III-Nitride Nanowires for Solid-State Lighting

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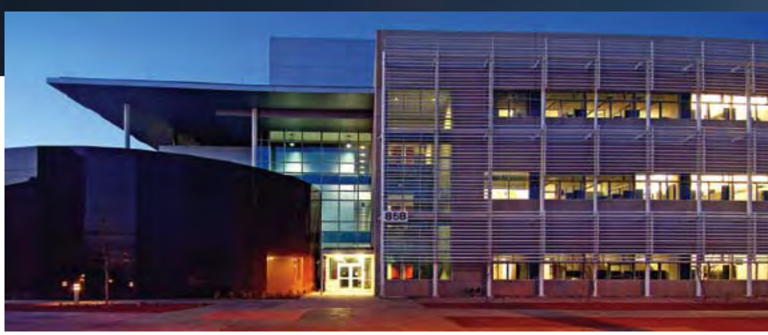

Sandia MESA Facility

Sandia Albuquerque

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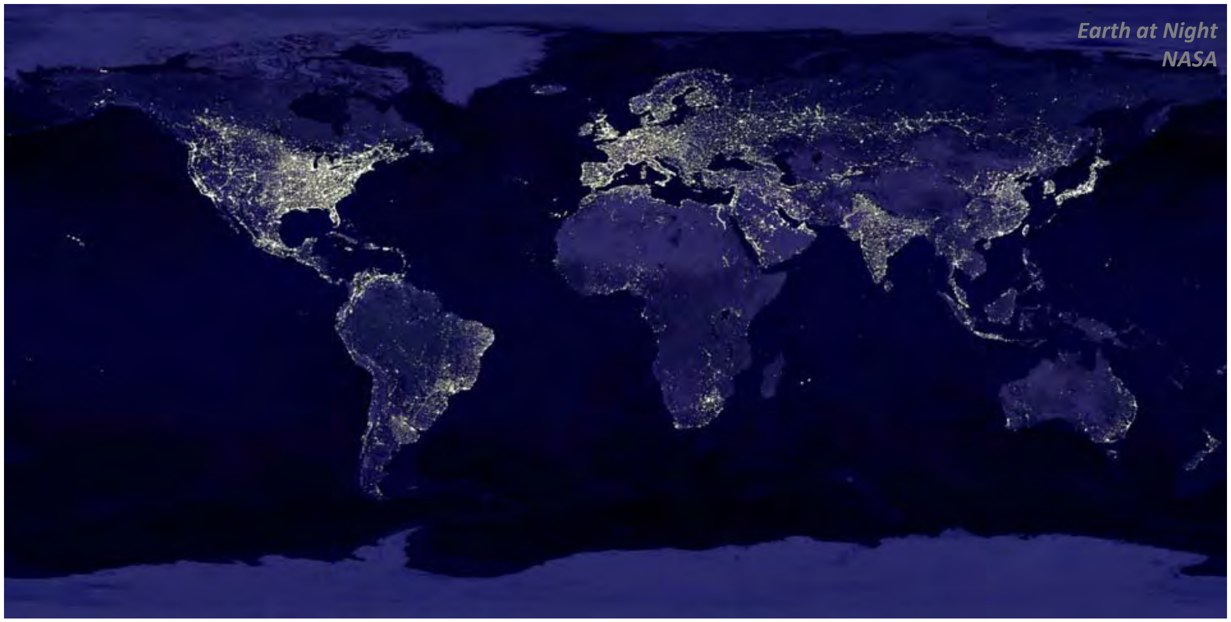
Lighting is a large fraction of energy consumption and is low efficiency



Efficiencies of energy technologies in buildings:

Heating:	70 - 80%
Elect. motors:	85 - 95%
Fluorescent:	**~17%**
Incandescent:	**~4%**

Lighting is one of the most *inefficient buliding* energy technologies → opportunity!

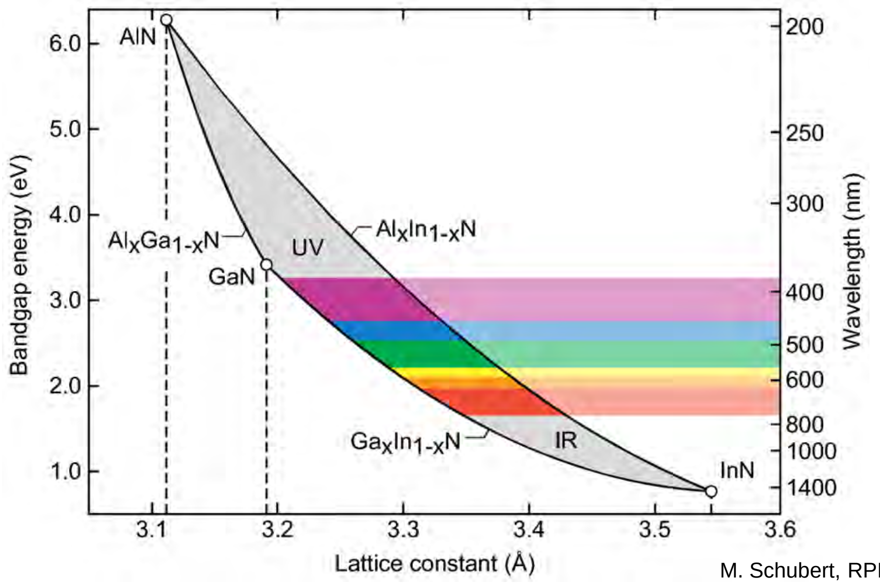
- ~22% of US electricity consumption is for general illumination (~1/15 world's energy, $330B in 2005)
- Achieving 50% efficient lighting would have tremendous global impact:

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

- **decrease electricity consumed by lighting by > 50%**
- **decrease total electricity consumption by 10%**

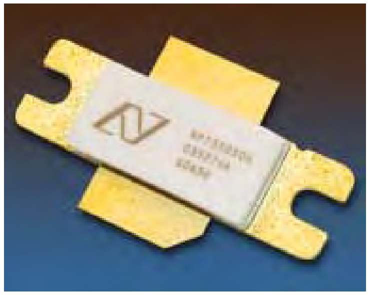
Foundation of SSL: III-Nitride (AlGaInN) Semiconductors

- **Direct RT bandgaps: ~0.7-6.2 ev**
- Solid alloy system (tuneable bandgaps)
- High breakdown field, mobility, thermal conductivity, melting temperature
- Radiation resistant and chemically inert
- InGaN covers entire visible & bulk of solar spectrum (PV material?)



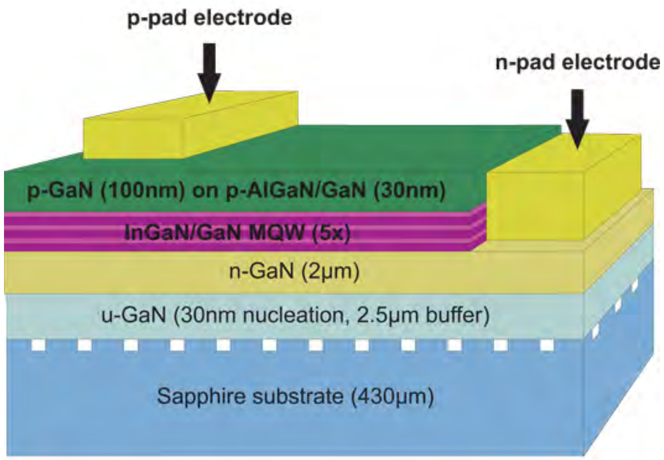
M. Schubert, RPI

- *Used in LEDs, blue laser diodes, high power transistors, HEMTs*

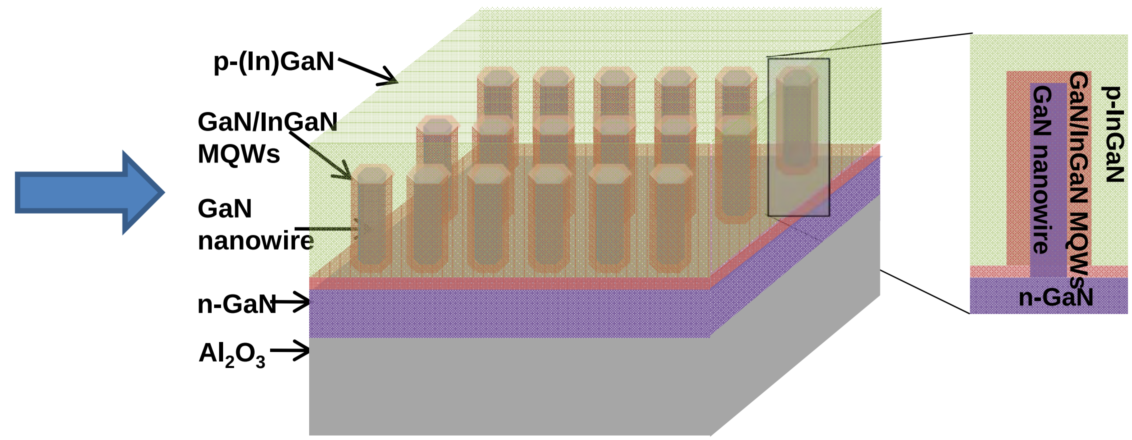


Nitronex GaN power transistor

Why III-nitride (AlGaInN) nanowires for SSL?



Y.S. Lin and J. A. Yeh, Appl. Phys. Express, vol4, p092103, 2011

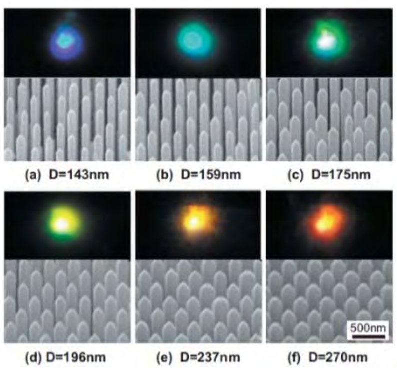


Why III-nitride (AlGaInN) nanowires for SSL?

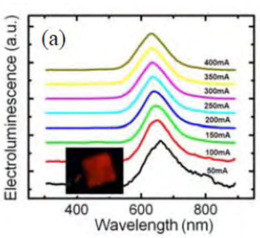
Advantages due to enhanced strain accommodation in nanowires

elastic strain relaxation at surface

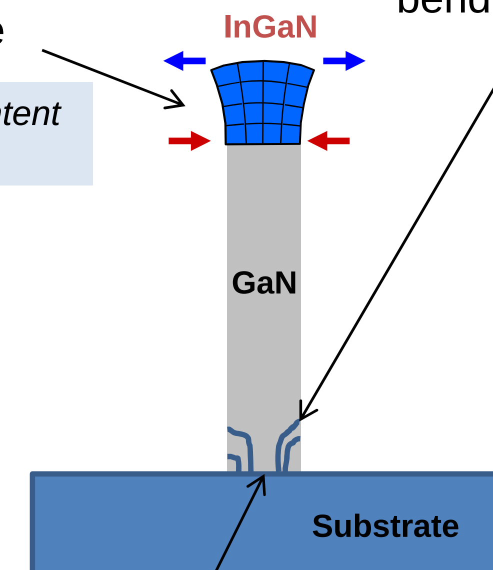
benefit: *heterostructures with high In content (e.g. green-yellow-red gap)*



Sekiguchi et al., APL **96**, 231104 (2010) – Sophia U.

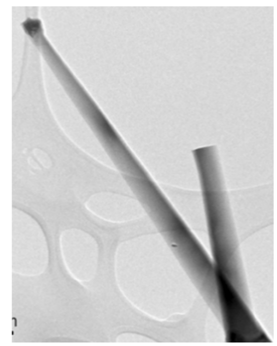


Nguyen et al., PTL IEEE **24**, 321 (2012) - McGill

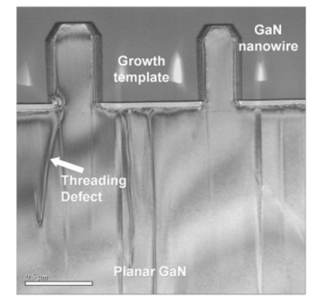


bending of dislocations (TDs) toward surface

benefit: *reduced TDs, higher IQE*



VLS-grown TD-free GaN NWs - Sandia

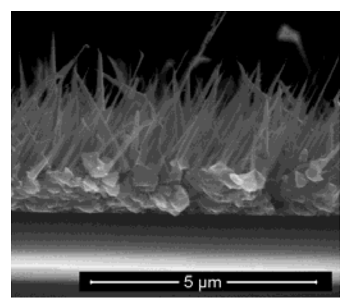


Hersee et al., J. Mat. Res. **17**, 2293 (2011) - UNM

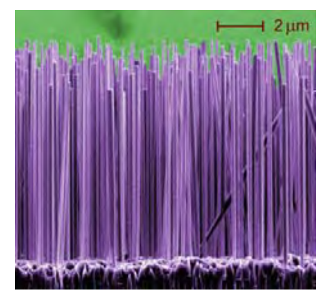
Bending & termination of TDs at nanowire base

small interfacial area

benefit: can grow on cheaper, lattice mismatched substrates; integration with Si devices



GaN NWs on tungsten foil - Sandia

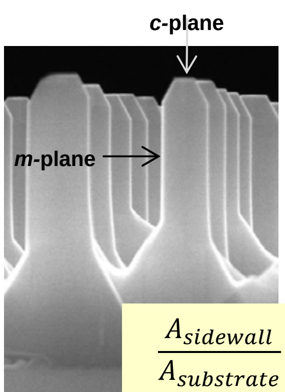


GaN NWs on Si - NIST

Why III-nitride (AlGaInN) nanowires for SSL?

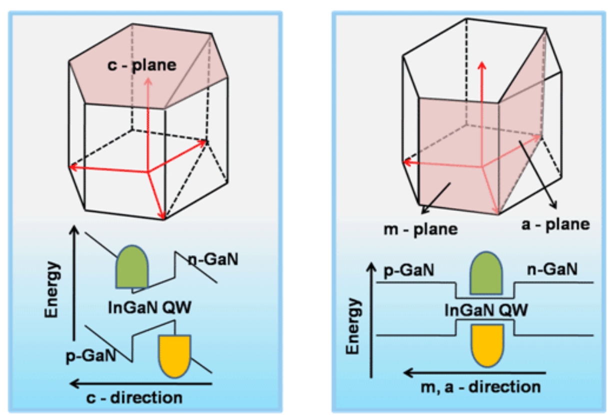
Size, geometry (applies to the first four rows)

Nanowire attribute	Benefit
Vertical 3D device integration (radial)	High surface/device area = lower cost;
Non-polar, semi-polar facets	Reduced polarization effects (IQE)
2D arrangements, e.g. photonic crystals	Enhanced/controlled light extraction, wavelength tuning, rad. recombination (IQE)
Nanolasers	Lower threshold lasers; reduced eff. droop
Discrete; (and all of the above)	Scientific test system



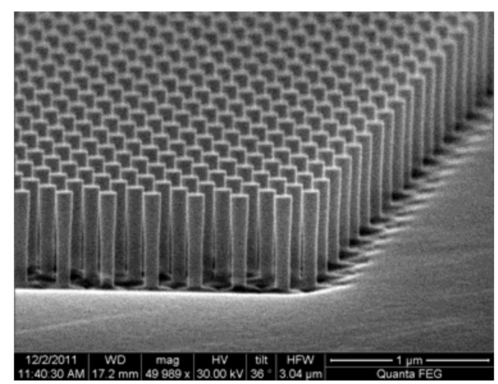
Radial GaN/InGaN MQW NW

$$\frac{A_{sidewall}}{A_{substrate}} = \frac{6rh}{2.6R^2} \cong \mathbf{4.6F \cdot AR}$$



