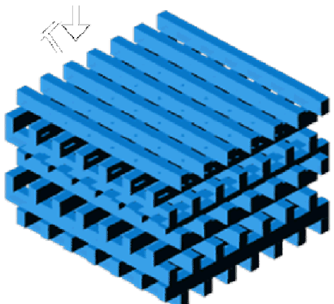
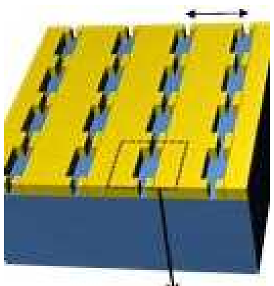
SAND2013-8502P

Nanostructures for Control of Nansocale Light-matter Interaction

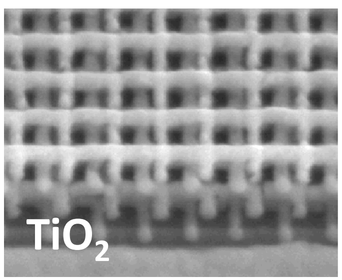


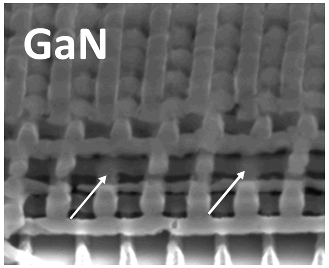


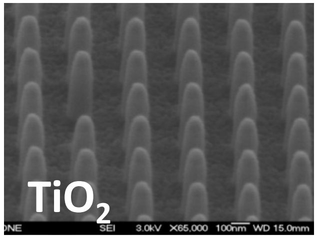
Ganapathi Subramania

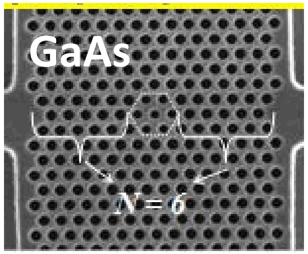
Advanced Material Science 01126
Sandia National Laboratories, Albuquerque, NM

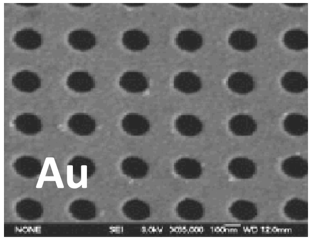
Material Science and Engineering Conference, Las Vegas, NV
October 7-9, 2013











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SSLS EFRC
SOLID-STATE LIGHTING SCIENCE
ENERGY FRONTIER RESEARCH CENTER

U.S. DEPARTMENT OF ENERGY

NNSA
National Nuclear Security Administration

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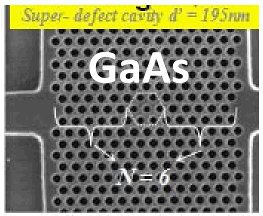
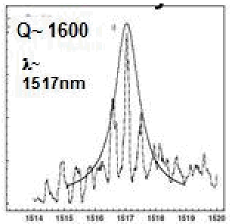
- John Nogan
- Doug Pete
- Carlos Sanchez
- Tony Coley
- Travis Young
- Luke Lester
- Stavroula Foteinopoulou
- Bob Biefeld
- Jerry Simmons

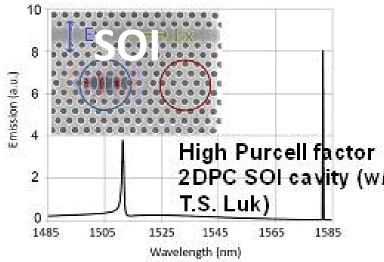
Nanophotonics Activity

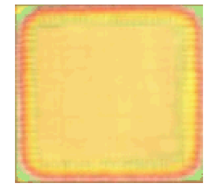
Sandia National Laboratories

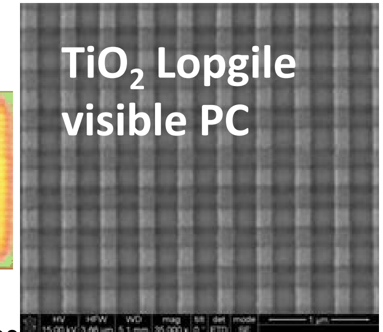
Integrated Photonics, Nanoscale lasing, Strong coupling, Full 3D emission control, Thermal control, Solid State lighting, Energy conversion

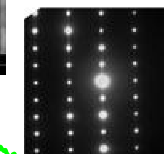


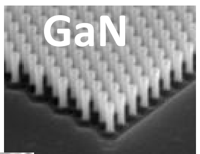




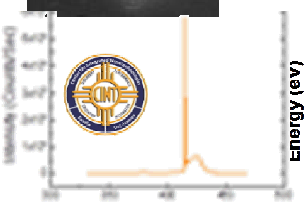


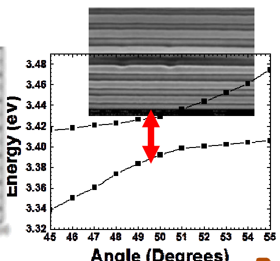






Strong coupling (w/A. Fischer)



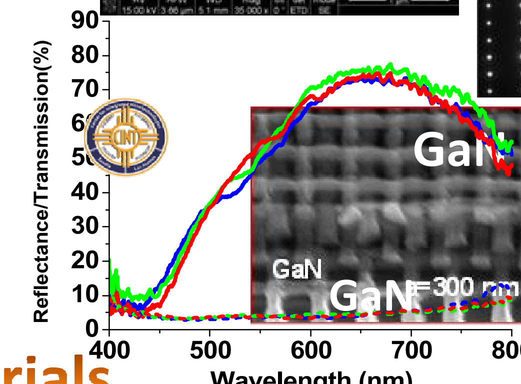


GaN 2DPC laser w/ G. Wang, I. Brener

1,2D PC

Photonic Nanostructures

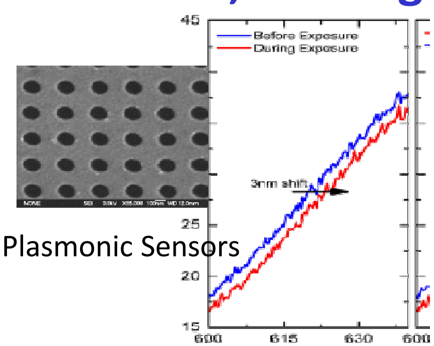
3D PC



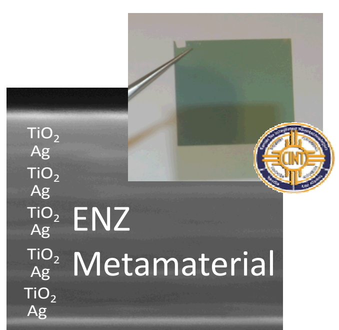
(w/ G.Wang, A. Fischer, D. Koleske)

Metal optics and Metamaterials

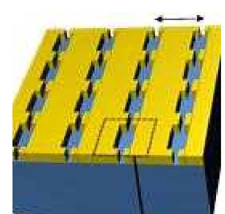
Ultrasubwavelength light control, Detection, Sensing , Nanocircuitry



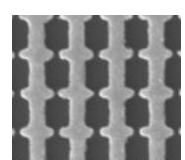
Plasmonic Sensors

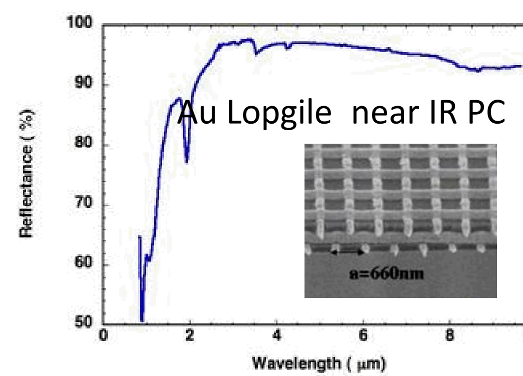


ENZ Metamaterial



Double groove Metal Nanostructure





Energy Frontier Research Center for Solid-State-Lighting Science

SSLS EFRC
SOLID-STATE LIGHTING SCIENCE
ENERGY FRONTIER RESEARCH CENTER

- **Duration: Aug 2009 – Jul 2014**
- **One of 46 EFRC centers nationwide**
- **Budget: $3.6M/year for 5 years**

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Mike Coltrin
Chief Scientist:
Jeff Tsao

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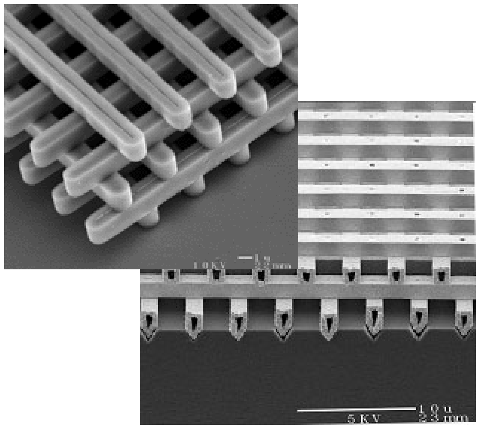
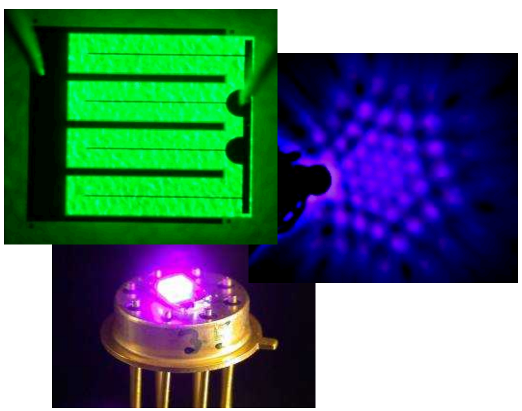
Objective : Understand Spontaneous Emission Control to Benefit SSL

Sandia National Laboratories

3D photonic crystals

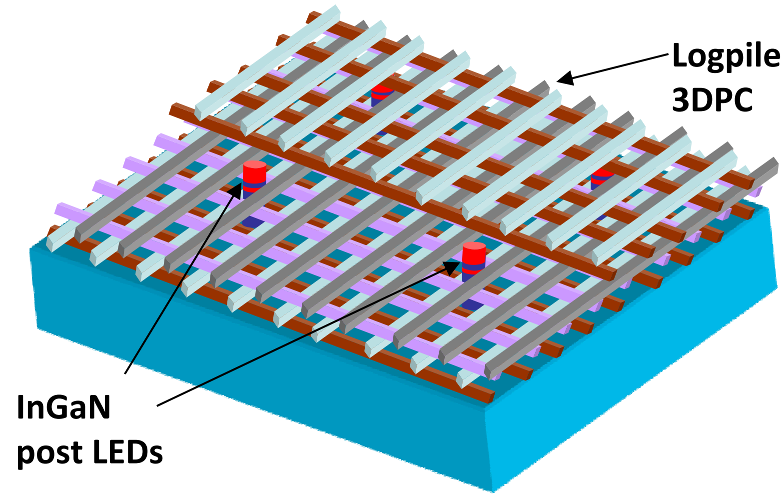


InGaN LEDs



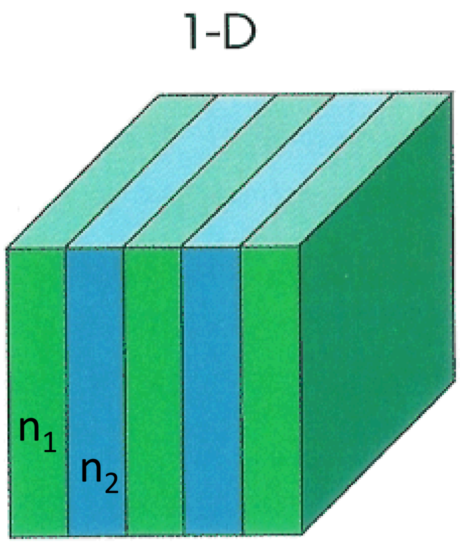
- Sandia has strong capabilities in 3DPC and Nitrides
- Merge InGaN LED research with 3D photonic crystal research.

InGaN LED array incorporated into a 3DPC

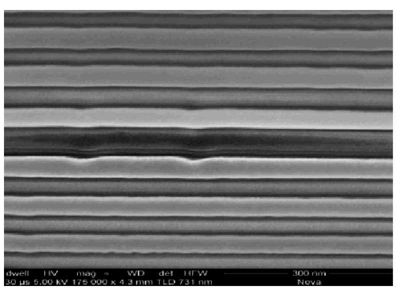


Improving InGaN LED efficiency enhancements will benefit SSL – e.g. Green emission.

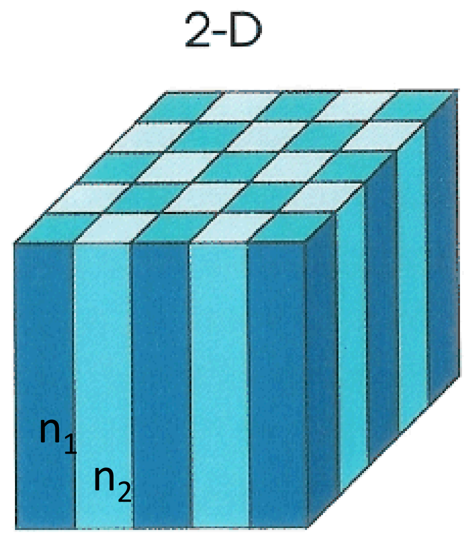
Periodic Sub-wavelength Structures for EM Control: Photonic Crystals (PC)



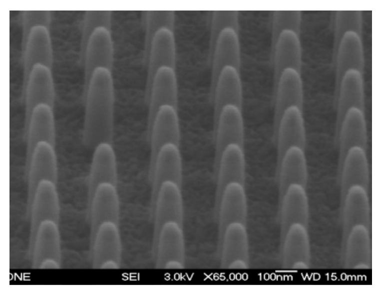
Periodic in 1 dimension



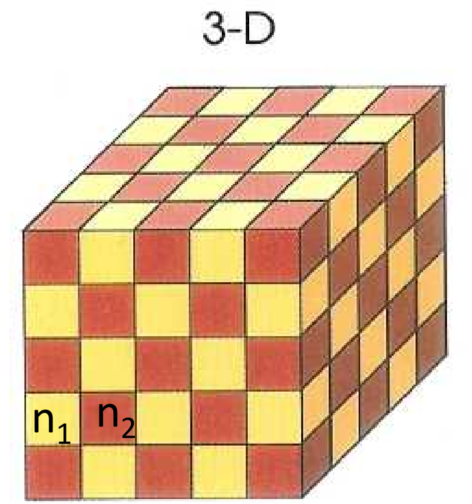
HfO_2/SiO_2 DBR cavity



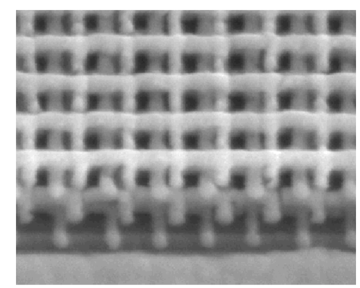
Periodic in 2 dimensions



Array of TiO_2 rods



Periodic in 3 dimensions



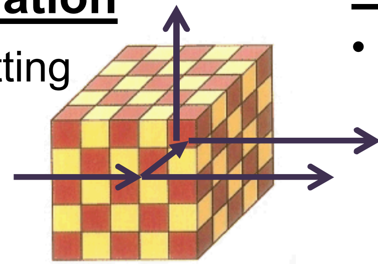
TiO_2 logpile PC

Reference: "Photonic crystals: Molding the flow of light", J.D. Joannopoulos, R.D. Meade, J. Winn, Princeton Univ.Press, NJ(1995)

Photonic Crystals: A Great Platform for Light Manipulation

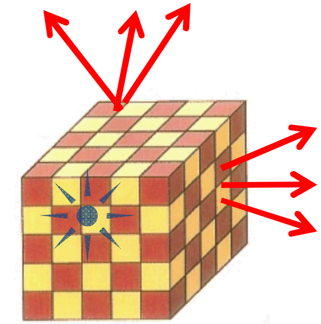
Control of light propagation

- Guiding, bending and splitting
- Negative refraction
- Self-collimation
- Localization
- Slow light
- Cloaking

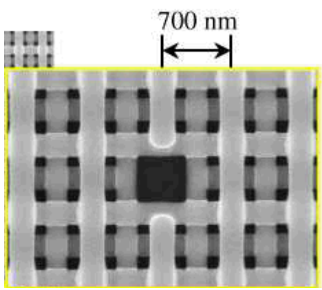


Control of light emission

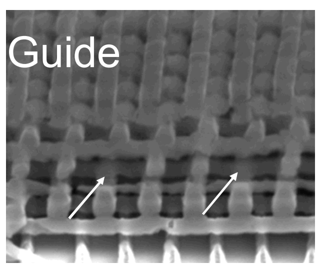
- Full 3D confinement effect
  - Photon – Atom Bound states, Non- Morkovian emission process



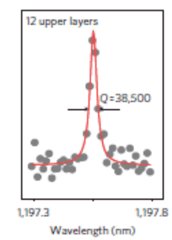
- Photonic Density of States effect
  - Spontaneous emission enhancement

