Control of spontaneous emission in complex materials

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artwork: Ivo Vellekoop

Outline

- Introduction with cartoons
- Photonic band gap crystal fabrication
- Emission in photonic band gap crystals
- Emission in random media
- Some other recent results (brief)
- Summary and outlook

Many thanks to

Allard Mosk, Pepijn Pinkse
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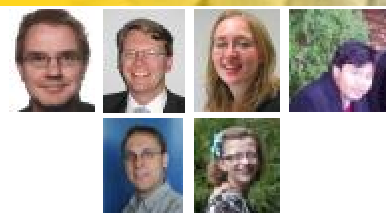
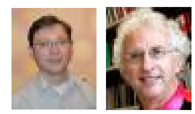
Danang Birowosuto (to NTT, Japan)
Bart Husken (to Lightmotif)
Ivan Nikolaev (to ASML)



Femius Koenderink (FOM Inst. Amolf, Amsterdam)
Ad Lagendijk

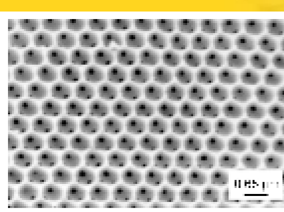


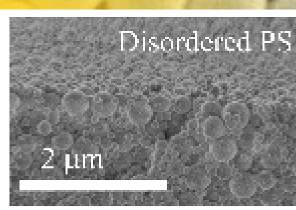
Jean-Michel Gérard, Julien Claudon (CEA, Grenoble, France)
Sergey Skipetrov (CNRS, Grenoble, France)
Peter Lodahl (DTU, Denmark)

Introduction: Nanophotonics

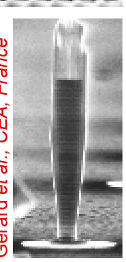
Manipulate light at length scales L ≤ wavelength λ.

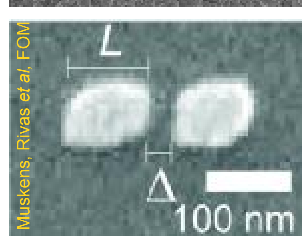
Important classes:
- photonic crystals
- random photonic media
- cavities
- plasmonics, antennae
  etc.





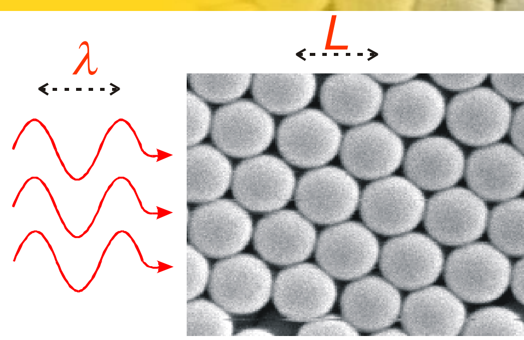
Gérard *et al.*, CEA, France





Photonic crystals

"**Crystal**"(*):
ordered dielectric composite.



Dielectric function varies with position $\varepsilon(\boldsymbol{r})$
⇒ light is scattered.

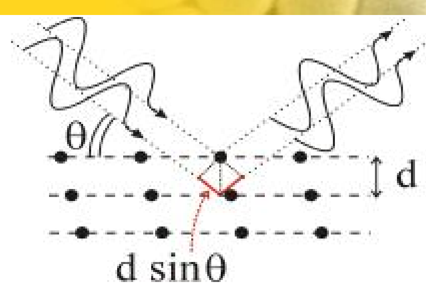
Photonic:
$L \approx \lambda$ ⇒ multiple scattered light interferes: Bragg diffraction, speckle

(*) In strict crystallographic sense: true crystal has identical building blocks.
Not so in Nanophotonics → More appropriate: "ordered metamaterial".

Crystal diffraction that you can see

Bragg interference.

