Novel Materials
Room Ballroom A - Session NM-MoM2

Nitrides

Moderator: Dr. Lutz Geelhaar, Paul-Drude-Institut für Festkörperelektronik Leibniz-Institut im Forschungsverbund Berlin

10:15am **NM-MoM2-11 Molecular Beam Epitaxy of Binary and Ternary Manganese and Chromium Nitrides**, *K. Vallejo, K. Gofryk,* Idaho National Laboratory*; S. Gutierrez-Ojeda,* Universidad Nacional Autónoma de México*; G. Cocoletzi,* Benemérita Universidad Autónoma de Puebla, Mexico*;* ***Brelon May,*** Idaho National Laboratory

Transition metal nitrides have exceptional properties and are used in a wide variety of electrochemical, structural, photochemical, and plasmonic applications. Among these compounds Mn- and Cr- nitrides have shown exceptional potential for magnetic sensing and spintronics. The Mn_xN_y system is complex with several different metastable phases both predicted and experimentally realized. Cr_xN_y has two primary phases, cubic (CrN) and hexagonal (Cr_2N), which exhibit desirable mechanical, thermal, wear, anti-corrosion, thermoelectric properties. Recent studies have provided valuable insights into the growth and formation of phases of both materials using various vapor deposition techniques. However, there are conflicting reports on the electrical and magnetic properties of Cr_xN_y which could be attributed to impurities, nitrogen vacancies, substrate effects, and strain. This controversy calls for a more detailed study and preparation of high-quality monocrystalline CrN to investigate he intrinsic physical properties. This study uses molecular beam epitaxy to synthesize epitaxial thin films of different Mn-N and Cr-N phases. The electrical and magnetic properties of these films are investigated with the rocksalt MnN and CrN both showing metallic behavior, with the latter showing a magnetic transition ~280K. However, when combining these materials at similar growth conditions, instead of maintaining the rocksalt structure, a new ternary cubic phase of Mn_xCr_yN is obtained which shows narrow-gap semiconducting behavior. This work presents an avenue for the epitaxial integration of metallic, magnetic, and semiconductor materials.

10:30am **NM-MoM2-12 Achieving Atomically Ordered GaN/AlN Quantum Heterostructures: The Role of Surface Polarity**, ***Yuanpeng Wu***, *P. Zhou, Y. Xiao, K. Sun, D. Wang, P. Wang, Z. Mi,* University of Michigan, Ann Arbor

A central goal of modern material physics and nanotechnology is the control of materials and their interfaces to atomic scales. However, for interfaces between polar layers, this goal is thwarted by the atomic substitution process among cations with different ionicities. In traditional semiconductor heterostructures, such as InAs/GaAs, Si/Ge, AlGaN/GaN and ABO_3 perovskites, diffusive interfaces have been widely observed, which deteriorates the performance of electronic and optoelectronic devices. Interfacial diffusion also prohibits achieving atomically ordered quantum heterostructures for applications such as quantum light sources and sensors. The studies on the origin of interfacial diffusion are often compounded by factors such as various synthesis parameters, available epitaxial substrates, strain distribution and surface reconstruction while a vital solution for achieving a perfect heterointerface remains elusive.

In this work, we discovered a strong dependence of interfacial diffusion on surface polarity in GaN/AlN quantum heterostructures. Atomically ordered quantum interface can be readily synthesized on the semipolar plane instead of the conventional c-plane of GaN/AlN heterostructures. The underlying mechanism of this dependence is explored through first-principles density functional theory calculations and it is found that the chemical bonding configurations at the semipolar plane can effectively eliminate the cation substitution process, which leads to an atomic sharp interface. The near-perfect interface quality ensures extreme quantum confinement and superior optical properties including record-high internal quantum efficiency of ~75% in the deep ultraviolet wavelength regime. We developed a scalable and robust fabrication method and demonstrated that electroluminescence energies of interdiffusion-free GaN are free from the quantum-confined Stark effect. In addition, we demonstrated a unique strategy of controlling surface polarities through different strain relaxation mechanisms in a core-shell nanostructure platform. This work provides, for the first time, a viable path for the synthesis of interdiffusion-free polar quantum heterostructures, which is paramount for high-performance devices across various material platforms.