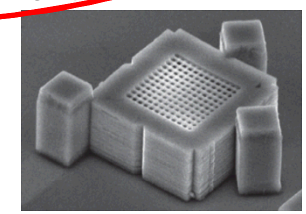


S. Kawashima et. al.., Opt. Exp. **18**, 386 (2010)

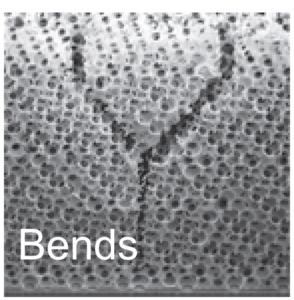


G. Subramania et. al, Nano Lett. **11**, 4591 (2011).

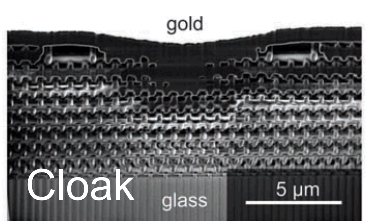




A. Tandaechanurat et. al., Nat Photon **5**, 91 (2011).

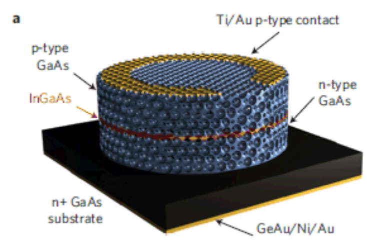


S.A. Rinne et. al., Nat. Phot. **2**, 52(2008)



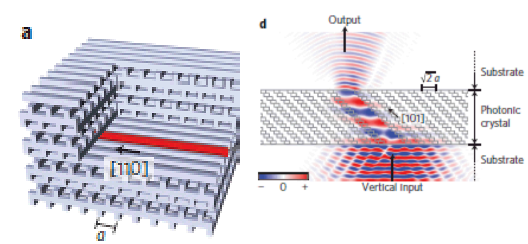
T. Ergin et. al.,Science **328**, 337 (2010)

Electrical Injection



E. C. Nelson, et al., Nat Mat **10**, 676 (2011)

Emission enhancement



K. Ishizaki et.al. , Nat. Phot.,**7**, 133 (2013)).

Spontaneous Emission Control using PC

Sandia National Laboratories

Radiative rate of an emitting dipole in an EM field (Fermi's Golden Rule)

$$\Gamma_r(\omega) = \frac{\pi\omega\mu^2}{3\varepsilon_0\hbar}\rho(\omega)$$

Photonic Density of States (PDOS)

$$\rho_f(\omega) = \frac{\omega^2}{\pi^2 c^3}$$

← PDOS in free space

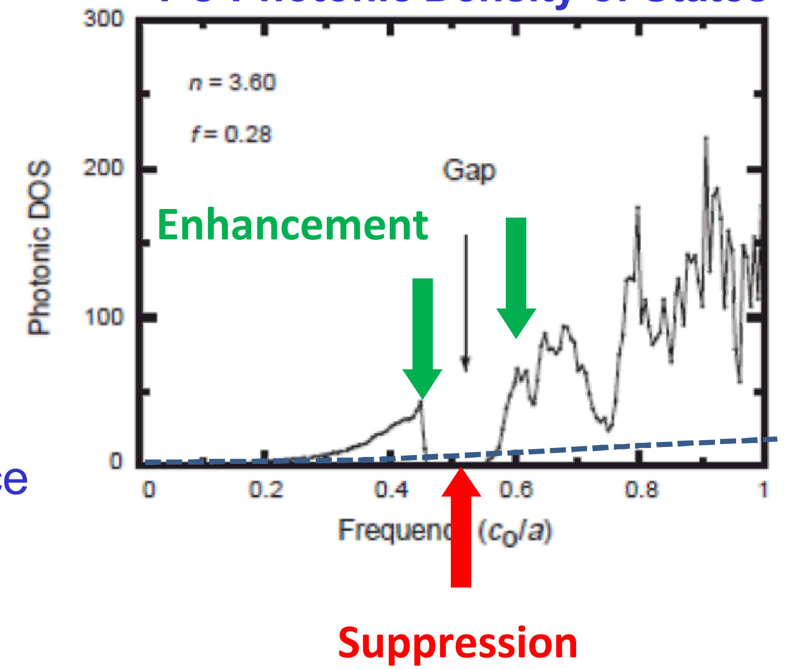
Radiative rate modification

$$\frac{\Gamma_{r\_PC}(\omega)}{\Gamma_{r\_free}(\omega)} = \frac{\rho_{PC}(\omega)}{\rho_f(\omega)}$$

Local Photonic Density of States (LDOS)

$$\rho_L(\mathbf{r},\omega) = \sum_n \int_{1stBZ} d^3k \, |\bar{\mu}\cdot\mathbf{E}_{n,k}(\mathbf{r})|^2 \, \delta(\omega^2 - \omega_{n,k}^2)$$

PC Photonic Density of States



Also Important :

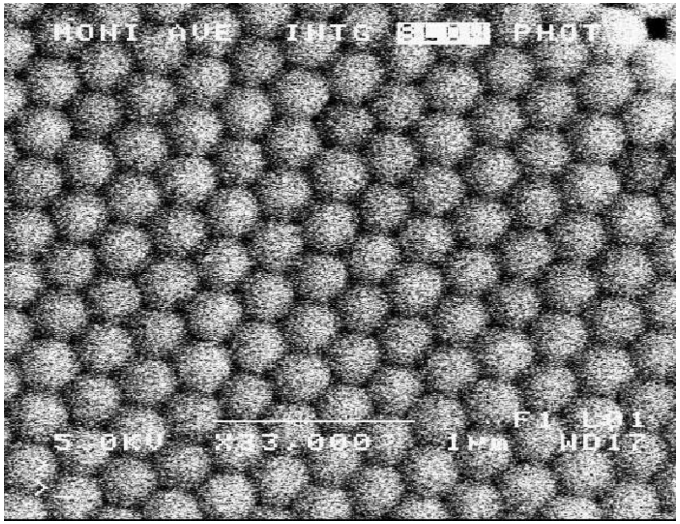
- Where the emitters are located in the unit cell
- The orientation of the emitter dipole w.r.t to the eigenfield

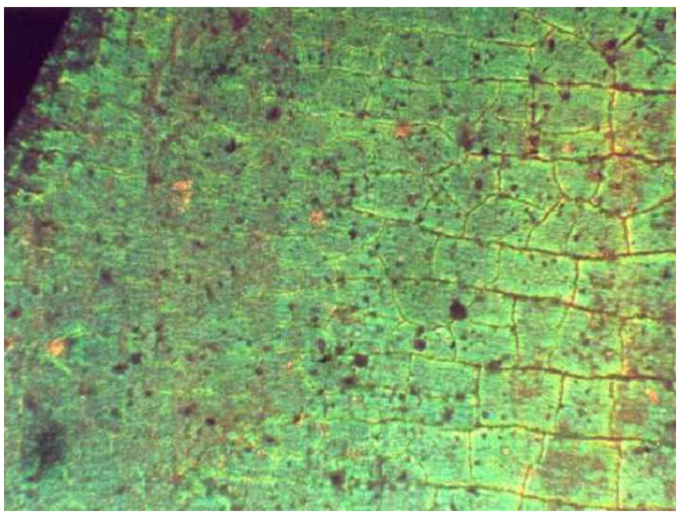
K. Busch and S. John, Phys. Rev. E,**58**, 3896(1998)

- TiO_2 inverse opal photonic crystal
- TiO_2 logpile photonic crystal
- GaN logpile photonic crystal
- GaN nanowire photonic crystal lasers

TiO2 Inverse Opal 3D Photonic Crystal

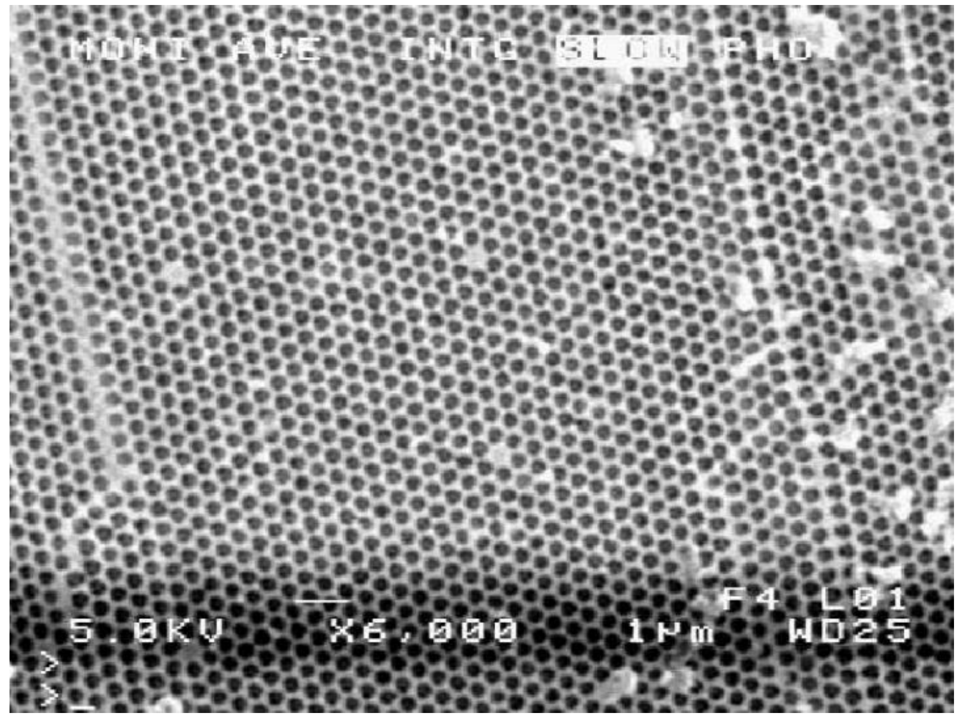
Sandia National Laboratories





Periodicity achieved using close packed nanospheres -> FCC lattice

High index material: Titania (TiO_2)

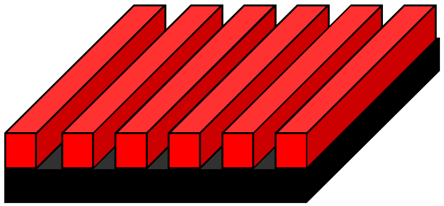


1. G. Subramania et. al. Applied Physics Letters **74**, 3933-3935(1999,).
2. G. Subramania et. al. , Synthetic Metals, **116**, 445-448 (2001).
3. G. Subramania et.al. , Physical Review B, **63**, 235111-235117(2001).

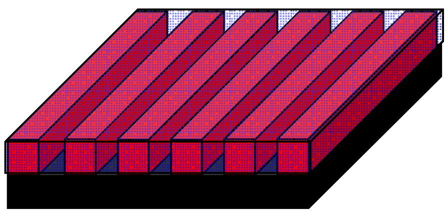
Multilevel E-beam Fabrication Process

PC fabricated one layer at a time.

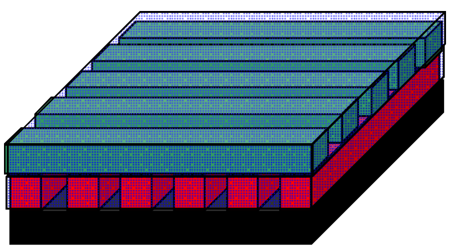
1.Fabricate first level



2. Fill and planarize



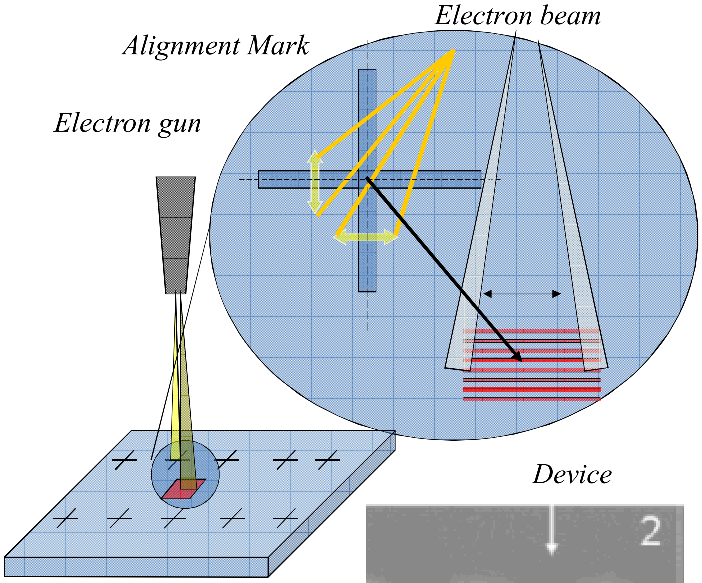
3. Make next level



4. Repeat process to make additional levels

Alignment

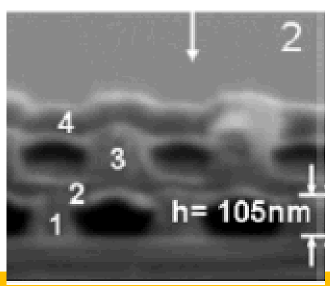
- Lattice constants < 400nm
- Alignment precision to within 10% of lattice constant needed
- High quality Au registration marks



- Alignment
- Planarization

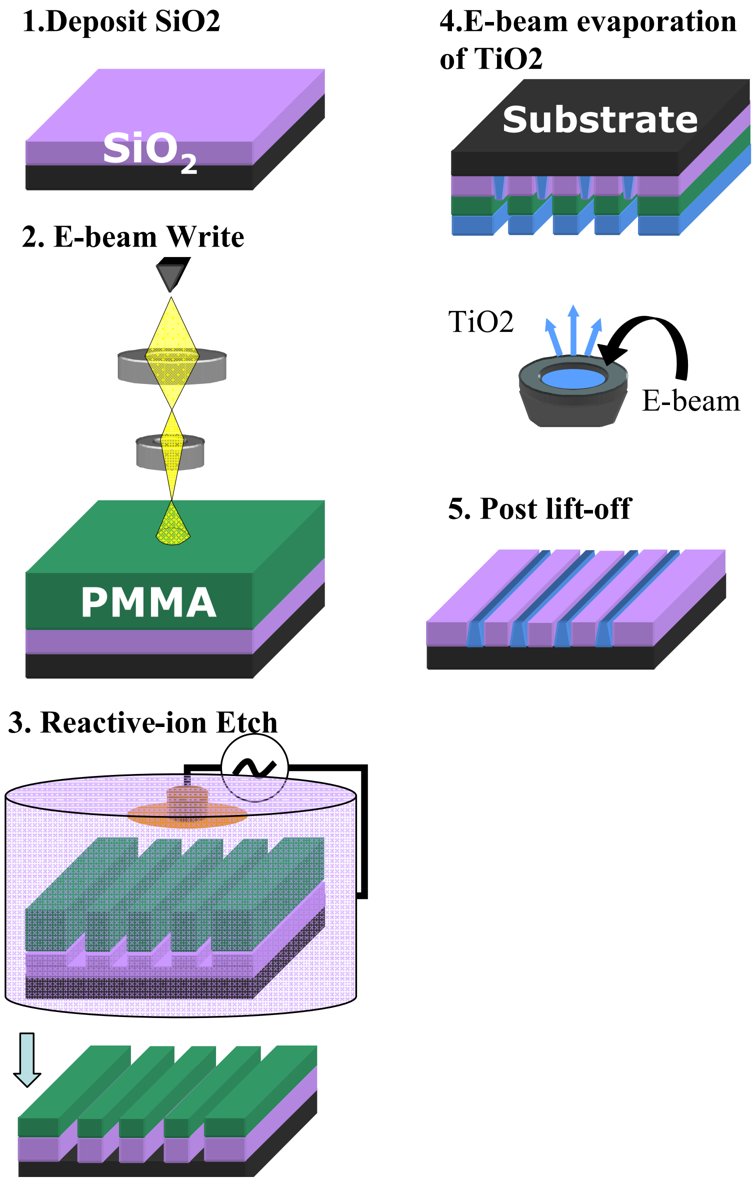
G. Subramania, Nanotechnology **18** , 035303 (2007).

