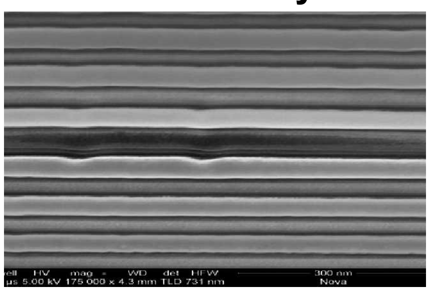
SAND2010-5215P

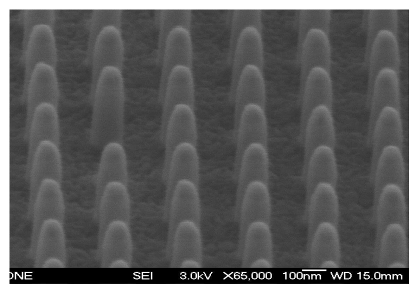
Enhanced Spontaneous Emission Using Visible 3D Logpile Photonic Crystal Structures

Arthur J. Fischer, Ganesh Subramania, Yun-Ju Lee, Qiming Li, George T. Wang, Daniel D. Koleske

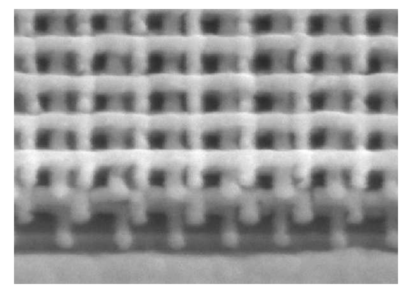
1D Photonic crystal



2D Photonic crystal



3D Photonic crystal



Work at Sandia National Laboratories was supported by Sandia's Solid-State-Lighting Science Energy Frontier Research Center, funded by the U.S. Department of Energy, Office of Basic Energy Sciences. 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.

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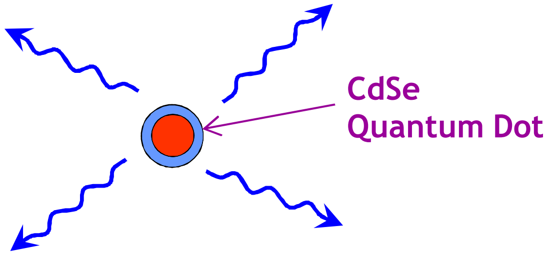
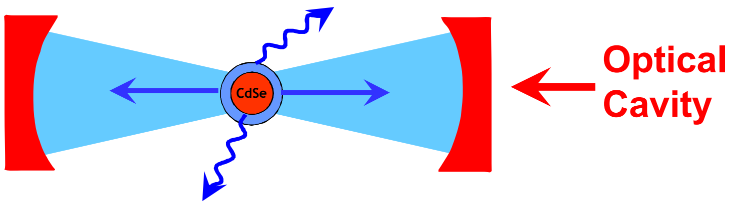
Photonic Density of States and Light Emission

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 (λ_{if})

Matrix Element (M_{if})

Density of Final States (ρ_f)

Purcell Effect:

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

n = index
g = degeneracy
λ = wavelength
Q = quality factor
V = mode volume

Ultimate control over when and if a photon is emitted !

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Light Control In Nature using Photonic Crystals

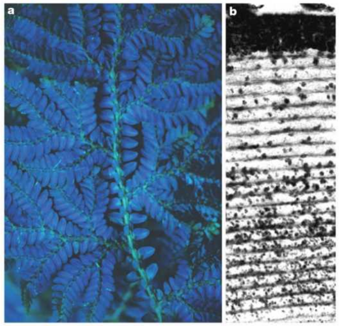
Morpho butterfly wings

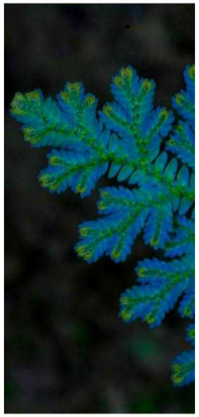
(M. menelaus) (M. sulkowskyi)

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

Opal stones

Fern leaves

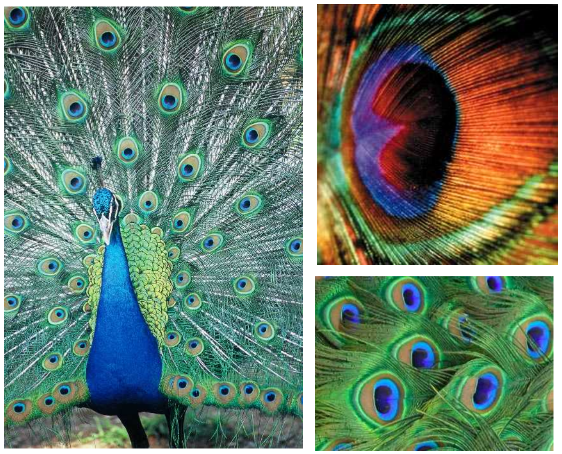




(Diplazium tomentosum)

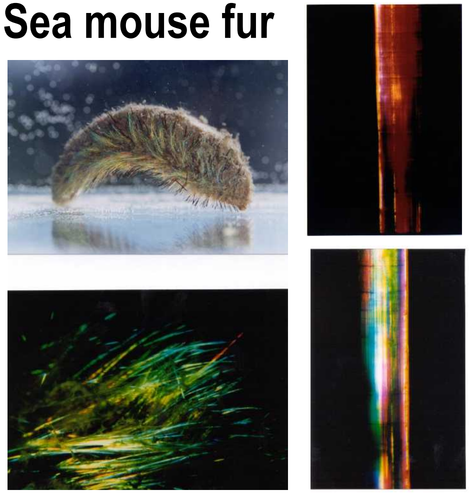
P. Vukusic et. al. , Nature **424,** 852 (2003)

Peacock feathers



Y. Li et al., Phys. Rev. E **72**, 010902 (2005)

Sea mouse fur



Dr. G. Rouse, School of Biological Sciences
University of Sydney

Beetles

(Chrysochroa vittata)

(Lamprocyphus augustus)

M.H. Bartl, Department of Chemistry, University of Utah

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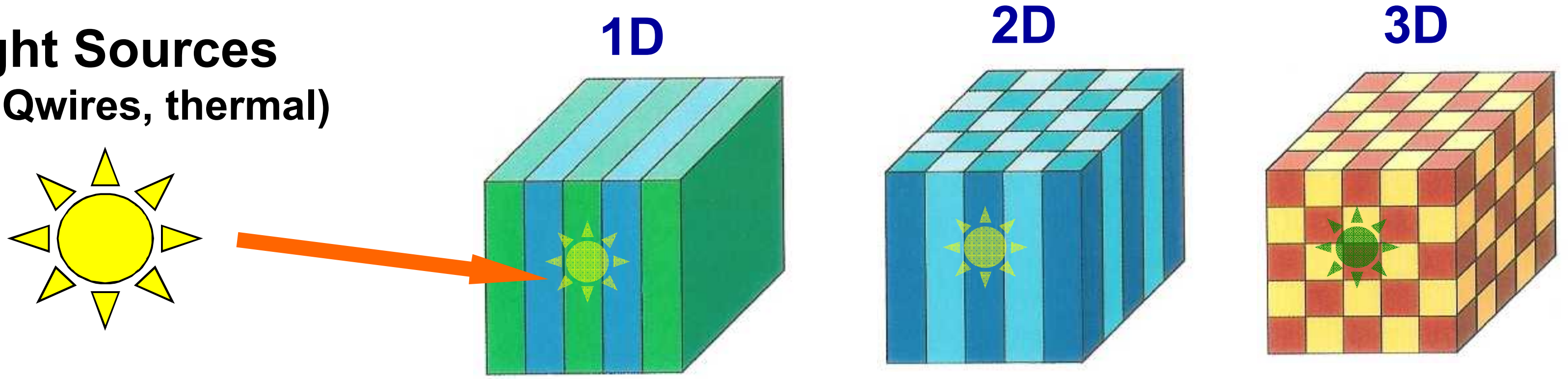
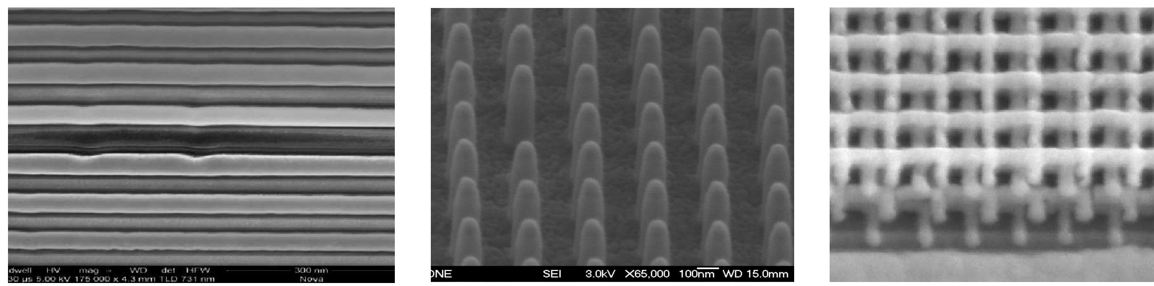
Light emission control with photonic crystals

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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3D photonic crystal: density of states

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

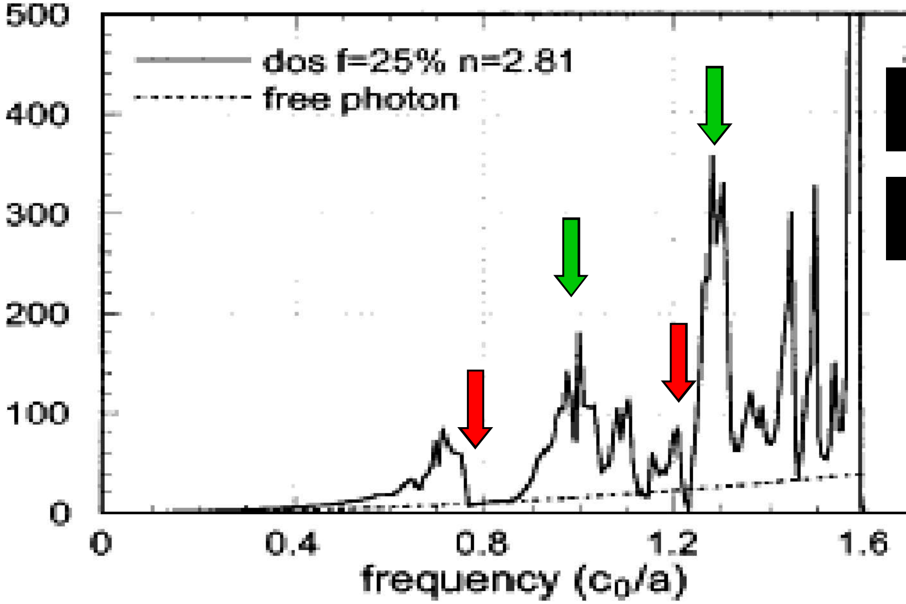
$$\Gamma_{\mathrm{rad}}(\boldsymbol{r}) = \frac{\pi\omega\mu^2}{3\hbar\epsilon_0} N_{\mathrm{rad}}(\boldsymbol{r},\, \boldsymbol{d},\, \omega)$$

Local radiative density of states (LRDOS)

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

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

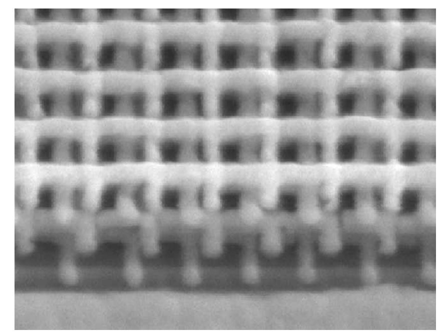
Photon Density of States in a 3D PC*



❖ 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)

