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Understanding GaN Nucleation Layer Evolution on Sapphire and its Impact on GaN Dislocation Density

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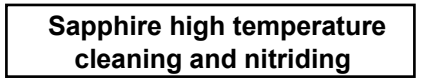
“Two step” heteroepitaxy of GaN on sapphire

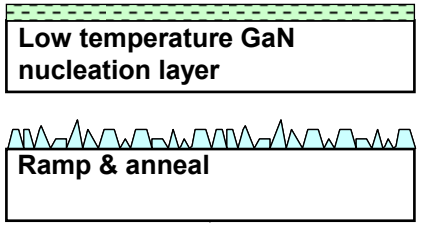
Based on work from K. Hiramatsu, et al., Journal of Crystal Growth 115, 628 (1991).

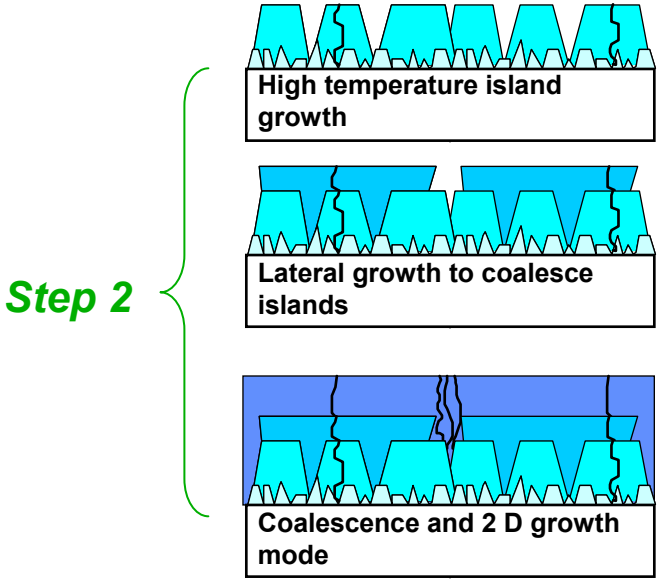
1) Sapphire heated to high temperature (1050 – 1100 ºC) in H_2 to “clean” the surface.



2) Grow 20 – 30 Å GaN NL at 500 – 600 ºC (NL contains “cubic” material with stacking faults).

3) Heat GaN NL to 1000 – 1080 ºC in flowing H_2, N_2, and NH_3 (Wurtzite nuclei form).





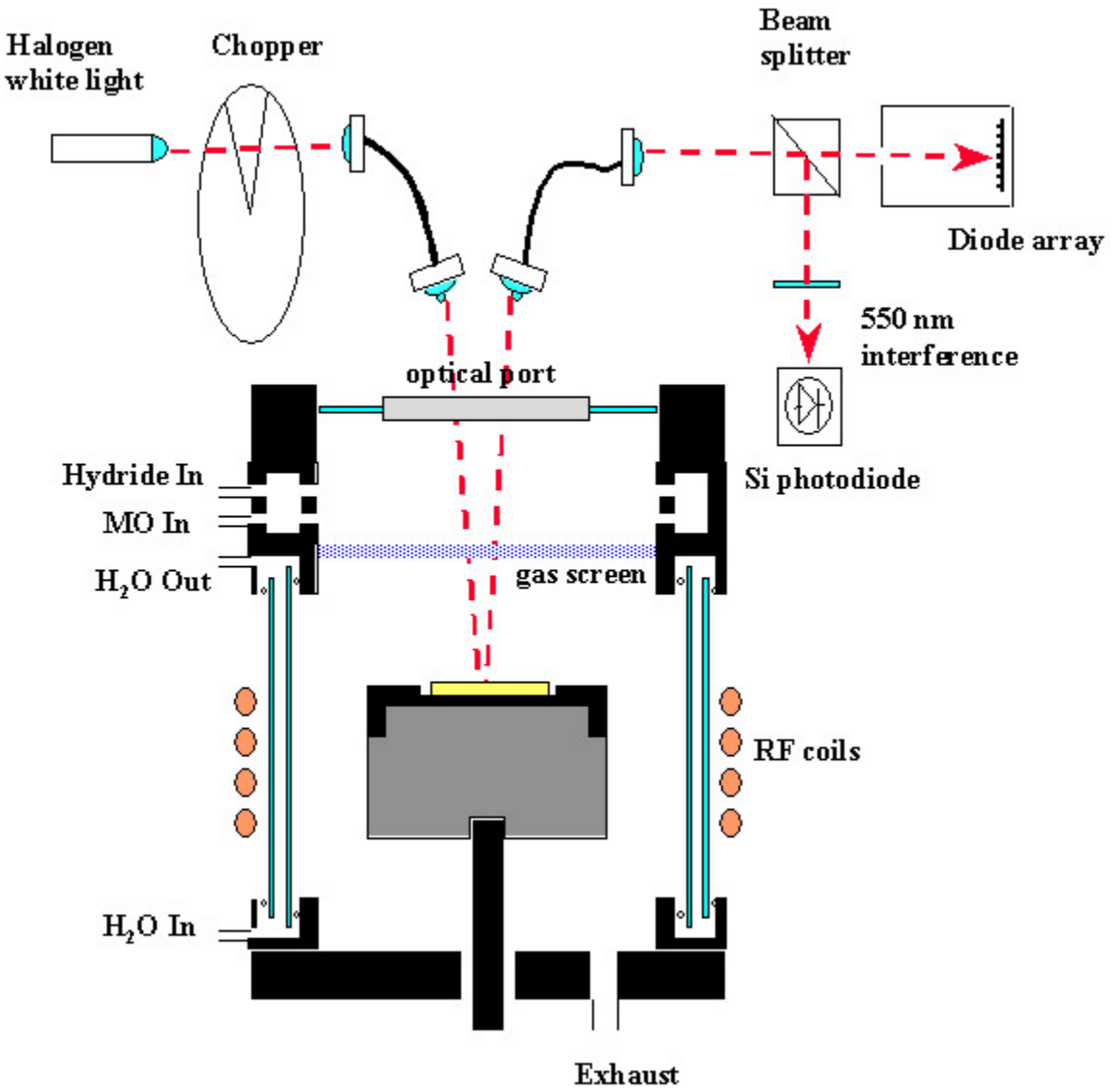
4) Ga source is turned on (high T). Isolated GaN grains grow on the GaN nuclei. Dislocations propagate up from some GaN nuclei.

5) Laterally / vertical growth; grain coalescence begins.

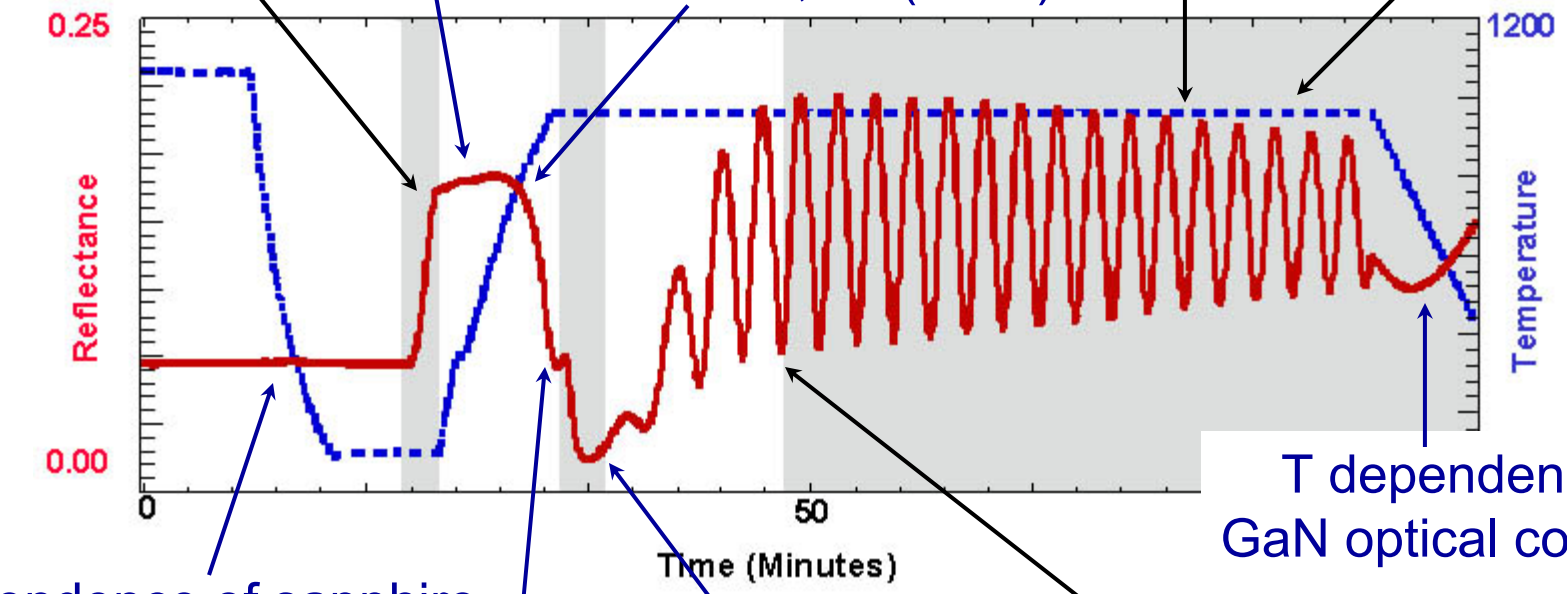
6) Dislocations form as GaN grains coalesce to accommodate grain twist / tilt.

