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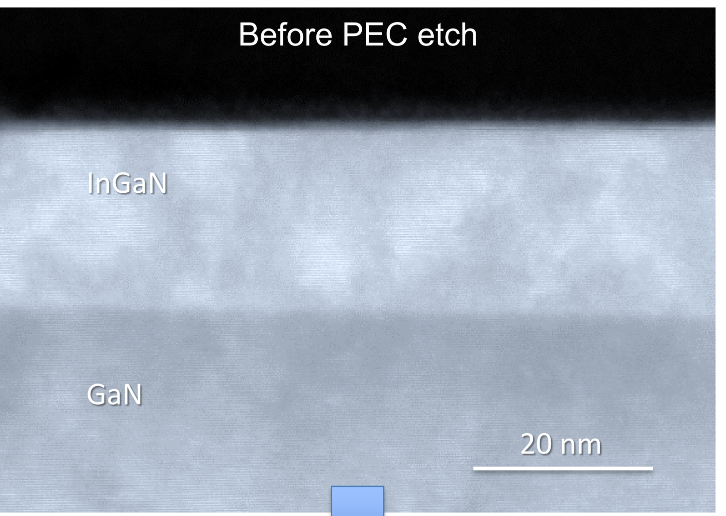

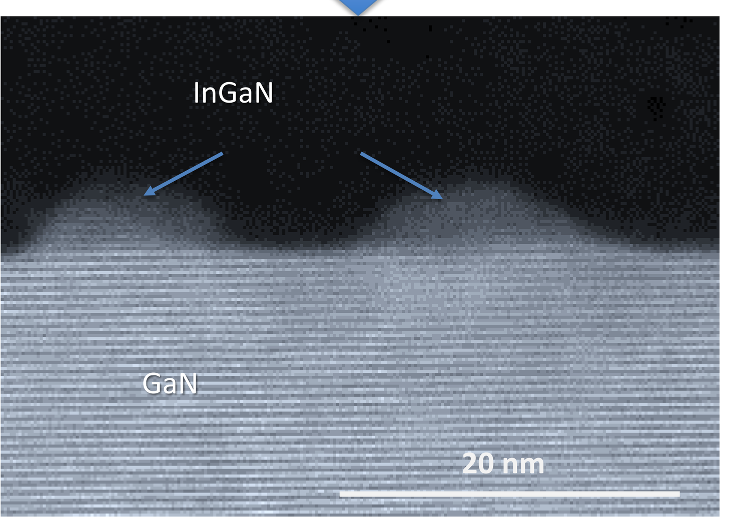

Abstract

III-nitride quantum dots (QDs) have significant potential for single-photon sources or gain media for low threshold and high efficiency visible and UV lasers, among others. "Bottom-up" Stranski–Krastanov growth is widely used--however both the size distribution and densities are difficult to precisely control. Here, we show a self-terminating top-down fabrication process, called quantum size controlled photo-electrochemical (QSC-PEC) etching, that can be controlled by the evolving properties of the nanostructures being fabricated.

Summary

- Demonstrated controlled fabrication of InGaN QDs from epitaxial layer using quantum-size controlled PEC etching
- AFM data indicates extremely high QD density (~10^{11} per cm^2)
- PL linewidth reduced from 24 nm to less than ~6 nm, indicating improved QD size distribution vs. Stranski-Krastanov grown InGaN QDs (~30-40 nm)
- PL measurements show significant enhancement of IQE for etched QDs vs initial QW
- **QD size and PL emission determined by PEC etch wavelength used!**
- Multiple layers of QDs demonstrated, but with lower per-layer density

Quantum Size Controlled Etching of InGaN Quantum Dots

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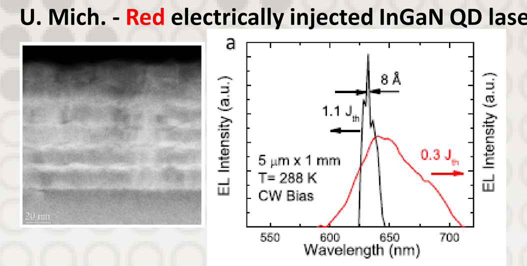
Motivation: InGaN Quantum Dots (QDs) as emitters

Potential advantages vs quantum wells

- Single photon emission
- Reduced polarization fields (reduced QCSE)
- Longer wavelength emission
- Lower lasing thresholds
- Higher efficiencies

