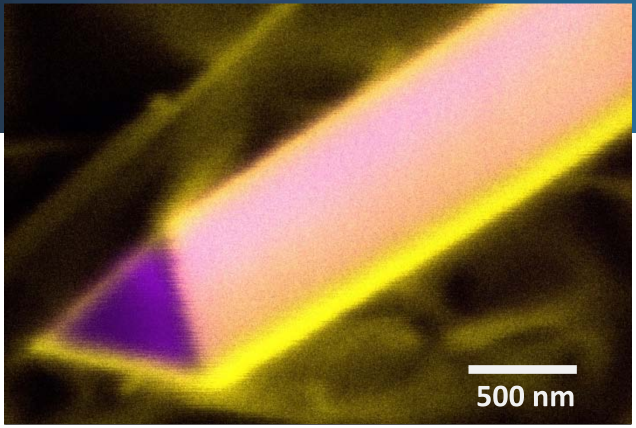
SAND2010-6495P

Spatially-Resolved Study of Luminescence and In Incorporation in GaN and High-In Content InGaN/GaN Nanowires

George T. Wang (gtwang@sandia.gov) and Qiming Li

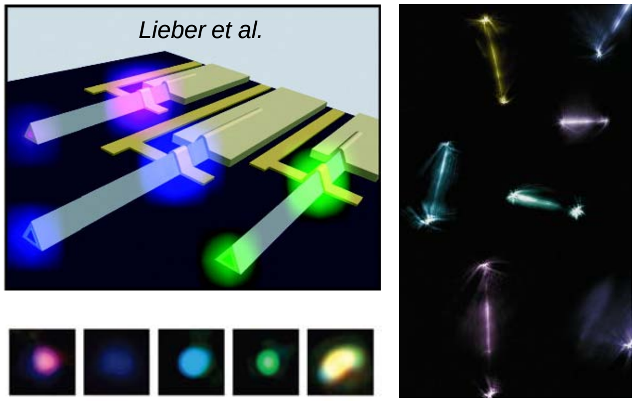
Sandia National Laboratories
Albuquerque, NM



Sandia National Laboratories is a multi-program laboratory operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin company, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

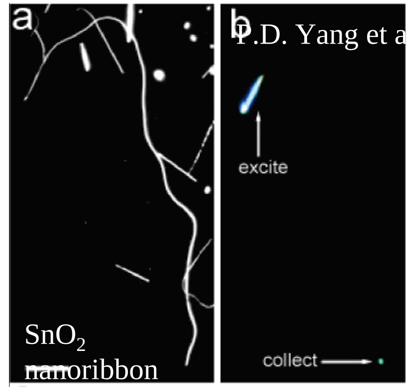
Semiconductor Nanowires (NWs)

Reduced dimensionality, high crystalline quality, high atomic surface/bulk ratio, size (intersects physical characteristic length scales) can lead to enhanced & novel properties



LEDs and lasers

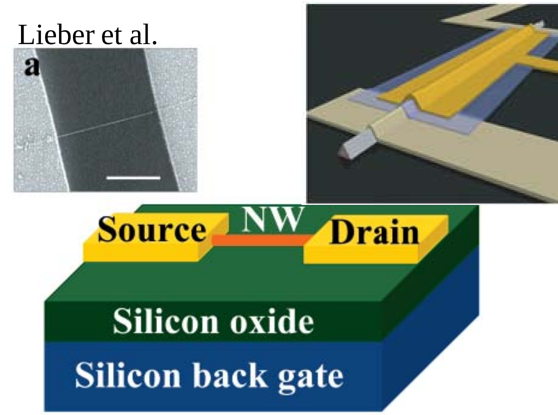
- Nanosized light sources
- Higher efficiency due to lack of defects
- High light extraction



Waveguides and Filters

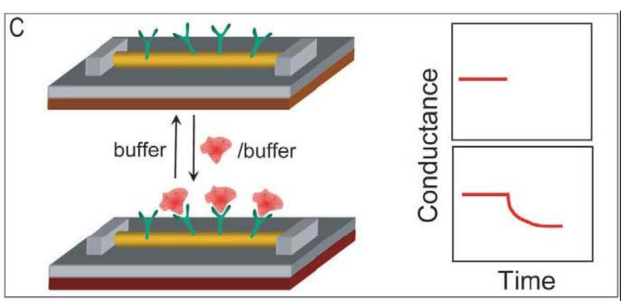
- coupled with nanowire light sources, building blocks for nanophotonics circuitry

Lieber et al.



Transistors/HEMTs

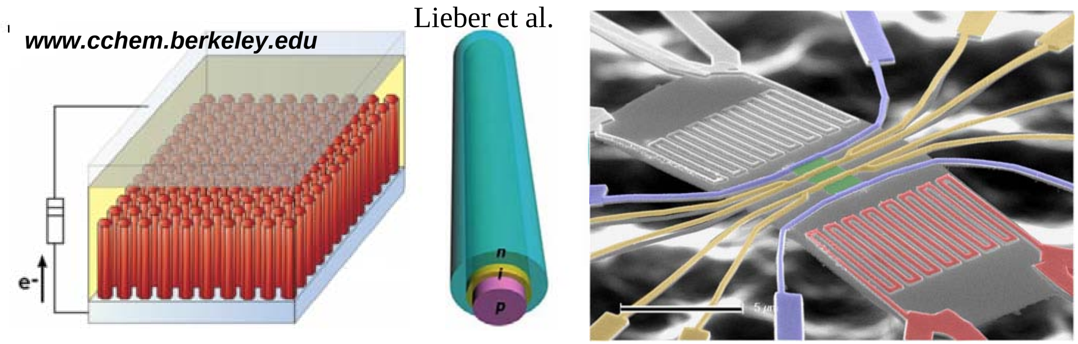
- improved performance characteristics
- small size



Lieber et al.

Chem/bio-sensors

- large atomic surface/bulk ratio leads nanowire depletion & ultrahigh sensitivity



Heath et al.

Energy Harvesting

- Nanowire Photovoltaics
- Thermoelectrics
- Piezoelectric energy generation

Sandia National Laboratories

III-Nitride (AlGaInN) Nanowires

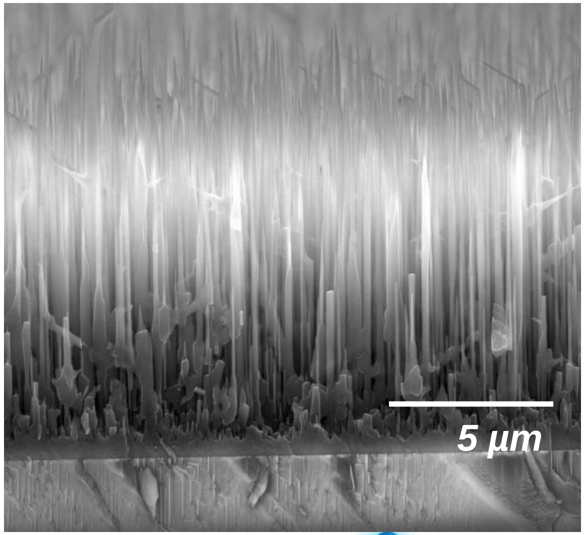
III-Nitride (AlGaInN) Properties

Direct RT bandgaps spanning very wide energy range from ~0.7-6.2 ev

- Form solid alloy system
- High breakdown field
- High mobility
- High thermal conductivity and temperature
- Radiation resistant and chemically inert
- *Used in LEDs, blue laser diodes, UV photodetectors, HEMTs*

