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III-Nitride Nanowires: From the Bottom-Up to the Top-Down

Solid-State Lighting Science Energy Frontier Research Center

Sandia National Laboratories, Albuquerque, NM

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Outline

- Why nanowires for SSL?
- Science challenges for nanowire-based SSL
  - Synthesis & fabrication
  - Characterization & properties
  - Electroluminescent structures
- Recent research highlights
  - Nanowire-templated growth
  - GaN/InGaN core-shell nanowires
  - Top-down fabrication of GaN nanorods and nanorod LEDs/lasers
- Summary

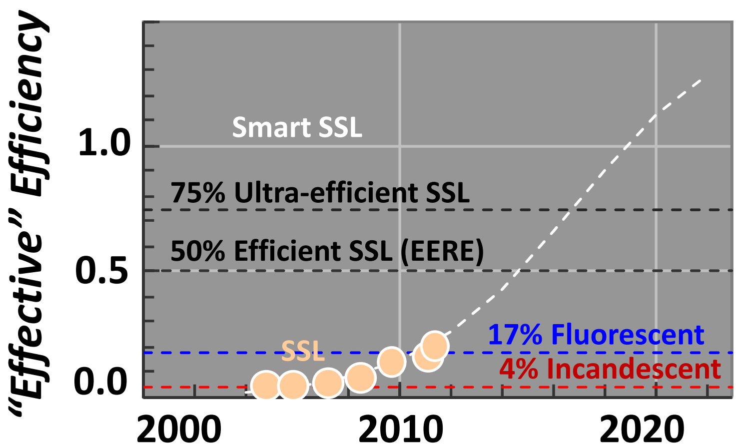
Lighting is a large fraction of energy consumption and is low efficiency

- **~22% of electricity consumption is for general illumination**
- **Lighting is one of the most *<u>inefficient</u>* energy technologies in buildings → opportunity!**
- **Achieving 50% efficient lighting would have tremendous global impact:**
  - **decrease electricity consumed by lighting by > 50%**
  - **decrease total electricity consumption by 10%**

<u>Efficiencies of energy technologies in buildings</u>:

Heating:	**70 - 80%**
Elect. motors:	**85 - 95%**
Fluorescent:	**~20%**
Incandescent:	**~3-5%**

US DOE target: 50%
"Ultra-efficient" SSL*: ≥ 70%

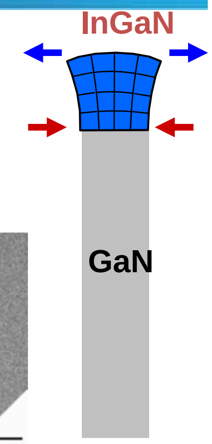


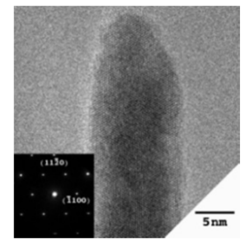
J.Y. Tsao

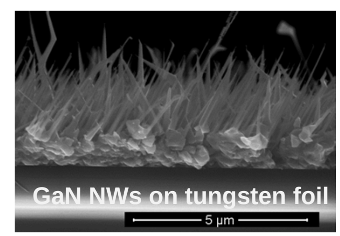
Why III-nitride (AlGaInN) nanowires for SSL, PV, etc.?

Strain accomodation (applies to rows: Few or no threading dislocations; Wider range of alloy compositions possible; Single crystal growth on any substrate; Reduced piezoelectric strain)

Nanowire attribute	SSL Benefit
Few or no threading dislocations	IQE, reliability
Wider range of alloy compositions possible T. Kuykendall et al. *Nat. Mat.* **6**, 951 (2007) Q. M. Li, G. T. Wang, "*Appl. Phys. Lett.*, **97**, 181107 (2010).	Wider color space (yellow-red)
Single crystal growth on any substrate	Cost, flexibility
Reduced piezoelectric strain C. Y. Wang et al. Opt. Expr. *16*, 10549 (2008)	IQE (reduced QCSE)
Reduced efficiency droop (?) *W. Guo et al., Nano Lett.*, **2011**, *11* (4),1434 *H. Nguyen et al.,Nano Lett.*, **2011**, *11* (5), 1919	Reduced droop
Controlled light extraction(?), lasing	EQE, droop?
Non-polar, semi-polar facets	IQE (Reduced fields)
Discrete	Good scientific test system

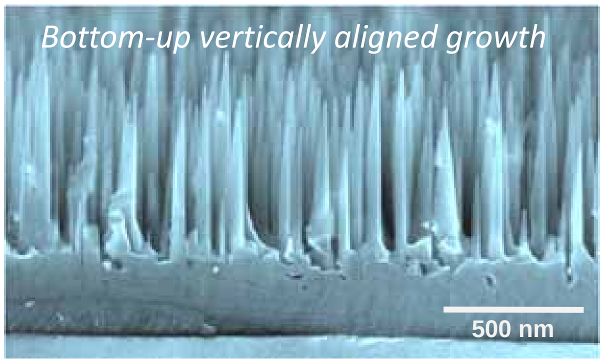






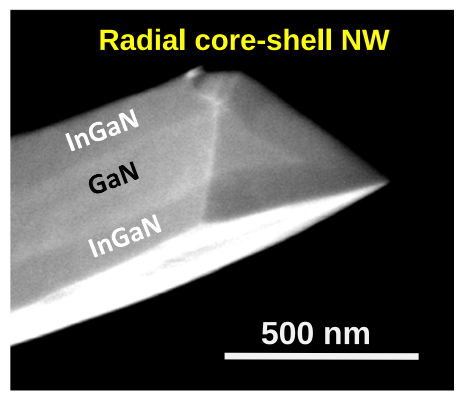
Challenge: Synthesis & Fabrication

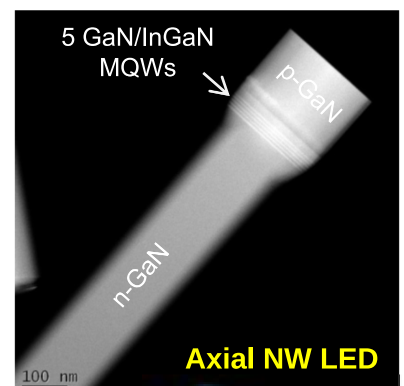
- “Bottom-up” catalyzed growth (VLS) vs. “top-down” fabrication



G. T. Wang et al., *Nanotechnology 17 5773-5780* (2006)
Q. Li, G. T. Wang, *Appl. Phys. Lett. 93, 043119* (2008)

- InGaN/GaN nanowire heterostructures for visible light emission
  - Facet-dependent growth
  - Limits of In incorporation
  - Radial vs axial heterostructures

