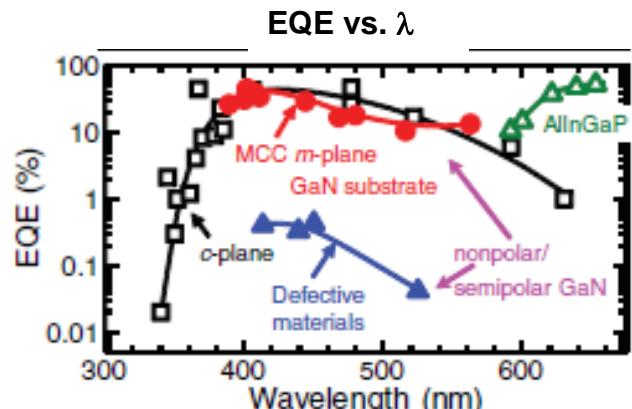

Role of defects in limiting the optical efficiency of InGaN/GaN quantum wells

Andrew Armstrong, Mary H. Crawford and Daniel D. Koleske
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.

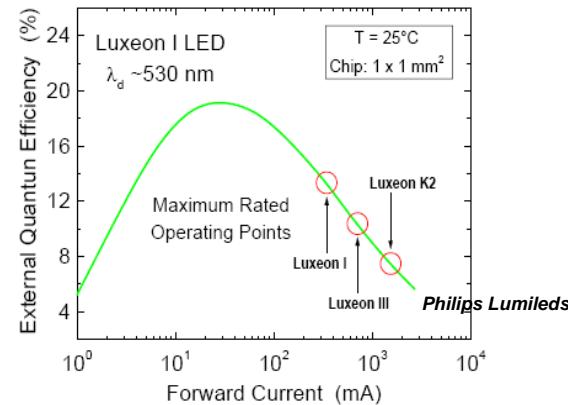
Potential impact of defects in InGaN luminescence

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

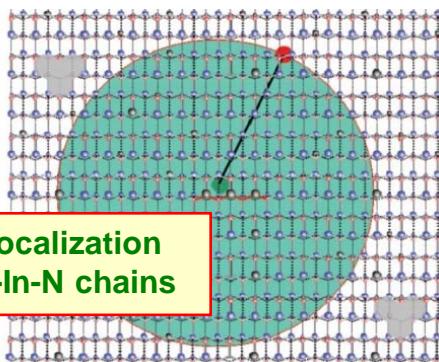


Speck et al. MRS Bullet. 34, 304 (2009)

“Efficiency Droop” of InGaN LEDs

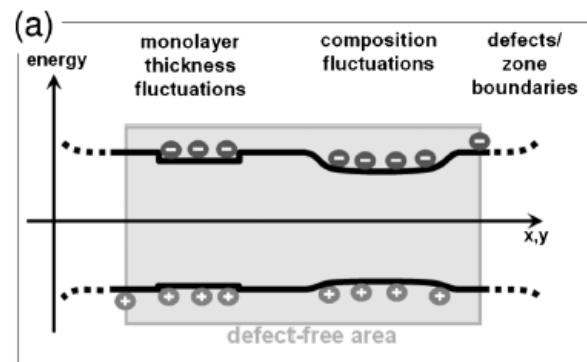


Defects and localization



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

Defects and localization

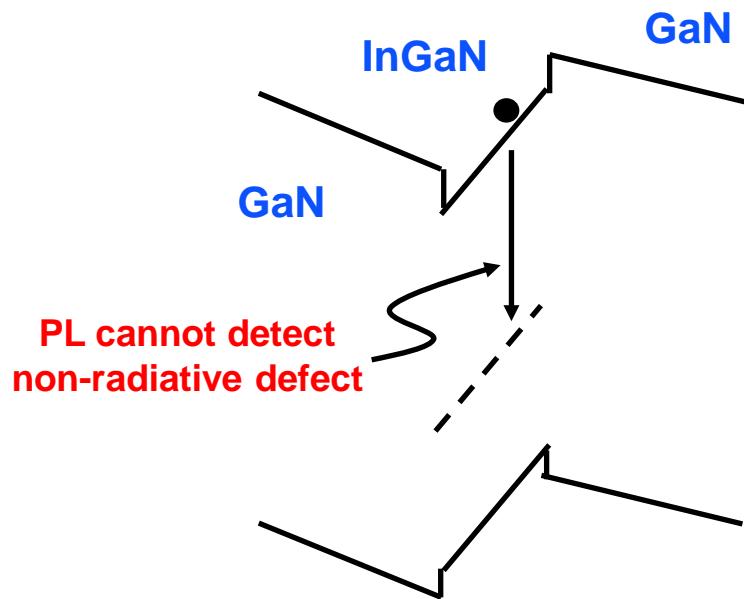


Hader et al. APL 96 221106 (2010)

➤ Connect InGaN QW growth conditions, defects and IQE

Defect Study of InGaN QW structures

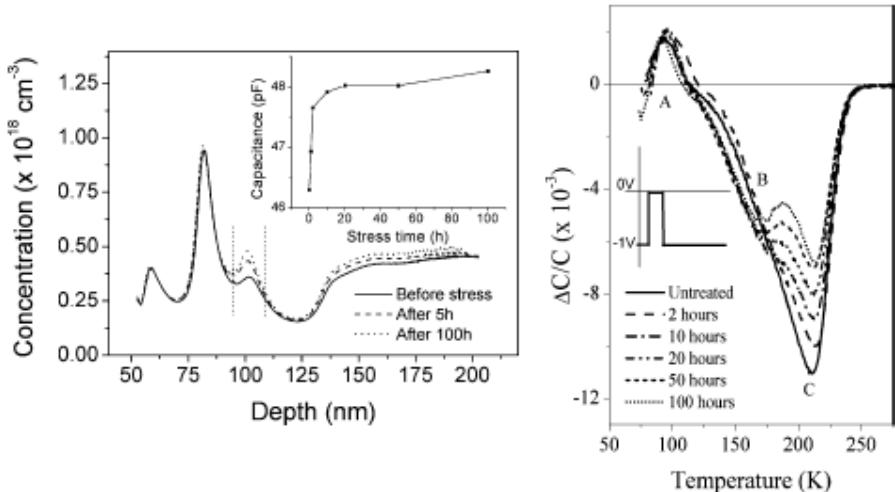
Resonant Photoluminescence



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^{1,2}

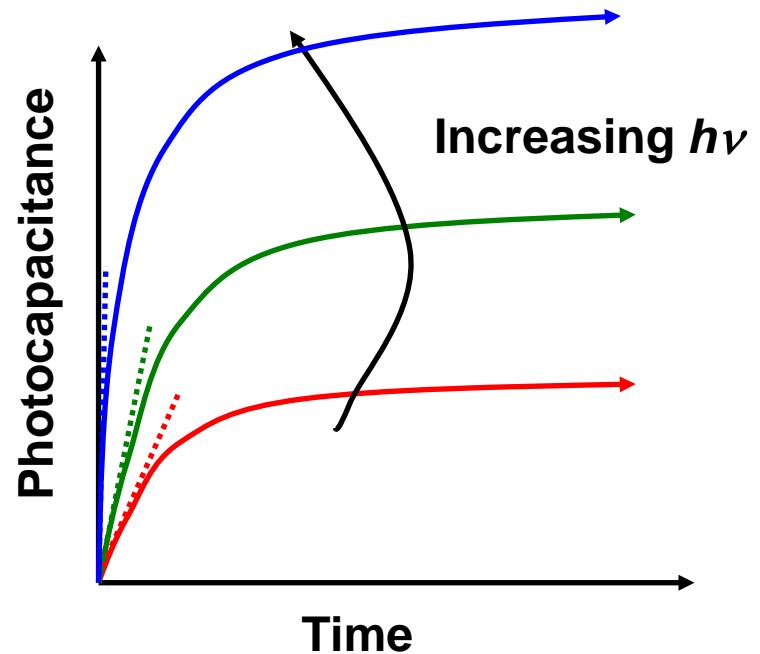
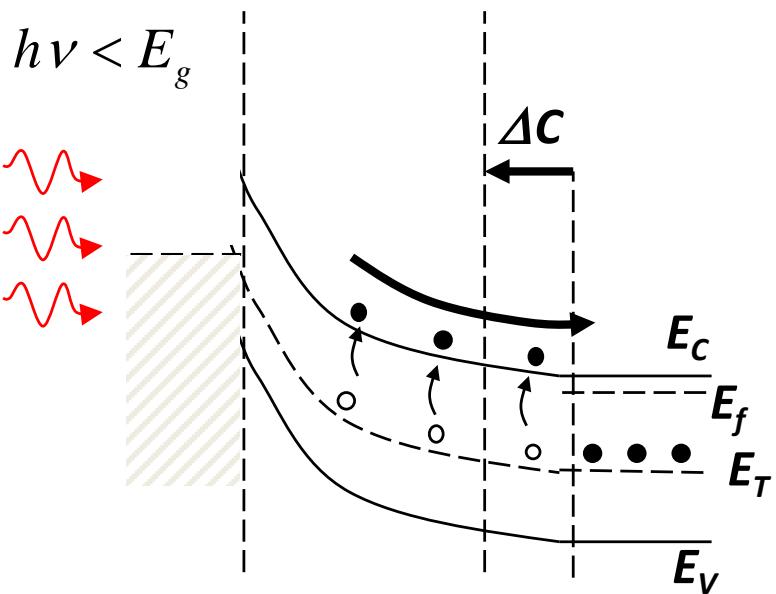
QW-based techniques not quantitative or have insufficient depth resolution

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

Deep Level Optical Spectroscopy

Deep Level Optical Spectroscopy (DLOS)¹

- 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)