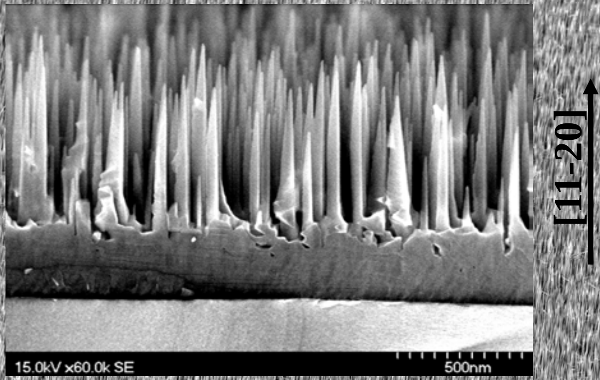
Compared to planar films, III-nitride nanowires …

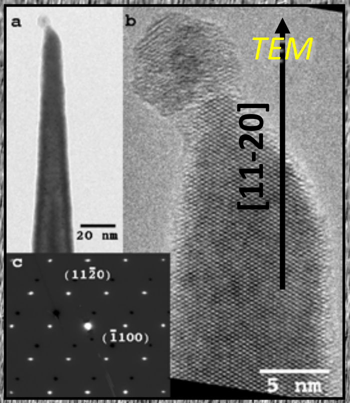
- Have high aspect ratio and high surface/bulk ratio
- Are strain relaxed
  - can accommodate wider range of alloy compositions
  - typically free of threading dislocations
  - grow on any substrate
- Are "discrete" – entire structure & changes to the structure can be investigated

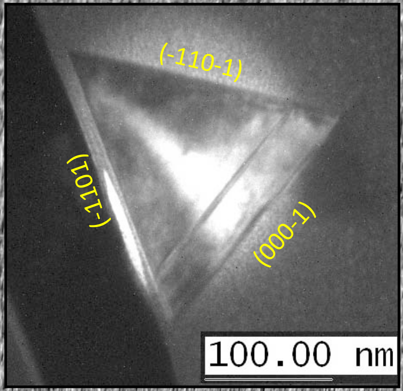


Aligned GaN NW growth

Epitaxial vertically aligned growth







- Nanowires grown by Ni-catalyzed MOVPE (VLS)
- Highly-aligned, uniform 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)

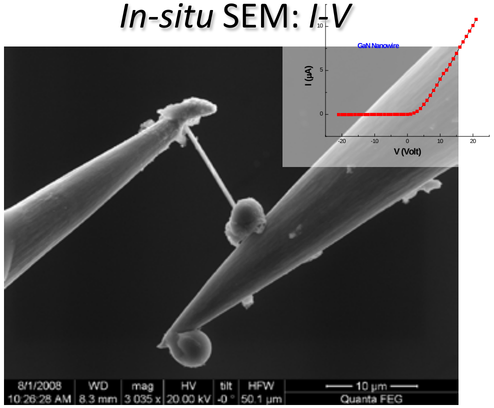
Structure & Properties - Vertically Aligned GaN Nanowires

In-situ SPM-TEM

In-situ NW decomposition/failure

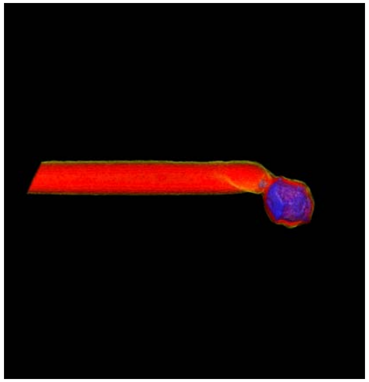
T. Westover, R. Jones, J. Y. Huang, G. Wang, E. Lai, A. A. Talin, *Nano Lett.*, **9**, 257 (2009).

In-situ SEM: I-V



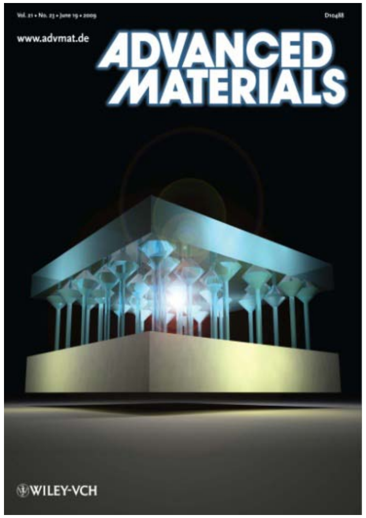
Y. Lin, Q. Li, A. Armstrong, and G. T. Wang, *Solid State Commun.*, ***149**, 1608 (2009)*

3D STEM tomography



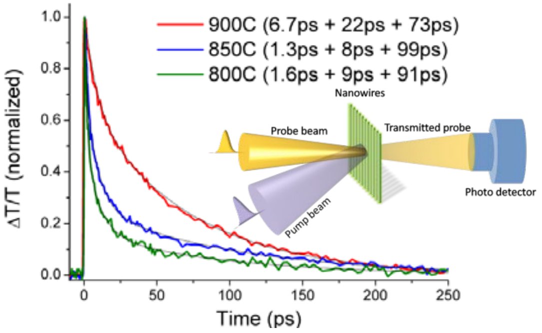
I. Arslan, A. A. Talin, G. T. Wang, *J. Phys. Chem. C* **112**, 11093. (2008)

NW-templated GaN



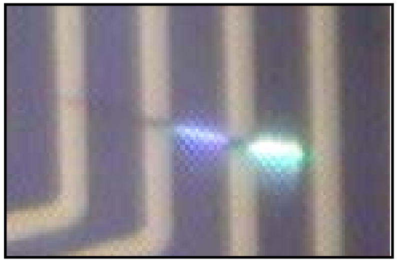
Q. Li, Y. Lin, J. R. Creighton, J. J. Figiel, and G. T. Wang, *Advanced Materials*, **21**, 2416–2420 (2009)

Ultrafast optical spectroscopy



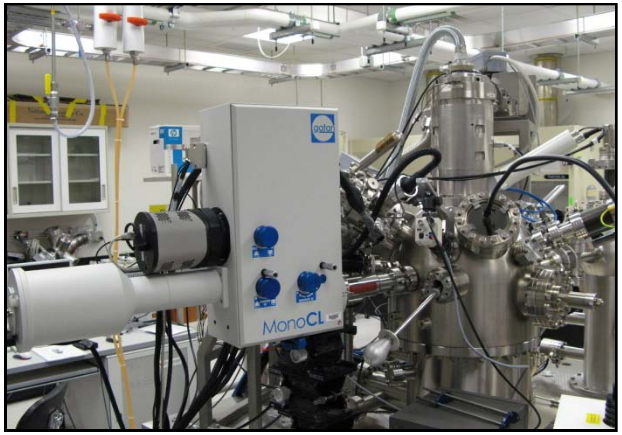
P.C. Uppadhya et al. *Semicond. Sci. Tech.* **25** 024017 (2010)