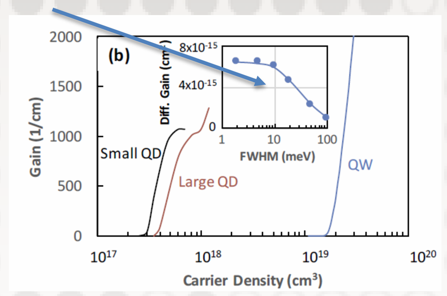
Schulz & Reilly, PRB 82, 033411 (2010)

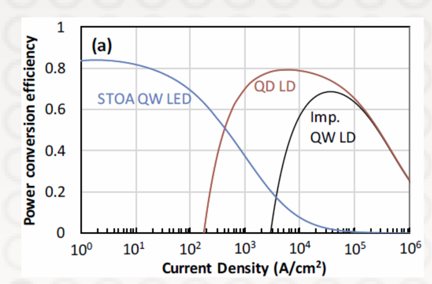
U. Mich. - Red electrically injected InGaN QD laser



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

Quantum nanostructure





Jonathan J. Wierer, Jr., Nelson Tansu, Arthur J. Fischer, and Jeffrey Tsao, "III-nitride quantum dots for ultra-efficient solid-state lighting," *Laser and Photonics Reviews*, 10, 612 (2016) / DOI 10.1002/lpor.201500332

Challenge: Find methods to synthesize QDs to meet the required sizes, inhomogeneous broadening, and densities

(In)GaN Photoelectrochemical (PEC) Etching

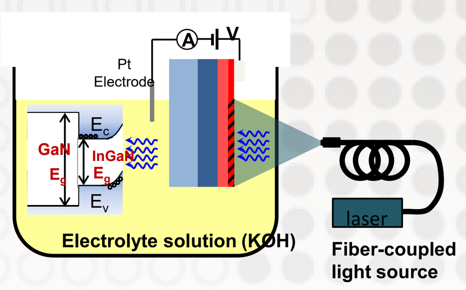
- GaN relatively inert to wet etching, particularly (0001) c-plane
- PEC etching: photogenerated holes oxide surface which is then dissolved

in 1 M H_2SO_4: $GaN + 3h^+ \rightarrow Ga^{3+} + 0.5N_2$

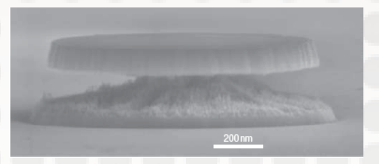
in 1 M KOH: $GaN + 6OH^- + 3h^+ \rightarrow GaO_3^{3-} + 0.5N_2 + 3H_2O$

Huygens et al., J. ECS 147, 1797 (2000)

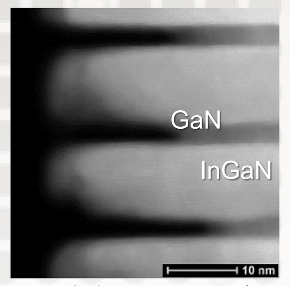
- Laser or lamp excitation (Xe arc lamp, tunable ps Ti:S)
- KOH (~0.1M) typically used as electrolyte for GaN
- Band gap selective (etch InGaN over GaN) based on wavelength used



Stonas et al., JVST B 19, 2838 (2001)



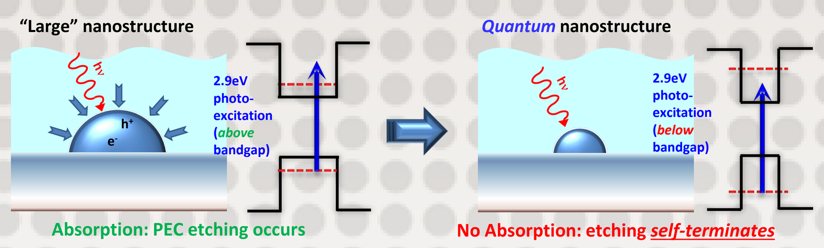
Tamboli et al., Nat. Phot. 1, 61 (2007)