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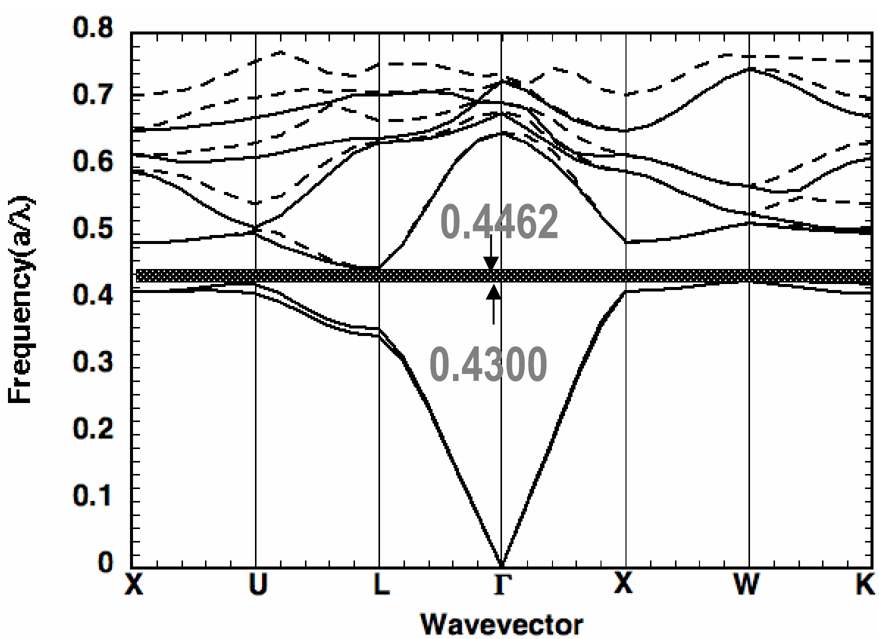
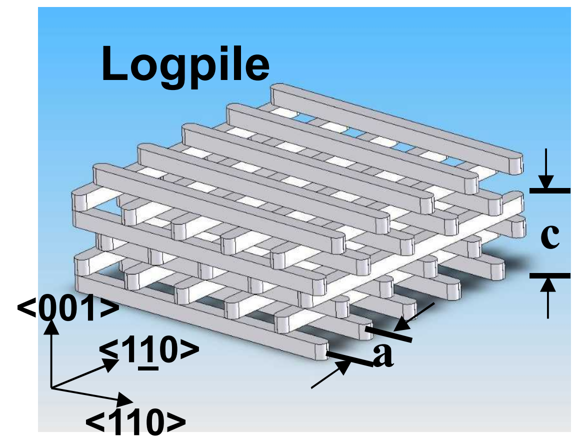
3D Photonic Crystals: Logpile Structure

- Dielectrics at visible frequencies have lower refractive indices (2.0-2.7) than in the near-IR
- Inverse Opal not suitable for use in the visible
- Logpile geometry enables complete 3D gap with n>1.9*
- TiO_2 : Suitable for visible PC/ low loss high index (2.3-2.7)





- 4% bandgap
- ~ 25nm $\delta\lambda$ at λ ~ 600nm

* K.M.Ho et.al. , Solid State Comm., **89** ,413(1994)

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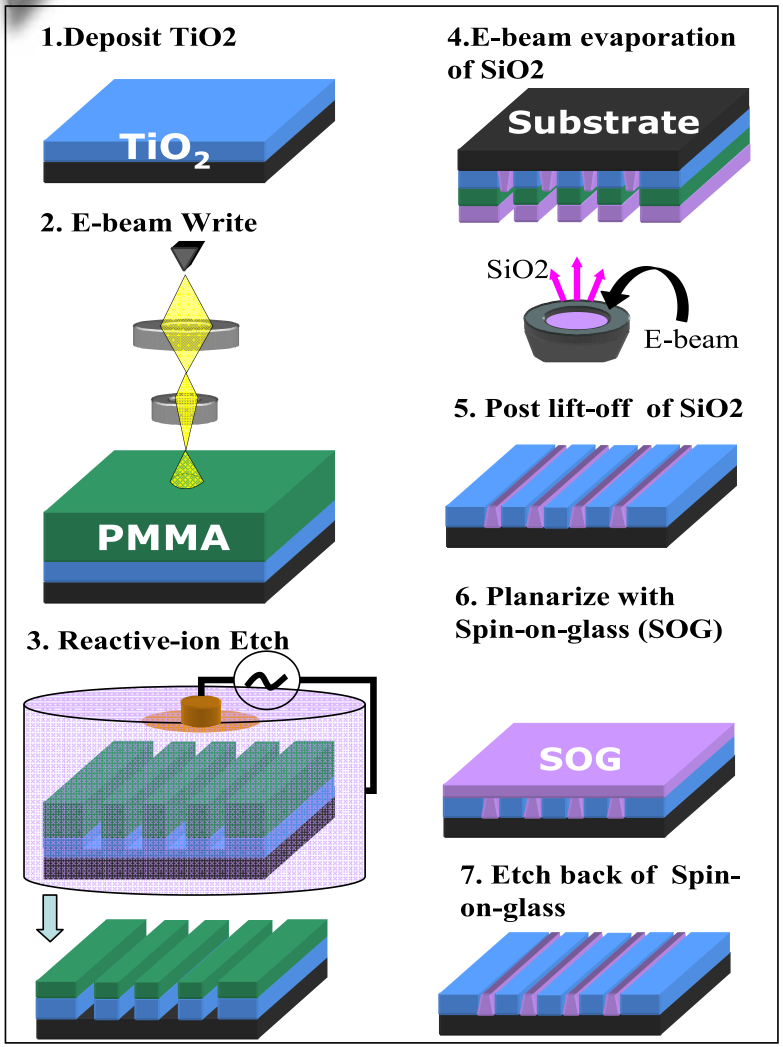
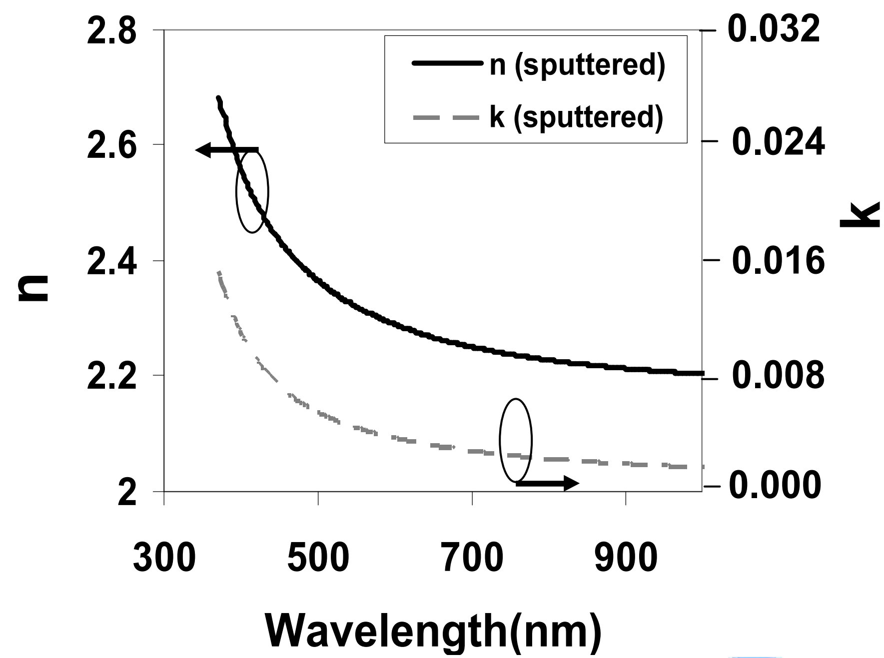
Logpile PC Fabrication: Sputtered TiO_2

Fabrication steps for one layer



- Thicker e-beam resist (PMMA) is required due poor etch selectivity of TiO_2
- Thicker PMMA results in poor dose control to obtain rod width uniformity between layers
- Reasonable index at visible wavelengths



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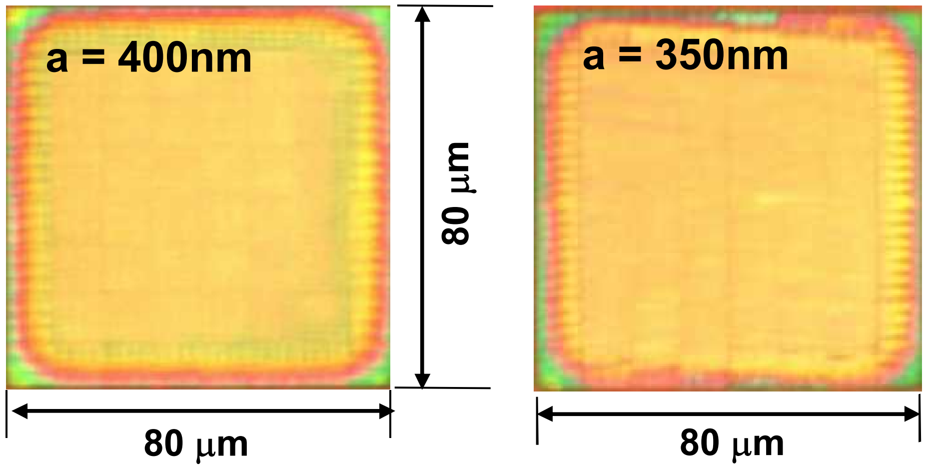
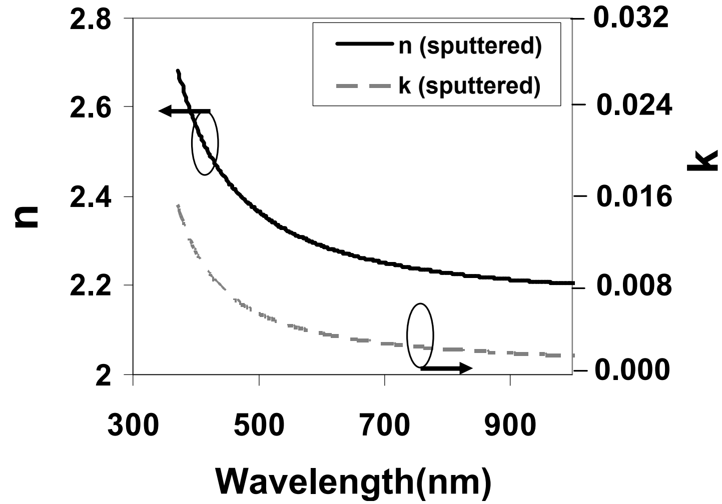
Visible TiO_2 Logpile 3D Photonic Crystals

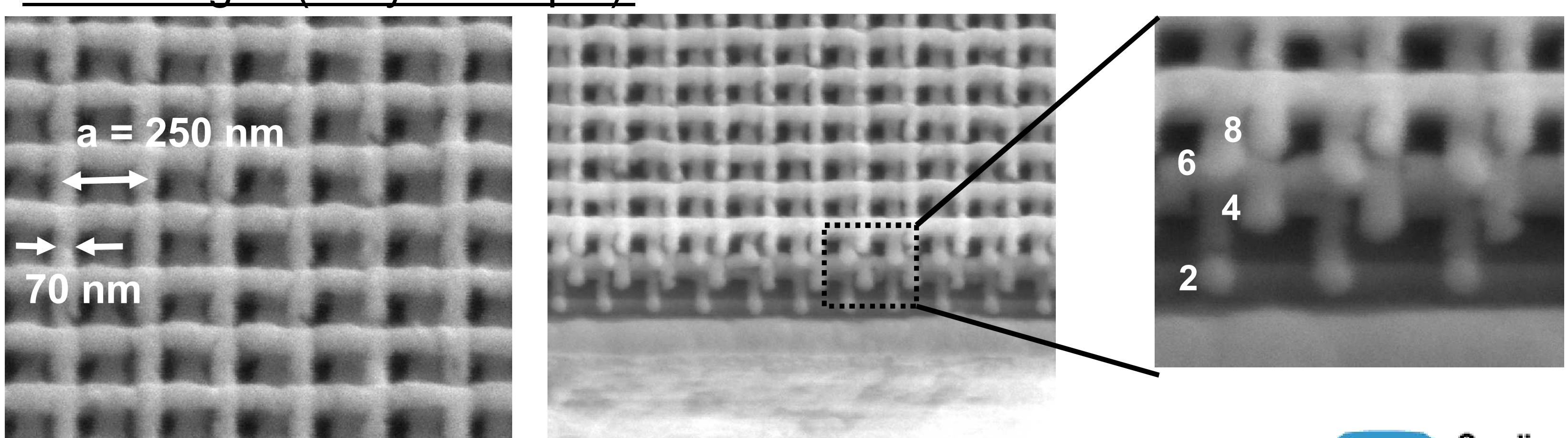
White light microscope images (5 layer sample)