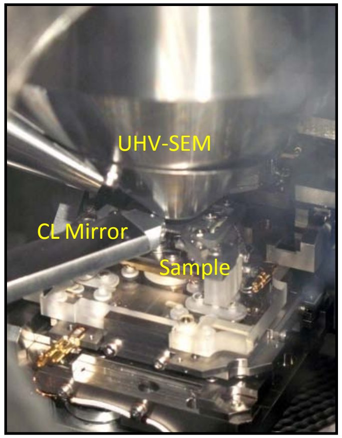
Correlated I-V/μ-PL



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

Cathodoluminescence (CL) Experimental Setup

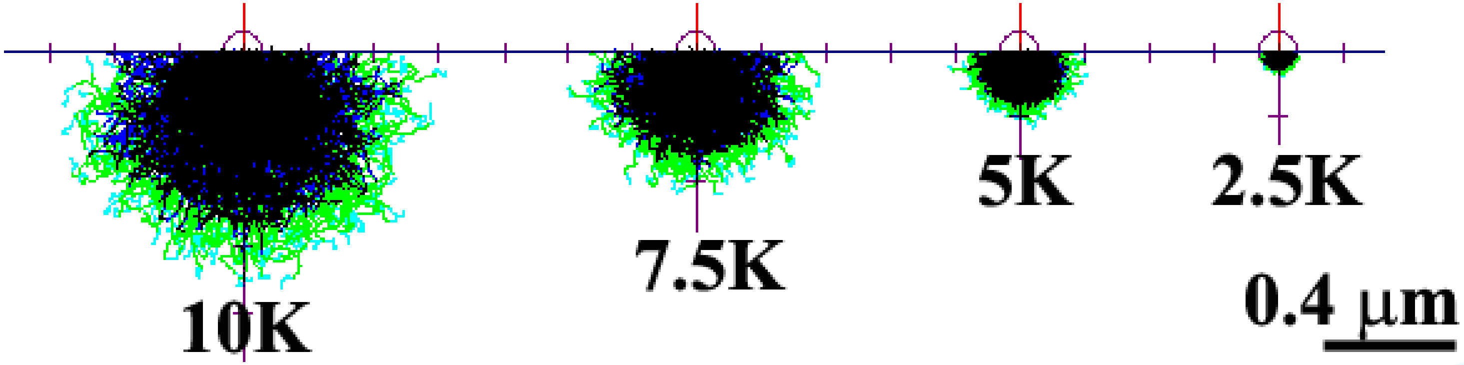




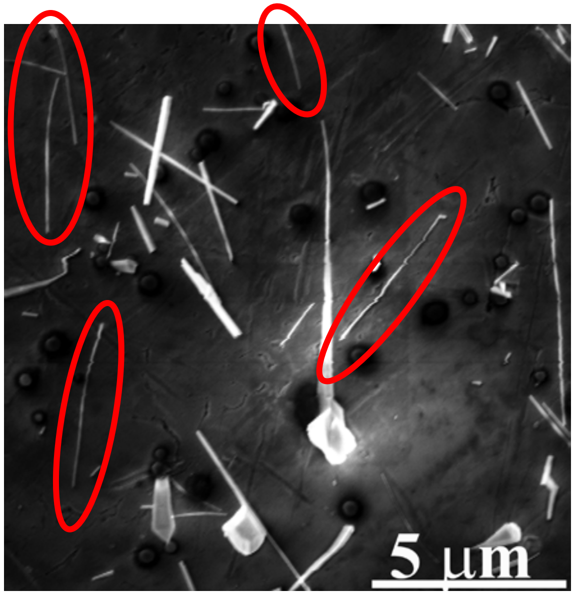
CL system on MESA Advanced Nanotechnology Tool (ANT)

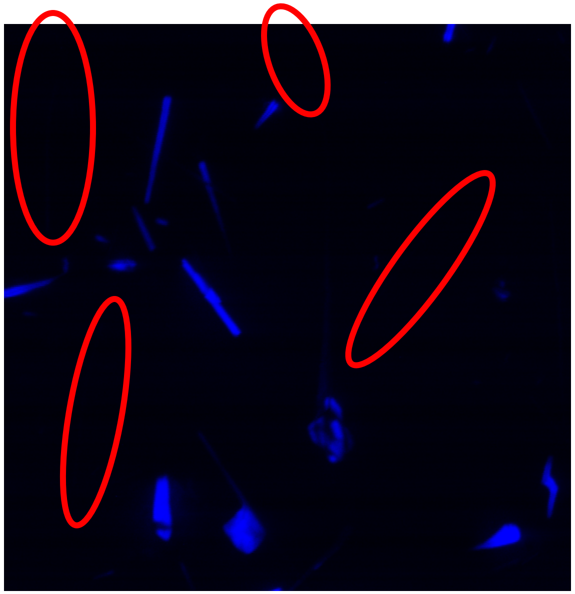
- CL source: UHV Zeiss Gemini FE-SEM
- CL detector: Gatan MonoCL3 (GaAs:Cs PMT)
- Environment: Room-temperature, UHV
- CL Resolution: <80 nm at 2.5KeV

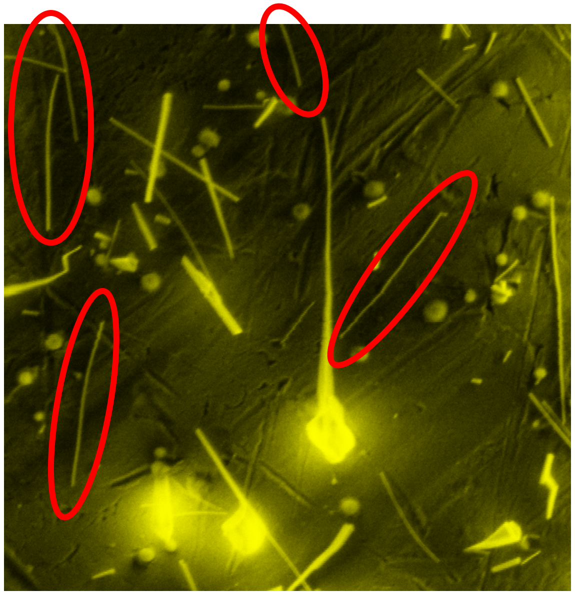
Spatial Resolution of CL – Monte Carlo Simulation for GaN