Diffraction condition:
$m\lambda = 2\ d \sin\theta$
[m = integer]



See: Blum, Mosk, Nikolaev, Subramaniam & WLV Small 4 (2008) 492

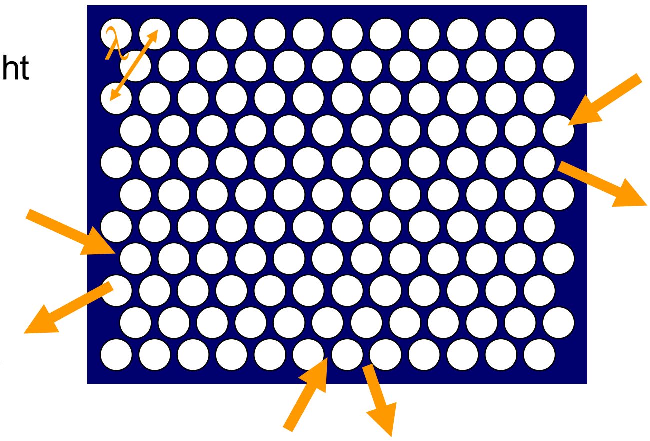
"Opalescence": color determined by lattice spacing *d*.

Bragg can also be terribly wrong, see:
WLV, Sprik, v Blaaderen, Imhof, Lagendijk & Wegdam, PRB **53** (1996) 16231

Photonic band gap crystal (cartoon)

Let's suppose:
frequency range of light is Bragg reflected ***irrespective*** of incident direction.

At these frequencies it's dark inside (multiple interference)

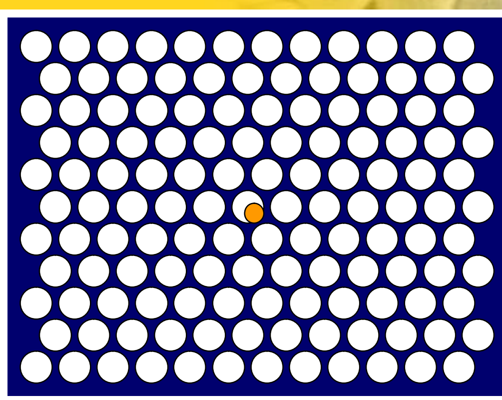


Pioneers: Eli Yablonovitch, PRL **58** (1987) 2059; Sajeev John, *Ibid*. 2486,
Earlier work by Vladimir Bykov (1972) Kazuo Ohtaka (1979), Philip Russell (1984)

Light source in photonic band gap

Perfect 3D crystal with light source (●): emission inhibited, it's still dark inside.

Light source remains excited ***forever***.



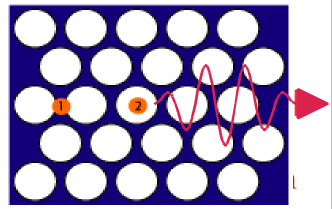
Role of band gap:

- Shield quantum systems from fluctuations.
- Efficient dipole-dipole coupling (→ couple Qubits?)
- Anderson localize photons by (intentional) disorder.

Emission rate of quantum emitter

Excited emitter emits photons at rate γ_{rad}:

$$\gamma_{rad} = \frac{\pi\omega}{3\hbar\varepsilon_0} \left|\langle \varphi_f | d | \varphi_i \rangle\right|^2 \cdot N_{rad}(\vec{r},\omega,\vec{e}_d)$$



Quantum emitter:
transition dipole

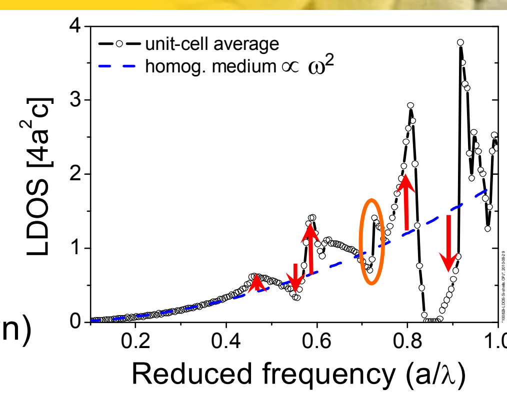
Environment:
local radiative density of states (LDOS)

Goals: Control emission rate by placing emitter in tailored nanophotonic media to control the LDOS.
Understand LDOS in ordered & random photonic media.

