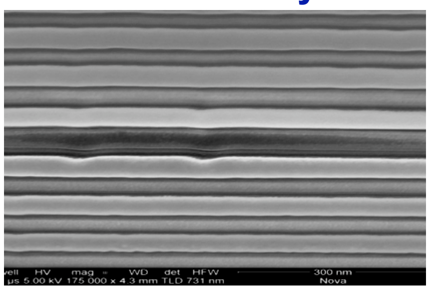
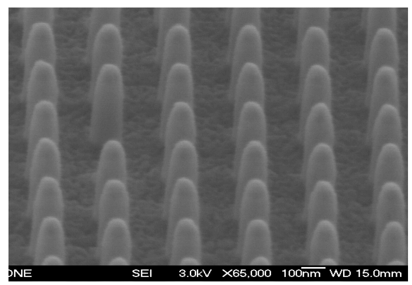
Enhanced spontaneous emission rate in visible III-nitride LEDs using 3D photonic crystal cavities.

Project Manager: James Hudgens
Principal Investigator: Arthur Fischer
Investment area: Electronics and photonics

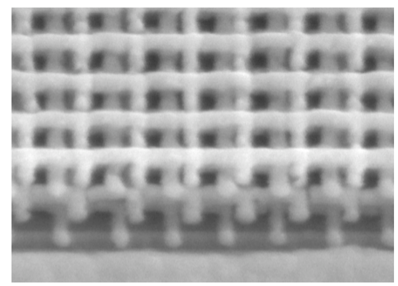
1D Photonic crystal



2D Photonic crystal



3D Photonic crystal



<u>Co-investigators:</u> Ganesh Subramania, Ting Shan Luk, Dan Koleske, Yun-Ju Lee, Qiming Li, George Wang, Kris Fullmer, and Anthony Coley.

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SAND Number: 2009-5157 P

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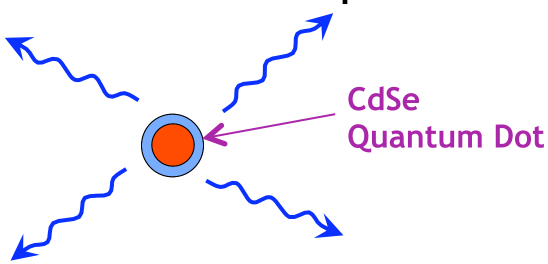
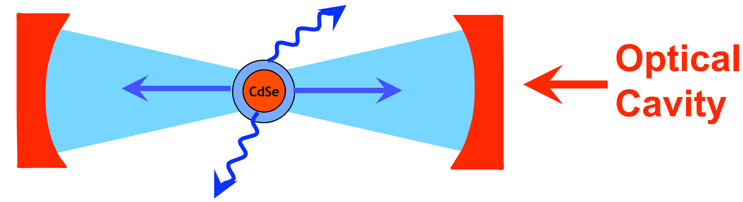
Photonic Density of States and Light Emission

Emitter in free space



- Emitter couples to free space radiation modes
- Emitter decays with natural, fixed radiative rate (R_r)

Emitter in environment with altered photonic density of states



- Emitter couples to different radiation modes
- Radiative rate can be increased or decreased
- Photonic crystals, surface plasmons, etc…

Fermi's Golden Rule:

$$\lambda_{if} = \frac{2\pi}{\hbar} \left| M_{if} \right|^2 \rho_f$$

Transition Probability

Matrix Element

Density of Final States

Purcell Effect:

$$F = \frac{3Q_m g(\lambda/2n)^3}{2\pi V_{eff}}$$

n = index
g = degeneracy
l = wavelength

Q = quality factor
V = mode volume

Ultimate control over when and if a photon is emitted !

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3D photonic crystal: density of states

Radiative decay rate
(dipole in a PC at coordinate ‘r’)

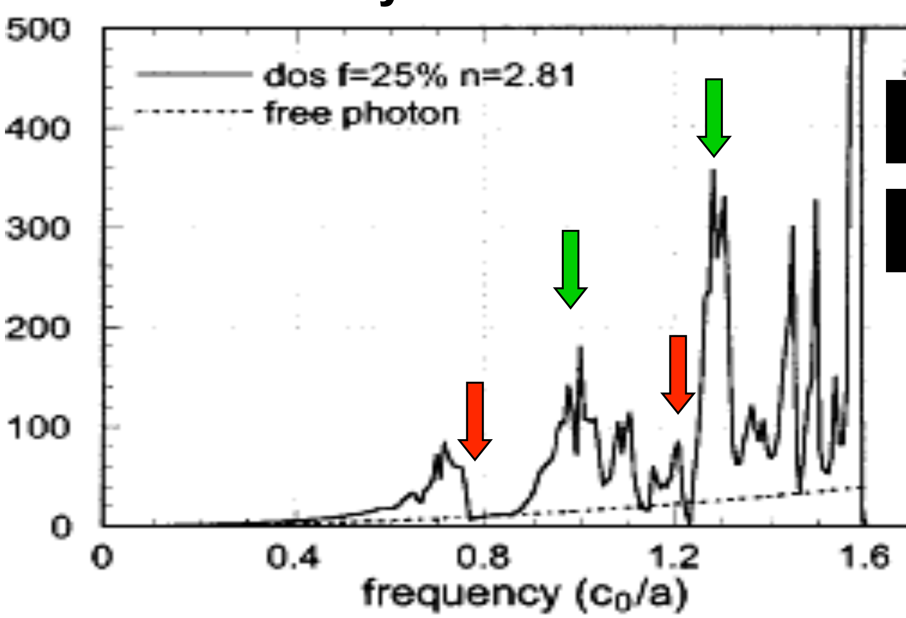
$$\Gamma_{\mathrm{rad}}(\boldsymbol{r}) = \frac{\pi\omega\mu^2}{3\hbar\epsilon_0} N_{\mathrm{rad}}(\boldsymbol{r},\, \boldsymbol{d},\, \omega)$$

Local radiative density of states (LRDOS)

$$N_{\mathrm{rad}}(\boldsymbol{r}, \boldsymbol{d}, \omega) = \frac{1}{\epsilon(\boldsymbol{r})} \sum_{n,\boldsymbol{k}} 2\omega\delta(\omega^2 - \omega_{n,\boldsymbol{k}}^2)\, |\boldsymbol{d} \cdot \boldsymbol{A}_{n,\boldsymbol{k}}(\boldsymbol{r})|^2$$

$$\boldsymbol{A}_{n,\boldsymbol{k}}(\boldsymbol{r}) = (\nabla \times \boldsymbol{H}_{n,\boldsymbol{k}}(\boldsymbol{r}))/(\sqrt{\epsilon(\boldsymbol{r})}\, i\omega_{n,\boldsymbol{k}})$$

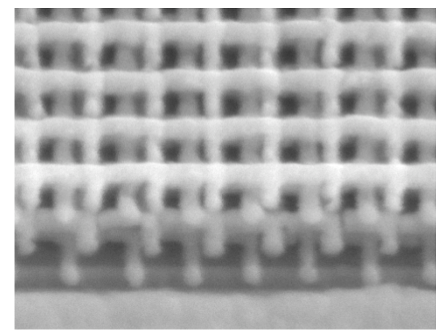
Photon Density of States in a 3D PC*



❖ **Enhancement**

❖ **Suppression**

3D photonic crystal



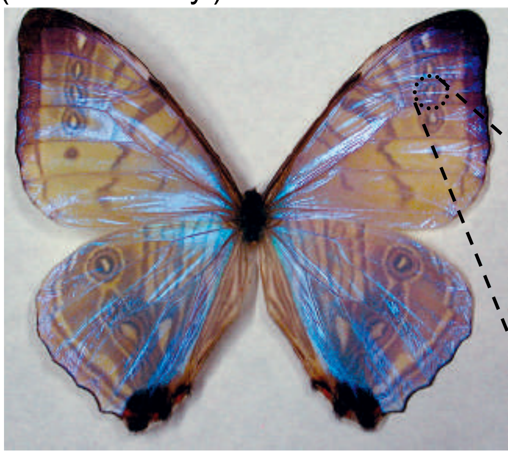
→ **3D PCs offers complete light control**
→ **Density of states is position dependent**
→ **Emitter placement inside PC is crucial**

*T.Suzuki et. al., JOSA-B, **12**, 570 (1995)