Band-edge luminescence "quenched" for thin NWs

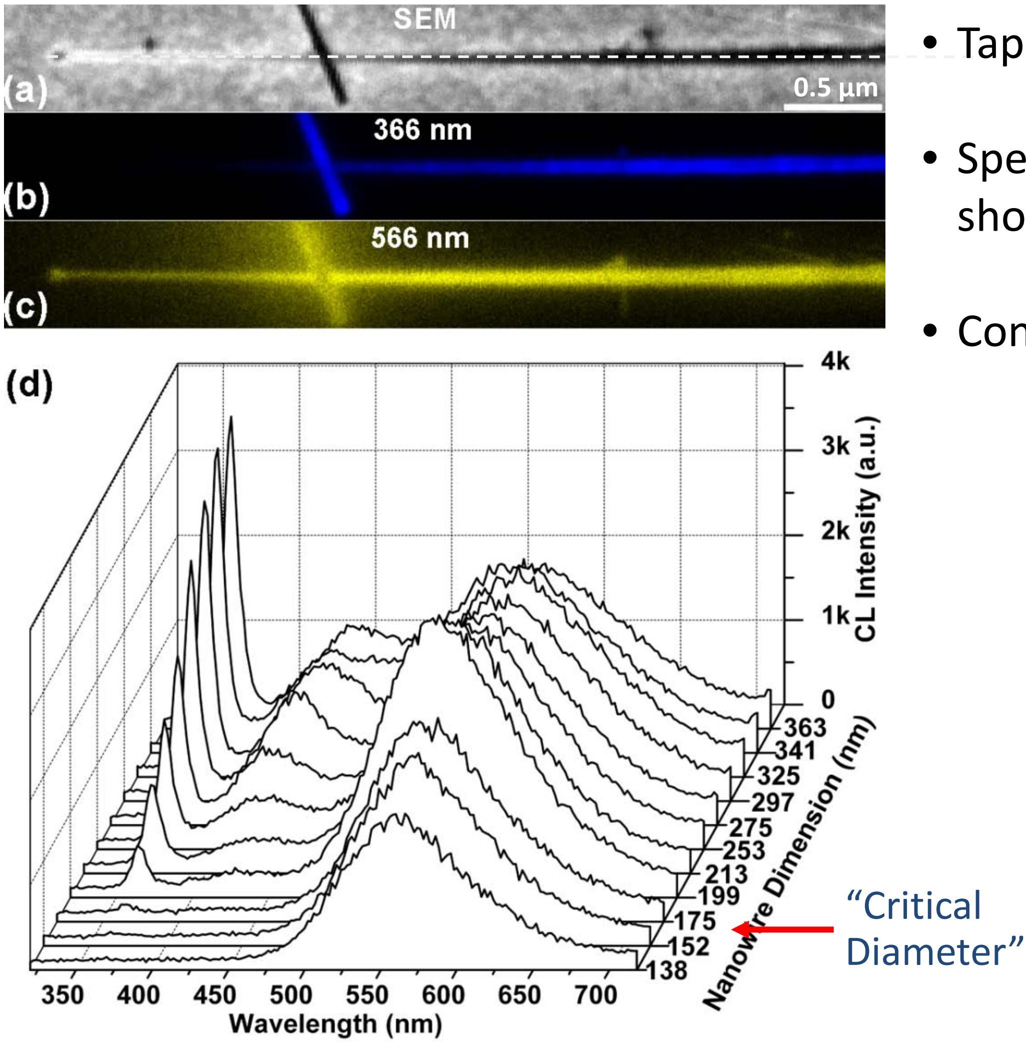






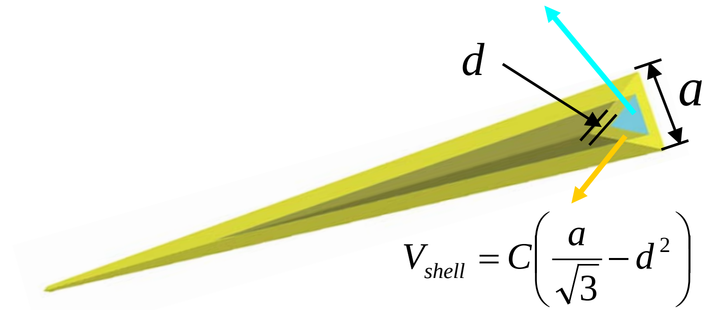
Band-edge luminescence (BEL) not observed for skinnier nanowires

YL surface layer causes "pinch-off" of BEL



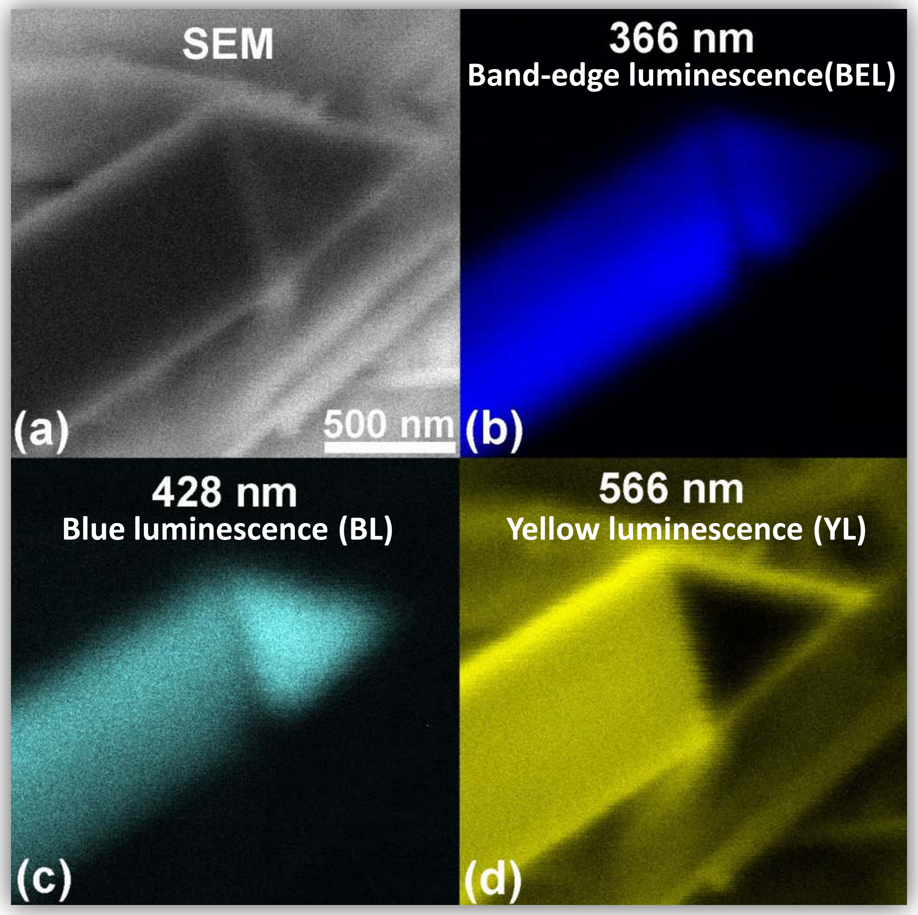
- Tapered nanowire shows "pinch-off" effect
- Spectra taken along tapered NW length, shows "critical diameter" of ~160 nm
- Consistent with YL surface layer model