10:45am **NM-MoM2-13 Epitaxial Cubic Boron Nitride Grown by Ion Beam-Assisted Molecular-Beam Epitaxy on Diamond**, ***David Storm***, *S. Maximenko, A. Lang, N. Nepal, T. Feygelson, B. Pate, D. Meyer,* US Naval Research Laboratory

Cubic boron nitride (c-BN) shares several properties with diamond, including high mechanical hardness; high thermal conductivity, second only to diamond; and an ultra-wide band gap (E_g ~ 6.2 eV, indirect). In addition, c-BN can be doped both *n*- and *p*-type, and the lattice mismatch between c-BN and diamond is only ~1.3%. These similarities suggest the potential for novel electronic devices based on c-BN/diamond heterostructures for high temperature and high power applications. However, the growth of device-quality layers of c-BN is challenging: boron nitride occurs in multiple phases; the desired cubic phase is metastable at pressures and temperatures typical of vapor-phase growth; and the absence of large-area bulk c-BN crystals necessitates heteroepitaxial growth on non-native substrates.

Single crystal epitaxial cubic boron nitride films were grown on (100) oriented IIa diamond substrates by ion beam-assisted molecular-beam epitaxy (MBE) in a custom MBE system equipped with an Ar ion source, a N_2 plasma source, and an electron beam evaporator for supplying elemental boron. The films are fully cubic, as indicated by Fourier transform infrared spectroscopy and corroborated by x-ray photoelectron spectroscopy. Transmission electron microscopy reveals an epitaxial c-BN film with the presence of isolated misfit dislocations but no indication of h-BN. The interface between the c-BN layer and the diamond substrate is structurally abrupt, and no interlayer between the c-BN film and diamond substrate is seen. It was found that trace amounts of impurities, such has Mg, Be, and Si, facilitate the growth of c-BN on diamond by ion-assisted MBE.

11:00am **NM-MoM2-14 Optical Properties of ScN Layers Grown on Sapphire Using Plasma-Assisted Molecular Beam Epitaxy**, ***Duc V. Dinh***, Paul-Drude-Institut für Festkörperelektronik, Leibniz-Institut im Forschungsverbund Berlin e.V., Germany*; F. Peiris,* Kenyon College*; J. Lähnemann, O. Brandt,* Paul-Drude-Institut für Festkörperelektronik, Leibniz-Institut im Forschungsverbund Berlin e.V., Germany

Since the report of rock-salt scandium nitride (ScN) on sapphire 50 years ago [1], ScN has been mostly used in combination with other materials. In particular, ScN has been combined with wurtzite AlN to form (Al,Sc)N for surface acoustic wave [2] and high-electron mobility transistor applications [3]. However, very recent studies have shown that ScN itself can also be used for electronic [4] and infrared opto-electronic applications [5].

Here, we report on the optical properties of (3–250)-nm-thick ScN(111) layers grown on sapphire Al2O3(0001) substrates using plasma-assisted molecular beam epitaxy. The optical properties of the layers were investigated by three different techniques, namely, spectroscopic ellipsometry, confocal Raman and photoluminescence spectroscopies. The optical constants of the layers are investigated by variable-angle spectroscopic ellipsometry covering a spectral range from far infrared to far ultraviolet (0.045–8.5 eV). Refractive indices (n, k) of the layers are determined by fitting ellipsometry data using a parametric semiconductor model, taking into account the effects of surface roughness. Fits of ellipsometry data return the energies of four oscillators representing the band-to-band transitions. These correspond to the high-symmetry points in the band structure of ScN including 2.03 eV at the X point and three transitions (3.89, 5.33, and 6.95 eV) at the Γ point. These three oscillators are associated with direct transitions that occur between the degenerate heavy and light hole-bands and the first, second and third conduction bands at the Γ point, respectively. These energy transition values and refractive indices are consistent with theoretical studies previously reported for ScN [6]. Despite the rocksalt structure of ScN, Raman spectra of all the layers reveal several first-order phonon modes with an LO(L) mode at 675–680 cm-1. The appearance of these first-order modes is attributed to defects and impurities in the layers. Room-temperature photoluminescence measurements of the layers are dominated by a band with a peak energy decreasing from 2.3 to 2.2 eV, attributed to a reduction of the oxygen concentration in the thicker layers.

References

[1] N. Sclar, J. Appl. Phys. 35, 1534 (1964)

[2] M. Akiyama et al, Adv. Mater., 21, 593 (2009).

