Photonics West, January 25th, 2012

SAND2012-0763P

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Steps toward understanding the growth and physics of visible group III-N LEDs

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

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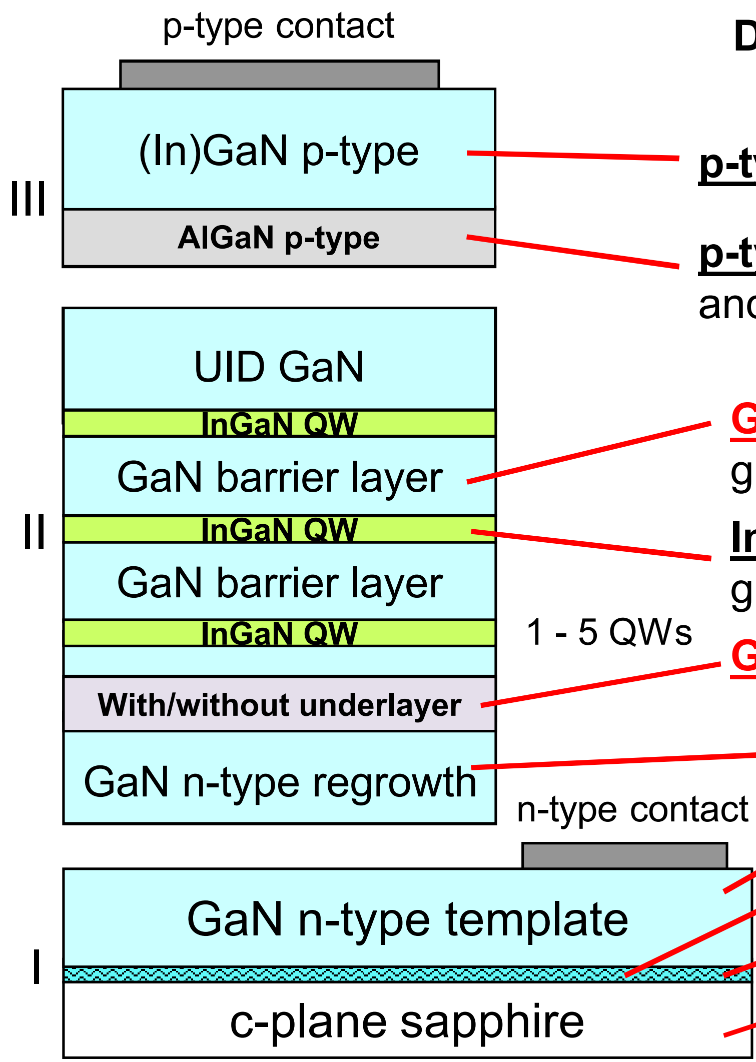
Outline

- Growth and PL emission intensity of InGaN MQWs
  - As the number of QWs increases
  - Using lower temperature layers under the QWs (underlayers).
  - GaN barrier growth temperature.
- AFM studies of InGaN/GaN growth morphology
  - Interplay of two smoothing mechanisms on morphology.
  - Commonality of morphology that leads to brighter QWs.
- Possible relationship between
  - QW efficiency,
  - growth morphology
  - non-radiative recombination centers (NRCs) reduction?

Typical LED structure and growth procedure



During growth changes in temperature, gas flows, molar flow rates, and pressure

p-type GaN – same temperature as electron block.

p-type AlGaN electron block grown at ~ 970 °C for blue and 930 °C for green – add H_2, 200 torr.

GaN barrier raise/lower temperature then back to QW growth temperature –Si doped.

InGaN QW growth at 770 °C for blue and 730 °C for green LEDs – use only N_2 and NH_3, 300 torr.

GaN or InGaN underlayer (UL) growth at 790 to 880 ºC

GaN bulk on top of annealed NL.

Heat NL to 1050 °C in NH_3, H_2, and N_2.

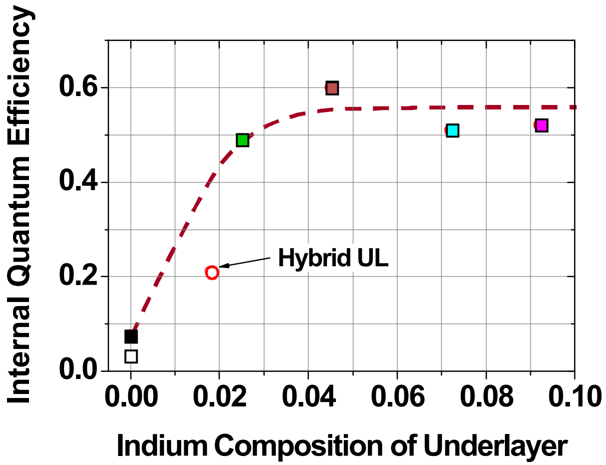
GaN nucleation layer at 530 °C, 500 torr.

Anneal sapphire in H_2 to 1050 °C.

Focus on section II

The "magic" of underlayers

- Underlayer (UL) - Lower temperature layer underneath the LED light emitting region.
- In many papers called "strain relief layer".
- Can be composed of lower temperature GaN, InGaN, multiple-QWs, or superlattice structures.
- Needs to be independently optimized for best QW emission intensity.

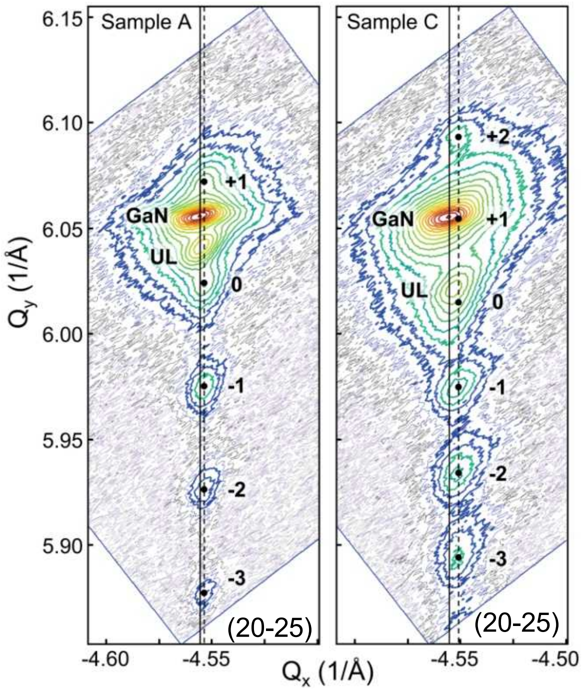


Improvement in the IQE of 450 nm MQWs when GaN and InGaN ULs are used.

Arrhenius analysis of the temperature dependent PL suggested that free carriers encounter **fewer non-radiative recombination centers (NRCs).**

S. Lee, et al., J. Appl. Phys. in review

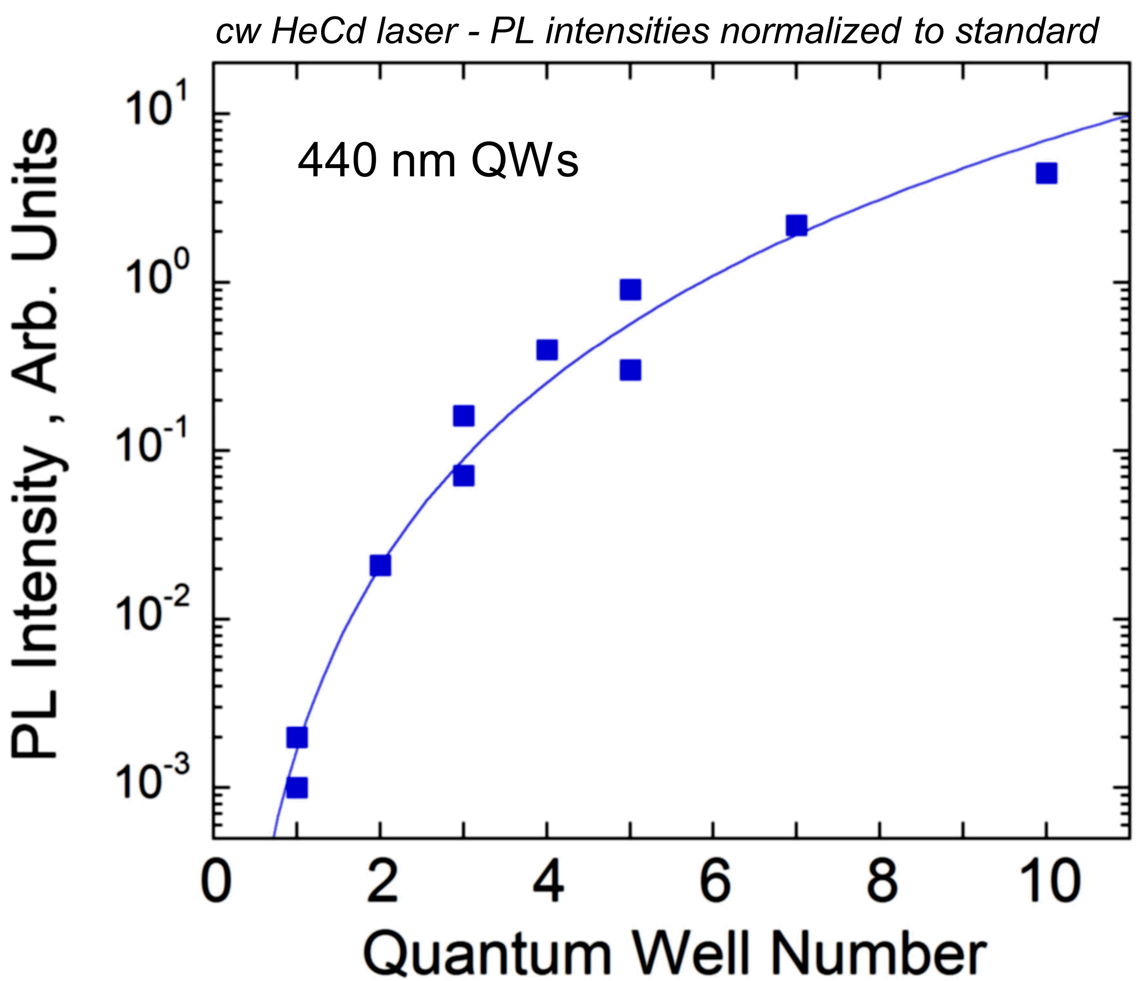
Strain relaxation is observed in k-space maps for MQWs on ULs.



Sample A - InGaN UL and a 5-period $In_{0.17}Ga_{0.83}N/GaN$ MQW – 2.8% per QW.

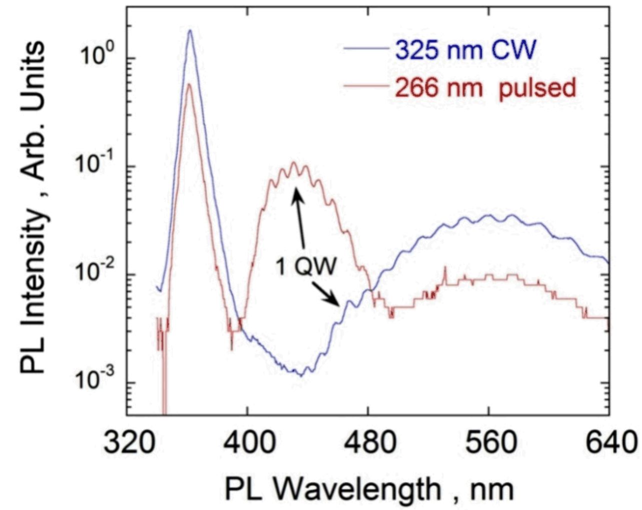
Sample C - InGaN UL and a 10-period $In_{0.20}Ga_{0.80}N/GaN$ MQW – 4.2% per QW.

