MOCVD Growth and Characterization of $ZnSnN_2$

Benthara Hewage Dinushi Dinushi Jayatunga

Case Western Reserve University
Dept. of Physics

B. H. D. Jayatunga[1], M. R. Karim[2], Menglin Zhu[3], Jinwoo Hwang[3], H. Zhao[2] and K. Kash[1]

[1]Department of Physics, Case Western Reserve University
10900 Euclid Avenue, Cleveland, Ohio 44106-7079, USA
[2]Department of Electrical and Computer Engineering
[3]Department of Materials Science and Engineering
The Ohio State University, 2015 Neil Avenue, Columbus, Ohio 43210

Motivation

All earth abundant elements

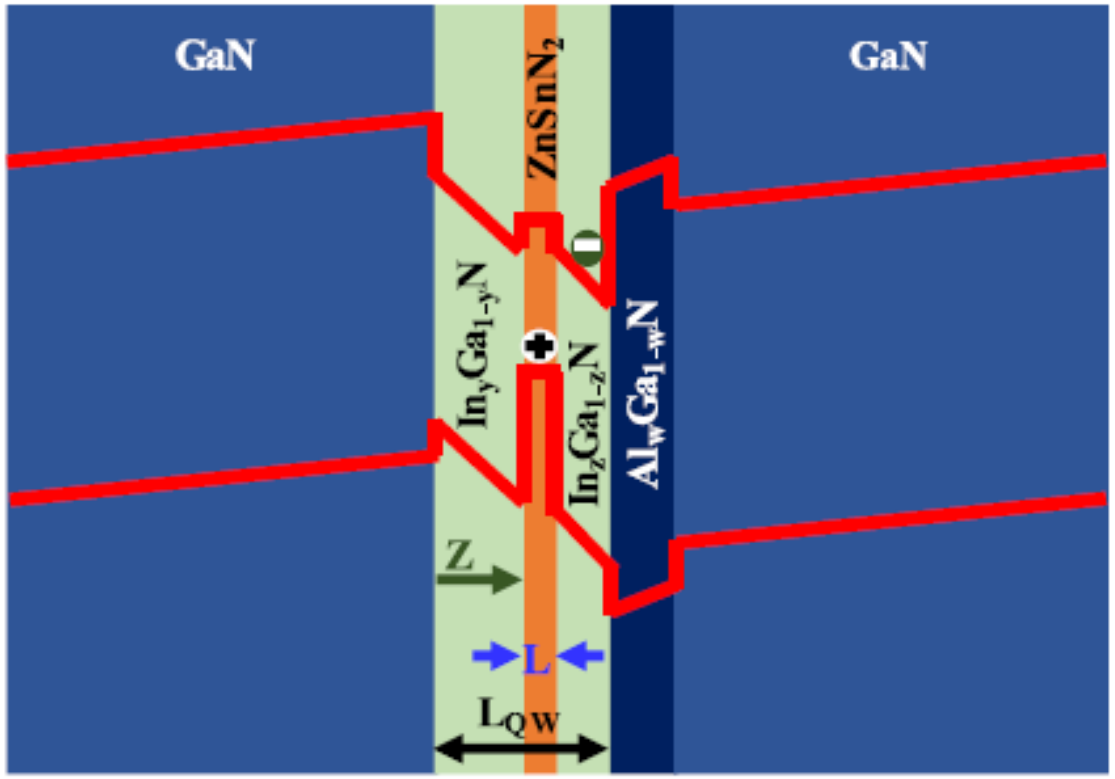
Diverse applications

- Photovoltaics
- visible light photocatalysis
- optoelectronics

Example

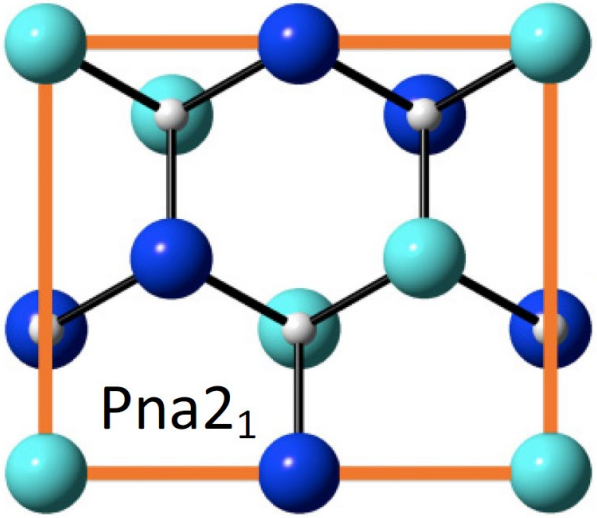
- **Proposed amber LED structures**
  - Lower In content
  - Improved electron-hole overlap

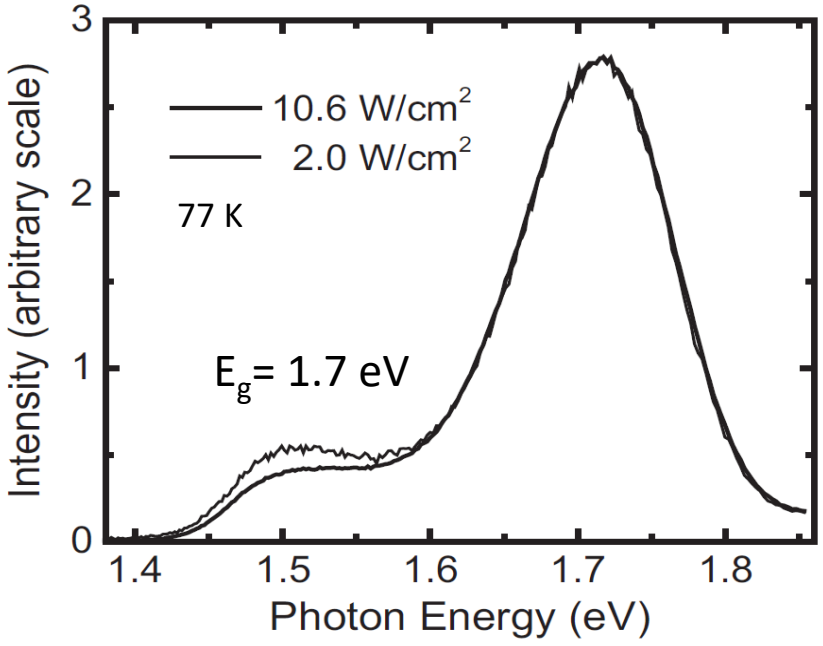
  (8% → 60%)
  - Improved radiative recombination rate (x 255)

