SAND2015-6660C

Deterministic Placement of Fabricated InGaN Quantum Dots in Photonic Structures

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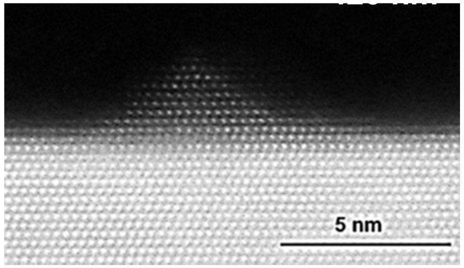
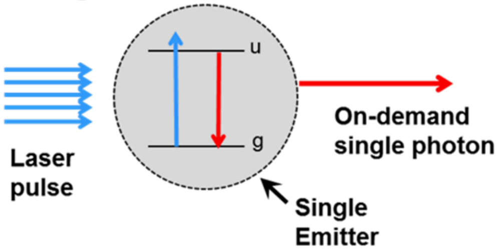
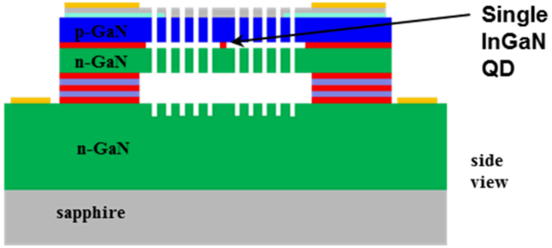
Outline:

1. **Motivation: InGaN Quantum Dots**
2. **InGaN QD fabrication- ensembles**
    - Quantum size controlled PEC etching
    - AFM/TEM analysis
    - Photoluminescence
3. **Deterministic placement of InGaN QDs**
    - Design of InGaN single photon source
    - FDTD calculations
4. **InGaN post samples**
    - Microphotoluminescence
    - g2 measurements
5. **Summary/Conclusions**

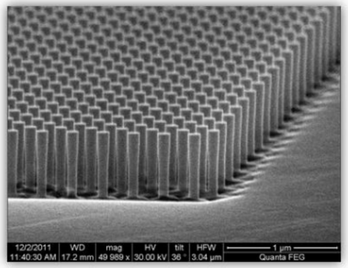


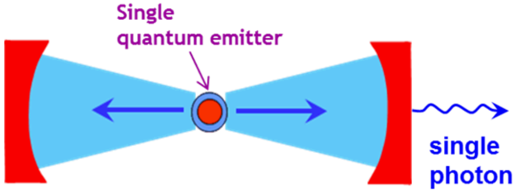
Single Quantum Emitter





InGaN QD Single Photon Source





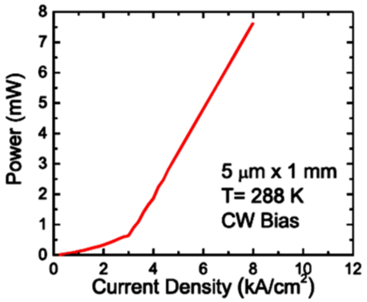
Motivation for InGaN Quantum Dot (QD) Emitters

- **Long wavelength visible emitters:**
  - **Nanostructure (NWs, QDs) can incorporate more indium**
  - **High efficiency yellow, orange, and red emission**
  - **RGB and RYGB emitters require high efficiency yellow to red emitters**
- **Visible QD diode lasers:**
  - **Lasers for lighting is gaining momentum**
  - **Low threshold, high efficiency, better temperature performance**
  - **Monodisperse QDs**

InGaN QD laser:

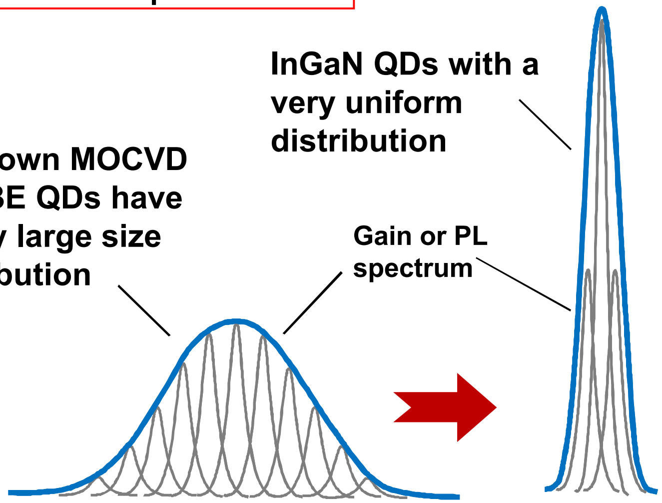
- **University of Michigan**
- **Electrically injected**
- **630 nm**
- **T_o = 236K**

Frost et al., IEEE JQE, **49,** 923 (2013).



Monodisperse QD Distributions

Impact on device performance

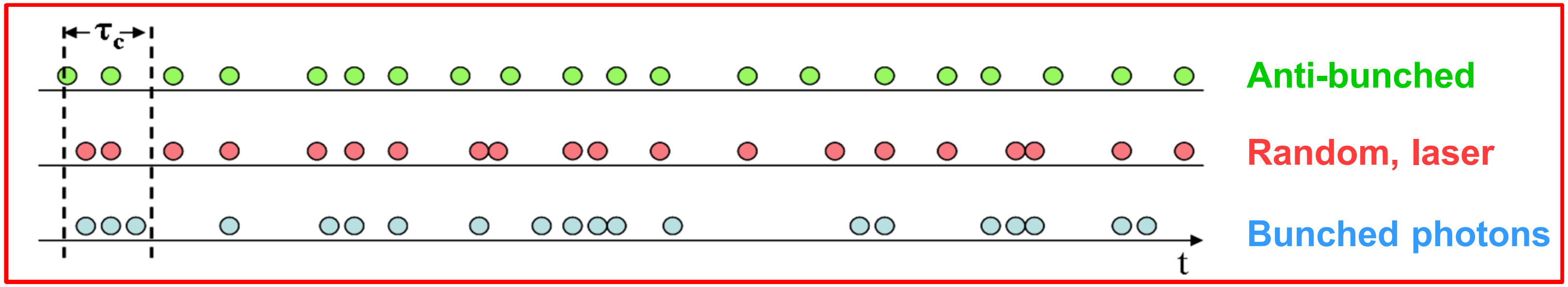


As grown MOCVD or MBE QDs have a very large size distribution

InGaN QDs with a very uniform distribution

Gain or PL spectrum

Single Photon Source: Photon Statistics



source: J.S. Lundeen

Classical light sources: **g(2)(t = 0) ≥ 1**

- Bunched or random photons (Laser, LED, thermal source)

Quantum light source: **g(2)(t = 0) < 1**

- Anti-bunched photons (Quantum optics theoretical treatment)

Applications:

- **Quantum key distribution**
  - **Attenuated laser can be used**
  - **SPS is the gold standard for QKD**
  - **Future QKD involving quantum repeaters**
- **Quantum metrology**
- **Quantum computing with photons**
- **True random number generation**

Single Photon Sources in the Literature

Faint Laser

- Not a quantum light source ($g^{(2)}(0) = 1$)

On-Demand Single Photon Sources:

- Single atoms/ions/molecules
  - $g^{(2)}(0) = 0.015$, cryogenic temperatures
- Color centers (nitrogen vacancy)
  - $g^{(2)}(0) = 0.07$, 300K
- InAs quantum dots
  - $g^{(2)}(0) = 0.02$, 5K, electrically injected
- CdSe/ZnS quantum dots
  - $g^{(2)}(0) = 0.004$, 300K, 5% extraction
- GaN-based SPSs
  - $g^{(2)}(0) = 0.4$, 200K
  - $g^{(2)}(0) = 0.16$, 10K, Electrically injected
  - $g^{(2)}(0) = 0.13$, 300K

Heralded Single Photon Sources:

- Parametric down conversion
  - $g^{(2)}(0) < 0.01$, 300K, pulsed laser
- FWM in optical fibers
  - $g^{(2)}(0) \sim 0.01$, 300K, pulsed laser

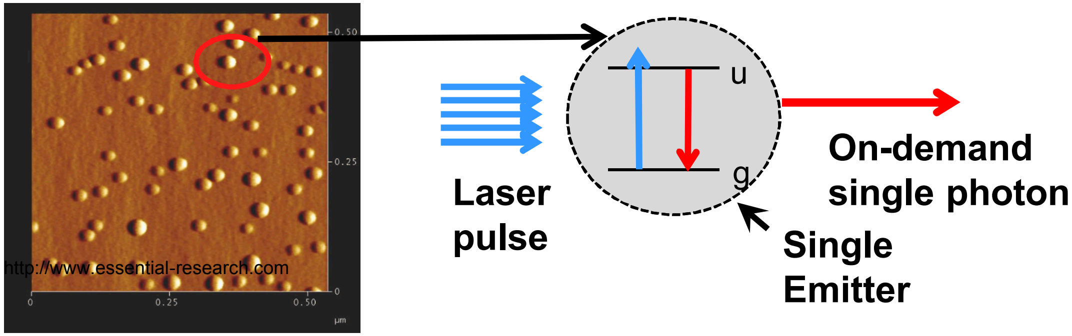
*M.D. Eisaman, et al. Rev. Sci. Instrum. **82,** 071101 (2011)

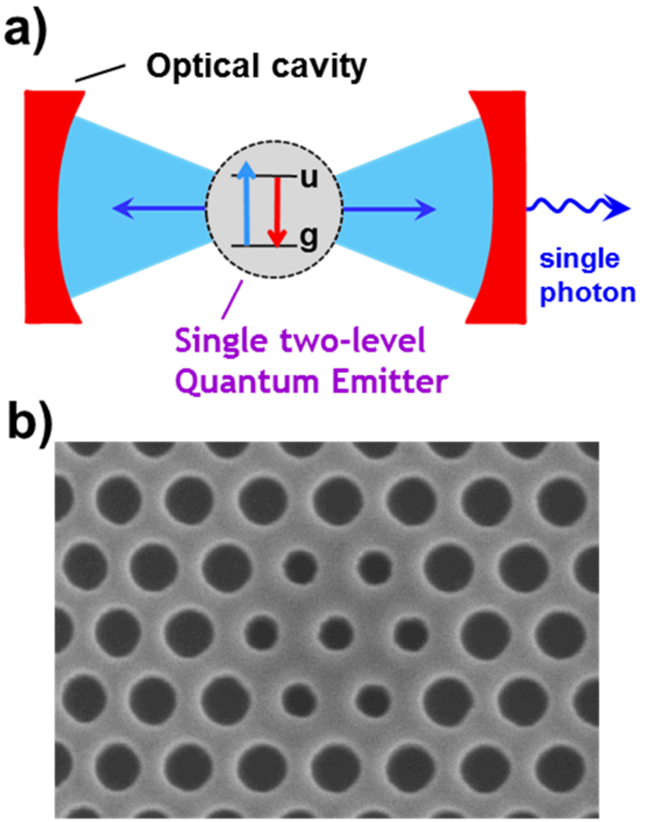
Ideal Single Photon Source:

- ➢ Triggered (on-demand)
- ➢ $g^{(2)}(0) = 0$
- ➢ Indistinguishable photons
- ➢ High repetition rate
- ➢ Room temperature
- ➢ Electrically injected
- ➢ High repetition rate
- ➢ Chip-scale

Single Photon Source based on Quantum Dots

AFM image of InAs QDs





Sandia 2D photonic Crystal Cavity

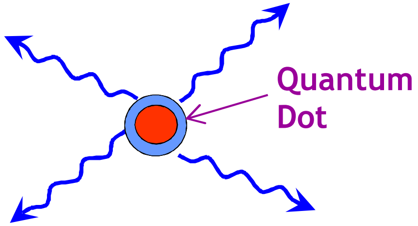
QD Single Photon Source:

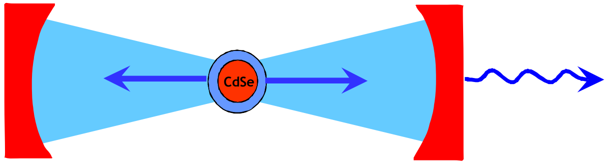
- Isolate emission from a singe quantum dot
- Only one photon can be emitted
  - Absorption of one photon saturates transition
  - Can be excited using many photons
  - **Deterministic Source: on demand**
- Triggered emission within radiative lifetime
  - Short emitter lifetime (~ 1 ns) → Fast rep. rate
- **Path to electrical-injection/chip-scale integration**

Single Emitters Inside and Optical Cavity

Why do we need an optical cavity?

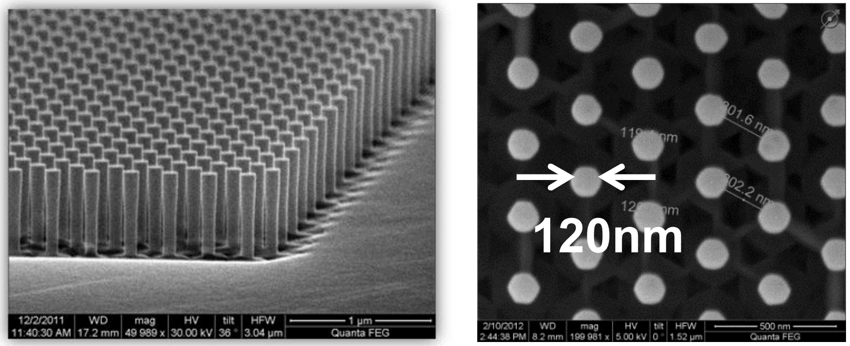
- Improve photon collection efficiency
- Light collection: emission into specific spatial modes
- Limit spectral content→ indistinguishable photons
- Generate photons with a specific polarization
- Increase radiative rate, efficiency via Purcell effect
- Theoretically study QD strong coupling physics
    - Coherent and incoherent interaction with phonon bath



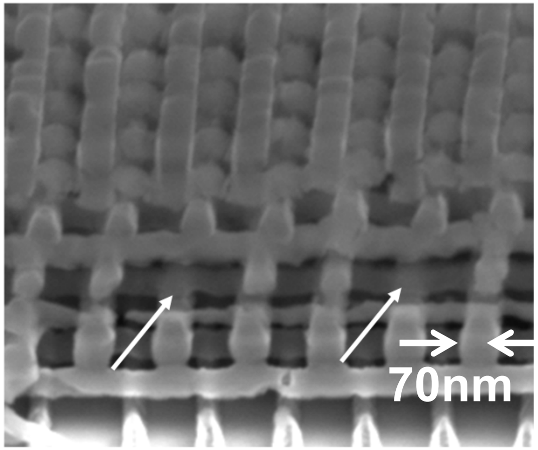


Directional emission

Sandia III-Nitride Nanostructure Fabrication



Scientific Reports **3,** 2982 (2013)

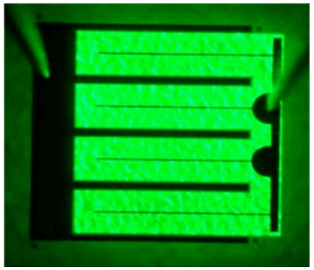


Nanoletters **11,** 4591 (2011)

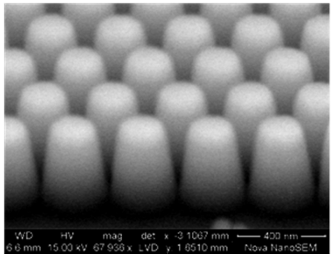
InGaN Quantum Dot SPS Development

- ***Goal:*** Room temperature, electrically-injected, chip-scale single photon source
- Our approach:
  - **MOCVD-grown InGaN QWs → PEC etched InGaN QDs**
  - **Design a PhC cavity for InGaN QD SPSs**
- Advantages to this approach
  - **InGaN QDs can operate at high temperatures**
  - **Si detectors work well in the blue/green**
  - **Free space data links: Reduce the size of optical telescopes**
  - **Plastic optical fiber at blue/green wavelengths**
  - **Deterministic placement of InGaN QDs**
  - **Leverage expertise in III-nitrides and photonic crystals**

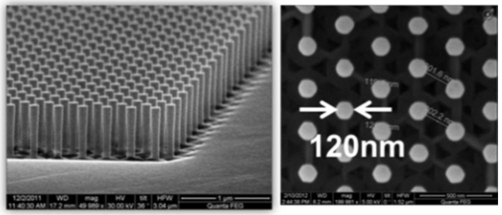
Green InGaN LEDs



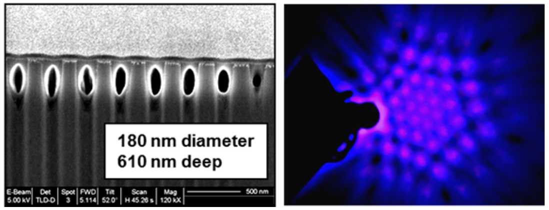
InGaN nano-LED array



GaN nanowire arrays



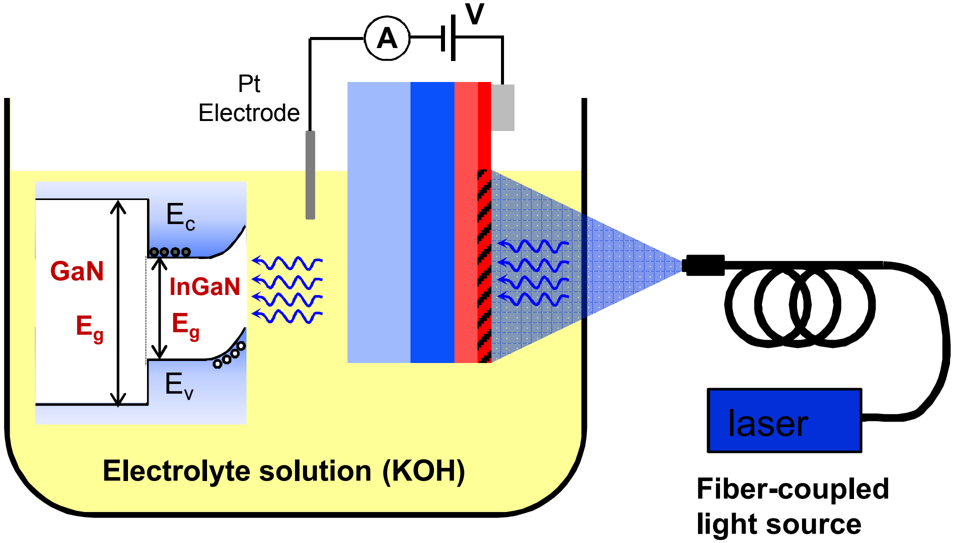
InGaN photonic crystal LEDs



Introduction to InGaN PEC Etching

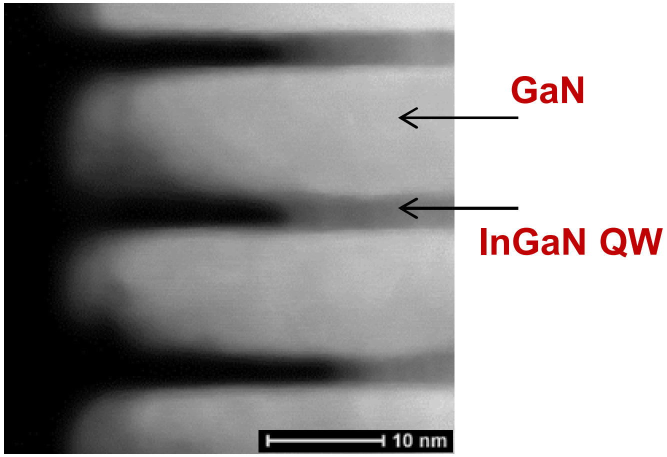
Photoelectrochemical (PEC) Etching:

- Very few wet etches work for III-nitrides
- Band gap selective (Etch InGaN over GaN)
- Dopant selective, light intensity dependent, etch current can be monitored
- Laser or lamp excitation (Xe arc lamp, tunable ps Ti:S)
- KOH (~0.1M) typically used as electrolyte



