

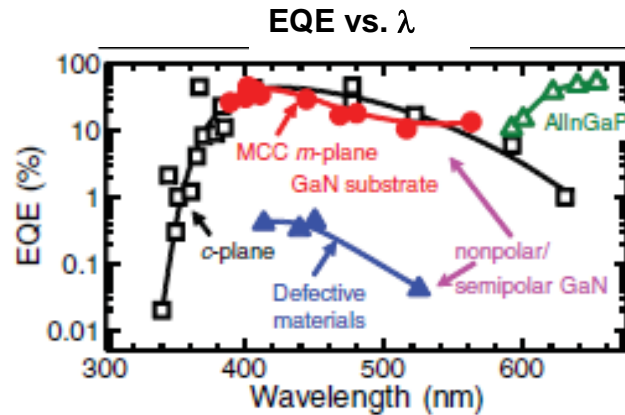
# **Role of defects in limiting the optical efficiency of InGaN/GaN quantum wells**

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Sandia National Laboratories  
Albuquerque, NM

Work at Sandia National Laboratories was supported by Sandia's Solid-State Lighting Science Energy Frontier Research Center, funded by the U.S. Department of Energy, Office of Basic Energy Sciences and by EERE/NETL, United States Department of Energy under project number M6802094, Sean Evans, Program Manager. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the U.S. Department of Energy's National Security Administration under Contract DE-AC04-94AL85000.

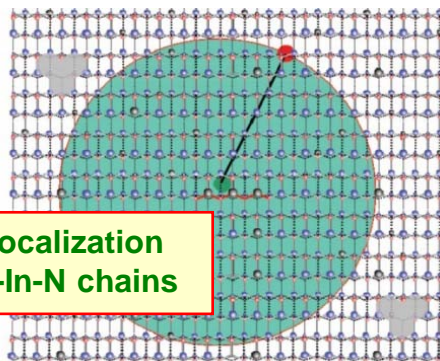
# Potential impact of defects in InGaN luminescence

How do InGaN defects impact the “green gap” and “efficiency droop” problems?



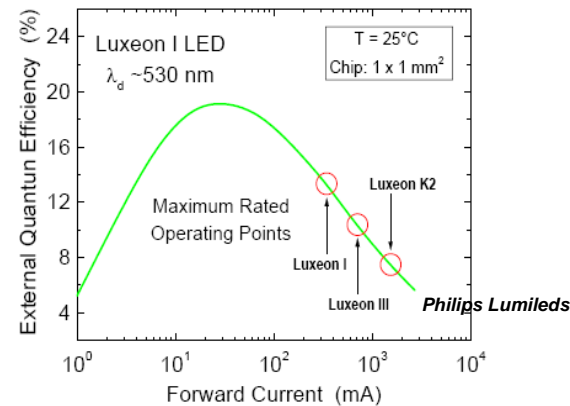
Speck *et al.* MRS Bullet. 34, 304 (2009)

## Defects and localization

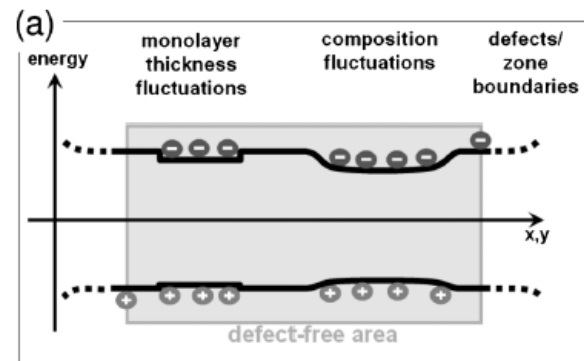


Chichibu *et al.* Philos. Mag. 87 2019 (2007)

## “Efficiency Droop” of InGaN LEDs



## Defects and localization

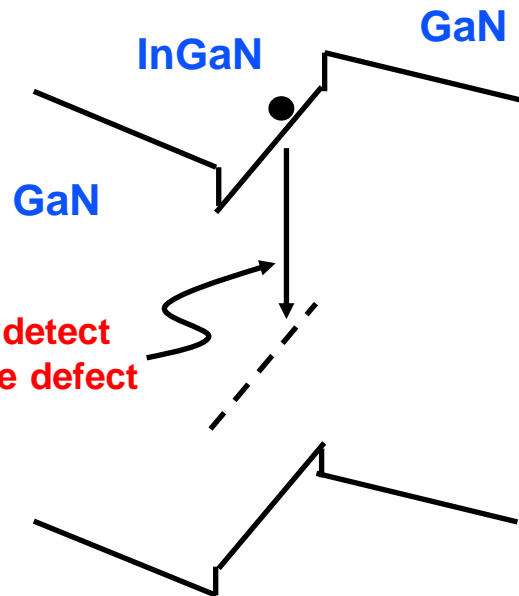


Hader *et al.* APL 96 221106 (2010)

➤ Connect InGaN QW growth conditions, defects and IQE

# Defect Study of InGaN QW structures

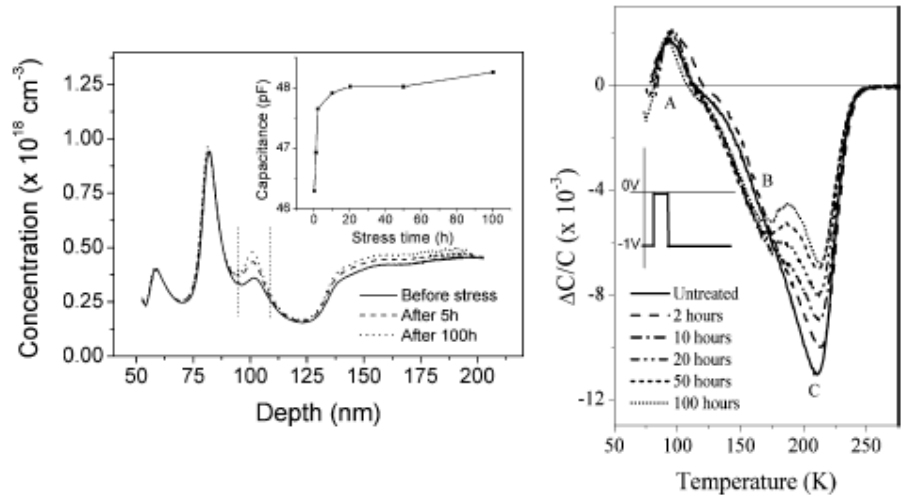
## Resonant Photoluminescence



### Drawbacks:

1. Non-radiative centers?
2. Quantify deep level energy, concentration?

## DLTS of InGaN LEDs



1. F. Rossi *et al.* JAP 99, 053104 (2006).

### Drawbacks:

1. DLTS cannot probe mid-gap states
2. Cannot determine defect density
3. No InGaN-related traps were found<sup>1,2</sup>

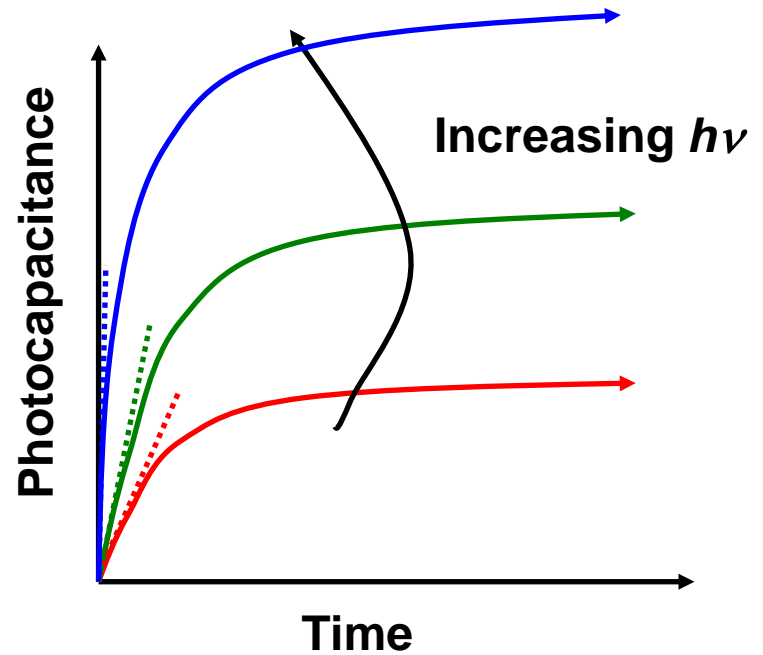
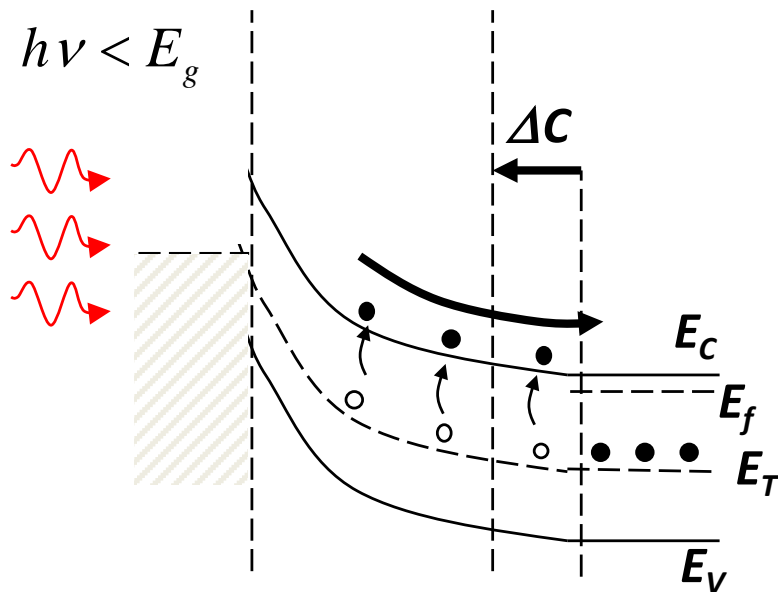
**QW-based techniques not quantitative or have insufficient depth resolution**

2. L. Rigutti *et al.* PRB 77, 045312 (2008).

# Deep Level Optical Spectroscopy

## Deep Level Optical Spectroscopy (DLOS)<sup>1</sup>

- Photocapacitance technique
  - Sub-band gap optical stimulation to photoionize defect levels (reverse of PL)
  - Quantify non-radiative defect level energy and density (difficult for PL)



