High Efficiency InGaN LEDs Emitting in Green, Amber and Beyond

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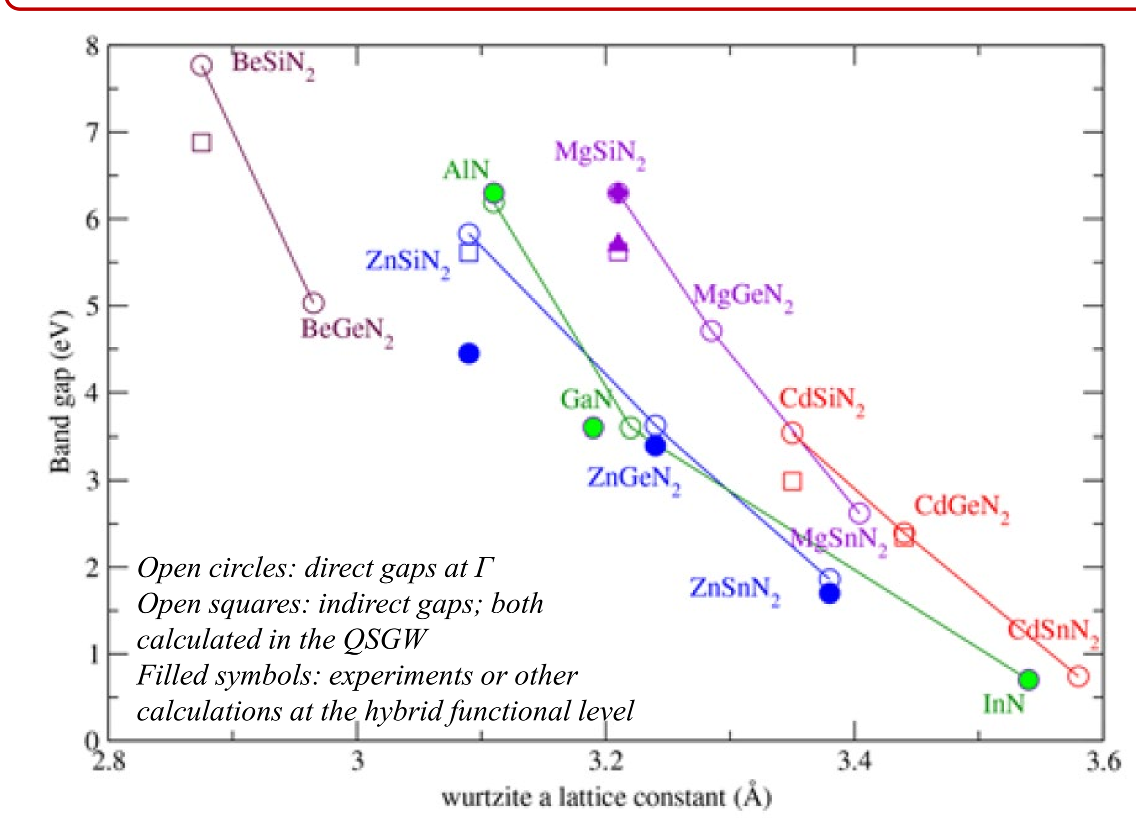
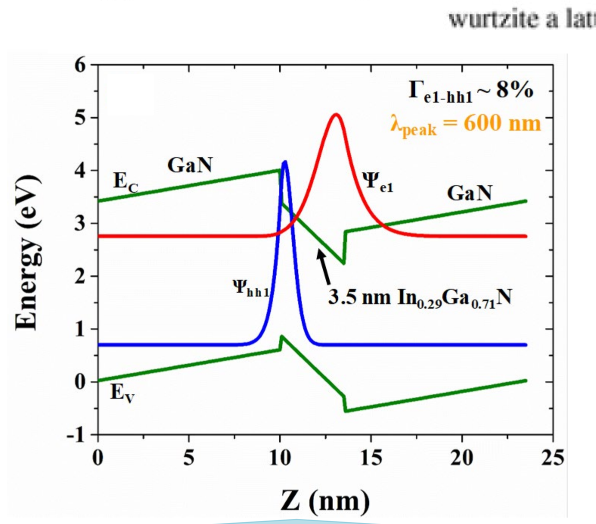
Objectives

