Chapter 9

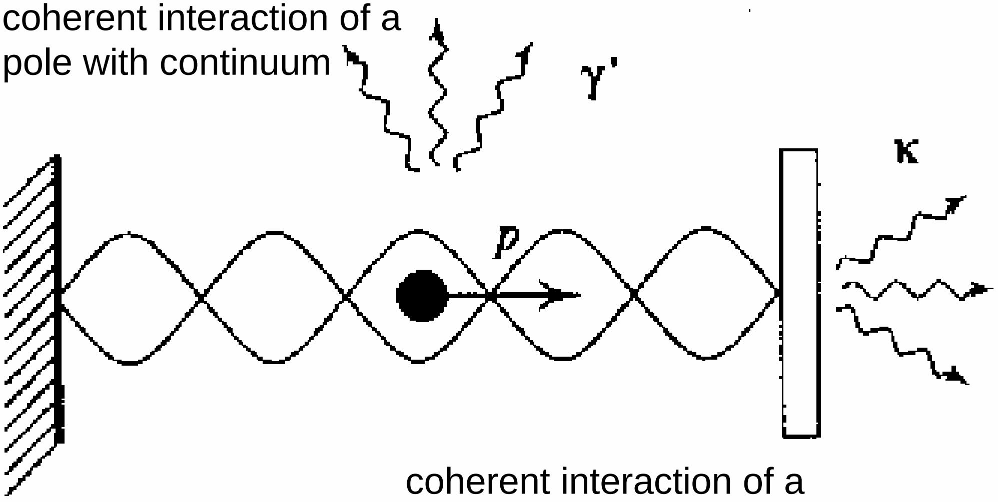
Aspects of Nano-Quantum Optics

9.1. Review of Quantum Electrodynamic Effects

Quantum electrodynamics describes the interaction of quantized matter with a quantized electromagnetic field.

A specific problem is the interaction of a single emitter (atom, molecule, quantum dot) with resonant structures, e.g., a cavity. This problem is addressed in cavity quantum electrodynamics or **cavity QED**.

A typical configuration is given below:



incoherent interaction of a dipole with continuum

incoherent field damping

coherent interaction of a dipole with cavity field

The dynamics can be described in two limiting regimes:

1. The **strong coupling regime**, i.e., the coherent interaction of the emitter with the cavity field described by a coupling constant *g* is the dominant interaction.

   $g \gg \kappa, \gamma'$

2. The **weak coupling regime**, i.e., interaction of the emitter is basically incoherent and dominated by the damping rates κ , γ'

   $g \ll \kappa, \gamma'$

In the theoretical description it is often appropriate to describe the emitter as a two-level system and to consider the interaction with a single cavity mode in dipole approximation.

Cavity QED has long been a domain of atomic physics, but the solid state systems (e.g., involving single quantum dots) have recently entered the stage.

9.1.1. The Strong Coupling Regime

In a first approximation it is possible to neglect damping at all. Then, the interaction of the single emitter with a single mode of the cavity is determined by the **Jaynes-Cummings-Hamiltonian**.

$$\boxed{H_{JC} = \frac{1}{2}\hbar\omega_0\sigma_z + \hbar\omega a^+ a + \hbar g(a\sigma^+ + a^+\sigma^-)}$$

transition frequency

Pauli spin operator

cav. resonance frequency

coupling constant

atomic state lowering operator

This Hamiltonian only couples states |*n,e*> with |*n+1,g*> where we denote with |*e*> and |*g*> the excited and ground state of the two-level system and |*n*> is a photon number state.

It thus suffices to describe *H* in this basis and define:

$$H_n = \hbar\left(n + \frac{1}{2}\right)\omega\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + \hbar\begin{pmatrix} -\delta/2 & g\sqrt{n+1} \\ g\sqrt{n+1} & \delta/2 \end{pmatrix}$$

with the detuning $\delta = \omega_0 - \omega.$

The general state is $|\psi(t)> = c_{en}(t)|e,n> + c_{gn+1}(t)|g,n+1>$

In resonance the **eigenenergies** are:

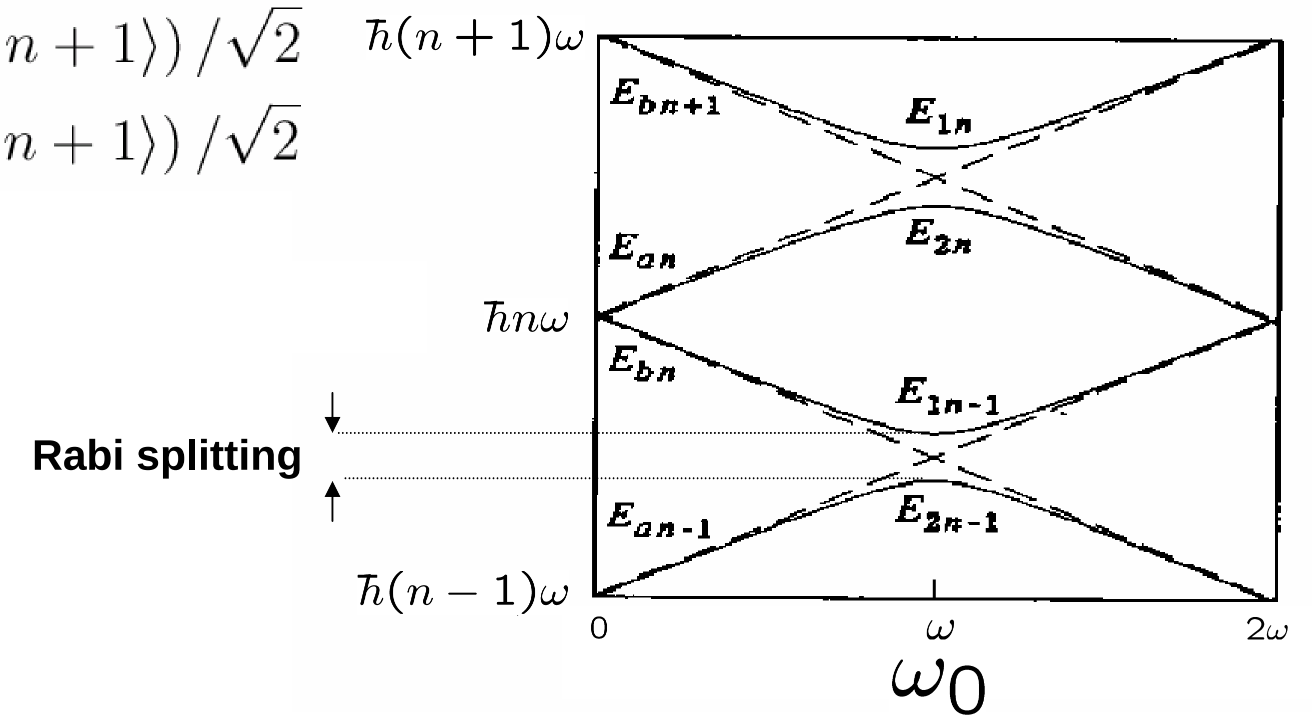
$$E_{2n} = \hbar\left(n+\frac{1}{2}\right)\omega - \hbar g\sqrt{n+1}$$

$$E_{1n} = \hbar\left(n+\frac{1}{2}\right)\omega + \hbar g\sqrt{n+1}$$

The **eigenstates** are called the **dressed states**. In resonance they are::

$$|2n\rangle = \left(|e,n\rangle - |g,n+1\rangle\right)/\sqrt{2}$$

$$|1n\rangle = \left(|e,n\rangle + |g,n+1\rangle\right)/\sqrt{2}$$



The coupled equation of motion for the coefficients $c_{en}(t)$ and $c_{gn+1}(t)$ are:

$$\dot{c}_{en} = -i\frac{\delta}{2}c_{en} - ig\sqrt{n+1}c_{gn+1}$$
$$\dot{c}_{gn+1} = i\frac{\delta}{2}c_{gn+1} - ig\sqrt{n+1}c_{en}$$

This gives for a state initially in the upper state:

$$|c_{en}(t)|^2 = \cos^2(g\sqrt{n+1}t)$$
$$|c_{gn+1}(t)|^2 = \sin^2(g\sqrt{n+1}t)$$

Even if $n = 0$ (no photon or interaction with the vacuum) there is:

$$\boxed{|c_{e0}(t)|^2 = \cos^2(gt) \quad \text{g is called vacuum Rabi-frequency}}$$

Thus there is a coherent exchange of one energy quantum between the emitter and the field mode, the so-called **vacuum Rabi oscillation**, in striking difference to the irreversible exponential decay into free space of an excited atom.
The periodic energy exchange has an analogy with two coupled pendula.

