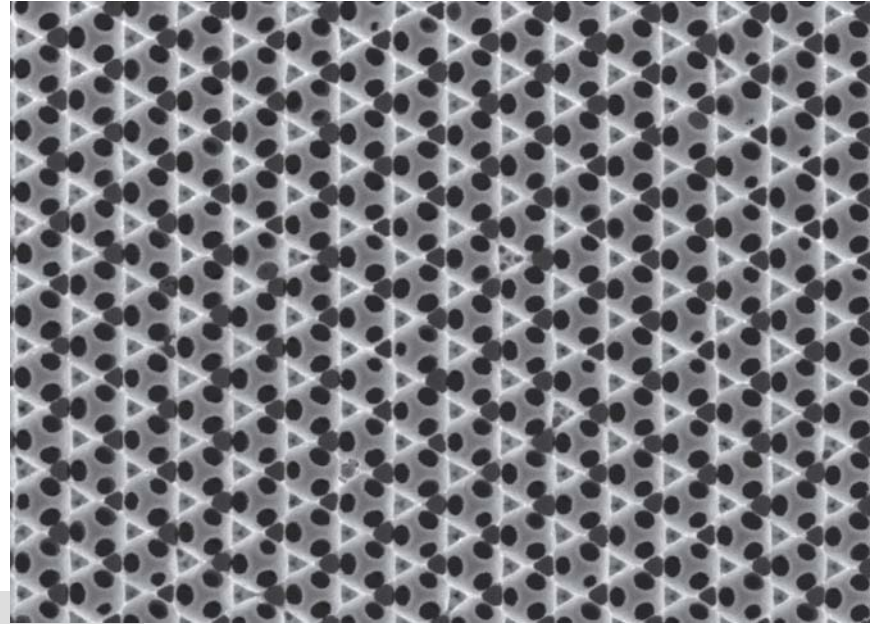


The Photonic Band Gap and Colloidal Crystals

David J. Norris

Chemical Engineering & Materials Science
University of Minnesota



Focus: Photonic Band Gap

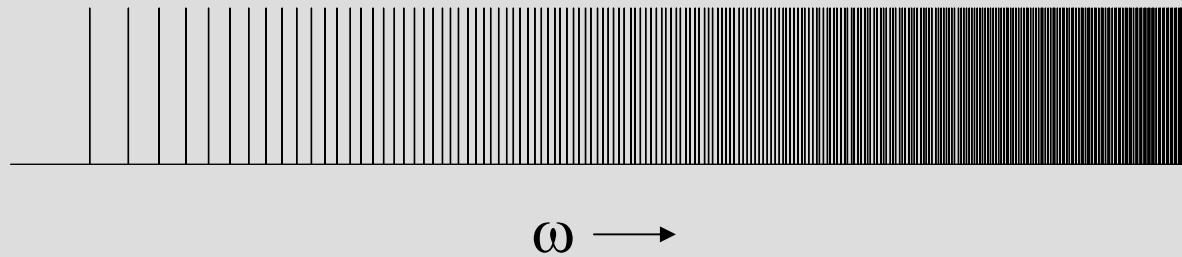
- What is it?
- Why is it interesting?
- How do colloidal particles fit in?

Your job is to ask questions as we go!

To understand the photonic band gap:
start from a basic concept . . .

Density of States (DOS)

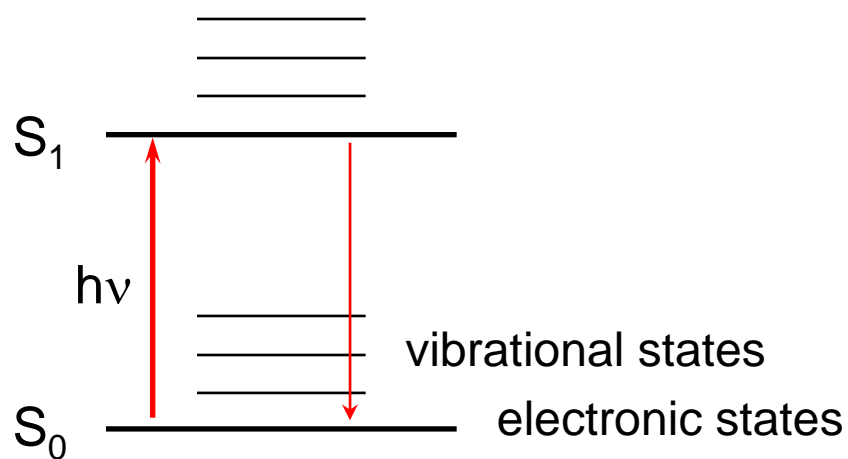
$$\rho(\omega) = \frac{\text{Number of states}}{\text{Unit frequency} \times \text{Unit Volume}}$$



States can be electronic, vibronic (phonons), or optical (photonic)

Why does the electronic DOS matter?

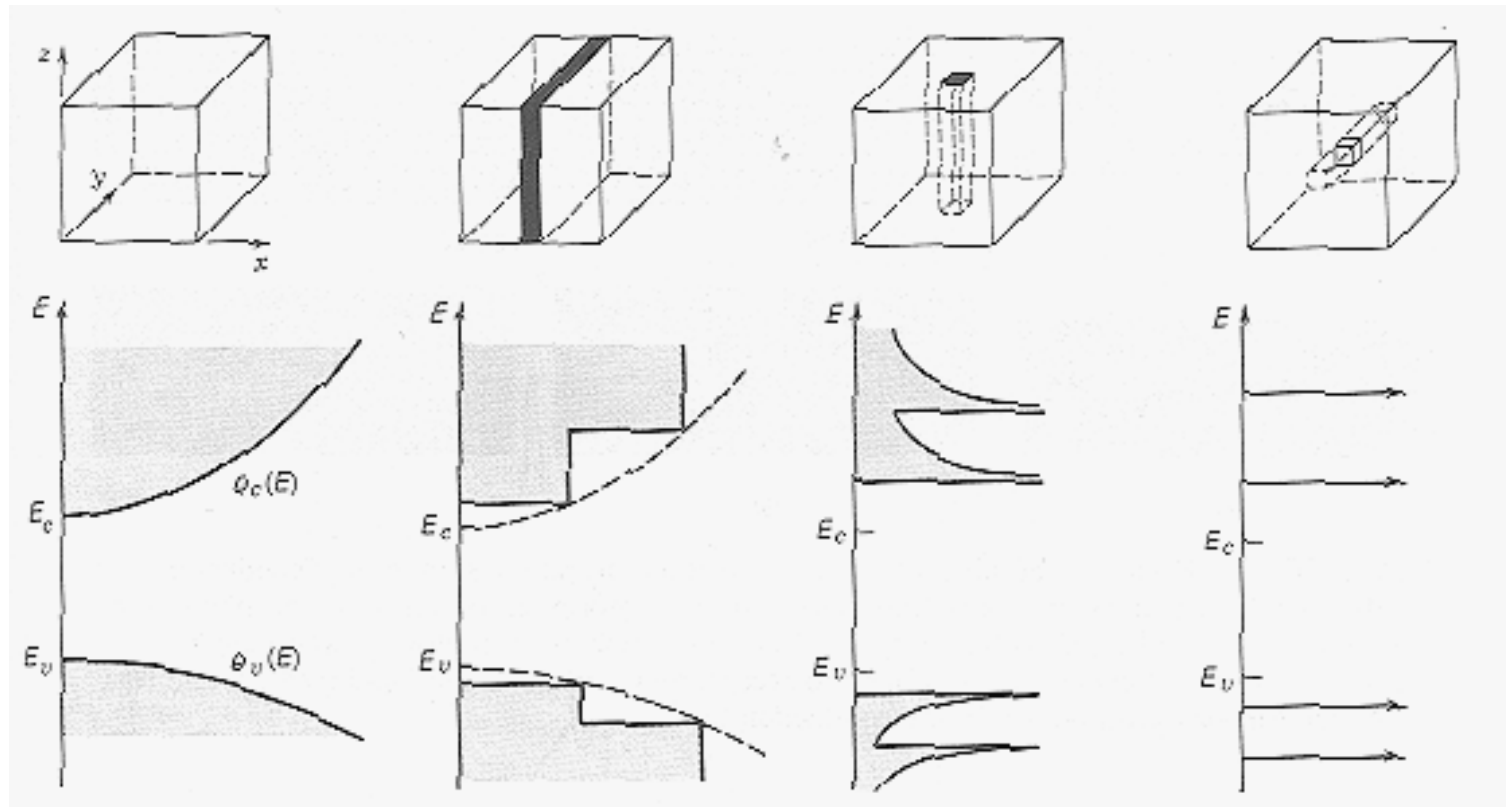
$$\text{Fermi's Golden Rule} \\ \text{Rate} \equiv \Gamma_0 \propto \rho(\omega)$$



i.e., “speed” of a process depends on the number of available states

Controlling Electronic DOS:

Early motivation for nano-structures

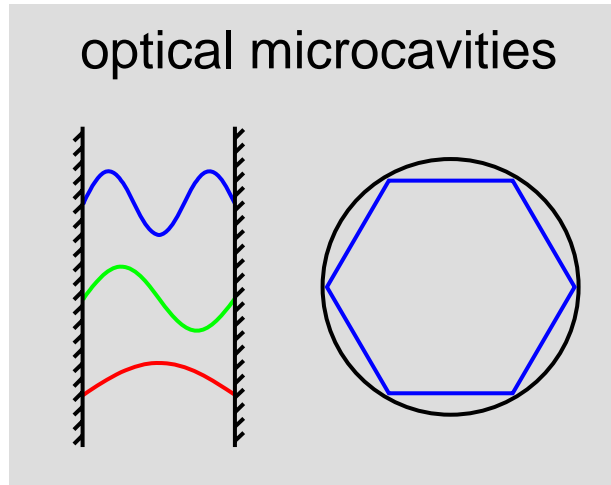


modification of electronic density of states

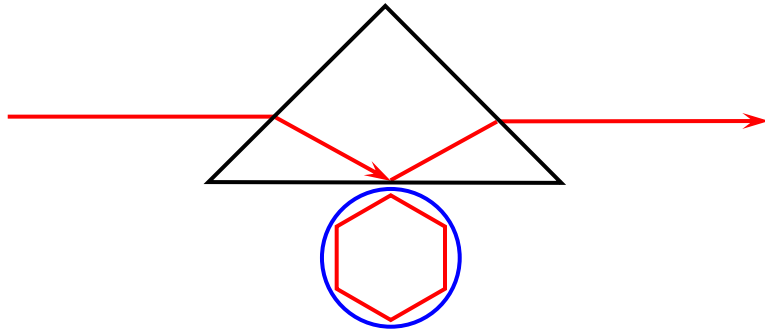
Saleh

What about the photonic density of states?

$$\rho_{\text{phot}}(\omega) = \frac{\text{Number of photon states}}{\text{Unit frequency} \times \text{Unit Volume}}$$



Cool lab demo: “marble” microcavity



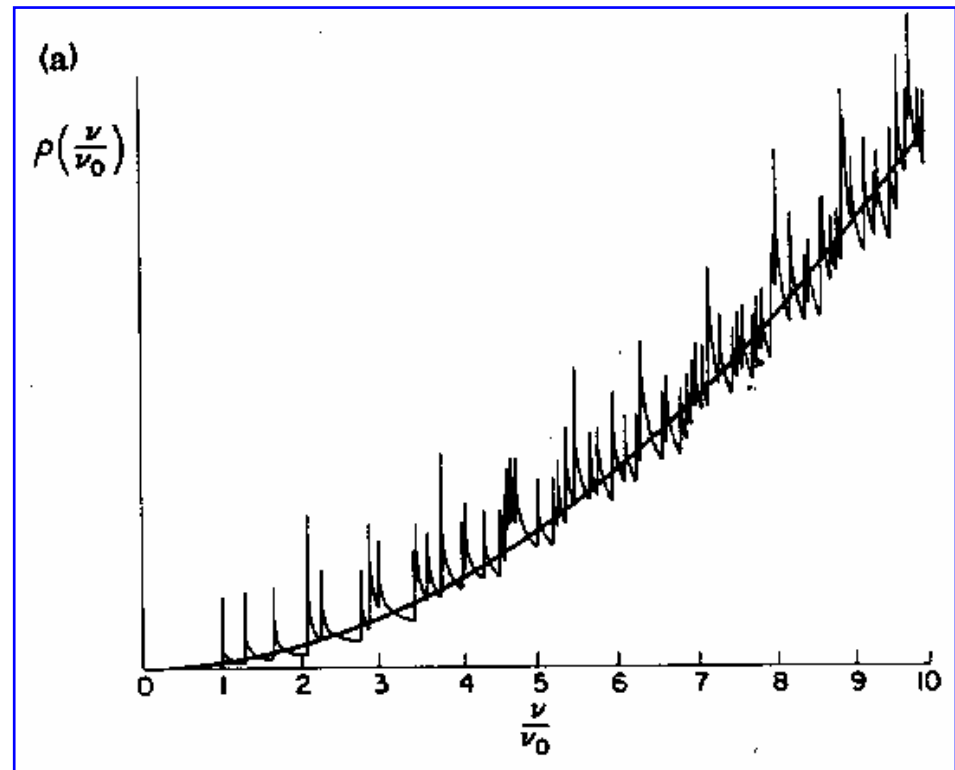
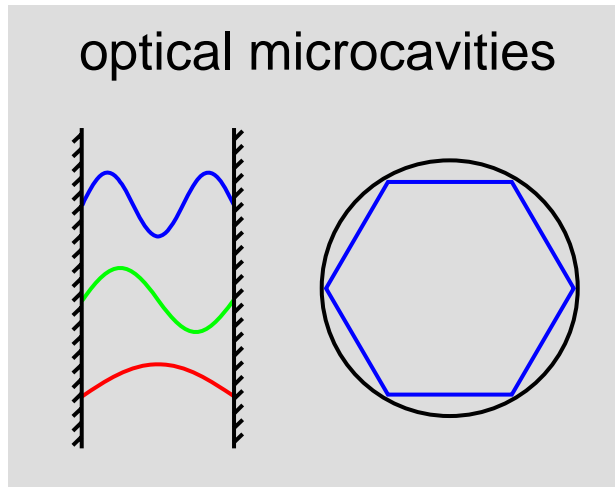
- Total internal reflection off prism
- Photons leak into the cavity
- Circulate in cavity mode



Controlling Photonic Density of States

Early motivation for photonic microstructures

$$\rho_{\text{phot}}(\omega) = \frac{\text{Number of photon states}}{\text{Unit frequency} \times \text{Unit Volume}}$$



Kleppner, Phys. Rev. Lett. **47**, 233 (1981).

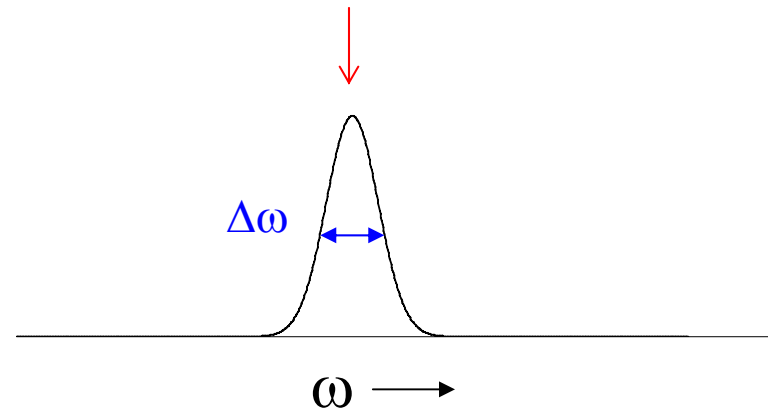
Purcell Effect - Control Spontaneous Emission

B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. PURCELL, *Harvard University*.—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

$$A_p = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2) \text{ sec.}^{-1},$$

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu = 10^7 \text{ sec.}^{-1}$, $\mu = 1$ nuclear magneton, the corresponding relaxation time would be 5×10^{21} seconds! However, for a system coupled to a resonant electrical circuit, the factor $8\pi\nu^2/c^3$ no longer gives correctly the number of radiation oscillators per unit volume, in unit frequency range, there being now *one* oscillator in the frequency range ν/Q associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor $f = 3Q\lambda^3/4\pi^2V$, where V is the volume of the resonator. If a is a dimension characteristic of the circuit so that $V \sim a^3$, and if δ is the skin-depth at frequency ν , $f \sim \lambda^3/a^2\delta$. For a non-resonant circuit $f \sim \lambda^3/a^3$, and for $a < \delta$ it can be shown that $f \sim \lambda^3/a\delta^2$. If small metallic particles, of diameter 10^{-3} cm are mixed with a nuclear-magnetic medium at room temperature, spontaneous emission should establish thermal equilibrium in a time of the order of minutes, for $\nu = 10^7 \text{ sec.}^{-1}$.