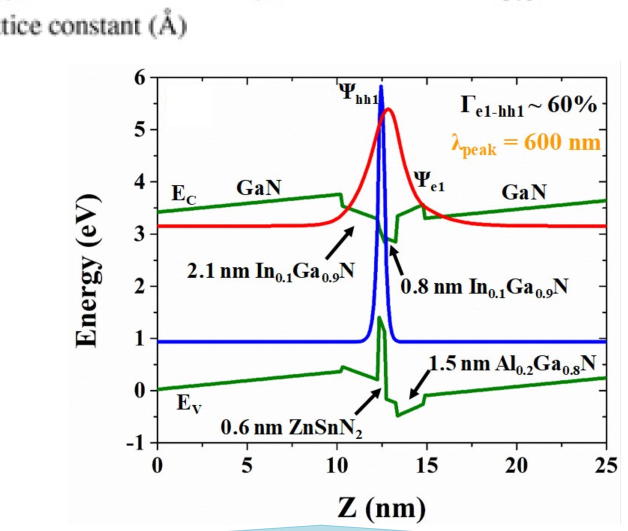
The objective of the proposed work is to develop and demonstrate MOCVD growth of InGaN-$ZnGeN_2$, InGaN-$ZnSnN_2$ QWs LEDs emitting at green, amber and longer wavelengths with high IQE (>50%). The goal is to achieve high efficiency blue, green, amber and red LEDs that enable color-mixed true white LEDs based on the same materials platform with wall plug efficiency exceeding the current state-of-the-art.

Conceptual Design





Low electron-hole wavefunction overlap in conventional GaN/InGaN LED designed for, e.g. 600 nm emission wavelength



Enhanced electron-hole wavefunction overlap in InGaN/II-IV-N_2 base LED designed for same (600 nm) emission wavelength with less In content

- Significantly enhanced e-h wavefunction overlap
- Lower In required to achieve emission beyond green

Strategies and Techniques

- Development of growth recipes for $ZnGeN_2$ and $ZnSnN_2$
  - Control over cation composition
  - Obtain stoichiometric materials
  - Growth conditions compatible with InGaN
- Demonstration of III-N/II-IV-N_2 based LEDs
  - Growth of the heterostructures
  - Insertion of $ZnGeN_2$ or $ZnSnN_2$ layer inside GaN/InGaN quantum wells
  - Evaluation of performance metrics
- Growth technique: Metalorganic chemical vapor deposition (MOCVD)
  - Used for LED manufacturing
  - Unique dual chamber MOCVD at the Ohio State University