1st paper on LDOS: Sprik, van Tiggelen & Lagendijk, Europhys. Lett. **35** (1996) 265.

Features of LDOS in photonic crystal

- Strong enhancements (arrows)
- Strong reductions (arrow)
- Sharp cusps (circle): van Hove singularities (Fermi golden rule breakdown)



and one most important feature...

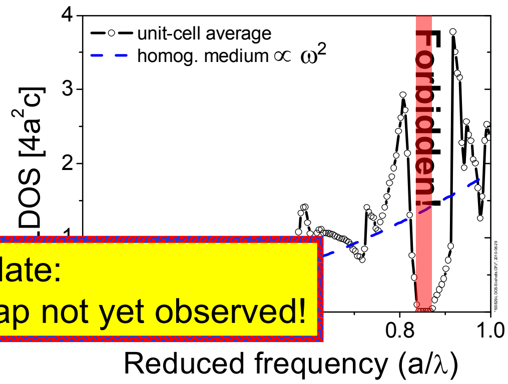
Calculation see:
Nikolaev, WLV & Koenderink
J. Opt. Soc. Am. B **26** (2009) 987

3D Band gap in the LDOS

Range where LDOS=0
→ *Photonic band gap*.

Extreme situation:
vacuum fluctuations inhibited.

Conseque
Spontane
Energy transfer (Förster)
Dispersion forces (van der Waals)
Lamb shift



To date:
3D photonic band gap not yet observed!

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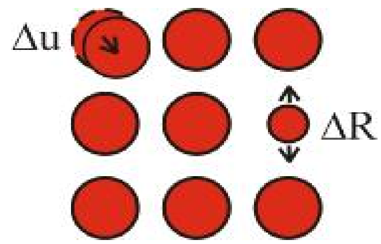
Required for photonic band gaps

- High refractive index contrast: $m=n_1/n_2$ > 2 to 3

air	1.0	glass	1.5	titania	2.7
water	1.3	plastics	1.6	Si, GaAs	3.5

- Ordered structure:
  small size variations
  $\Delta R << R_{avg.}$,
  small displacements
  $\Delta u << a_{lattice}$.



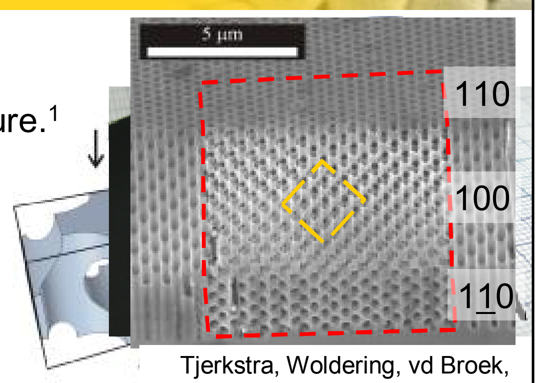
- Low absorption [light should scatter many times.]
- Materials interconnected [helps greatly, no simple reason.]

New: diamond structure crystals in Si

We have developed new Si inverse woodpile crystals with 3D cubic diamond structure.[1]

CMOS compatible methods: deep UV step-scan and *twice* RIE etching[2,3].

High-purity Si (no deposition)
High reflectivity and broad gaps[4]

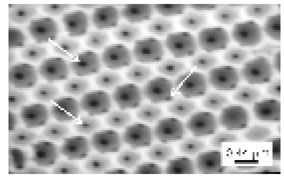


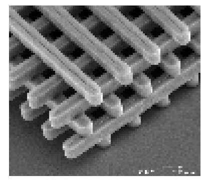
Tjerkstra, Woldering, vd Broek, Roozeboom, Setija & WLV (2010)