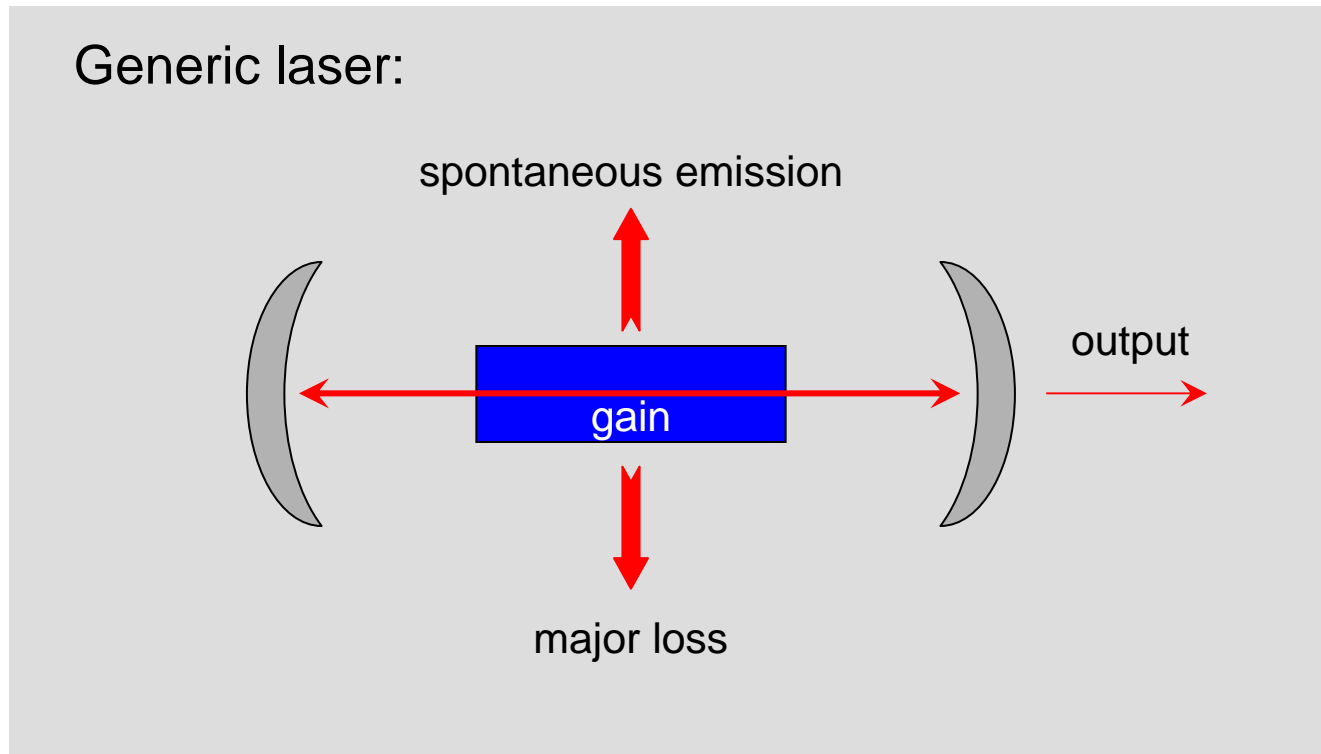
$$\Gamma_o \propto \rho_{\text{elec}}(\omega) \cdot \rho_{\text{phot}}(\omega)$$



$$\eta = \frac{\Gamma_c}{\Gamma_o} \propto \frac{Q \lambda^3}{V_c}$$

Purcell, Phys. Rev. **69**, 681 (1946).

Device Implications:



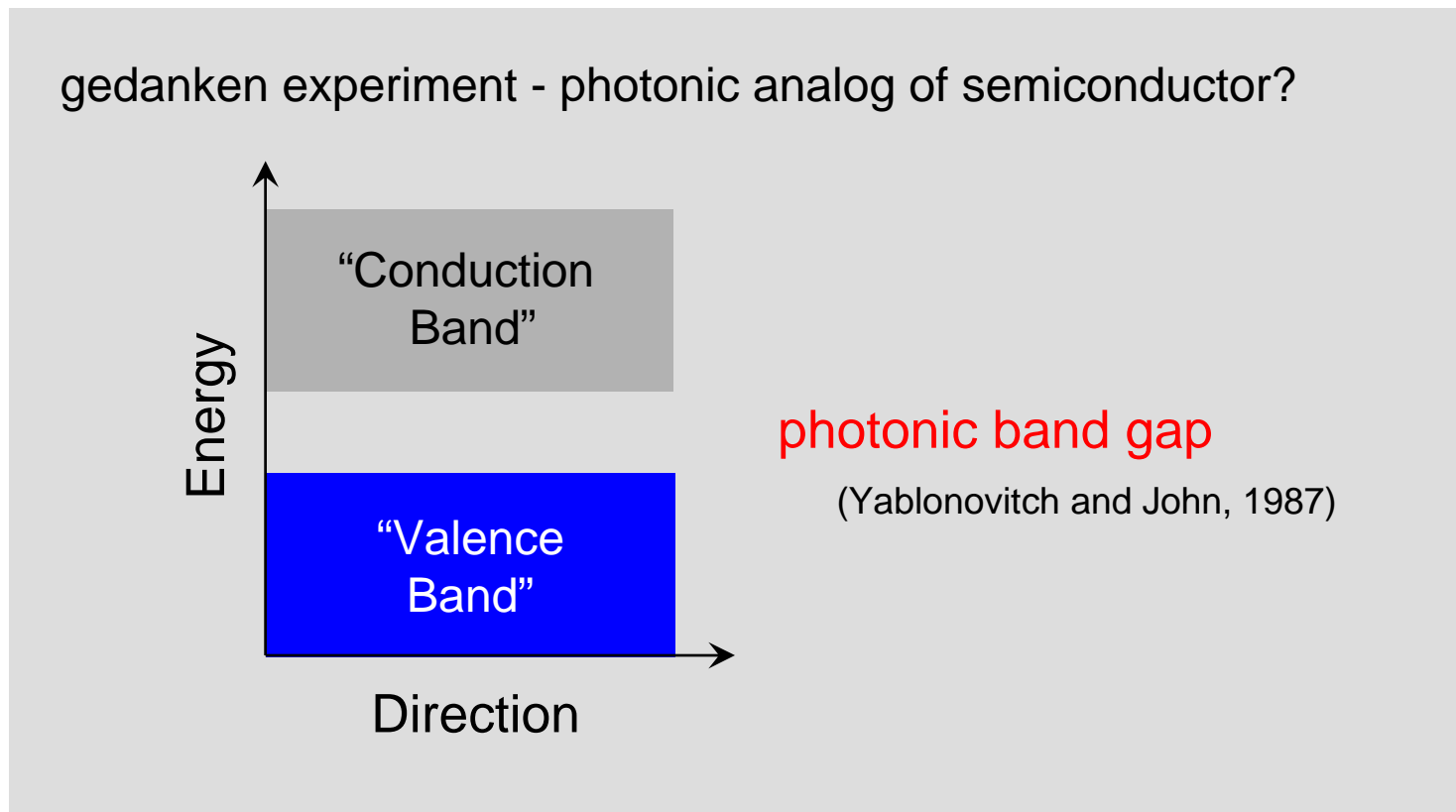
Early idea:

Control spontaneous emission = better laser

How do we do this?

Optical microcavities are one option . . .

but is there another approach?

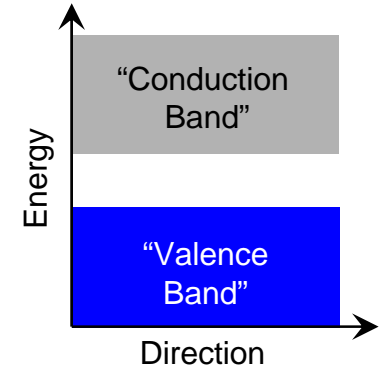
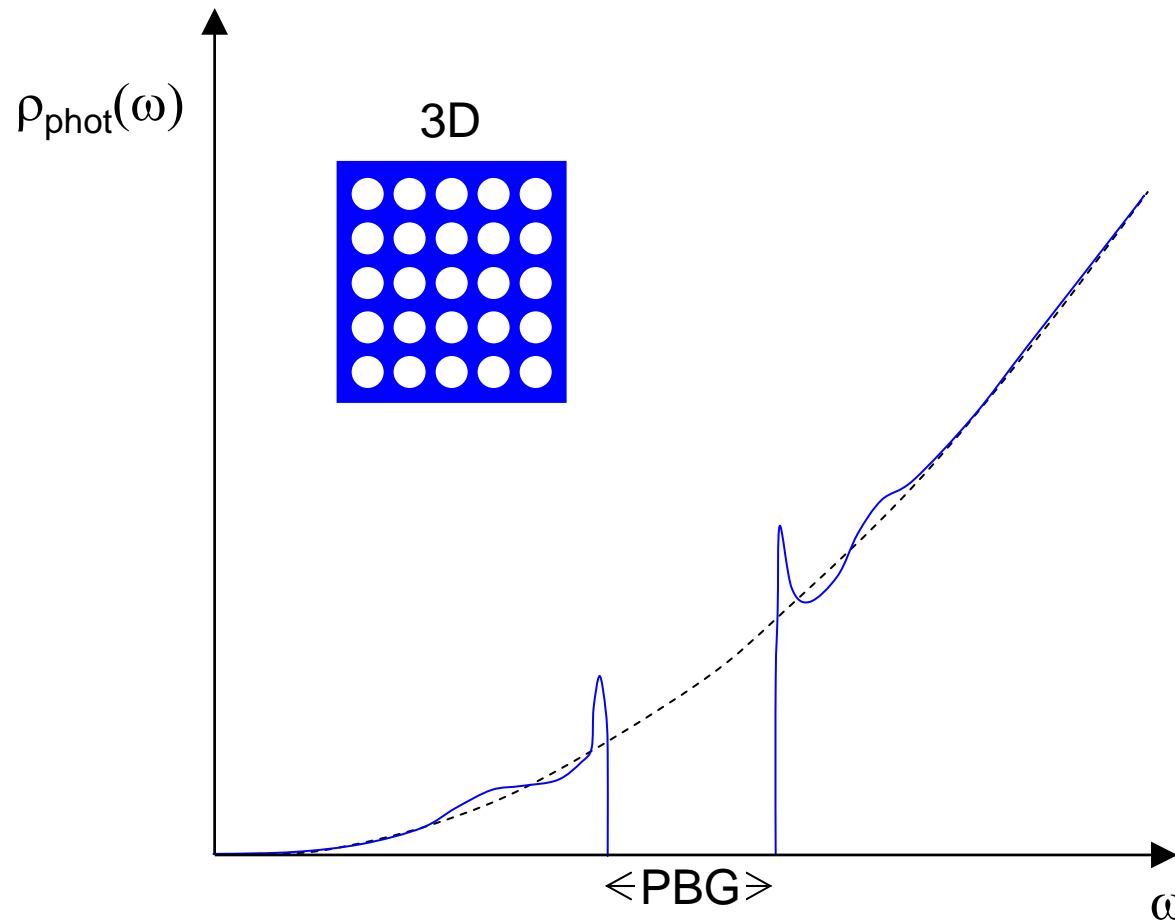


Can such materials exist?

What are the implications of a photonic band gap?

Photonic Band Gap:

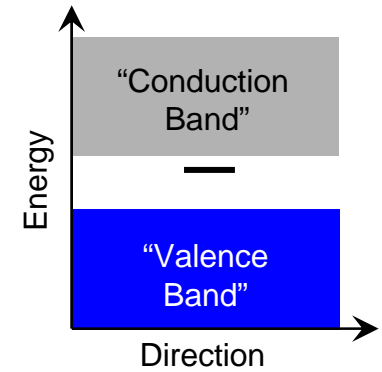
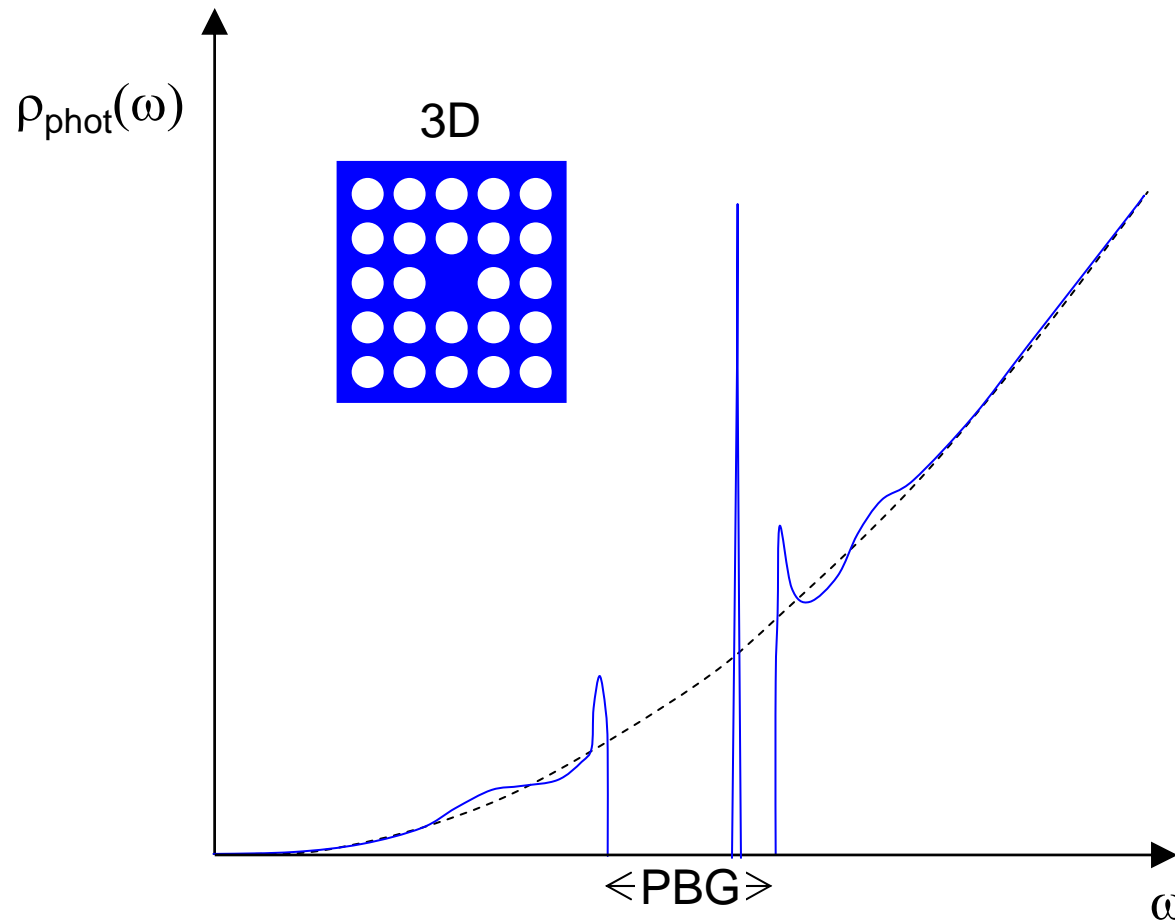
Ultimate control over the photonic density of states



$$\Gamma_o \propto \rho_{\text{elec}}(\omega) \cdot \rho_{\text{phot}}(\omega)$$

Photonic Band Gap:

Ultimate control over the photonic density of states



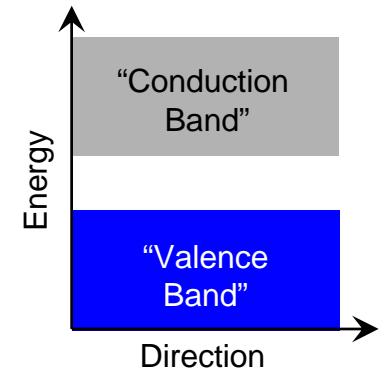
$$\Gamma_o \propto \rho_{\text{elec}}(\omega) \cdot \rho_{\text{phot}}(\omega)$$

Photonic Band Gap:

Ultimate control over the photonic density of states

Implications:

Control photonic density of states
From zero to extremely high values!

