SAND2014-20400C

III-nitride nanowires for future optoelectronics

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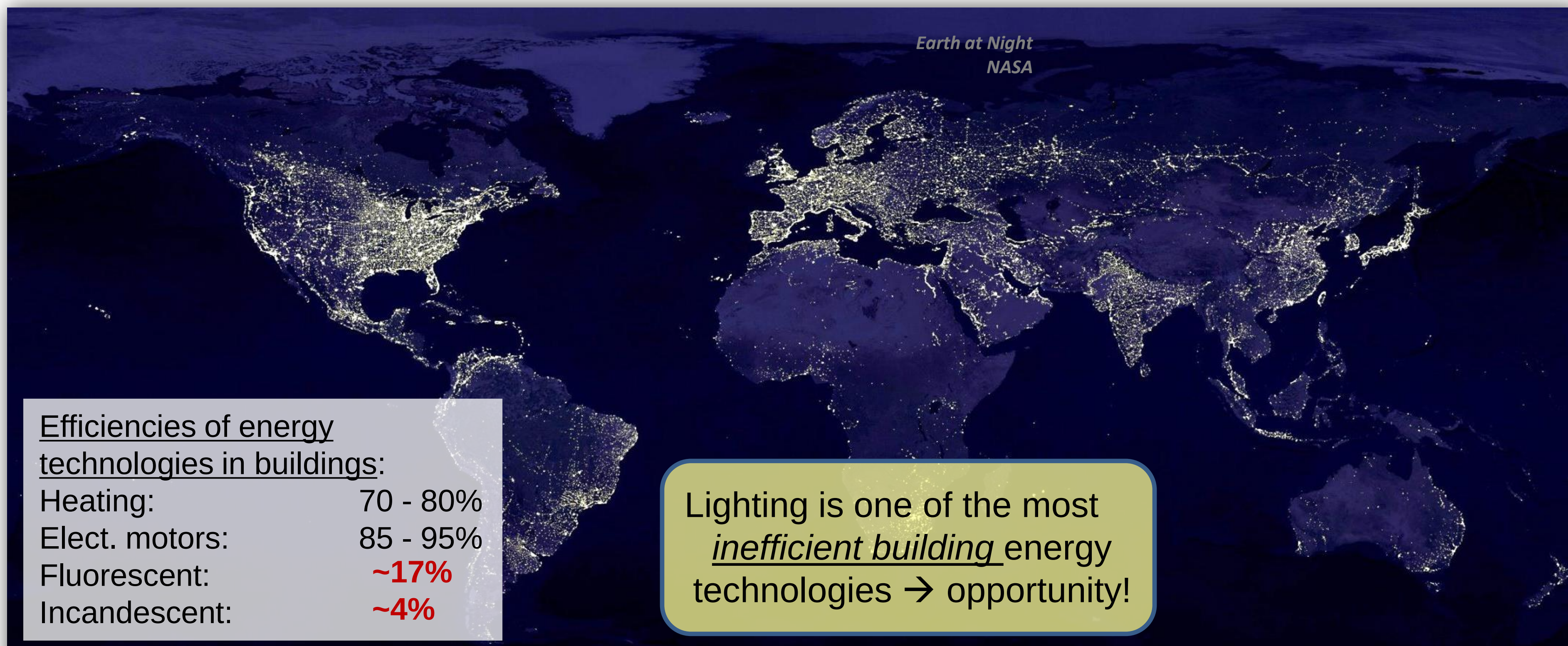
Lighting: large fraction of energy use and low efficiency



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

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

Lighting is one of the most *<u>inefficient building</u>* energy technologies → opportunity!

- ~22% of US electricity 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% efficiency

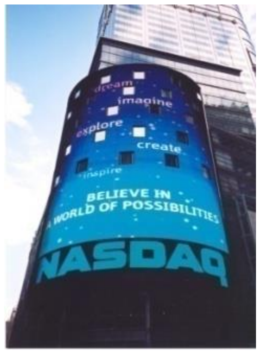
- **decrease electricity consumed by lighting by > 50%**
- **decrease total electricity consumption by 10%**

A Progression of Productive Uses for Colored and White Solid-State (LED) Lighting

COLORED

Center high-mount stop light (CHMSL). http://www.honda-tech.com/showthread.php?t=2413558

NASDAQ's Giant Video Display in Times Square, New York (Jeff Tsao)



Sharp QuadPixel RGBY LED-backlit LCD Display. http://www.macworld.com/article/145541/2010/01/sharp_quadpixel.html

SMART

hue
PHILIPS

Philips HUE Connect Bulb http://store.apple.com

http://uncrate.com/stuff/philips-hue/

WHITE

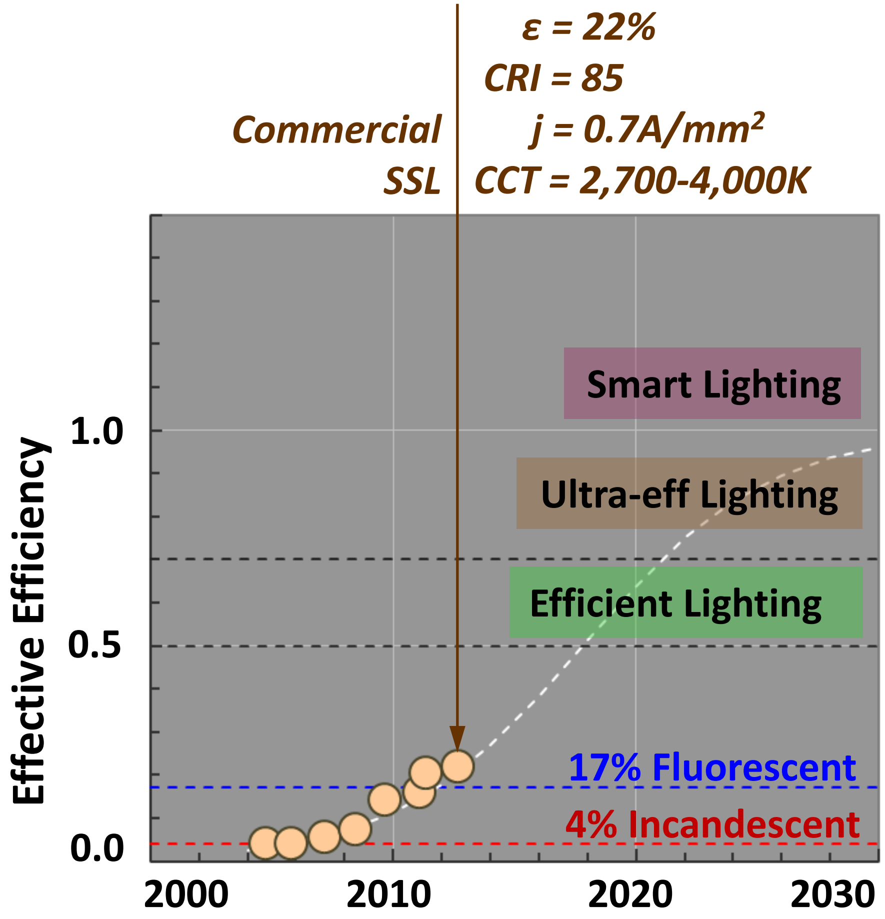
Surefire U2 flashlight. http://en.wikipedia.org/wiki/File:SurefireU2JPG.jpg

Nokia camera phone with LED flash. http://www.itechnews.net/wp-content/uploads/2009/07/Nokia-3720-Classic-the-most-rugged-mobile-phone.jpg

2011 Philips L-prize 60W equivalent 10W LED light bulb replacement http://www.lightingprize.org/philips-winner.stm

after M.E. Coltrin, Sandia

Efficiency, Ultra-efficiency, and Beyond



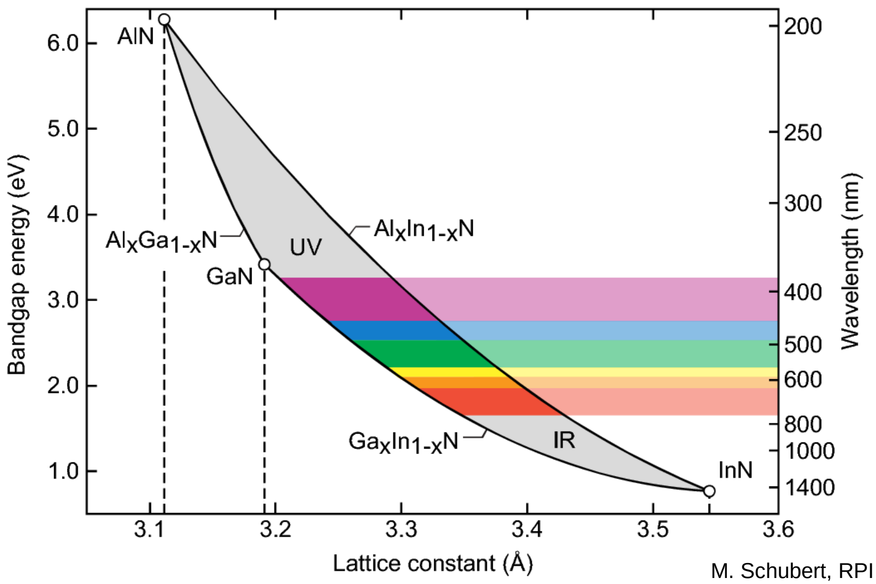
"Ultra-efficient" SSL: > 70%

Achieves the greatest savings in energy consumption and gains in human productivity

Requires fundamentally new architectures and understanding, *enabled* by basic research

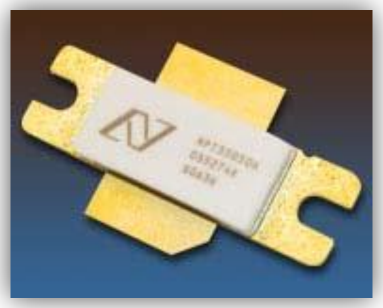
Foundation of SSL: III-Nitride (AlGaInN) Semiconductors

- **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?)



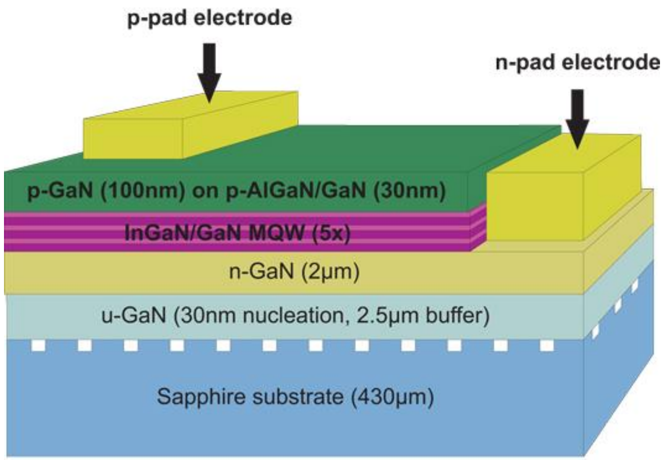
M. Schubert, RPI

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

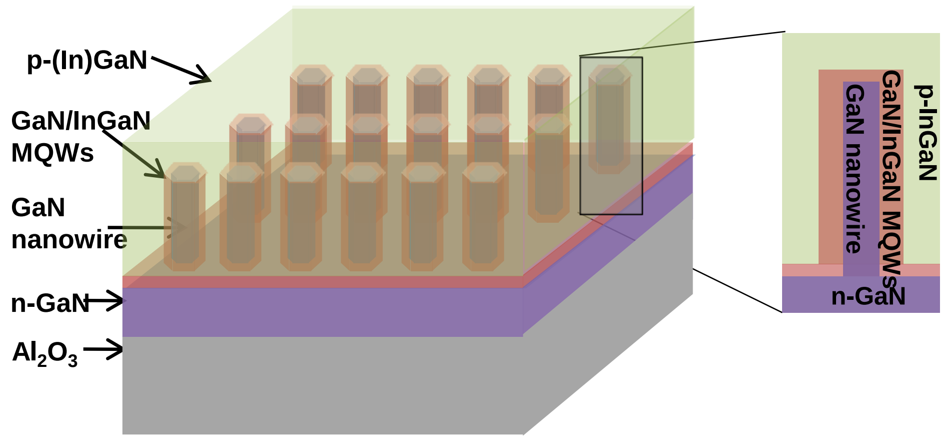


Nitronex GaN power transistor

Why III-nitride (AlGaInN) nanowires for SSL?



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



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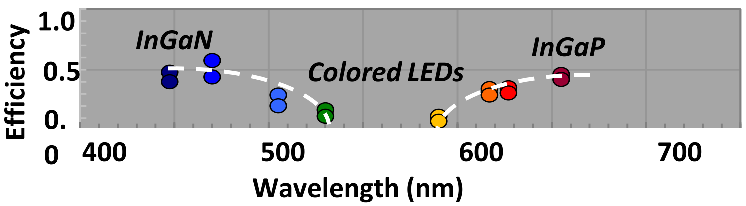
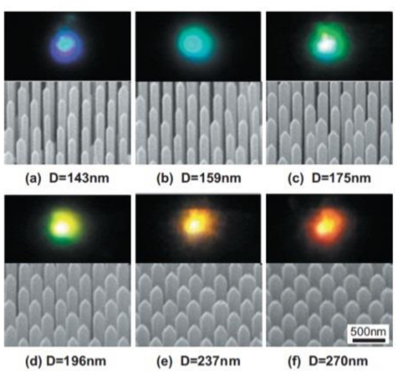
Why III-nitride nanowires for SSL?

Advantages due to enhanced strain accommodation in nanowires

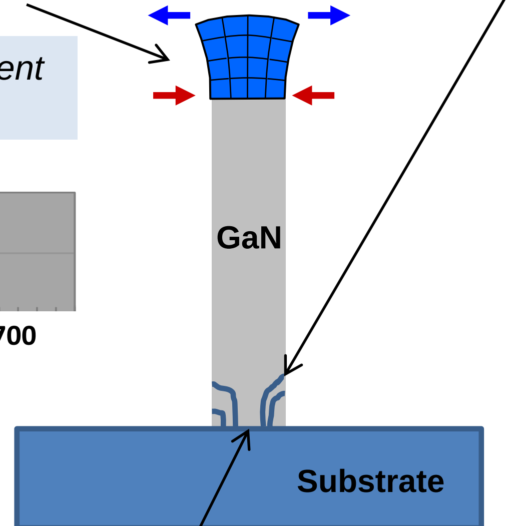
elastic strain relaxation at surface

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





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

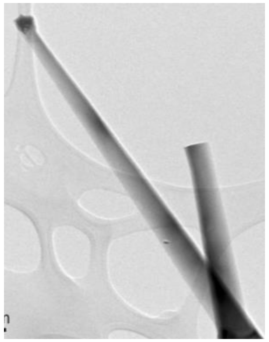


small interfacial area

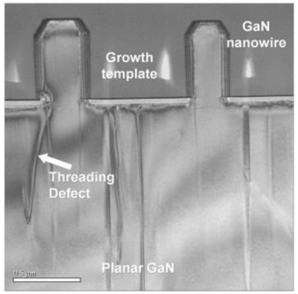
benefit: can grow on cheaper, lattice mismatched substrates; integration with Si devices

bending of dislocations (TDs) toward surface

benefit: reduced TDs, higher IQE

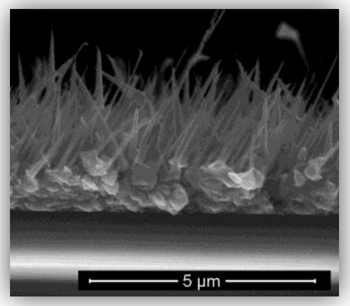


VLS-grown TD-free GaN NWs - Sandia

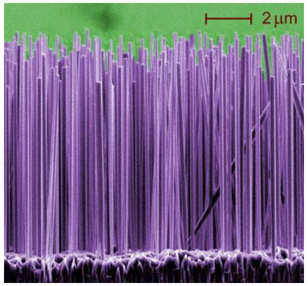


Hersee et al., J. Mat. Res. **17**, 2293 (2011) - UNM

Bending & termination of TDs at nanowire base



GaN NWs on tungsten foil - Sandia

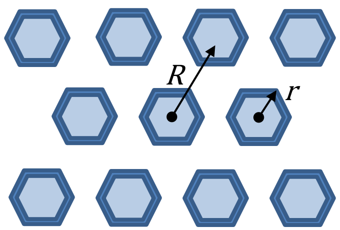


GaN NWs on Si - NIST

Why III-nitride nanowires for SSL?

vertical device integration

benefit: *higher device area per chip (cost/droop)*

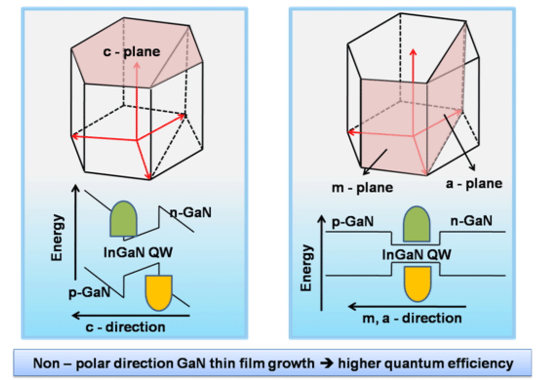


$$\frac{A_{sidewall}}{A_{substrate}} = \frac{6rh}{2.6R^2}$$
$$\cong \mathbf{4.6F \cdot AR}$$

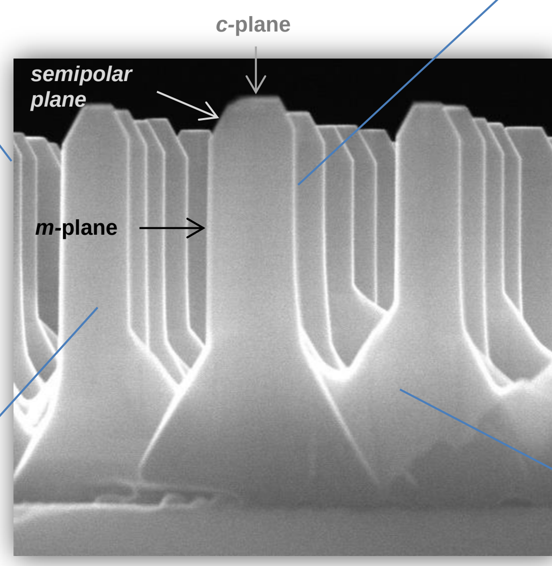
*example: 4.6·0.5·3=**6.9× increase!***

access to nonpolar & semipolar planes

benefit: *higher IQE, reduced wavelength shift*



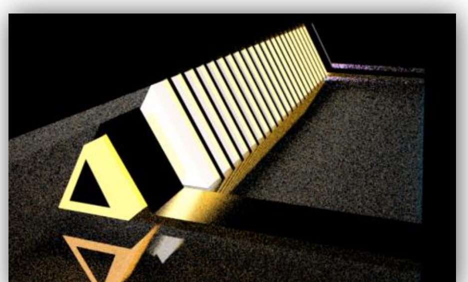
http://csel.snu.ac.kr/research/LED.php



Radial GaN/InGaN MQW NW (Sandia)

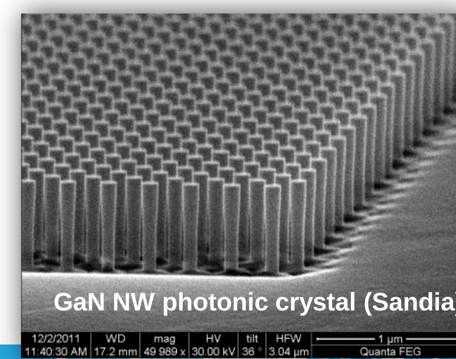
nanolasers

benefit: *ultracompact, low power coherent light source*



2D arrangements (photonic crystals)

benefit: *higher light extraction, IQE, wavelength tuning*



GaN NW photonic crystal (Sandia)

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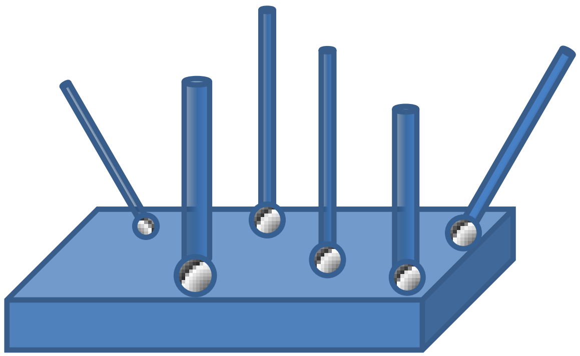
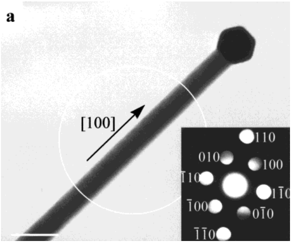
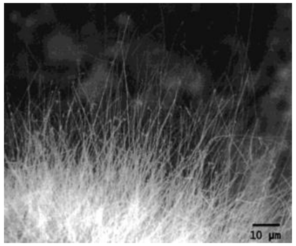
Bottom-up growth of GaN-based nanowires

Metal catalyzed (vapor-liquid-solid)





C. Lieber, 2000

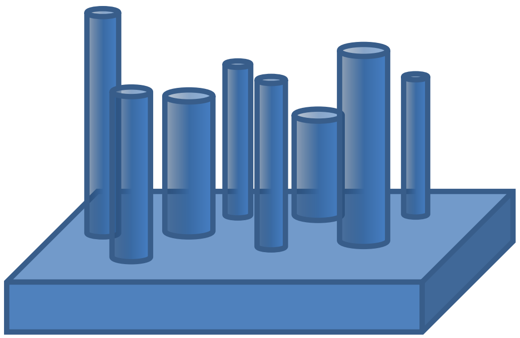


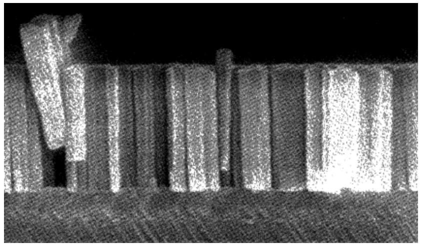
C.C. Chen, 2001

Methods: PLD, thermal evaporation, (MO)CVD

Historically popular (ease & low cost of entry), but falling out of favor due to limitations

"Self-catalyzed"



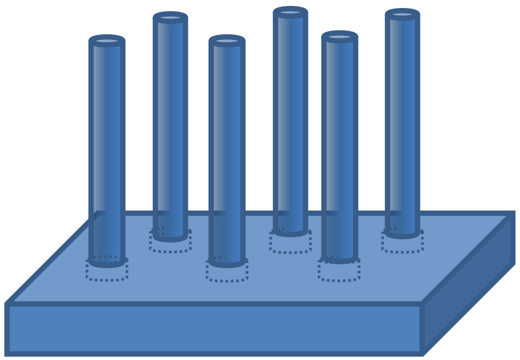


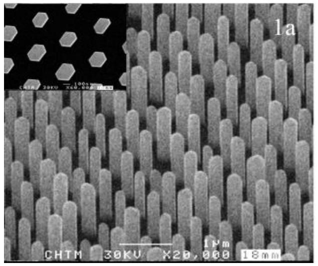
K. Kishino, 1997

Methods: (PA)MBE, HVPE, (MO)CVD

Cons include slow growth rates and random ordering

Selective area growth (SAG)





S. Hersee, 2006

Methods: (PA)MBE, MOCVD

Currently popular, but may require growth conditions that limit materials & architectural flexibility

Worldwide GaN-based nanowire SSL-related research

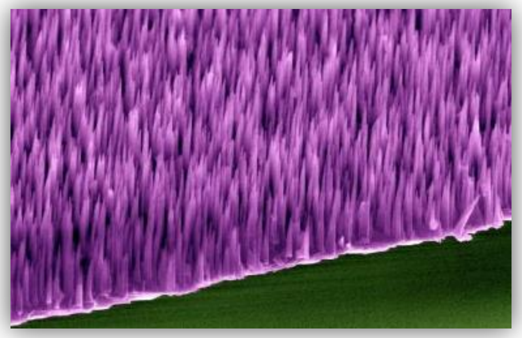
North America

Europe

Asia

Previous SNL research: bottom-up III-nitride nanowires

Vertically-aligned growth



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

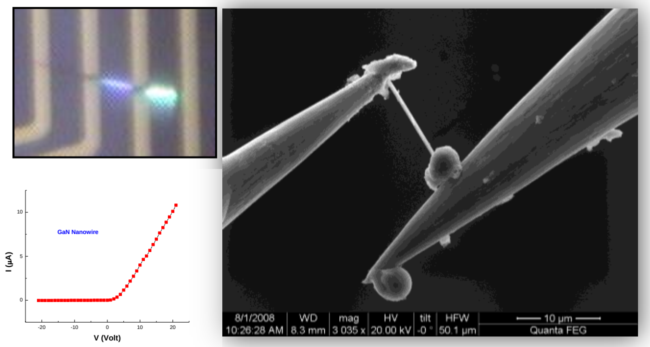
Q. Li, G. T. Wang, *Appl. Phys. Lett. 93, 043119* (2008)

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)

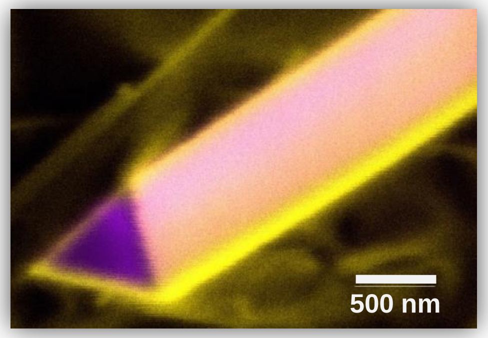
Electrical characterization

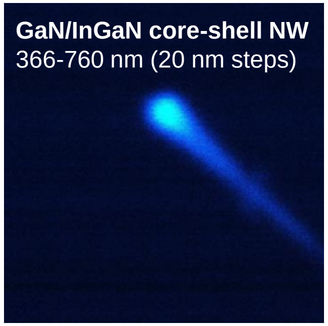


A. A. Talin, G. T. Wang, E. Lai, R. J. Anderson, *Appl. Phys. Lett.* **92** 093105 (2008)

Y. Lin, Q. Li, A. Armstrong, and G. T. Wang, *Solid State Commun.*, ***149***, *1608 (2009)*

Optical imaging and spectroscopy





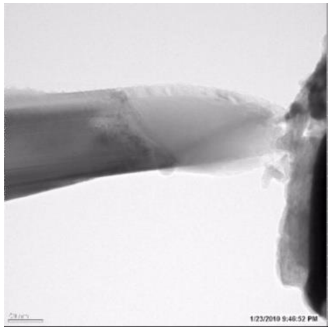
Q. Li, G. T. Wang, *Nano Lett.*, 2010, 10 (5), 1554 [GaN defect CL]

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]

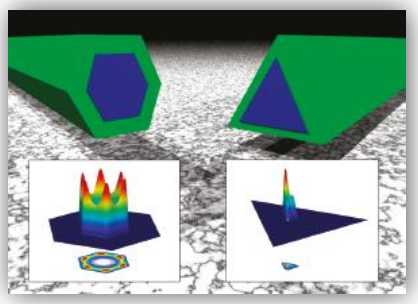
In-situ TEM



T. Westover et al., *Nano Lett.*, **9**, 257 (2009). [*in-situ* NW breakdown]

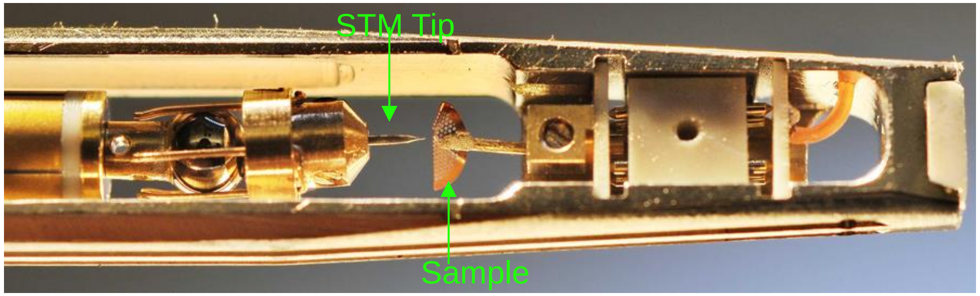
J. Y. Huang et al., *Nano Lett,* ***11 (4),*** *1618* ***(2011).*** [*in-situ* nanomechanics]