Fiber-coupled light source

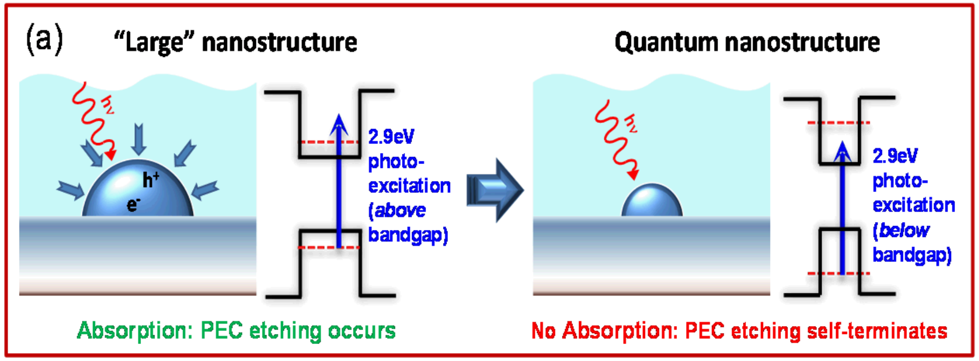
PEC etched InGaN/GaN QWS



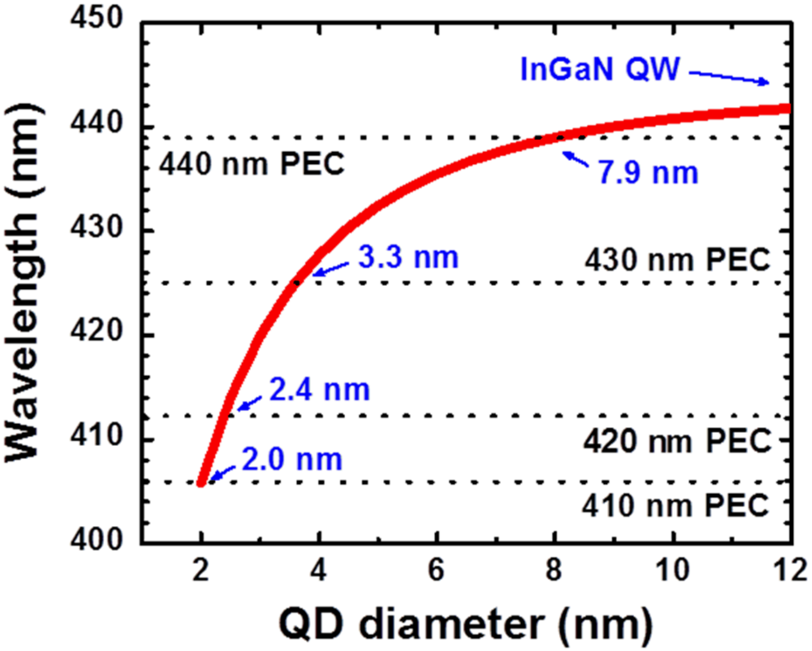
Fabrication of InGaN QDs via PEC etching

Quantum Size Control: Use size quantization to control QD size

Self-limiting PEC etch process:



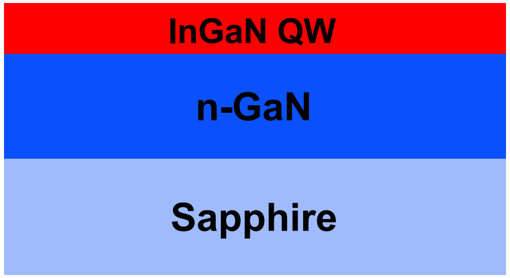
- For QDs, band gap depends on size
- As etch proceeds,
  - QD size gets smaller, band gap goes up
  - Etch terminated for $E_g > E_{photon}$ pump
- Self-terminating etch process
- QD size depends on PEC wavelength
- Monodisperse QD distributions ??
- Prof. Torimoto- colloidal QDs



G. Pellegrini, et al., Journal of Applied Physics 97, 073706 (2005).

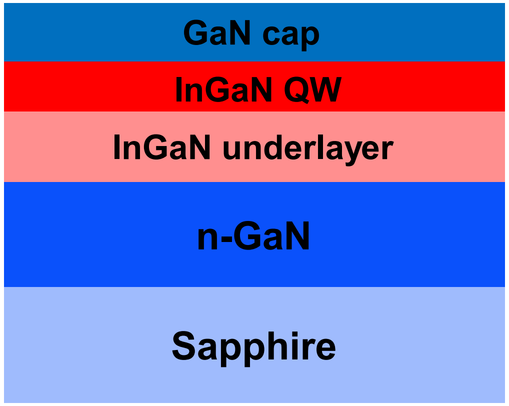
MOCVD-grown InGaN samples

Uncapped single InGaN QW



- Grown by MOCVD on sapphire substrates
  - 5 - 20 nm thick InGaN layer
- No InGaN underlayer is used in this sample
- Uncapped single QW sample (14% In):
  - Amenable to surface characterization of QDs
  - TEM, AFM characterization
  - Luminescence weaker than capped sample

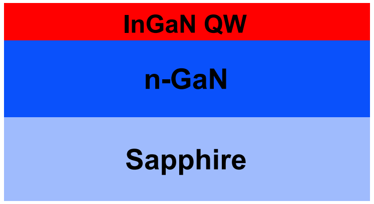
Capped single InGaN QW



- Grown by MOCVD on sapphire substrates
  - 3 nm InGaN QW, 10 nm GaN cap
- InGaN underlayer (~2% In) used in this sample
- Capped single QW sample (14% In):
  - AFM is not useful for capped samples
  - Luminescence brighter than uncapped samples
- Etch is thought to proceed via pits, dislocations

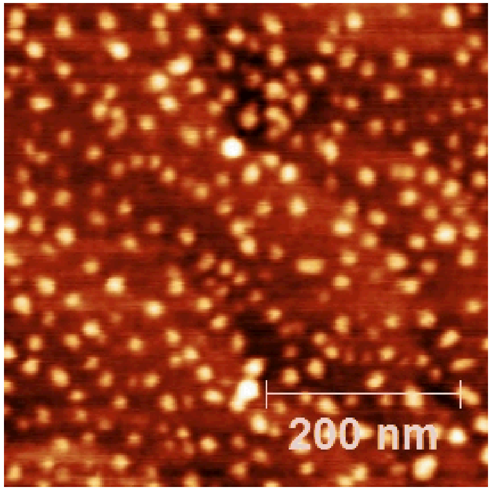
Atomic Force Microscope (AFM) Measurements

Uncapped InGaN QW

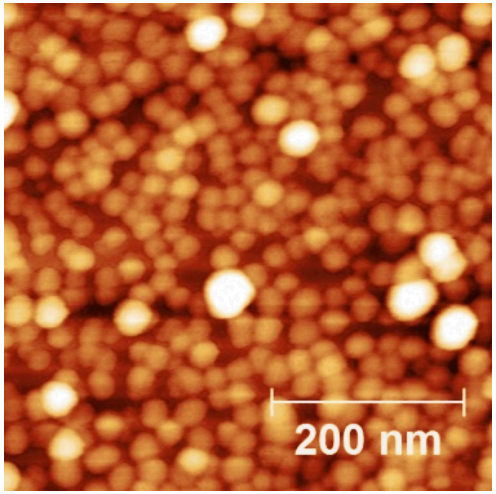


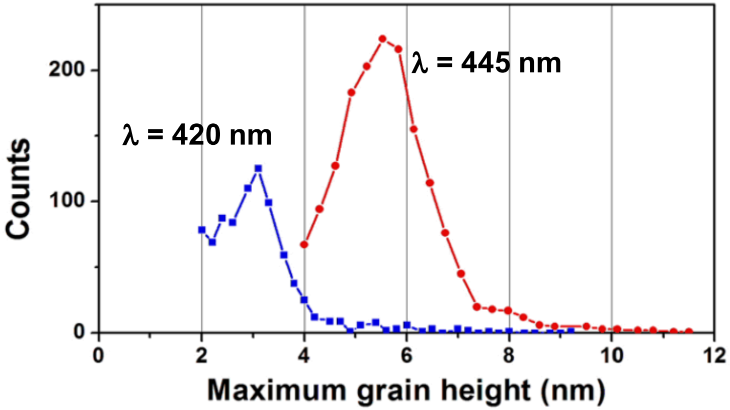
- Samples etched for two hours at 420 nm and 445 nm
  - Laser power density: ~ 3 mW/cm^2
- High dot density: 10^{11}/cm^2
- Some big dots (10-20 nm) remain: due to dislocations?
- QD size depends on PEC etch wavelength

PEC etch→ λ = 420 nm



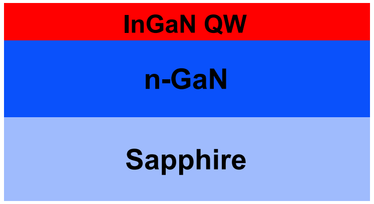
PEC etch → λ = 445 nm



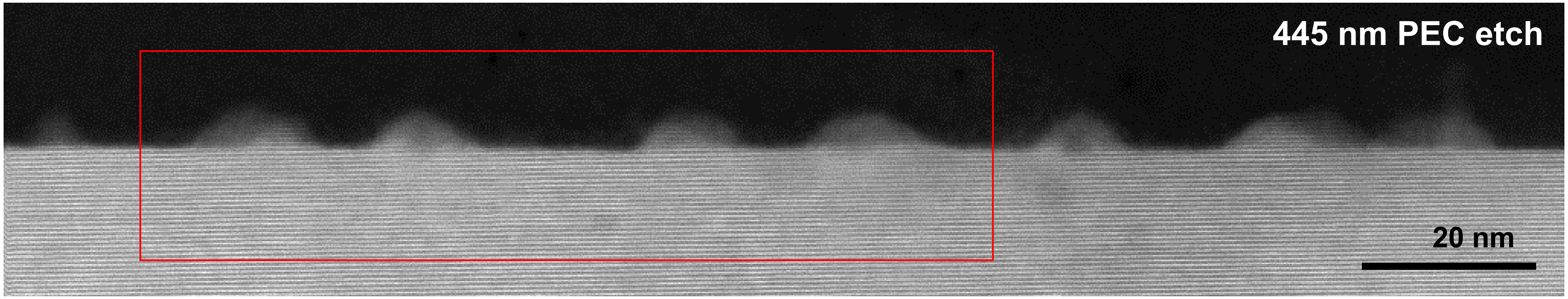


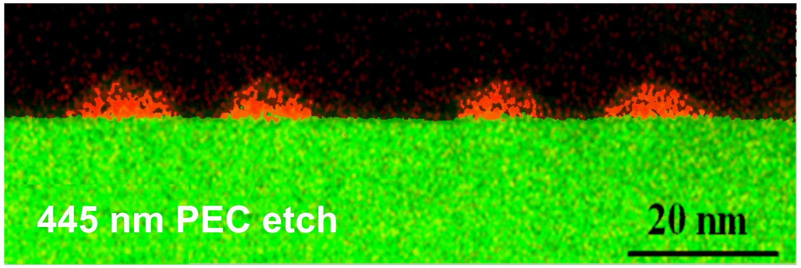
Transmission Electron Microscope (TEM) Images

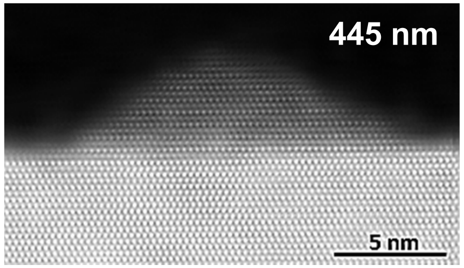
Uncapped InGaN QW

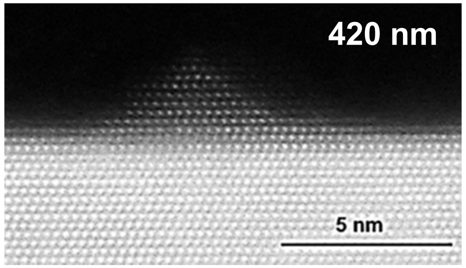


- High-angle annular dark-field (HAADF) TEM images
- Samples etched at 420 nm and 445 nm
- Energy dispersive x-ray mapping
  - QDs on surface are InGaN
  - Red = indium, green=gallium
- InGaN QDs are epitaxial to the underlying GaN
- No underlayer, no cap → PL is not very bright