9.1.2. The Weak Coupling Regime

This regime is conveniently described by Fermi‘s golden rule:

The transition rate of an excited emitter (due to its interaction with the continuum of vacuum modes) is:

$$\boxed{\frac{dP_e(t)}{dt} = \left(\frac{2\pi}{\hbar^2}\right) |\langle \mu_{12} E \rangle|^2 D_{free}(\omega)}$$

dipole moment

with $D_{free} = \dfrac{\omega^2 V}{\pi^2 c^3}$ free space density of states

For spontaneous emission in free space this gives:

$$\boxed{\Gamma_{free} = \frac{\omega_0^3 \mu_{12}^2}{3\pi\varepsilon_0 \hbar c^3}}$$

If the emitter is surrounded by a cavity, the density of states of the vacuum is changed. One replaces D_{free} with D_{cav} and puts instead:

$$\boxed{D_{cav}(\omega) = \frac{\kappa}{2\pi V}\frac{1}{(\kappa/2)^2 + (\omega_{cav} - \omega)^2}}$$

This results in

$$\boxed{\Gamma_{cav} = \frac{3Q\lambda^3}{4\pi^2 V}\frac{\kappa^2}{4(\omega-\omega_{cav})^2+\kappa^2}\frac{|\mu_{12}\hat{E}|^2}{|\mu_{12}|^2}\Gamma_{free}}$$

$$Q = \omega_{cav}/\kappa$$

Cavity Q-factor

In resonance (and perfect matching of dipole orientation)

$$\boxed{\Gamma_{cav} = \frac{3}{4\pi^2}\left(\frac{\lambda_0^3}{V}\right) Q\ \Gamma_{free}}$$

enhancement of spontaneous emission

Off resonance (and perfect matching of dipole orientation)

$$\boxed{\Gamma_{cav} = \frac{3}{16\pi^2}\left(\frac{\lambda_0^3}{V}\right) Q^{-1}\ \Gamma_{free}}$$

suppression of spontaneous emission

The important result is that the „natural“ lifetime of an excited emitter is not a constant.
It can be modified using **high-Q cavities**.

The fraction

$$\boxed{\Gamma_{cav}/\Gamma_{free} = \frac{3Q\lambda^3}{4\pi^2 V} \quad \text{is called Purcell-factor.}}$$

In order to obtain a large Purcell factor:

- the Q-factor of the cavity should be large
- the mode volume should be small
- the spectral width of the emitter should be smaller than the cavity width

In particular the last requirement is difficult to fulfill in the solid state system.
It requires narrow-band (atom-like) emitters such as semiconductor quantum dots.

9.1.3. Intermediate Regime

The general solution for $c_e(t)$ for arbitrary g, γ', κ is of the form

$$c_e(t) = c_{e1} e^{\alpha_1 t} + c_{e2} e^{\alpha_2 t}$$ with

$$\alpha_{1,2} = -\frac{1}{2}\left(\frac{\gamma'}{2} + \frac{\kappa}{2} + i\delta\right) \pm \frac{1}{2}\left[\left(\frac{\gamma'}{2} + \frac{\kappa}{2} + i\delta\right)^2 - 4g^2\right]^{1/2}$$

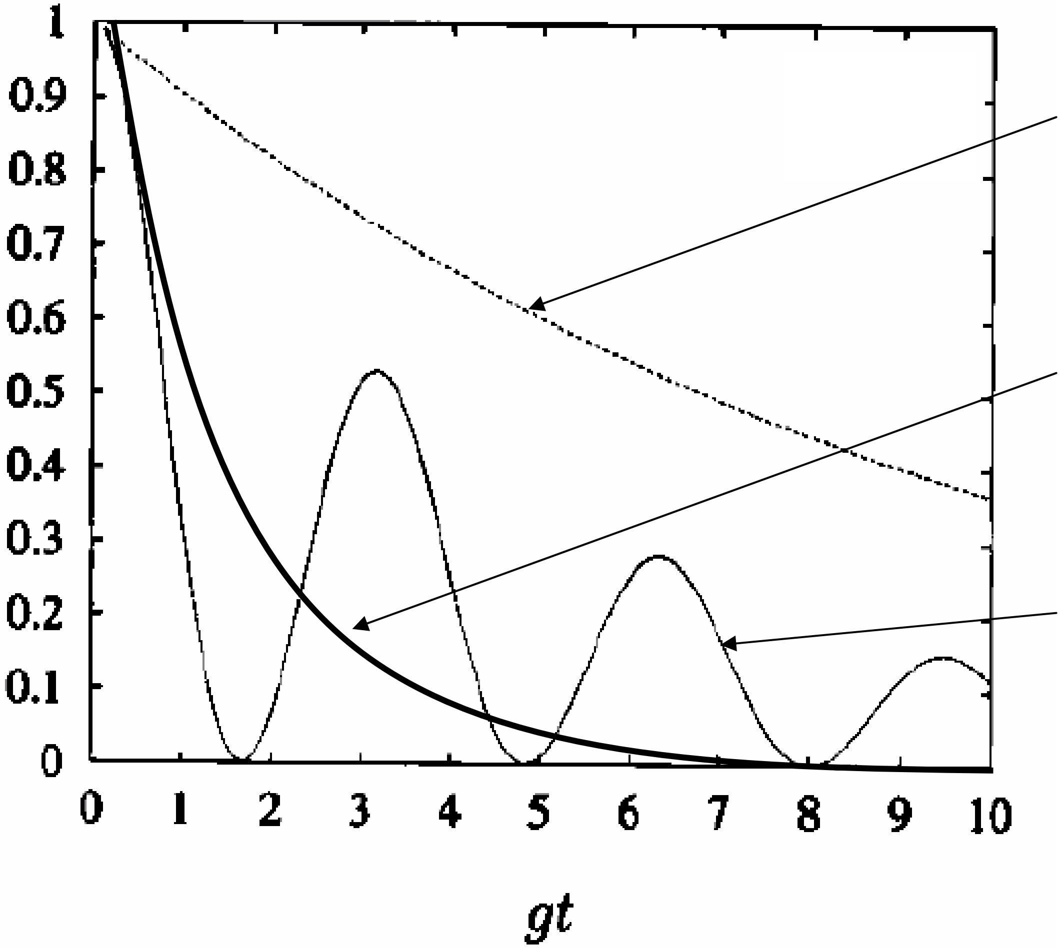
with some coefficients c_{e1} and c_{e2}.