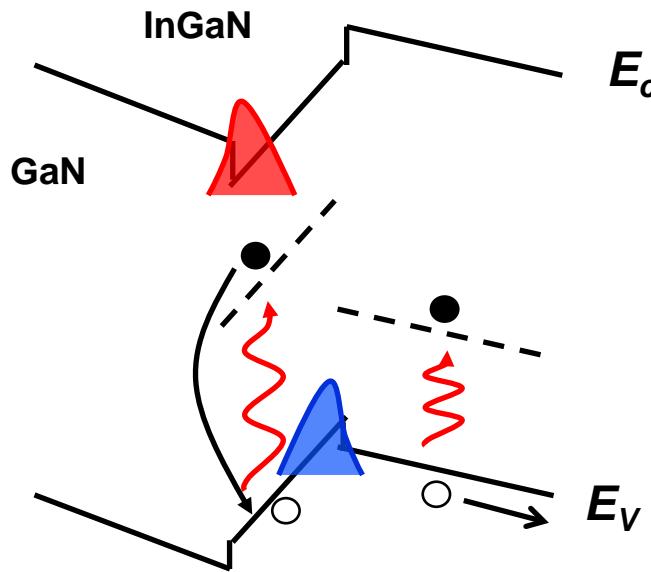
- DLOS only sensitive to depleted regions
- Enables nano-scale depth resolution

- 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

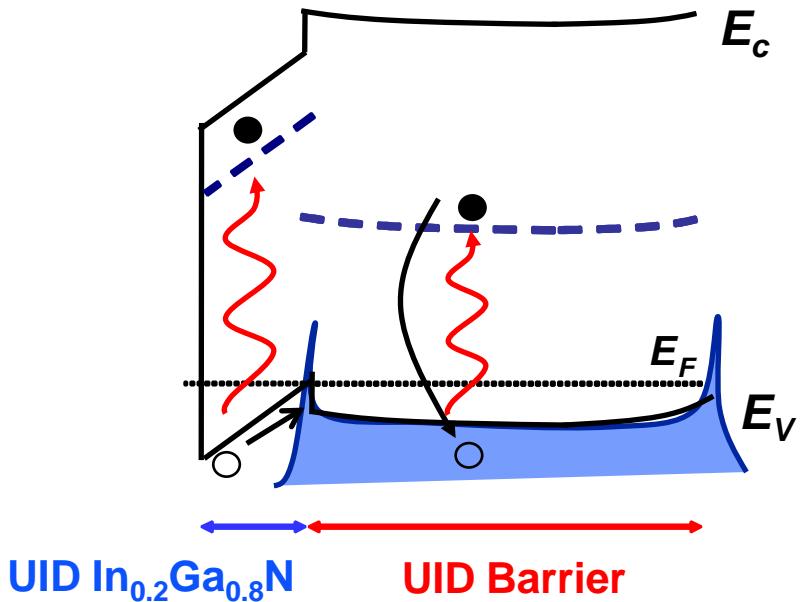
Designed InGaN/GaN heterostructure for depth-resolved DLOS

InGaN/GaN QW



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

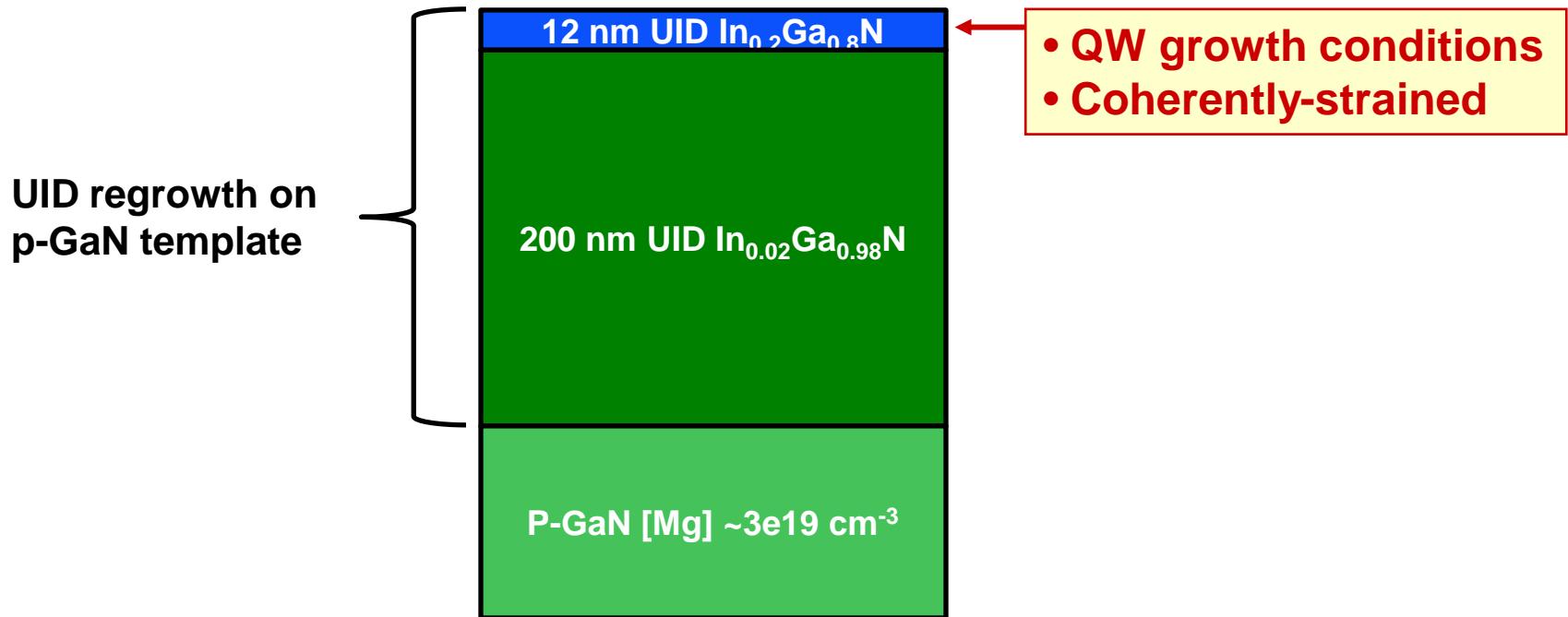
InGaN/GaN “half QW”



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

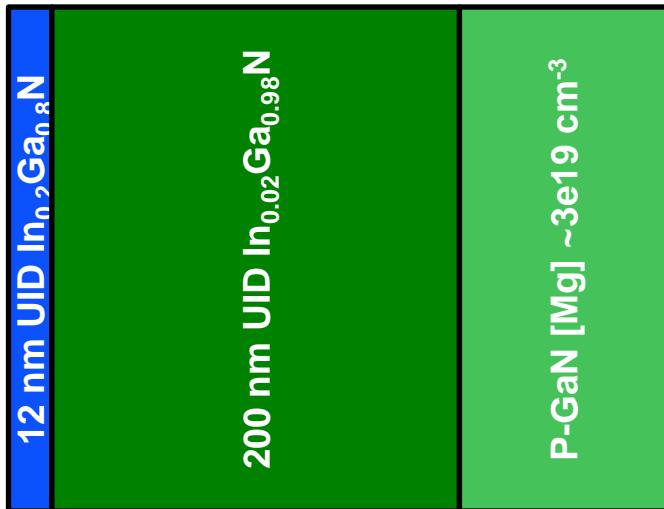
Same $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$ growth conditions for both structures

Depth-resolved DLOS of InGaN

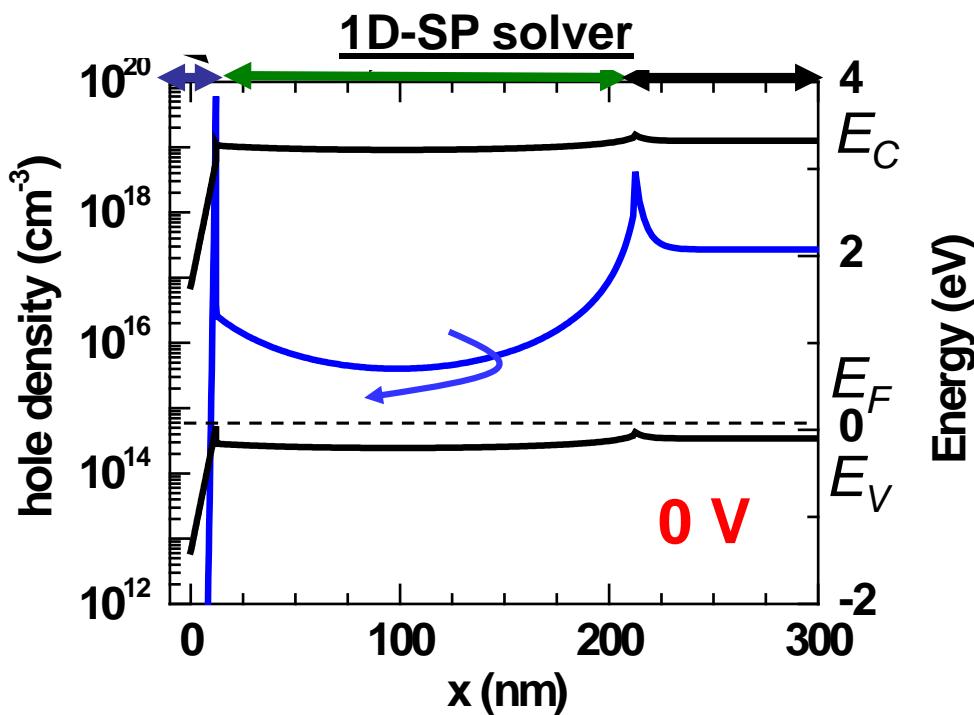


- Grown by MOCVD in a D125 short-jar Veeco reactor
- 300 torr, 15 SLM NH_3 , 10 SLM N_2 , no H_2
- In content controlled through growth temperature
- $\text{In}_{0.02}\text{Ga}_{0.83}\text{N}$: $24.3 \mu\text{moles/min Ga}$, $72.8 \mu\text{moles/min In}$
- $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$: $10.4 \mu\text{moles/min Ga}$, $55.8 \mu\text{moles/min In}$