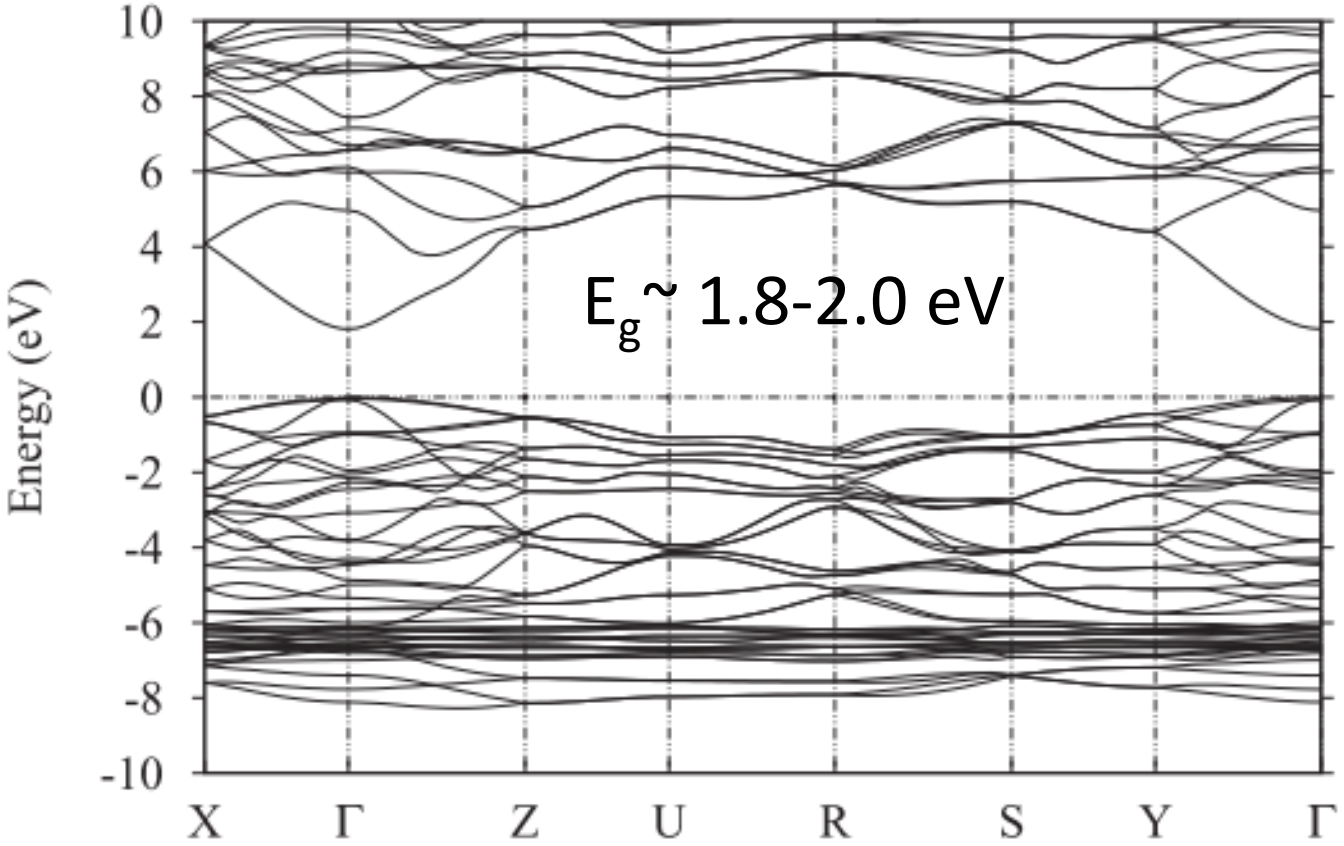


M.R. Karim & H. Zhao, J. Appl. Phys. 124, 034303 (2018)

Lattice and Band Structure







Quayle et al, Physical Review B 91 (2015).

Background

- Previous growth techniques:
  - plasma-assisted vapor–liquid–solid[1]
  - Magnetron Sputtering[2]

  } Polycrystalline

  - RF Sputtering [3][4]
  - plasma-assisted MBE[5][6]

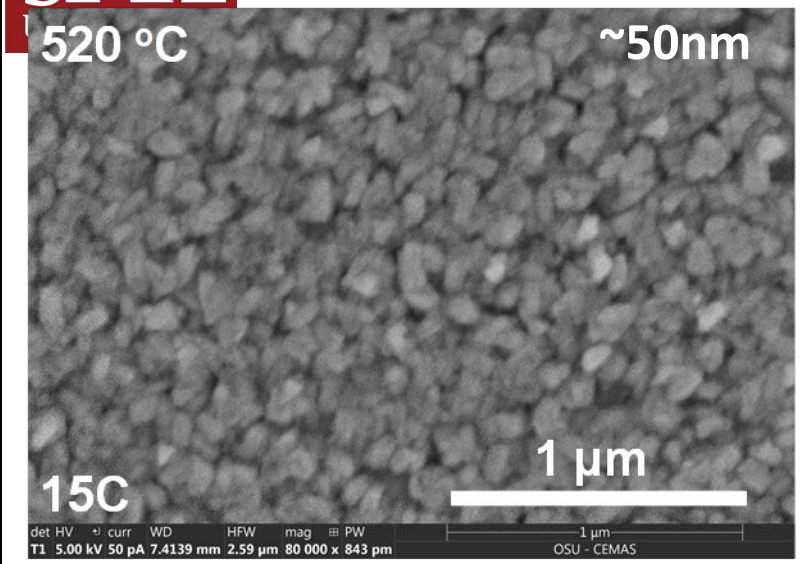
  } Single Crystalline

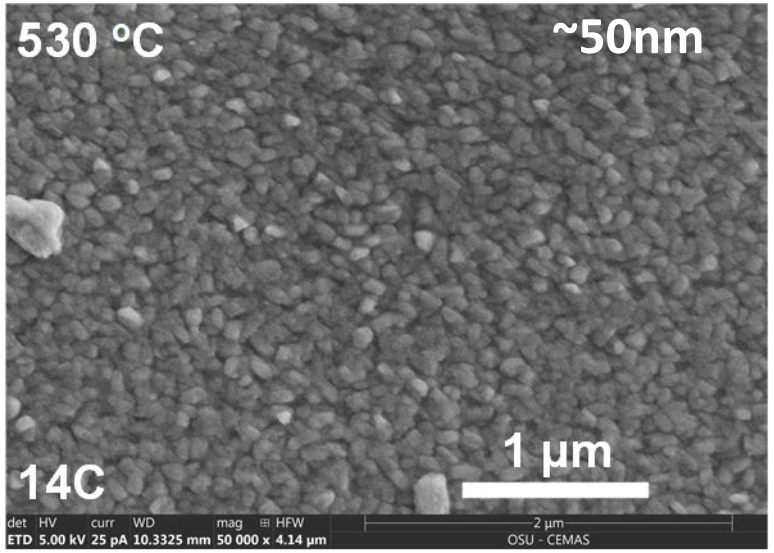
[1] Quayle et al. MRS Comm. **3**, 135 (2013)
[2] Qin et al. App. Phys. Lett. **108**, 142104 (2016)
[3] Narang et al. Adv Mater **26**, 1235 (2014)
[4] Lahourcade et al. Adv Mater **25**, 2562 (2013)
[5] Feldberg et al. 38th IEEE Photovoltaic Specialists Conference (2012)
[6] Le et al. Cryst. Growth Des. **18** 1385 (2018)