If $g \gg \gamma', \kappa$ it follows:

$$\alpha_{1,2} = -\frac{1}{2}\left(\frac{\gamma'}{2} + \frac{\kappa}{2} + i\delta\right) \pm ig$$

This leads to a damped oscillation of $|c_e(t)|^2$ with the Rabi frequency g and whith a damping constant $(\gamma' + \kappa)/4$.

The time evolution of the probability to find the (initially excited) emitter in its upper state is:



free space decay

enhanced decay rate
in the weak coupling regime

oscillatory behaviour
(with some damping)
in the strong coupling regime

The time evolution corresponds to a characteristic spectrum of the emitted light:

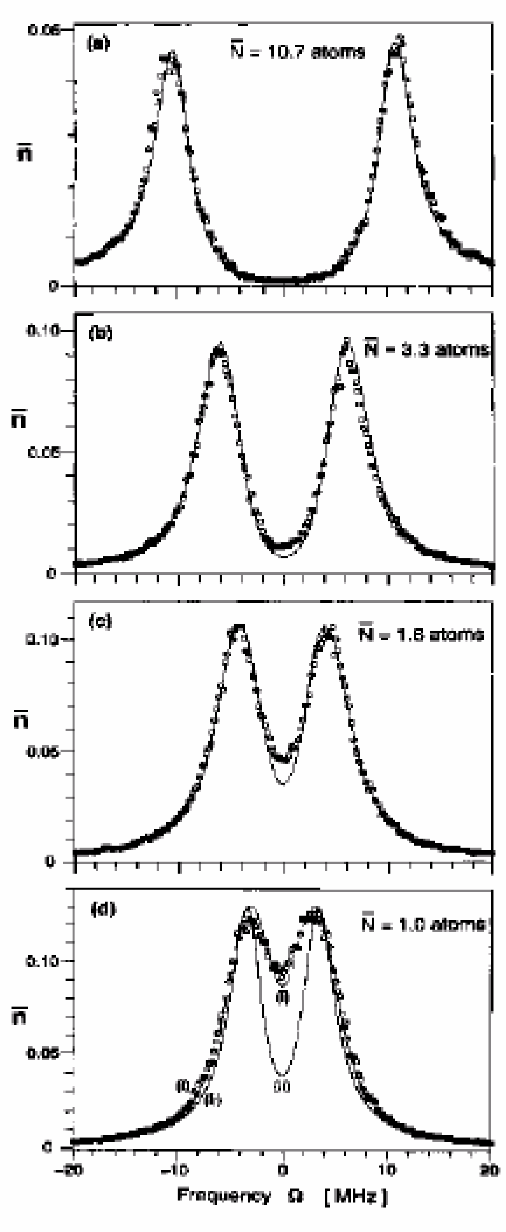
Weak coupling regime:

Lorentzian with a width proportional to the modified decay rate

Strong coupling regime:

Double peak of two Lorentzians with a width proportional to the damping rate

The right figure shows the normal mode splitting a coupled atom-cavity system with increasing coupling strength (from bottom to top).



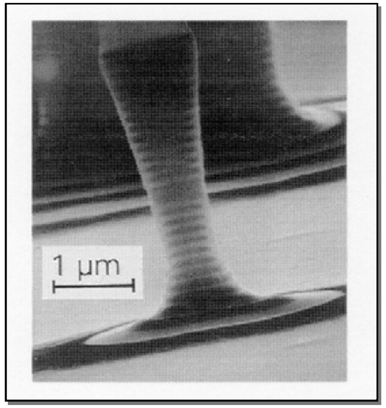
increasing effective g

9.2. Cavities in Solid State Cavity QED

In order to observe cavity QED effects cavities of high Q-factor and small mode volume are required.

In solid state systems basically three types of cavities are used:

1. Pillar cavities
2. Whispering-Gallery mode cavities
3. Photonic crystal cavities



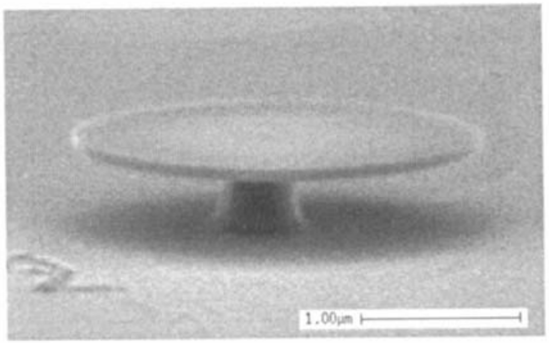
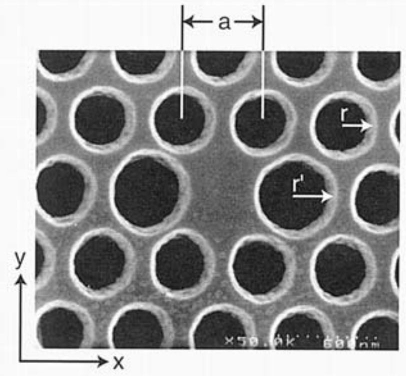


FIG. 1. Scanning electron micrograph of a microdisk

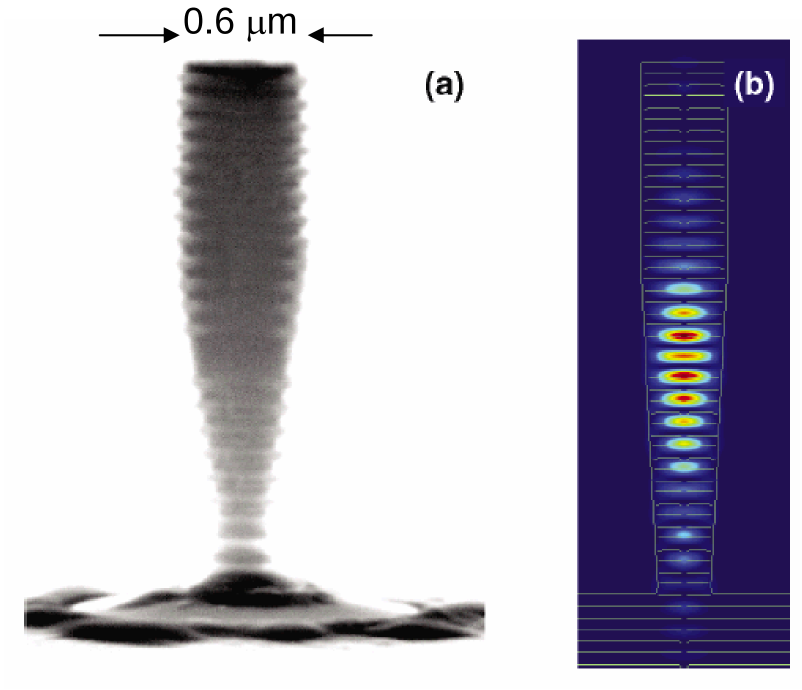


9.2.1. Pillar Cavities

Pillar cavities are etched out of pairs of Bragg mirrors.

They have first been exploited for VCSLs (vertical cavity surface emitting lasers)

The following picture shows a pillar cavity together with the field distribution of a resonant mode.



