Emerging Materials
Room Grand Ballroom E-G - Session EM3-WeM
Epitaxial Growth and III-V Materials

Moderator: John Ekerdt, University of Texas at Austin

10:45am **EM3-WeM-12 Atomic Layer Epitaxy of Zinc Oxide on C-plane Sapphire from Diethylzinc and Water using Pulsed-Heating Atomic Layer Deposition***, **Brandon Piercy**, M Losego,* Georgia Institute of Technology

A challenge to atomic layer epitaxy (ALEp) is the mismatch between low-temperature windows for certain ALD precursors and the higher thermal budgets required for epitaxial growth. A common solution is to use precursors with higher ALD temperature windows, but these do not necessary exist for all desired chemistries. Therefore, ALEp is limited to material combinations having low lattice mismatch and high temperature precursor stability. To circumvent this challenge, we are exploring a new technique called pulsed-heating ALD (PH-ALD) that interleaves individual ALD growth cycles with a fast, high-temperature heat pulse using a high-power resistive heater. As proof-of-concept, we study epitaxial growth of ZnO on c-plane sapphire using a diethylzinc (DEZ) / water chemistry. DEZ is known to decompose above about 180˚C, and the DEZ-H_2O system cannot be grown epitaxially on c-sapphire with traditional thermal ALD approaches. Here, we explore the use of pulse heating to temperatures of up to 900˚C and for various ALD cycle ratios. X-ray diffraction shows the best epitaxial alignment and ZnO rocking curve, with a FWHM of <0.5˚, at a pulse temperature of 900˚C and a 1:1 ALD:heat pulse ratio. Crystal quality and intensity drop significantly with pulse temperatures less than 800˚C. Reducing the ALD cycles:heat pulse ratio also lowers crystal quality, but epitaxial growth is retained down to 5 ALD cycles per heat pulse. Photoluminescence spectroscopy of the D0x band shows that the 1:1 PH-ALD samples have 1.3x the intensity of post-annealed ZnO ALD and 10x the intensity of as-deposited ZnO ALD. Finally, we study growing a few layers of PH-ALD ZnO followed by standard thermal ALD with no pulsed heating. We find that a template layer of only 20 PH-ALD cycles is sufficient to template ZnO epitaxial films up to 100 nm thick.

11:00am **EM3-WeM-13 Growth of AlN Barriers in Al/AlN/Al SIS Josephson Junctions by Low Temperature Atomic Layer Epitaxy***, **Charles Eddy, Jr.**,* U.S. Naval Research Laboratory*; D Pennachio, J Lee, A McFadden,* University of California, Santa Barbara*; S Rosenberg,* U.S. Naval Research Laboratory*; Y Chang, C Palmstrom,* University of California, Santa Barbara

Superconductor-Insulator-Superconductor (SIS) structures are of increasing interest for the creation of Josephson junctions that can serve as the basis for quantum qubit transmons, which hold significant promise for quantum computing technologies. Traditionally, these devices have been developed using amorphous AlO_x in Al/AlO_x/Al structures and have enabled fundamental demonstrations of transmon performance. However, improved performance may be expected with an epitaxial insulator. Even in these structures, the nature of the superconductor/substrate interface and the superconductor/ambient interface limits coherence and, consequently, qubit performance.