Non – polar direction GaN thin film growth → higher quantum efficiency

http://csel.snu.ac.kr/research/LED.php



GaN NW photonic crystal

Worldwide GaN-based nanowire SSL-related research

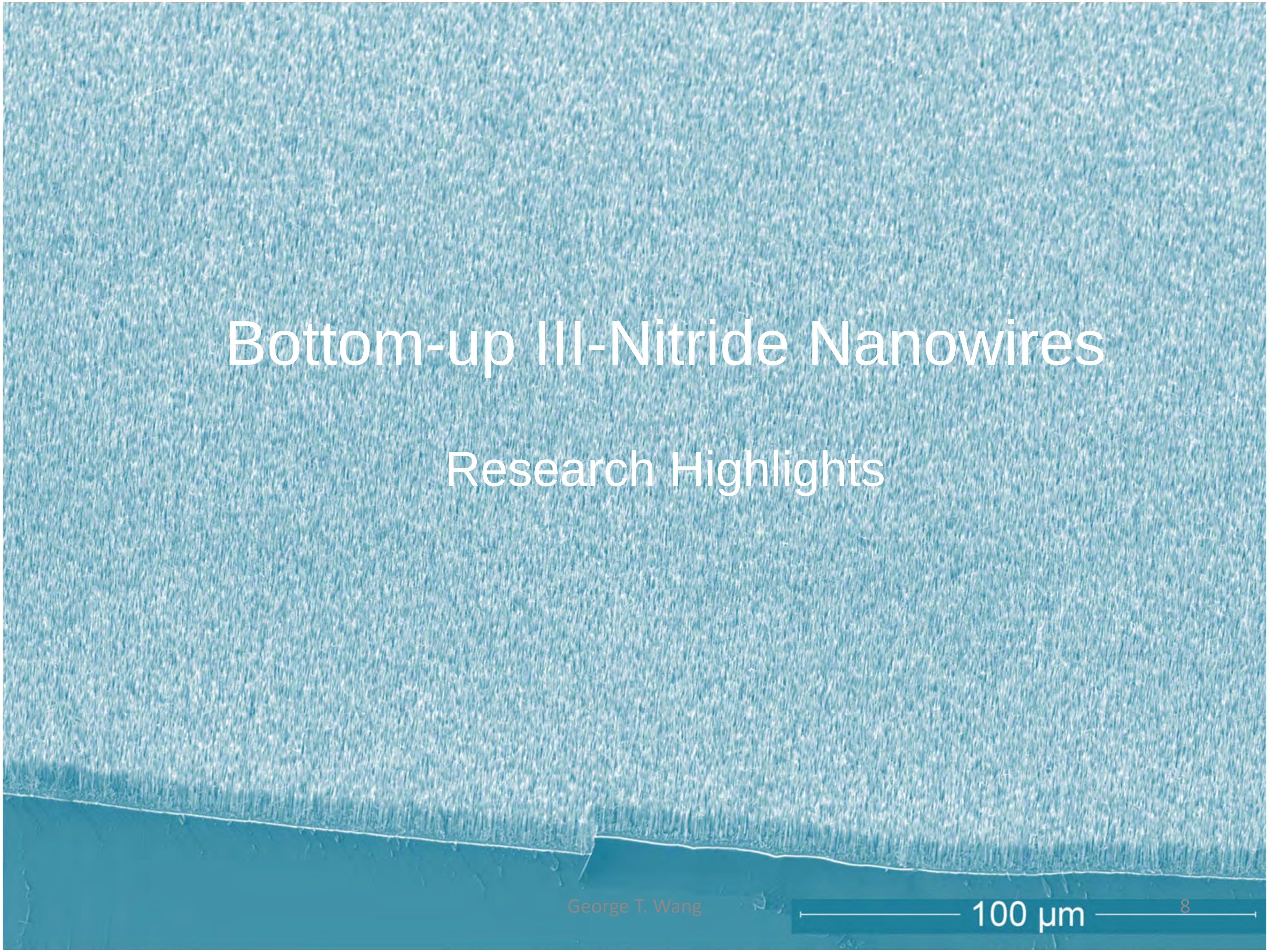
North America

Europe

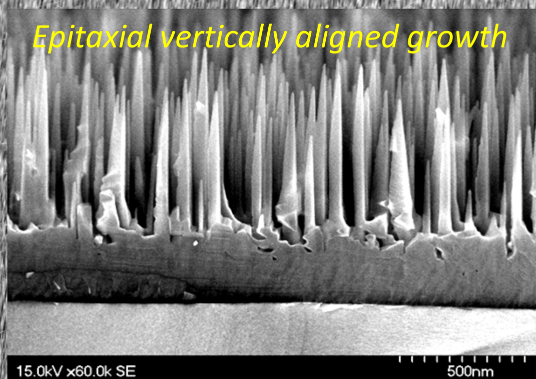
Asia

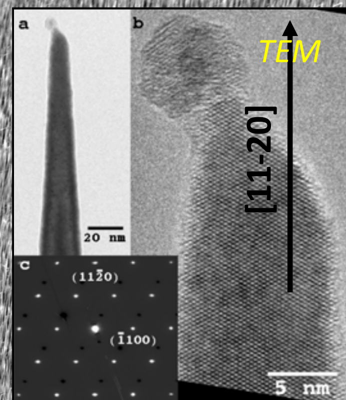
Bottom-up III-Nitride Nanowires

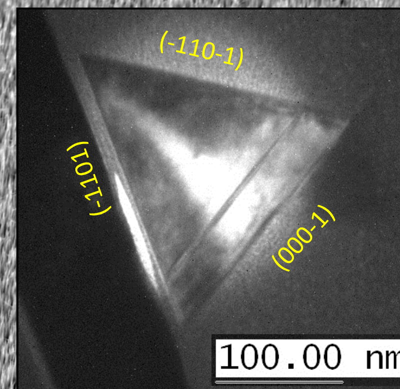
Research Highlights



"Bottom Up" aligned GaN nanowire growth



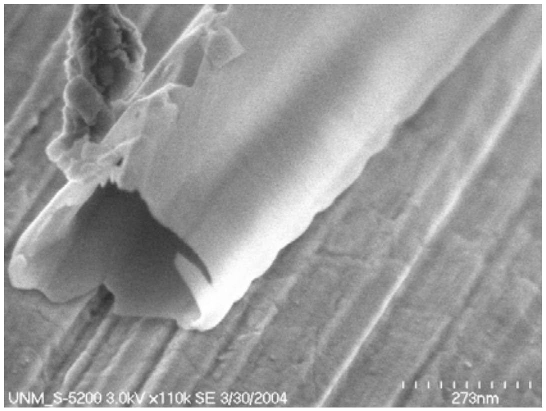




- Nanowires grown by Ni-catalyzed MOVPE/MOCVD (VLS)
- Highly-aligned vertical growth over large areas (2" r-sapphire wafer)
- Controllable densities as high as ~150 nanowires μm^{-2}
  - **Q. Li, G. T. Wang, *Appl. Phys. Lett. 93, 043119* (2008)**
  - **Q. Li, J. R. Creighton, G.T. Wang. *J. Crys. Growth 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)**

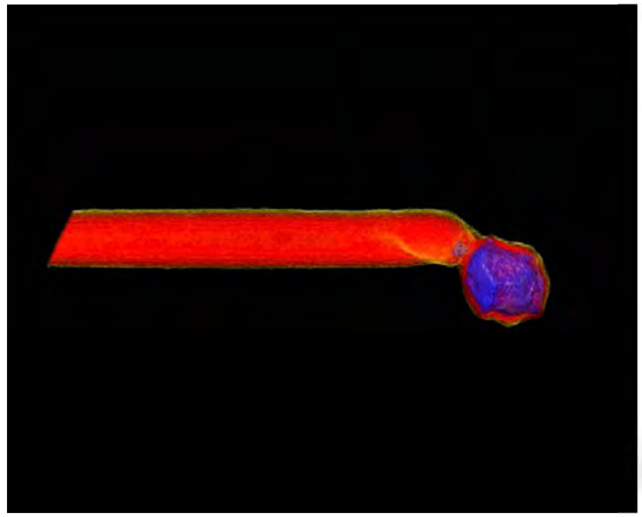
100 µm

Radial heterostructure nanowire growth

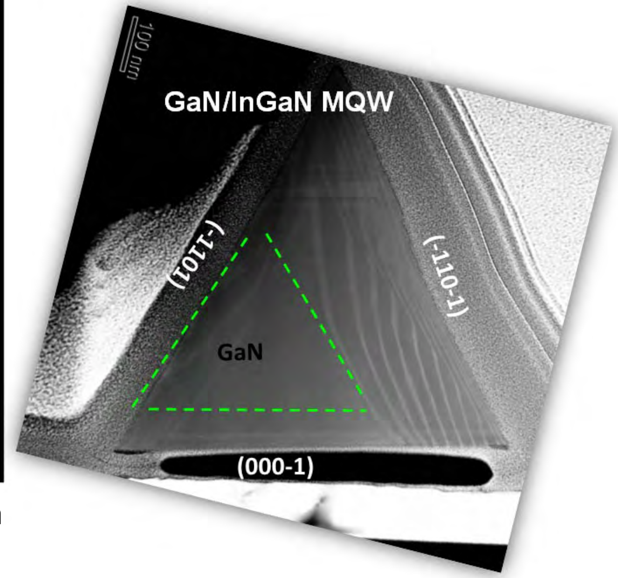


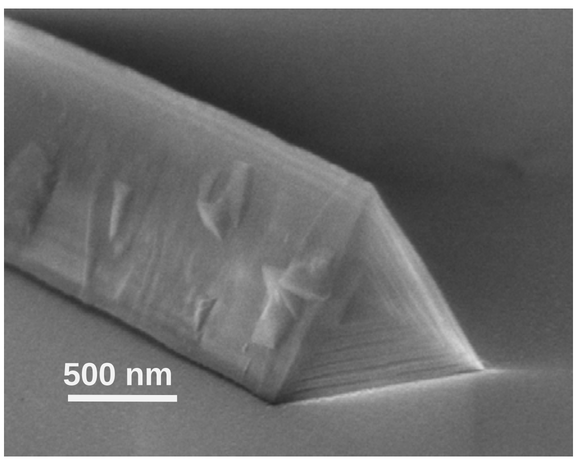
G. Stan et al. , *Nanotechnology*, ***20***, **2009**

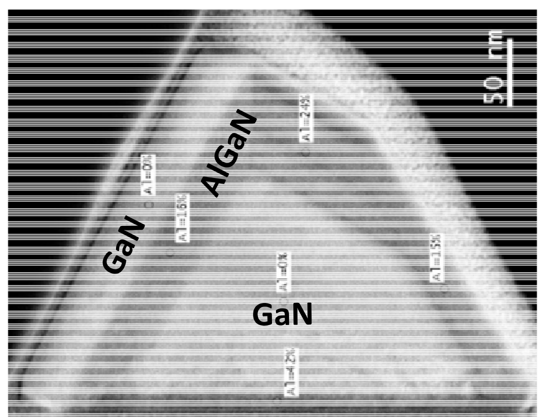
AlN Nanotubes



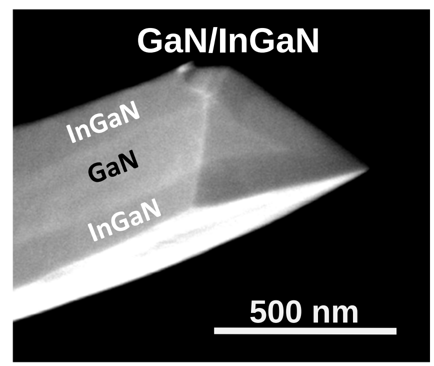
GaN/AlN NW I. Arslan



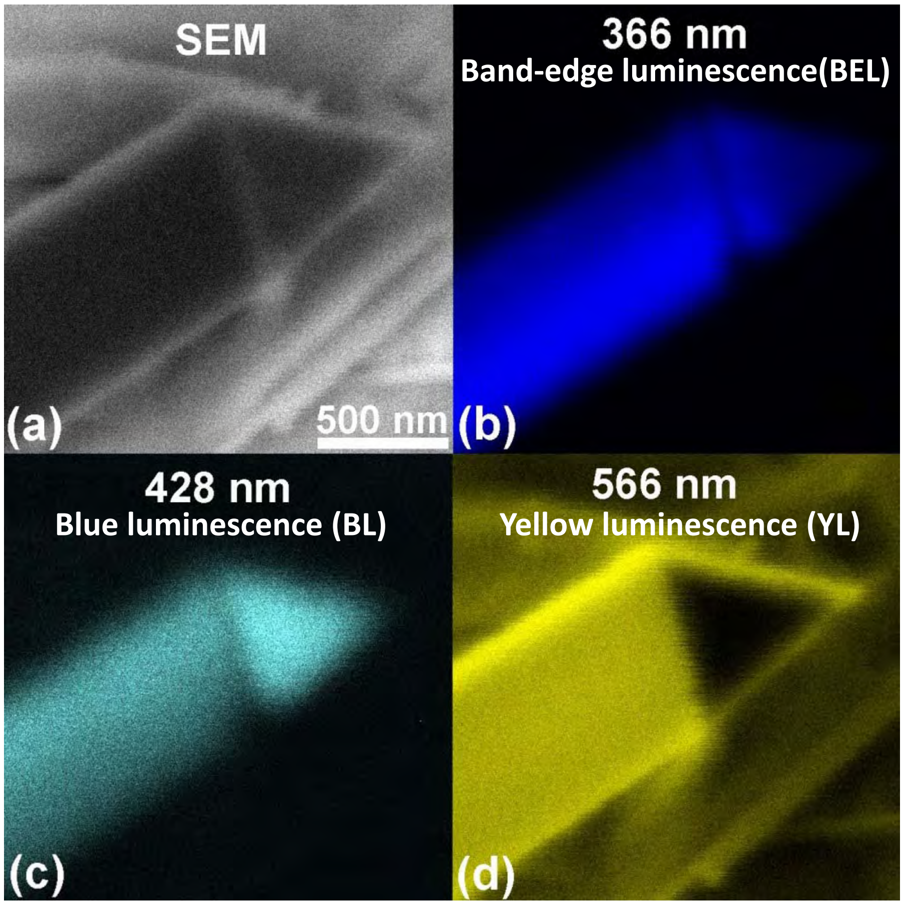




GaN/AlGaN/GaN
Double heterostructure



Spatial distribution of luminescence in GaN NWs



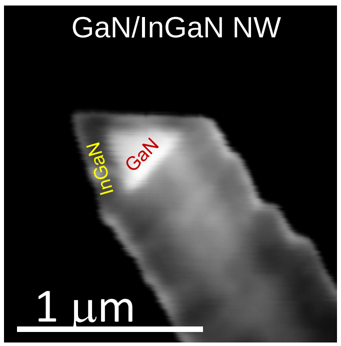
Nanoscale Cathodoluminescence (CL) imaging: **Cross-section GaN NW**

- Band-edge luminescence (BEL) at ~366 nm and defect-related blue luminescence (BL) at ~428 nm observed in NW core/bulk
- **Defect-related yellow luminescence (YL) exhibits strong surface component** -- associated with surface states or concentrated near surface region
- YL in GaN attributed to many possible sources (C, O impurities, Ga vacancies, etc.)
- Isolated Ga vacancies have low diffusion barrier (~1.5 eV) & may migrate toward surface during growth
- BL linked to V_{Ga}-O_N (D ~2.2 eV), less mobile

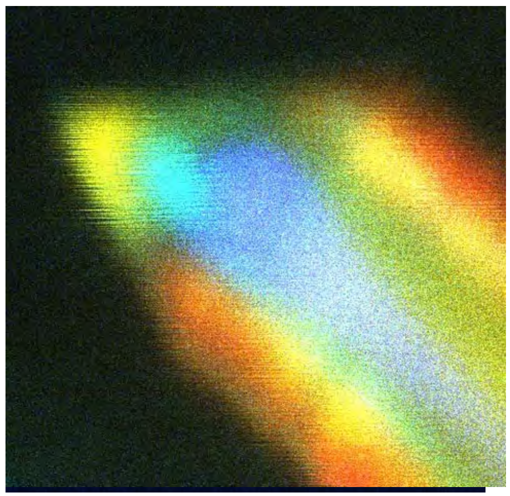
Q. Li, G. T. Wang, *Nano Lett.*, 2010, 10 (5), 1554

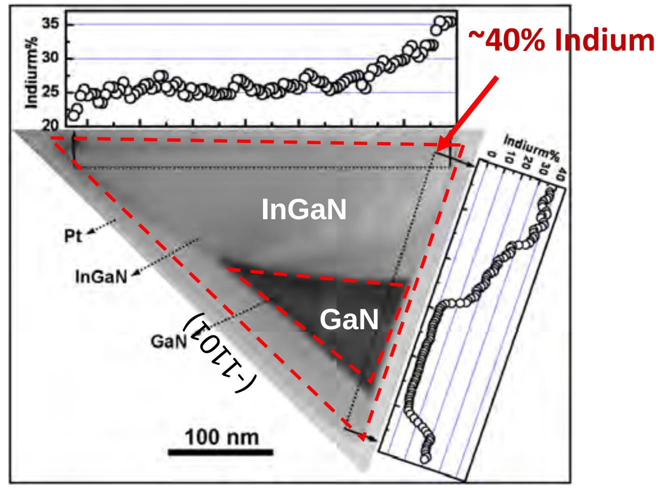
Core-shell NWs – A good platform for high In InGaN?

