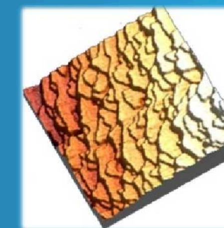
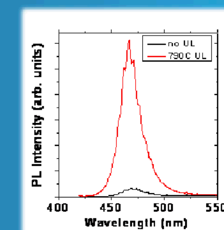
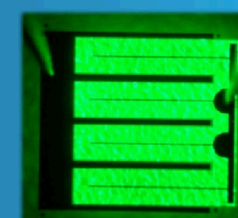
APS March Meeting, Portland, OR, March 15-19, 2010

SAND2010-1562P

InGaN Growth Morphology and Its Relationship to Luminescence for Solid State Lighting







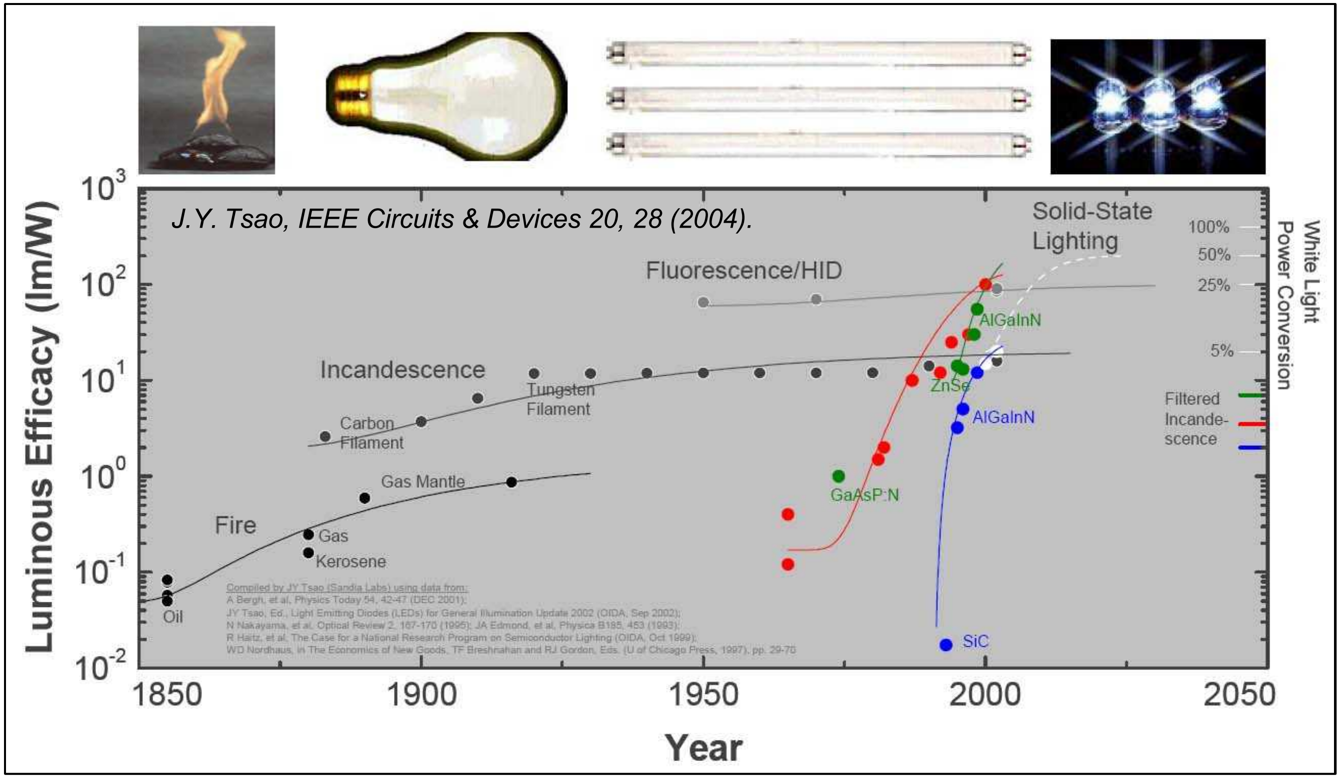
Daniel D. Koleske

Sandia National Laboratories

Work supported by the US Department of Energy, Office of Basic Energy Sciences

Why Solid State Lighting?

Luminous efficacy from conventional light sources has stagnated for the last 50 years



Power Conversion Efficiency

Incandescent ~4%

Fluorescent ~ 18%

SSL Goal ~ 50%

LED package mid 1990's

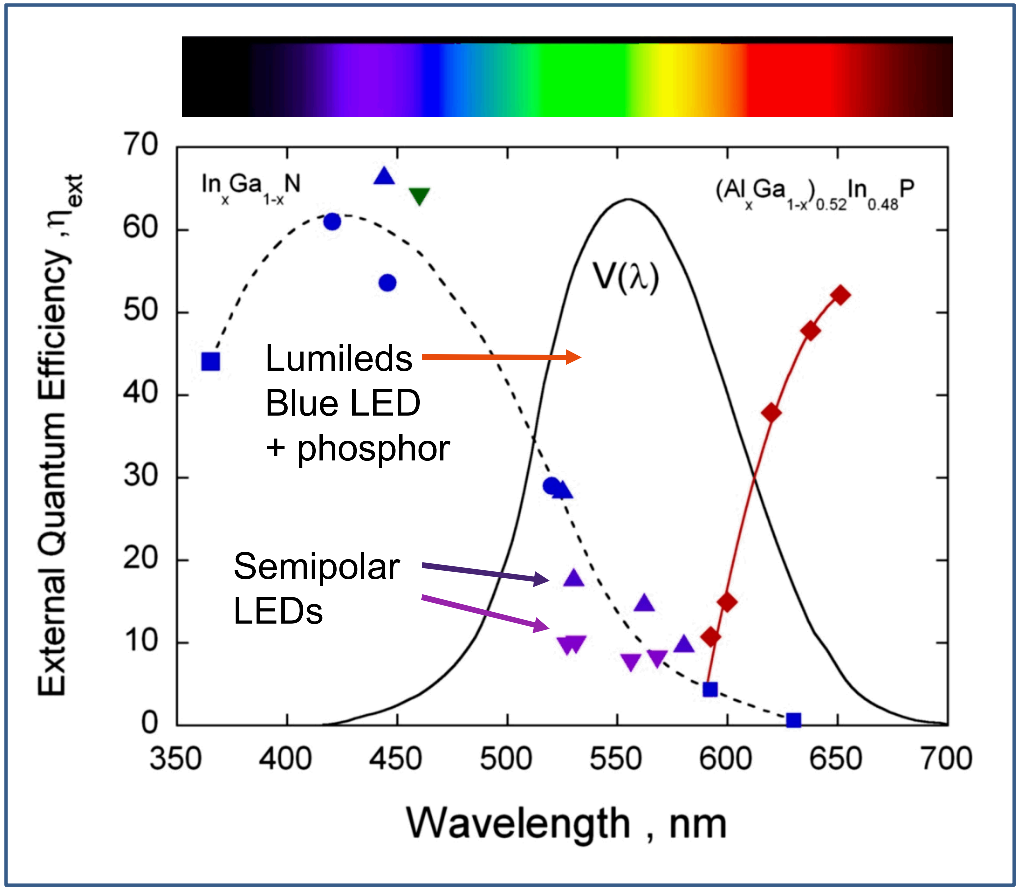
Lumileds 5 W package, 2009

DOE has a stated goal of producing SSL with 50% wall-plug efficiency by 2025 (> 200 lm/W)

On Feb. 2, 2010, Cree reported a 208 lm/W cool white LED.

DOE Goal: Improve IQE of All Visible Emitters to 90%

Current EQE of SOA LEDs, V(λ) = CIE standard eye response



InGaN for blue & green LEDs
AlGaInP for red LEDs

Green LEDs on semipolar GaN are showing promise.

white LEDs are obtained using blue + yellow phosphor (have ~30% Stokes loss).

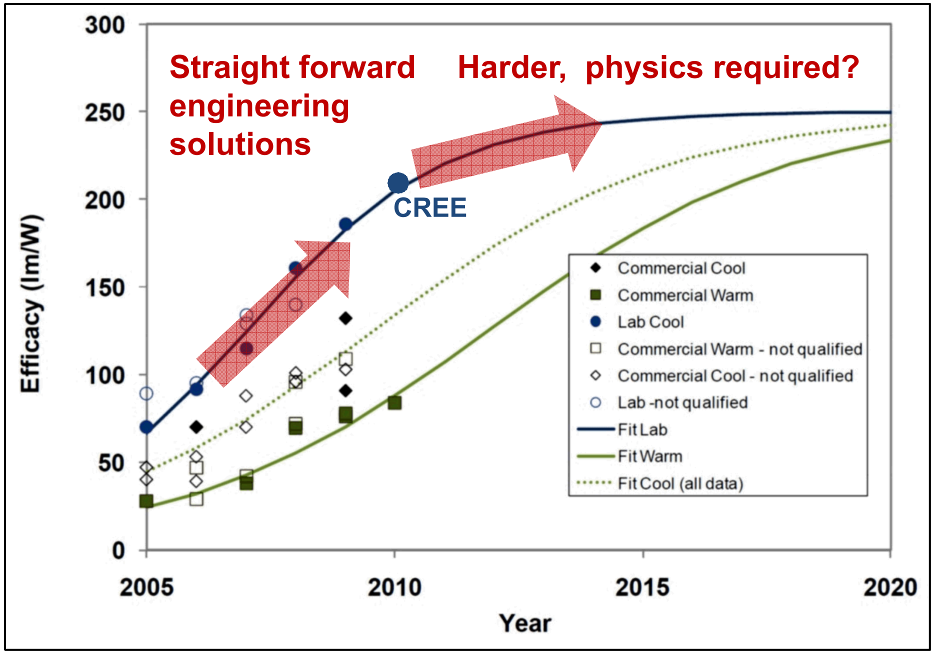
Most efficient white LEDs will be obtained using red + green + blue approach or a four color solution.

Monochromatic LEDs can be made using a blue LED pumping a longer wavelength phosphor.

If IQE ≤ 90 %, extraction efficiency ≤ 90 %, maximum EQE ≤ 80 %

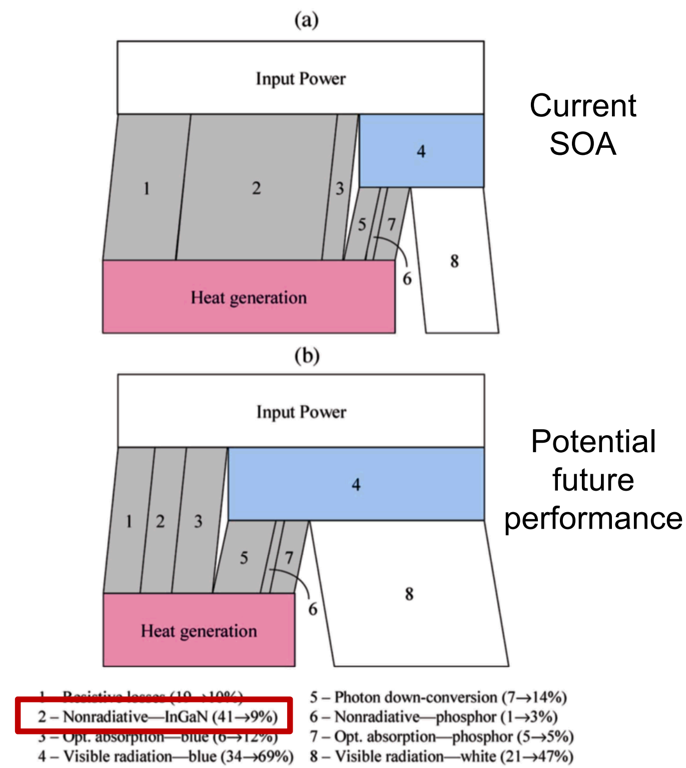
Despite huge success, LED progress is getting more difficult

From the DOE SSL 2010 Multi-Year Plan Technology R&D



Beyond 2010, improving LEDs may be limited by physical understanding.

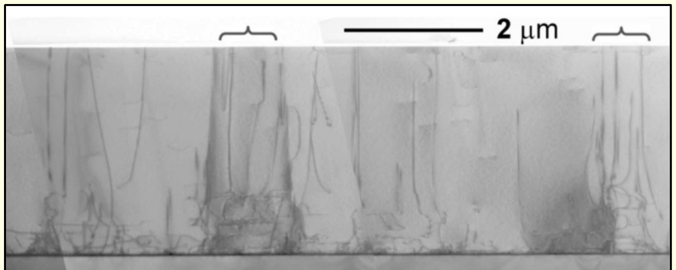
From Krames, et al., J. Display Tech. 3, 160 (2007)



2 - Nonradiative – InGaN (41 → 9%)

What are the nonradiative defects and how do we remove them?

With high dislocation density, how can InGaN LEDs be so bright?



