The Photonic Band Gap and Colloidal Crystals

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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?

Fermi’s Golden Rule

\[ \text{Rate} \equiv \Gamma_0 \propto \rho(\omega) \]

\[ i.e., \text{“speed” of a process depends on the number of available states} \]
Controlling Electronic DOS:

Early motivation for nano-structures

modification of electronic density of states
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

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

optical microcavities

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

\[ A_r = (8\pi \nu^2/c^3)\hbar(8\pi^2\mu^2/3\hbar^2) \text{ sec}^{-1}, \]

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for \( \nu = 10^7 \) sec\(^{-1} \), \( \mu = 1 \) nuclear magneton, the corresponding relaxation time would be \( 5 \times 10^4 \) 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 \( \nu/Q \) 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 \( V \sim a^2 \), and if \( \delta \) is the skin-depth at frequency \( \nu \), \( f \sim \lambda^3/a^2 \). For a non-resonant circuit, \( f \sim \lambda^3/a^2 \), and for \( a < \delta \) it can be shown that \( f \sim \lambda^3/a^2 \). 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 \( \nu = 10^7 \) 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

\[ \Gamma_0 \propto \rho_{\text{elec}}(\omega) \cdot \rho_{\text{phot}}(\omega) \]
Photonic Band Gap:

Ultimate control over the photonic density of states

\[ \rho_{\text{phot}}(\omega) \]

\[ \Gamma_0 \propto \rho_{\text{elec}}(\omega) \cdot \rho_{\text{phot}}(\omega) \]
Photonic Band Gap:
Ultimate control over the photonic density of states

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

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
Yablonovitch’s original idea:

Device inside photonic crystal:

Complete control over wasteful spontaneous emission in unwanted directions
Photonic crystals: experiment

**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


Photonic crystal laser
Photonic Crystal Fibers

Guiding light in air:
13dB/km - Venkataraman et al.

Enhanced optical nonlinearities:
Low Threshold Stimulated Raman
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
Unfortunately, nature does not provide this . . .

Brillouin zone in reciprocal space

Most sphere-like
Experimental realization

fcc with non-spherical atoms


Ho, Chan, & Soukoulis, PRL 65, 3152 (1990).
Measurement of the photonic band gap

*Microwave Structures:*

probe transmission for all directions of the Brillouin zone

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

Experimental difficulty:
Demonstration of a photonic band gap

*Microwave Structures:*

What about an optical photonic band gap?

*Photon physics is exactly the same*

Requirements:

- 3D periodicity
- size of unit cell on the order of $\lambda$
- ideal structure is mostly “air”
  \[ \sim 80\% \]
- large refractive index necessary
  \[ > 2 \quad (\text{ideal case}) \]
  \[ > 3 \quad (\text{typical}) \]

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?

“Layer by Layer” Nanofabrication:

Advantages: excellent control of structure
optical band gap demonstrated

Challenges: expensive

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


Advantages: simple “one shot”, good control
Challenges: makes polymer template
to date: no band gap
Alternative Approaches: Block Copolymers

UBAS, MALDOVAN, DeReGE, and THOMAS; ADV. MAT. 14, 1850 (2002).

Advantages: simple technique
Challenges: makes polymer structure disorder?
to date: no band gap
Alternative Approaches: Glancing Angle Deposition

Advantages: simple technique
band gap demonstrated

Challenges: disorder under control?
Our Approach: Colloidal Self-Assembly

- Infiltration: sub-micron colloidal spheres
- Template: (synthetic opal)
- Infiltration
- Remove Template
- “Inverted Opal”

Literature Results:


Theory: inverted opals have a PBG
- 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

SiO$_2$
250nm

Sediment

Colloidal Crystal

dry and collect sediment → synthetic opal

8 microns
What about disorder? Self-assembly makes mistakes!

**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"  

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

1μm silica spheres

infiltrate with semiconductor → photonic band gap in the infrared

(potentially useful for applications at λ=1.55μm)
But First, What are the Optical Properties?

planar template

870nm silica spheres

Measure transmission . . .
Silicon Inverted Opal  
in collaboration with X. –Z. Bo and J. Sturm (Princeton)

850nm silica spheres

Si wafer

850nm silica spheres

amorphous silicon

Si deposition

reactive ion etch

acid etch
Si inverted opals

Testing the photonic band gap:

Measure reflection spectra
Testing the Photonic Band Gap

Once we have these photonic band gap coatings . . .

\[\text{photoresist mask} \rightarrow \text{photolithography} \]

\[\text{reactive ion etch} \]

\[\text{remove photoresist} \rightarrow \text{patterned photonic crystal} \]
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

\[ \Gamma_0 \propto \rho_{\text{elec}}(\omega) \cdot \rho_{\text{phot}}(\omega) \]

Enhance Both Electronic and Photonic DOS?

quantum dots + photonic crystals?
Combine with quantum dots?

Challenge:

• IR emitting quantum dots?
PbSe Nanocrystals

Acknowledgements:

Collaborators:
Y. Vlasov
Prof. J. Sturm (Princeton)
X.-Z. Bo (Princeton)
Y. Jun
H. Wei

Financial Support:
University of Minnesota, IPRIME
NSF MRSEC, NSF CTS
PRF