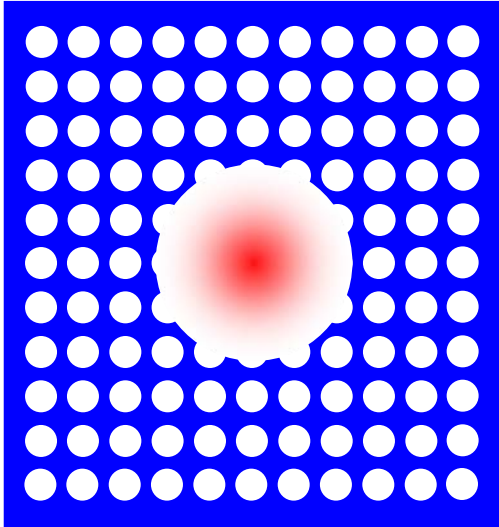


$$\Gamma_o \propto \rho_{\text{elec}}(\omega) \cdot \rho_{\text{phot}}(\omega)$$

But what causes a material to have a PBG?

Photonic Band Gap:

3D PBG Material



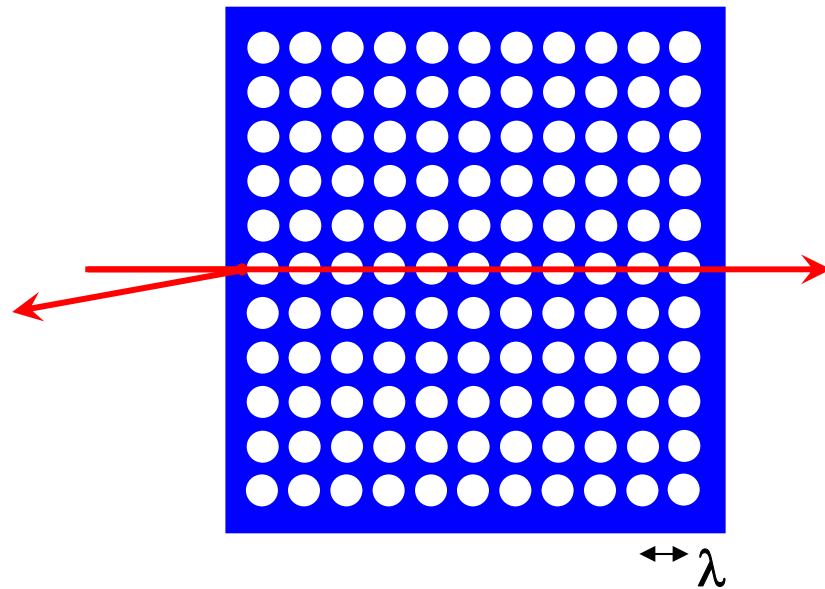
Structure always scatters light backward

- Light cannot travel through the material
- Because density of states is zero
- Instead, light is Bragg diffracted backwards
- Light can become trapped at a defect site where the density of states is high

Strong Bragg Diffraction:

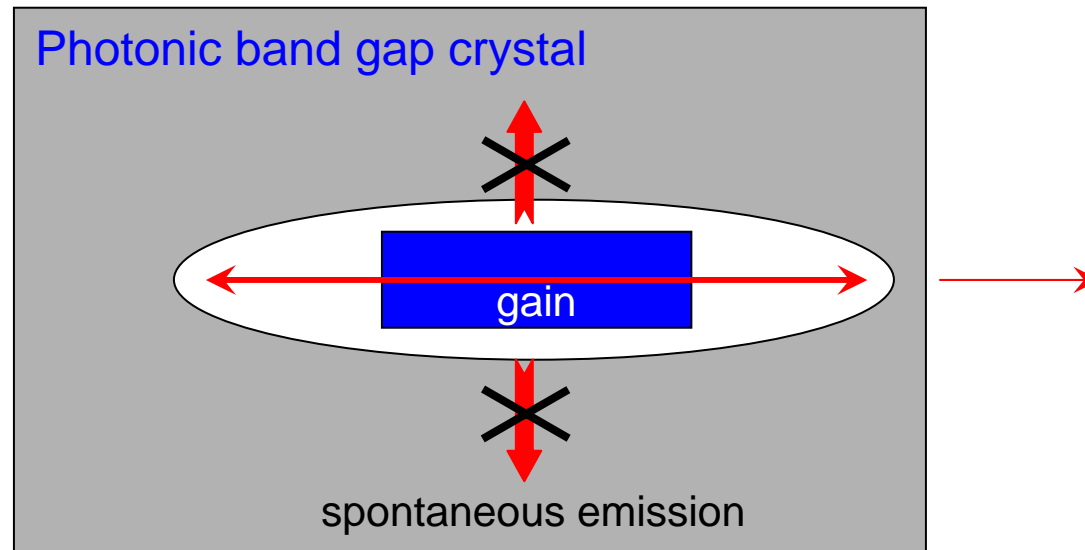
Implies periodic structure is necessary:

- Analogy with X-ray diffraction
- Here diffracted wave is an optical beam
- Thus, photonic crystal must be periodic on an optical length scale
- Need strong scattering, so must have a high refractive index contrast



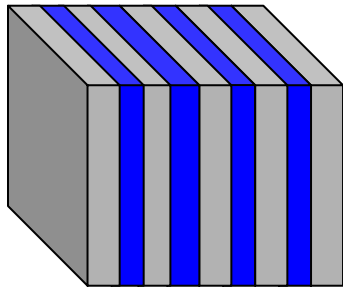
Yablonoitch's original idea:

Device inside photonic crystal:

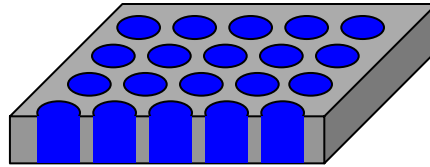


Complete control over wasteful spontaneous emission in unwanted directions

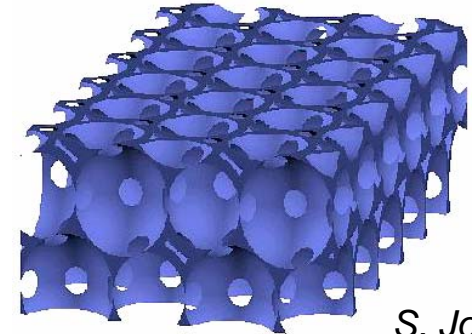
Photonic crystals: experiment



1D



2D



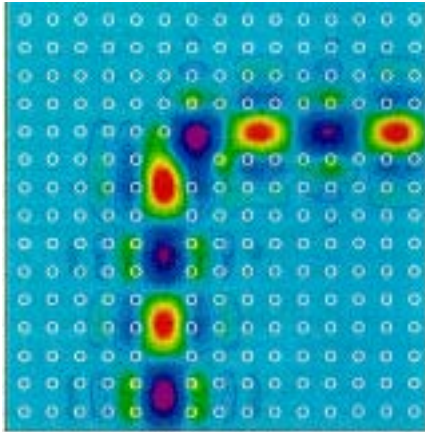
S. John

3D

Original Idea: 3D Crystal

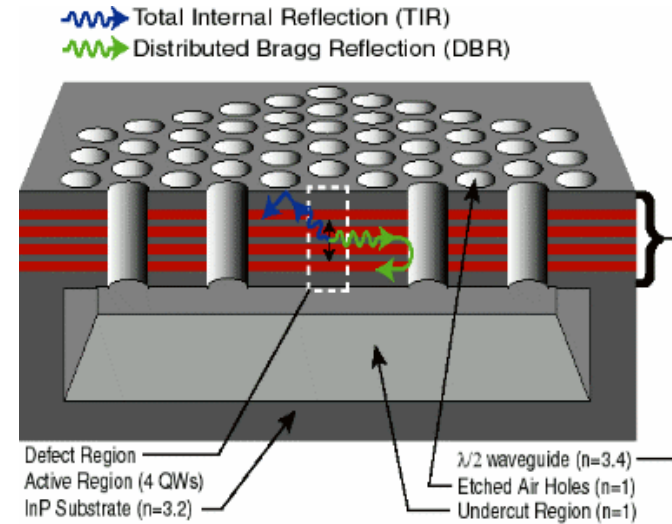
- Only 3D crystal can have a “complete” PBG
- But difficult to fabricate
- Researchers explored 1D and 2D crystals

Tremendous progress in 2D photonic crystals



photonic crystal
waveguides

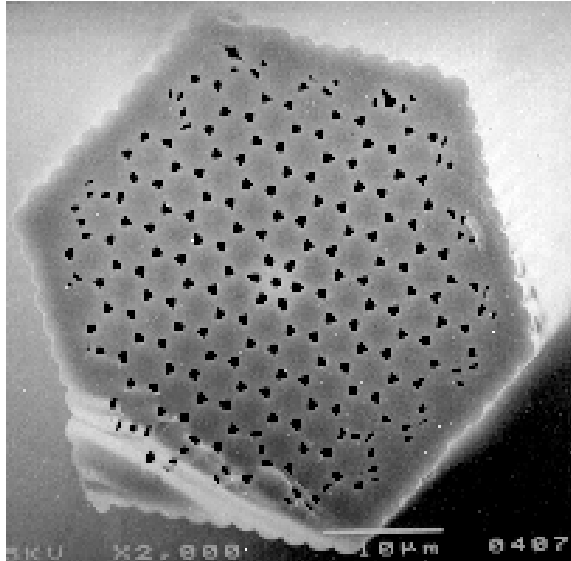
Mekis et al., PRL 77, 3787 (1996)



photonic crystal
laser

Painter et al., Science 284, 1819 (1999)

Photonic Crystal Fibers



Russell Group (Bath)

photonic crystal fibers

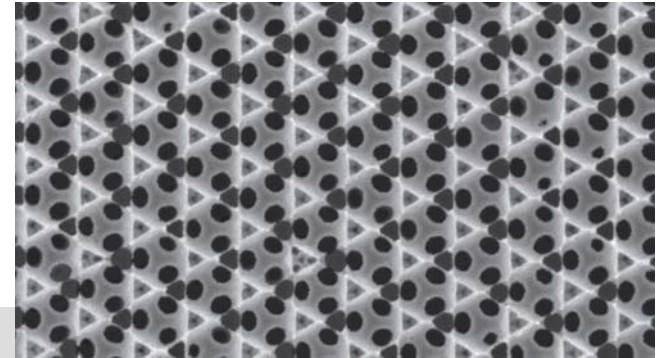
Guiding light in air:

13dB/km - *Venkataraman et al.*

Enhanced optical nonlinearities:

Low Threshold Stimulated Raman
Benabid et al., Science **298**, 399 (2002)

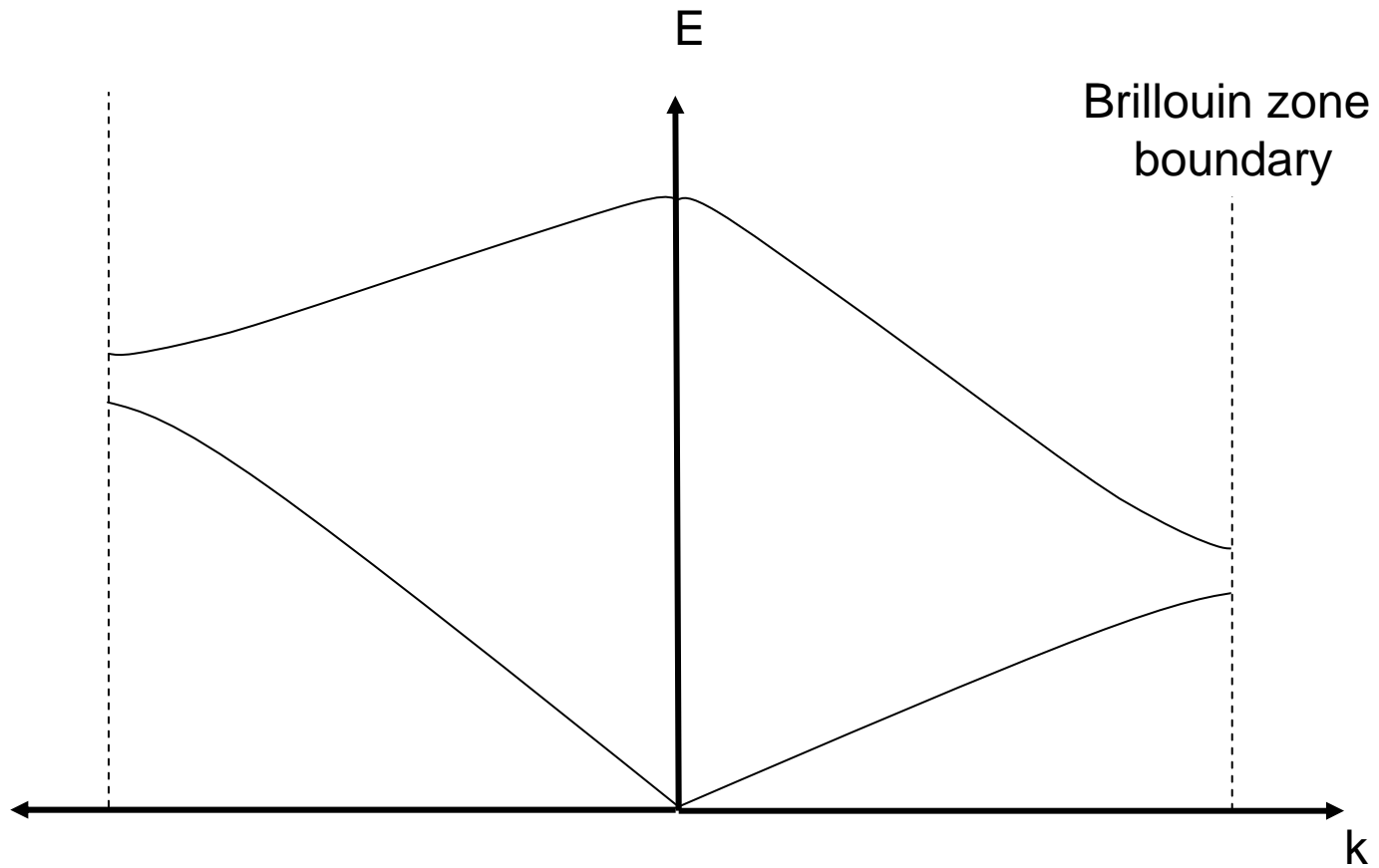
Beyond 2D Photonic Crystals:



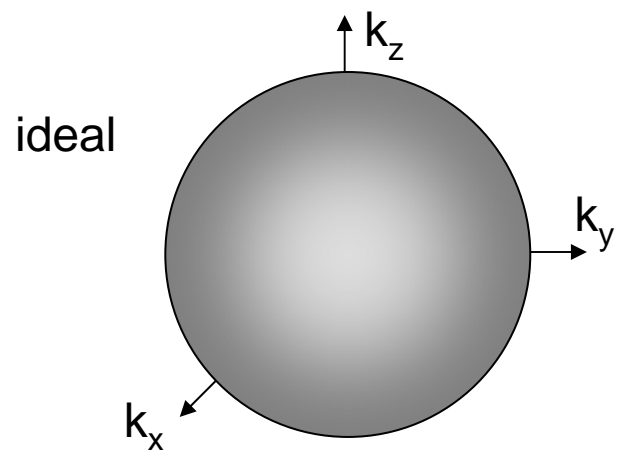
- Complete photonic band gap requires 3D photonic crystal
- Much more challenging to fabricate
- What structure do we need?
- Best approach to make?

Which Crystal Structure?