Theory

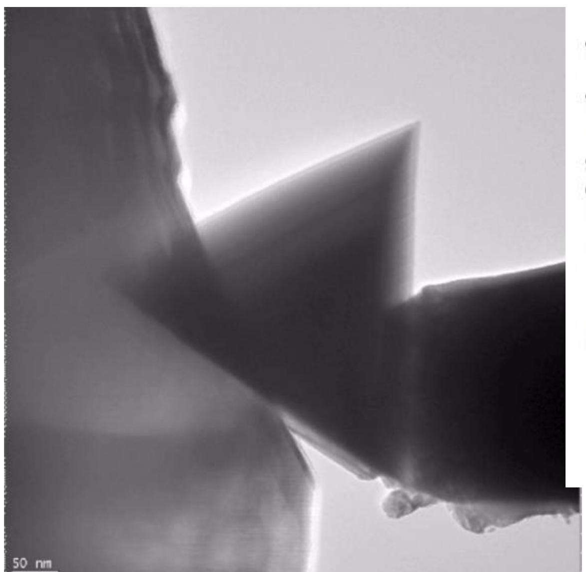


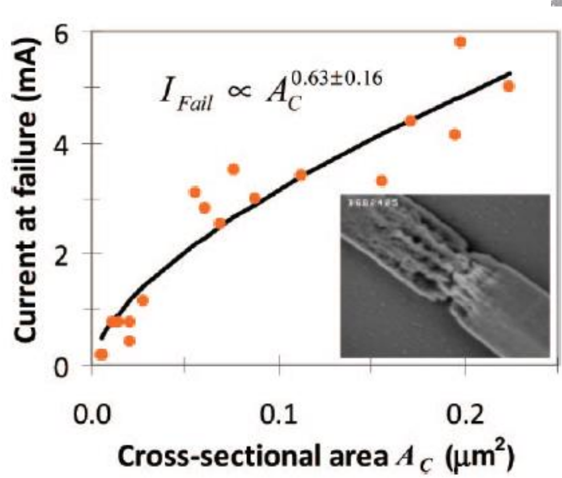
B. Wong et al., *Nano Lett* ***11 (8)***, *3074, 2011*

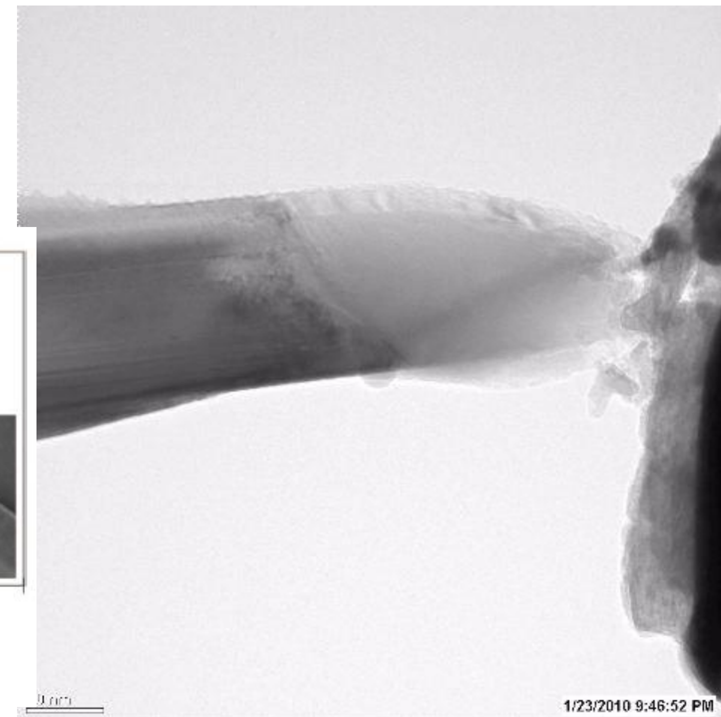
In-situ TEM studies – nanowire electrical breakdown



NW decomposition via Joule heating
(relevant for NW devices)







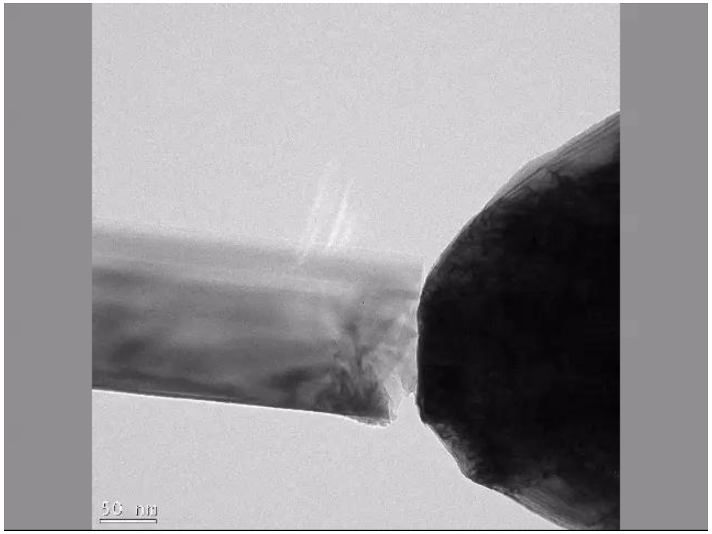
NW breakdown at 60V, 20 µA
(avg. breakdown I ~3000 kW/cm^2)

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

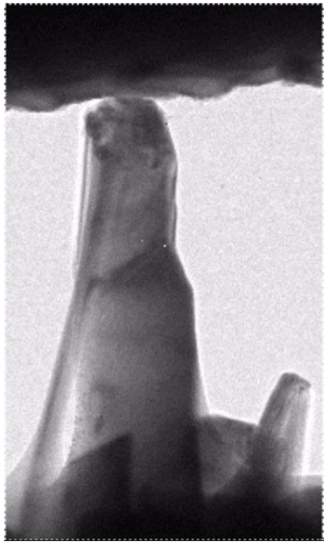
PI: J. Huang

SSLS EFRC

Real time *In-situ* TEM nanomechanics of GaN NWs



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



- Plastic deformation often observed before fracture
- Plasticity is local, not global

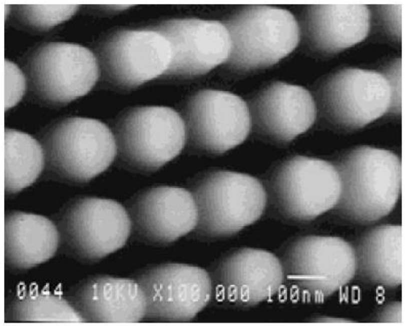
Study provides unique insight into the nanomechanical properties which may impact nanowire device processing and operation

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

PI: J. Huang

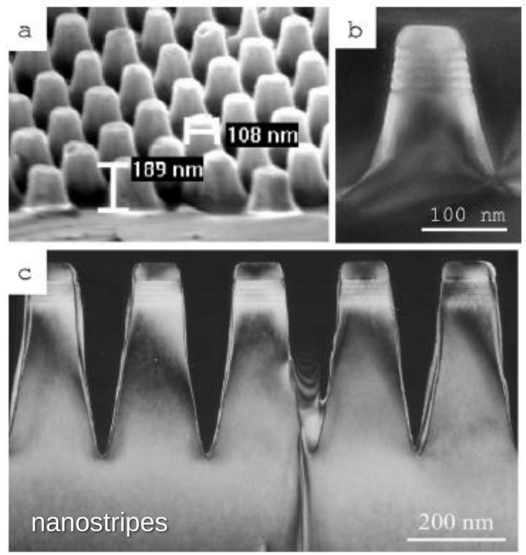
Previous top-down nanorod/nanopillar/nanopost work

InGaN/GaN MQW nanoposts/nanopillars



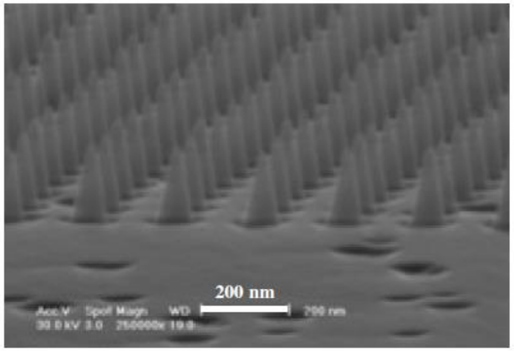
L. Chen et al., PSSa **188** 135 (2001) (Brown, Yale)

- E-beam Ni mask, Cl-based RIE etch



S. Keller et al., J. Appl. Phys. **100** 054314 (2006) (USCB)

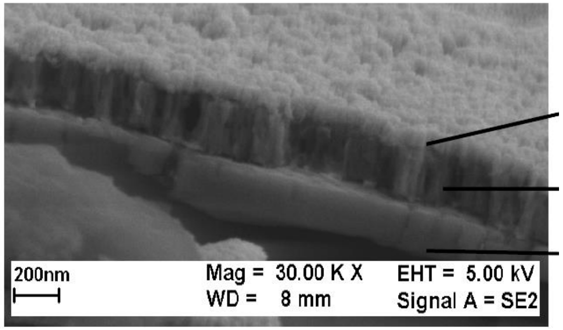
Holographic lithography, SiO_2 mask, CHF_3 ICP etch to make strips and pillars

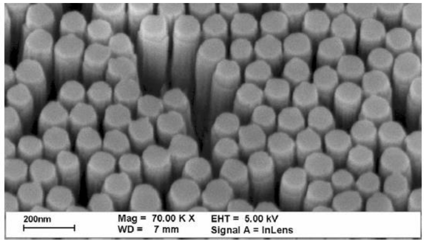


H. S. Chen et al., Nanotechnology **17** 1454 (2006) (NTU)

- E-beam Cr mask, Cl-based ICP-RIE etch
- Observed PL blue-shift w/decreasing diameter due to strain relaxation (reduction of QCSE)

InGaN/GaN MQW nanorod LED





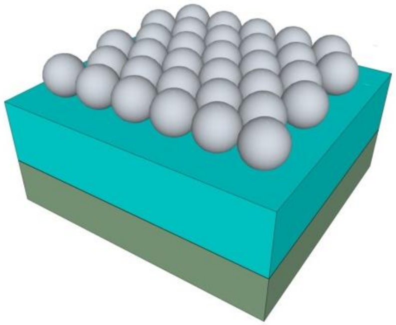
C. Y. Wang et al., Opt. Expr. *16,* 10549, 2008. (NTU)

M. Y. Ke et al., IEEE J. Sel. Top. Quantum Elec. **15** 1242 (2009) (NTU)

SiO_2 nanosphere lithography, Cl-based ICP-RIE etch; electrically injected array

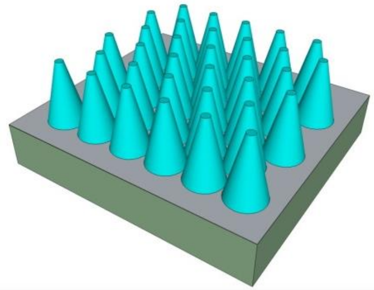
- Tapered, short aspect ratio nanopillars via plasma etching
- Plasma etch causes sidewall damage

New dry + wet top-down ordered nanowire fabrication process



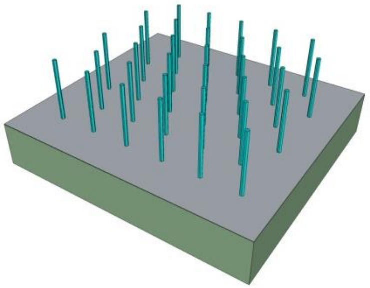
(0001) GaN on sapphire

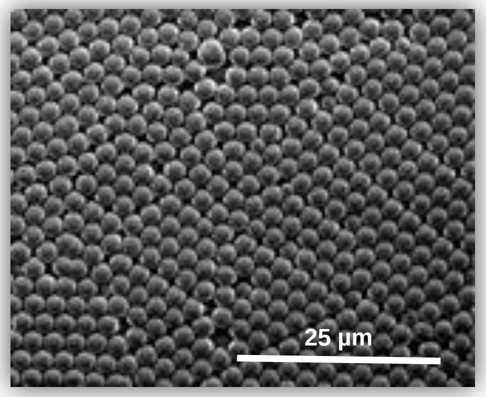
ICP etch



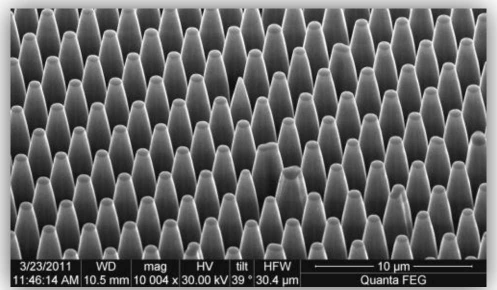
Selective KOH-based wet etch

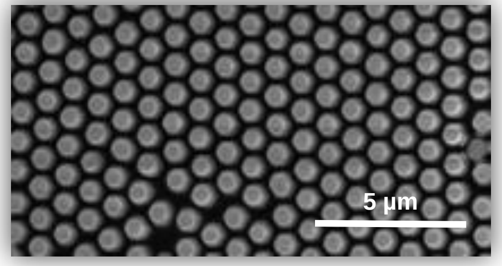
(AZ-400K developer)





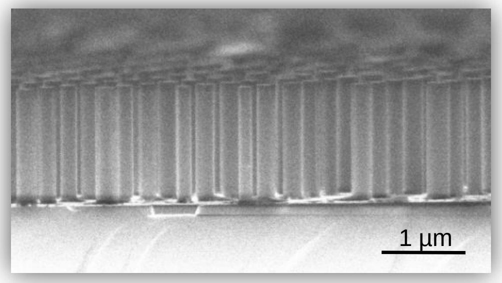
Q. Li, J. J. Figiel, G. T. Wang, Appl. Phys. Lett., **94**, 231105 (2009).

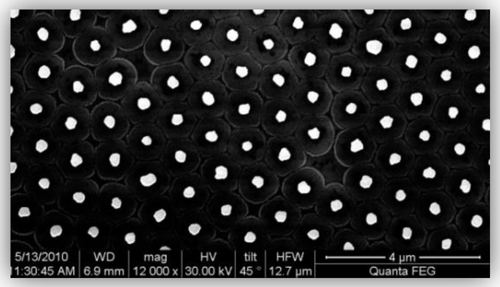




Plasma etch causes sidewall damage

Tapered; no well-defined facets





Wet etch: straight sidewalls, removes sidewall damage

Straight GaN nanowires with controllable geometries

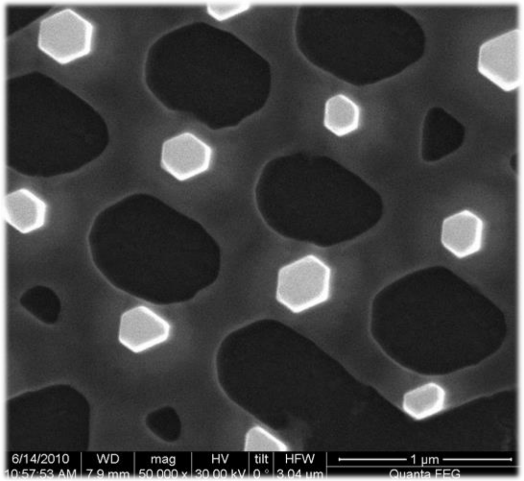
Wet etch rate negligible for top (Ga-polar) *c*-face & fast for [10-10], leads to nominally hexagonal NWs with **straight & smooth** *m*-facets

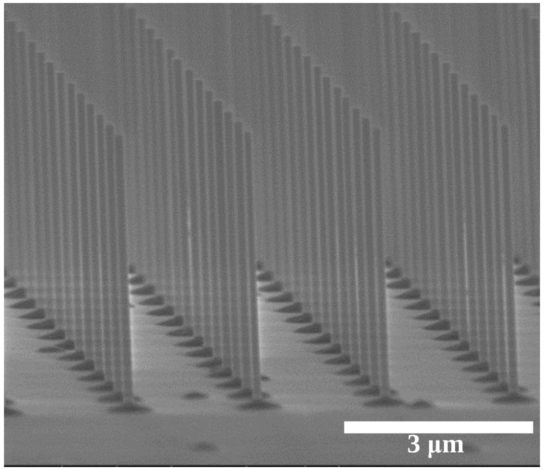
Superior and independent control of:

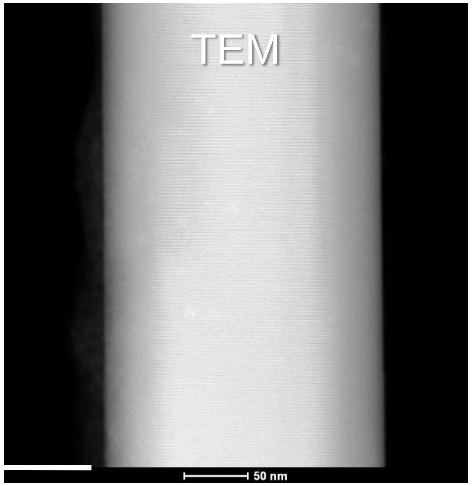
Height (dry etch depth)

Diameter (wet etch time)

Pitch/arrangement (defined by masking template)







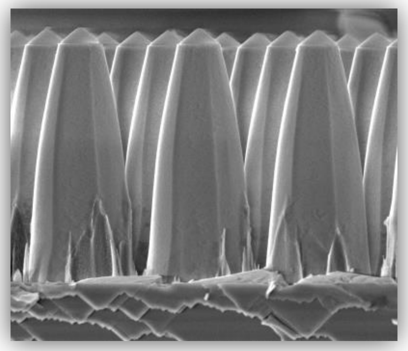
Smooth sidewall created by wet etch

TEM: Ping Lu, SNL

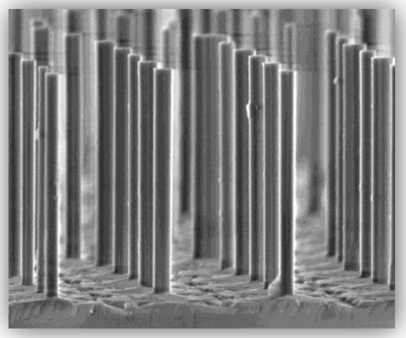
See: Q. Li et al., *Optics Express* **19**, 25528 (2011)

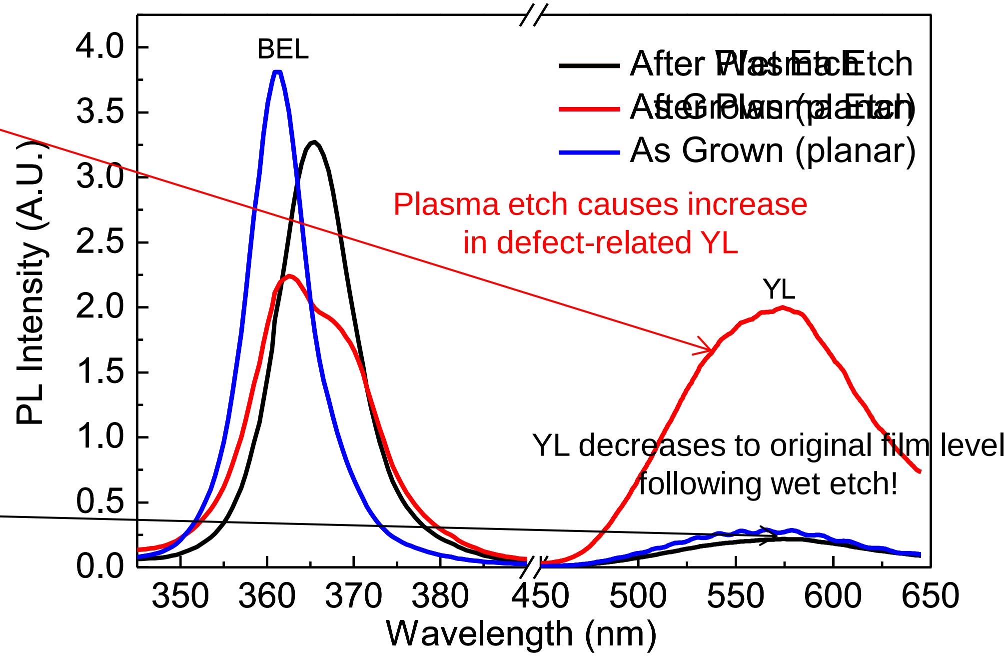
Wet etch step removes plasma etch damage

Plasma etch only

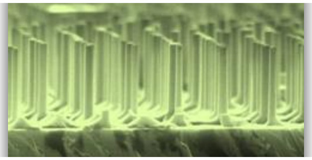


After wet etch

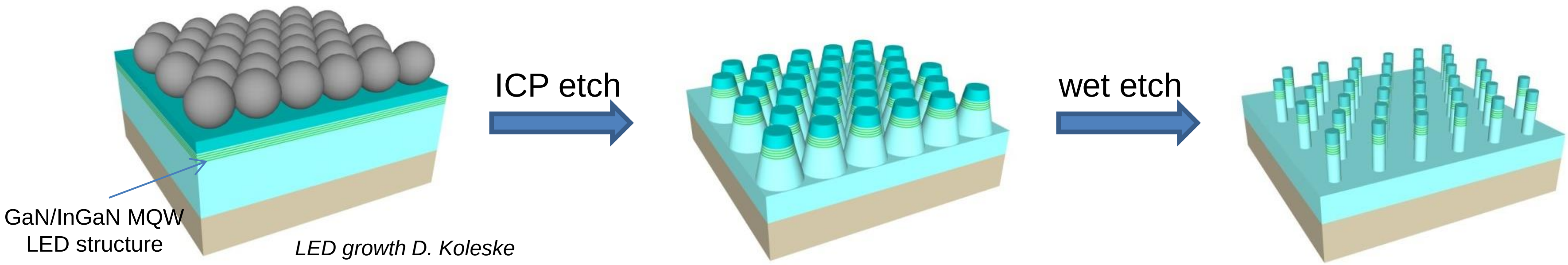


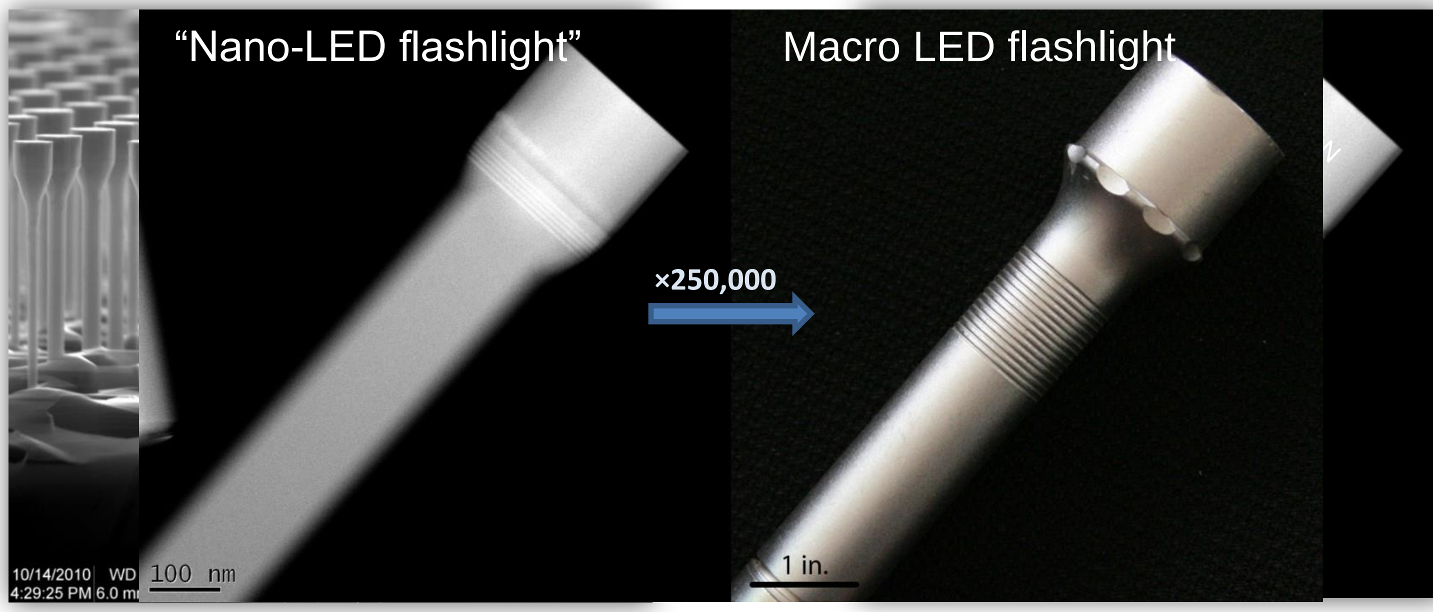


Bottom-up vs. Top-down nanowires

	Bottom-up 	Top-down (+ regrowth) 
Length	Up to hundreds of μm	Several μm
Diameter	~30 nm to microns	~10 nm to microns
Dislocations	Few or none	Depends on density in film
Radial architectures	Yes, typically by MOCVD	Yes, by MOCVD regrowth
Axial architectures	Yes, typically by MBE	Yes, but not strain relaxed
Substrate	Lattice matching less critical	Lattice matched
Characterization	Some properties difficult to measure (e.g. doping)	Starting material can be measured using standard techniques
Material Quality:	Point defect density may be high due to growth conditions needed	Starting material can be grown under optimal growth conditions
Uniformity	Wire-to-wire variations may occur based on pitch, diameter	Good, although regrowth has same issues as bottom-up

Axial GaN/InGaN nanowire LEDs

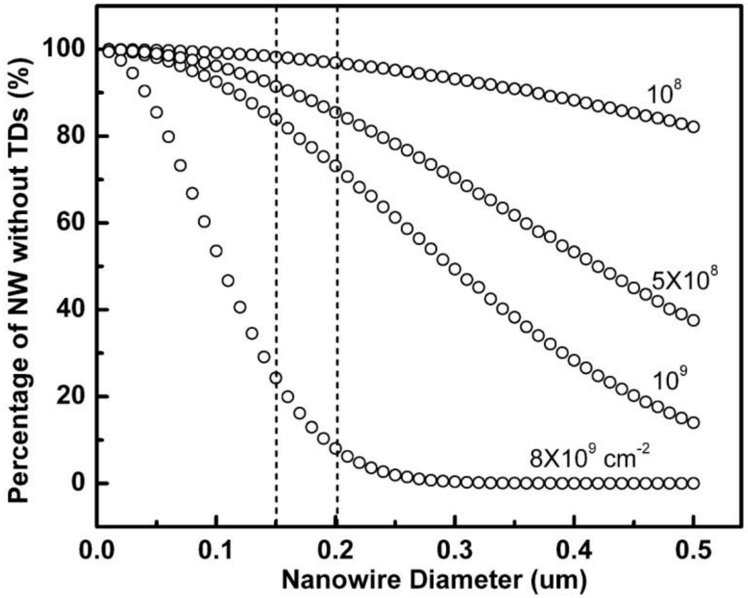




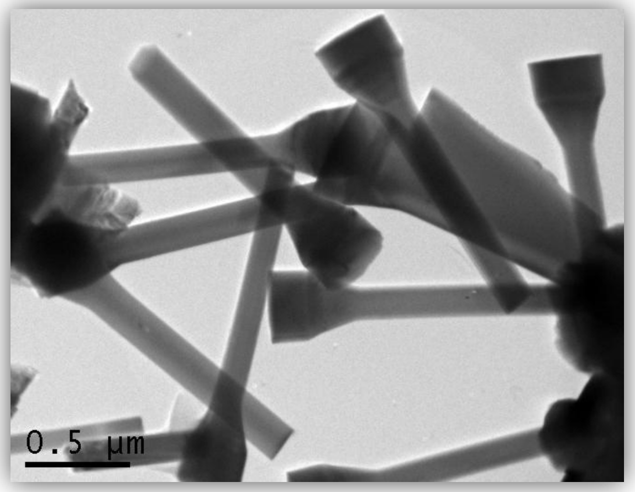
Q. Li et al., *Optics Express* **19**, 25528 (2011)

Top-down nanowire threading dislocations

Monte Carlo simulation



Bright-field TEM

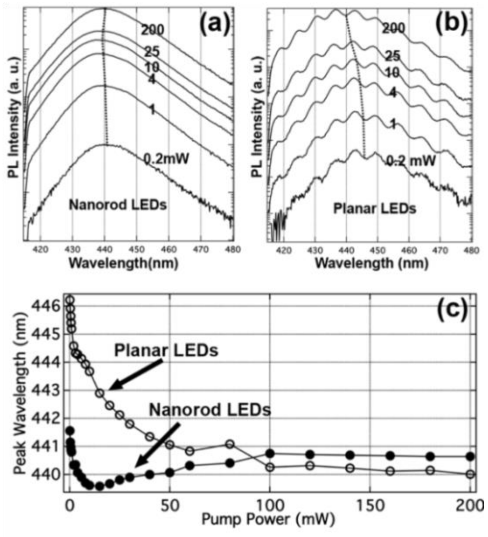


Nanowires etched from ~5e8 cm^{-2} planar LED

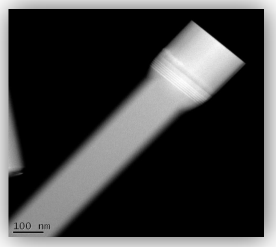
- Etched nanowires inherit the dislocation density of the parent film
- However, as the diameter approaches zero, the *fraction* of nanowires with one or more dislocations also approaches zero! [# TDs per rod ~ (TDD)×($A_{cross\text{-}section}$)]
- ~94% of nanowires ~150 nm in diameter from TDD~5e8 cm^{-2}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