- ***Issue: Strain limits In incorporation in InGaN thin films (green-yellow-red gap)***
- Radial core-shell NWs: much higher active region area than axial NW or planar heterostructures



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

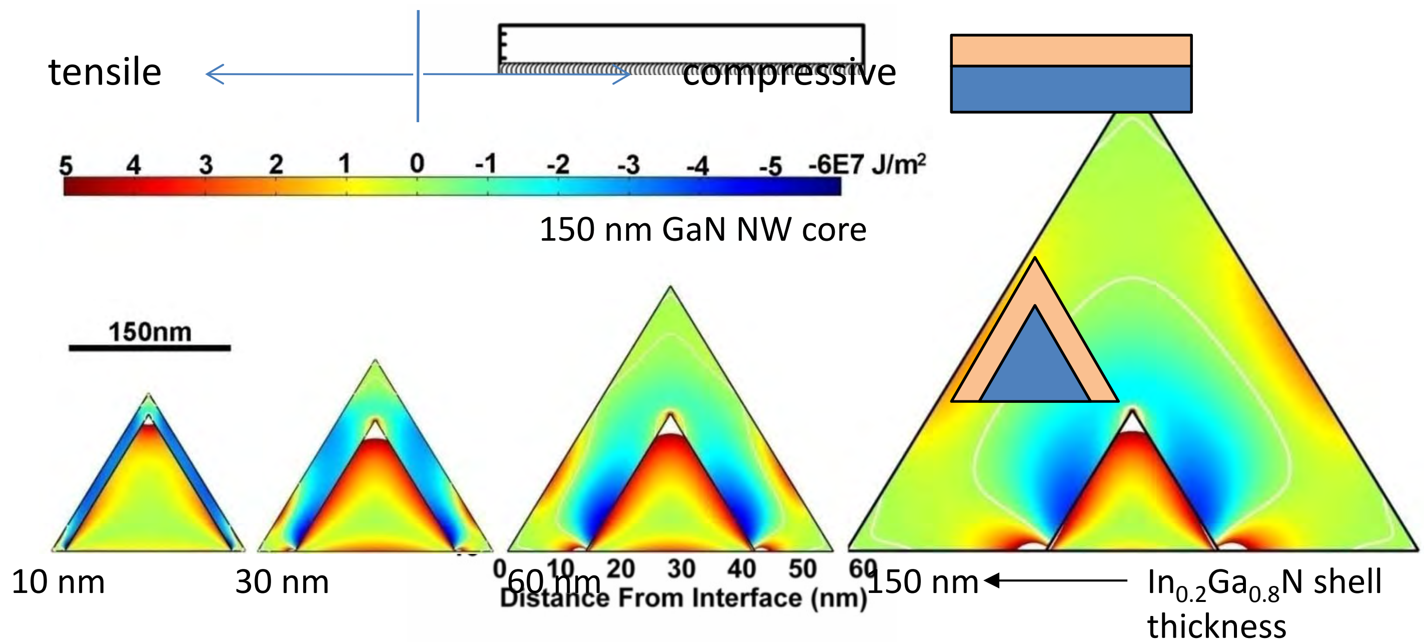




CL shows strong multicolor emission up into the IR!
- **high In incorporation with high material quality**
- nonuniform In composition?

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

Reduced strain in nanowires allows higher In incorporation

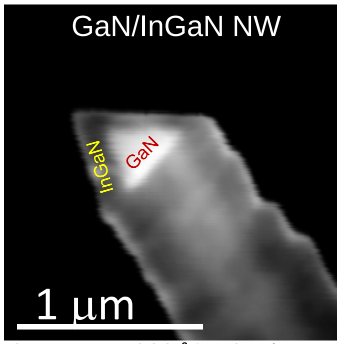


- Strain lower for GaN/InGaN nanowire than for GaN/InGaN thin film
- Compressive strain decreases away from GaN/InGaN interface, lowest in corners

Q. M. Li, G. T. Wang, "*Appl. Phys. Lett.*, ***97***, 181107 **2010**.

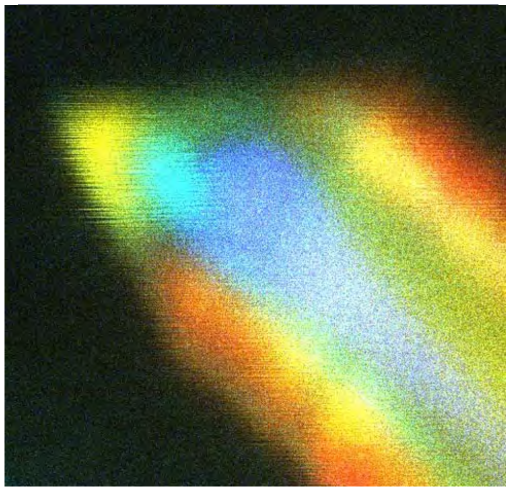
Strain-dependent In incorporation in GaN/InGaN core-shell NWs

- ***Issue: Strain limits In incorporation in InGaN thin films (green-yellow-red gap)***
- Radial core-shell NWs: much higher active region area than axial NW or planar heterostructures



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

Q. M. Li, G. T. Wang, "*Appl. Phys. Lett.*, ***97***, 181107 **2010**.



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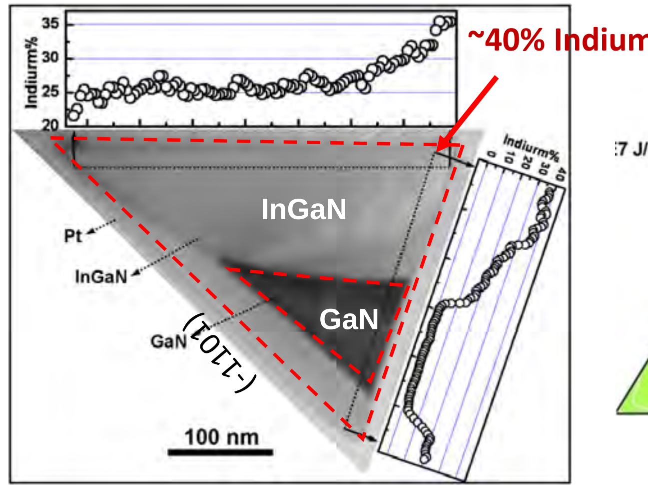
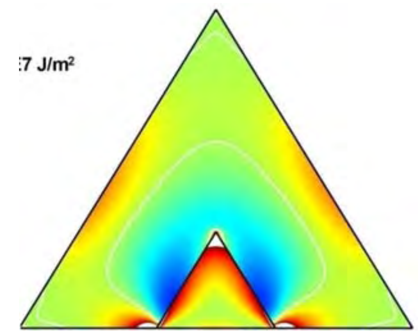


STEM/EDS shows In distribution, **highest at surface/corners**

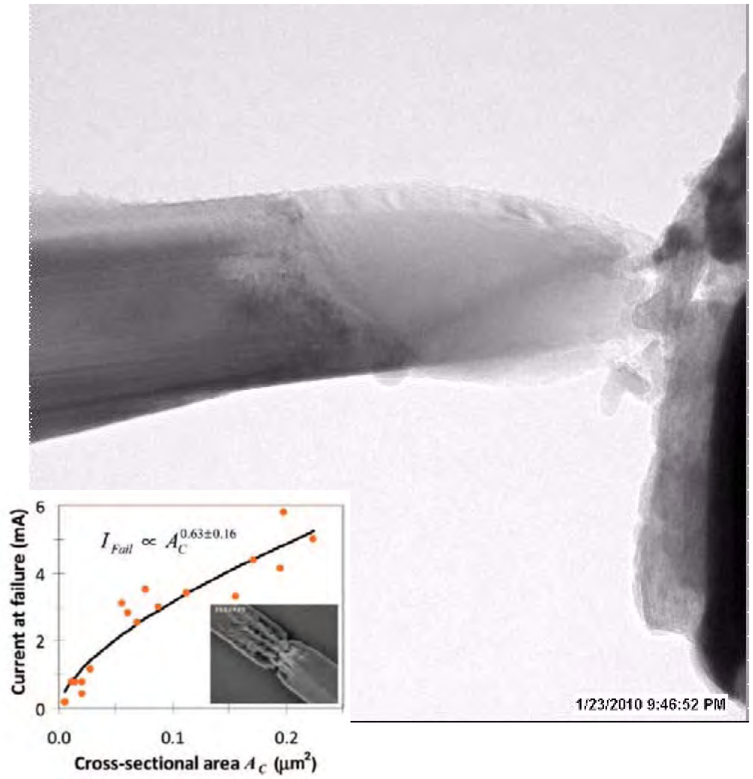


2D FEA: highest In regions correlated w/lowest compressive strain regions

Radial InGaN/GaN nanowires promising for addressing green-yellow-red gap

In-situ TEM studies

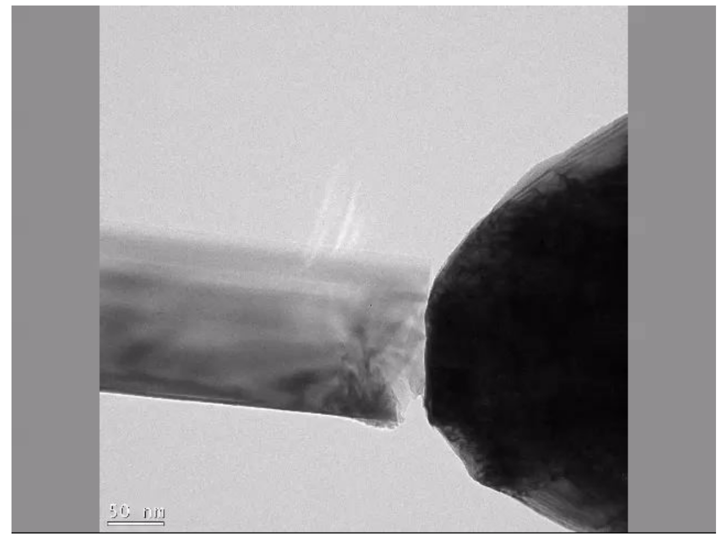
GaN NW decomposition via Joule heating
(relevant for NW devices)



NW breakdown at 60V, 20 µA
(avg. breakdown I ~3000 kW/cm^2)

T. Westover, R. Jones, J. Y. Huang, G. Wang, E. Lai, A. A. Talin, *Nano Lett.*, **9**, 257 (2009).

Local plastic deformation of GaN NW
(relevant for NW devices)

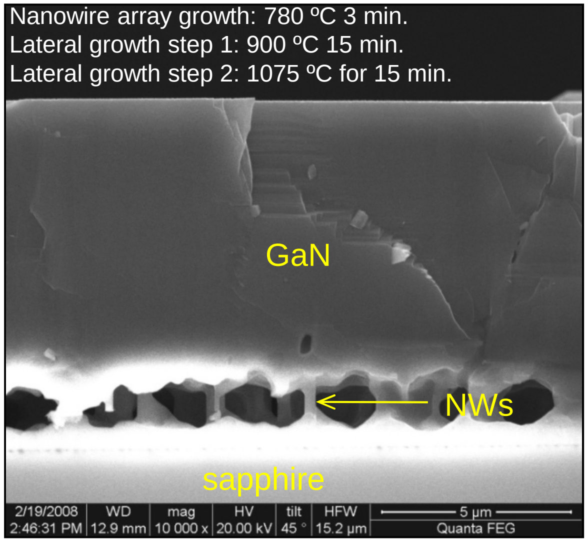


- Dislocation-free NW shows significant surface plastic deformation
- Mediated by dislocation nucleation & pile up, grain boundary sliding

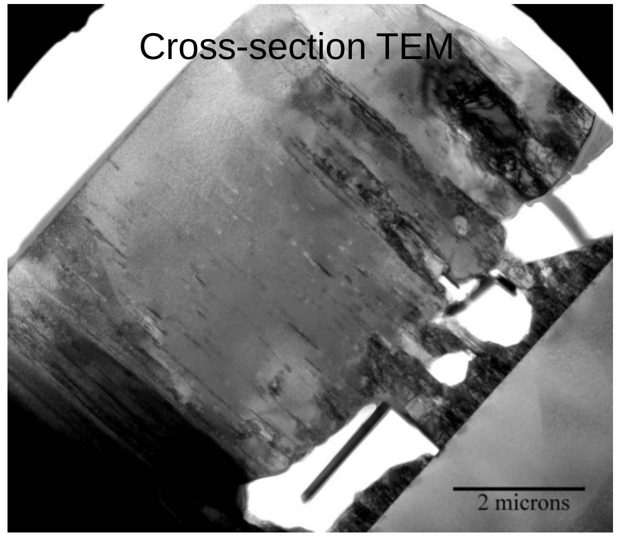
J. Y. Huang, H. Zheng, S. X. Mao, Q. Li, and G. T. Wang, *Nano Lett*, **11 (4),** 1618 (2011).

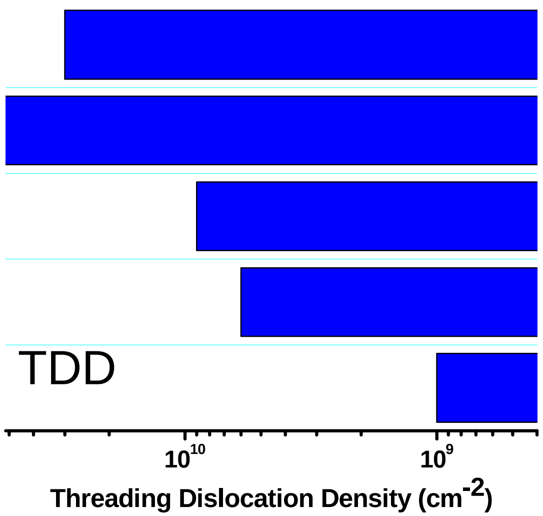
Nanowire-templated lateral epitaxial growth (NTLEG) of GaN

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



Cross-section SEM





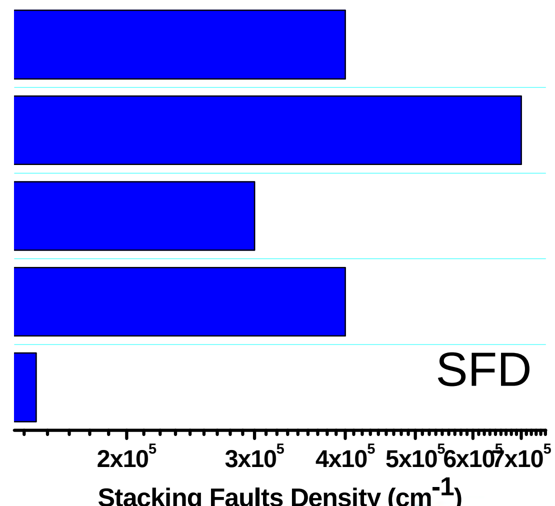
Craven et al., *APL* 81(2002), 469 (LT nucl. Layer)

Chakraborty et al. *APL* 89(2006), 041903 (LT nucl. Layer)

Chakraborty et al. *APL* 89(2006), 041903 (SiNx Nanomask)

Qian et al, *JAP* 106(2009), 123519 (HT AlN nucl. Layer and 3-step growth)

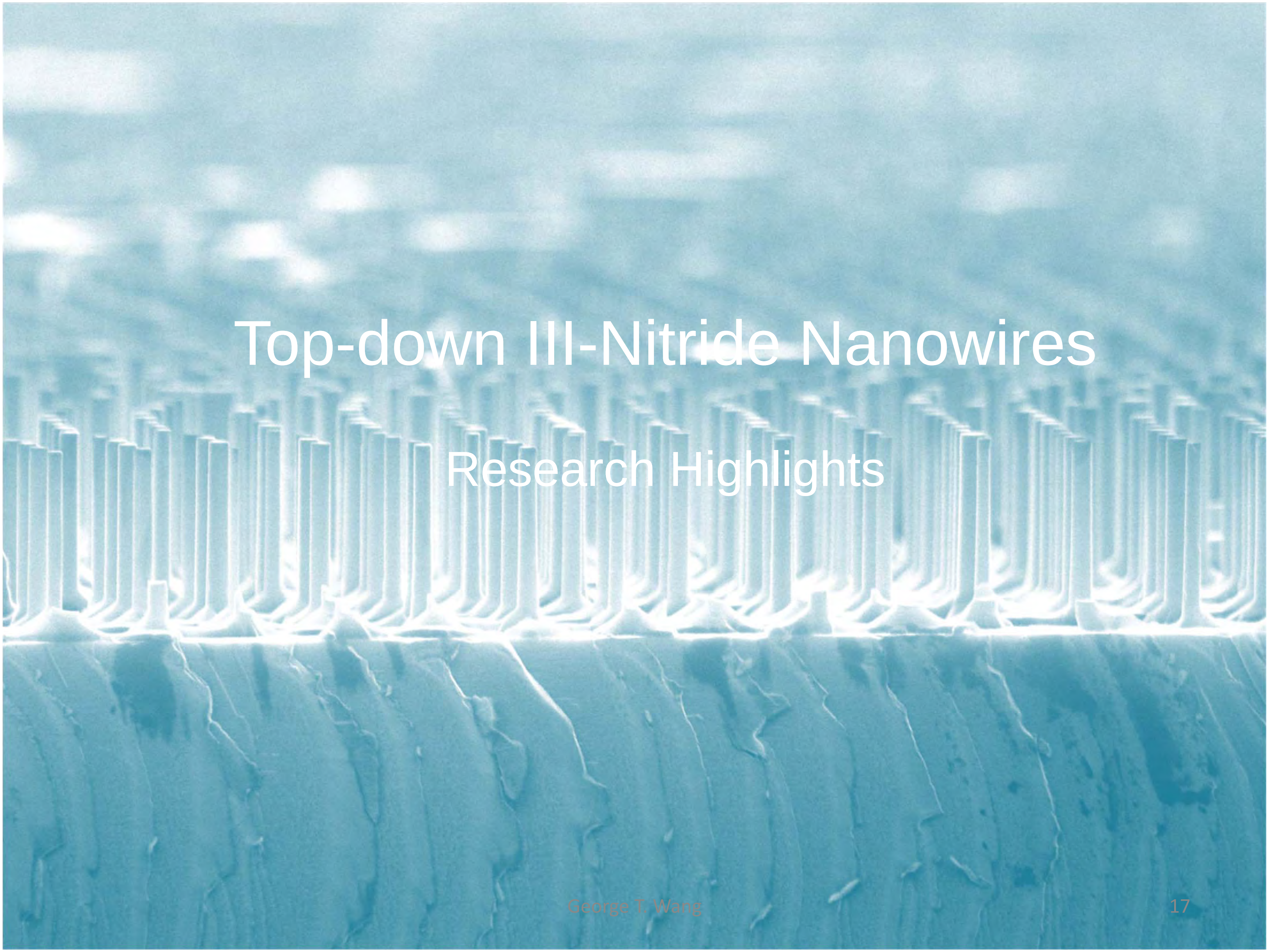
Li et al. *Adv. Mat.*, 21(2009), 2416 (NTLEG) THIS WORK