Key Progresses

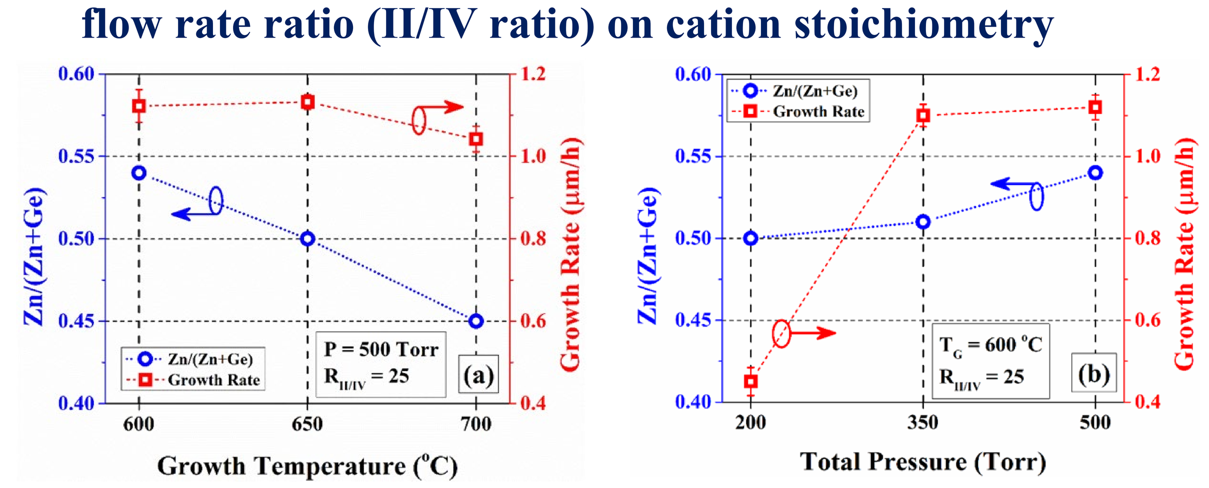
- Growth of $ZnGeN_2$ and $ZnSnN_2$ thin films by MOCVD
  - Establishing optimal MOCVD growth conditions for single crystalline $ZnGeN_2$ and $ZnSnN_2$ thin films
  - Understanding structural, electrical and optical properties of the grown films

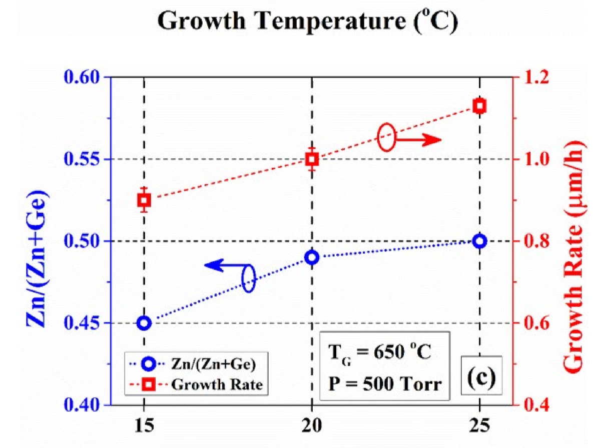
Experimental details

- Substrates
  - GaN-on-sapphire templates
  - InGaN/GaN/c-sapphire templates
  - Sapphire (c-, r- and a-plane)
- Precursors
  - Diethylzinc (DEZn)
  - Tetramethyltin (TMSn)
  - Germane (GeH_4)
  - Ammonia (NH_3)

Results – $ZnGeN_2$ MOCVD

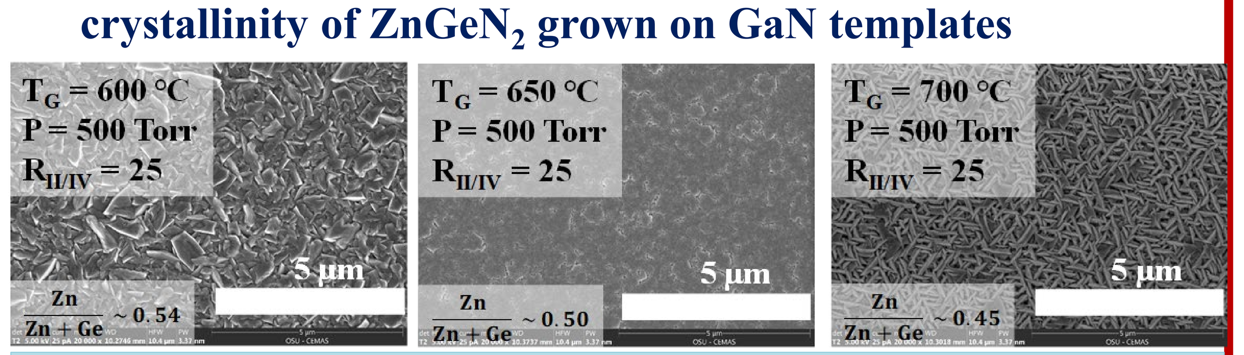
- Effects of growth temperature, pressure and DEZn/GeH_4 flow ratio (II/IV ratio) on cation stoichiometry