Optical performance – axial nanowire LEDs vs. planar LED

InGaN peak position vs pump power

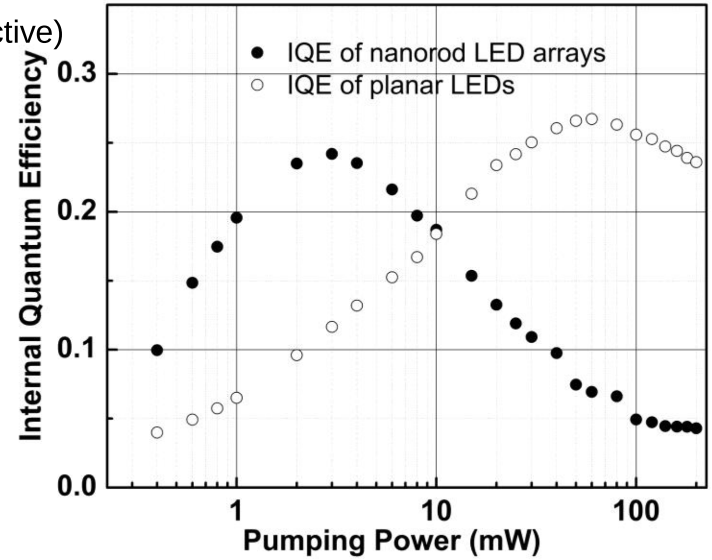


413 nm pump (InGaN selective)



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

IQE – nanowire vs Film



PL, IQE: K. Westlake, M. Crawford

- 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)

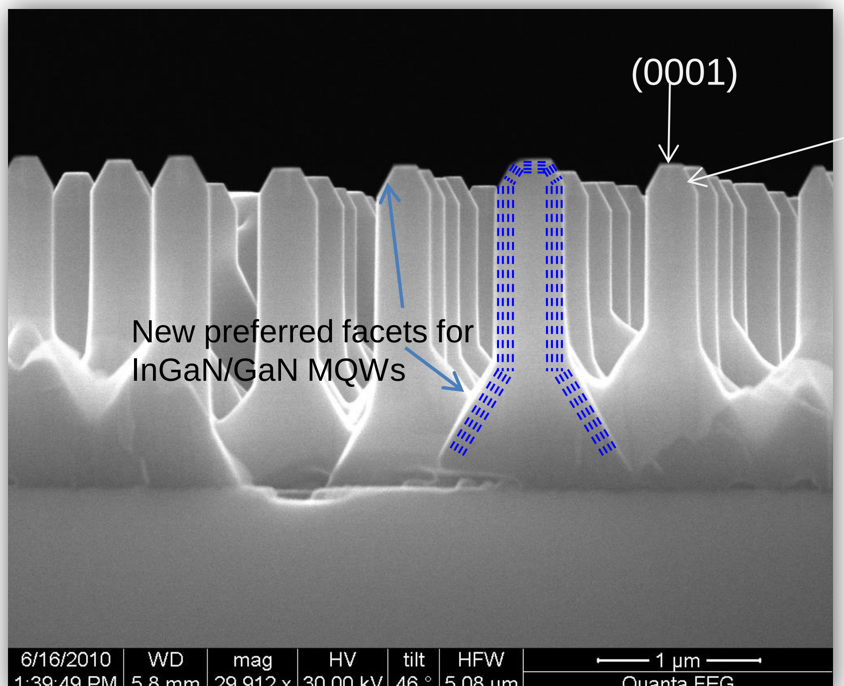
No obvious benefit for LED performance/cost ratio (loss of device area), but possibility for very high IQE single NW emitters for nanophotonic applications

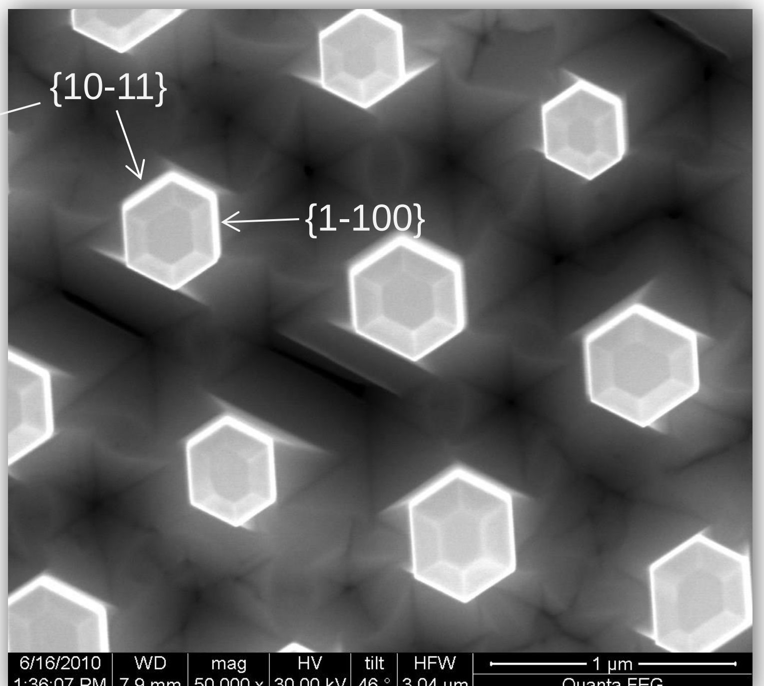
Radial core-shell InGaN/GaN MQWs on top-down NWs

<u>Radial core-shell NW LEDs:</u>

- Much higher active area than axial NW or planar structures
- Reduced strain InGaN growth for higher In incorporation
- Being pursued by a number of groups/companies (MOCVD)

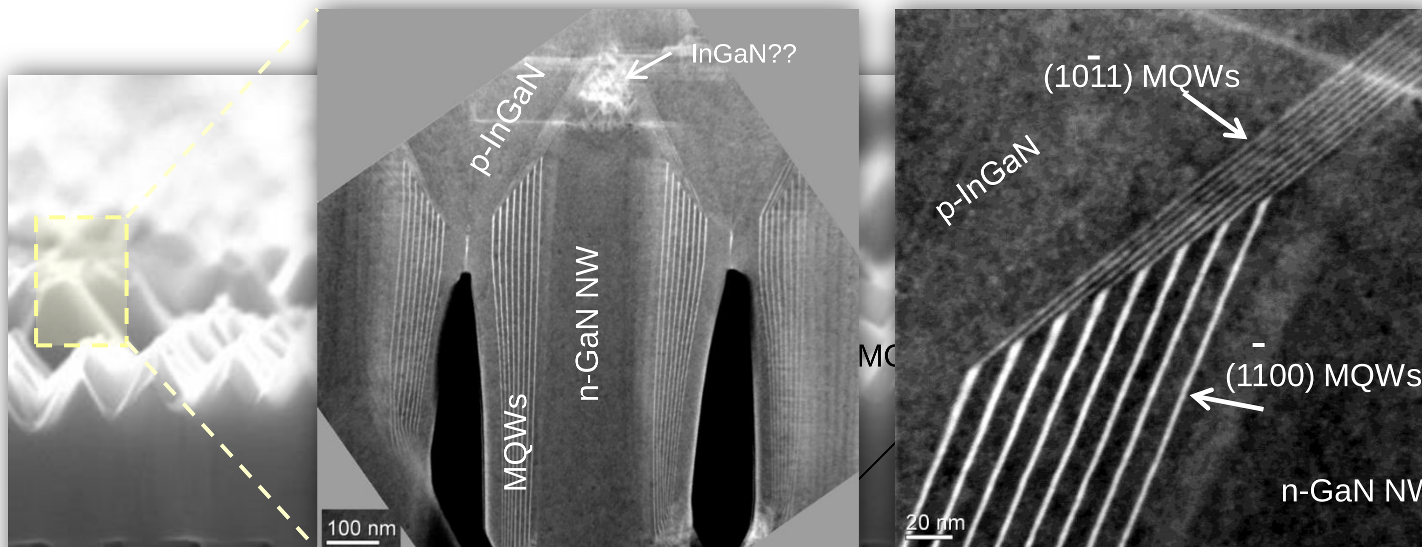
After 5-period MQW GaN/InGaN shell growth



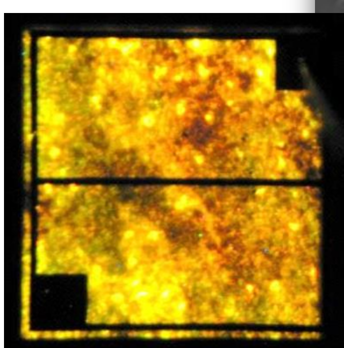


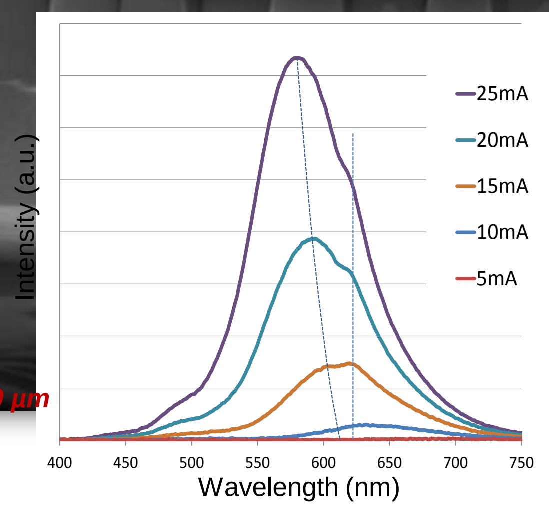
New semipolar facets form with InGaN/GaN MQW growth

Electrically injected core-shell nanowire based "3D" LED emitting at yellow-red wavelengths



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



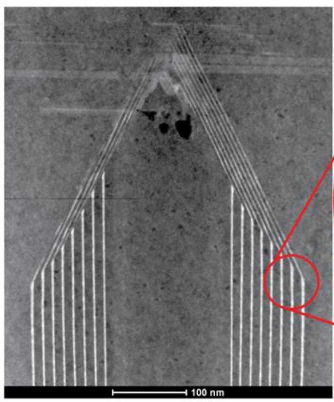


- Yellow-red electroluminescence!
- Two EL peaks:
  - 615 nm (const) red
  - 600-565 nm (shift) yellow

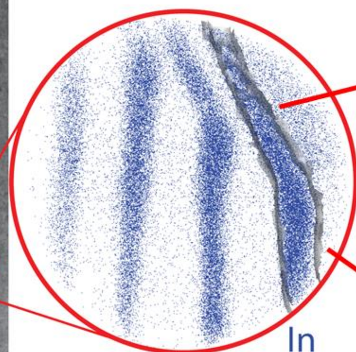
G. T. Wang et al., *Phys. Stat. Solidi A,* **211**, 748 (2014)

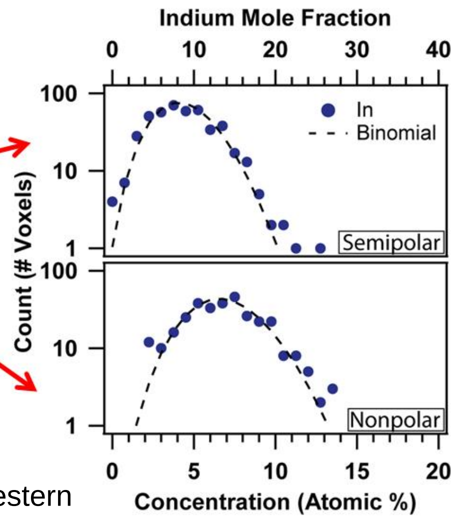
3-D Mapping of Quantum Wells in a GaN-InGaN Core-Shell Nanowire – a Correlated Study

STEM-EDS



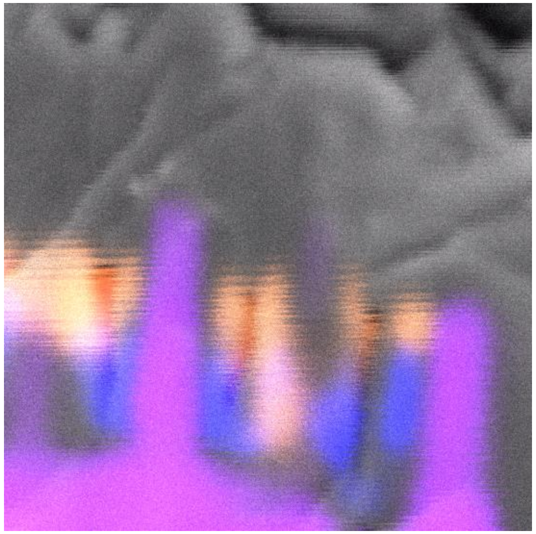
Atom probe tomography (APT)





APT: L. Lauhon, Northwestern

Cathodoluminescence



365 nm, 402 nm, 442 nm

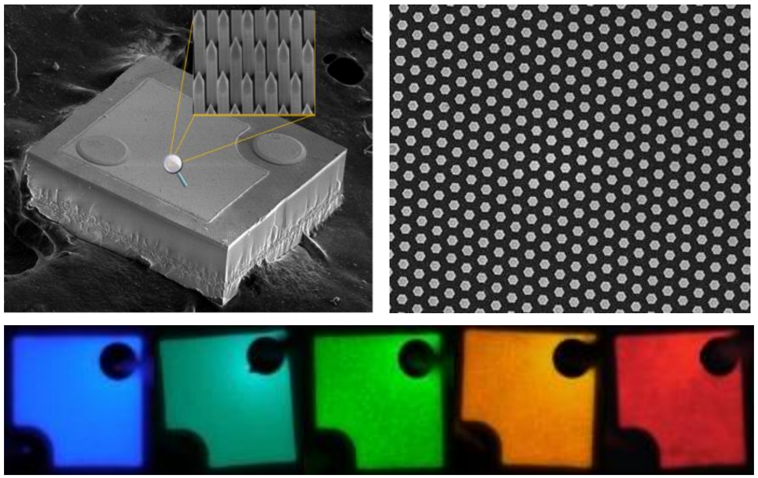
Riley, J.; Padalkar, S.; Li, Q.; Lu, P.; Koleske, D. D.; Wierer, J. J.; Wang, G. T.; Lauhon, L. J., *Nano Lett.* **14**, 4317 (2013).

- Nonpolar and semipolar QWs from radial NW LED were imaged by APT and correlated with scanning TEM and cathodoluminescence data
- Results suggest yellow-red emission originates from In-rich tip regions, not sidewalls
- How to get long wavelength emission from m-plane sidewalls?

Nanowire LED Commercialization/Industry Efforts

glō

- Spun out of Lund University, Sweden
- Engineering Center in Sunnyvale, CA



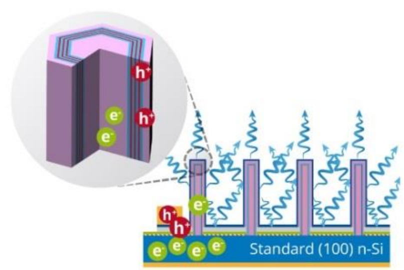
www.glo.se Bottom-up grown nanowires with *m*-plane MQWs

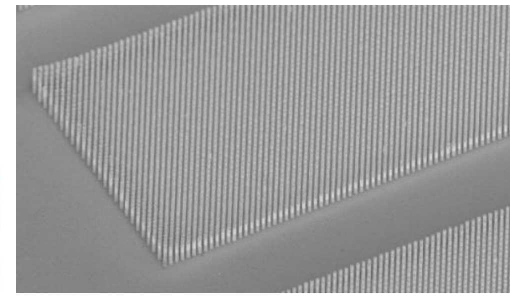
From ICNS-10 (Aug 2013):

- Claims ~80% QW IQE (via LT PL)
- Estimated 80% light extraction eff. Based on FDTD models, device data
- EL: 459 nm (1 mA), 452 nm (20 mA)
- Showed green, amber, red NW LED images (no performance data)

Aledia SA

- Spun out of CEA-LETI (Grenoble, France)

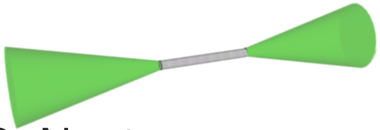




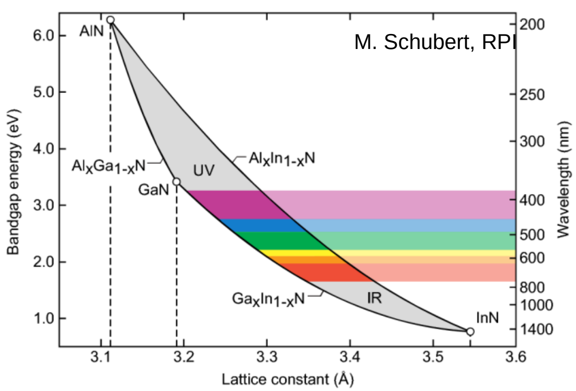
http://www.eetimes.com/document.asp?doc_id=1280676

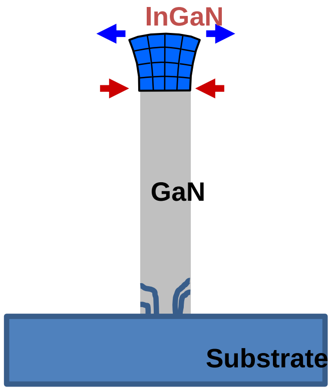
- goal is to manufacture 3D WireLED^{TM} for on thin silicon wafers ≥ 8 inches in diameter
- developed over a six-year period at the LETI research institute in Grenoble and the claim is production cost will be one quarter that of conventional planar LEDs.
- Announced 200-mm diameter wafers and raised $13 million in a first round of venture capital finance (Mar '13)
- Claims three times more light per square millimeter of planar area
- Claims red, green WireLEDs possible and enable phosphor-free LEDs.
- Purchased Veeco's TurboDisc K465i MOCVD reactors

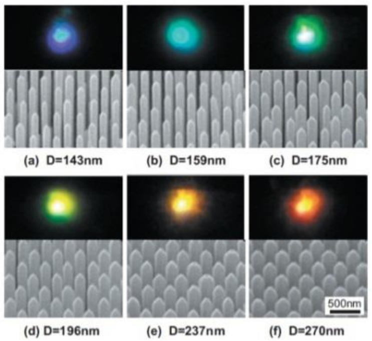
Nanowire Lasers



- Nanowire forms a freestanding, low loss optical cavity
- Optically pumped nanowire lasers: ZnO, CdS, GaAs, GaSb, GaP, GaN, etc.
- Most commonly Fabry-Perot type lasing from end facets
- Compact and low power due to small mode volume
- Strain accommodation in GaN nanowires opens up possibility of high efficiency lasers over a wide spectral range (incl. green and yellow) through facile In incorporation







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

See also:

Ma, Y.; Guo, X.; Wu, X.; Dai, L.; Tong, L. "Semiconductor nanowire lasers," *Adv. Opt. Photon.* **2013,** 5, (3), 216-273

C.Z. Ning, Semiconductor nanowire lasers, Semiconductors and Semimetals v. 86 p455, Chapter 12 (2012)

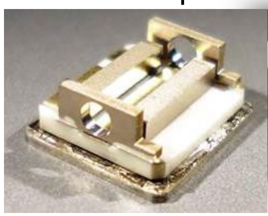
S. Arafin, X. Liu, Z. Mi, "Review of recent progress of III-nitride nanowire lasers," *J. Nanophotonics* **7,** 074599-1 (2013)

Why Nanowire Lasers?

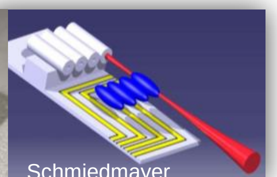
Integrated coherent nanophotonic elements for communications, sensing, imaging, lithography, lighting, etc.

Integrated nanophotonics, atom trapping, optical MEMS

atom trap



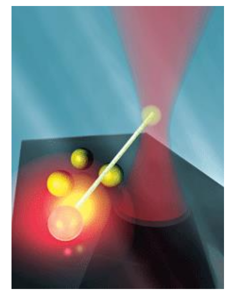
P. Schwindt (1725)

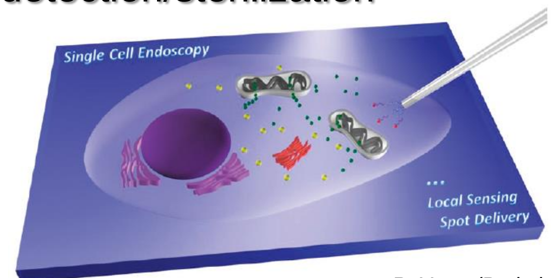


J. Ford, Lucent

Small UV-visible laser for trapping, interconnects, switching, etc.

Optical nanoprobes & chem/bio detection/sterilization

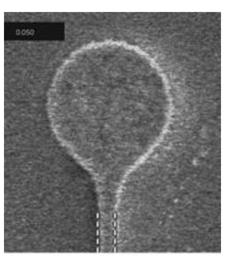




P. Yang (Berkeley)

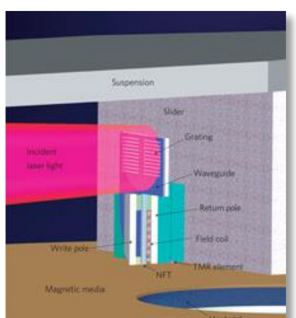
Sub-λ resolution probes for localized excitation, detection

Nanolithography & Heat-assisted Magnetic Recording



Nature Photonics 3, 220 (2009)

Seagate

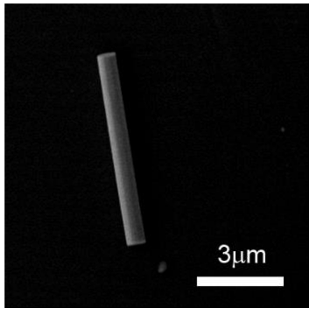


Subwavelength & efficient laser spot

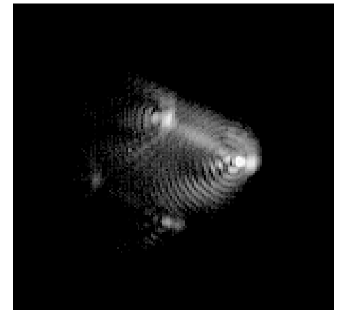
Lighting, projection, & holographic displays

Low-power, speckle-free low coherence (random laser), multicolor pixels

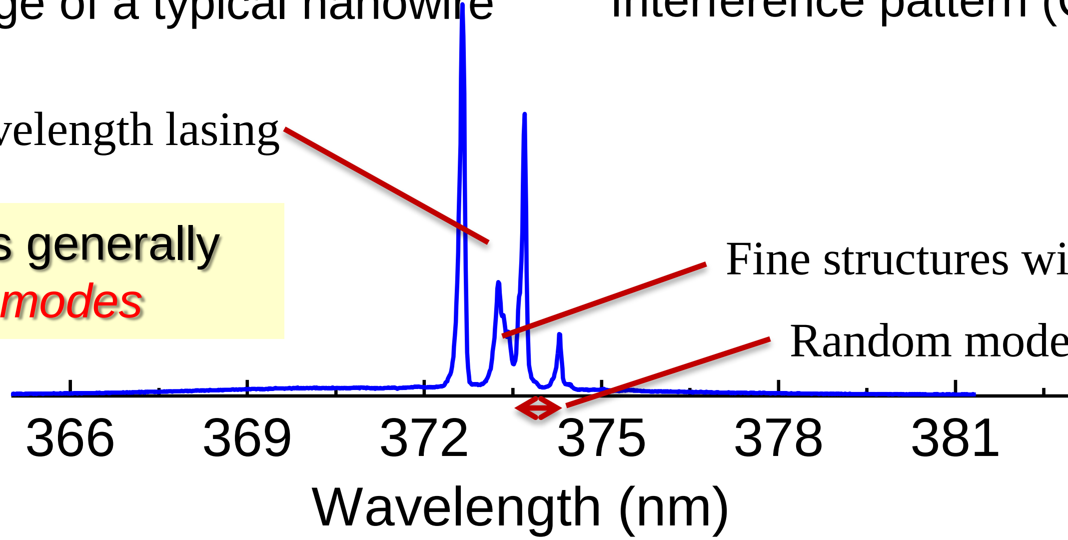
Typical GaN Nanowire Lasing: Multimode Operation



SEM image of a typical nanowire



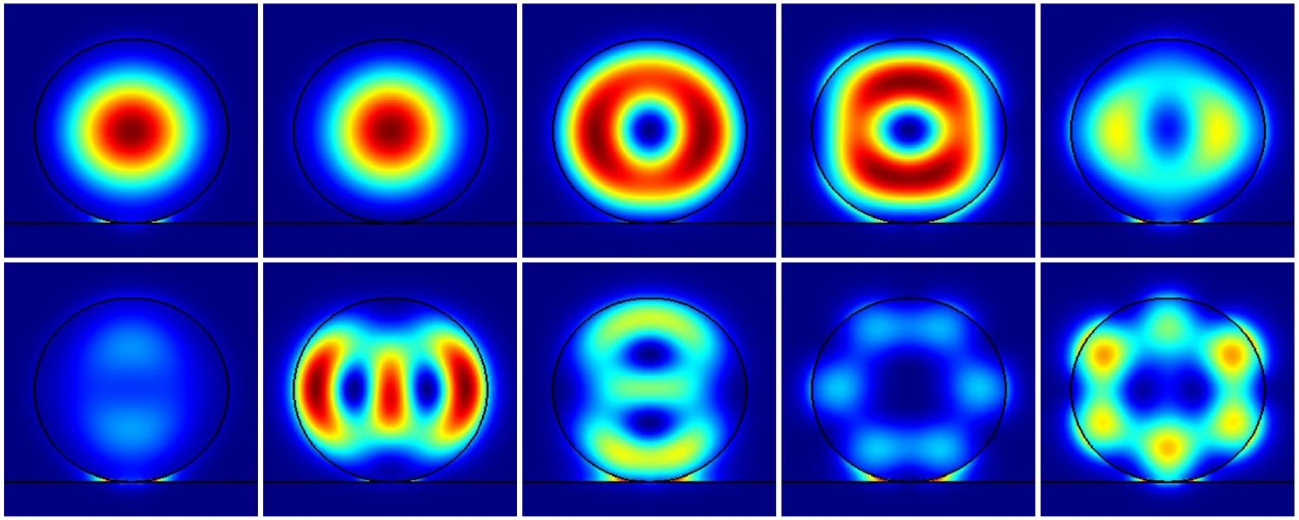
Interference pattern (CCD image)



Nanowire lasers generally exhibit *multiple modes*

- *Single mode* behavior desired for highest resolution and beam quality; may also reduce lasing threshold due to reduced mode competition

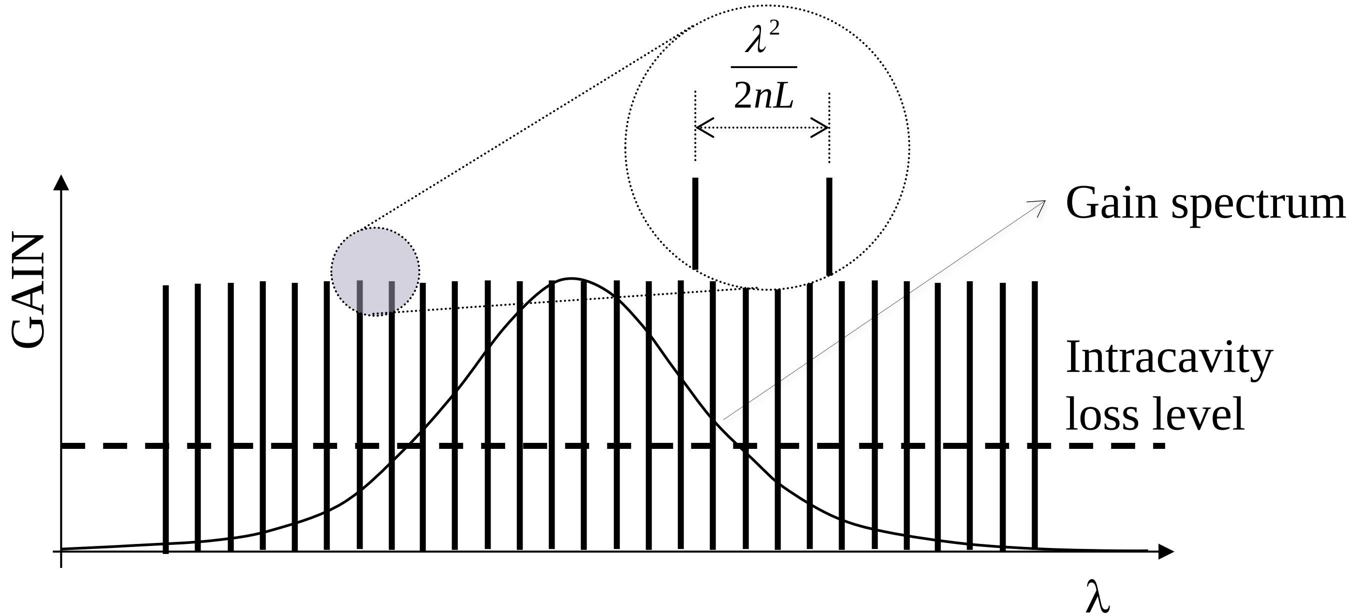
Why Multimode Lasing Occurs



Calculated transverse modes supported by a 300 nm diameter GaN nanowire.