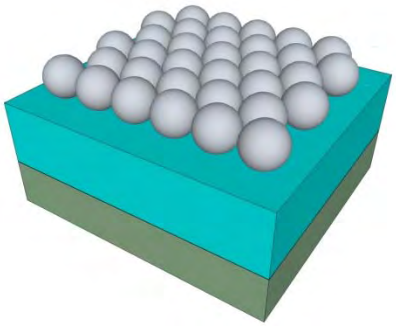
Q. Li, Y. Lin, J. R. Creighton, J. J. Figiel, and G. T. Wang, *Adv. Mat.*, **21**, 2416–2420 (2009)

Top-down III-Nitride Nanowires

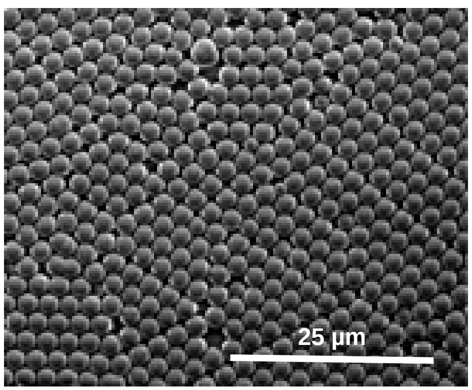
Research Highlights



New dry + wet top-down ordered nanowire fabrication process

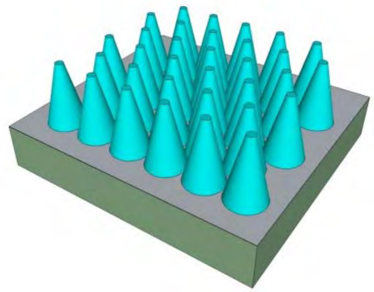


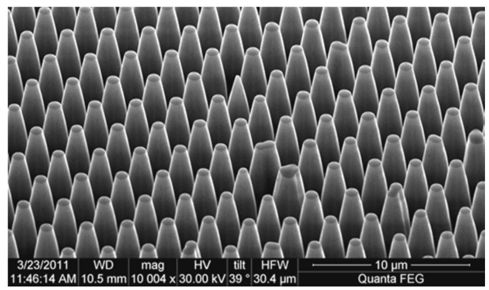
(0001) GaN on sapphire

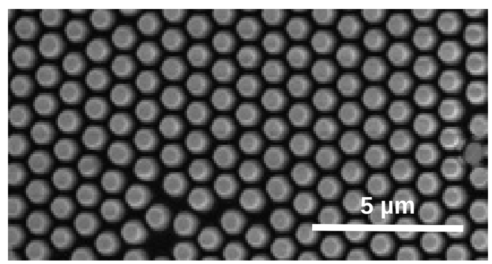


Q. Li, J. J. Figiel, G. T. Wang, Appl. Phys. Lett., **94**, 231105 (2009).

ICP etch







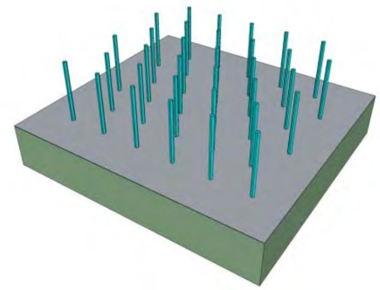
Plasma etch causes sidewall damage

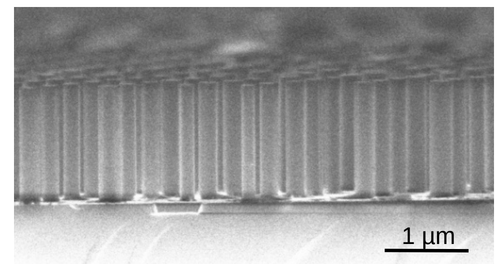
C. Y. Wang et al., Opt. Expr. *16,* 10549–10556, 2008.

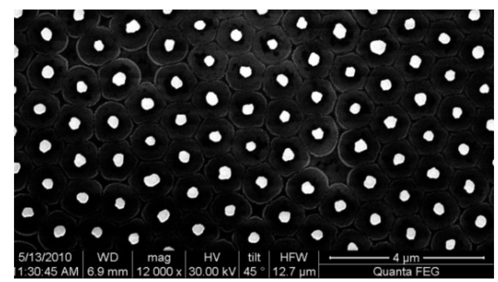
Tapered; no well-defined facets

Selective wet etch

(AZ-400K developer)





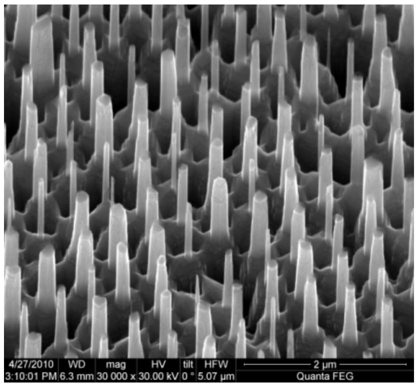


Wet etch: straight sidewalls, removes sidewall damage

Straight GaN nanowires with controllable geometries

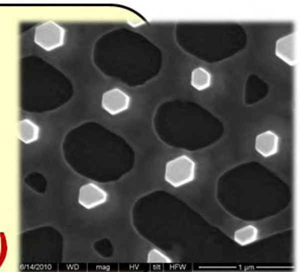
Wet etch rate negligible for top (Ga-polar) *c*-face & fast for [10-10], leads to hexagonal NWs with **straight & smooth** *m*-facets

0.5 µm sphere size

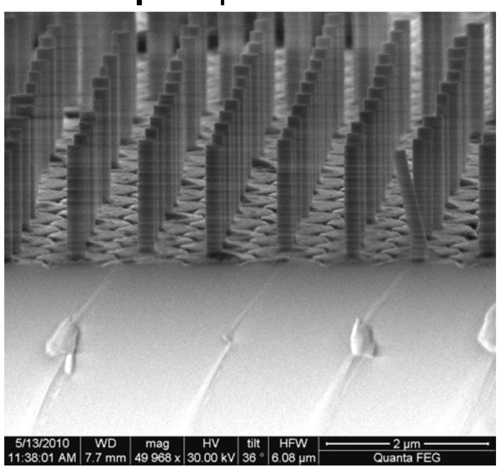


Superior and independent control of:

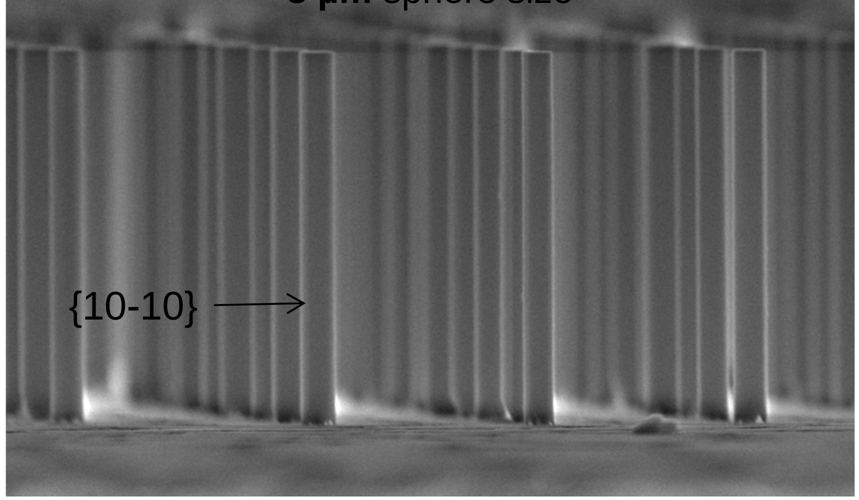
- ***Height (dry etch depth)***
- ***Diameter (wet etch time)***
- ***Pitch/arrangement (defined by masking template)***

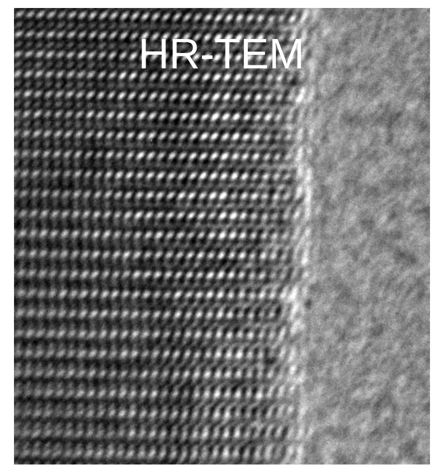


1 µm sphere size



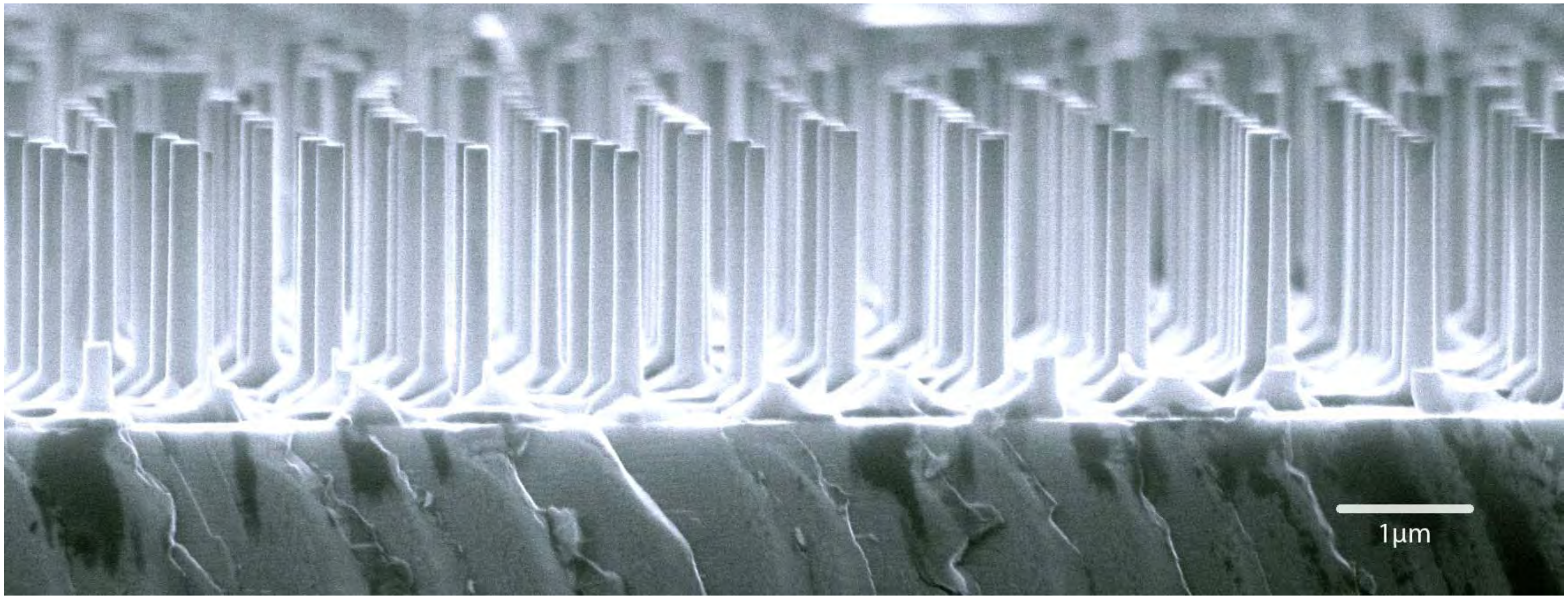
3 µm sphere size





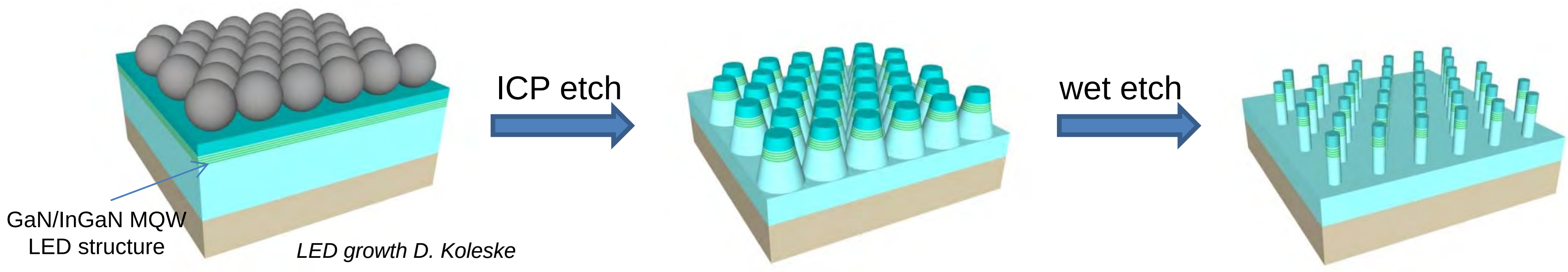
Smooth sidewall created by wet etch

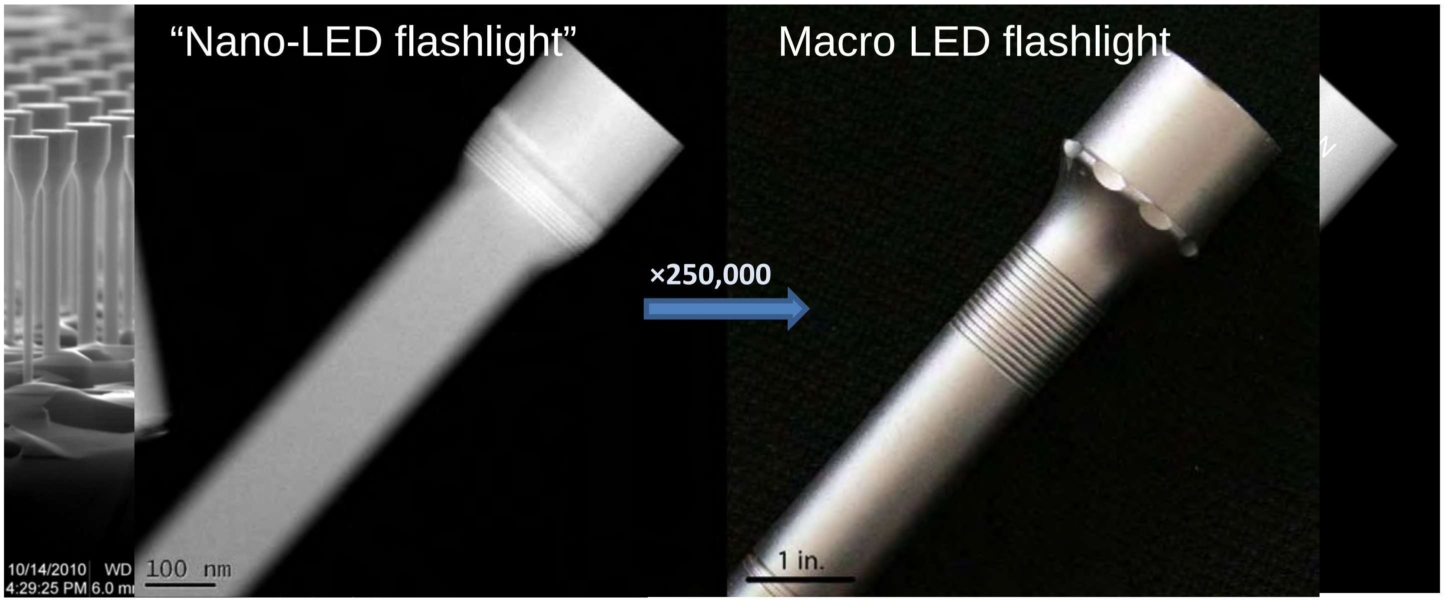
Advantages of new top-down nanowire fabrication method



