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InGaN Quantum Dots for Room Temperature Single Photon Sources

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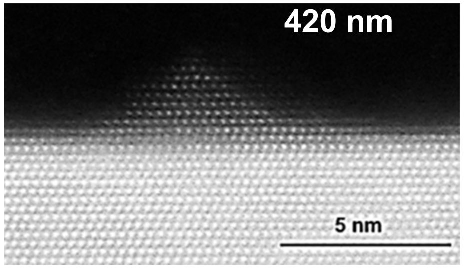
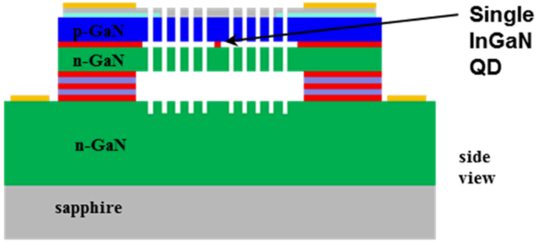
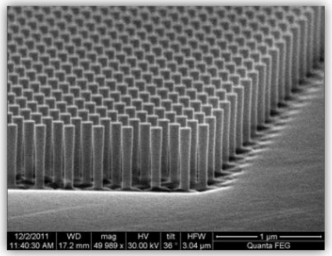
Outline:

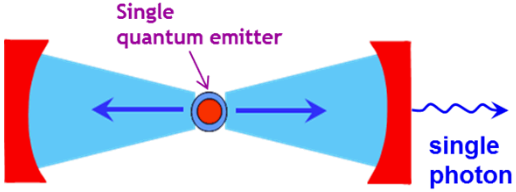
1. **Motivation: QD single photon sources**
2. **InGaN QD fabrication**
   - Quantum size controlled PEC etching
3. **AFM/TEM analysis of InGaN QDs**
4. **Optical characterization of QDs**
   - Photoluminescence, TRPL
5. **Deterministic placement of InGaN QDs**
   - Emission from single InGaN QDs
   - Photonic crystal based single photon source
6. **Calculated g2 for multi-dot systems**
   - Studying the transition from classical to quantum
7. **Summary/Conclusions**



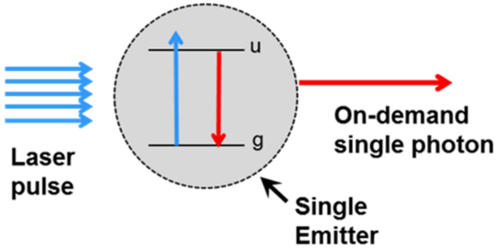


InGaN QD Single Photon Source

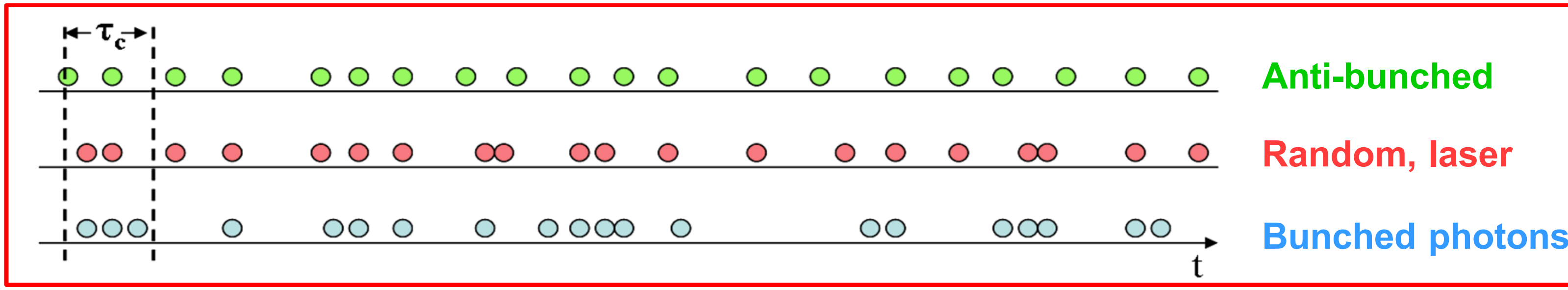




Single Quantum Emitter



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

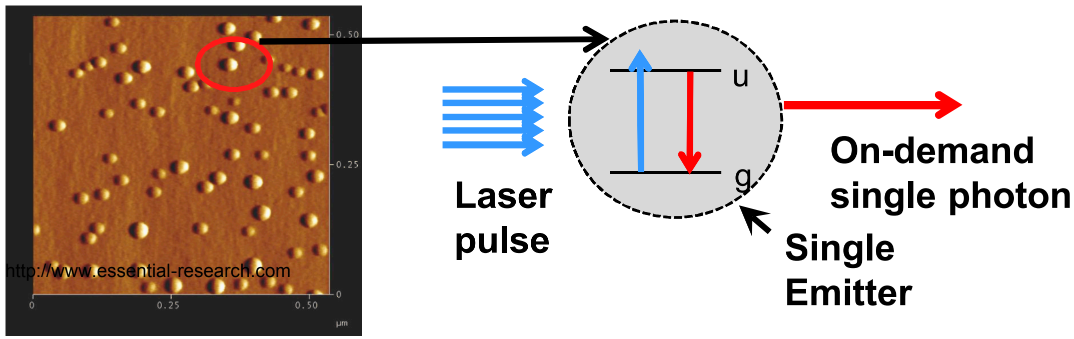
- Anti-bunched photons (Quantum optics theoretical treatment)

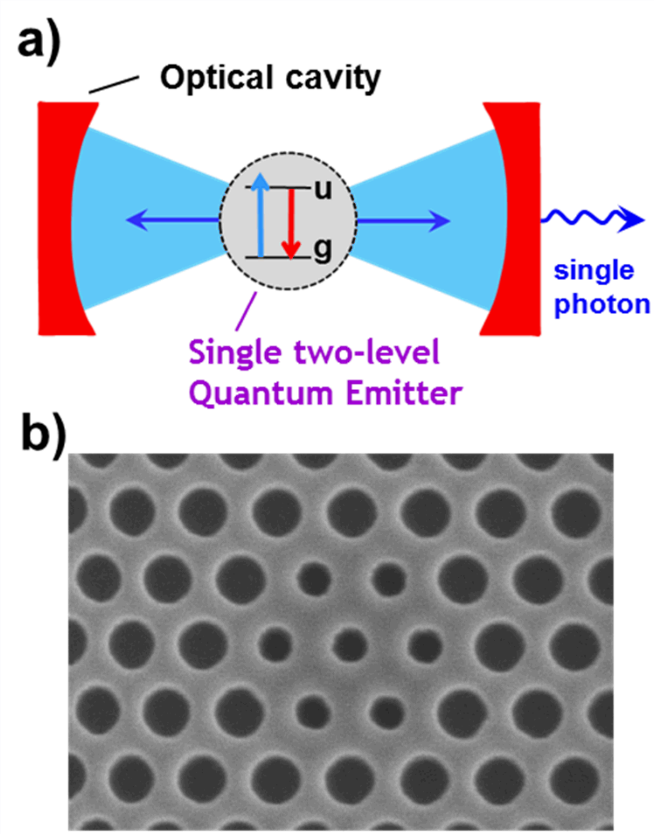
Applications:

- **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 Source based on Quantum Dots

AFM image of InAs QDs



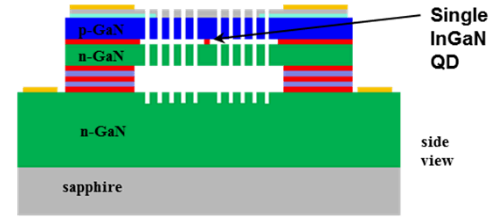


QD Single Photon Source:

- 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**

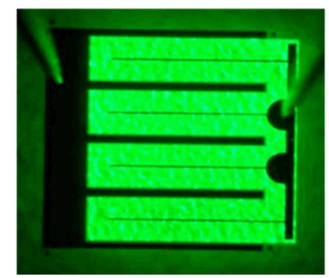
InGaN Quantum Dot SPS Development

- ***Goal:*** Room temperature, electrically-injected, chip-scale single photon source
- Our approach:
  - Develop a III-nitride QD capability at Sandia
  - **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**
  - **Deterministic placement of InGaN QDs**
  - Leverage previous work in III-nitride optoelectronics
  - Leverage SNL PhC design, fabrication expertise