First few quantum wells not very bright



Growth conditions – *n* period InGaN MQWs at 770 ºC, GaN barriers at 800 ºC, pressure 300 torr. Structure verified using XRD.

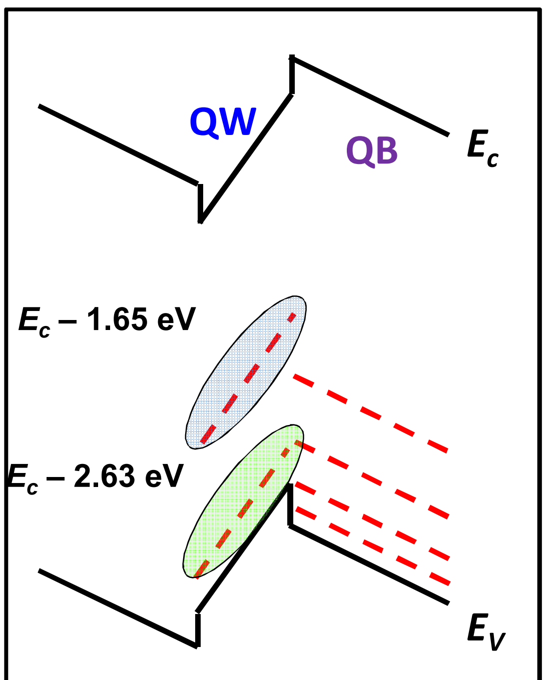
First QW optically dead
Barely observable using low intensity HeCd, brighter using pulsed 266 nm laser. Suggests first QW contains many non-radiative recombination centers (NRCs).

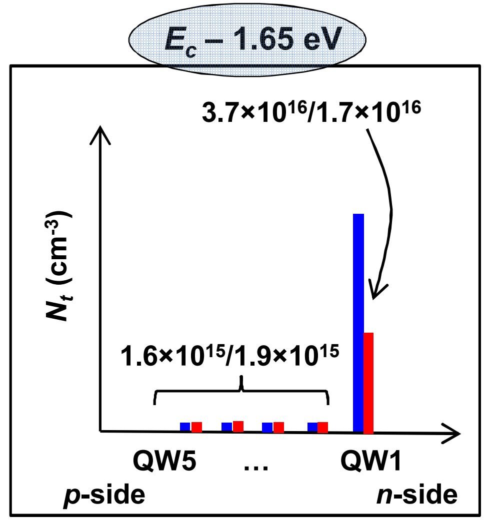


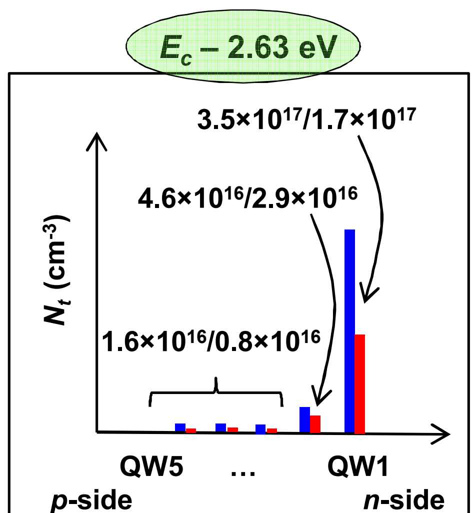
DLOS study confirms more defects in first QW

Deep Level Optical Spectroscopy (DLOS) by Andy Armstrong and Tania Henry for 440 nm LEDs grown on two different dislocation density GaN templates.

TDD = 7 × 10^8 cm^{-2}
TDD = 3 × 10^9 cm^{-2}







E_C – 1.65 eV defect level

Potential non-radiative recombination center (NRC)

More in 1st vs 5th QW.

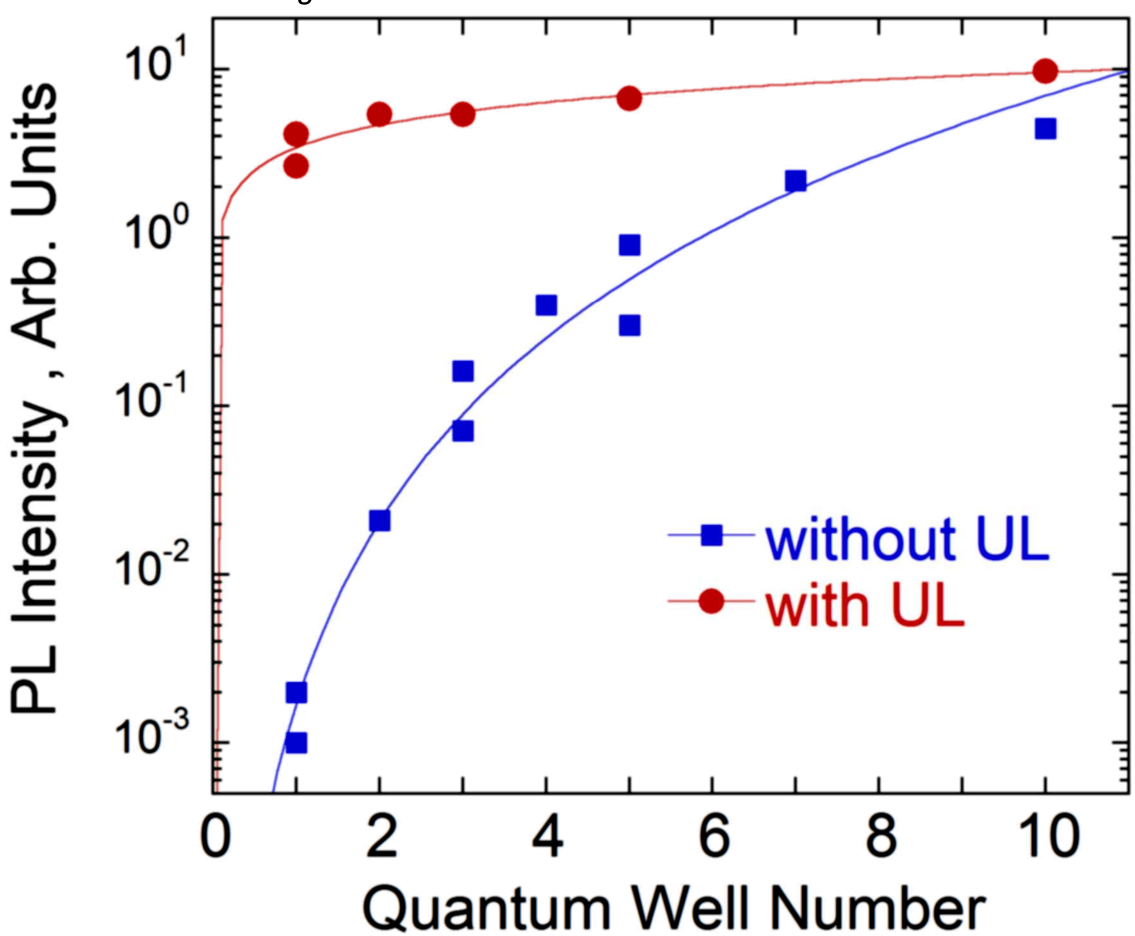
E_C – 2.63 eV defect level

More in 1st vs 5th QW.

More NRCs in 1st QW than subsequent QWs

Same QWs on dilute indium InGaN layer

Using 325 CW HeCd

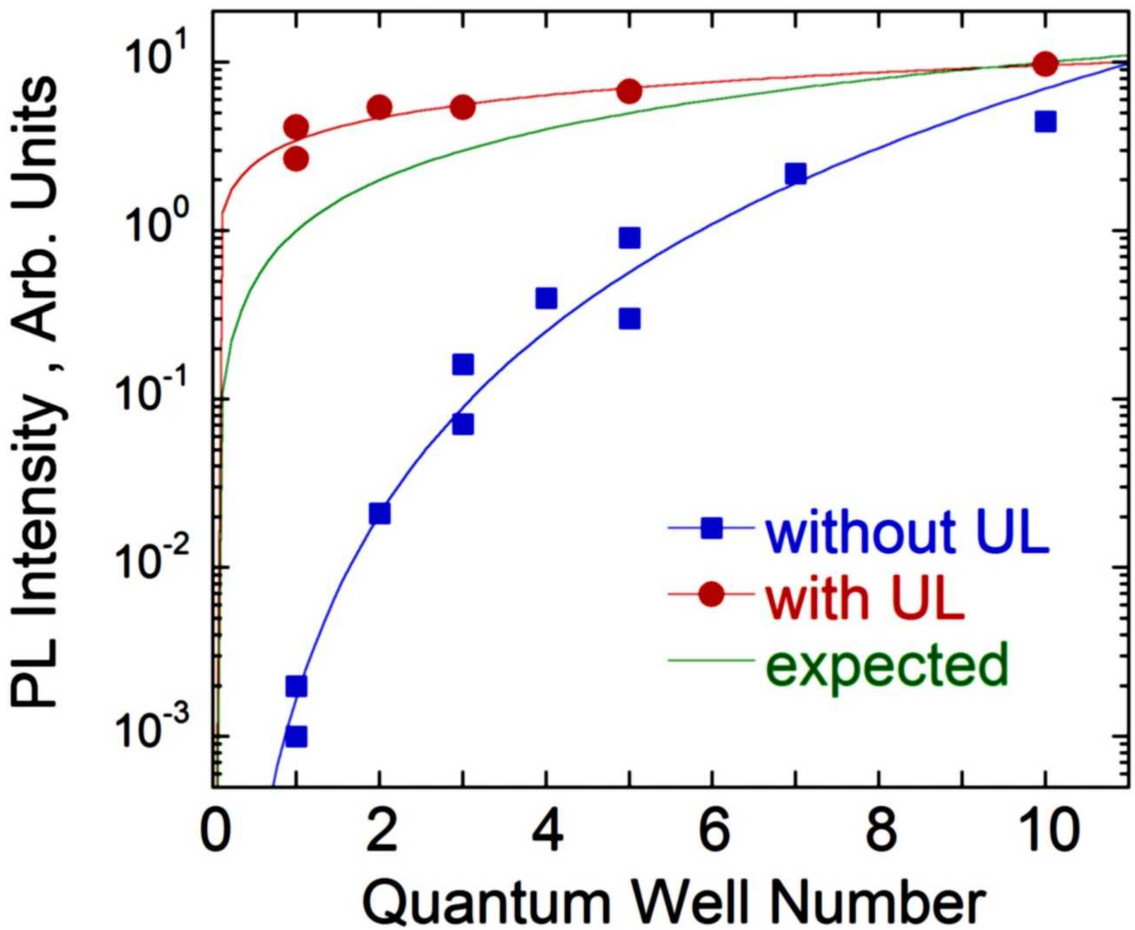


<u>Growth conditions</u> - InGaN underlayer (UL) at 850 ºC, ~ 180 nm thick, 3 - 4% indium, pressure 300 torr. Verified with XRD.

Single QWs on ULs can be as bright as 10 QWs.

Same QWs on dilute indium InGaN layer

Using 325 CW HeCd



Growth conditions - InGaN underlayer (UL) at 850 ºC, ~ 180 nm thick, 3 - 4% indium, pressure 300 torr. Verified with XRD.

Single QWs on ULs can be as bright as 10 QWs.