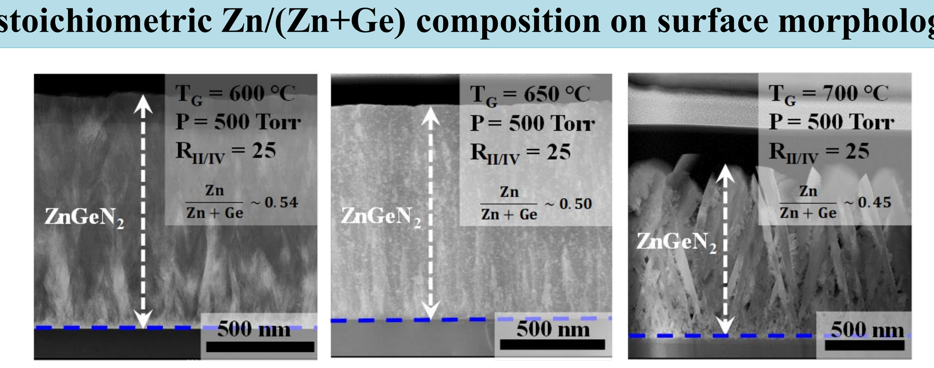


- T_G ↑ → Zn/Ge ratio ↓
- P ↑ → Zn/Ge ratio ↑
- II/IV ratio ↑ → Zn/Ge ratio ↑

- Effects of cation compositions on morphology and crystallinity of $ZnGeN_2$ grown on GaN templates



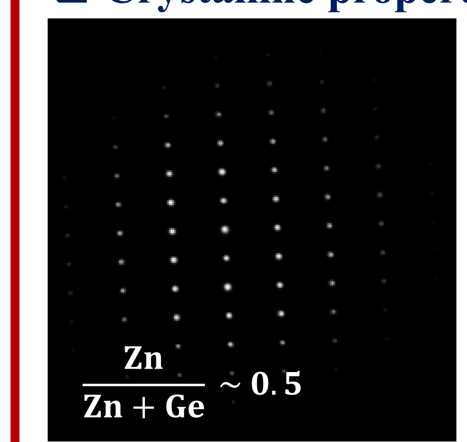
Plan-view FESEM images of $ZnGeN_2$ films showing the effect of stoichiometric Zn/(Zn+Ge) composition on surface morphologies



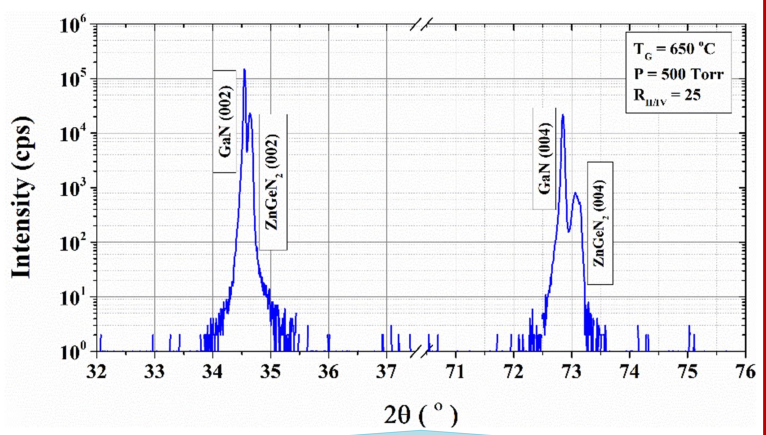
STEM-LAADF images showing dramatically different cross-sectional morphologies in $ZnGeN_2$ films having different cationic compositions