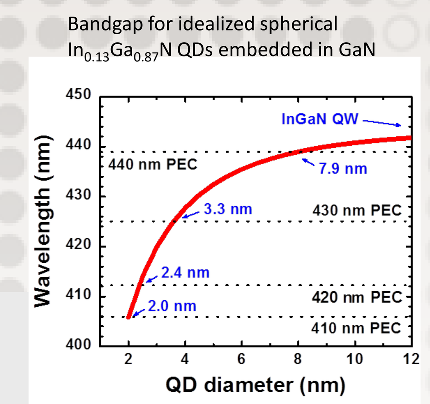


Xiao et al., Elec. Acta 162, 163 (2015)

Quantum-Size Controlled fabrication of QDs

Quantum Size Control: Use size quantization to control QD size





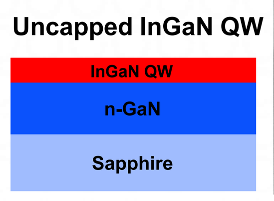
- For QDs, band gap depends on size
- As PEC etch proceeds,
  - QD size gets smaller, band gap goes up
  - Etch should terminate for $E_g > E_{photon}$ pump (no more carriers needed for etch)
- Self-terminating etch process
- Final QD size (band gap) depends on PEC etch wavelength used
- "Monodisperse" QD distributions ??
- Earlier work on "size selective photocorrosion" to shrink size of colloidal CdS QDs using monochromatic light[1,2]

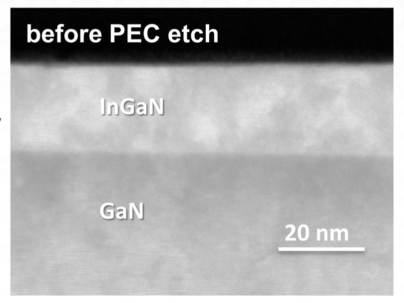
[1]Matsumoto et al., J. Phys. Chem. 100, 13781 (1996) [2]Torimoto et al., J. ECS 145, 1964 (1998)

PEC Etching of InGaN films into QDs

Uncapped InGaN QW

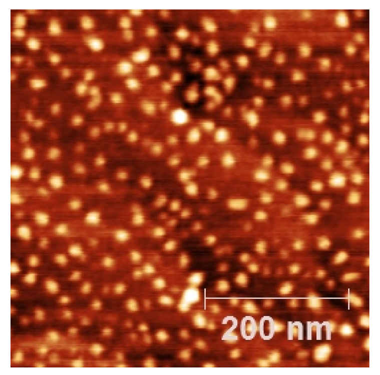
Uncapped 5 - 20 nm thick $In_{0.14}Ga_{0.86}N$ layer:



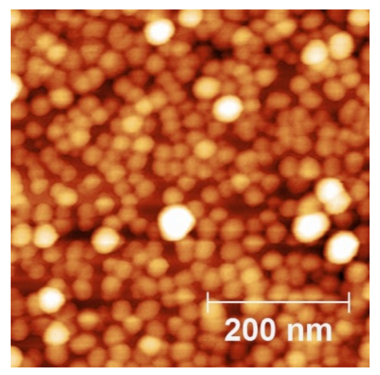


AFM

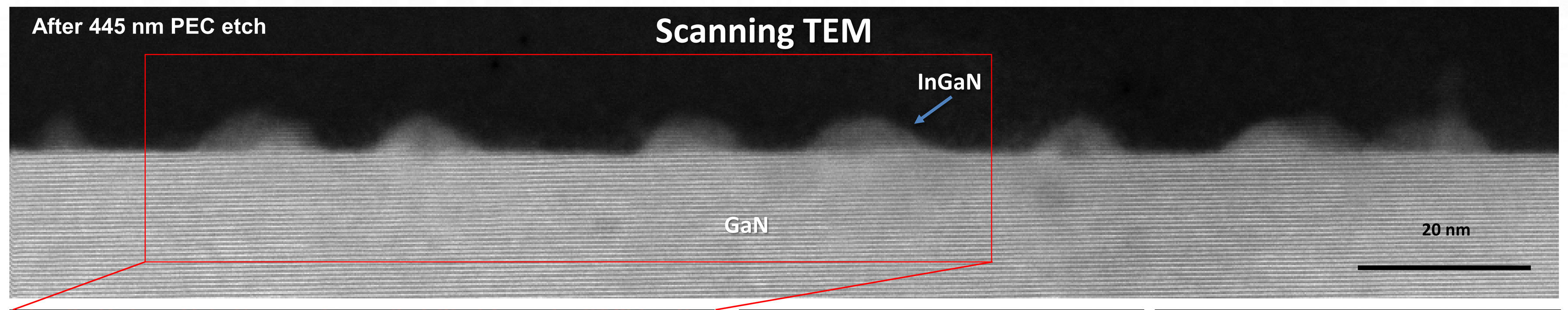
PEC etch→ λ = 420 nm

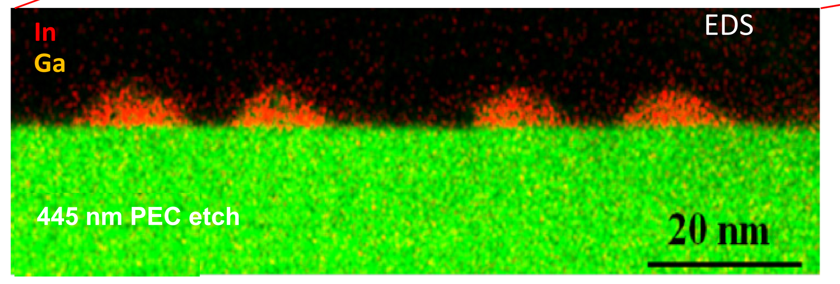


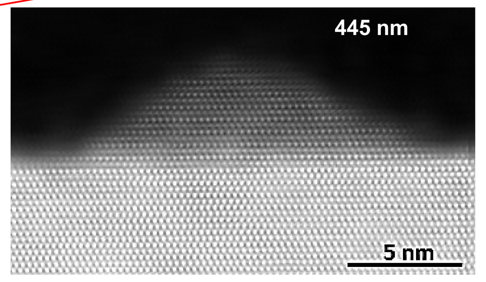
PEC etch → λ = 445 nm

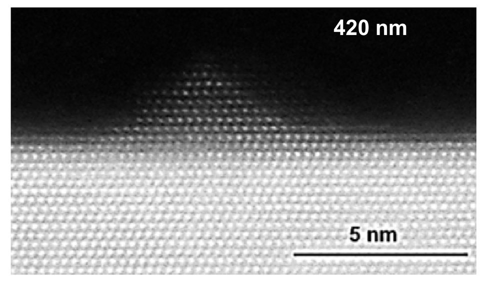


- Samples etched two hours at 420 nm and 445 nm
  - 0.2M H_2SO_4 solution (no dark etch)
  - Tunable Ti:sapphire laser (2 ps pulse width, < 1 nm linewidth,82 MHz pulse repetition rate), Laser power density: ~ 3 mW/cm^2
- Observe formation of (quantum) dots!
  - Very high dot density: ~10^{11}/cm^2
- Some big dots (10-20 nm) remain: due to dislocations?
- QD size depends on PEC etch wavelength!