Expect

I_{PL} ~ number of QWs or $I_{PL} \sim n^1$

Instead without UL

$I_{PL} \sim n^{3.6}$

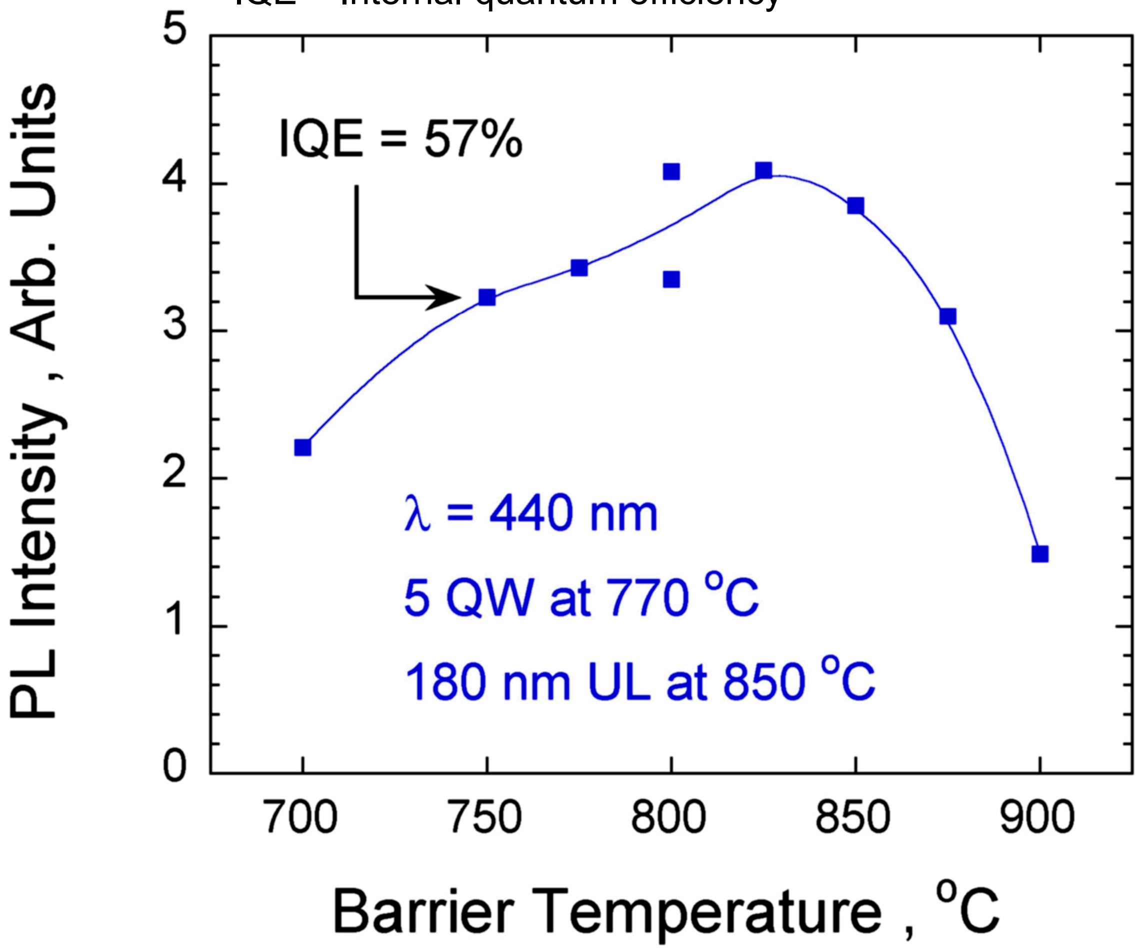
And with UL

$I_{PL} \sim n^{0.45}$

Without ULs – gradual I_{PL} improvement.
With ULs – immediate I_{PL} improvement.

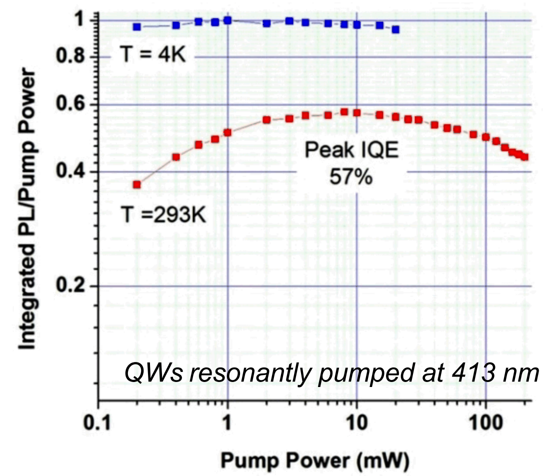
Influence of GaN barrier growth temperature

IQE = Internal quantum efficiency



Growth conditions – 5 MQW structures grown on InGaN UL at 770 ºC using different GaN barrier growth temperatures. Verified with XRD.

IQE = I_{PL} at T=300K/ I_{PL} at T = 4K

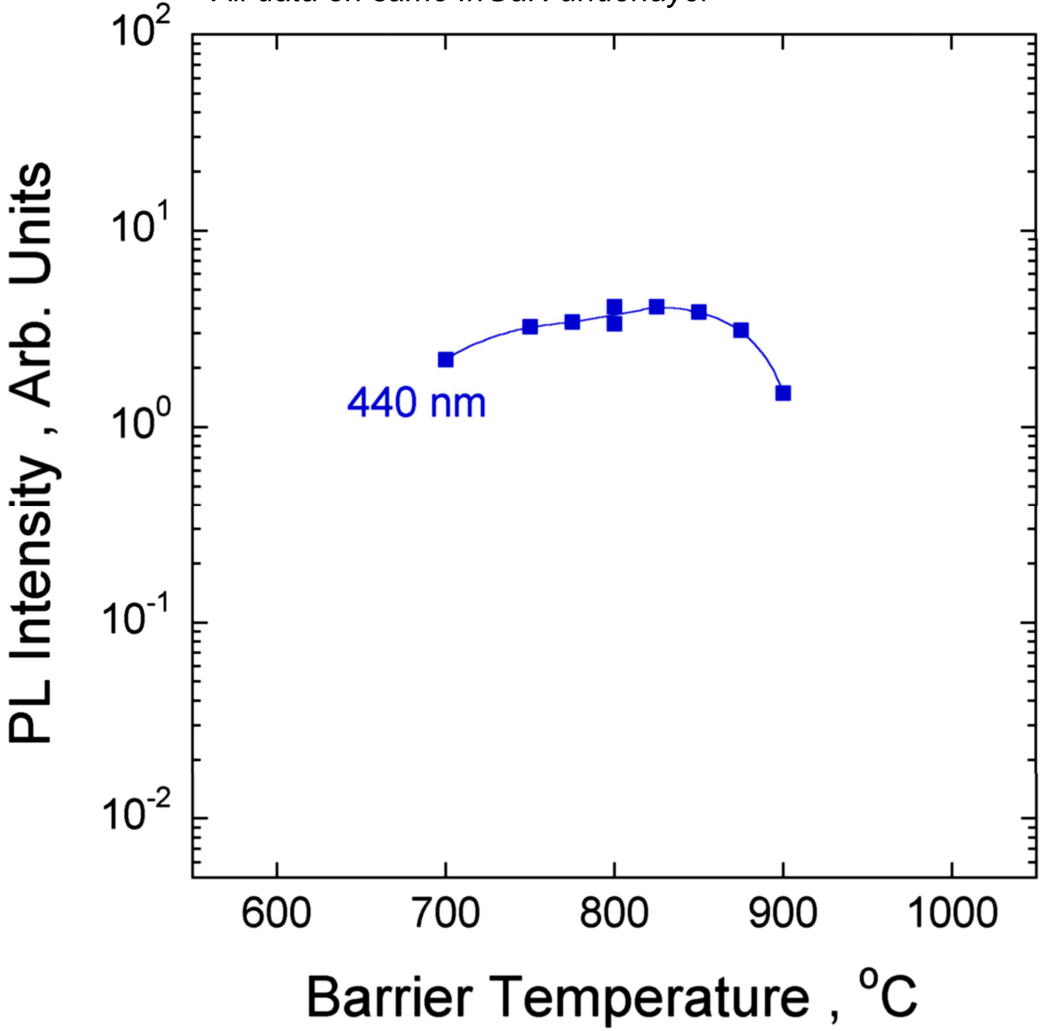


IQEs of ~70% at 440 nm

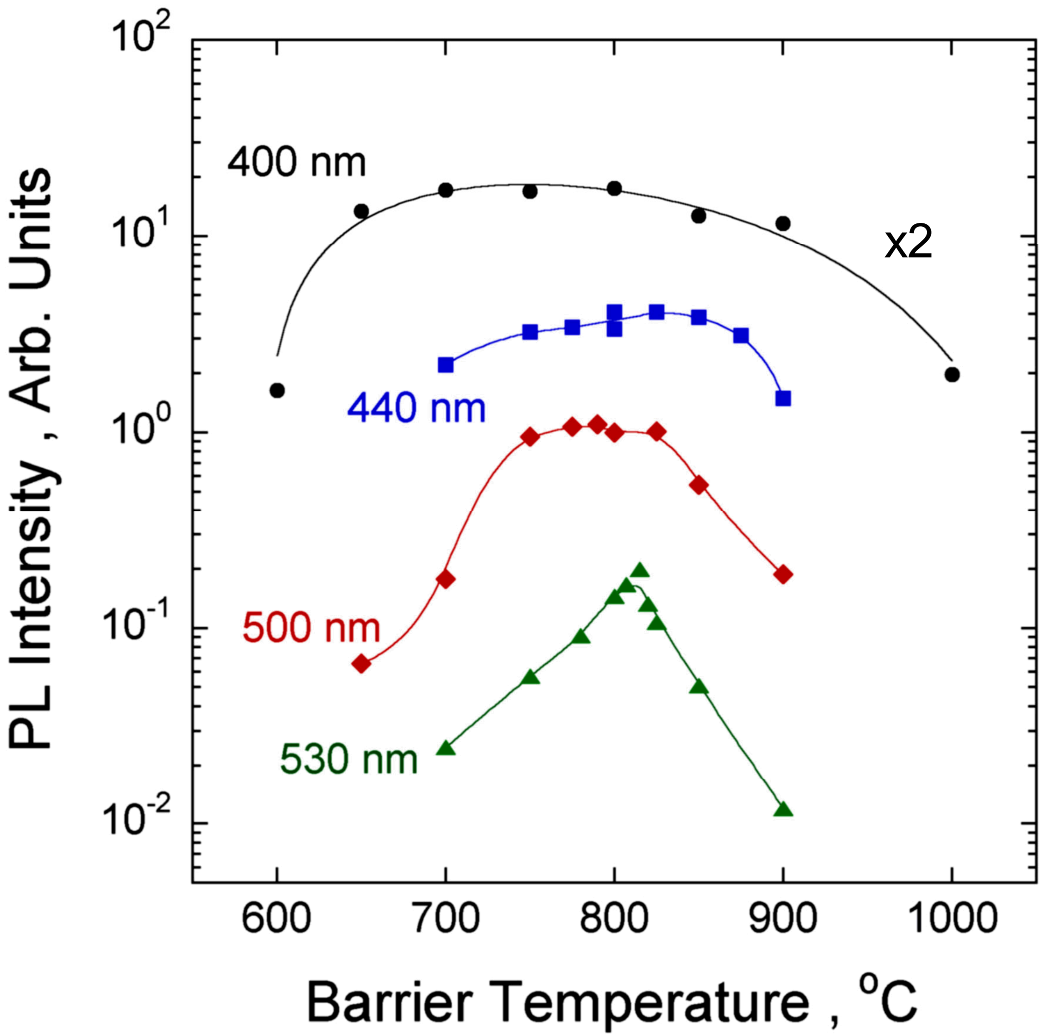
PL intensity changes by a factor of 2.5x and is fairly constant from 750 to 875 ºC.