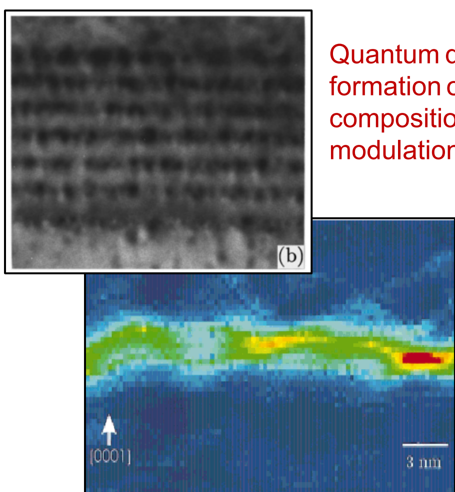
TEM courtesy of D. M. Follstaedt

- High brightness LEDs are typically grown on high dislocation density GaN (10^9 cm^{-2}).
- With this large dislocation density LEDs in the other III-V materials would not work.

Dislocations ↔ nonradiative recombination sites

High efficiency suggests some type of carrier localization; the exact nature of which is unknown!

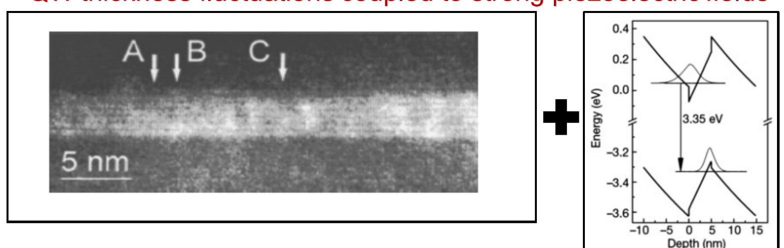
Narukawa, APL 70, 981 (1997)



Quantum dot formation or compositional modulation

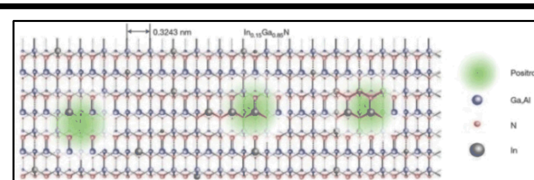
Gerthsen, Phys. Stat. A Sol. 177, 145 (2000).

QW thickness fluctuations coupled to strong piezoelectric fields

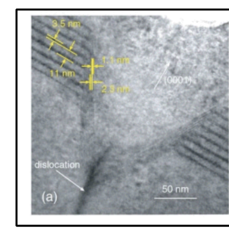


Graham, JAP 97, 103508 (2005)

Holes localized at In-N valence states, followed by exciton formation and light emission.

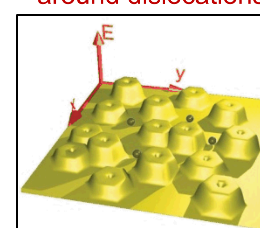


Chichibu, Nat. Mat. 5, 810 (2006)



Thinner QWs around v-defects

Energetic screening around dislocations



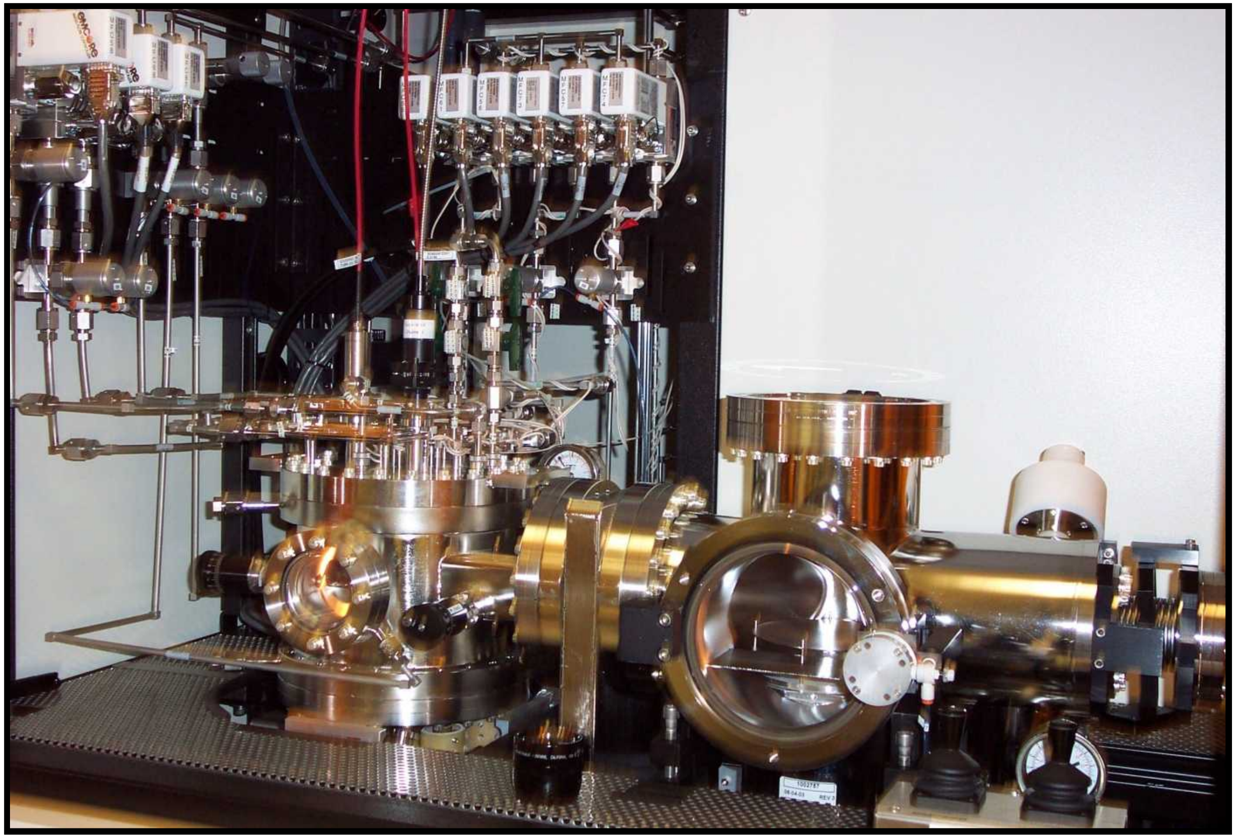
Hangleiter, PRL 95, 127402 (2005).

Focus of this talk

Localization models suggest that InGaN film structure somehow influences quantum efficiency.

- Quantify InGaN step structure using AFM.
  - multiple vs. single layer steps as InGaN thin films and QW grow on GaN.
- Show how multiple-layer steps develop on InGaN – strain relief.
- Use Power Spectral Density analysis to determine mechanisms for InGaN growth – Herring smoothing mechanisms.
- Explain how two different smoothing mechanisms can be used to influence InGaN interface morphology.
- Show two examples where improved PL intensity occurs when increased multiple-layer steps are observed.

InGaN films grown using MOCVD

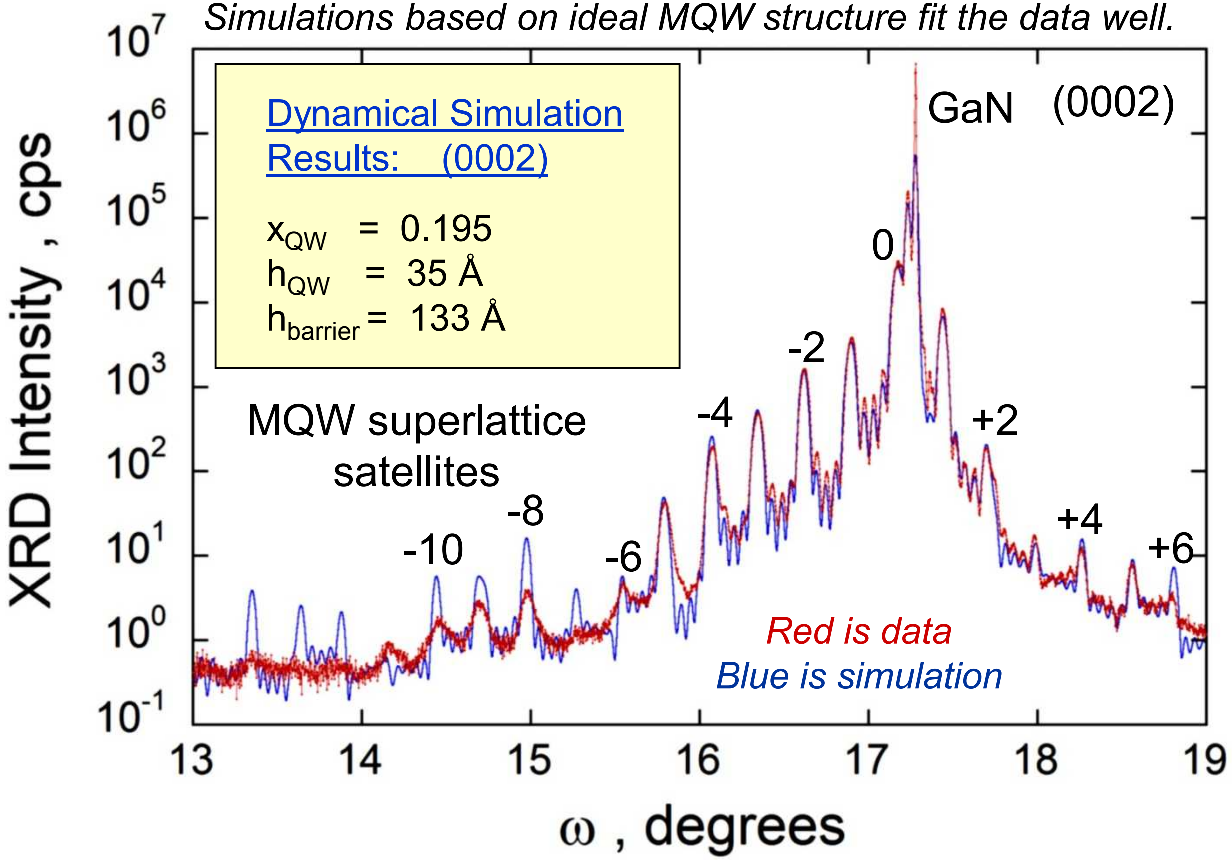


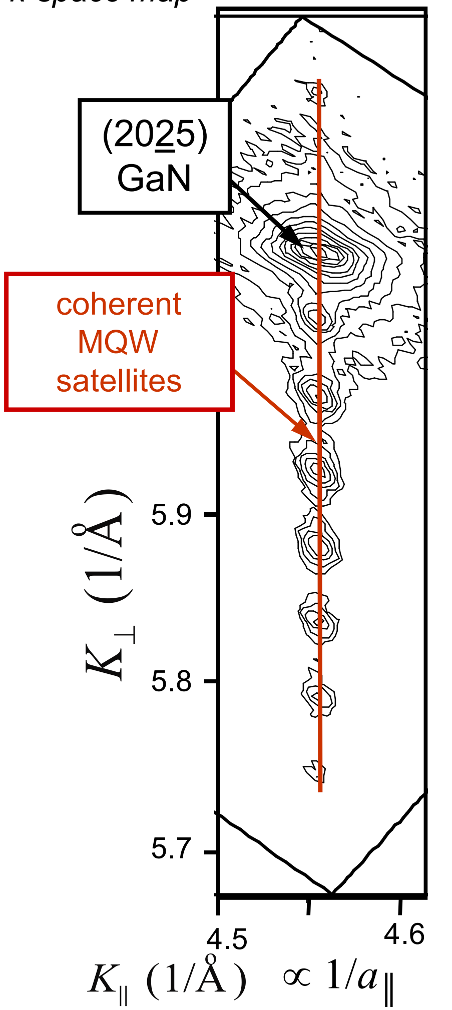
- **MOCVD reactor** – Veeco D125 short-jar - 3-2” wafers simultaneously.
- **Precursors –** trimethyl sources of In, Al, and Ga, and $Mecp_2Mg$ and SiH_4 for p- and n-type doping.
- **Gases –** NH_3, N_2, H_2 (no H_2 for InGaN)
- **Temperature –** GaN at 1050 °C, InGaN at 680 – 880 °C.

Growth differences between InGaN and GaN are:
lower temperature, higher NH_3, no H_2, slower growth rate (less total MO).

X-ray diffraction is used extensively to determine InGaN structural information

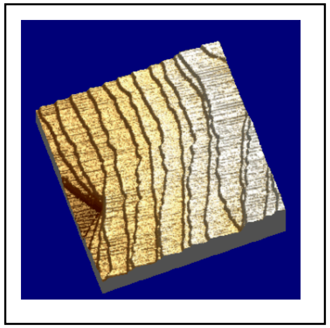
Simulations based on ideal MQW structure fit the data well.

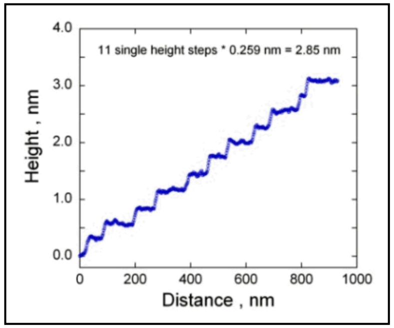




Both the dynamic diffraction fit and lack of change in $K_\parallel$ in the k-space map indicate that the InGaN QW are coherently strained.