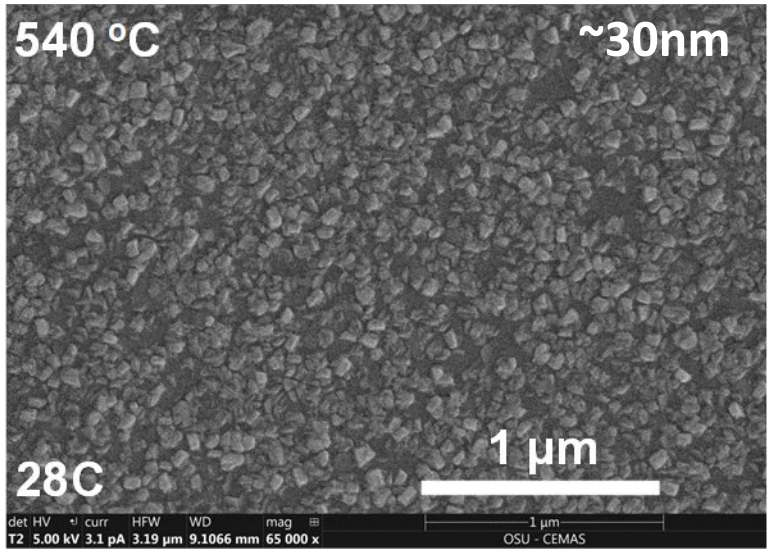
Experimental Procedure

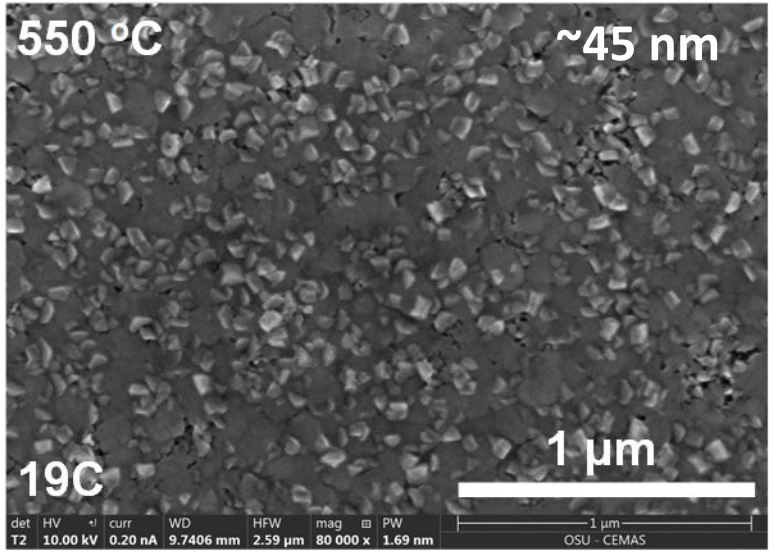
- Substrates → r- and c-sapphire, GaN-on-sapphire, InGaN
- Precursors → tetramethyltin (TMSn), diethylzinc (DEZn), NH_3
- Growth →
  - 500 -560 ^{0}C
  - 200 -500 torr
  - DEZn 20-70µmol/min
  - TMSn 20-70 µmol/min
- Characterization → SEM, XRD, AFM, TEM, Hall, APT

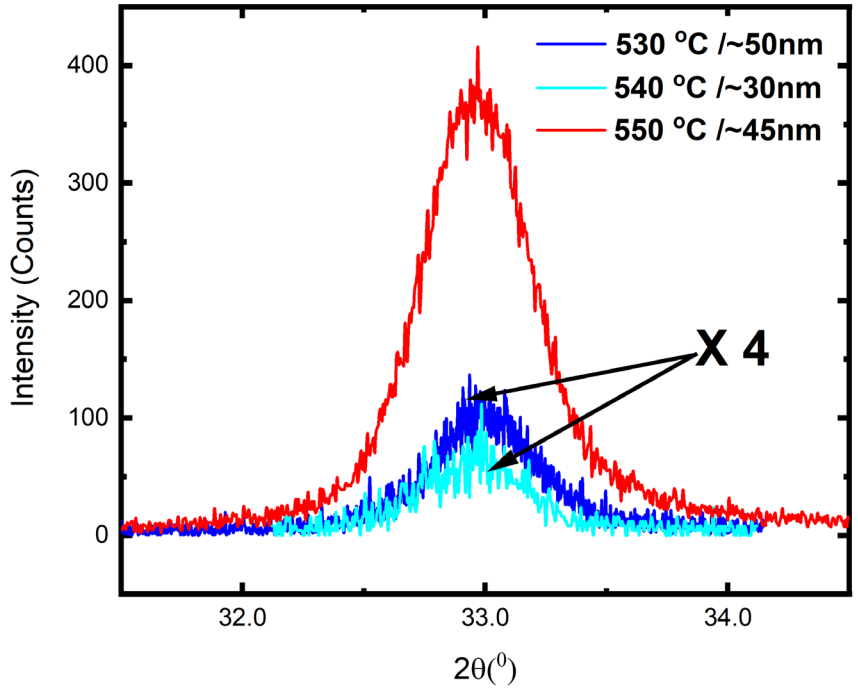
Higher Temperature → Increased smoothness