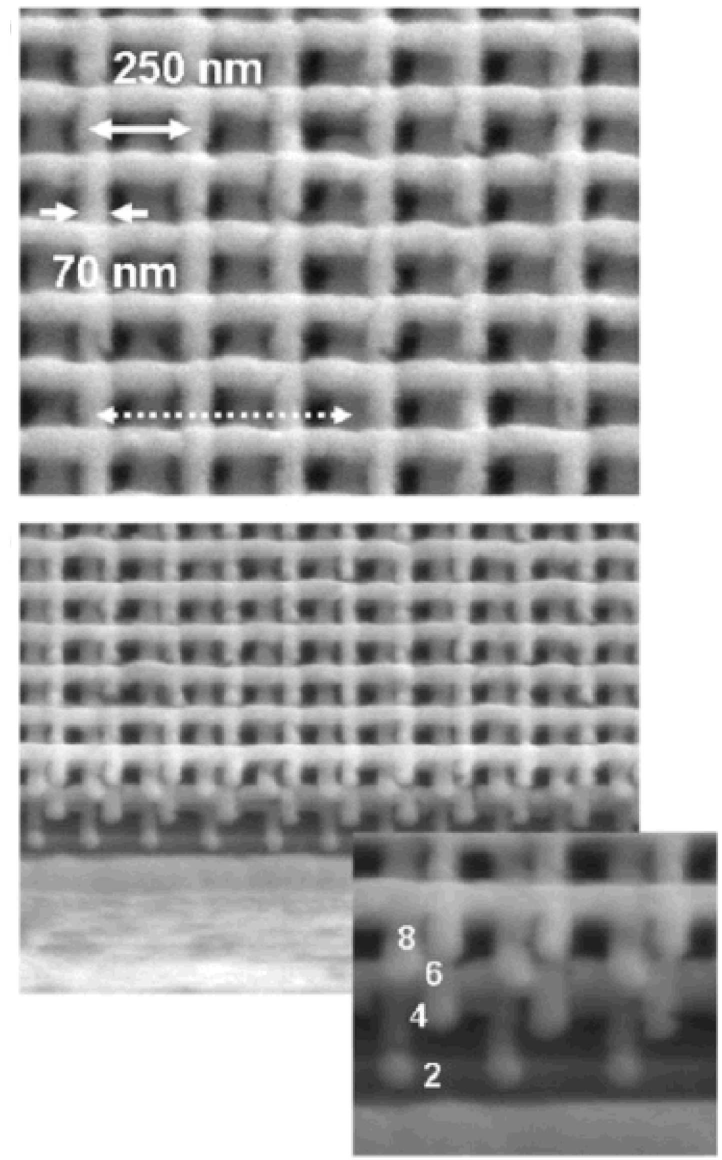
Device



Excellent alignment was obtained with both JEOL- JBX5E and 9300FS

TiO_2 Logpile Photonic Crystal Fabrication Process

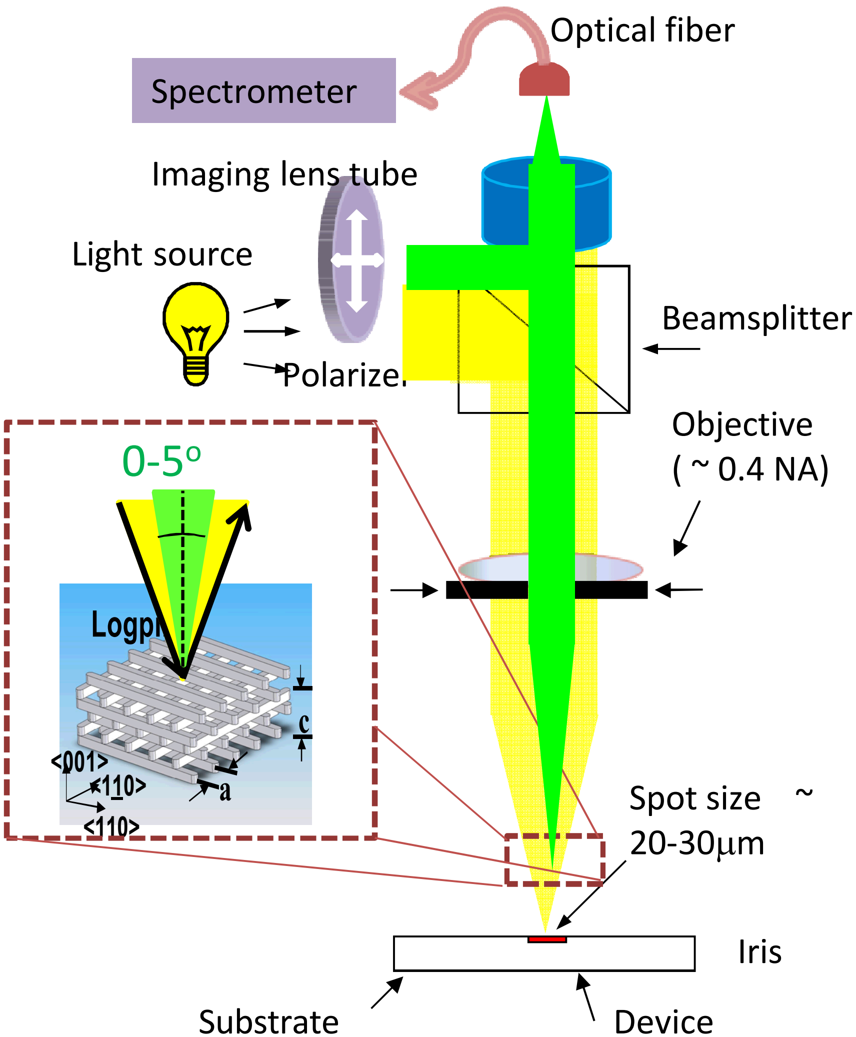


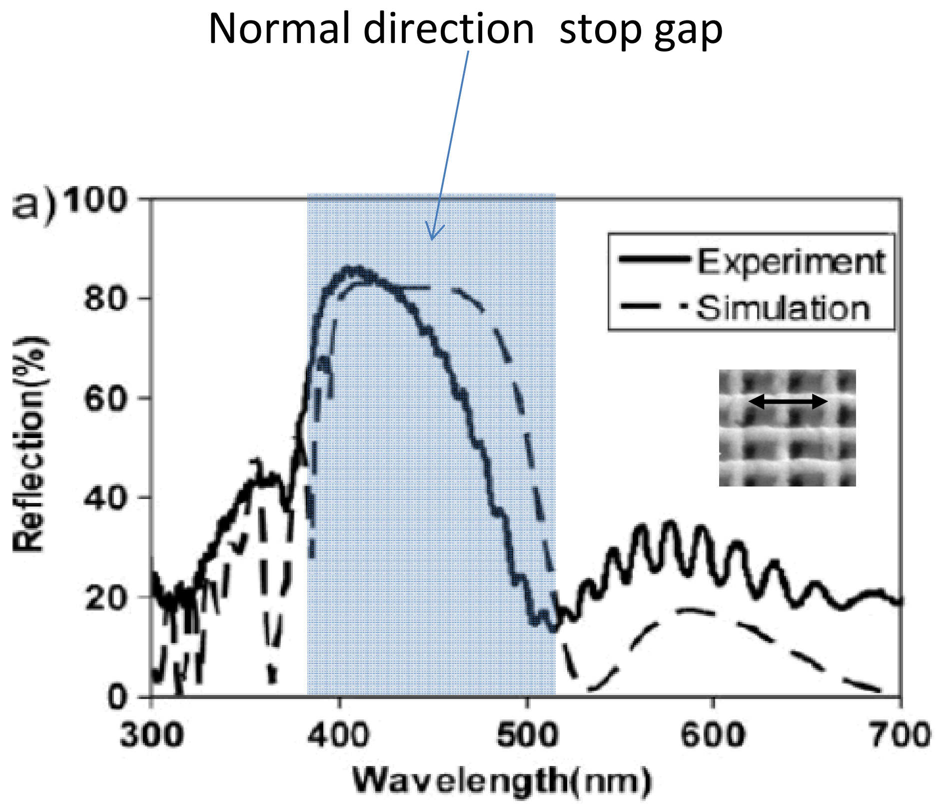


G. Subramania et. al., Adv. Mat. **22**, 487-491 (2010)

Visible- Near UV 9 Layer TiO_2 Logpile

Sandia National Laboratories





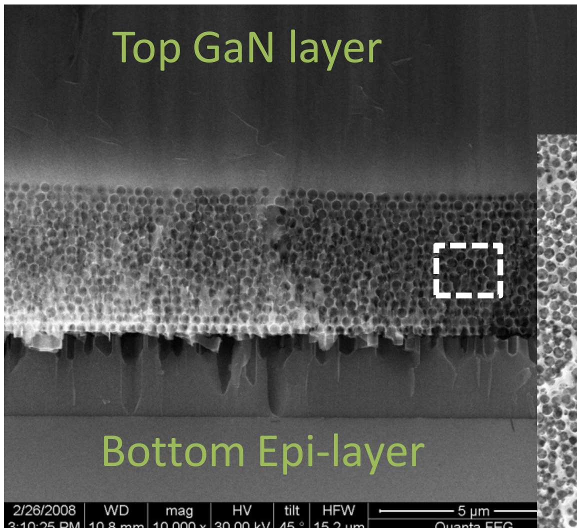
G. Subramania, Y.-J. Lee, A. J. Fischer, and D. D. Koleske, Advanced Materials **22**, 487-491 (2010)

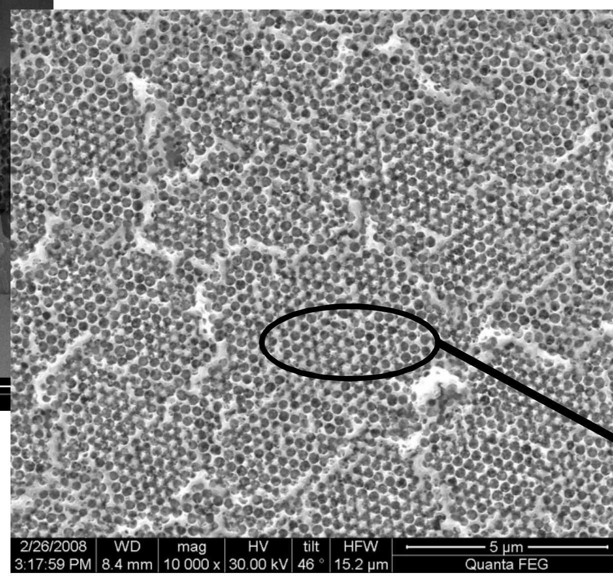
All GaN Logpile Photonic Crystal

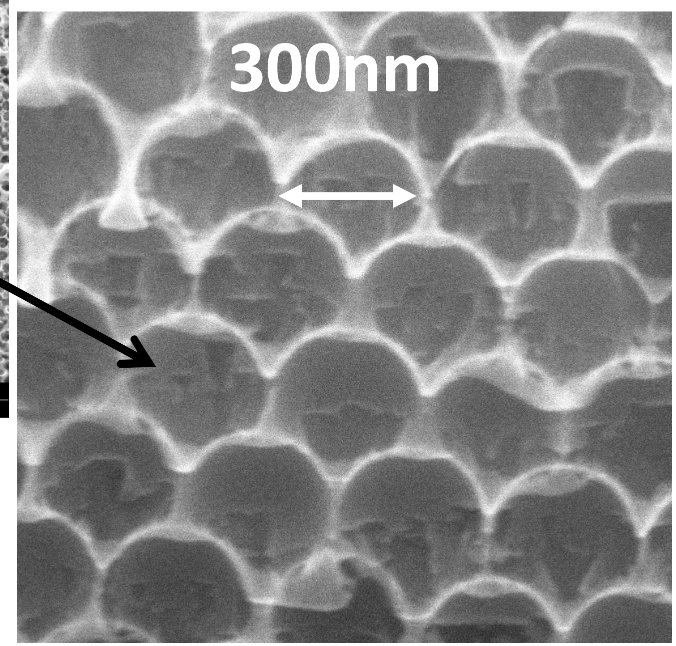
Sandia National Laboratories

Epitaxial GaN grown though nanostructured template reduces dislocation density

(George T. Wang, Qiming Li)

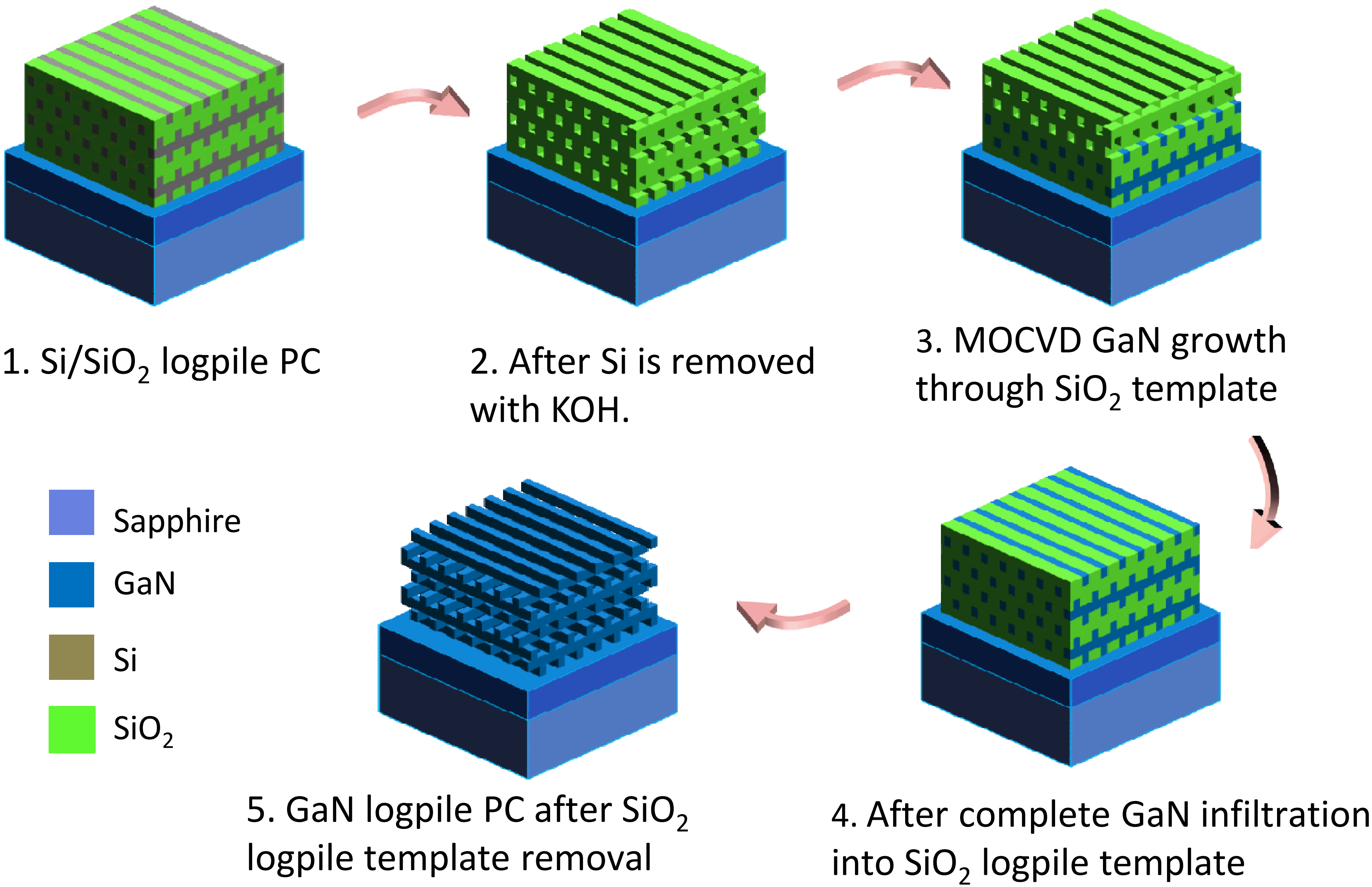




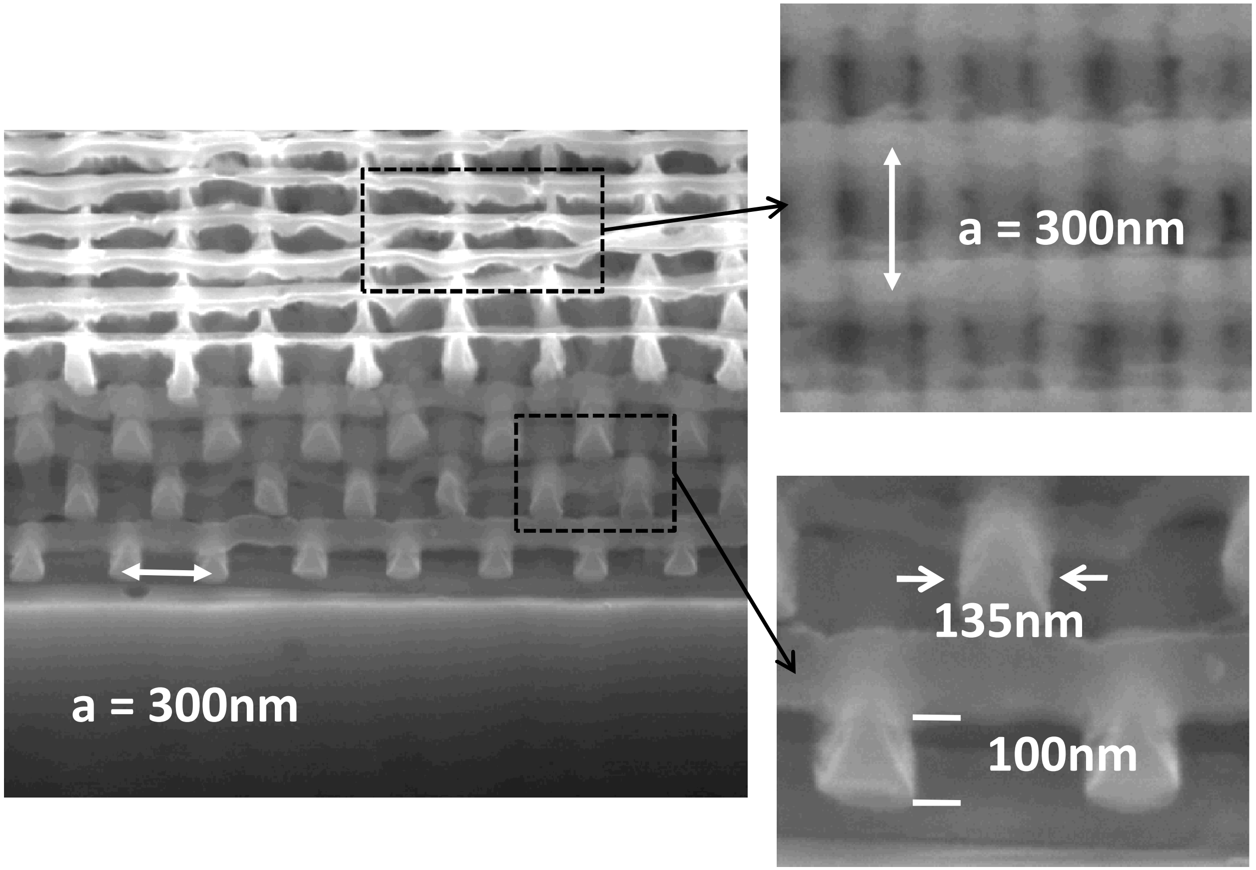


Qiming Li et. al., Appl. Phys. Lett. 94, 231105(2009).