- Wider range of growth conditions, material tunability (1D growth cndns not needed)
- Lower point defect densities (higher growth temperature)
- Ordered/periodic arrays (difficult with catalyst/VLS-based methods)
- Axial III-nitride nanowire heterostructures possible by MOCVD
- Better control of geometry (independent control over height, width, & pitch)
- Improved uniformity
- Easier vertical device integration (height uniformity, base GaN-layer)

Axial GaN/InGaN nanowire LEDs

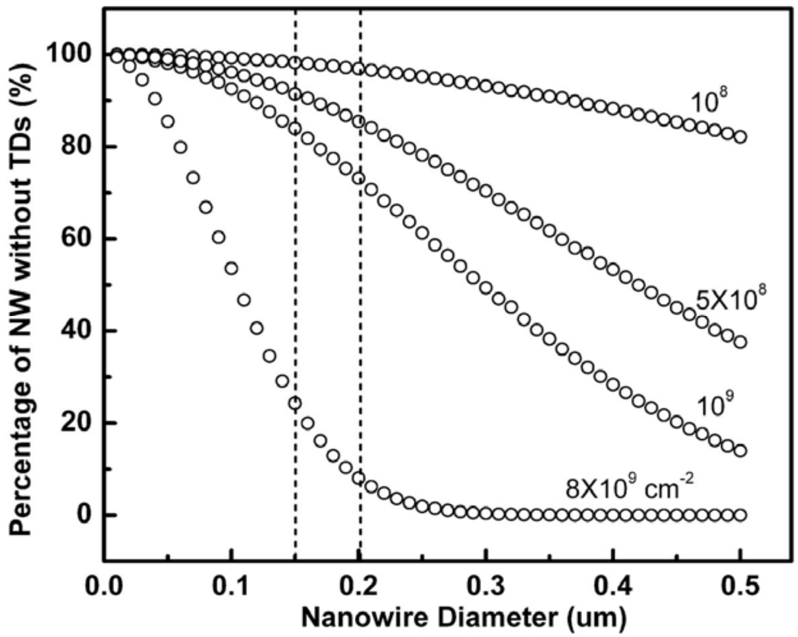




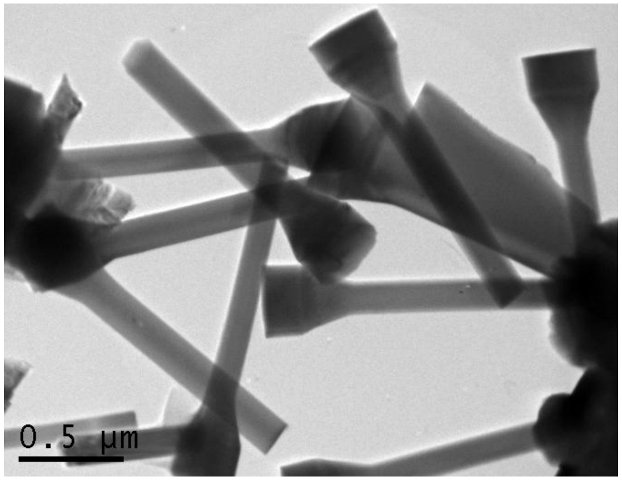
Q. Li et al., *Optics Express* **19**, 25528 (2011)

Top-down nanowire threading dislocations

Monte Carlo simulation



Bright-field TEM



Nanowires etched from ~5e8 cm^{-2} planar LED

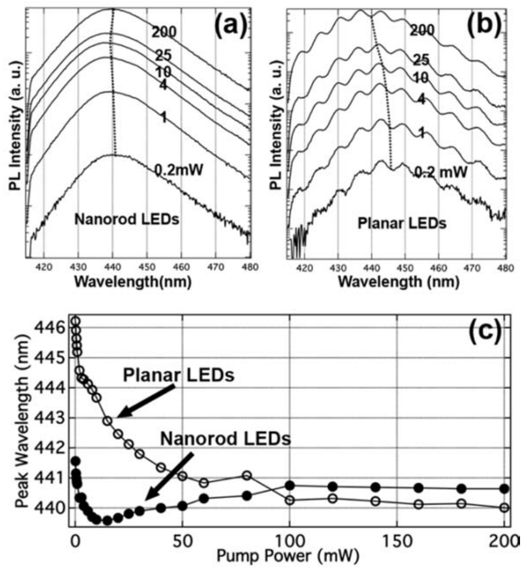
- Etched nanowires inherit the dislocation density of the parent film
- However, as the diameter approaches zero, the *fraction* of nanowires with one or more dislocations also approaches zero! [# TDs per rod ~ (TDD)×($A_{cross\text{-}section}$)]
- *~94% of nanowires ~150 nm in diameter from TDD~5e8 cm^{-2} film dislocation free!*

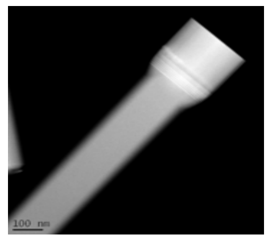
Optical performance – axial nanowire LEDs vs. planar LED

413 nm pump (InGaN selective)

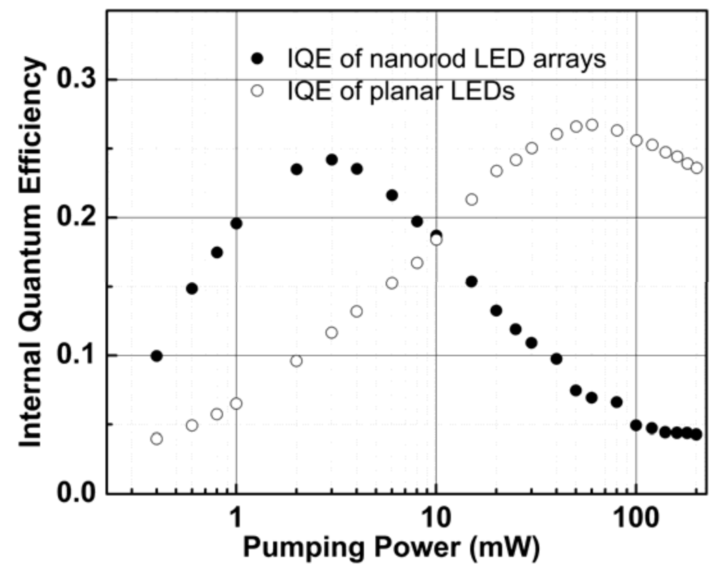
PL, IQE measurement K. Westlake, M. Crawford

InGaN peak position vs pump power





IQE – nanowire vs Film



- XRD shows ~16±4% strain reduction in InGaN QWs in nanowire LEDs

  XRD measurement courtesy Steve Lee

- Little wavelength shift at higher pump powers for nanowire LEDs (no/reduced QCSE)

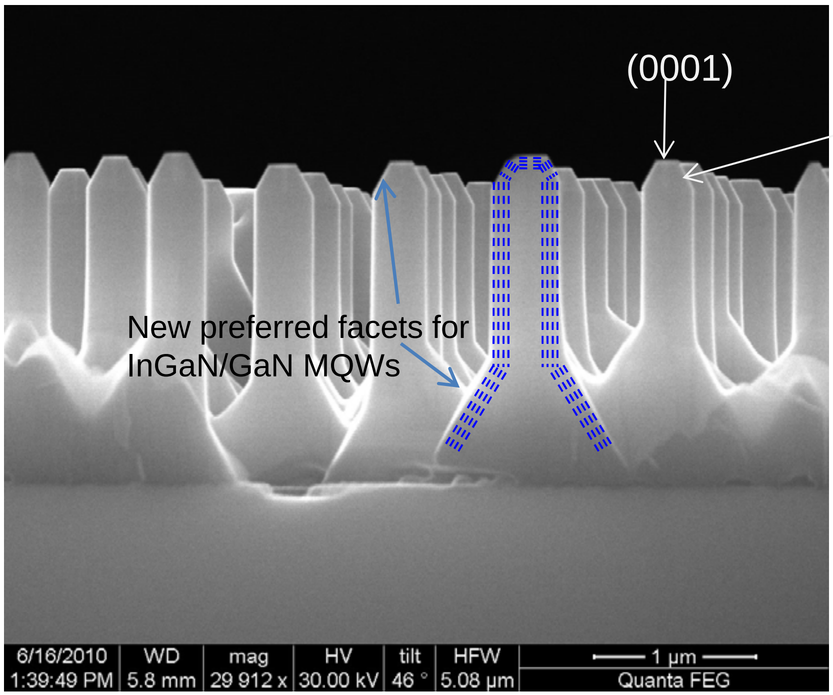
- nanowire LED: comparable IQE to planar LED but peak IQE occurs at much lower pumping power (enhanced light absorption, heating)

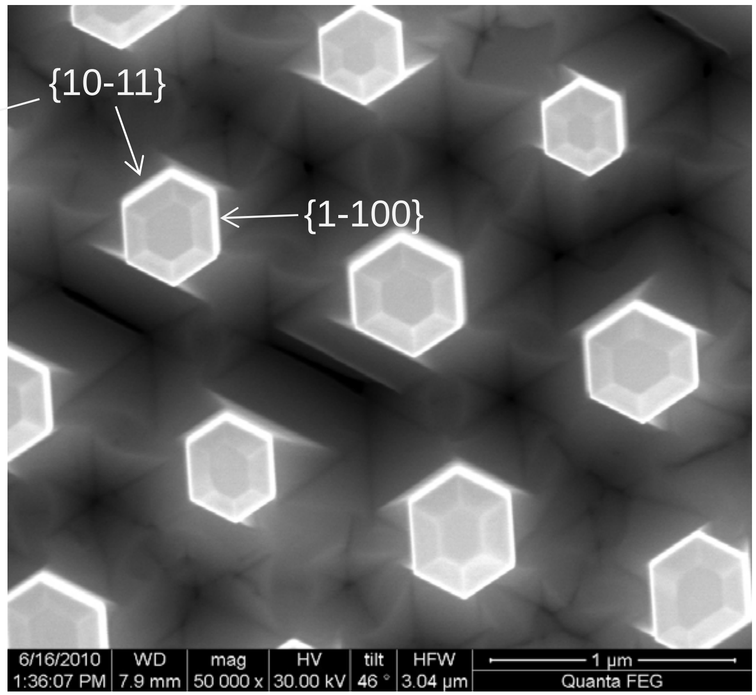
Q. Li et al., *Optics Express* **19**, 25528 (2011)

Radial core-shell InGaN/GaN MQWs on top-down NWs

Radial core-shell heterostructures
- Much higher active area than axial or planar structures
- Reduced strain InGaN growth for higher In incorporation

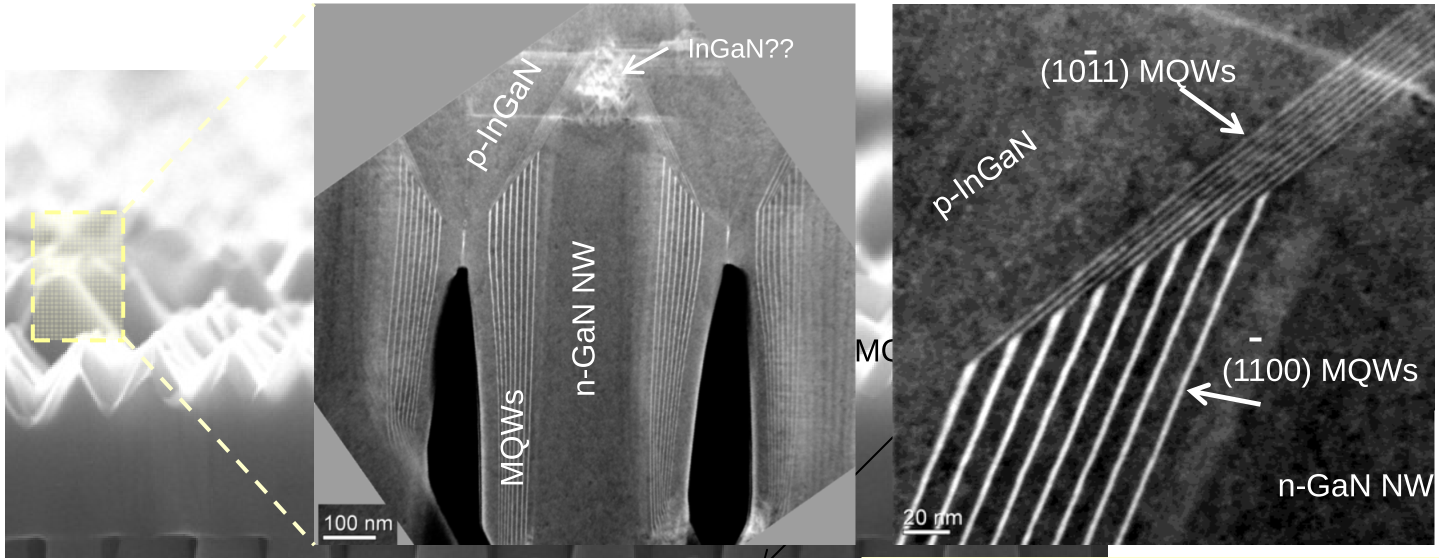
After 5-period MQW GaN/InGaN shell growth



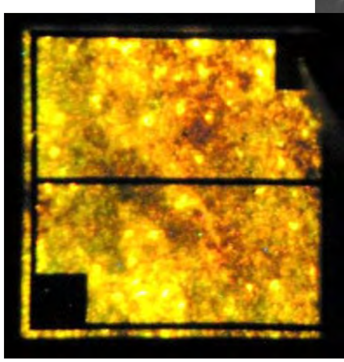


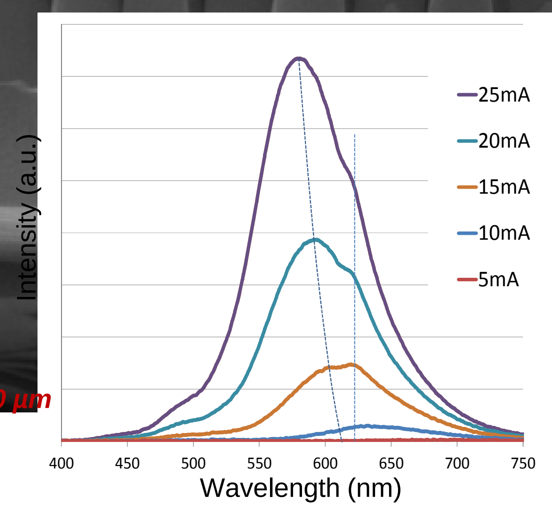
New semipolar facets form with InGaN/GaN MQW growth

Highlight: Electrically injected core-shell nanowire based “3D” LED emitting at yellow-red wavelengths



J. Wierer et al., *Nanotechnology* **23** 194007 (2012)





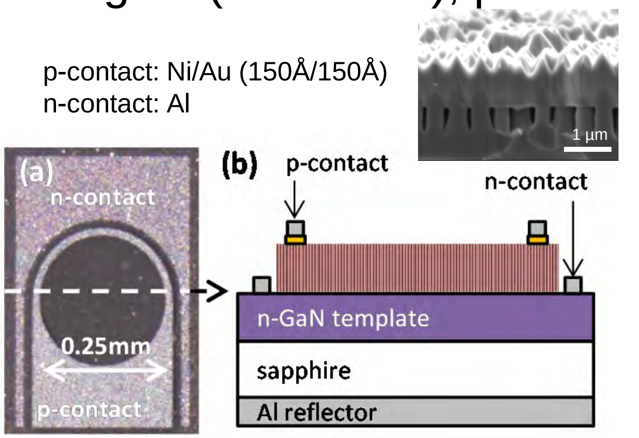
- Yellow-red electroluminescence!
- Two EL peaks:
  - 615 nm (const) red
  - 600-565 nm (shift) yellow

SSLS EFRC

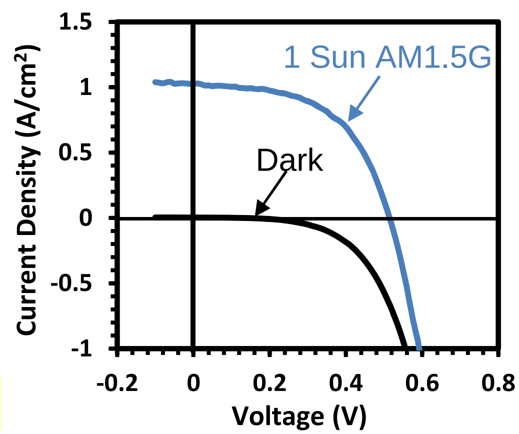
III-nitride nanowire arrayed solar cell

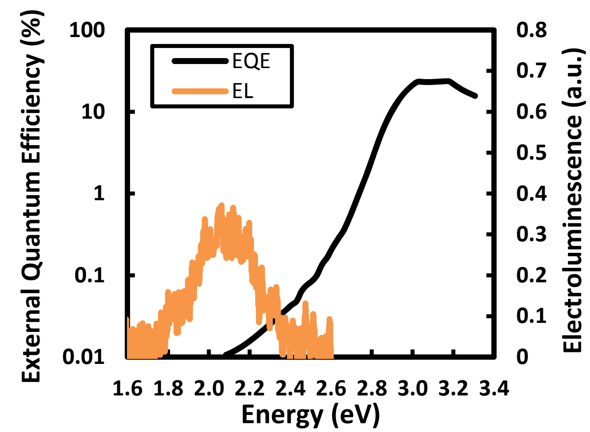
- <u>III-nitride solar cells</u>: InGaN bandgap (0.7-3.4 eV) covers solar spectrum; high rad. resistance