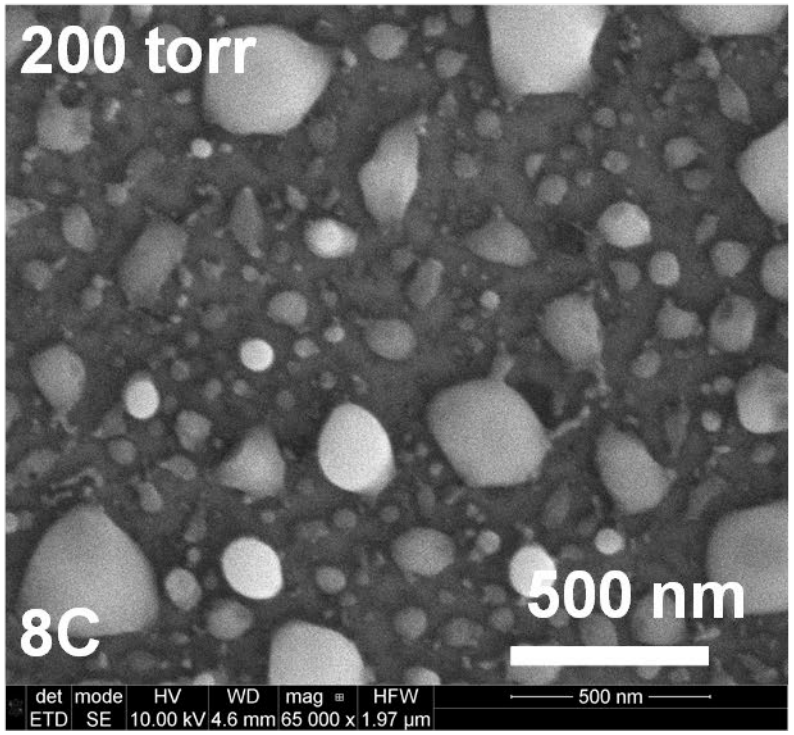


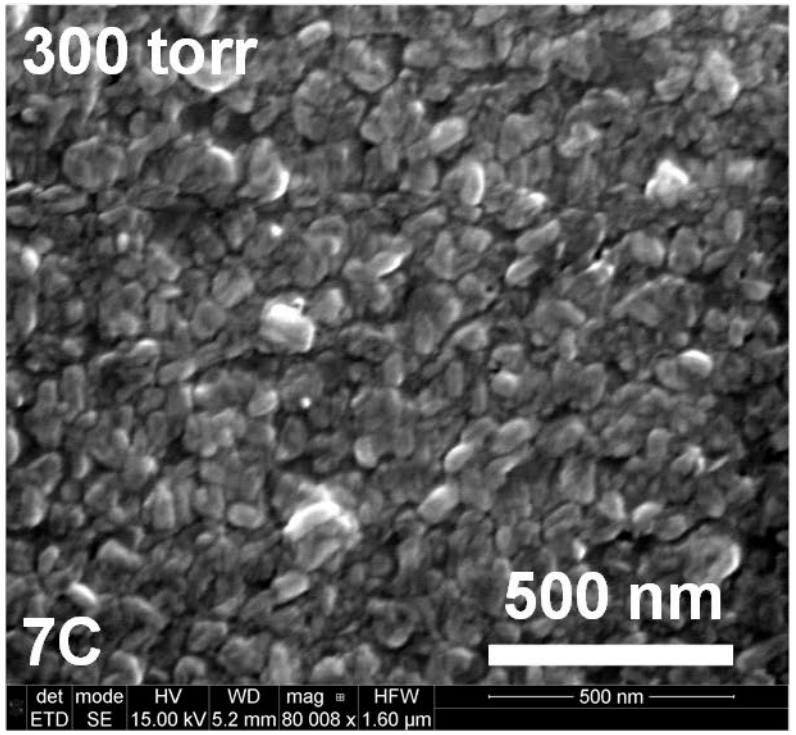


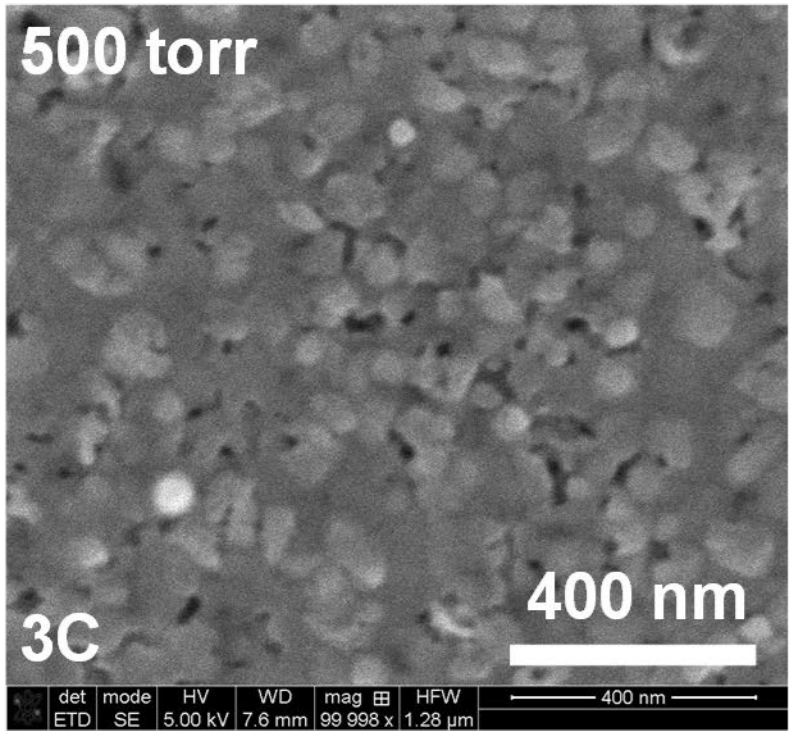
Ga contamination after growth 16

Substrate – c sapphire

Higher Pressure facilitates growth



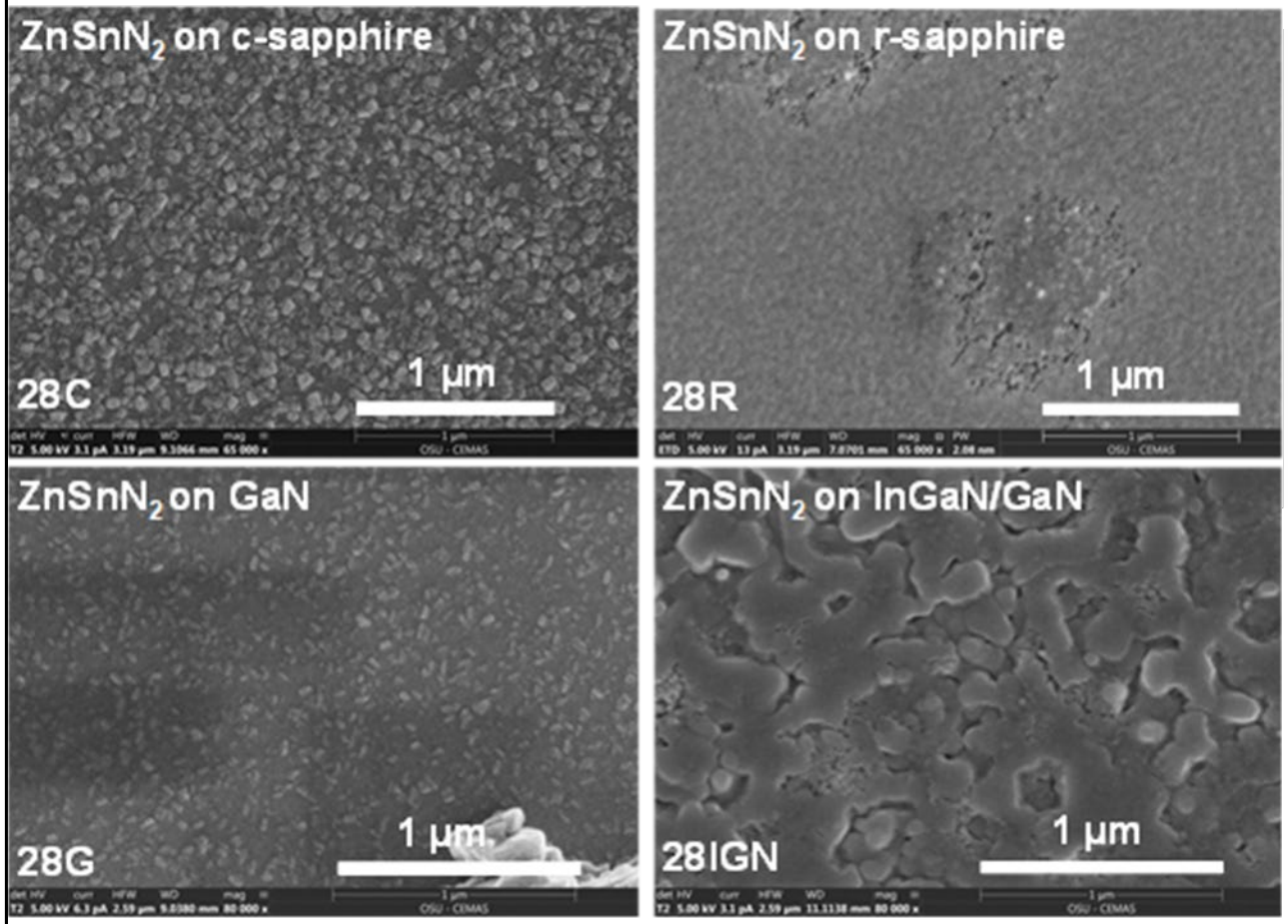


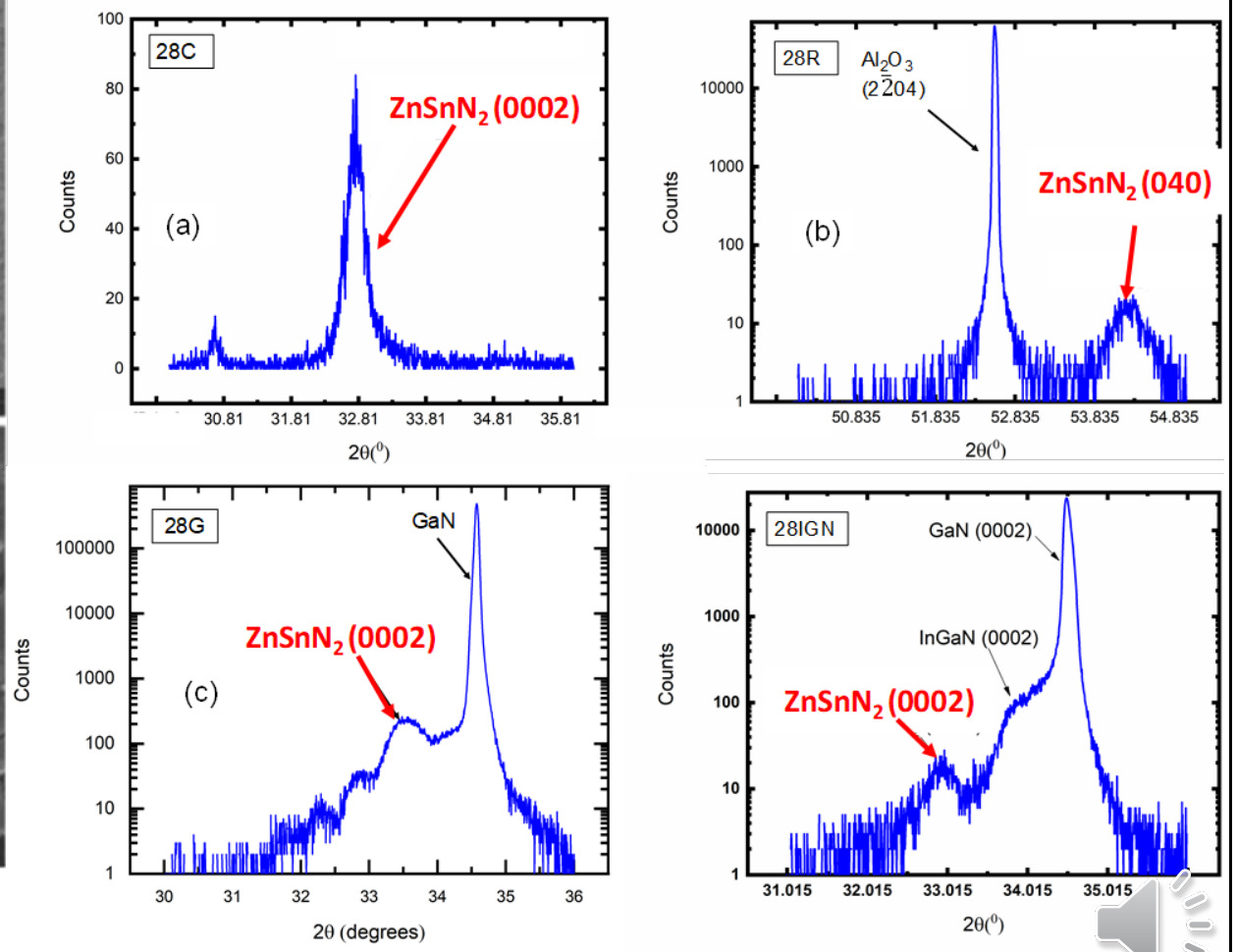


Sn droplets