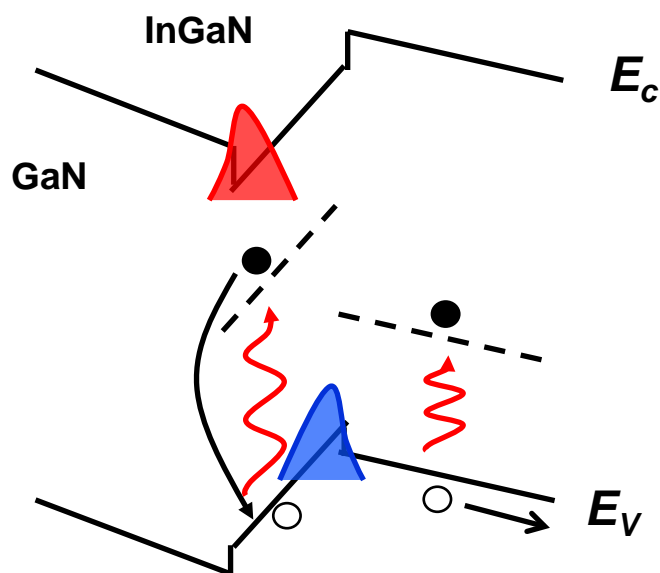
- DLOS only sensitive to depleted regions
- Enables nano-scale depth resolution

- Optical cross-section  $\sigma^o = e^o / \Phi(h\nu)$ 
  - $dC(t)/dt|_{t=0} \propto \sigma^o(h\nu)$
- Optical ionization energy  $E^o$
- Defect density  $D_t$  from lighted C-V

# Defect Study of InGaN QWs

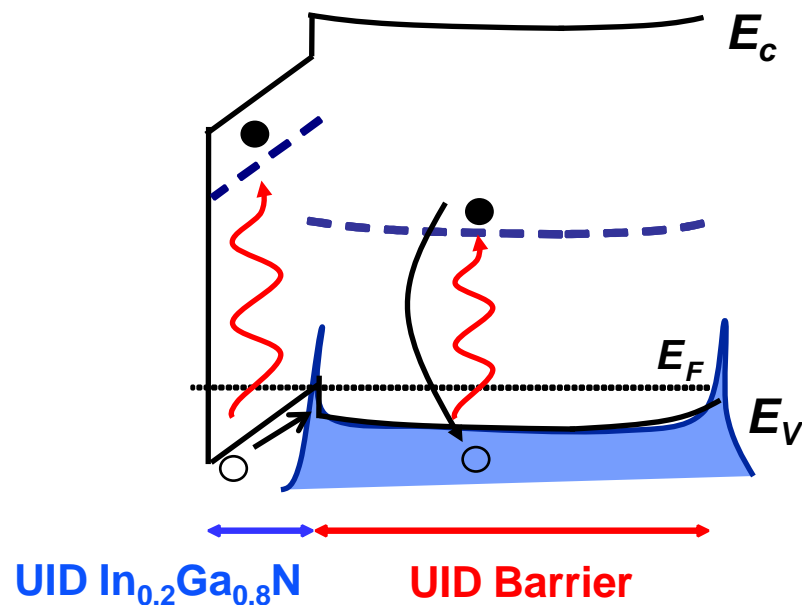
Designed InGaN/GaN heterostructure for depth-resolved DLOS

## InGaN/GaN QW



Not sensitive to  $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$  well defects

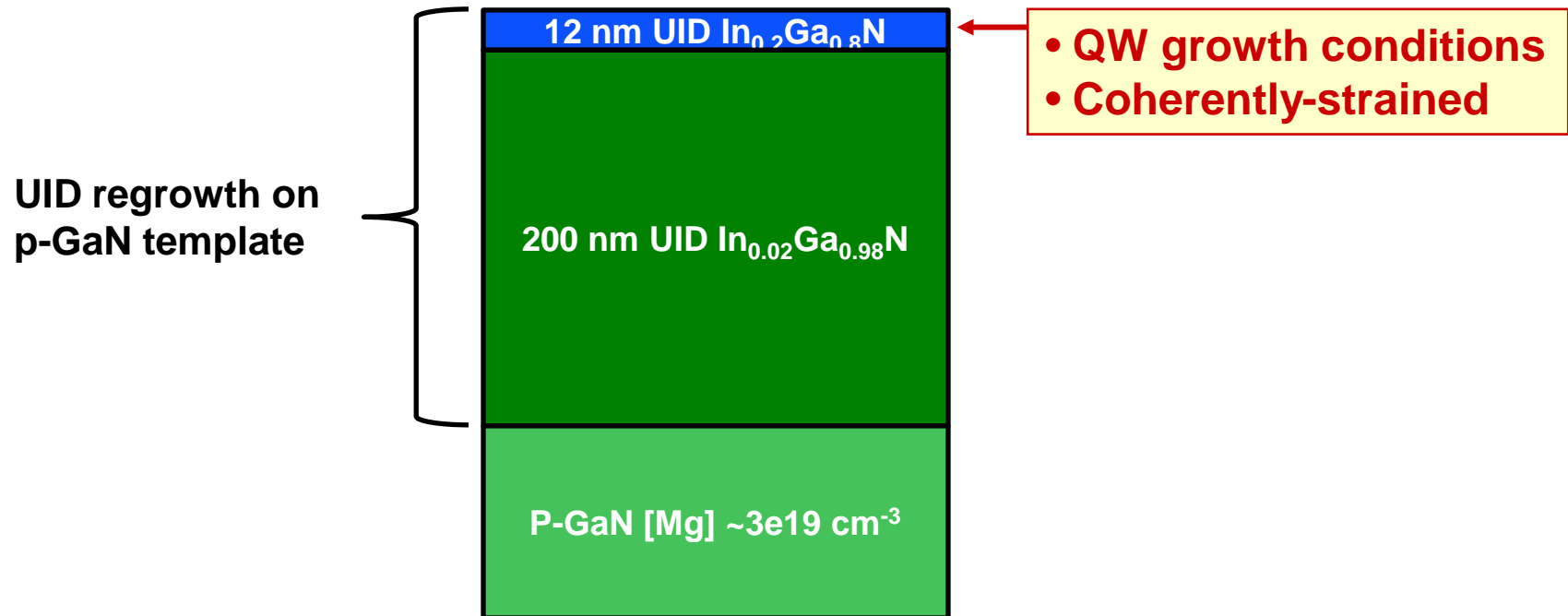
## InGaN/GaN “half QW”



Mainly sensitive to  $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$  cap defects

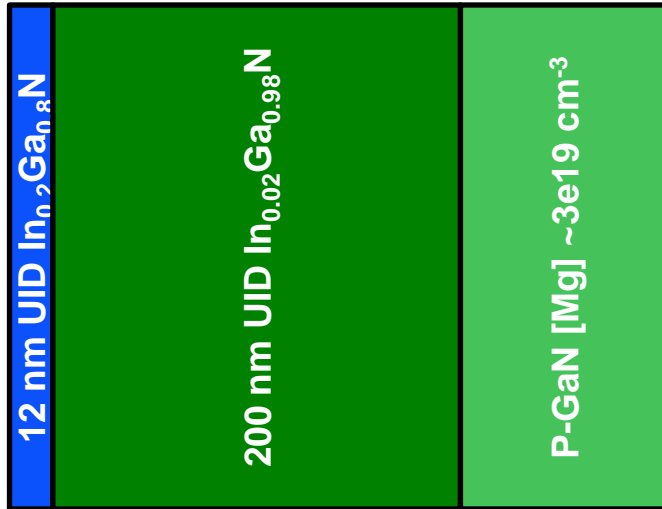
**Same  $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$  growth conditions for both structures**

# Depth-resolved DLOS of InGaN

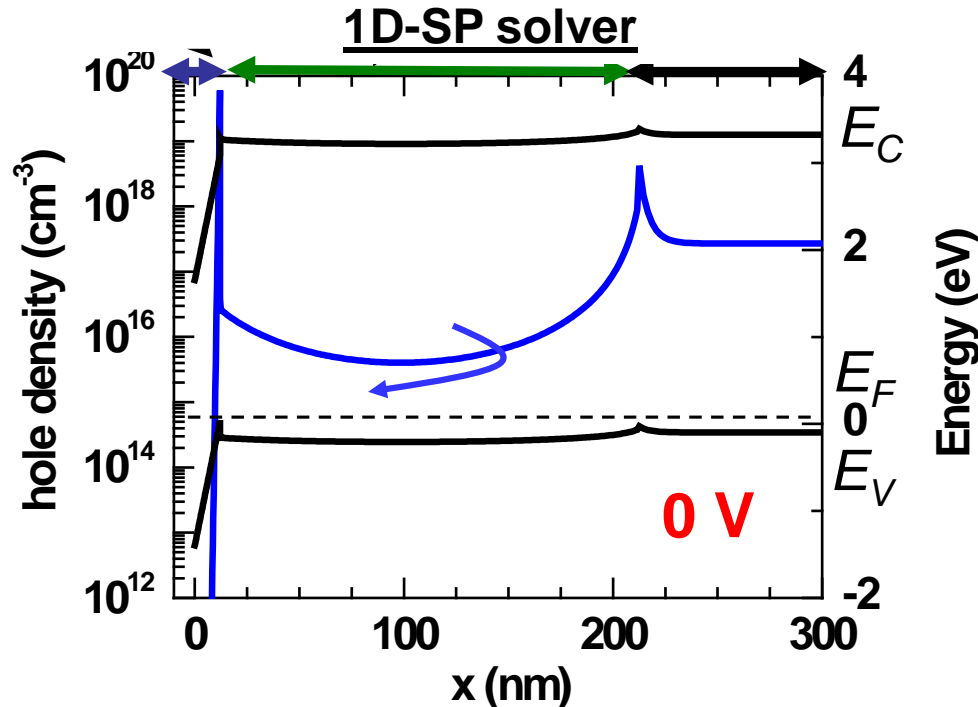


- Grown by MOCVD in a D125 short-jar Veeco reactor
- 300 torr, 15 SLM  $\text{NH}_3$ , 10 SLM  $\text{N}_2$ , no  $\text{H}_2$
- In content controlled through growth temperature
- $\text{In}_{0.02}\text{Ga}_{0.83}\text{N}$ : 24.3  $\mu\text{moles/min Ga}$ , 72.8  $\mu\text{moles/min In}$
- $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$ : 10.4  $\mu\text{moles/min Ga}$ , 55.8  $\mu\text{moles/min In}$