GaN growth through logpile template

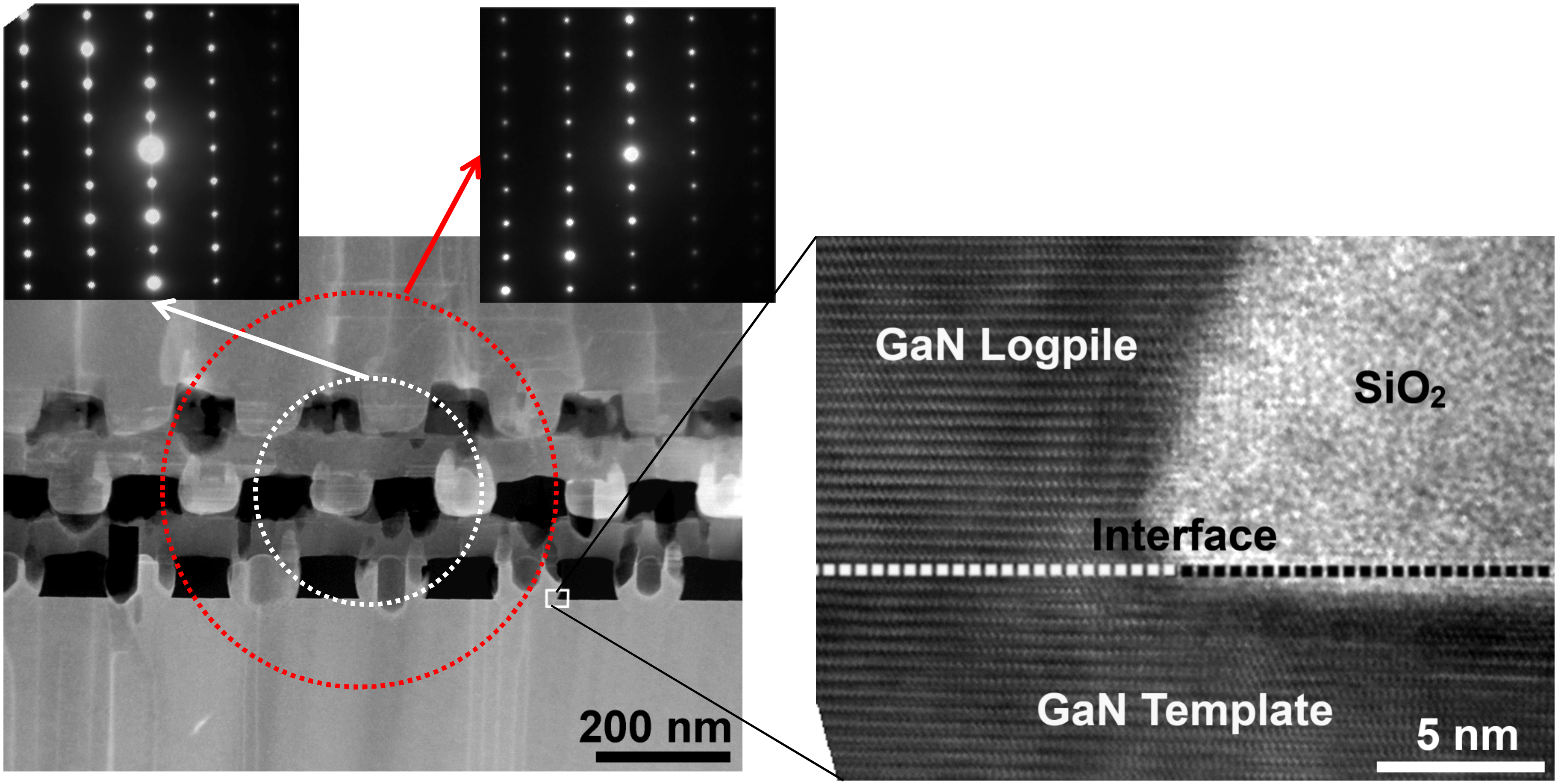


A nine layer all GaN logpile PC



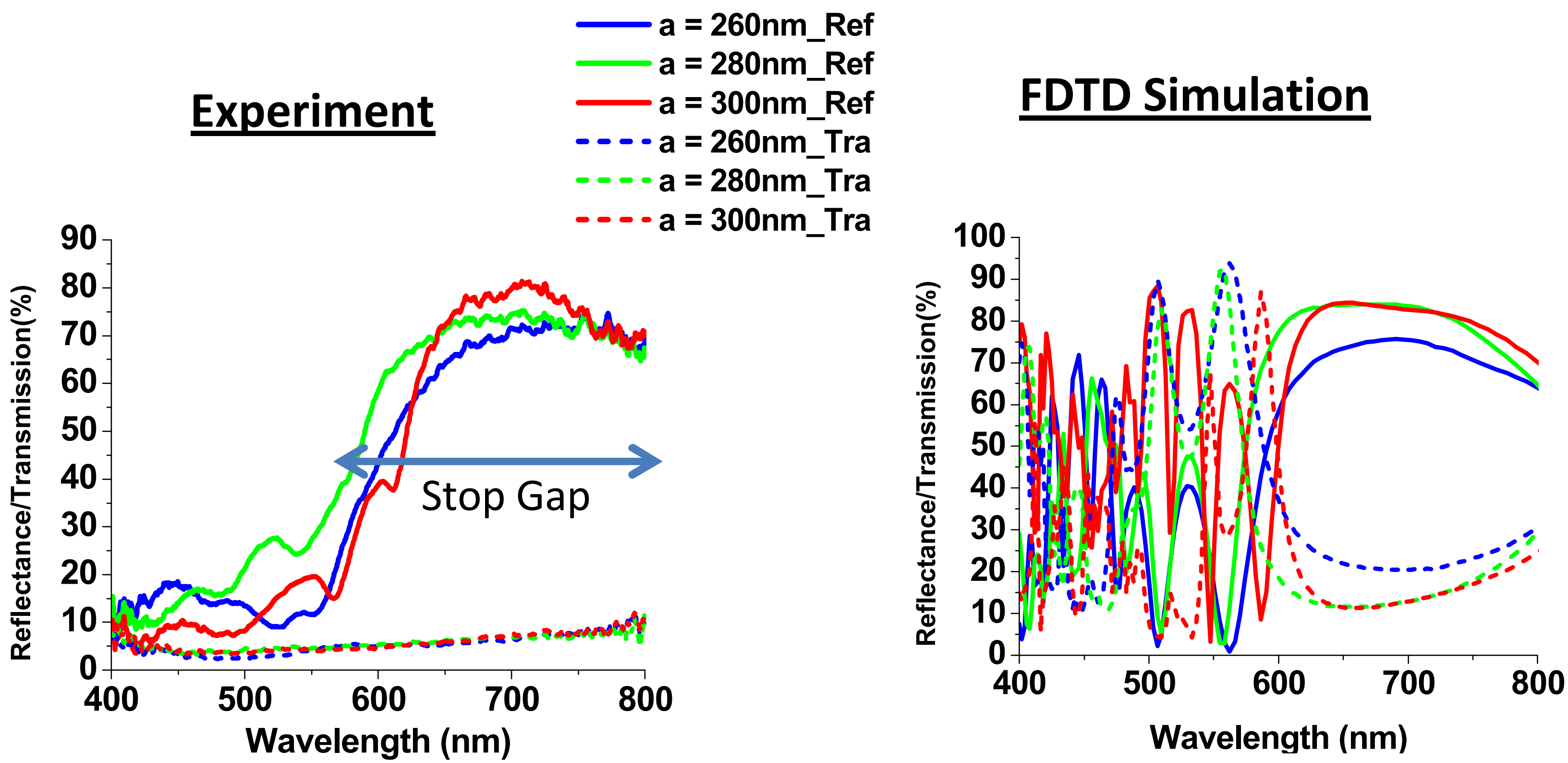
G. Subramania, Q. Li, Y.-J. Lee, J. J. Figiel, G. T. Wang, and A. J. Fischer, Nano Lett. **11**, 4591 (2011).

Bottom-up epitaxial growth of crystalline GaN



G. Subramania et. al. , Nano Lett. **11**, 4591 (2011).

Optical Response

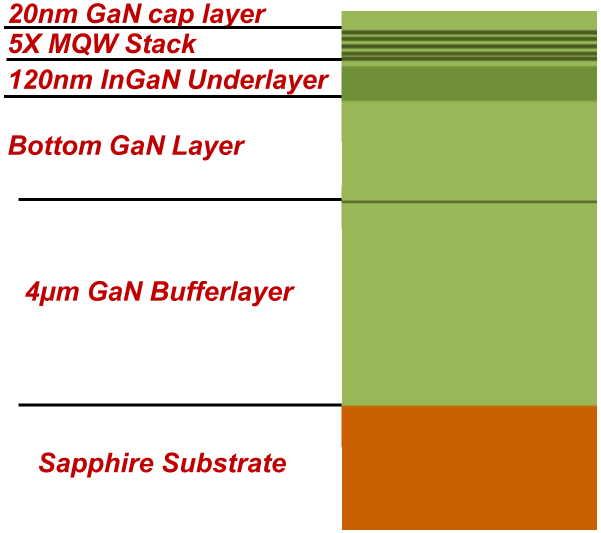


G. Subramania et. al. , Nano Lett. **11**, 4591 (2011).

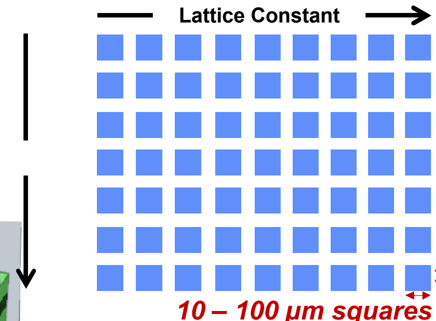
Photonic Crystal III-N nanowire laser array

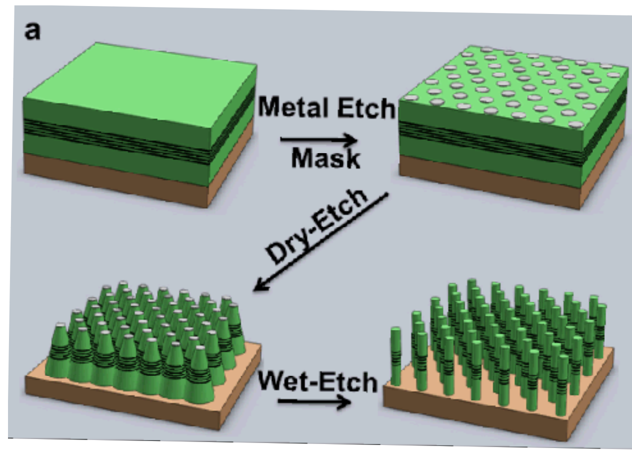
Sandia National Laboratories

Growth of III-N active material

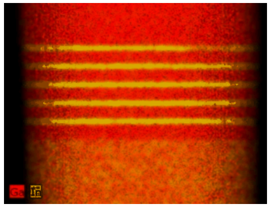


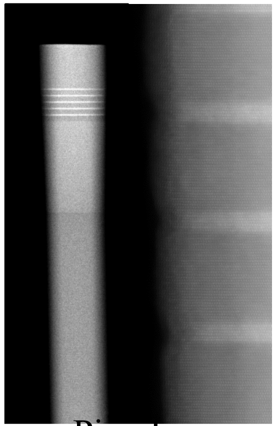
Patterning of 2DPC using e-beam lithography





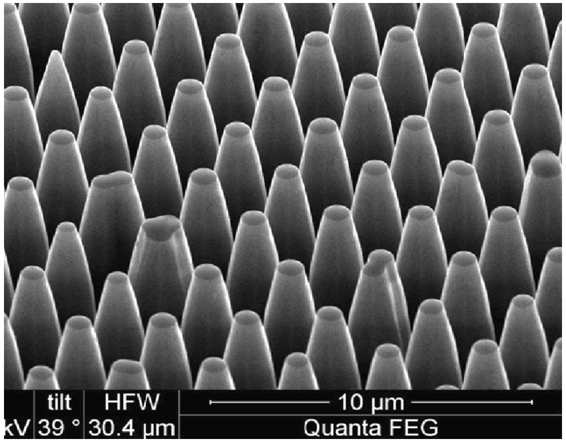
TEM images

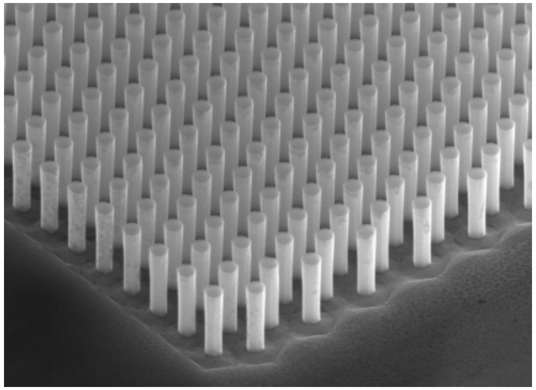




Ping Lu, Sandia

Inductively coupled plasma dry etch





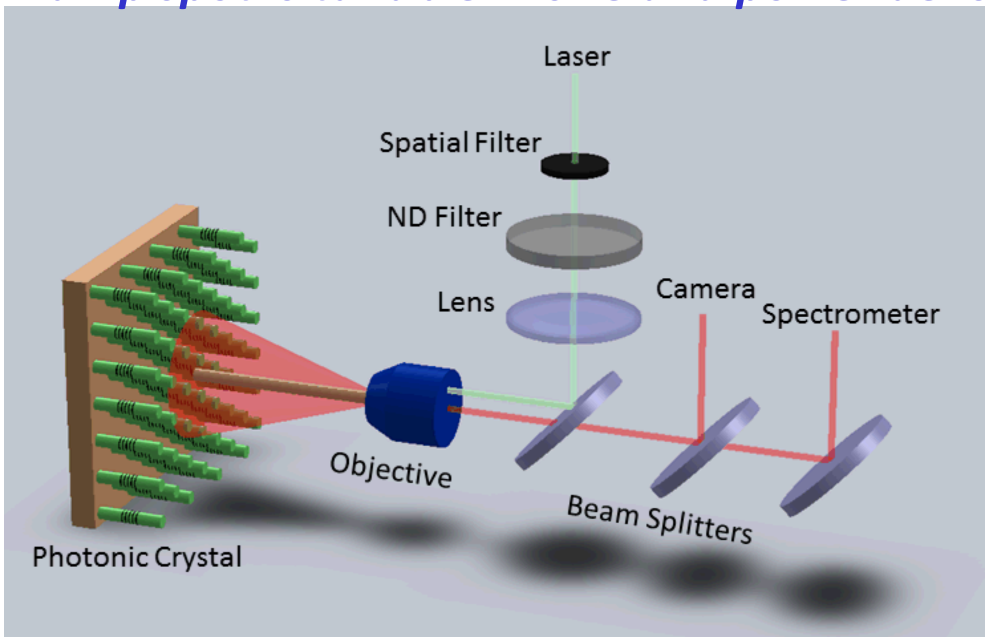
Wet etch using KOH process

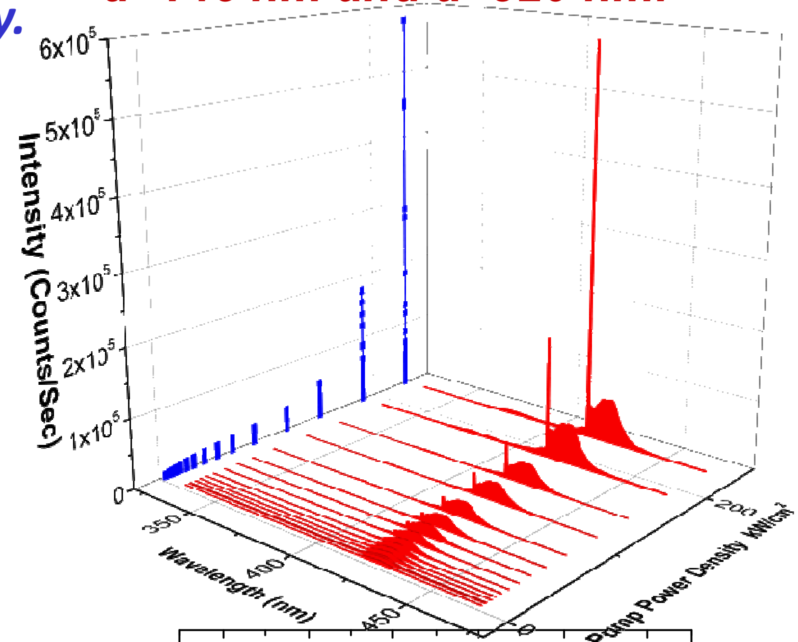
Optical response – low threshold lasing

Sandia National Laboratories

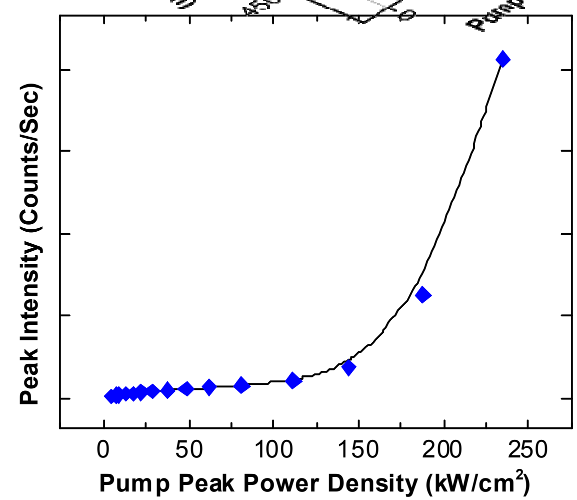
Array Element Dimensions: d=145 nm and a=320 nm.

Pump spot is tunable in size and power density.