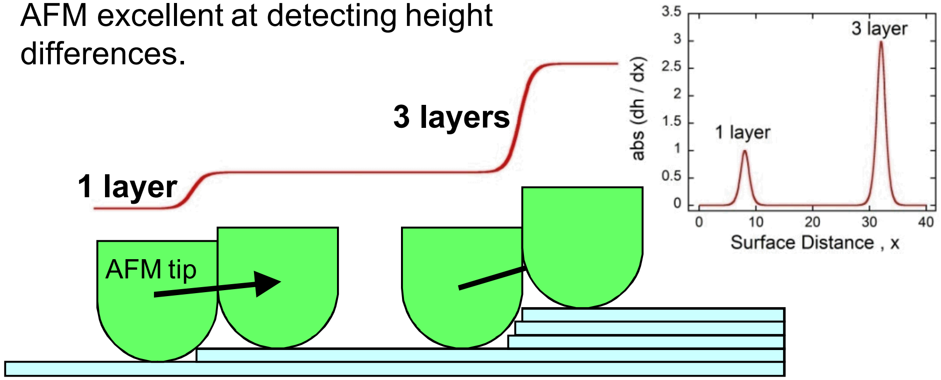
Counting step height distributions



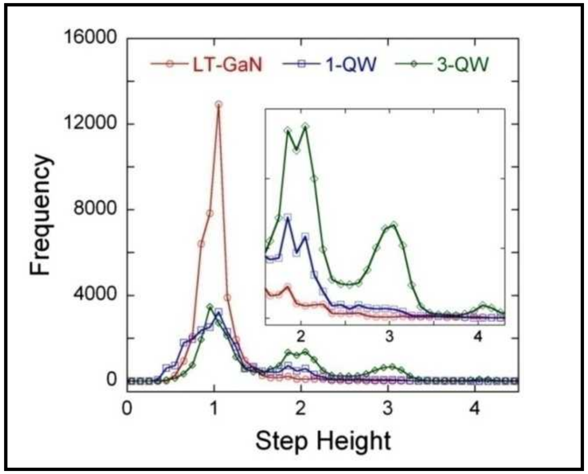


Can use AFM image to quantify the step heights, single layer steps, double layer steps, triple layer steps, etc...
1). Calculate the first and second derivative.
2). Magnitude of 1st derivative gives number of layer steps.
3). Second derivative = 0 gives step location.

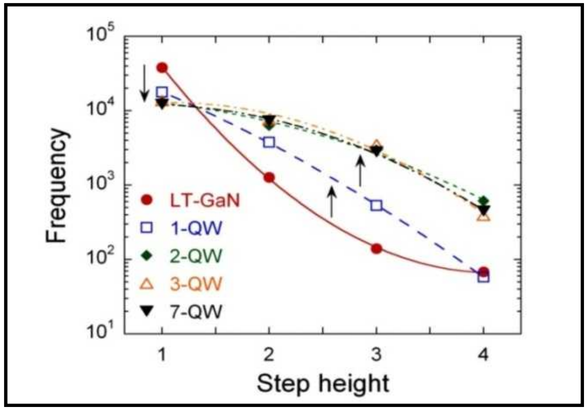
AFM excellent at detecting height differences.



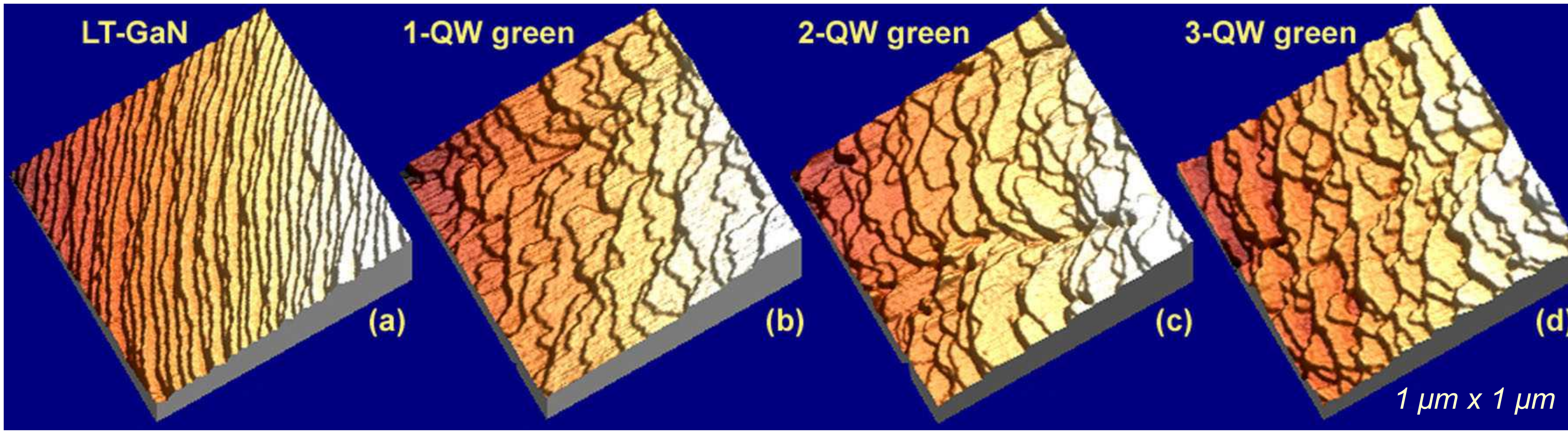
Histogram of the 1st derivative heights



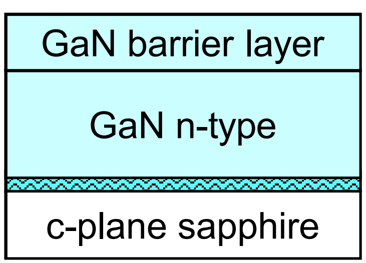
Binned distribution of step heights



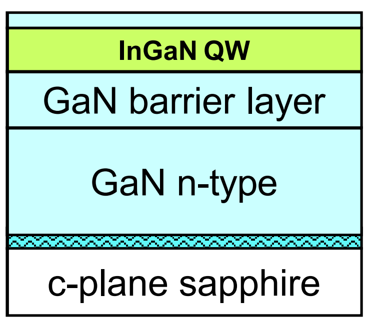
Observation of increased multi-layer steps in green MQWs

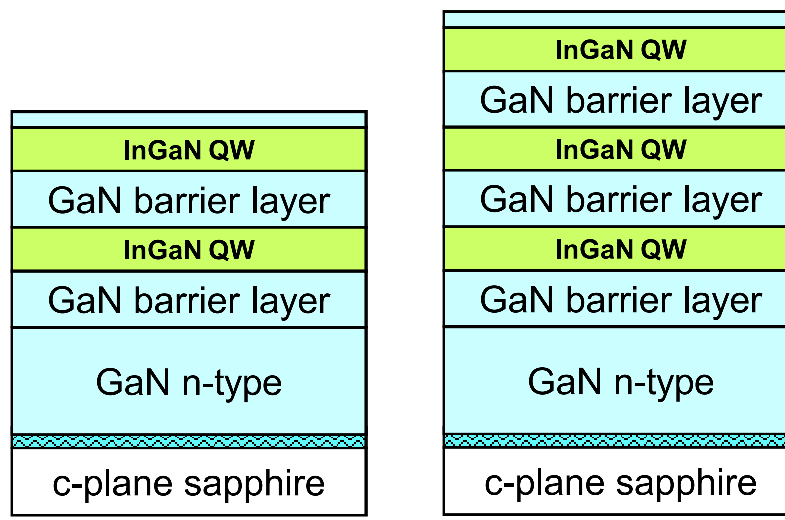


10 nm thick LT GaN barrier layer has the same step structure as the underlying HT GaN.

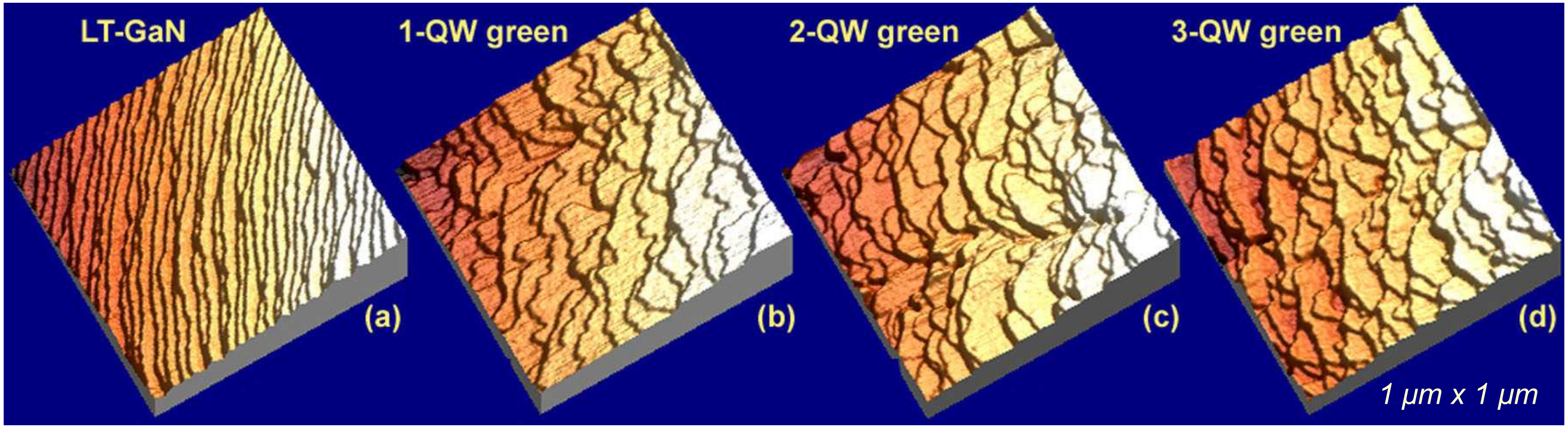


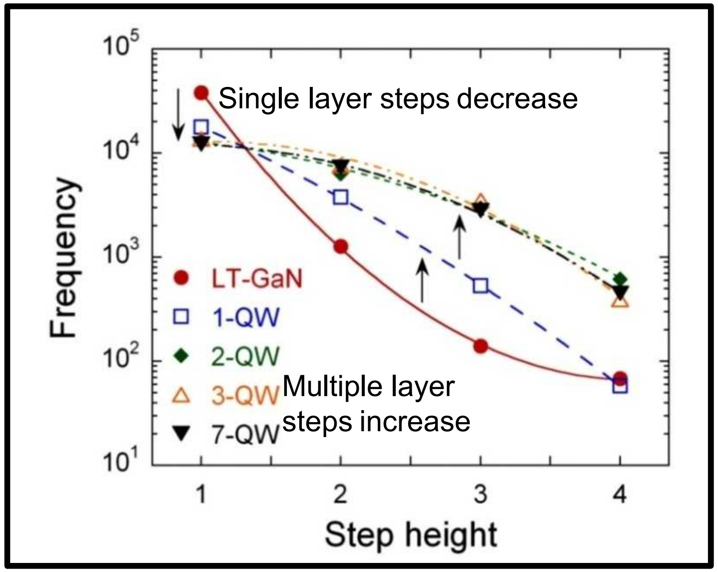
Addition of single 3 nm InGaN QW capped with 1.5 GaN barrier layer





Observation of increased multi-layer steps in green MQWs



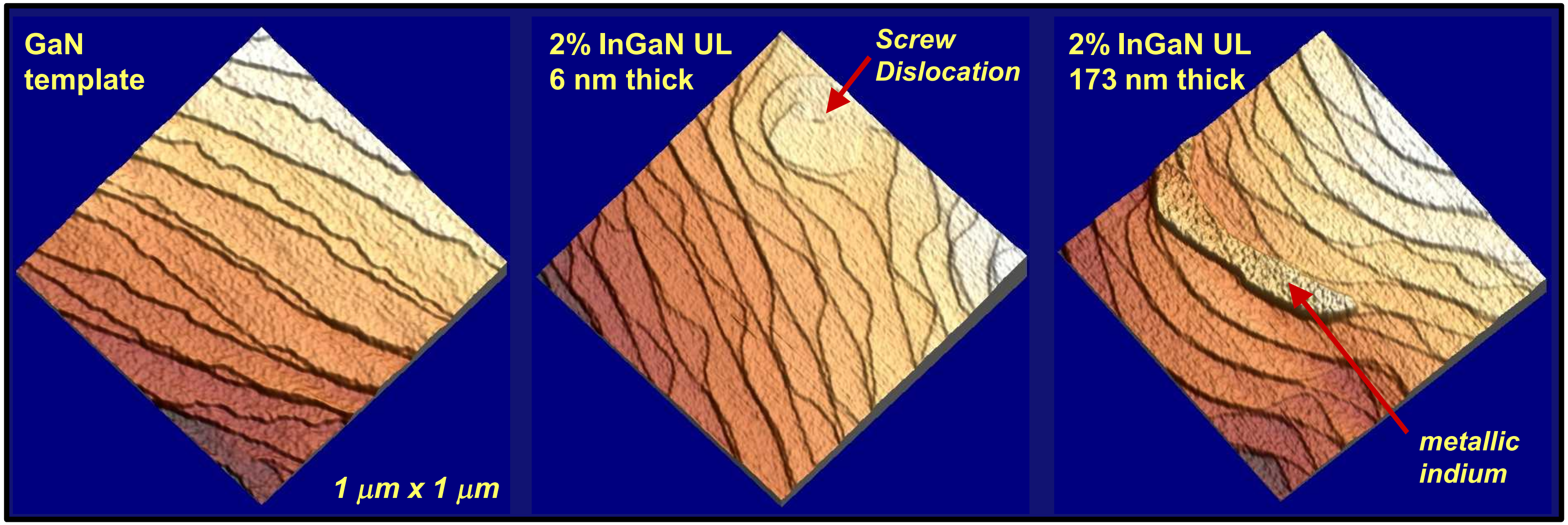


As the number of QWs increases the number of multiple layer steps increases, but reaches a steady configuration after the 2nd QW.