GaN barrier temperature influence on PL intensity for different wavelengths

All data on same InGaN underlayer

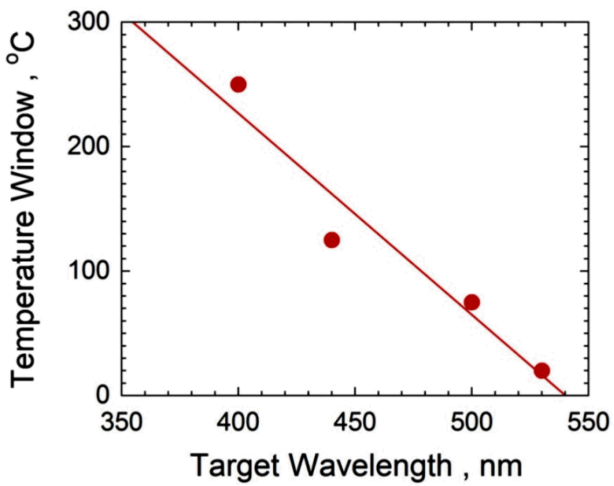


GaN barrier temperature influence on PL intensity for different wavelengths



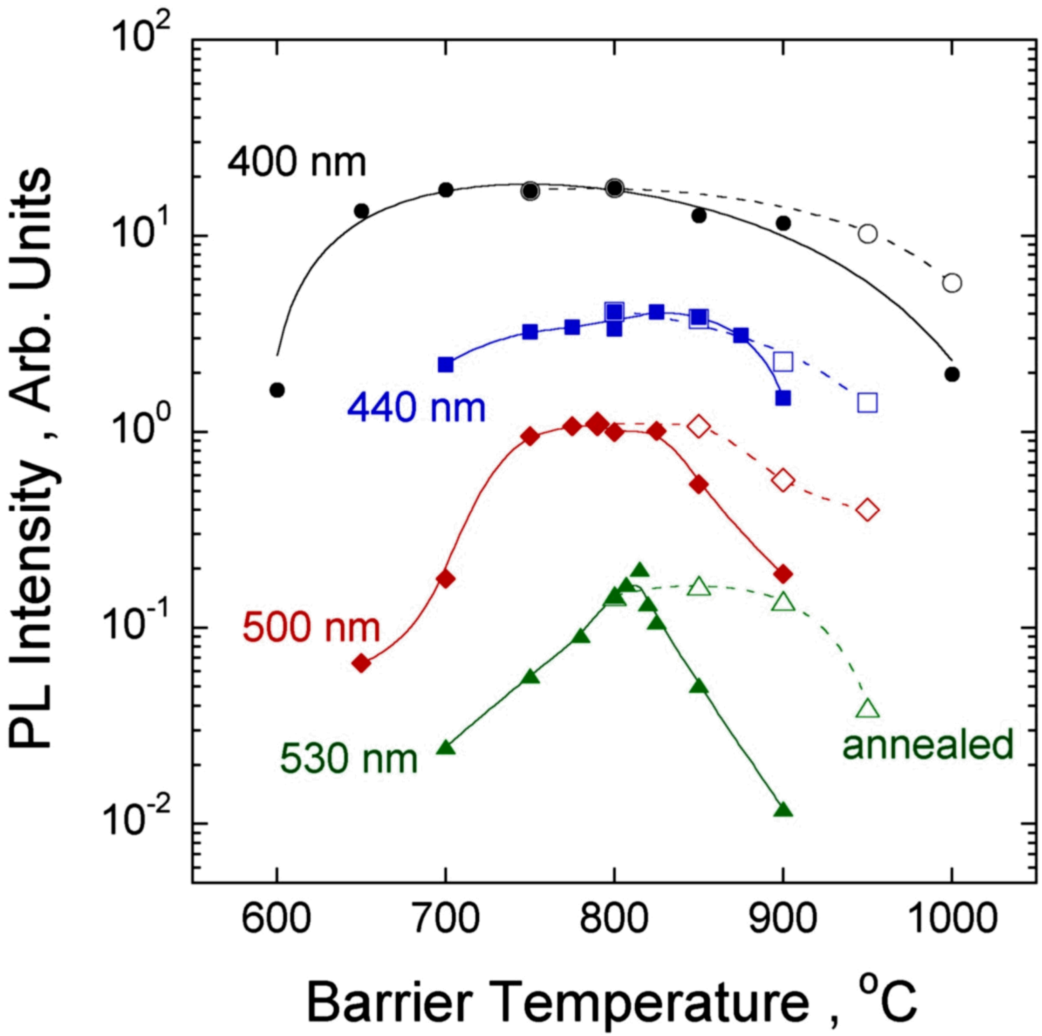
Growth conditions

QW growth temperature varied
T_{QW} = 790 ºC for 400 nm, 2 min.
T_{QW} = 770 ºC for 440 nm, 2 min.
T_{QW} = 750 ºC for 500 nm, 2.1 min.
T_{QW} = 740 ºC for 530 nm, 2.2 min.



Larger growth temperature window at shorter wavelengths – barrier conditions not as critical at these wavelengths?

Testing if high temperature GaN barriers degrade QW emission intensity



<u>InGaN decomposes as the annealing temperature increases</u>

(Thaler et al. JCG 311, 2933 (2010))

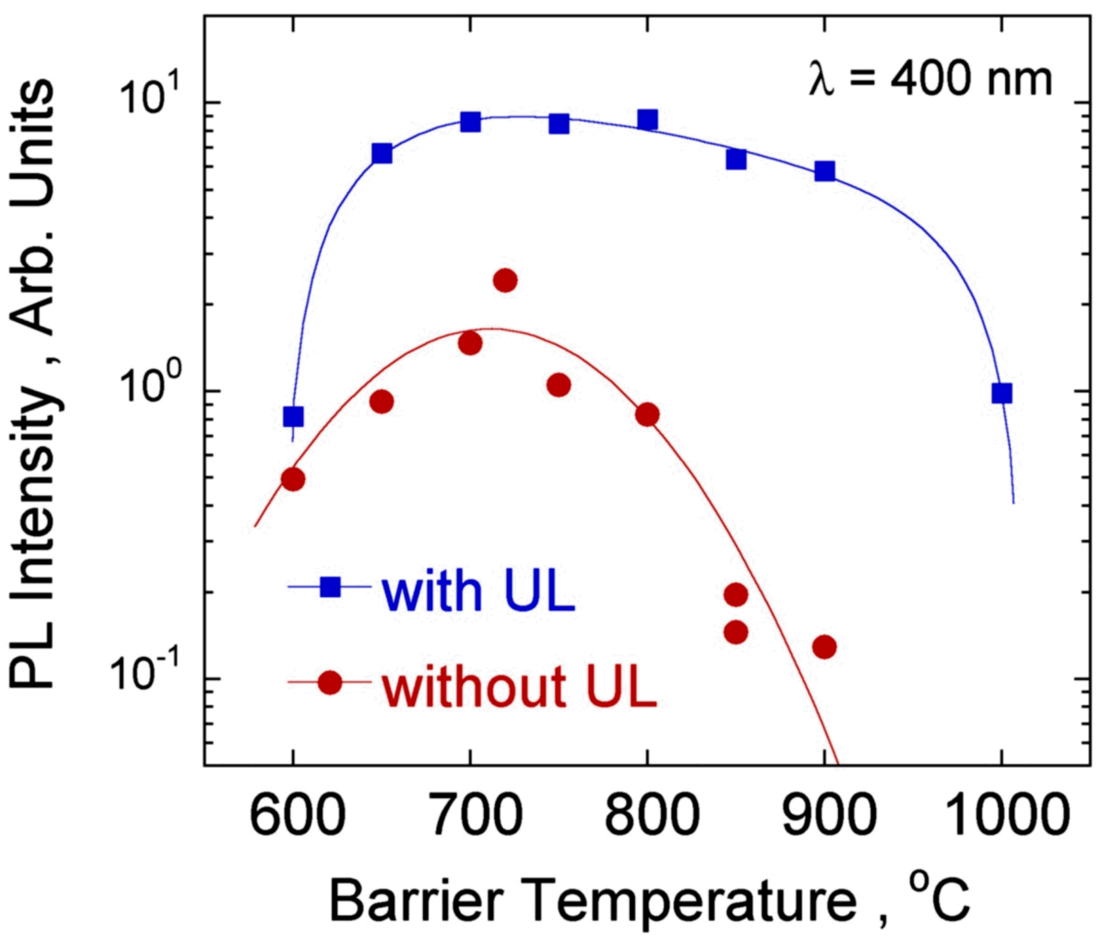
For bulk InGaN films, the PL intensity decreased prior to any measurable decrease in XRD peak intensities as the annealing temperature increased. Suggests NRC are formed during annealing.

Best QW wafers were quartered and annealed for 30 additional minutes at temperatures higher than the initial GaN barrier growth conditions.

Observe some degradation in the PL intensity for QWs annealed T ≤ 900 ºC but does not account for entire decrease in PL intensity.

Surprisingly observe least degradation for 530 nm QWs for T ≤ 900 ºC.

Lower temperature barrier growth improves QW intensity



Growth conditions – 5 period MQWs at 790 ºC with and without InGaN underlayer (UL), only GaN barrier growth temperature varied.

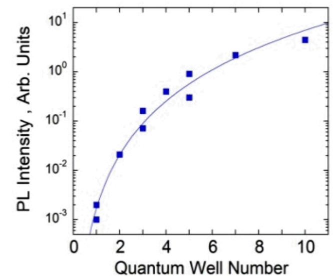
With UL – GaN barrier temperature range is 650 to 900 ºC

Without UL – Best GaN barrier temperature is ~ 700 ºC.

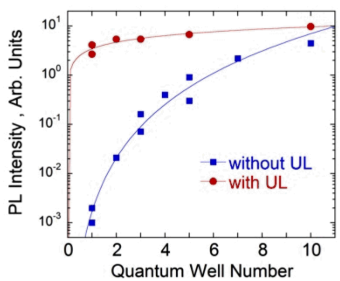
Suggests – Without ULs, lower temperature GaN barriers somehow improve InGaN QW intensity.

However – **Seems Inconsistent!** Lower GaN barrier temperatures should have more NRCs in the GaN (ex. carbon). How could increasing defects improve the PL intensity?

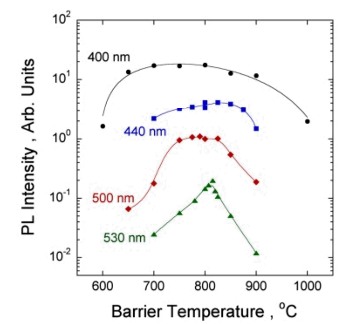
What correlates with improvement in QW intensity?



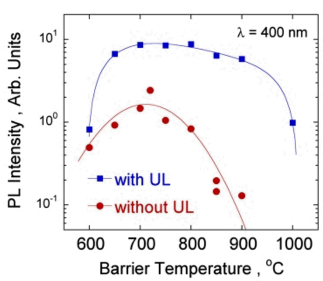
- Use same QW growth conditions with/without UL.
  - QWs and barriers are formed in a “growth loop” – n-times.
  - Suggests that QWs improve during growth – naturally.



- Underlayers immediately improve PL intensity of QWs.
  - Doesn’t take as many QW periods to improve quality?



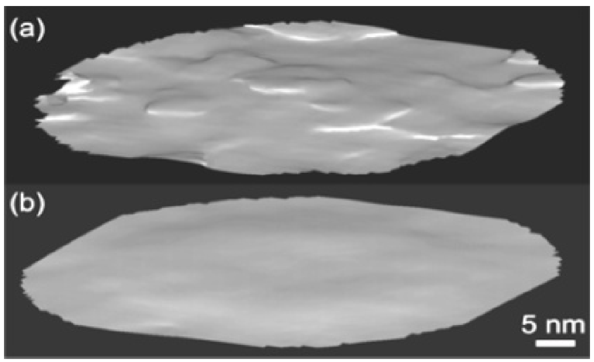
- For thicker InGaN, V-defects form – NRCs screened.
  - Observe more V-defects for QWs at 530 nm vs. 440 nm; suggests V-defects not playing a major role.



