-matched InGaAs alloy films were deposited at three temperatures (400, 450, and 500 ºC) by MBE on a (001) InP substrate. Using TEM, APT and XRD, we have found phase separation in all three growths to varying degrees.

According to the kinetic compositional instability (KCI) model developed by Glas [1], the critical temperature for kinetic spinodal phase separation in InGaAs is 814 ºC, a temperature well above the growth temperatures commonly used in InGaAs growths. Our XTEM measurements found that the amplitude of composition modulations averaged over the thickness of the XTEM sample are 0.7, 0.5, and 0.4 atomic percent for the growth temperatures 400, 450, and 500 ºC, respectively. APT indicates that the amplitude of composition modulation for the 400 ºC growth is approximately 1 atomic percent, a value that compares favorably with the 0.7 atomic percent measured by XTEM.

We have used KCI theory to evaluate the average amplitude of composition modulation for a given growth temperature by integrating the KCI vertical composition profile over thickness. The KCI model explicitly addresses the kinetics of the volatile near-surface region of the film, where surface undulations driven by surface diffusion introduce the kinetic component that undermines compositional stability beyond the point dictated by thermodynamics alone. This analysis finds that the kinetically unstable layer is approximately 2 nm thick when the lateral composition modulation wavelength is 3 nm. The thickness of this kinetically unstable layer corresponds to features marking both lateral and vertical composition, providing good evidence for the kinetic origins of the observed phase separation in the material.

4:15pm **MBE-MoA-12 Investigation of Gallium-related Defects in III/V Epitaxial Layers**, ***Yossi Cohen***, *O Klin, I Grimberg, N Yaron, E Weiss,* SemiConductor Devices Company, Israel

III/V materials are among the most common materials for the production of IR detectors. Gallium and indium droplets in MBE grown material are long-time known to be a major cause for decrease in detector operability (percentage of good pixels). In this work we present the investigation of gallium-related defects formed in an InAs/GaSb strained layer superlattice (SLS) structure. The SLS structure allows us to understand, in details, the mechanism in which the defect is formed and evolves.

Based on TEM analysis shown in figure 1 and other results (AFM, SEM, cross-section EDS mapping), we conclude that after a gallium droplet reaches the epilayer, it etches and dissolves several hundreds of nanometers below its landing point. Gallium from the droplet migrates

sideways on the surface (at different rates along the [01-1] and [011] directions) for few microns, increasing temporarily the growth rate of the epilayer around the droplet and changing its composition (figure 1c). The incoming fluxes together with the dissolved material enrich the Ga droplet with Sb, As and In. In our growth conditions, the Ga droplet top surface solidifies, forming a GaAs shell [1]. High threading dislocation density is formed in the InAs-GaSb SLS grown on such surface due to the large mismatch between the SLS and the GaAs shell. The InGaAsSb solution inside the droplet separates, at some point, to the thermodynamically stable InSb and GaAs phases. In some parts of the core we see pure gallium that probably solidifies only when the sample is cooled down.

4:30pm **MBE-MoA-13 Acoustic Nanostructures for Charge Carrier Confinement in GaAs/$Al_xGa_{1-x}As$ Multiple Quantum Wells**, ***Kevin Vallejo***, *C Schuck, T Garrett,* Boise State University*; Z Hua, D Hurley,* Idaho National Laboratory*; P Simmonds,* Boise State University

Quantum confinement of charge carriers in semiconductors is at the heart of next generation energy conversion technologies, as well as new encryption and computation paradigms. We propose a novel approach that uses picosecond-duration surface acoustic phonon pulses to produce lateral carrier confinement (2D and 3D confinement) in III-V (i.e. polar) semiconductor quantum wells. Strain generated by the phonon pulses varies with depth below the sample surface (Fig. 1), locally deforming the valence and conduction bands to produce lateral confinement in the plane of a quantum well. This approach offers the prospect of continually modifying confinement in a manner that can be externally controlled. Using molecular beam epitaxy, we grew the GaAs/AlGaAs structure consisting of three quantum wells of width 5, 7, and 10 nm, buried beneath the sample surface at depths of 14, 49, and 112 nm respectively. These wells are positioned so as to coincide with different conditions of shear strain and dilatation, and hence piezoelectric field strength. We will present results from preliminary studies showing carrier transport at the speed of sound in the sample with extended lifetimes due to acoustic confinement. This approach could find useful applications in nanocircuitry.

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