How many dislocations evolve from grains (4) vs. nuclei coalescence (6)?

In-situ optical reflectance to monitor film thickness and morphology



What can we learn from optical reflectance?



T dependence of NL GaN optical constants

NL thickness

NL decomposition rate APL 82, 1170 (2003) and JCG 279, 37 (2005).

bulk growth rate

bulk layer roughening and uniformity

T dependence of GaN optical constants

T dependence of sapphire optical constants

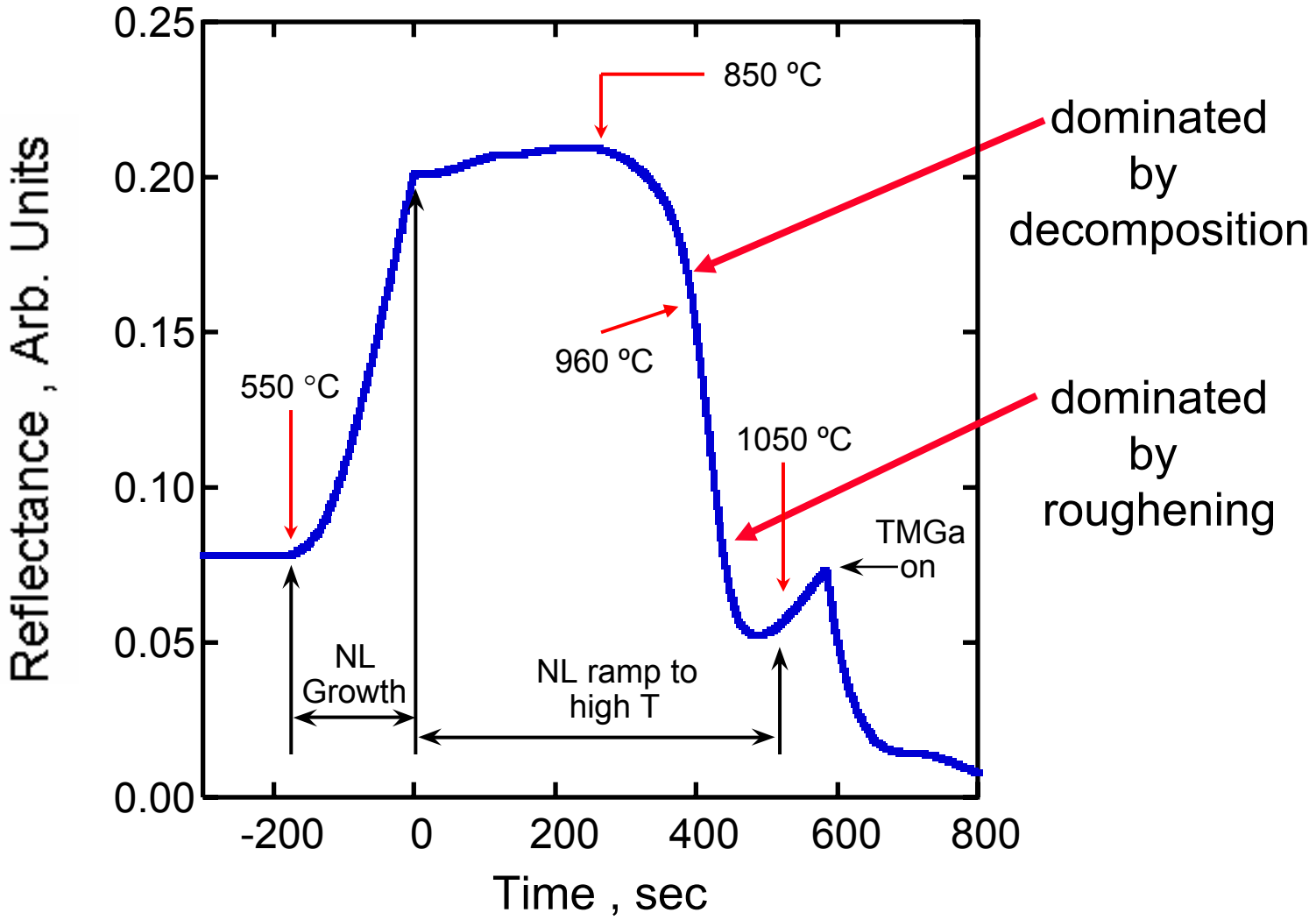
NL roughness and nuclei size

3D island roughening and size

3D to 2D recovery time APL 81, 1940 (2002)