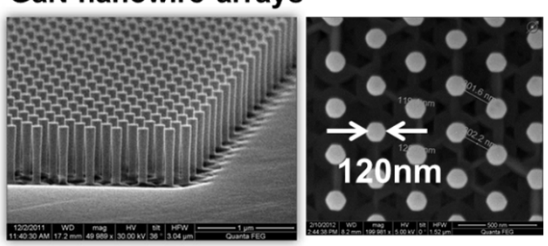


InGaN QD Single Photon Source

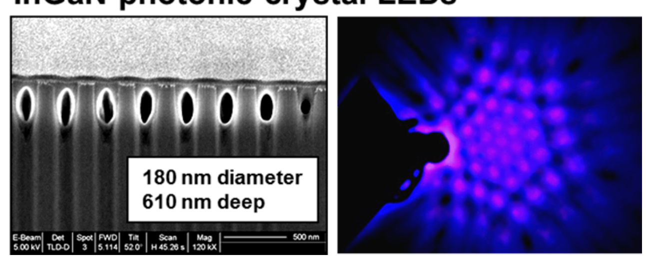
Green InGaN LEDs



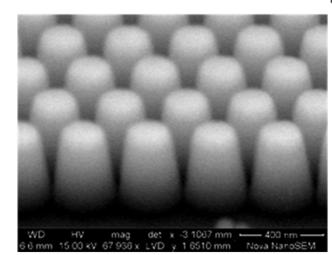
GaN nanowire arrays



InGaN photonic crystal LEDs



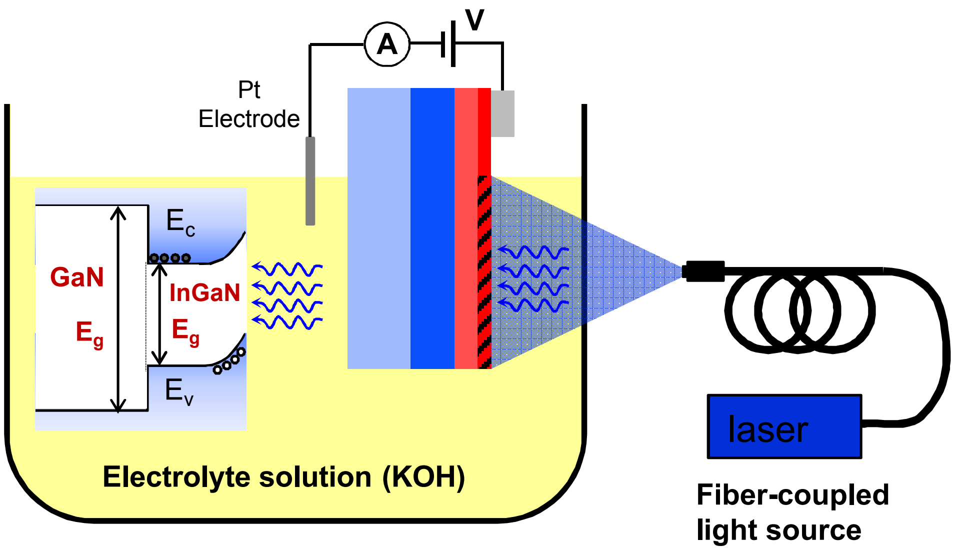
InGaN nano-LED array



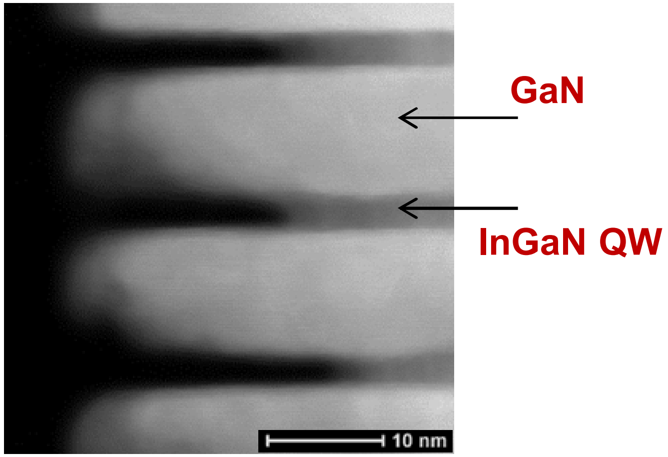
Introduction to InGaN PEC Etching

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



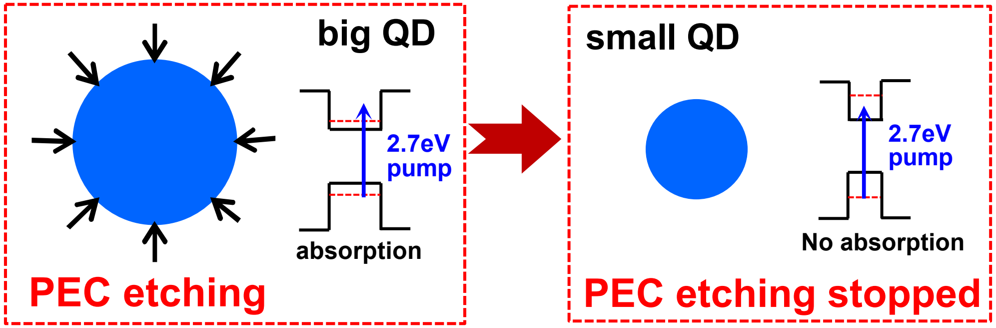
PEC etched InGaN/GaN QWS

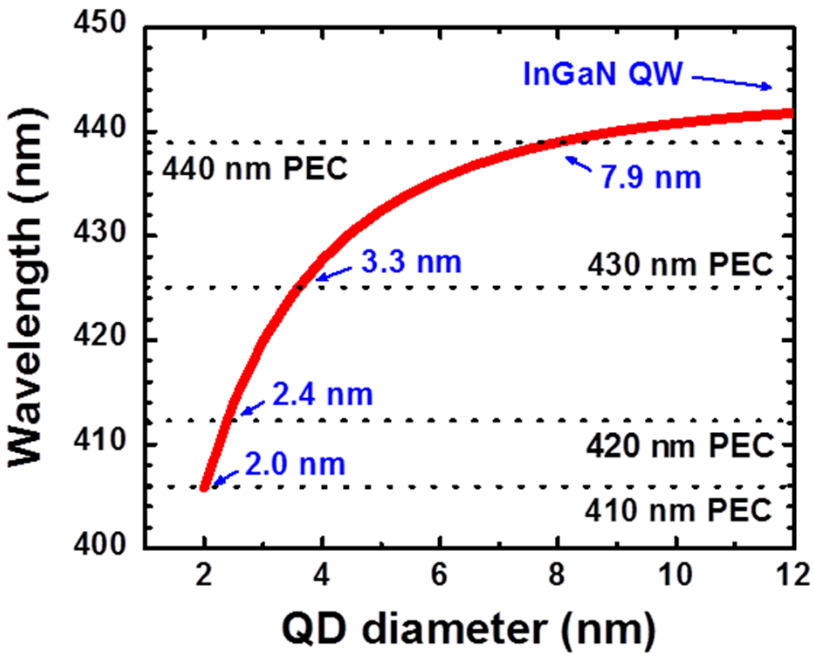


Fabrication of InGaN QDs via PEC etching

Quantum Size Control: Use size quantization to control QD size

Self-limiting PEC etch process:



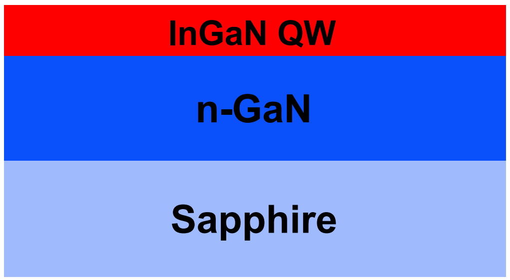


G. Pellegrini, et al., Journal of Applied Physics 97, 073706 (2005).

- 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 ??**

MOCVD-grown InGaN samples

Uncapped singe InGaN QW



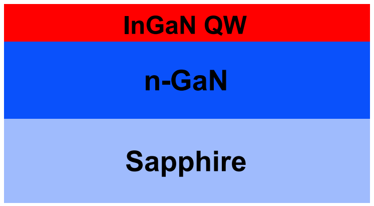
- Grown by MOCVD on sapphire substrates
  - 5 - 20 nm thick InGaN layer
- No InGaN underlayer is used in this sample
- Uncapped single QW sample (14% In):
  - Amenable to surface characterization of QDs
  - TEM, AFM characterization
  - Luminescence weaker than capped sample

Capped singe 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

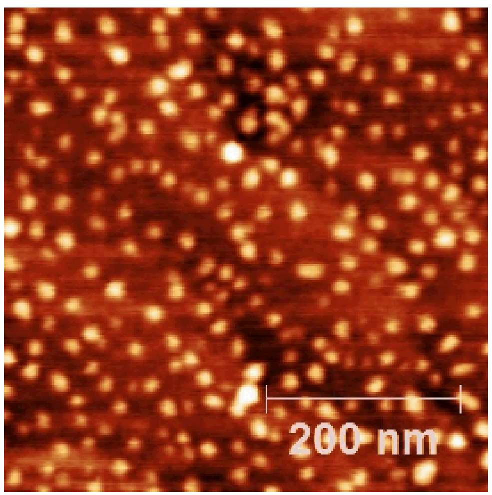
Atomic Force Microscope (AFM) Measurements

Uncapped InGaN QW

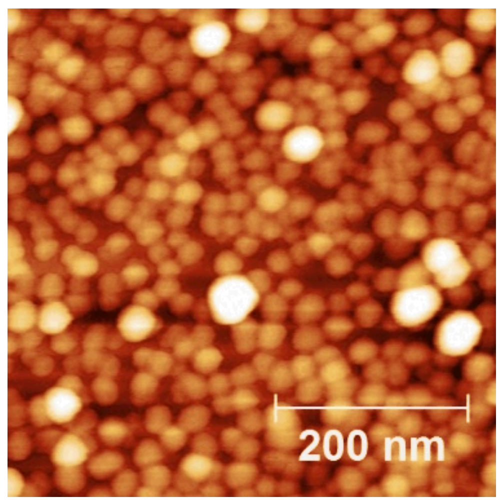


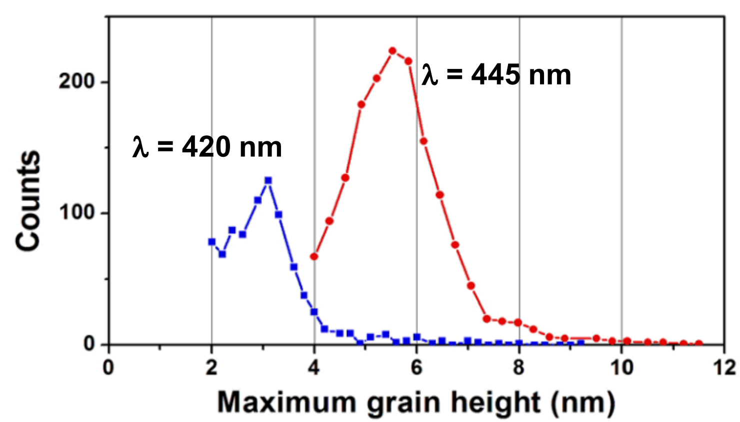
- Samples etched for two hours at 420 nm and 445 nm
  - Laser power density: ~ 3 mW/cm^2
- High dot density: 10^{11}/cm^2
- Some big dots (10-20 nm) remain: due to dislocations?
- QD size depends on PEC etch wavelength