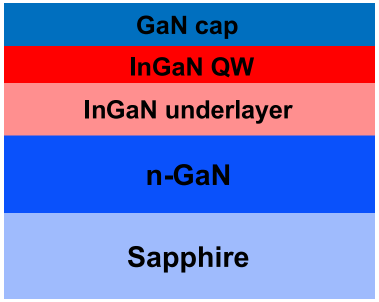




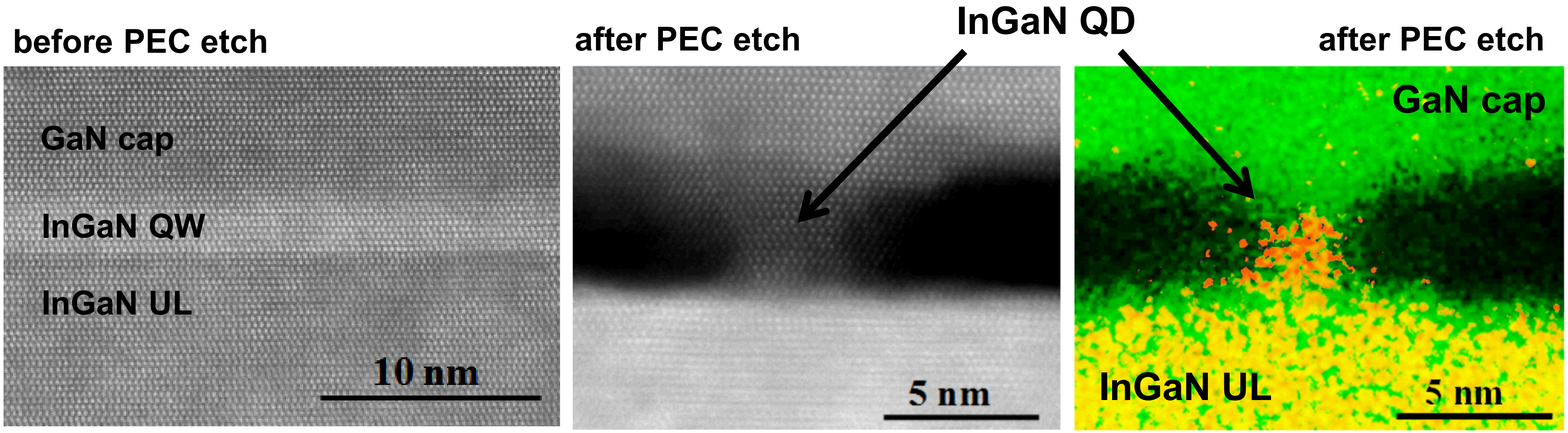


Transmission Electron Microscope Images

Capped InGaN QW

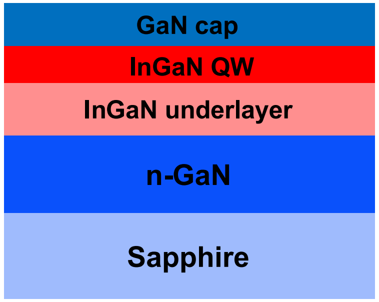


- High-angle annular dark-field (HAADF) TEM images
- Sample etched at 420 nm
- EDX mapping shows that dots are InGaN
- InGaN QDs are epitaxial to the underlying GaN
- 2% InGaN underlayer + GaN cap → PL is much brighter
- GaN cap provides partial passivation



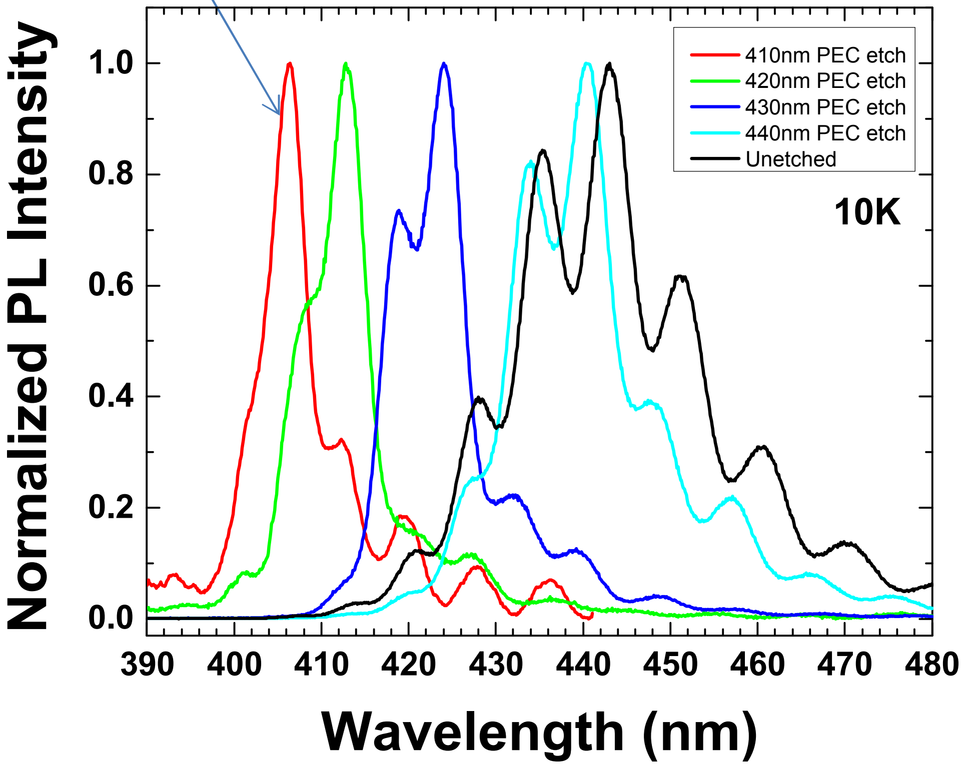
Photoluminescence from fabricated InGaN QDs

Capped InGaN QW



GaN cap
InGaN QW
InGaN underlayer
n-GaN
Sapphire

As narrow as **6 nm** FWHM is consistent with a narrowing of the QD size distribution



Photoluminescence (PL) data:

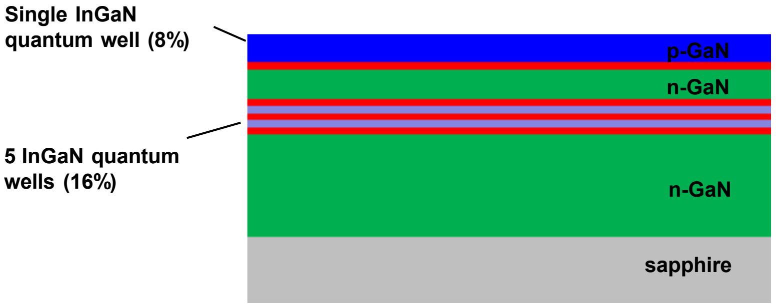
- **375 nm pump (ps pulsed)**
- **10K PL data**
- **PL wavelength determined by PEC etch wavelength**
- **PL linewidth: 24 nm → 6 nm**
- **Quantum size-controlled PEC etching works!**

Nanoletters **14,** 5616 (2014)

Deterministically positioned InGaN QDs

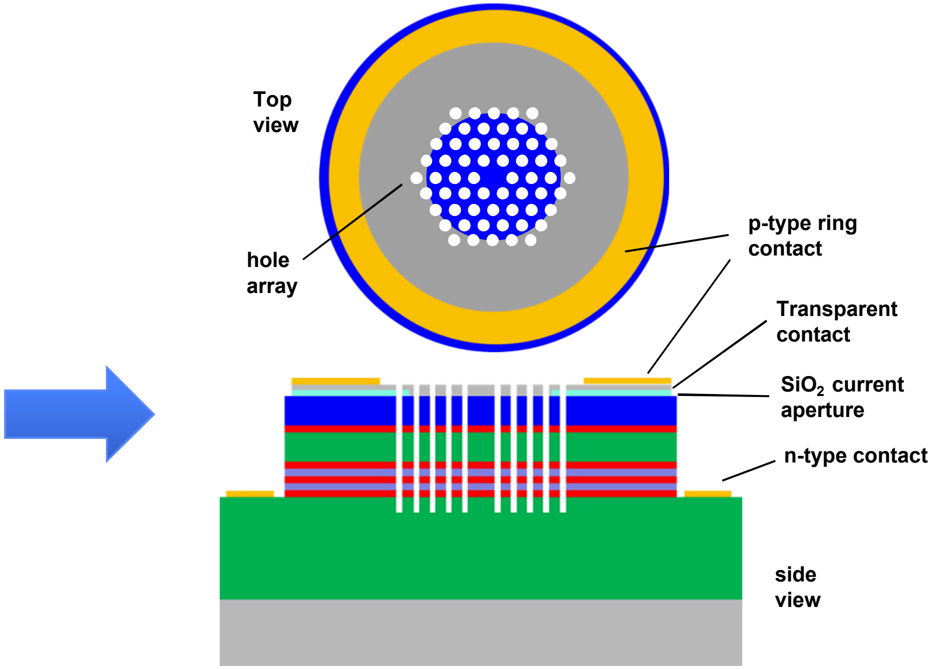
Standard III-nitride device fabrication

1.) Epitaxial layer structure



- Specially design epitaxial structure for PEC etching
- InGaN QWs with 8% and 16% indium

2.) Fabrication of device plus PhC etch

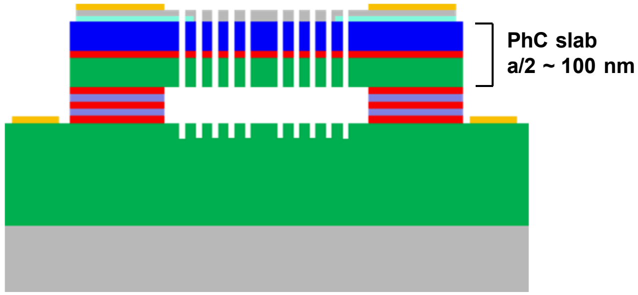


- Standard LED fabrication
- Etch PhC triangular array
- defect cavity at the center
- Requires current aperture
- Use transparent contact