M. Pelton et al., Phys. Rev. Lett. 89, 233602 (2002)

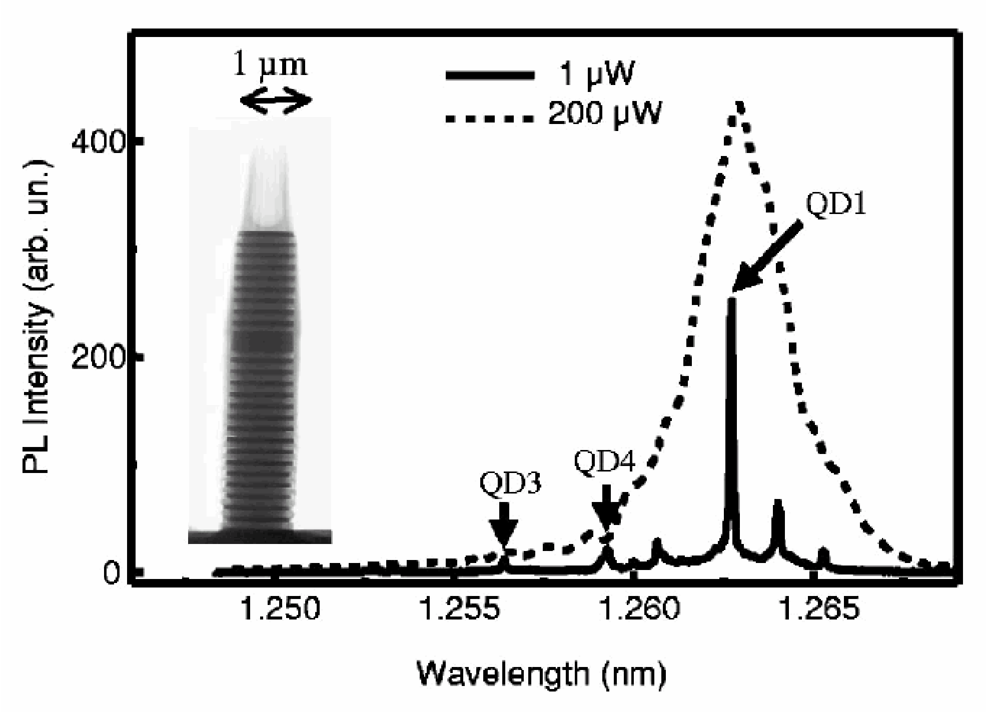
The confinement in radial direction is provided by the index contrast (total internal reflection).

Due to scattering loss Q-factors are limited to a few 1000s.

The mode volume is a few $(\lambda/n)^3$

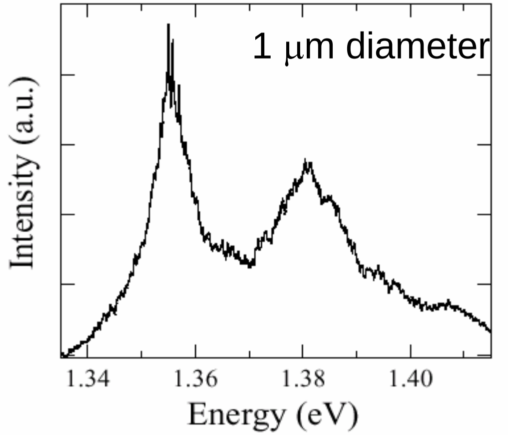
Purcell-factors of about 30 have been obtained.

- Quantum dots are grown in between the two Bragg mirrors of a pillar cavity before etching.
- With a low dot density, small pillar diameter, and wavelength matching coupling to single dots is possible.
- Tuning of the dot to a resonance is obtained by changing the sample temperature.
- The cavity mode structure can be studied by embedding broad-band emitters (quantum wells, quantum dots of high density, or quantum dots under strong excitation)

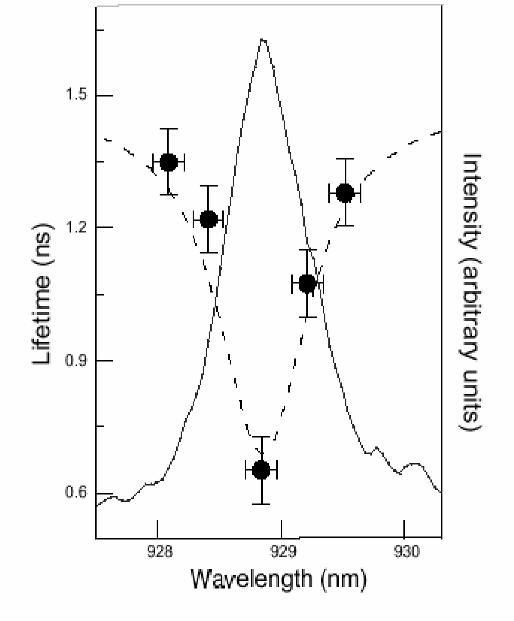


Enhanced luminescence from quantum dots. Emission lines are from different dots, but only dot 1 is in resonance with the cavity.

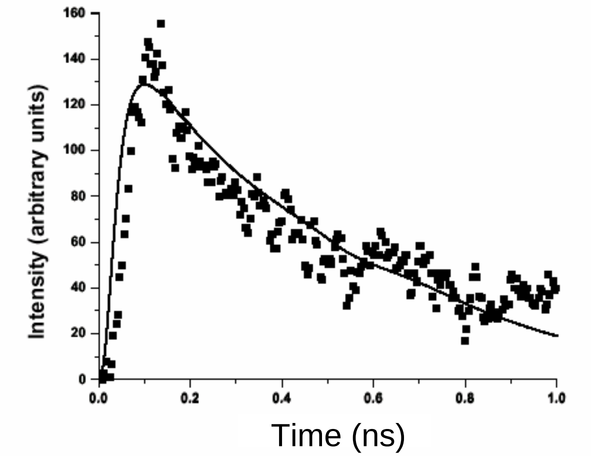
Moreau et al., Appl. Phys. Lett 79, 2865 (2001) spectrum of a small pillar cavity; two modes are visible



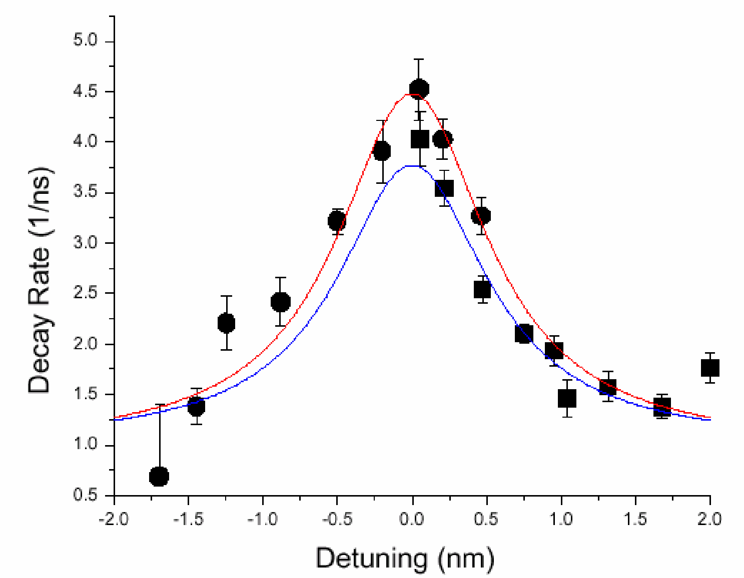
lifetimes of 5 different (emission wavelength) dots within a cavity