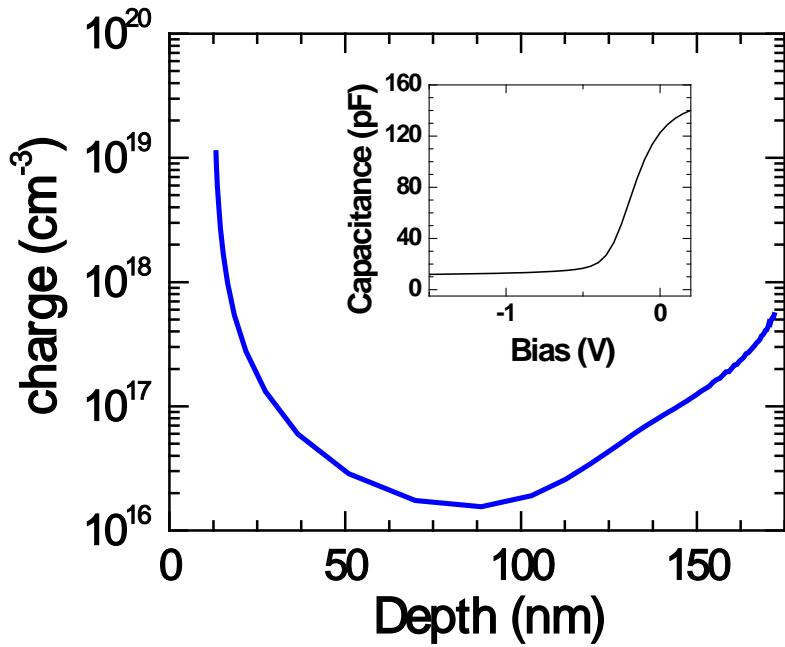
Depth-resolved DLOS of InGaN



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

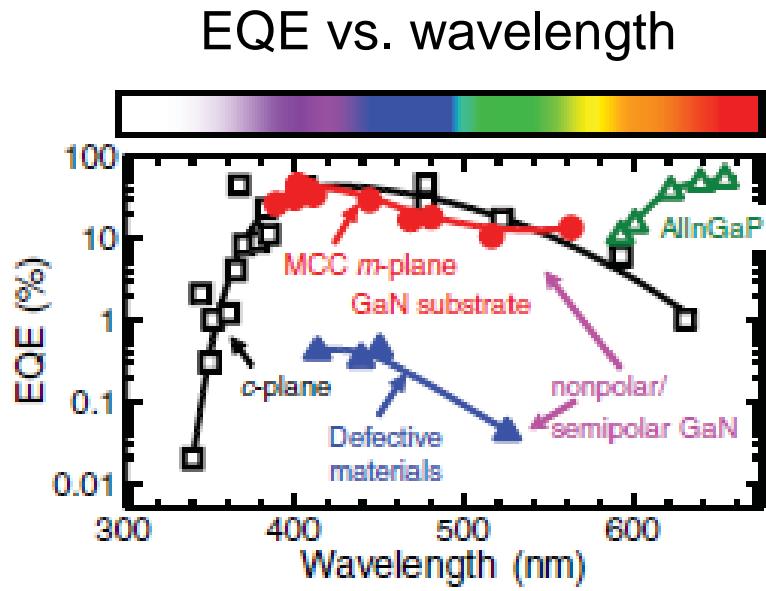
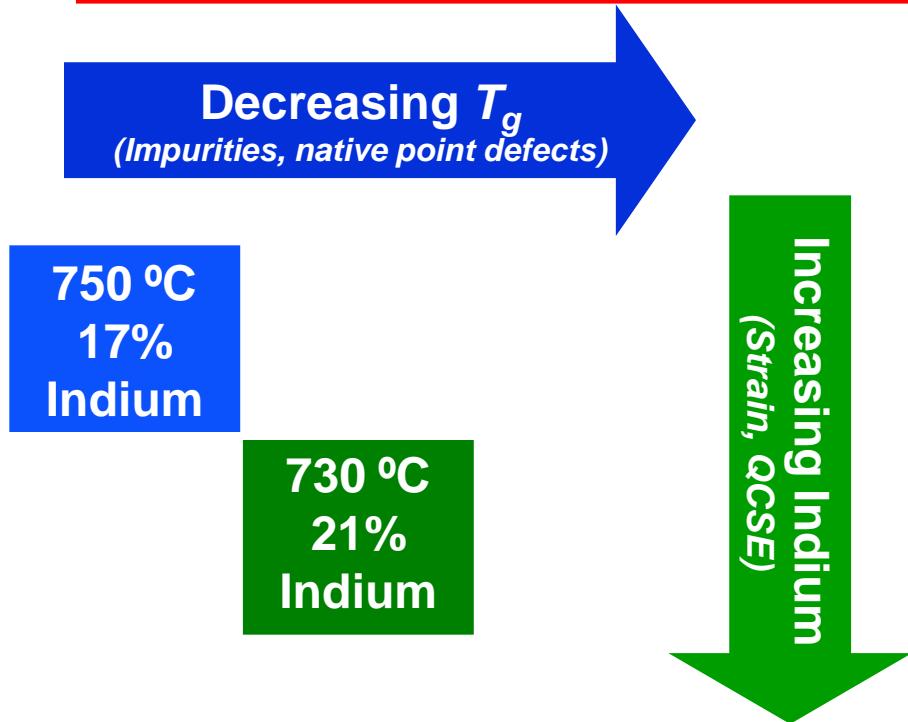


Diode C-V and carrier density



Influence of T_g and Indium alloying

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



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

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

- Decouple T_g vs. indium alloying

Effect of T_g on MQW IQE

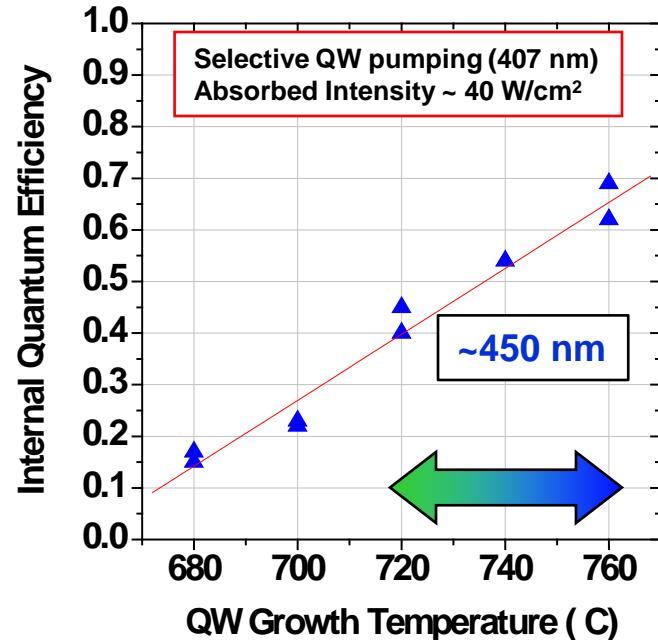
Decreasing T_g
(Impurities, native point defects)

750 °C
17%
Indium

690 °C
17%
Indium

Reduced T_{QW} at fixed Indium

Estimated IQE from Temp. Dep. PL

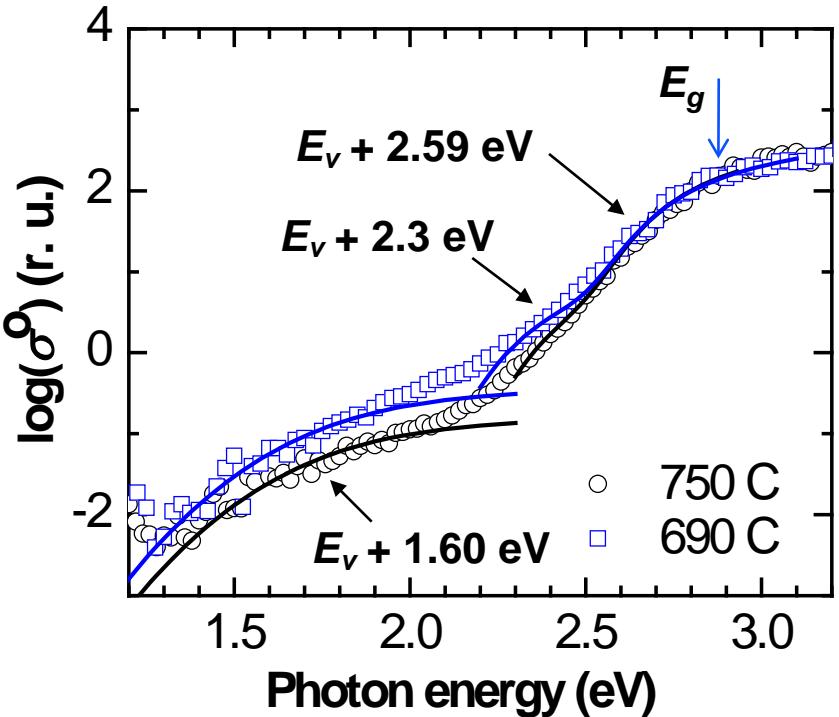


- Isolate impact of lower T_g
 - Vary TMIn flow only for similar Indium composition at various T_g
- Fixed MQW structure, strain, QCSE
- Fixed [V-defect], TDD

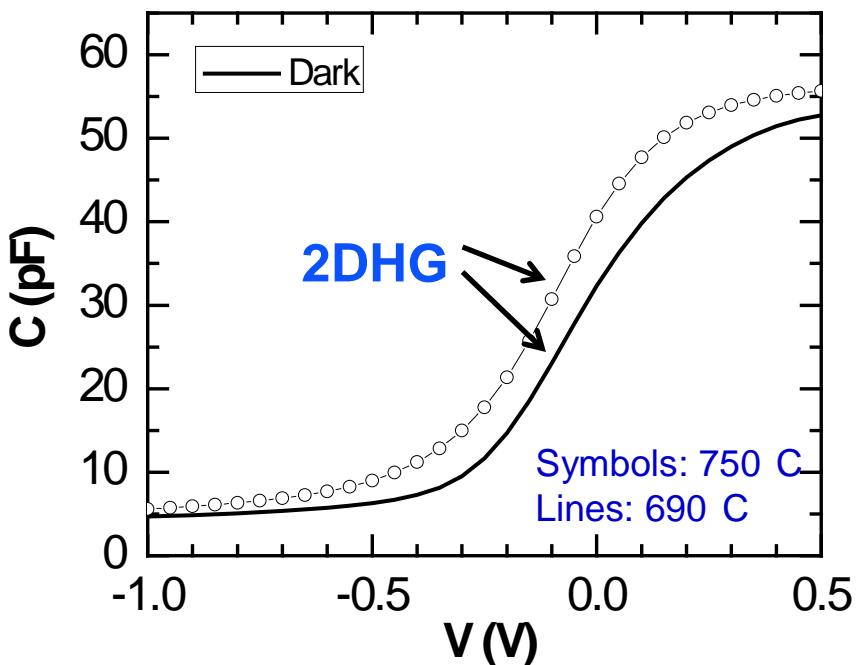
- Decreasing IQE with decreasing T_{QW}
- Point defects are suspected cause