Deterministically positioned InGaN QDs

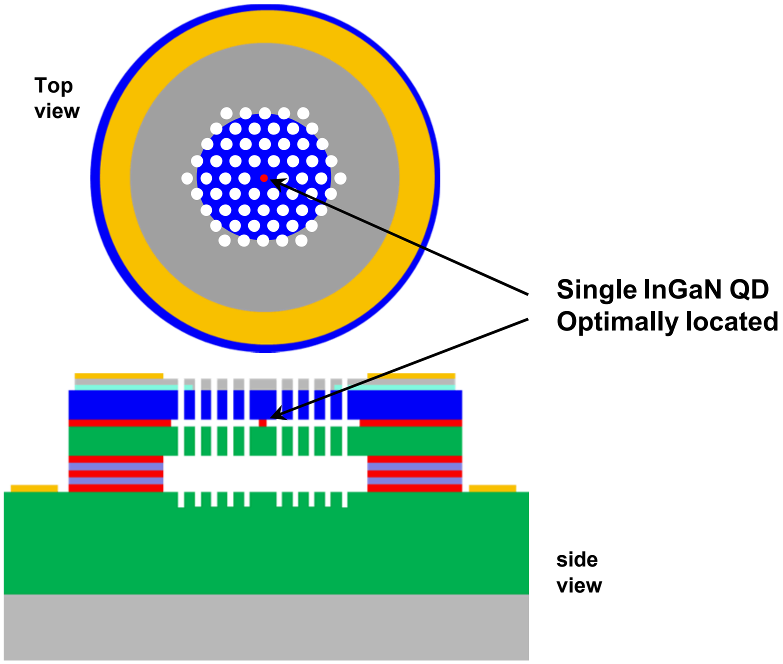
PEC etch of InGaN QWs

3.) First photoelectrochemical etch



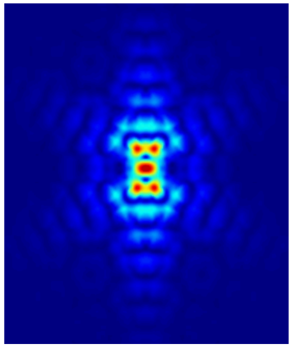
- Use 450 nm laser for PEC etch
- Area under contacts will not etch
- 8% InGaN QW will not etch

4.) Second photoelectrochemical etch

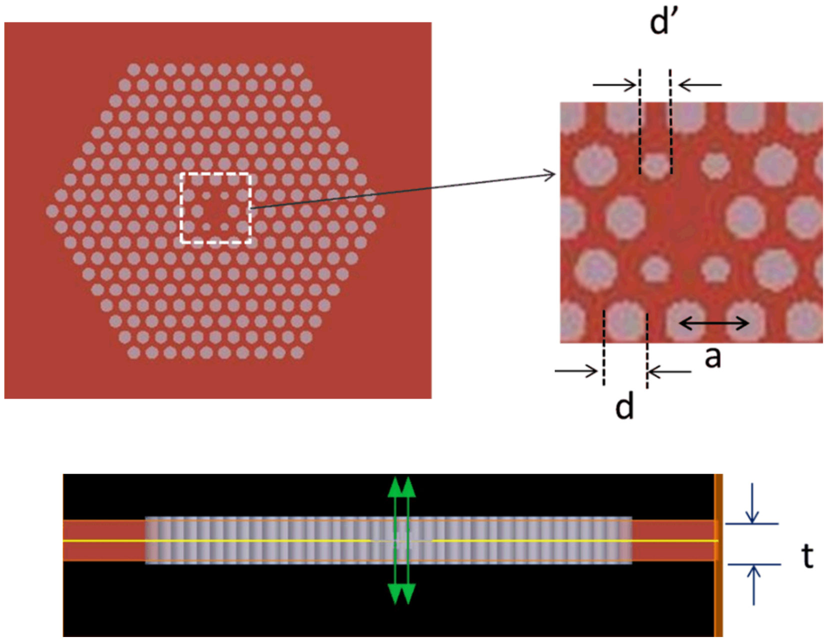


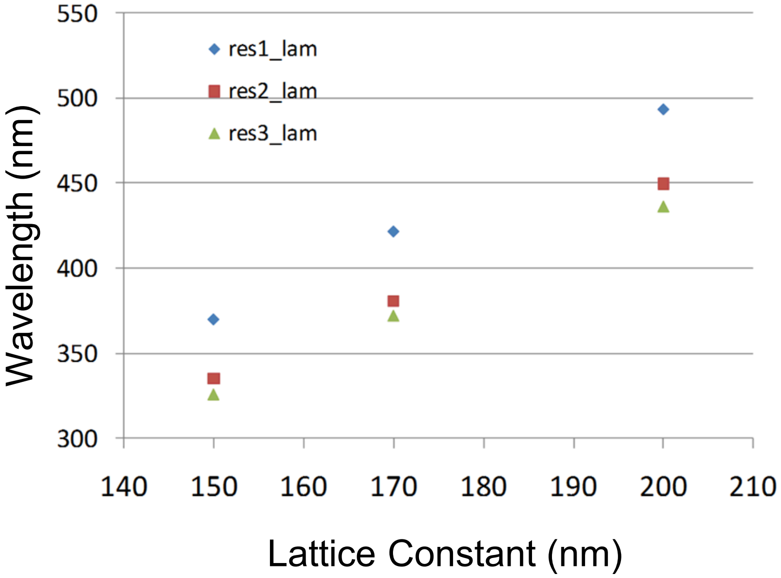
- Use 405 nm laser for PEC etch
- Self limiting PEC etch
- Size quantization in QD will raise energy level above 405 nm

FDTD Photonic Crystal Modeling

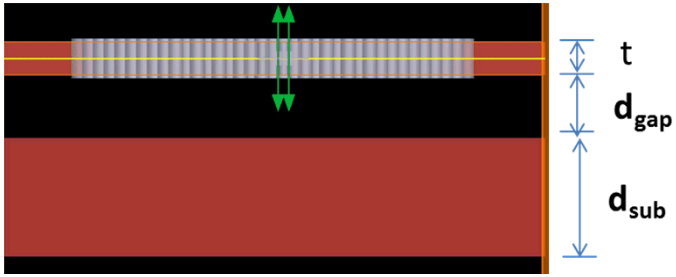


- Calculations done using Lumerical
- Need resonance with anti-node at center (res1)
- No Substrate: Q = 941 for this design
  - a=170 nm, d=0.7a, d’=0.45a, t=0.5a
- Design is not fully optimized (higher Qs are possible)
- Need lattice constant in the range from 170 – 200 nm





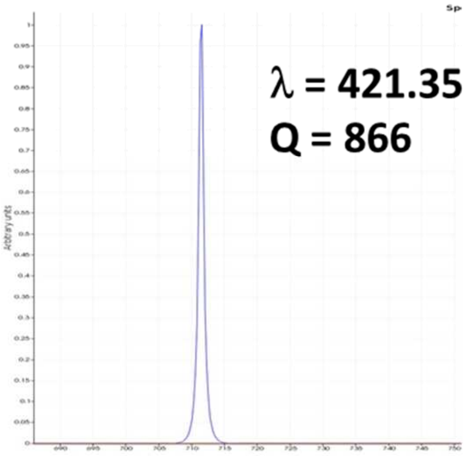
FDTD Modeling: Spacer Layer Thickness



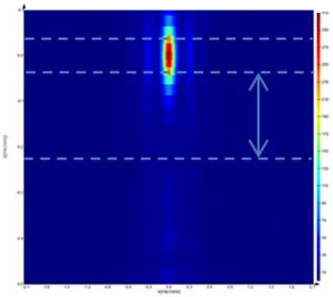
- Determine thickness for air gap
- Calculations done using Lumerical
- For d(gap) < a mode leaks out into substrate
- 10 or more sacrificial InGaN QWs required
- Air gap should be at least one lattice constant in thickness

a = 170nm
d = 0.7a
d' = 0.45a
t=0.5a

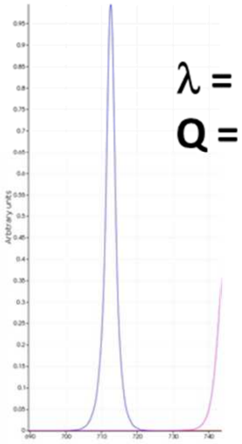
d_{gap} = 0.95a



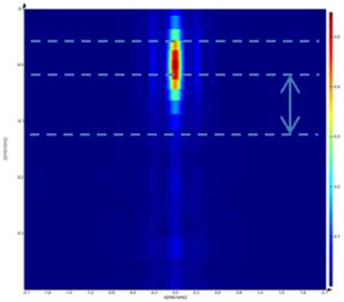
λ = 421.35 nm
Q = 866



d_{gap} = 0.6a



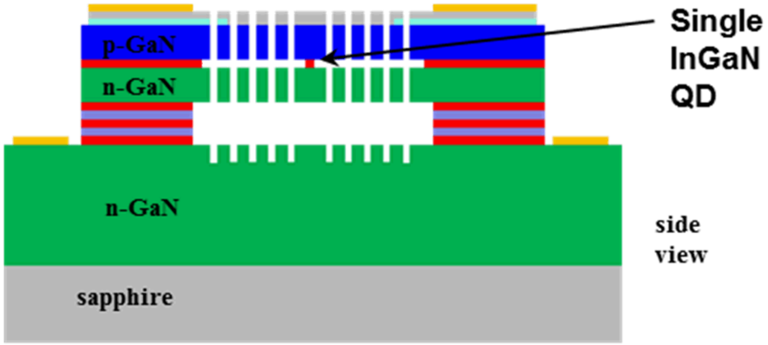
λ = 420.765 nm
Q = 237



Calculated far-field patterns for InGaN SPS

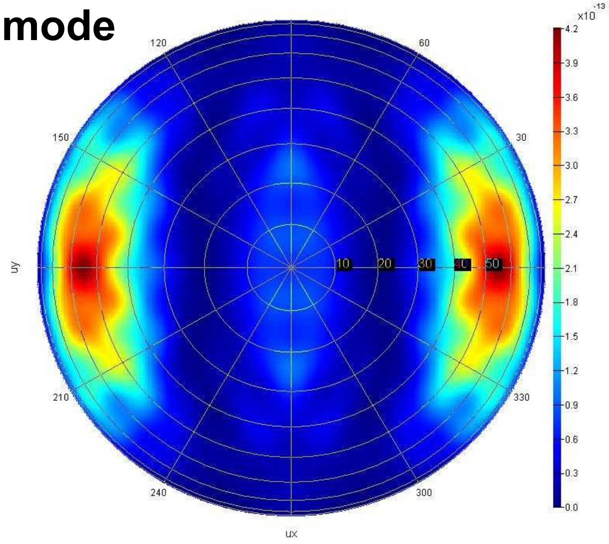
- Performed FDTD calculation for different cavities
- Calculations show that the highest Q cavities tend to have a resonance with a node at the center
- Cavities for PEC-etched InGaN QDs should have a field maximum at the center of the cavity
- FDTD calculation show that light extraction can be enhanced (~ 5X) by collecting at large angles relative to surface normal