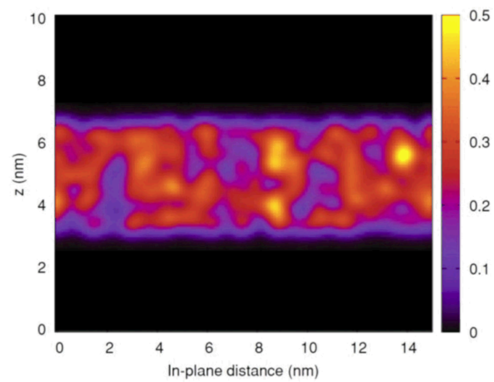
- Lower temperature GaN barrier growth without ULs.
  - Suggests that material surrounding InGaN QW is important.

InGaN/GaN morphology likely influences localization

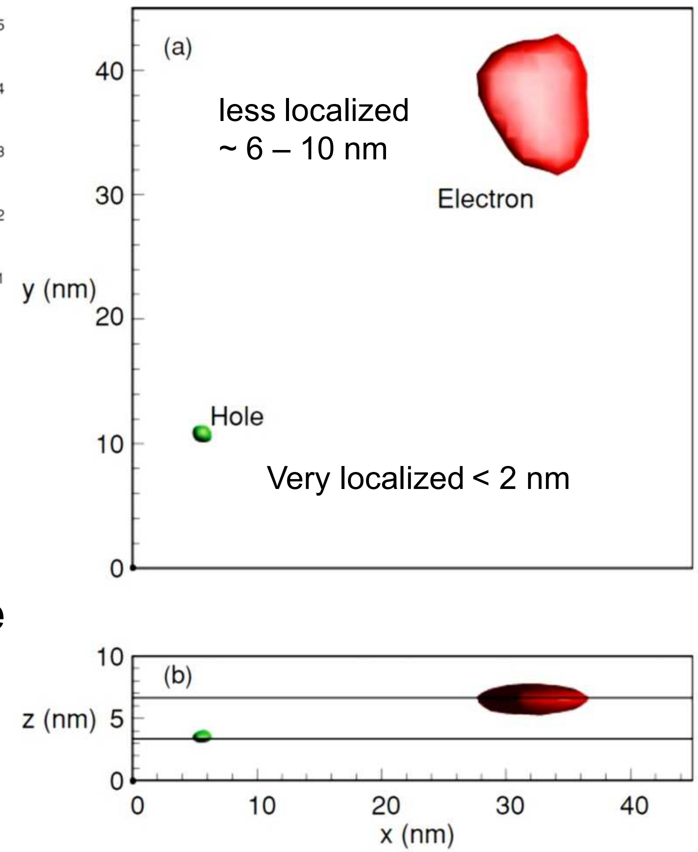
Watson-Parris (Manchester), et al., Phys. Rev B ***83, 115321 (2011)***



An isoconcentration surface based on atom probe tomography data illustrating the lower and upper interface of an InGaN QW.



Indium fraction in a 3 nm thick QW with an average indium concentration of 25%.



The calculated ground state probability density of an electron (red) and a hole (green).

Are random alloy fluctuations sufficient to localize carriers?

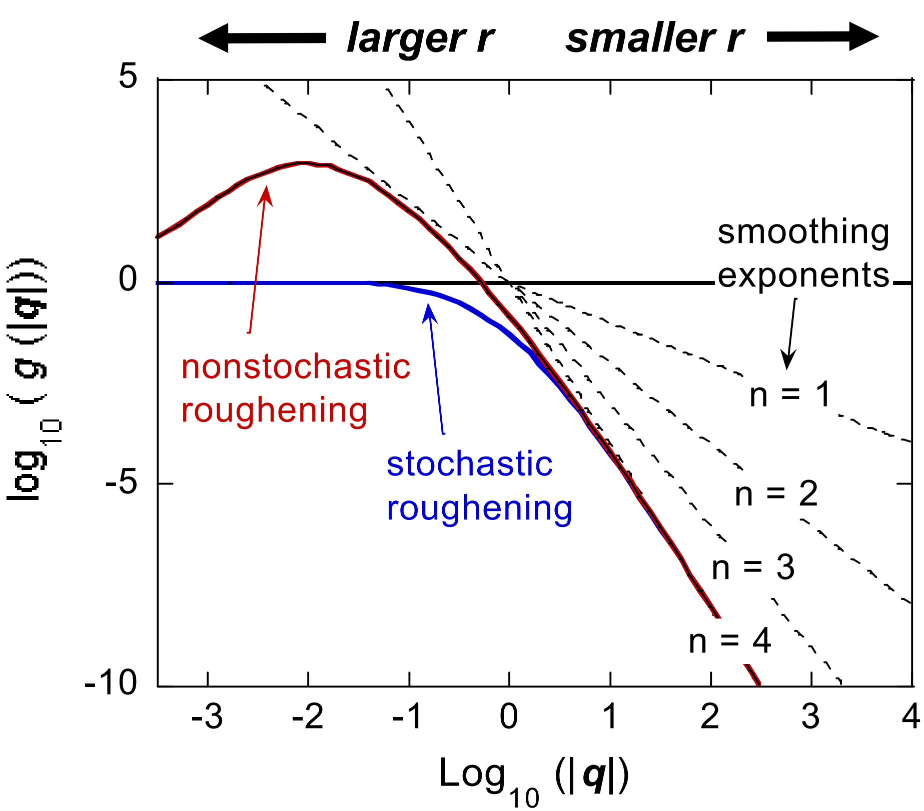
Holes strongly localized in regions of higher indium content near bottom interface.

Electrons are localized by mono-layer well width fluctuations near top interface.

Power Spectral Density (PSD)

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

From h(*x*,*y*) of the AFM image, calculate the height-height correlation function, *g(**q**)*, as a function of ***q***, *where **q** = 1/r*. AFM Software package

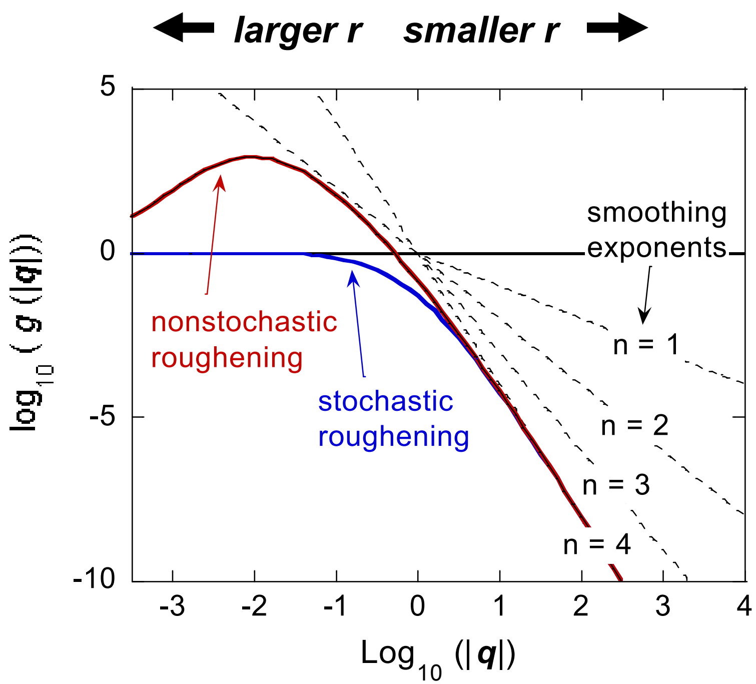


RMS roughness is calculated from *g*(***q***)

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

Put simply – PSD is a measure of surface roughness vs. length

Using PSD to determine smoothing mechanisms at large q



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

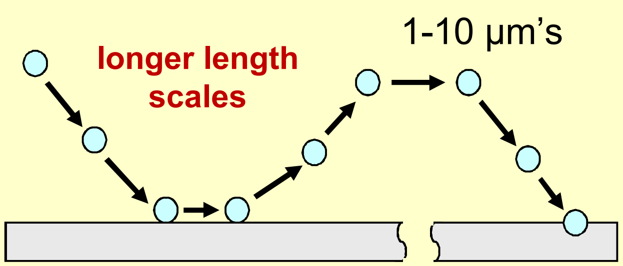
At large ***q***

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

n = 2

evaporation and recondensation

(GaN for T > 900 °C)

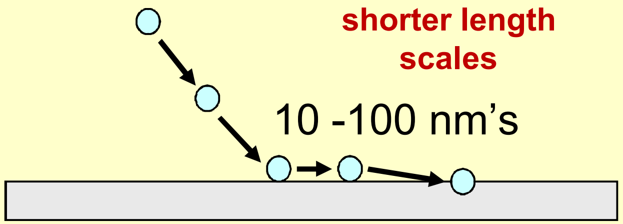


See Mitchell et al., JCG 222, 144 (2001).

n = 4

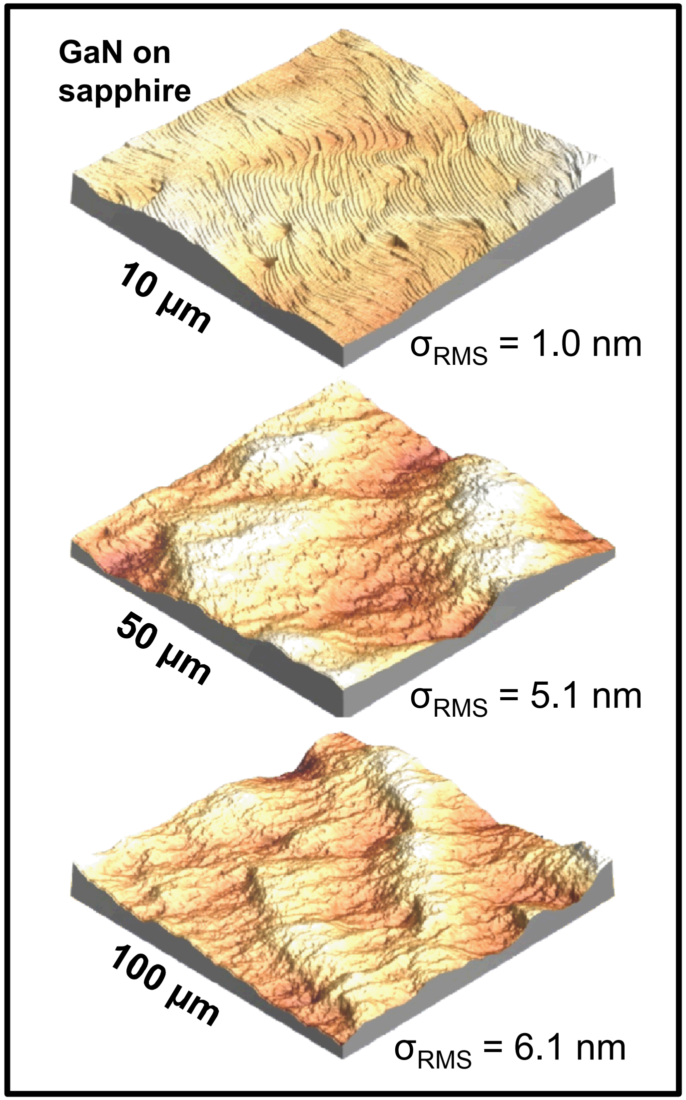
surface diffusion

(InGaN and GaN T < 900 °C)