- <u>Nanowire solar cells</u>: increased light scattering/absorption, short carrier collection lengths (core-shell), potentially smaller bandgap cell (higher In content InGaN layers)



J. Wierer et al., *Nanotechnology* **23** 194007 (2012)





- *First vertically integrated III-nitride nanowire solar cell*

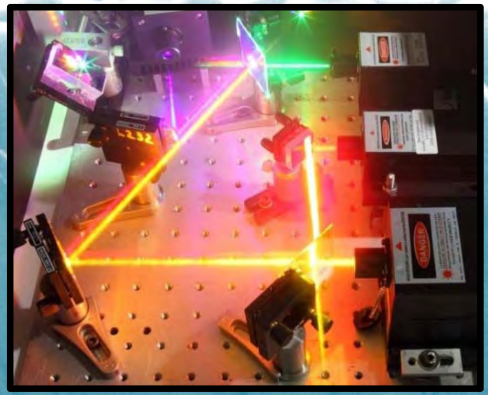
Previous Work: Single III-nitride NW solar cell:
Dong, Y. et al., *Nano Lett.* **9** 2183 (2009)

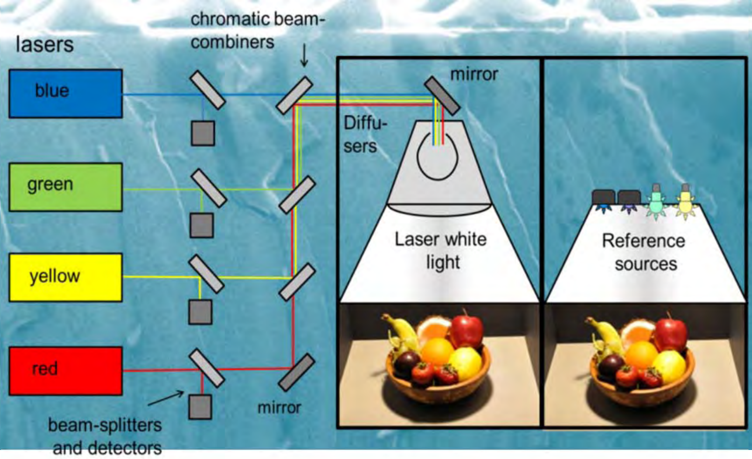
- Peak EQE ~23% at 3.0 eV; Photoresponse to 2.1 eV (590 nm), lowest bandgap reported for III-nitride solar cell; V_{OC} ~ 0.5 V; FF ~ 54%; Power conversion efficiency ~0.3% (shorting from defects in nanowire templates)

Nanowire Lasers

Why Nanowire Lasers?

- *Nanowire forms a freestanding, low loss optical cavity*
- *Compact and low power due to small mode volume*
- *Possibility of high efficiency lasers at green and yellow wavelengths*
- *Potential applications including electronic/optical integration, sensing, imaging, lithography, lighting*
- *Lasers may circumvent droop problem in LEDs*



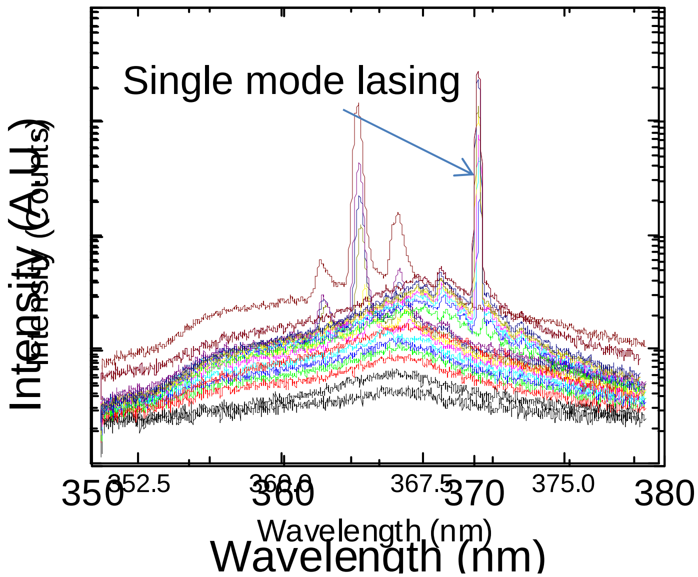


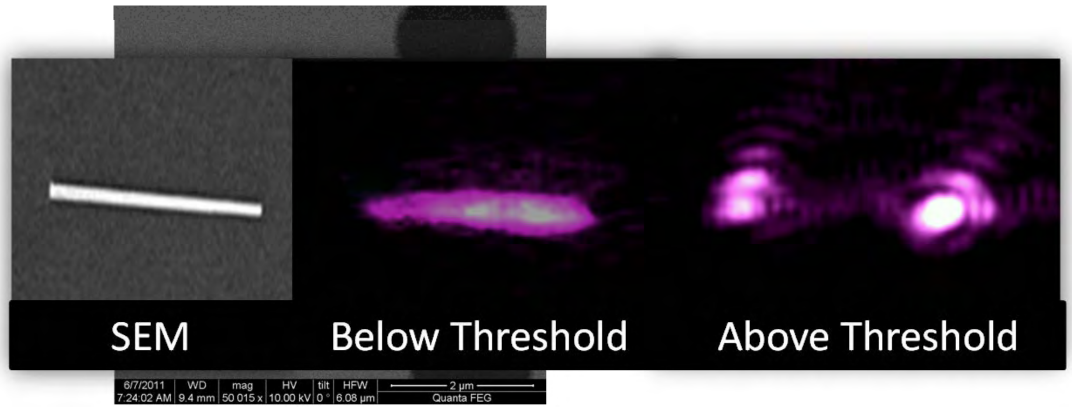
A. Neumann et al, *Optics Express*, **19**, A982-A990 (2011)

Mode control (i.e. How to make a single-mode GaN nanowire laser)

Method 1: Single-mode GaN nanowire laser via geometry control

- Single-mode lasers are desirable (higher resolution, lower threshold & noise)





Nanowire dimensions: ~**130nm** x 4.7µm
Nanowire dimensions: ~**500 nm** x 4.7µm

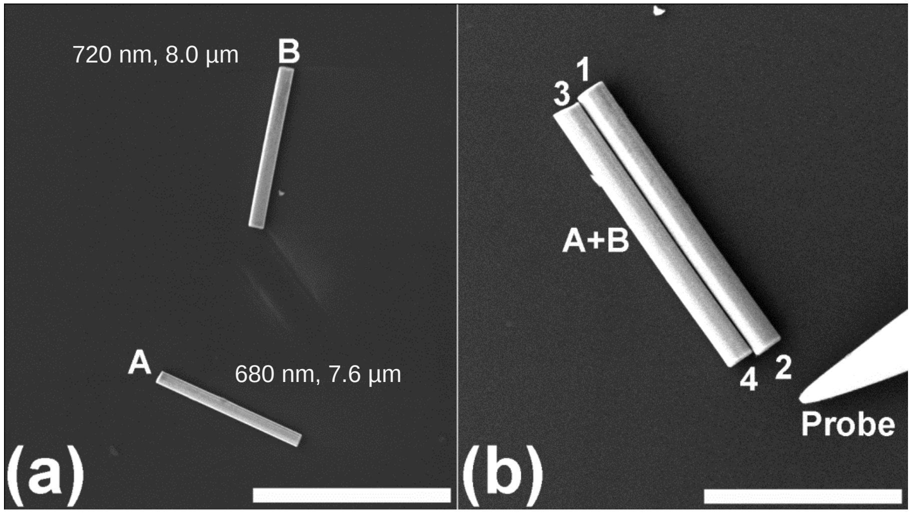
Q. Li et al., *Opt. Exp.* ***20*** *17873 (2012)*

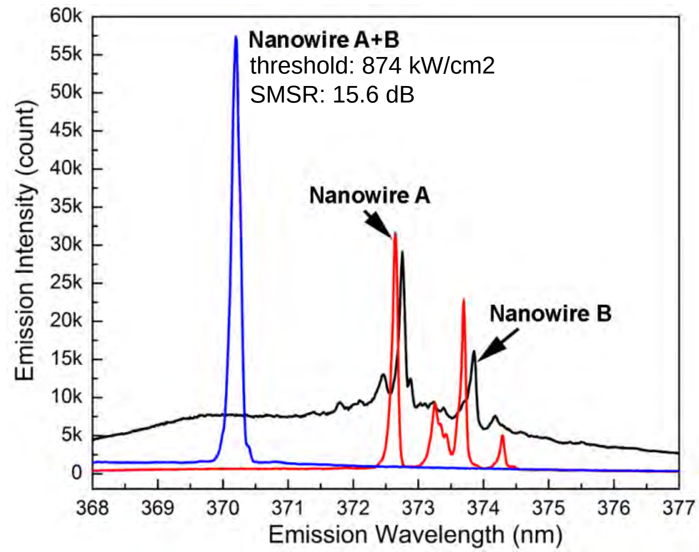
Single-mode: Narrow linewidth (<0.1 nm), ~15dB Side Mode Suppression Ratio, and **Low Threshold** (~250 kW/cm^2)

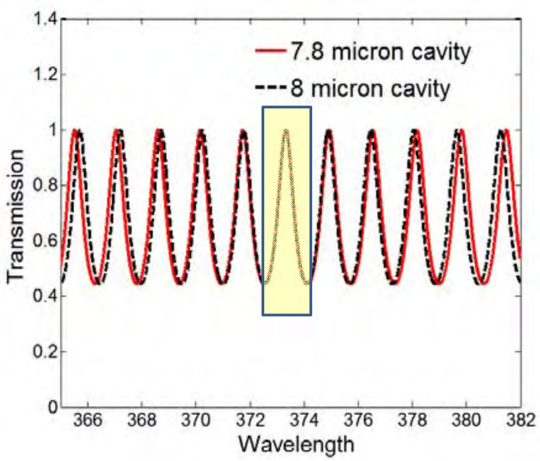
Multi-mode: Threshold ~500 kW/cm^2

Reducing the dimensionality of the wire (< ~130 nm diameter and < ~6 µm length) lowers the number of competing modes, leading to single-mode lasing.

Method 2: Single-mode lasing via coupled nanowire cavities





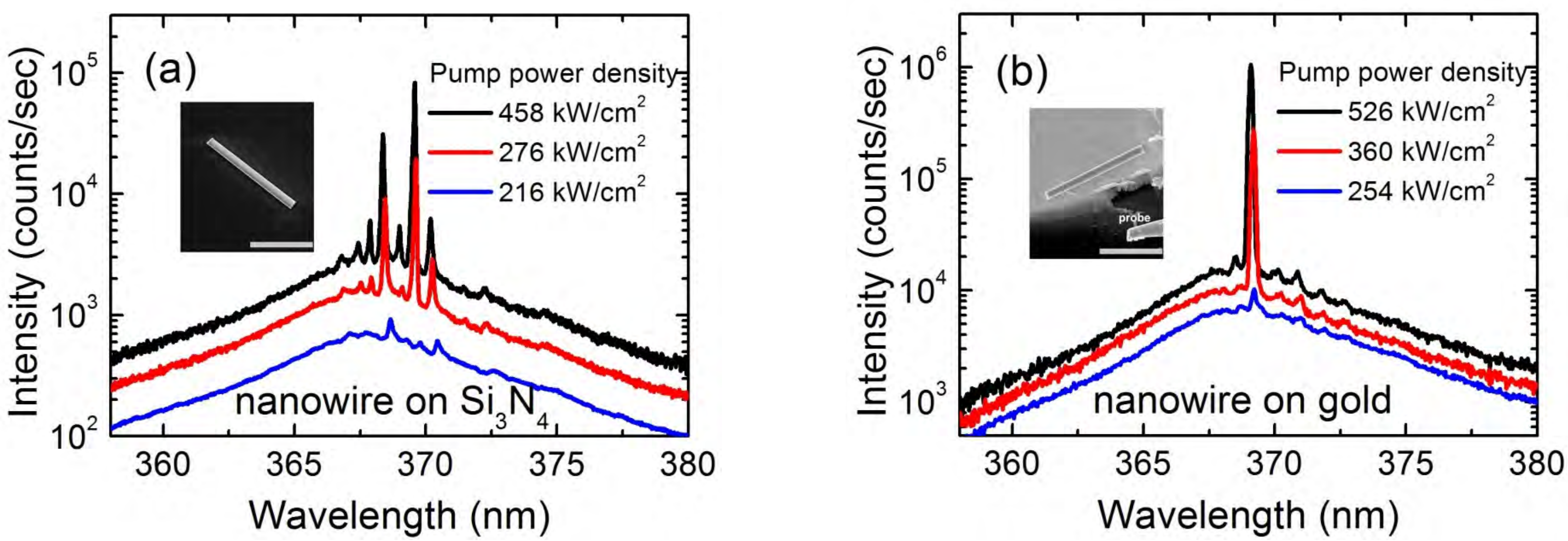


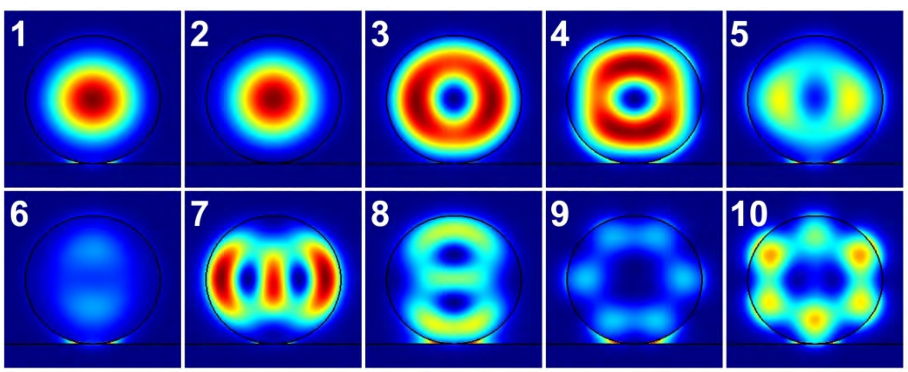
- GaN NWs need to be <~130 nm for single transverse mode behavior; larger single-mode NWs?
- Individual large NWs shows multiple modes.
- Coupled nanowires show single mode!
- Vernier effect – only resonant modes survive

modeling: Huiwen Xu (UNM)

H. Xu et al., *Appl. Phys. Lett.* **101** 113106 (2012)

Method 3: Metal substrate induced single-mode nanowire lasing



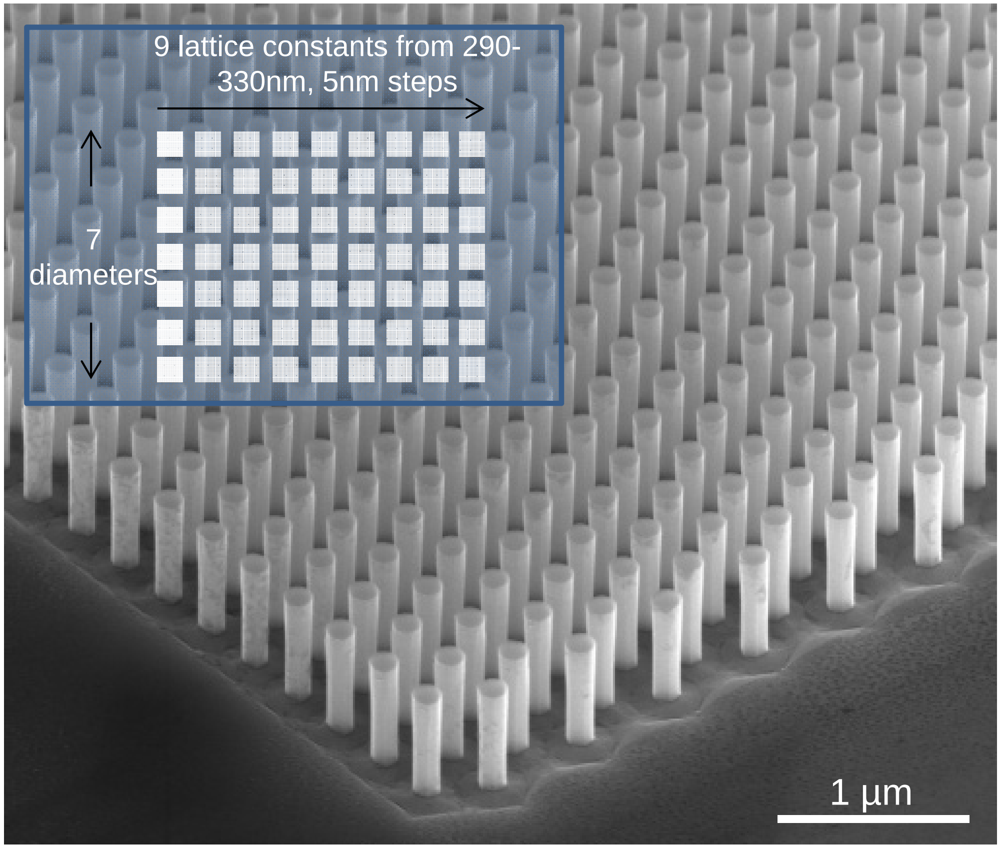


- NWs on Si_3N_4 show multi-mode lasing
- Same NWs moved onto gold-coated spot show single-mode lasing!
- Metal substrate induces mode-dependent loss mechanism