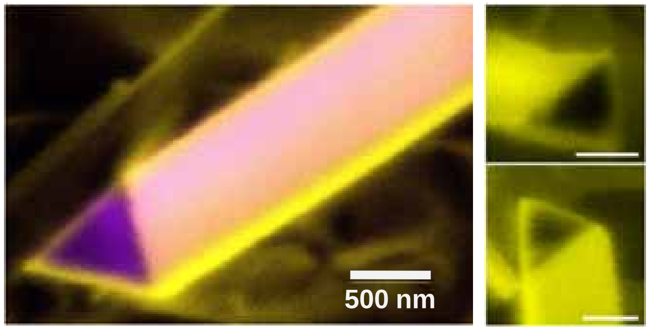




Challenge: Properties – Understand, control, optimize

Defect luminescence

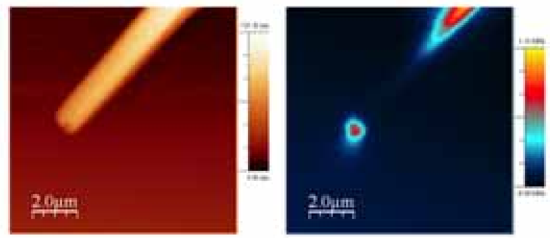
Spatially-resolved cathodoluminescence (CL)



Q. Li, G. T. Wang, *Nano Lett.*, **10** (5), 1554 (2010)

Carrier transport

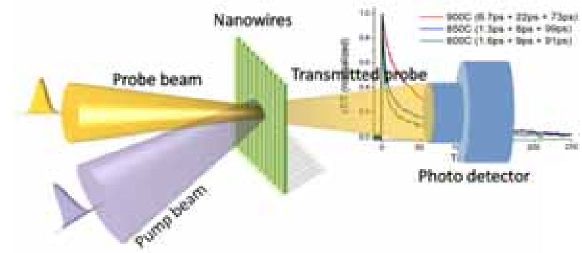
Transport imaging (NSOM/EBIC)



L. Baird et al., "*Appl. Phys. Lett.*, **98**, 132104, **2011**.

Carrier dynamics

Ultrafast pump-probe spectroscopy



P.C. Uppadhya et al., *Semicon. Sci. Tech.* **25,** 024017 (2010)

Electrical properties

In-situ TEM electrical breakdown

T. Westover et al. *Nano Lett.*, **9**, 257 (2009).

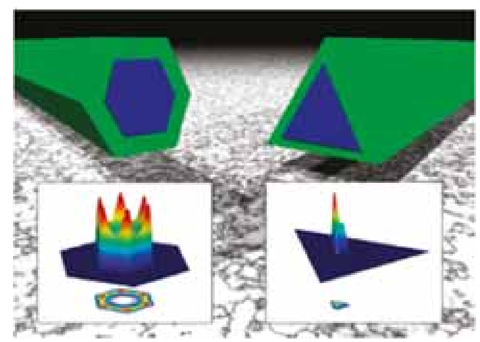
Mechanical properties

In-situ TEM nanomechanics

J. Y. Huang et al. *Nano Lett*, **11 (4),** 1618 (2011)

Electronic structure

Theoretical modeling



B. Wong et al., *Nano Lett* ***11*** ***(8)****, 3074, 2011*

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Challenge: Electroluminescent structures

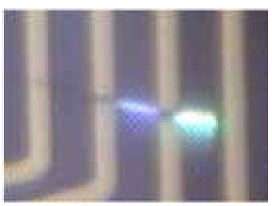
Goal: ***Investigate light generation & emission via nanowire electrical injection***

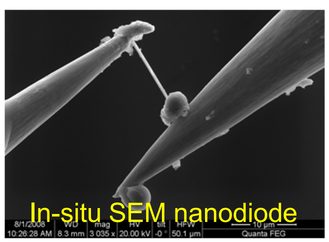
Focus: Electrical injection – contacts, localization, efficiency, carrier/leakage paths

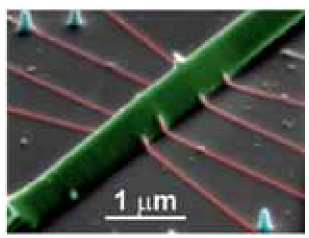
Electroluminescence – waveguiding, extraction, scattering, efficiency (EQE)