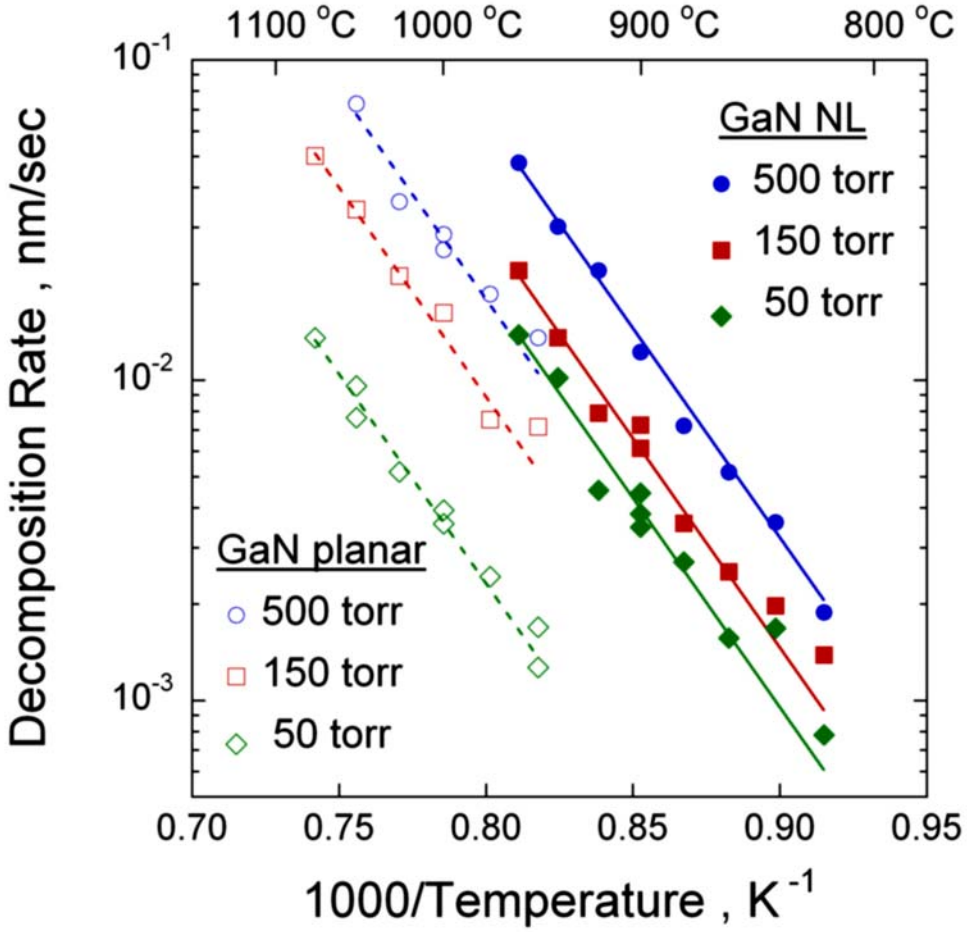
Blue = temperature
Red = reflectance

Analyzing nucleation layer reflectance waveforms



Reflectance measurement of GaN NL and bulk GaN decomposition rates

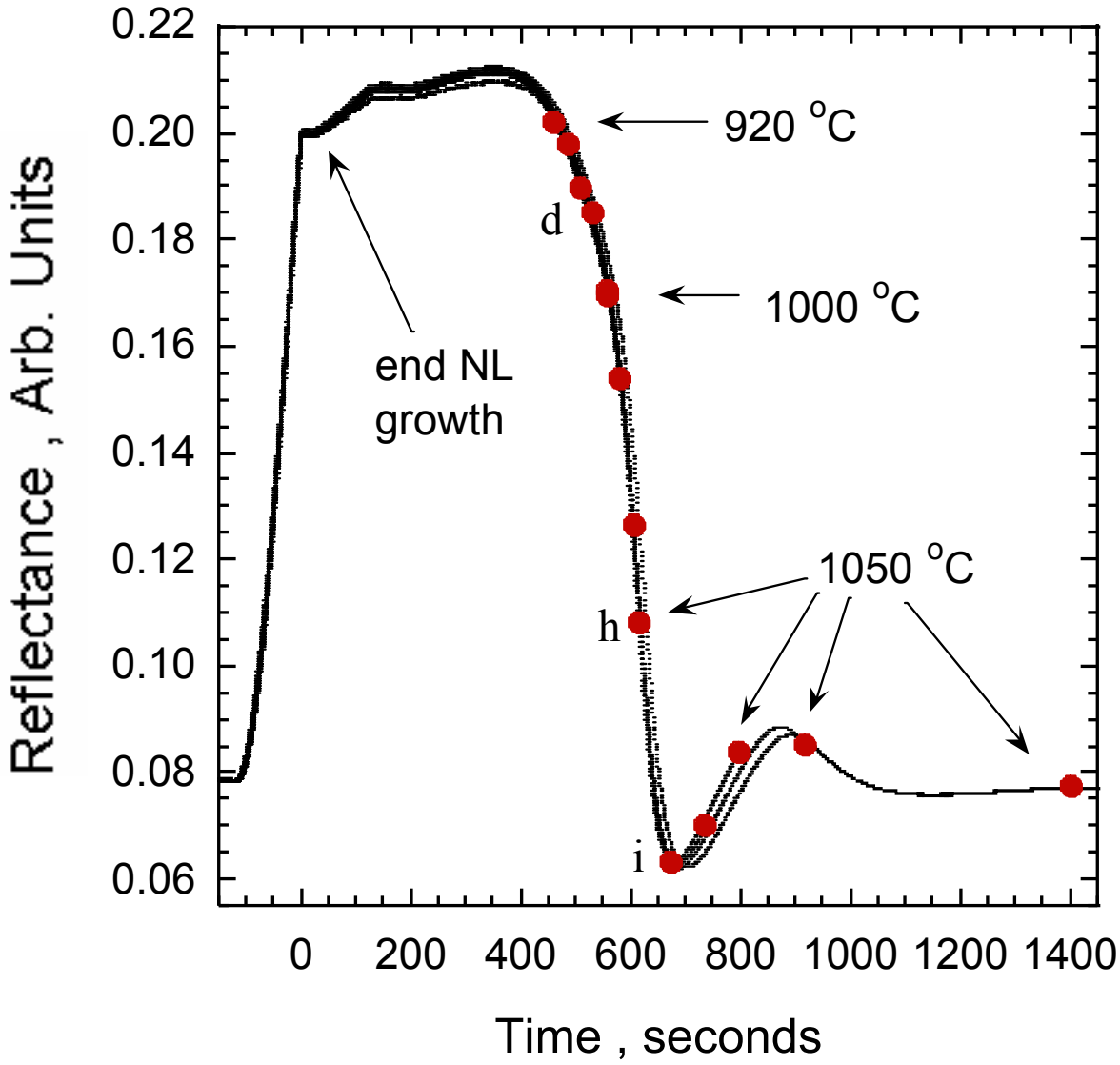
D. D. Koleske, et al., Appl. Phys. Lett. 82, 1170 (2003).



$E_A = 2.7 \pm 0.1$ eV → Ga atom desorption

A_0 4 to 9 times larger for NL compared to bulk

Evolution of the reflectance waveform as the GaN NL is annealed



13 NLs (30 nm thick) were grown and annealed for different times (red circles). Annealed NLs were quenched in N_2 and NH_3 to freeze the morphology.

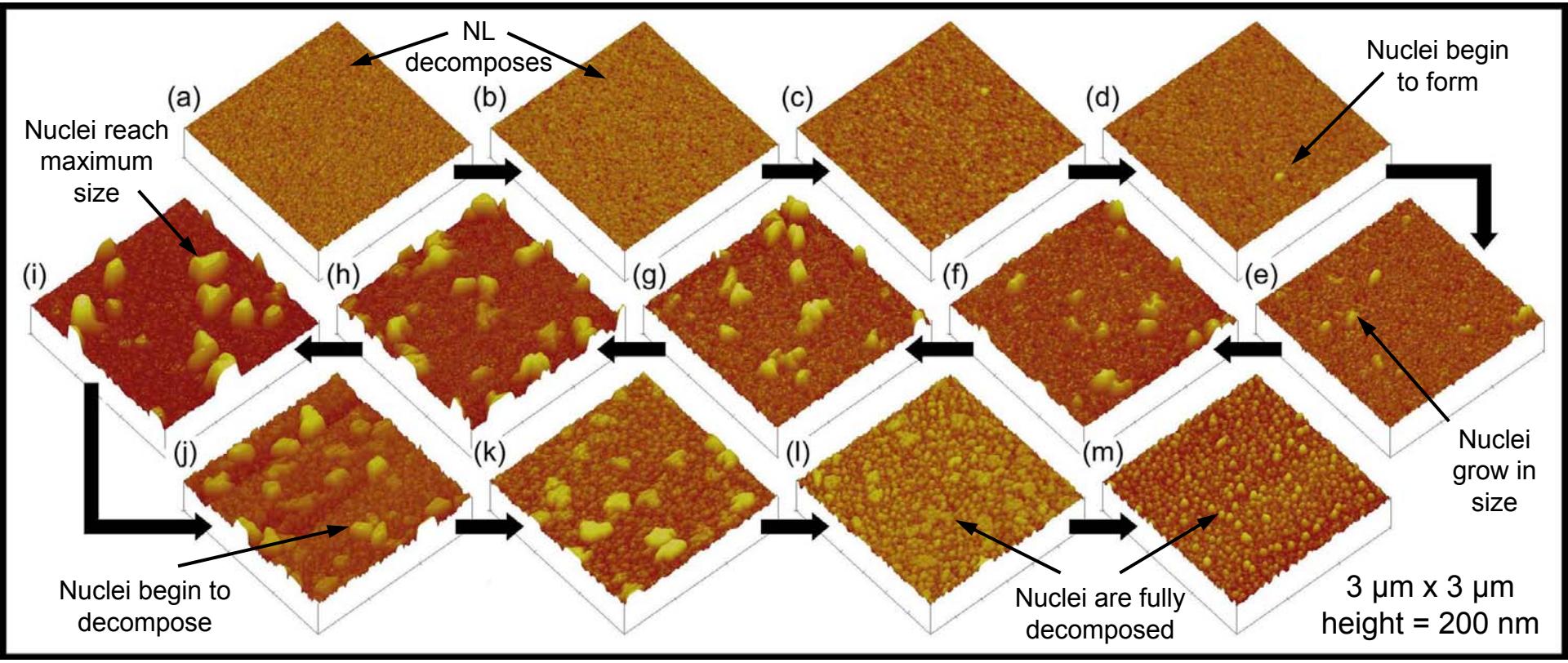
The overlap in waveform shapes indicates reproducibility of the NL evolution.

AFMs of annealed NLs shown next.

Details in Koleske et al., J. Crystal Growth 272, 227 (2004).

Mechanism for GaN nuclei formation as the nucleation layer is annealed

Arrows indicate increasing annealing temperature + time in H_2, N_2, and NH_3.



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

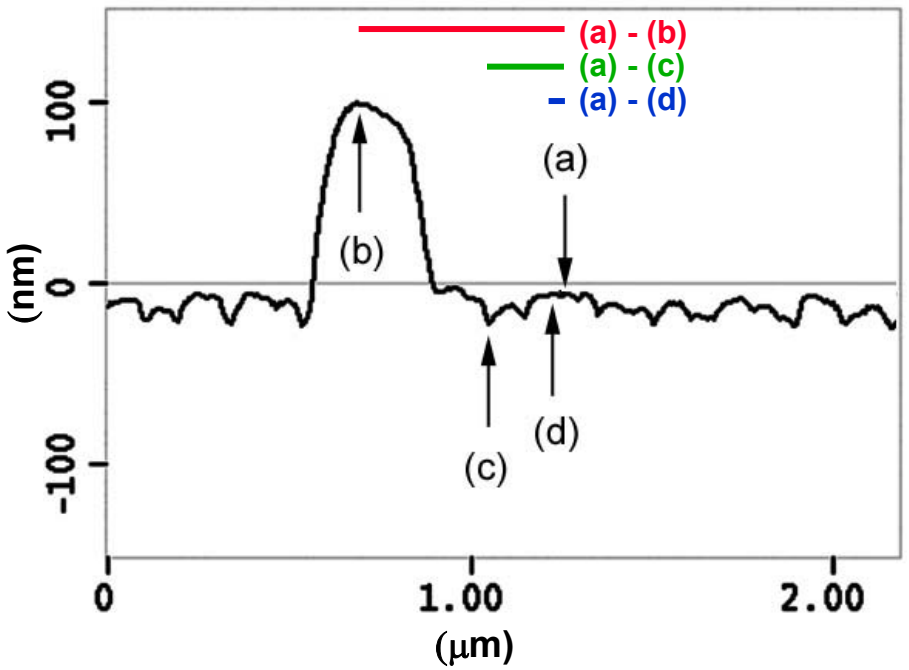
Mechanism for GaN nuclei formation → NL decomposes during annealing, Ga atoms desorb into the gas phase and reincorporate with NH_3 to form GaN nuclei.