Light Control In Nature Via Nanostructuring

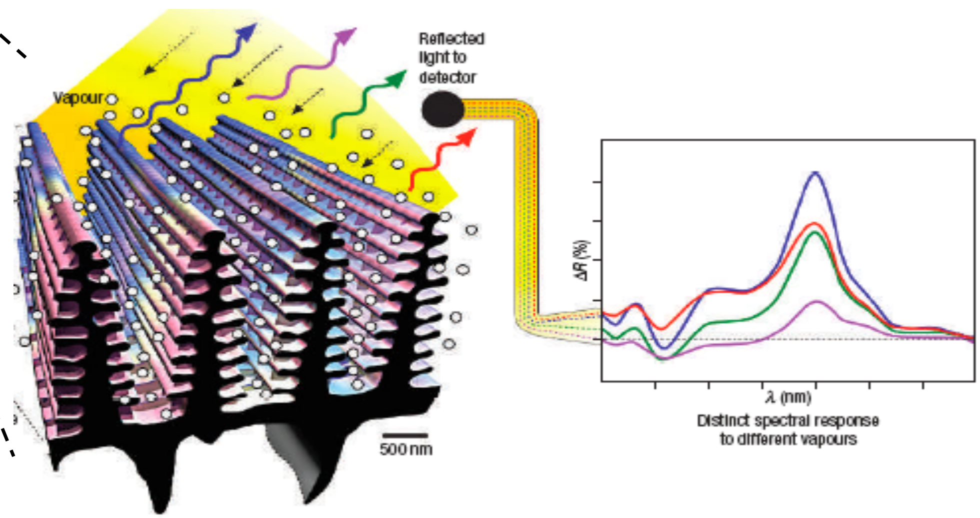
Morpho Butterfly

(M. Sulkowskyi)



(M. menelaus)

- **Photonic Crystal structures in butterfly wings.**
- **Color response highly sensitive to different vapors**
- **Signals by giving off colors by strong reflection of sunlight : visible half mile away**



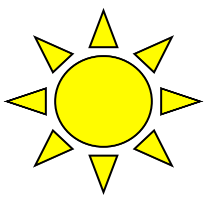
Passive control of light sources to suit one's requirements

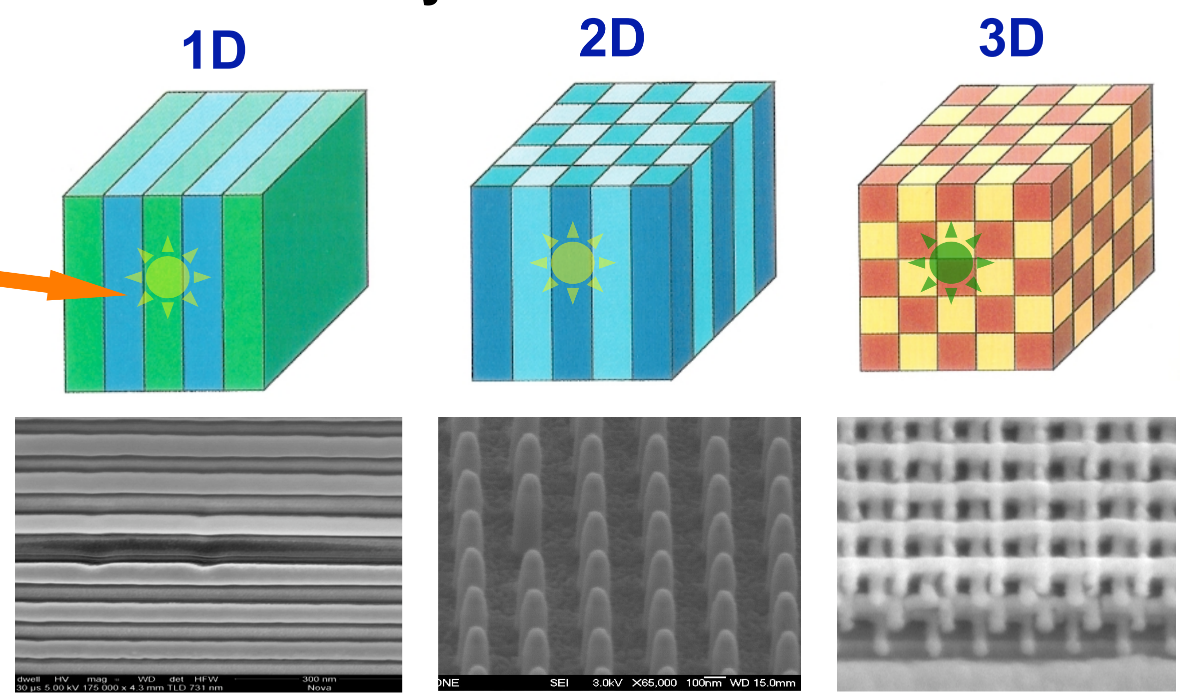
*** R.A. Potyrailo et. al. , Nature Photonics, 1, 123 (2007)**

Light emission control with photonic crystals

Photonic Crystal Nanostructures

Light Sources
(QDs, Qwires, thermal)





HfO_2/SiO_2 DBR cavity **Array of TiO_2 rods** **TiO_2 logpile PC**

Modify emission behavior: Sources with novel and improved functionalities

- **High brightness/efficiency**
- **Tunable**
- **Directional, compact**
- **Anti-bunched**
- **Entangled**

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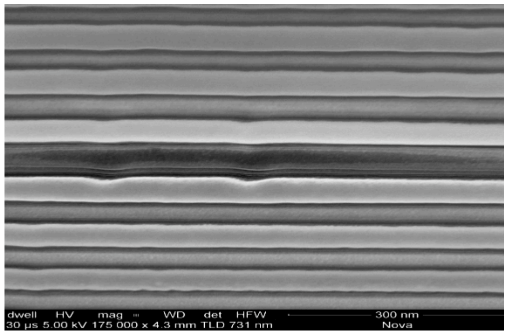
1D microcavity with GaN emitter

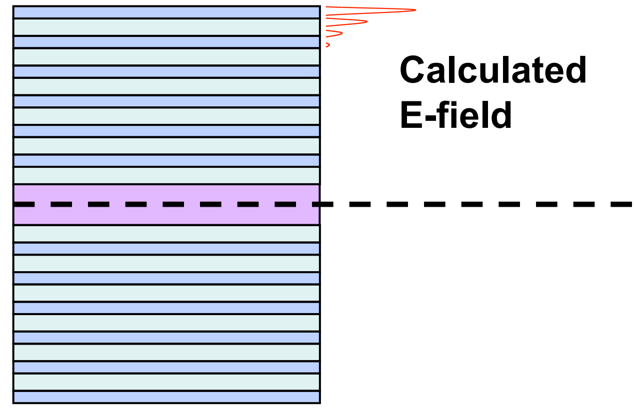
FIB-SEM image → 1D HfO_2/SiO_2 cavity



SiO_2/HfO_2 DBR

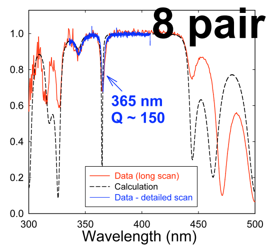
GaN epilayer

SiO_2/HfO_2 DBR

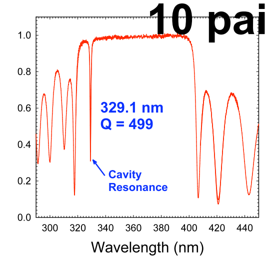


Calculated E-field

8 pair GaN microcavity



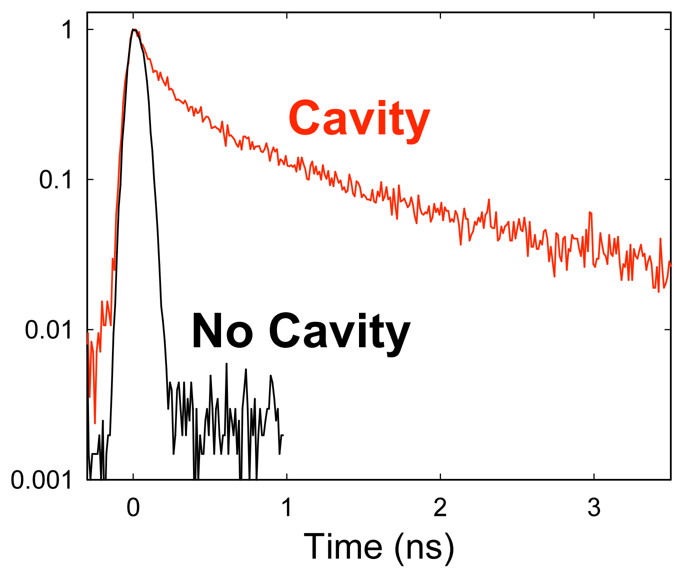
10 pair GaN microcavity



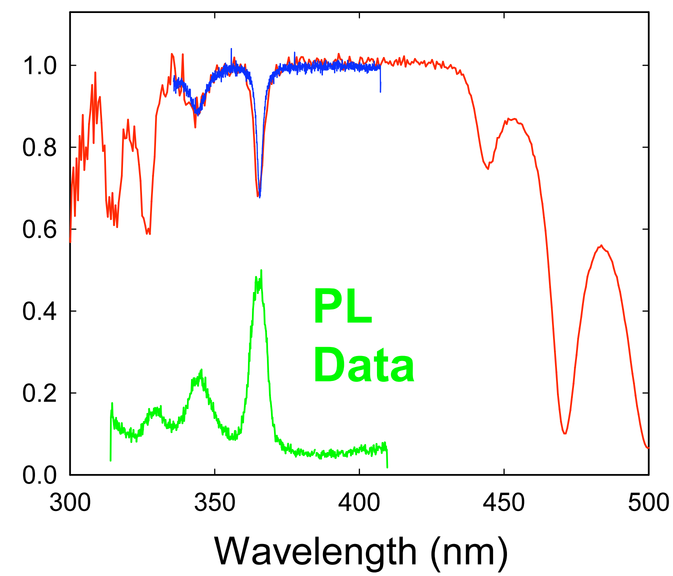
1D microcavity with GaN emitter: photoluminescence