Single nanowire devices

Individual lighting element

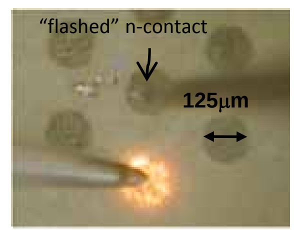


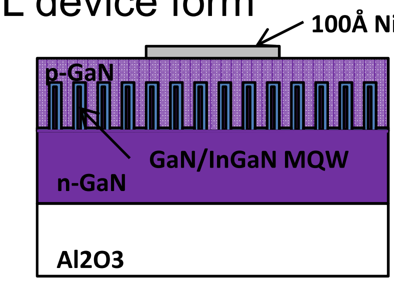




Integrated nanowire devices

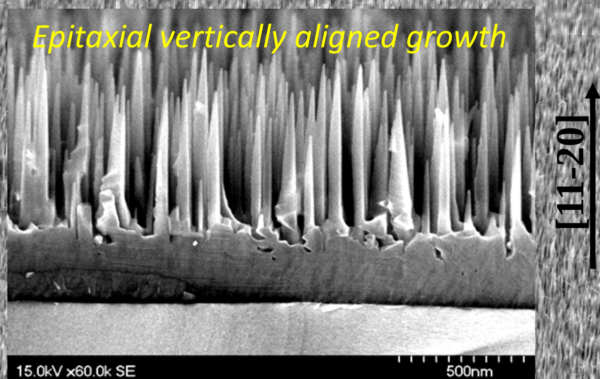
Ultimate SSL device form

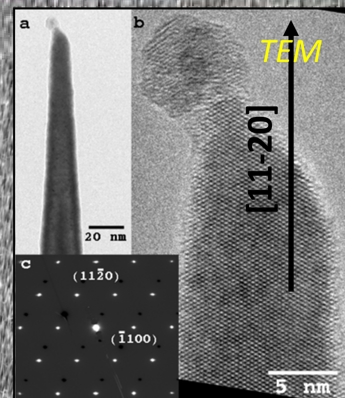


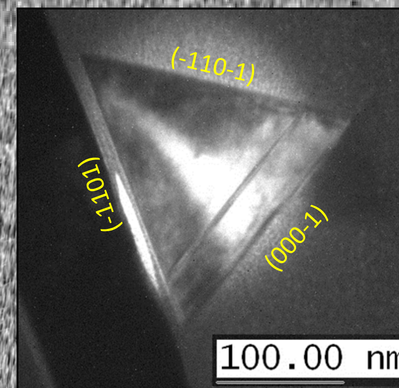


SSLS EFRC

"Bottom Up" aligned GaN nanowire growth







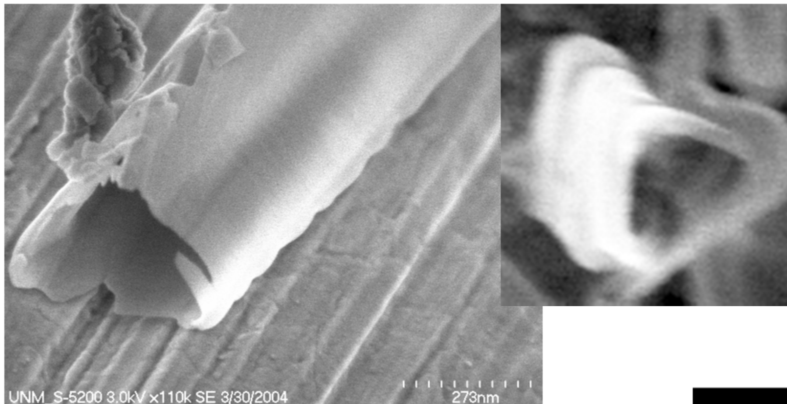
- Nanowires grown by Ni-catalyzed MOVPE/MOCVD (VLS)
- Highly-aligned vertical growth over large areas (2" r-sapphire wafer)
- Controllable densities as high as ~150 nanowires μm^{-2}

  Q. Li, G. T. Wang, *Appl. Phys. Lett. 93, 043119* (2008)

  Q. Li, J. R. Creighton, G.T. Wang. *J. Crys. Growth 310 3706-3709* (2008)
- Primary [11-20] growth orientation (⊥ to (11-20) *a*-plane)
- Triangular faceted -- (000-1) and equiv. (-1101) and (-110-1)
- TEM: Single crystal, dislocation free; *c*-plane stacking faults

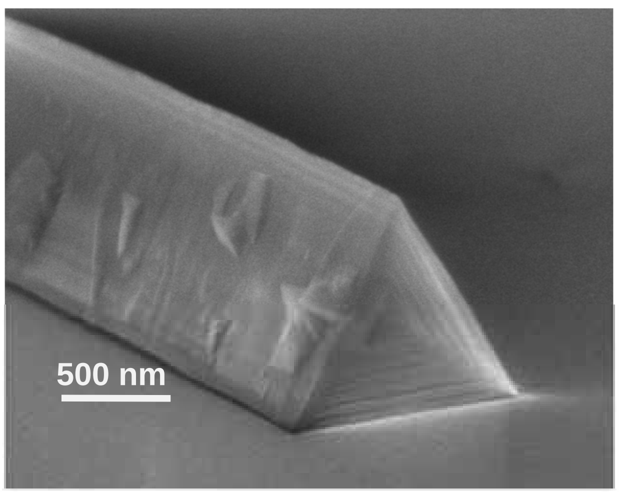
  G. T. Wang et al., *Nanotechnology 17 5773-5780* (2006)

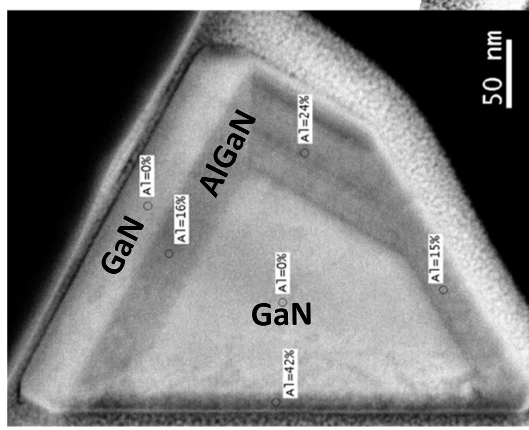
Radial heterostructure nanowire growth



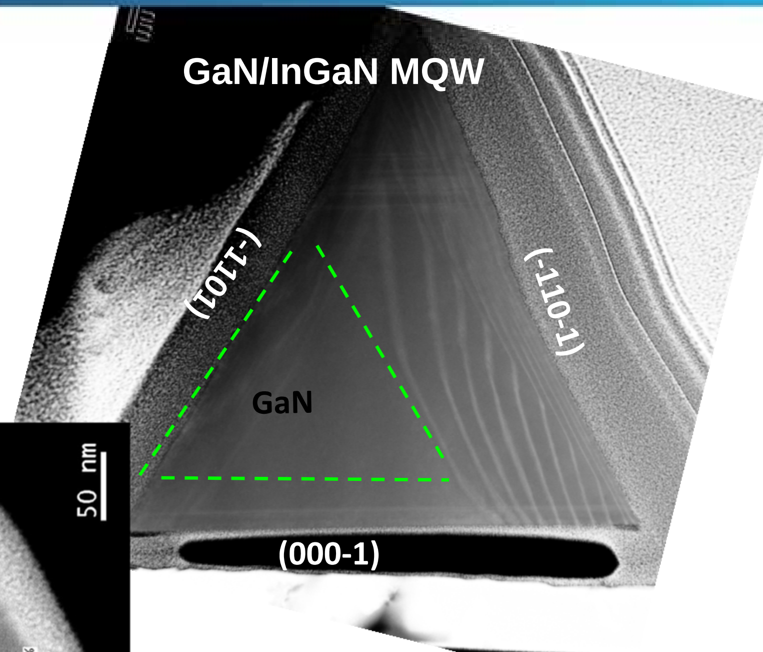
G. Stan et al. , *Nanotechnology*, ***20***, **2009**