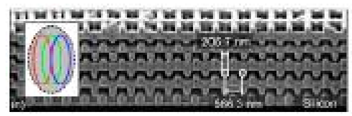
[1] Ho, Chan, Soukoulis, Biswas & Sigalas, Solid State Comm. **89** (1994) 413
[2] Woldering, Mosk, Tjerkstra & WLV, J. Appl. Phys. **105** (2009) 093108
[3] Woldering, Tjerkstra, Jansen, Setija & WLV, Nanotechnology **19** (2008) 145304
[4] Huisman, Nair, Woldering, Leistikow, Mosk & WLV, Arxiv.org (2010)

Many 3D crystals around the world

- Opals, inverse opals.
  [> 100s groups worldwide.]
  [Wijnhoven & WLV, Science **281** (1998) 802]
- Etching (e.g. Halle, Kiel).
- Layer by layer fabrication (Sandia, Kyoto)
- Laser writing (Oxford, Karlsruhe)
- Holography, robotics, etc. etc.







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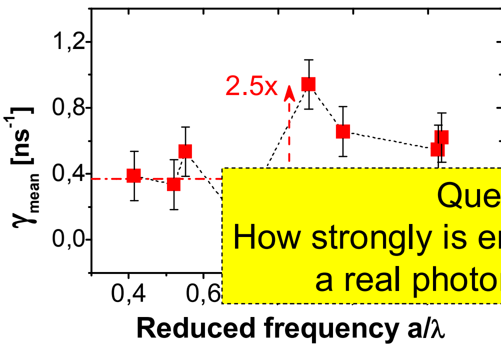
Early results on photonic crystals



Time-resolved emission of QDots in TiO_2 inverse opal, without a band gap:

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Question:
How strongly is emission inhibited in a real photonic band gap?

First ever lifetime control with photonic crystals.

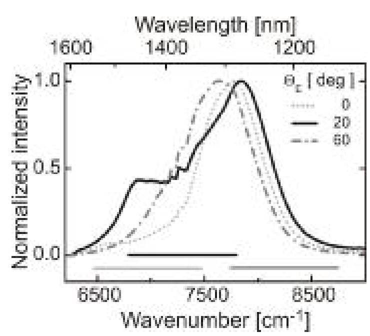
Lodahl, v.Driel, Nikolaev, Irman, Overgaag, Vanmaekelbergh & WLV, Nature **430** (2004) 654
Nikolaev, Lodahl, van Driel, Koenderink & WLV, Phys. Rev. B **75** (2007) 115302

Emitters: near infrared quantum dots

Efficient emitters compatible with silicon (λ>1100 nm):
PbS and PbSe quantum dots.

In our experiments,
quantum efficiency ~80%.

Study quantum dots in suspension:
keep the dots in stable chemical state (reduce blinking).



See also: Husken, Koenderink & WLV, Arxiv.org/abs/0910.5749

Optical geometry and time-resolved

Crystal is sitting in suspension.

Minimize background signal:
excite the crystal at 90°.

(sorry, images temporarily removed, will be published by *Leistikow et al.*)

“Problem” due to band gap:
low emission rate → long averaging.
(We love this kind of problem!)

Time-resolved emission
of good quality.

Emission above the band gap

We observe clear enhancement:
Quantum dots emit faster
(>3x) than outside crystal.

(sorry, images temporarily removed, will be published by *Leistikow et al.*)

Good agreement between
theory and experiment.

Vacuum fluctuations are
strongly enhanced.

Emission from inside the band gap

We observe very long lifetimes:
Quantum dots remain excited
>11x longer than outside gap.

Excellent agreement between
theory and experiment.

Vacuum fluctuations are
strongly inhibited.

Future: probe long-lived
population of excited states.

(sorry, images temporarily removed, will be published by *Leistikow et al.*)