- Laser-lift-off process is used to remove sapphire substrate
- 8 pair SiO_2/HfO_2 mirror used to form cavity
- GaN high Q cavities can lead to exciting new physics studies
    - Strong coupling, polaritons
    - Room temperature Bose-Einstein condensates
- **DBR microcavity suppresses emission from GaN epilayer**
- **Dramatic change in lifetime observed**

Time-resolved Photoluminescence

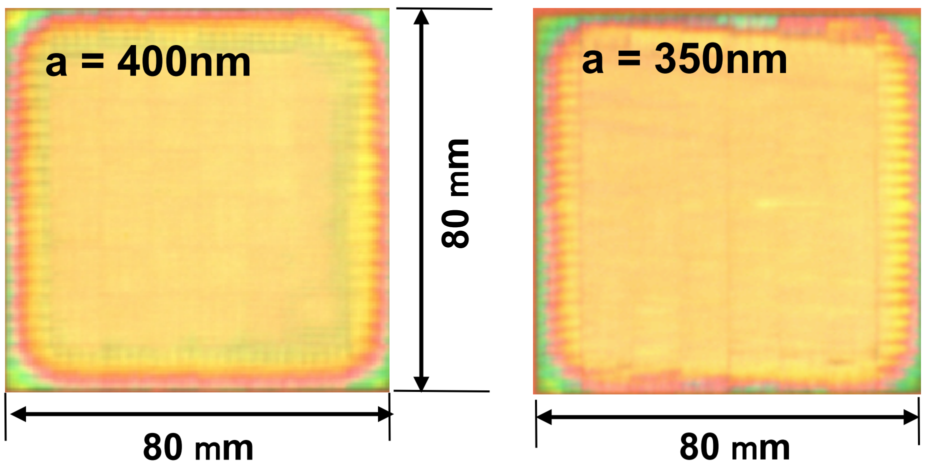


Time-integrated Photoluminescence

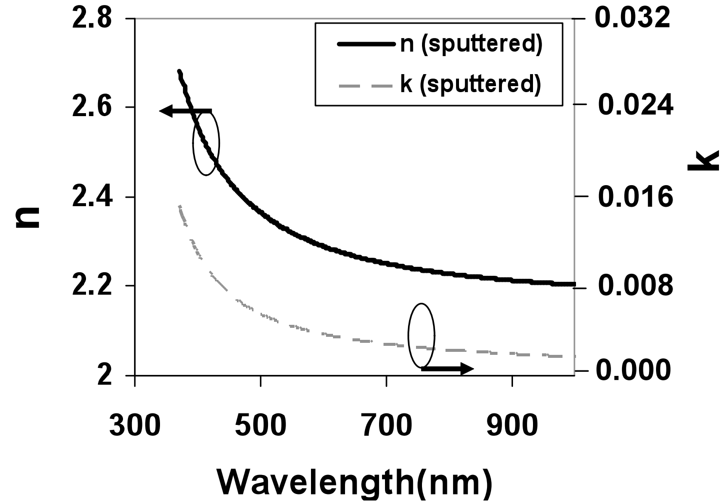


Visible TiO_2 Logpile 3D Photonic Crystals

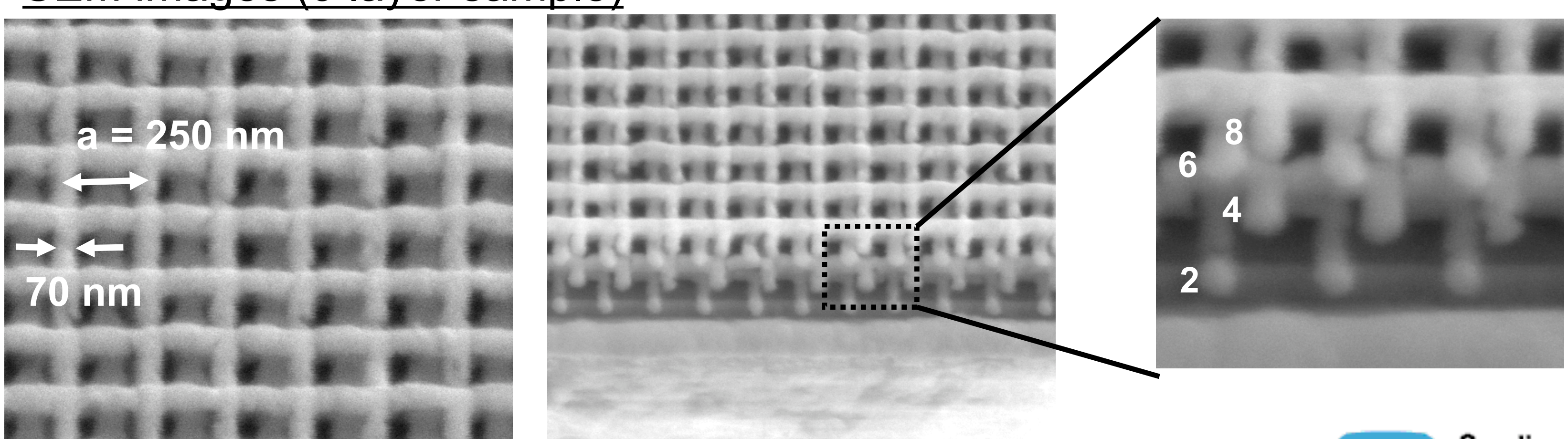
White light microscope images (5 layer sample)



TiO_2 index of refraction

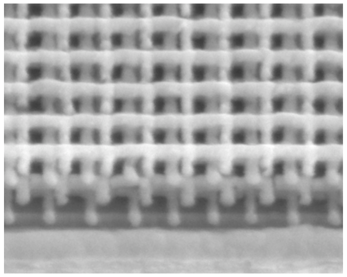


SEM images (9 layer sample)



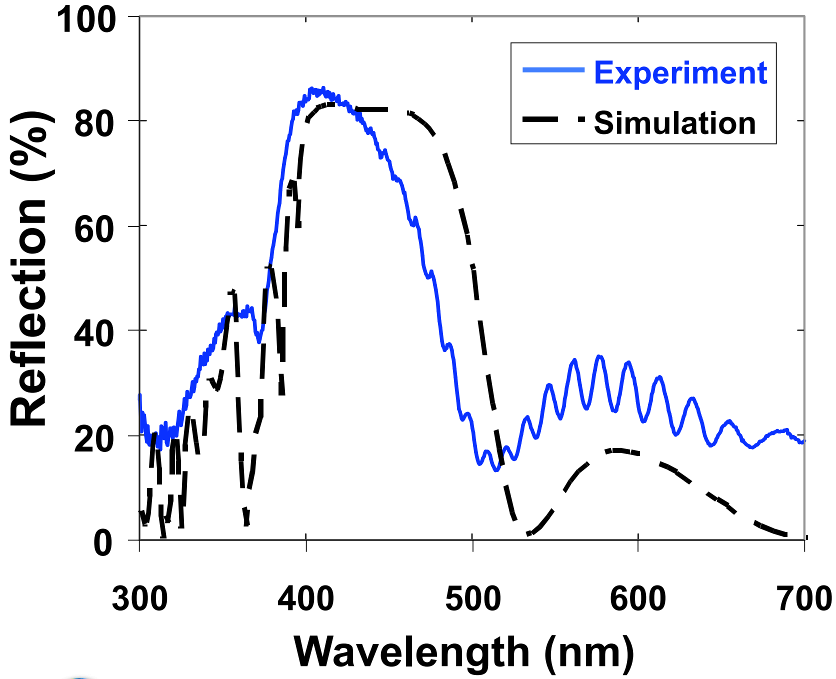
Reflectance from 9-Layer Visible TiO_2 Logpile