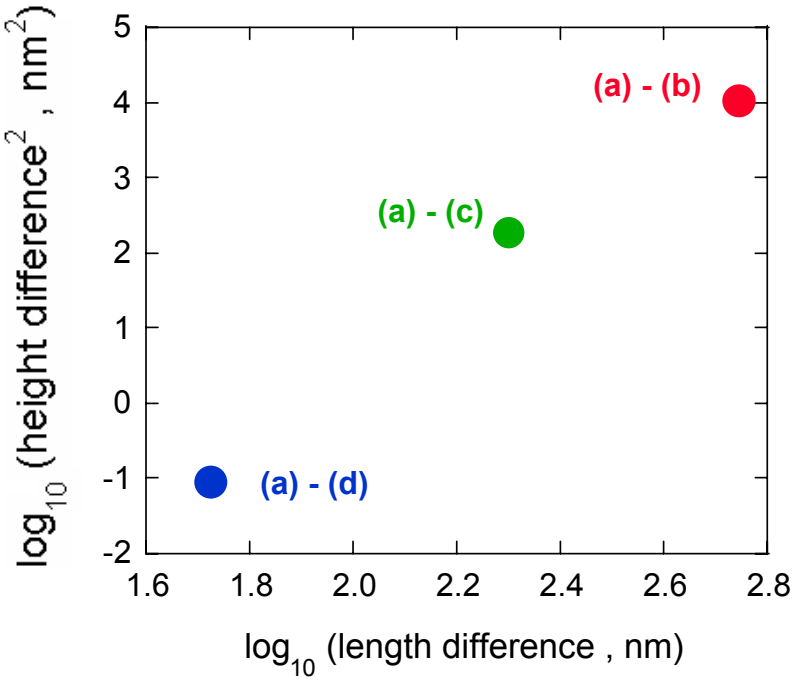
Gas phase desorption and reincorporation; not surface diffusion

Roughening / smoothing mechanisms can be extracted from AFM Power Spectral Density analysis

W. M. Tong and R. S. Williams, Annual Review Physical Chemistry 45, 401 (1994).

Calculate height-height correlation function = $g(r)$ or $g(|q|)$ from $h(x, y)$

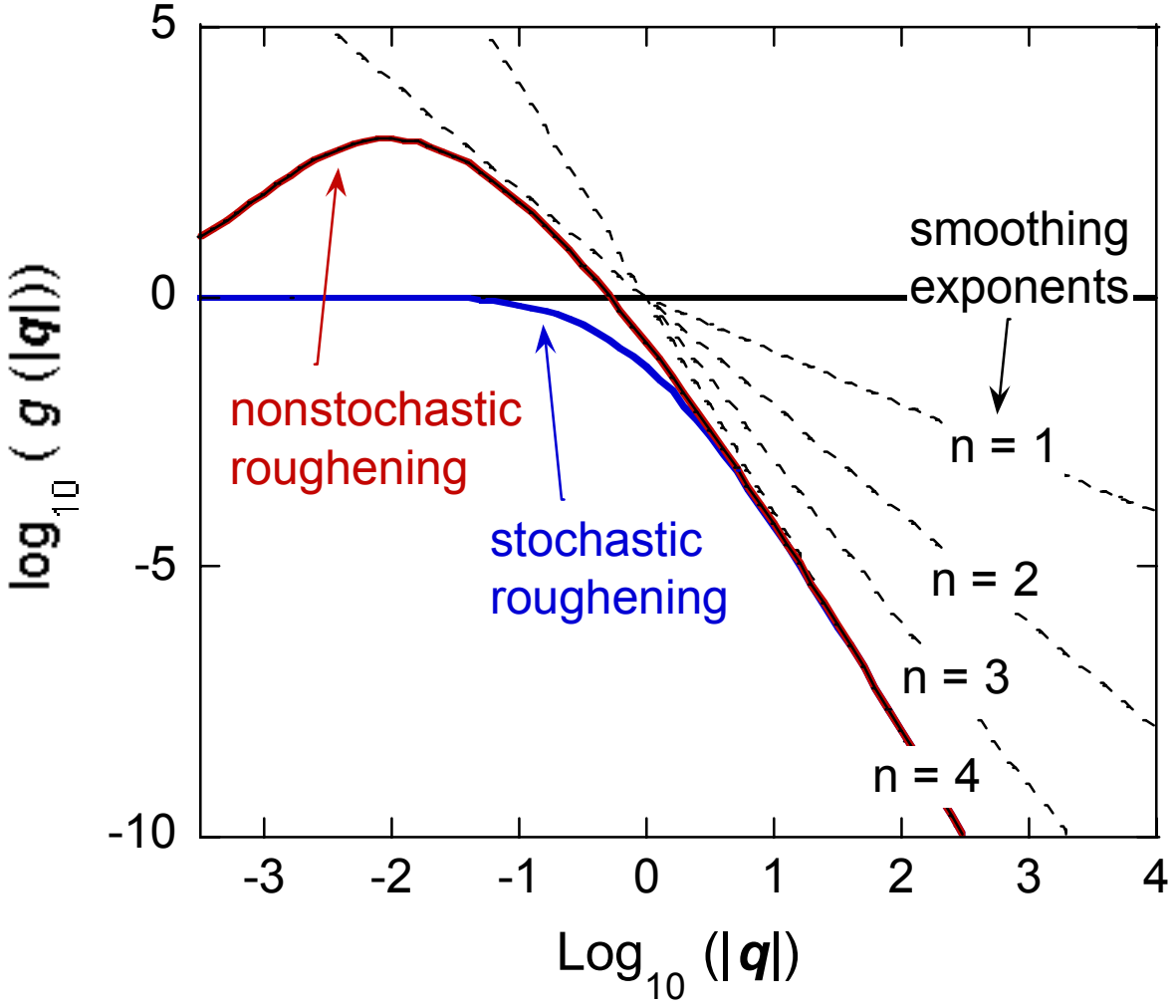




$$\sigma_{RMS} = \left(\sum g(r)\right)^{1/2}$$

Stochastic and non-stochastic roughening with smoothing

W. M. Tong and R. S. Williams, Annual Review Physical Chemistry 45, 401 (1994);
C. Herring, Journal of Applied Physics, 21, 301 (1950).



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

Stochastic roughening
- random deposition with length scale independence

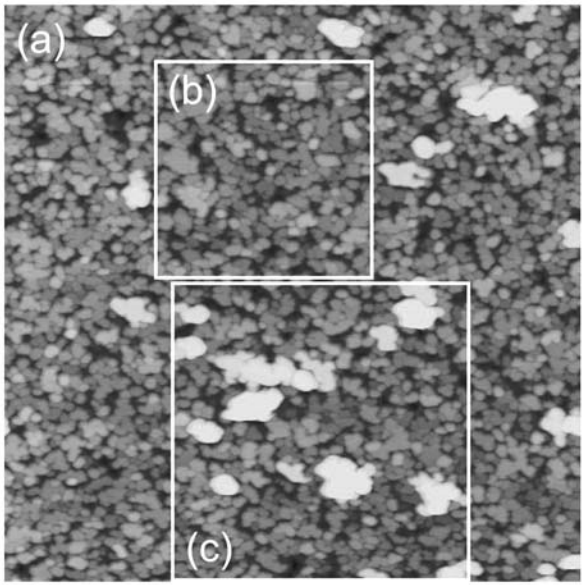
Stochastic roughening and smoothing by n = 4

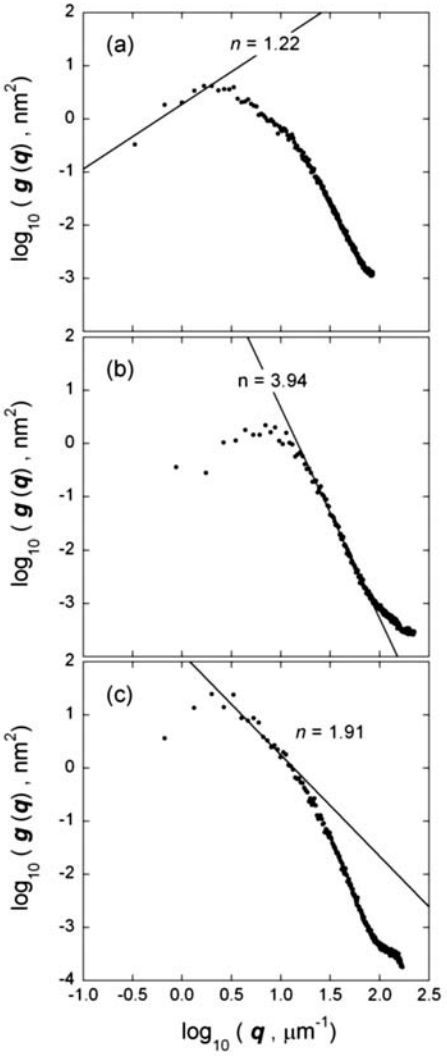
Non-stochastic roughening and smoothing by n = 2 and n = 4

Nucleation layer evolution mechanism was determined from analysis of AFM power spectral density

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

Calculate height-height correlation function = $g\,(r\,)$ or $g\,(|q\,|\,)$ from $h\,(x\,,y\,)$