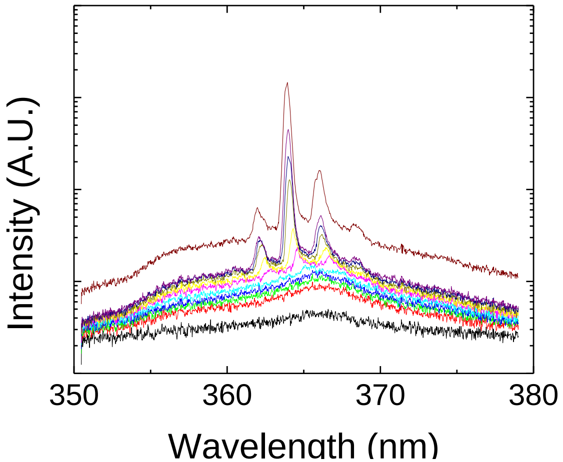
- ✓ Multiple transverse mode waveguide;
- ✓ Single-mode cutoff diameter ~120 nm;
- ✓ Lasing in multiple transverse mode operation
- ✓ Polarization is mode-dependent

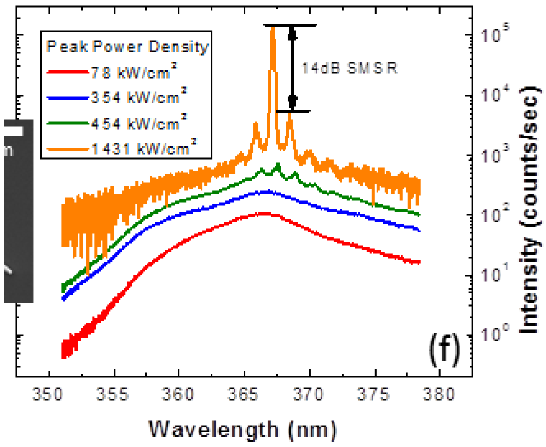
Why Multimode Lasing Occurs

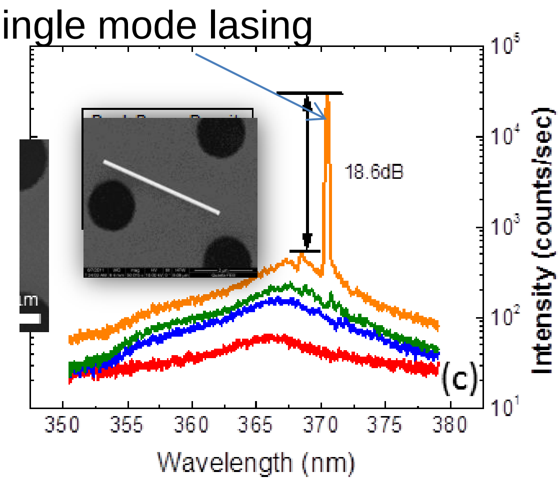


- ✓ Gain bandwidth ~7 nm;
- ✓ Longitudinal mode spacing ~1 nm;
- ✓ Several modes located in the gain spectrum;
- ✓ Multiple longitudinal mode oscillation

Method 1: Single-mode NW laser via geometry control







~500 nm x **4.7µm (short)** | **145 nm** x 7.2 µm **(skinny)** | **~130nm x 4.7µm (short & skinny)**

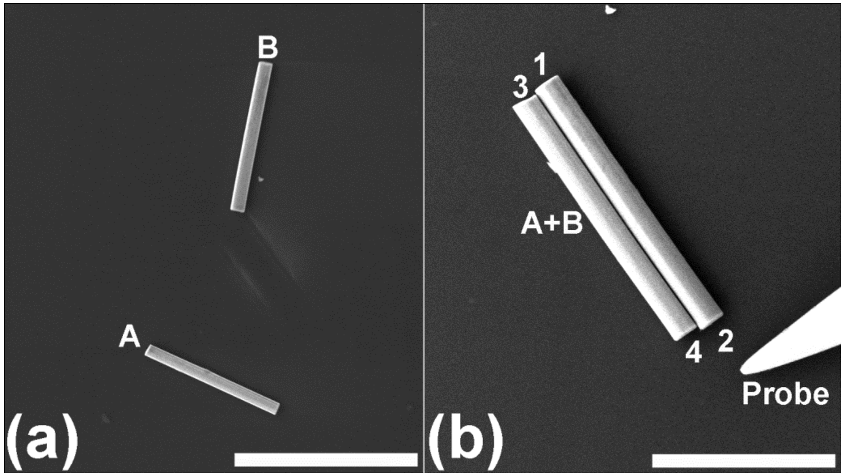
NW diameter x length

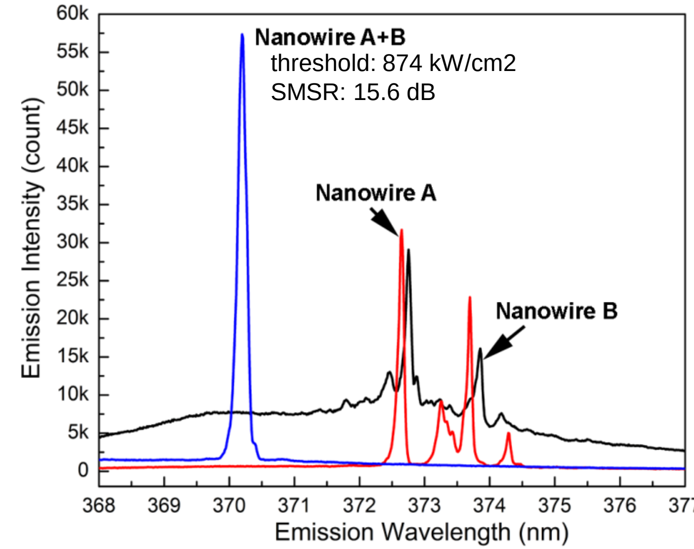
Q. Li et al., *Opt. Exp.* ***20*** *17873 (2012)*

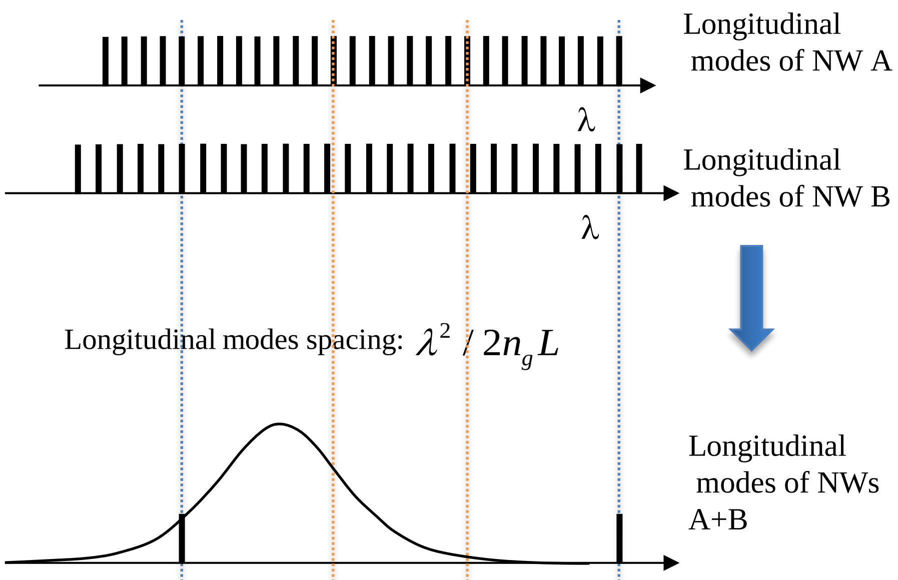
Single-mode: Narrow-Linewidth (<0.1 nm), 18.6 dB Side Mode Suppression Ratio

- Reduction of diameter (<~130 nm): reduction of transverse modes
- Reduction of length: (<~6 µm length): reduction of longitudinal modes
- **Reduced dimensionality leads to single mode lasing**

Method 2: Single-mode lasing via coupled nanowire cavities







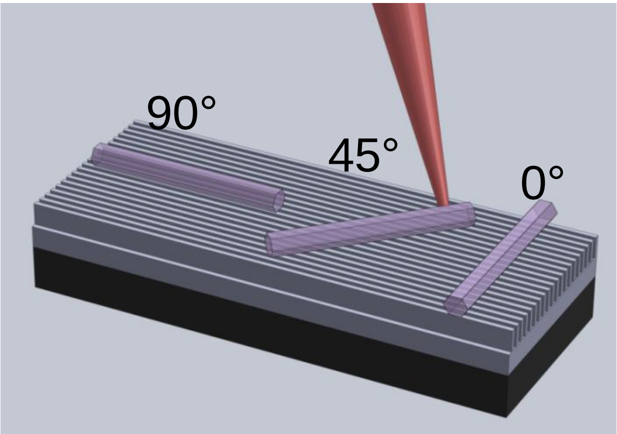
Longitudinal mode spacing greatly enhanced!

- Individual large NWs shows multiple modes.
- Coupled nanowires show single mode!
- Vernier effect – only resonant modes survive

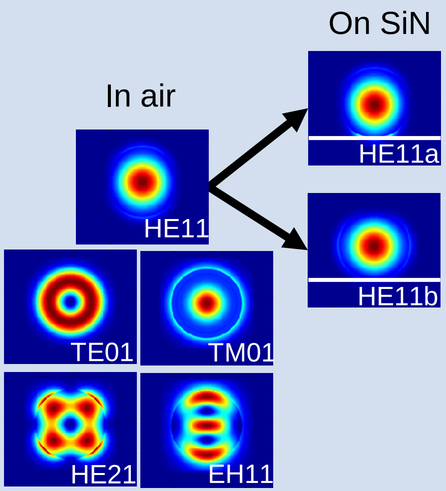
H. Xu et al., *Appl. Phys. Lett.* **101** 113106 (2012)

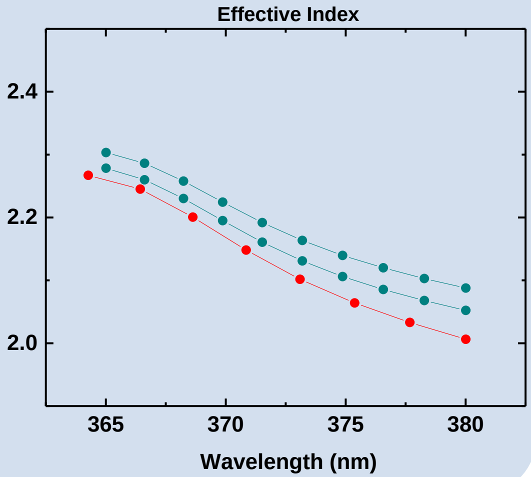
Method 3: Distributed Feedback (DFB) Nanowire Laser

- Standard method for mode control in semiconductor lasers
- Here, single nanowires are coupled to grating substrate to achieve distributed feedback
- Stop-band position and width tuning via nanomanipulation of the angular alignment

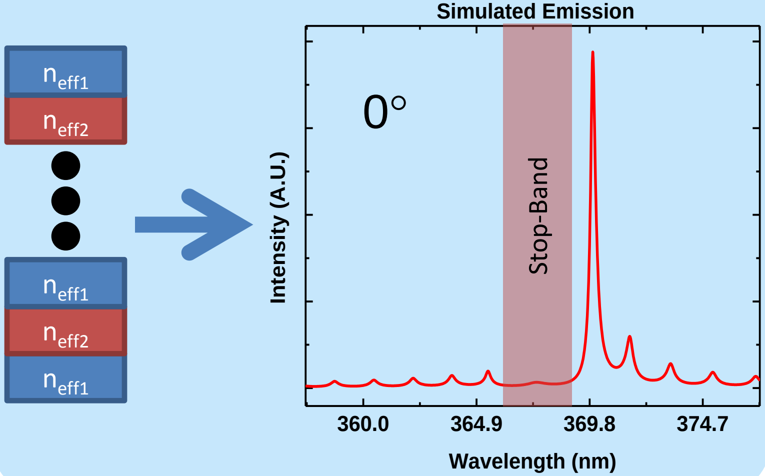


Mode Calculations

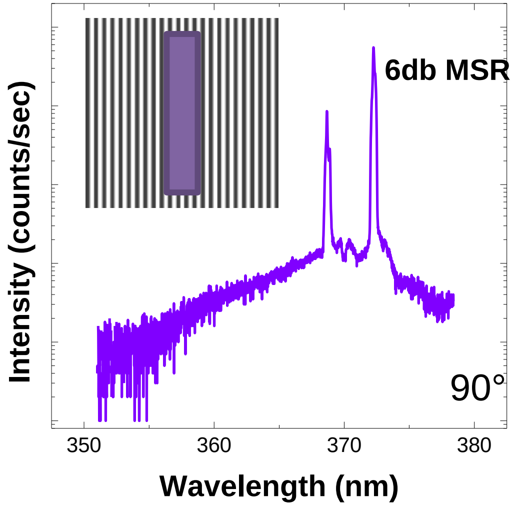


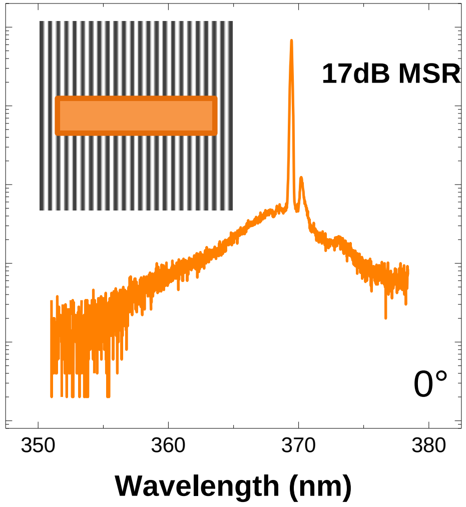


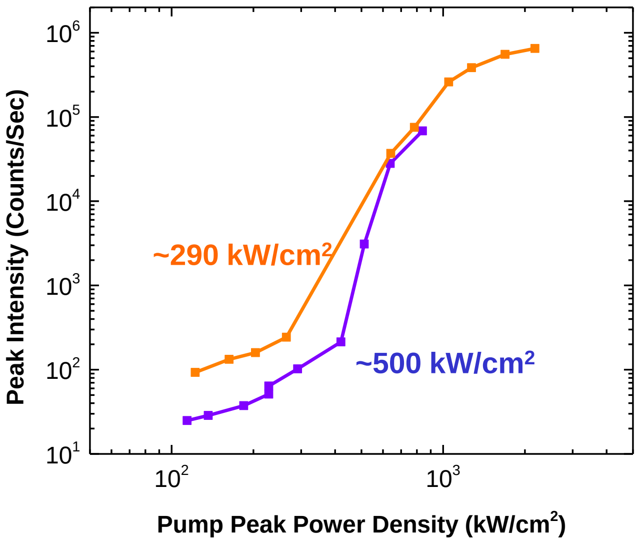
Transfer Matrix Method

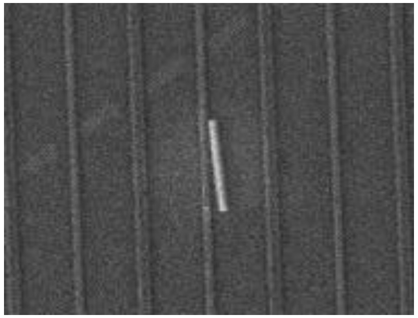


Method 3: DFB Nanowire - Optical Results

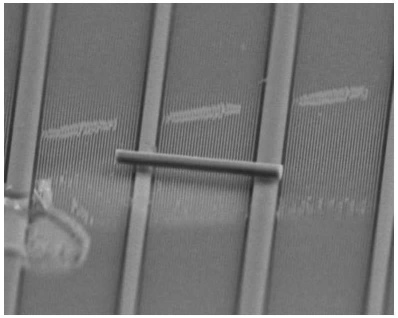








NW diameter ~180 nm

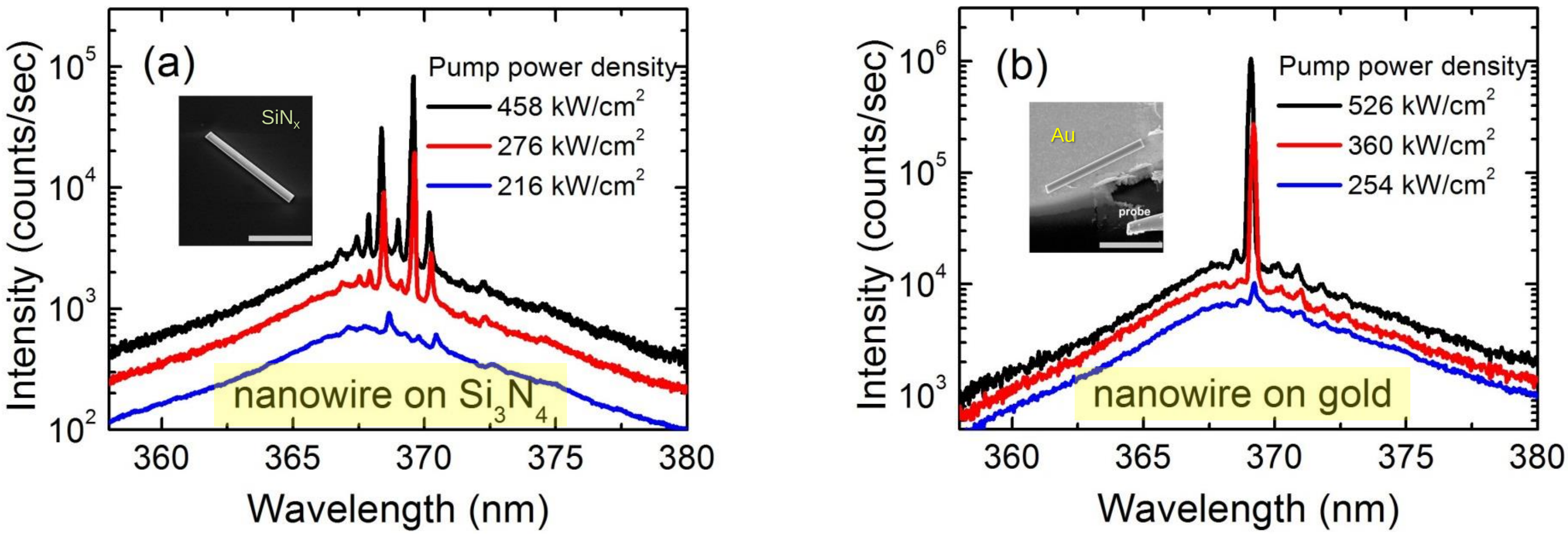


- At the designed alignment single-mode lasing was achieved with a 17dB mode suppression ratio.
- Observed reduction in the lasing threshold

J.B. Wright, S. Campione, S. Liu, J.A. Martinez, H Xu, T.S. Luk, Q. Li, G.T. Wang, B.S. Swartzentruber, L.F. Lester, and I. Brener, APL 104 (4), 041107 2014.

Method 4: Metal substrate induced single-mode nanowire lasing

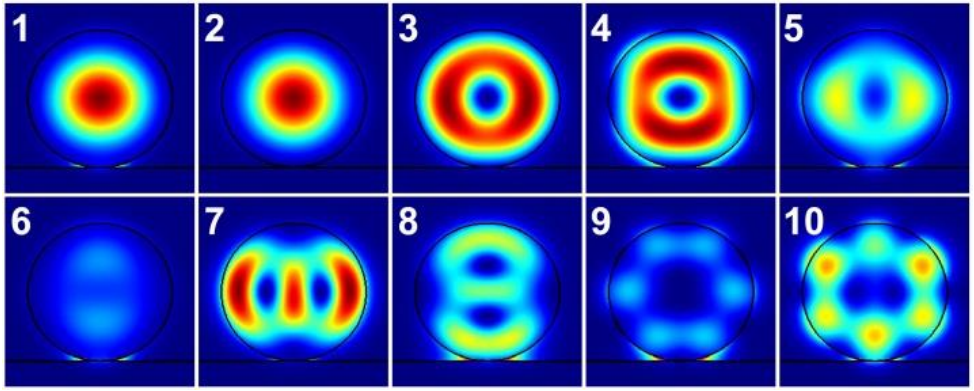
diameter~350 nm, length ~5.3 µm



- NWs on Si_3N_4 show multi-mode lasing
- **Same NWs moved onto gold-coated spot show single-mode lasing**
- Increase in lasing threshold of only ~13%

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

Gold-substrate induced single-mode lasing



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

Transverse modes supported by a 300nm diameter nanowire on gold substrate.

Only surviving mode (Mode 2)

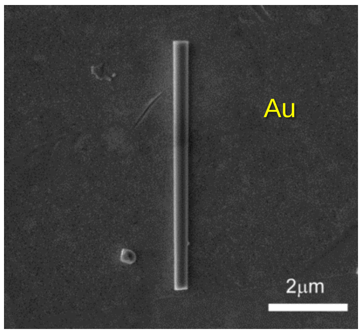
	Mode 1	Mode 2	Mode 3	Mode 4	Mode 5
Loss (dB/cm)	8151	1730	4806	7109	16041
	Mode 6	Mode 7	Mode 8	Mode 9	Mode 10
Loss (dB/cm)	34858	7551	22205	29175	28706

Propagation loss of different transverse modes.

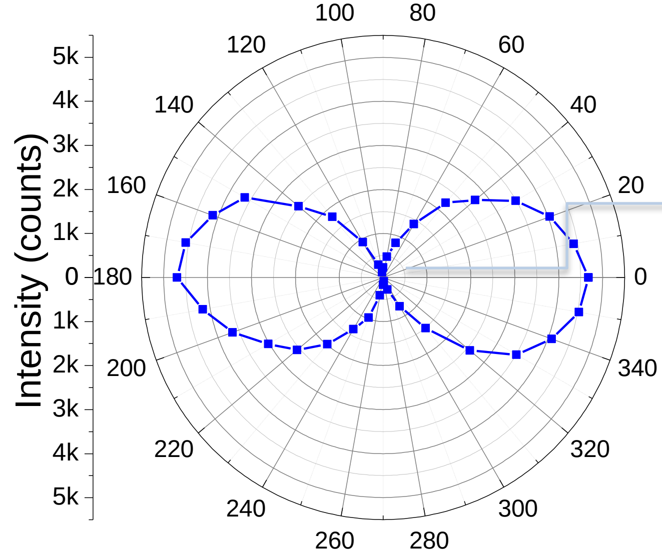
Metal substrate generates a mode-dependent propagation loss

Polarization Control: Nanowire on Gold Substrate

Polarization control (linear polarization) desired for many applications

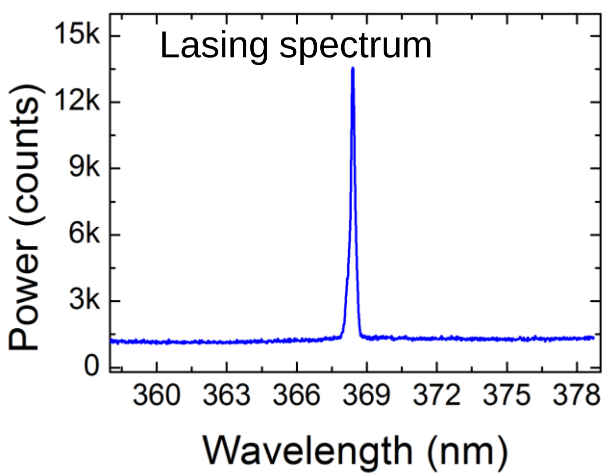


nanowire on **gold**



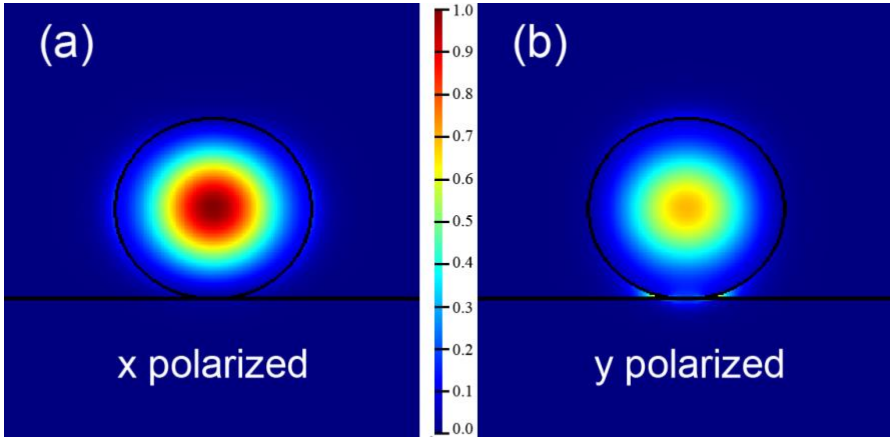
Indicative of linear polarization

Intensity vs. the rotation of the polarizer



- ✓ Single-wavelength lasing
- ✓ Linear polarization
- ✓ Large cross-polarization suppression ratio (CPSR): 21:1
- ✓ 0^{o} polarization angle

Why Does Linear Polarization Happen on Gold?