SEM - 9 layer logpile

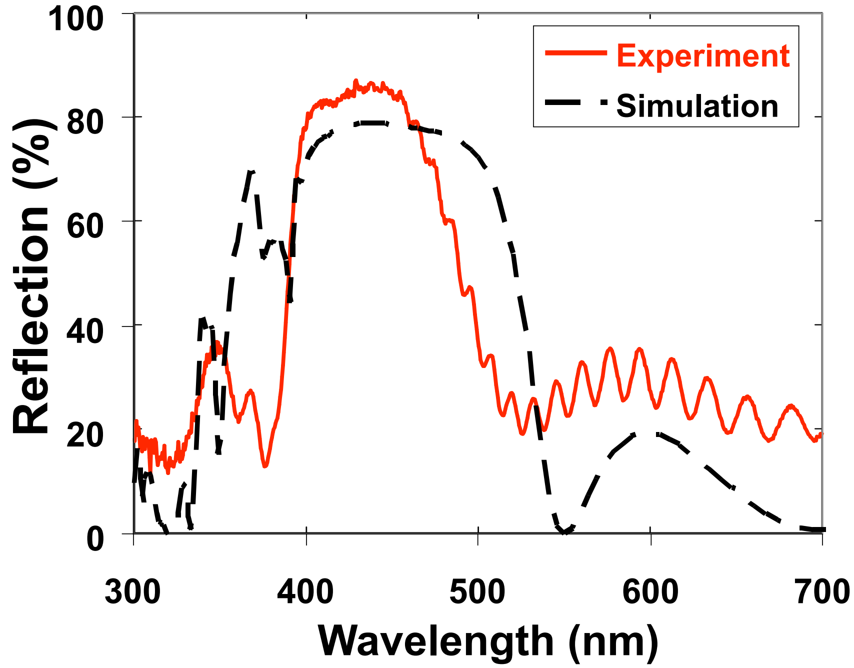


- Near normal incident reflection data
- Nine layer structure with lattice constant a=250 nm
- Bandgap extends from 400 to 500 nm
- Reflection greater than 80% → excellent morphology
- Demonstrates that TiO_2 PCs can be used at 400 nm

Reflection- Parallel Polarization



Reflection- Perpendicular Polarization



3D photonic crystal 'air defect' microcavity

Geometry: 9 layer structure with microcavity defect in the 5th layer

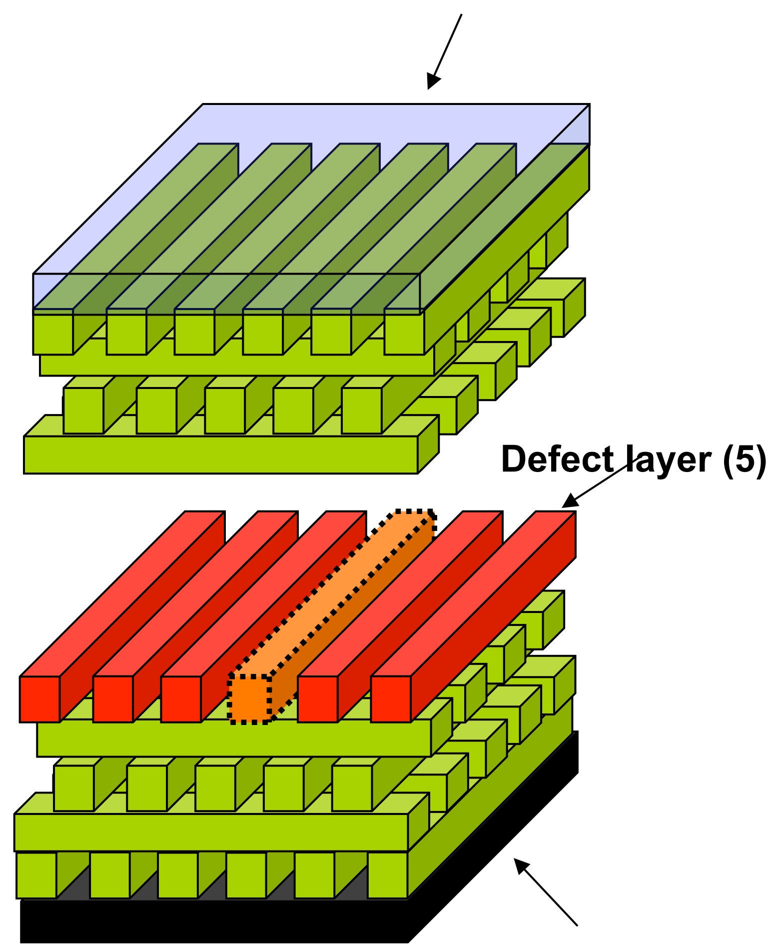
• Portions of rods removed to create 'air' defect cavities

• Fabricate bottom 5 layers first and then the top 4 layer separately on a transparent substrate (sapphire)

•Enables introduction of light emitters (e.g. QDs) post fabrication using chemical functionalization (already developed)

•Evaluate cavity modes: resonance frequency, Q factor

• Small modes volumes and high Q are desired



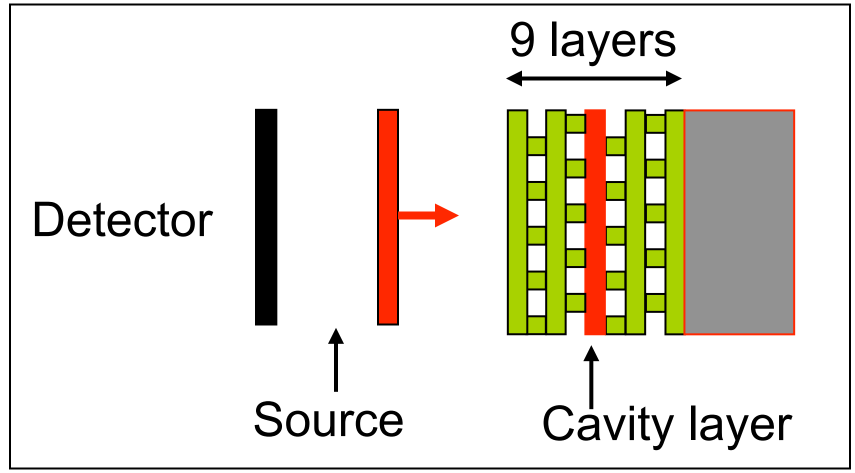
FDTD modeling of microcavity reflectance

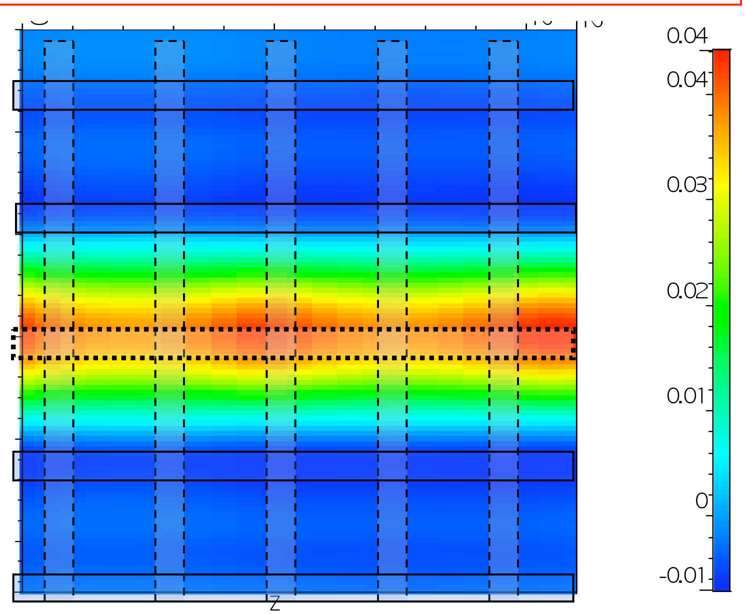
FDTD conditions:

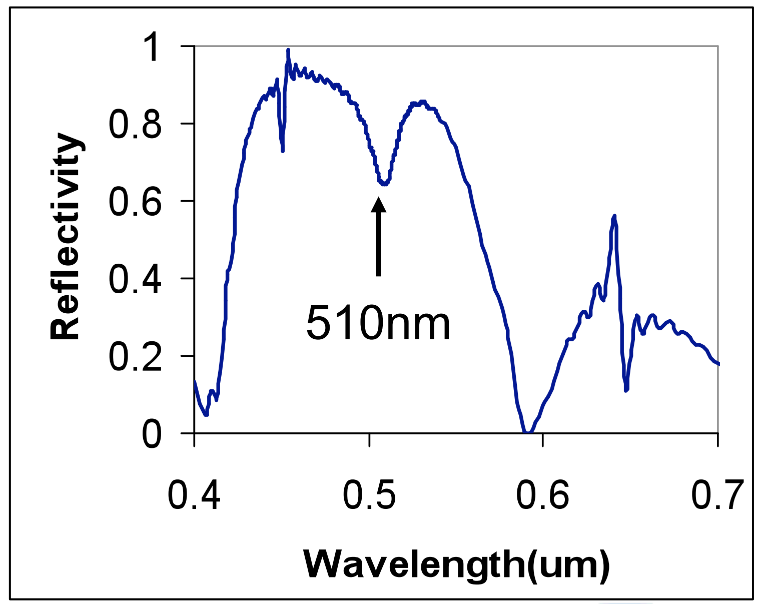
- Supercell: 5 unit cells
- Cavity defect periodic with 5 unit cells
- Periodic boundary conditions in the transverse
- Gaussian source