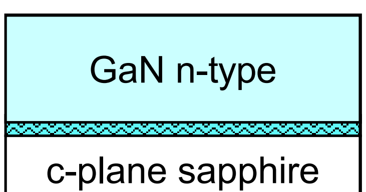
Suggests that while the InGaN QWs roughen the surface, the GaN barrier layer smooth the surface.

What are the smoothing mechanisms?

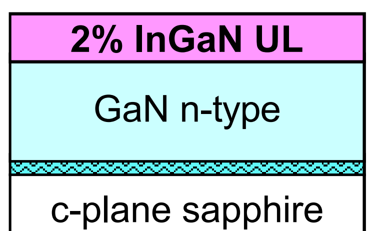
Adding indium to GaN growth changes the step structure

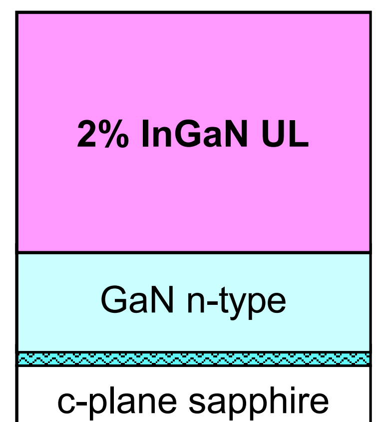


Same GaN template used for each growths.

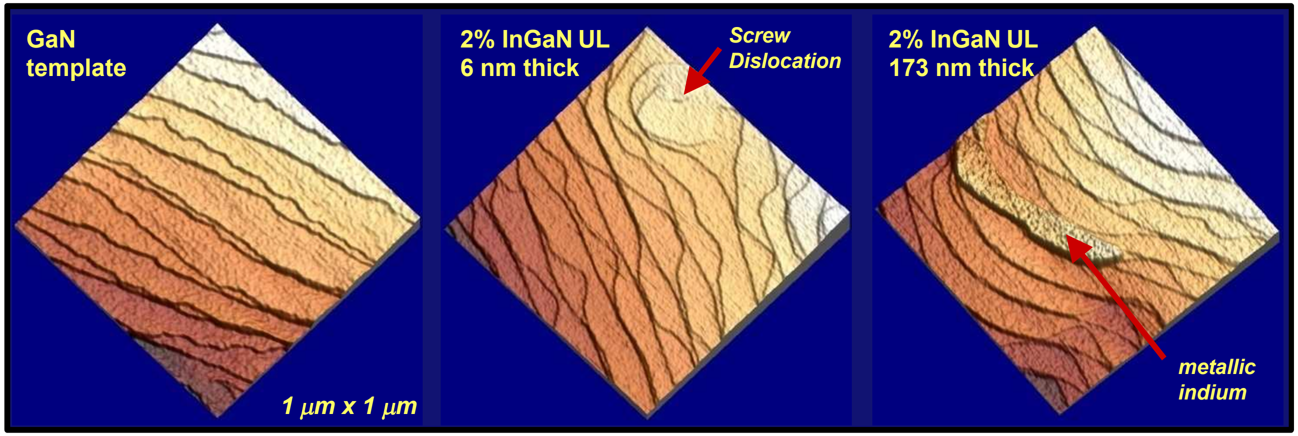


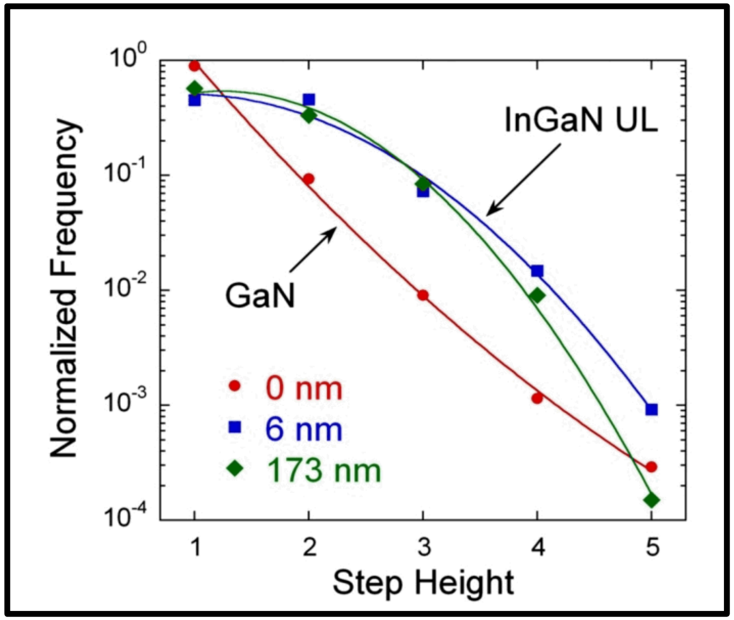
InGaN growth at 880 ºC using a high flow rate of indium





Adding indium to GaN growth changes the step structure

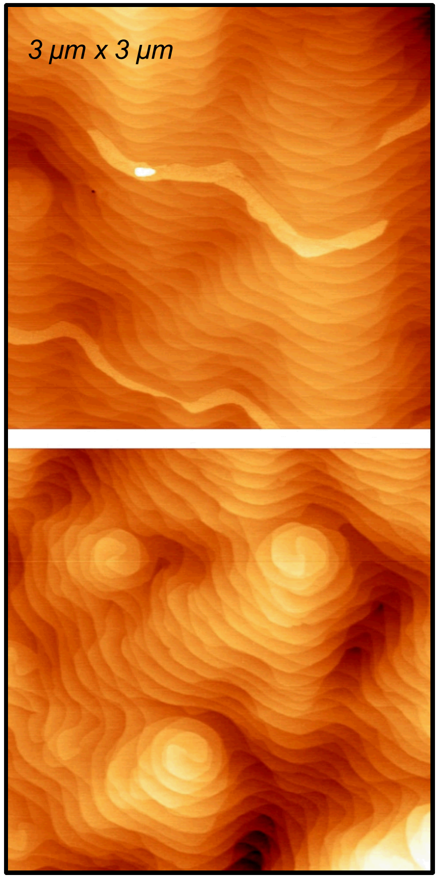




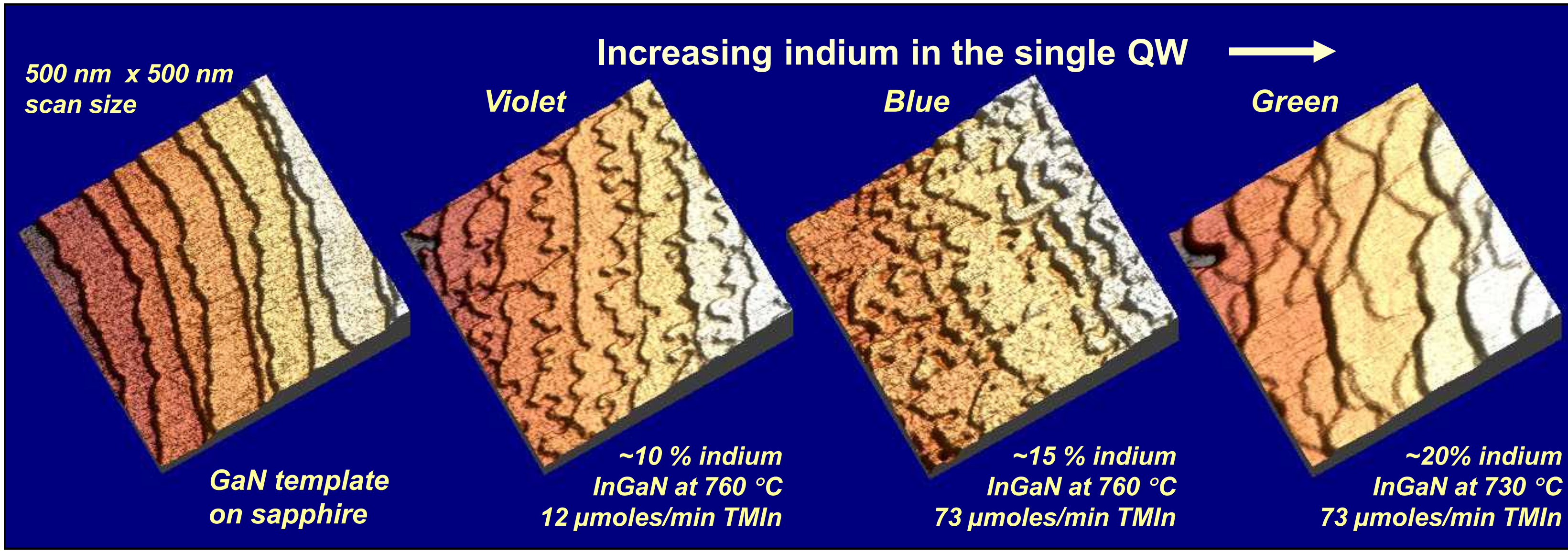
- The InGaN step height distribution changes after 6 nm of growth and remains similar as the thickness increases.

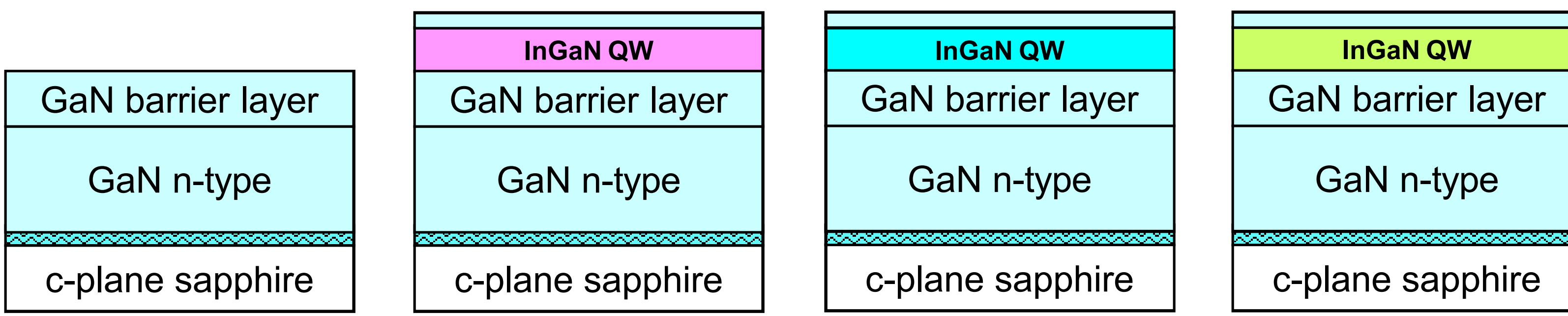
- Increased step edge meandering implying a reduction in the step-step repulsion energy.

Metallic indium can be removed using dilute nitric or hydrochloric acid.