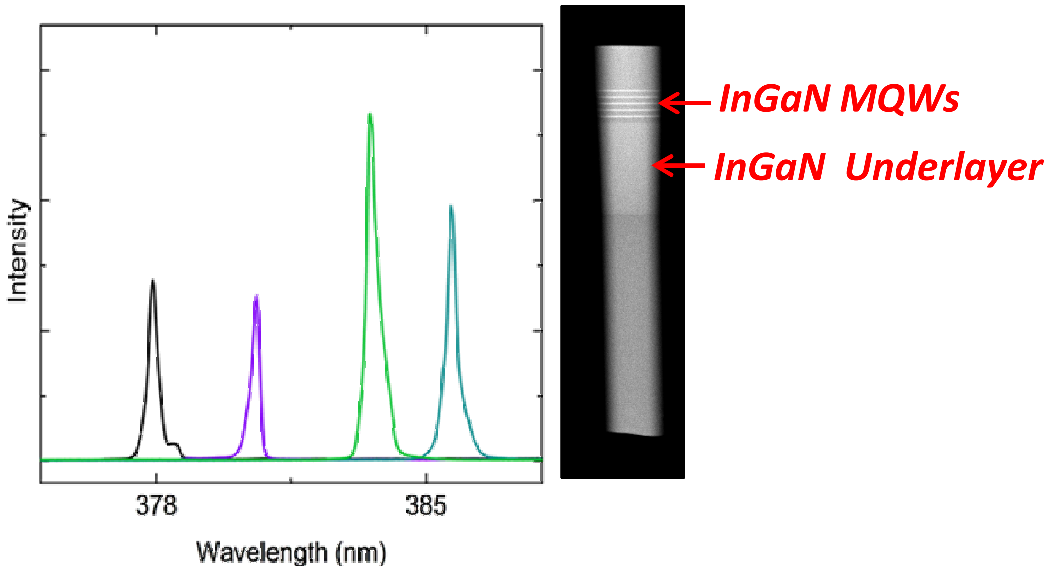


A low lasing threshold is achieved with < 500kW/cm2 for all PC lasers fabricated.



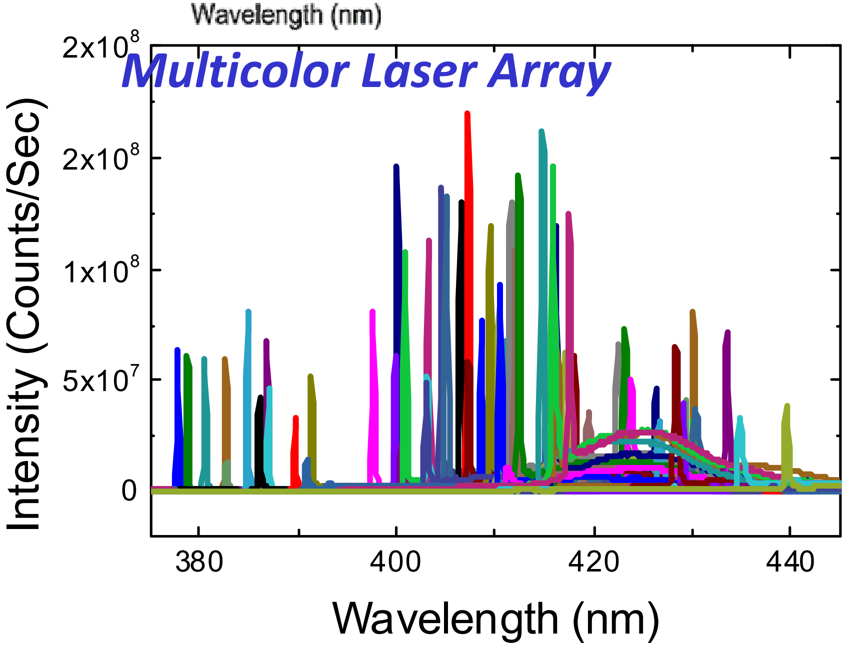
Results

Gain from multiple gain sections



- ***Multiple gain regions can simultaneously lase***
- ***Pathway for enhancing the available lasing spectral range***

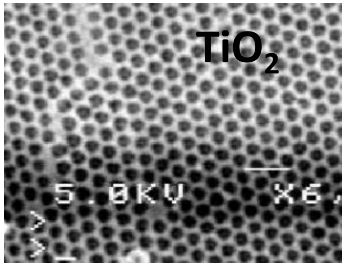
Multicolor Laser Array

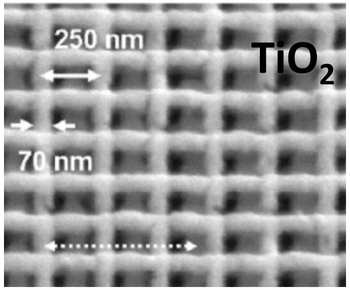


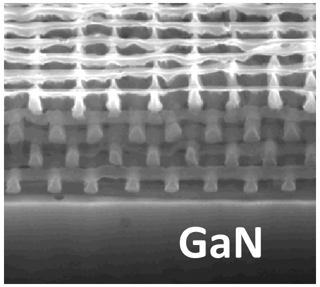
- ***Wavelength tunability (lattice constant and nanowire diameter).***
- ***Multiple color laser emission on a single epitaxial wafer covering a spectral range from 380-440nm.***

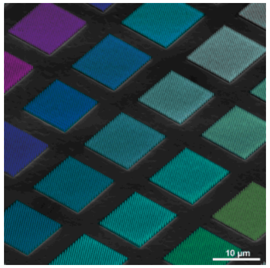
Summary and Outlook

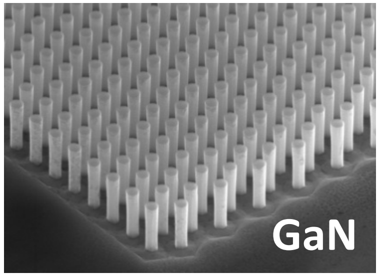
- *Photonic Crystals are very effective in controlling light matter interaction*
- *Enhances photonic density of states*
- *Low threshold lasing*
- *Demonstration of PCs in the visible with different material systems*
    - *TiO_2, GaN, Si*











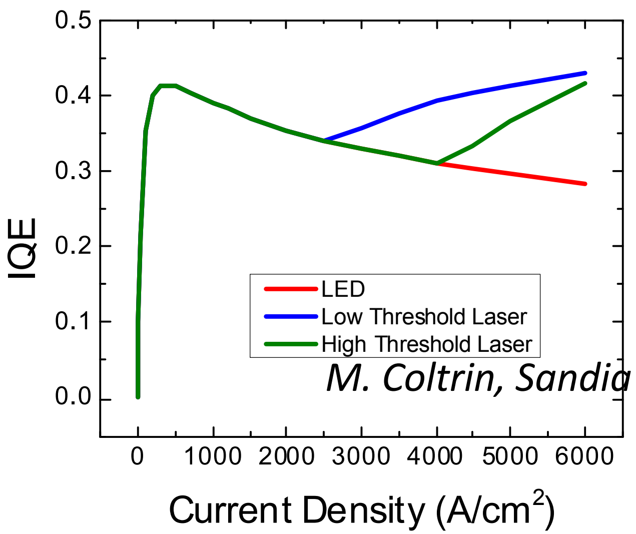
Thank you for your attention!

Backup slides

Nanowire Photonic Crystal Lasers

(Jeremy B. Wright, Sheng Liu, Alexander Benz, George T. Wang, Qiming Li, Daniel D. Koleske, Ping Lu, Huiwen Xu, Luke Lester, Ting S. Luk, Igal Brener)

Why Lasers in Solid State Lighting?

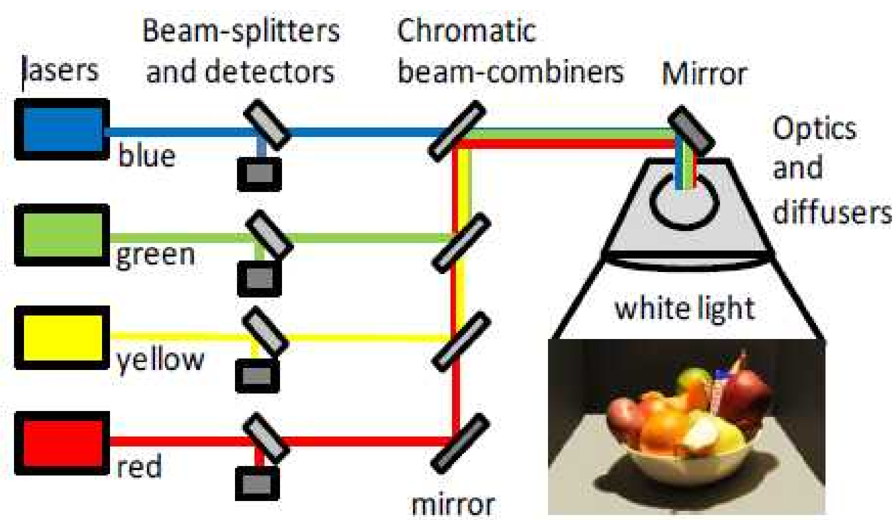


The LED "efficiency droop" problem:

- ***InGaN LEDs exhibit a decrease in efficiency at high drive currents***
- ***limits the operating current density***
- ***increases the cost per lumen***

➢ ***4 color lasers suitably combined can achieve high color rendering***

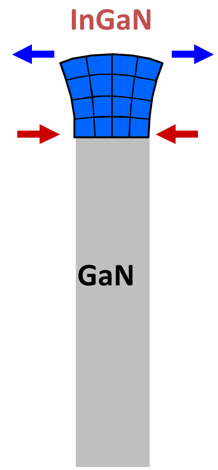
➢ ***Lasers can potentially mitigate the efficiency droop problem !***

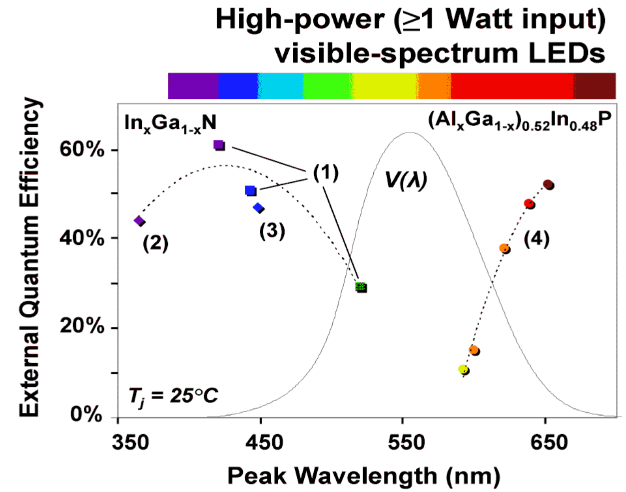


A. Neumann et al., "Four-color laser white illuminant demonstrating high color-rendering quality," Opt. Express, vol. 19, pp. A982-A990, 2011.

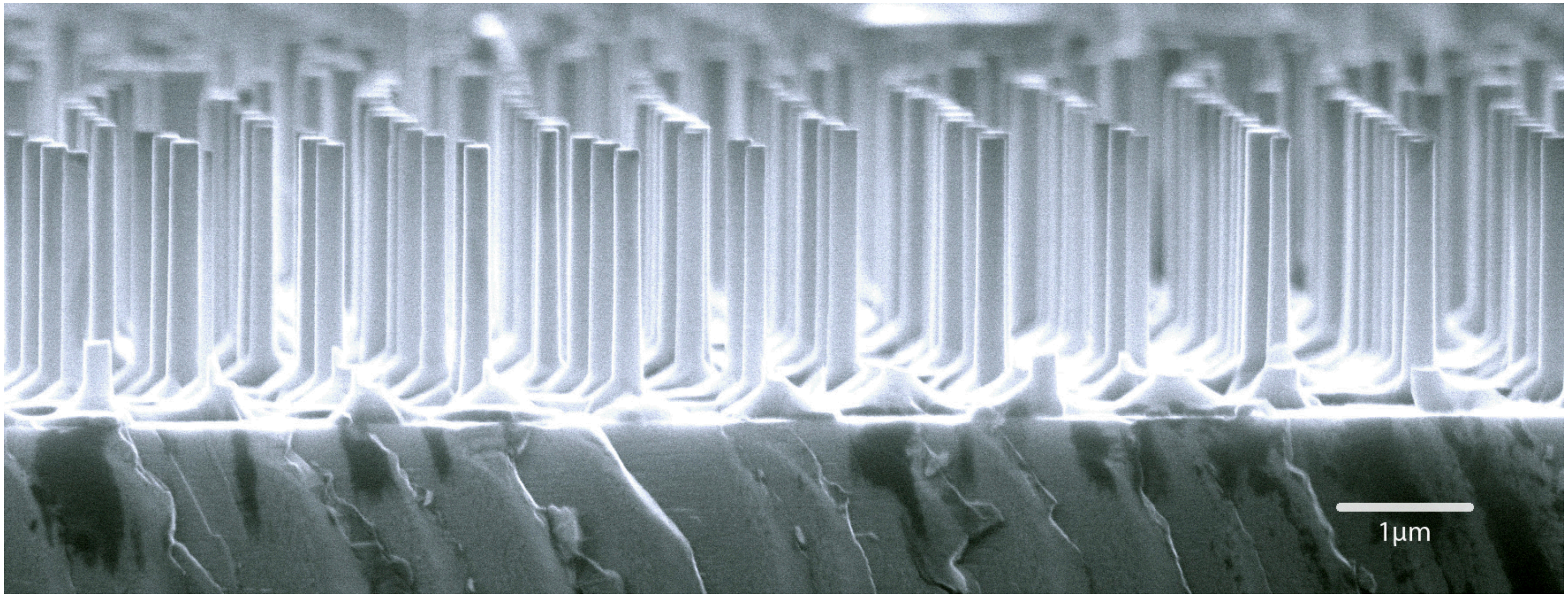
Why Nanowires for Lasing?

- **Reduced strain**
  - **Greater range of alloy compositions**
- **Easily integrated into two-dimensional arrays**

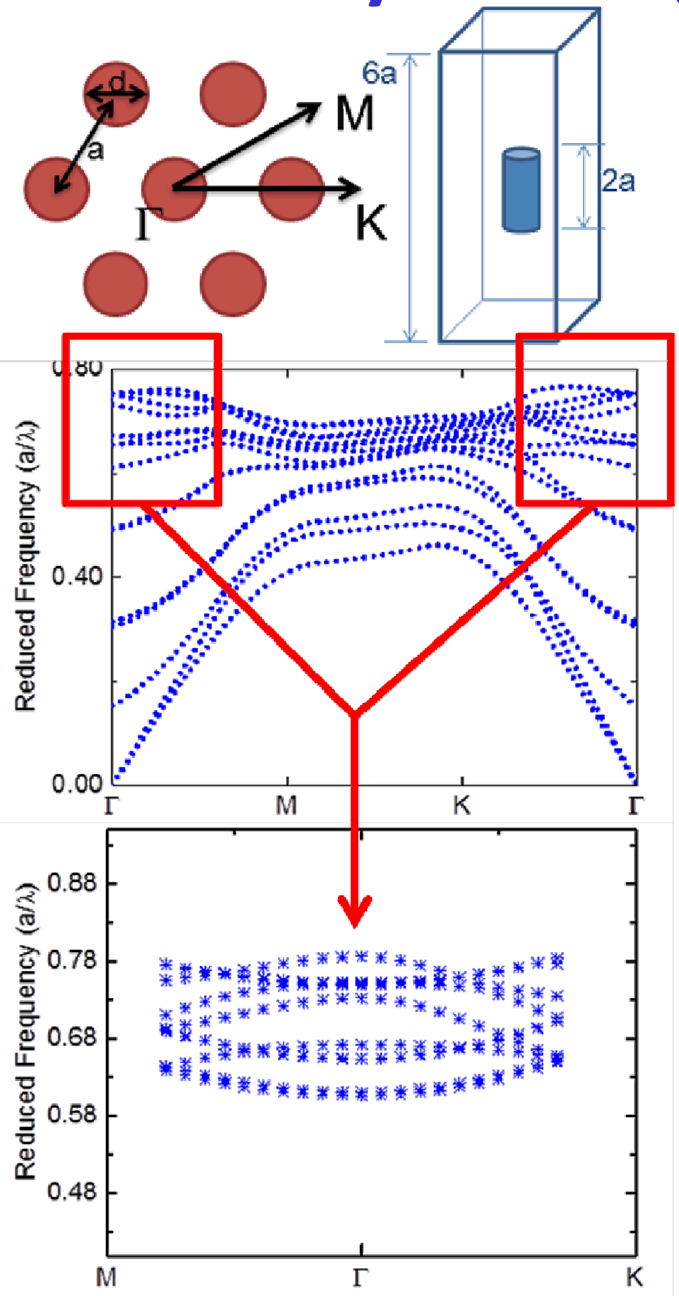




M. Krames, Philips Lumileds



Vertically emitting photonic crystal lasers - Theory



- ***Low lasing threshold***
  - ***Increase interaction of light with the gain region***
  - ***Enabled by 'Low group velocity modes' in photonic crystal***
- ***Vertical Emission***
  - ***At higher bands there are low group velocity modes near the Gamma point***
  - ***Near the Gamma point the in-plane wave vectors are small resulting in near-normal emission***
- ***Wavelength Tunability***
  - ***Achieved through lattice constant and nanowire diameter variation***

Visible Frequency Logpile Photonic Crystal

Sandia National Laboratories

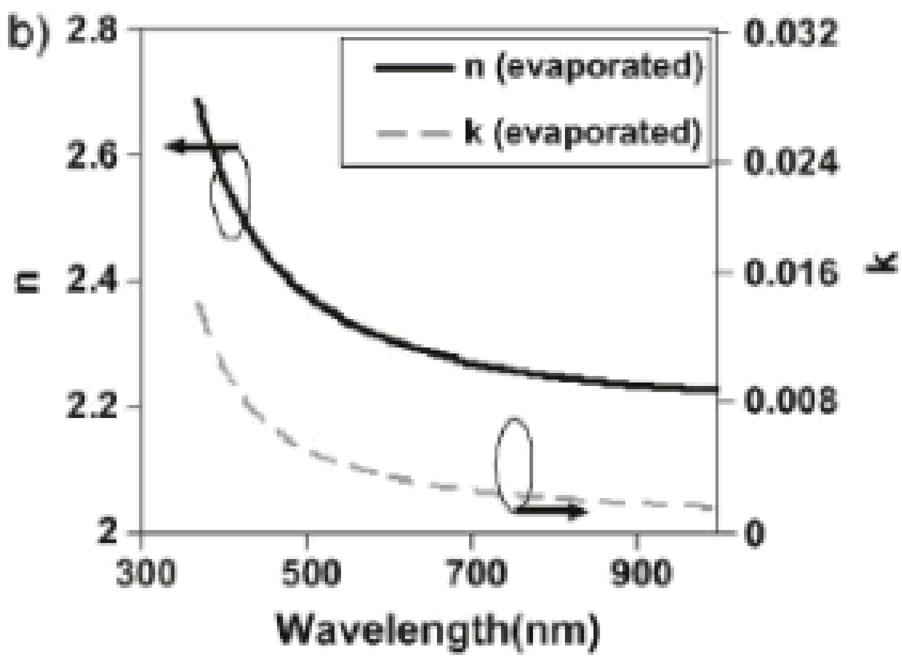
Material Choice : TiO_2

Requirements:

- **High enough index with low loss in the visible :**
  - n ~ 2.3-2.4 in the visible /w k< 0.015
- **Easy to deposit :**
  - Sputtering, evaporation, solgel, Atomic Layer Deposition

Method of fabrication:

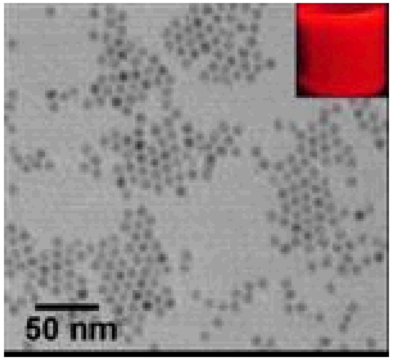
- **Nanofabrication based on electron beam lithography**



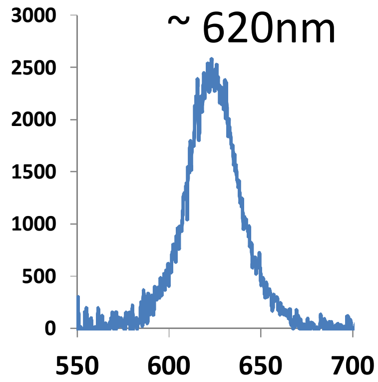
Emission Modification in TiO_2 Logpile

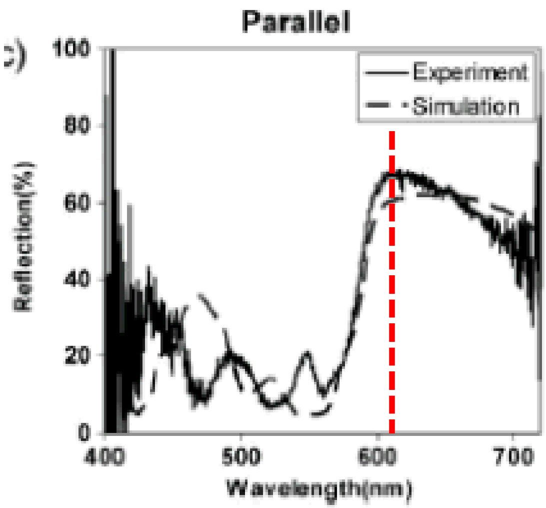
Sandia National Laboratories

CdSe red emitting quantum dots

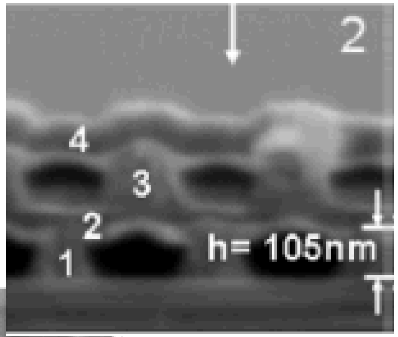


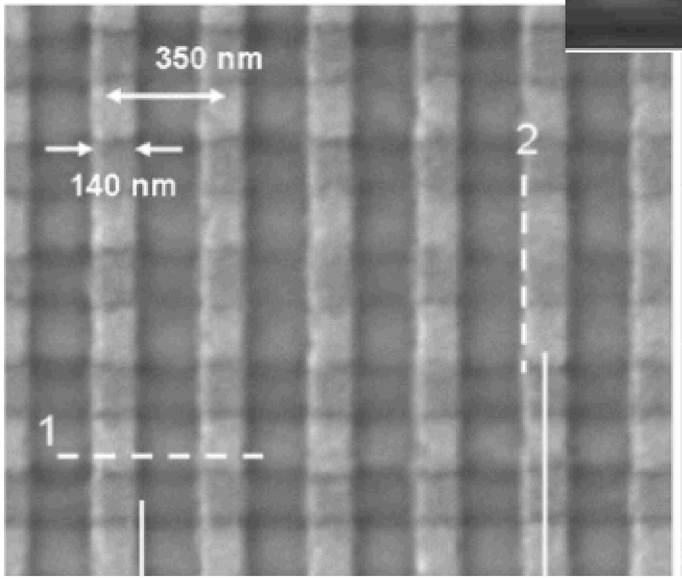
(www.nn-labs.com)





Larger lattice constants a= 350,400 with 4 Layers





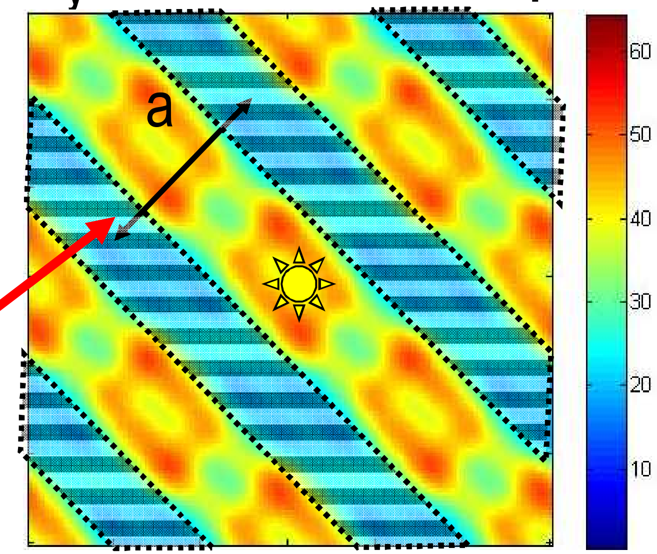
G. Subramania, Y. J. Lee, A. J. Fischer, T. S. Luk, C. J. Brinker, D. Dunphy, Appl. Phys. Lett. , **95**, 151101(2009)

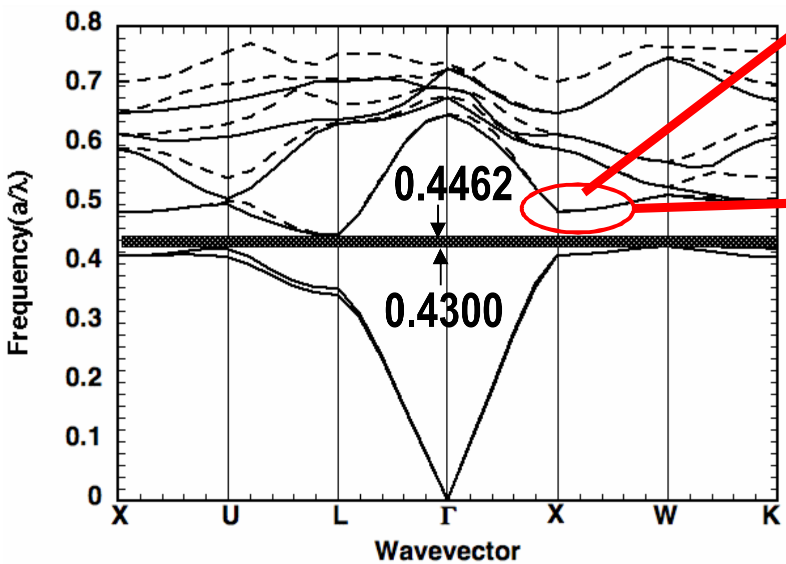
Effect of LDOS- Emitter Placement Inside the PC

Sandia National Laboratories

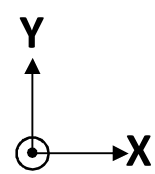
- Thus far emission control studies typically done for emitters in high index region – probing dielectric band
- Effect of 'air band' modes have not been probed
- Advantages
  - Post introduction of light source
  - Choice of light source (QDs or dyes or gas)
  - Potentially higher density of states enhancement

Ey field distribution at Xpt

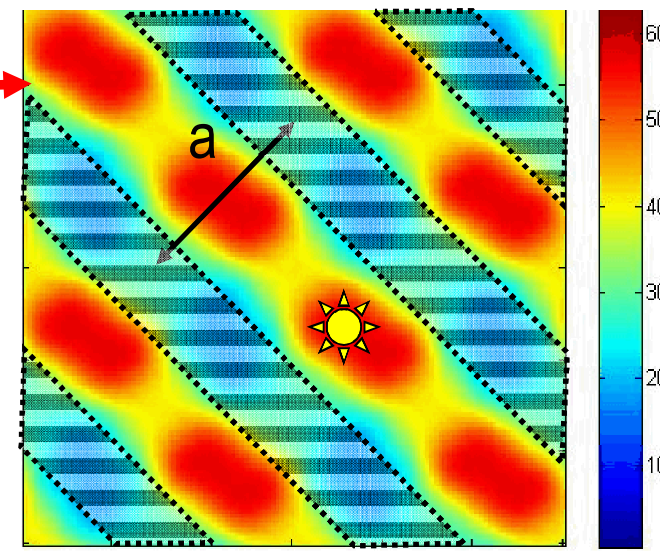




'Air' band



Ez field distribution at Xpt



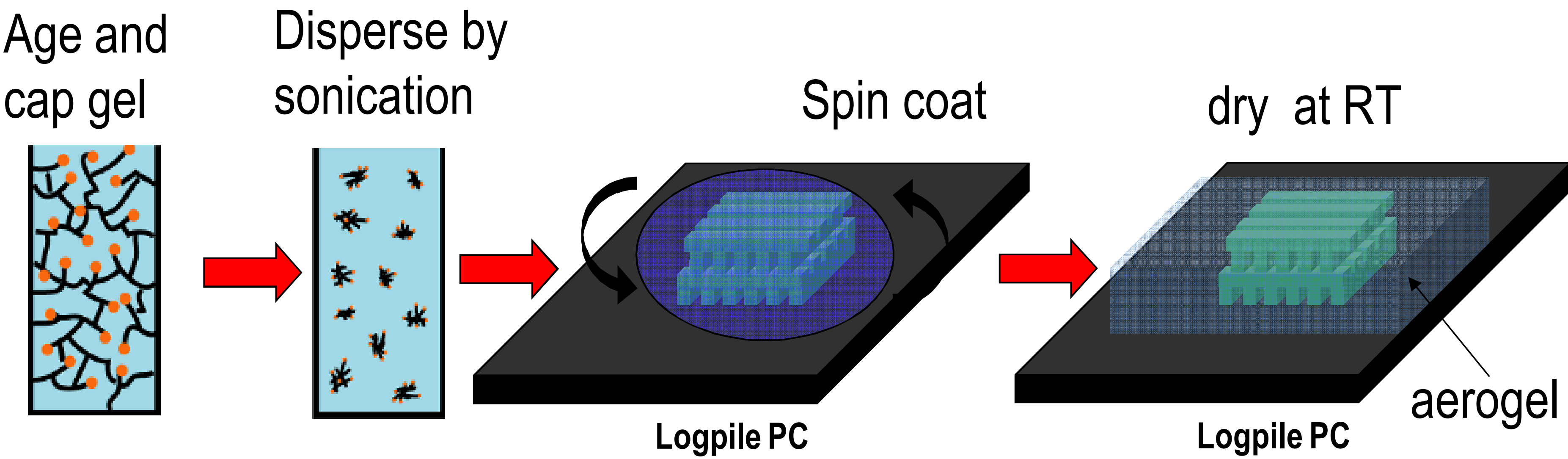
Field Distribution for the 'air band' using PWE method

Requirements for Accessing the 'Air' Void Region of the PC

- **Key Challenge: Locating the QDs in the void region without affecting the photonic bandgap**
- Physical access to the 'air' region with ability to attach QDs
- Low refractive index: minimal disturbance to photonic bandgap

Room temperature Spin Coatable Aerogel

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



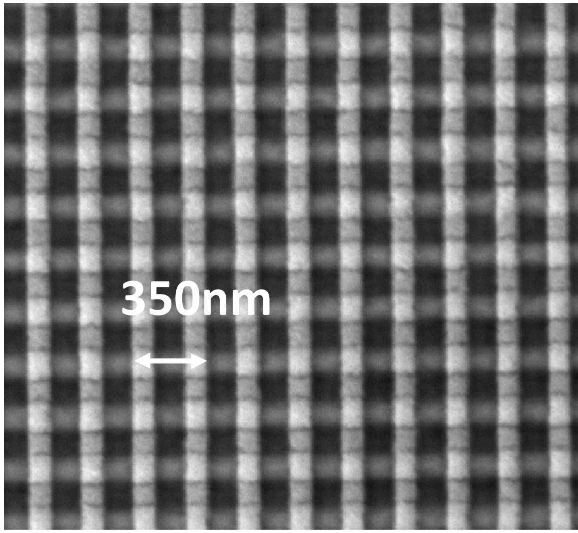
Introduce QDs through chemical attachment (3-mercaptopropyltrimethoxysilane chemistry)

Developed by Alex Lee

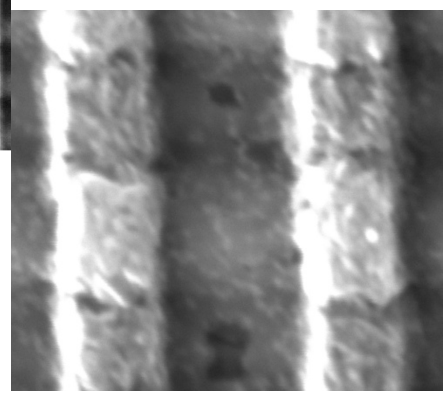
G. Subramania, Y. J. Lee, A. J. Fischer, T. S. Luk, C. J. Brinker, D. Dunphy, Appl. Phys. Lett. , **95**, 151101(2009)

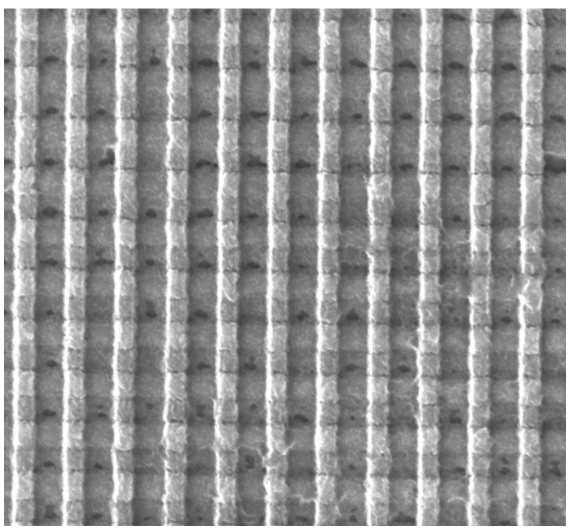
Effect of Aerogel and CdSe QD infiltration