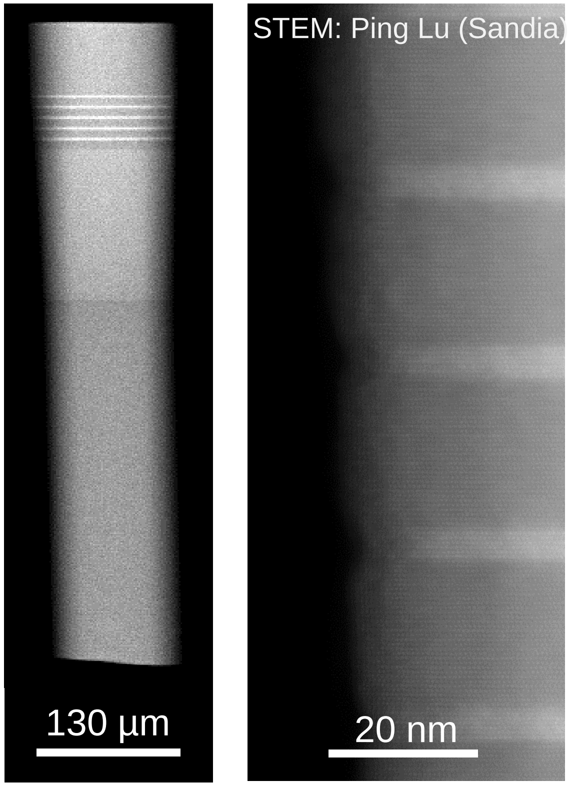
H. Xu et al., *Appl. Phys. Lett.* **101** 221114 (2012)

III-N Nanowire Photonic Crystal (PC) Lasers

Motivation: Achieve single-mode, tunable lasing on same chip. Applications in optical information processing, biology, solid state lighting, displays, etc.



Nanowire PCs fabricated by top-down method using e-beam lithographic mask

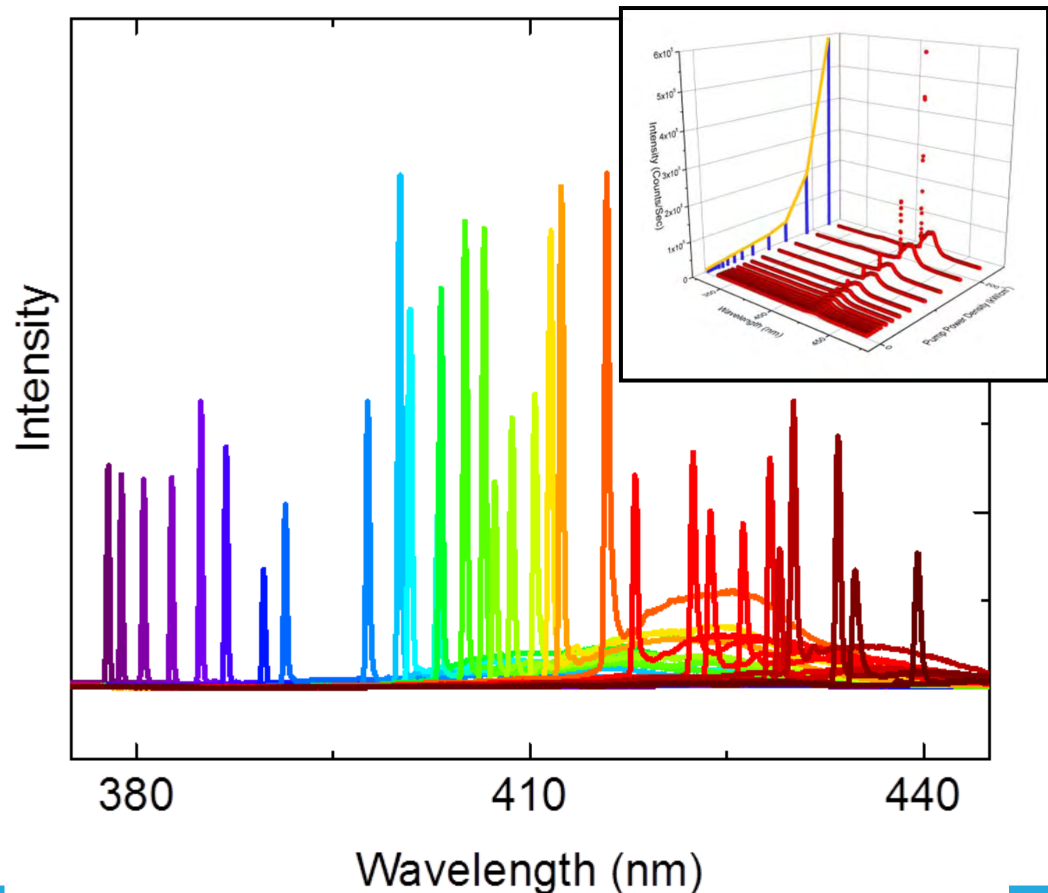


NW STEM images: 5x MQW InGaN emission centered at 420nm, $In_{0.02}GaN$ underlayer

III-N Nanowire Photonic Crystal Lasers

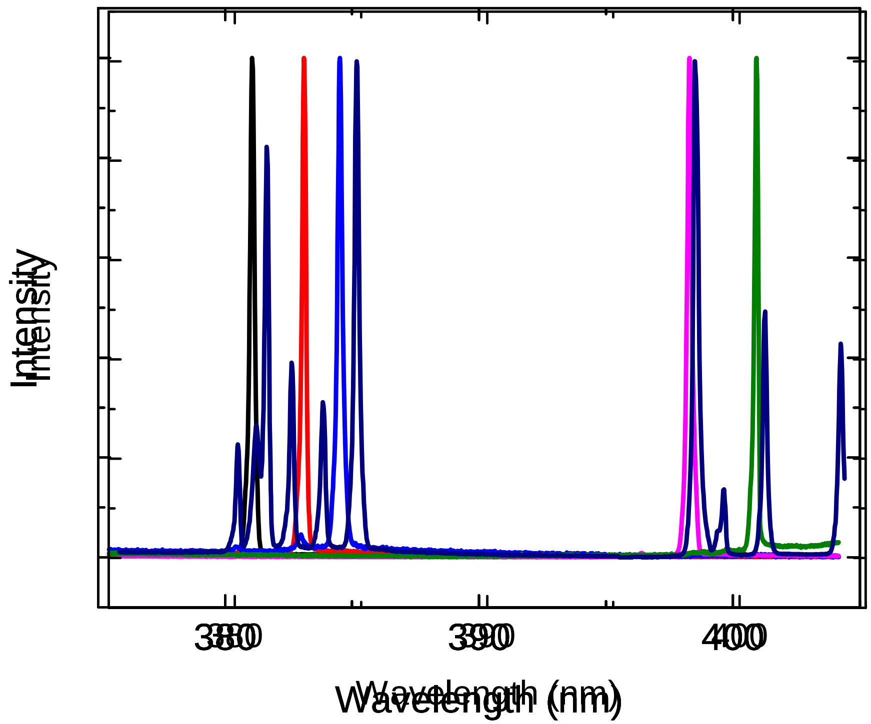
Broad gain width of InGaN MQWs with PC design allows for tunable single mode lasing over large range on same chip

61 color nanowire laser array



- High-yield >95% (2 of the PCLs were accidentally removed during sample handling.)
- Spectral Coverage from 380-440nm.
- Emission wavelength increases with the diameter and the lattice constant
- Thresholds are reasonable compared to other optically pumped III-N nanowire devices. (<500kW/cm^2 for all devices)

III-N Nanowire Photonic Crystal Lasers

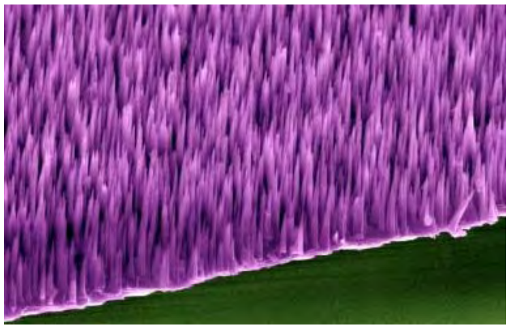


pitch: 320 nm
diameter: 130-140nm

10 um

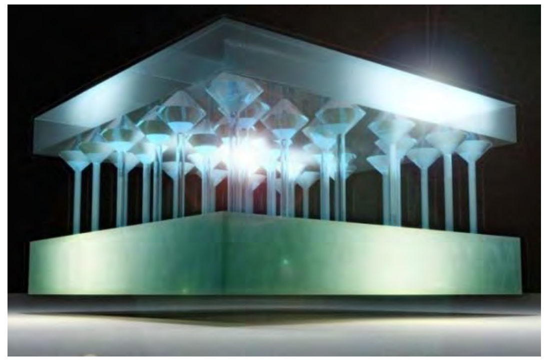
Summary - Bottom-up III-nitride nanowires

Vertically-aligned growth



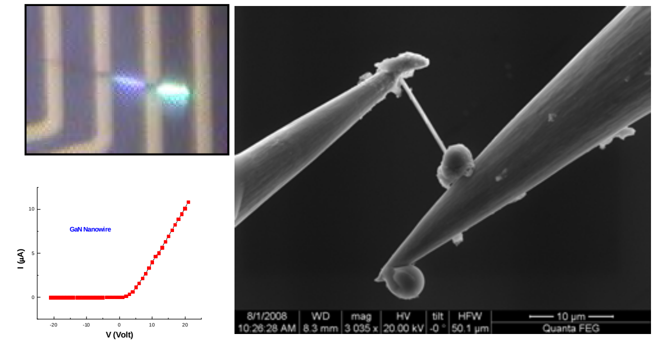
G. T. Wang et al., *Nanotechnology 17 5773-5780* (2006)
Q. Li, G. T. Wang, *Appl. Phys. Lett. 93, 043119* (2008)
Q. Li, J. R. Creighton, G.T. Wang. *J. Crys. Growth 310 3706-3709* (2008)

Nanowire-templated growth



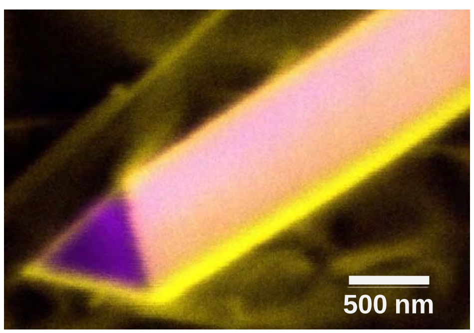
Q. Li, Y. Lin, J.R. Creighton, J. Figiel, G.T. Wang, *Adv. Mat.*, **21** 2416–2420 (2009)

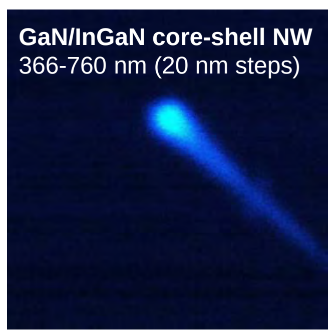
Electrical characterization



A. A. Talin, G. T. Wang, E. Lai, R. J. Anderson, *Appl. Phys. Lett.* **92** 093105 (2008)
Y. Lin, Q. Li, A. Armstrong, and G. T. Wang, *Solid State Commun.*, ***149***, *1608 (2009)*

Optical imaging and spectroscopy



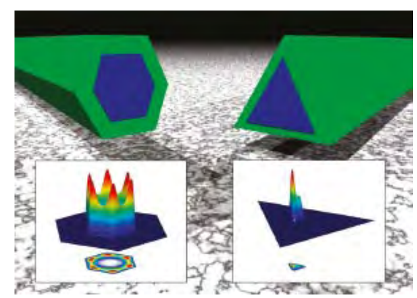


Q. Li, G. T. Wang, *Nano Lett.*, 2010, 10 (5), 1554 [GaN defect CL]
Q. M. Li, G. T. Wang, "*Appl. Phys. Lett.*, ***97***, 181107, 2010. [Gan/InGaN]
P.C. Uppadhya et al. *Semicond. Sci. Tech.* **25** 024017 (2010) [Ultrafast]
A. Armstrong, Q. Li, Y. Lin, A. A. Talin, G. T. Wang, APL **96**, 163106 (2010). [DLOS]

In-situ TEM

T. Westover et al., *Nano Lett.*, **9**, 257 (2009). [*in-situ* NW breakdown]
J. Y. Huang et al., *Nano Lett*, ***11 (4)***, *1618* ***(2011).*** [*in-situ* nanomechanics]

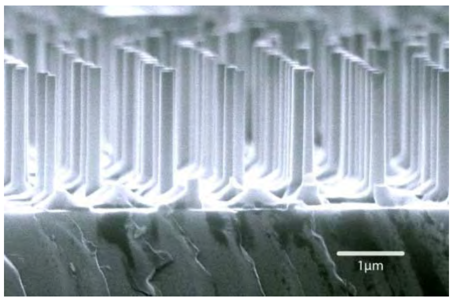
Theory



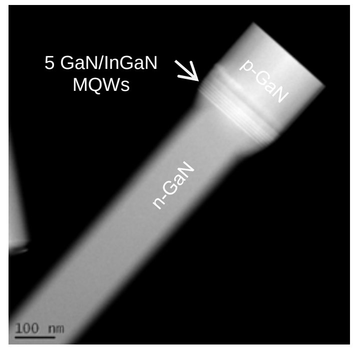
B. Wong et al., *Nano Lett* ***11 (8)***, *3074, 2011*

Summary – Top-down III-nitride nanowires

Top-down fabrication

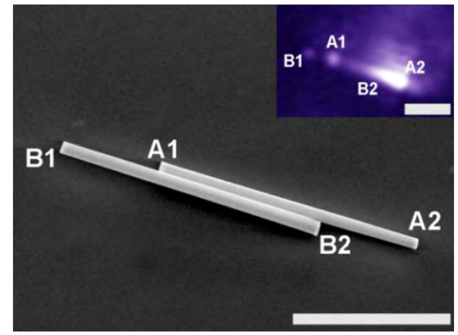


Nanowire LED "flashlight"



Q. Li et al., *Optics Express* **19**, 25528 (2011)

Single-mode GaN nanowire lasers

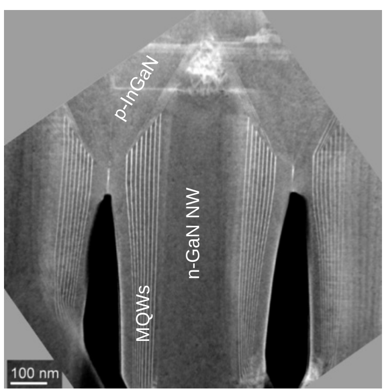


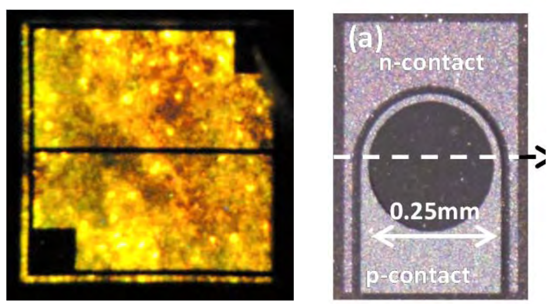
Q. Li et al., *Optics Express* **20** 17874 (2012)

H. Xu et al., *Appl. Phys. Lett.* **101** 113106 (2012)

H. Xu et al., *Appl. Phys. Lett.* **101** 221114 (2012)

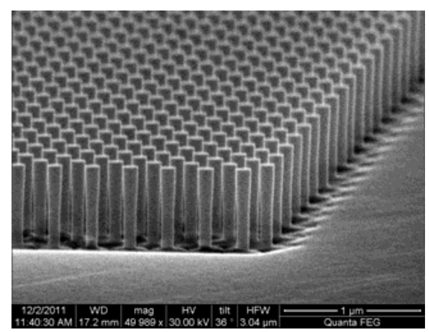
Vertically integrated radial nanowire LEDs and solar cells