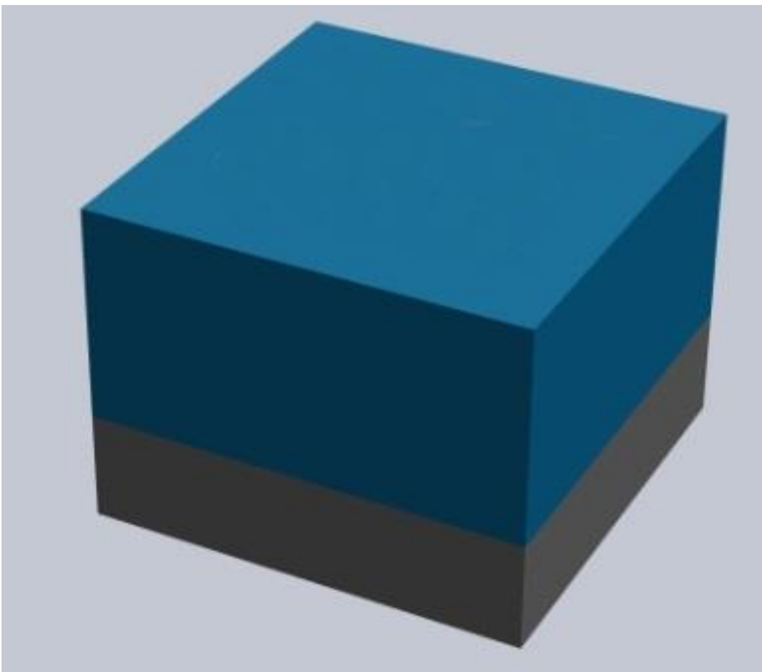
Fundamental modes supported by the nanowire-gold geometry

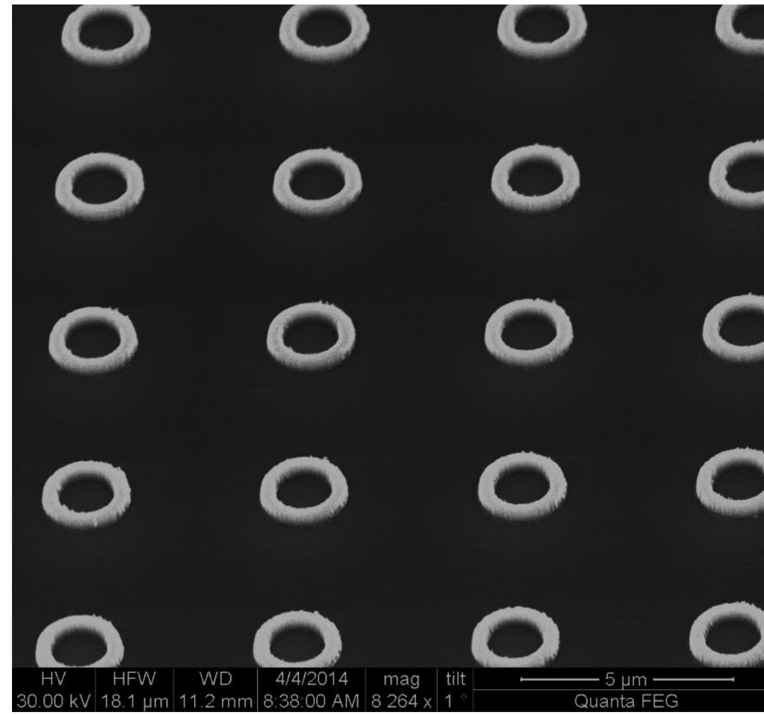
- ✓ Differently polarized modes experience different cavity losses
- ✓ Loss for x-polarized mode: 0.36 dB/mm
- ✓ Loss for y-polarized mode: 2.11 dB/mm

A nanowire on gold substrate experiences a larger cavity loss for the perpendicularly polarized mode, leading to *linear polarization* with a fixed polarization parallel to the substrate

Cross-sectional shape control- GaN nanotubes

EBL Patterning PMMA Lift-off





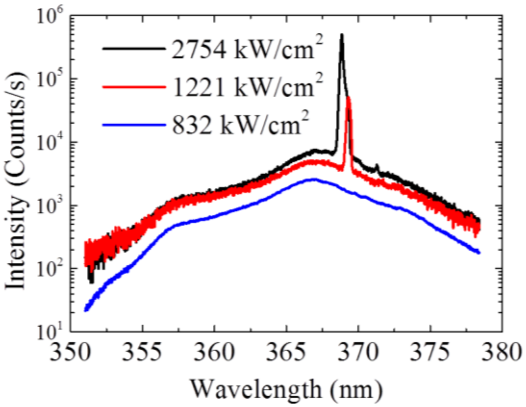
Details of the process, see

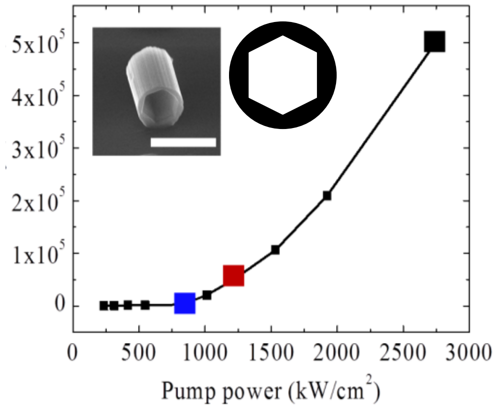
Li, Qiming, et al. *Optics express* 19.25 (2011): 25528-25534.

Li, Qiming, et al. *Optics express* 20.16 (2012): 17873-17879.

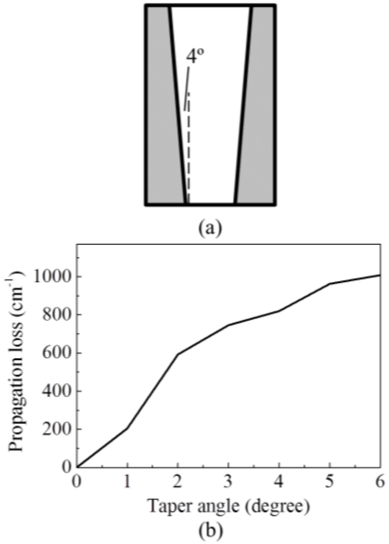
GaN nanotube lasers

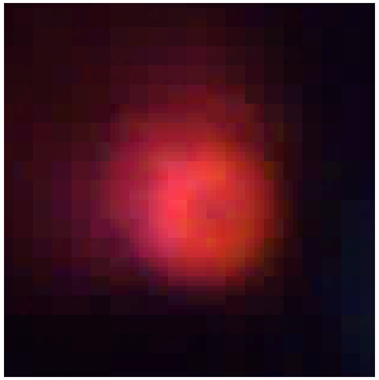
- Length: 4µm
- Outer Diameter: 1.3µm
- Thickness: 150nm





Threshold pump density = 1055 kW/cm^2





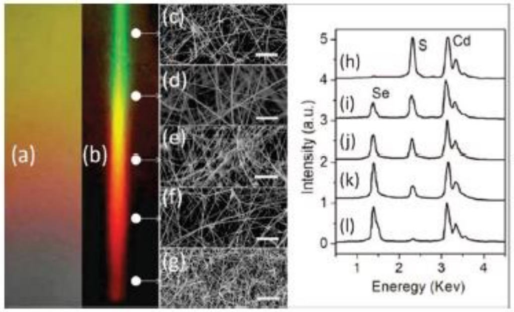
CCD image far field lasing emission

- Higher threshold of nanotube vs. nanowire may be due in part to inner taper
- Hollow nanotube geometry potentially useful for optofluidic/biosensing applications
- Observed annular shaped emission may have potential applications for atom trapping, stimulated emission depletion (STED) microscopy, etc.

C. Li et al., in preparation

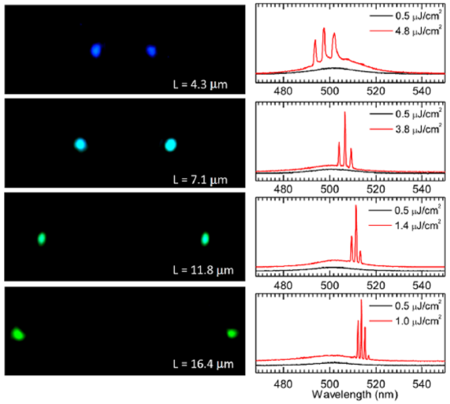
Previous wavelength “tuning” (selection) in nanowire lasers

CdSSe NWs with different compositions across substrate



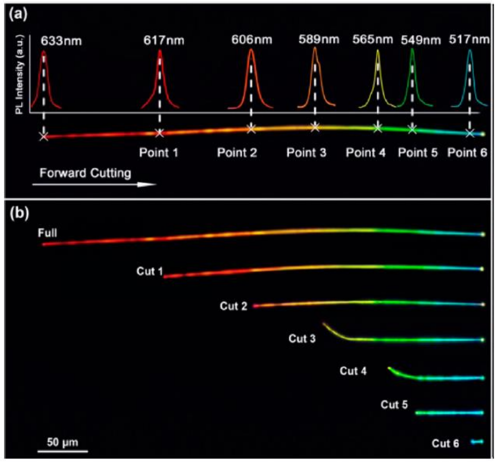
Pan, A. et al., *Nano Letters* **9**, 784 (2009). (ASU)

Different length CdS NWs (intrinsic self absorption effect)



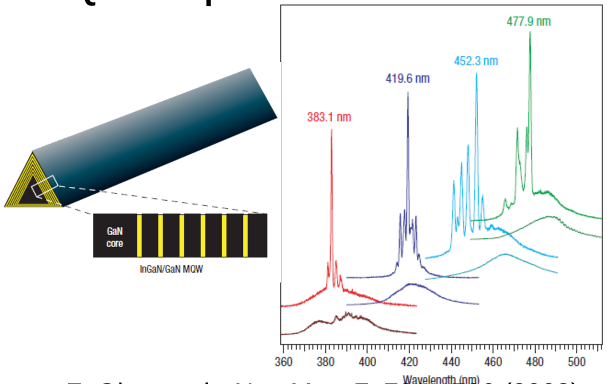
Liu, X et al., *Nano Letters* **13**, 1080 (2013) (Nanyang Tech. U.)

Cutting bandgap graded CdSSe NWs



Yang, Z. et al. *Nano Letters* **14**, 3153 (2014)

Core-shell NWs with different InGaN MQW compositions

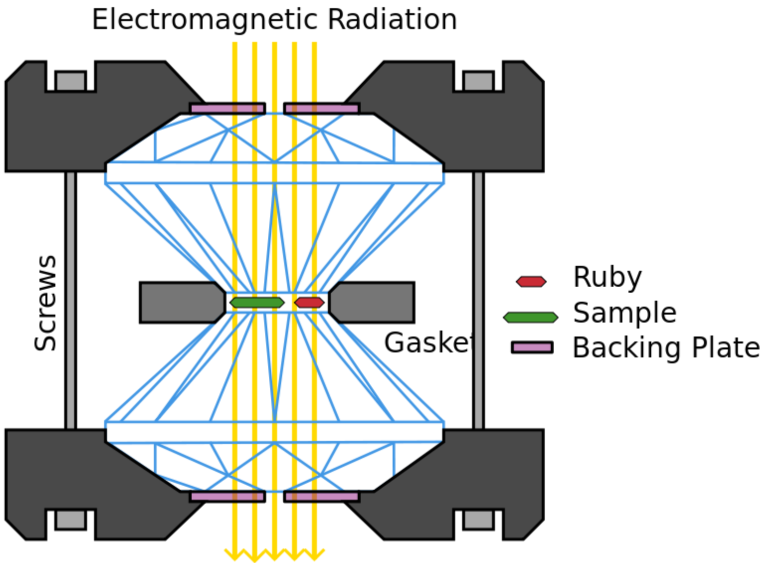


F. Qian et al., *Nat. Mat.*, **7**, 701 - 706 (2008) (Harvard)

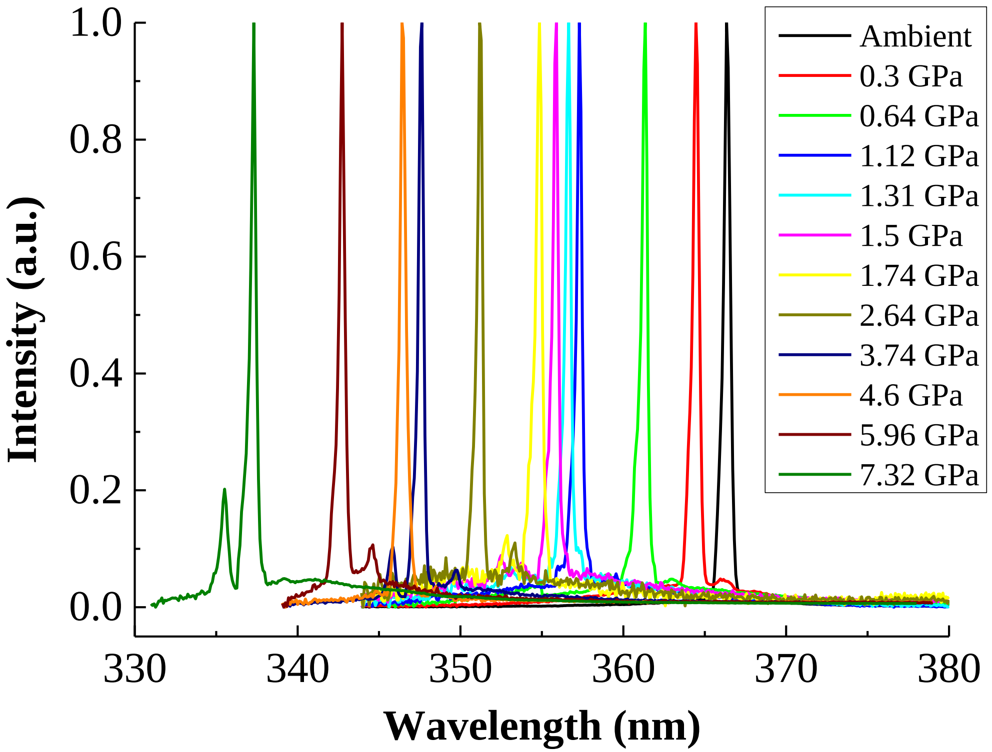
Wavelength selection in nanowire lasers has been demonstrated, but not true continuous and reversible tuning within the same nanowire

Continuous & dynamic bandgap/wavelength tuning in GaN NW lasers via hydrostatic pressure

Hydrostatic pressure medium (silicone oil) transmits pressure isotropically

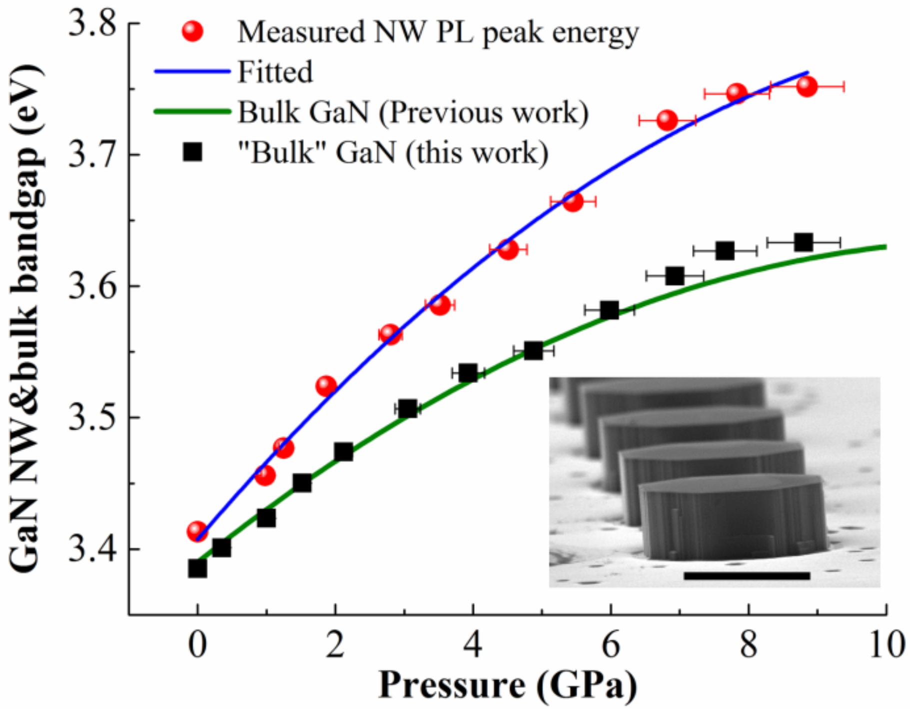


http://en.wikipedia.org/wiki/Diamond_anvil_cell



- Applied pressure induces bandgap increase
- Continuous and reversible tuning ~30 nm lasing tuning demonstrated over ~7.3 GPa (previous works were wavelength selection)
- Well defined fine tuning (<0.1nm) possible with current setup

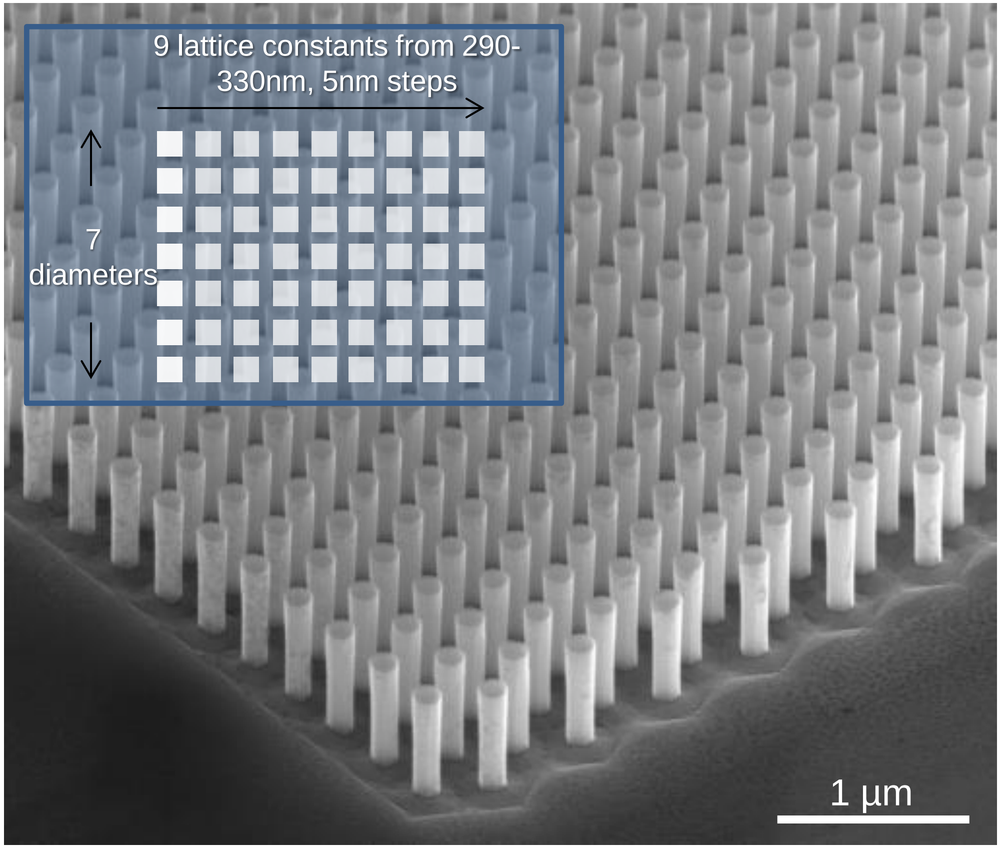
Continuous & dynamic bandgap/wavelength tuning in GaN NW lasers via hydrostatic pressure



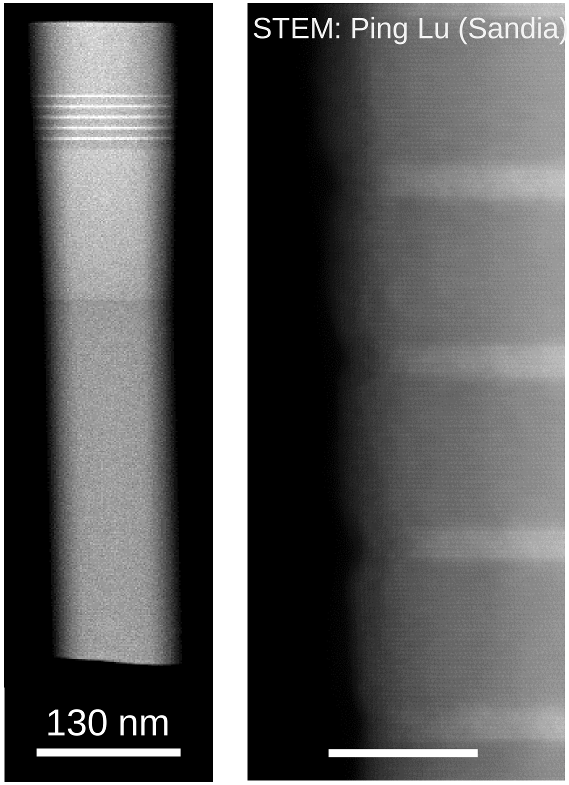
- Pressure coefficient ~40% higher for GaN NWs vs GaN micropillars or bulk GaN, allows for larger tuning range.

Wavelength tuning in III-N Nanowire Photonic Crystal Lasers

<u>Motivation</u>: 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

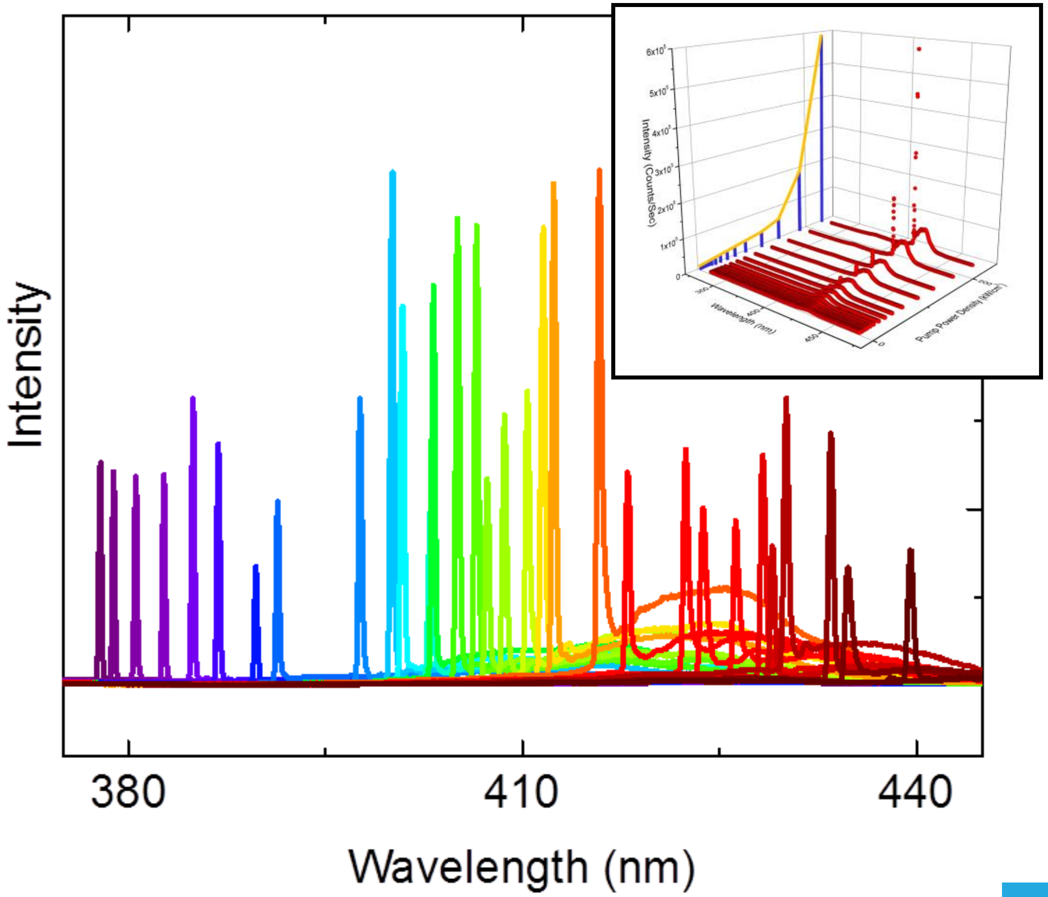


NW STEM images: 5x MQW InGaN emission centered at 420nm, $In_{0.02}GaN$ underlayer

Wavelength tuning in 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

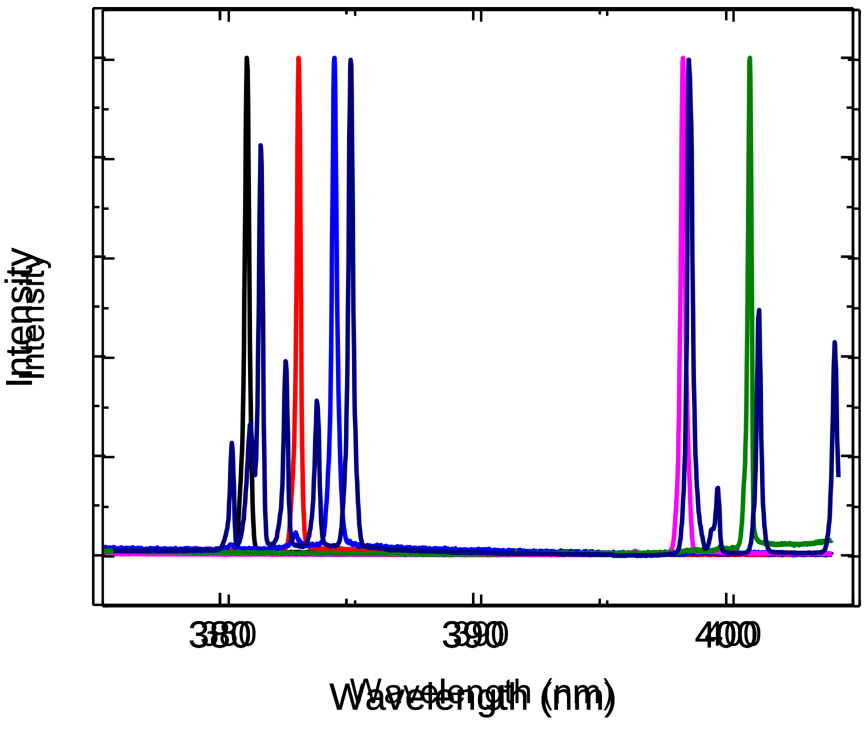
61 color nanowire laser array



- 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)

J.B. Wright et al., Scientific Reports 3, Article number: 2982 (2013) doi:10.1038/srep02982

III-N Nanowire Photonic Crystal Lasers



J.B. Wright et al., Scientific Reports 3, Article number: 2982 (2013) doi:10.1038/srep02982

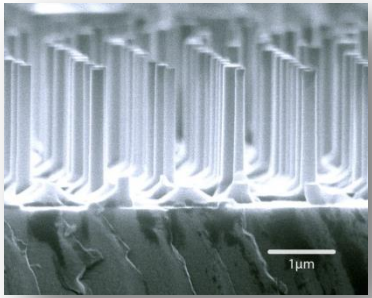
pitch: 320 nm
diameter: 130-140nm

10 um

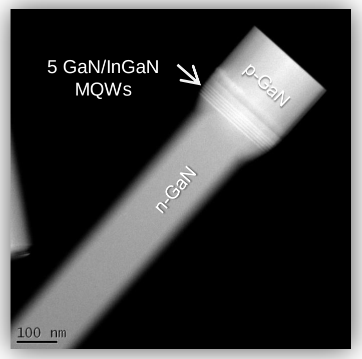
SSLS EFRC

Summary – Top-down III-nitride nanowires

Precision top-down fabrication *controlled geometries*

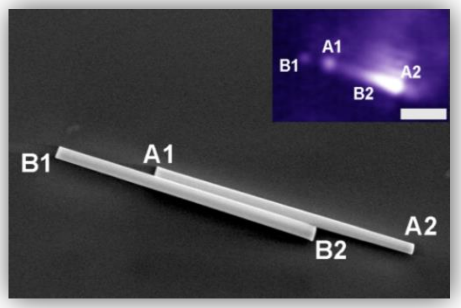


Axial nanowire LED "flashlight"



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

Mode & polarization control in GaN nanowire lasers

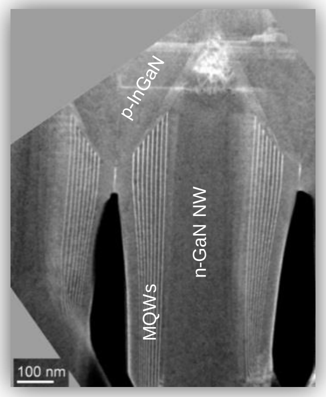


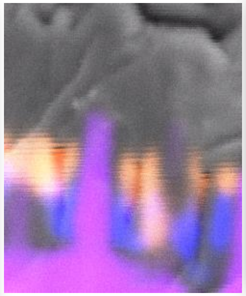
Q. Li et al., *Optics Express* **20** 17874 (2012)

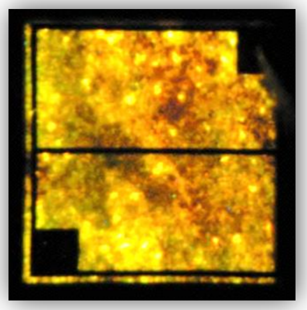
H. Xu et al., *Appl. Phys. Lett.* **101** 113106 (2012)

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

Vertically integrated radial nanowire LEDs

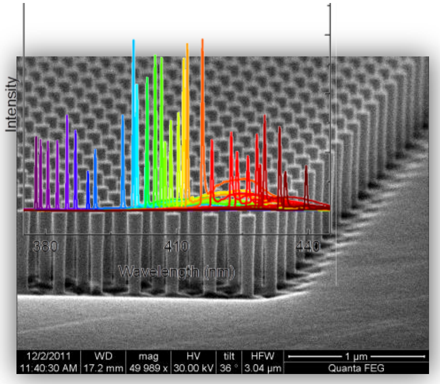


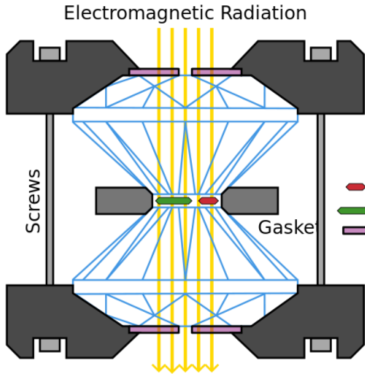




J. Wierer et al., *Nanotechnology* **23** 194007 (2012)
Riley, J.; *Nano Lett.* **14**, 4317 (2013).
G. T. Wang et al., *Phys. Stat. Solidi A,* **211**, 748 (2014)