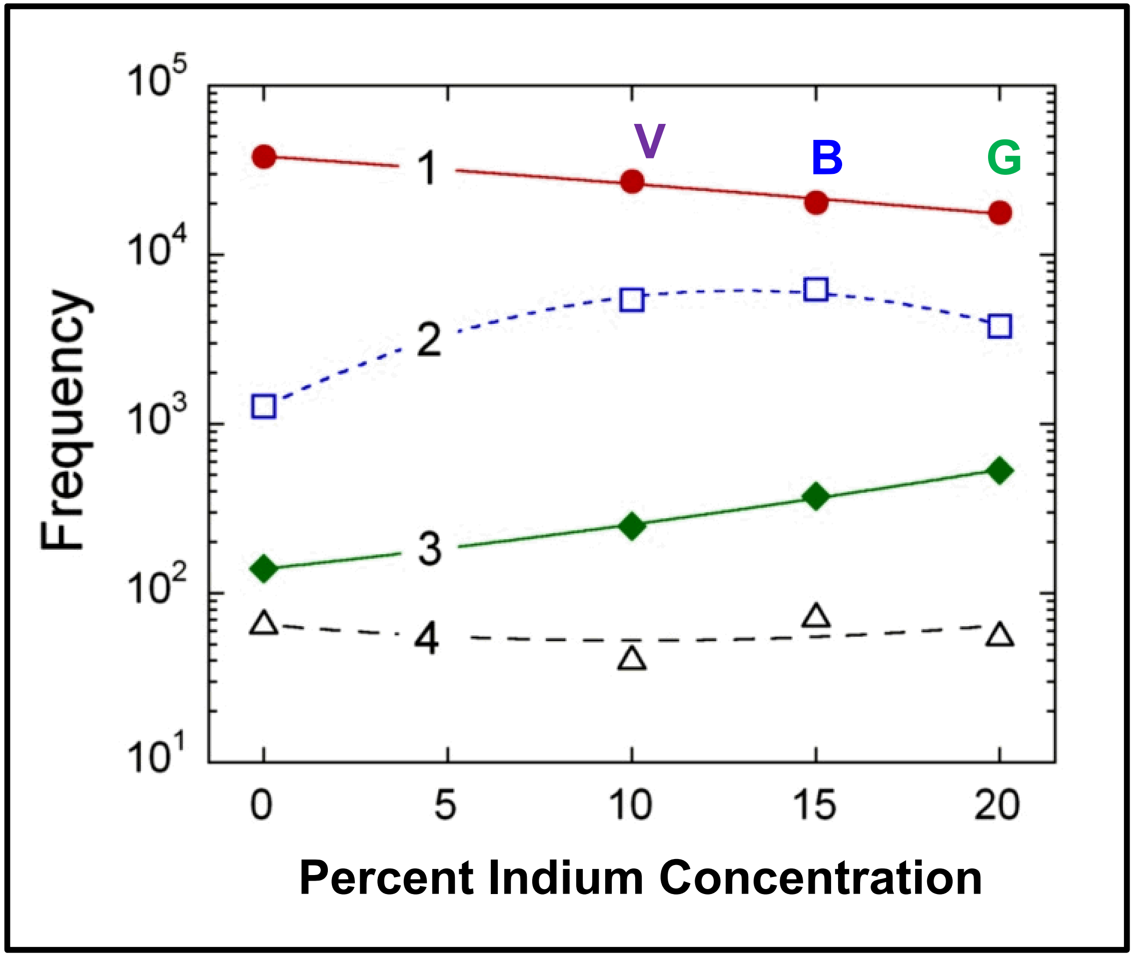
Increased indium in single QWs increases multi-layer steps





The multiple-layer step density increases as the indium incorporation increases

Increased indium in single QWs increases multi-layer steps



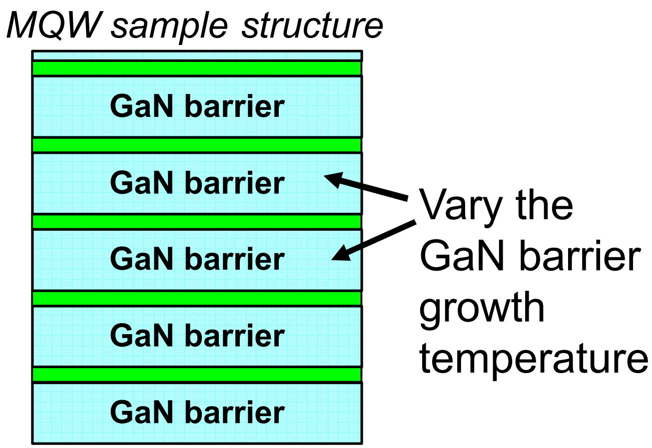
Indium concentration is measured using XRD of MQW structures.

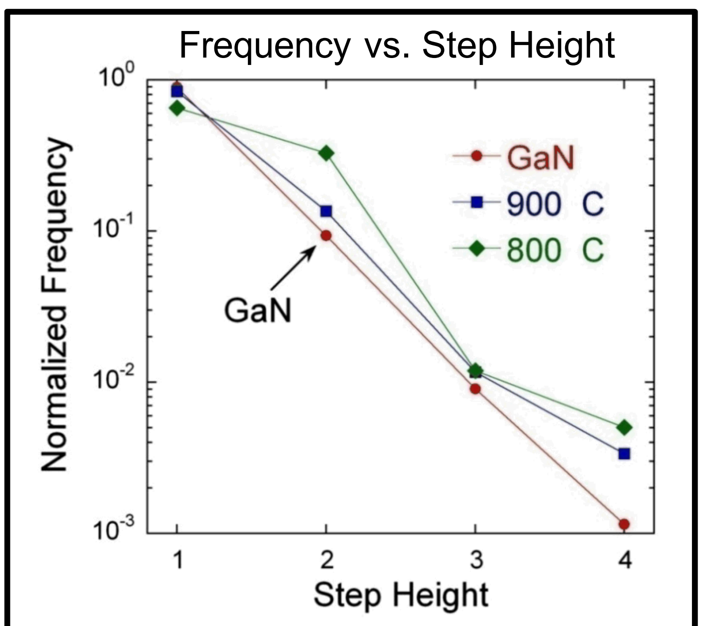
As the indium increases the frequency of single-layer steps decreases and the frequency of multiple-layer steps increases.

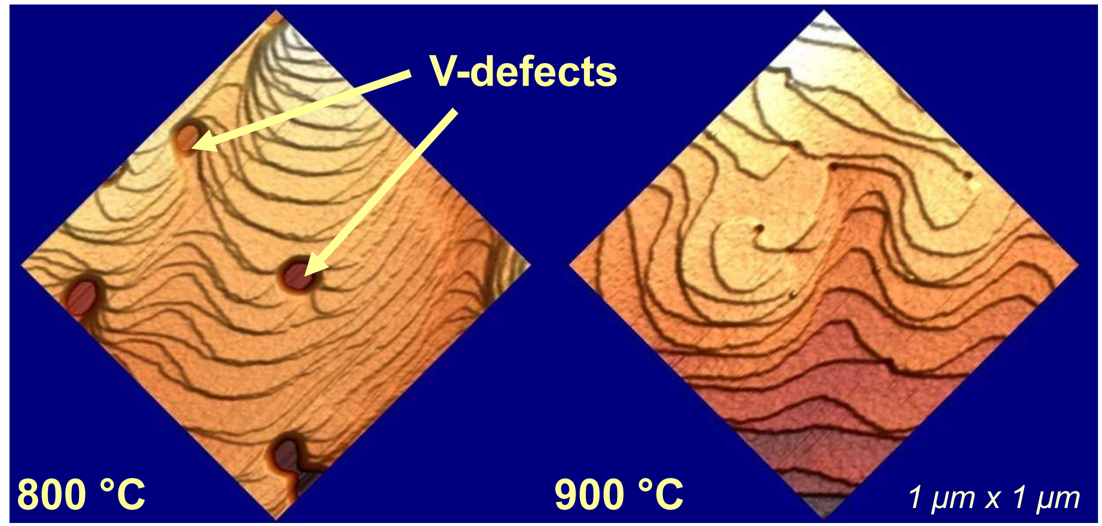
Direct correlation between the increased indium concentration and the increase in multiple-layer steps.

Increasing strain in the InGaN QW →

Control of InGaN steps using GaN barrier growth temperature







Increase in the frequency of double-layer steps at 800 °C compared to 900 °C.

The 900 ºC step height frequency distribution is closer to the starting GaN template than 800 ºC.

Implication: Higher temperature GaN barrier growth can be used to control the step morphology and control the surface smoothness.

Summary of InGaN step morphology

- Frequency of multiple-layer steps increase…
  - As the amount of indium increases (V → B → G).
  - As the number of QWs increases (up to 2).
  - In thin InGaN underlayers with only 2% indium.
  - As the GaN barrier temperature is lowered.

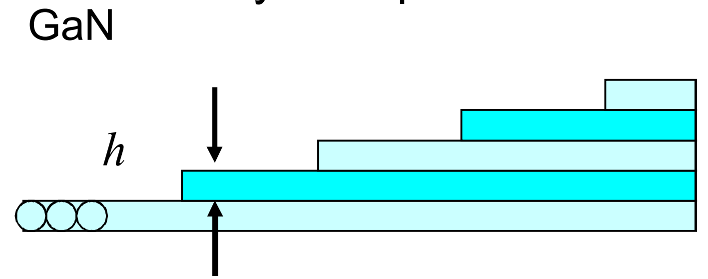
- Two influences on InGaN step structure.
  - <u>Strain relief</u> by forming a multiple-layer step edge. Not sufficient for bulk InGaN strain relief (XRD).

InGaN compressive strain will increase as the indium concentration increases and/or the film thickness increases. Strain relief by point defect generation is also possible.

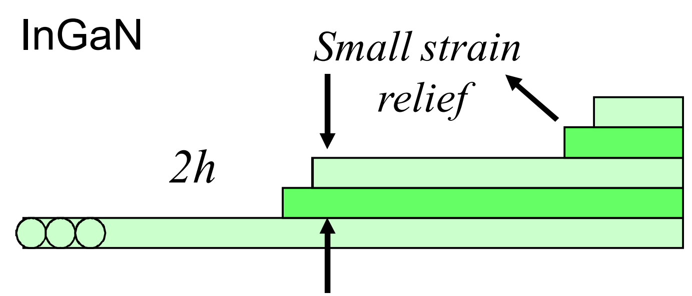
Explore possible changes in <u>growth mechanism</u> in comparing GaN to InGaN growth.

Measure the different smoothing mechanisms responsible for changes in growth morphology.

As-grown, GaN has single-layer steps.



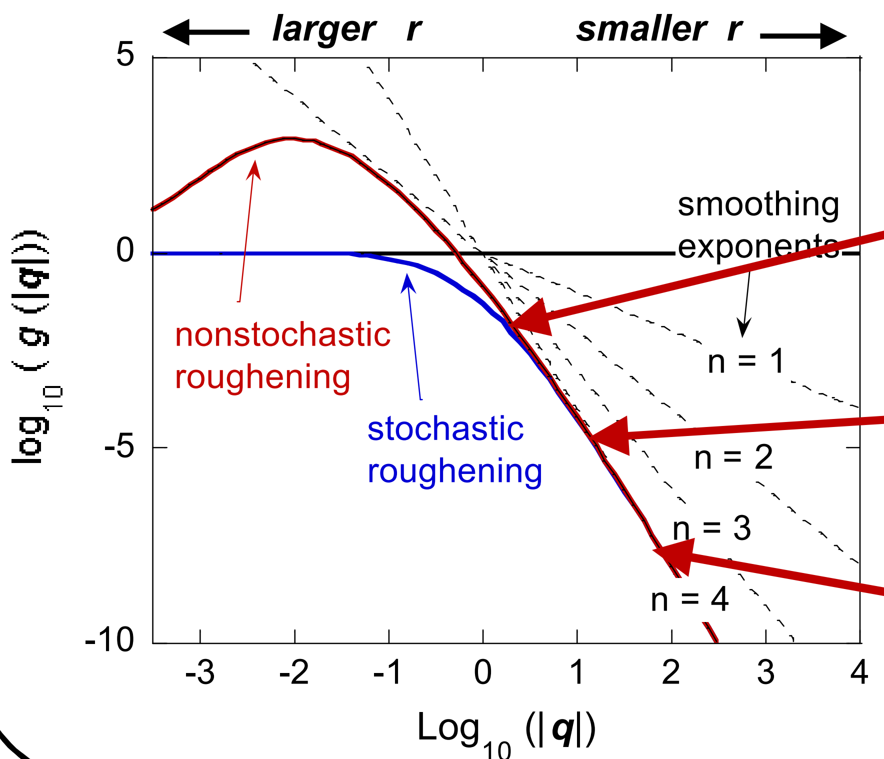
InGaN growth increases the frequency multiple-layer steps



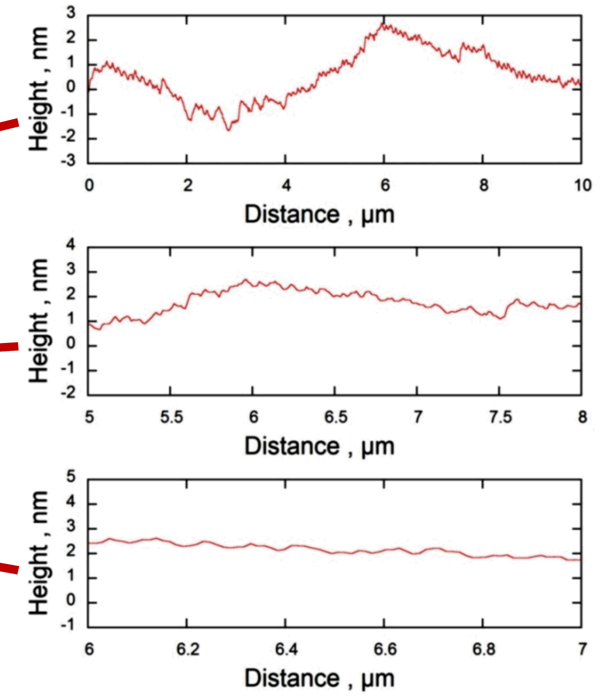
Smoothing mechanisms can be obtained from PSD analysis

Power spectral density (PSD) is the height-height correlation function from AFM
PSD or *g* can be calculated from h(*x*,*y*) as a function of *q, where q = 1/r*.