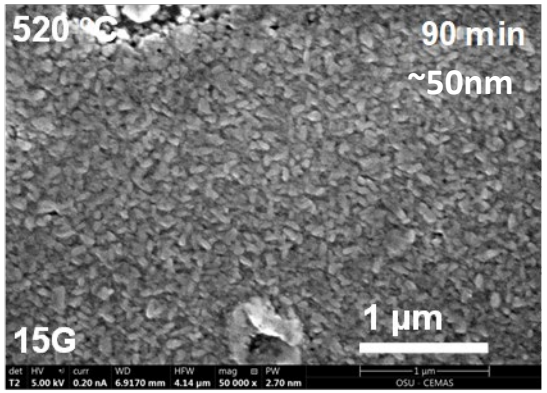
Continuous film

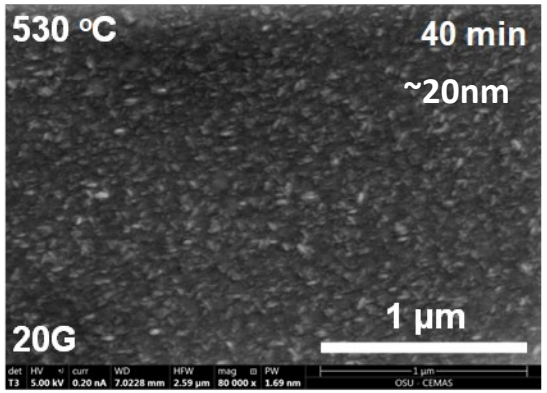
Choice of Substrate – GaN template is best

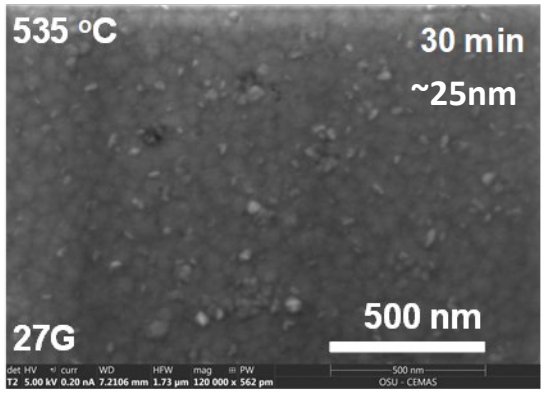


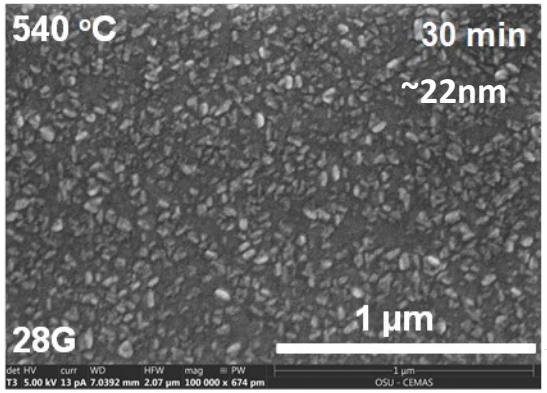


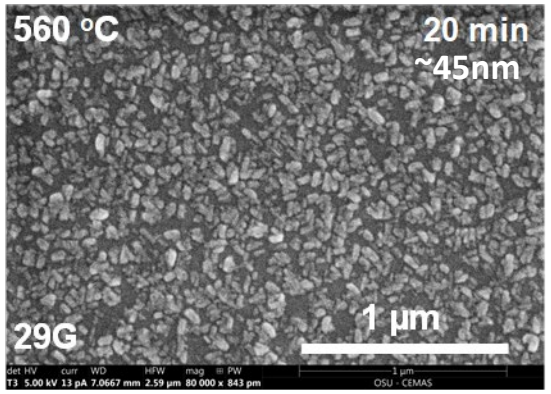
$ZnSnN_2$ growth on GaN – Satellite peaks and Better film quality