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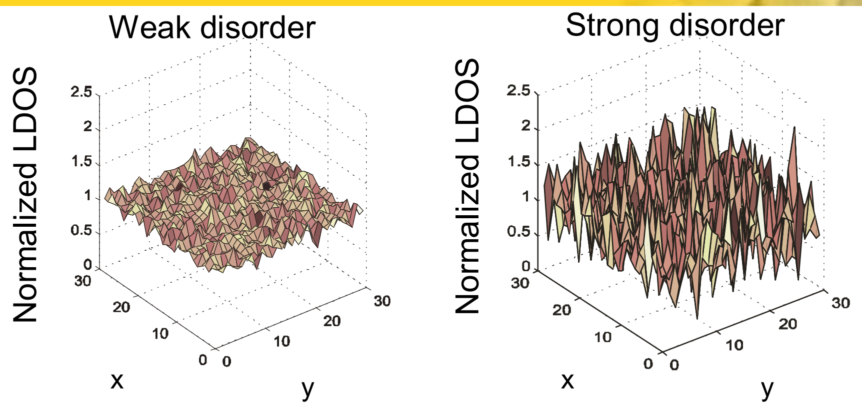
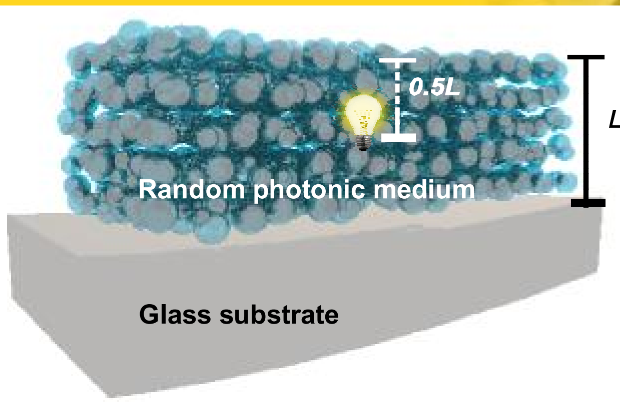
Emission rates in random media?



LDOS fluctuates spatially in random photonic media (speckle).
How is the LDOS related to the disorder, *i.e.*, (1/kℓ)?

B. Shapiro, PRL (1999);
B. van Tiggelen & S. Skipetrov, PRB (2006);
D. Mirlin, Phys. Rep. (2000);
R. Carminati *et al*, PRA (2007).

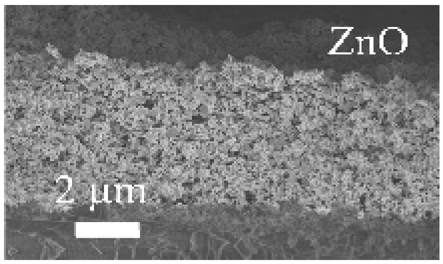
Experimental method

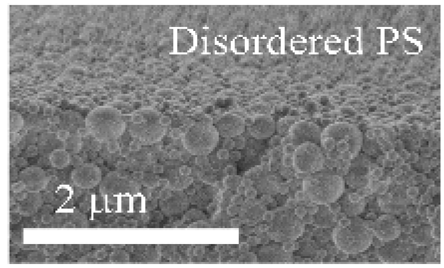


Measure histograms of emission rates from single emitters, embedded *deep inside* random photonic media.

Random photonic media

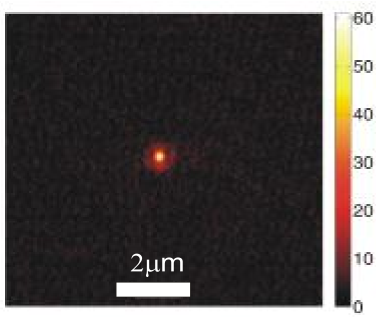
Photonic media: ZnO pigment & polystyrene spheres.
Thickness L = 4 to 20 μm

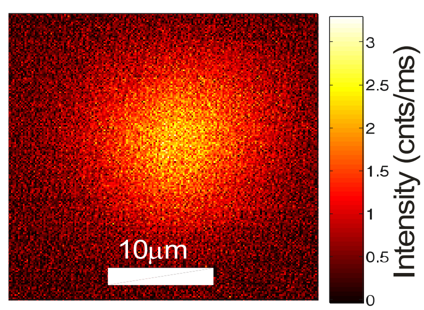




Single probes: D=25 nm polystyrene spheres with dye.
Short lifetime, high quantum efficiency (96%), no blinking.