Influence of T_g on defect incorporation

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



Lighted C-V



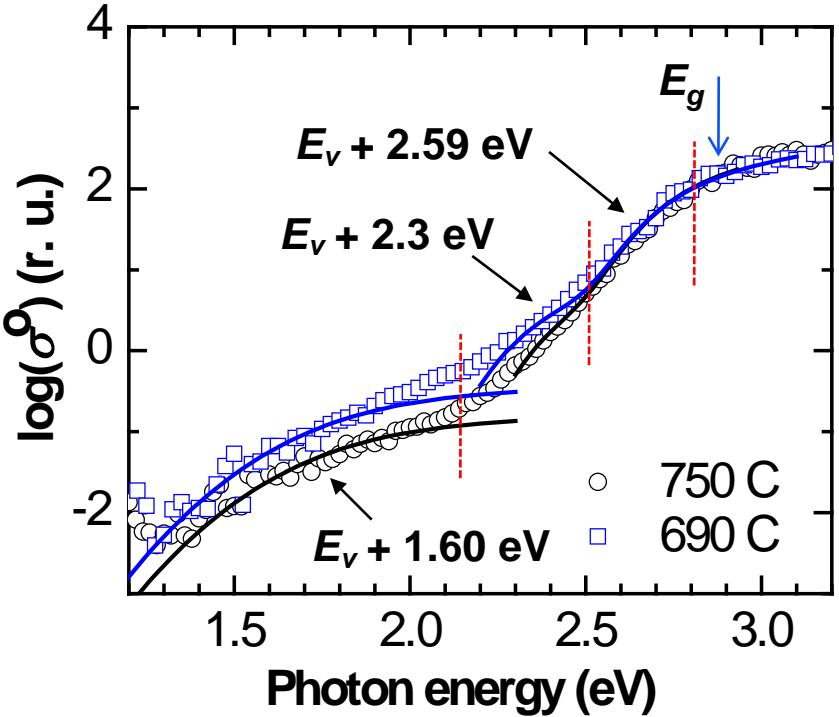
- Similar $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$ defect levels
 ➤ 60 C reduction in T_g does not introduce new deep levels

- Larger dark “threshold voltage” at 750C
 ➤ 60 C reduction in T_g introduces excess deep levels

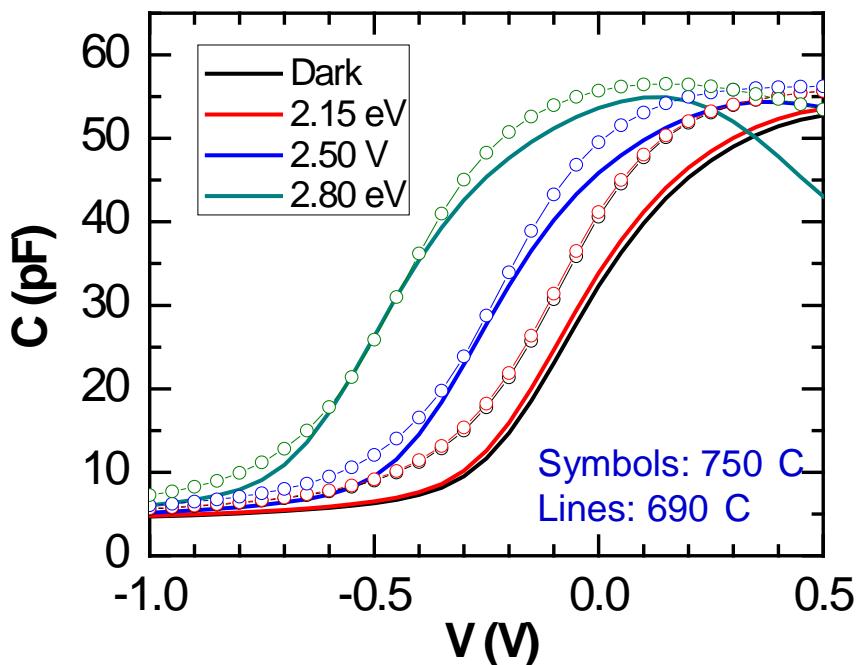
Model used to fit DLOS data:
 Paessler JAP 96, 715 (2004)

Influence of T_g on defect incorporation

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



Lighted C-V



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

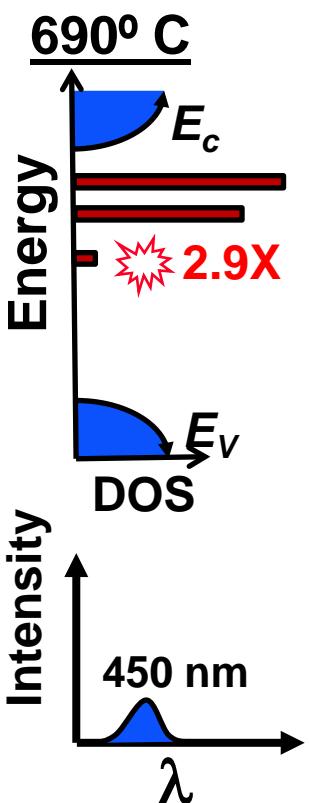
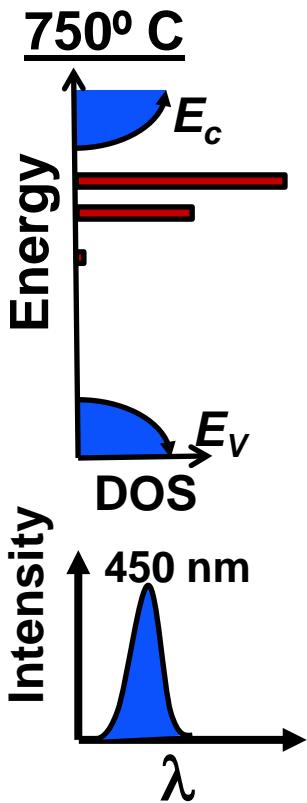
Model used to fit DLOS data:
 Paessler JAP 96, 715 (2004)

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

Influence of T_g on defect incorporation

Areal Densities of Deep Levels

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



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

➤ **Quantified** $\uparrow D_t$, $\downarrow \text{IQE}$ for $\downarrow T_g$

Possible Deep Level Candidates:

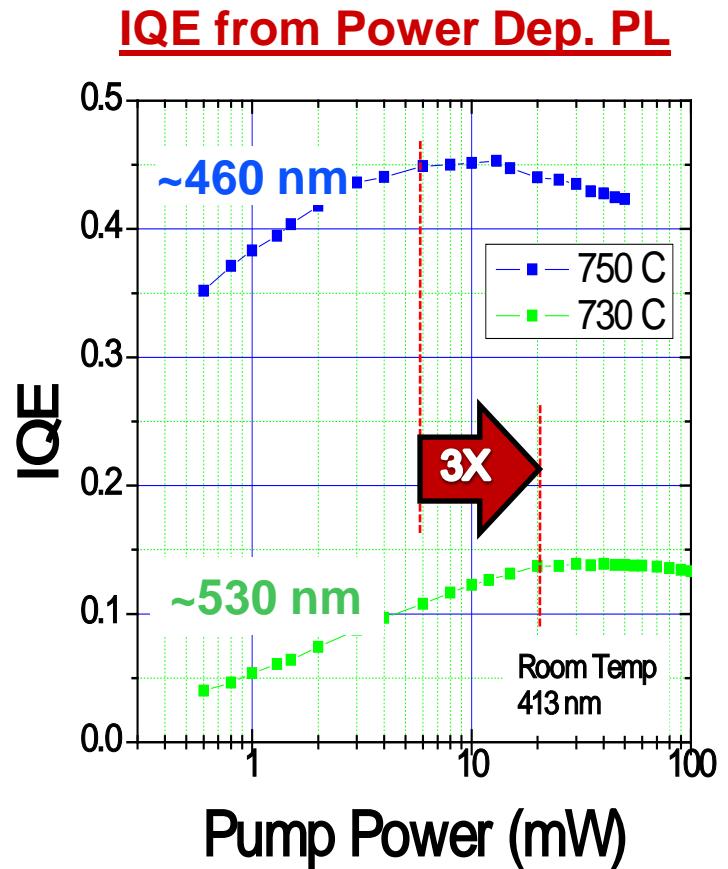
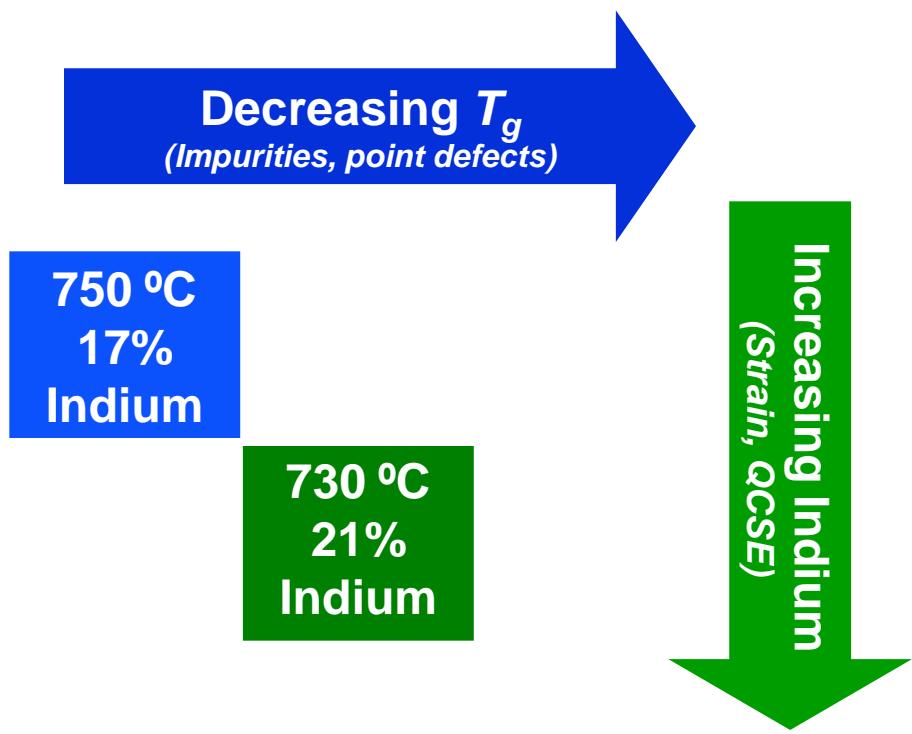
1. Carbon related (\uparrow with lower T_g ¹, nonradiative²)
2. V_{III} and Oxygen related (\uparrow with lower T_g , low formation energy of $V_{\text{III}}\text{-O}$ complexes³)