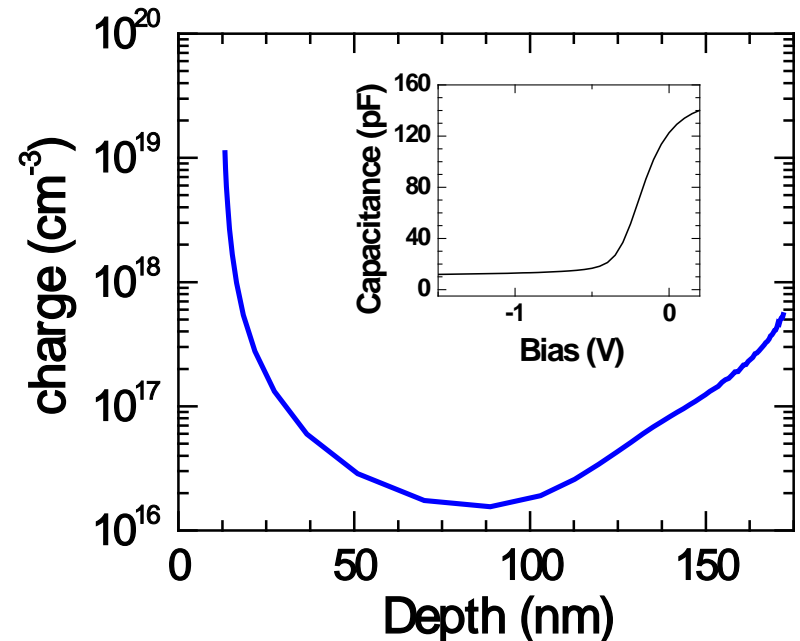
# Depth-resolved DLOS of InGaN



➤ Sensitivity to  $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$  defects



## Diode C-V and carrier density



# Influence of $T_g$ and Indium alloying

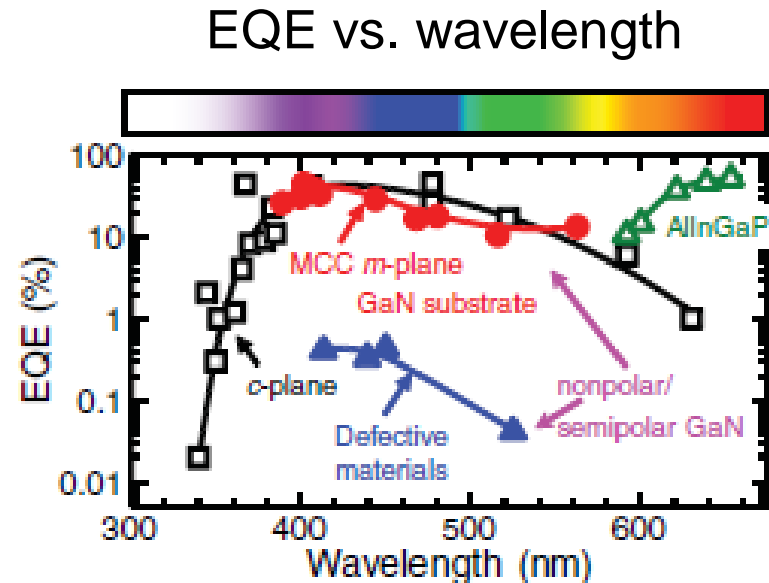
- Quantitatively study defects and IQE as function of same InGaN growth conditions

Decreasing  $T_g$   
(Impurities, native point defects)

750 °C  
17%  
Indium

730 °C  
21%  
Indium

Increasing Indium  
(Strain, QCSE)



Speck et al. MRS Bullet. 34, 304 (2009)

- Decrease in EQE/IQE with increasing  $\lambda$
- Partly QCSE, well width, and defects....?

- Decouple  $T_g$  vs. indium alloying



# Effect of $T_g$ on MQW IQE

Decreasing  $T_g$

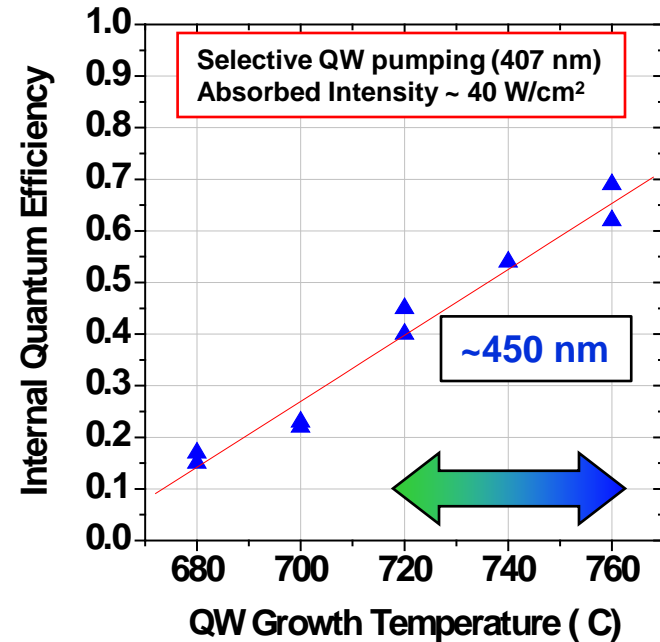
(Impurities, native point defects)

750 °C  
17%  
Indium

690 °C  
17%  
Indium

Reduced  $T_{QW}$  at fixed Indium

Estimated IQE from Temp. Dep. PL

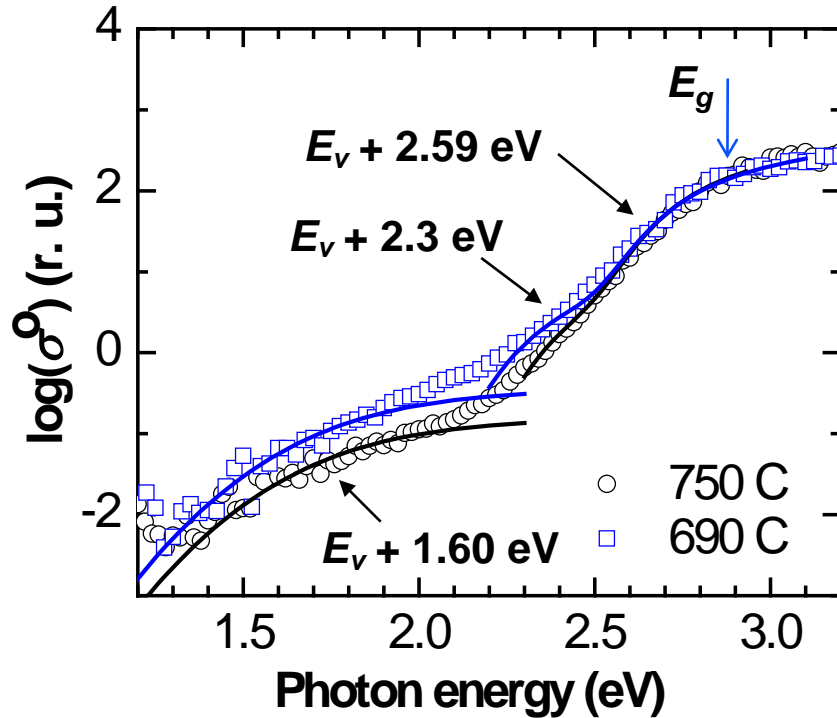


- Isolate impact of lower  $T_g$ 
  - Vary  $T_{MIn}$  flow only for similar Indium composition at various  $T_g$
- Fixed MQW structure, strain, QCSE
- Fixed [V-defect], TDD

- Decreasing IQE with decreasing  $T_{QW}$
- Point defects are suspected cause

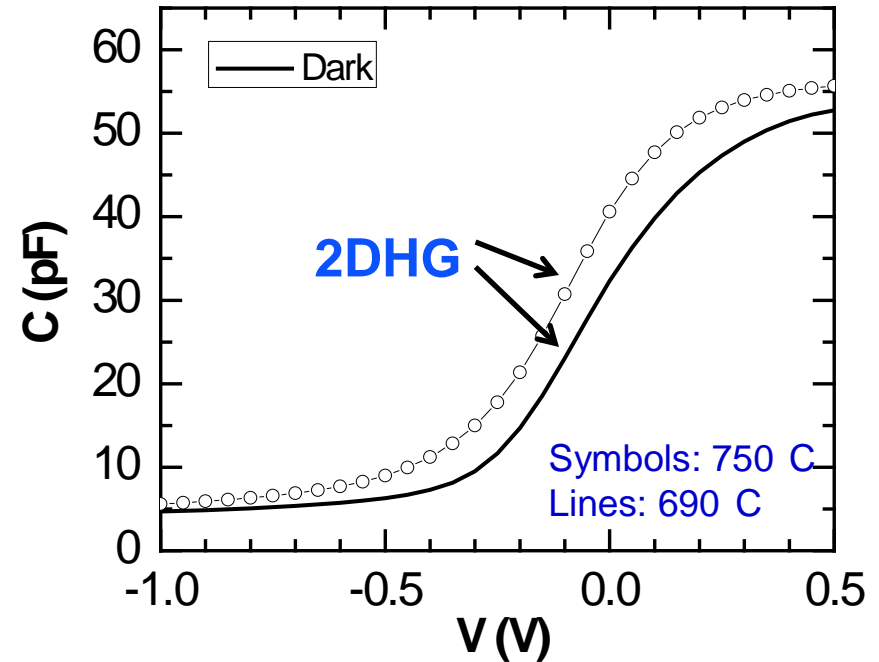
# Influence of $T_g$ on defect incorporation

## DLOS of $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$



- Similar  $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$  defect levels  
 $\rightarrow$  60 C reduction in  $T_g$  does not introduce new deep levels