TiO_2 index of refraction

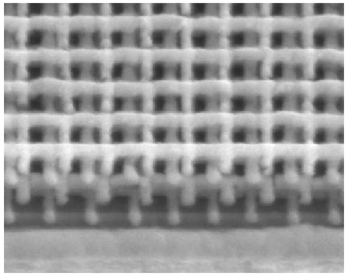


SEM images (9 layer sample)



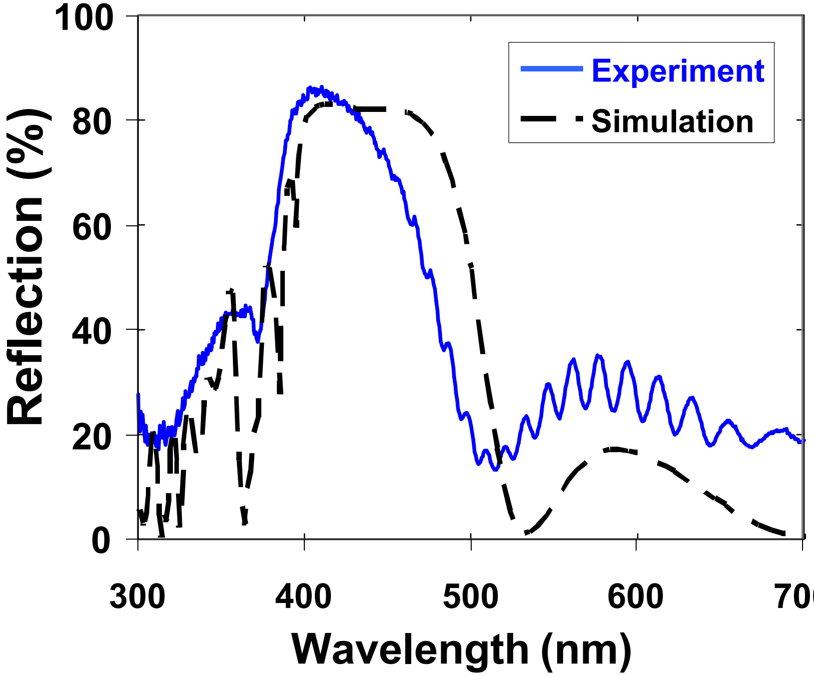
Reflectance from 9-Layer Visible TiO_2 Logpile

SEM - 9 layer logpile

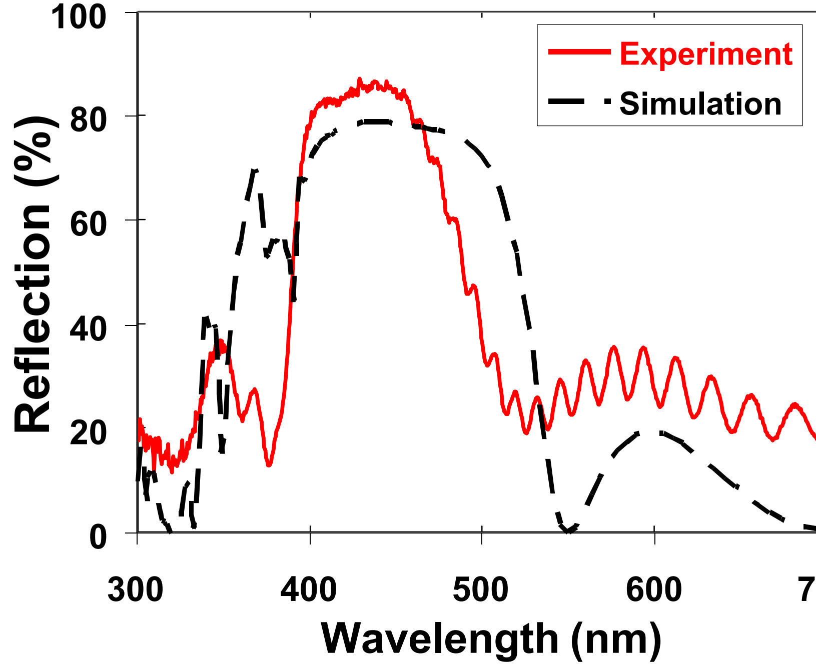


- Near normal incident reflection data
- Nine layer structure with lattice constant a=250 nm
- Bandgap extends from 400 to 500 nm
- Reflection greater than 80% → excellent morphology
- Demonstrates that TiO_2 PCs can be used at 400 nm

Reflection- Parallel Polarization



Reflection- Perpendicular Polarization



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3D photonic crystal ‘air defect’ microcavity

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

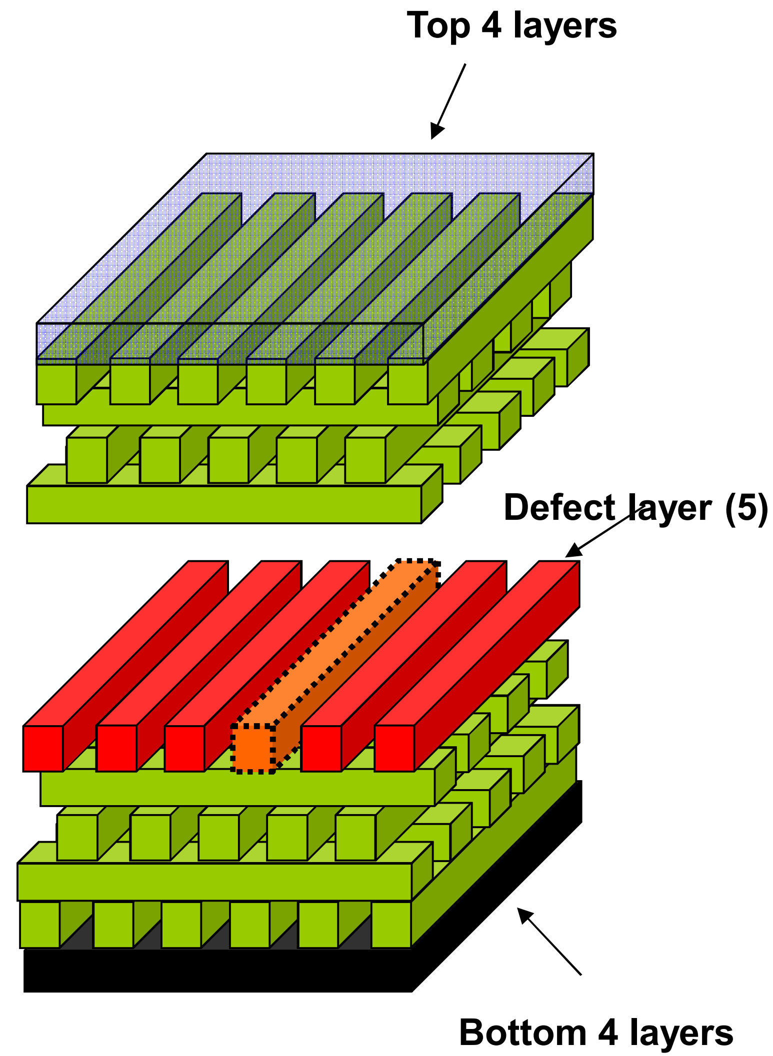
• Portions of rods removed to create ‘air’ defect cavities

• 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



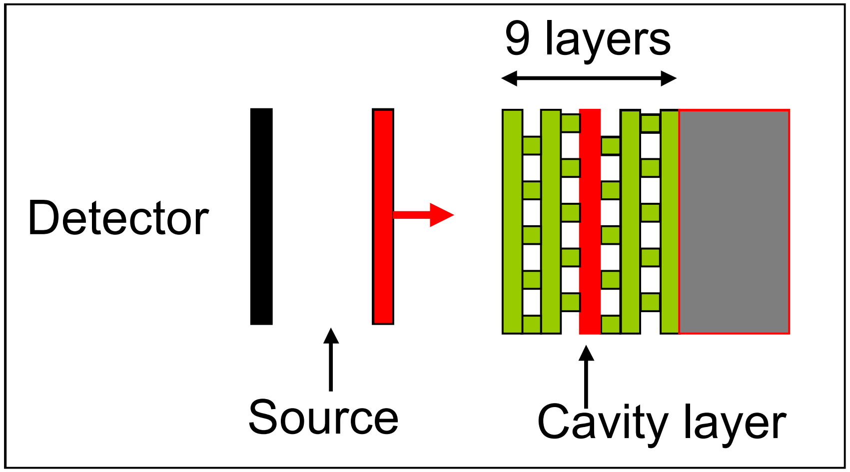
FDTD modeling of microcavity reflectance

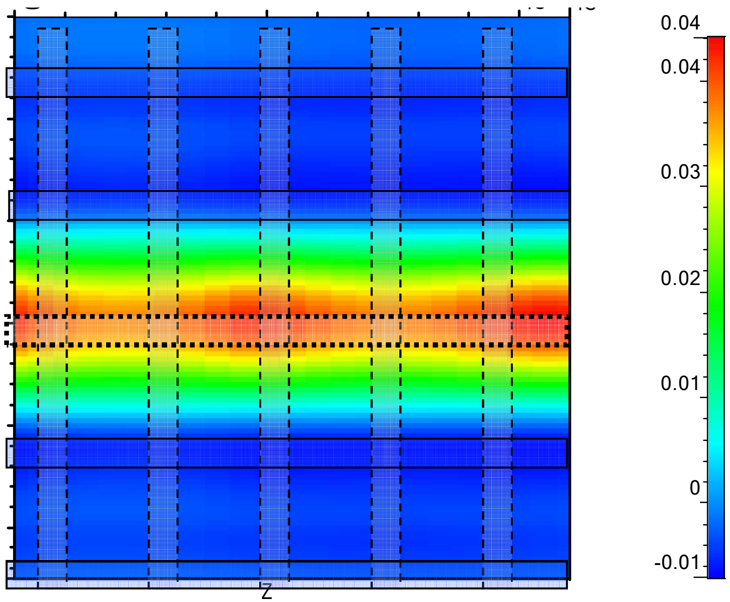
FDTD conditions:

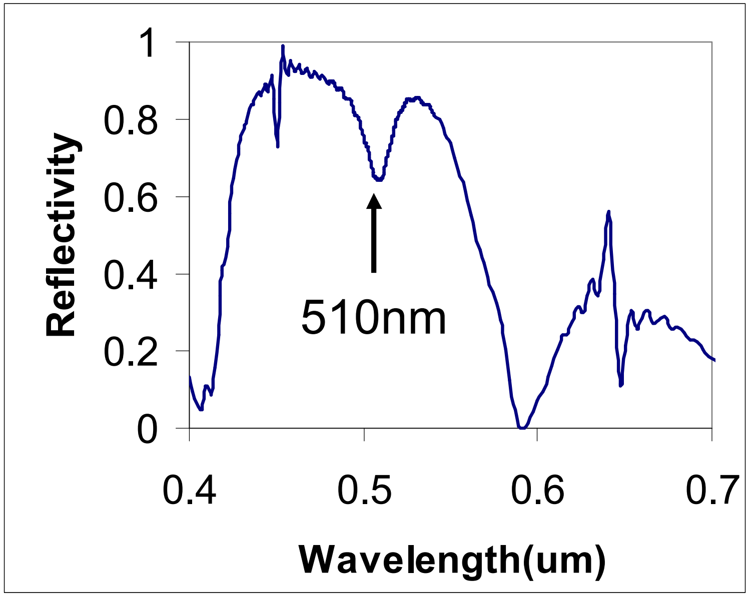
- Supercell: 5 unit cells
- Cavity defect periodic with 5 unit cells
- Periodic boundary conditions in the transverse
- Gaussian source