time resolved PL from a single dot coupled to a cavity mode



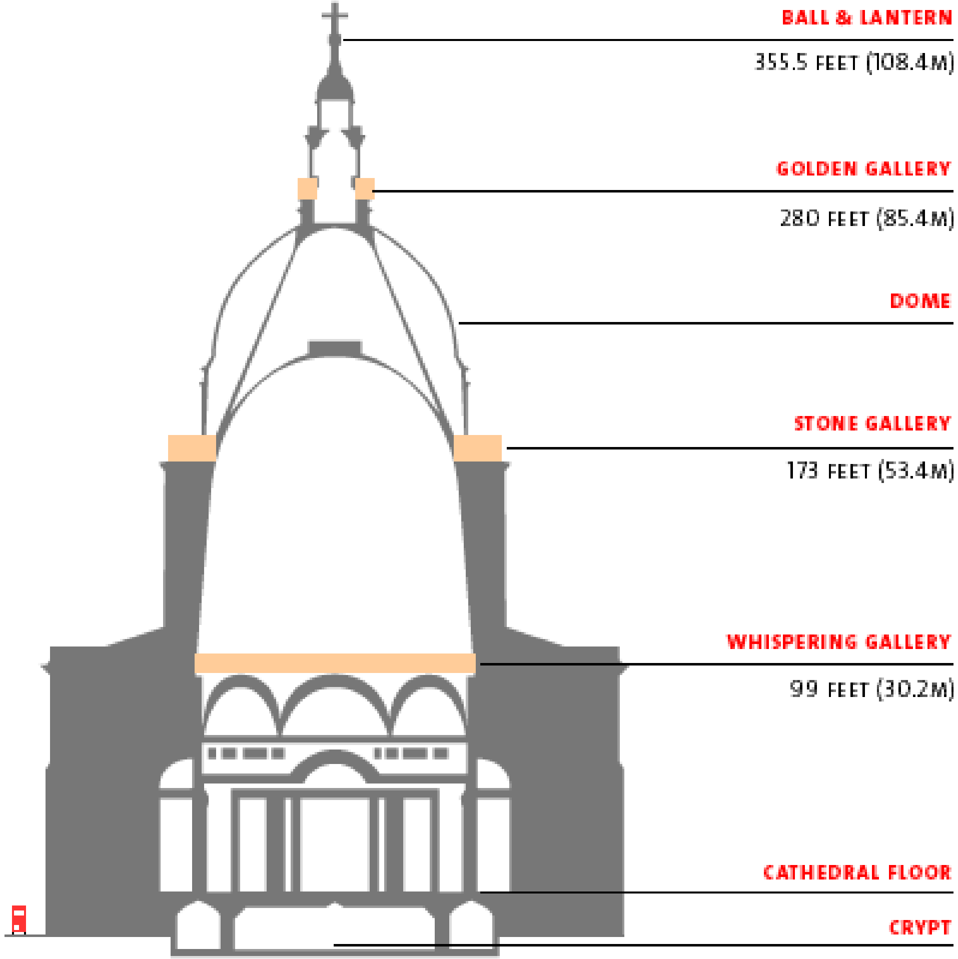
tuning of a single dot through resonance by changing the temperature

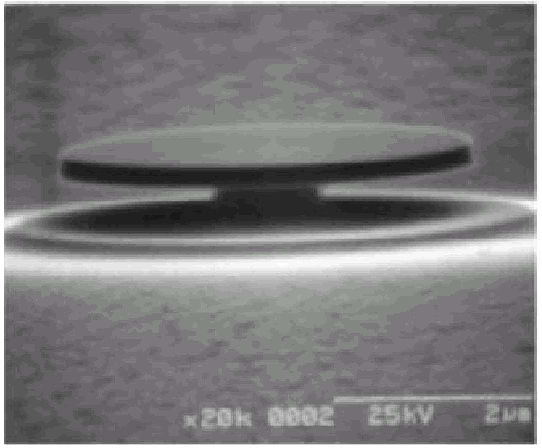


[M. Pelton, thesis, Stanford 2002]

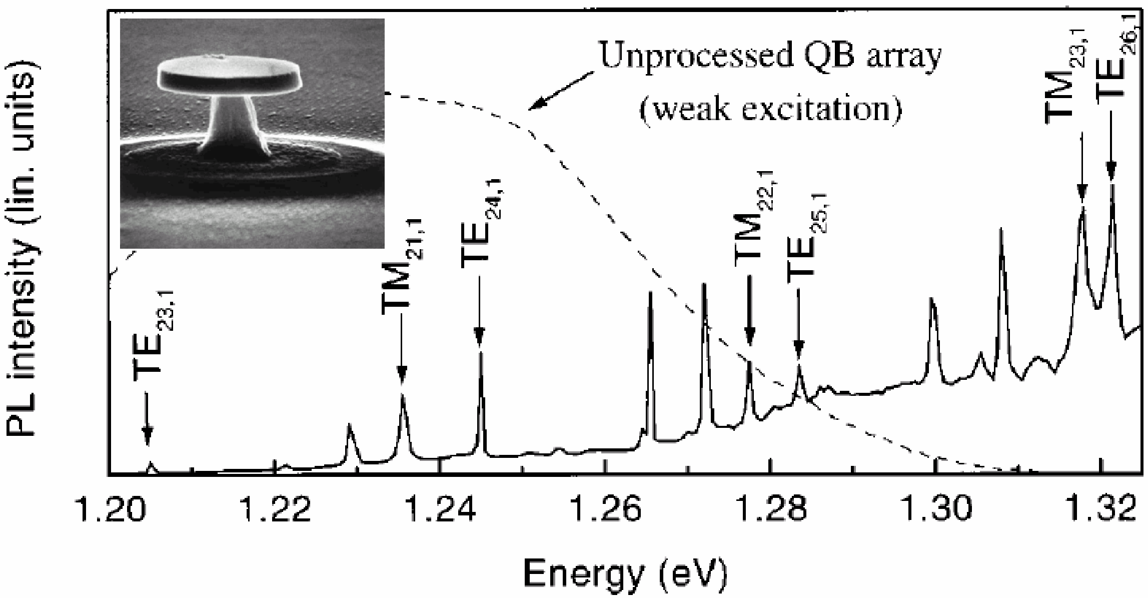
9.2.2. Whispering-Gallery mode cavities

The name Whispering-Gallery modes (WGM) was coined by Rayleigh (1878) in his studies of certain acoustic modes, e.g., in St. Paul‘s cathedral





SEM picture of a microdisc with diameter 5 µm [Michler et al., Science 290, 2282 (2000)]



Spectrum of a microdisc with a diameter of 3 µm [Gayral et al., Appl. Phys. Lett. 75, 1908 (1999)]

In WGM cavities the light is confined by total internal reflection. It travels along a rim inside the cavity.

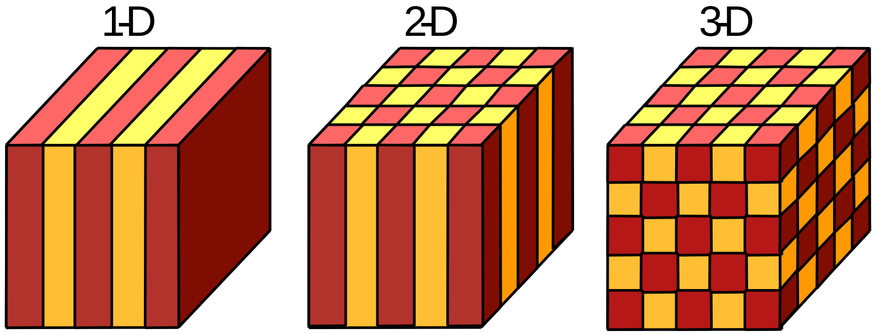
The mode-volume of small micro-discs is on the order of $(\lambda/n)^3$. The Q-factor is limited by surface roughness (etching process), but can still be as high as 12.000.

Micro-discs provide a very high Purcell-factor (exceeding 150).