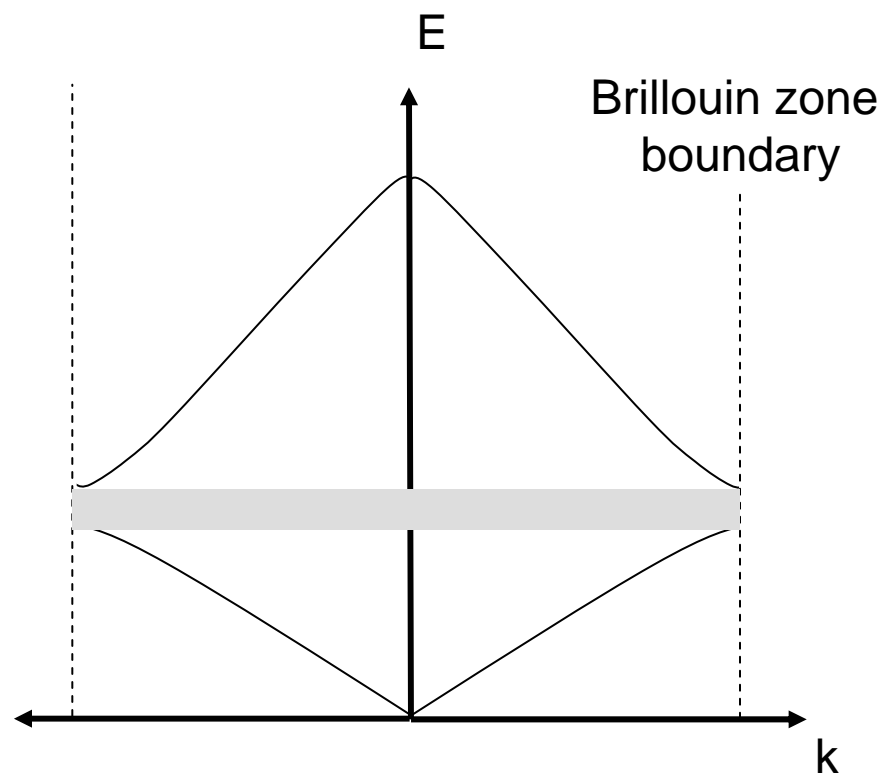
We need intuition . . .



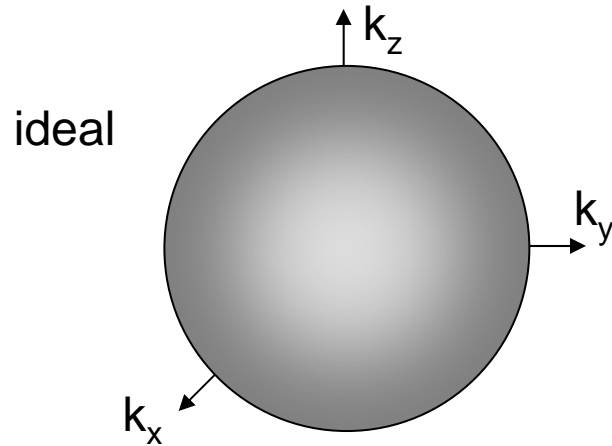
For complete gap want spherical Brillouin zone



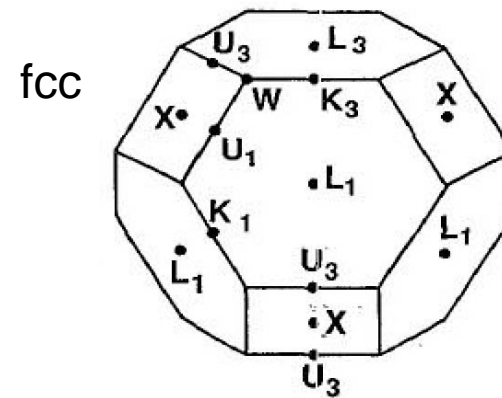
*Brillouin zone
in reciprocal space*



Unfortunately, nature does not provide this . . .

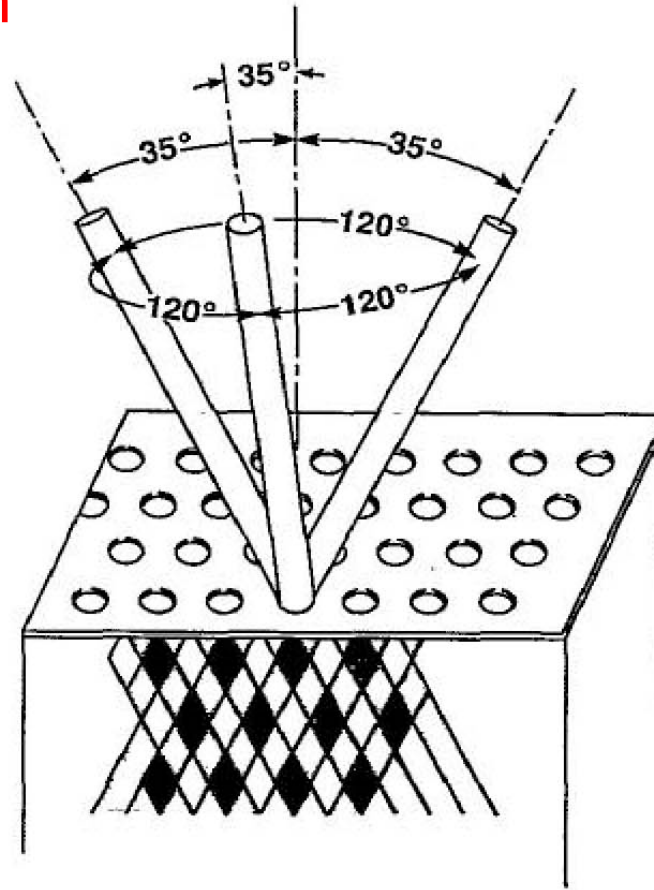


*Brillouin zone
in reciprocal space*



Most sphere-like

Experimental realization

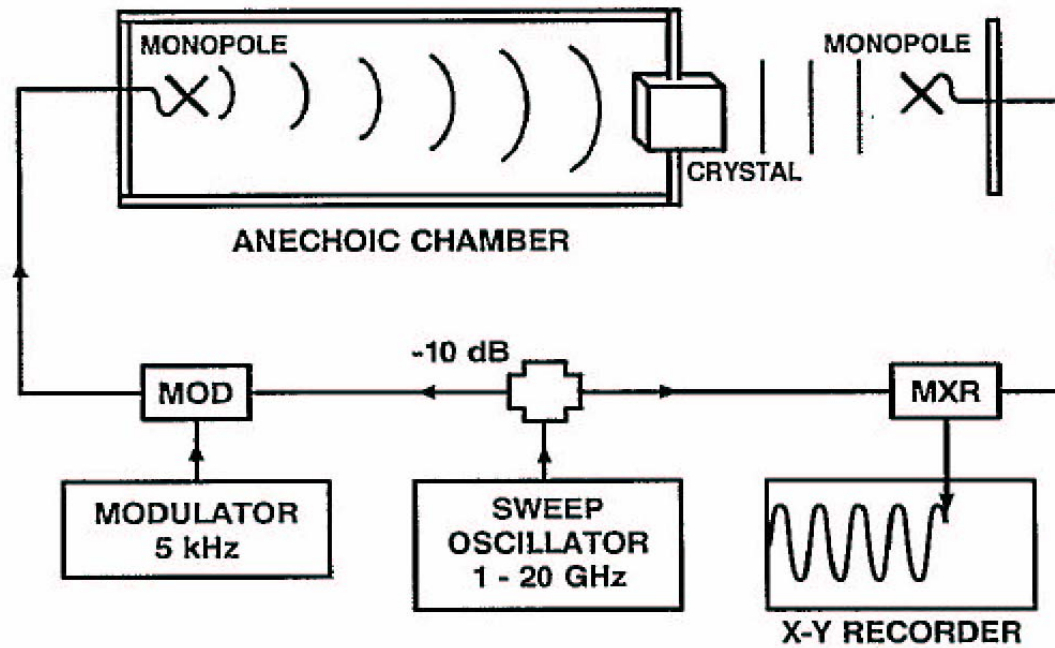


fcc with non-spherical atoms

- Expt: Yablonovitch & Gmitter; PRL **61**, 1950 (1989).
Yablonovitch, Gmitter, & Leung; PRL **67**, 2295 (1991).
- Theory: Leung & Liu; PRL **65**, 2646 (1990).
Zhang & Satpathy; PRL **65**, 2650 (1990).
Ho, Chan, & Soukoulis, PRL **65**, 3152 (1990).

Measurement of the photonic band gap

Microwave Structures:

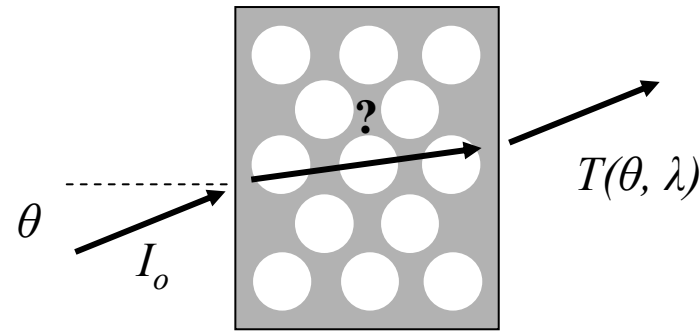


probe transmission for all directions of the Brillouin zone

Yablonovitch, Gmitter, & Leung; PRL **67**, 2295 (1991).

Experimental difficulty:

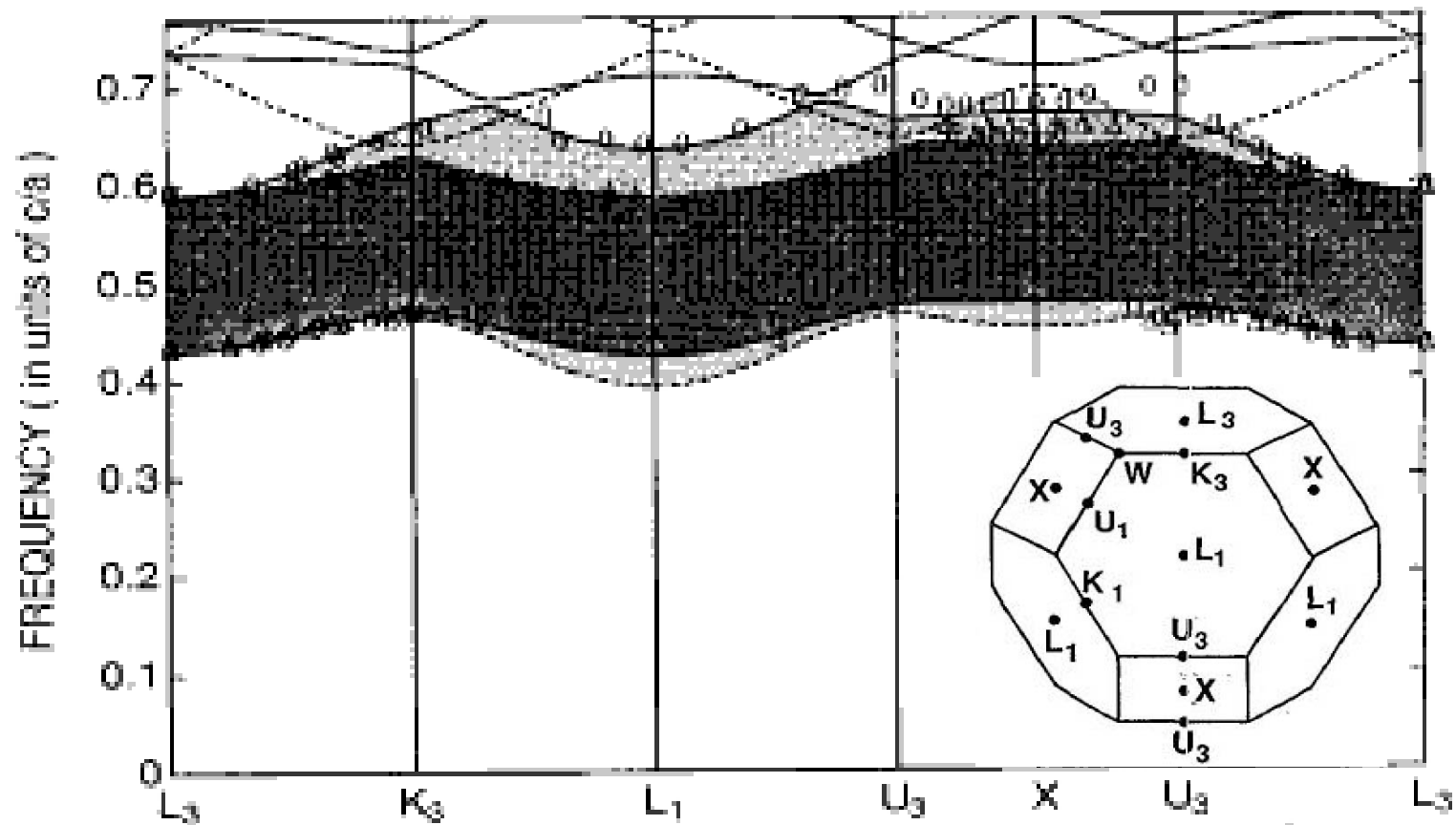
Breakdown of Snell's law



- measure transmission vs. propagation direction
- refractive index varies near photonic band gap
- don't know internal propagation direction
- solution: measure phase of transmitted photons

Demonstration of a photonic band gap

Microwave Structures:



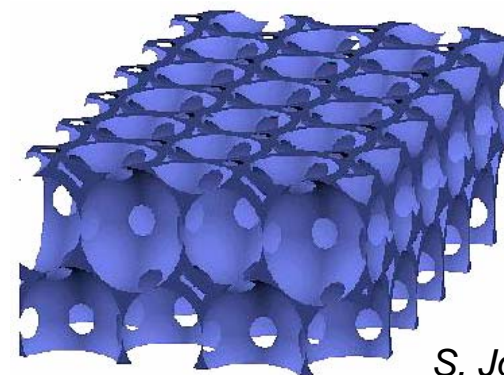
Yablonovitch, Gmitter, & Leung; PRL **67**, 2295 (1991).

What about an optical photonic band gap?

Photon physics is exactly the same

Requirements:

- 3D periodicity
- size of unit cell on the order of λ
- ideal structure is mostly “air”
~80%
- large refractive index necessary
>2 (ideal case)
>3 (typical)

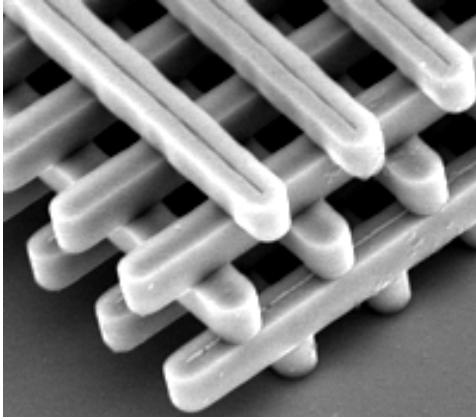


S. John

Best material: **semiconductors**

- high refractive index
- convenient electronic and optical properties
- integrate photonic crystals into optoelectronics

But how do we make 3D photonic band gap crystals?