Cavity A: Every 5th rod removed

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







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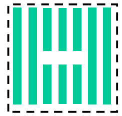
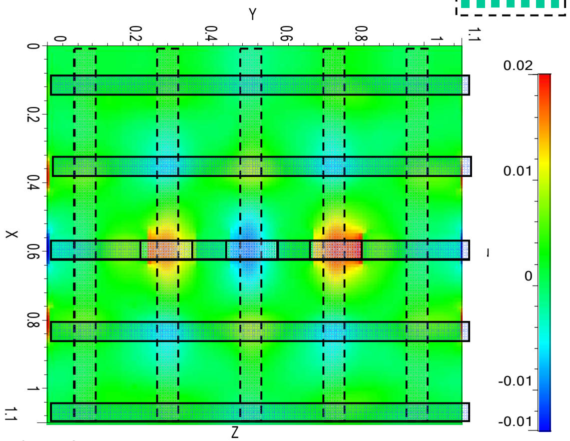
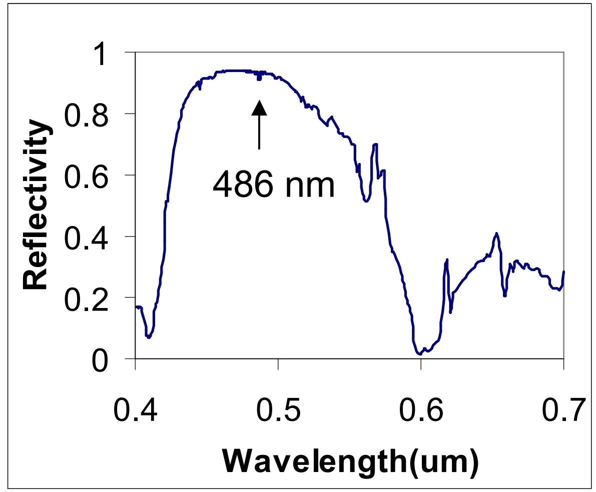
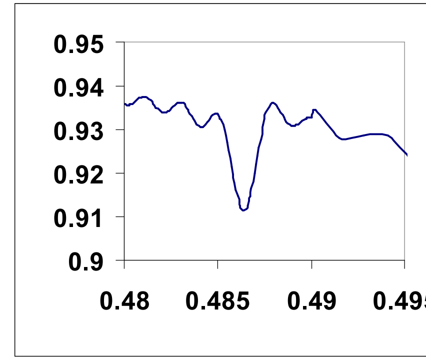
FDTD modeling of microcavity reflectance

Cavity B: →



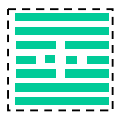


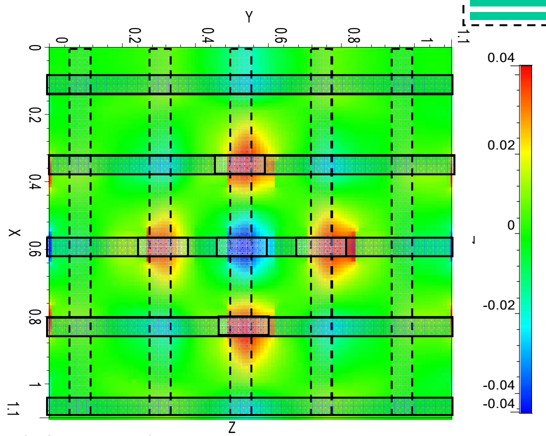


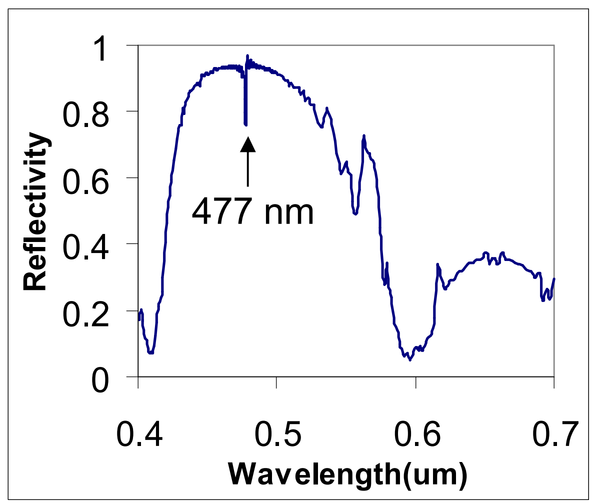


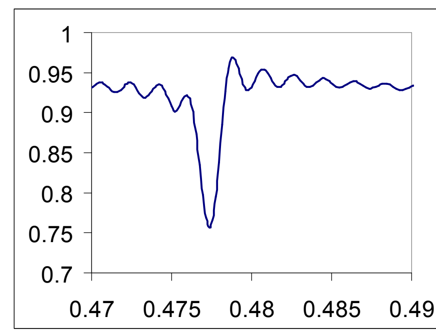
- Q ~ 250
- MV : 0.25(λ/n)^3

Cavity C: →





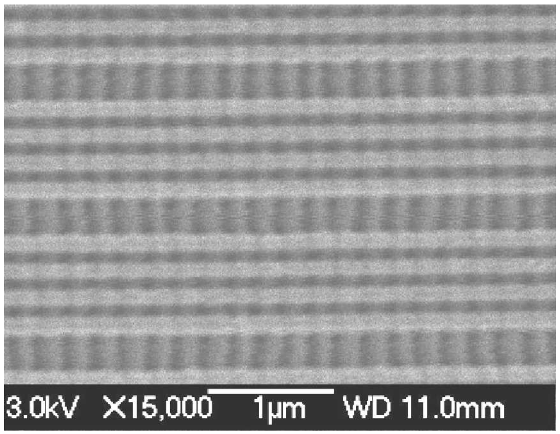


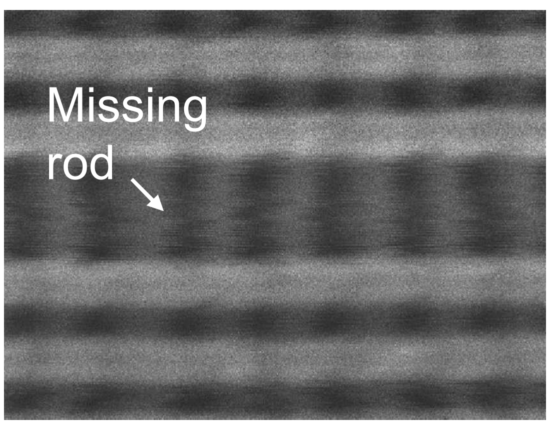


- Q ~ 300
- MV : 0.42(λ/n)^3

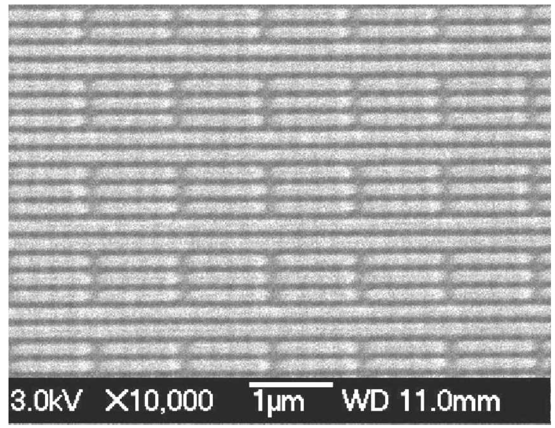
Fabricated TiO_2 Photonic Crystal Cavities