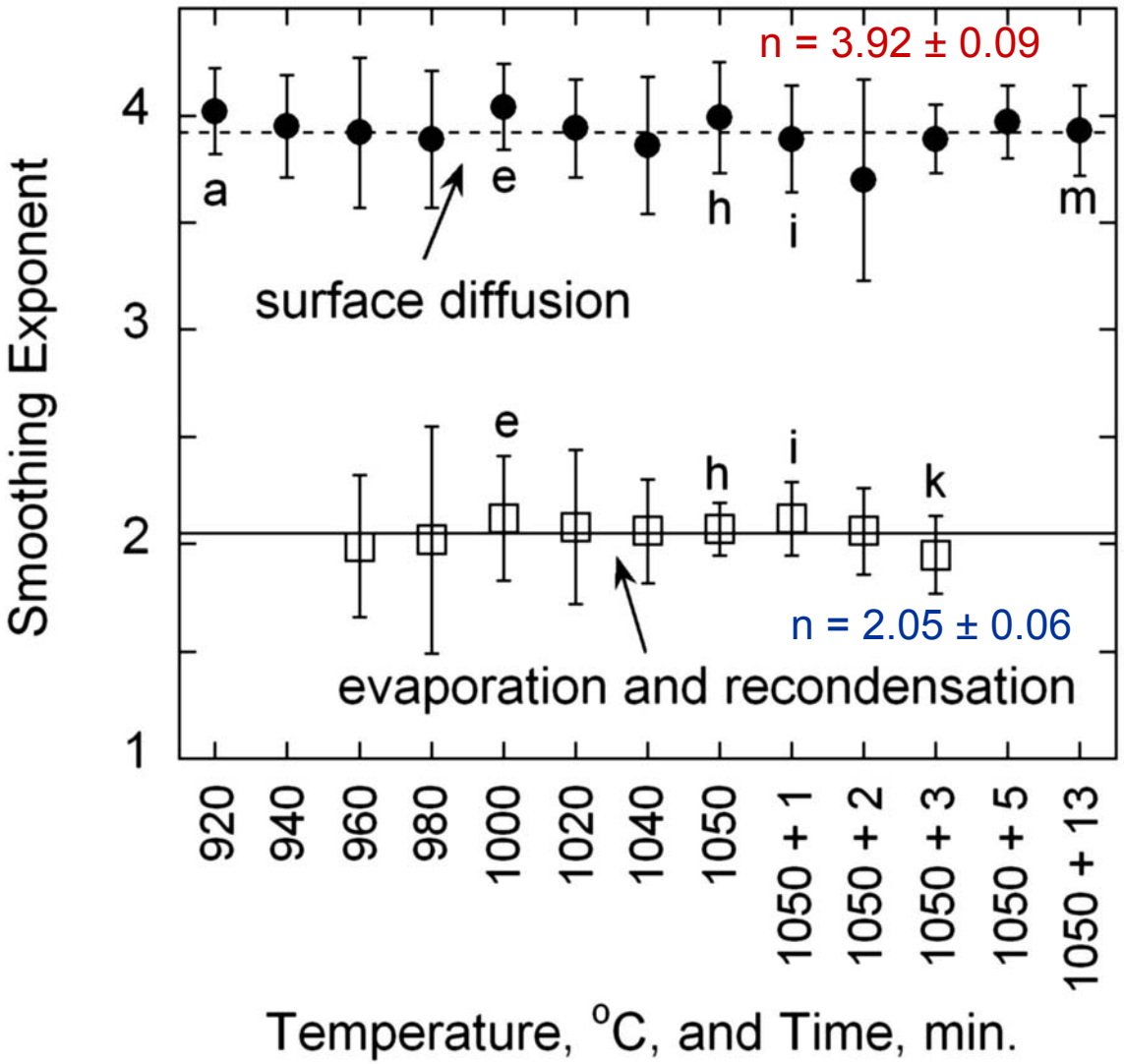




The g (r) is related to the RMS roughness σ_{RMS} *by*

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

Change in smoothing mechanism as NL is annealed



n = 1, plastic flow driven by surface tension

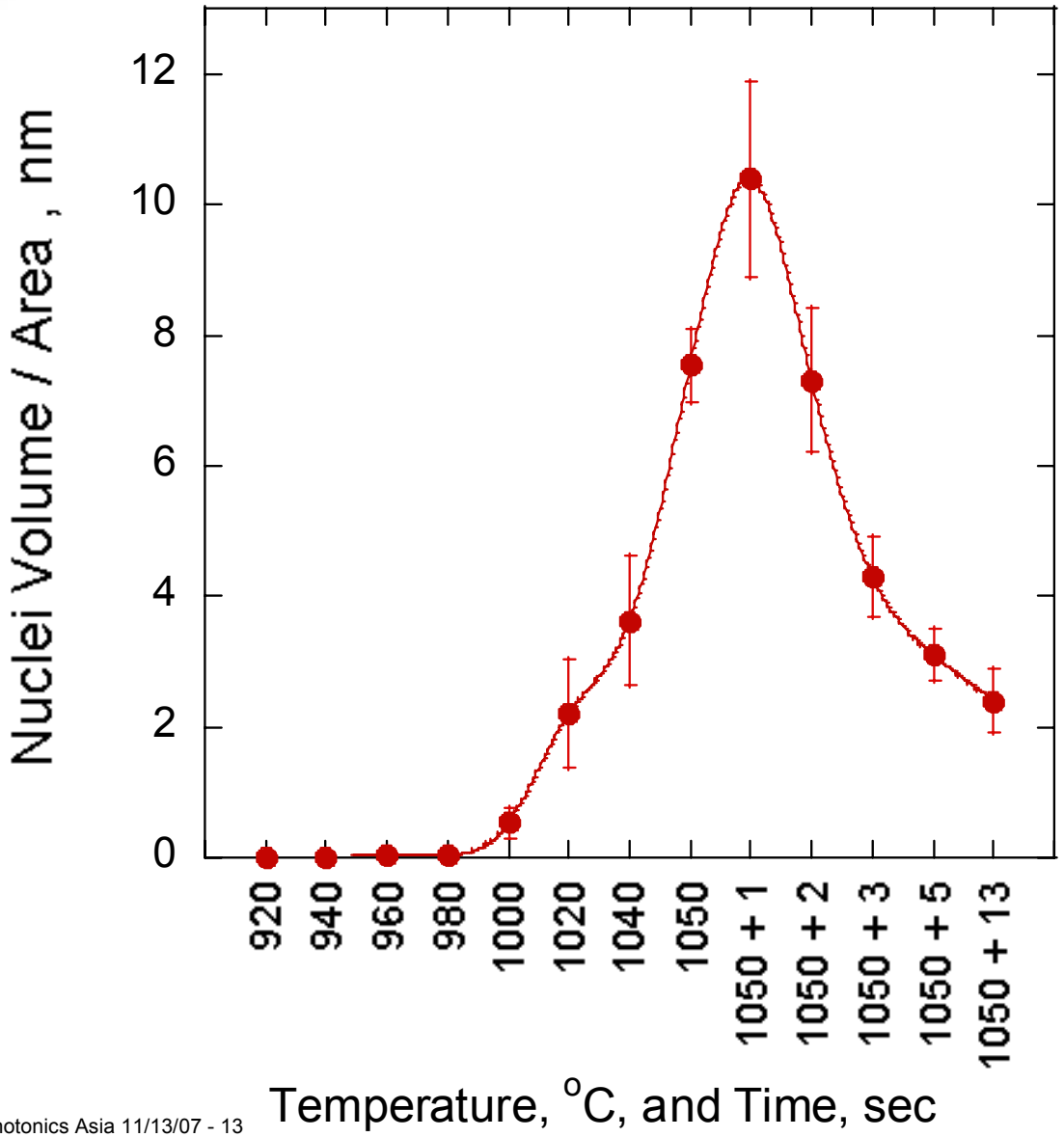
n = 2, evaporation and recondensation

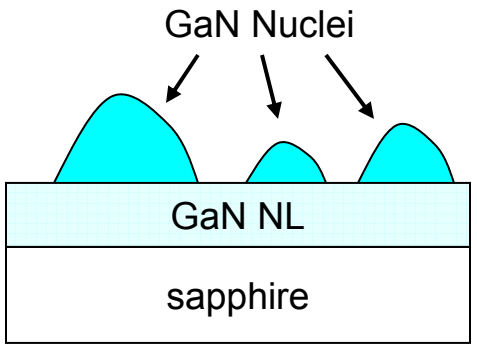
n = 3, volume diffusion

n = 4, surface diffusion

Consistent with gas phase transport model of Mitchell, Coltrin, and Han, J. Crystal Growth 222, 144 (2001).

Redistribution of mass from NL into nuclei

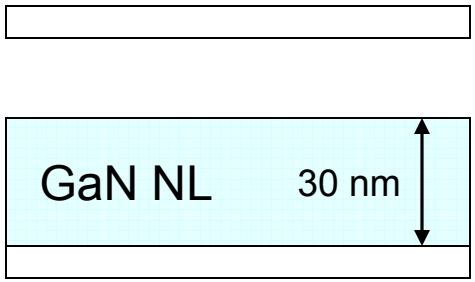




From the AFM images the GaN nuclei volume can be measured.

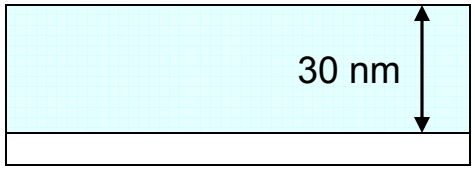
At 1050 °C + 1 min. anneal, the nuclei mass is 1/3 the original NL mass

Summary of NL evolution before high-T growth

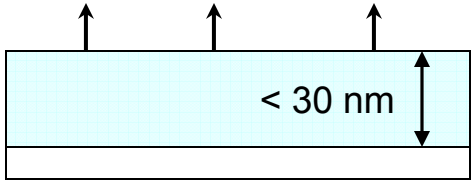


1. Sapphire annealed in H_2 1080 °C, cooled to 550 °C

2. Grow GaN NL, reflectance signal increases and can be used to control NL thickness.

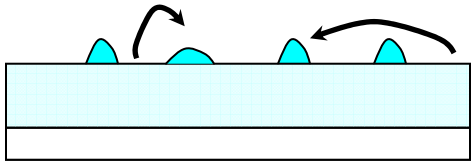


3. Heat NL to 850 °C, reflectance signal increases slightly due to T dependent change in optical constants. No NL decomposition.

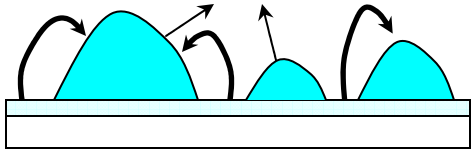


4. Above 850 °C, GaN NL starts to decompose, follows Arrhenius kinetics, E_A = 2.7 eV, A_0 varies. Rate limiting step is Ga desorption from the surface.