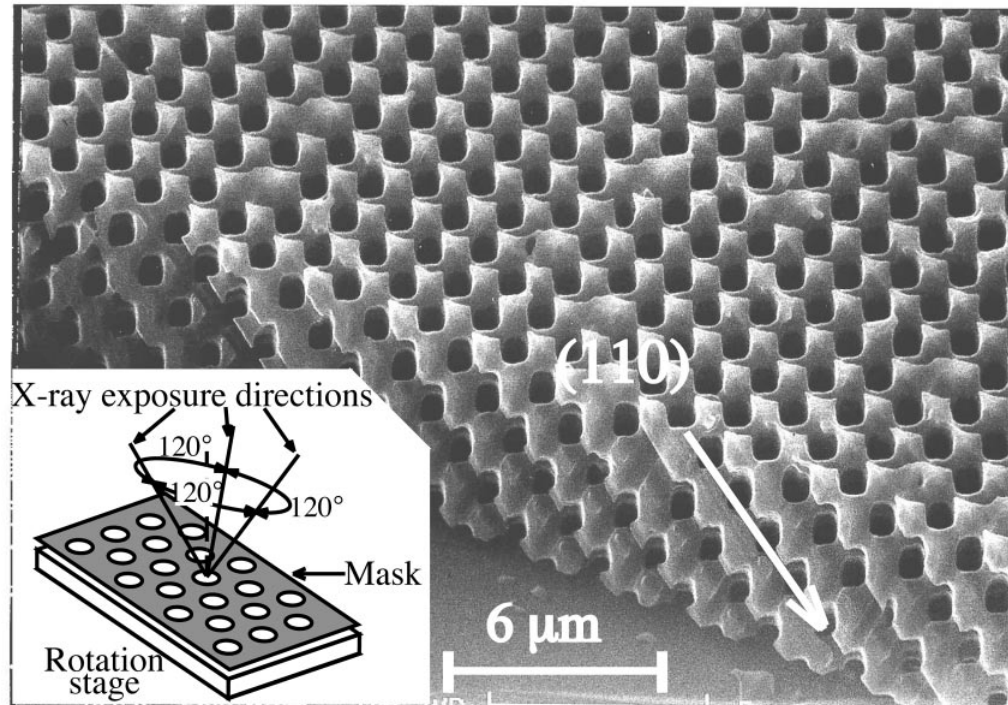
S. Lin et al., Nature **394**, 251 (1998)

“Layer by Layer” Nanofabrication:

Advantages:	excellent control of structure optical band gap demonstrated
Challenges:	expensive

Alternative Approaches: X-ray Lithography (LIGA)

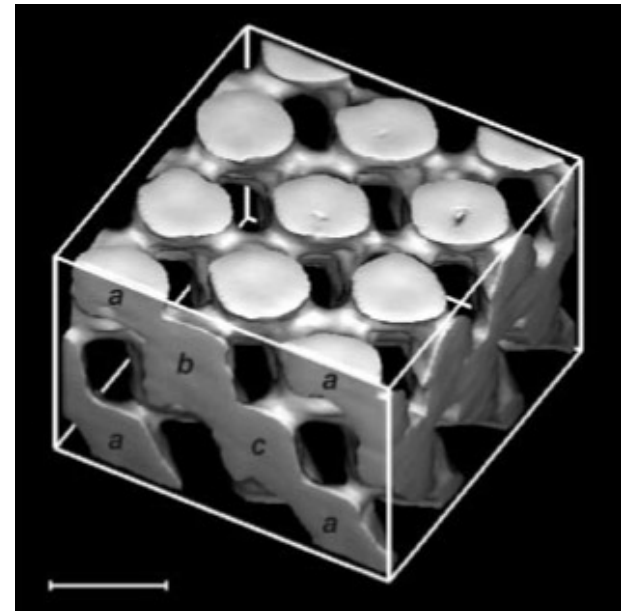
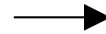
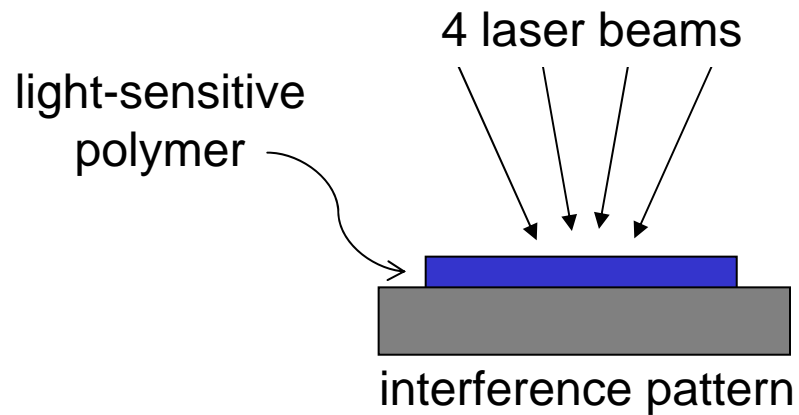
Feiertag et al., APL 71, 1441 (1997); C. Cuisin et al., APL 77, 770 (2000).



Advantages:	good control; makes Yablonovite
Challenges:	difficulties with mask makes polymer template to date: no photonic band gap

Alternative Approaches: Laser Holography

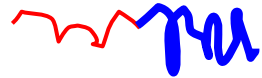
M. Campbell et al., Nature 404, 53 (2000).



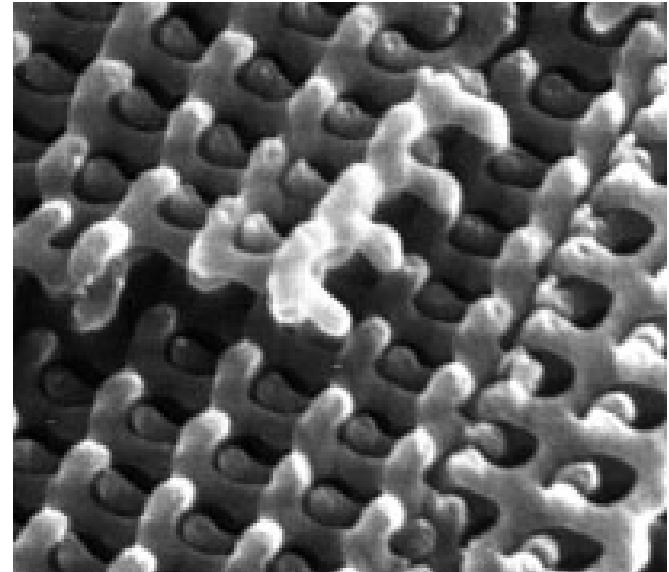
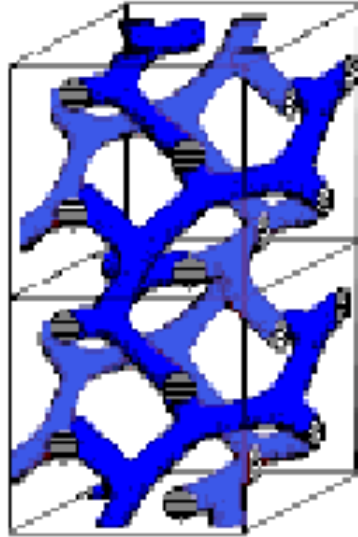
Advantages:	simple "one shot", good control
Challenges:	makes polymer template to date: no band gap

Alternative Approaches: Block Copolymers

Urbas, Maldovan, DeRege, and Thomas; Adv. Mater. 14, 1850 (2002).



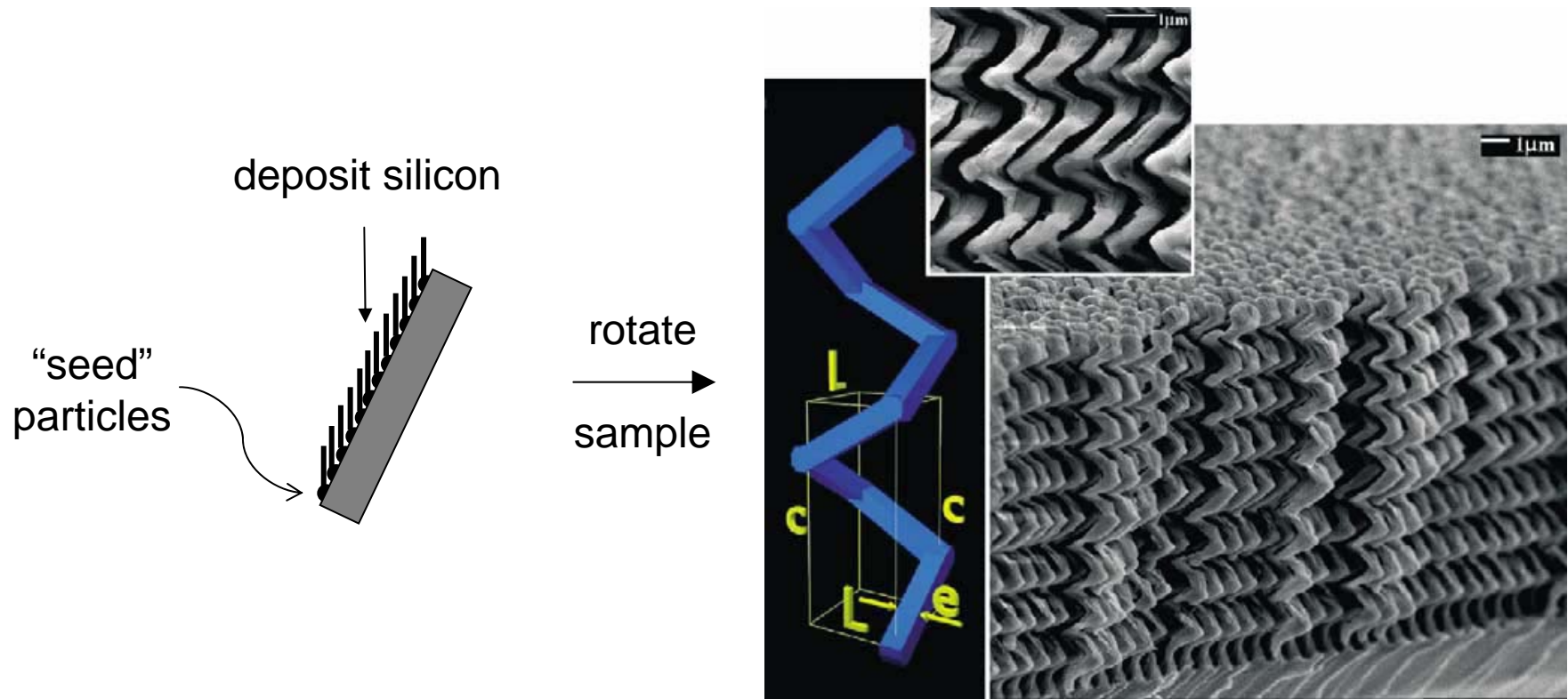
AB diblock



Advantages:	simple technique
Challenges:	makes polymer structure disorder? to date: no band gap

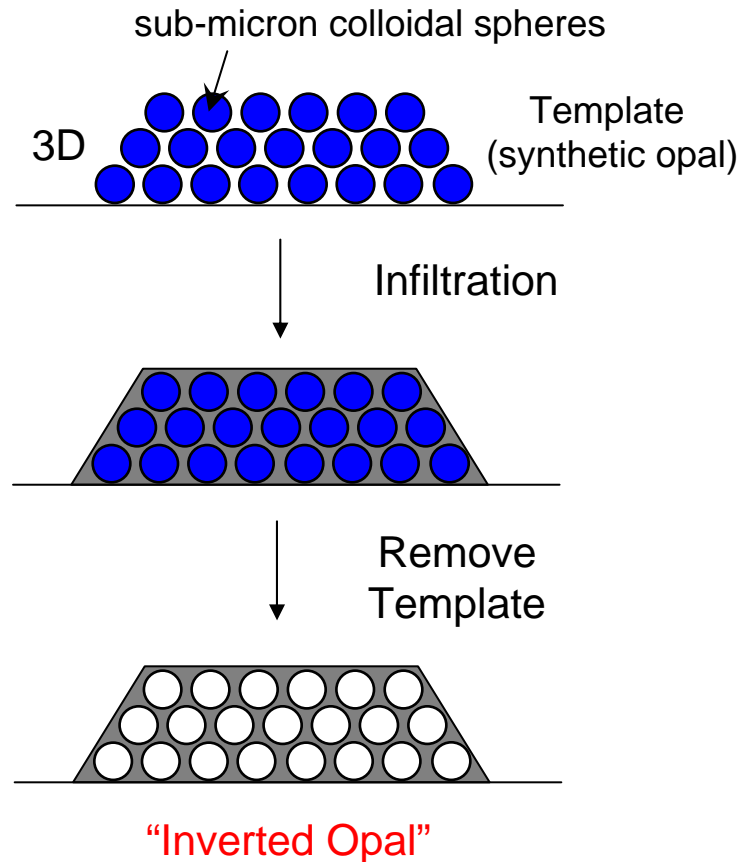
Alternative Approaches: Glancing Angle Deposition

Kennedy, Brett, Miguez, Toader, and John; Photonics and Nanostructures 1, 37 (2003).



Advantages:	simple technique band gap demonstrated
Challenges:	disorder under control?

Our Approach: Colloidal Self-Assembly



Literature Results:

- O. D. Velev *et al.*, *Nature* **1997**, 389, 447.
- D. Pine *et al.*, *Nature* **1997**, 389, 948.
- G. Stucky *et al.*, *Science* **1998**, 279, 548.
- A. Stein *et al.*, *Science* **1998**, 281, 538.
- W. Vos *et al.*, *Science* **1998**, 281, 802.
- A. A. Zakhidov *et al.*, *Science* **1998**, 282, 897.
- T. E. Mallouk *et al.*, *Science* **1999**, 283, 963.
- V. L. Colvin *et al.*, *PRL* **1999**, 83, 300.
- A. Blanco *et al.*, *Nature* **2000**, 405, 437.

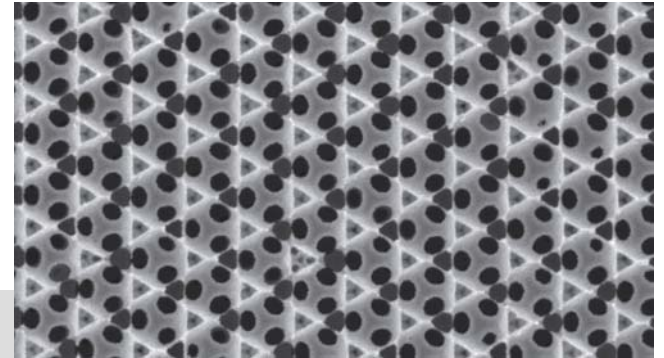
Theory: inverted opals have a PBG

Haus *et al.*, *PRB*, **45**, 13962 (1992).

Busch and John, *PRE*, **58**, 3896 (1998).

Requires refractive index > 2.85

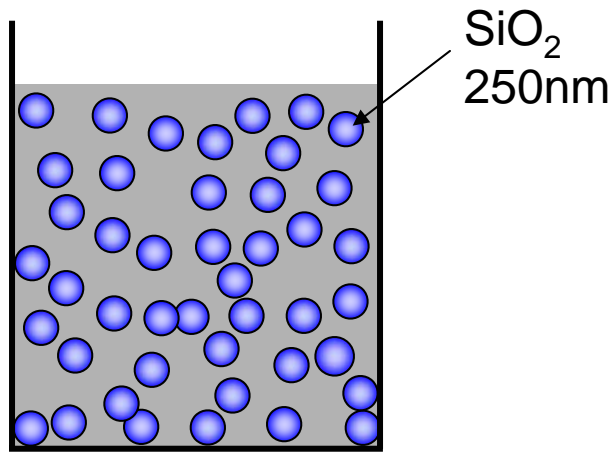
Colloidal Self-Assembly:



Challenges: control disorder?
devices?