## Lighted C-V

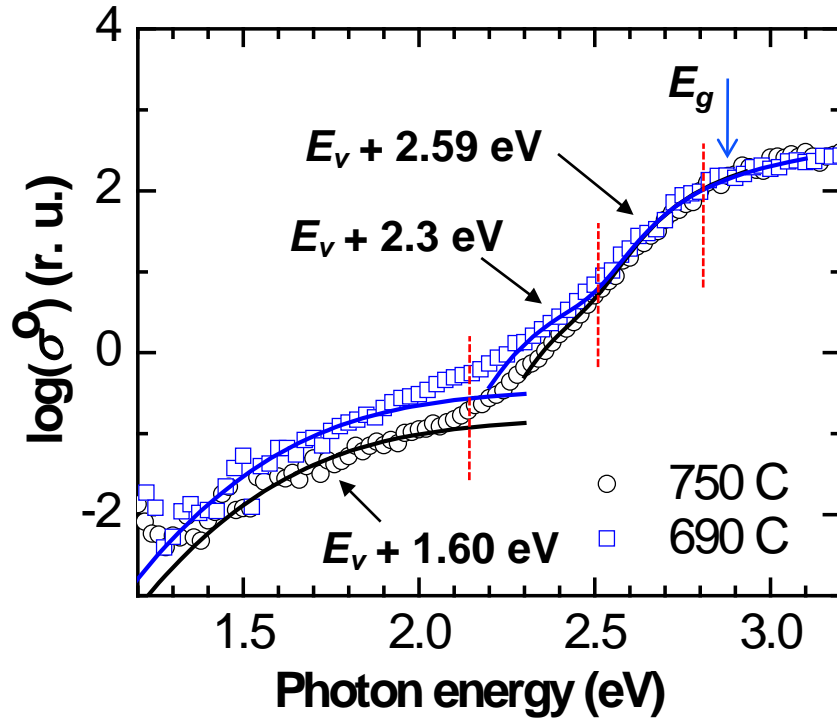


- Larger dark "threshold voltage" at 750C  
 $\rightarrow$  60 C reduction in  $T_g$  introduces excess deep levels

Model used to fit DLOS data:  
 Paessler JAP 96, 715 (2004)

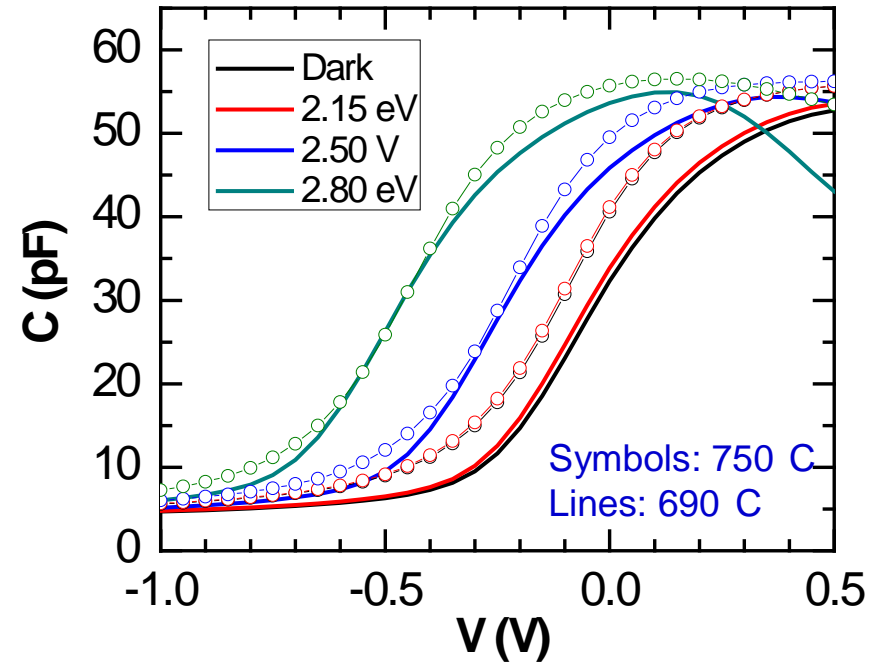
# Influence of $T_g$ on defect incorporation

## DLOS of $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$



- Similar  $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$  defect levels  
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## Lighted C-V



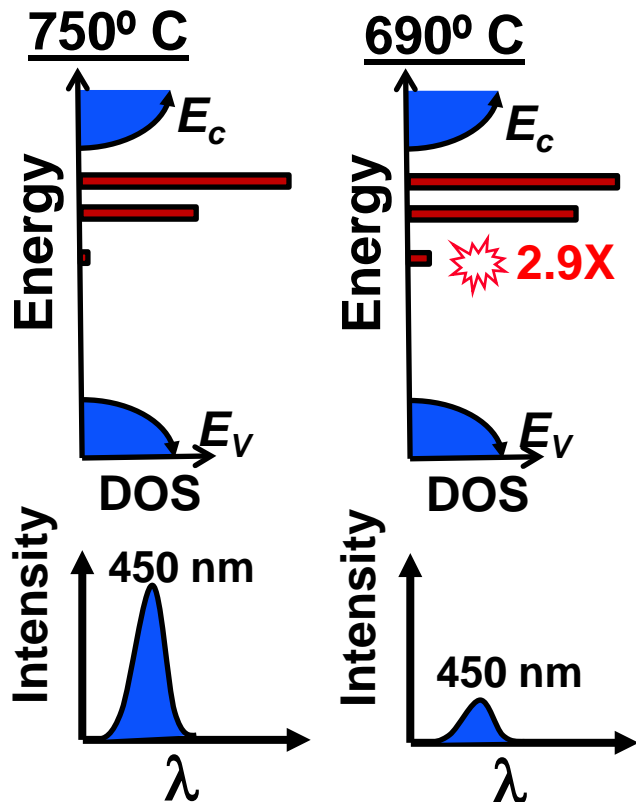
- Larger dark “threshold voltage” at 750C  
 $\rightarrow$  60 C reduction in  $T_g$  introduces excess deep levels
- Individual areal deep level densities ( $D_t$ ) using selective photon excitation energy

Model used to fit DLOS data:  
 Paessler JAP 96, 715 (2004)

# Influence of $T_g$ on defect incorporation

## Areal Densities of Deep Levels

	690 C	750 C	Change
$E_v + 1.60$ eV	$2.0 \cdot 10^{11}$	$0.70 \cdot 10^{11}$	<b>2.9X</b>
$E_v + 2.3$ eV	$1.6 \cdot 10^{12}$	$1.1 \cdot 10^{12}$	<b>1.4X</b>
$E_v + 2.59$ eV	$2.0 \cdot 10^{12}$	$2.0 \cdot 10^{12}$	--



- $[E_v + 2.59$  eV] largest but no change for  $\downarrow T_g$
- $[E_v + 1.60$  eV] greatest increase for  $\downarrow T_g$

➤ Quantified  $\uparrow D_t$ ,  $\downarrow$  IQE for  $\downarrow T_g$

### Possible Deep Level Candidates:

1. Carbon related ( $\uparrow$  with lower  $T_g$ <sup>1</sup>, nonradiative<sup>2</sup>)
2.  $V_{III}$  and Oxygen related ( $\uparrow$  with lower  $T_g$ , low formation energy of  $V_{III}$ -O complexes<sup>3</sup>)

[1] D. D. Koleske, et al., JCG 242, 55 (2002)

[2] C. H. Seager et al., JAP 92, 6553 (2002)

[3] J. Neugebauer and C. G. Van de Walle, APL 69, 503 (1996)

# Influence of $T_g$ and indium alloying

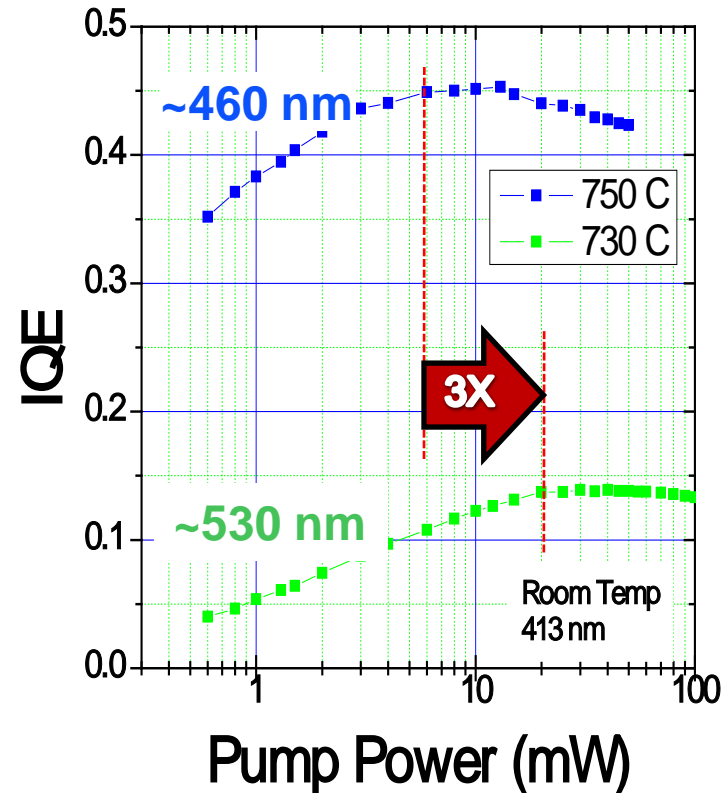
Decreasing  $T_g$   
(Impurities, point defects)

750 °C  
17%  
Indium

730 °C  
21%  
Indium

Increasing Indium  
(Strain, QCSE)