$$V_{Core} = C\left(\frac{a}{2\sqrt{3}} - d\right)^2$$



$$V_{shell} = C\left(\frac{a}{\sqrt{3}} - d^2\right)$$

Spatially-resolved CL study of GaN NWs



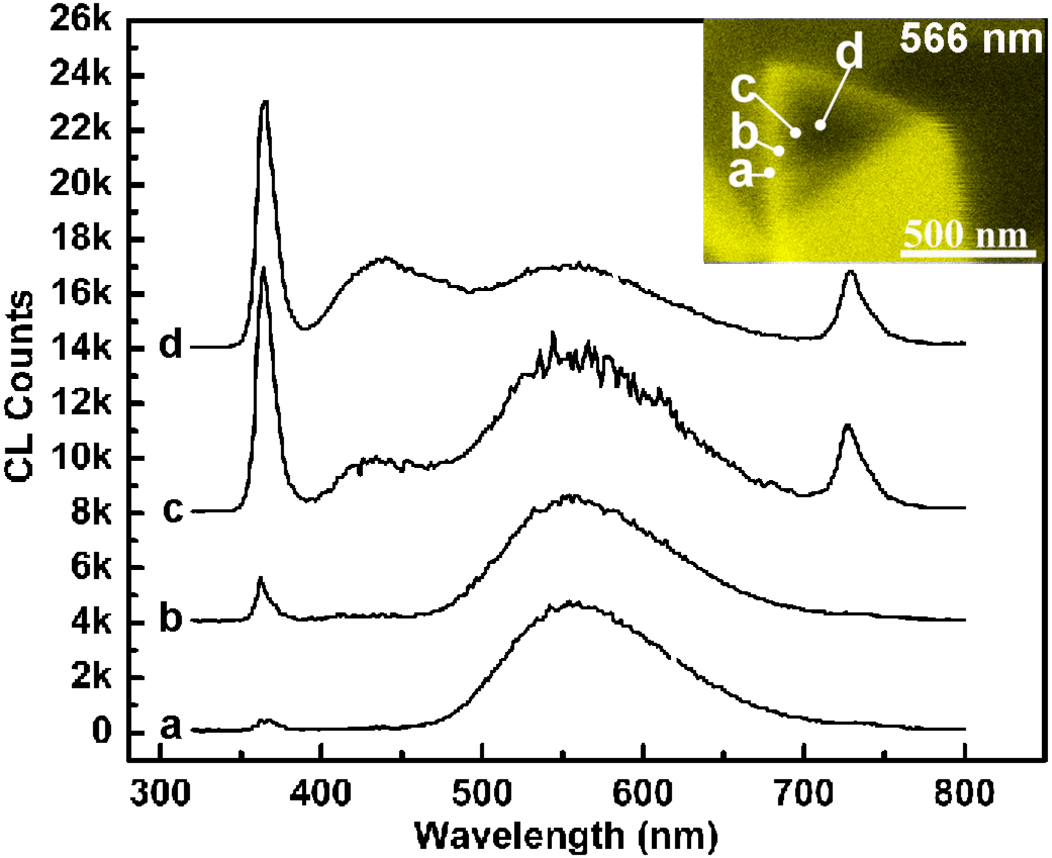
Nanoscale CL imaging: **Cross-section GaN NW**

- 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
- Well-known 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

Sandia National Laboratories

CL study of GaN NWs – 'Spot mode' spectroscopy

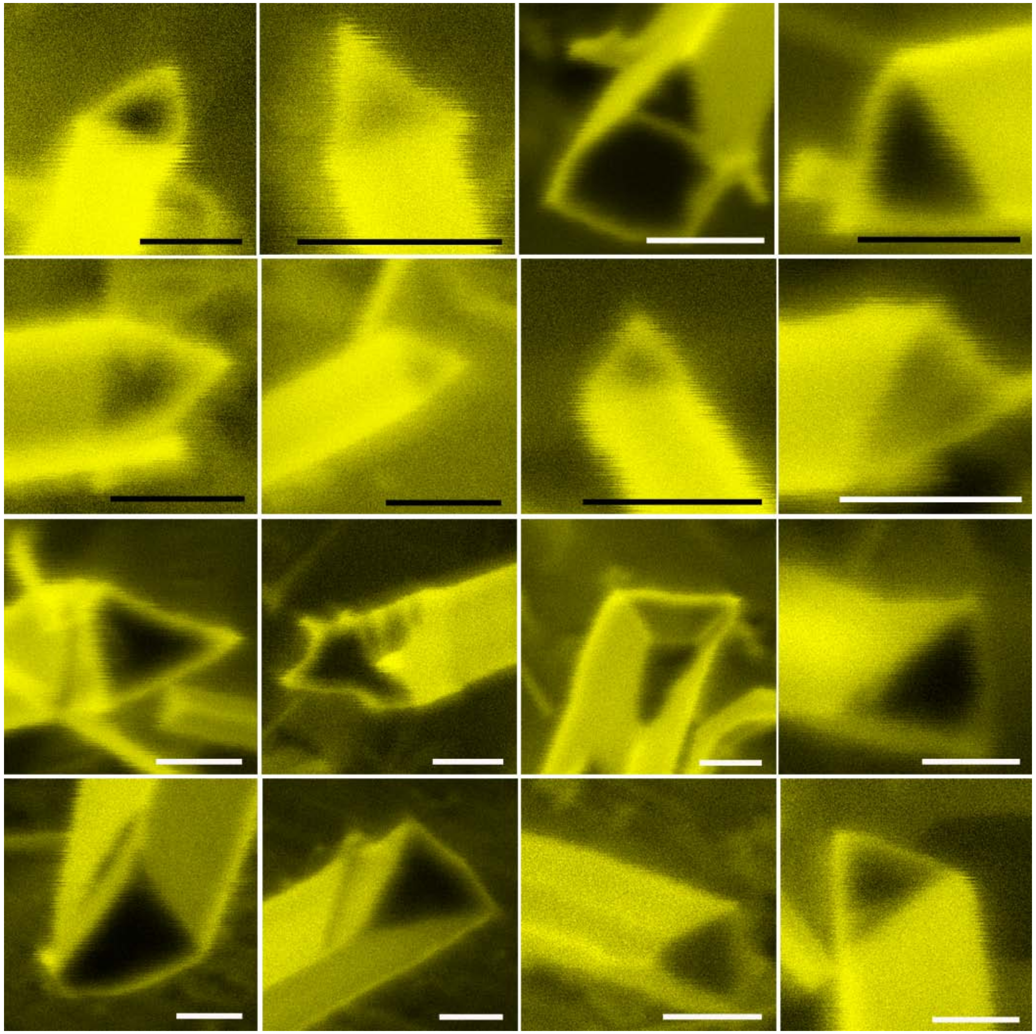


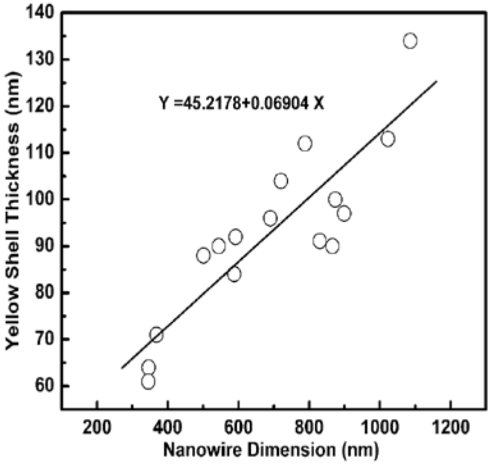
- Spot-mode: electron beam focused on different regions of NW cross-section, emission spectra collected
- YL strongest at surface
- BEL and BL strongest at NW center, quench at surface
- YL-related defects weaker but still present in the nanowire core (potentially less mobile C or O related impurities)