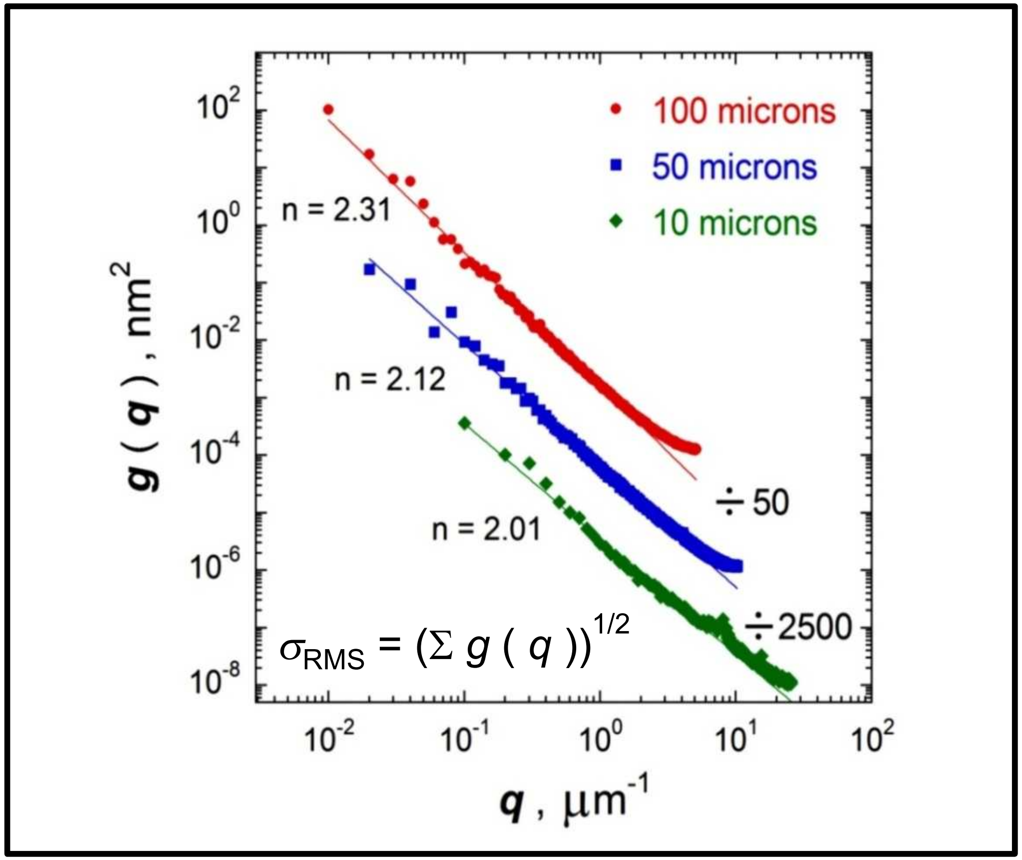


See Koleske et al., JAP 84, 1998 (1998).

PSD analysis of GaN films on sapphire



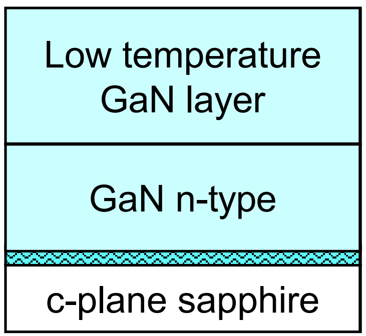
σ_{RMS} depends on scan size

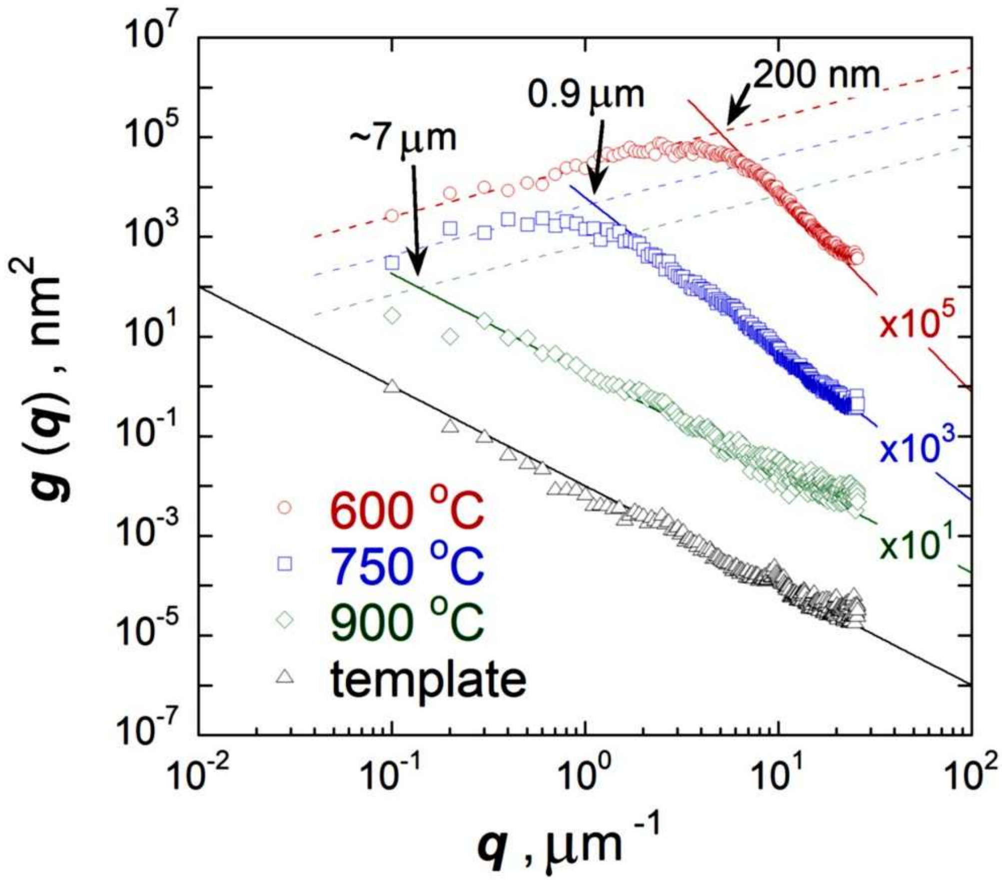


***n* ~ 2 - implies the smoothing mechanism is evaporation and recondensation of Ga atoms**

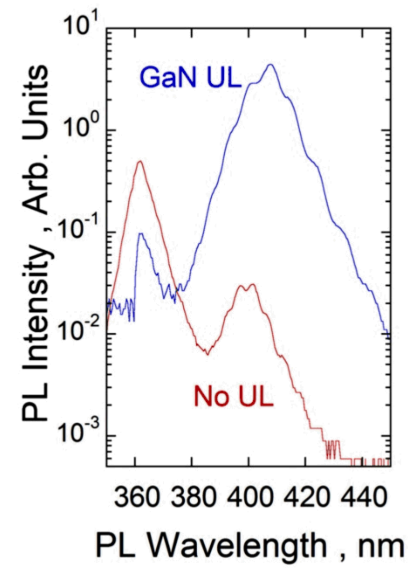
PSD of lower temperature GaN growth

Growth of 200 nm GaN on high temperature GaN templates



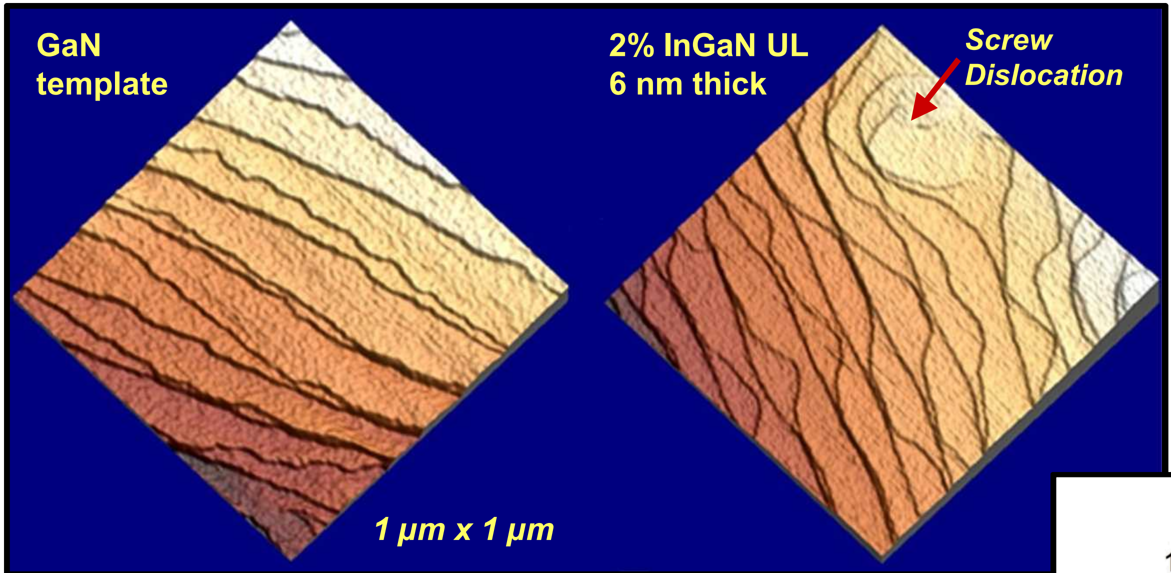


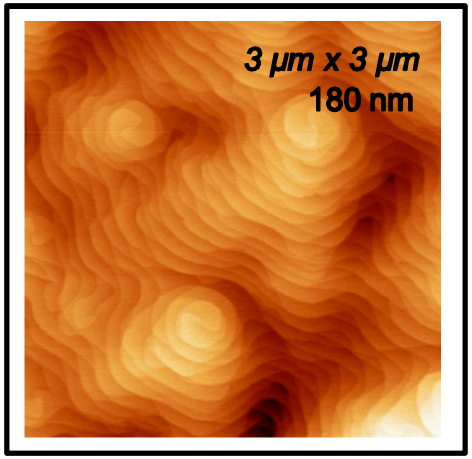
Have used 790 ºC GaN as an underlayer to improve 400 nm 3 period MQWs

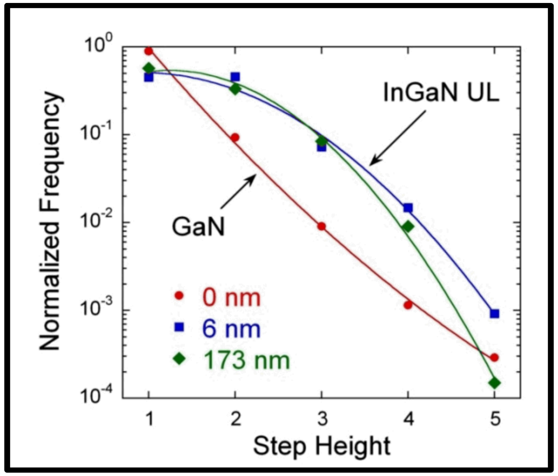


As the GaN temperature is reduced, get increased roughness ~ 0.05 to 1 µm.