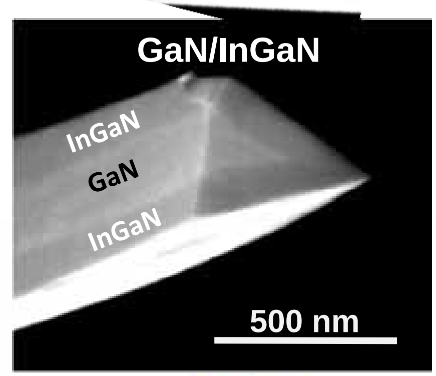
AlN Nanotubes





GaN/AlGaN/GaN
Double heterostructure



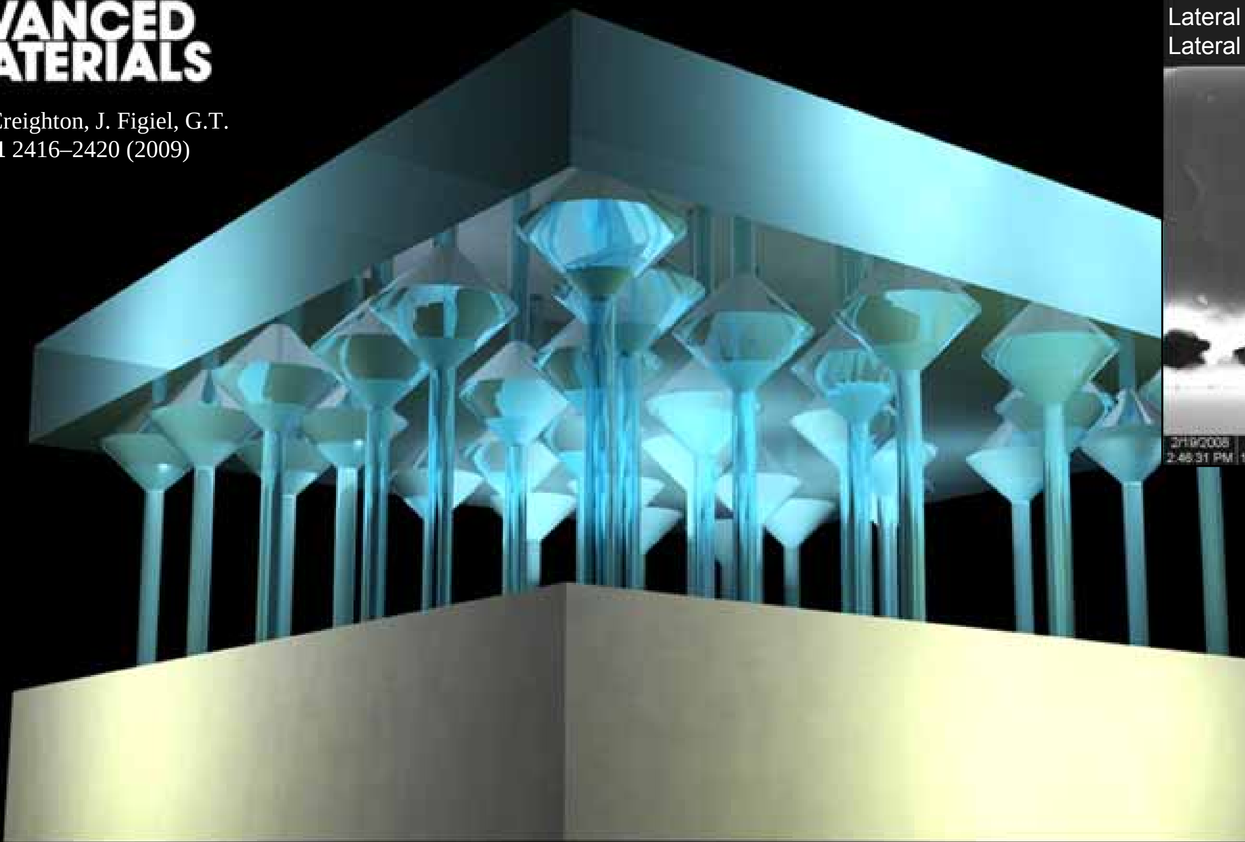


Nanowire-templated lateral epitaxial growth (NTLEG) of GaN

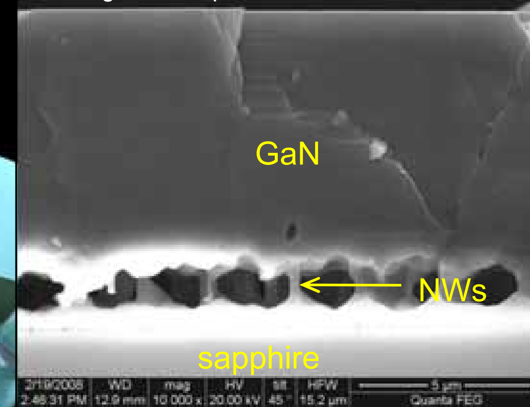
Inexpensive method to reduce dislocation density in GaN films growth on lattice mismatched substrates

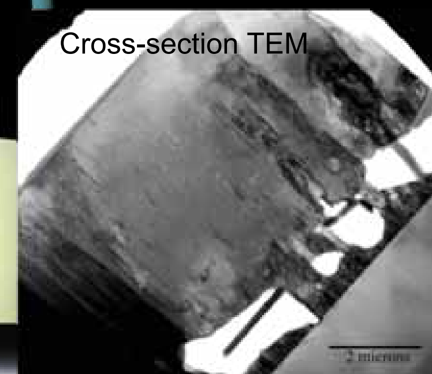
ADVANCED MATERIALS

Q. Li, Y. Lin, J.R. Creighton, J. Figiel, G.T. Wang, *Adv. Mat.*, **21** 2416–2420 (2009)



Nanowire array growth: 780 °C 3 min.
Lateral growth step 1: 900 °C 15 min.
Lateral growth step 2: 1075 °C for 15 min.

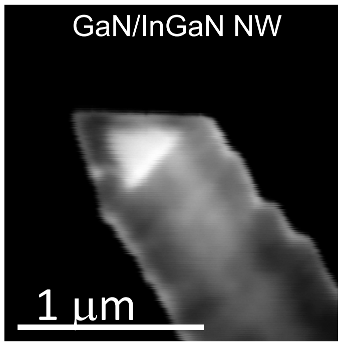




NTLEG films shows ~20 times reduction in dislocation density compared to LT nucleation layer

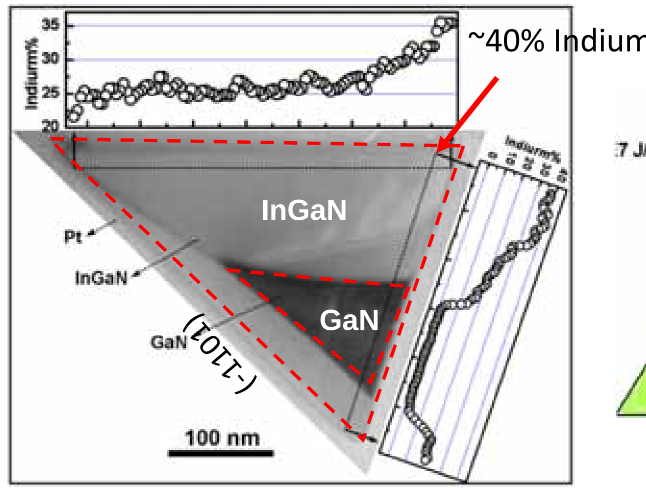
High In incorporation in GaN/InGaN core-shell NWs