Tunable wavelength nanowire [photonic crystal] lasers





J.B. Wright et al., *Scientific Reports* 3, Article number: 2982 (2013) doi:10.1038/srep02982

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

Jeffrey Figiel, Randy Creighton, Karen Cross –MOCVD growth, device processing & support

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

Karl Westlake, Mary Crawford – PL, IQE measurements

Daniel Koleske, Jonathan Wierer – LED growth, LED device fabrication

Igal Brener, Willie Luk, Weng Chow, Jeremy Wright, (Ph.D student), Huiwen Xu (Ph. D student), Changyi Li (Ph. D student), Sheng Liu, Steven Brueck, Ganesh Subramania – NW lasers

Jim Riley (Ph.D student), Sonal Padalkar, Lincoln Lauhon (Northwestern) – Atom Probe Tomography

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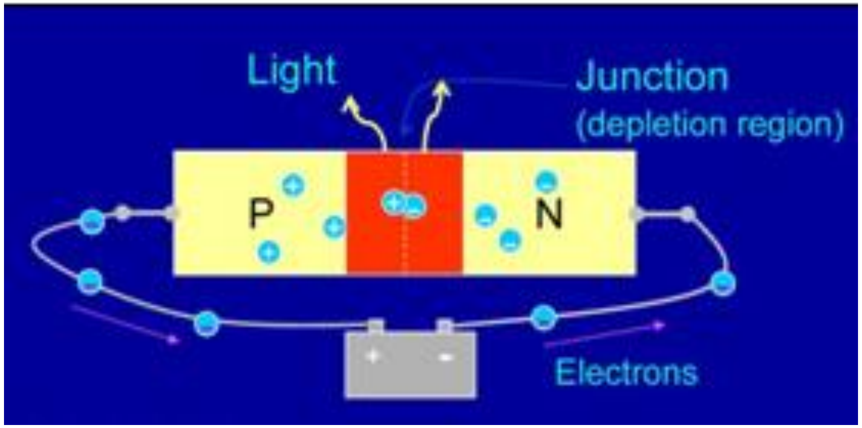
Contact: e-mail: gtwang@sandia.gov http://www.sandia.gov/~gtwang

Backup/Extra Slides

How LEDs work

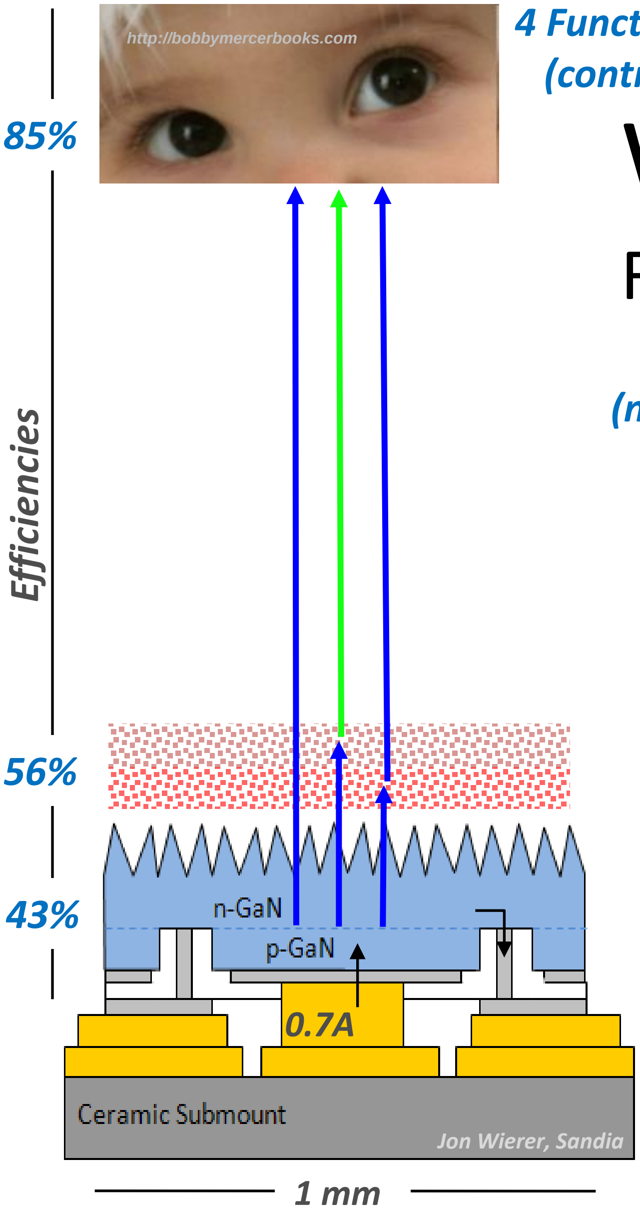


- An LED is a chip of semiconducting material treated to create a structure called a p-n (positive-negative) junction.

- Current flows from the p-side or anode to the n-side, or cathode. Charge-carriers (electrons and electron holes) flow into the junction.

- When an electron meets a hole, it falls into a lower energy level, and releases energy in the form of a photon (light).

- Depending on the composition of the semiconducting material, different colors of light are emitted.



4 Functional Light
(control of light in intensity, chromaticity, time, and space)

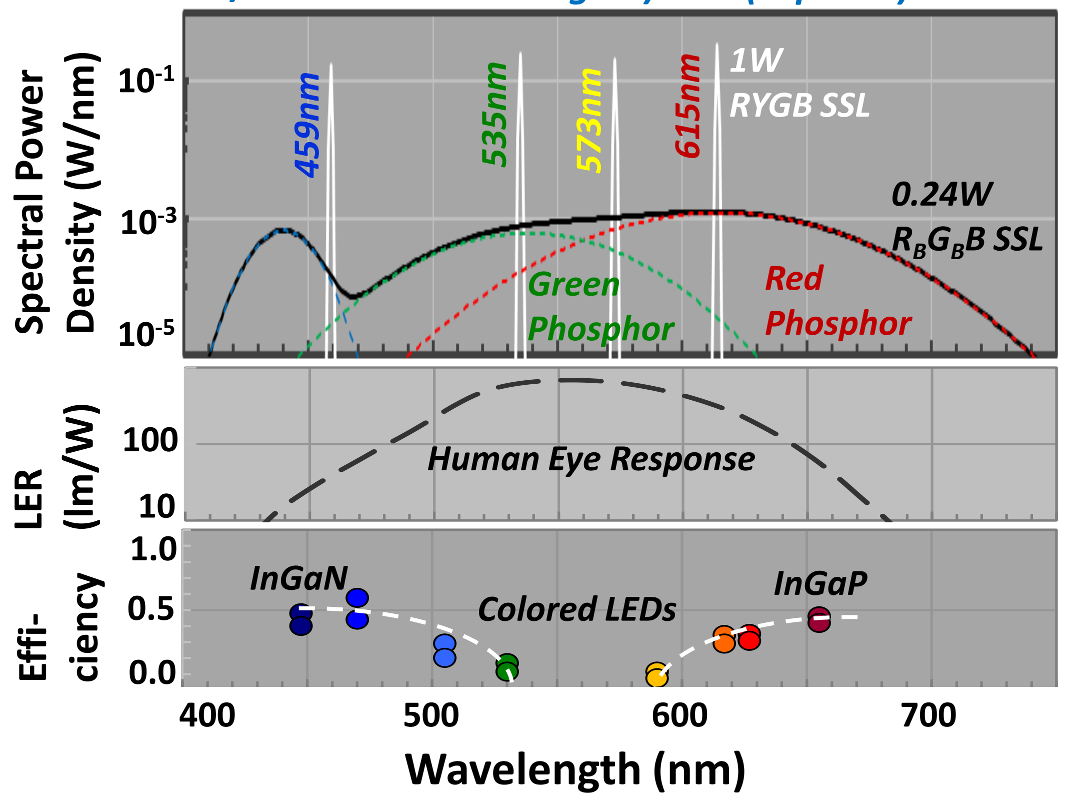
Why Only 20% Efficient?

Four SSL Technology Grand Challenges

1 Efficiency Droop (near-100% efficiency at all currents)

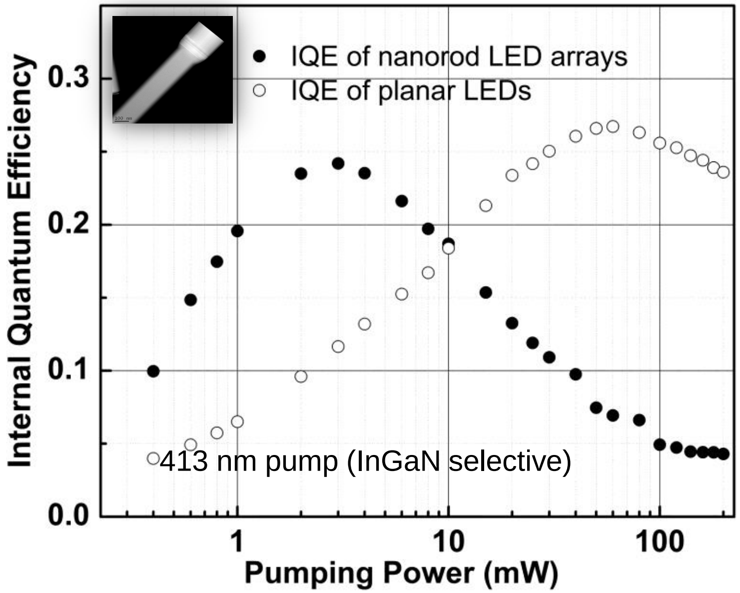
2 Green-Yellow Gap (near-100% efficiency at all wavelengths)

3 Narrow-linewidth λ downconversion (esp. red)



Optical performance – axial nanowire LEDs vs. planar LED

IQE – nanowire vs Film



PL, IQE: K. Westlake, M. Crawford

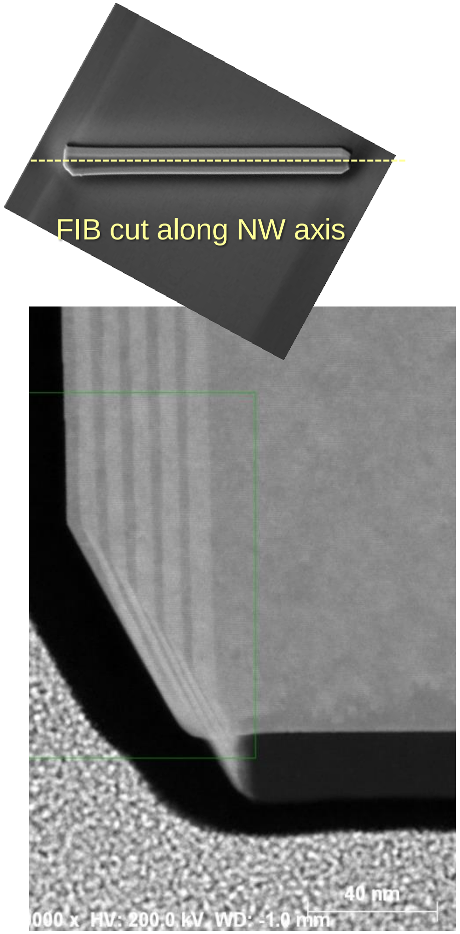
Q. Li et al., *Optics Express* **19**, 25528 (2011)

- XRD shows ~16±4% strain reduction in InGaN QWs in nanowire LEDs *XRD measurement :Steve Lee, Sandia*
- TEM shows ~94% of nanowire LEDs are dislocation free
- Little wavelength shift at higher pump powers for nanowire LEDs (no/reduced QCSE)
- But nanowire LED shows only comparable IQE to planar LED; peak IQE occurs at much lower pumping power (enhanced light absorption, heating)

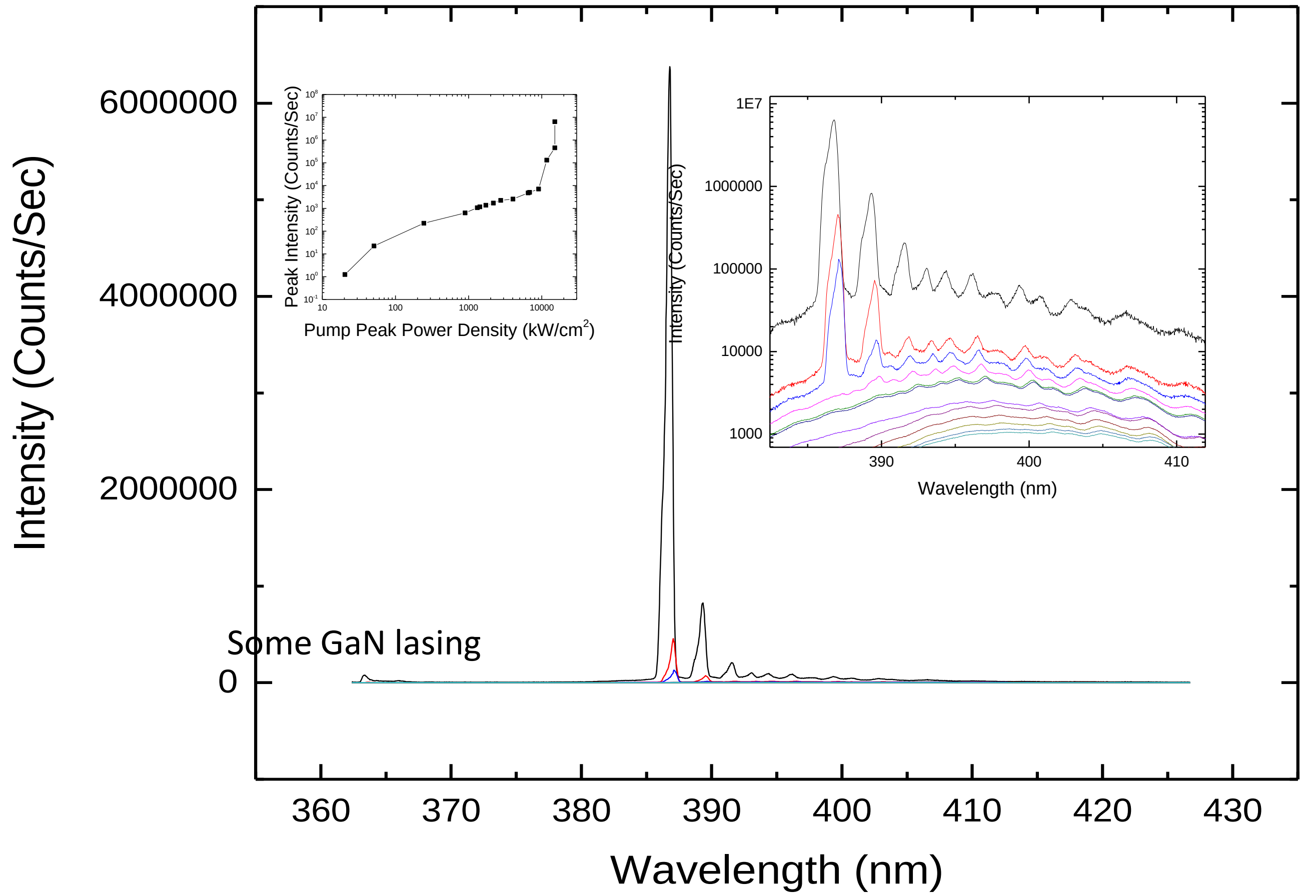
How much is surface passivation likely to improve IQE of NW LED?

No obvious benefit for blue LED performance/cost ratio (loss of device area), but possibility for very high IQE single NW emitters for nanophotonic applications by starting with commercial quality planar LEDs

Lasing in single NWs with nonpolar *m*-plane InGaN/GaN MQWs

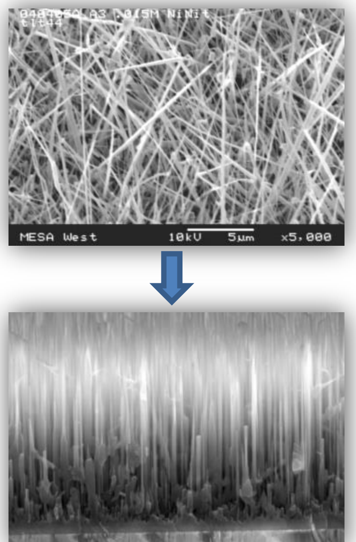


InGaN gain region needed to reach visible wavelengths



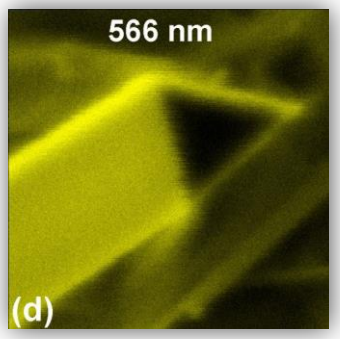
First known demonstration of optically-pumped lasing in radial nonpolar m-plane GaN/InGaN MQW nanowires

Challenges for nanowire-based SSL



Synthesis/Fabrication

- Ordering, alignment, density
- Size control (uniformity, diameter, height)
- Doping, heterostructures
- Growth on different facets
- Manufacturability

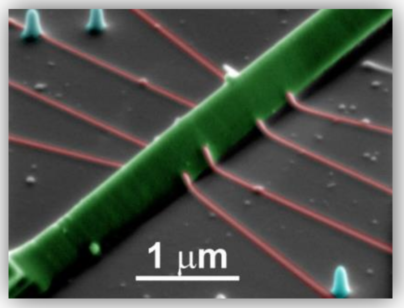


surface defect luminescence

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

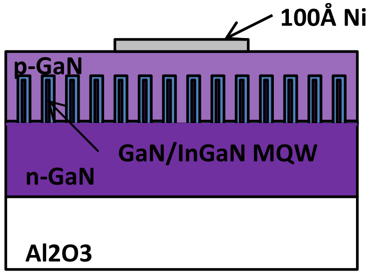
Properties (electrical, optical, thermal, mechanical)

- Role of defects & surfaces
- Control/uniformity
- Differences from bulk/planar material

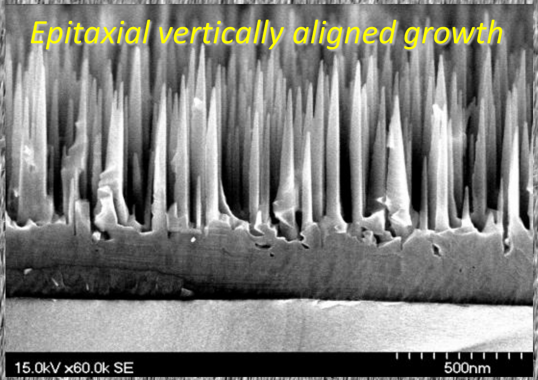


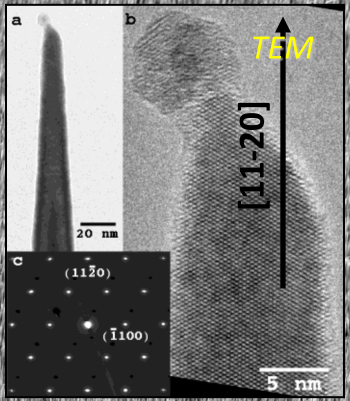
Electrically-integrated devices

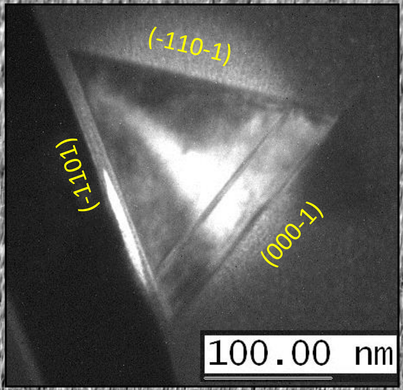
- Contacts (single NW & vertically integrated)
- Surface states
- Charge injection and transport



"Bottom Up" aligned GaN nanowire growth



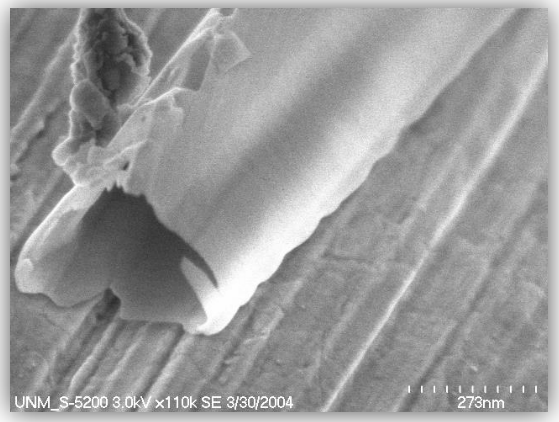




- Nanowires grown by Ni-catalyzed MOVPE/MOCVD (VLS)
- Highly-aligned vertical growth over large areas (2" r-sapphire wafer)
- Controllable densities as high as ~150 nanowires µm^{-2}
  - **Q. Li, G. T. Wang, *Appl. Phys. Lett. 93, 043119* (2008)**
  - **Q. Li, J. R. Creighton, G.T. Wang. *J. Crys. Growth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

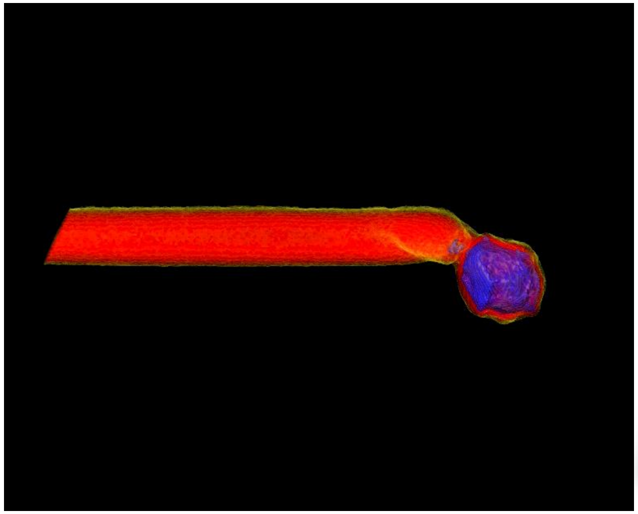
100 µm

Radial heterostructure nanowire growth

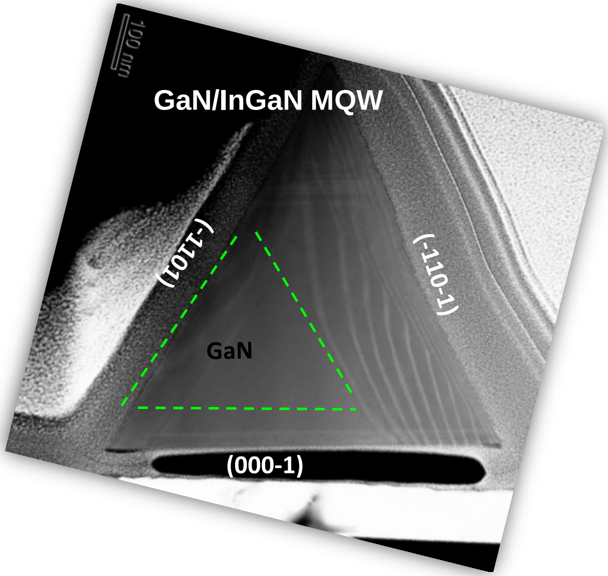


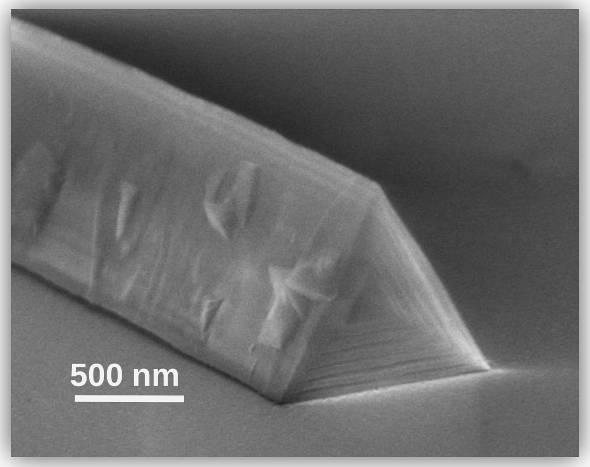
G. Stan et al. , *Nanotechnology*, ***20***, **2009**

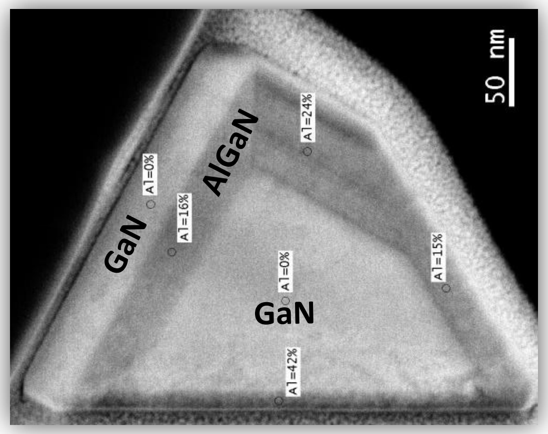
AlN Nanotubes



GaN/AlN NW I. Arslan

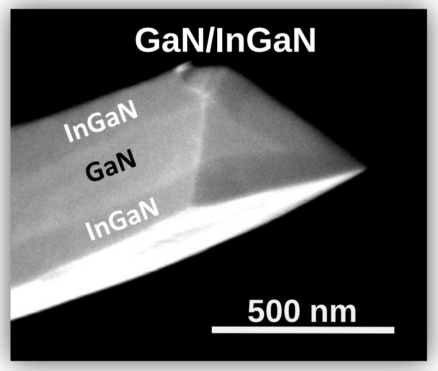






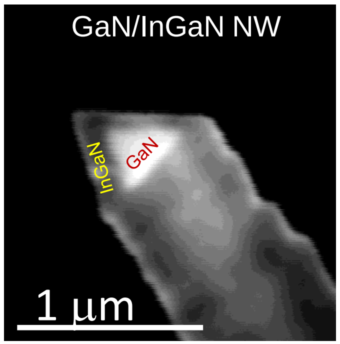
GaN/AlGaN/GaN

Double heterostructure

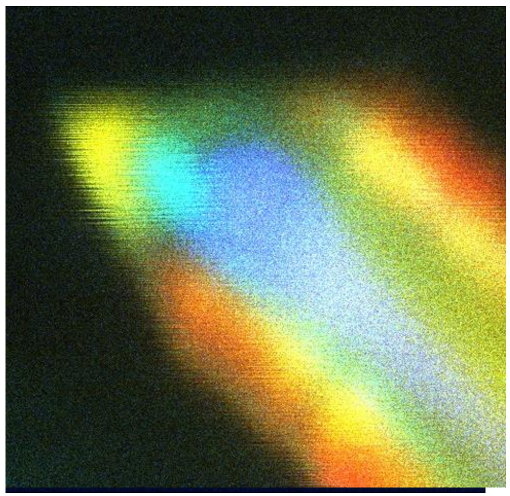


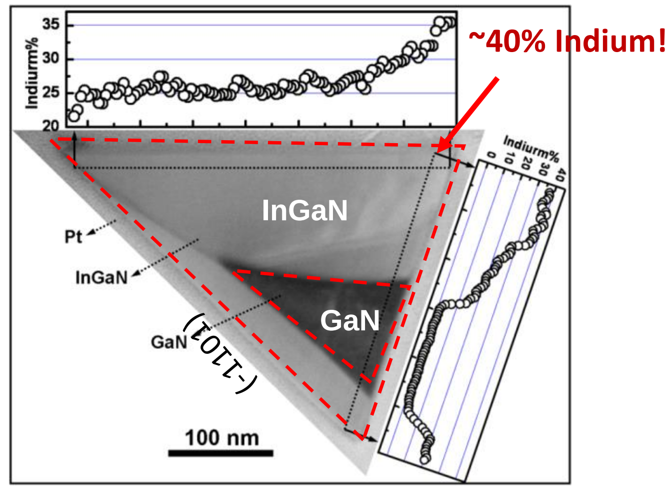
Core-shell NWs – A good platform for high In InGaN?

- ***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.