[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

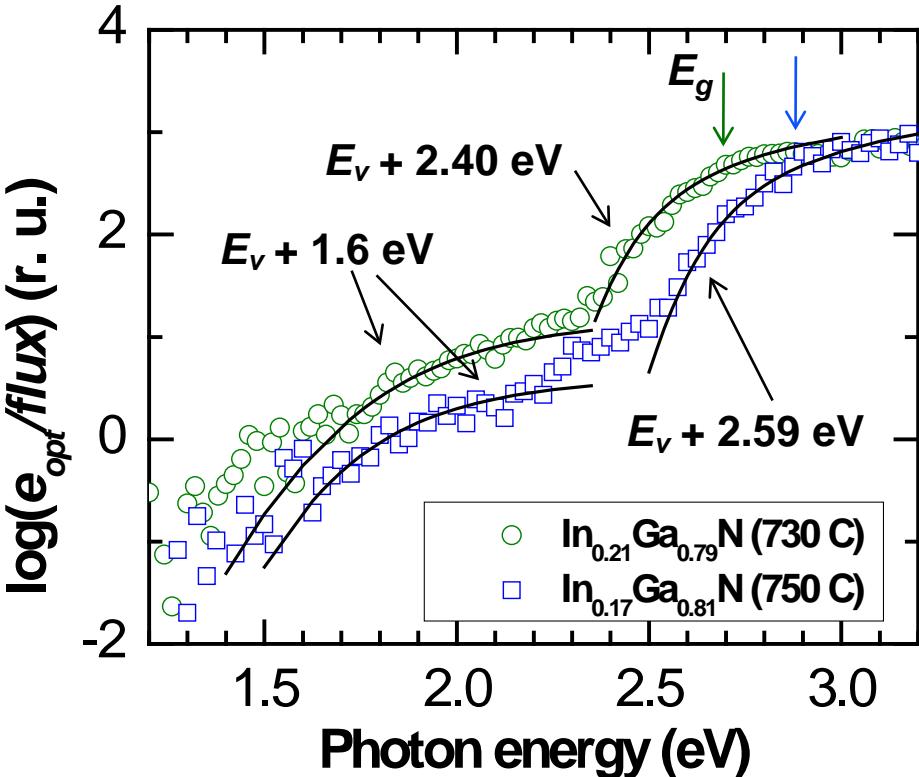


- Strongly reduced IQE
- 3X saturation power suggests more defect recombination for green MQW

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

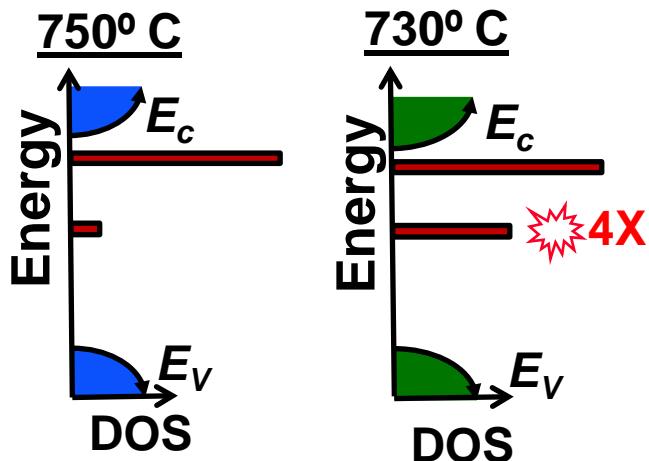
Influence of T_g and indium alloying

$\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$ vs. $\text{In}_{0.21}\text{Ga}_{0.79}\text{N}$



Densities of Deep Levels

	$\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$	$\text{In}_{0.21}\text{Ga}_{0.79}\text{N}$
$E_v + 1.6 \text{ eV}$	$7\text{e}10 \text{ cm}^{-2}$	$28\text{e}10 \text{ cm}^{-2}$
$E_v + 2.40 \text{ eV}$	$5\text{e}11 \text{ cm}^{-2}$	--
$E_v + 2.59 \text{ eV}$	--	$5\text{e}11 \text{ cm}^{-2}$

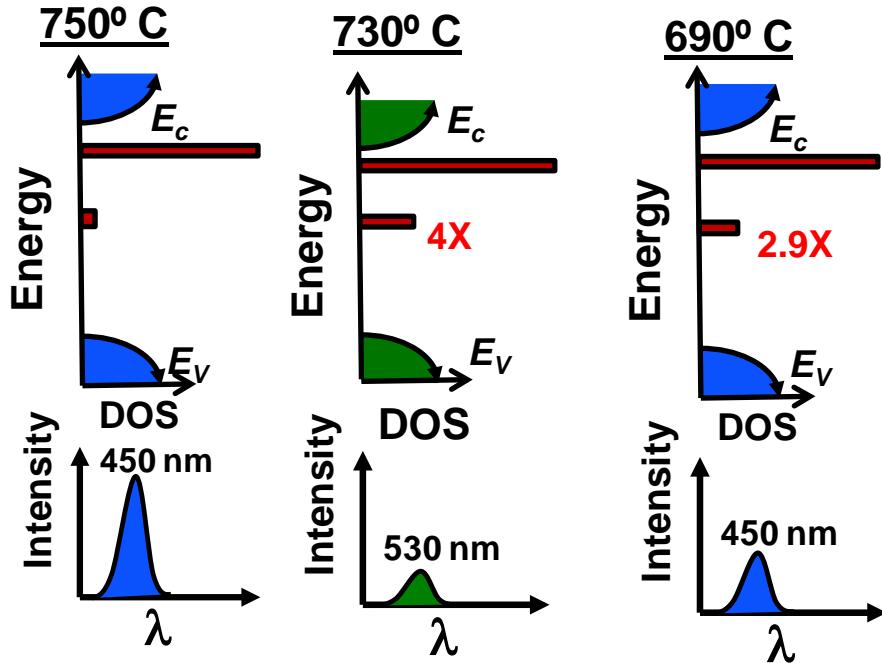


- Near- E_c level shifts with E_g
- Similar $E_v + 1.6 \text{ eV}$ deep level
- 2.3 eV level missing

→ Accelerated IQE drop and greater $[E_v + 1.6 \text{ eV}]$ with $\downarrow T_g$ and \uparrow 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):

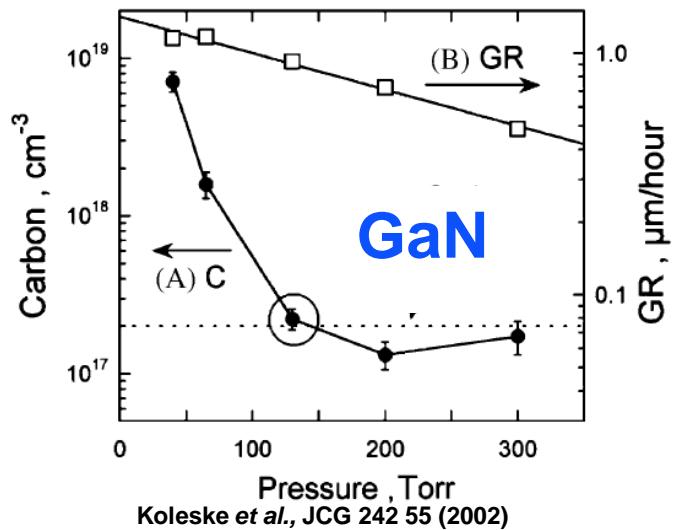
- Does not incorporate new deep levels
- Enhanced [$E_v + 1.6$ eV] correlates with strongly reduced IQE

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

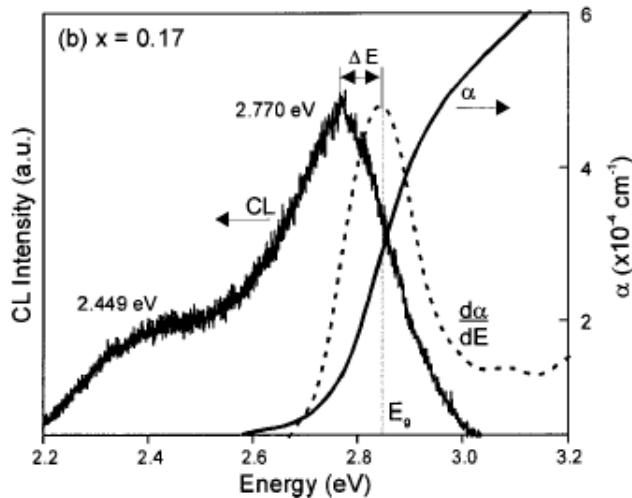
Impurity incorporation and growth pressure