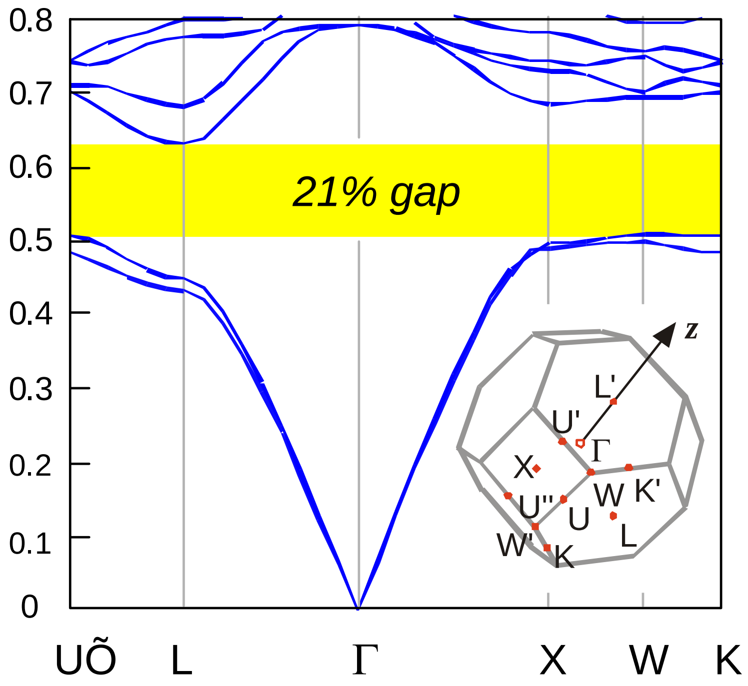
9.2.3. Photonic Crystal Cavities

Photonic crystals are electromagnetic media with a modulated index of refraction (a strong index contrast is obtained, e.g., by drilling holes in a dielectric medium).

Similar as for the electron wave, e.g. in a crystal, there exist forbidden energy gaps (photonic bandgaps) where light cannot propagate.



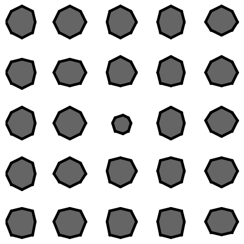
1D, 2-D, and 3-D photonic crystals
[from J. D. Joannopoulos homepage, MIT]

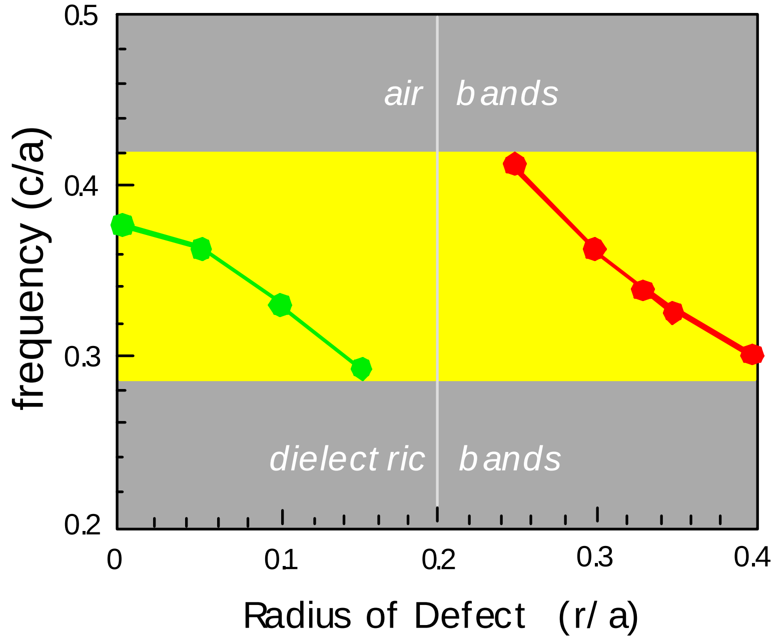


Complete bandgap calculated for a 3D structure
[S. G. Johnson et al., Appl. Phys. Lett. **77**, 3490 (2000)]

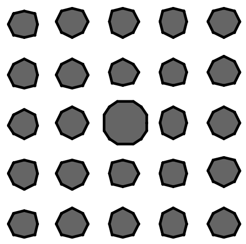
Light can be localized very tightly close to a defect in a photonic crystal structure (e.g., a smaller, larger, or missing hole). This is the principle of a photonic crystal cavity. Advanced structure designs allow Q-factors > 10^6 and mode volumes ~ $(\lambda/n)^3$

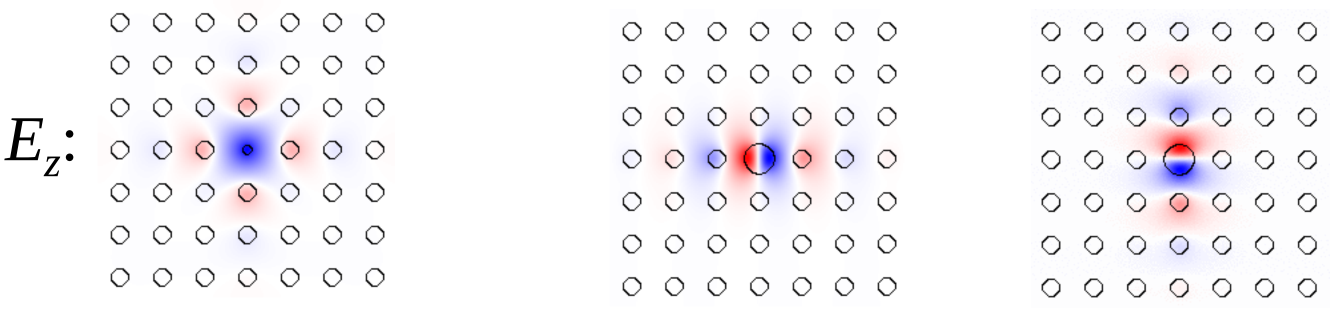
Air Defect





Dielectric Defect





monopole

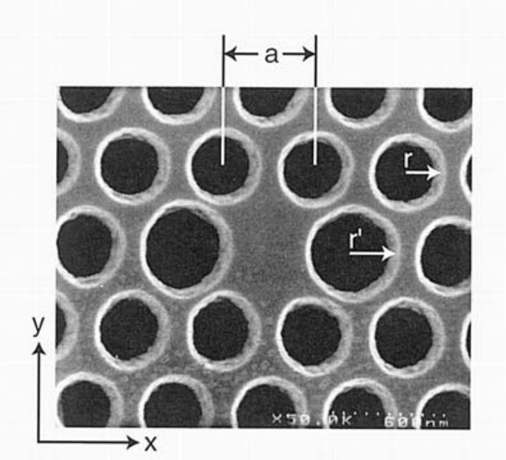
dipole

[from J. D. Joannopoulos homepage, MIT]