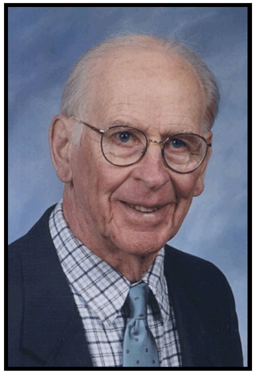
Discussed by Tong and Williams in Ann. Rev. Phys. Chem. 45, 401 (1994).



$$\sigma_{RMS} = (\Sigma\, g\,(\,q\,))^{1/2}$$



Various smoothing mechanisms calculated by Herring



Conyers Herring
1914 - 2009

J. Appl. Phys. 21, 301 (1950).

The PSD can be smoothed by various mechanisms that decrease *g*(*q*) at large *q*,

$$g(|\mathbf{q}|,t) \propto \frac{\Omega}{c_n|\mathbf{q}|^{n}}$$

Smoothing mechanisms

n = 1 - plastic flow driven by surface tension

***n* = 2 - evaporation and recondensation**

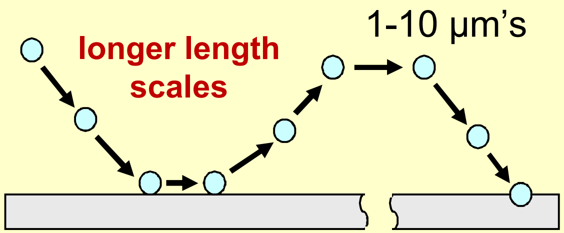
n = 3 - volume diffusion

***n* = 4 - surface diffusion**

Geometric details of mechanisms could influence the values of *n* by as much as 0.5.

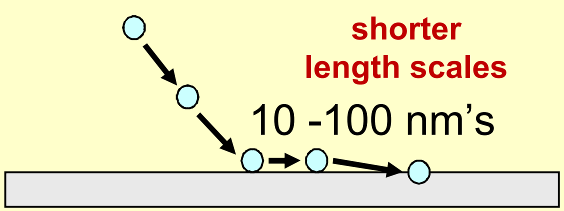
Mechanism influences length scale over which the smoothing occurs.

***n* = 2**
evaporation and recondensation
(GaN for T > 900 °C)



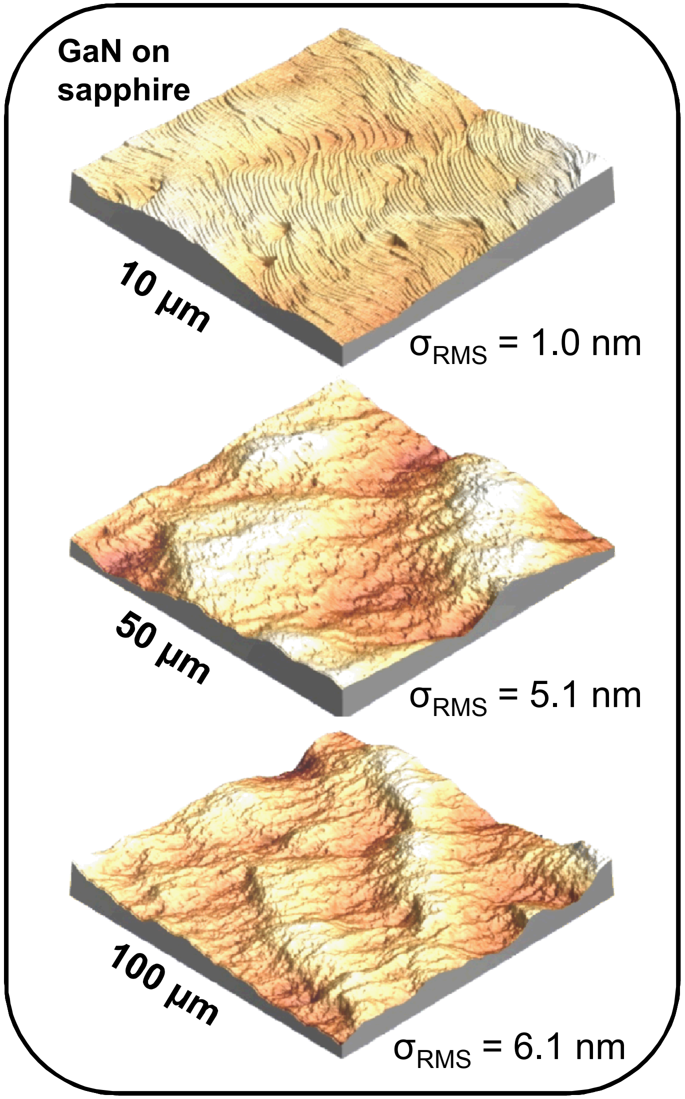
See Mitchell et al., JCG 222, 144 (2001).

***n* = 4**
surface diffusion
(InGaN and GaN T < 900 °C

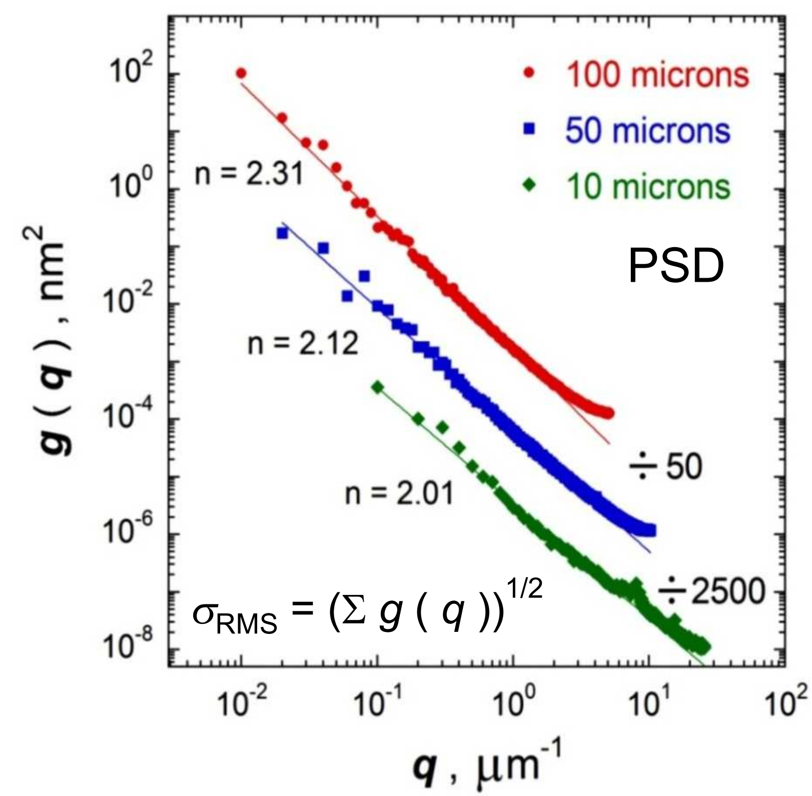


See Koleske et al., JAP 84, 1998 (1998).

PSD analysis of GaN films on sapphire



σ_{RMS} typically depends on scan size

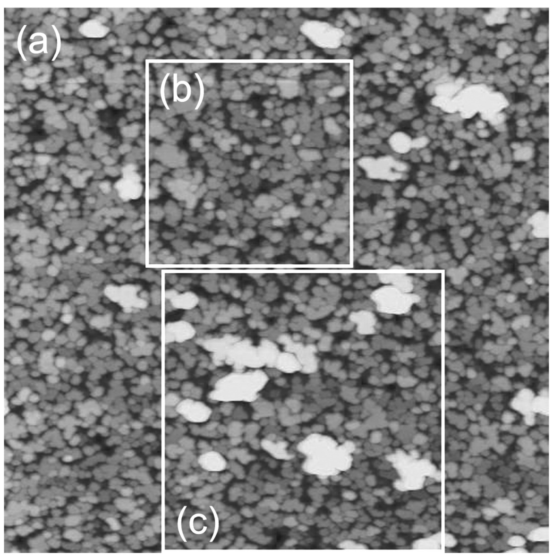


***n* ~ 2 - implies the smoothing mechanism is evaporation and recondensation of Ga atoms**

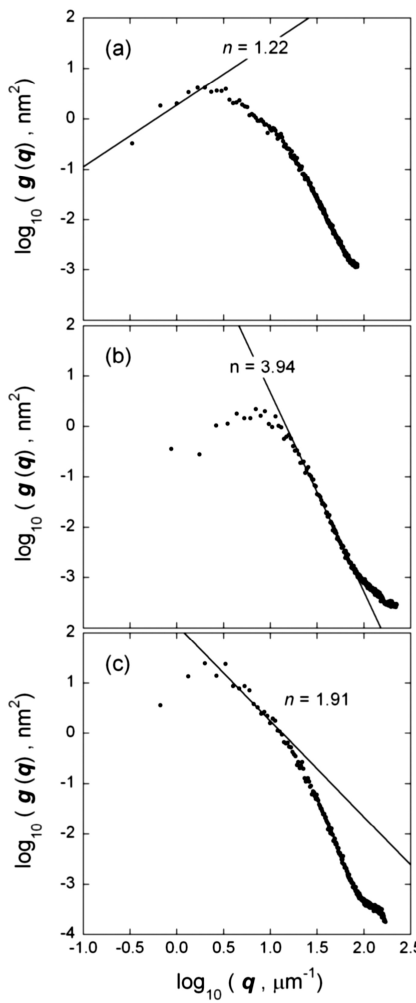
PSD analysis of GaN nucleation layer evolution

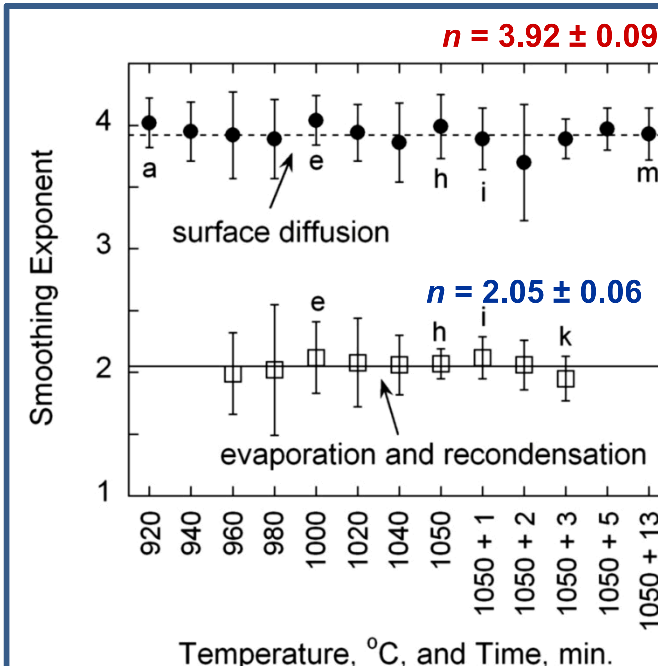
AFM scan of NL at 1000 °C

Low temperature deposited GaN NL is smoothed via surface diffusion, *n* = 4.



GaN nuclei form out of deposited GaN NL via evaporation and reconden-sation mechanism, *n* = 2.



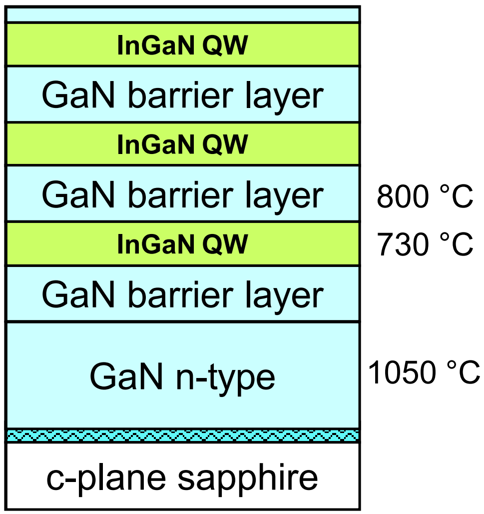


Consistent with gas phase transport model of Mitchell, *et al.*, J. Crystal Growth 222, 144 (2001).

Koleske et al., J. Crystal Growth 273, 86 (2004).