Cavity A: Every 5th rod removed

- Q factor ~ 30
- Mode volume : 2*(l/n)^3







LDRD
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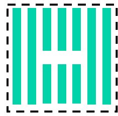
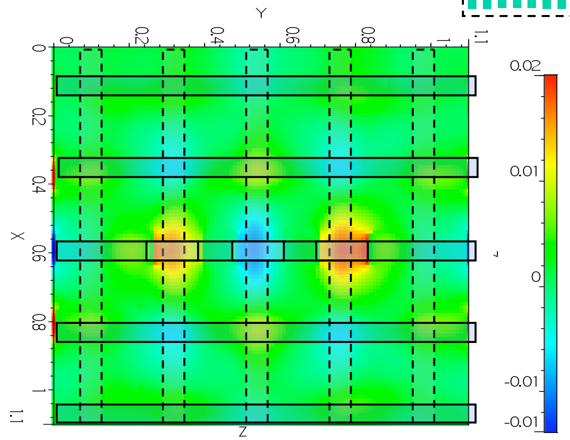
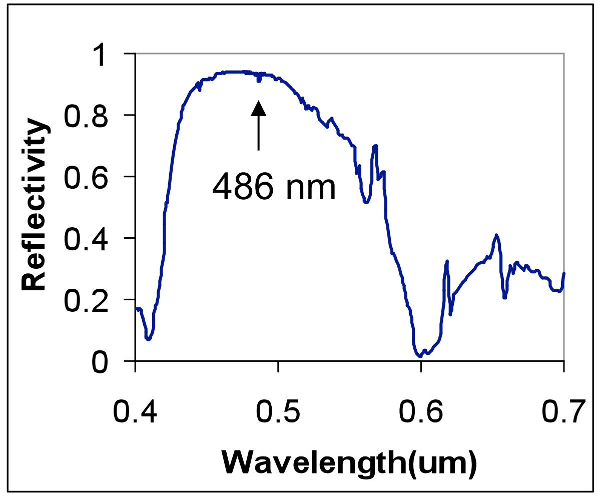
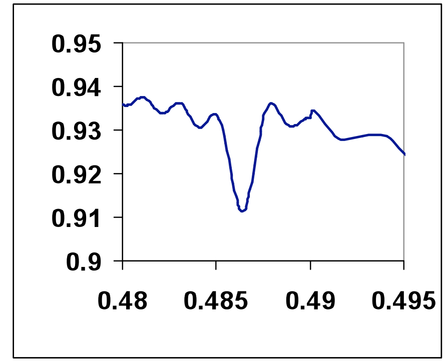
FDTD modeling of microcavity reflectance

Cavity B: →



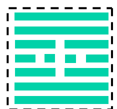


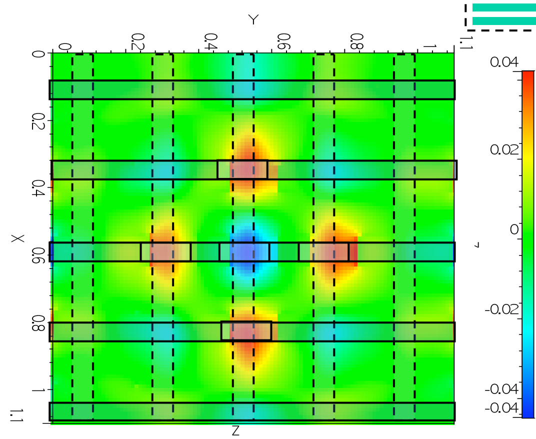


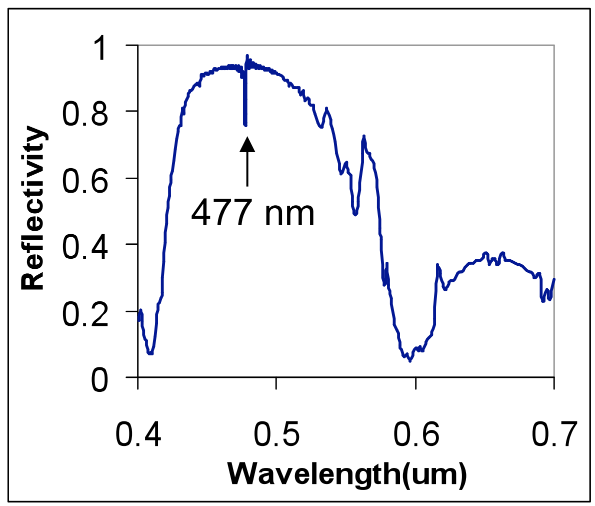


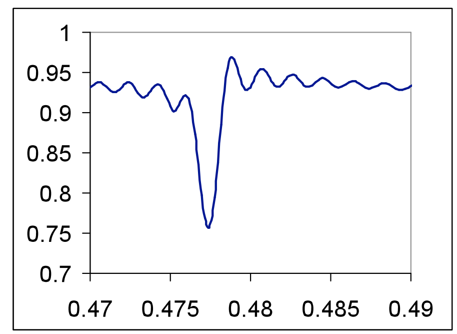
- Q ~ 250
- MV : 0.25(l/n)^3

Cavity C: →









- Q ~ 300
- MV : 0.42(l/n)^3

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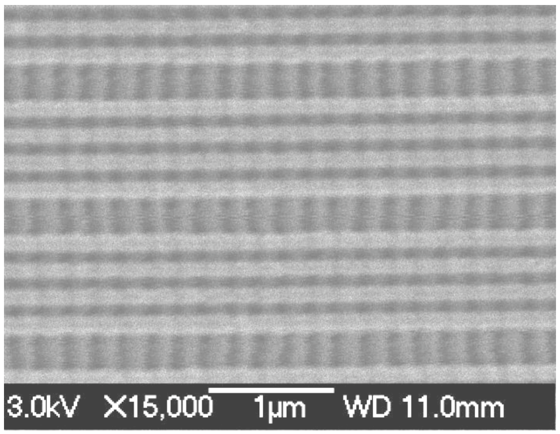
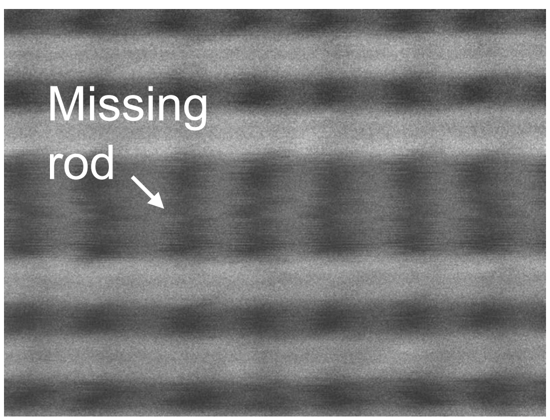
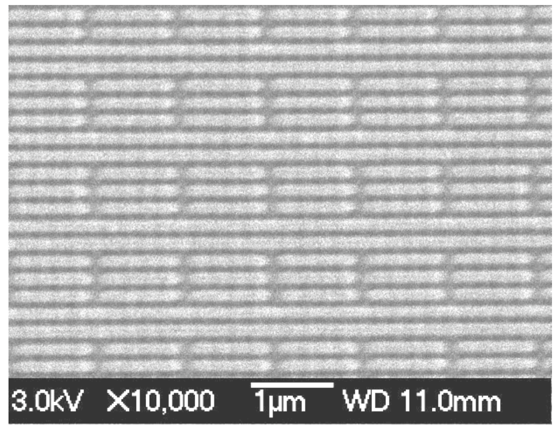
Fabricated TiO_2 Photonic Crystal Cavities