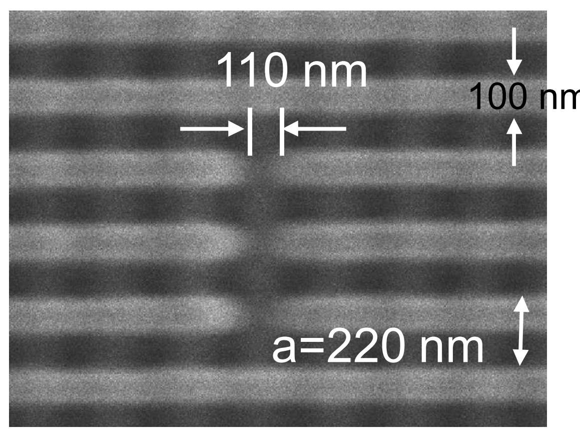
Cavity A



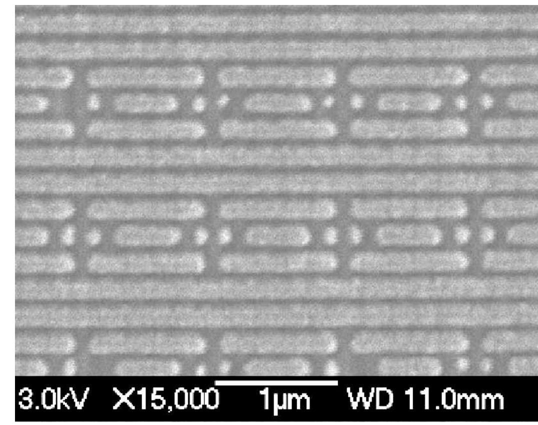


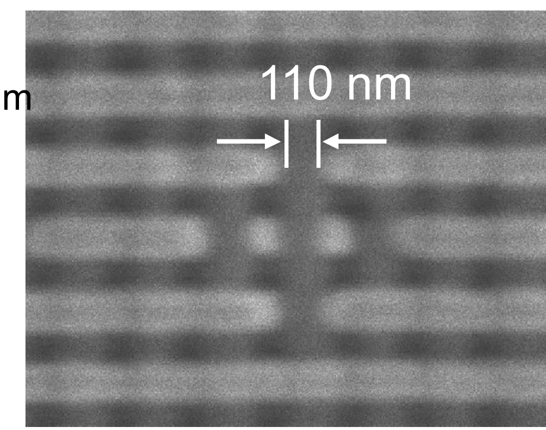
Cavity B





Cavity C





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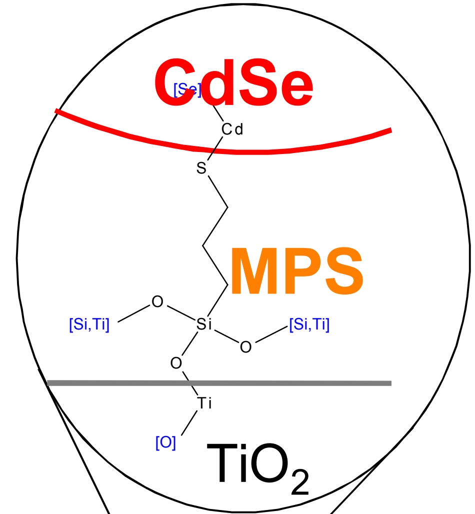
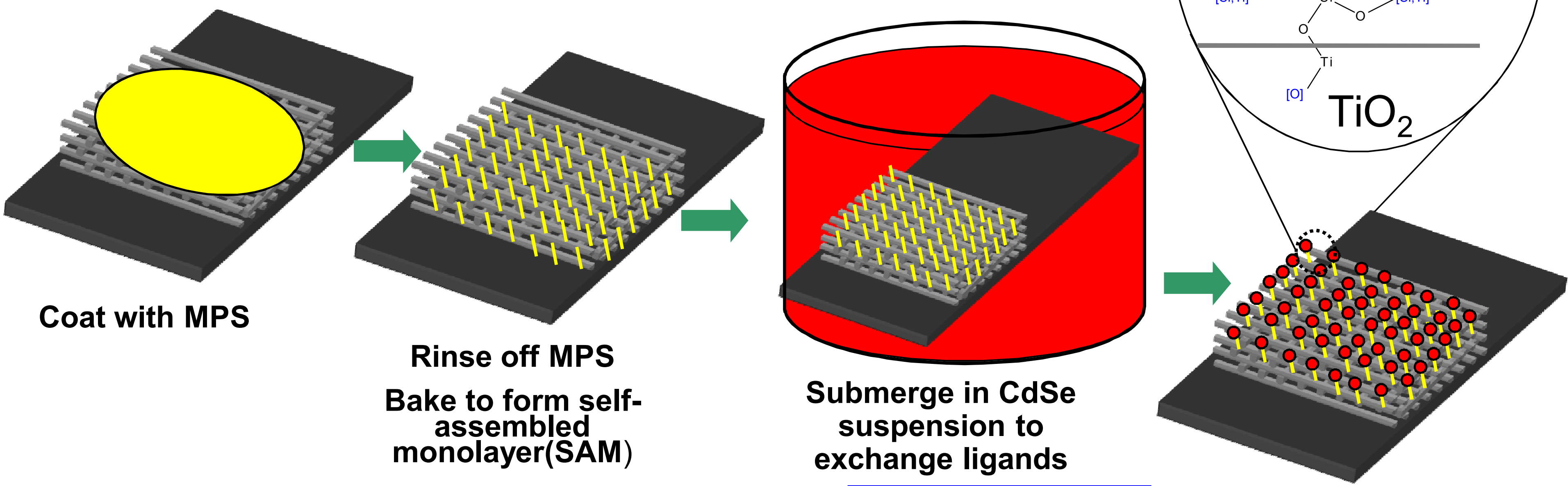
Self-assembled Monolayer of CdSe on TiO_2

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)





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)

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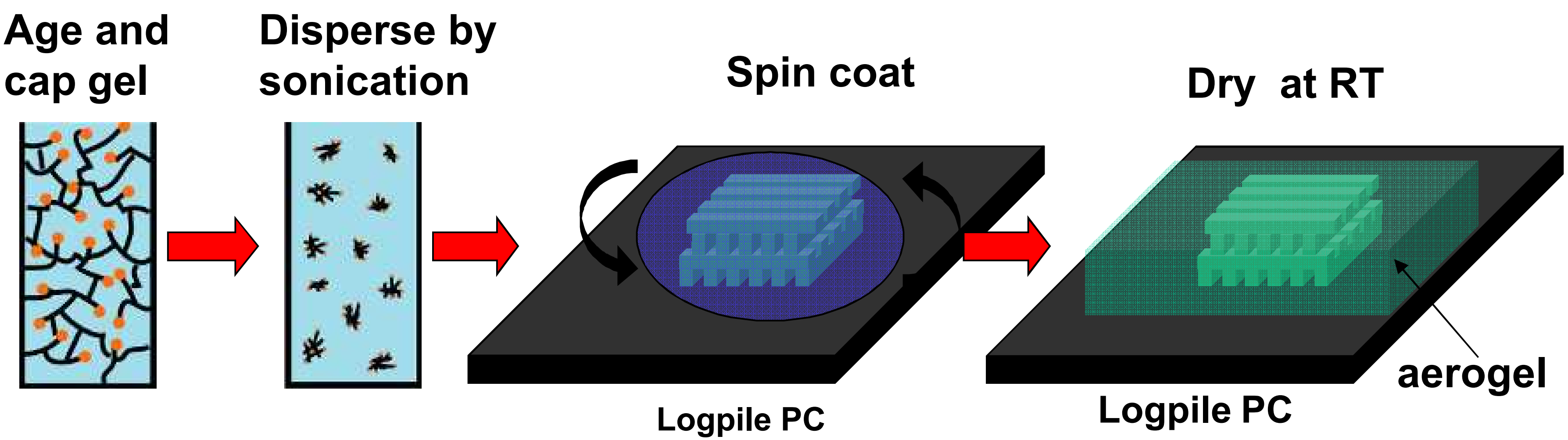
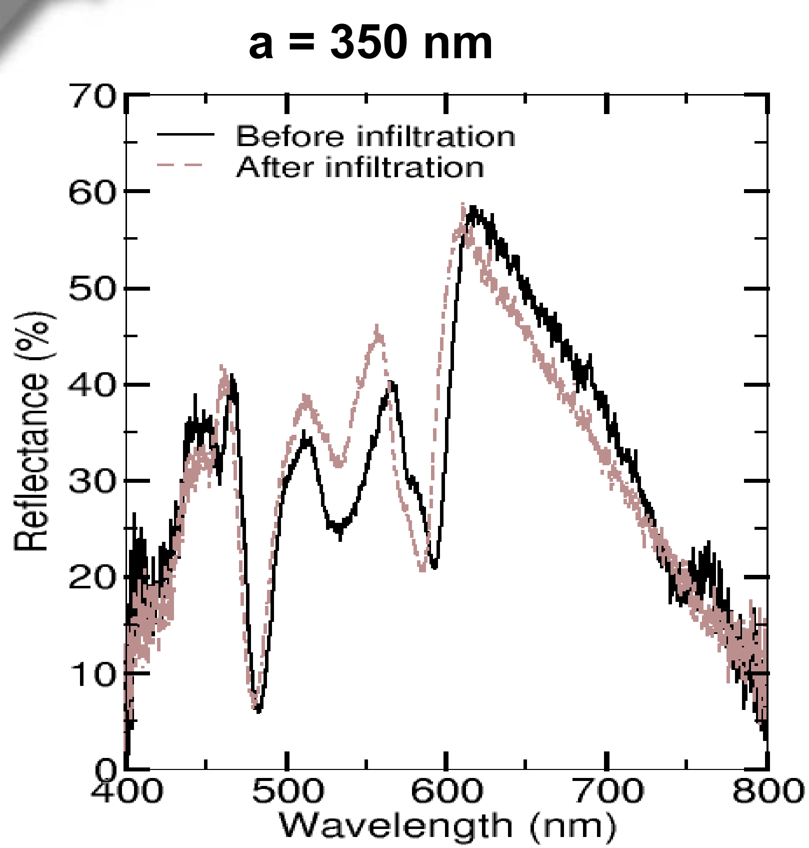
Improving QD uniformity using Silica Aerogel

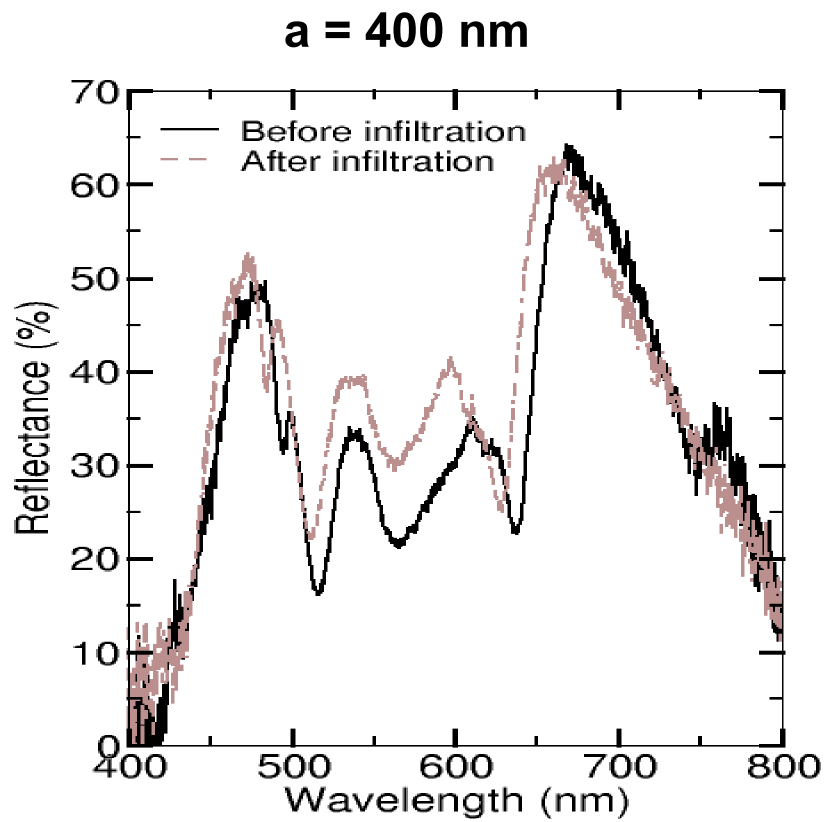
- High surface to volume ratio of silica aerogel
  - Increased surface area for QD attachment
  - Higher probability of overlap of the QD emission with PC LDOS
  - Potentially minimal disturbance of the bandstructure
- Sol-gel approach requiring no special drying procedure
  - Capping agent halts condensation, allows spring-back after drying
- **Successfully incorporated QDs into 3D logpile structures**



S. S. Prakash et al., Nature 374, 439 (1995)

Effect On Bandgap Due To Aerogel + CdSe Infiltration





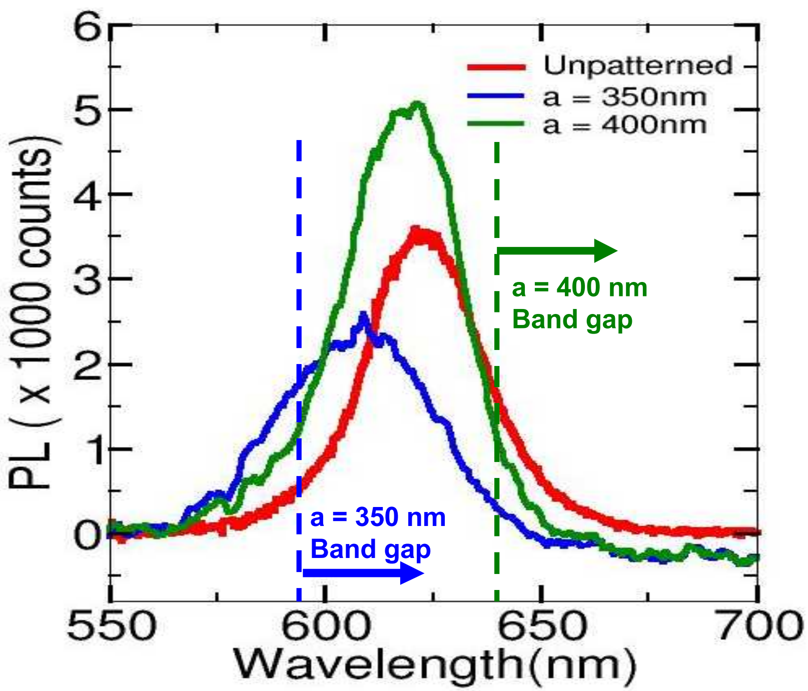
For rod filling fraction f ~ 0.4±0.05 ; n_{aero} ~ 1.1±0.05

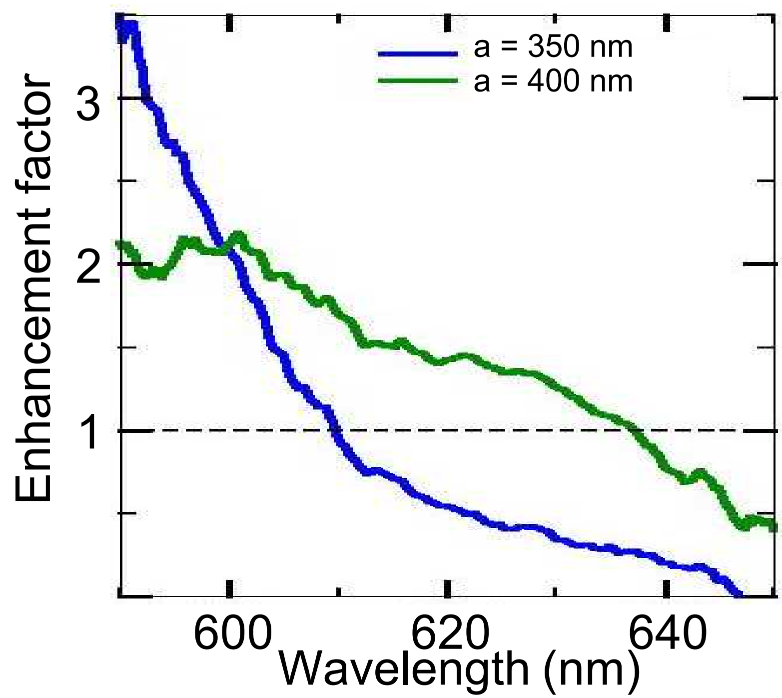
Minimal effect on the bandgap!