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

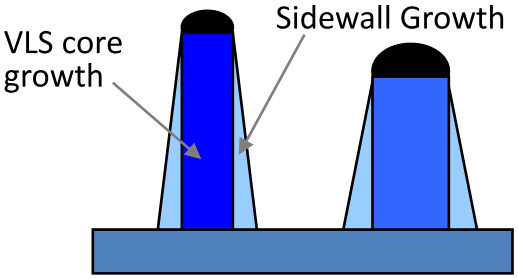
SSLS EFRC
SOLID-STATE LIGHTING SCIENCE ENERGY FRONTIER RESEARCH CENTER

CL study of GaN NWs – YL surface layer



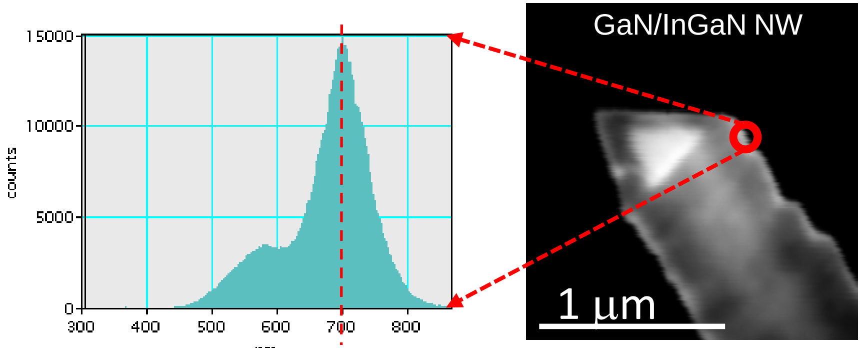


- YL thickness ∝ to the NW diameter
- Indicates YL-related defects incorporate though sidewall growth

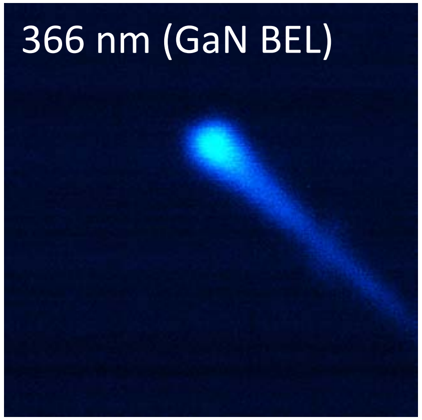


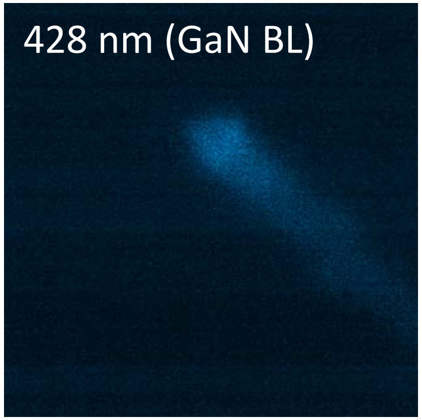
High Indium Incorporation in GaN/InGaN core-shell NWs

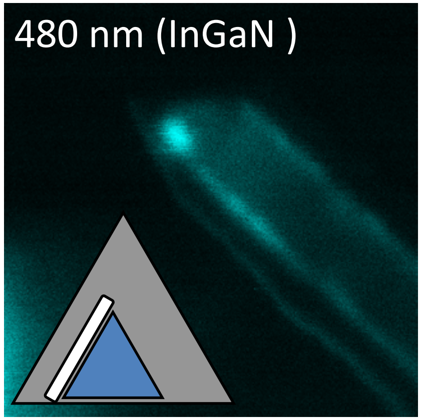
- InGaN: visible wavelengths for solid-state lighting, PV, etc.
- Strain limits practical In incorporation in InGaN thin films (e.g., green-yellow-red gap)

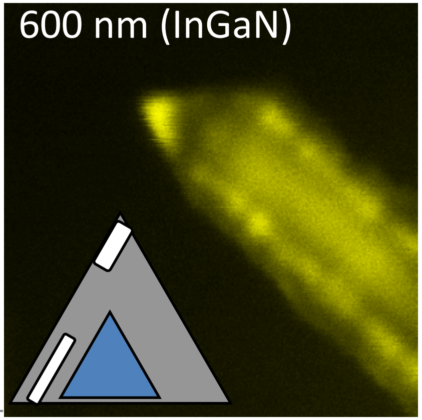


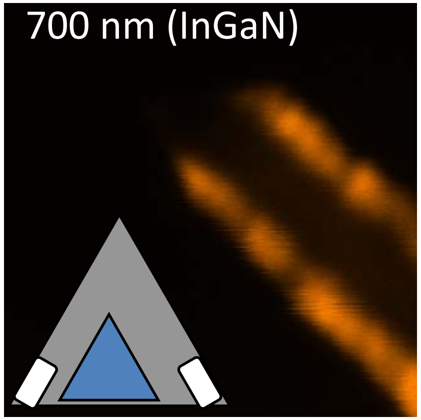
Growth conditions: GaN core – 900 ˚C, 10 min.
InGaN shell – 760 ˚C, 60 min.