- Achieved $ZnGeN_2$ films with RT mobility of 50 cm^2/Vs

Crystalline properties

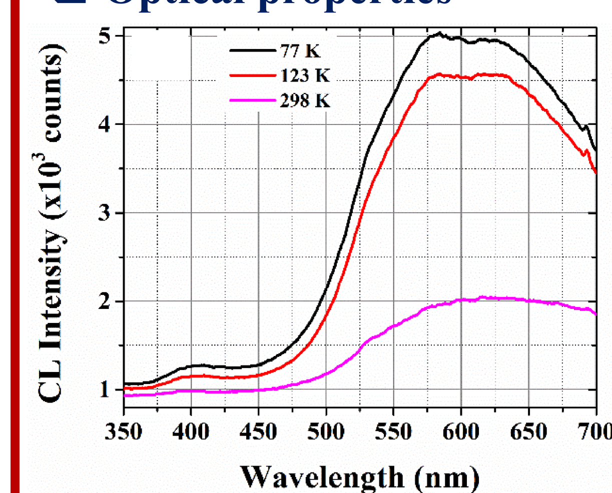


TEM nano-diffraction pattern of a stoichiometric $ZnGeN_2$ film

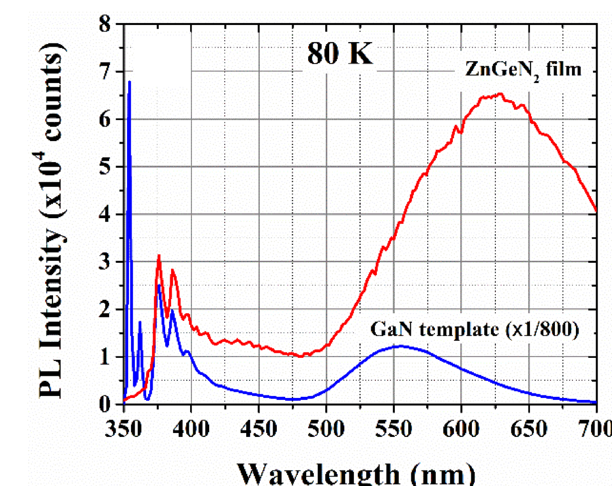


XRD 2θ-ω scan profile of a stoichiometric $ZnGeN_2$ film grown on GaN template

Optical properties



CL spectra of a near-stoichiometric $ZnGeN_2$ film measured at different temperature



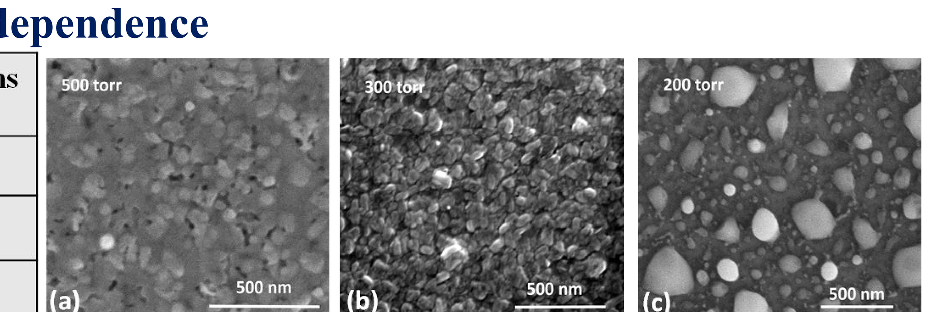
PL spectra of a near-stoichiometric $ZnGeN_2$ film and the GaN substrates measured at 80K