## IQE from Power Dep. PL

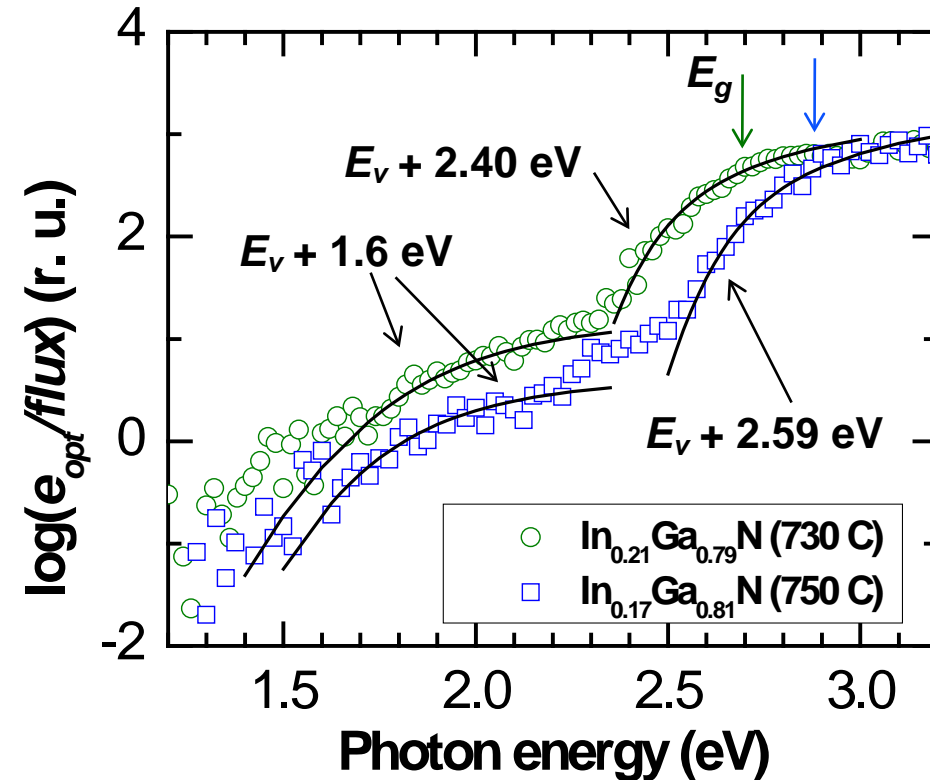


- Strongly reduced IQE
- **3X saturation power suggests more defect recombination for green MQW**

**Are the  $E_v + 1.60$  eV,  $E_v + 2.3$  eV levels relevant for green MQWs?**

# Influence of $T_g$ and indium alloying

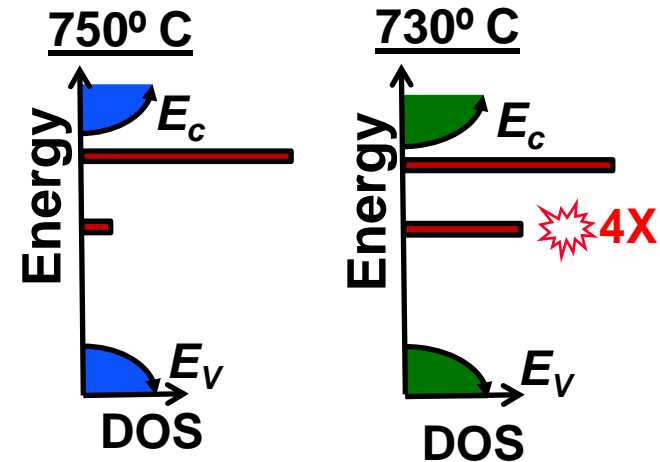
In<sub>0.17</sub>Ga<sub>0.83</sub>N vs. In<sub>0.21</sub>Ga<sub>0.79</sub>N



- Near- $E_c$  level shifts with  $E_g$
- Similar  $E_v + 1.6$  eV deep level
- 2.3 eV level missing

## Densities of Deep Levels

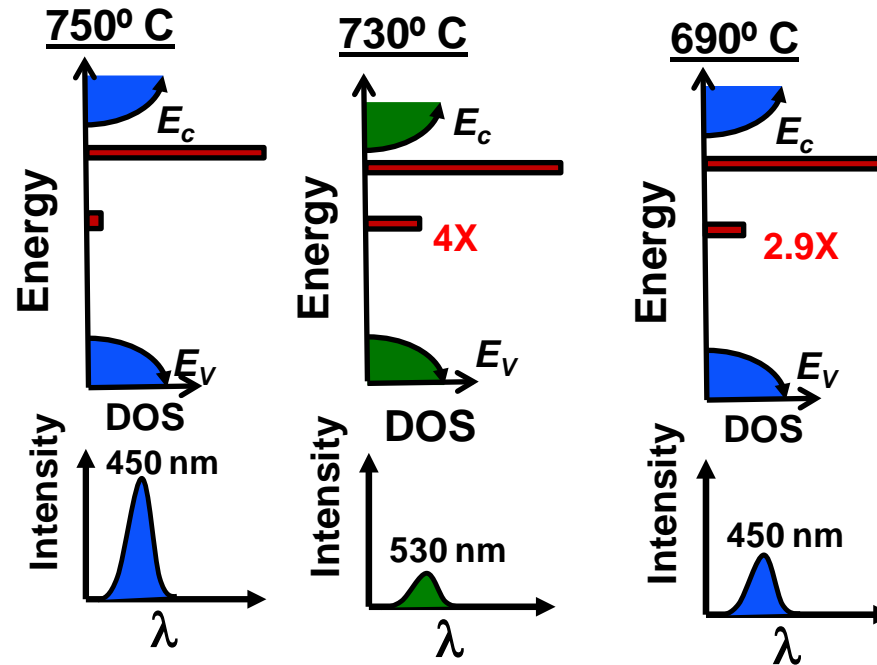
	In <sub>0.17</sub> Ga <sub>0.83</sub> N	In <sub>0.21</sub> Ga <sub>0.79</sub> N
$E_v + 1.6$ eV	7e10 cm <sup>-2</sup>	28e10 cm <sup>-2</sup>
$E_v + 2.40$ eV	5e11 cm <sup>-2</sup>	--
$E_v + 2.59$ eV	--	5e11 cm <sup>-2</sup>



→ **Accelerated** IQE drop and greater  
[ $E_v + 1.6$  eV] with ↓  $T_g$  and ↑ indium

# Conclusions and summary

Used DLOS, Lighted C-V and PL to quantitatively study how QW growth conditions and alloying influence defect incorporation and IQE



Reduced  $T_g$  at fixed indium (blue MQWs):

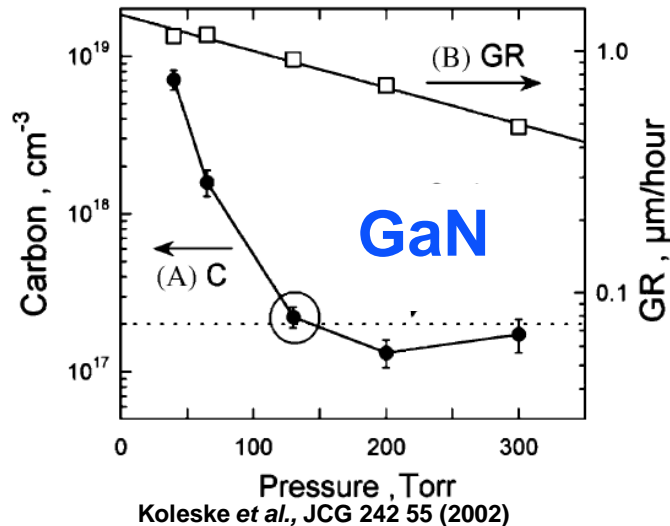
- Does not incorporate new deep levels
- Enhanced [ $E_v + 1.6$  eV] correlates with strongly reduced IQE

Reduced  $T_g$  to increase indium (green MQWs):

- IQE degradation and [ $E_v + 1.6$  eV] further enhanced
- Identify  $E_v + 1.6$  eV deep level as an effective NRC contributing to efficiency roll-off with increasing wavelength

# Future work

## Impurity incorporation and growth pressure

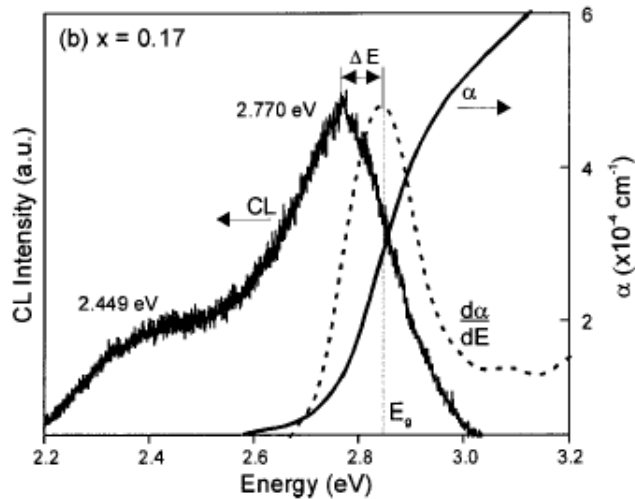


- MQW growth optimization
  - Expect that  $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$  growth pressure is important
  - Examine influence of precursor point and defect incorporation
- Study  $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$  thermal decomposition
  - Design “half-QWs” with high temp. GaN caps
  - In-situ thermal cycling to mimic LED growths