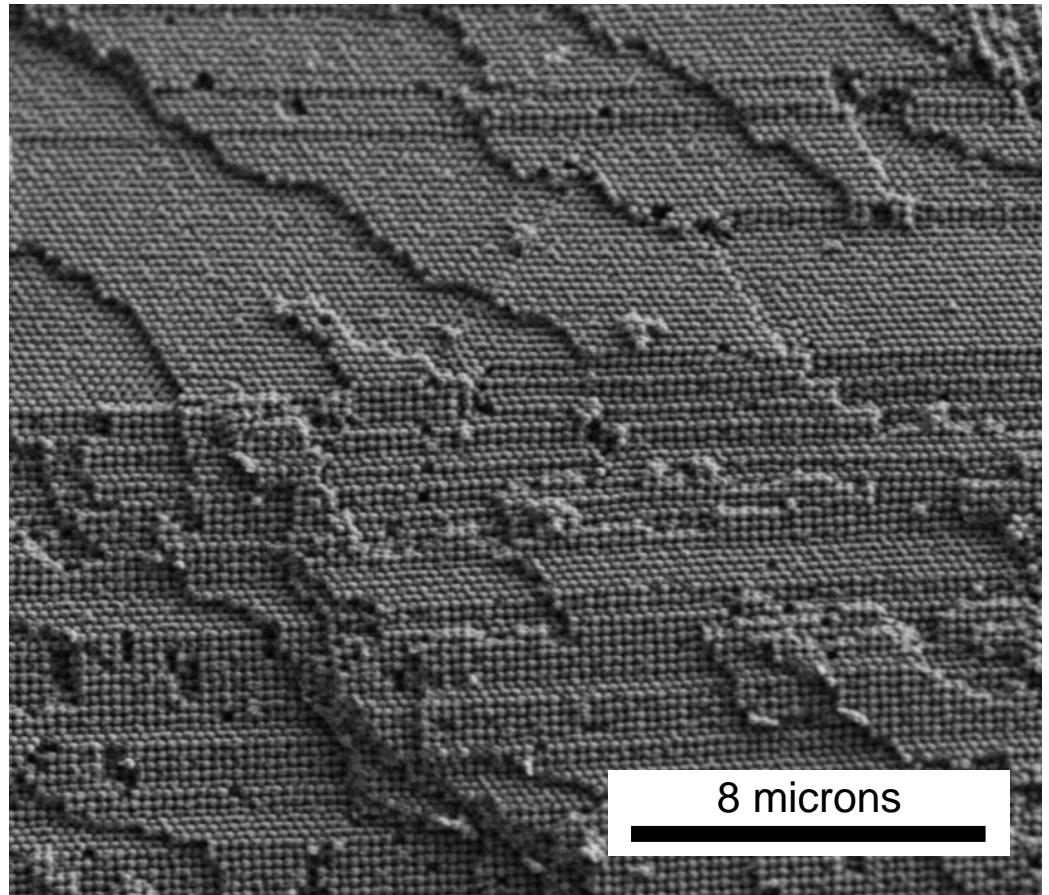
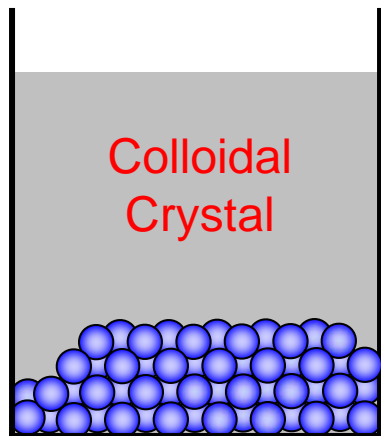
Can this approach lead to useful photonic band gaps?

Initial approach to opals: sedimentation



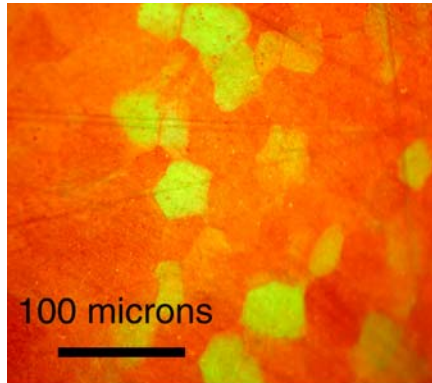
Sediment

A downward-pointing arrow is positioned to the left of the word 'Sediment'.



dry and collect sediment → synthetic opal

What about disorder? Self-assembly makes mistakes!



synthetic opal

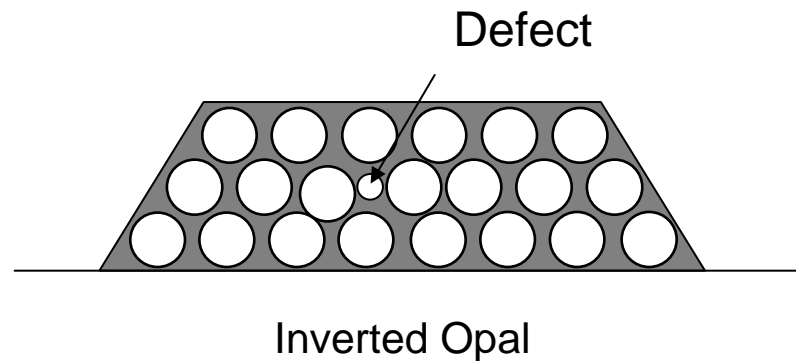


natural gemstone opal

Problems

- Opals have disorder
- First, opals are polycrystalline
- Beautiful in gemstone; bad for photonics
- Also, other defects (vacancies, stacking faults, *etc.*)

Inverted Opals and Disorder:



Serious question:

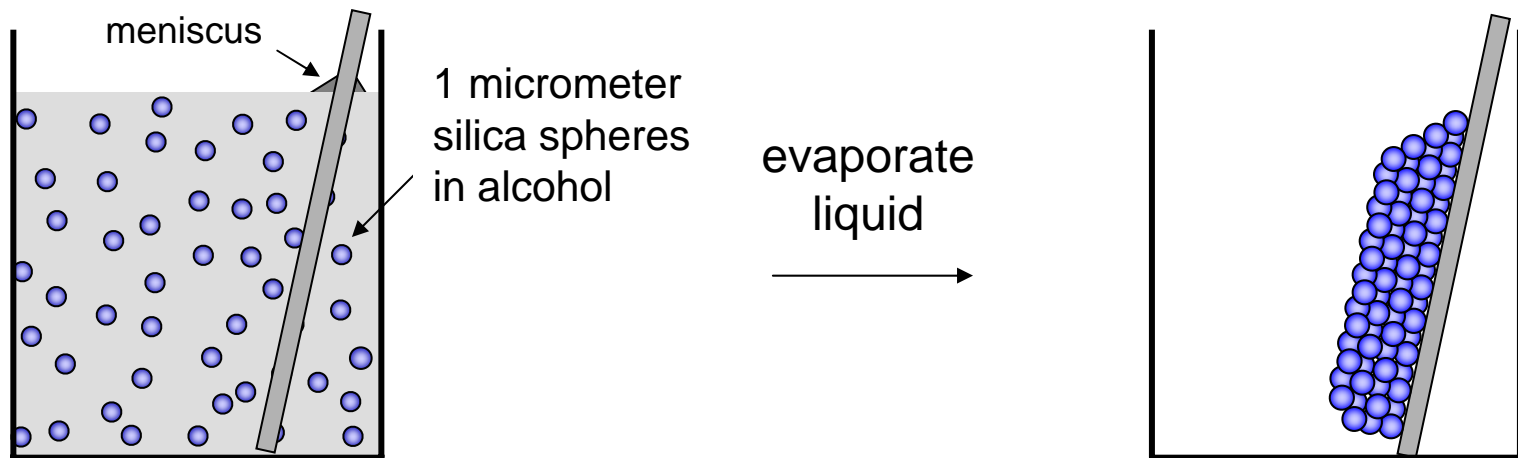
- Will photonic band gap exist in self-assembled materials?
disorder can destroy photonic band gap

Theory: 4% deviation in “bubble spacing” closes band gap!
Li & Zhang, PRB 62, 1516 (2000).

To reduce disorder: make better opals

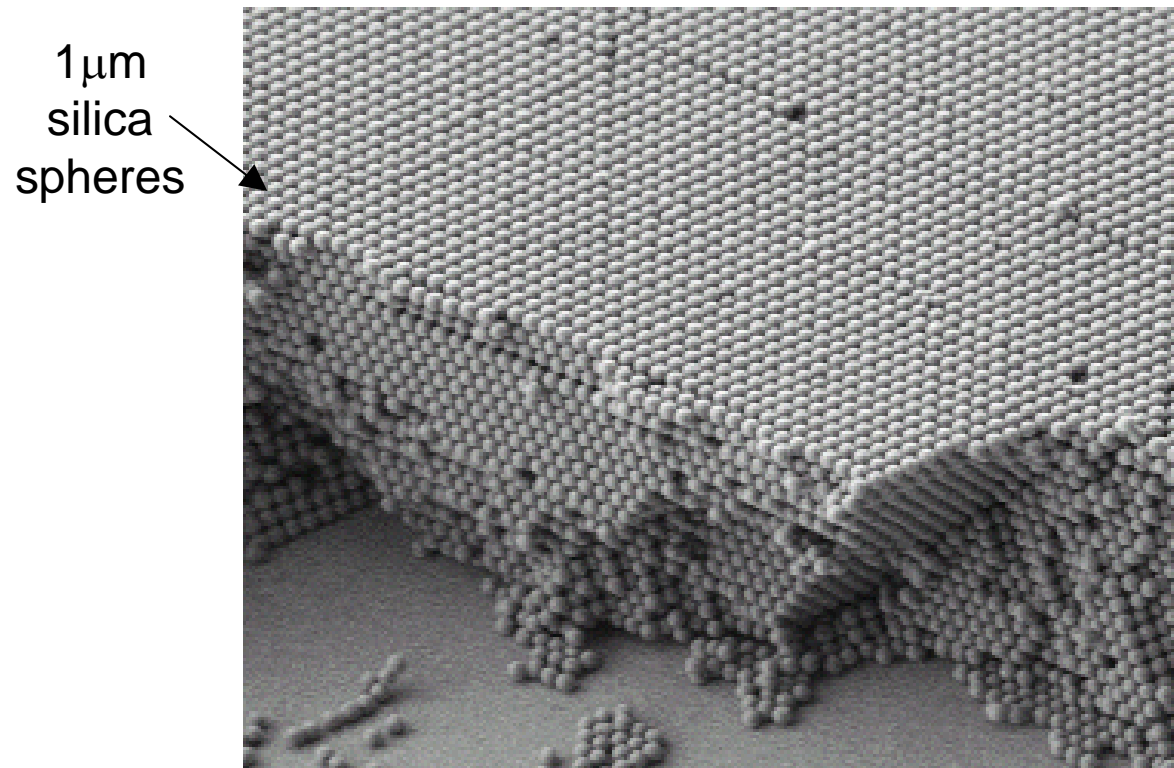
“Convective Assembly”

Nagayama, Velev, *et al.*, *Nature* (1993)
Colvin *et al.*, *Chem. Mater.* (1999)
Norris *et al.*, *Nature* (2001)



- Evaporating the liquid causes assembly in the meniscus
- Extremely flat, large-area opals of controllable thickness

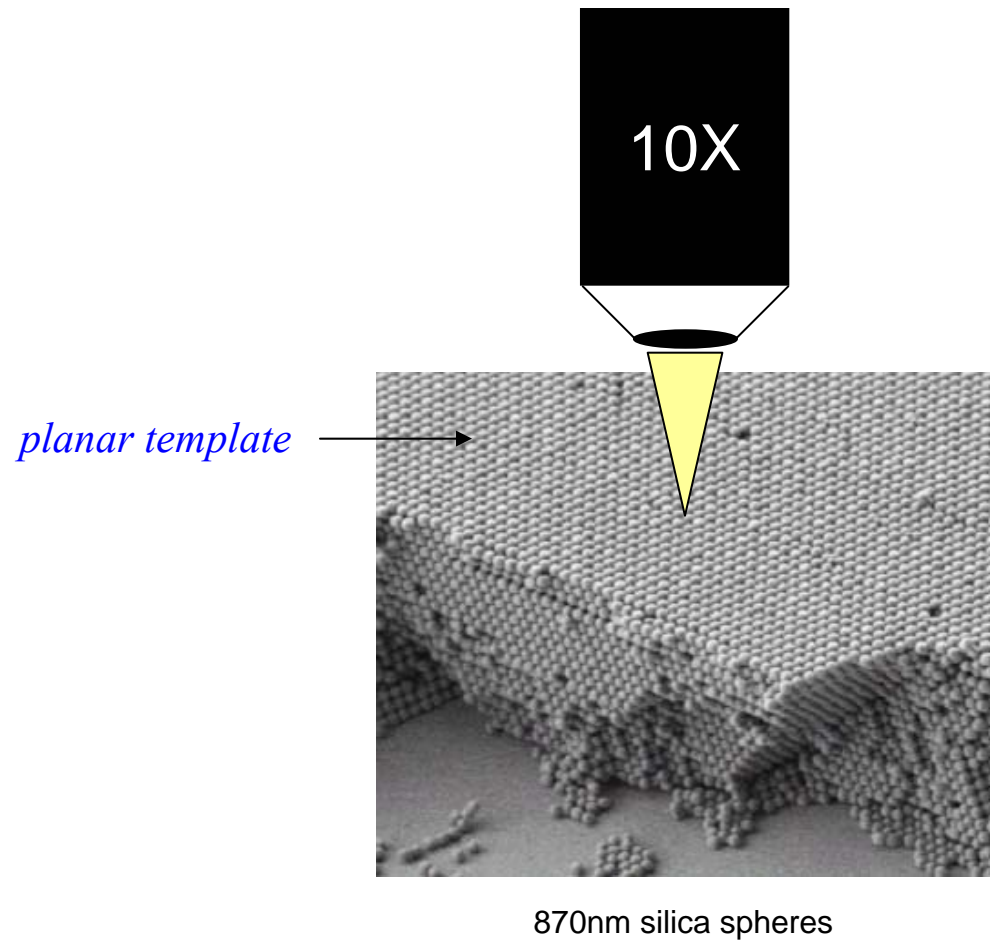
Thin opaline coatings



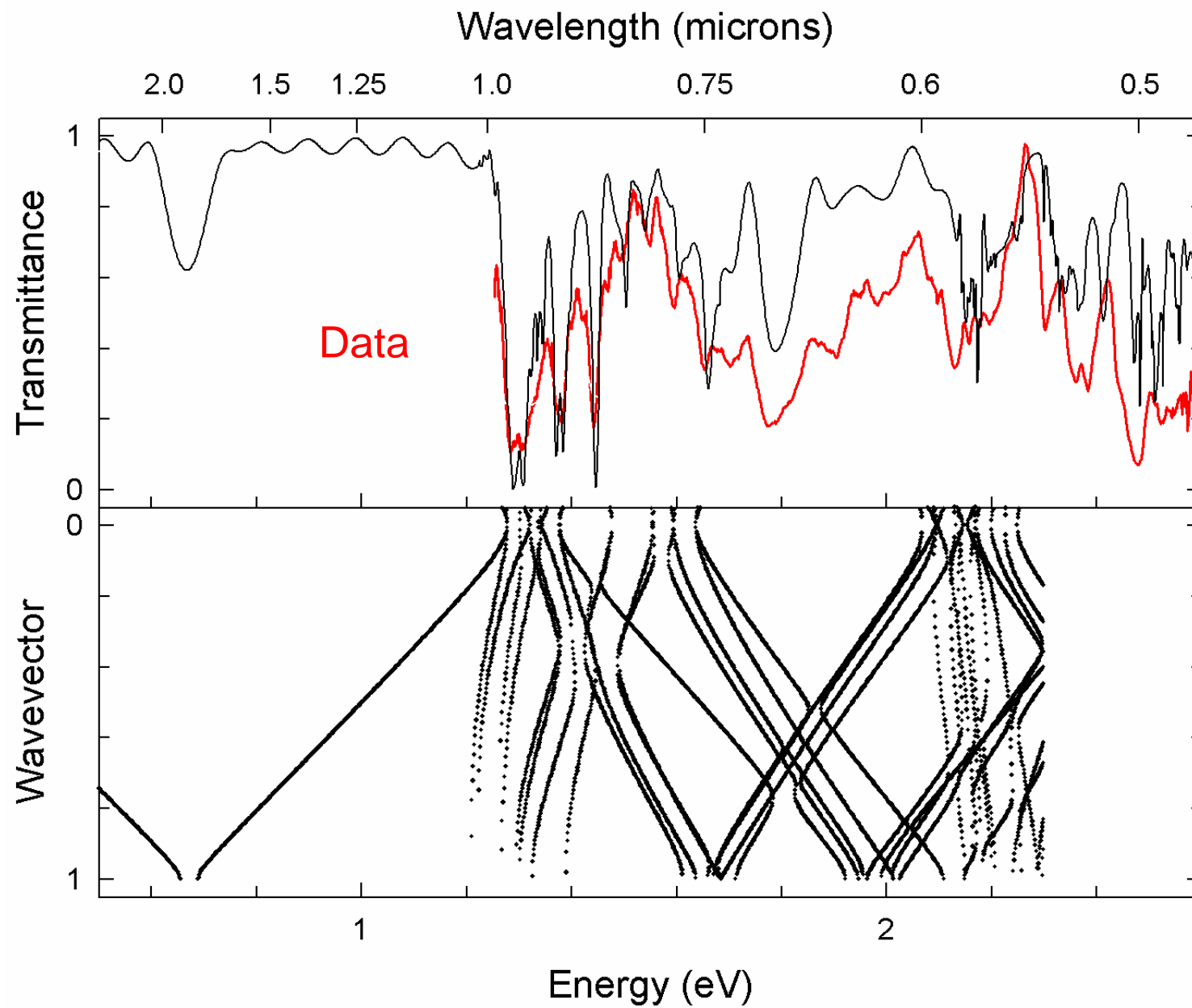
infiltrate with semiconductor \rightarrow photonic band gap in the infrared

(potentially useful for applications at $\lambda=1.55\mu\text{m}$)

But First, What are the Optical Properties?