PEC etch→ λ = 420 nm



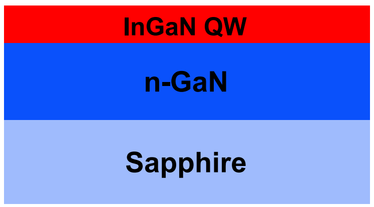
PEC etch → λ = 445 nm



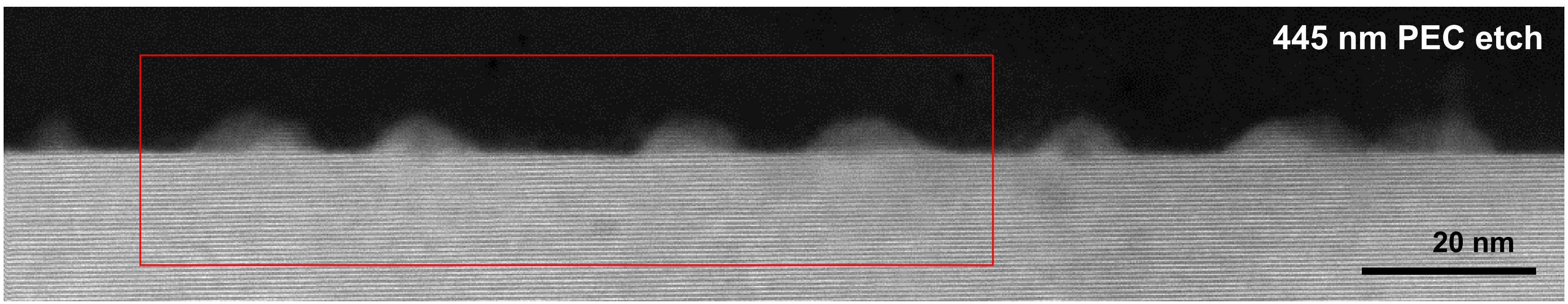


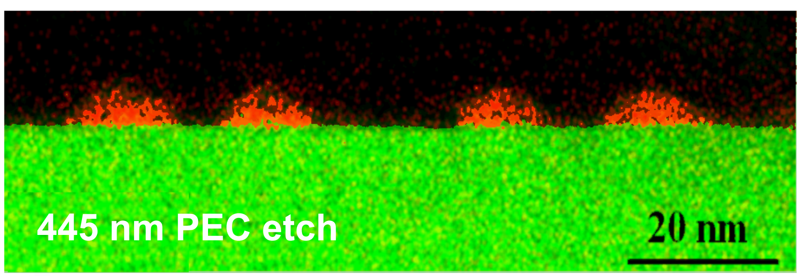
Transmission Electron Microscope (TEM) Images

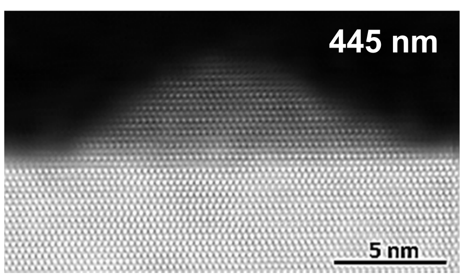
Uncapped InGaN QW

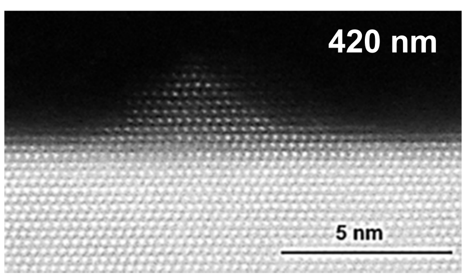


- High-angle annular dark-field (HAADF) TEM images
- Samples etched at 420 nm and 445 nm
- Energy dispersive x-ray mapping
  - QDs on surface are InGaN
  - Red = indium, green=gallium
- InGaN QDs are epitaxial to the underlying GaN
- No underlayer, no cap → PL is not very bright



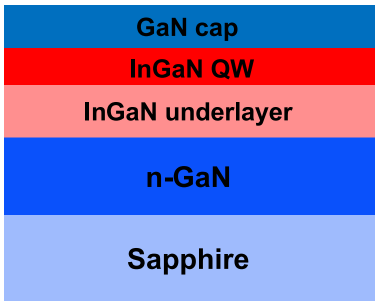




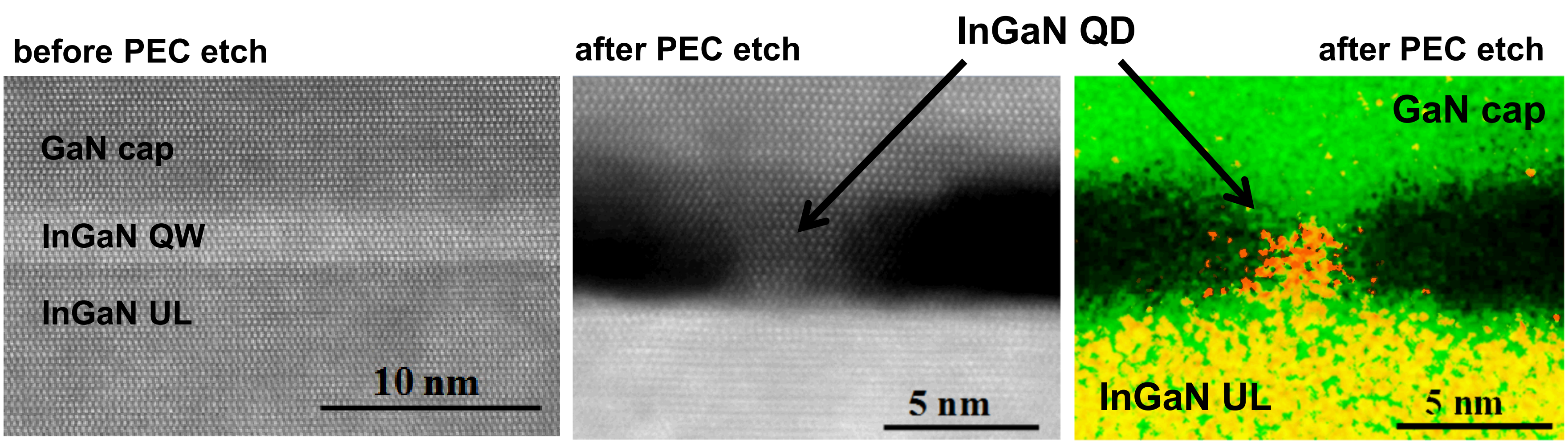


Transmission Electron Microscope Images

Capped InGaN QW

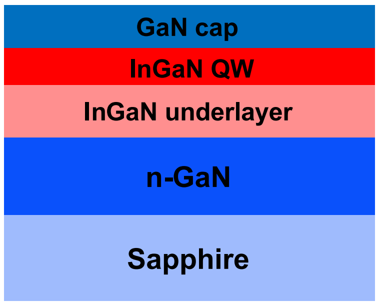


- High-angle annular dark-field (HAADF) TEM images
- Sample etched at 420 nm
- EDX mapping shows that dots are InGaN
- InGaN QDs are epitaxial to the underlying GaN
- 2% InGaN underlayer + GaN cap → PL is much brighter
- GaN cap provides partial passivation

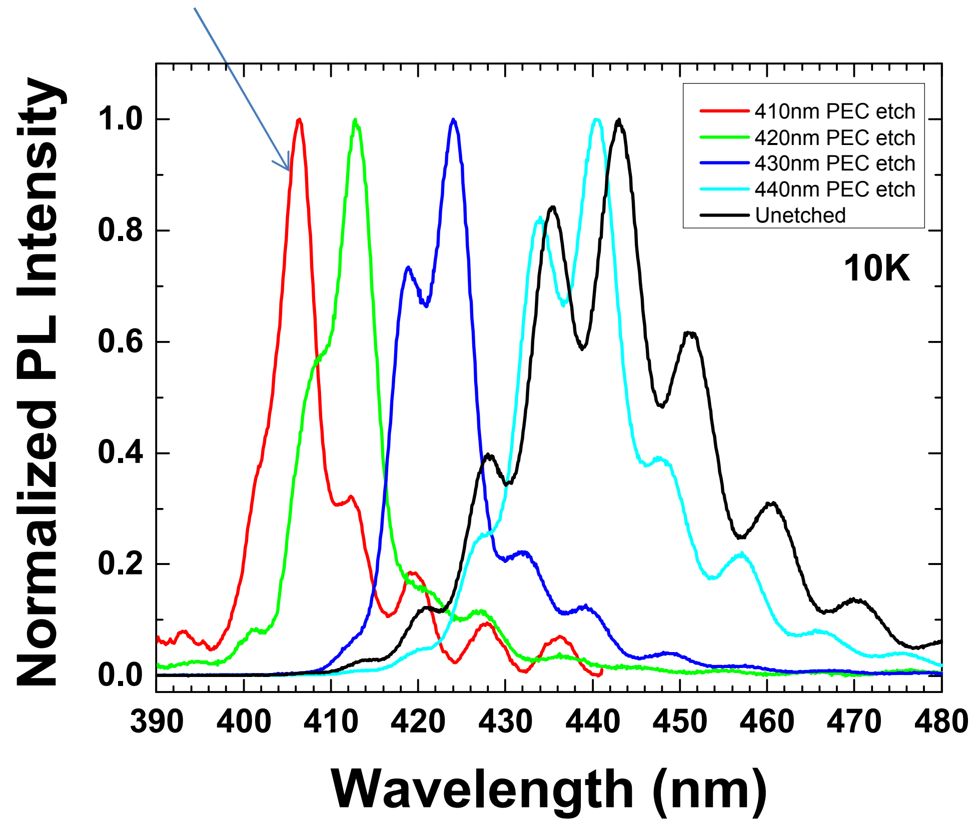


Photoluminescence from fabricated InGaN QDs

Capped InGaN QW



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



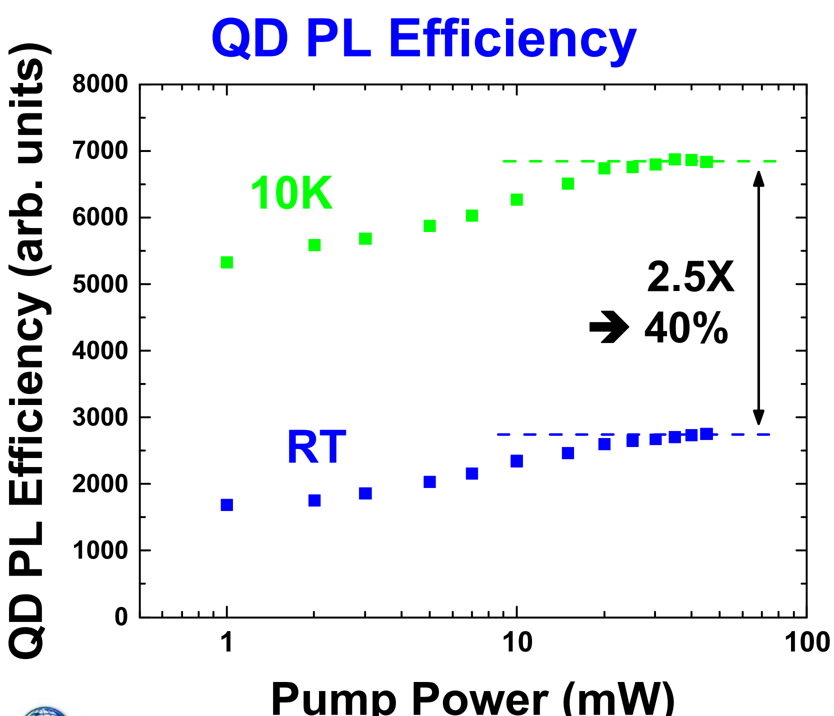
Photoluminescence (PL) data:

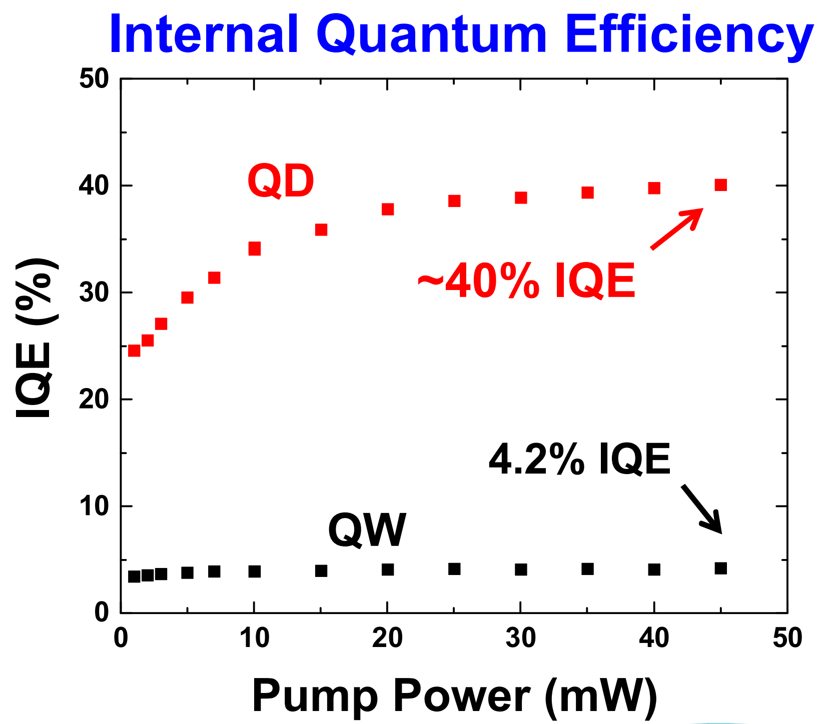
- **375 nm pump (ps pulsed)**
- **10K PL data**
- **PL wavelength determined by PEC etch wavelength**
- **PL linewidth: 24 nm → 6 nm**
- **Quantum size-controlled PEC etching works!**

InGaN QD internal quantum efficiency

Capped InGaN QW

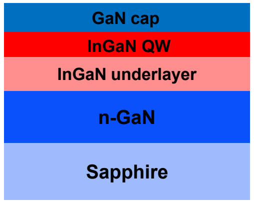
- Compare 10K and RT PL efficiency
- Assumes 10K PL is 100% efficient
- PL Intensity drops by >100X after QD etching
- IQE goes up by almost 10X after QD etching
- QDs are expected to have better IQE

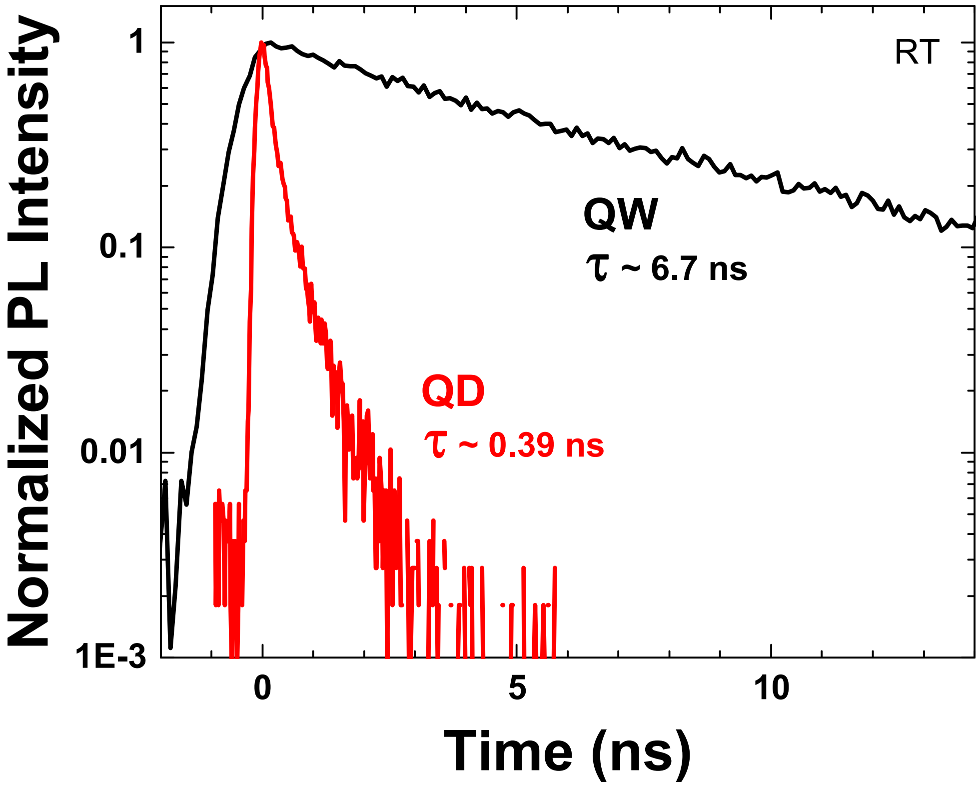




Time-resolved PL data from InGaN QDs

Capped InGaN QW





TRPL data:

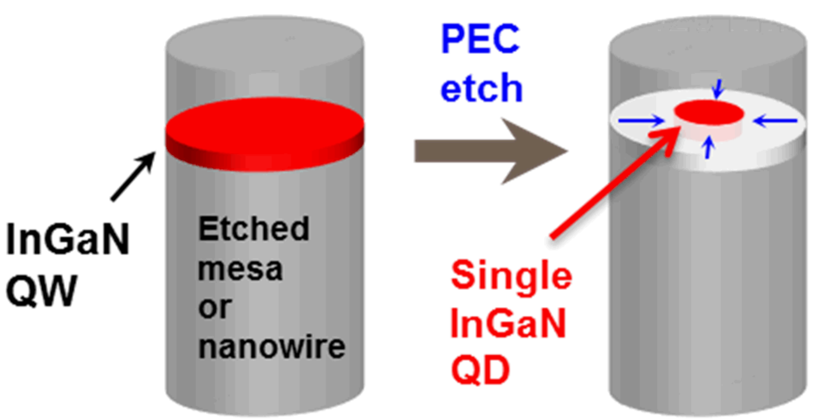
- 405 nm pump (~ 2 ps, pulsed)
- Resonant pumping into InGaN
- Room temperature TRPL data
- Hamamatsu streak camera data
- 17X change in PL lifetime
- Lifetime is expected to be much shorter for QDs
- Shows that we have fundamentally changed the InGaN material
- QW → QDs

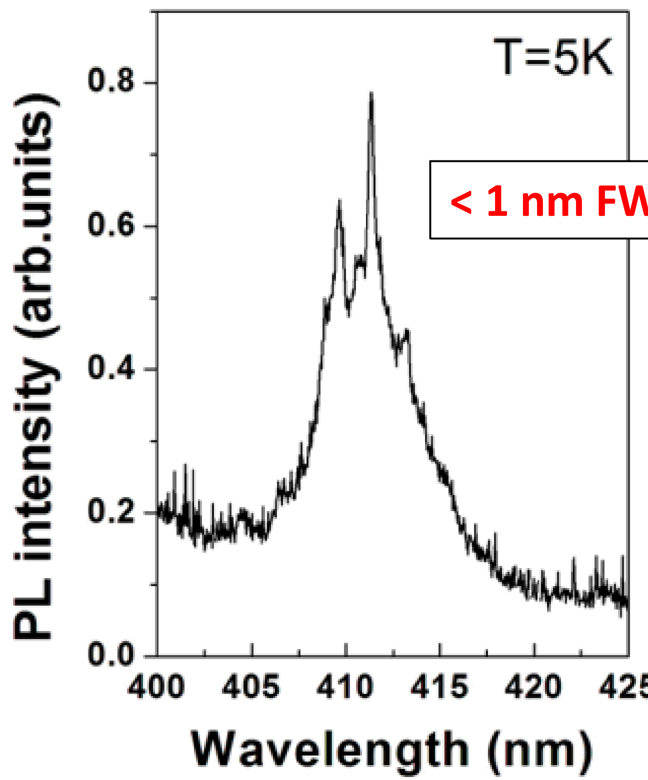
Emission from single InGaN QDs

Capped InGaN QW

- Posts (150 – 200 nm) patterned with e-beam lithography
- Narrow PL emission (<1 nm FWHM) observed
- Fabricate InGaN QDs at deterministic locations
- InGaN QD single photon source