Results – $ZnSnN_2$ MOCVD

$ZnSnN_2$ on c-sapphire

- Pressure dependence

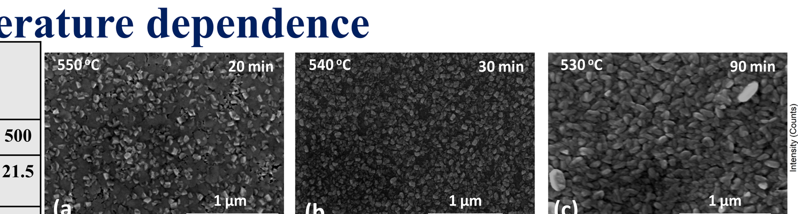
Growth Conditions	
Temperature (°C)	550
DEZn (μmol/min)	24
TMSn (μmol/min)	68
NH_3 (L/min)	4.0



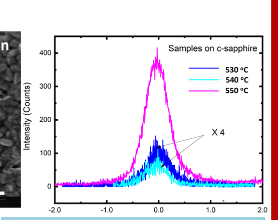
SEM images of $ZnSnN_2$ films grown on c- sapphire at the chamber pressure of (a) 500 torr ,(b) 300 torr and (c) 200 torr in Sn rich environment. Higher pressure promotes film formation.

- Temperature dependence

Growth Conditions	
Pressure (torr)	500
DEZn(μmol/min)	21.5
TMSn(μmol/min)	22.7
NH_3 (L/min)	4.0

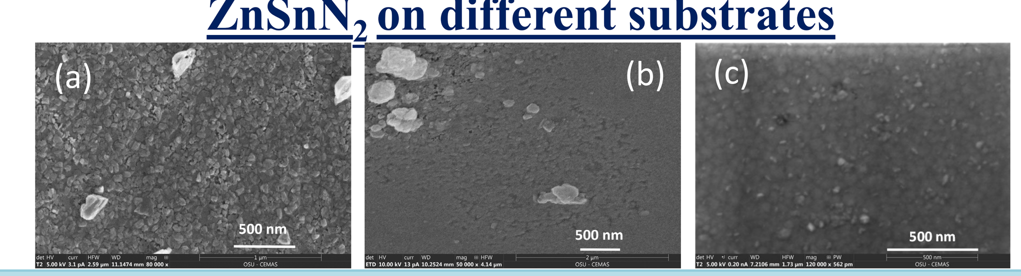


SEM images of $ZnSnN_2$ films grown on c-sapphire at growth temperatures of (a) 550 °C, (b) 540 °C and (c) 530 °C with a Zn-to-Sn flux ratio of ~1. Film surface morphology improves as the temperature increases. Major changes in the film quality resulted from small changes in the growth temperature.

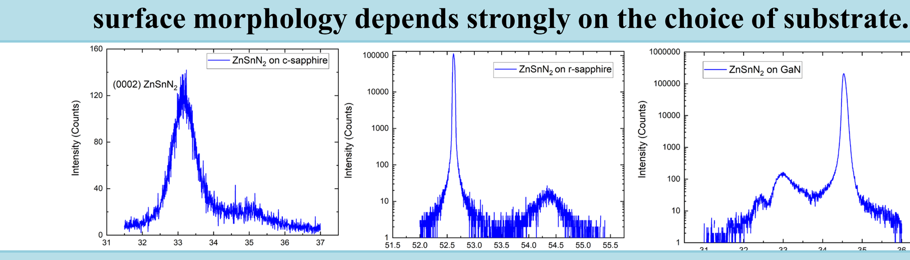


XRD 2θ-ω scan profile of the films shown at the left. The higher peak intensity for the film grown at higher temperature is an indication of improved crystallinity.

$ZnSnN_2$ on different substrates



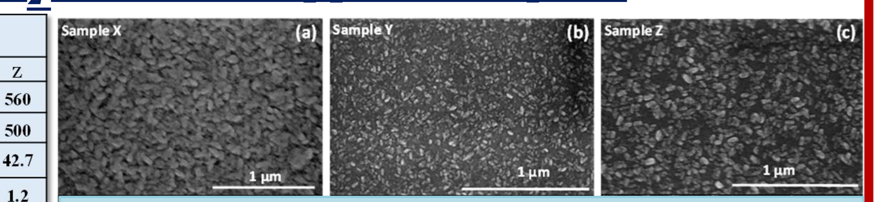
SEM images of $ZnSnN_2$ films grown simultaneously on (a) c- sapphire, (b) r-sapphire and (c) GaN/c-sapphire substrates at 535 °C and 500 torr with a Zn-to-Sn flux ratio of ~1. The film surface morphology depends strongly on the choice of substrate.