[3] D.V. Dinh et al, Appl. Phys. Lett. 122, 152103 (2023).

[4] N.L. Adamski et al, Appl. Phys. Lett., 115, 232103 (2019).

[5] K.C. Maurya et al, Nano Lett. 22, 5182 (2022).

[6] R. Deng et al, Phys. Rev. Lett., 91, 045104 (2015).

11:15am **NM-MoM2-15 Epitaxial Growth of High ScN Fraction ScAlN on NbN and SiC**, ***Matthew Hardy***, *A. Lang, E. Jin, N. Nepal, B. Downey, V. Gokhale, S. Katzer, V. Wheeler,* U.S. Naval Research Laboratory

ScAlN thin films have attracted significant attention due to their factor of five increase in piezoresponse over AlN for $Sc_{0.43}Al_{0.57}N$. Integration of metallic epitaxial NbN with ScAlN using molecular beam epitaxy (MBE) enables a pathway towards a highly conductive lower electrode while preserving high crystal quality even in relatively thin ScAlN films suitable for use at or above X-band frequencies. Maintaining phase-pure and high crystal quality $Sc_xAl_{1-x}N$ at high *x* is critical to improve resonator bandwidth and reduce insertion loss.

In this work, we show the importance of layer nucleation—both an AlN interlayer, and the initial ScAlN layer—to the final crystal quality of MBE-grown ScAlN films on SiC and NbN/SiC. With the inclusion of a 5-nm AlN interlayer, the $Sc_{0.32}Al_{0.68}N$ XRD FWHM decreases from 2.0° to 1.13°. An AlN interlayer is also critical to growth of ScAlN on NbN thin films. A two-step AlN growth process can effectively encapsulate the NbN layer while providing a smooth surface on which to nucleate ScAlN growth, and is critical to maintaining high crystal quality for ScAlN grown on NbN.

The ScAlN initiation steps also have a strong impact on the final quality of the film. Instead of a two-step ScAlN growth we previously demonstrated, initiation using a linear composition grade from $Sc_{0.32}Al_{0.68}N$ to $Sc_{0.40}Al_{0.60}N$ over 100 nm leads to further improvements in the RHEED pattern, including a narrowing of the spots early in the growth, as well as elimination of remaining ring-like character in the final RHEED pattern after an additional 40 nm of growth, and a XRD FWHM as low as 1.22° for ScAlN films grown on SiC. Transmission electron micrographs show near elimination of cubic grains that otherwise form in the initial layers of the $Sc_{0.40}Al_{0.60}N$. The graded sample has the same average ScN fraction and thickness as the two-step sample. The grade thickness can be reduced to 25 nm (with the remaining 125 nm $Sc_{0.40}Al_{0.60}N$) without degrading the XRD FWHM or RHEED pattern, increasing the average ScN fraction from 0.373 to 0.393. Finally, a 500-nm-total-thickness sample (100 nm $Sc_{0.32}Al_{0.68}N \rightarrow Sc_{0.40}Al_{0.60}N$, 400 nm $Sc_{0.40}Al_{0.60}N$) was grown to show the impact of defect annihilation in thicker films, resulting in a reduction of XRD FWHM to 0.89°. Employing the same 25-nm grade followed by 125-nm of $Sc_{0.40}Al_{0.60}N$ grown on AlN/NbN/SiC results in a FWHM of 1.97°. The improved layer initiation shows that more gradual changes in surface energy and strain reduces the nucleation of undesirable cubic grains, and may point to a general strategy for elimination of anomalous grains in high ScN fraction ScAlN.

11:30am **NM-MoM2-16 High Efficiency Micrometer Scale Green and Red Light Emitting Diodes**, ***Yixin Xiao***, *R. Maddaka, Y. Wu, Y. Malholtra, Y. Guo, S. Yang, J. Liu, K. Sun, A. Pandey, J. Min, Z. Mi,* University of Michigan