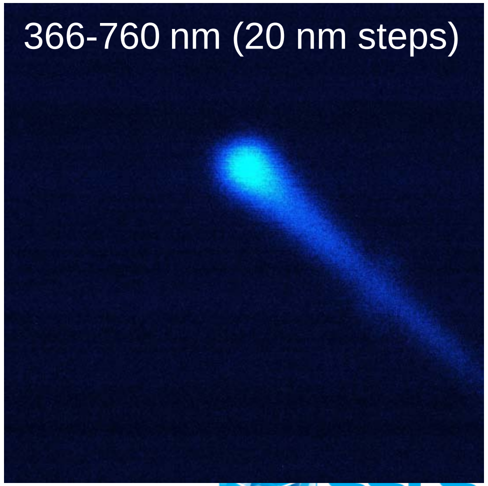






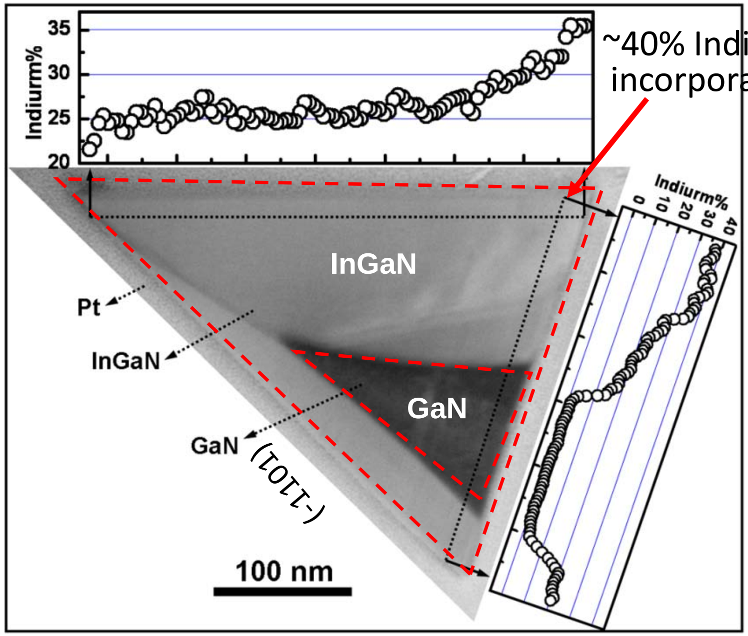




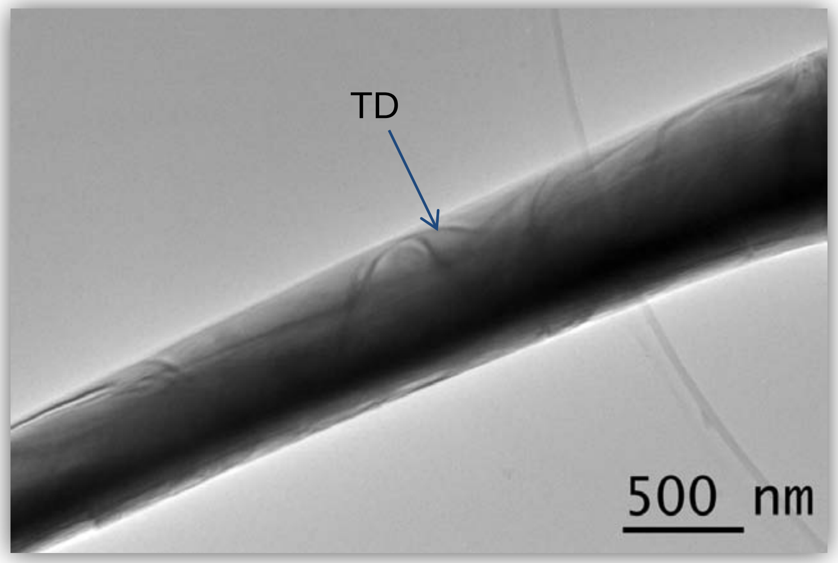


In incorporation in GaN/InGaN core-shell NWs

EDX(STEM)

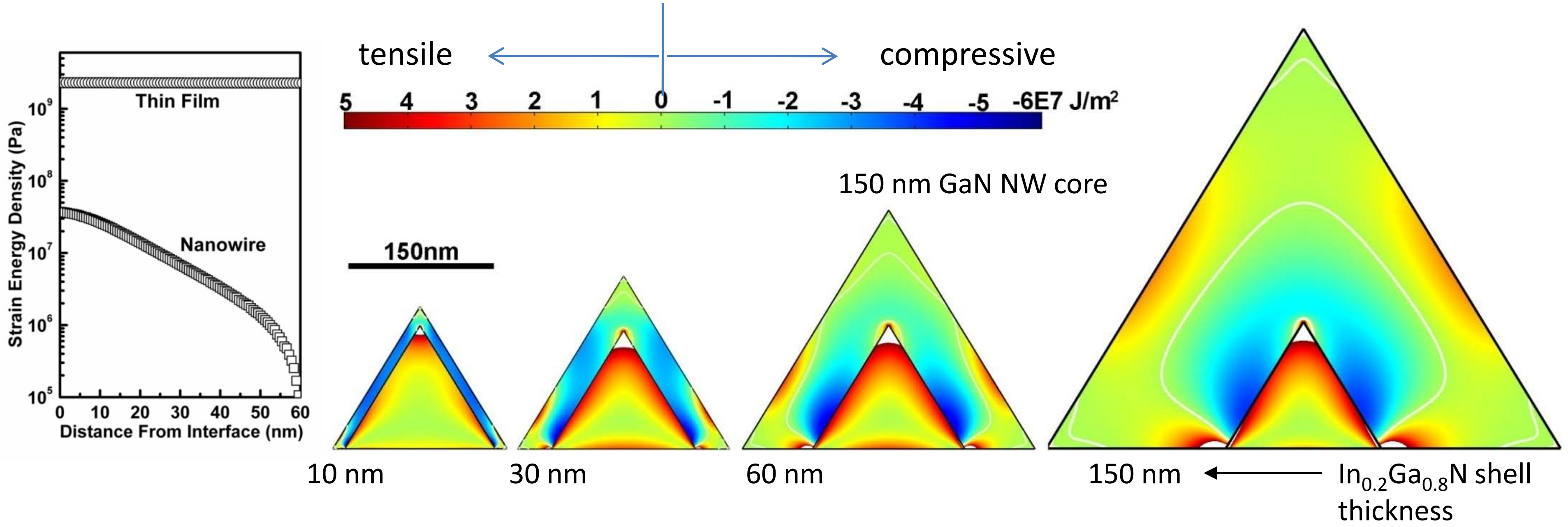


TEM



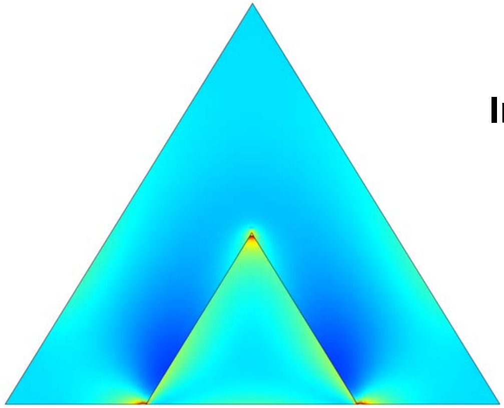
- InGaN shell growth highly facet-dependent -- no growth on (000-1) c-plane facet
- In concentration increases away from GaN/InGaN interface, highest at corners
- Strain in InGaN NW shell much lower than for InGaN thin film
- Threading dislocations observed via low resolution TEM

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



- Strain lower for InGaN/GaN NW than for thin film
- Finite element models show compressive/tensile strain in GaN core and InGaN shell
- Compressive strain dominates in thinner shells, decreases away from interface and becoming tensile for thicker shells
- Higher In incorporation correlated with lower (compressive) strain regions