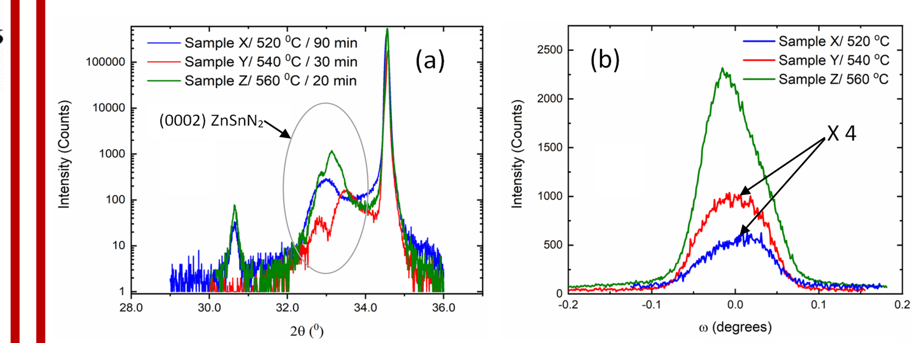
XRD 2θ-ω scan profiles of the $ZnSnN_2$ films shown above. The presence of satellite peaks in the XRD 2θ-ω spectrum of the film grown on GaN indicates higher crystalline quality.

$ZnSnN_2$ on GaN/c-sapphire template

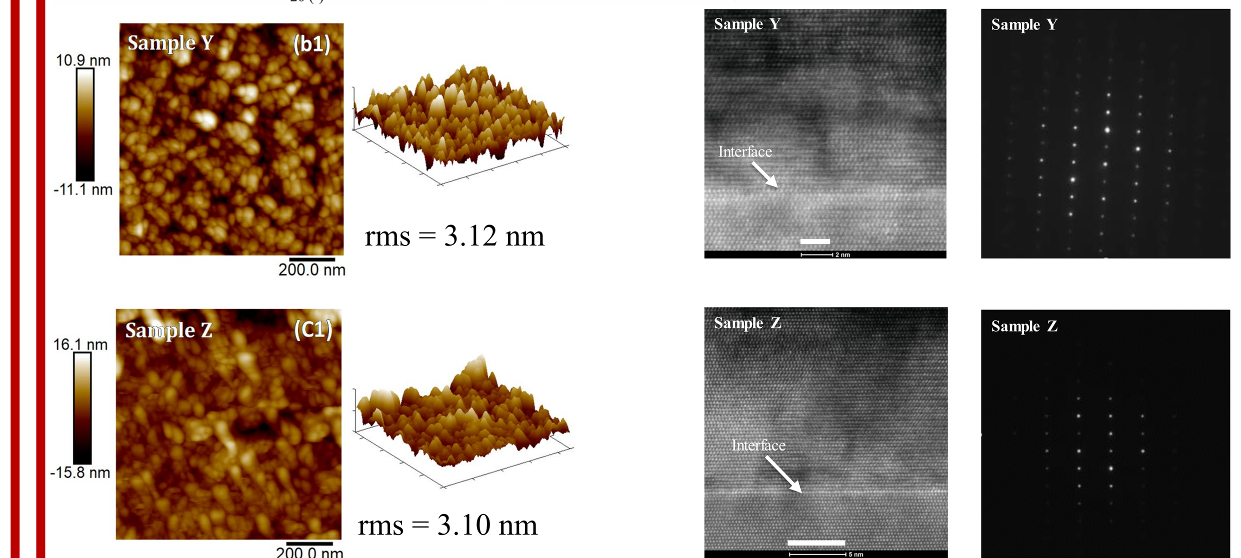
Growth Conditions	Sample		
	X	Y	Z
Temperature (°C)	520	540	560
Chamber Pressure (Torr)	500	500	500
DEZn Flow rate (μmol/min)	22.7	22.0	42.7
Zn/Sn flux ratio	1.1	1.1	1.2
Duration (min)	90	30	20