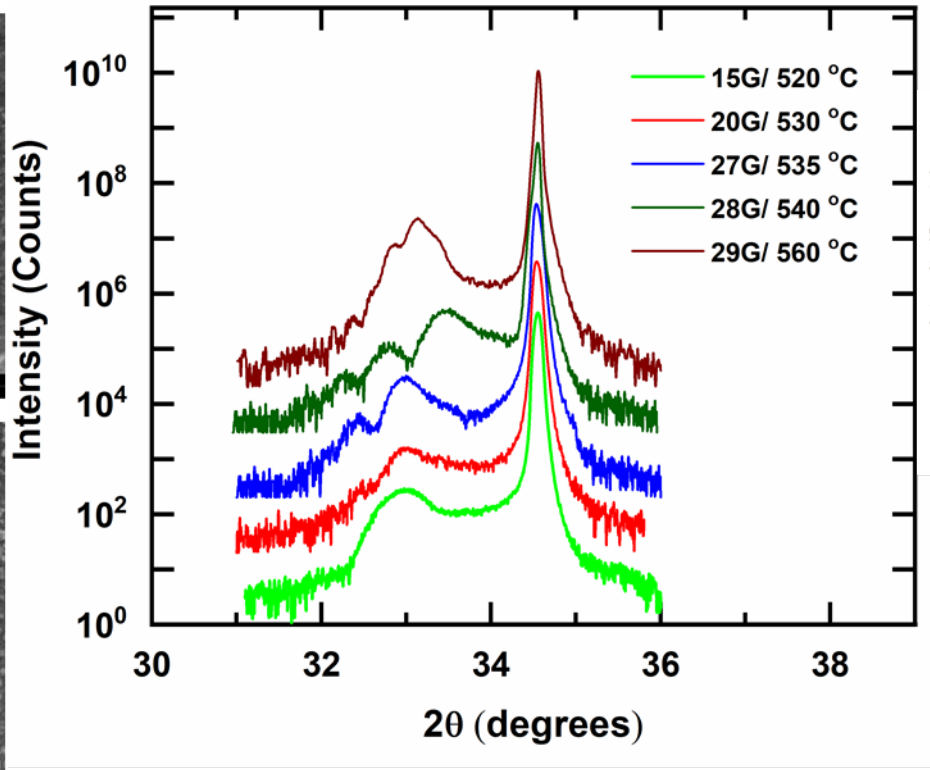


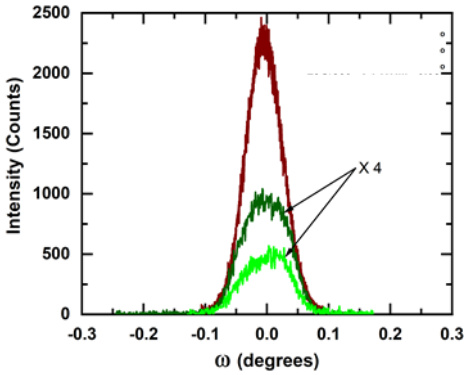












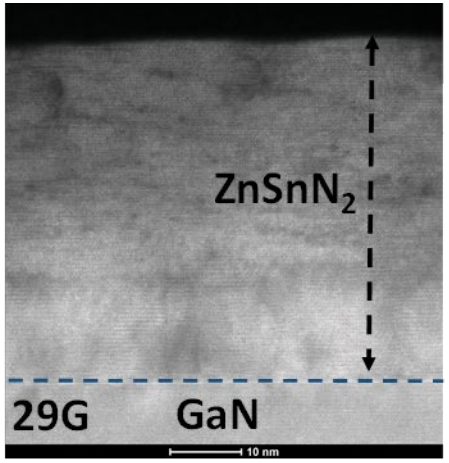
15G/520 °C-FWHM= 0.10°
28G/540 °C-FWHM= 0.09°
29G/560 °C-FWHM= 0.08°

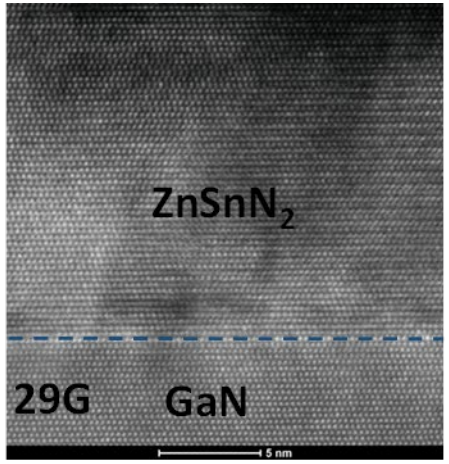
Growth Parameters

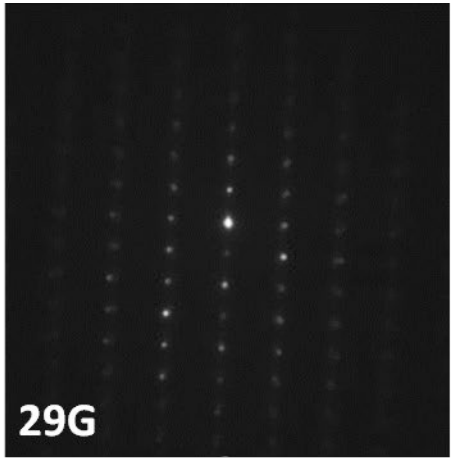
Sample	Temp. (°C)	Duration (min)	DEZn (µmol/min)	TMSn (µmol/min)	Zn:Sn flux ratio	NH_3 ($Lmin^{-1}$)
15G	520	90	22.7	21.5	1.1	4
20G	530	40	22.7	21.5	1.1	4
27G	535	30	22.7	21.5	1.1	4
28G	540	30	19.1	17.17	1.1	4.5
29G	550	20	42.7	36.48	1.2	4.5