- InGaN: visible wavelengths for solid-state lighting, PV, etc.
- Strain limits practical In incorporation in InGaN thin films (e.g., green-yellow-red gap)

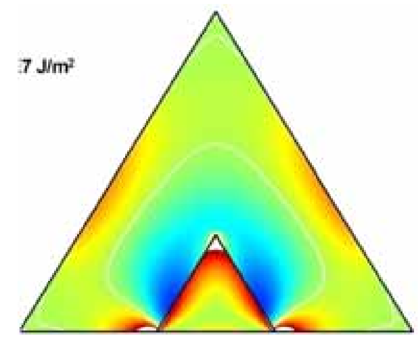


GaN core – 900 ˚C, 10 min.
InGaN shell – 760 ˚C, 60 min.

CL shows multicolor emission up to the IR! (nonuniform InGaN composition?)



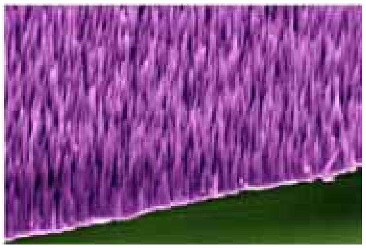
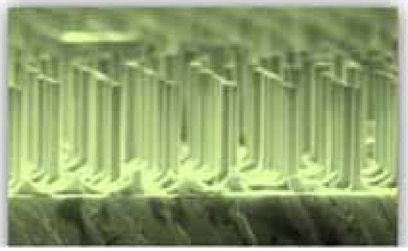
STEM/EDS shows In distribution, highest at surface/corners



2D FEA: highest In regions correlated w/lowest compressive strain regions

Nanowires good platform for high quality, high In content InGaN alloys

Bottom-up nanowires vs. Top-down nanorods

	Bottom-up nanowires	Top-down etched nanorods
		
Length:	up to hundreds of µm	Few µm
Diameter:	~30 nm to hundreds of nm	~30 nm to hundreds of nm
Dislocations	Few or none	Depends on film, wire diameter
Template/mask?	Yes, for ordered arrays or to direct vertical growth, otherwise no	Yes, to etch rods
Substrate	Any (?)	Crystallographically matched
Growth conditions	Narrow range, due to VLS or anisotropic growth conditions	Flexible, any used for planar film/device growth
Material quality	Tuneability may be limited by narrow growth conditions	Depends on starting material

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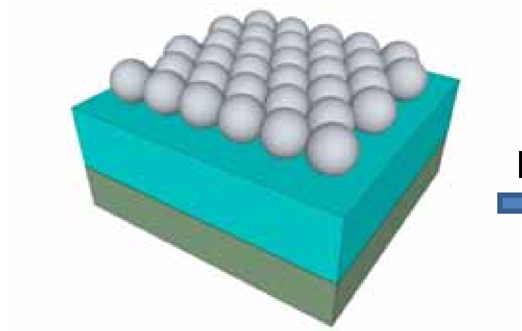
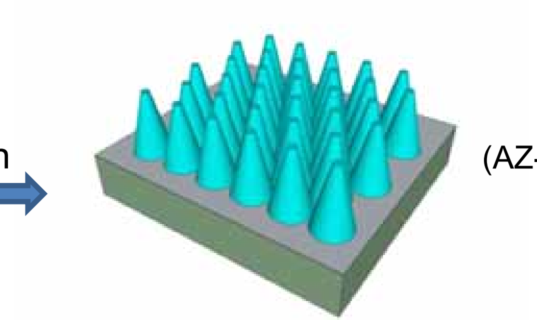
Highlight: New 2-step top-down ordered nanorod fabrication process

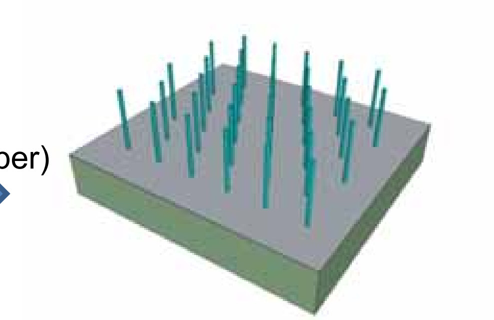


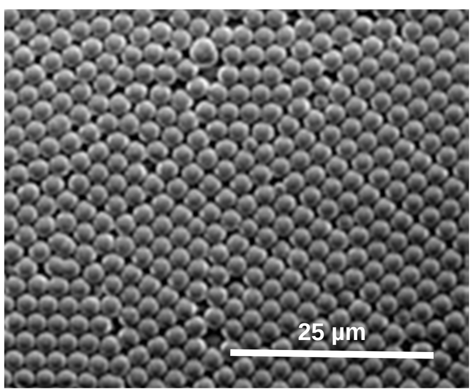
(0001) GaN on sapphire

ICP etch

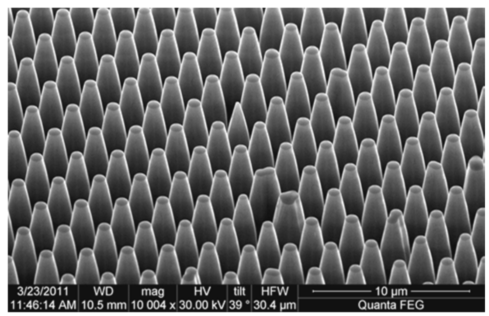


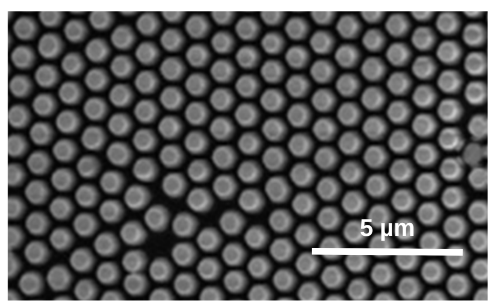
wet etch
(AZ-400K developer)





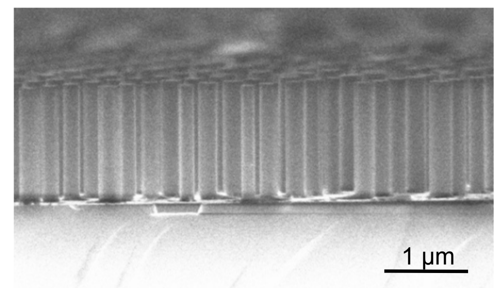
Q. Li, J. J. Figiel, G. T. Wang, Appl. Phys. Lett., **94**, 231105 (2009).