SEM images of $ZnSnN_2$ films grown on GaN/c-sapphire under the conditions presented in the left table. Samples Y and Z are continuous films with embedded nano-crystallites.

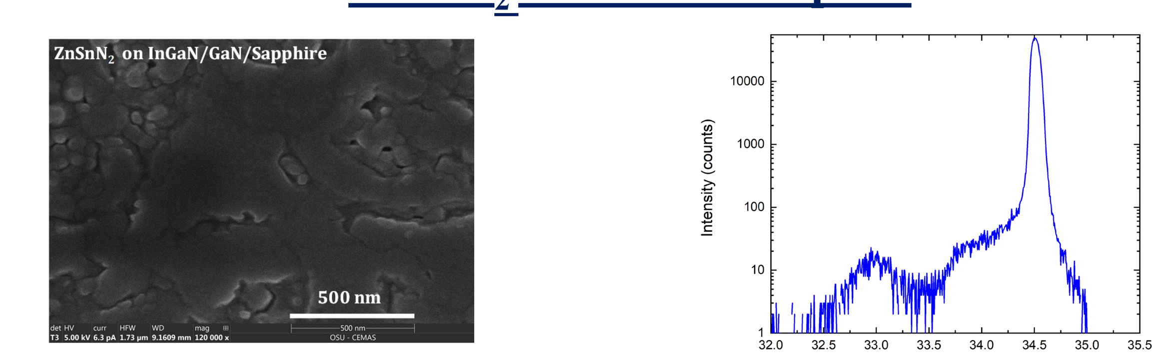


XRD (a) 2θ-ω scan profiles and (b) rocking curves of the main 2θ-ω peak of the $ZnSnN_2$ films shown above. The measured FWHM are 0.10°, 0.09° and 0.08° for samples X, Y and Z, respectively.



AFM images of sample Y and Z (left) and TEM measurements of these samples (right). The surface roughnesses are almost the same for the two samples. The TEM images show smooth interfaces. The TEM diffraction patterns are characteristic of wurtzite single crystal diffraction.

$ZnSnN_2$ on InGaN template



The SEM surface morphology and XRD 2θ-ω scan profile of a $ZnSnN_2$ film grown on InGaN/GaN/Sapphire. This sample was grown simultaneously with Sample Y. The XRD profile of the sample shows that the crystallinity of the film is poorer than that of the film grown on a GaN template. The recipes for growth of $ZnSnN_2$ films on InGaN have yet to be optimized.

Summary

Key accomplishments to date

- ✓MOCVD for single crystalline $ZnGeN_2$ and $ZnSnN_2$ films
- ✓Control over stoichiometry → from Zn-rich to Zn-poor
- ✓Films with decent surface morphology and roughness
- ✓Characterization of crystalline and optical properties
- ✓$ZnGeN_2$ films with RT electron mobility of 50 cm^2/Vs

Ongoing and future work

- Develop InGaN/GaN QW LEDs emitting beyond green
- Develop InGaN/$ZnGeN_2$ and InGaN/$ZnSnN_2$ QW LEDs

Acknowledgement

- U.S. Department of Energy (DE-EE0008718)
- Project Term: April 17, 2019 – April 16, 2022
- Prior support: NSF (DMREF-SusChEM-1533957)
- Team: OSU and CWRU. Both universities will contribute towards growth of materials and devices using the MOCVD system physically located at the OSU as well as towards characterization.