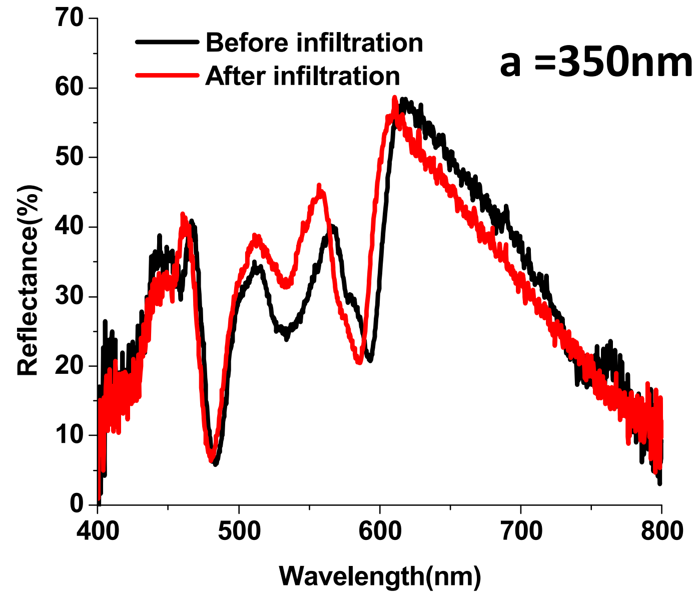
Sandia National Laboratories

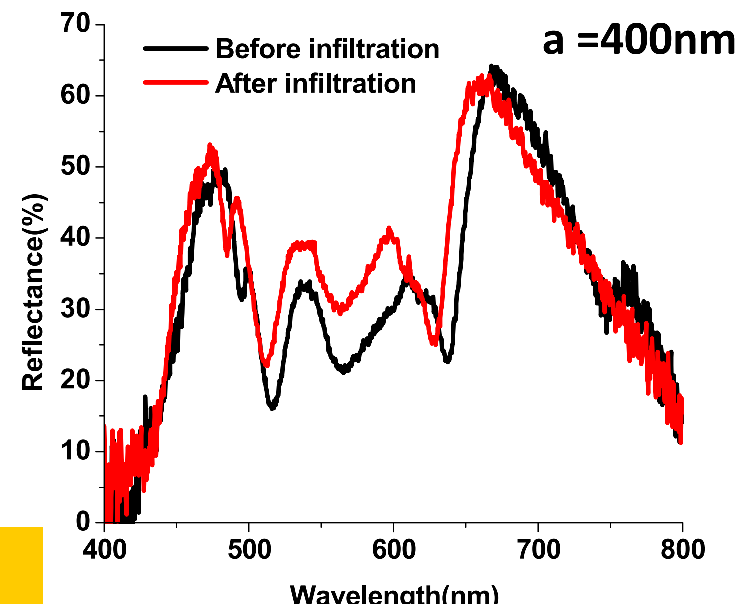


Aerogel Ref. index ~1.05-1.1



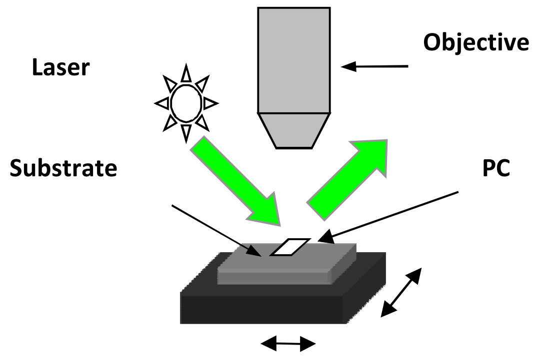






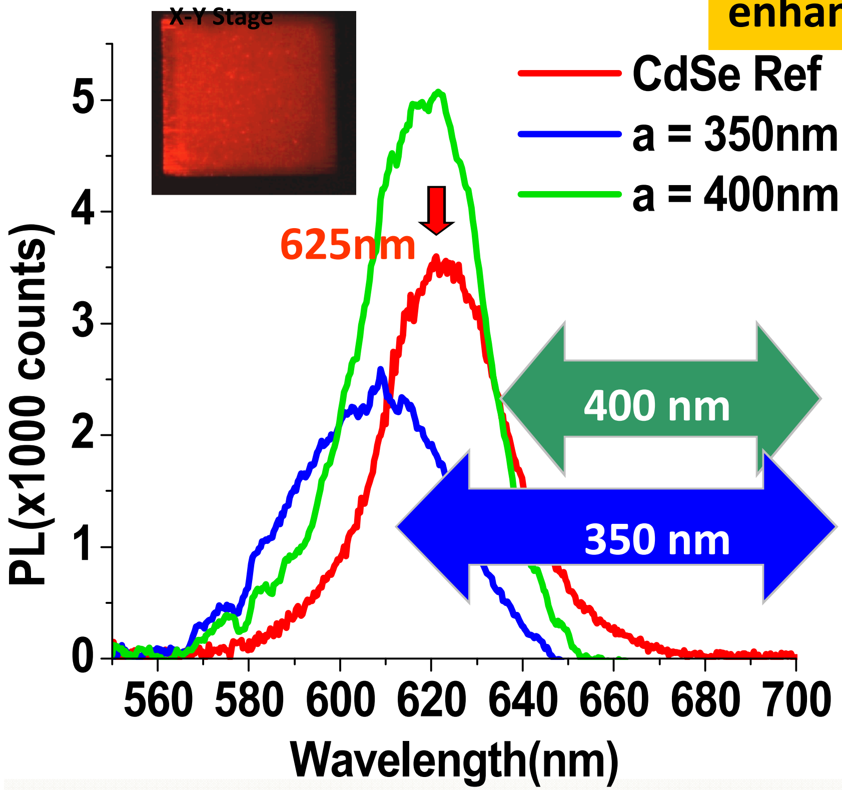
Aerogel and CdSe QD infiltration has negligible effect on the bandgap!

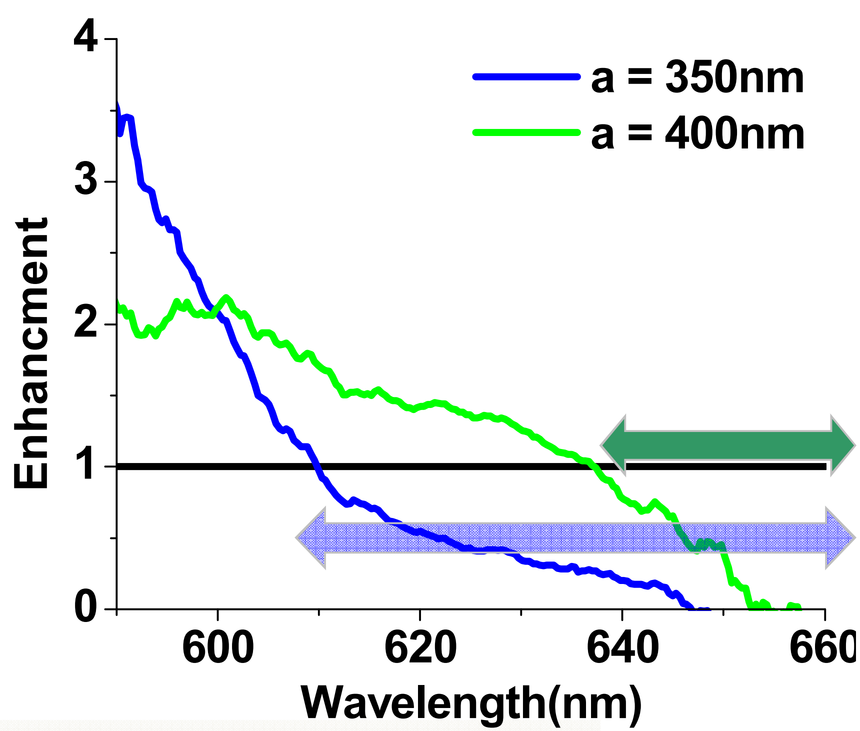
Spontaneous Emission Modification



- ❑ Microphotoluminsecence with 532nm (5mW CW pump)
- ❑ Equivalent volume of unpattened CdSe infiltrated aerogel as reference

Light emission suppression inside the bandgap and enhancement at the band edge

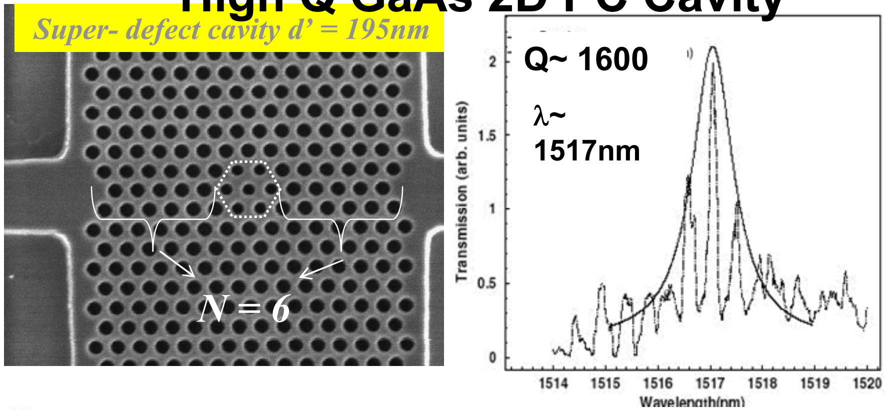




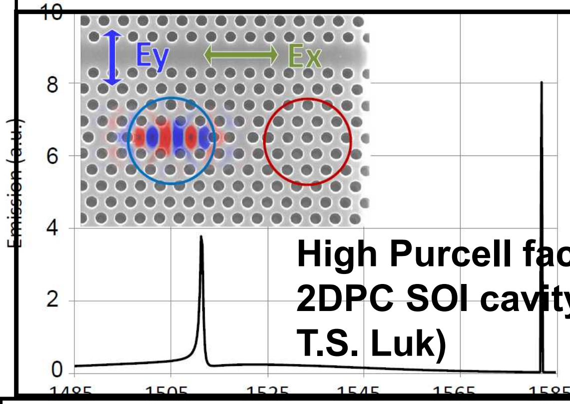
G. Subramania, Y. J. Lee, A. J. Fischer, T. S. Luk, C. J. Brinker, D. Dunphy, Appl. Phys. Lett. , **95**, 151101(2009)

L-M Interaction in 2DPC

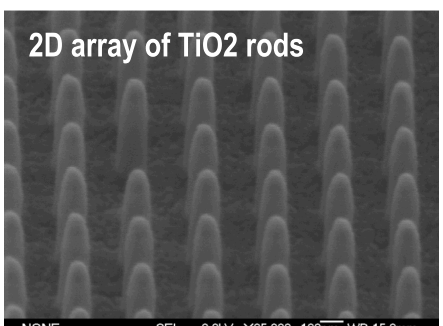
High Q GaAs 2D PC Cavity

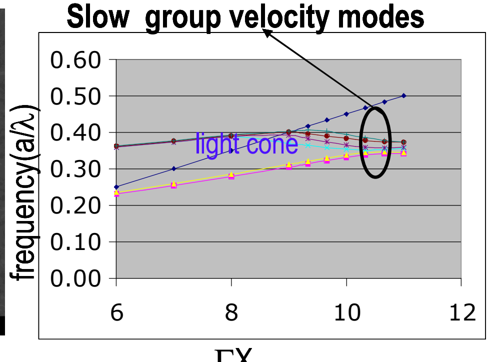


High Purcell factor 2DPC SOI cavity (w/ T.S. Luk)

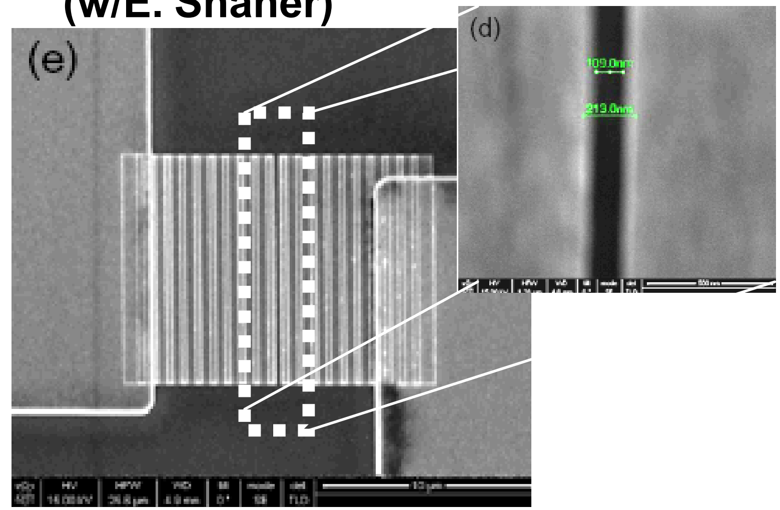


Slow group velocity mode lasing

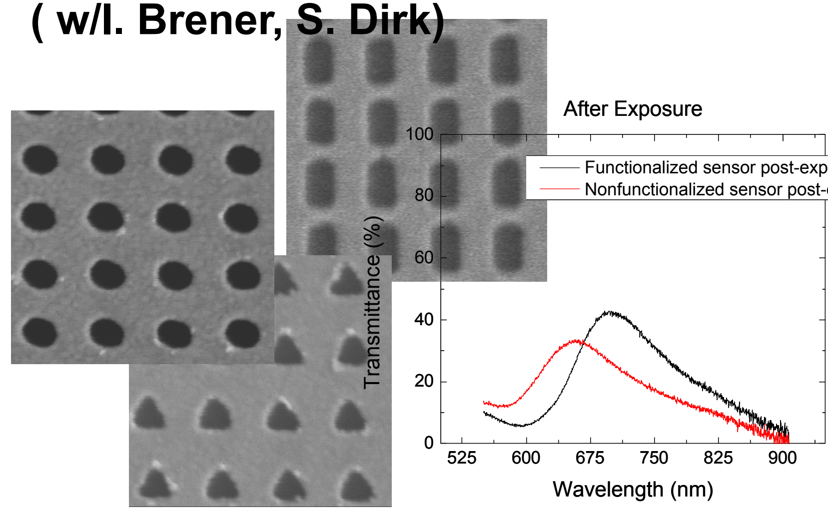




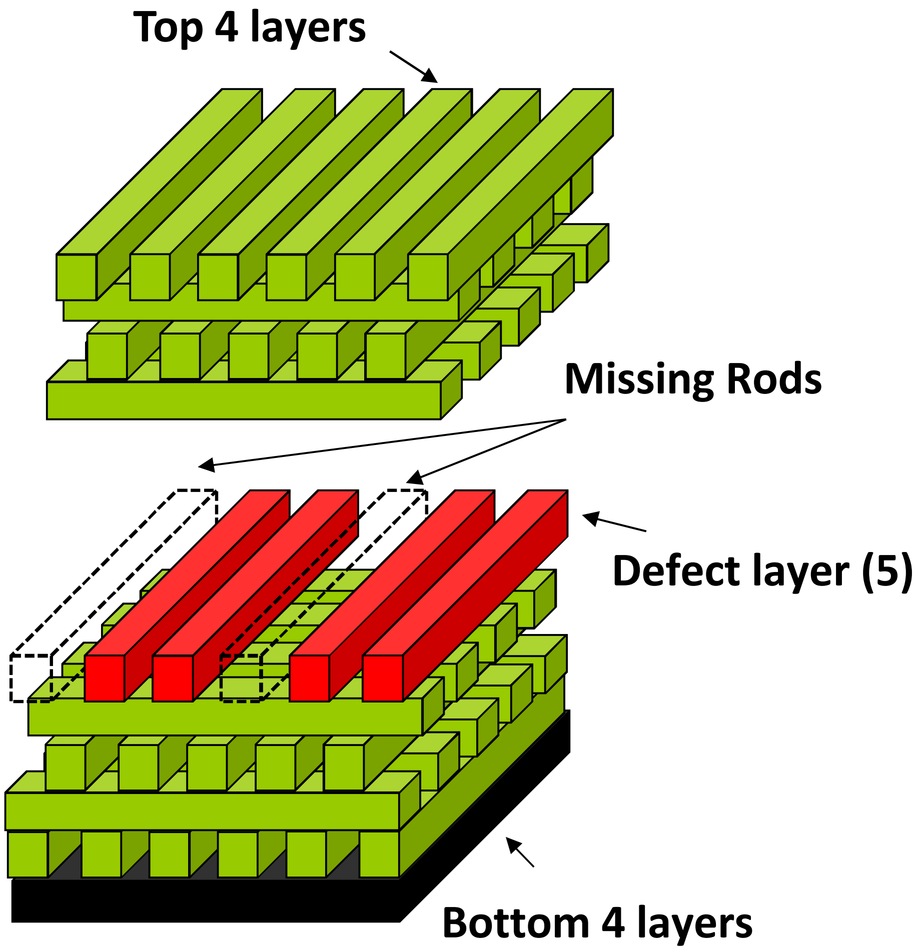
Surface plasmon Auston Switch (w/E. Shaner)

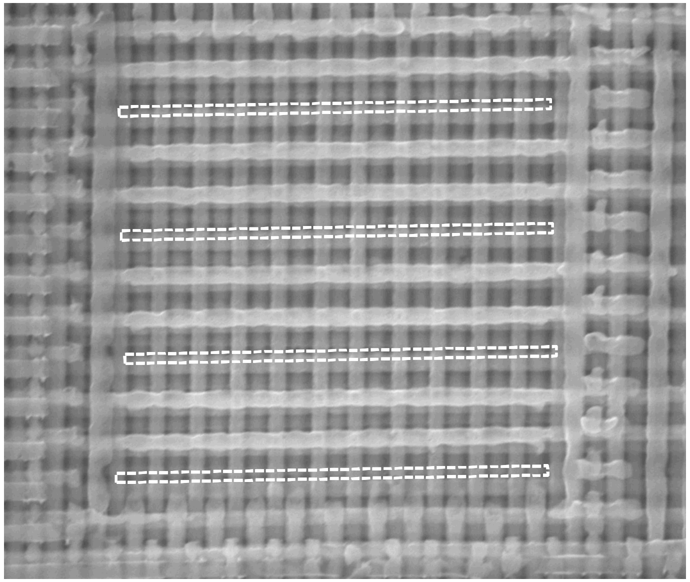


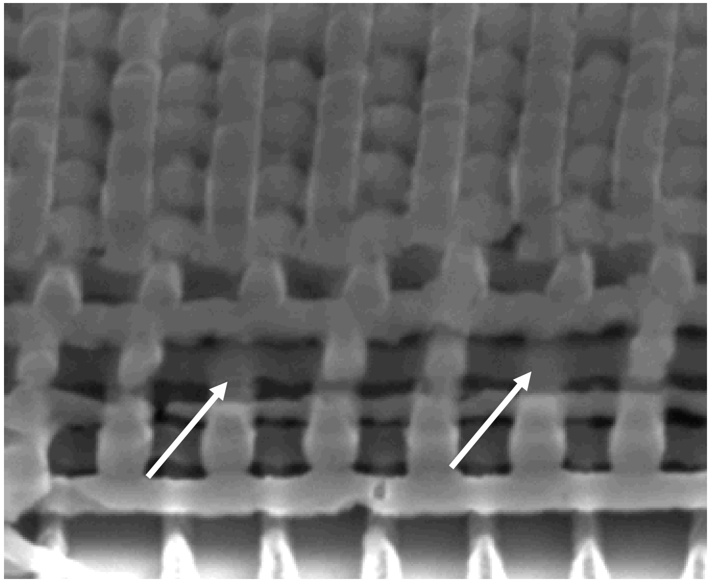
Nanhole EOT based sensors (w/I. Brener, S. Dirk)