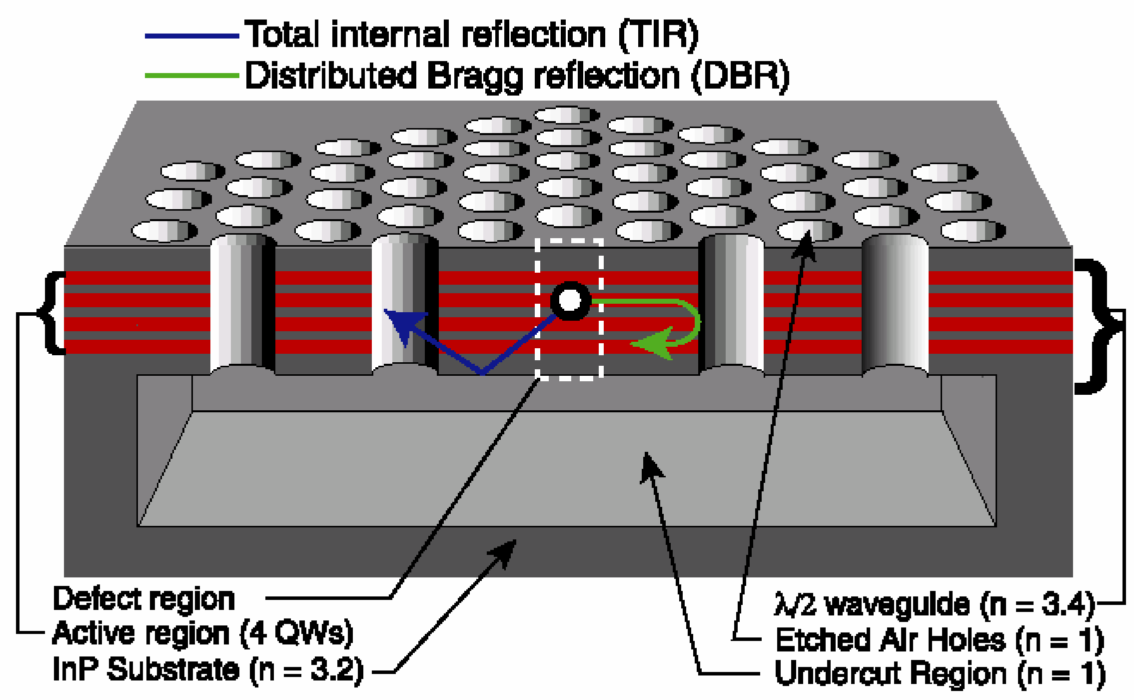
A specific photonic crystal cavity is a 2D slab cavity.

Out of plane confinement is provided by the high index contrast (total internal reflection).

Slab cavities can be produced by e-beam lithography and etching techniques.



PC slab cavity



PC cavity with active material for a PC defect mode laser [Painter et al., Science 284, 1819 (1999)]

9.3. Realization of Strong Coupling

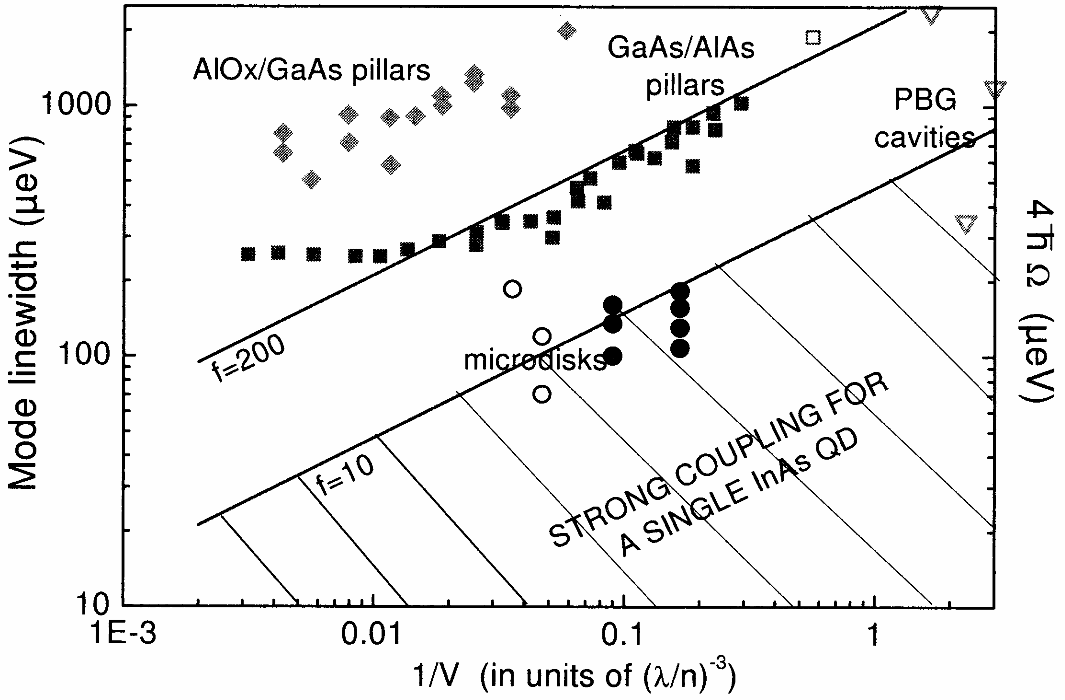
Quantum dots in solid state cavities are good candidates to observe strong coupling:

- Tight confinement leads to a large vacuum field, e.g., 2×10^5 V/cm with $V_{mode} = (\lambda/n)^3$
- Quantum dots have a large electric dipole moment (about 10^{-28} Cm) which is even larger than typical values for atomic transitions (e.g. the Rb 5s-5p transition)

However, broadening of transitions in a solid state system, e.g., due to phonon interaction is much more significant than in the perfectly isolated atomic system.

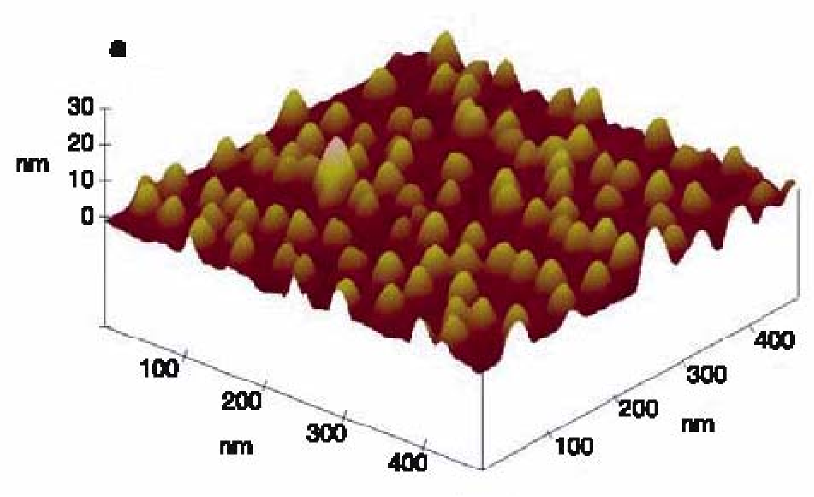
There is a trade-off between cavity size and Q-factor when choosing the best cavity to obtain strong coupling.

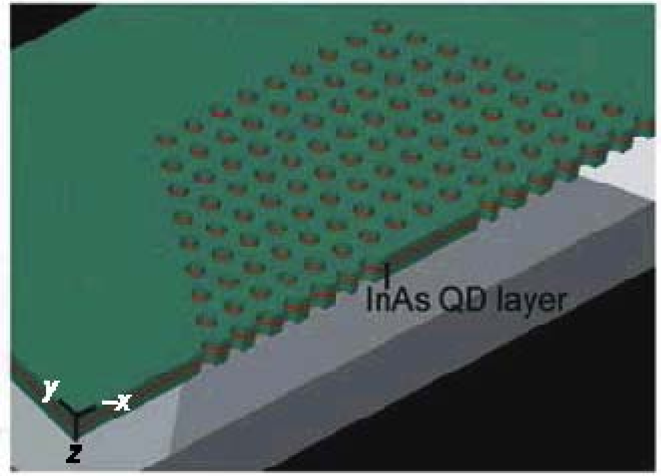
J.M. Gerard in „Single Quantum Dots“, eds. P. Michler



9.3.1. Strong Coupling in a Photonic Crystal Cavities

Interestingly, strong coupling has been demonstrated with three different cavities. It is proven by detecting the splitting of the coupled atom-field states (vacuum Rabi splitting).