View single nanosphere emitters



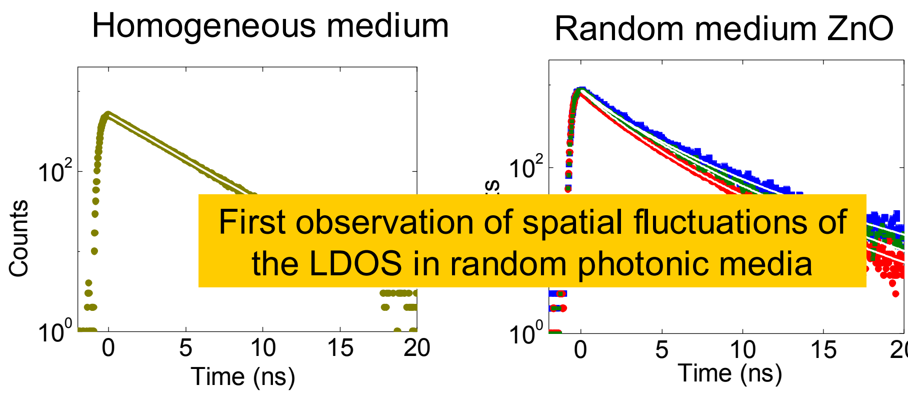


Emitter in homogeneous medium
(PVA polymer layer)

Emitter in random photonic medium
(in ZnO medium)

Diffuse spot size helps to determine source depth.

Time-resolved emission random media



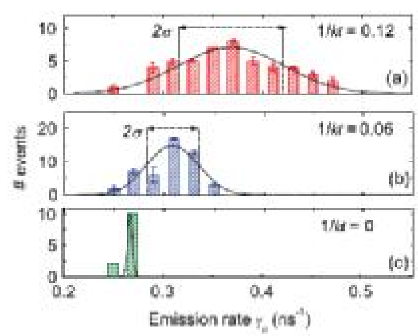
First observation of spatial fluctuations of the LDOS in random photonic media

- Different decay curves for single probes at different positions.
- Non-exponential decay: variation of dye dipole orientations.

Distributions of emission rates

By modeling(*) time-resolved emission obtain average rate.

Obtain distributions of emission rates.

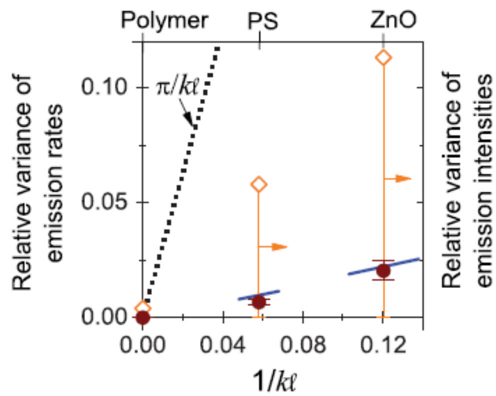


Relative variance (width) of distributions ***increases*** with (1/kℓ)!

(*) Nikolaev, Lodahl, van Driel, Koenderink & WLV, Phys. Rev. B **75** (2007) 115302

Variance increases with disorder

Theory (π/kl) 20x too high:
limitation of scalar waves
& point scatterers.



Good agreement with
new theory that considers
only the closest scatterer.

Diamonds: measured
upper bound to C0
(intensity correlation function): equivalent to LDOS.

Birowosuto, Skipetrov, WLV & Mosk, Phys. Rev. Lett. **105** (2010) 013904

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  Emission versus dipole orientation
  Ultrafast cavity switching
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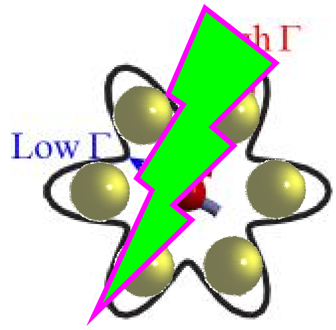
1) Emission rate versus orientation?

How does spontaneous emission decay rate depend on orientation of any transition dipole?

Consider polar plots $\Gamma = \Gamma(\theta, \varphi)$

Can polar plots take on any shape?