Logpile PC Cavity



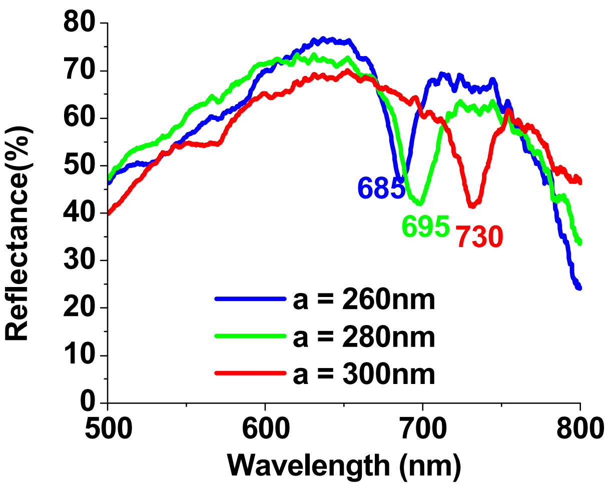




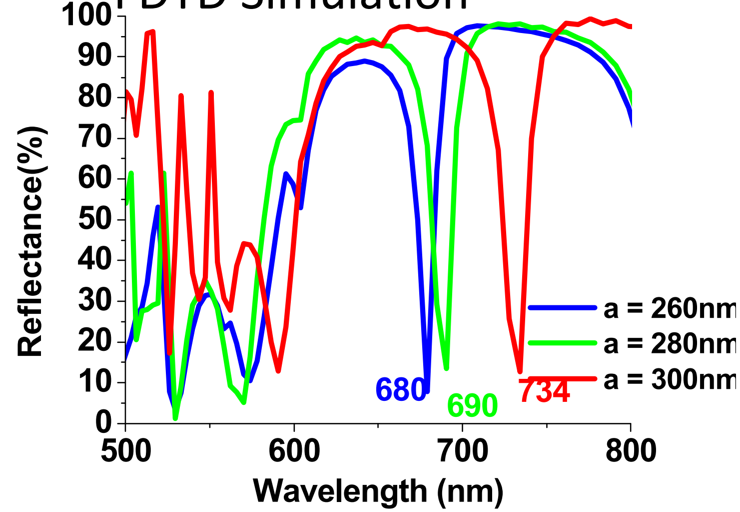
Logpile PC Cavity Optical Response

Reflection

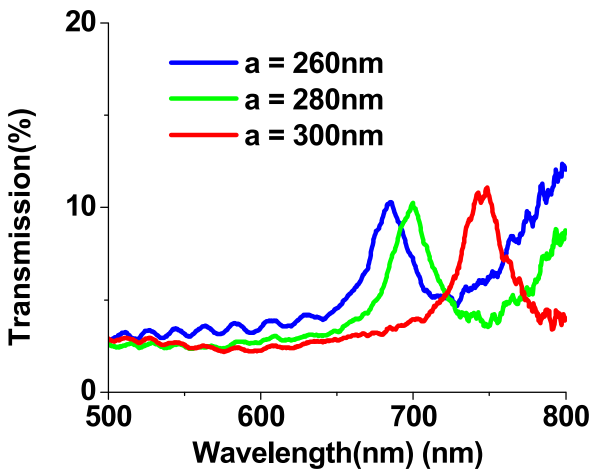
Experiment

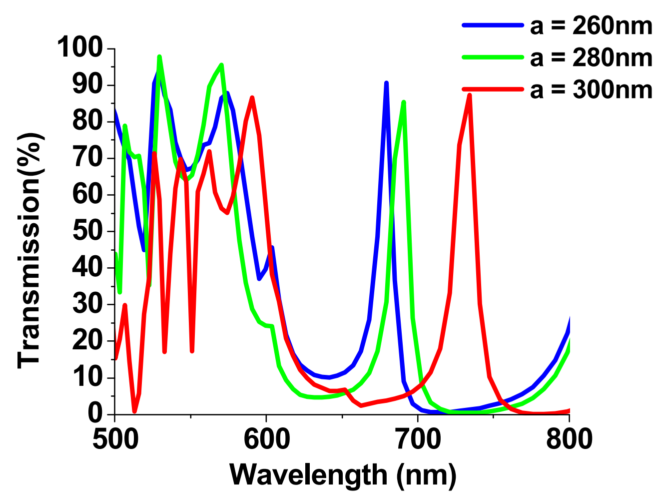


FDTD Simulation



Transmission

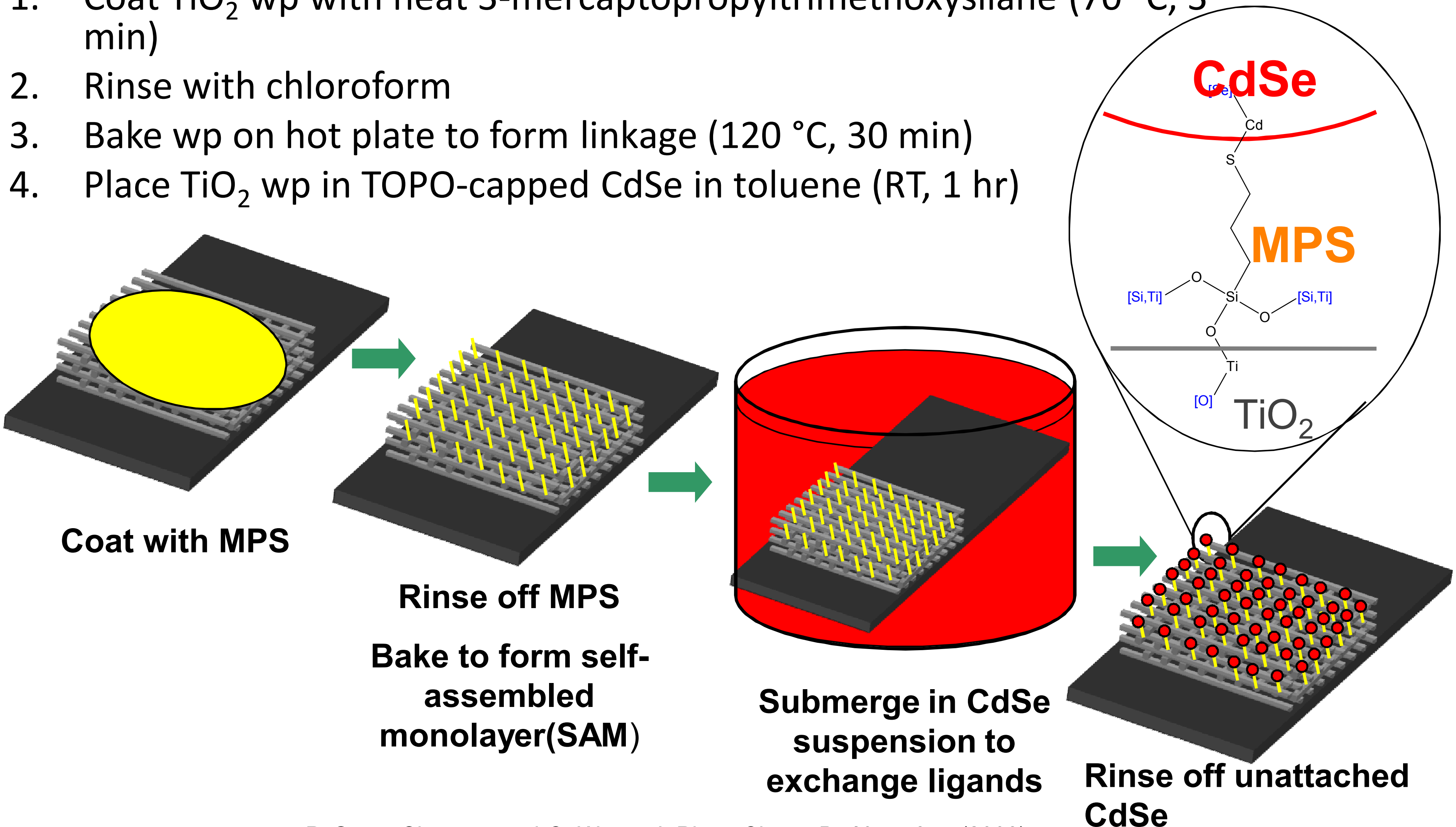




G. Subramania et. al. , Nano Lett. **11**, 4591 (2011).

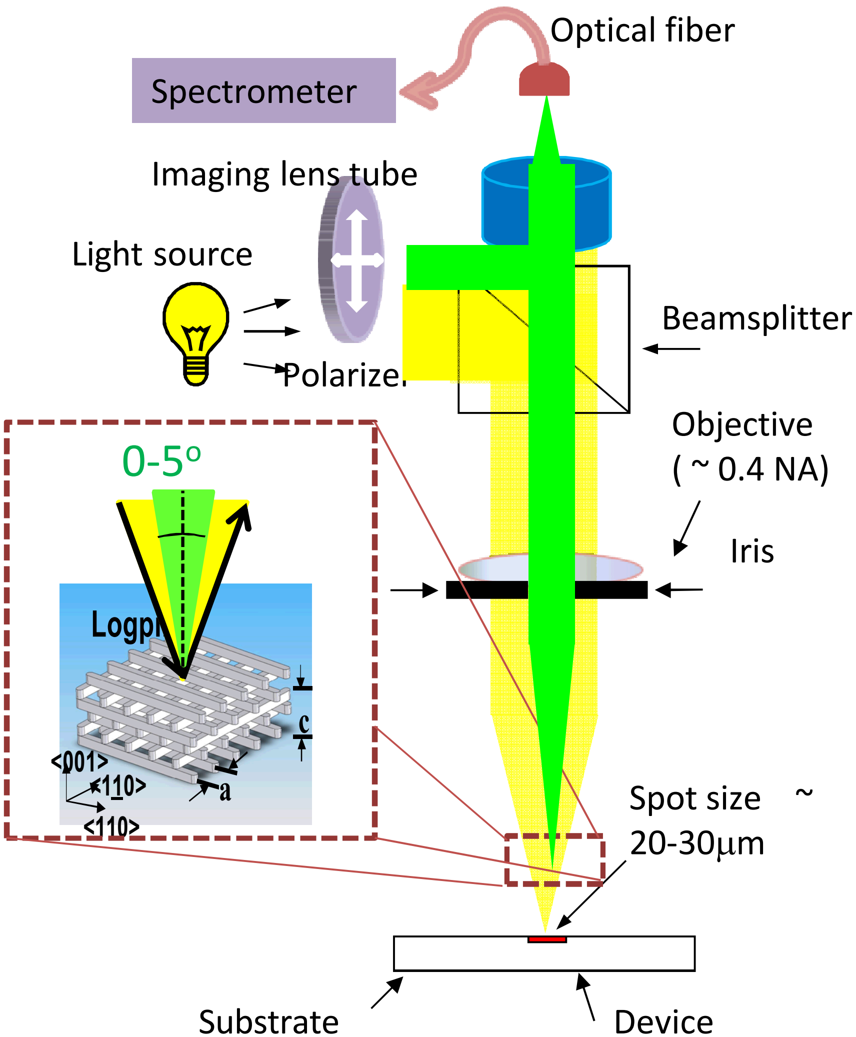
Development of Controlled Coating of Cdse Self-assembled Monolayer On TiO_2 Rods

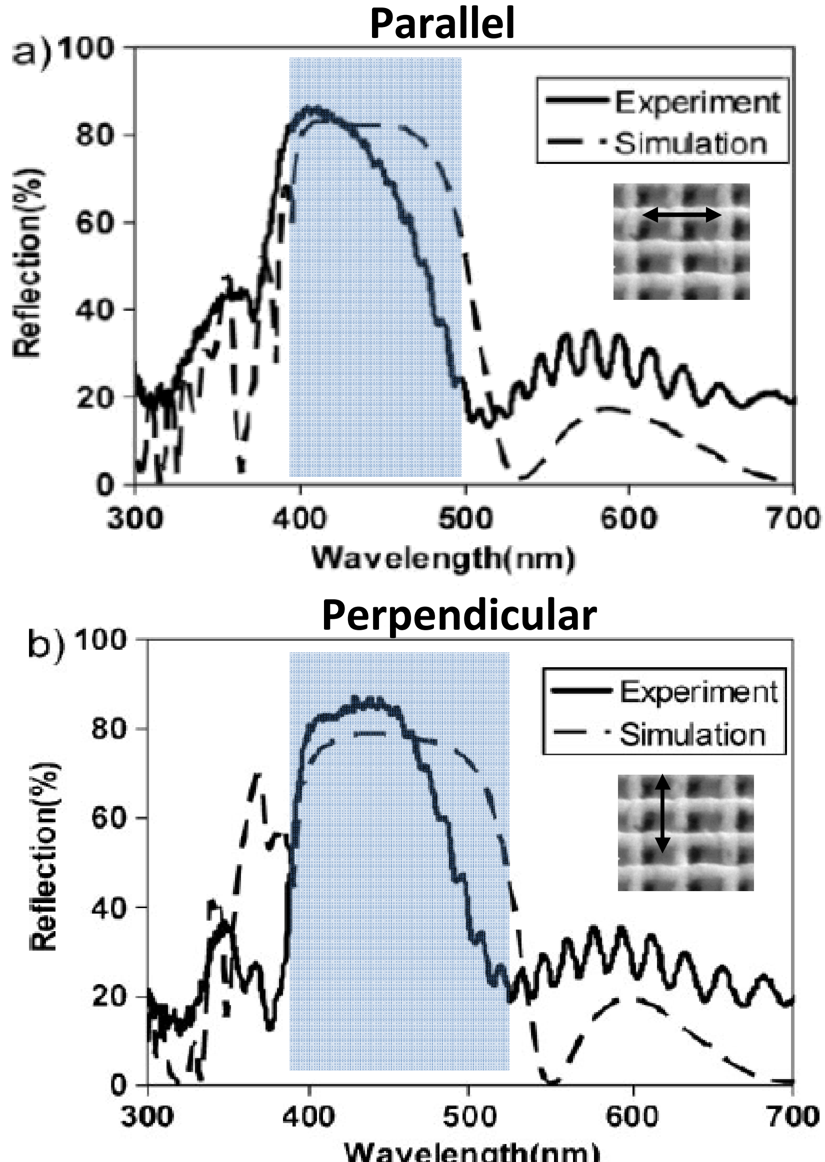
1. Coat TiO_2 wp with neat 3-mercaptopropyltrimethoxysilane (70 °C, 3 min)
2. Rinse with chloroform
3. Bake wp on hot plate to form linkage (120 °C, 30 min)
4. Place TiO_2 wp in TOPO-capped CdSe in toluene (RT, 1 hr)



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)

Visible- Near UV 9 Layer TiO_2 Logpile





G. Subramania, Y.-J. Lee, A. J. Fischer, and D. D. Koleske, Advanced Materials **22**, 487-491 (2010)

Band Structure Calculation

Solve Electromagnetic wave equation for Plane wave

$$\begin{cases} E(r,\ t) \propto e^{-i\omega t} \\ H(r,\ t) \propto e^{-i\omega t} \end{cases}$$

$$\nabla \times \left(\frac{1}{\varepsilon(r)} \nabla \times H(r) \right) = \left(\frac{\omega}{c} \right)^2 H(r)$$

In a periodic potential

$$\varepsilon(r + R) = \varepsilon(r)$$

$$R = a_1\hat{x} + a_2\hat{y} + a_3\hat{z}$$

Apply Bloch's Theorem

$$E, H(r + R) = E, H(r)$$

Plane Wave Expansion Method

Sandia National Laboratories

Reciprocal (K) space

Reciprocal Lattice vector

$$\varepsilon(r) = \sum_G \bar{\varepsilon}(G) e^{iG.r} \qquad E,H(r) = \sum_G \overline{E},\overline{H}(G) e^{-i(-k+G).r} \qquad G \cdot R = 2\pi n$$

Bloch Waves $\overline{H}(G) = H_x(G)\hat{x} + H_y(G)\hat{y} + H_z(G)\hat{z}$

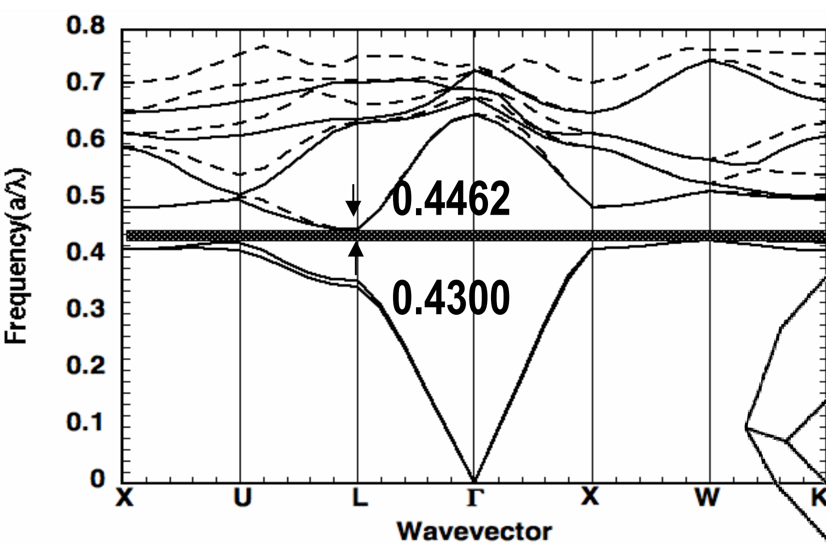
Eigen value equation

$$\nabla \times \left(\frac{1}{\varepsilon(r)} \nabla \times H(r) \right) = \left(\frac{\omega}{c} \right)^2 H(r) \quad \Rightarrow \quad A\overline{H}_n(k) = -\frac{a^2 \omega_n^2}{c^2} \overline{H}_n(k)$$

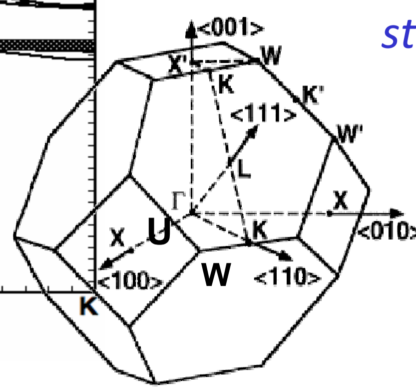
Band index

Wave vector within the first BZ

Photonic Band Structure for TiO_2 logpile



1st BZ



Eigen value equation becomes a matrix equation for which you can write a code and solve for band structure within the first Brillouin zone.

- **n ~ 2.3**
- **4% bandgap**
- **~ 20nm @ 500nm**