PSD analysis of high temperature InGaN underlayer growth



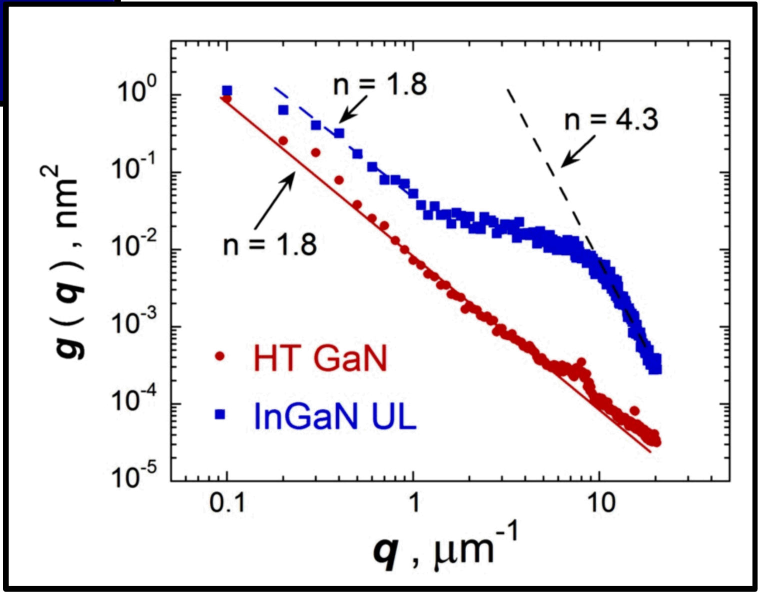




InGaN grown at 870 ºC and has few V-defects

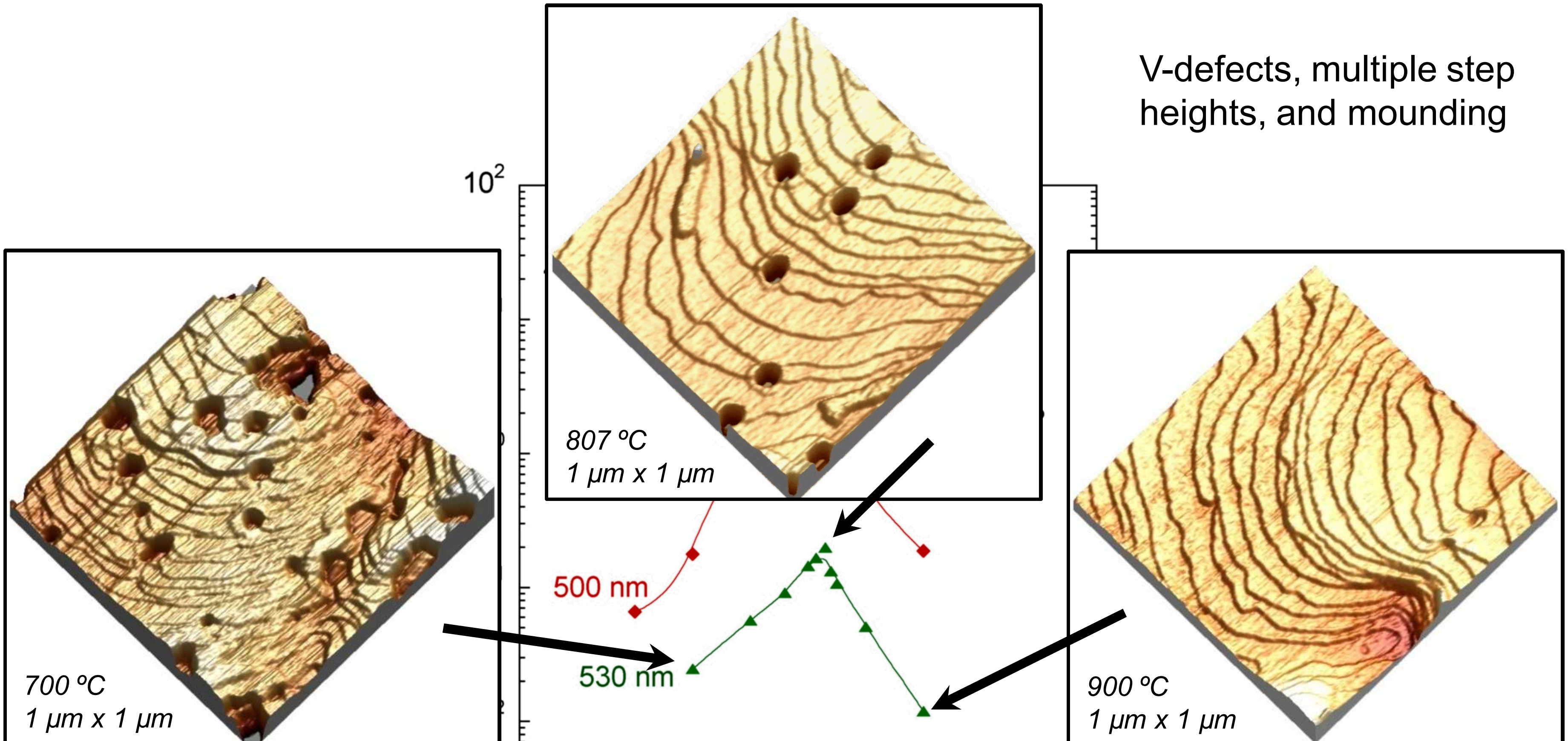
Increase in multiple step heights after only 6 nm of growth.

Increase in roughness from 0.05 to 1 µm with the InGaN UL



AFMs of 530 nm QWs with different barrier temperatures

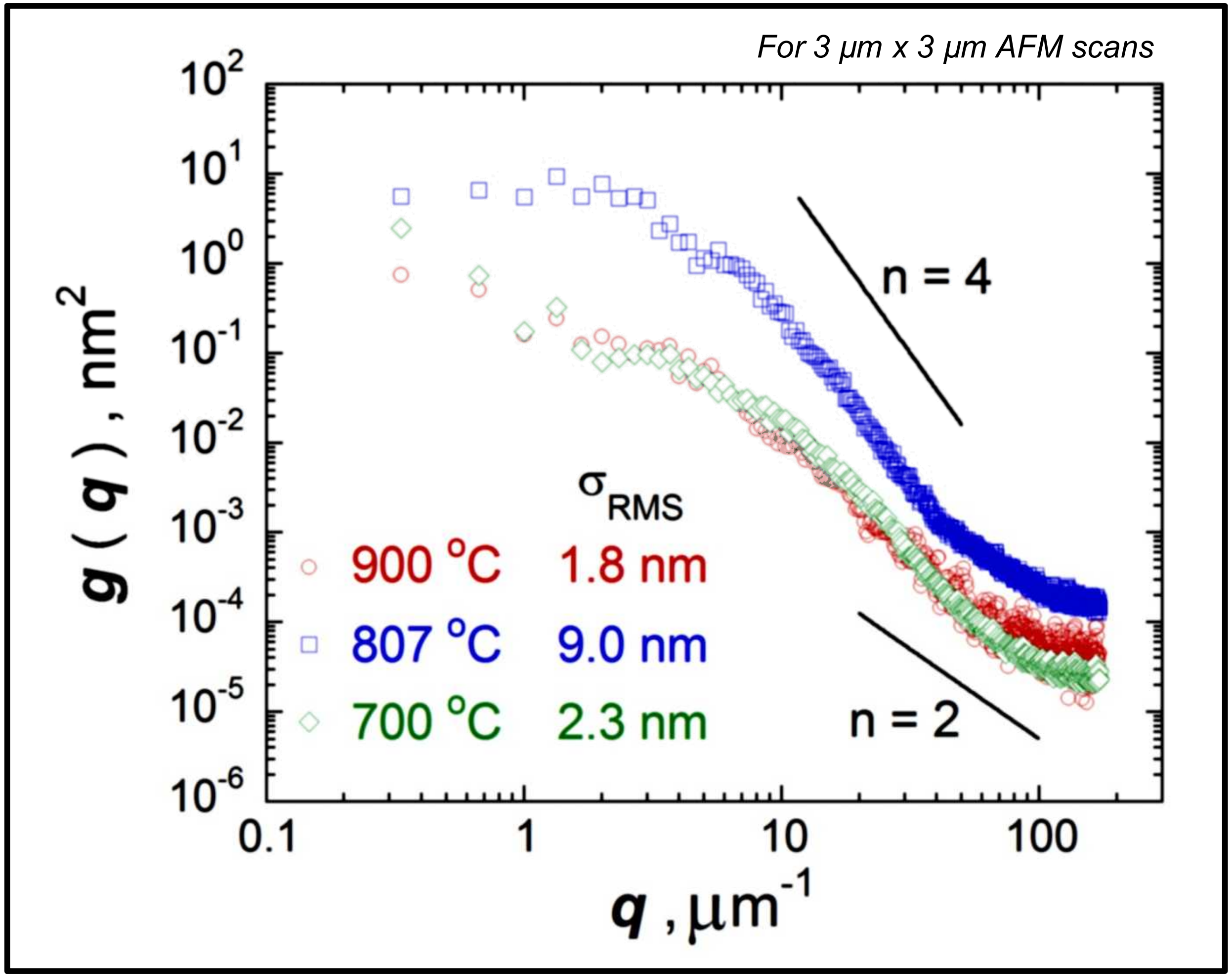
V-defects, multiple step heights, and mounding



V-defects, multiple step heights, and mounding

fewer V-defects and smoother

PSDs confirm surface roughening for 530 nm QWs



Similar to GaN and InGaN ULs and MQW structures, observe increase in roughness from 0.05 to 1 μm

Barrier Temp.	PL intensity	σ_{RMS}
900 ºC	0.010	1.8
807 ºC	0.200	9.0
700 ºC	0.025	2.3

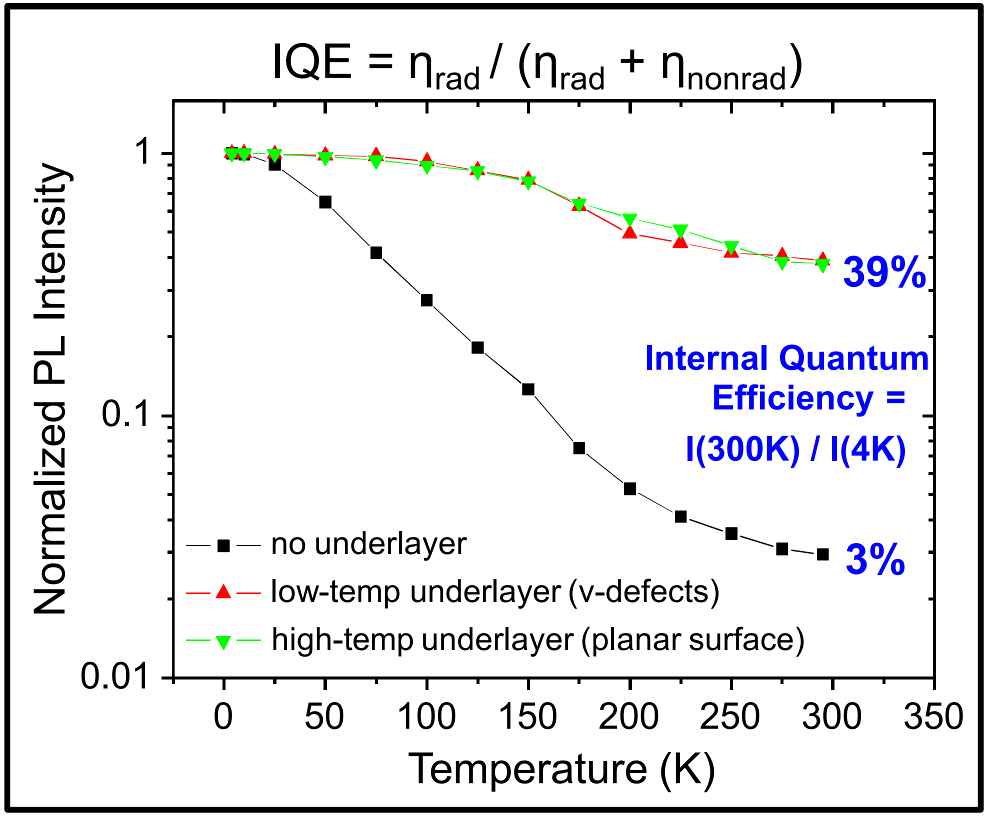
Apparent correlation between the PL intensity and the increased observed in the PSD and by σ_{RMS}.

Same IQE with and without V-defects

Temperature dependent PL studies by Mary Crawford

ULs grown at low temperature, 790 ºC have many V-defects.

ULs grown at high temperature, 880 ºC have few V-defects.



5 period blue QWs

Suggests that V-defects are not correlated to NRCs concentration!