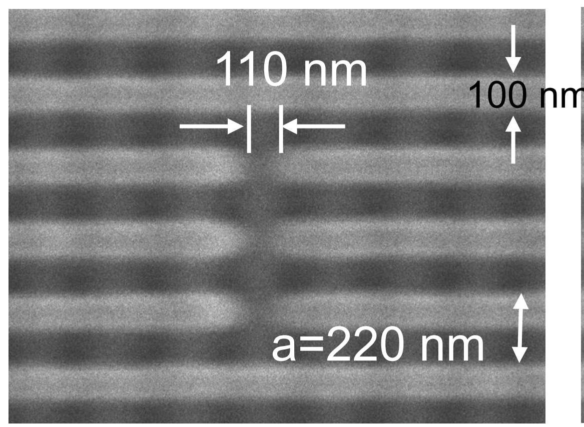
Cavity A



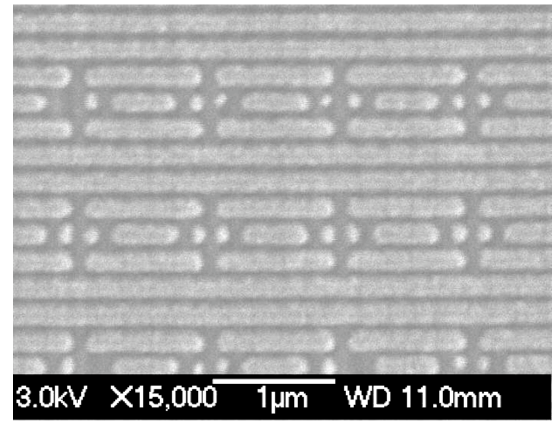


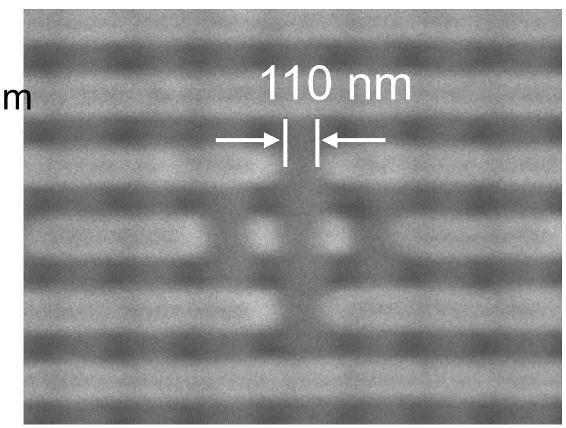
Cavity B





Cavity C





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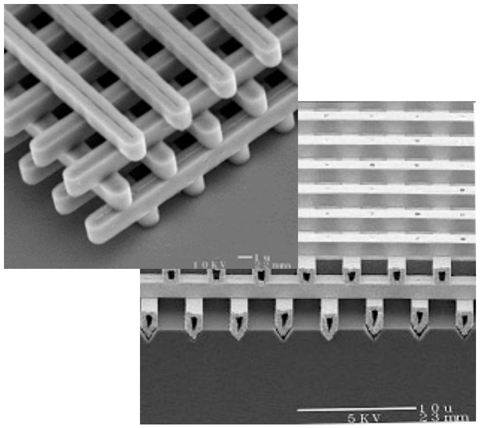
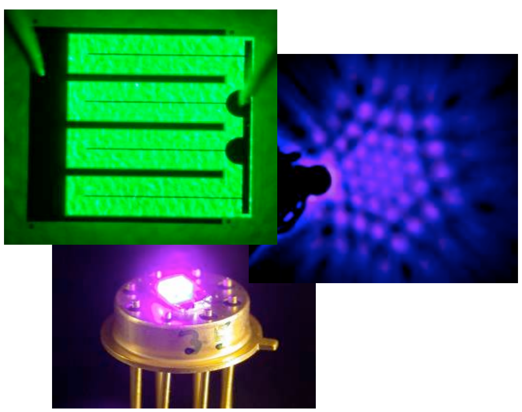
Combine 3D photonic crystals and InGaN LEDs

- **Use InGaN LEDs as a test case for enhanced spontaneous emission.**
- **Merge InGaN LED research with 3D photonic crystal research.**
- **Sandia is a recognized world leader in both areas.**
- **Both research areas will benefit.**
  - **GaN is an attractive dielectric for 3D PBG work.**
  - **InGaN LED efficiency enhancements benefit SSL**

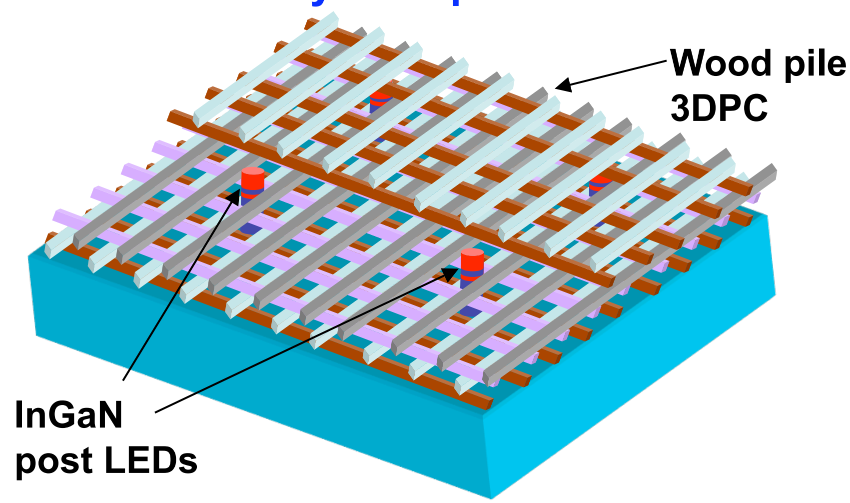
3D photonic crystals



InGaN LEDs

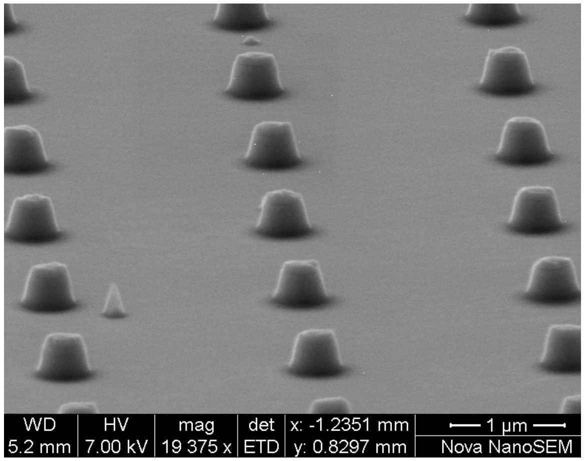


InGaN LED array incorporated into a 3DPC

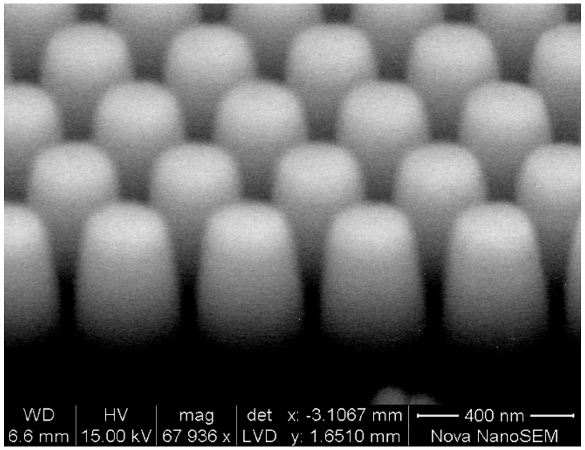


Incorporation of emitters: InGaN nanopost LEDs

loose-packed nanopost LED array

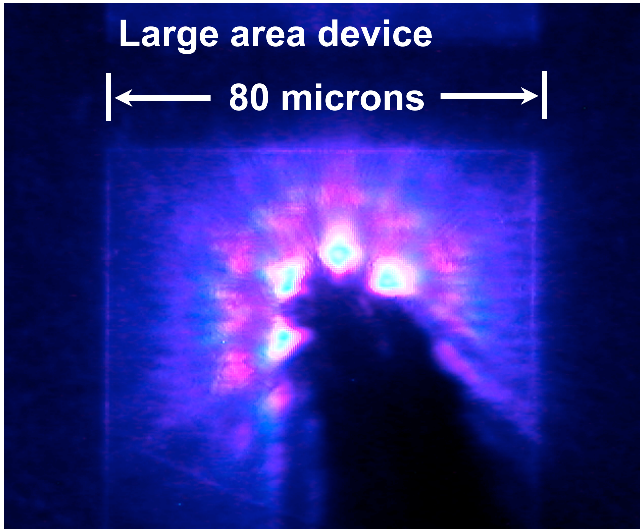


close-packed nanopost LED array



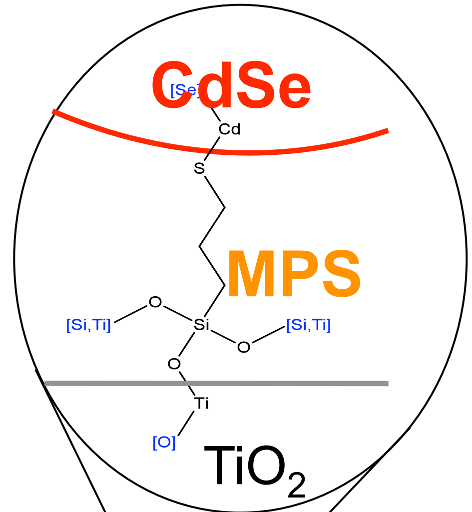
- Fabricated InGaN nanopost LEDs
- Arrays were fabricated such that a 3D logpile PC could be formed around posts.
- Difficult to electrically connect LEDs
- Original plan was to incorporate InGaN nanopost LEDs in 3D PCs
- Better methods of emitter incorporation were developed as a part of this program