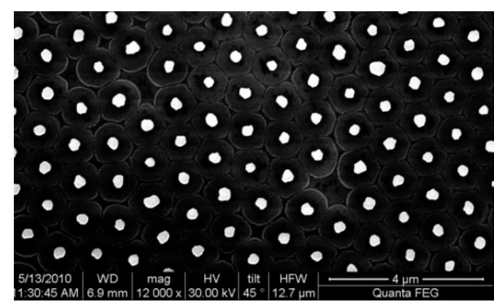




Plasma etch causes sidewall damage

C. Y. Wang et al., Opt. Expr. *16,* 10549–10556, 2008.

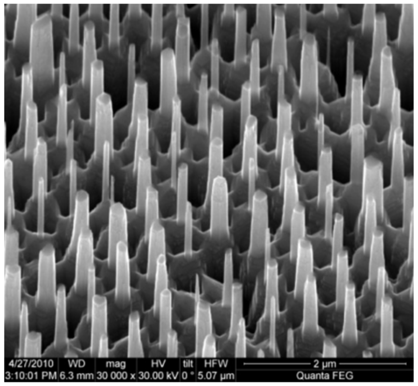




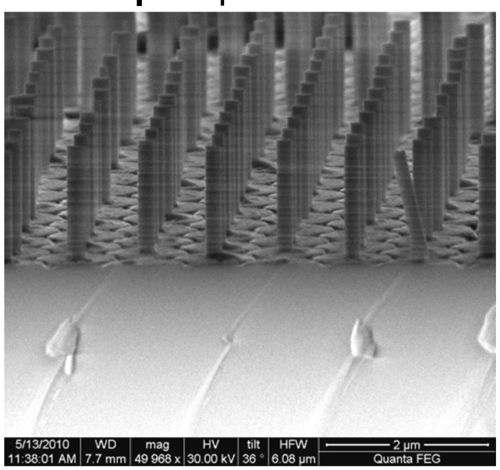
Wet etch removes sidewall damage

Selective wet etch – smooth and straight GaN nanorods

0.5 µm sphere size

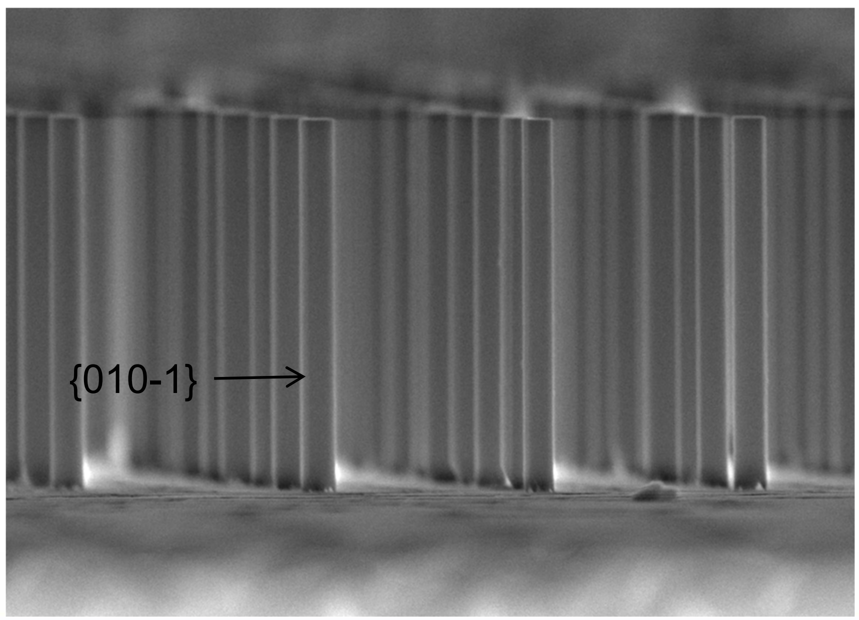


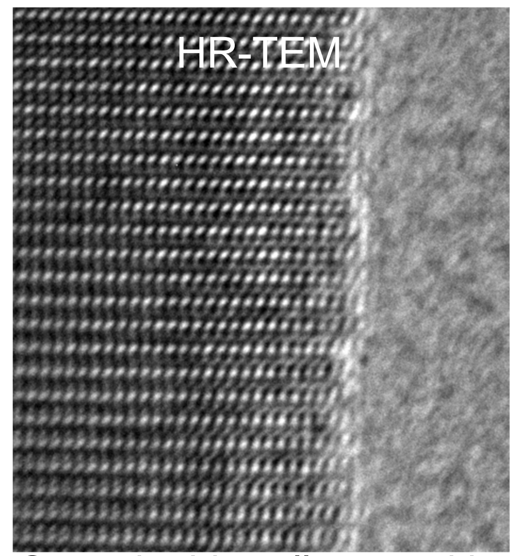
1 µm sphere size



- Etch rate negligible for top *c*-face & fast for {010-1} *m*-facets, leads to hexagonal nanorods with **straight & smooth** *m*-facets
- Sphere diameter: **pitch**, **max length** (~2x diameter)
- Wet etch time: nanorod **diameter**
- *PL experiments: wet etch removes plasma-induced damage*