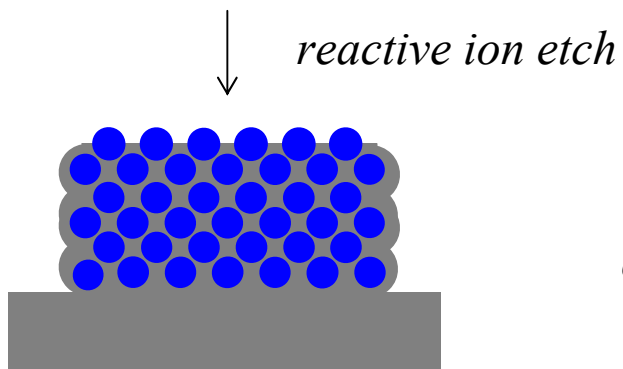
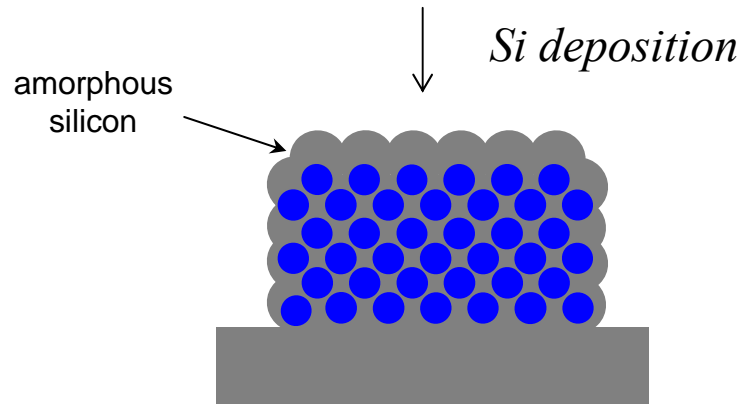
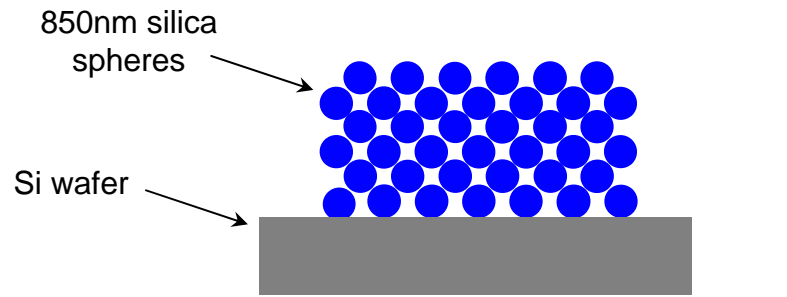


Measure transmission . . .



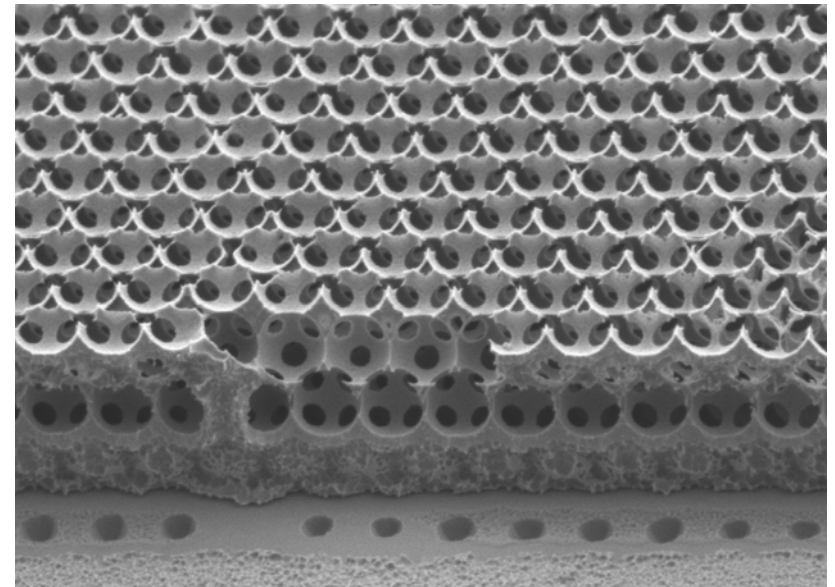
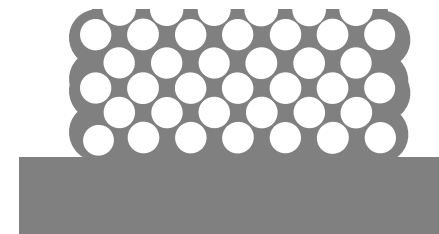
Silicon Inverted Opal

in collaboration with X.-Z. Bo and J. Sturm (Princeton)

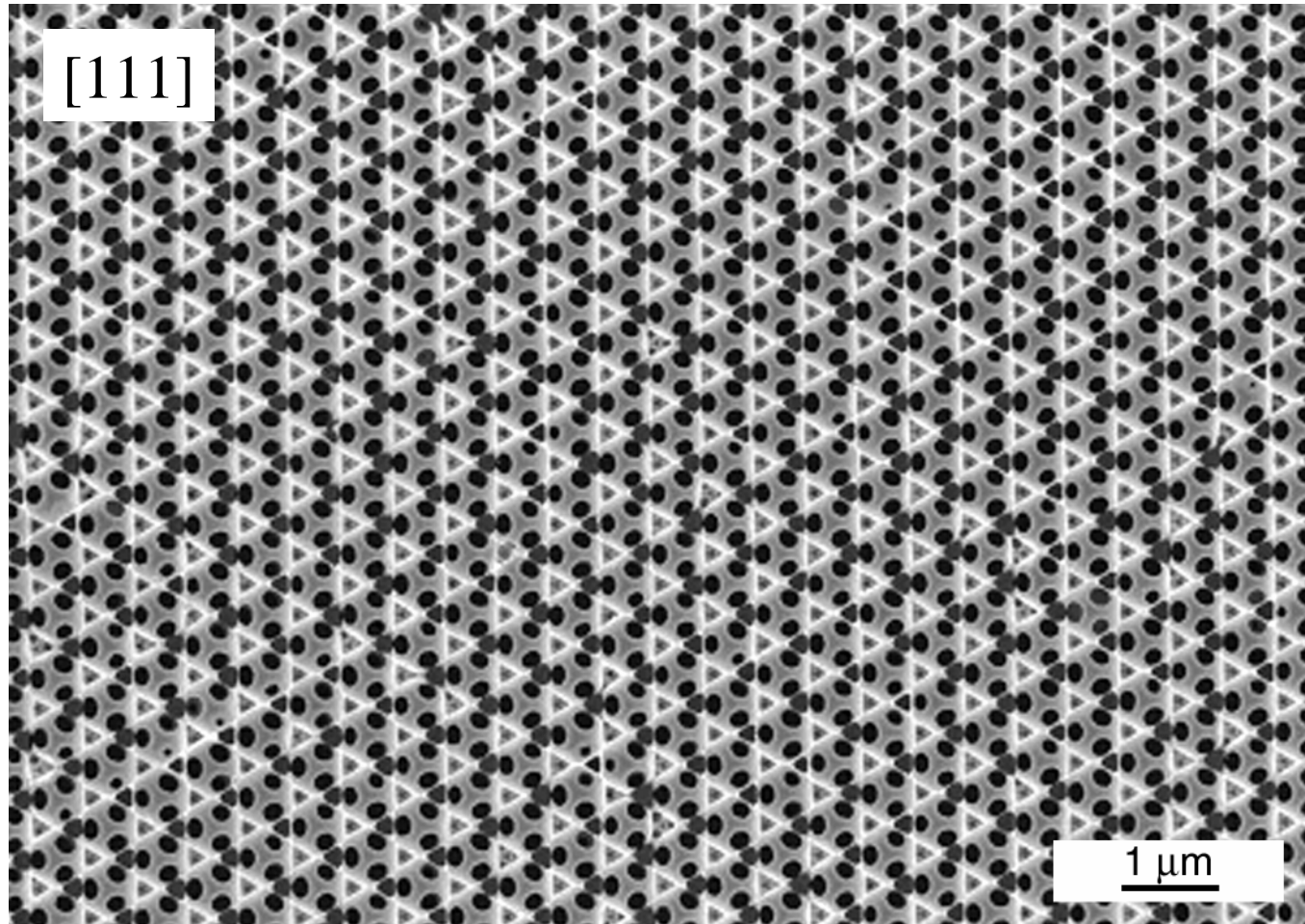


acid etch

→

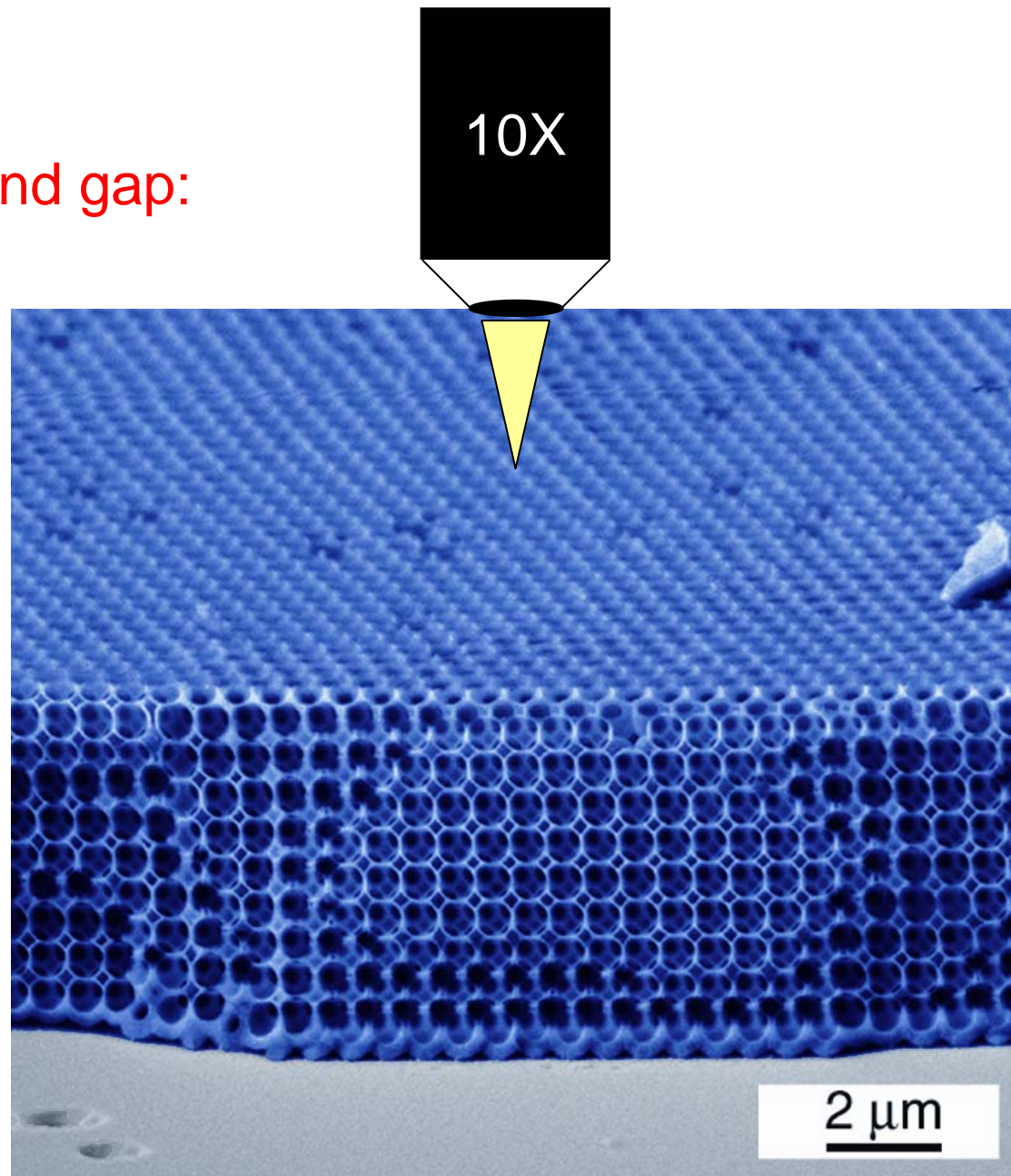


Si inverted opals



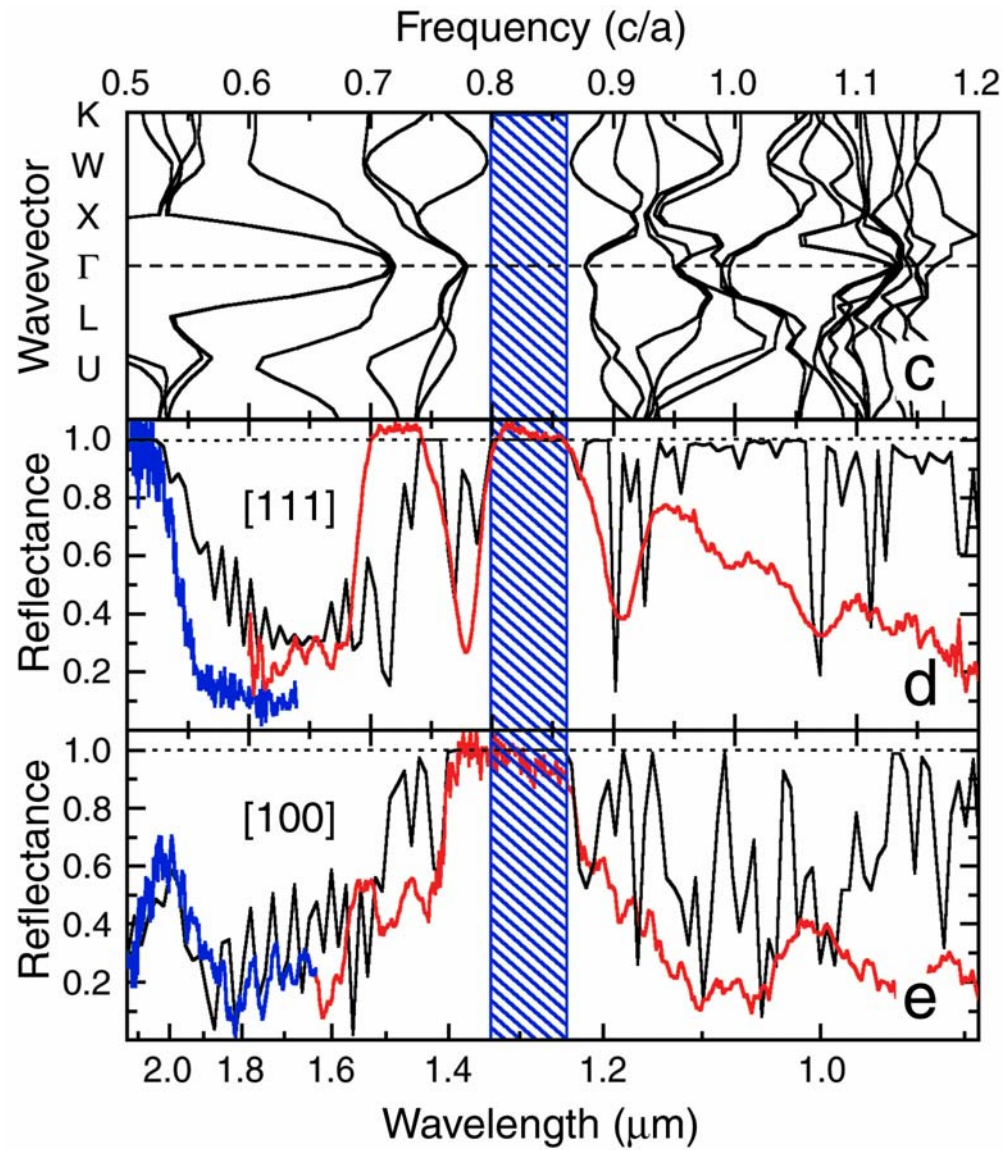
Vlasov, Bo, Sturm, & Norris; *Nature* **414**, 289 (2001).