Design for InGaN SPS:



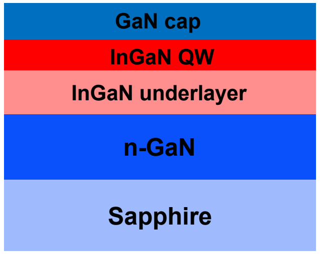
InGaN QD Single Photon Source

Far-field mode Profile:



PL from single InGaN QDs

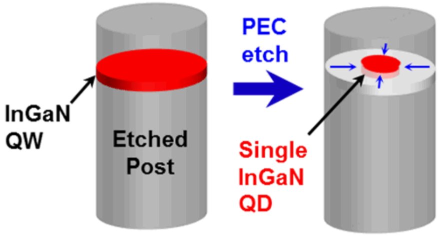
Capped InGaN QW



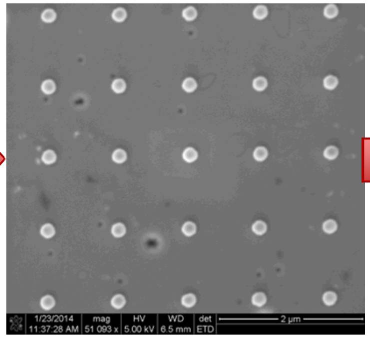
- Posts (150 – 200 nm) patterned with e-beam lithography
- Thicker GaN capping layer (~ 30 nm)
- Narrow PL emission (<1 nm FWHM) observed
- Better ratio of single QD mission to background
- May be pumping more than one post (2 micron pitch)

Submicron Post Samples

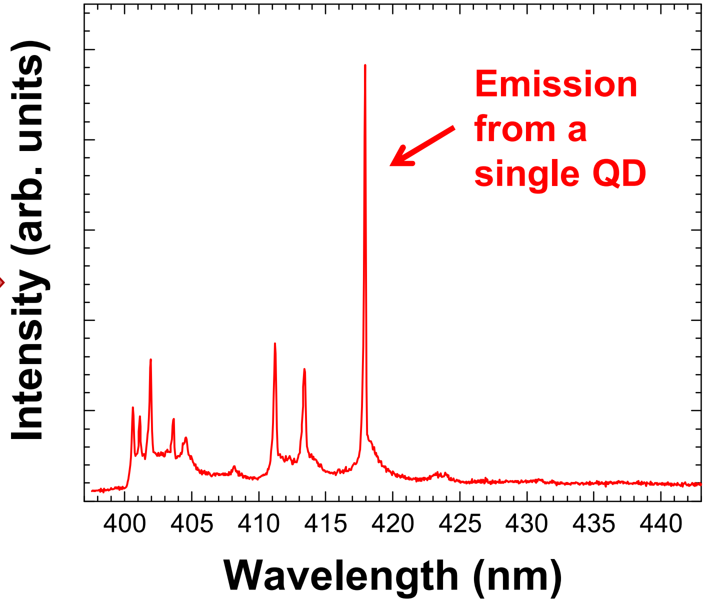
- Intermediate test structure
- Test PEC etching,, demonstrate single QDs.



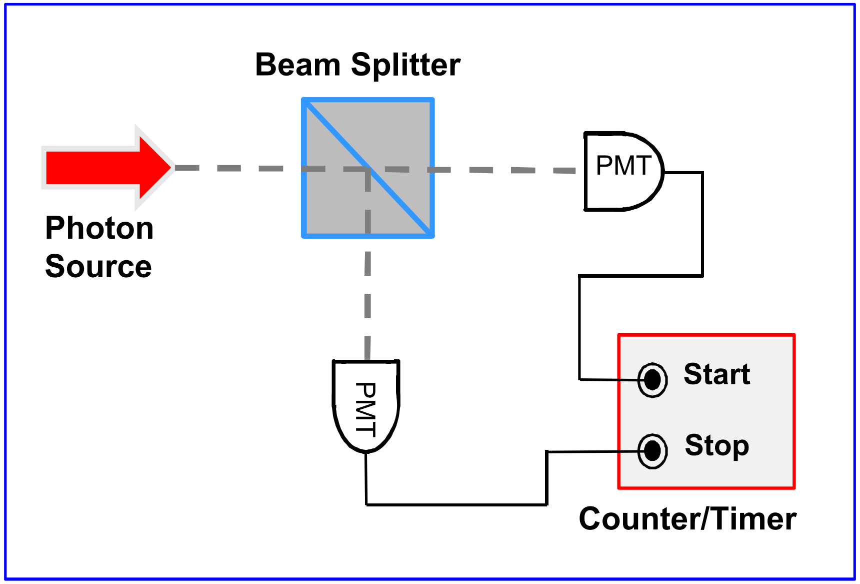
SEM image of posts (top view)



InGaN QD emission:



Single Photon Measurement: HBT experiment

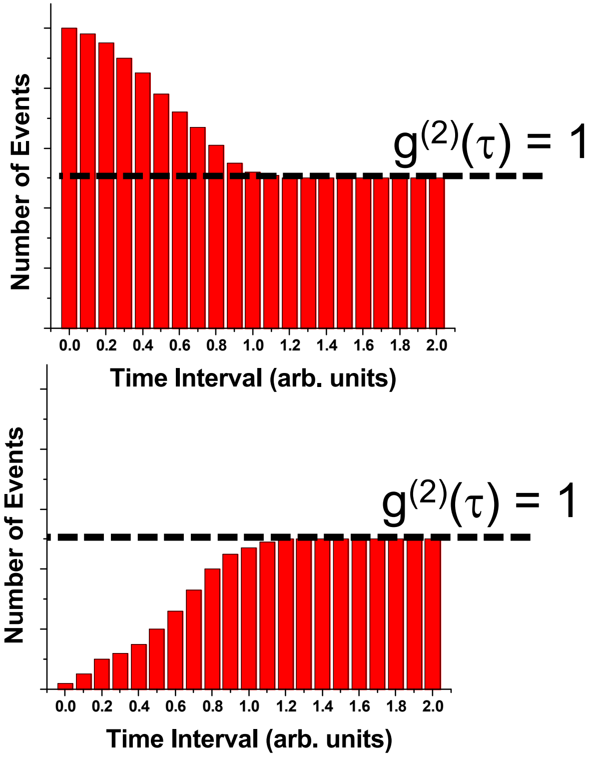


Thermal Source

$g^{(2)}(0) > 1$

Quantum Source

$g^{(2)}(0) < 1$

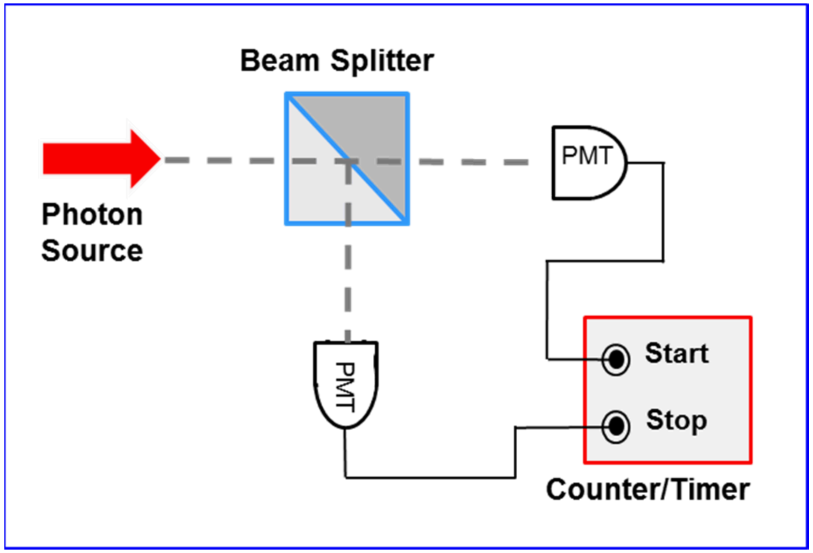


- Non-polarizing beamsplitter to split intensity between detectors
- Single photon counting required
  - Use PMTs or avalanche photodiodes
  - Single photon counting modules
- Measure correlation between detected photons
- Detectors: low QE, speed, and dark counts

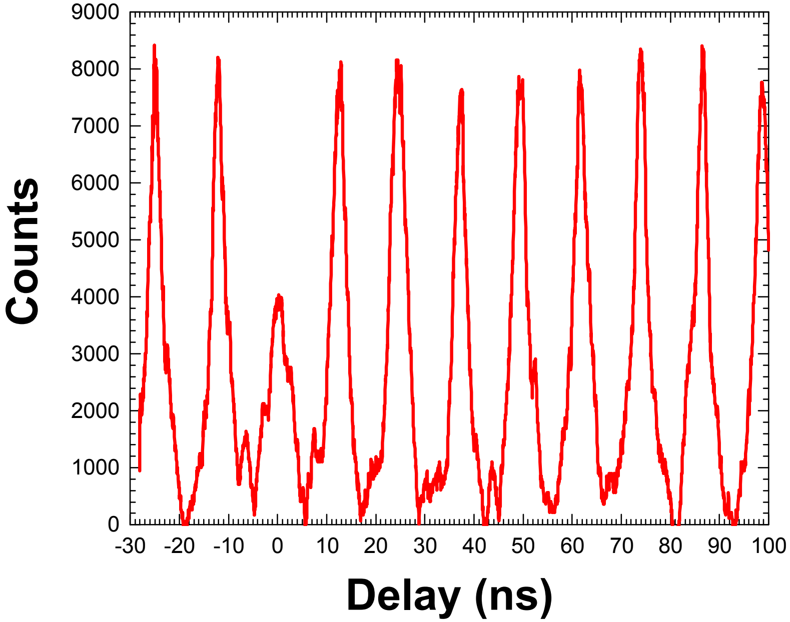
Single Photon Measurement: HBT experiment

- Set up a Hanbury-Brown and Twiss measurement
- Demonstrated a g2 ~ 0.5 for PEC-etched InGaN QDs
  - Signal-to-noise ratio could be better
  - Expect improvements with recent changes to g2 set-up
- Improved light collection will help measurement