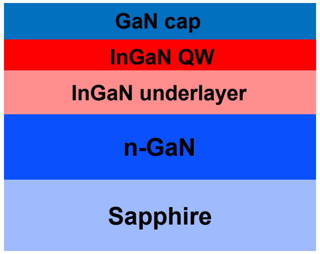
Fabrication of single InGaN QDs





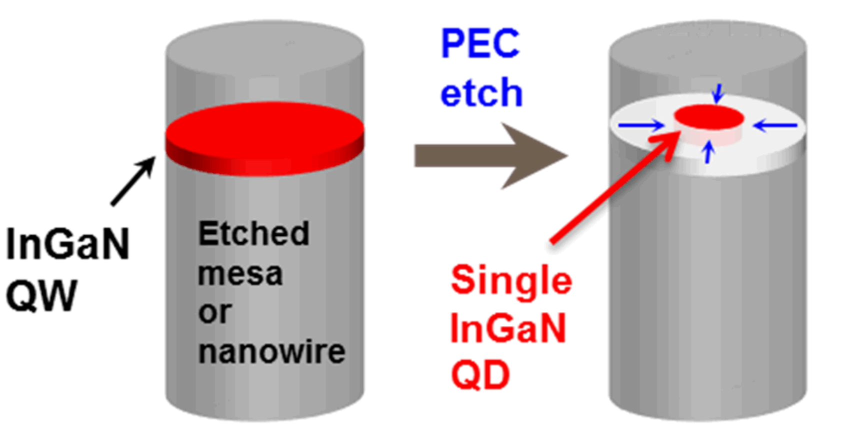
PL from InGaN QDs (newer data)

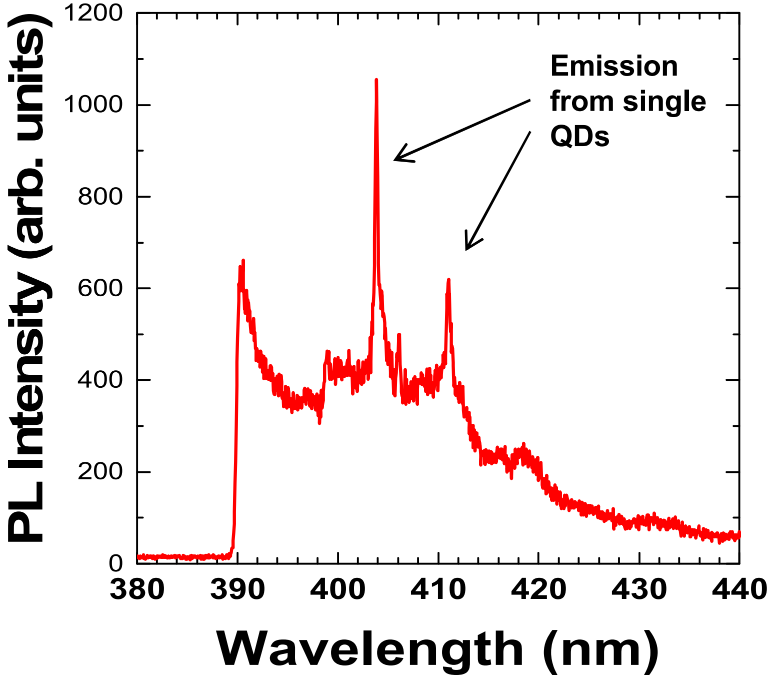
Capped InGaN QW



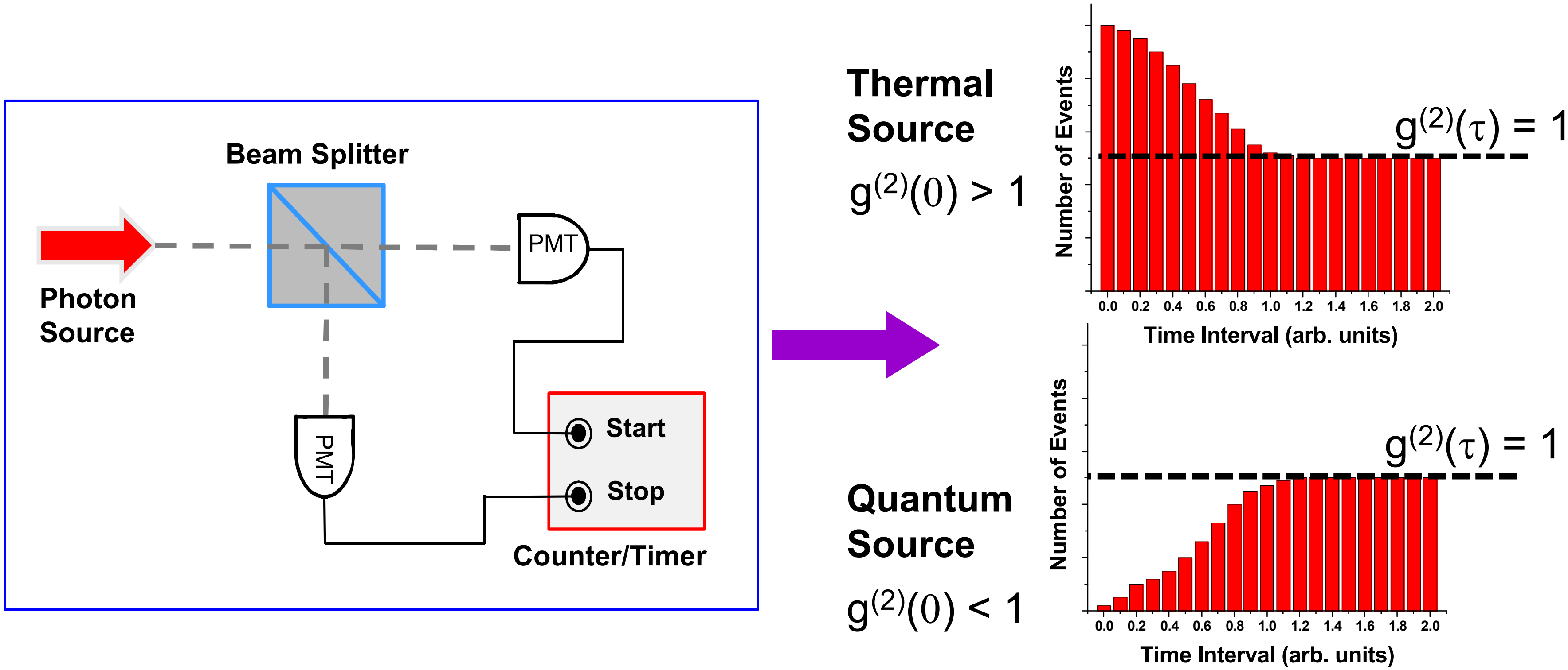
- Posts (150 – 200 nm) patterned with e-beam lithography
- Thicker GaN capping layer (~ 30 nm)
- Narrow PL emission (<1 nm FWHM) observed
- Better ratio of single QD mission to background
- Limited by motion of closed-cycle cryostat

Fabrication of single InGaN QDs





Single Photon Measurement: HBT experiment

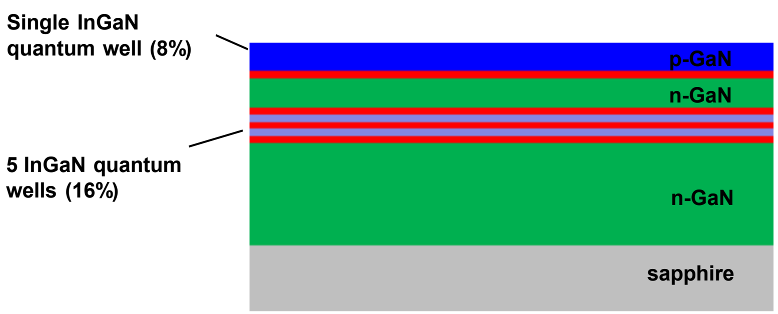


- Non-polarizing beamsplitter to split intensity between detectors
- Single photon counting required
  - Use PMTs or avalanche photodiodes
  - Single photon counting modules
- Measure correlation between detected photons
- Detectors: low QE, speed, and dark counts

Deterministically Positioned InGaN QDs

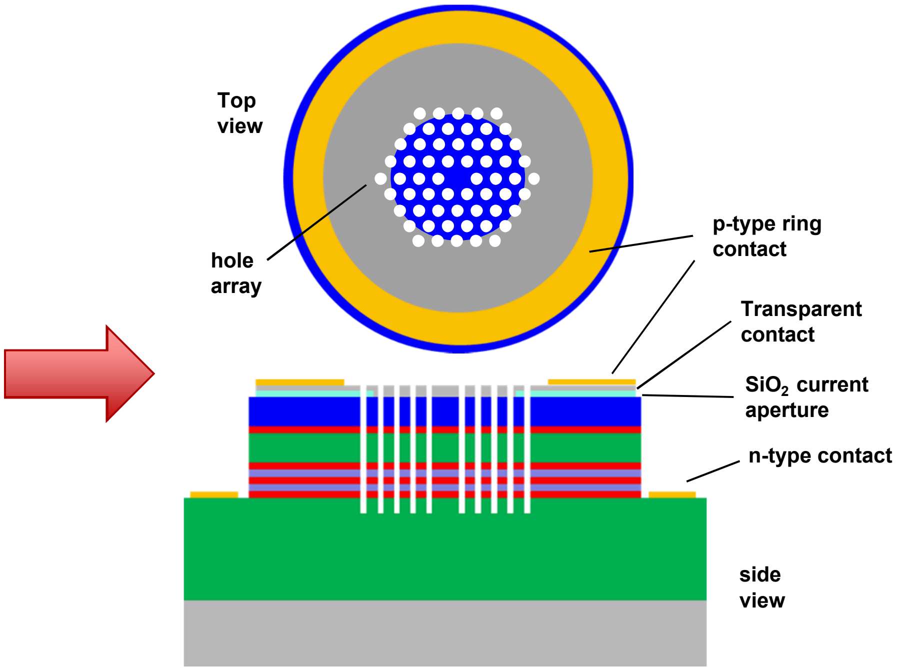
Standard III-nitride device fabrication

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

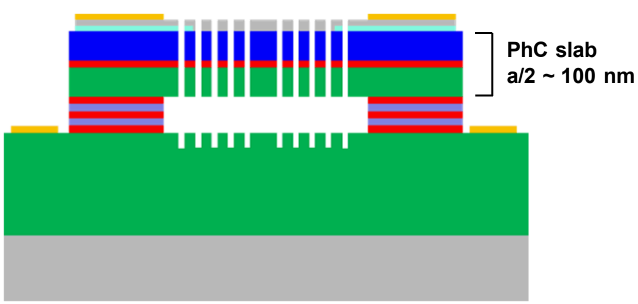


- Standard LED fabrication
- Etch PhC triangular array
- defect cavity at the center
- Requires current aperture
- Use transparent contact