G. Subramania et al., Appl. Phys. Lett. **95,** 151101 (2009).

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





G. Subramania et al., Appl. Phys. Lett. **95,** 151101 (2009).

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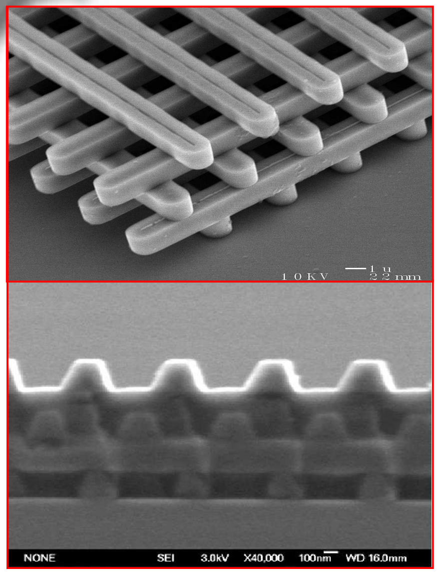
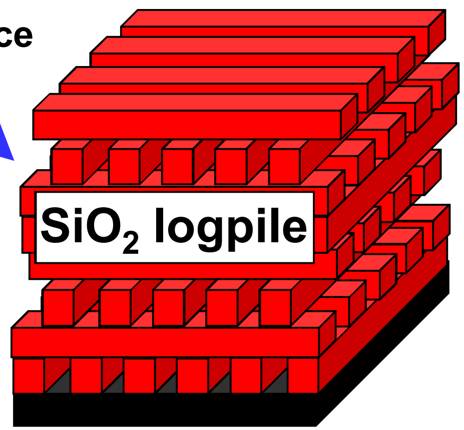
Fabrication of all GaN Vis-UV Logpile PC

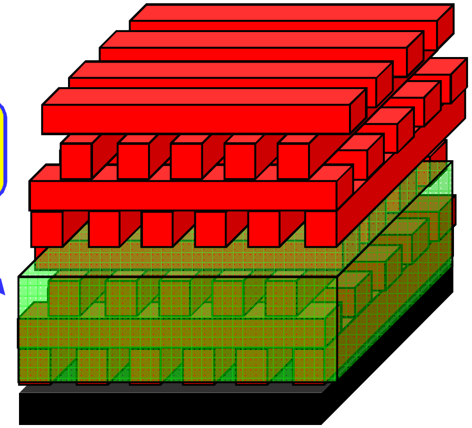


1. Fabricate Si logpile with SiO_2 background still in place



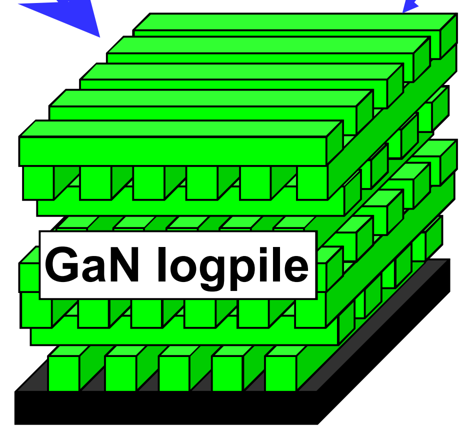
2. Etch out Si using KOH leaving SiO_2 skeleton in place

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

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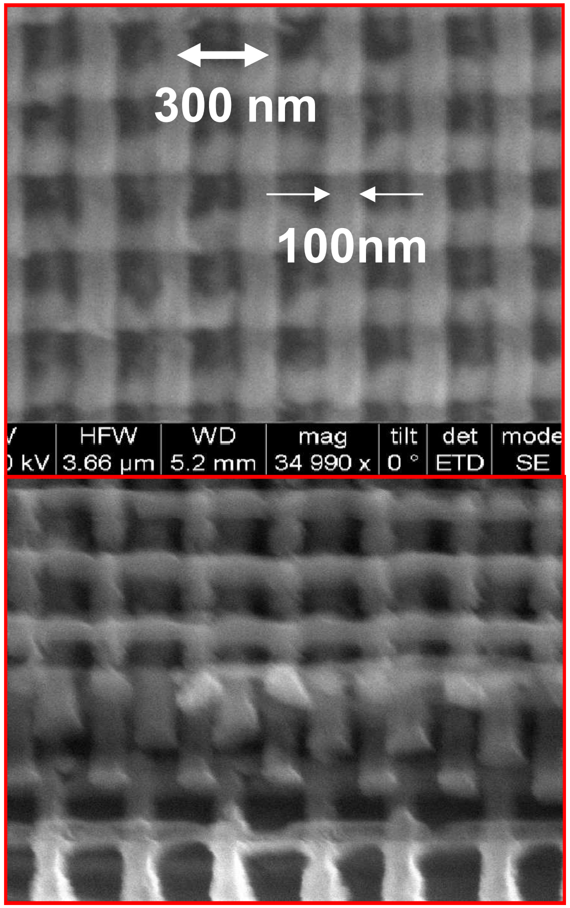
All GaN Vis-UV Logpile Photonic Crystal

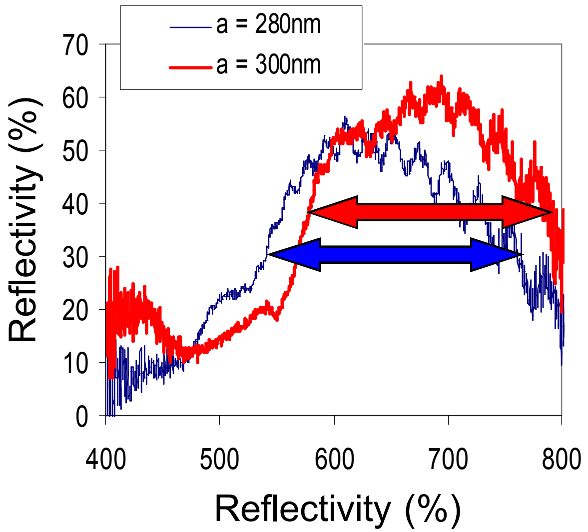
Implications of GaN logpile PC:

- **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**

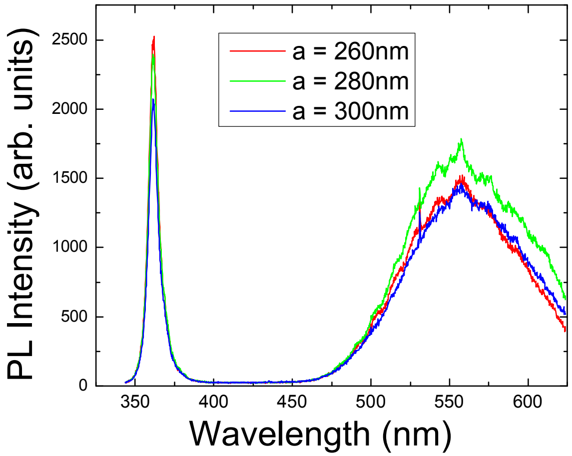
SEM images of GaN PC



Reflectivity from GaN PC sample



PL from GaN PC sample



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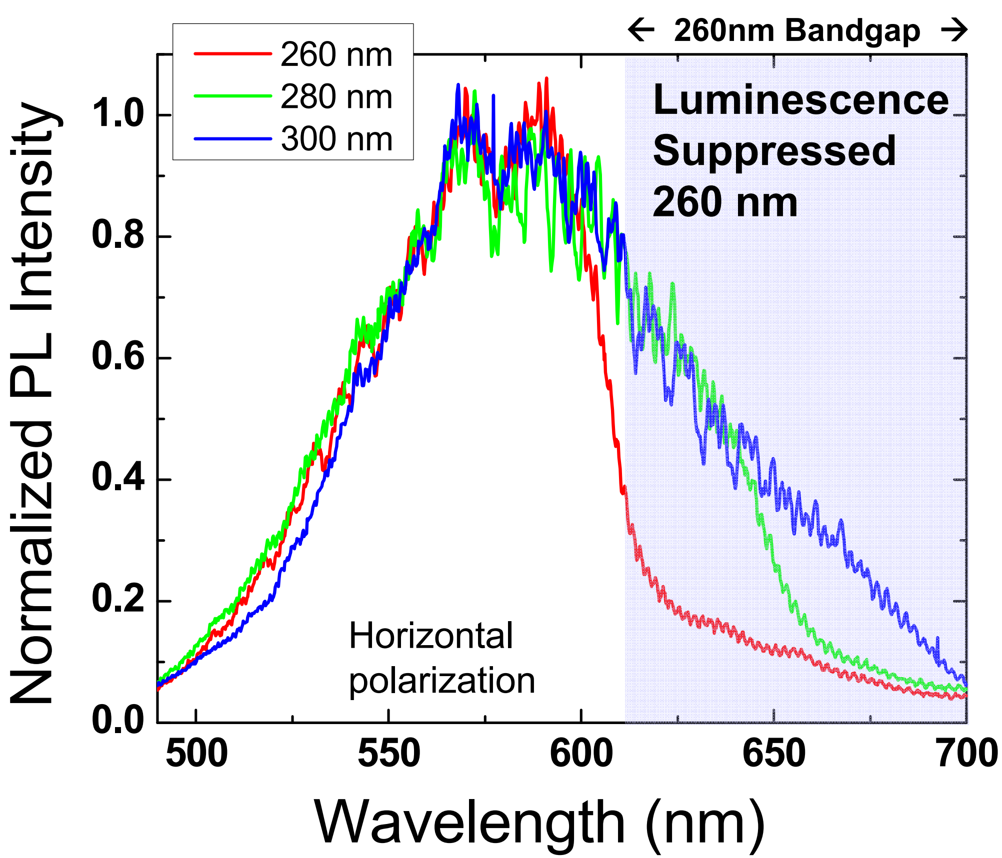
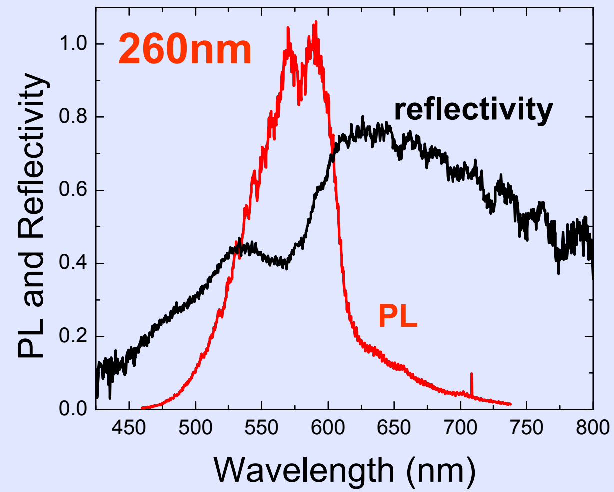
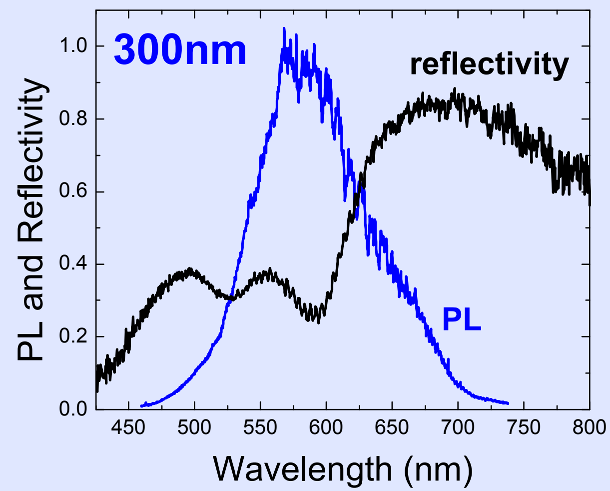
TIPL data from 5 layer GaN logpile: oxide removed