HBT Experimental Set-up

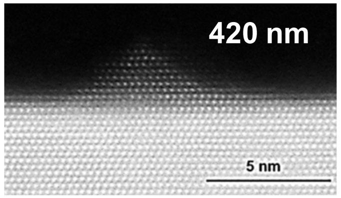


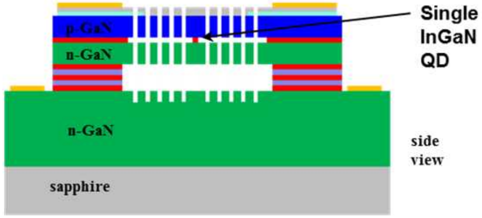
Pulsed g2 data



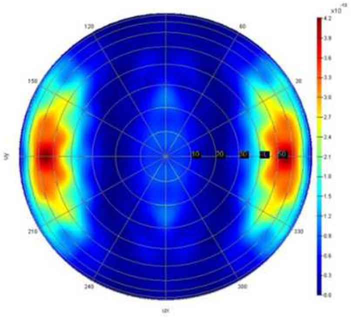
Summary/Conclusions

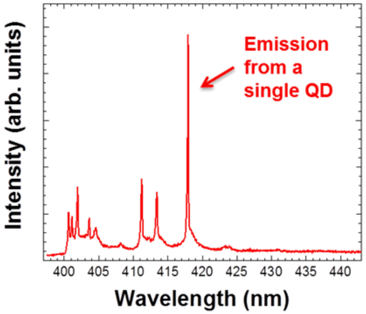
- **Quantum size controlled PEC etching**
  - TEM EDX mapping shows we have InGaN QDs
  - Quantum wells are etched to make QDs
  - QD size and emission λ determined by PEC wavelength
- **Design for InGaN QD single photon source**
  - Deterministic placement of a single InGaN QD in a photonic crystal cavity
  - FDTD modeling: cavity resonances and far field patterns
- **Emission from single InGaN QDs in post samples**
  - Narrow linewidth emission (<0.3 nm)
  - Pulsed g2 measurement showing g2 ~ 0.5





InGaN QD Single Photon Source



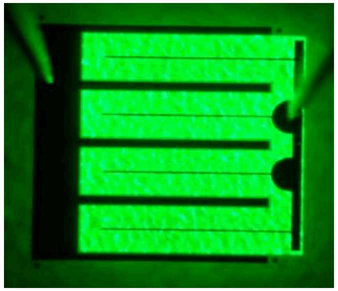


Extra slides →

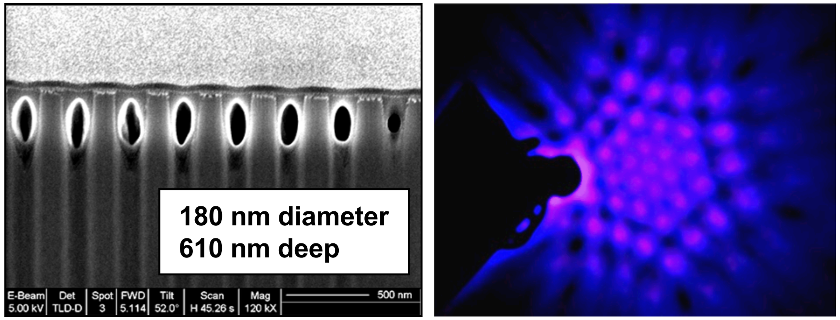
SNL III-Nitride Capabilities

- Leverage Work on Previous III-nitride Emitters
  - EFRC, Previous LDRDs, multiple EERE programs
- Three MOCVD reactors growing III-nitride materials
- Previous work in III-nitride photonic crystal structures

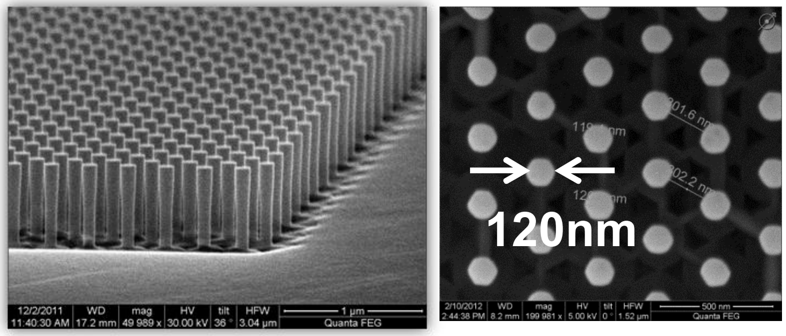
Green InGaN LEDs



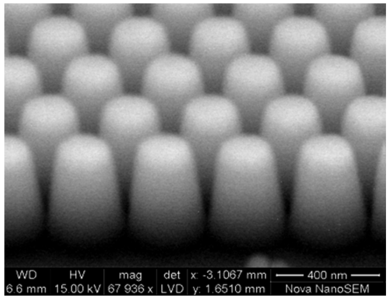
InGaN photonic crystal LEDs



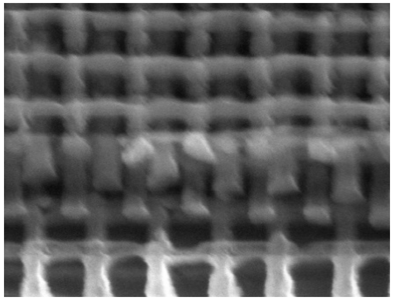
GaN nanowire arrays



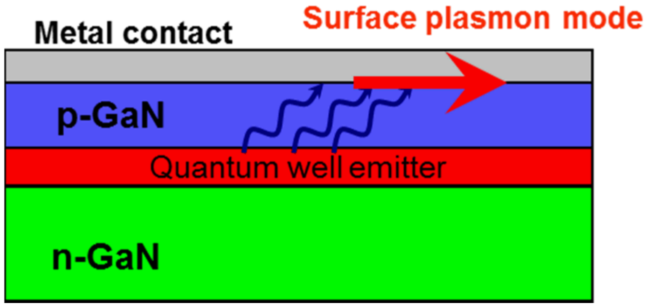
InGaN nano-LED array



GaN 3D logpile PhC



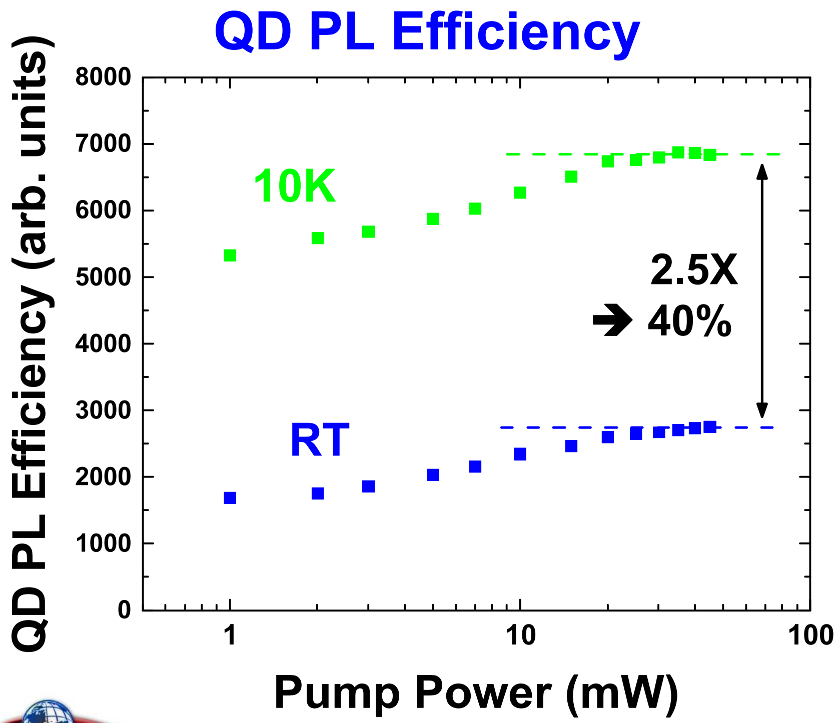
Surface plasmon enhanced LEDs

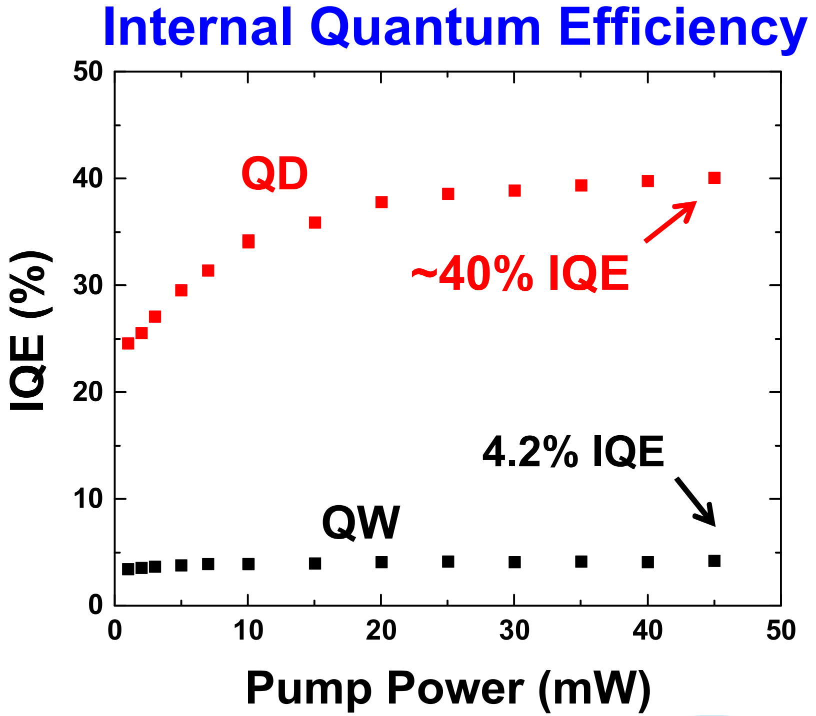


InGaN QD internal quantum efficiency

Capped InGaN QW

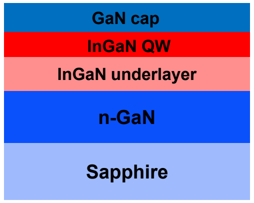
- Compare 10K and RT PL efficiency
- Assumes 10K PL is 100% efficient
- PL Intensity drops by >100X after QD etching
- IQE goes up by almost 10X after QD etching
- QDs are expected to have better IQE