Deterministically Positioned InGaN QDs

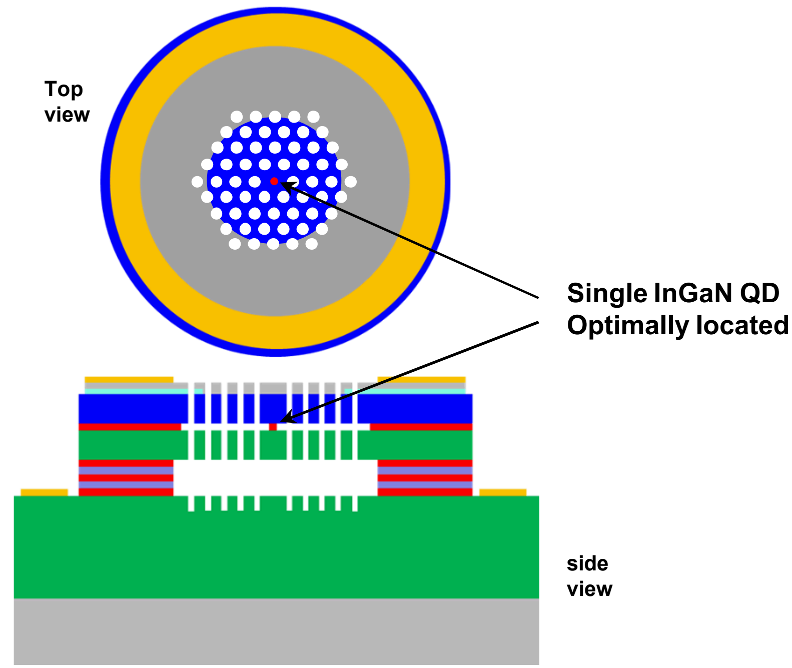
PEC etch of InGaN QWs

3.) First photoelectrochemical etch



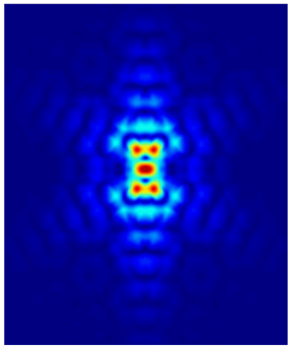
- Use 450 nm laser for PEC etch
- Area under contacts will not etch
- 8% InGaN QW will not etch

4.) Second photoelectrochemical etch

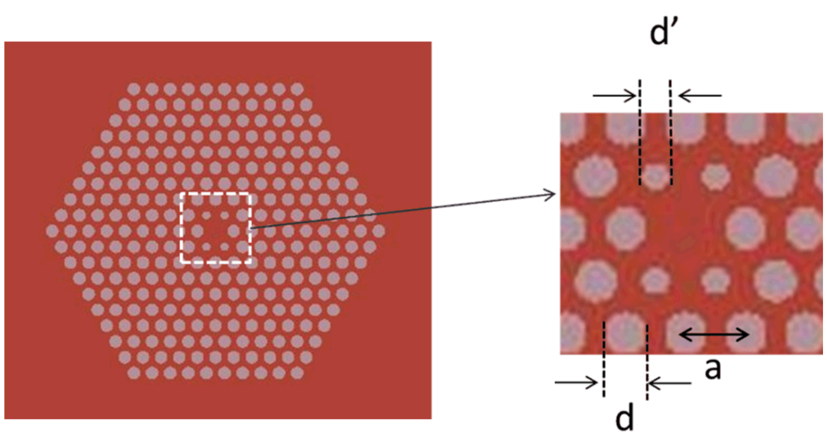


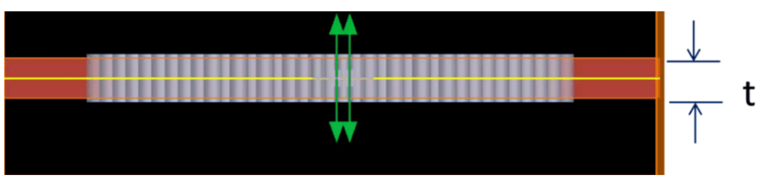
- Use 405 nm laser for PEC etch
- Self limiting PEC etch
- Size quantization in QD will raise energy level above 405 nm

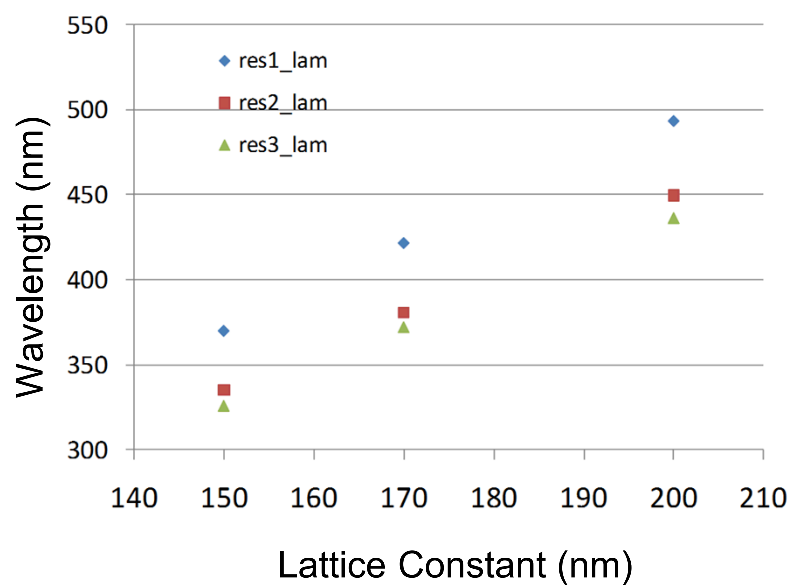
FDTD Photonic Crystal Modeling



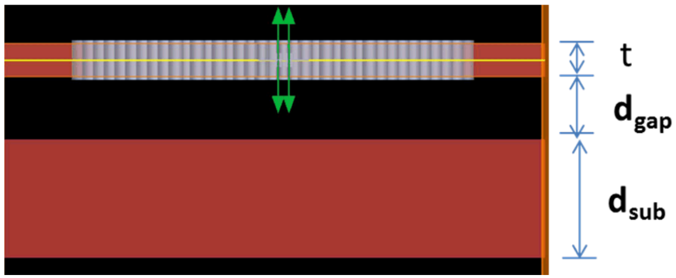
- Calculations done using Lumerical
- Need resonance with anti-node at center (res1)
- Calculated cavity Q is 941 for this design (no substrate)
- Design is not fully optimized (higher Qs are possible)
- Need lattice constant in the range from 170 – 200 nm







FDTD Modeling: Spacer Layer Thickness

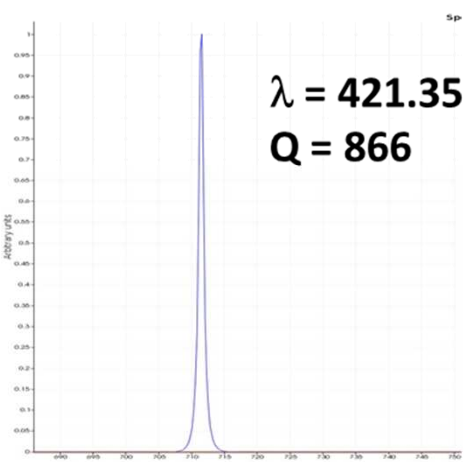


- High-angle annular dark-field (HAADF) TEM
- Sample etched at 420 nm
- EDX mapping shows that dots are InGaN
- InGaN QDs are epitaxial to the underlying
- 2% InGaN underlayer + GaN cap → PL is