- Focus on GaN yellow-band emission (570nm)
- PL change only for horizontal polarization
- 5 layer logpile with a = 260nm, 280nm, 300nm
- Largest effect for 260 nm lattice constant
- **PL suppressed inside band gap region**



PL vs. Reflectivity





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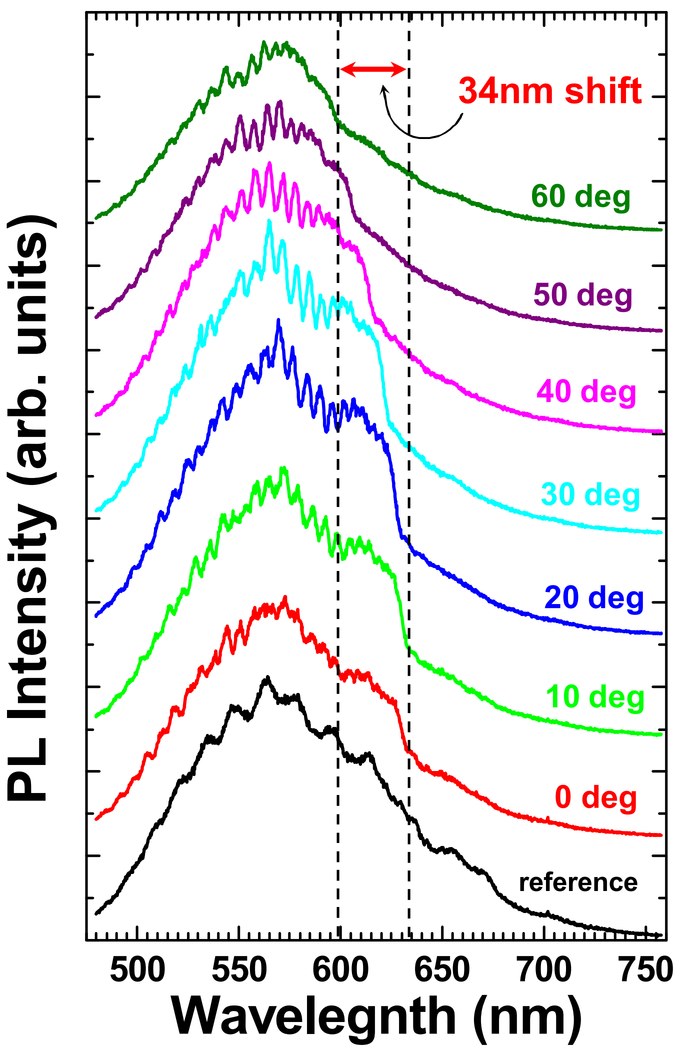
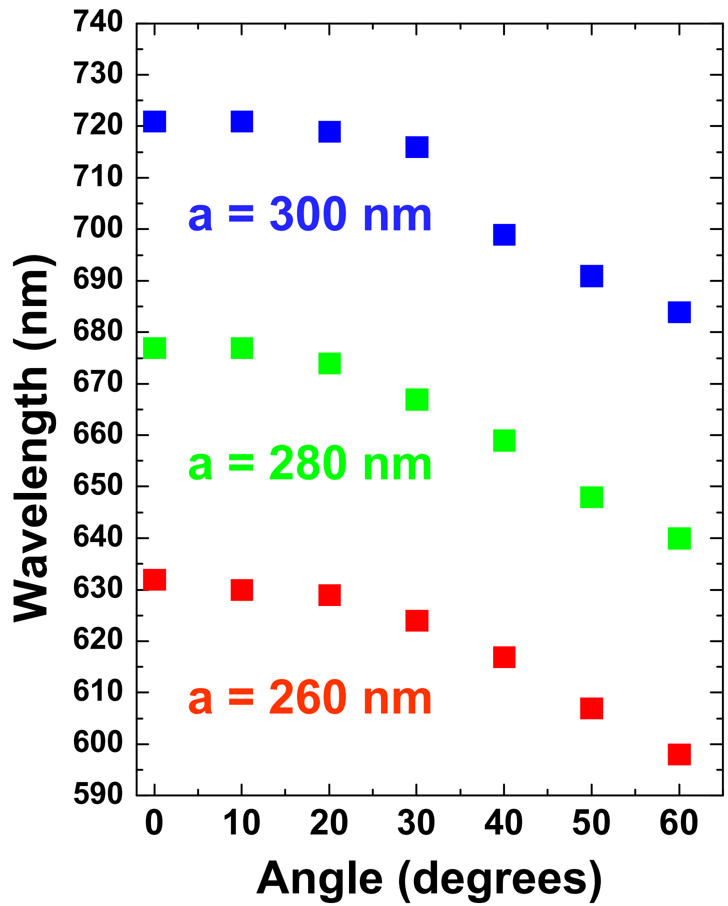
Angle dependent TIPL data from 5 layer GaN logpile: oxide removed

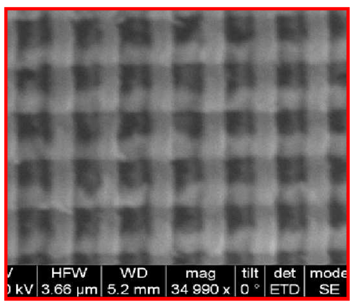
- Collect PL into a 2 degree solid angle
- Horizontal polarization (TM) shows bigger effect
- Band edge shifts with angle (~ 35 nm shift)
- Gives further evidence for GaN emission modification
- Need to compare to angle-dependent reflectivity

260nm lattice constant

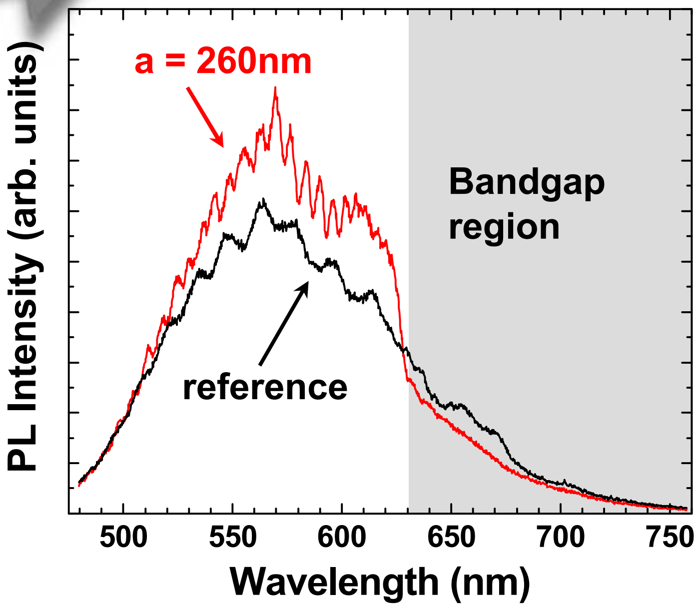




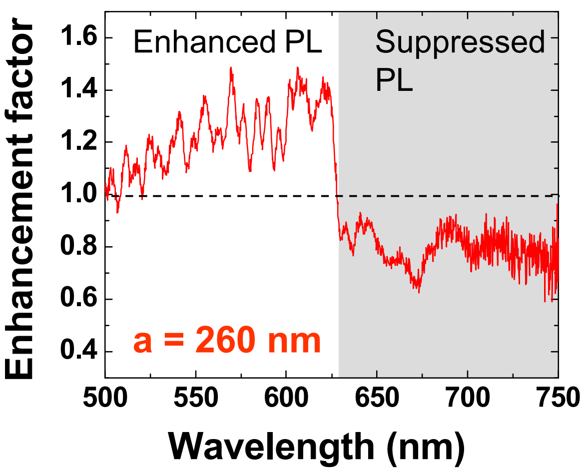
GaN 3D Logpile

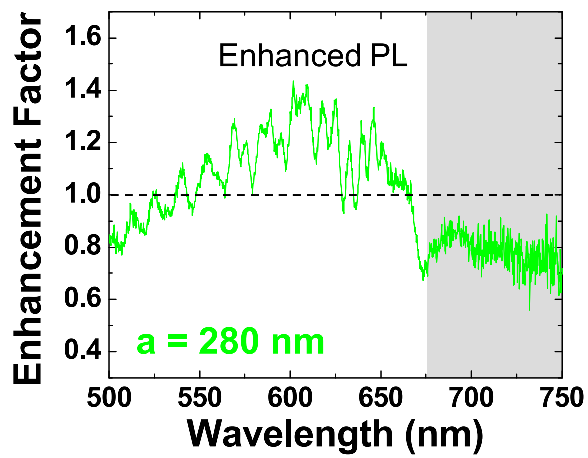


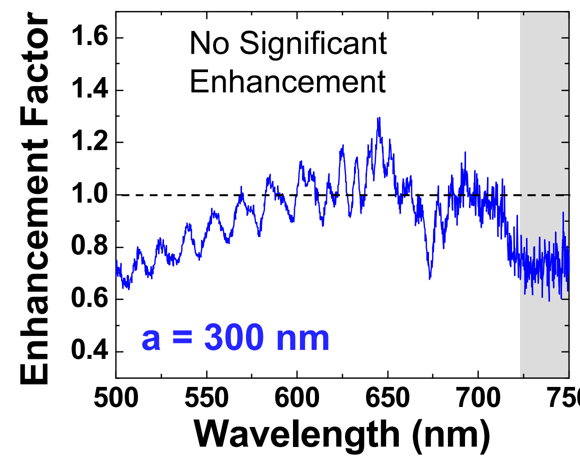
Angle dependent TIPL data from 5 layer GaN logpile: oxide removed



- Intensity comparison shows both enhanced and suppressed emission
- Reference curve is a representative GaN yellow band peak with characteristic lineshape
- Data shown for emission 20 degrees w.r.t. normal
- Horizontal polarization (TM) shows larger effect
- Emission enhancement as large at 1.4 X
- 20% reduction in PL intensity inside bandgap
- No significant effect for 300nm lattice constant