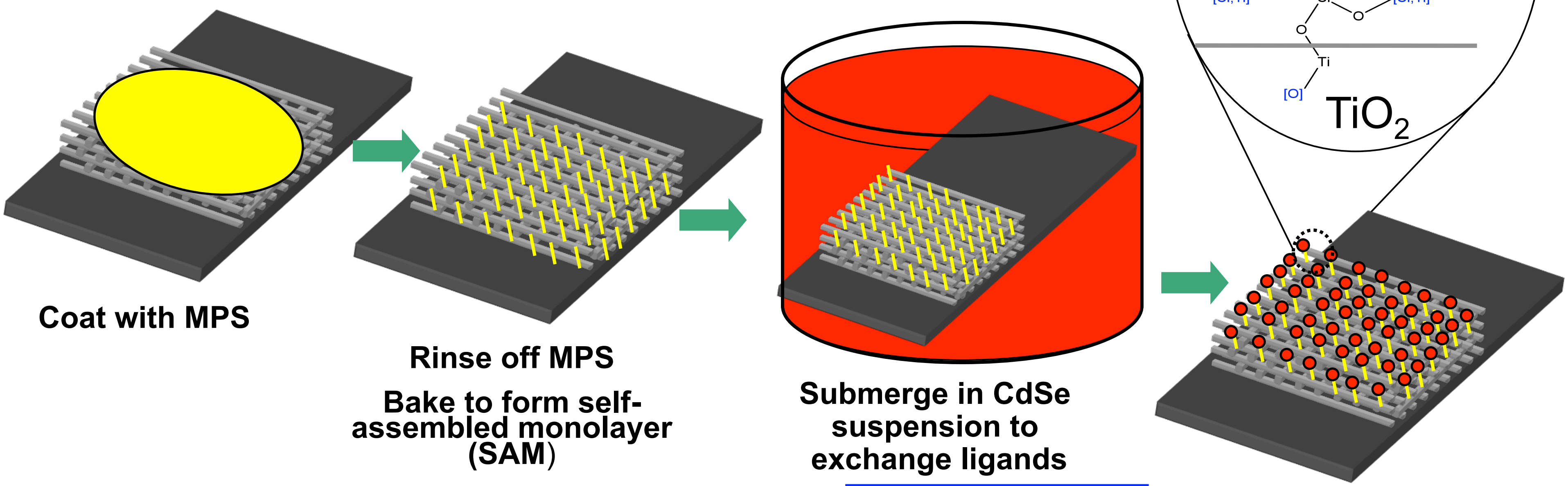
EL from one (or a few) nanopost LEDs



Self-assembled Monolayer of CdSe on TiO_2

1. Coat TiO_2 logpile with neat 3-mercaptopropyltrimethoxysilane (70 °C, 3 min)
2. Rinse with chloroform
3. Bake logpile on hot plate to form linkage (120°C, 30 min)
4. Place TiO_2 logpile in TOPO-capped CdSe in toluene (RT, 1 hr)





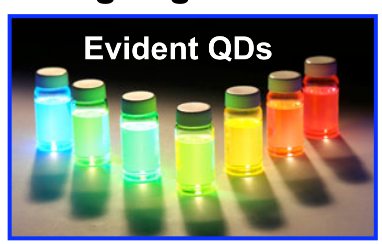
Coat with MPS

Rinse off MPS

Bake to form self-assembled monolayer (SAM)

Submerge in CdSe suspension to exchange ligands

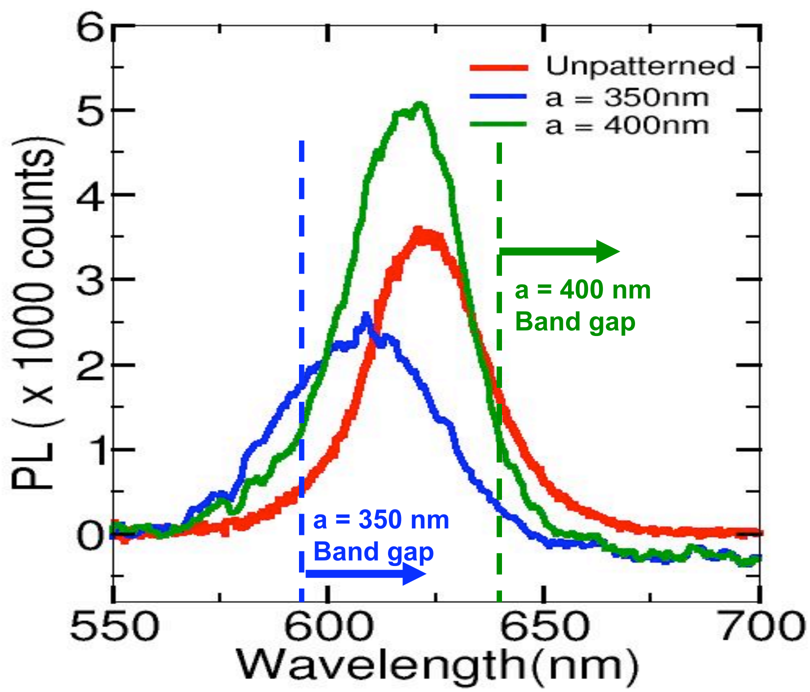
Rinse off unattached CdSe

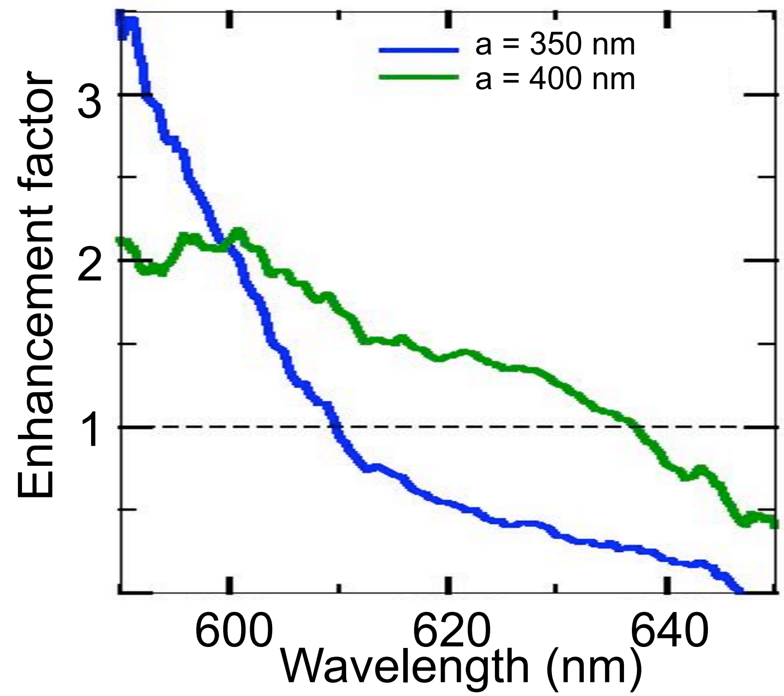


P. Guyot-Sionnest and C. Wang, J. Phys. Chem. B, 107, 7355 (2003)
J. Pacifico, D. Gomez, and P. Mulvaney, Adv. Mater. 17, 415 (2005)

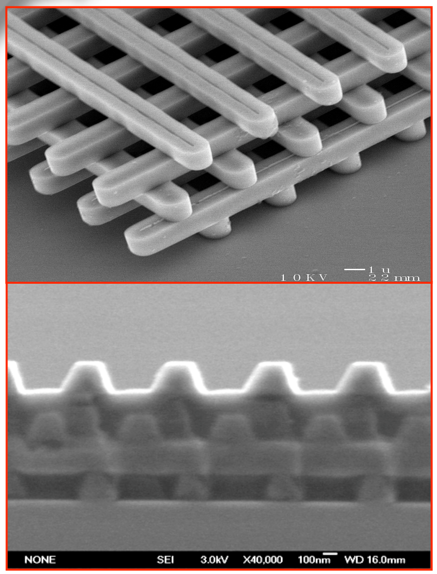
PL Response of CdSe QDs Inside the PC

- Incorporated CdSe quantum dots into TiO_2 3D logpile structures
- Developed Aerogel + quantum dot infiltration process
- PL is enhanced for 400 nm lattice constant (outside band gap)
- PL is suppressed for 350 nm lattice constant (inside band gap)
- 3D photonic crystal is effective at controlling photon emission

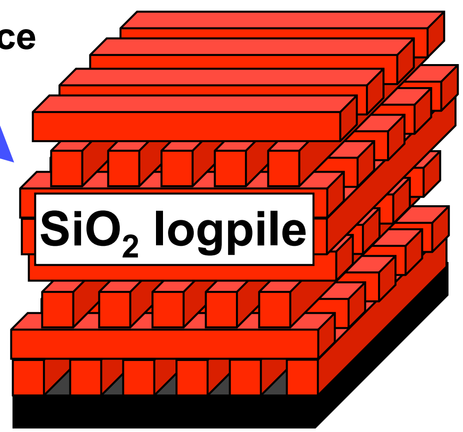




Fabrication of all GaN Vis-UV Logpile PC

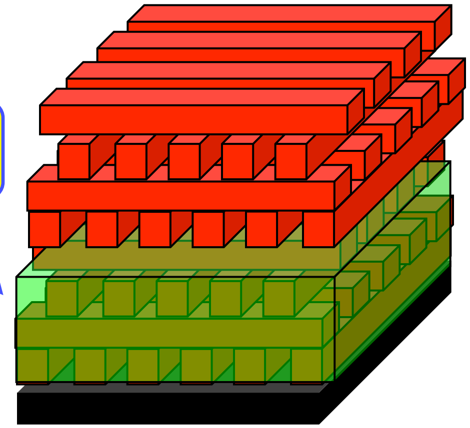


1. Fabricate Si logpile with SiO_2 background still in place



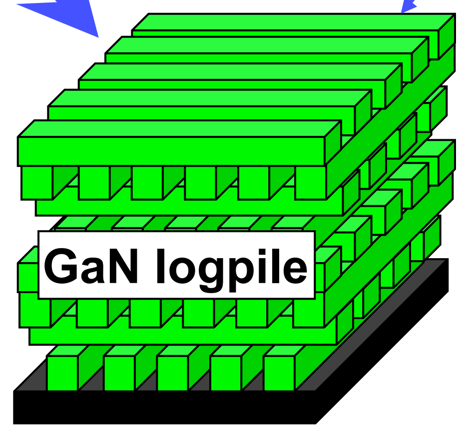
2. Etch out Si using KOH leaving SiO_2 skeleton in place