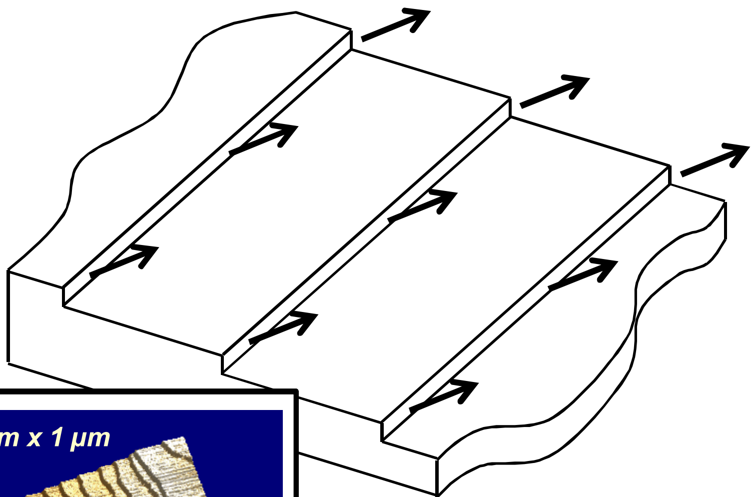
Relationship between morphology, PL intensity, and NRCs

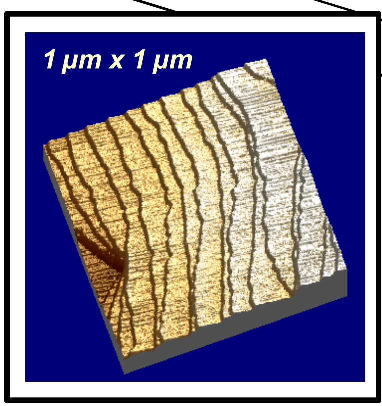
- Summary of Data
  - Observe increase in roughness ~ 0.05 - 1 μm.
  - Increased roughness observed for GaN and InGaN ULs, and MQWs.
  - Faster increase in roughness as the indium concentration increases.
  - Increased roughness driven by surface diffusion.
  - Steps structure changes
    - single to multiple layer step heights – increased step curvature.

- Possible mechanisms that may decrease NRCs
  - Layers "getter" point defects and impurities – depends on thickness.
  - Atomistic scale mechanism (surface reconstruction, indium bilayers).
  - Elastic relaxation at step edges – morphology influences NRCs.
  - Others?

Elastic relaxation at step edges

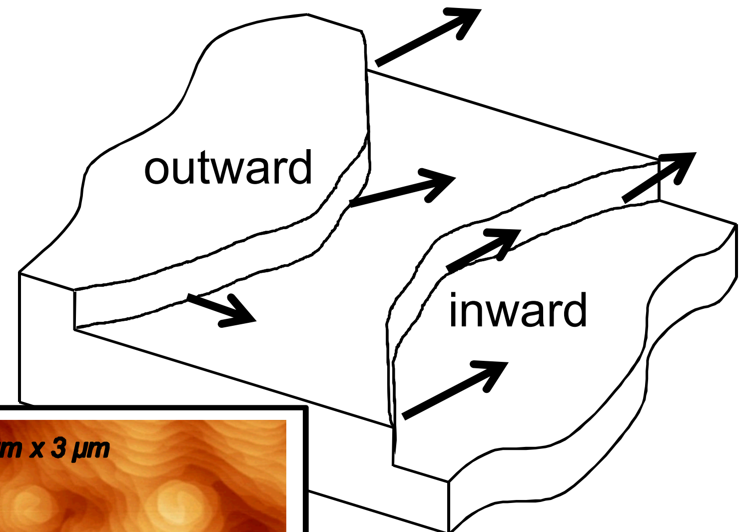
GaN substrate – straight edges with single step heights.

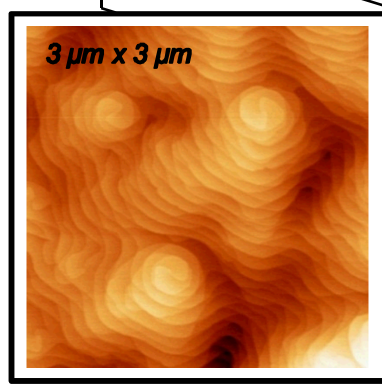




Steps curved only slightly

InGaN or GaN UL – curved step edges with multiple step heights.



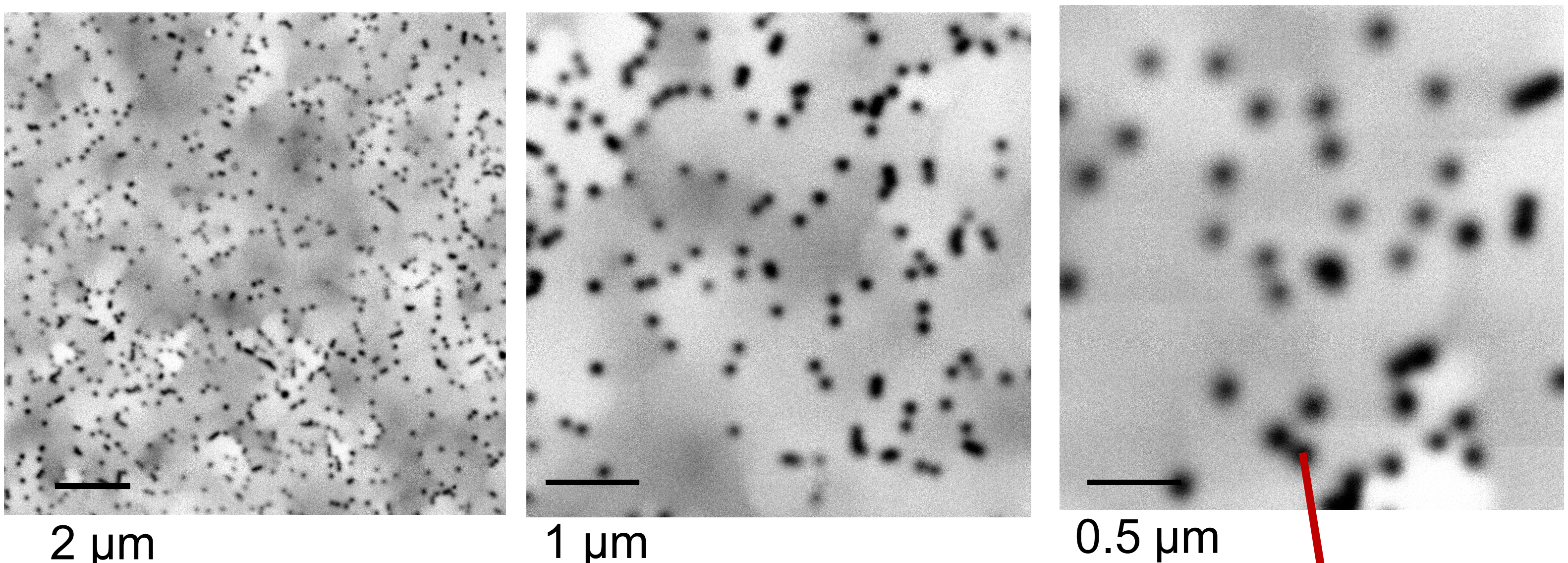


Steps curved outward contain more elastic relaxation than steps curved inward.

Observe larger density of outward curved steps
Increased elastic relaxation compared to GaN starting template

Spatial luminescence variations observed in RT cathodoluminescence (CL) data

Images normalized to maximum intensity - 450 nm, 5 period MQWs

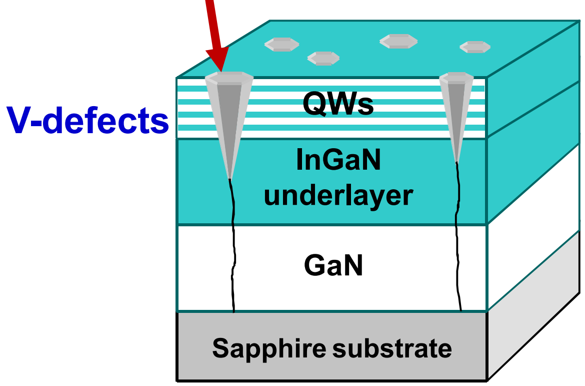


Observe strong spatial variations in CL intensity.

For samples with lower integrated CL intensity, spatial variations still observed.

CL emission variation not correlated to V-defects.

Variation in CL intensity might correlate to surface morphology, however SEM images appear flat.

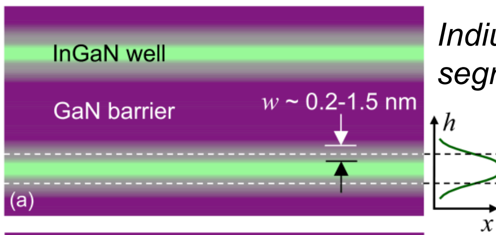


Summary and Implications

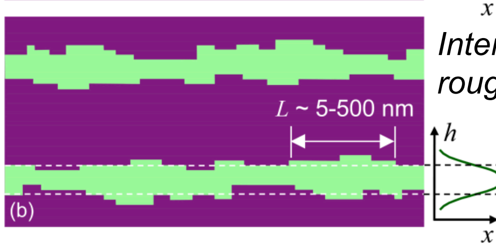
- Believe that morphology plays a role in MQW efficiency
  - Common increase in roughness at the 0.05 to 1 μm.
  - Surrounding material "more" important than QW – lower barrier temperatures, use of ULs, MQW emission gradually improves.
- Step edge elastic relaxation possibly decreases NRCs.
  - Consistent with DLOS studies – 1^{st} QW has more NRCs than 5^{th} QW.
  - Variation in CL emission intensity similar in length scale to developed morphology.
- Elastic relaxation might be sufficient to improve blue QWs, however more is needed for green QWs.
  - Increased need to explore other GaN orientations?, strain relaxed InGaN substrates?

Multiple quantum well InGaN/GaN morphology

Compositional grading along the surface normal direction

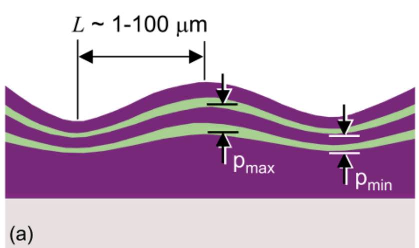


Indium segregation



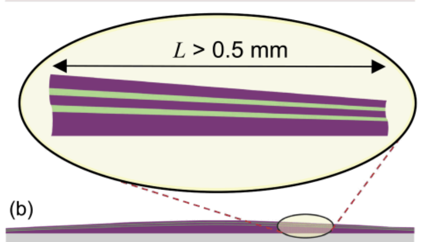
Interface roughening

lateral thickness variation



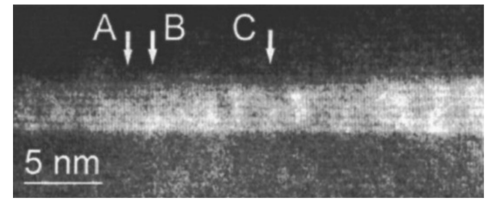
Island formation

Strain-induced surface instability

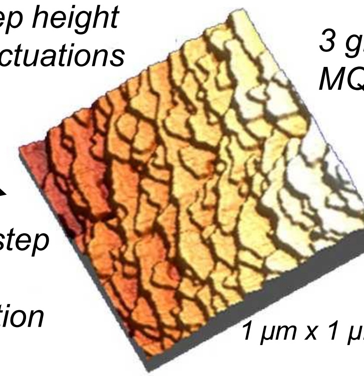


Across wafer non-uniformity

Graham et al., JAP 97, 103508 (2005).

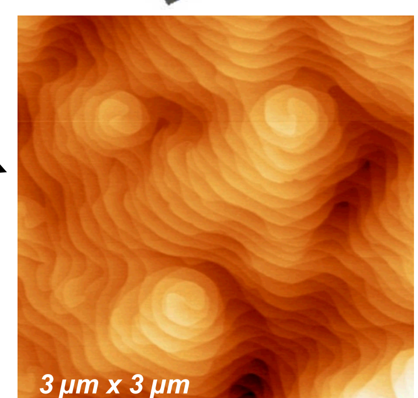


step height fluctuations



3 green MQWs

Multi-step height formation



WSxM software from www.nanotec.es