- 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

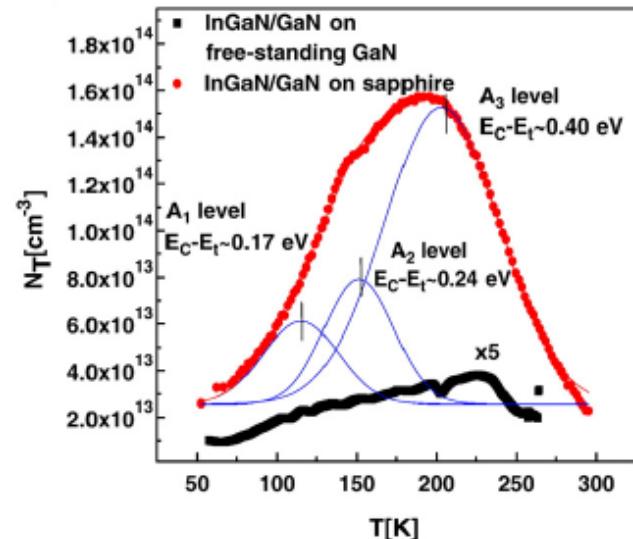
Defect Study of "bulk" InGaN films

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



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

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



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

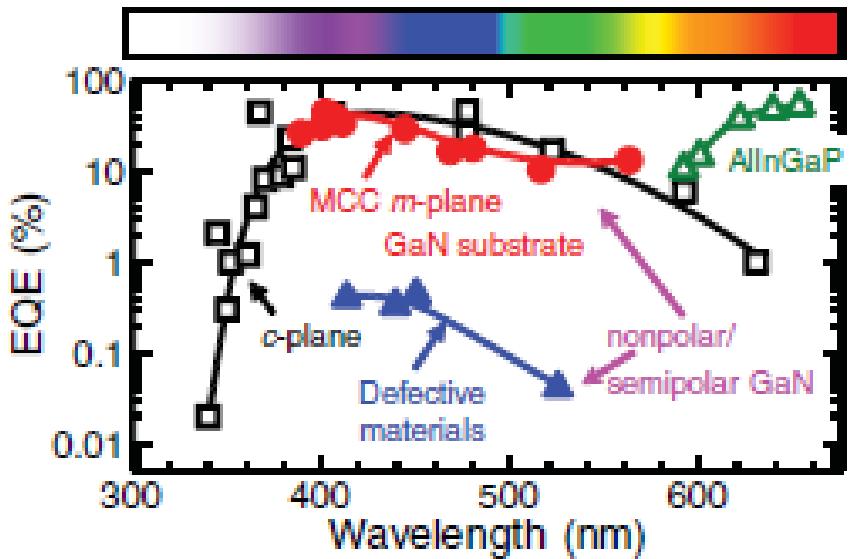
- $\text{In}_x\text{Ga}_{1-x}\text{N}$ film quality degrades for thick layers ($x > 0.15$)
 - Strain relaxation and surface roughening
 - V-defect expansion
 - Indium segregation

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



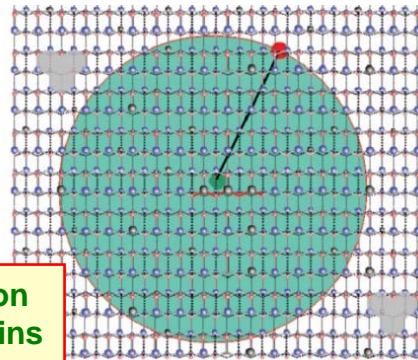
Potential impact of defects for InGaN LEDs

InGaN EQE vs. wavelength



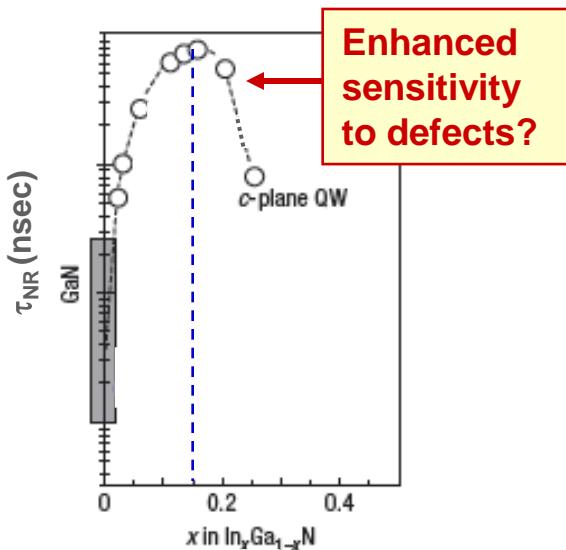
Speck *et al.* MRS Bullet. 34, 304 (2009)

InGaN defects and efficiency



Hole localization
at In-N-In-N chains

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

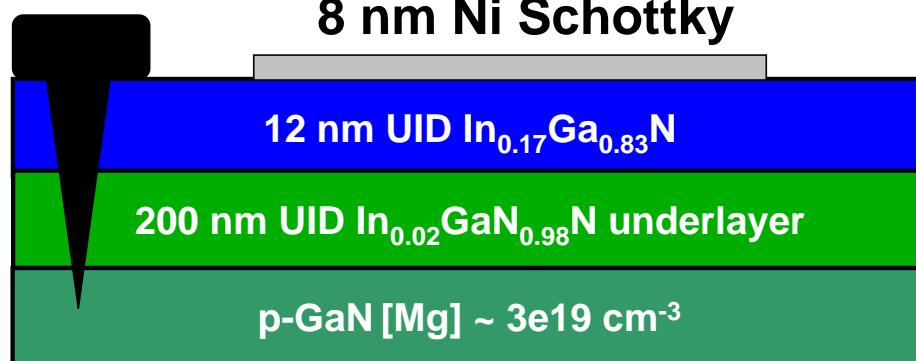


Efficiency roll-off not just QCSE

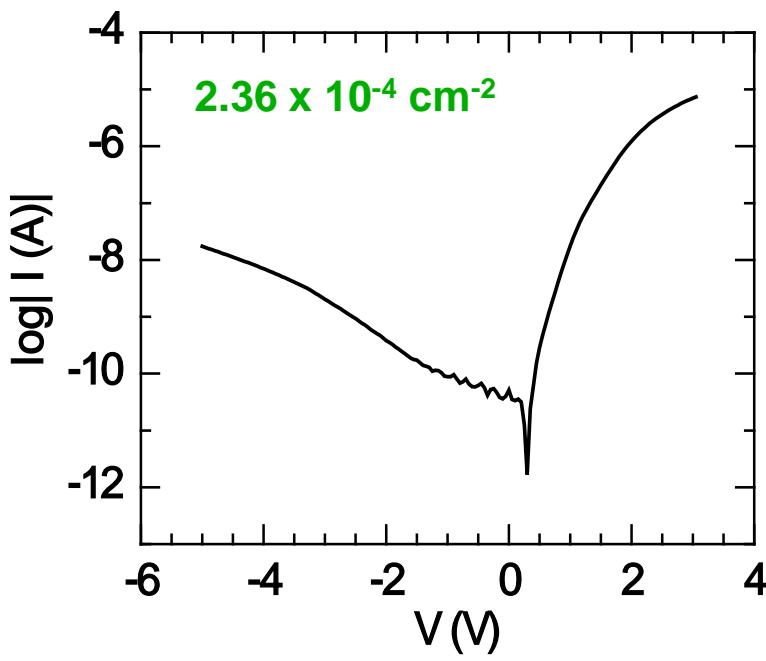
- Connect InGaN QW growth conditions, defects and IQE
- Deep Level Optical Spectroscopy (DLOS) for InGaN epilayers