GaN grows uniformly throughout structure



3. Grow GaN by MOCVD

Transparent to 365nm
Index ~ 2.5



4. Remove SiO_2 template to leave behind GaN logpile

LDRD
LABORATORY DIRECTED RESEARCH & DEVELOPMENT

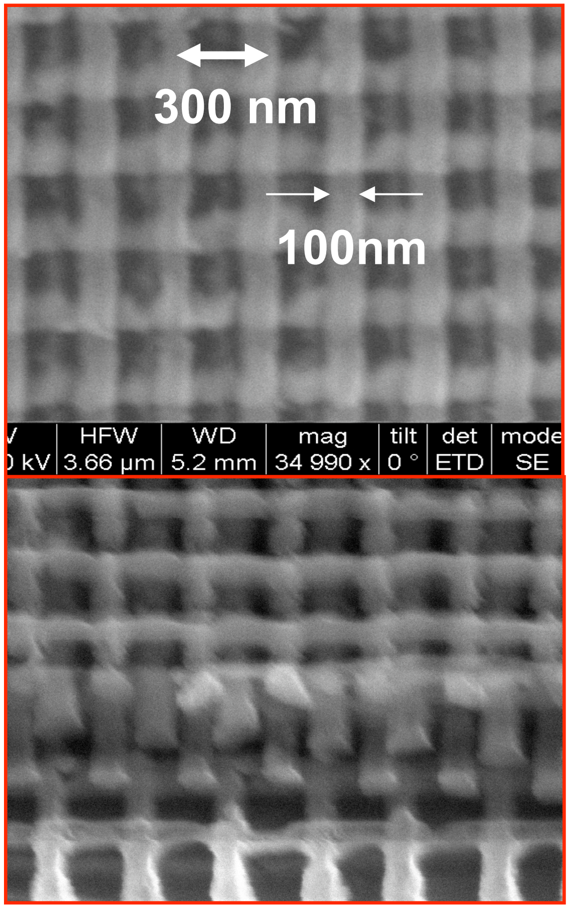
Sandia National Laboratories

All GaN Vis-UV Logpile Photonic Crystal

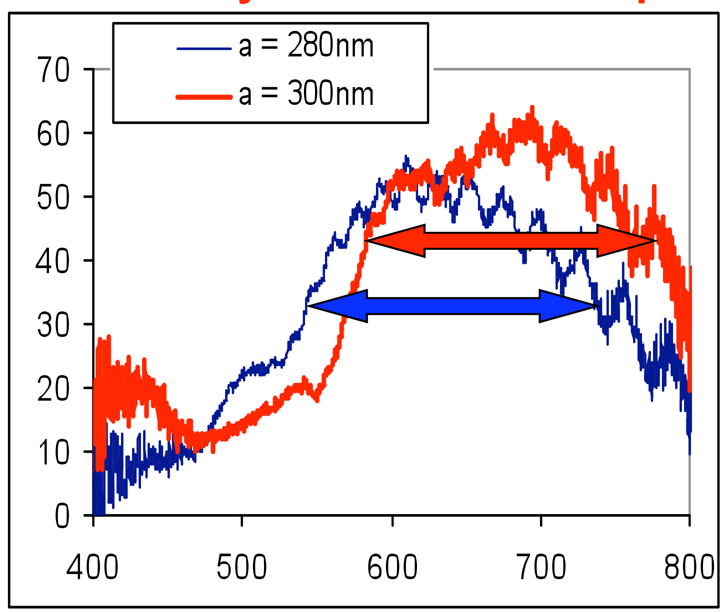
Implications of GaN logpile PC:

- Demonstrated all GaN 3D PC structure
- Important step forward for visible PCs
- Extremely good transparency across entire visible spectrum and into UV
- Potential to demonstrate shortest 3D PC
- Incorporate InGaN emitters, n-type and p-type
- Clear path to an electrically-injected 3D photonic crystal light source

SEM images of GaN PC



Reflectivity from GaN PC sample

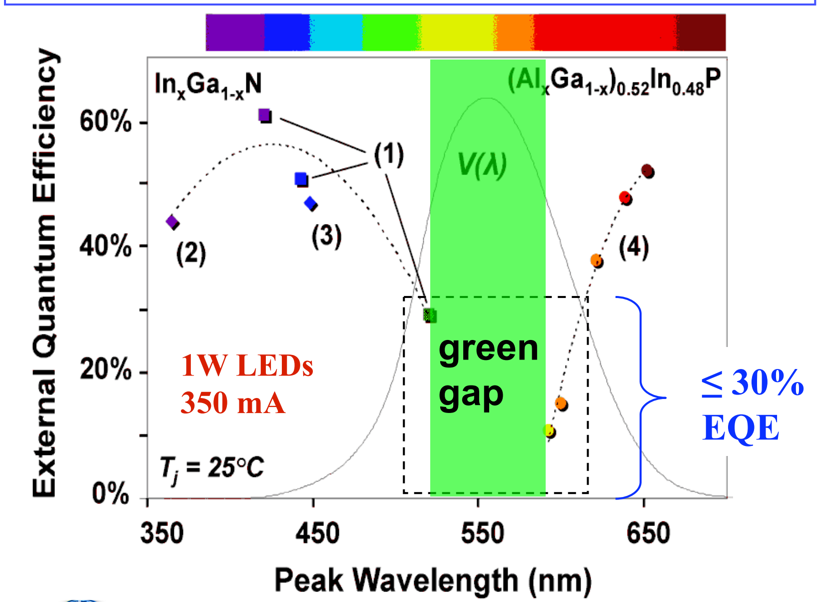


Impact of 3D photonic crystal research

Example Application Area:

→ **High efficiency lighting (SSL)**

- Efficiency gap between nitrides and phosphides (green gap)
- Utilize changes to photonic density of states to increase efficiency
- Increase radiative rate to better compete with non-radiative



Significance of Results

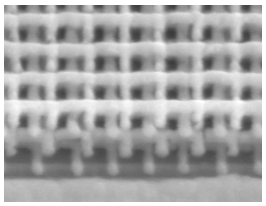
- Advance our understanding of interaction between semiconductor emitters and 3D photonic crystals.
- Develop new fabrication methods for visible 3D photonic crystals.
- Pave the way for investigation of exciting new areas of physics research:
  - Polariton dynamics, strong coupling
  - Solid state Bose-Einstein condensation
- Enable the development of advanced photon sources for applications such as optical and quantum computing.
- Control photon emission processes at an new and unprecedented level.
- Create solid state light sources with new functionality:
  - High Brightness/Efficiency
  - Single/Entangled Photon Sources
  - Directional, Compact
  - Anti-bunched Photons

Summary

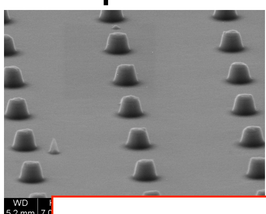
Accomplishments:

- Developed a method of fabricating 3D logpile photonic crystal structures made from TiO_2 for use in the visible.
- Incorporated GaN and quantum dot emitters in photonic crystal cavities and investigated luminescence properties.
- Demonstrated changes to luminescence intensity as well as radiative lifetime for emitters inside photonic crystal cavities
- Demonstrated control of light emission via altered photonic density of states.
- Developed a new method to fabricate UV/visible 3D photonic crystal structure made totally of GaN material.
- Funded follow-on project to investigate strong coupling and polariton condensates in GaN microcavities (BES-EFRC)
- 5 journal publications and 7 conference presentations

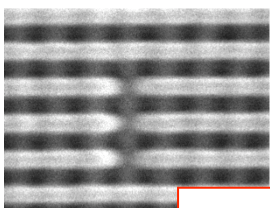
Visible 3D logpile



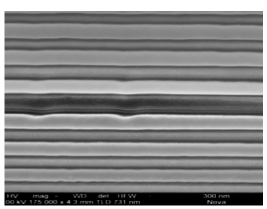
Nano-post LEDs



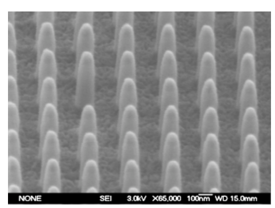
Defect cavities



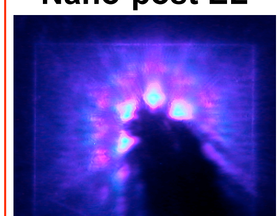
1D GaN cavity



Array of TiO_2 rods



Nano-post EL



Defect cavities