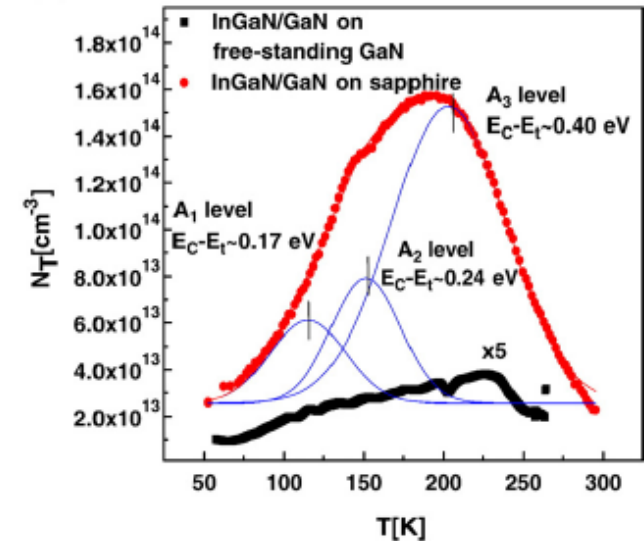


# Defect Study of "bulk" InGaN films

## Cathodoluminescence of 100 nm thick $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$



## DLTS of 200 nm thick $\text{In}_{0.14}\text{Ga}_{0.86}\text{N}$



1. S. Srinivasan *et al.* APL 80, 550 (2002).

2. C. Soh *et al.* Thin Solid Films 515, 4509 (2007).

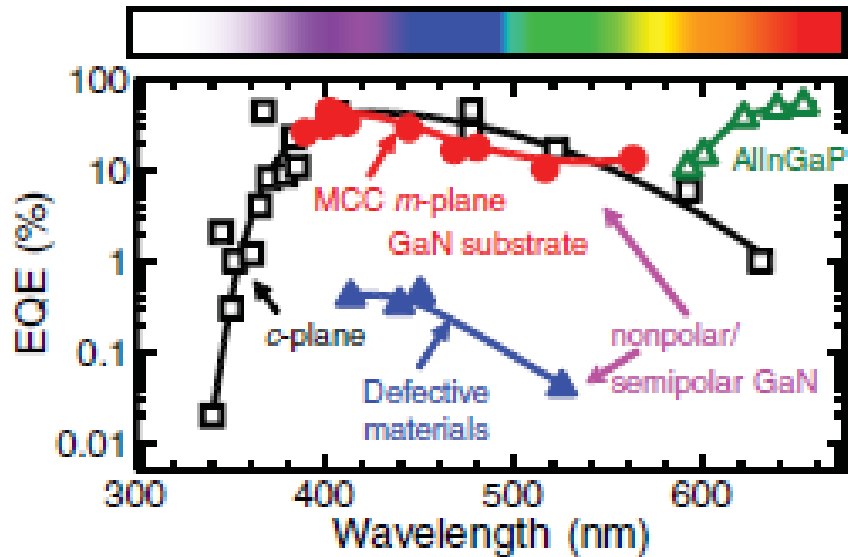
- $\text{In}_x\text{Ga}_{1-x}\text{N}$  film quality degrades for thick layers ( $x > 0.15$ )
  - Strain relaxation and surface roughening
  - V-defect expansion
  - Indium segregation

**Unclear that "bulk"  $\text{In}_x\text{Ga}_{1-x}\text{N}$  reflects optical quality InGaN wells**

# Potential impact of defects for InGaN LEDs

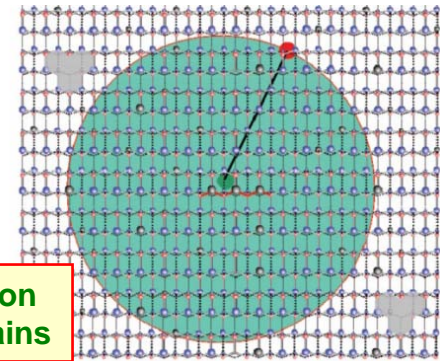
## InGaN defects and efficiency

### InGaN EQE vs. wavelength

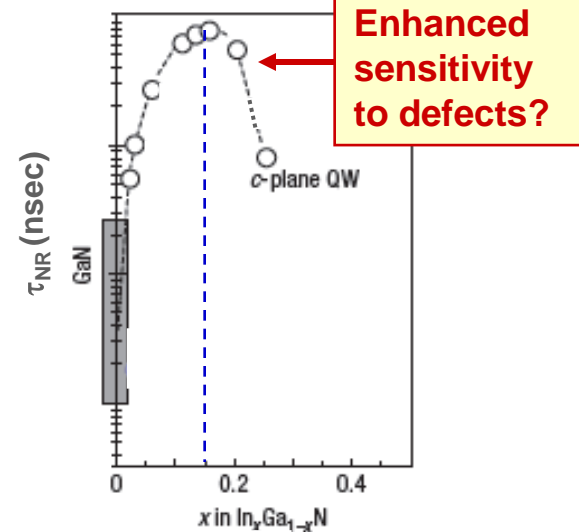


Speck *et al.* MRS Bullet. 34, 304 (2009)

Hole localization  
at In-N-In-N chains



Chichibu *et al.* Philos. Mag. 87 2019 (2007)



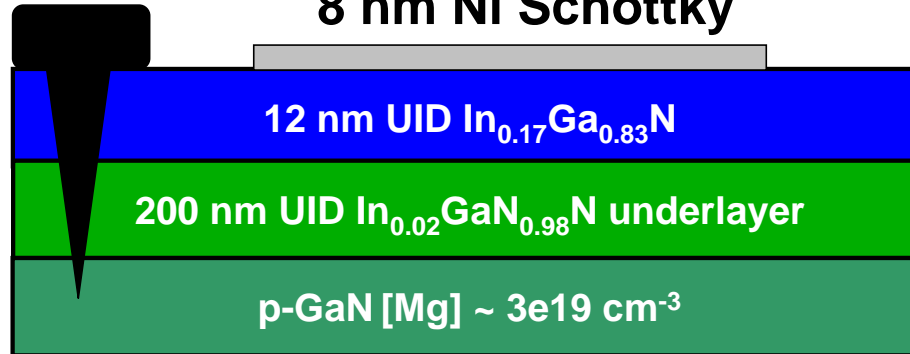
## Efficiency roll-off not just QCSE

- Connect InGaN QW growth conditions, defects and IQE
- Deep Level Optical Spectroscopy (DLOS) for InGaN epilayers