Diode structure

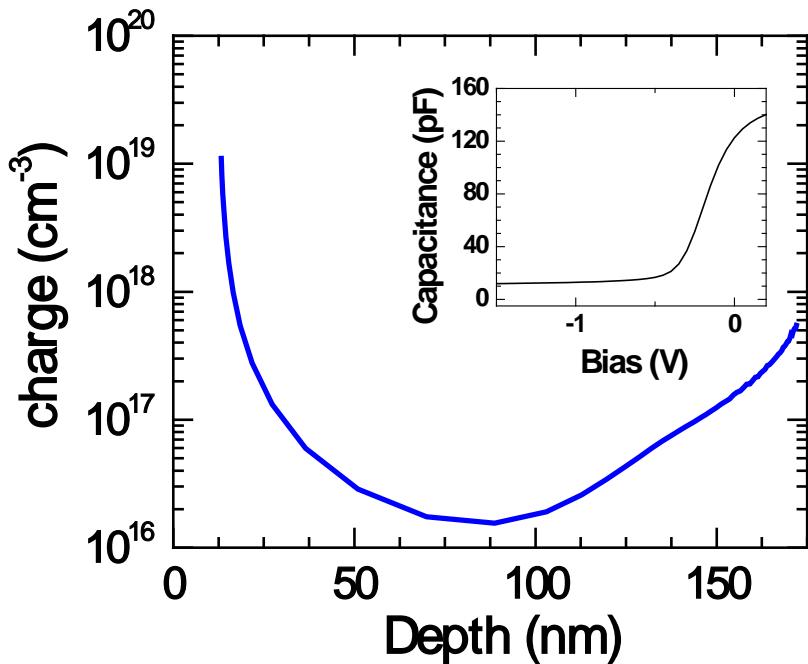
In contact



I-V

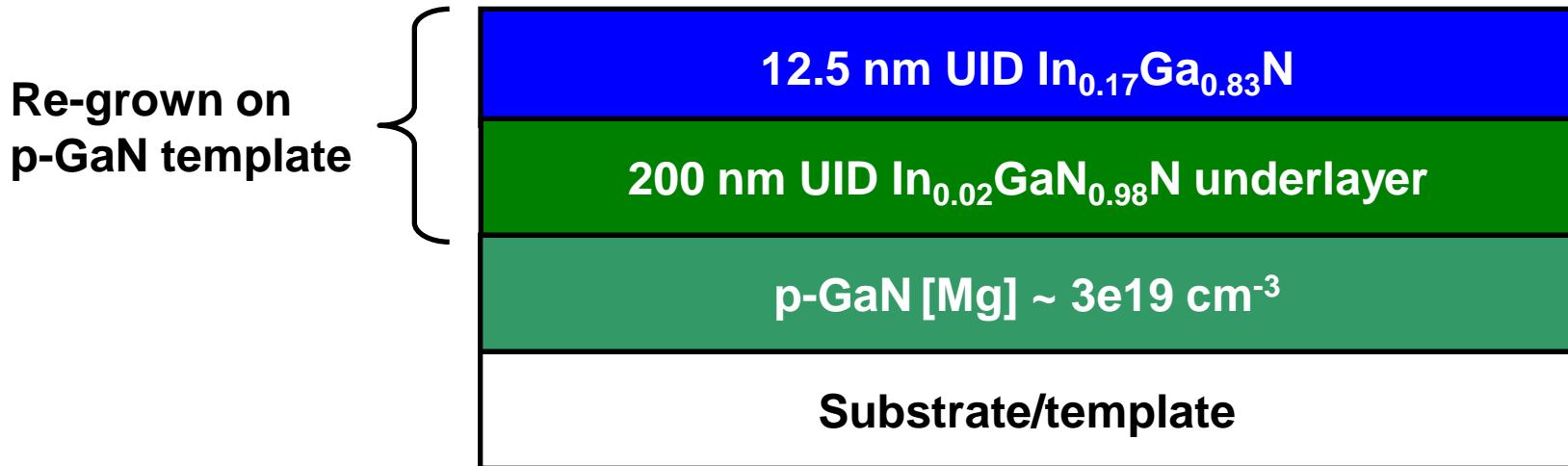


C-V and carrier density



Growth of InGaN/GaN DLOS structure

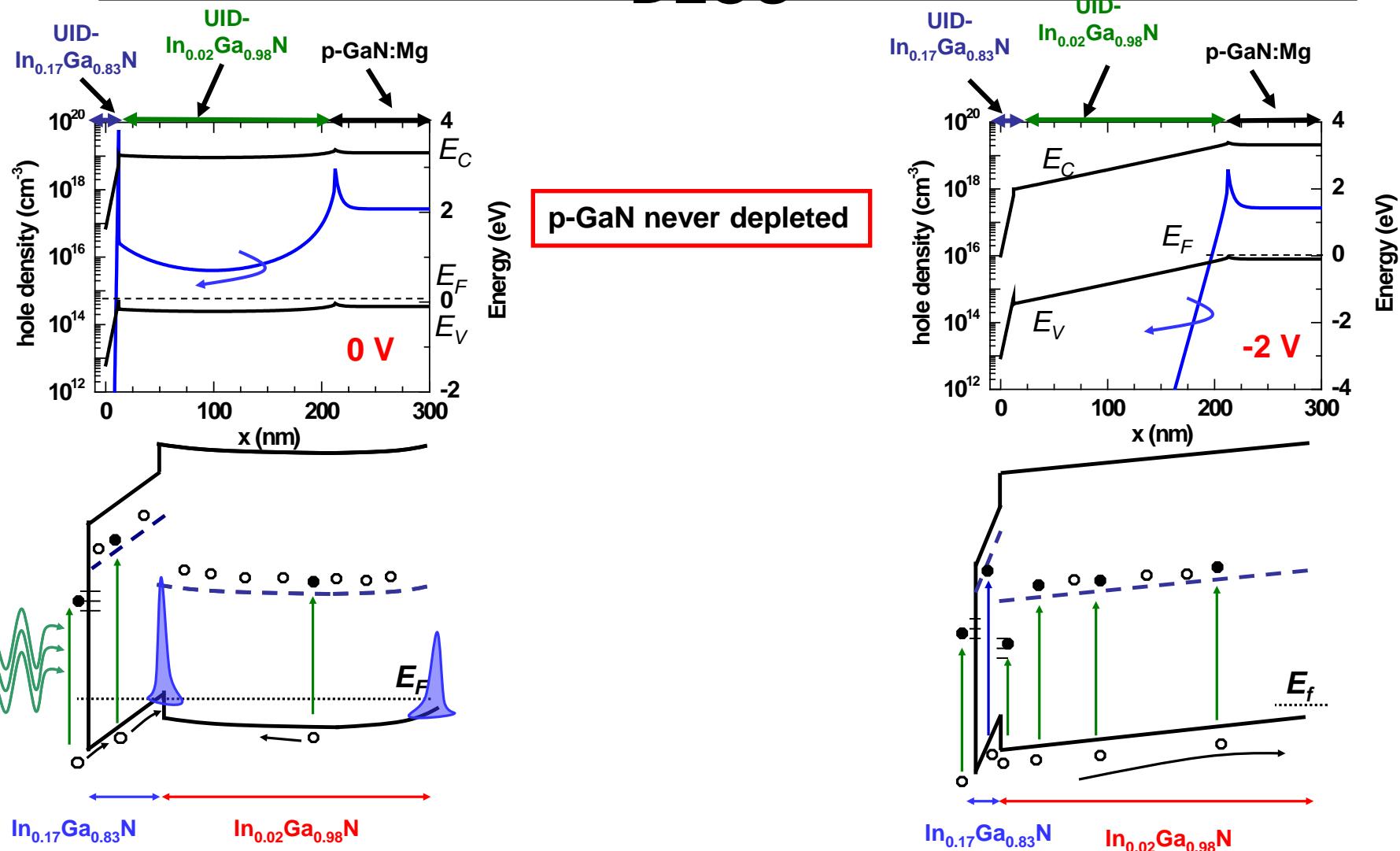
$\text{In}_x\text{Ga}_{1-x}\text{N}$ layers grown under nominal QW conditions



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

- Grown by MOCVD in a D125 short-jar Veeco reactor
- 300 torr, 15 SLM NH_3 , 10 SLM N_2 , no H_2
- In content controlled through growth temperature
- $\text{In}_{0.02}\text{GaN}_{0.83}\text{N}$: 24.3 $\mu\text{moles}/\text{min}$ Ga, 72.8 $\mu\text{moles}/\text{min}$ In
- $\text{In}_{0.17}\text{GaN}_{0.83}\text{N}$: 10.4 $\mu\text{moles}/\text{min}$ Ga, 55.8 $\mu\text{moles}/\text{min}$ In

Simulate Depth-resolved DLOS



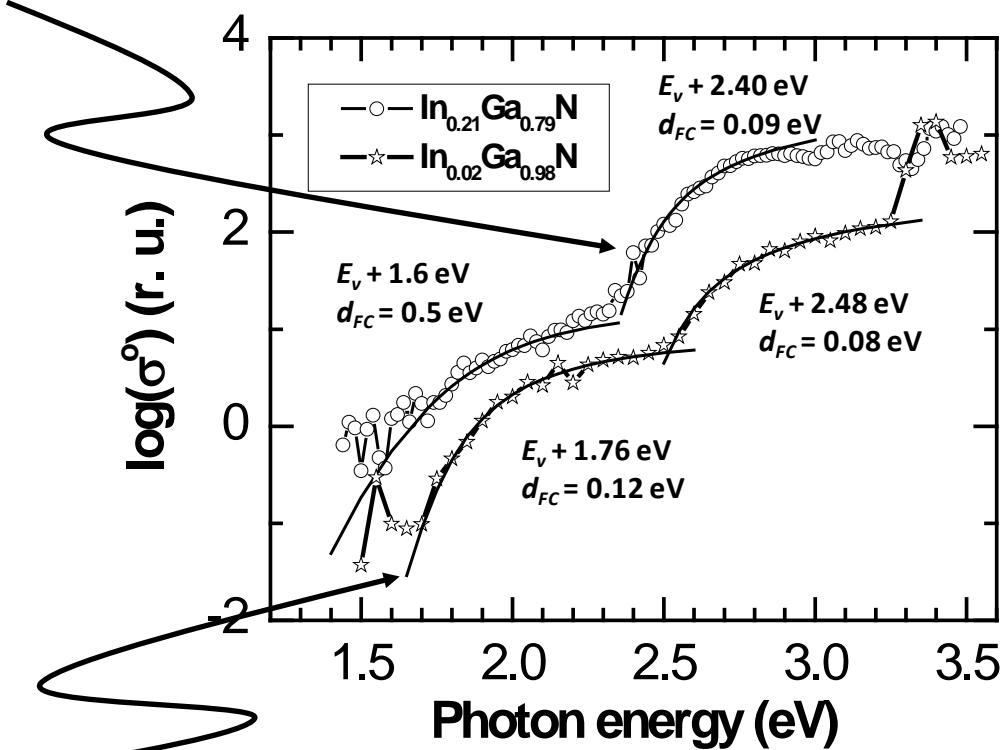
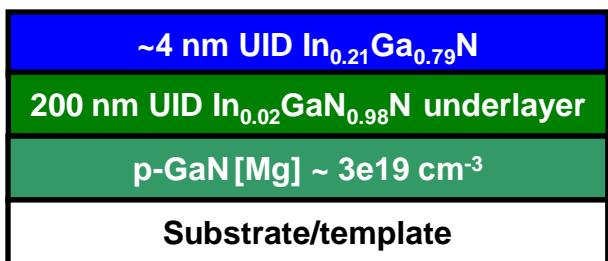
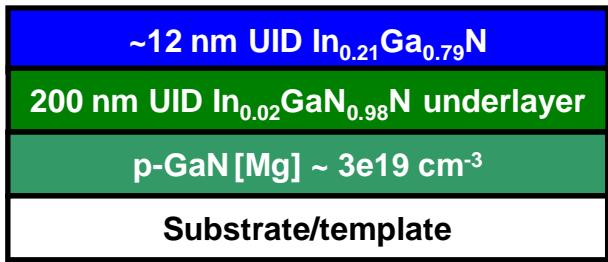
Accumulation:

- "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:

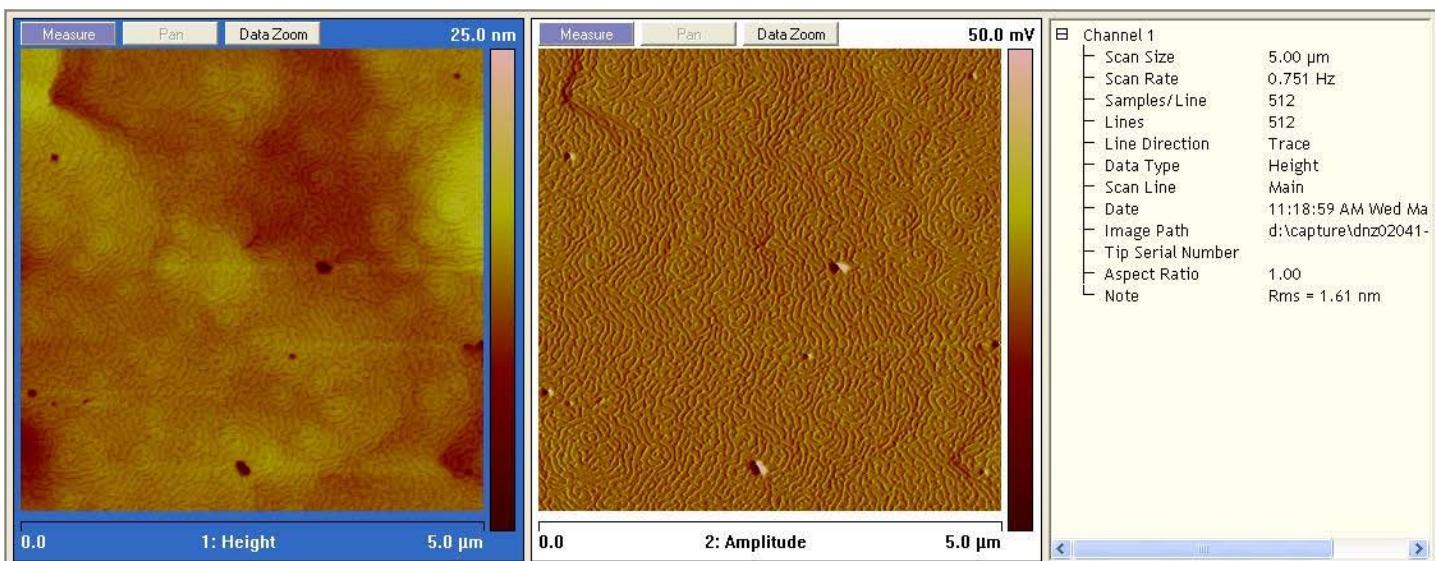
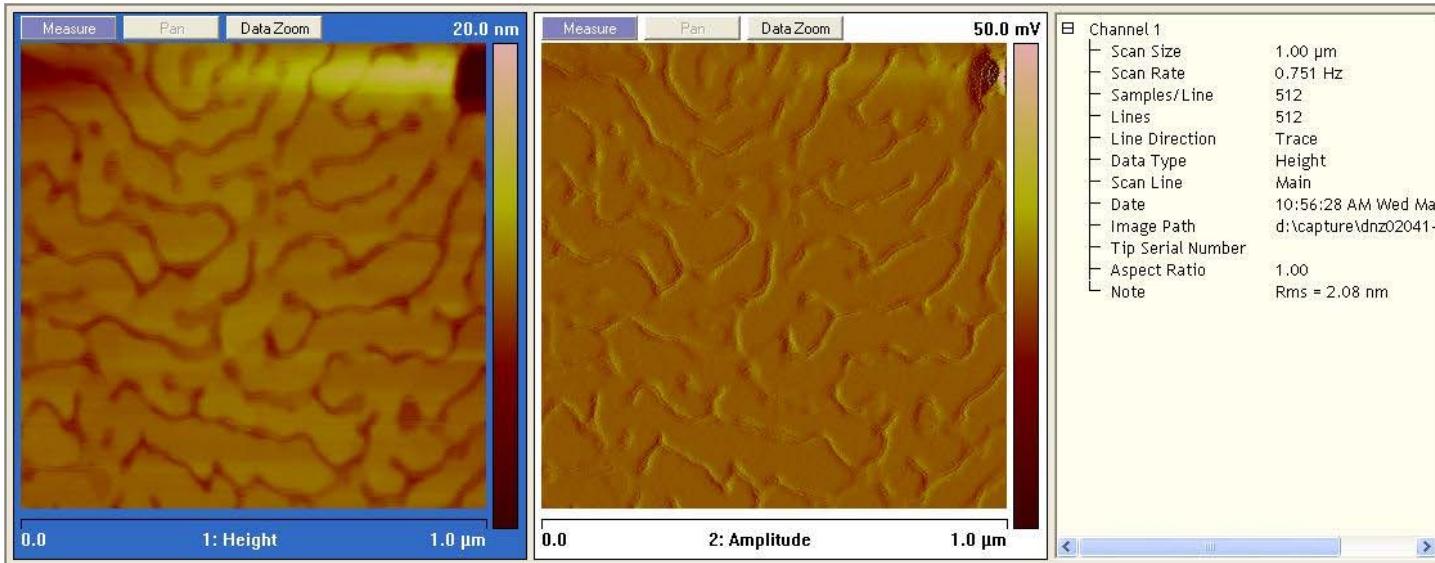
- "well", interface, and UL depleted
- UL defects dominate DLOS
- Contribution from all InGaN regions

Confirmed DLOS depth-resolution

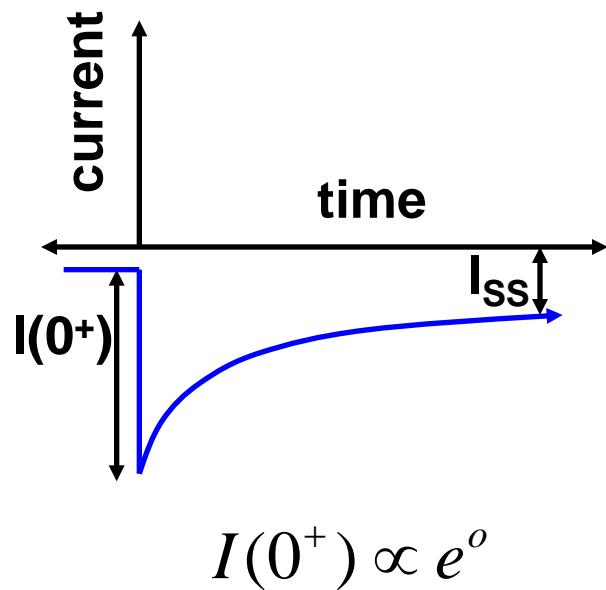
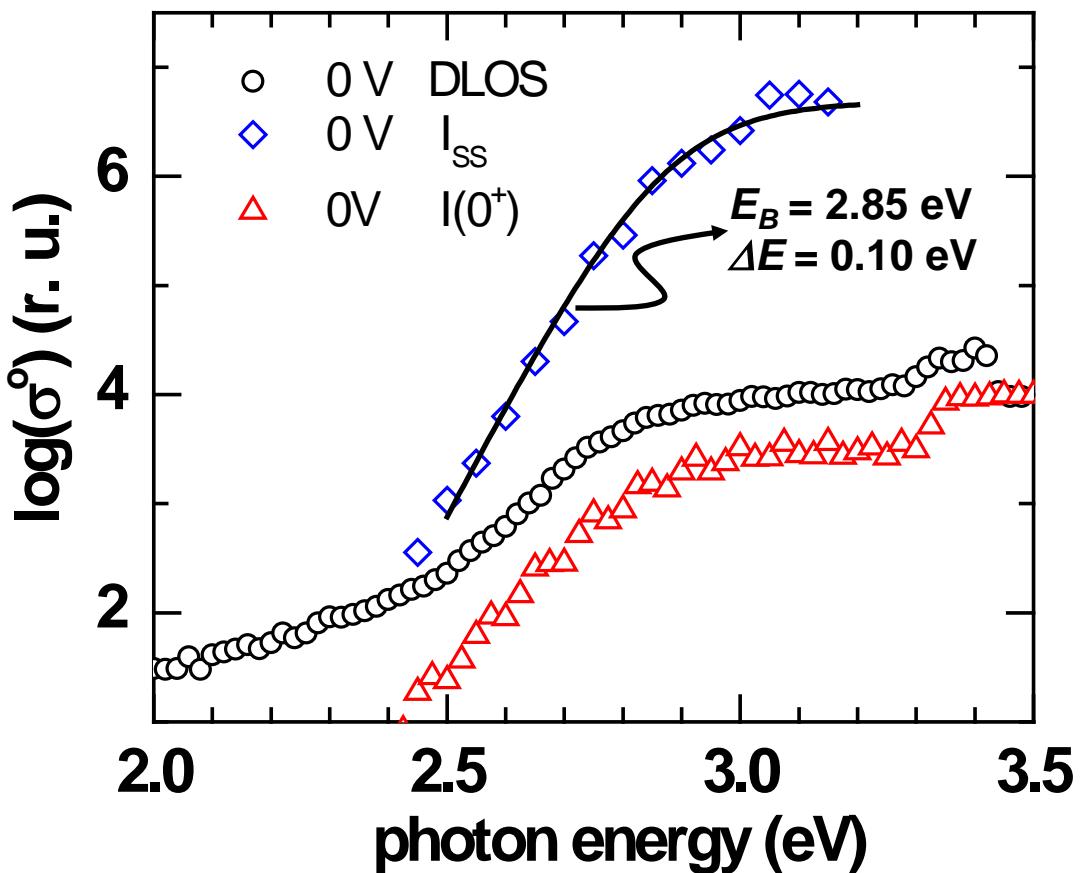




AFM of $\text{In}_{0.17}\text{Ga}_{0.83}\text{N}$ surface



Photocurrent



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).

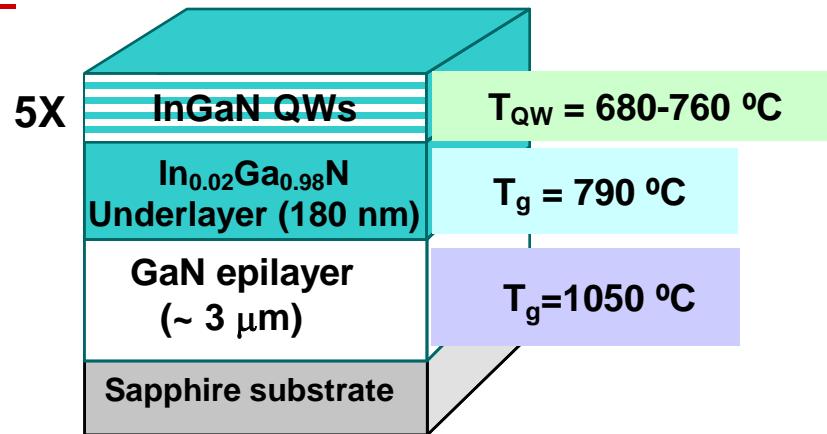
- Steady-state photocurrent suggests band tailing
- Photocurrent deep level spectrum agrees with DLOS

T_{QW} Study: Sample Designs and Structural Data

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

- Green QWs ($\text{In} \geq 0.20$); limited range of T_{QW} 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_{QW}

MOCVD-grown InGaN QW Samples



Structural Parameters from XRD and Modeling

T_{QW} (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_g for LEDs:

Blue ~750-780 C
Green ~700-740 C

- GaN barrier T_g = 850 C
- Threading dislocation densities ~1e9 cm⁻² (XRD)

QW In comp: 16.9 +/- 0.1

QW Width: 2.2 +/- 0.3