PSD analysis of green MQWs as number of QWs increases

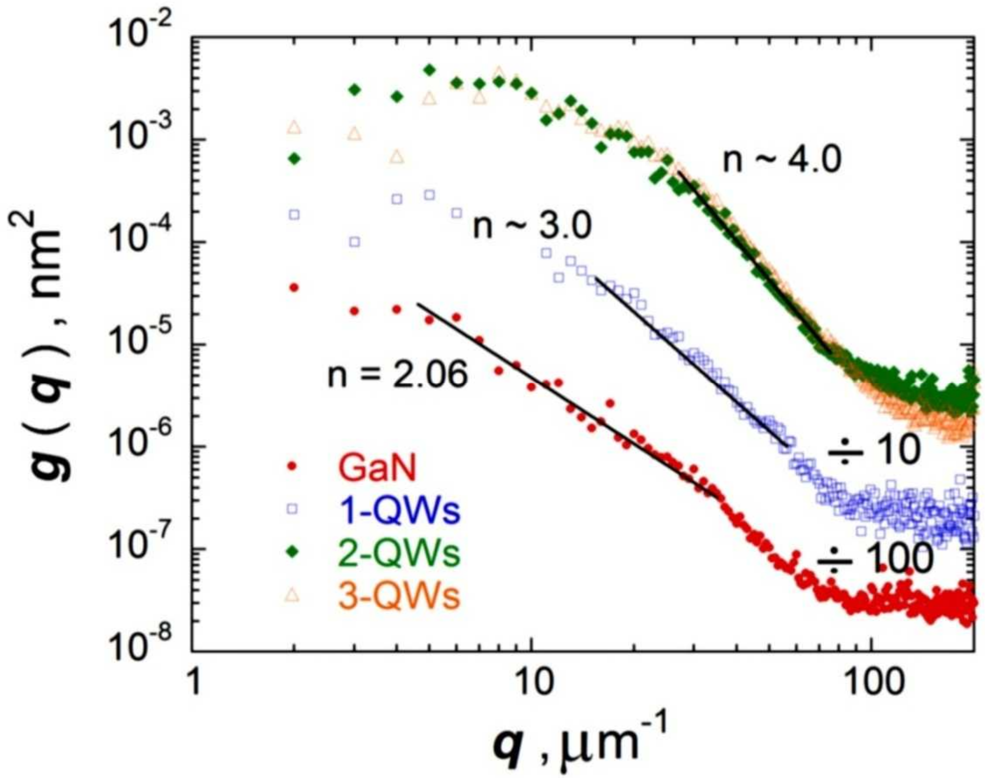
Green MQW structure



n = 2 = evaporation / recondensation

n = 4 = surface diffusion

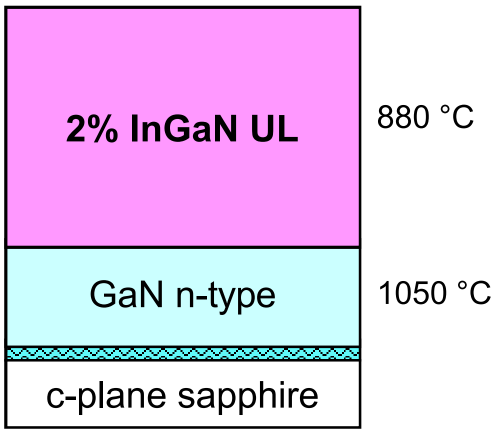
n = 3 = average 2 & 4?



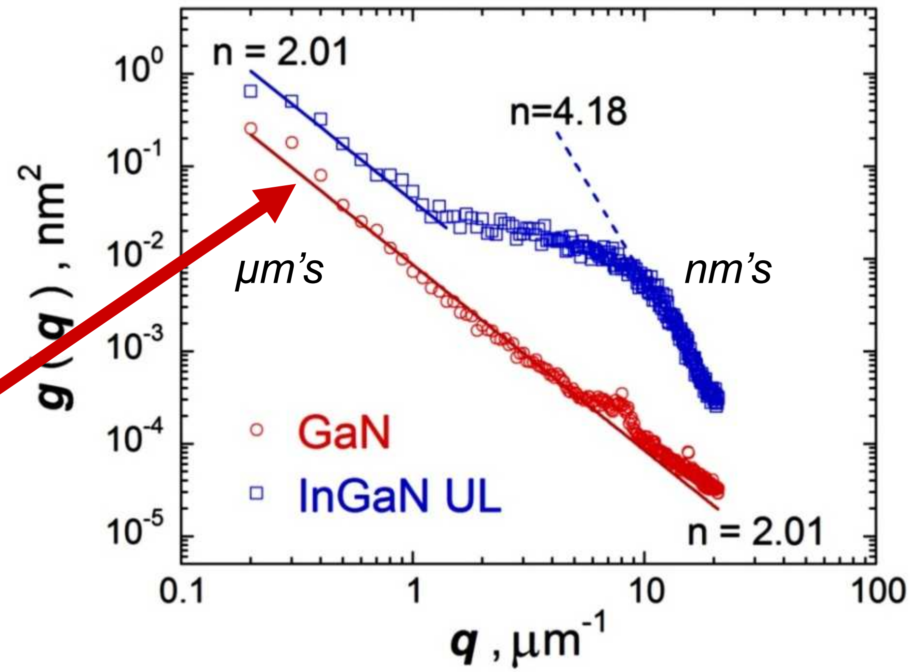
Smoothing exponent increases from 2 to 4 as the number of QWs increases.

PSD analysis of 2% indium concentration InGaN (ULs)

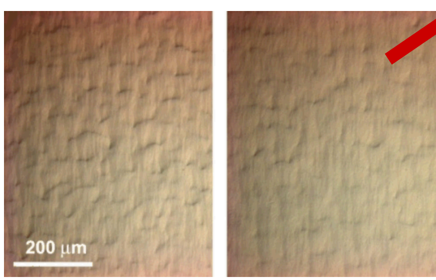
InGaN (2%) UL structure



PSD for GaN and 173 nm thick InGaN



Starting GaN template

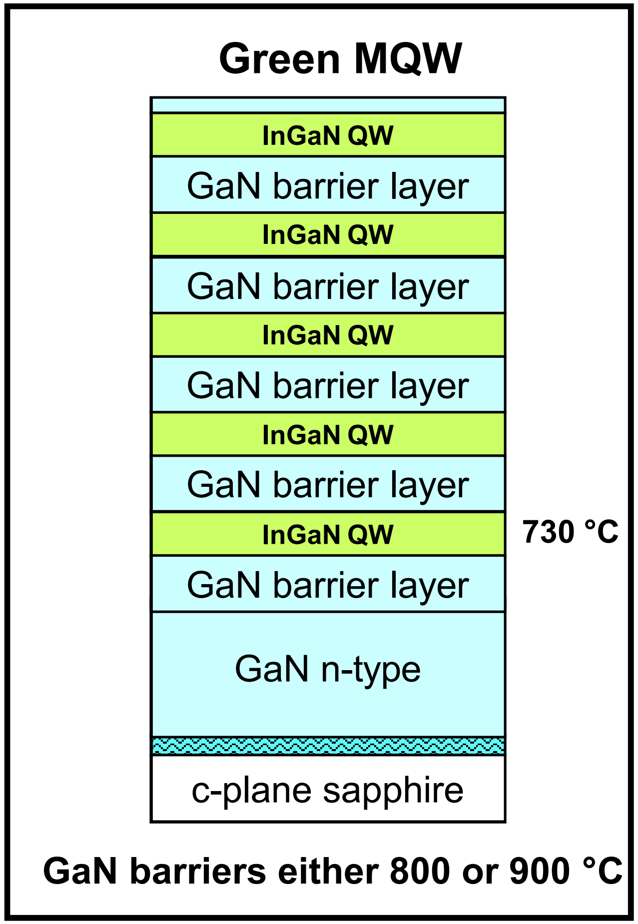


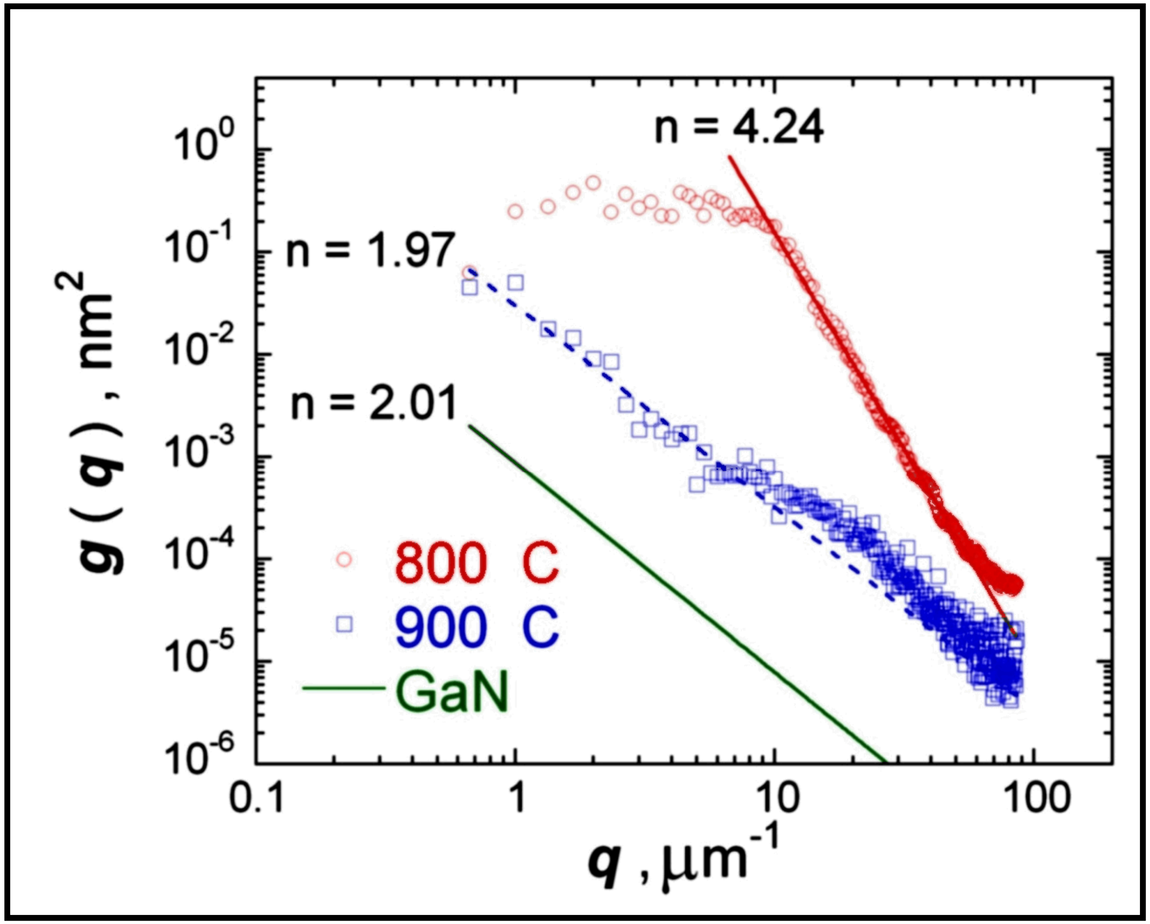
After growth of 173 nm InGaN UL

No change in large scale roughness

2% InGaN UL roughening is caused by surface diffusion (short length scale).

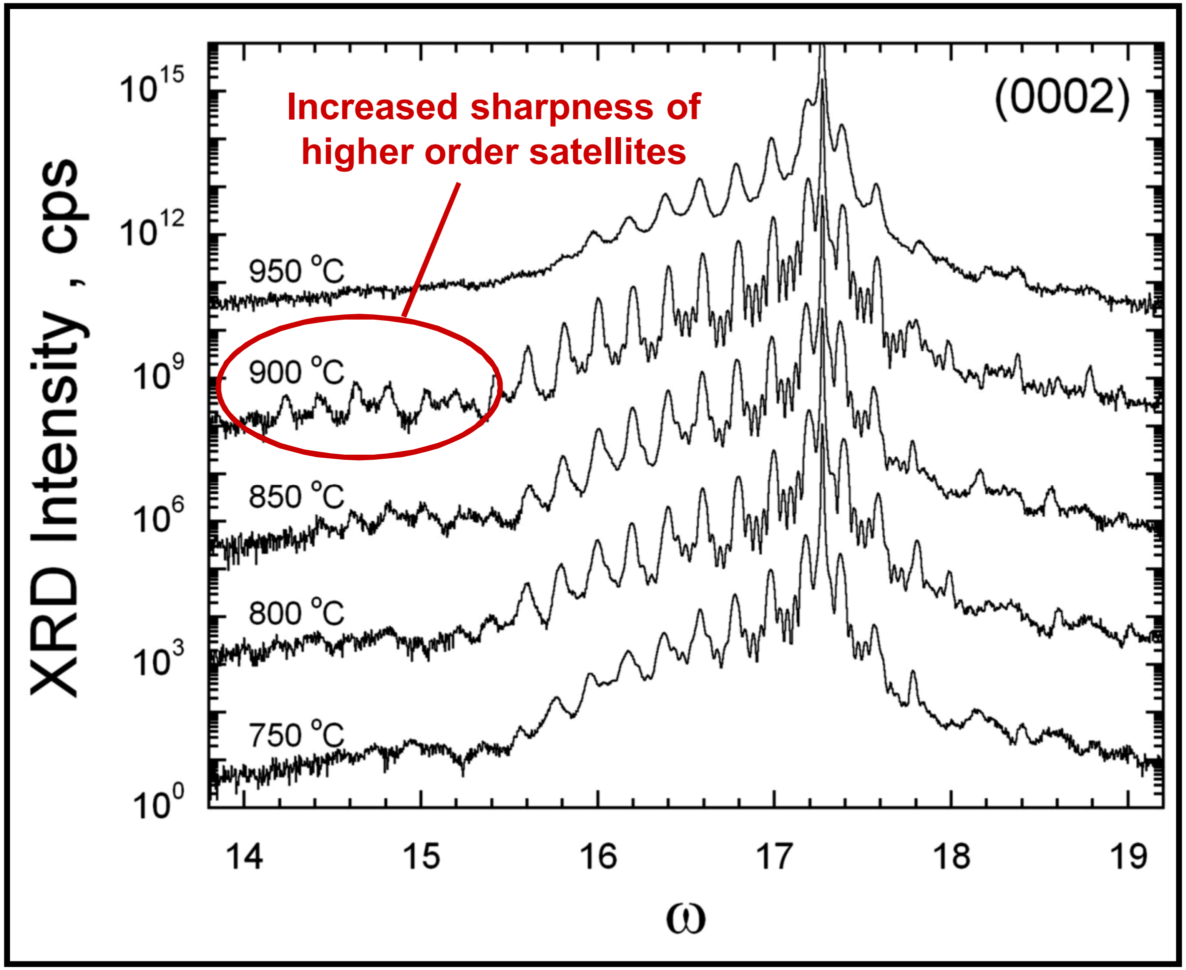
PSD analysis of the green MQWs with different GaN barrier growth temperature





As the GaN barrier growth temperature is increased, the surface is smoothed increasingly by surface diffusion (n ~2 (900 °C) for to n ~4 (800 °C)).