3 µm sphere size

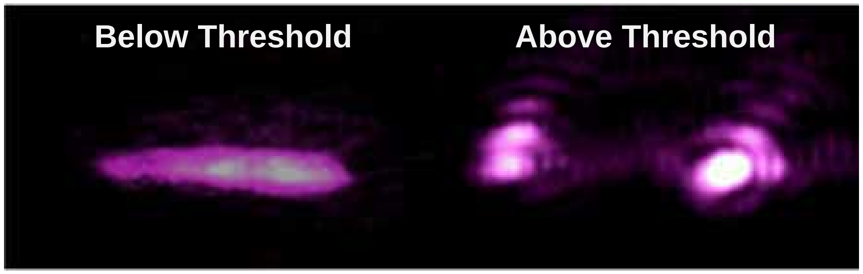




Smooth sidewall created by wet etch

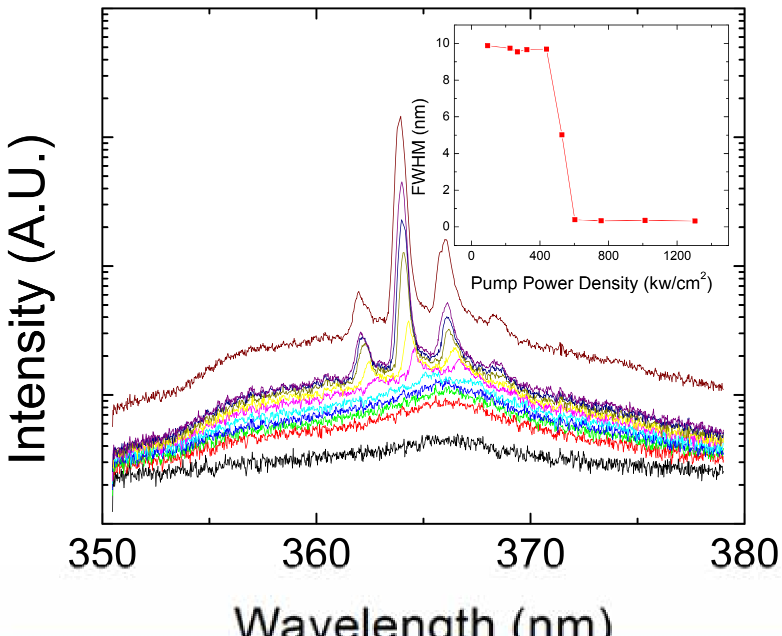
Single mode GaN nanorod lasing

Single-mode lasers are desirable (no mode-hopping, lower noise, stable far-field)

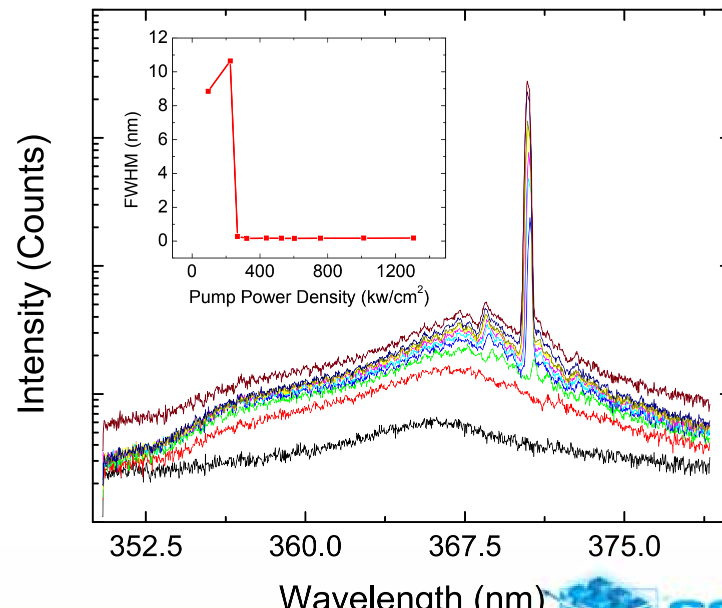


- μ-PL: 266 nm pump
- Narrow-Linewidth (<0.1nm), **Single-Mode**, 15dB Side Mode Suppression Ratio

- Nanorod dimensions: **250 nm** x 4.7μm
- Threshold ~ 500 kW/cm^2



- Nanorod dimensions: **130 nm** x 4.7μm
- Threshold ~ 250 kW/cm^2



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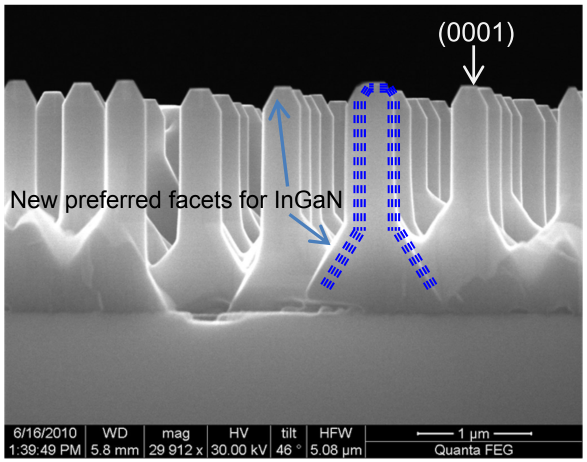
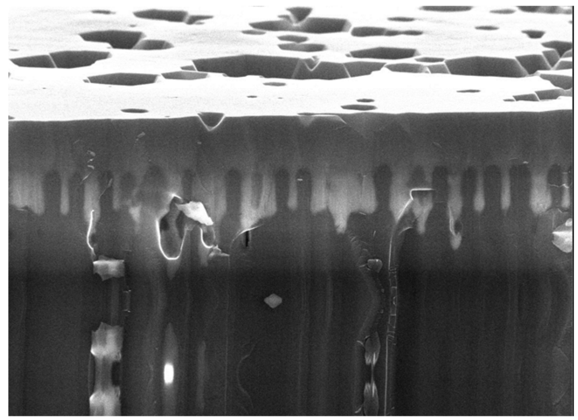
Electroluminescent radial GaN/InGaN nanorod LEDs