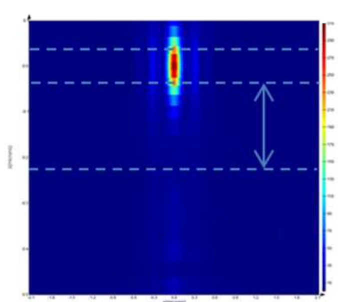
d_{gap} = 0.95a

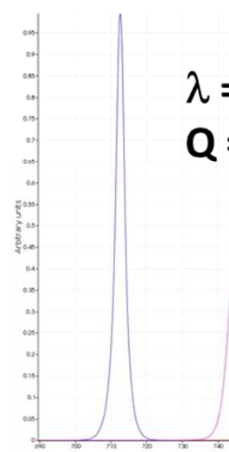
a = 170nm
d = 0.7a
d' = 0.45a
t=0.5a

d_{gap} = 0.6a

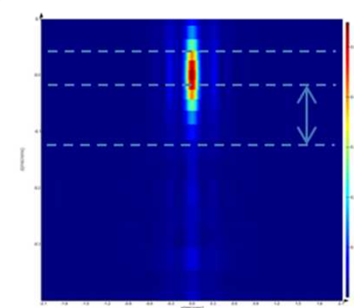


λ = 421.35 nm
Q = 866

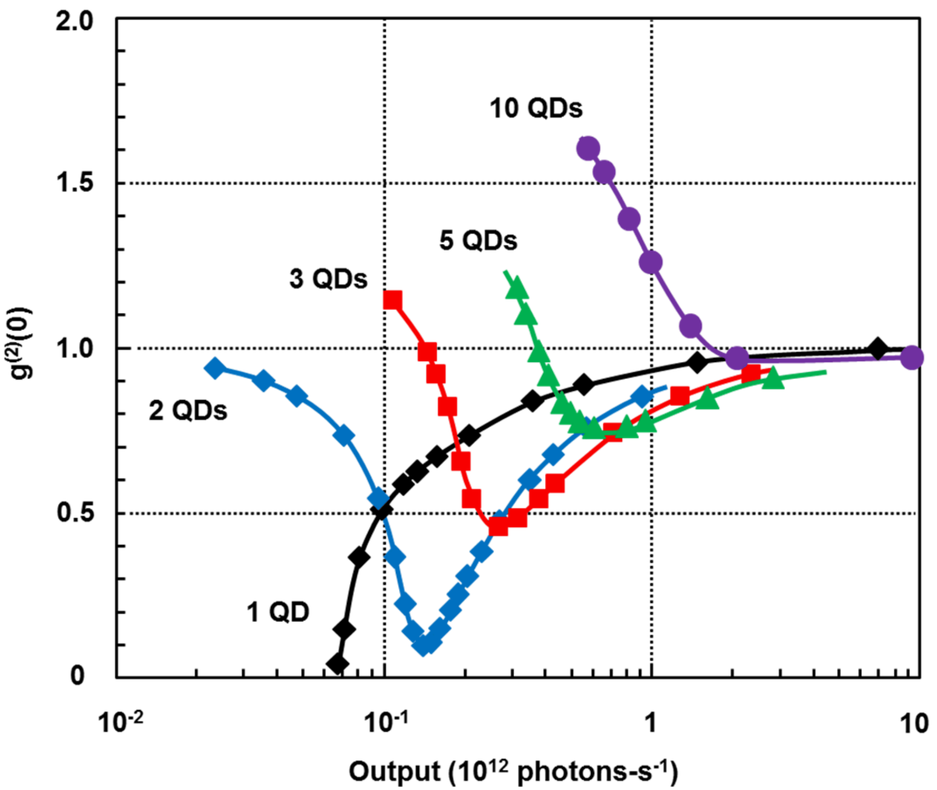




λ = 420.765 nm
Q = 237



Calculated $g^{(2)}(0)$ for systems with multiple QDs

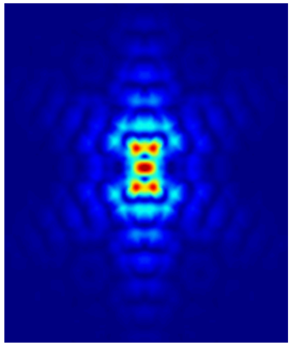


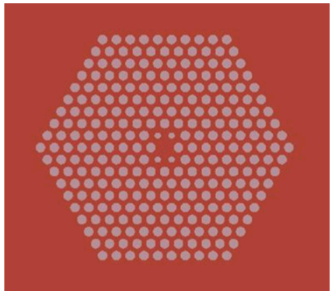
- Study the transition region between classical and quantum light sources
- Calculate g2 using many body theory for a cavity - QD system
- Calculated for InAs QD system, but should be generally applicable
- Vary cavity Q at a constant pump rate
- **Shows that we can get non-classical light for multi-QD systems**
- **Trade-off between low g2 and higher photon output rate.**

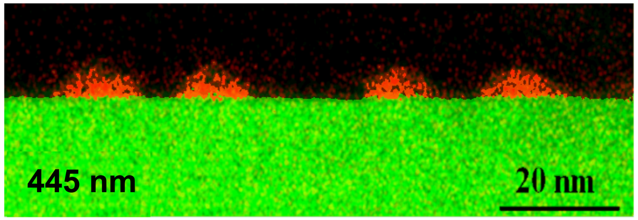
Details on theory: Chow, Jahnke, Gies. To appear in Light: Science and Applications

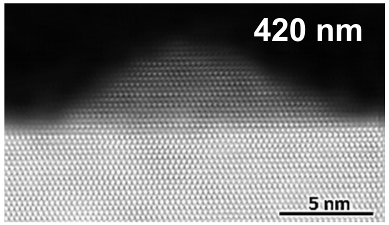
Summary/Conclusions

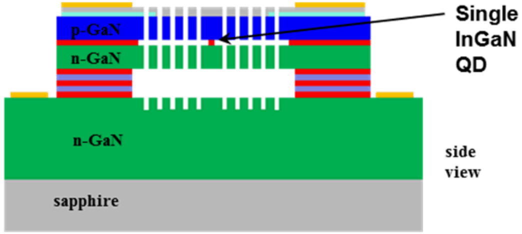
- **Demonstrated InGaN QDs using PEC etching**
  - TEM EDX mapping shows we have InGaN QDs
- **Quantum size-controlled etching of InGaN QDs**
  - QD size and emission λ determined by PEC wavelength
  - 40% IQE for InGaN dots emitting at 435 nm
- **Demonstrated emission from single InGaN QDs**
- **Design for PhC single photon source**
  - Deterministic placement of InGaN QDs
  - FTDT modeling of cavity resonances
- **Calculated g2 for multi-QD systems**
  - Non-classical light from multi-QD systems











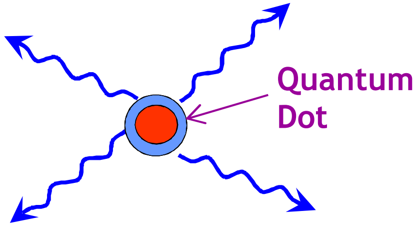
InGaN QD Single Photon Source

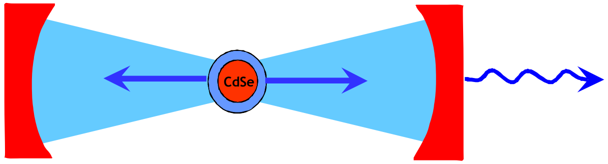
Extra slides →

Single Emitters Inside and Optical Cavity

Why do we need an optical cavity?

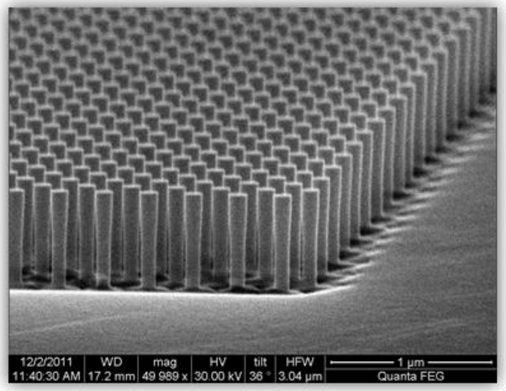
- Improve photon collection efficiency
- Restrict emission to specific spatial modes
- Limit spectral content→ indistinguishable photons
- Generate photons with a specific polarization
- Increase radiative rate, efficiency via Purcell effect
- Theoretically study QD strong coupling physics
  - Coherent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

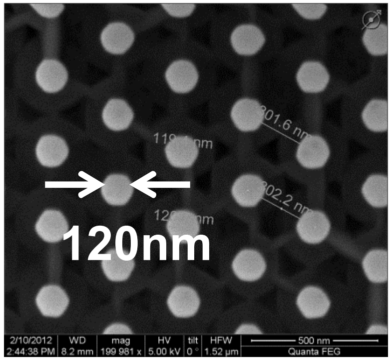


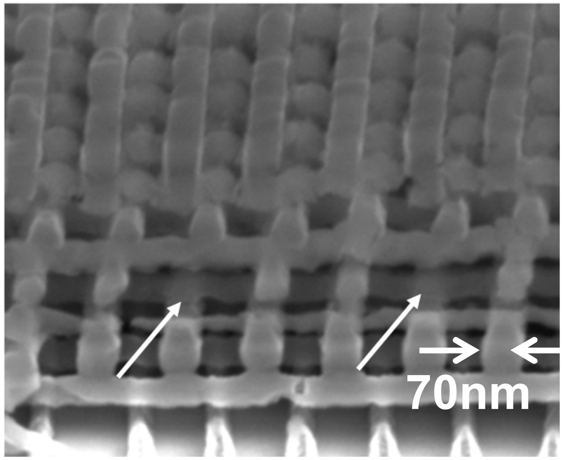


Directional emission

Sandia III-Nitride Nanostructure Fabrication







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
  - $g^{(2)}(0) = 0.16$, 10K, electrically-injected
  - $g^{(2)}(0) = 0.4$, 200K

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

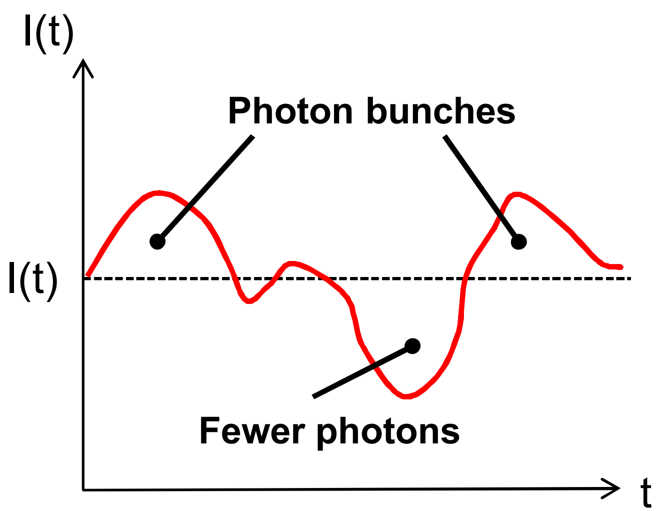
*M.D. Eisaman, et al. Rev. Sci. Instrum. **82,** 071101 (2011)

Ideal Single Photon Source:

- Triggered (on-demand)
- 100% probability of emitting one photon
- 0% probability of emitting multiple photons
- $g^{(2)}(0) = 0$
- Indistinguishable photons
- High repetition rate
- Room temperature
- Electrically injected

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

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

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

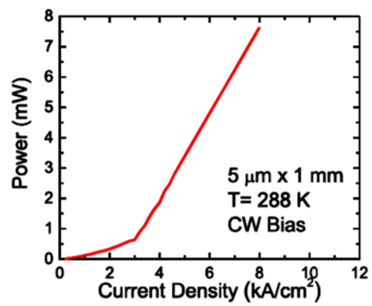
Motivation for InGaN Quantum Dot (QD) Emitters