In an effort to address this challenge, we employ low temperature atomic layer epitaxy (ALEp) to grow crystalline AlN insulators on crystalline aluminum films. Smooth epitaxial aluminum films are grown by evaporation on cryogenically-cooled, buffered GaAs(001) substrates [1]. These epitaxial surfaces are “frozen” using a low temperature nitridation atomic layer process (ALP) before the samples are ramped to 300° C for low temperature ALE of AlN using semiconductor grade trimethylaluminum and UHP argon and nitrogen inductively coupled plasmas (ICPs). In this study, we evaluate the structural effects of variations in the initial nitridation ALP, growth conditions of ALEp AlN barriers, and SIS barrier thickness using transmission electron microscopy. We have found that at one end of the spectrum, a simple 5 cycle nitridation ALP of epitaxial aluminum at ~90° C, where each cycle is a 30 second exposure to 300W UHP argon/nitrogen (200/75 sccm) ICP , consumes a significant fraction of the aluminum to make an amorphous AlN insulator that is roughly 2 nm thick. When this surface is subjected to another low temperature Al evaporation, the top Al films are a mixture of amorphous and polycrystalline. When the same nitridation ALP is employed and followed by 5nm of ALEp AlN growth at 300° C, a similar amount of the aluminum film is consumed and an amorphous ALEp AlN layer results. Finally, when the nitridation ALP is reduced to a single cycle of nitridation, less of the aluminum film is consumed and the 5nm AlN ALEp film shows polycrystallinity with small regions demonstrating sharp, potentially epitaxial interfaces. This result suggests that proper ALP nitridation of the epitaxial aluminum can support epitaxial growth of AlN by ALE. Further studies of the influence of number of cycles, cycle duration, plasma chemistry and plasma power on both the nitridation ALP and AlN ALEp will be presented.

[1] S. Gazibegovic et al., Nature 548, 434 (2017).

11:15am **EM3-WeM-14 Investigating Plasma Parameters and Influence of Argon to the Crystallinity of GaN Films Grown by Plasma-Assisted ALD***, **Deepa Shukla**, I Saidjafarzoda, A Mohammad, B Brian Willis, N Biyikli,* University of Connecticut

Gallium nitride has attracted significant attention in (opto)electronic and RF-chip industry mainly due to its wide bandgap of about 3.4 eV,which revolutionized lighting as well as high-power and high-frequency electronic devices. However, conventional crystal growth methods employ substrate temperatures typically around ~1000 °C, which is incompatible with post-CMOS integration and other temperature-sensitive substrates. To broaden the application spectrum of GaN, lower temperature growth methods are highly needed. Plasma-assisted ALD (PA-ALD) is a strong candidate which also provides unmatched conformality and uniformity performance. In this work, GaN thin films were grown on Si (100) substrates using PA-ALD. Trimethylgallium (TMG) and N2 /H2/Ar plasma were used as metal precursor and co-reactants. GaN deposition experiments were performed with and without Ar plasma at different plasma powers (150W, 175W, 200W) and different temperatures ranging within 120-240°C. An optimal growth per cycle (GPC) of ~0.7Å was observed at 150°C using in-situ ellipsometer and was confirmed by ex-situ x-ray reflectivity (XRR) analysis. The hexagonal wurtzite crystal peaks were observed in the grazing-incidence x-ray diffraction (GI-XRD) analysis. Among the films deposited, GaN samples grown at 240°C showed crystalline character with a preferred orientation along the (002) plane signifying GaN crystal formation temperature above 200°C. Moreover, the absence of Ar gar in the plasma showed a notable reduction in the peak intensities of (100) and (101) crystal domains, resulting in a predominantly preferred (002) orientation. Effect of plasma power and plasma duration on the quality of GaN was carried out with N2/H2-plasma during deposition cycles. No considerable change in crystal orientation was observed with change in plasma power, however increase in plasma duration from 10 to 40 sec suppressed the formation of (100) and (101) domains, resulting in a dominant (002) diffraction peak.