TEM and AFM on ZTN29G

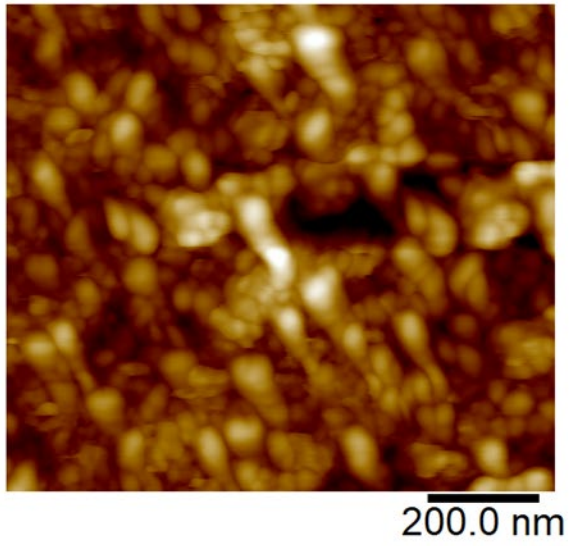
TEM

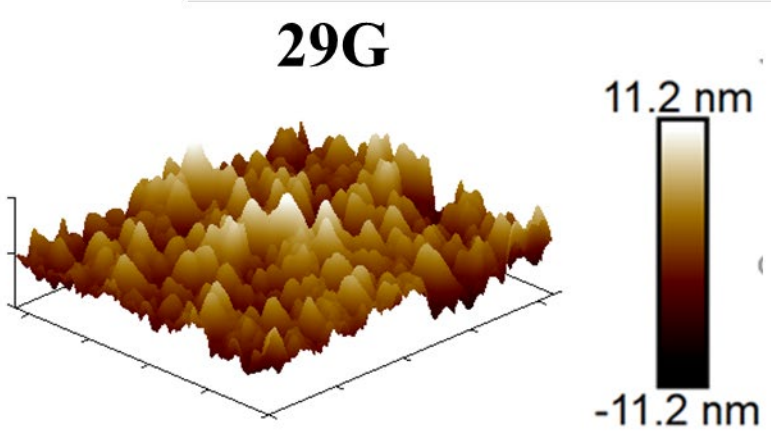






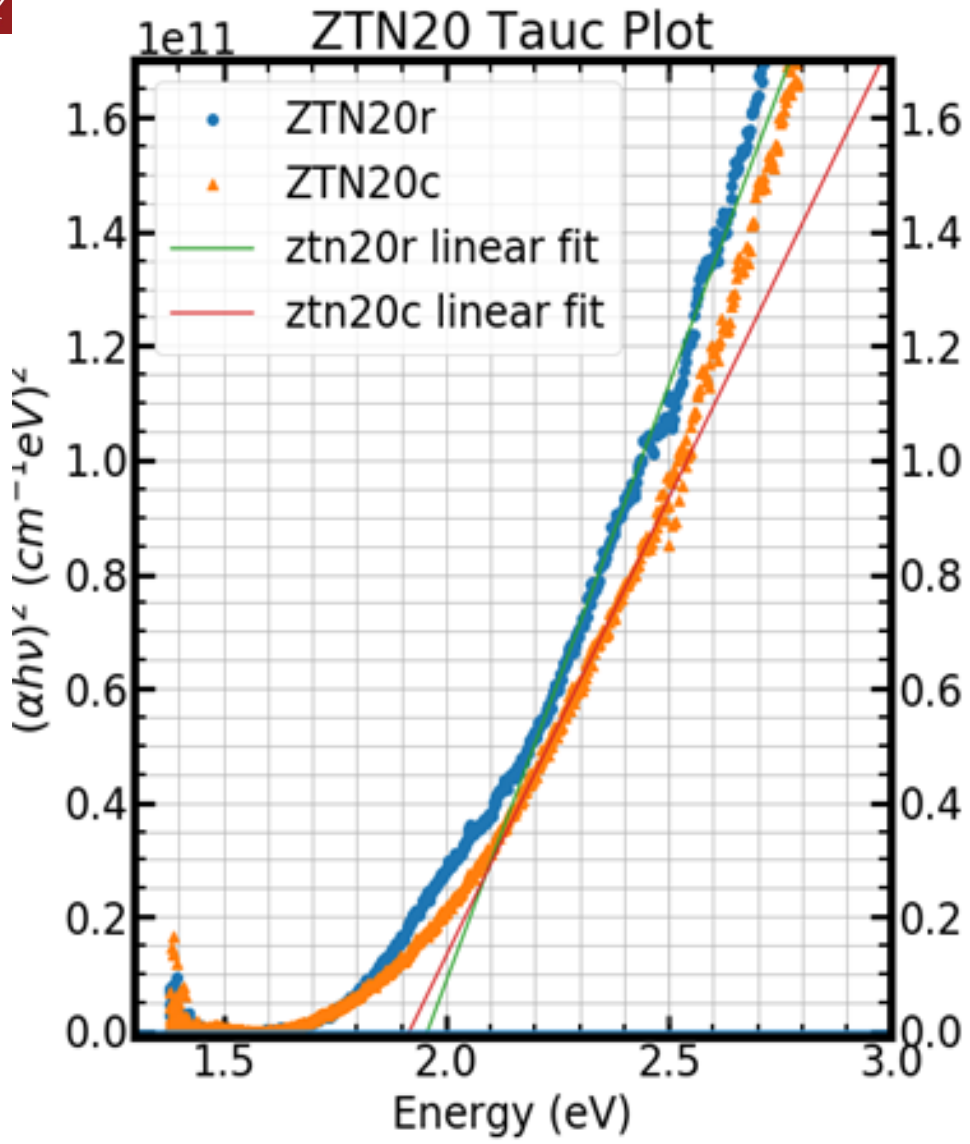
AFM

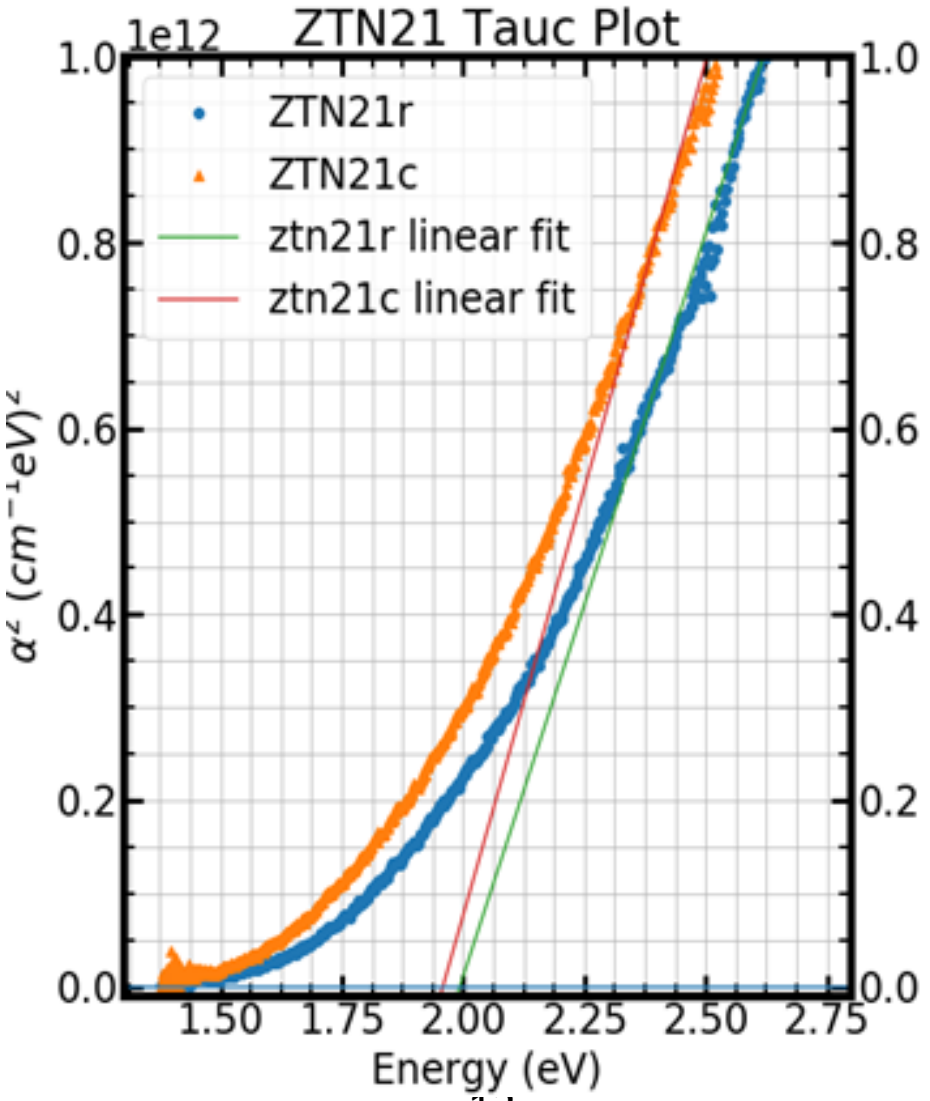




	28G	29G
Thickness (nm)	22.5±0.3	55.7±0.5
Growth rate (nm/hour)	45.0±0.6	167.1±1.0

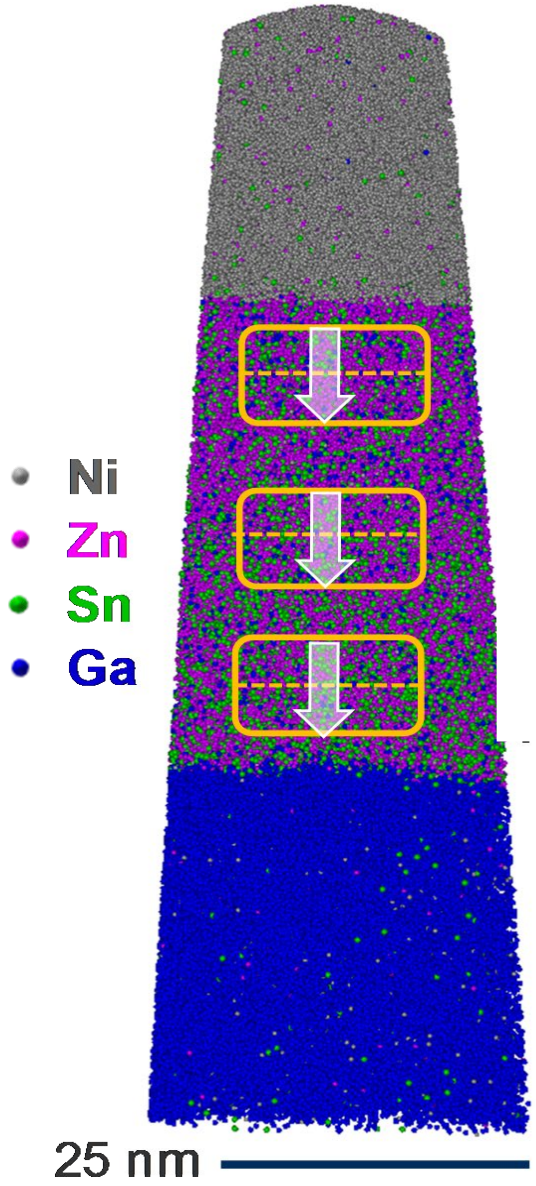
Deduce bandgap by Absorption spectra

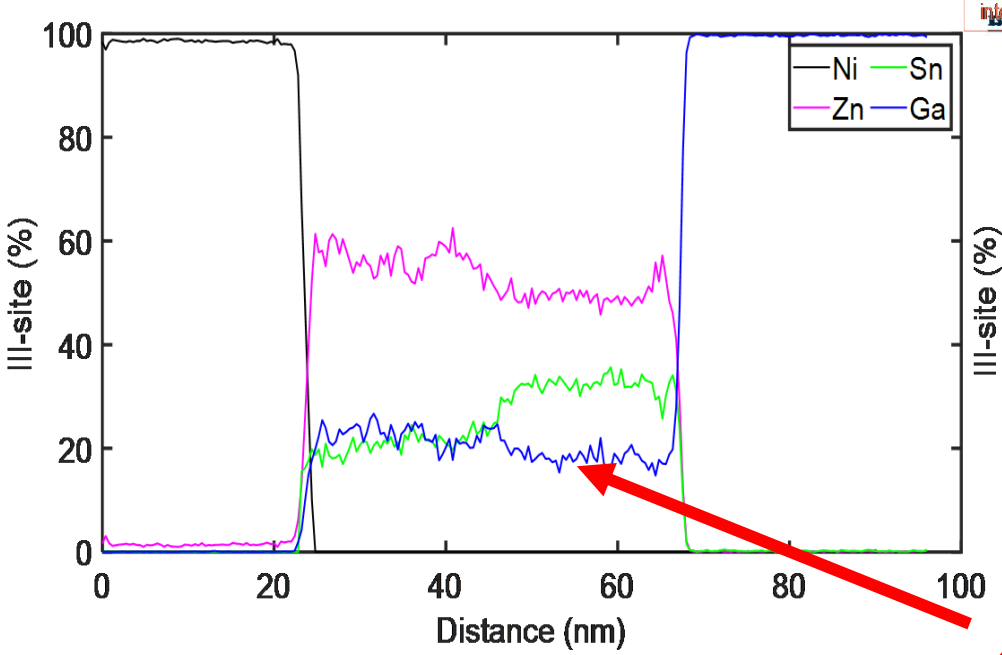




APT results show Ga contamination during growth of sample 33

Ni cap
$ZnSnN_2$-15nm 530 C
$ZnSnN_2$-15nm 540 C
$ZnSnN_2$-20nm 560 C
GaN template





Ga

	Concentration
Ga	17.5 at%
Sn	31.8 at%
Zn	50.7 at%
Zn/(Zn+Sn)	61.5 %
Sn/(Zn+Sn)	38.5 %

Future Work

- Optimize $ZnSnN_2$ growths on higher quality, locally-grown InGaN templates
- Overcome the Ga contamination issue
- Measure the composition accurately by XPS
- Produce $ZnSnN_2$-based LED device structures

Benthara Hewage Dinushi Dinushi Jayatunga

Case Western Reserve University
Dept. of Physics

ACKNOWLEDGMENTS

- DMREF: SusChEM: 1533957 and by the U.S. Department of Energy under grant no. DOE SSL: DE-EE0008718.
- Rebecca Lalk, Absorption spectroscopy , Case Western Reserve University