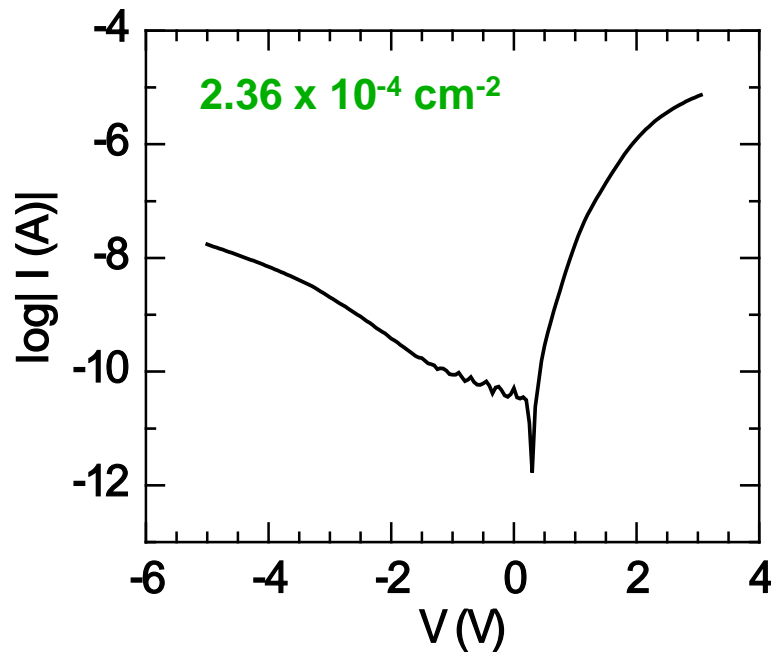
# Diode structure

In contact

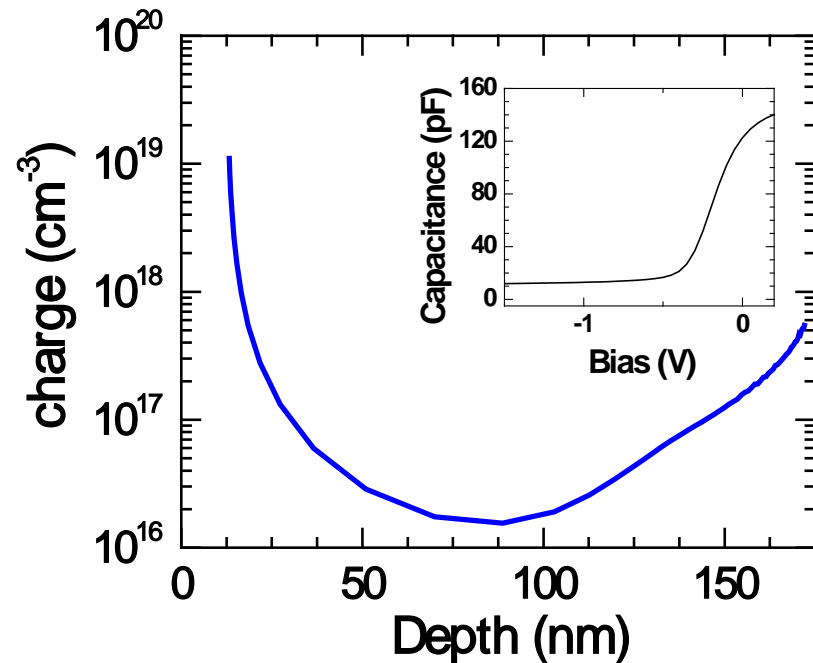
8 nm Ni Schottky



I-V

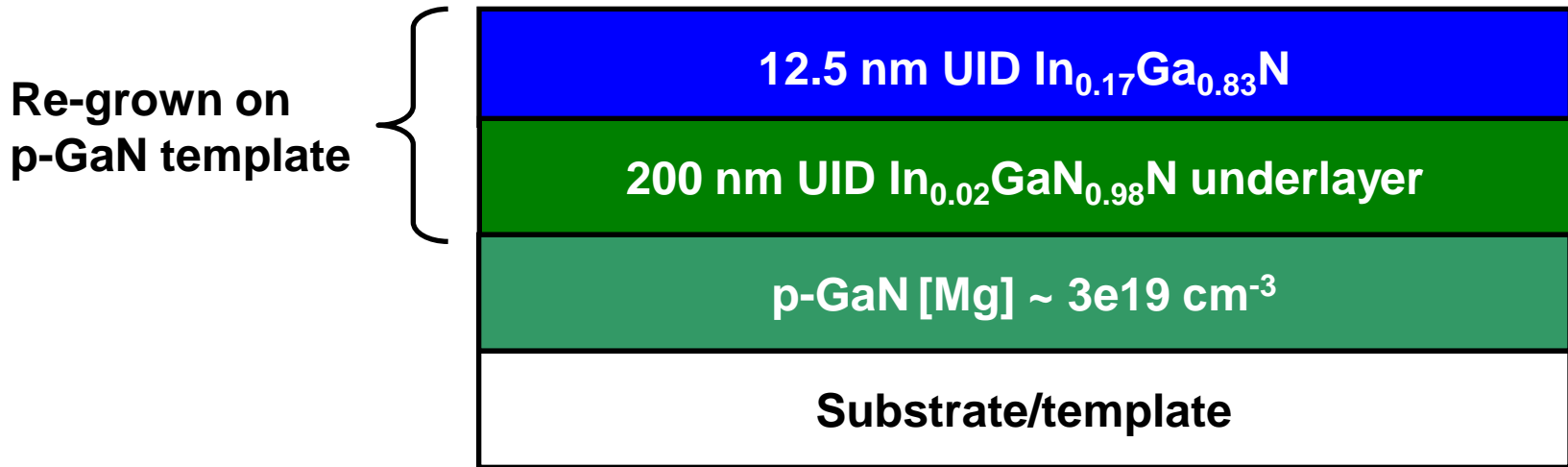


C-V and carrier density



# Growth of InGaN/GaN DLOS structure

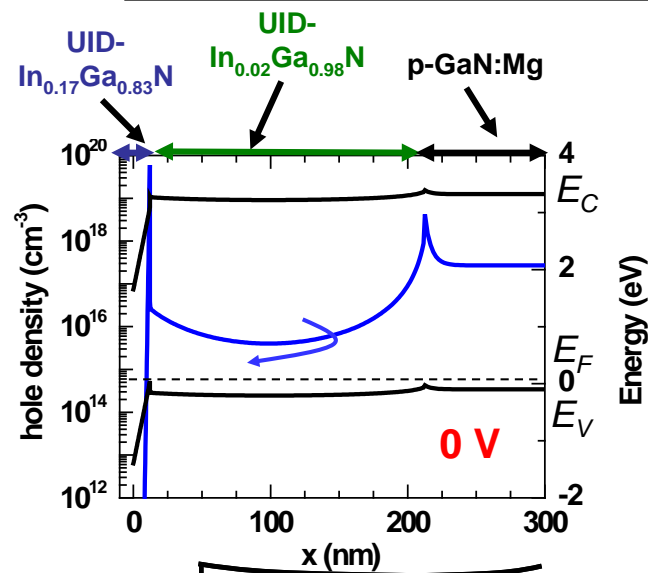
$\text{In}_x\text{Ga}_{1-x}\text{N}$  layers grown under nominal QW conditions



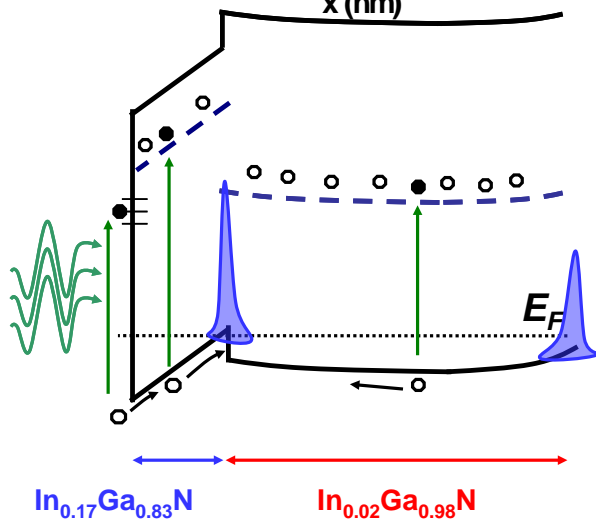
- MOCVD UID-InGaN re-grown on MOCVD p-GaN:Mg template
- $\text{In}_{0.02}\text{Ga}_{0.83}\text{N}$  underlayer (UL) grown at 880 C for 60 min.
- $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$  "well" grown at 760 C for 7 min.

- Grown by MOCVD in a D125 short-jar Veeco reactor
- 300 torr, 15 SLM  $\text{NH}_3$ , 10 SLM  $\text{N}_2$ , no  $\text{H}_2$
- In content controlled through growth temperature
- $\text{In}_{0.02}\text{Ga}_{0.83}\text{N}$ : 24.3  $\mu\text{moles/min}$  Ga, 72.8  $\mu\text{moles/min}$  In
- $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$ : 10.4  $\mu\text{moles/min}$  Ga, 55.8  $\mu\text{moles/min}$  In

# Simulate Depth-resolved DLOS

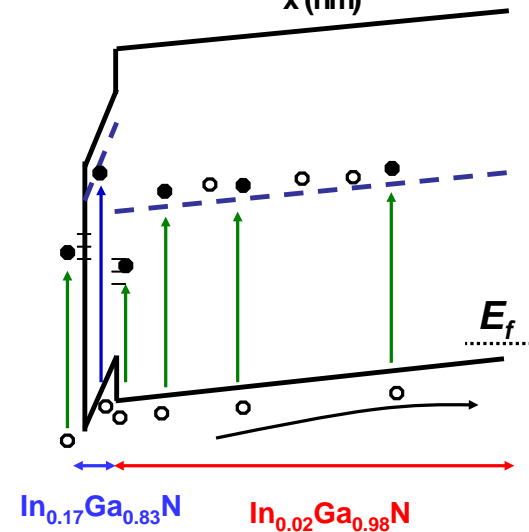
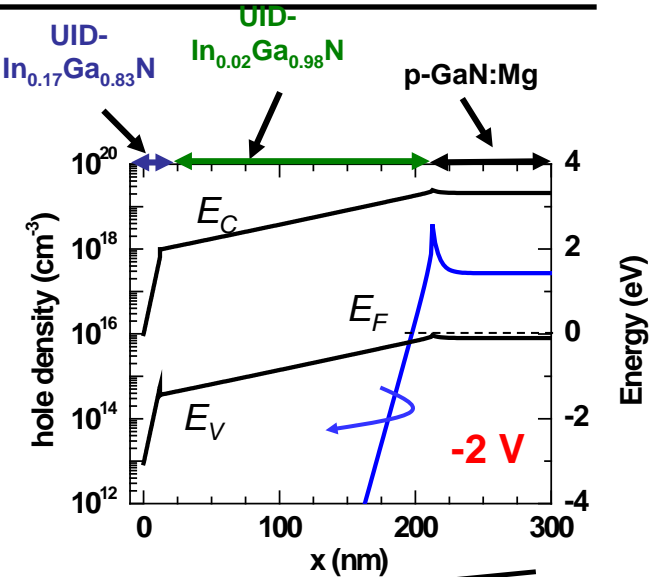


p-GaN never depleted



## Accumulation:

- "well" and portion of UL depleted
- $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$  defects dominate DLOS
- Defects near 2DHG remain filled



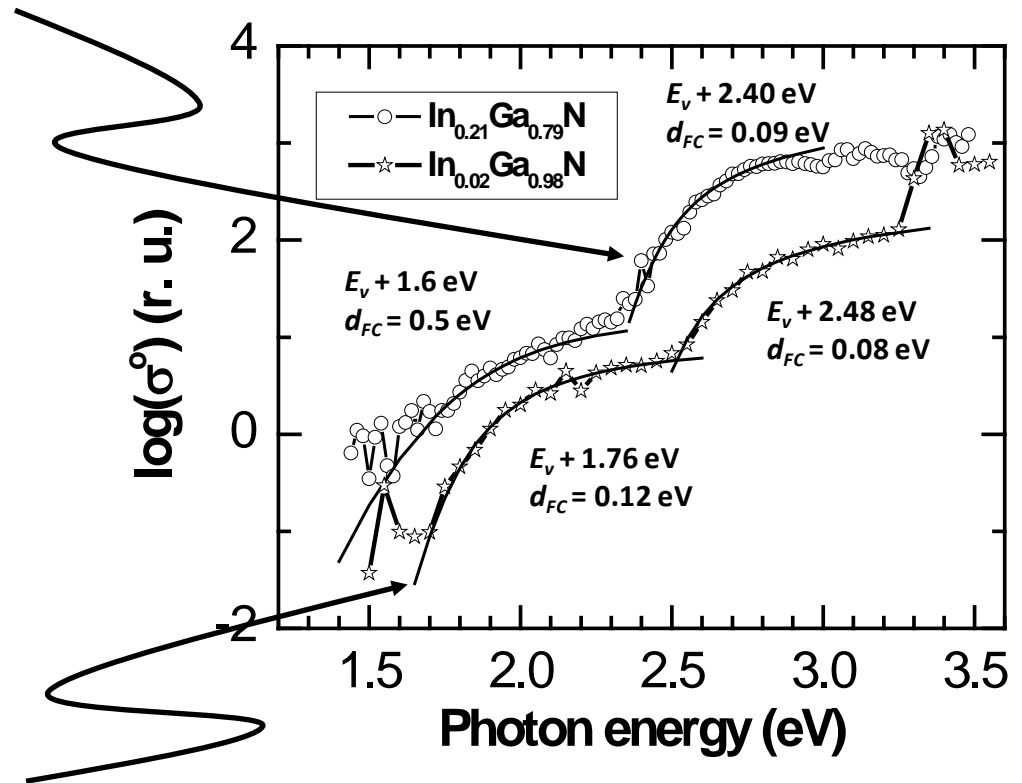
## Depletion:

- "well", interface, and UL depleted
- UL defects dominate DLOS
- Contribution from all InGaN regions