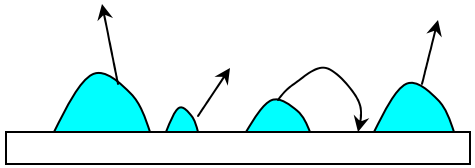
Summary of NL evolution before high-T growth



5. Near 960-1000 °C, NL begins to roughen. GaN nuclei are formed from Ga atoms generated during GaN NL decomposition. Reflectance continues to decrease.



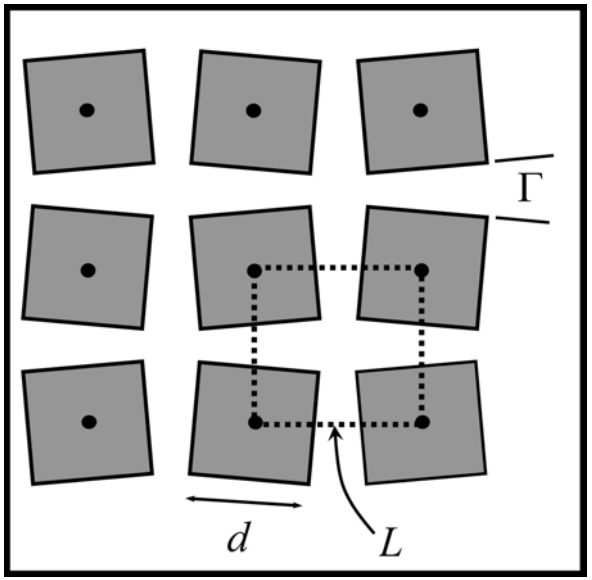
6. Near 1050 °C, nuclei reach maximum size, ≈ 1/3 of the NL Ga atoms are incorporated into the GaN nuclei. Nuclei reach maximum size at the same time the NL thickness reaches zero. Reflectance reaches a minimum.



7. With the original NL fully decomposed, further annealing at 1050 °C causes the GaN nuclei to decompose. Reflectance increases.

8. Further annealing at 1050 °C reduces the size of the GaN nuclei and the number density increases.

Geometric model relating nucleation density to the dislocation density



Γ is angle between grains, b is the Burgers vector, K is assumed to be unity and L is the distance between grains.

Note that independent of the dislocation generation mechanism lower nucleation density should lead to lower GaN films with lower dislocation densities.
Also, both generation mechanisms can be present to varying degrees.

Case 1: Dislocations are generated along tilt boundaries due to twist.

- The nucleation density, n_D, is related to the nuclei separation, L, by $n_D = 1/L^2$.
- For a small angle tilt boundary, the angle between grains, Γ, is related to L, by, $\Gamma \approx (Kmb)/L$.
- The number of dislocations, m, is $(L\Gamma)/(Kb)$ and along each dislocation edge, $m \approx 2L\Gamma/Kb$.
- The dislocation density, ρ, per grain area is $(2L\Gamma/Kb)(1/L^2) = (2\Gamma/Kb)(1/L)$, and since $1/L = n_D^{1/2}$, the dislocation density is:

$$\rho \approx (2\Gamma/Kb)\, n_D^{1/2}$$

Case 2: Dislocations are generated with in each nuclei.

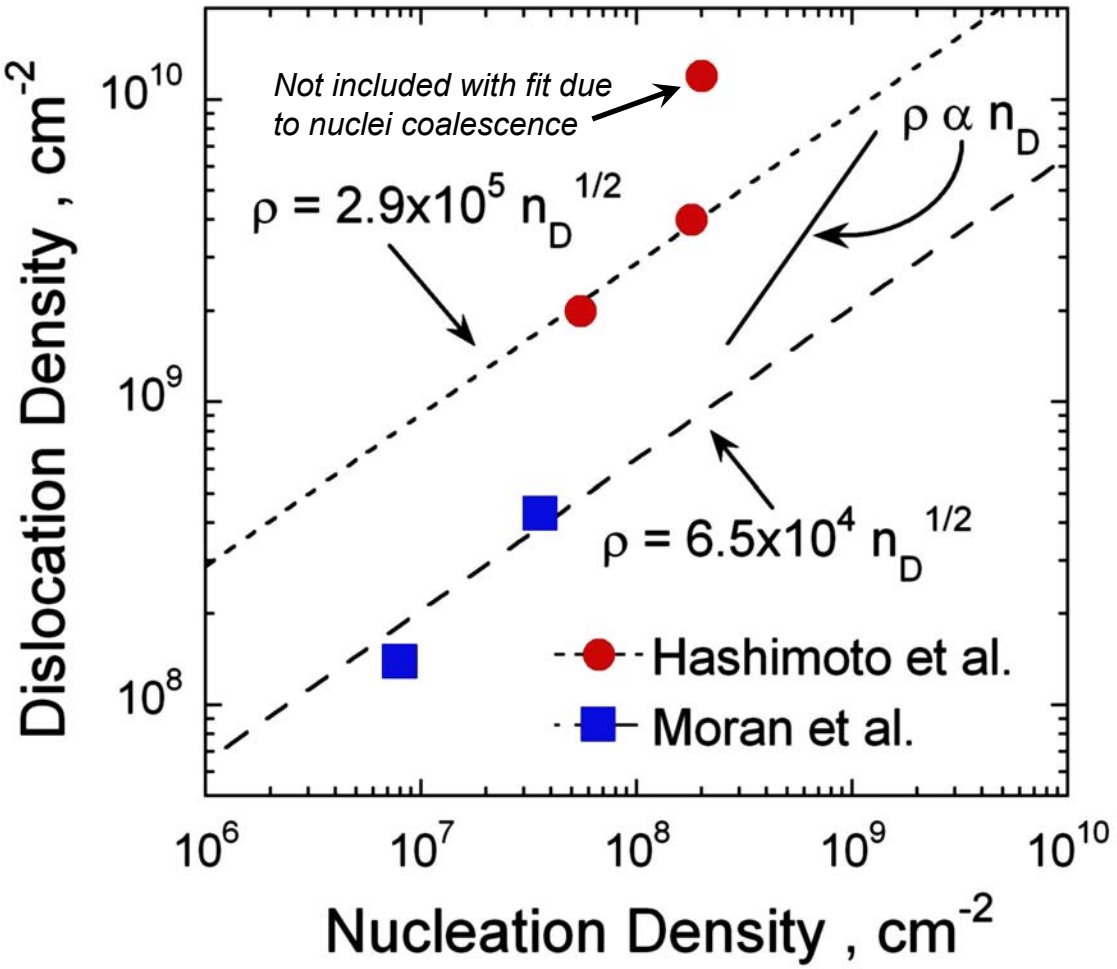
- If there is an a number dislocations associated with each nuclei, then on average:

$$\rho \approx n_D$$

Comparison of these two cases with data from the literature

Red circles from T. Hashimoto, Jpn. J. Appl. Phys. ***38****, 6605 (1999).*

Blue squares from B. Moran, J. Crystal Growth ***273****, 38 (2004).*



Fits assume $\rho \propto n^{1/2}$, and $\rho \propto n$ is shown for comparison.

One of the data points from Hashimoto excluded since the GaN nuclei are partly coalesced.

The difference in the prefactors (2.9×10^5 vs. 6.5×10^4) is due to the different substrates (sapphire vs. SiC) and growth pressures (150 torr vs. 760 torr) used for these growths.

Suggests that $\rho \propto n_D^{1/2}$

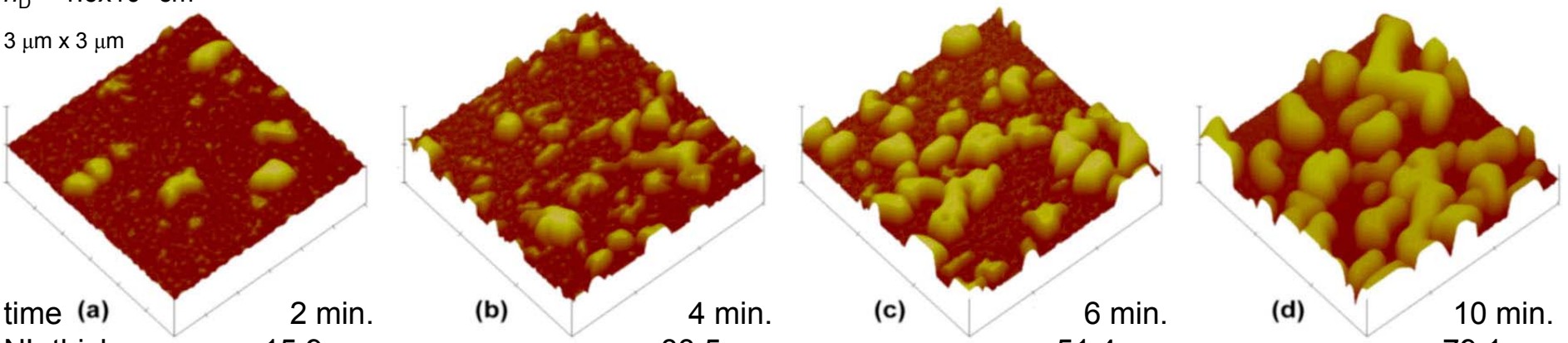
Nucleation layer thickness can be used to vary the nucleation density

← Individual GaN nuclei ┐

n_D = 1.6x10^8 cm^{-2}

3 μm x 3 μm

After growth each NL was ramped to ~1000 ºC in reduced NH_3 flow to develop the GaN nuclei. Annealing was stopped when the minimum in the reflectance was observed.



time	(a)	(b)	(c)	(d)
	2 min.	4 min.	6 min.	10 min.
NL thickness	15.9 nm	33.5 nm	51.4 nm	79.1 nm

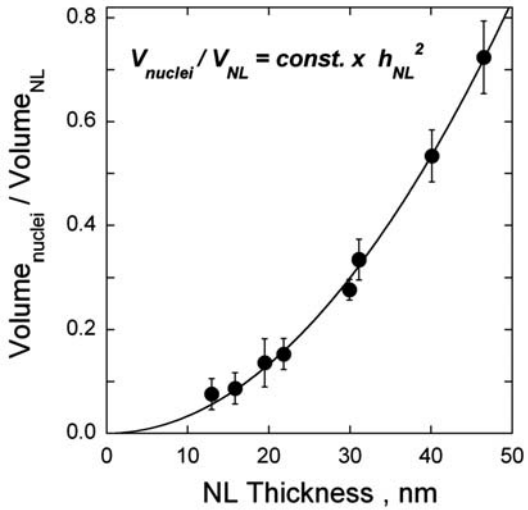
└ coalesced GaN nuclei →

Achieving lower nucleation density is difficult because:

Thinner NLs are required, which is harder to precisely control using reflectance.