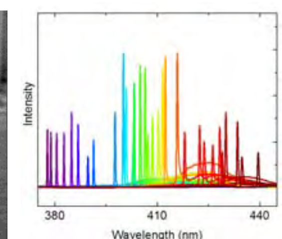




J. Wierer et al., *Nanotechnology* **23** 194007 (2012)

Tunable nanowire photonic crystal lasers





J.B. Wright et al., *in preparation*

Summary

- III-nitride nanowire based architectures have several potential advantages over planar-based devices for solid-state lighting, but numerous scientific & technical challenges in fabrication, performance, device contacts

- Sandia research highlights
  - Growth and properties of bottom-up III-nitride nanowires
  - New top-down (+ regrowth) fabrication for controlled geometries, flexible design, high quality, easier vertical device integration
  - Top-down nanowire LEDs, solar cells, lasers

Acknowledgments

Qiming Li - nanowire growth, nano-CL, TEM/EDXS, nanofabrication, strain-modeling

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Jianyu Huang – In-situ SPM-TEM for correlated structure-property studies

Jonathan Wierer – LED/solar cell device fabrication/characterization

Karl Westlake, Mary Crawford – PL, IQE measurements

Daniel Koleske – LED growth

Igal Brener, Willie Luk, Weng Chow, **Jeremy Wright, (Ph.D student)**, **Huiwen Xu (Ph. D student)**, Ganesh Subramania – NW [PC] lasers

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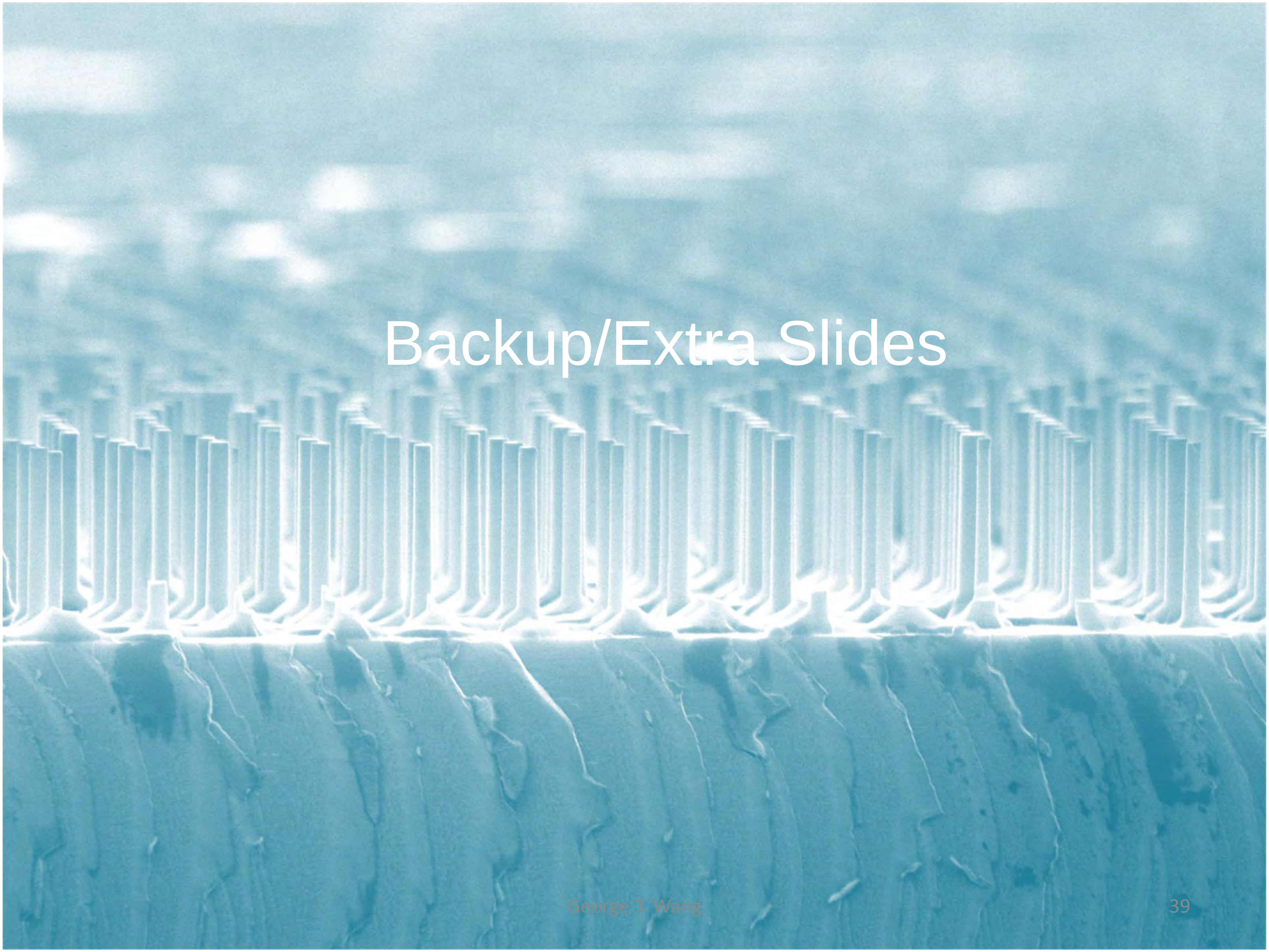
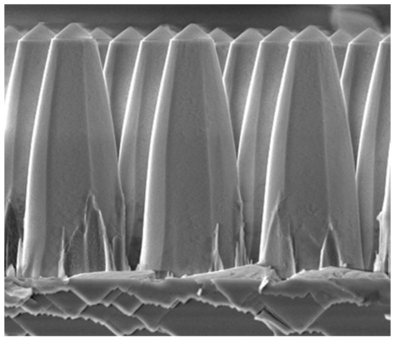
Contact: e-mail: gtwang@sandia.gov http://www.sandia.gov/~gtwang

Backup/Extra Slides

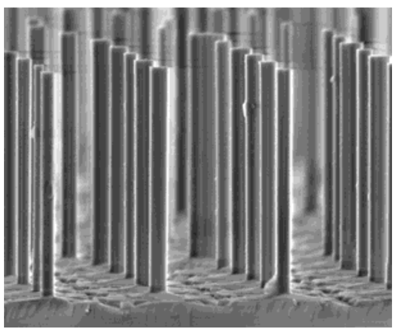


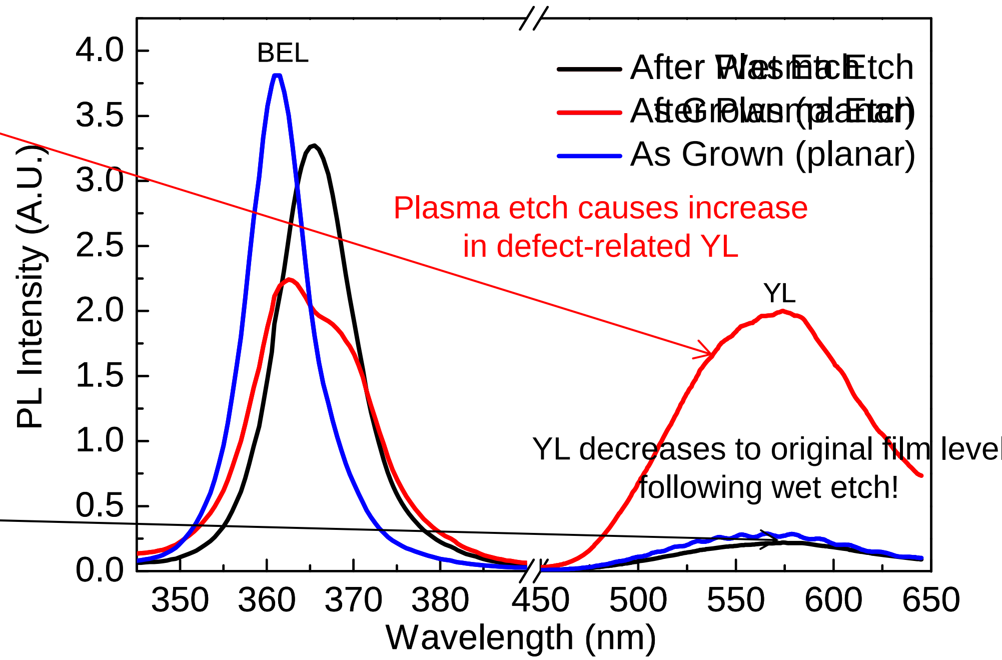
Wet etch step removes plasma etch damage

Plasma etch only



After wet etch





Lasers for Solid State Lighting

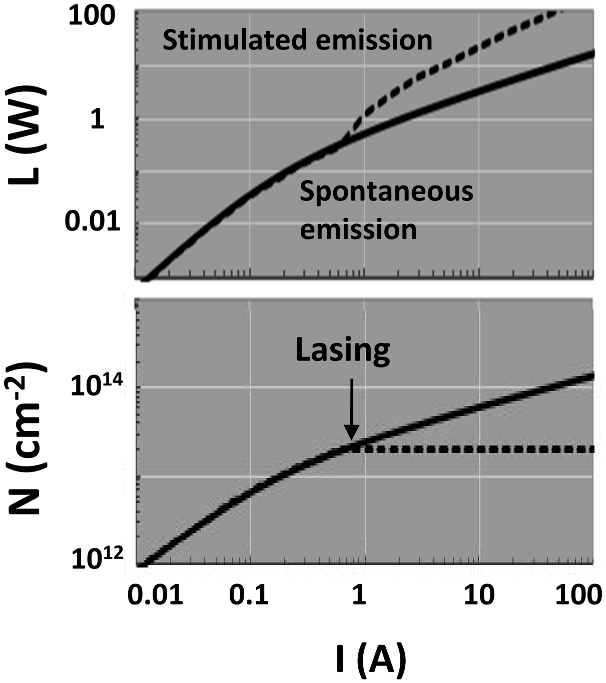
Advantages of lasers for lighting:

- Lasers show very high efficiency at high power
- LED and LD current densities are converging
- Carrier density is clamped at threshold
    - Circumvent the droop problem in LEDs
    - Need to reduce threshold to avoid losses
- After threshold slope efficiency is one
- Directionality, polarized emission, modulation

Laser Sources For SSL:

- High efficiency
- Low threshold
- Focus on III-nitrides
- Nanowire lasers
    - Low threshold
- Polariton lasers
    - Ultralow threshold
    - New physics

Clamped carrier density

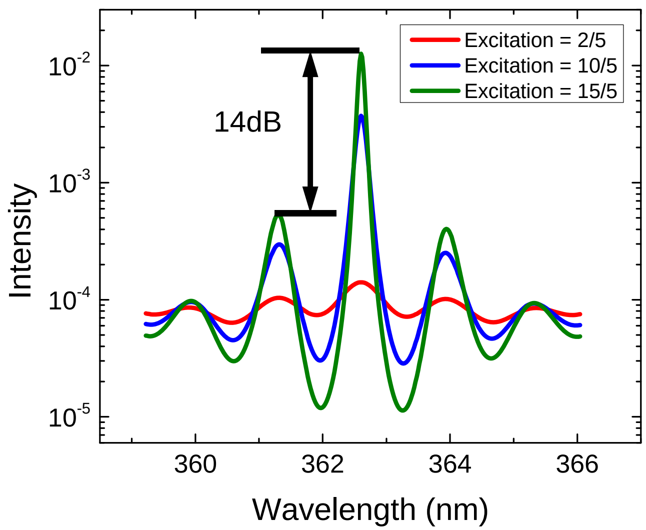


Are narrow linewidth sources acceptable?

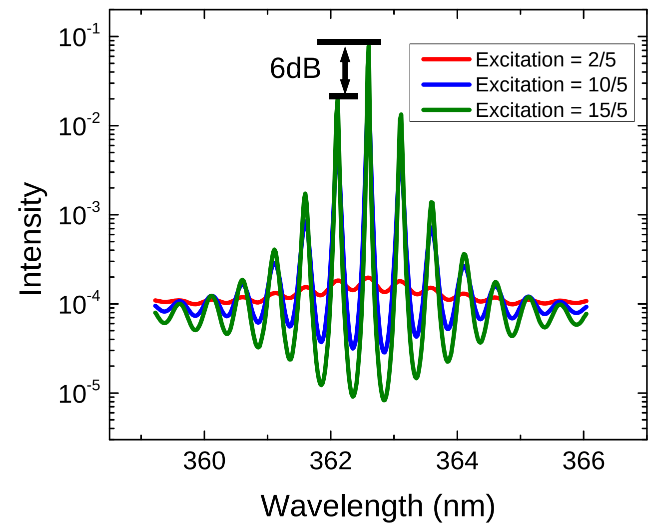
A.J. Fischer, Sandia

Single mode GaN nanowire laser

4 µm long, 140 nm dia. nanowire



12 µm long, 140 nm dia. wire



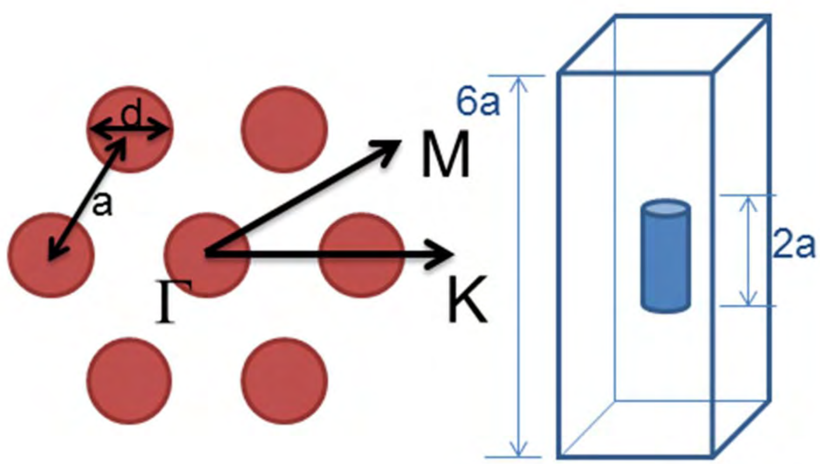
- Multimode laser theory calculations to determine which of the passive-cavity eigenmodes will be above lasing threshold for given experimental conditions.
- Modeling shows that by reducing the dimensionality of the wire we can reduce the number of competing modes, leading to single-mode lasing.

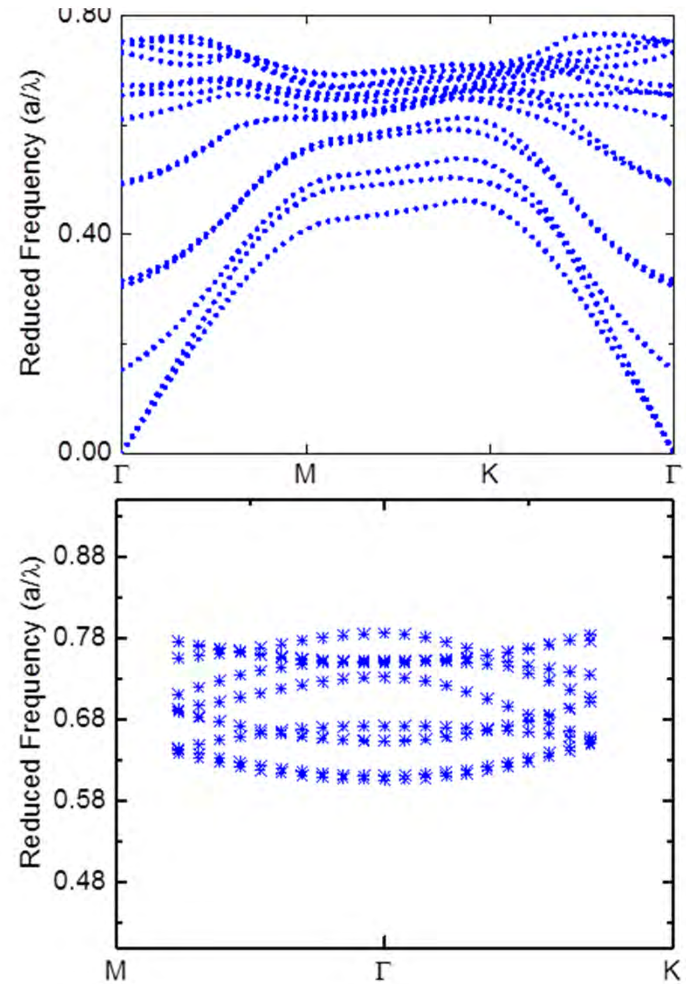
Outline

- **Why III-nitride nanowires for solid-state lighting, PV, etc. ?**
- **Bottom-up nanowires**
  - **Growth, characterization, nanowire-templated film growth**
- **Top-down nanowires**
  - **Fabrication, axial and radial LEDs, solar cell, lasers**
- **Summary**

Design Rationale

- Higher order bands have low dispersion
- We desire low group velocity to enhance the light matter interaction and the formation of standing waves within the gain medium, to allow low lasing thersholds in materials that exhibit reduced gain over larger bandwith





III-N Nanowire Photonic Crystal Lasers

Continuously tuned photonic crystal stripe