Testing the
photonic band gap:



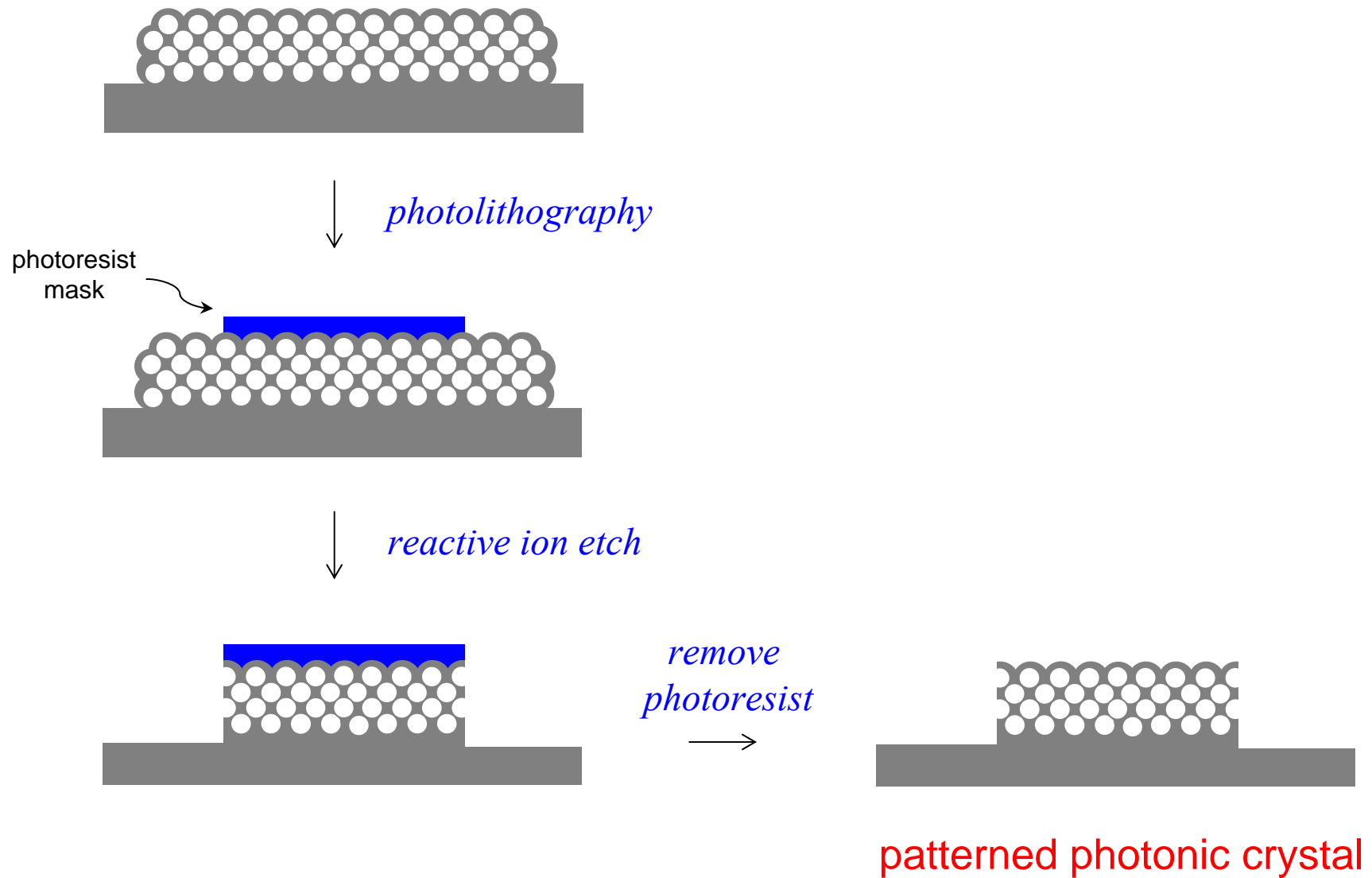
Measure reflection spectra

Testing the Photonic Band Gap

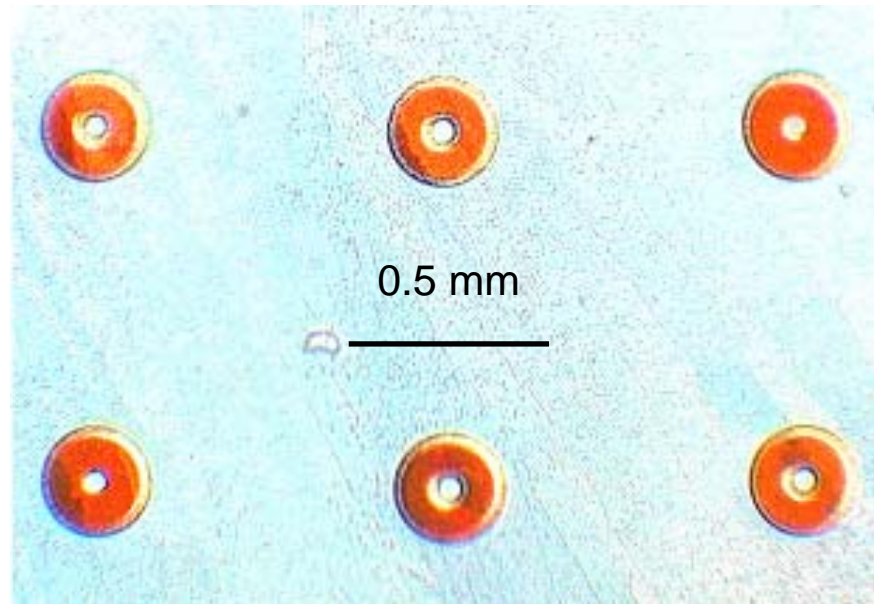
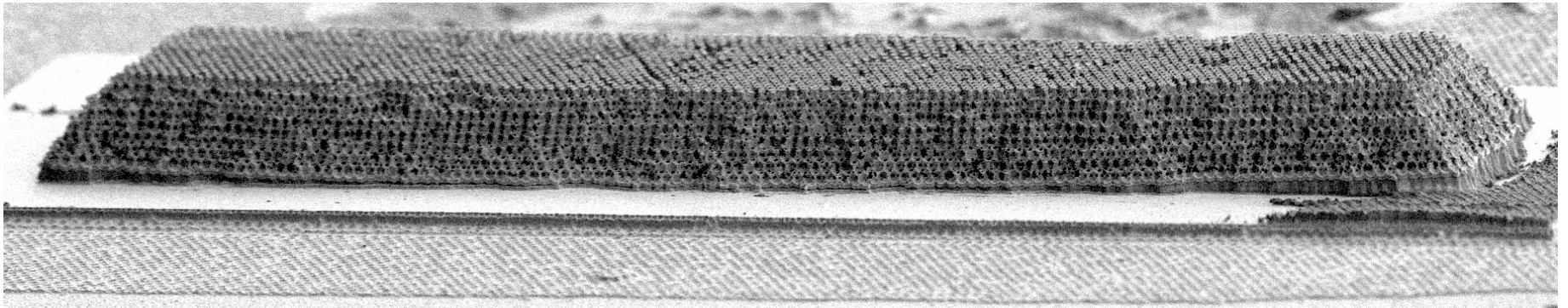


Nature (2001).

Once we have these photonic band gap coatings . . .

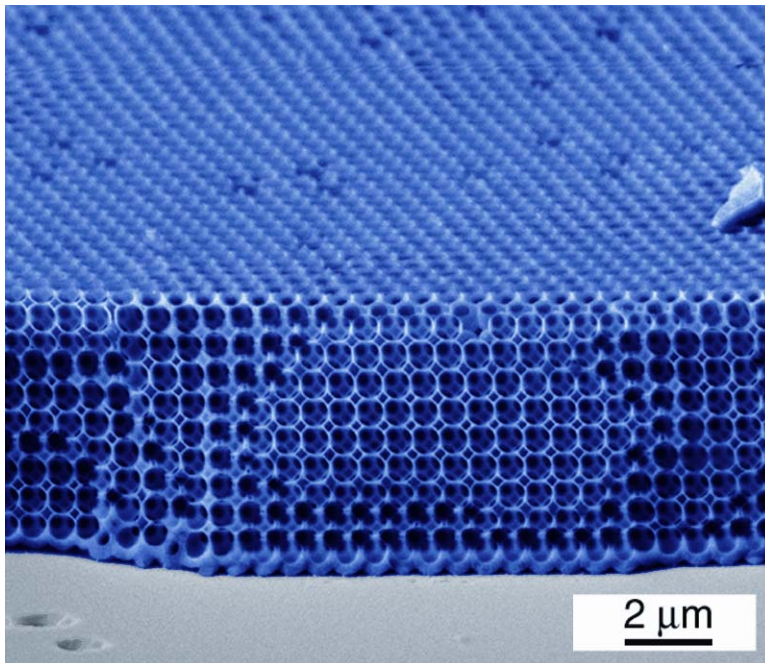


Patterned Si Photonic Band Gap Crystals



Conclusion:

- Self-assembly can yield structures that have optical properties consistent with a photonic band gap
- Photonic band gap coatings !



Questions:

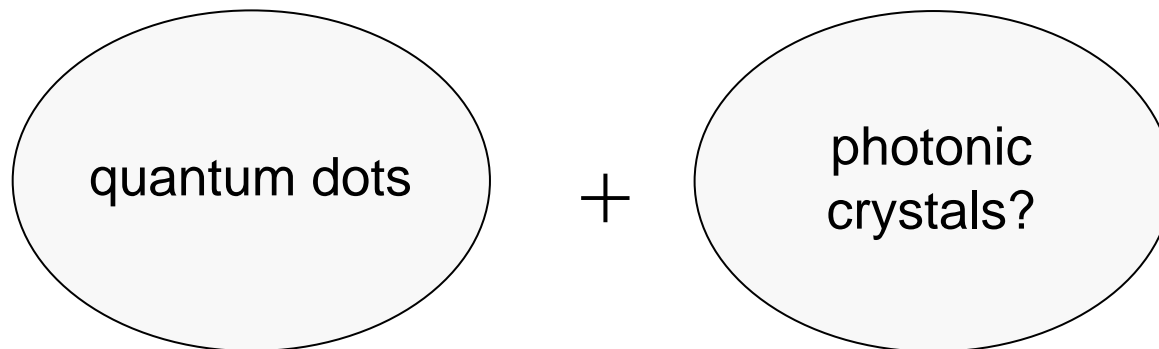
- How do these materials behave?
- How can we best utilize?
- How does self-assembly work?

Another Possibility:

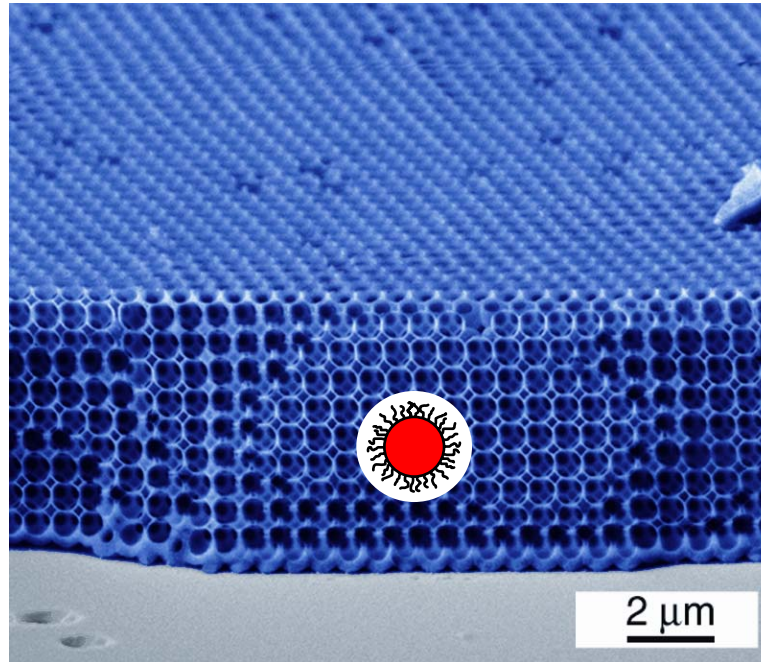
Back to Fermi's Golden Rule

$$\Gamma_o \propto \rho_{\text{elec}}(\omega) \cdot \rho_{\text{phot}}(\omega)$$

Enhance Both Electronic and Photonic DOS?



Combine with quantum dots?

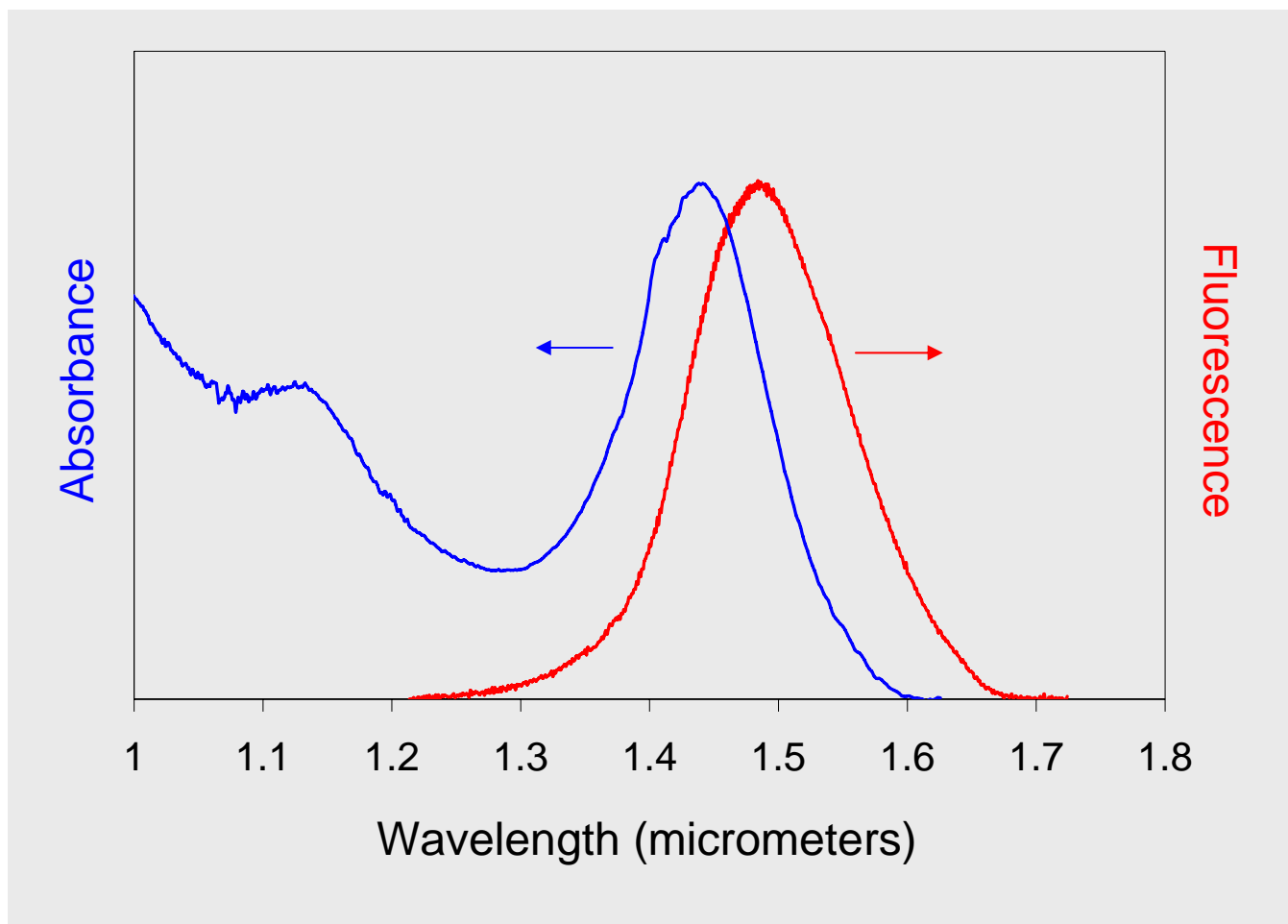


Challenge:

- IR emitting quantum dots?

PbSe Nanocrystals

F. Chen, K.L. Stokes, W. Zhou, J. Fang and C.B. Murray, *MRS Proc.* **685**.



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