High efficiency light emitting diodes (LEDs) with characteristic length scales on the order of microns or less, also known as µLEDs, have been under intense investigations for their immense promise in various display and communications scenarios. Among the many material systems investigated for µLEDs, the III-nitride family possesses many desirable material properties such as comparatively low surface recombination velocities and excellent wavelength tunability. To date, however, it has remained a challenge to achieve efficient green and red emitting µLEDs, largely due to the enhanced surface recombination and poor p-type doping related to the top-down etching. Moreover, it has remained difficult to achieve the high levels of indium incorporation required for a red emitting indium gallium nitride (InGaN) active region. Here we demonstrate that the efficiency bottleneck of µLEDs can be fundamentally addressed by utilizing bottom-up III-nitride nanostructures. We report on the demonstration of micrometer scale green and red LEDs with an external quantum efficiency of 25% and 8%, respectively, which are the highest values ever reported to the best of our knowledge. We employ selective area plasma-assisted molecular beam epitaxy as the material synthesis platform. Due to efficient strain relaxation, such bottom-up nanostructures are largely free of dislocations. By exploiting the large exciton binding energy and oscillator strength of quantum-confined InGaN nanostructures, we show that the external quantum efficiency of a green-emitting micrometer scale LED can be dramatically improved from ~4% to >25%. The dramatically improved efficiency is attributed to the utilization of semipolar planes in strain-relaxed nanostructures to minimize polarization and quantum-confined Stark effect and the formation of nanoscale quantum-confinement to enhance electron-hole wavefunction overlap. We have further developed a new approach that included an InGaN/GaN short period superlattice together with an InGaN quantum dot active region to achieve high efficiency red emission. A maximum quantum efficiency of >7% was measured. Our studies offer a viable path to achieve high efficiency micrometer scale LEDs for a broad range of applications including mobile displays, virtual/augmented reality, biomedical sensing, and high-speed optical interconnects, that were difficult for conventional quantum well based LEDs.

11:45am **NM-MoM2-17 AlN/AlGaN Short Period Superlattices With Sub 2 nm Layers Grown by MME**, ***Alexander Chaney***, Azimuth Corporation*; C. Bowers, K. Mahalingam,* UES INC*; S. Mou, K. Averett, T. Asel,* Air Force Research Laboratory, Materials and Manufacturing Directorate, USA

In this work we present a method for creating $AlN/Al_xGa_{1-x}N$ short period superlattices (SPSL) with individual layer thicknesses down to 3-4 ML. By taking advantage of the oscillatory nature of metal modulated epitaxy (MME), the formation of alternating AlN and $Al_xGa_{1-x}N$ layers can be achieved through the introduction of a constant Ga overpressure during the MME growth of AlN. However a typical MME process results in SPSLs with periods between 5 and 6 nm. In order to reduce the thickness of the AlN and $Al_xGa_{1-x}N$ layers, the shutter timings are reduced in order to limit the total amount of Al deposited on the surface. Initial results with this shortened shutter timing showed 0^{th} order SL peak with Pendellosung fringes in an XRD coupled scan. Such behavior is typically indicative of a high quality SPSL. However, TEM imaging showed a strong intermixing of between the AlN and $Al_xGa_{1-x}N$ layers, leading to a structure that more resembles a random $Al_xGa_{1-x}N$ alloy than a well ordered SPSL. In order to prevent intermixing at the interface layers, 3 altered growth processes were investigated: alternating Al and Ga shutters, inclusion of a N plasma only exposure step and reduction of the Ga flux. The goal with each of these changes was to limit the interaction of the Ga and Al during the final stages of the AlN layer formation. TEM examination of each growth's layer structures showed a significant reduction in layer intermixing for all samples. Alternating the Al and Ga shutter resulted in a SPSL with high order in its layer structure, with AlN and $Al_xGa_{1-x}N$ thicknesses of 4 ML and 3 ML respectively. Introduction of the plasma only step resulted similarly improved interfaces however the thicknesses of AlN and $Al_xGa_{1-x}N$ layers had changed to be 3 ML and 4 ML respectively. Finally, reducing the Ga flux led to a layer structure almost identical to what was obtained using alternating shutters. XRD coupled scans each sample showed Pendellosung fringes centered on a 0^{th} order peak. Fitting of the peaks enabled determination of the average Al composition for the SPSL, which ranged from a low of 84% for alternating shutters to a high of 90% for lower Ga flux. Because these 2 samples showed the same layer thicknesses in TEM, reducing the Ga flux resulted in higher Al content $Al_xGa_{1-x}N$ layers. A coupled scan of the N pause sample showed a broad peak to the left of the main alloy peak. The cause of this is a section of growth where the layered structure had non uniformity which can be seen in TEM indicating further optimization is needed. Based on these results, MME shows promise as method for forming digital alloys.

Author Index

Bold page numbers indicate presenter