11:30am **EM3-WeM-15 Ultrathin GaN Epilayer by Low-temperature Atomic Layer Annealing and Epitaxy***, **Wei-Chung Kao**, W Lee, Y Yin,* National Taiwan University, Republic of China*; J Shyue,* Academia Sinica*; H Lin, M Chen,* National Taiwan University, Republic of China

Conventionally, GaN epilayers have been grown by metal-organic chemical vapor deposition (MOCVD) at very high growth temperatures (>1000°C) in order to achieve high crystal quality. However, the high-temperature growth technique is challenged by thermal stress and cracking due to the large difference in thermal expansion coefficients between GaN and the substrate. In this research, high-quality GaN heteroepitaxy has been achieved by atomic layer annealing and epitaxy (ALAE) at a low growth temperature of 300°C. By introducing a layer-by-layer, *in-situ* He/Ar plasma treatment at a low plasma power in each cycle of atomic layer deposition, the crystal quality of the GaN is significantly enhanced. The significant improvement of the GaN crystal quality is attributed to the effective annealing effect due to ion bombardment from the He/Ar plasma to each as-deposited layer. The nano-beam electron diffraction, high-resolution transmission electron microscopy, and atomic force microscopy reveal a high-quality nanoscale single-crystal GaN epilayer with a very smooth surface. The full width at half-maximum of the X-ray rocking curve of the GaN epilayer is as low as 168 arcsec. This research demonstrates the impact of the low-temperature ALAE technique for growing high-quality nanoscale GaN epilayers for high-performance solid-state lighting, solar cells, and high-power electronics.

11:45am **EM3-WeM-16 High Quality ALD Formation of Group-III Nitrides and their Applications in FTO-based Thin Film Solar Cells***, **Xinhe Zheng**, H Wei, P Qiu, M Peng, S Liu, Y He, Y Song, Y An,* University of Science and Technology Beijing, China

Group-III nitrides semiconductors (GaN, AlN, InN), grown by high-temperature MOCVD or MBE methods, have been widely utilized in optoelectronic and microelectronic devices. However, due to some benefits like low deposition temperature, precise thickness control and good conformality, ALD has recently attracted much attention in the fabrication of group-III nitrides materials. Here, we report high-quality growth of nitrides using plasma-enhanced ALD (PEALD). By optimizing the ALD window parameters, a single-crystalline nitride thin film can be achieved. Their optical, electrical, structural properties and impurities content are in detail characterized.

For some thin film solar cells using FTO glass substrates, interface modification and/or carrier transportation have been mainly focused for the improvement of device performance. So far, various metal oxides (TiO_2, Al_2O_3, ZrO_2, SiO_2, etc) deposited by ALD have been used in the CIGS and perovskite solar cells. However, there are still no reports about the application of group-III nitrides in FTO-based thin film solar cells using quantum dots (QDs) and perovskite compounds as solar energy materials. In this study, we successfully fabricate high quality GaN and AlN onto the FTO glass substrates at a very low temperature by PEALD. It was found that the existence of ultrathin AlN coating can efficiently reduce quantum loss in the interface between TiO_2 and CdSeTe QDs. An enhancement of open-circuit from 0.642 V to 0.679 V, corresponding to an increase of conversion efficiency from 8.51 % to 9.31% under AM 1.5G irradiance, is achieved for an insertion of AlN. The results show that the ultra-thin AlN layer on TiO_2/QDs surfaces can create an energy barrier for electron injection from QDs into the electrolyte and the injected electron from TiO_2 into the electrolyte, thus effectively inhibiting photo-generated electron recombination. The ALD GaN thin film on FTO glass here could serve electron transport layer for perovskite solar cells. The results indicate that GaN thin-film thickness has a significate effect on the electron transport and collection of the photovoltaic cells. It is found that the devices based on the 50-cycle GaN thin-film (~4 nm) show the best cell performance with efficiency of 15.2%. The introduction of group-III nitrides has shown promise for improving solar cell performance and could open up a way in designing novel thin film solar cells.

References

1， H. Wei, X. Zheng*, et.al, Appl. Surf. Sci., to be issued.

2， S. Liu, X. Zheng*, et.al, Chin. Phys. B, to be issued.

3， M. Peng, X. Zheng*, et.al, Nanoscale, to be issued.

4， S. Liu, X. Zheng*, et.al, Nano. Res. Lett., 12(279), 1-6(2017).

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