For thinner NLs, less of the NL volume is converted into GaN nuclei, probably due to the reduced NL decomposition rate. (see Lang, et al., JCG 277, 64 (2005).)

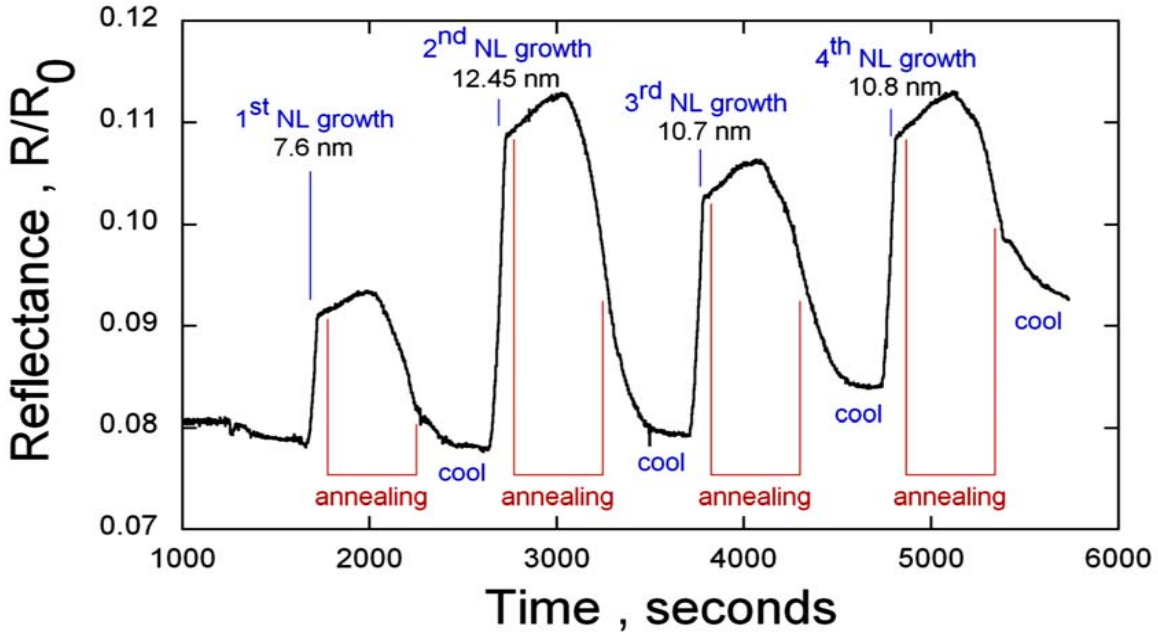
Conversion ∝ (NL thickness)^2 →

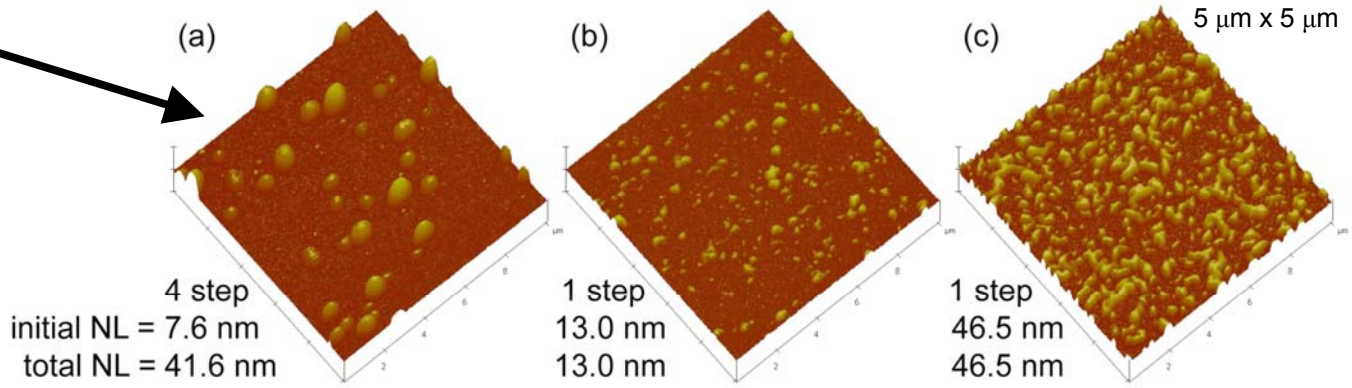


Lower nucleation densities can be achieved using the method of Lang *et al.*

See Lang et al., JCG ***277****, 64 (2005) for details on this method.*

- **Growth and annealing of multiple NLs.**
- **Reflectance is used to monitor GaN NL thickness during growth step and roughness during annealing.**
- **Result: larger nuclei and lower nucleation density.**
- **Nucleation density correlates with thickness of the first NL.**