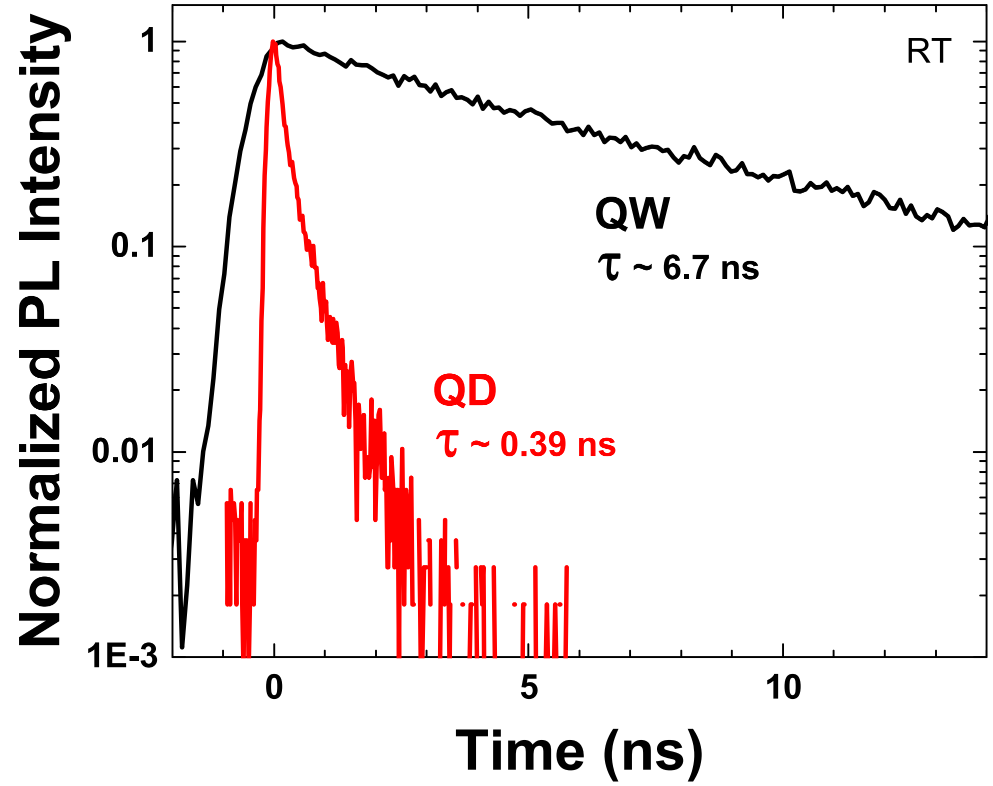




Time-resolved PL data from InGaN QDs

Capped InGaN QW



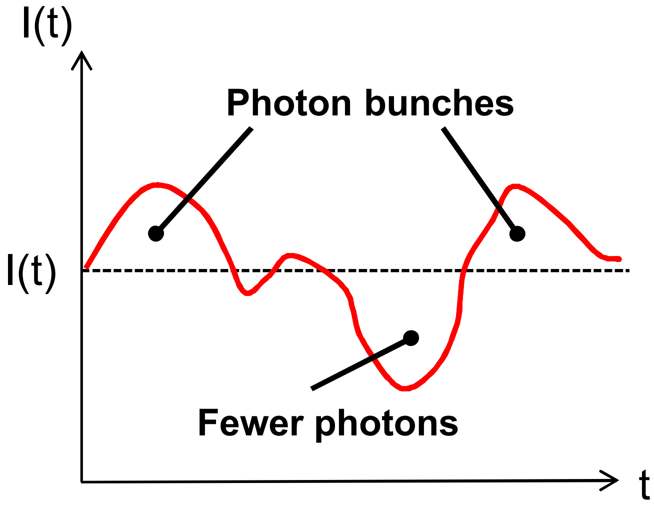


TRPL data:

- 405 nm pump (~ 2 ps, pulsed)
- Resonant pumping into InGaN
- Room temperature TRPL data
- Hamamatsu streak camera data
- 17X change in PL lifetime
- Lifetime is expected to be much shorter for QDs
- Shows that we have fundamentally changed the InGaN material
- QW → QDs

Single Photons for Weak absorption Measurements

- **Perfectly coherent light source has shot noise**
  - Poissonian photon statistics (random)
- **Single photon source can beat the photon shot limit**
  - Sub-Poissonian photon statistics (anti-bunched)
  - Regular emission of single photons is very low noise
- **Single photon sources deliver amplitude squeezed light**
  - Uncertainly relationship between amplitude and phase
  - Field amplitude if precisely known, but phase is unknown
- **Low noise source allows very precise absorption measurement**
  - For large transmissions and good detectors



Adapted from “Quantum Optics: An Introduction” by Mark Fox

$$St.Dev(Coherent) = \sqrt{xMT}$$

$$St.Dev(Single\ photon) = \sqrt{xMT(1-xT)}$$

M = number of incident photons
T = transmission coefficient
x = detector QE

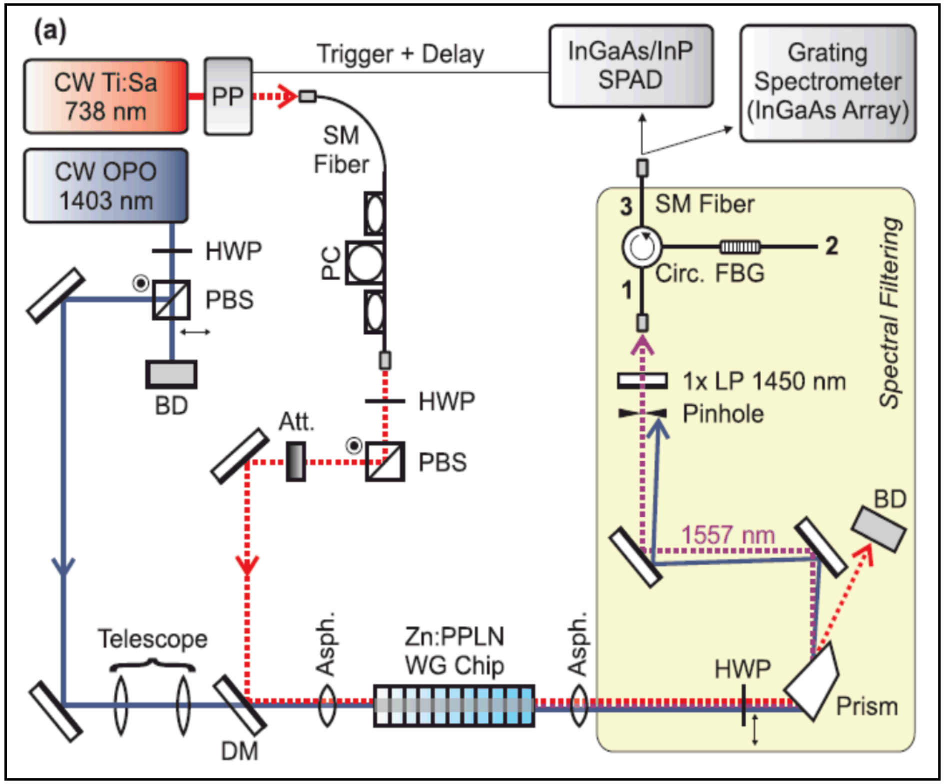
$$\boxed{\frac{Var\ T\ (SP)}{Var\ T\ (C)} = 1 - 2x\frac{T}{1+T}}$$

Analysis from B. Lounis et al., “Single Photon Sources,” Rep. Prog. Phys. **68,** 1129 (2005).

Frequency Conversion: 480 nm to 1.5 um

Quantum Frequency Conversion:

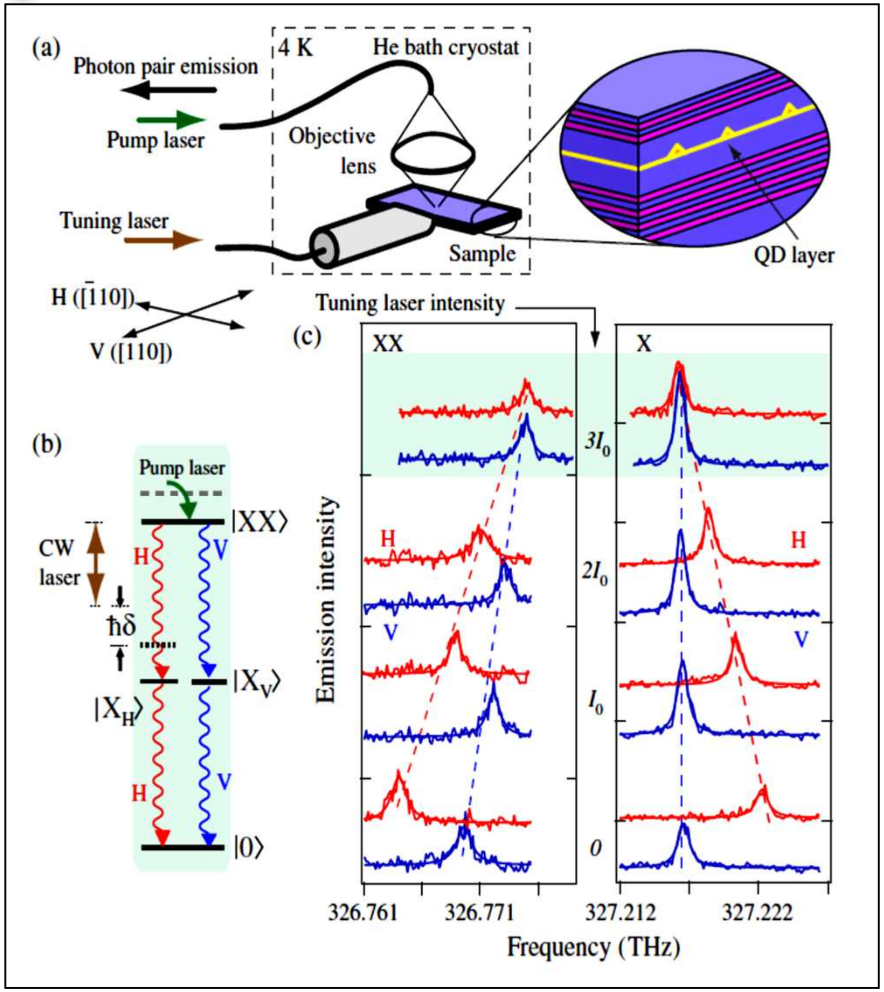
738 nm to 1557 nm
73% Quantum Efficiency



S. Zaske et al., Optics Express **19,** 12825 (2011)

- **Quantum frequency conversion is possible**
- 73% efficiency demonstrated
- Single photon statistics are preserved
- Conversion is desired since single photon detectors work best in the visible.
- Use periodically-poled lithium niobate
- Requires a high power pump laser
- QD-based SPS
  - 64% Quantum Efficiency
  - 711 nm → 1313 nm
  - S. Zaske et al., PRL **109,** 147404 (2012).
- Frequency conversion could be incorporated on chip

QD entangled photons: Biexciton cascade



A. Muller et al., PRL **103,** 217402 (2009)

- **Biexciton cascade: two bound excitons decay radiatively**
- Can create a polarization entangled photon pair
- Exciton fine structure can destroy entanglement
- Use external perturbation to align levels
  - Applied Electric field
  - Applied Stress
  - Incident CW Laser
  - Strong coupling- Cavity QED
- Only demonstrated for InAs QDs at low temperature
- **Should be possible for GaN (RT?)**
- **Potential for chip-based entangled photons**