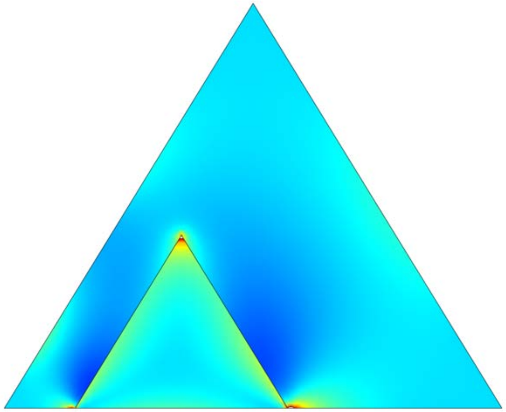
Strain – Symmetric vs. asymmetric InGaN shell



Mean:3.89E5 J/m2

$In_{0.2}GaN/GaN$

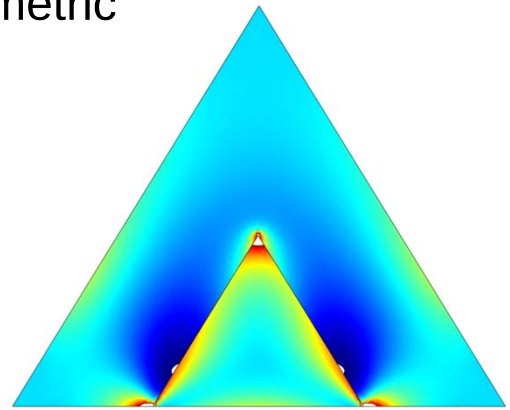
In=20%



Mean:1.16E5 J/m2

Symmetric

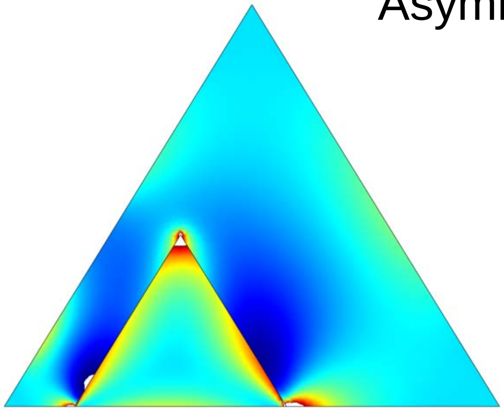
Asymmetric



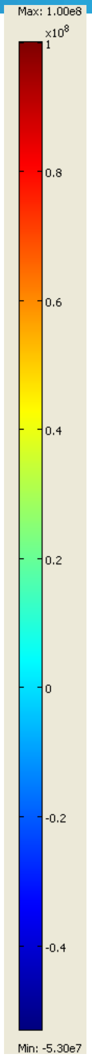
Mean:6.69E5 J/m2

$In_{0.3}GaN/GaN$

In=30%



Mean:1.34E5 J/m2



Asymmetric InGaN shell shows significiantly less strain

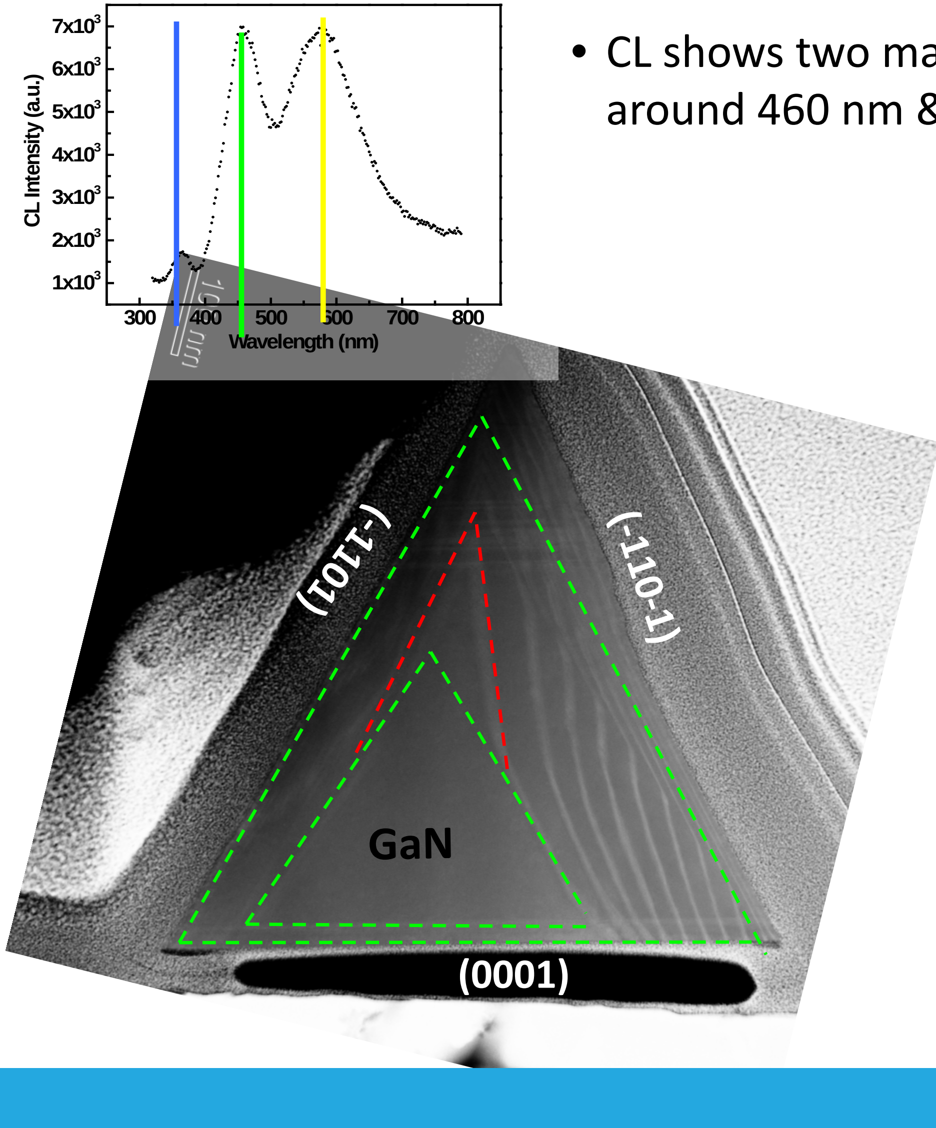
Conclusions

- Spatially resolved CL reveals a yellow luminescence (YL) surface “layer” in GaN NWs
- YL-related defects may be relatively mobile & migrate to the surface region during growth
- For InGaN shell growth, up to 40% In incorporation is observed
- CL reveals higher In incorporation at corners of InGaN/GaN NWs and asymmetric shell growth due to strain relaxation

<u>Funding Acknowledgment:</u> DOE Basic Energy Sciences (BES) DMSE, Sandia’s Solid-State-Lighting Science Energy Frontier Research Center (DOE BES), and Sandia's LDRD program

Further questions? gtwang@sandia.gov

Multiquantum well InGaN/GaN nanowires



- CL shows two main InGaN peaks around 460 nm & 580 nm