- **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**

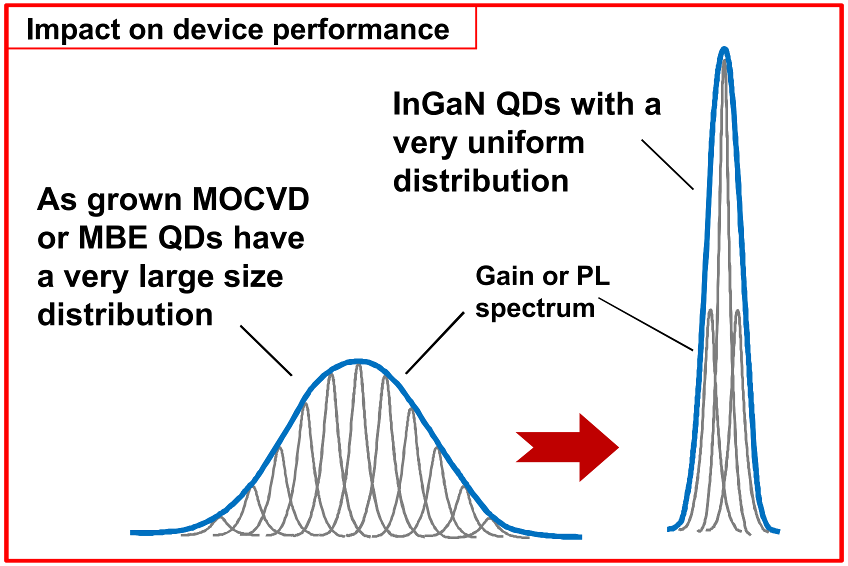
InGaN QD laser:

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

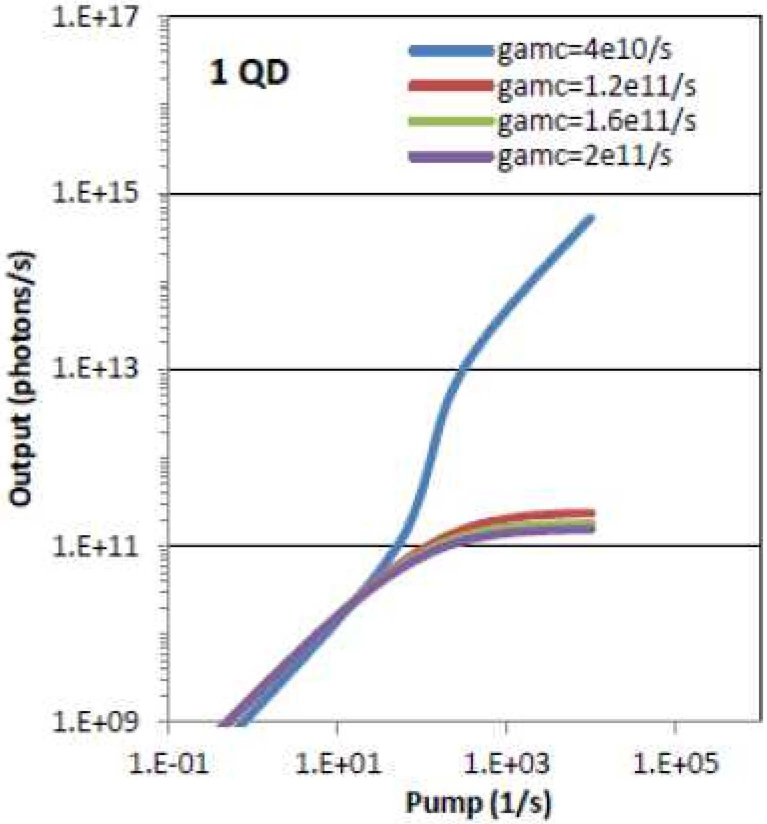
Frost et al., IEEE JQE, **49,** 923 (2013).

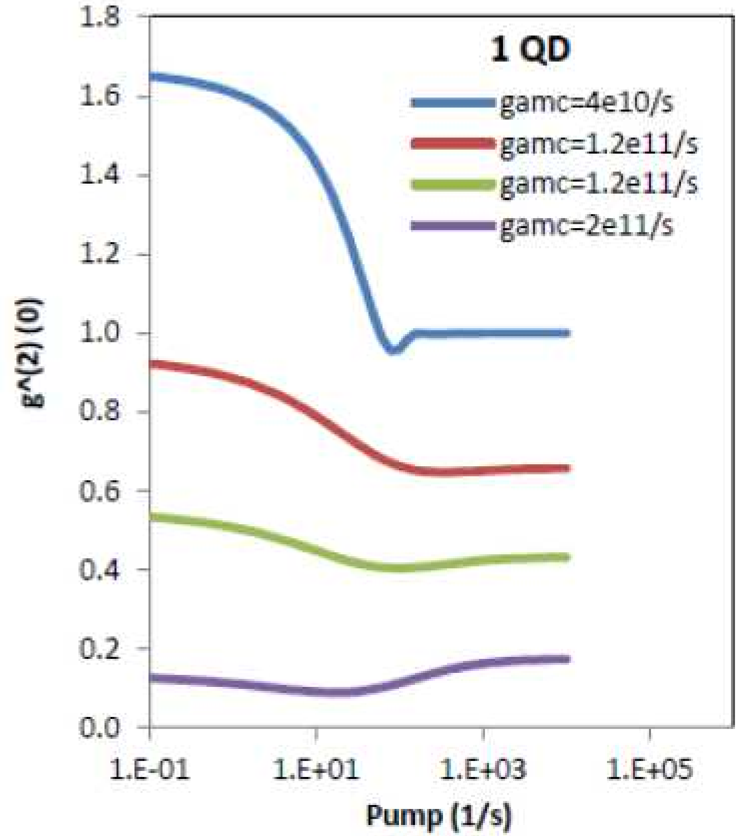


Monodisperse QD Distributions

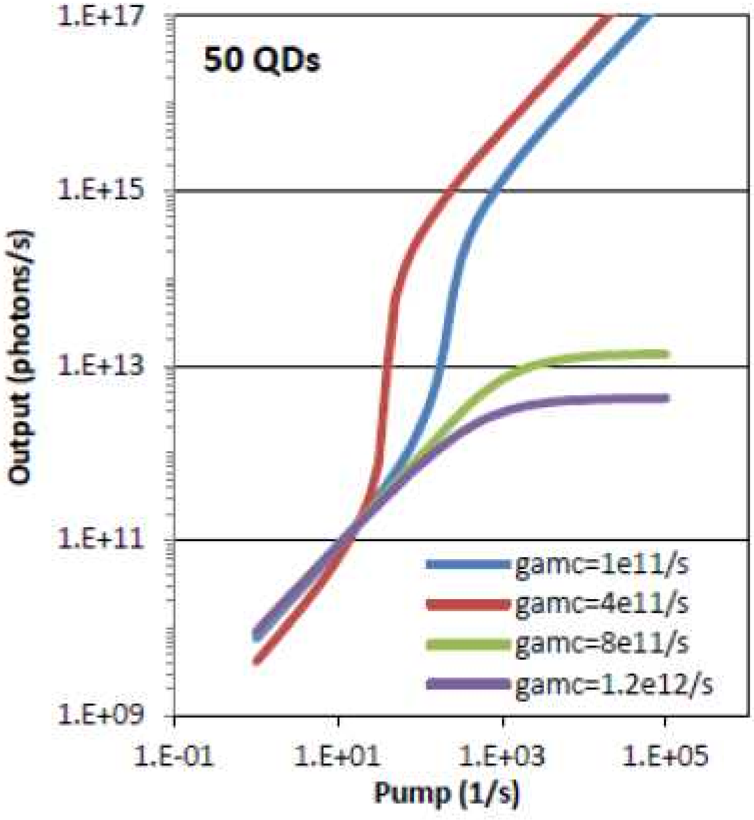


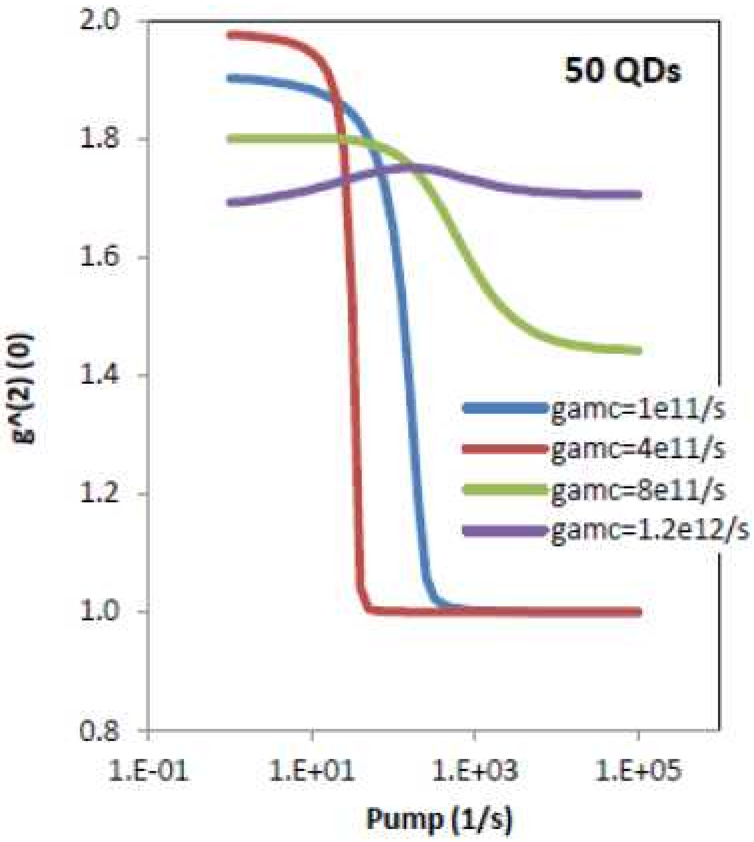
Calculated g2 for a single QD





Calculated g2 for 50 QDs





Calculated g2 for 5 QDs