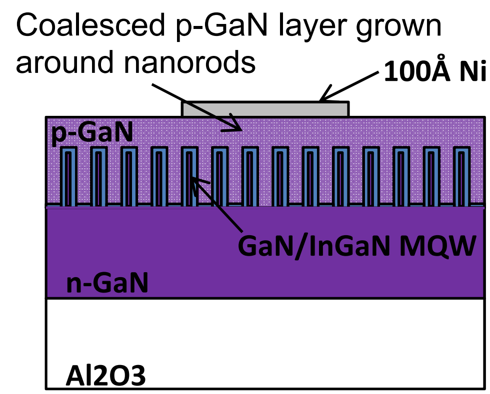
5-period MQW GaN/InGaN growth

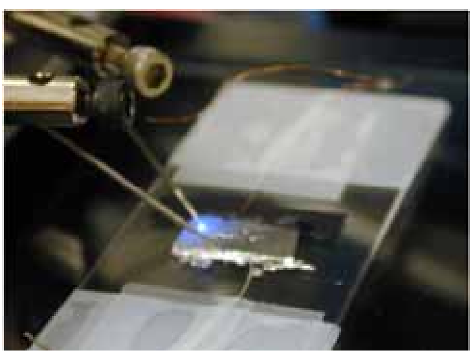


p-GaN coalescence

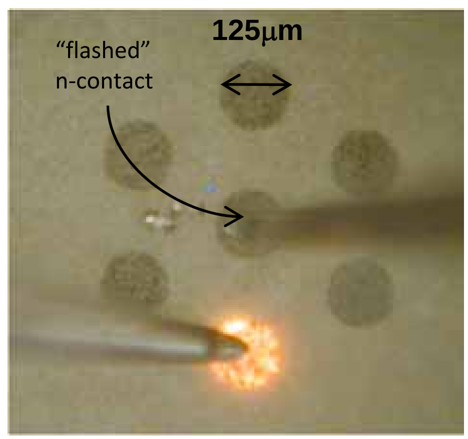


Nanorod LED

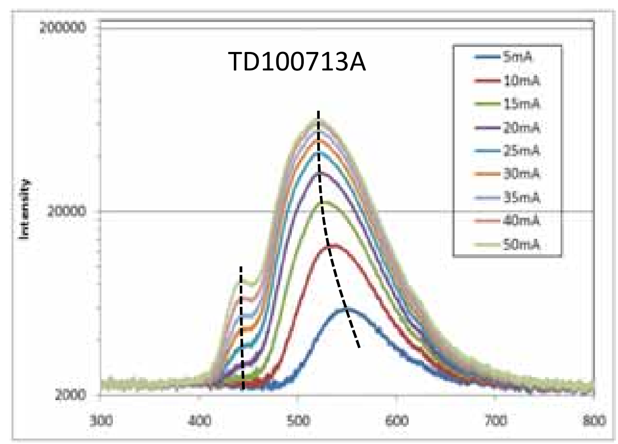




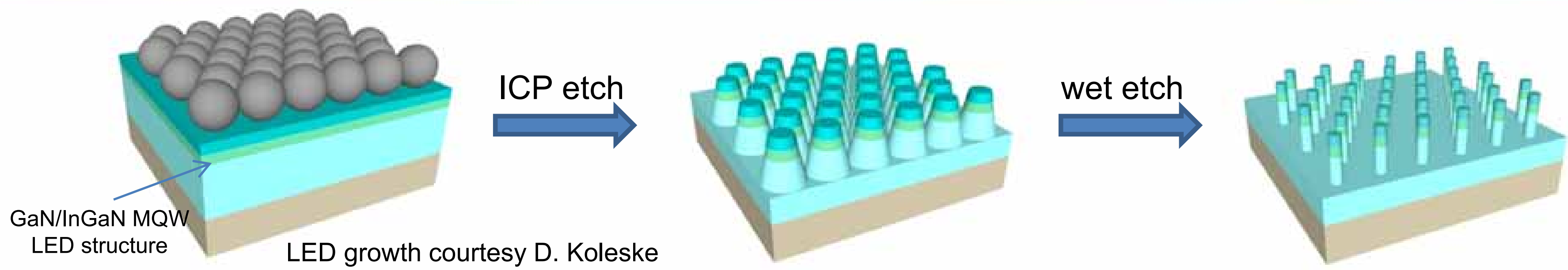
Bare wafer probe, In n-contact on side.

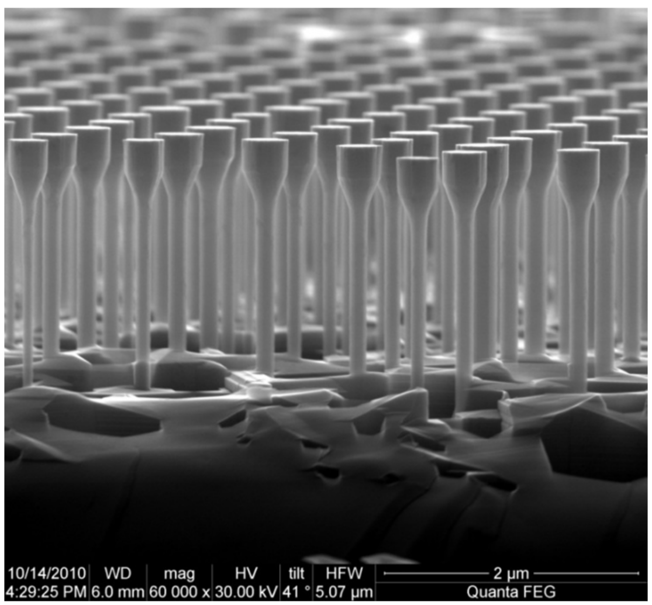


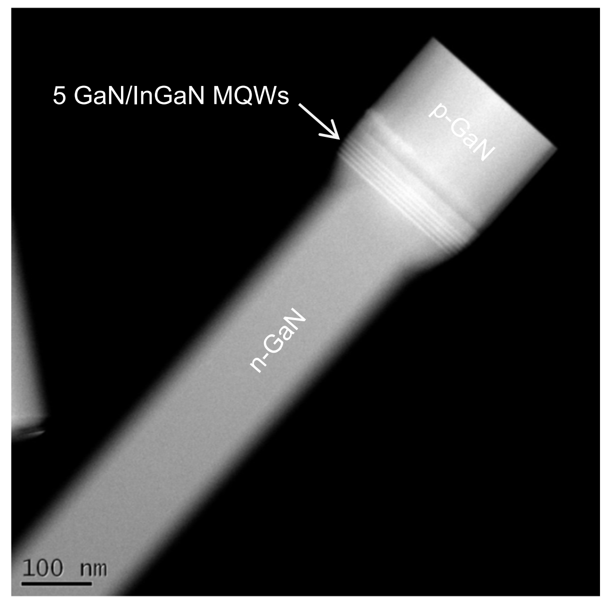
Electroluminescence



Axial GaN/InGaN LED Nanorods







"Nano-LED flashlight"

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Summary

- **Nanowire based architectures have several potential advantages over planar-based LEDs & may help to achieve 50% or higher efficiency SSL**
- **Numerous scientific & technical challenges for nanowire-based SSL**
- **Our efforts to address these challenges combine bottom-up and top-down nanowire synthesis/fabrication, nanocharacterization, and devices**

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Lincoln Lauhon, Jim Riley (Ph.D student) (Northwestern) – Atom probe tomography

Francois Leonard, Bryan Wong (SNL-CA) – Theoretical modeling

Jonathan Wierer – LED characterization

Andrew Armstrong –photoconductivity/DLOS – point defects

Karl Westlake, Mary Crawford – PL, IQE measurements

Dan Koleske – planar LED growth

Jeremy Wright, Igal Brener, Willie Luk (Ph.D student) – NW lasers

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