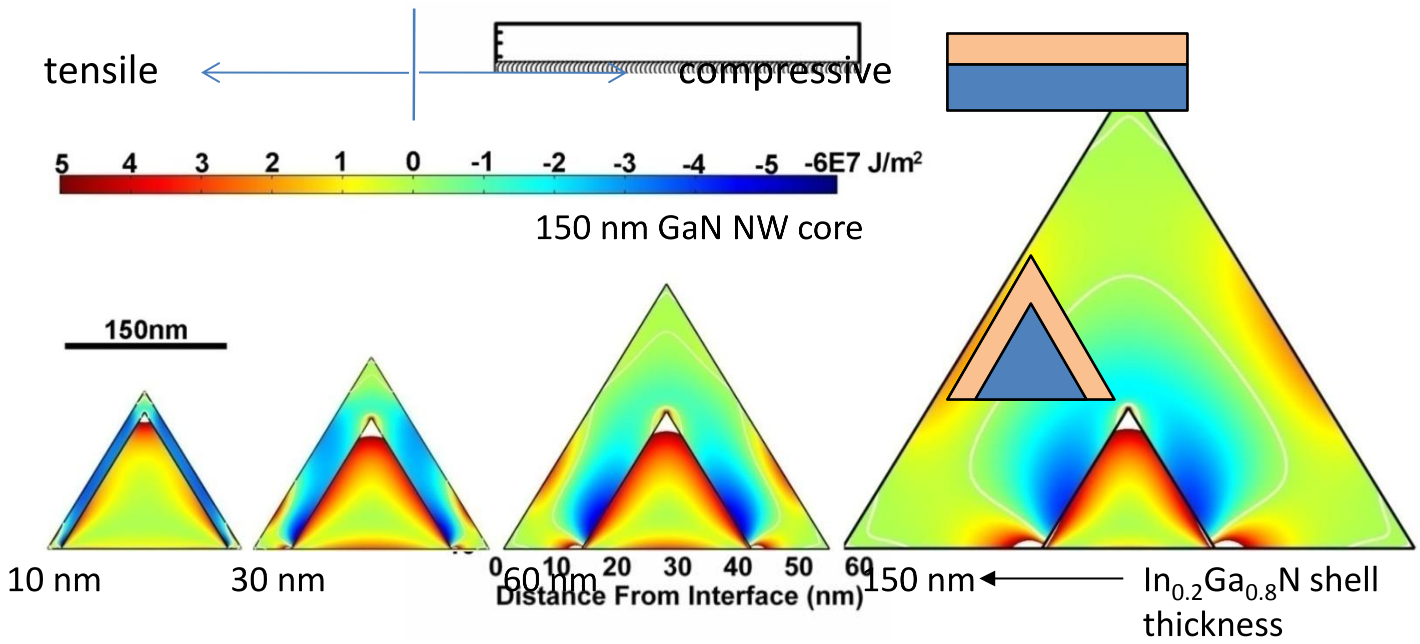


CL shows strong multicolor emission up into the IR!

- **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

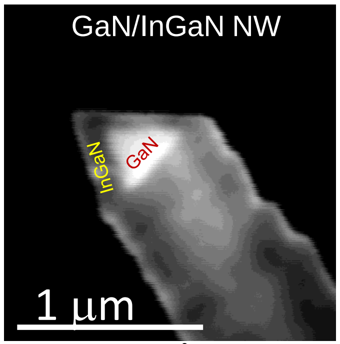


- Strain lower for GaN/InGaN nanowire than for GaN/InGaN thin film
- Compressive strain decreases away from GaN/InGaN interface, lowest in corners

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.
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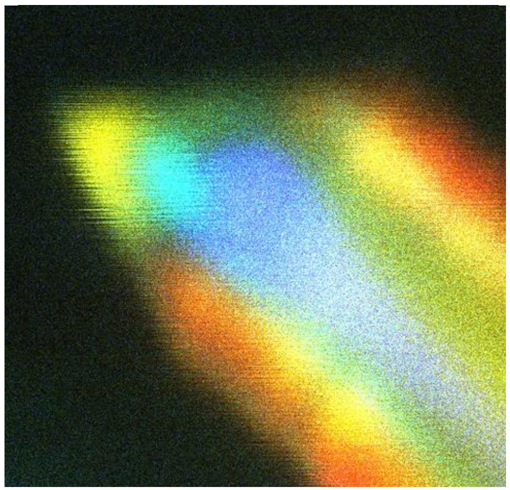

CL shows strong multicolor emission up into the IR!

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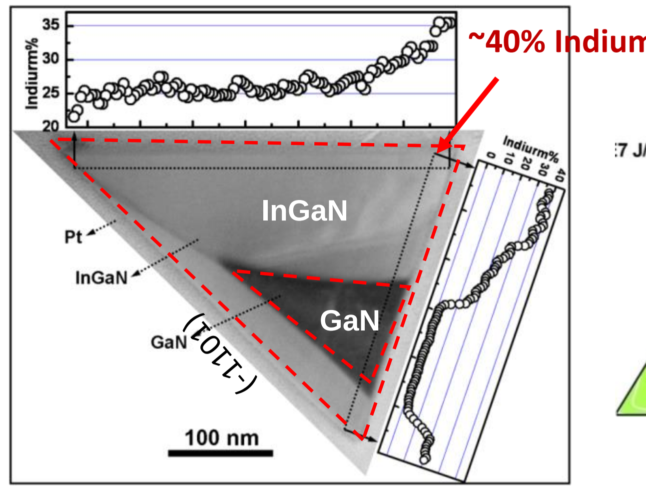
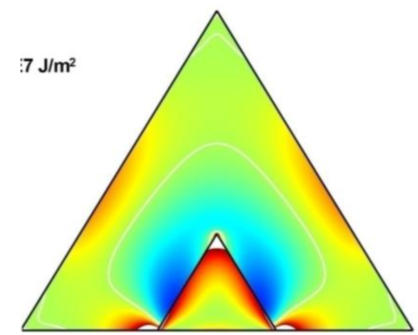


STEM/EDS shows In distribution, **highest at surface/corners**

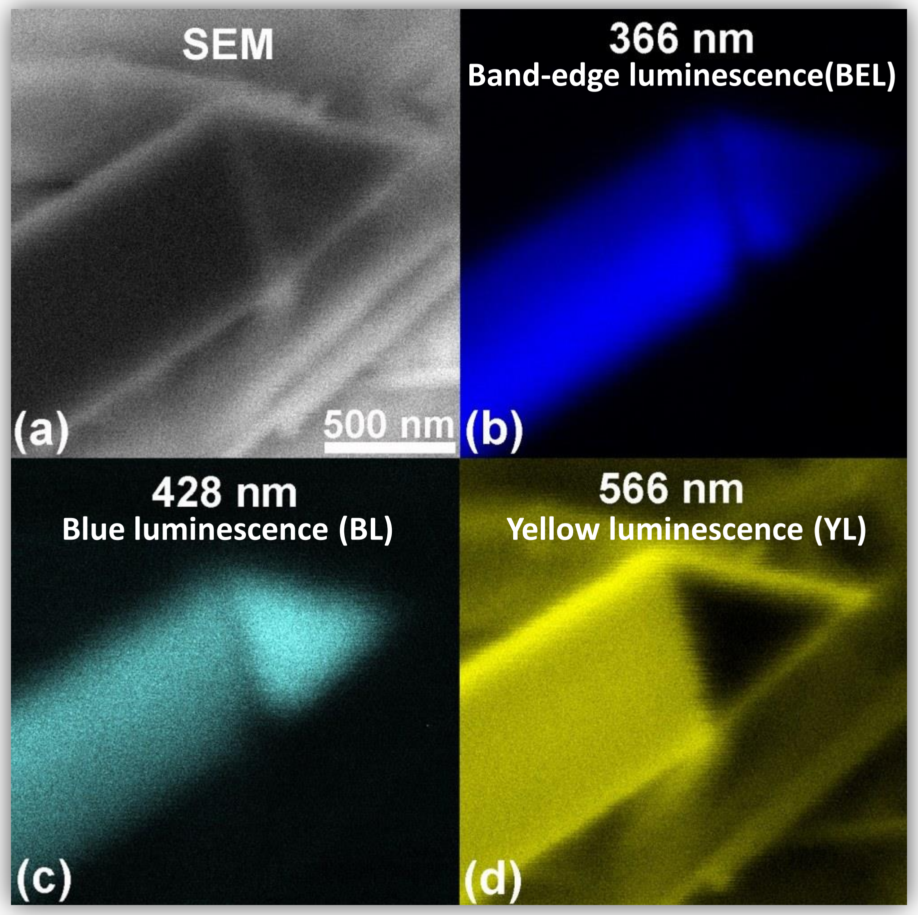


2D FEA: highest In regions correlated w/lowest compressive strain regions

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

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

Spatial distribution of luminescence in GaN NWs

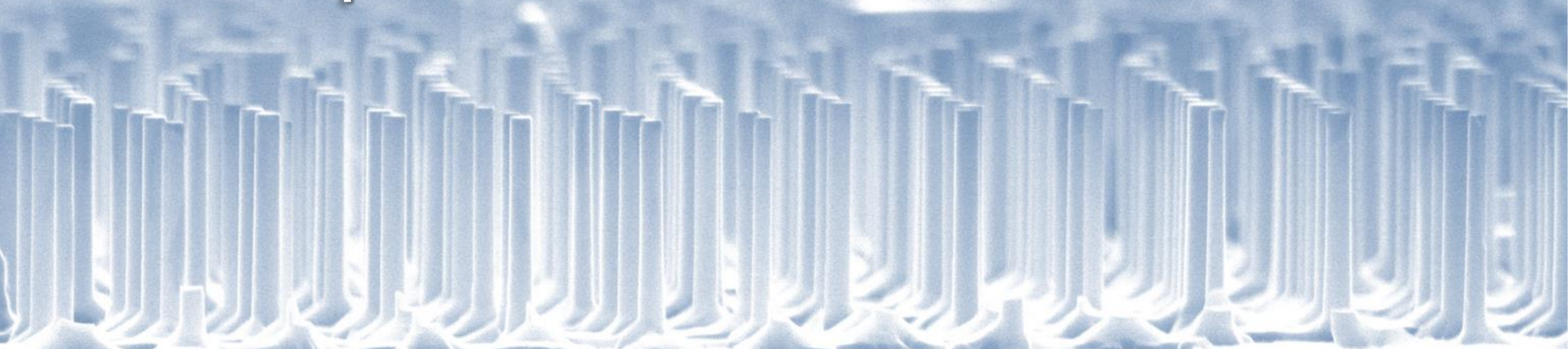


***Nanoscale Cathodoluminescence (CL) imaging:* Cross-section GaN NW**

- 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

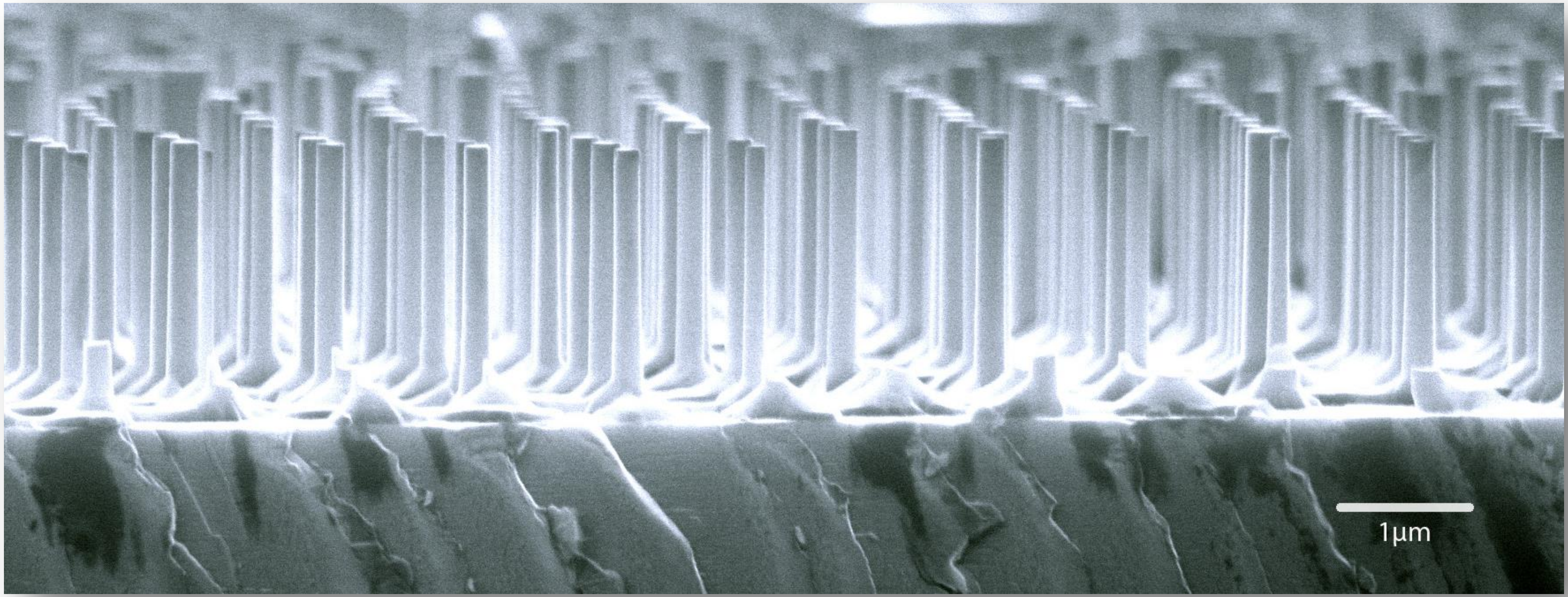
Q. Li, G. T. Wang, *Nano Lett.*, 2010, **10** (5), 1554

Top-down III-Nitride Nanowires



- Although dominant, bottom-up nanowire approaches suffer from some limitations
- Specific growth conditions needed for anisotropic growth which may not result in optimal material quality or desired material property
- Architectures may be limited depending on growth method (MBE vs. MOCVD)
- In response, around 2010 our group began investigating new top-down approaches for fabricating III-nitride nanowires…

Advantages of new top-down nanowire fabrication method

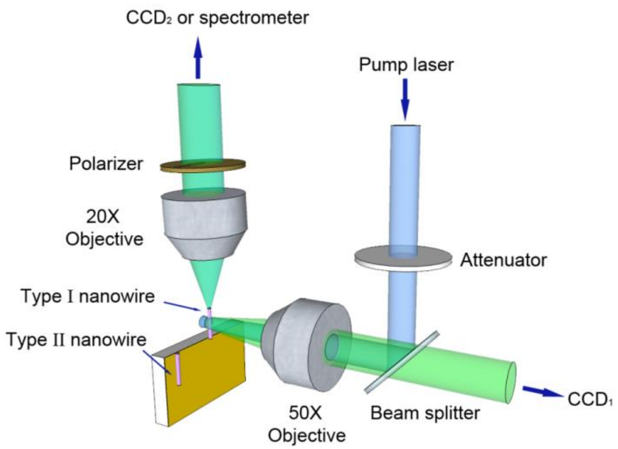


- Wider range of growth conditions, material tunability (1D growth cndns not needed)
- Lower point defect densities (higher growth temperature)
- Ordered/periodic arrays (difficult with catalyst/VLS-based methods)
- Axial III-nitride nanowire heterostructures possible by MOCVD
- Better control of geometry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

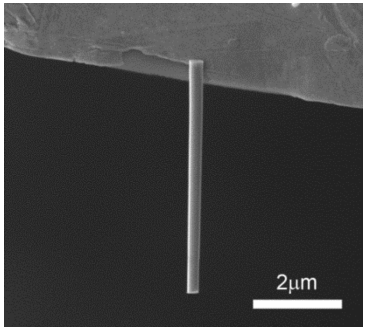
Mode control (i.e. How to make a single-mode GaN nanowire laser)

- Nanowire lasers generally exhibit *multiple modes*
- *Single mode* behavior desired for highest resolution and beam quality

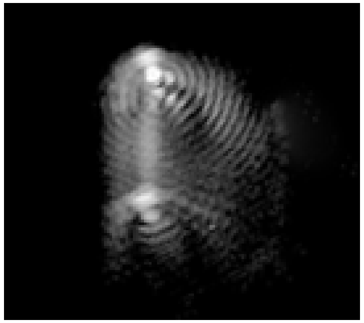
Lasing Polarization Properties of Freestanding GaN NWs



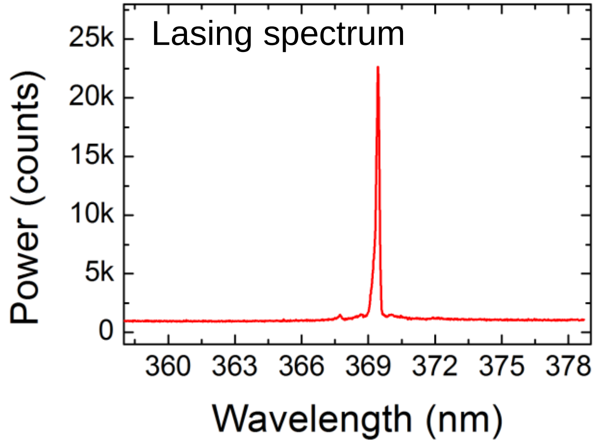
End-facet optical characterization setup

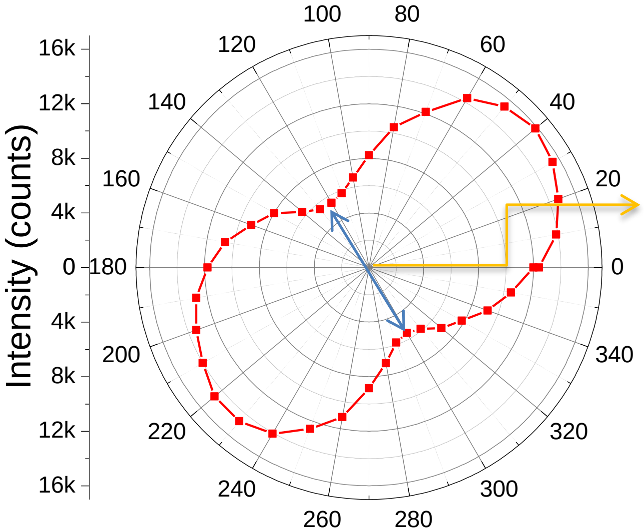


SEM image of a nanowire hanging off substrate



CCD image of lasing nanowire hanging off substrate





Indicative of elliptical polarization

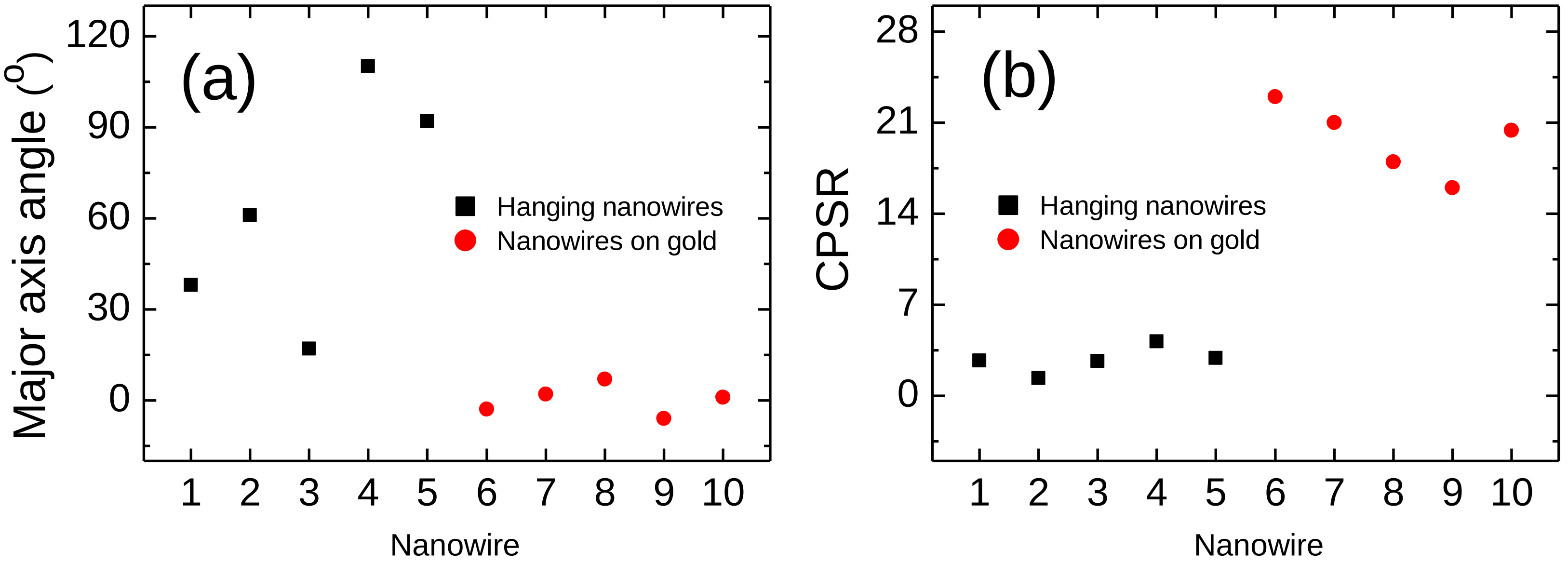
Polarization angle random wrt surface

Intensity vs. the rotation of the polarizer

Lasing Polarization Statistics of Free Standing and on-Gold Nanowires

Statistics on the polarization axis

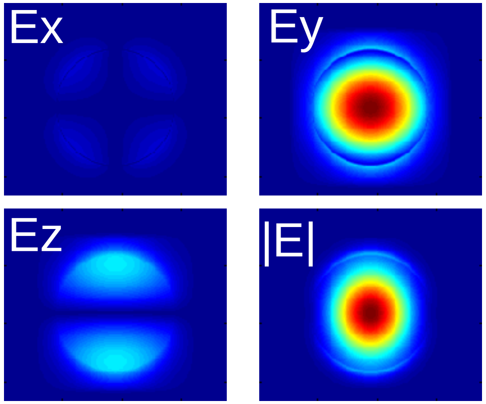
Statistics on cross-polarization suppression ratio (CPSR)



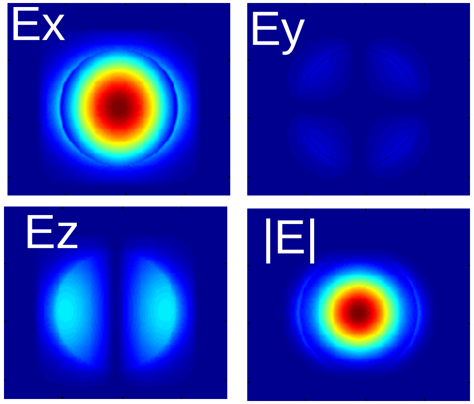
- ✓ Free standing nanowires are polarized with random axes and a small CPSR
- ✓ On-gold nanowires have fixed polarization angle and large CPSR

Why nanowire laser elliptically polarized?

Mode 1 HE11$_a$ -n$_{eff}$=2.1



Mode 2 HE11$_b$ - n$_{eff}$=2.1



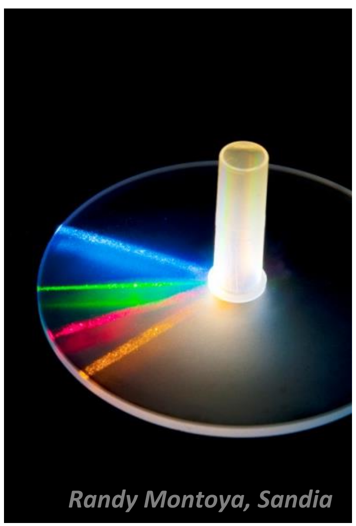
Two orthogonal polarized fundamental modes supported by a 200 nm diameter nanowire in air

- ✓ Both fundamental transverse modes
- ✓ Same electric field intensity
- ✓ **Two degenerate modes superimpose resulting in elliptical polarization**

Lasers for Solid State Lighting

Advantages of lasers for lighting:

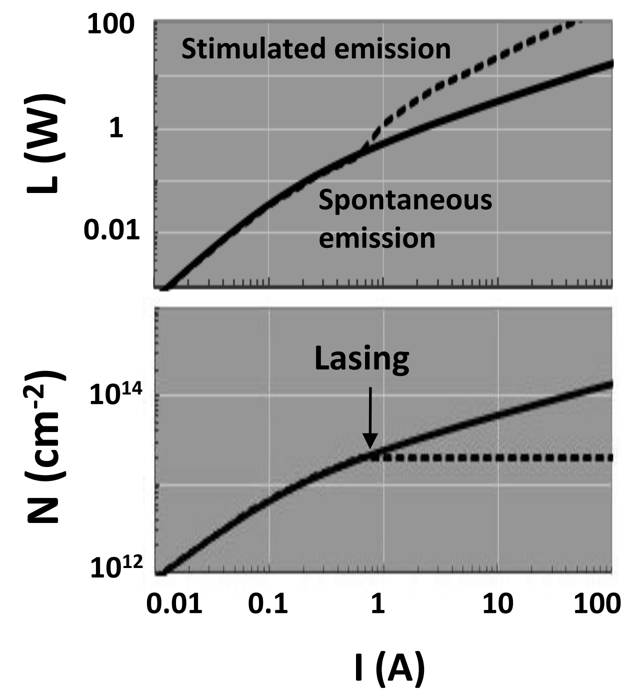
- Lasers show very high efficiency at high power
- LED and LD current densities are converging
- Carrier density is clamped at threshold
  - Circumvent the droop problem in LEDs
  - Need to reduce threshold to avoid losses
- After threshold slope efficiency is one
- Directionality, polarized emission,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

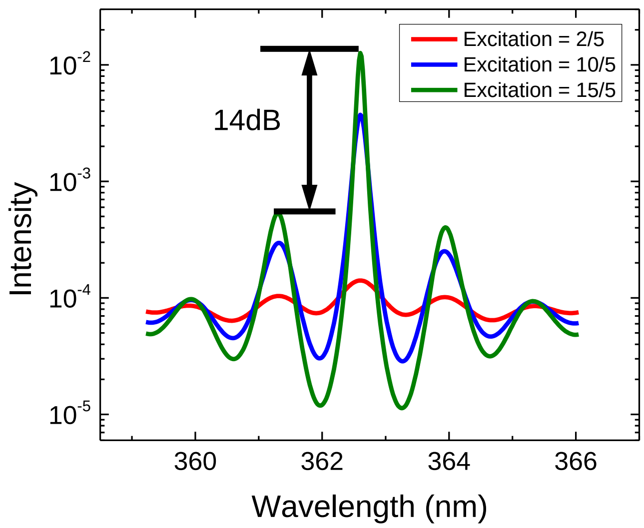


Are narrow linewidth sources acceptable?