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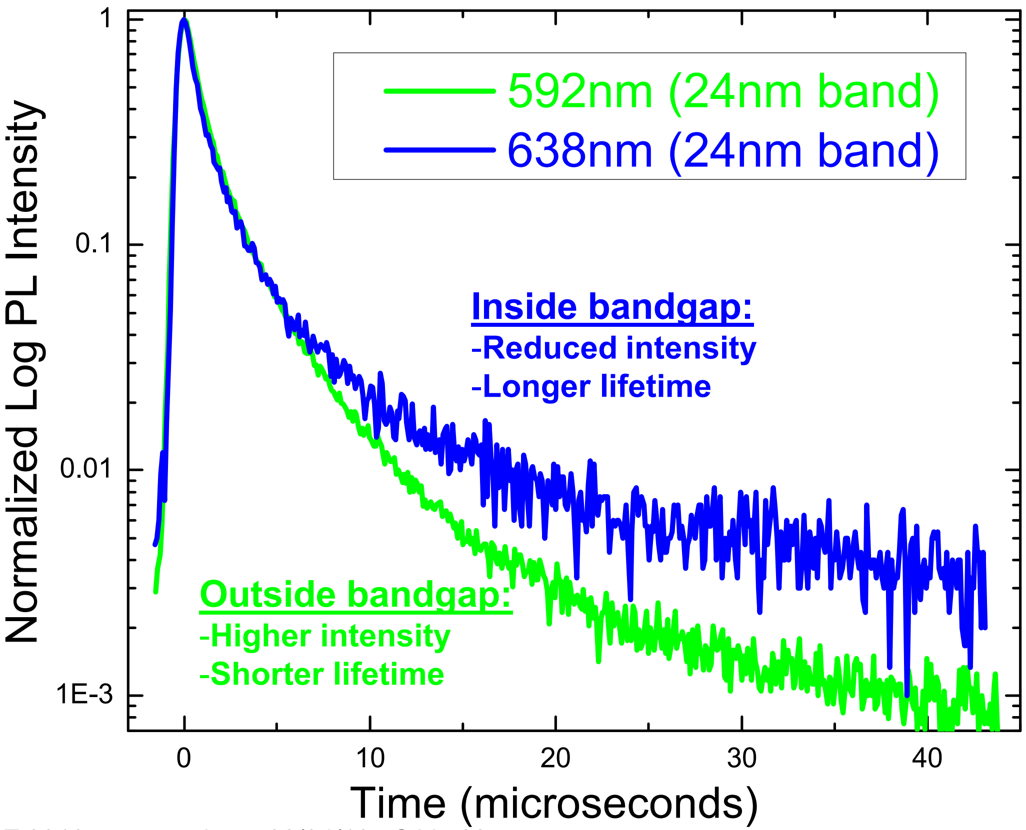
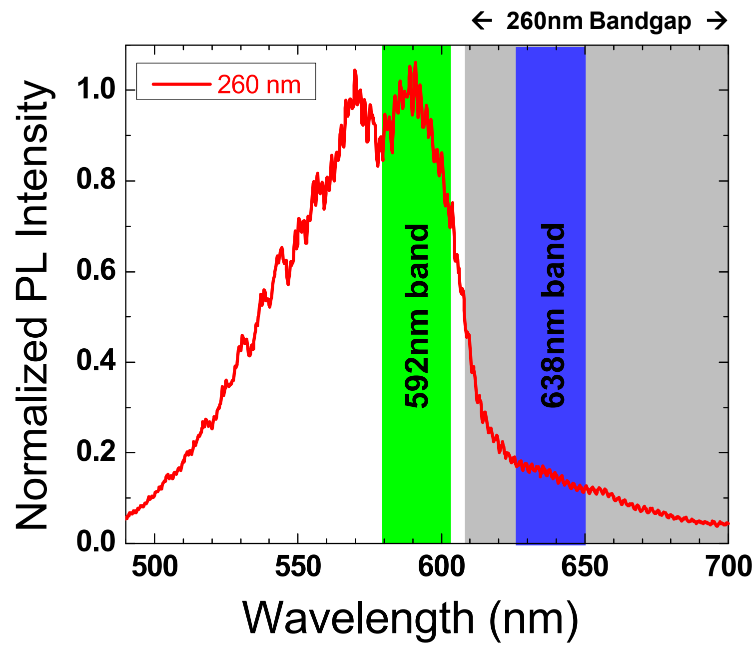
TRPL data from 5 layer GaN logpile: oxide removed

- Focus on 260nm lattice constant sample
- Small lifetime change for horizontal polarization only
- 100 fs pump at 340nm, streak camera with 15ps resolution
- **Longer lifetime and reduced intensity inside bandgap region**
- **Shows PL suppression is more than a passive effect**
- Larger change expected for emitters fully inside PC structure





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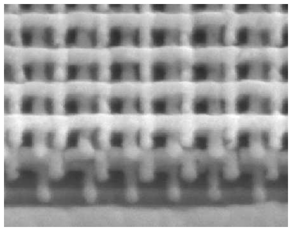
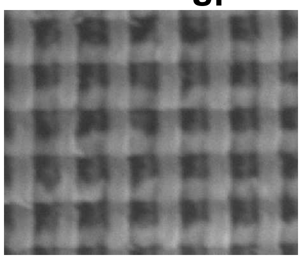
Summary

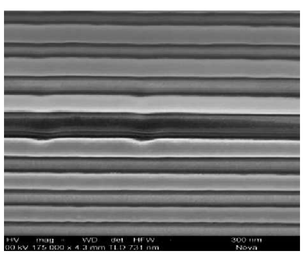
TiO$_2$ 3D logpile



GaN 3D logpile



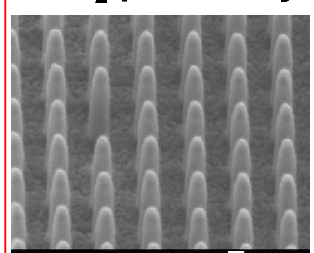
1D GaN cavity



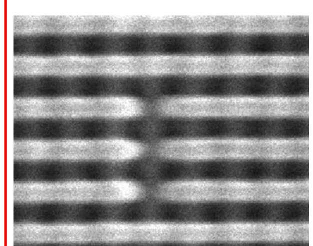
Accomplishments:

- Developed a method of fabricating 3D logpile photonic crystal structures made from TiO$_2$ for use in the visible.
- Incorporated quantum dot emitters in photonic crystal cavities and investigated luminescence properties.
- Demonstrated changes to luminescence intensity for TiO$_2$ 3D logpile structures.
- Developed a new method to fabricate UV/visible 3D photonic crystal structure made totally of GaN material.
- Demonstrated changes to luminescence intensity and lifetime for GaN 3D logpile structures.
- Demonstrated control of light emission via altered photonic density of states.

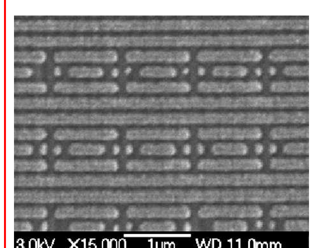
TiO$_2$ post array



Defect cavities



Defect cavities



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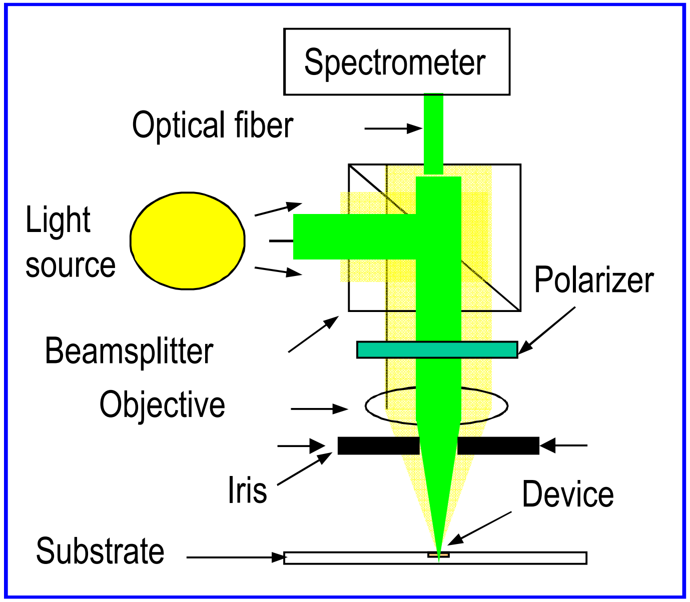
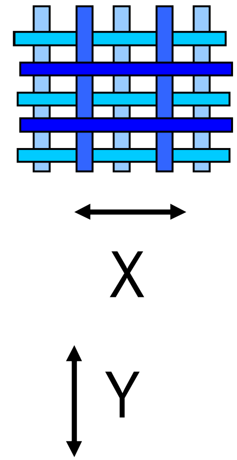
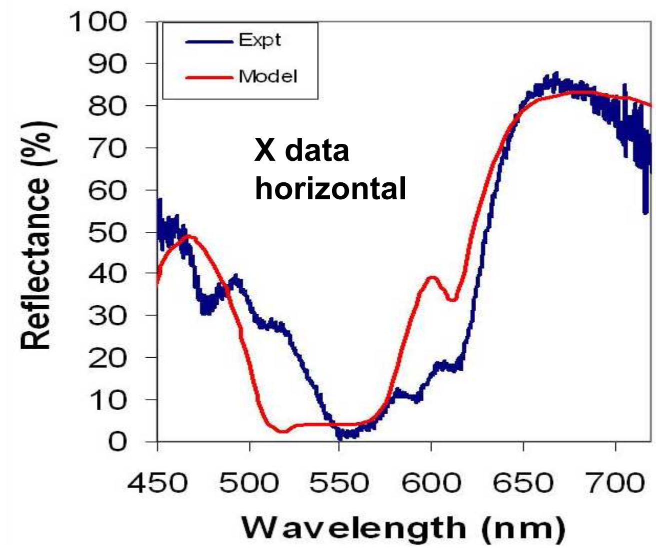
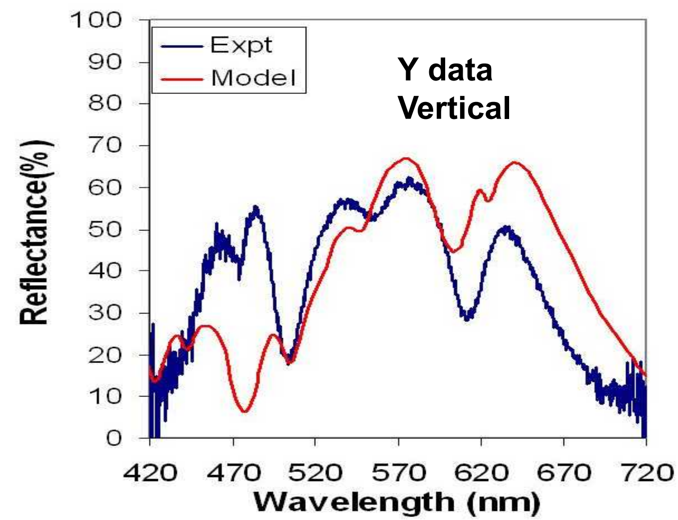
Extra Slides →

Reflectance Response: Near Normal Incidence









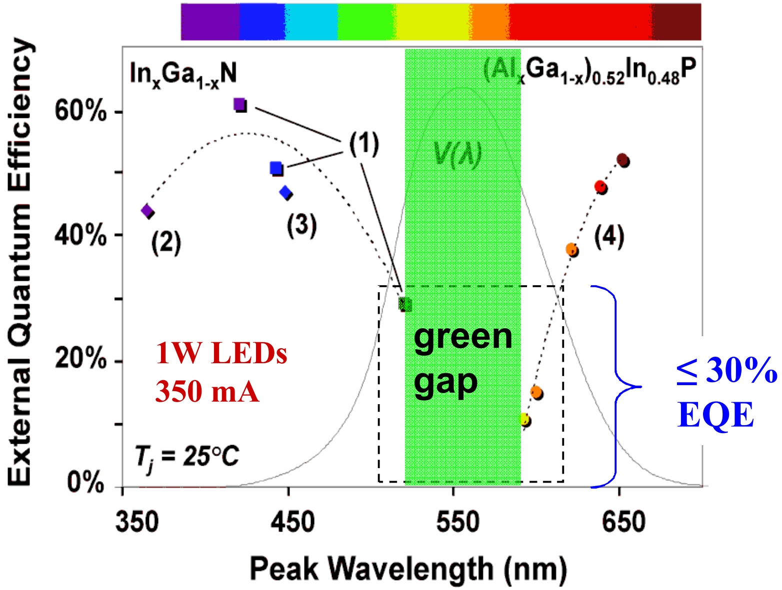
- **5 layer sample with a = 400 nm**
- **Entrance aperture at objective to reduce NA**
- **‘X’ polar spectrum different from ‘Y’ polar**
- **Surface termination**
- **Incomplete unit cell for Y polar**

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

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