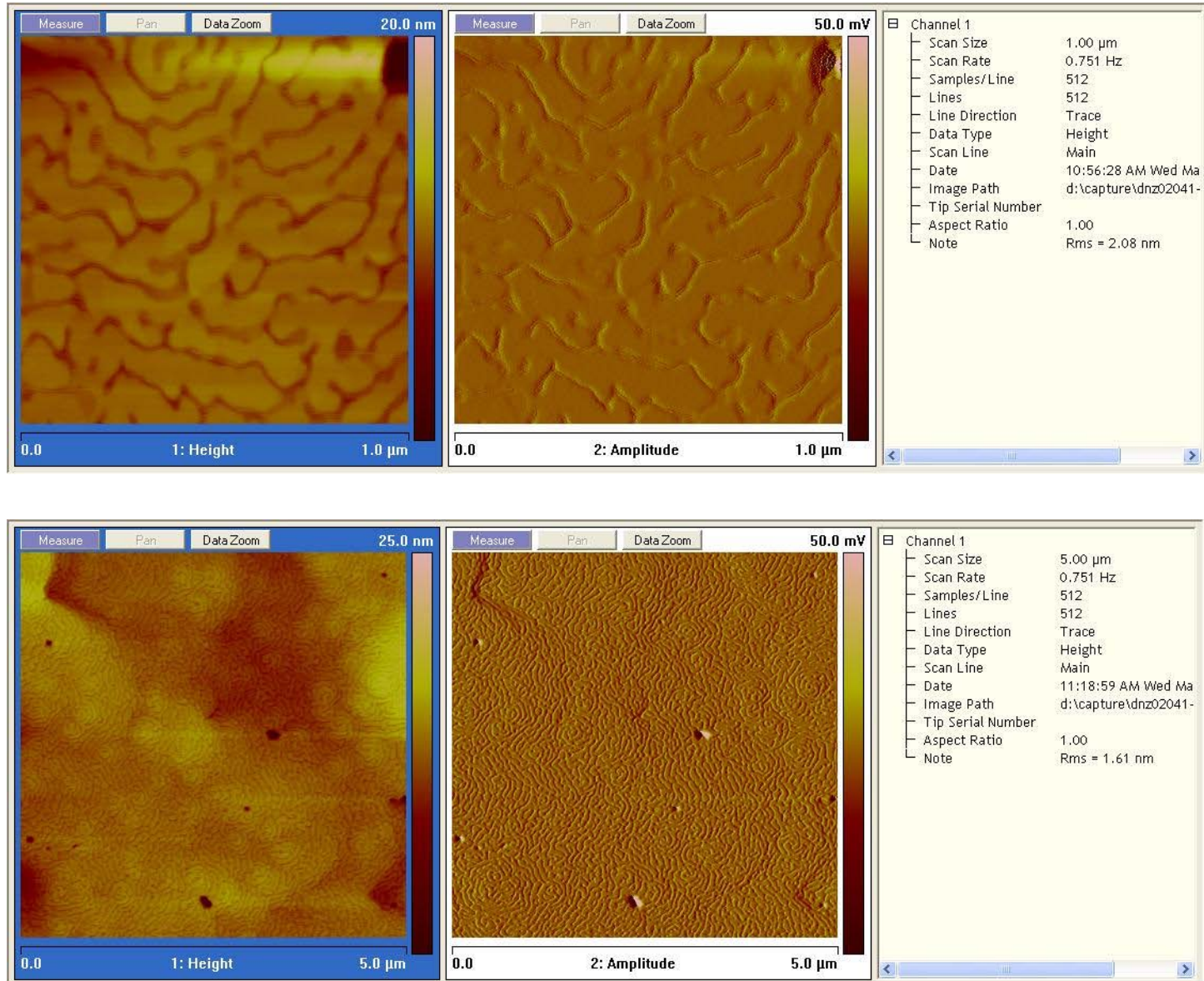
# Confirmed DL0S depth-resoltuion

~12 nm UID $\text{In}_{0.21}\text{Ga}_{0.79}\text{N}$
200 nm UID $\text{In}_{0.02}\text{Ga}_{0.98}\text{N}$ underlayer
p-GaN [Mg] ~ $3\text{e}19 \text{ cm}^{-3}$
Substrate/template

~4 nm UID $\text{In}_{0.21}\text{Ga}_{0.79}\text{N}$
200 nm UID $\text{In}_{0.02}\text{Ga}_{0.98}\text{N}$ underlayer
p-GaN [Mg] ~ $3\text{e}19 \text{ cm}^{-3}$
Substrate/template

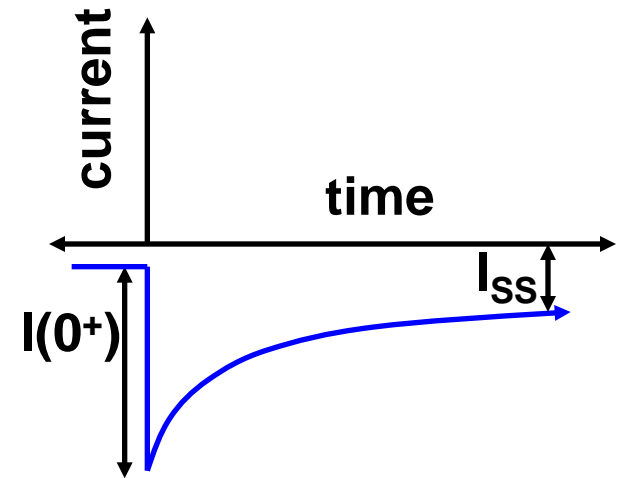
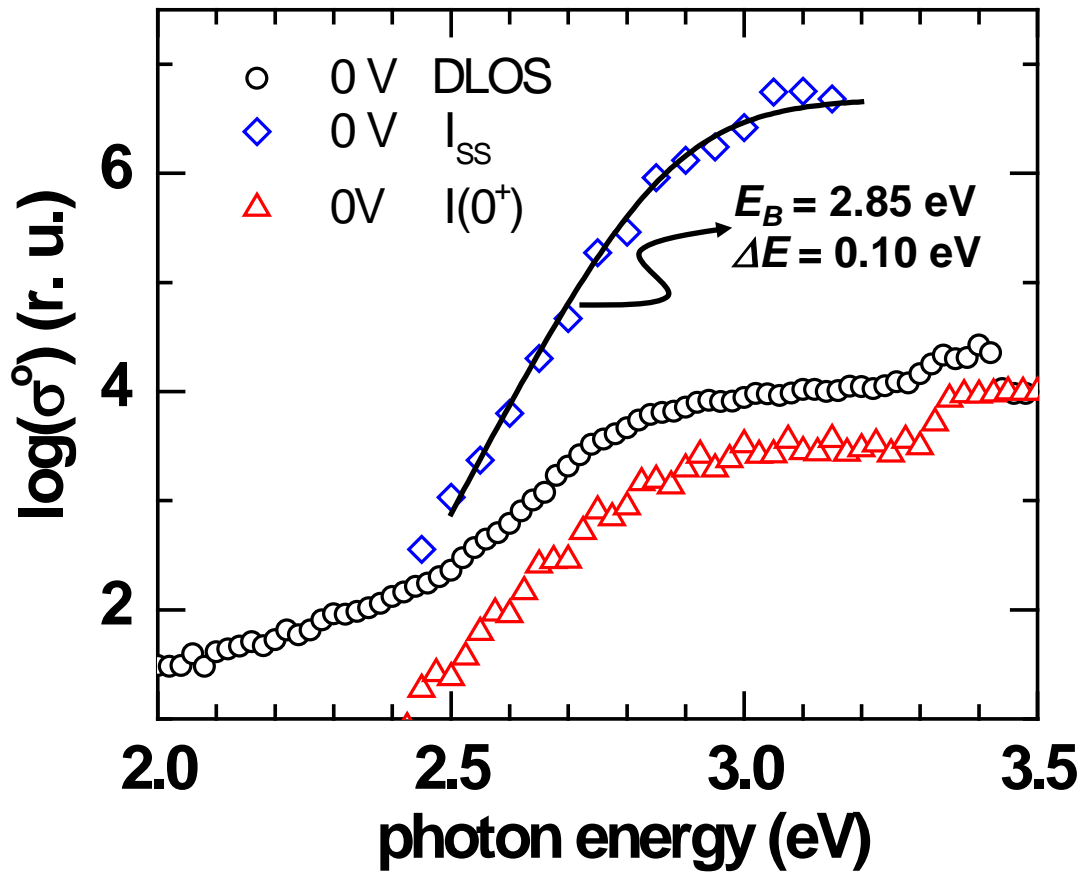


# AFM of $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$ surface





# Photocurrent



$$I(0^+) \propto e^0$$

1. C. T. Sah *et al.* SSE 13, 759 (1970).

$$I_{ss} = \frac{A}{1 + \exp\left(\frac{E_B - h\nu}{\Delta E}\right)}$$

2. R. W. Martin *et al.* APL 74, 263 (1999).

- Steady-state photocurrent suggests band tailing
- Photocurrent deep level spectrum agrees with DLOS

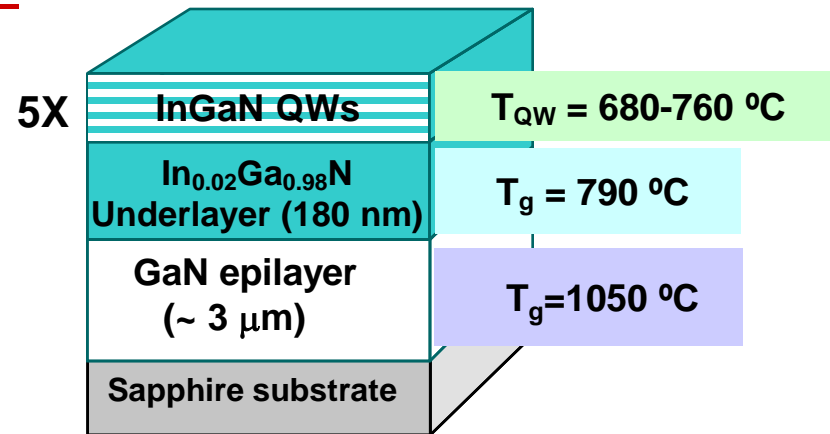


# T<sub>QW</sub> Study: Sample Designs and Structural Data

Goal: produce identical InGaN MQWs over a wide range of growth temperatures:

- Green QWs (In  $\geq 0.20$ ); limited range of T<sub>QW</sub> for sufficient indium incorporation
- Work with lower In comps (~17 %, blue emission wavelengths)
- Vary TMIn flow only to maintain similar QW In composition at various T<sub>QW</sub>

MOCVD-grown InGaN QW Samples



Structural Parameters from XRD and Modeling

T <sub>QW</sub> ( C )	QW Indium Composition	QW Thickness (nm)	GaN Barrier Thickness (nm)
680	0.161	2.56	10.3
700	0.172	2.20	10.0
720	0.166	2.20	10.1
740	0.177	1.89	8.4
760	0.167	2.31	9.4

Typical QW T<sub>g</sub> for LEDs:

Blue ~750-780 C

Green ~700-740 C

- GaN barrier T<sub>g</sub> = 850 C
- Threading dislocation densities ~1e9 cm<sup>-2</sup> (XRD)

QW In comp: 16.9 +/- 0.3

QW Width: 2.2 +/- 0.3