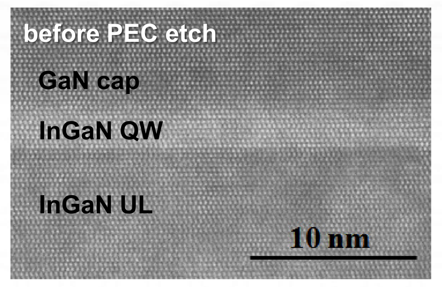




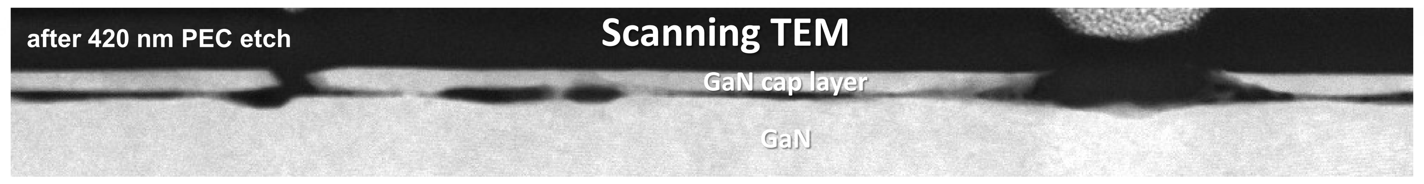


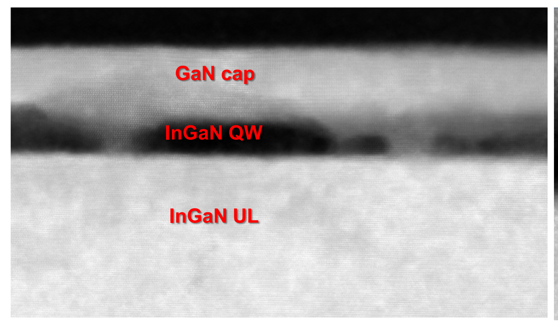
Capped InGaN QW

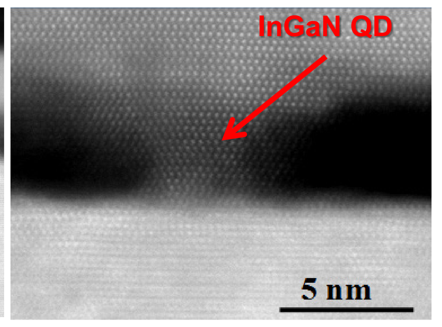
~3 nm $In_{0.14}Ga_{0.86}N$ QW, 10 nm GaN cap

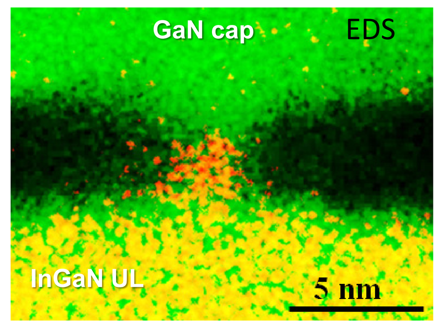


- Sample PEC etched at 420 nm
- Etch appears to break through GaN cap – defects?
- EDS mapping: dots are InGaN
- QD PL is ~100x brighter than uncapped QDs
- GaN cap provides (partial) passivation



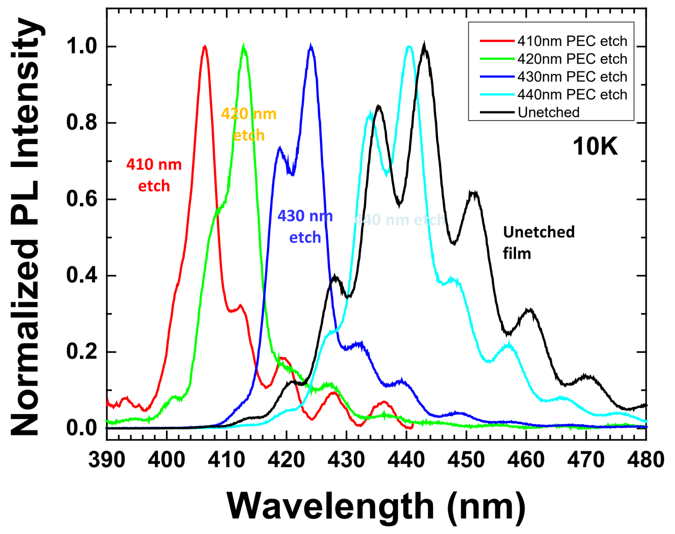






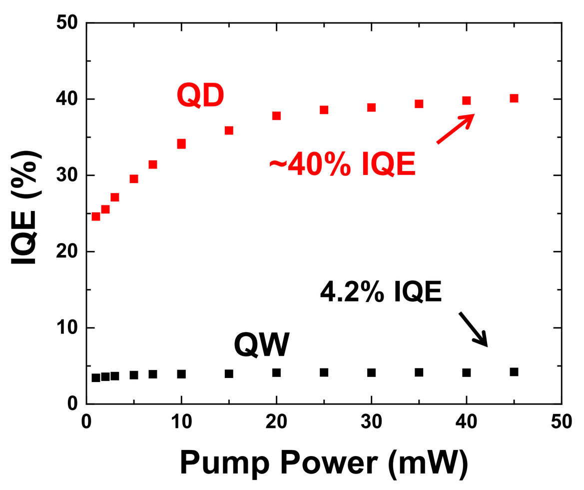
Optical Characterization of QDs

Photoluminescence (PL) of etched QDs



- 375 nm pump (ps pulsed), 10K
- Etched QD PL **wavelength correlates** (but doesn't exactly coincide) **with PEC etch wavelength**
- PL linewidth **decreases with decreasing etch wavelength**: 24 nm (film) → 6 nm (QDs etched at 410 nm)
- **Strong evidence for quantum-size controlled mechanism!**