Does spontaneous emission rate mimic local symmetry around source? [*e.g.*, emitter in hexagonal cluster.]



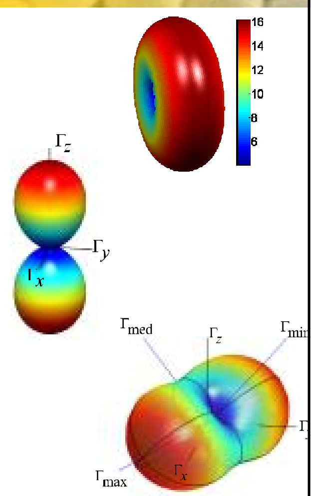
(... this guess is completely wrong!)

Rate vs orientation has high symmetry

Made new systematic classification for emission rate polar plots.

Only used symmetry of Green function:
- Know all with ***only 3*** special rates
- Only ***few*** limited polar plots occur
  (quadratic in orientation vector)

Found efficient method to calculate any orientation dependence, no Green function needed.



WLV, Koenderink & Nikolaev, Phys. Rev. A **80** (2009) 053802

2) Ultrafast switching of a microcavity

Information technology: High-speed all-optical data transfer
Cavity QED: Ultrafast strong coupling

Goal: control switch-on and -off times only by pulses and optics, not by material properties*.
Only achievable via the electronic Kerr effect!

* See: Harding, Euser, Nowicki-Bringuier, Gérard & WLV, APL **91** (2007) 111103

Ultimate fast cavity switching

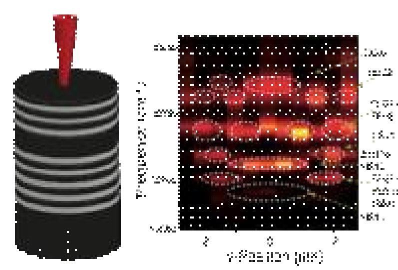
Ultrafast switching of a GaAs microcavity with the electronic Kerr effect

(sorry, images temporarily removed for reasons of priority...)

Fastest possible cavity switch!

Resonance shift can be tuned and controlled

Supra-THz data modulation feasible



Next: switch high-Q micropillar cavities

Ctistis, Hartsuiker, vd Pol, Claudon, WLV & Gérard, Phys Rev B 82 (Nov. 2010) 195330

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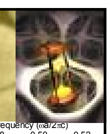
Summary



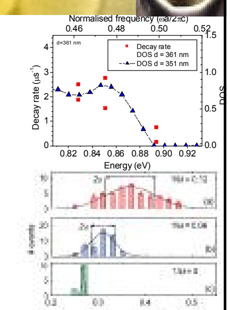
Spontaneous emission in nanophotonic media has interesting complex behavior;
depends on frequency, position, and orientation

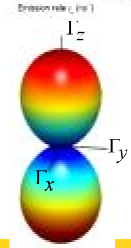
Observed first very long lifetimes in
3D photonic band gap crystals

Observed first long-range spatial fluctuations

Developed new classification for orientation effects

Demonstrate ultrafast <ps cavity switching





Outlook

Finite-size scaling of spontaneous emission:
first ever true experimental probe of band gap,
anticipate novel Anderson localization transition behavior.

Map Anderson modes with *near-field* scanning microscopy.

Engineer LDOS & apply to study properties of emitters:
- InAs quantum dots (Johansen et al, PRB 2008)
- CdSe quantum dots (Leistikow et al, PRB 2009)
- EGFP fluorescent proteins (Cesa et al, PCCP 2009)

Study collective photon behavior (analogy to Cond.Mat.)

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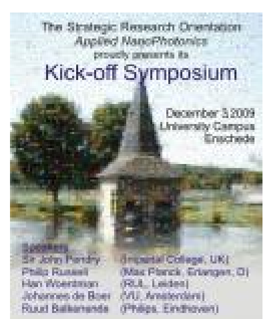
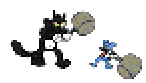


Thank you!

Thank you very much for your attention.

Questions?

Discussion?



(just kidding!)