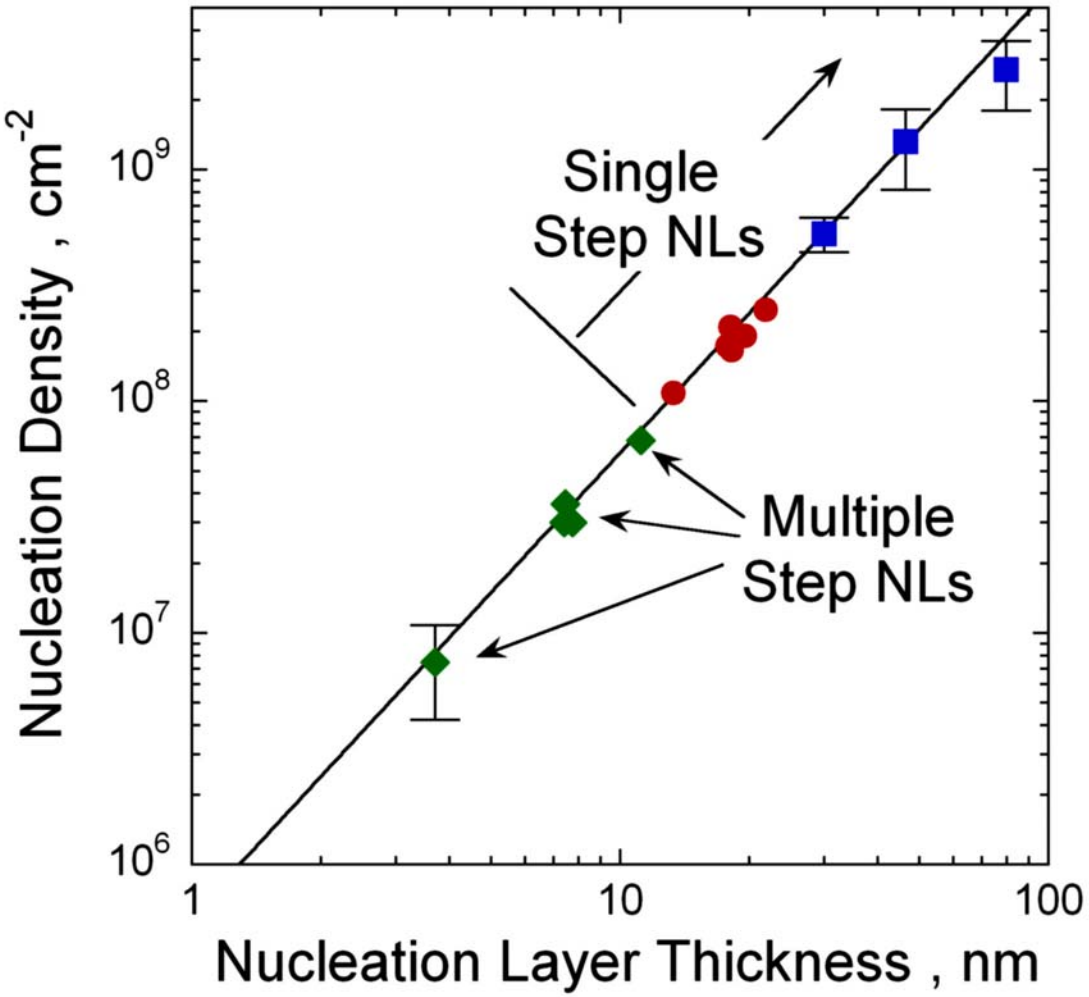




Correlation between the NL thickness and the resultant nucleation density

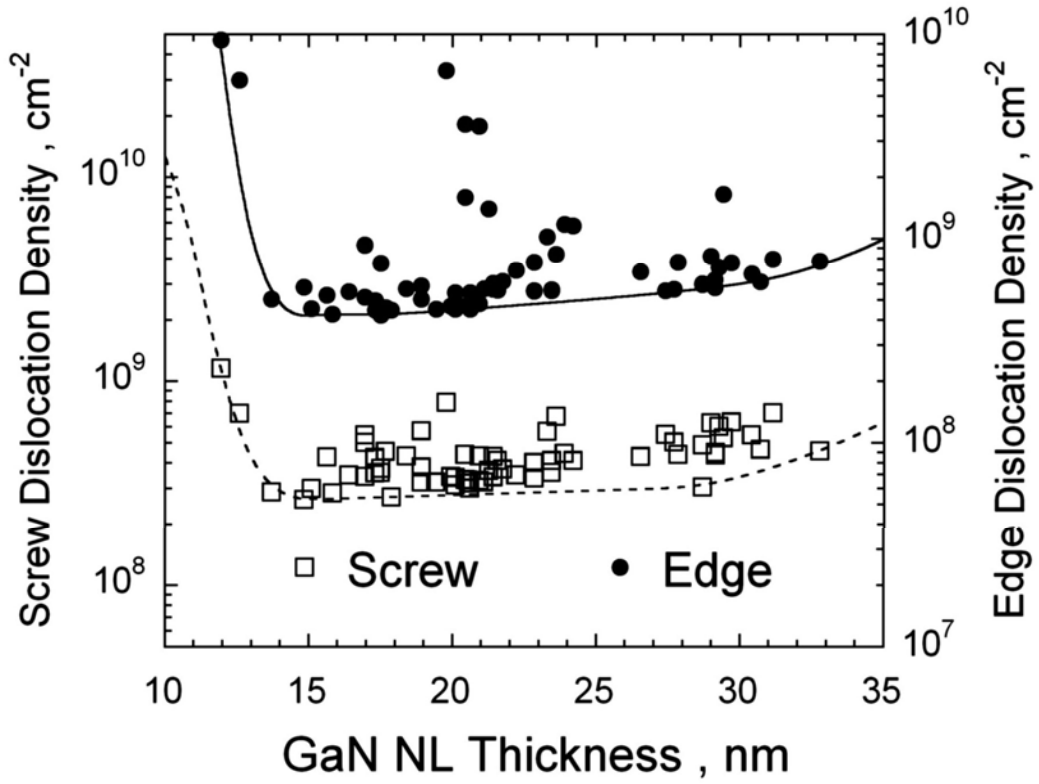
- Combining the data from
  - Single-step NL growths and anneals (red).
  - Single-step NL growth and anneals where were partially coalesced (blue).
  - Multiple NL growth and annealing steps (green). For these layers the first NL thickness is used.
- A power-law relationship is observed between nucleation density and NL thickness.

$$n_D = \text{const.} \times h_{NL}^2$$



Growth optimization to lower GaN dislocation density

Dislocation densities measured using XRD following Lee et al., APL 86, 241904 (2005).



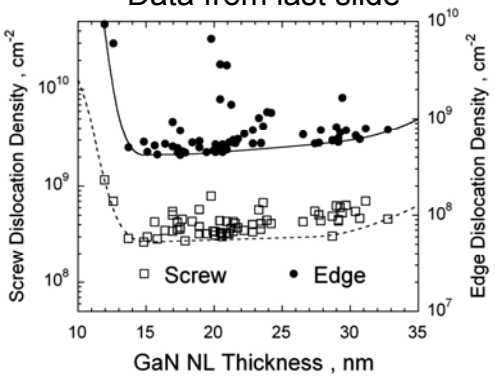
- Achieve dislocation densities of
  - 2.5×10^8 cm^{-2} for screw component
  - 4.0×10^8 cm^{-2} for edge component
- Lower DDs have been achieved
  - 1×10^8 cm^{-2} using SiN layers (Datta, *Superlattices and Microstructures* ***36****, 393 (2004)*.)
- Lower DDs for GaN on SiC
  - 1.5×10^8 cm^{-2} using HT AlN NL (Moran, *JCG* **273** 38, (2004)).

Goal: Routinely achieve dislocation densities of ~ 1×10^8 cm^{-2} on sapphire without too much additional growth overhead

Dependence of dislocation density on nucleation density

If the nucleation density depends on the square of the nucleation layer thickness we can re-plot the dislocation density vs. NL thickness data.

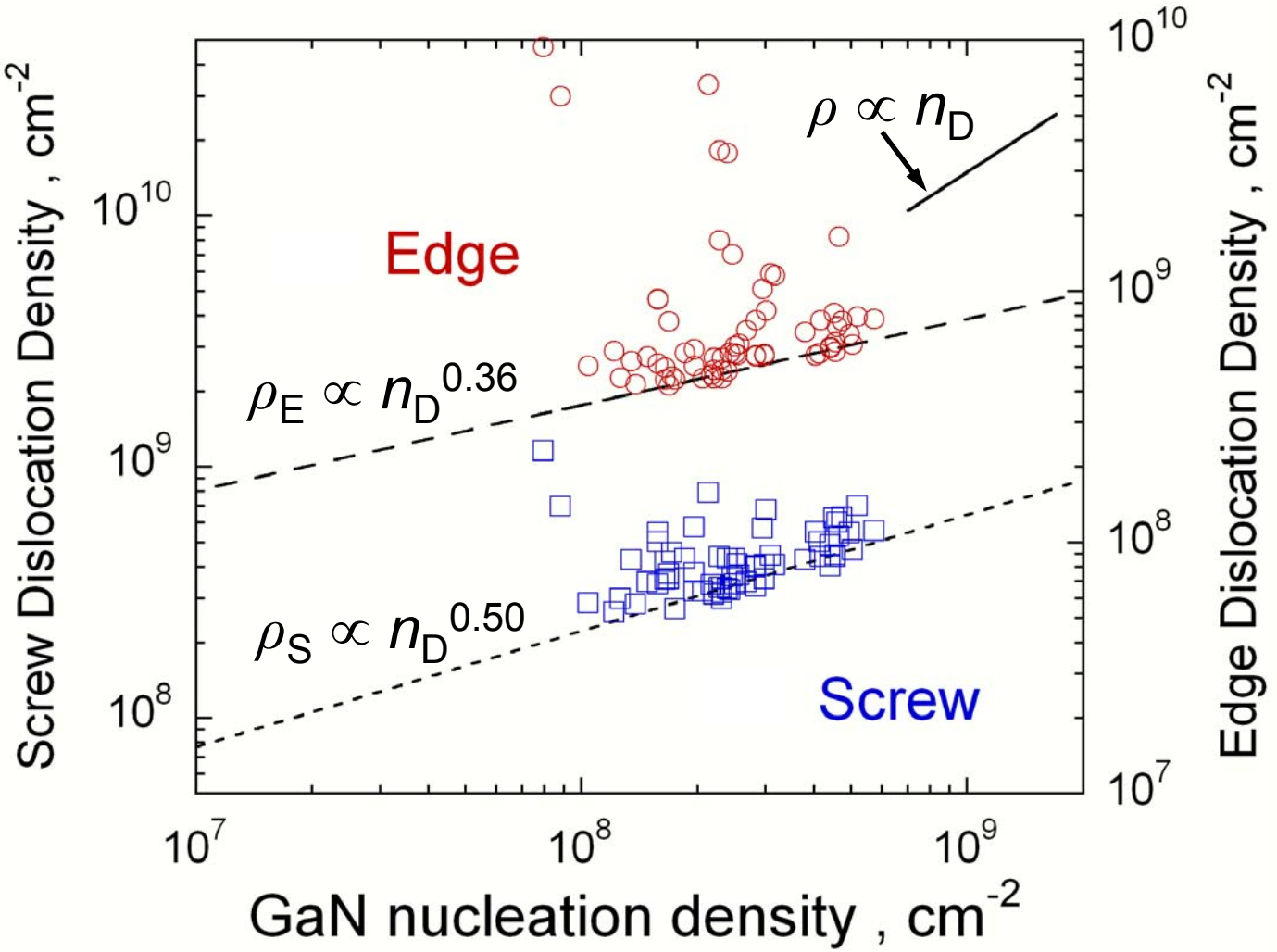
Data from last slide



And assume that

$$n_D = \text{const.} \times d_{NL}^{\,2}$$

Fits are through the lowest dislocation density films at each value of the nucleation density.



Summary and Conclusions

- Mechanism for GaN NL decomposition
  - Ga-atom evaporation, then re-incorporation to form nuclei
- Geometric model for correlating dislocation density, ρ, to nucleation density, n_D.
  - 1) Dislocations are generated along tilt boundaries, $\rho \propto n_D^{1/2}$.
  - 2) Dislocations are generated from each nuclei , $\rho \propto n_D$.
- Ultra-low nucleation densities achieved using the multi-step NL growth and annealing of Lang *et al*.
- Nucleation density can be varied by changing the NL thickness, h_{NL}, and $n_D \propto h_{NL}^2$.
- Plot of dislocation density vs. NL thickness shows that data follows $\rho \propto n_D^{1/2}$ more likely than $\rho \propto n_D$.

Next : Use ultra low nucleation densities to reduce GaN dislocation densities