Internal Quantum Efficiency (IQE)



- 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**

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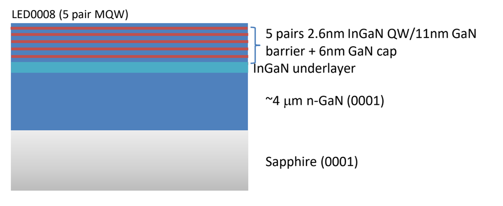
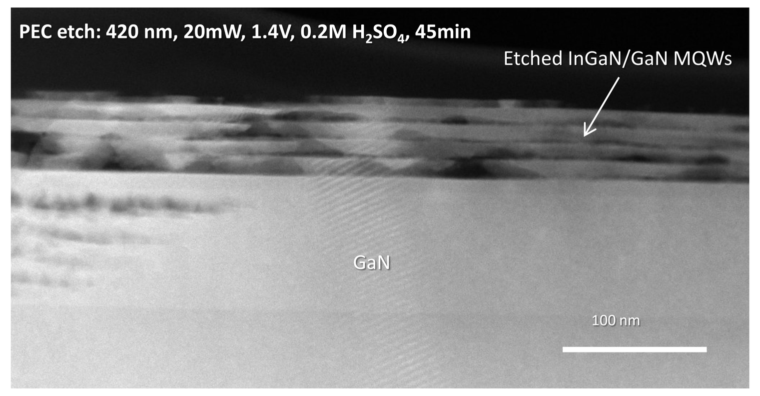
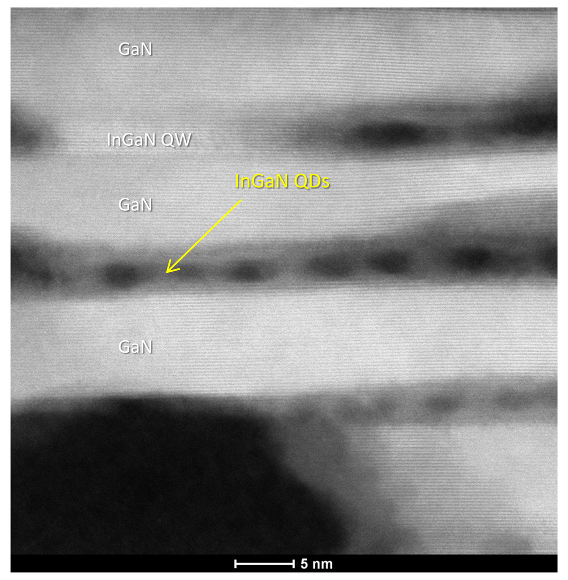
InGaN/GaN Multi-Quantum Wells (MQWs)

- QD lasers need multiple layers of QDs to provide sufficient gain -> PEC etch InGaN/GaN MQWs to provide **multiple layers of capped/passivated QDs?**
- Cross-sectional STEM shows **all InGaN layers etched after PEC etch at 420 nm**, with GaN voids present
- InGaN QD density per layer appears low compared to QSC-PEC etched single InGaN layer, potentially explaining relatively low PL of etched MQW sample







References

1. X. Xiao, A. J. Fischer, G. T. Wang, P. Lu, D. D. Koleske, M. E. Coltrin, . . . J. Y. Tsao, "Quantum-Size-Controlled Photoelectrochemical Fabrication of Epitaxial InGaN Quantum Dots", *Nano Lett.*, **14**, 5616-5620 **2014**.
2. X. Xiao, A. J. Fischer, M. E. Coltrin, P. Lu, D. D. Koleske, G. T. Wang, . . . J. Y. Tsao, "Photoelectrochemical etching of epitaxial InGaN thin films: self-limited kinetics and nanostructuring", *Electrochim. Acta*, **162**, 163-168 **2015**.
3. X. Xiao, P. Lu, A. J. Fischer, M. E. Coltrin, G. T. Wang, D. D. Koleske, J. Y. Tsao, "Influence of pH on the Quantum-Size-Controlled Photoelectrochemical Etching of Epitaxial InGaN Quantum Dots", *The Journal of Physical Chemistry C*, **119**, 28194-28198 **2015**.

Acknowledgements

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