XRD analysis of the green MQWs with different GaN barrier growth temperature



Using dynamic diffraction fits

Indium content = 0.21 ± 0.01

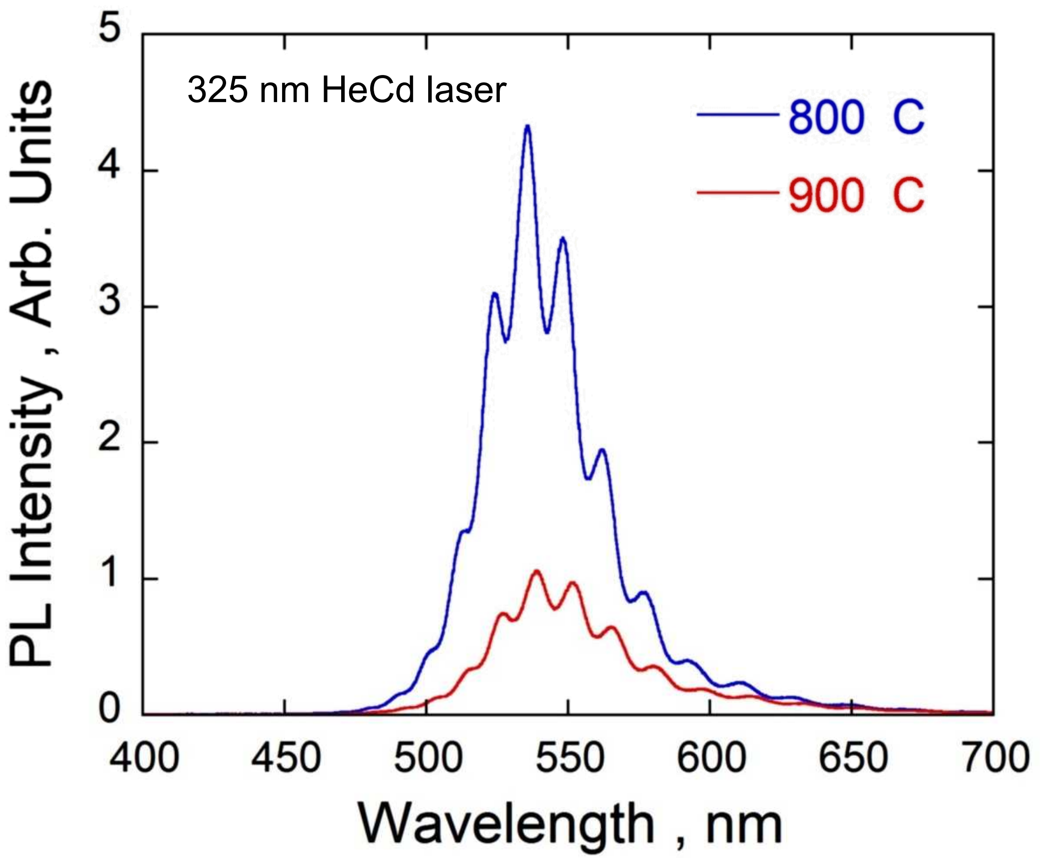
QW thickness = 3.5 ± 0.1 nm

GaN barrier = 19.6 ± 0.5 nm

The sharpness of the higher order satellites indicates better interface alignment for GaN barrier layers grown at 900 ºC compared to those grown at 800 ºC.

PL analysis of the green MQWs with different GaN barrier growth temperature

Higher PL intensity for GaN barriers at 800 ºC



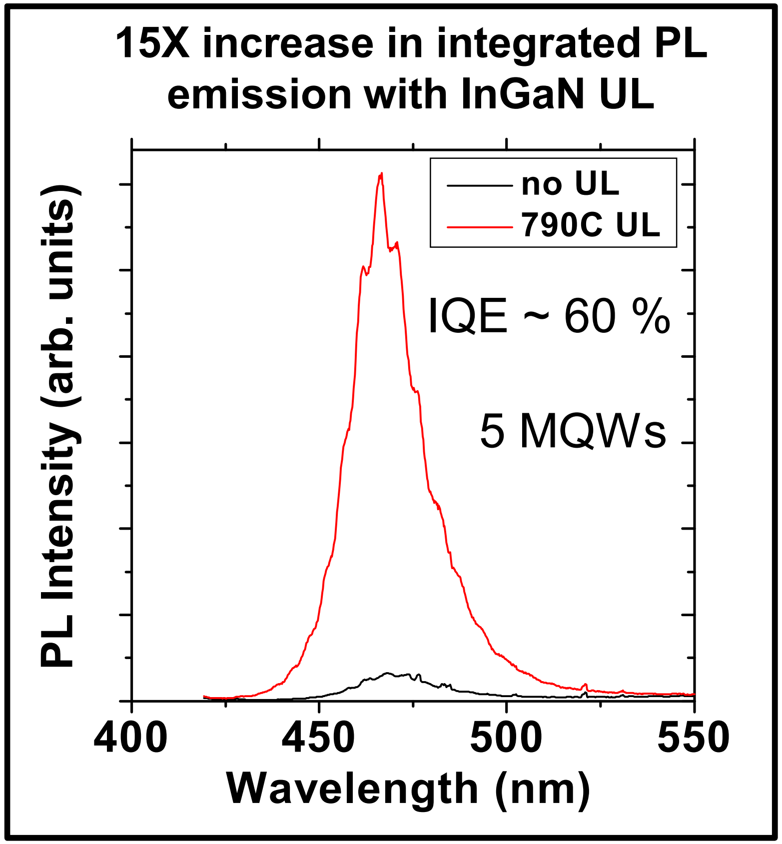
Same intensity and wave-length trends are observed for resonant optical pumping at 407 nm.

Suggests that the rougher step morphology produces QW with increased PL intensity.

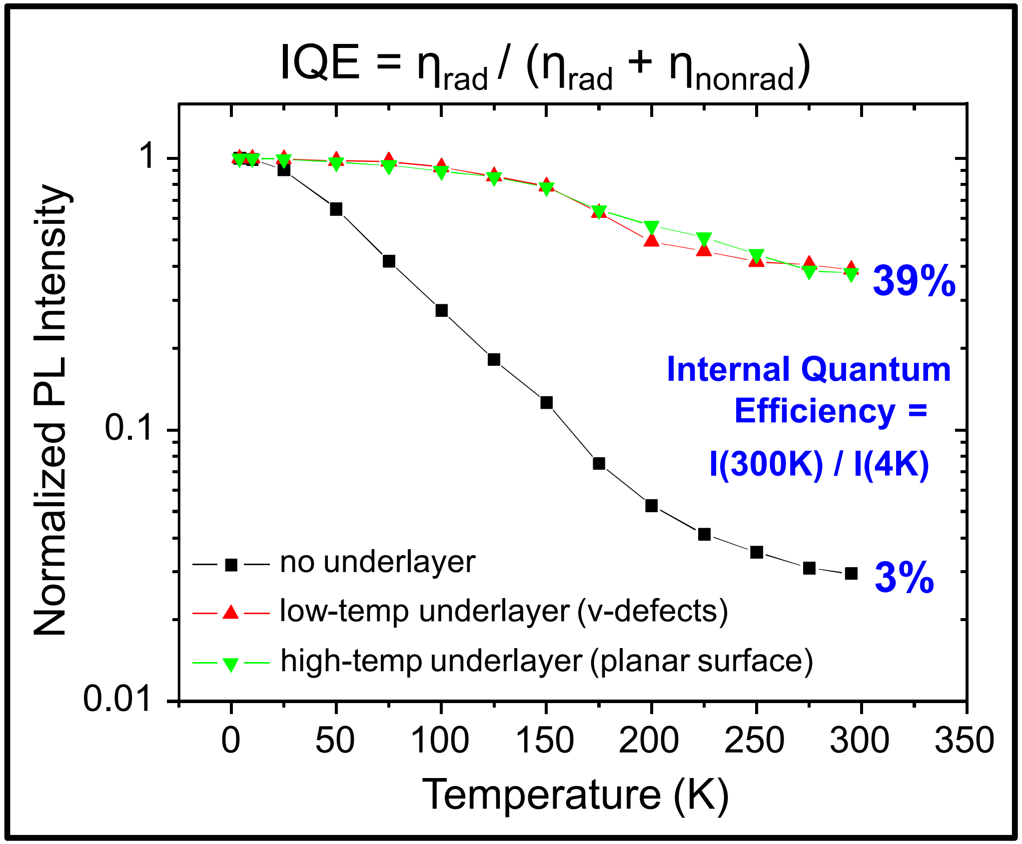
The morphology could produce QW thickness fluctuations coupled to the strong piezo-electric fields to produce electron/hole localization. Graham et al., JAP 97, 103508 (2005).

InGaN underlayers increase IQE of MQWs

Especially help emission of single QWs



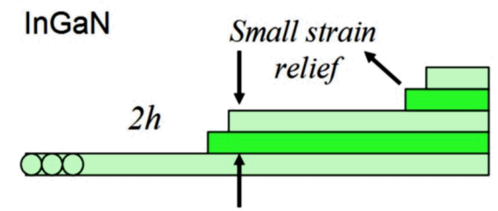
Temperature dependent PL studies by Mary Crawford



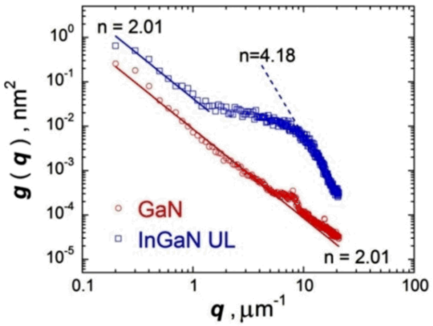
Underlayers enhance efficiency compared to same MQW with no underlayer.

Conclusions

InGaN growth increases the frequency of multiple-layer steps



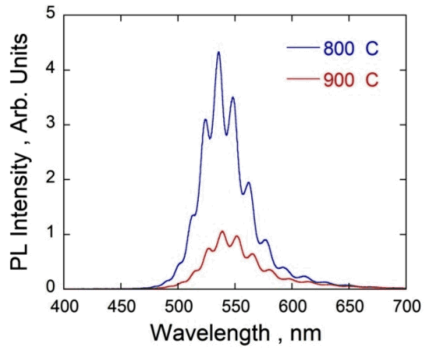
During InGaN growth the frequency of multi-layer steps increases – *strain relief*.



InGaN morphology can be controlled with temperature

- **Lower T – InGaN – multiple steps – surface diffusion**
- **Higher T – GaN – single steps evaporation/recondensation**

Demonstrated correlation between increased multi-layer steps and increased PL emission.
The exact physical reason for this correlation is currently under investigation.



Thanks to:

Sandia Colleagues:

Steve R. Lee – XRD analysis, strain relaxed InGaN
Mary H. Crawford – PL, LED studies
Mike Coltrin – GaN NL evolution studies
Karen Cross – AFM images.
Jerry Thaler – MOCVD growth now at Lasertec.
J. J. Figiel – MOCVD tech.
J. M. Kempisty – MOCVD tech.
R. M. Biefeld – Manager