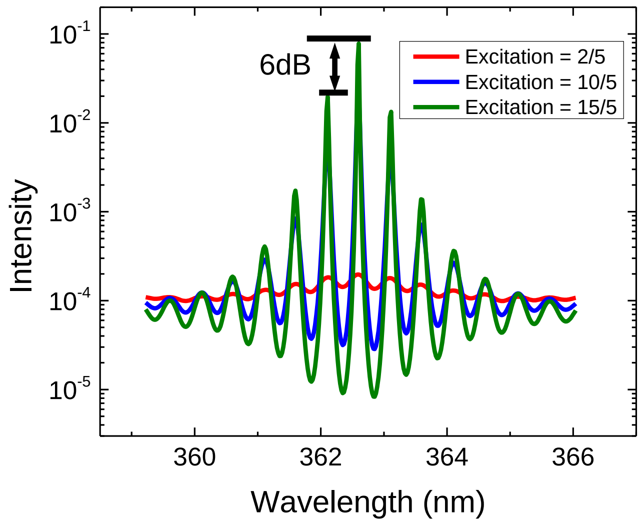
A.J. Fischer, Sandia

Single mode GaN nanowire laser

4 µm long, 140 nm dia. nanowire

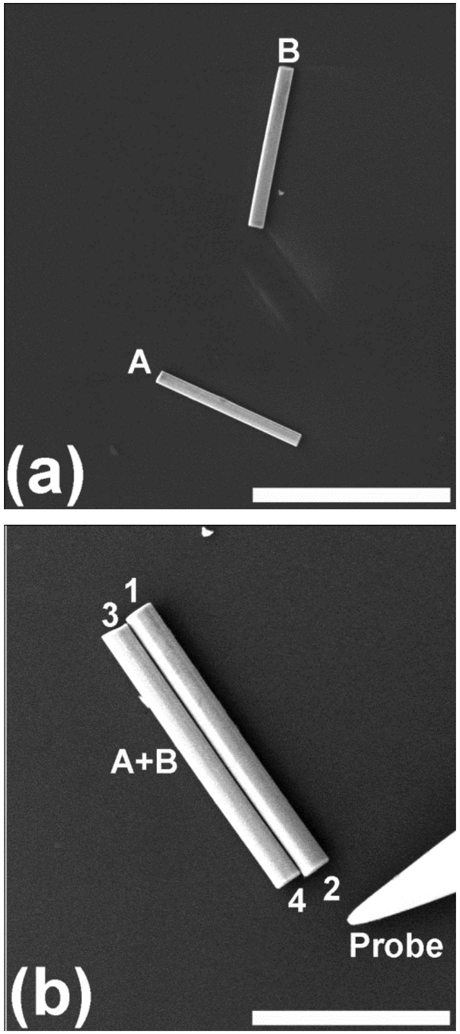


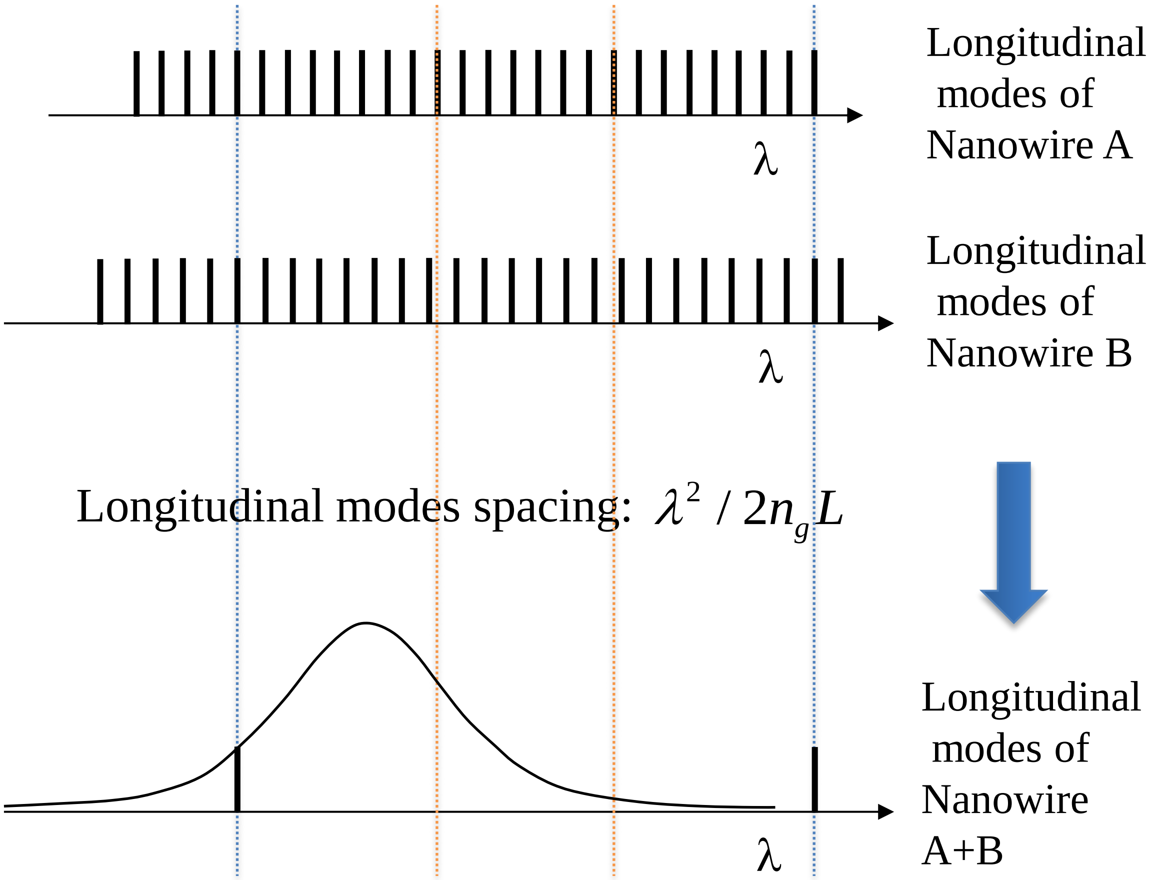
12 µm long, 140 nm dia. wire



- 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.

Method 2: Vernier Effect From a Coupled Cavity

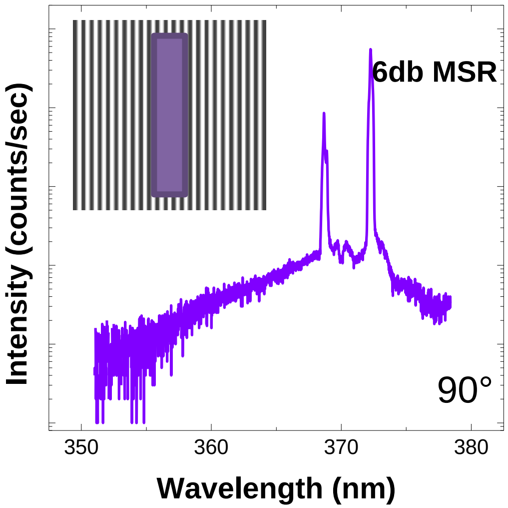


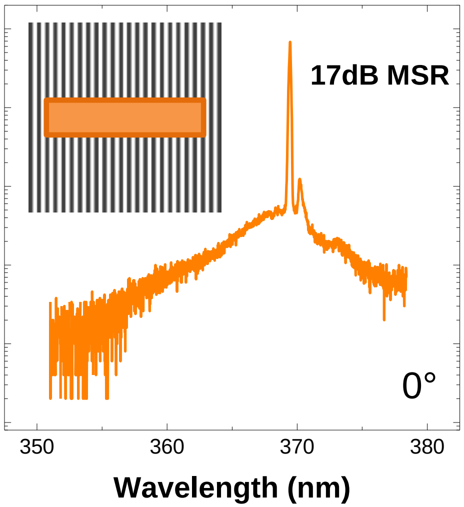


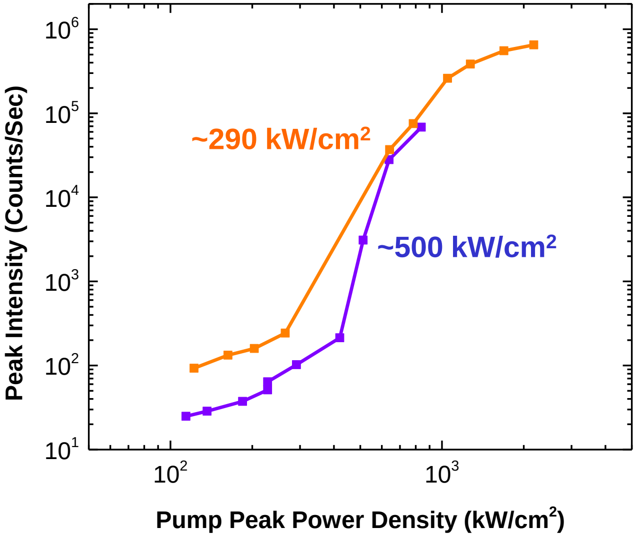
Longitudinal mode spacing greatly enhanced!
(10s of nanometers in this coupled cavity)

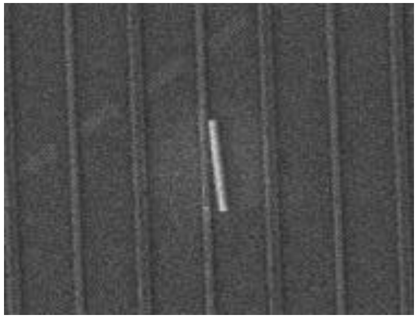
Method 3: Distributed Feedback (DFB) Nanowire Laser

- Standard method for mode control in semiconductor lasers
- Here, single nanowires are coupled to grating substrate to achieve DFB

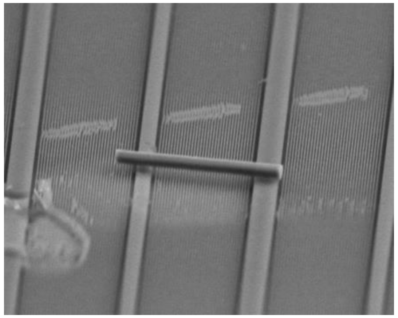








NW diameter ~180 nm

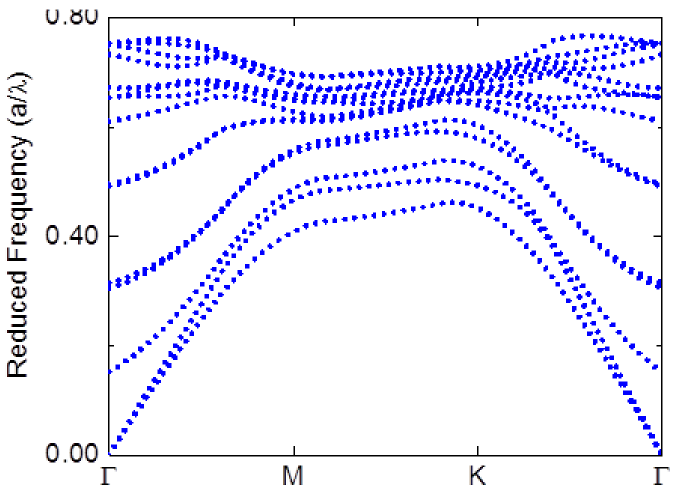


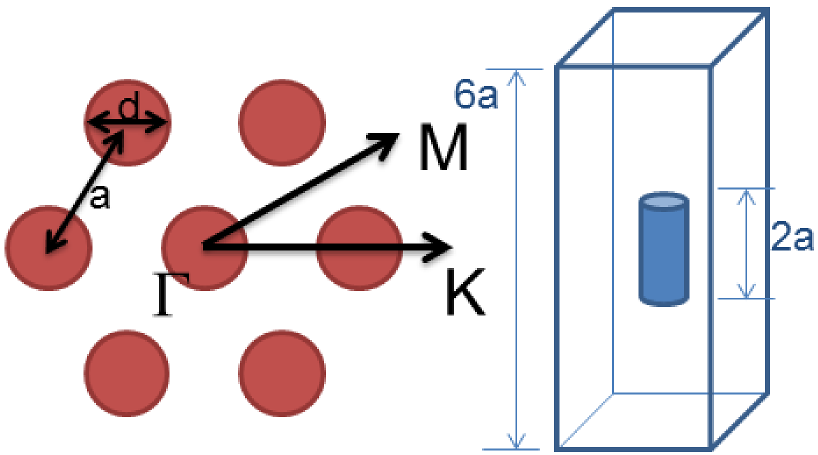
- At the designed alignment single-mode lasing was achieved with a 17dB mode suppression ratio.
- Observed reduction in the lasing threshold

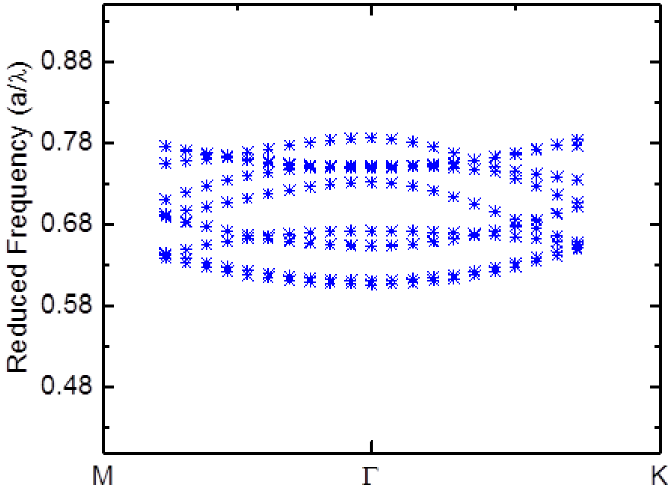
J.B. Wright, S. Campione, S. Liu, J.A. Martinez, H Xu, T.S. Luk, Q. Li, G.T. Wang, B.S. Swartzentruber, L.F. Lester, and I. Brener, APL 104 (4), 041107 2014.

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

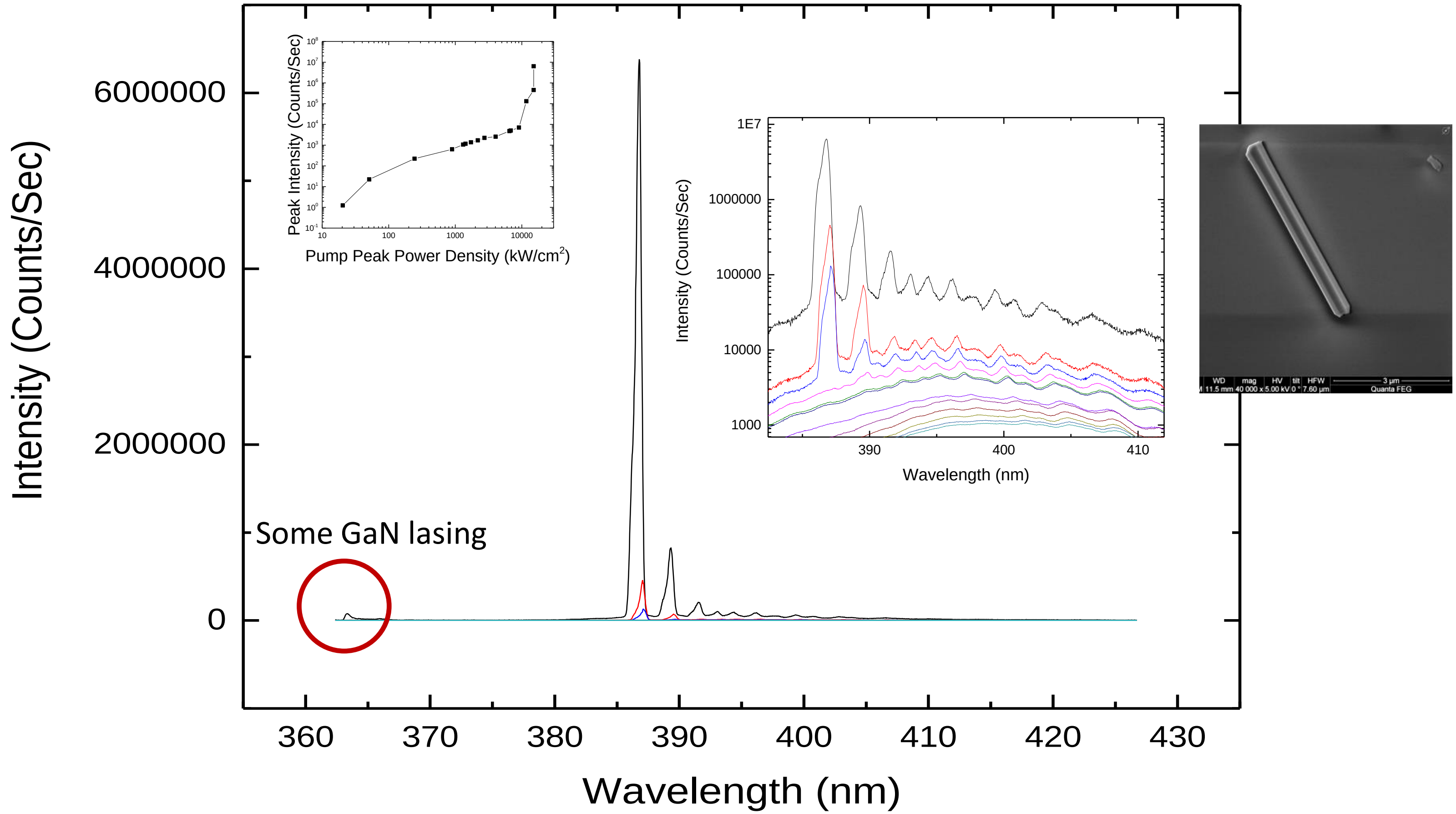






Key Accomplishments

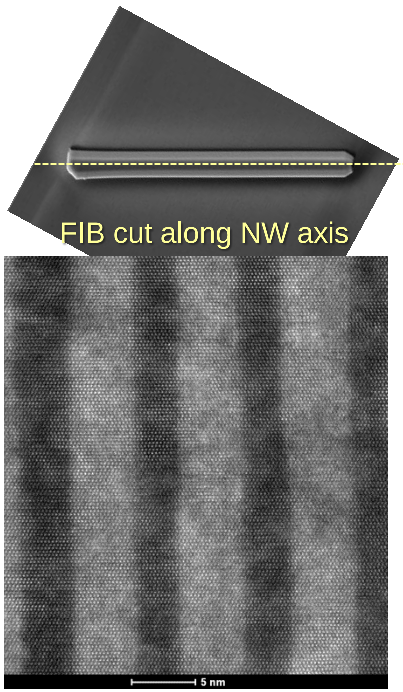
First known demonstration of optically-pumped lasing in radial m-plane GaN/InGaN MQW nanowires

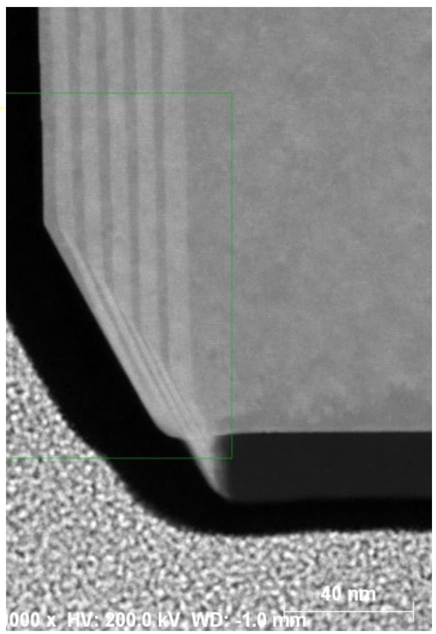


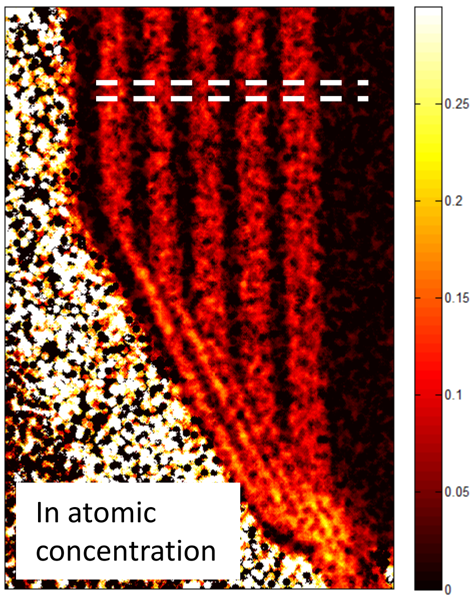
Result confirms radial InGaN/GaN MQW NW can lase (note: not optimized designs)

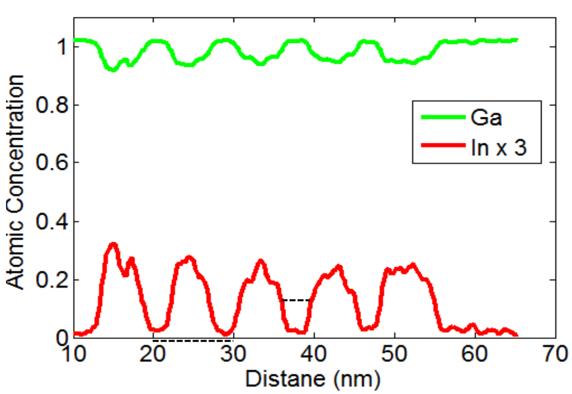
Key Accomplishments

Optically-pumped lasing in radial m-plane GaN/InGaN MQW nanowires – STEM/EDS analysis









Near tip region

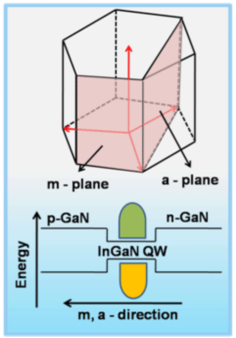
- In composition ~6-10%
- InGaN QWs ~4-6 nm
- GaN barriers ~2.5-4 nm

Cross-sectional STEM/EDS analysis enables post-growth analysis of radial InGaN/GaN NW structure needed to realize targeted design

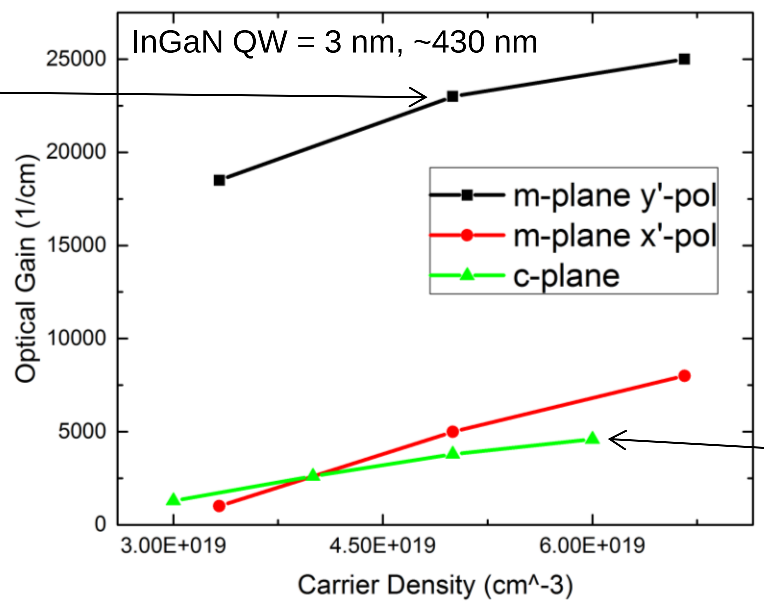
Key Accomplishments

Analysis of axial vs radial NW designs

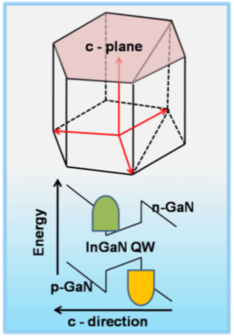
Radial NW



Optical Gain - Crystallographic Dependence



Axial NW



Much higher optical gain for m-plane QWs (benefits radial NW design)

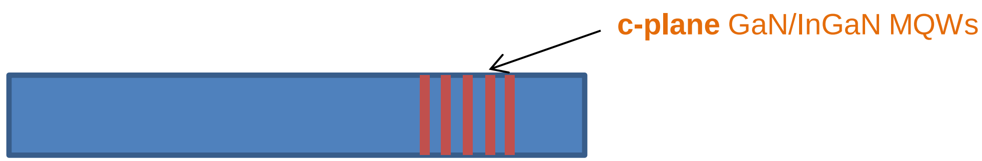
S.-H. Park and D. Ahn, "Optical polarization characteristics of m-plane InGaN/GaN quantum well structures and comparison with experiment," *Appl. Phys. Lett.*, **103**, *pp. -, (2013)*

J. Piprek, R. K. Sink, M. A. Hansen, J. E. Bowers, and S. P. DenBaars, "Simulation and optimization of 420-nm InGaN/GaN laser diodes," 2000, pp. 28-39.

Crystallographic figures from: http://csel.snu.ac.kr/research/LED.php

Key Accomplishments

Analysis of axial NW laser design – needed reflectivities



Nanowire Constants

$\lambda := 420\,\text{nm}$

$\text{Length} := 5\,\mu\text{m}$

$t_{QW} := 3\,\text{nm} \qquad N_{wells} := 5$

$\Gamma_{xy} := 1$

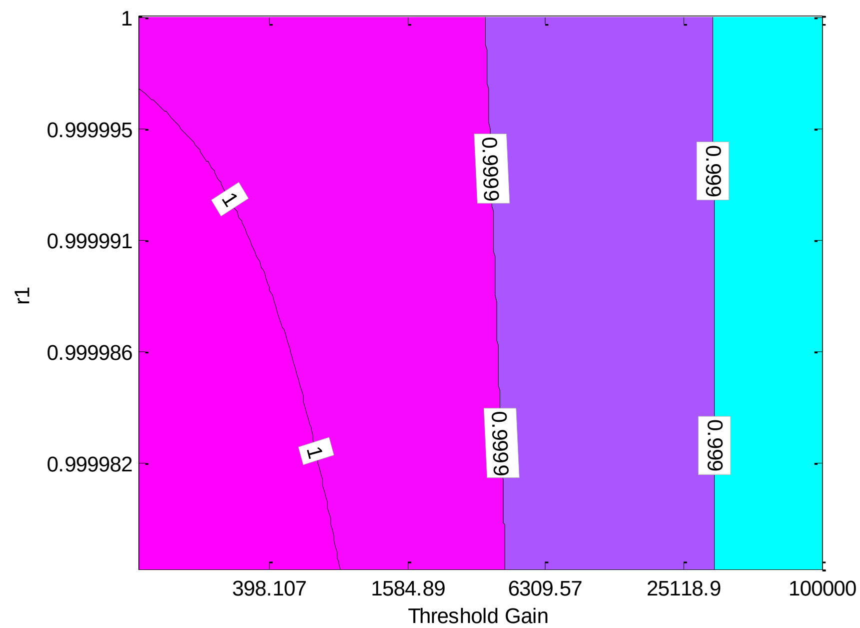
Overlap Calculation

$$\Gamma_z := \frac{t_{QW} \cdot N_{wells}}{\text{Length}} = 3 \times 10^{-3}$$

$$\Gamma_{tot} := \Gamma_{xy} \cdot \Gamma_z$$

Threshold Gain

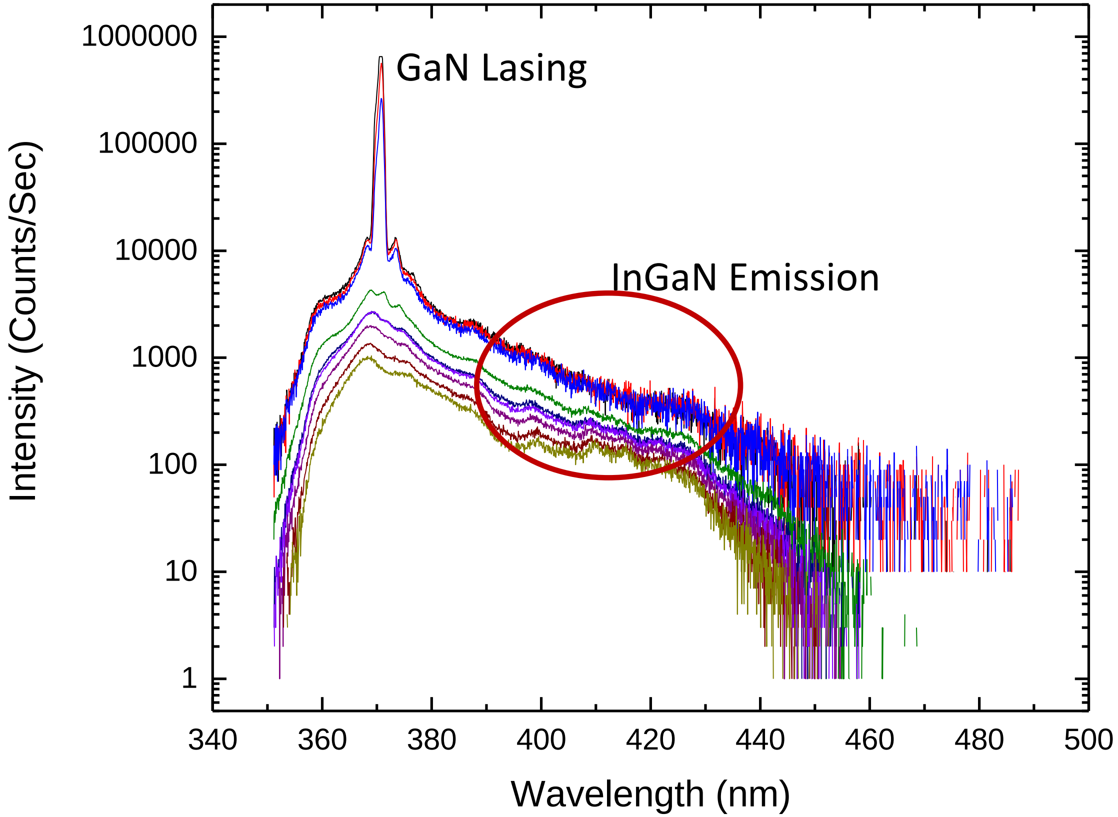
$$g_{th} := \frac{1}{\text{Length}} \cdot \ln\left(\frac{1}{r_1 \cdot r_2}\right) \cdot \frac{1}{\Gamma_{tot}}$$

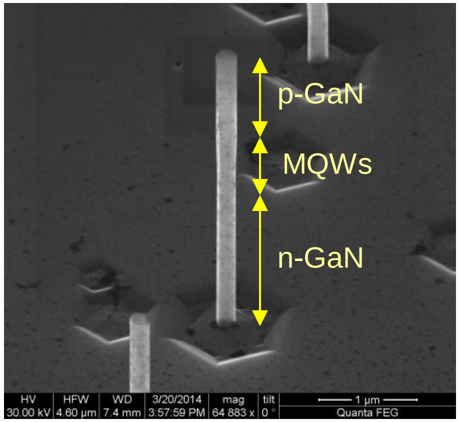


Need unrealistically high mirror reflectivities for axial NW design!
(focus on radial NW design)

Key Accomplishments

Fabrication and optical characterization of axial GaN/InGaN MQW NW diode





No InGaN lasing observed for axial NW laser diode structure