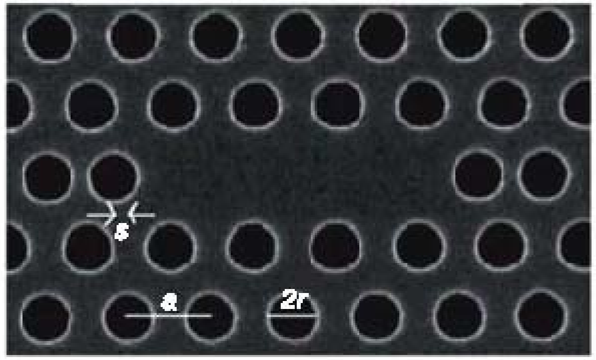




System: InAs quantum dots in a PC slab cavity

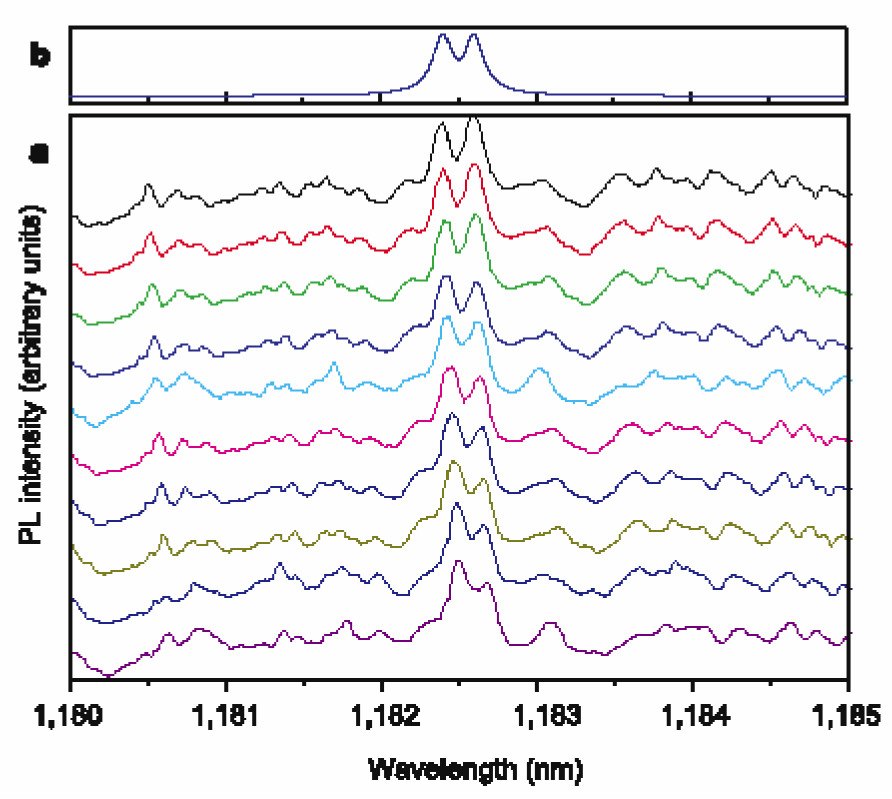
V_{mode}: 0.04 μm^3

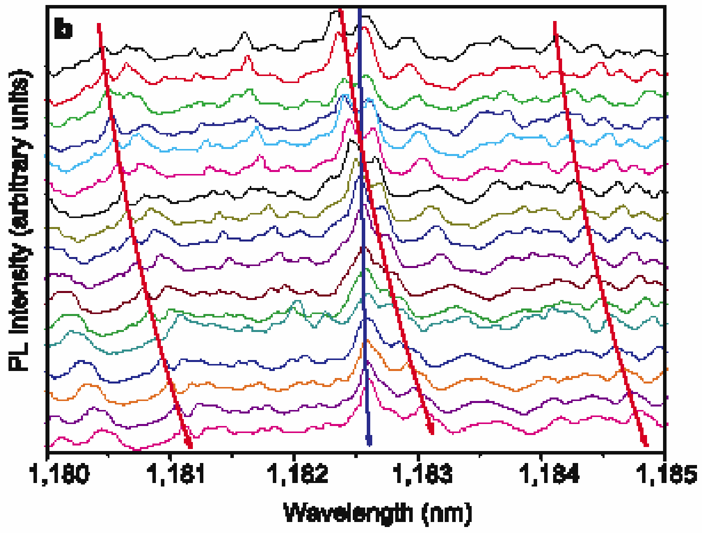
Q: 13,300

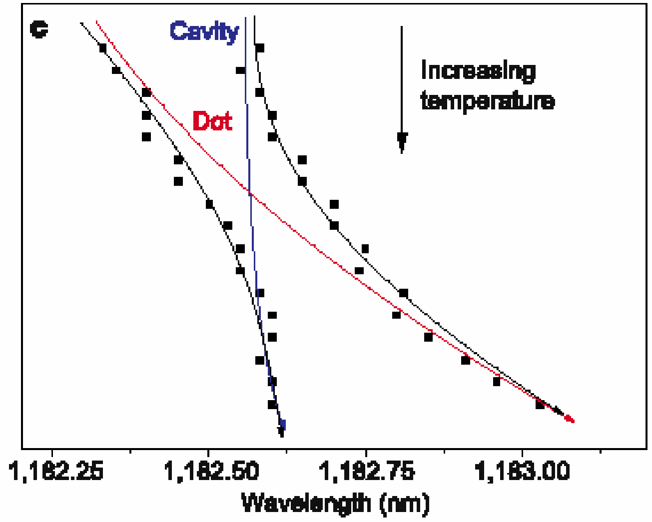


[Yoshie et al., Nature 432, 200 (2004)]

Observation of anti-crossing of quantum dot emission and cavity resonance line (Rabi splitting) when the temperature is changed.

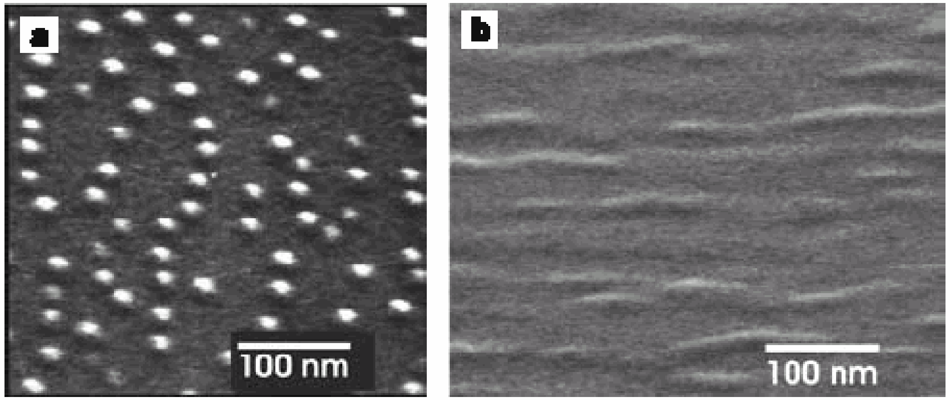




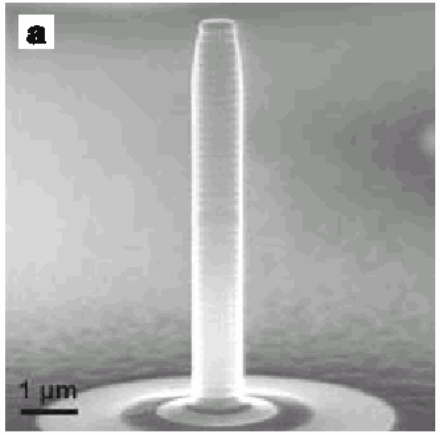


[Yoshie et al., Nature 432, 200 (2004)]

9.3.2. Strong Coupling in a Pillar Cavities



Enlarging quntum dots by higher Indium content

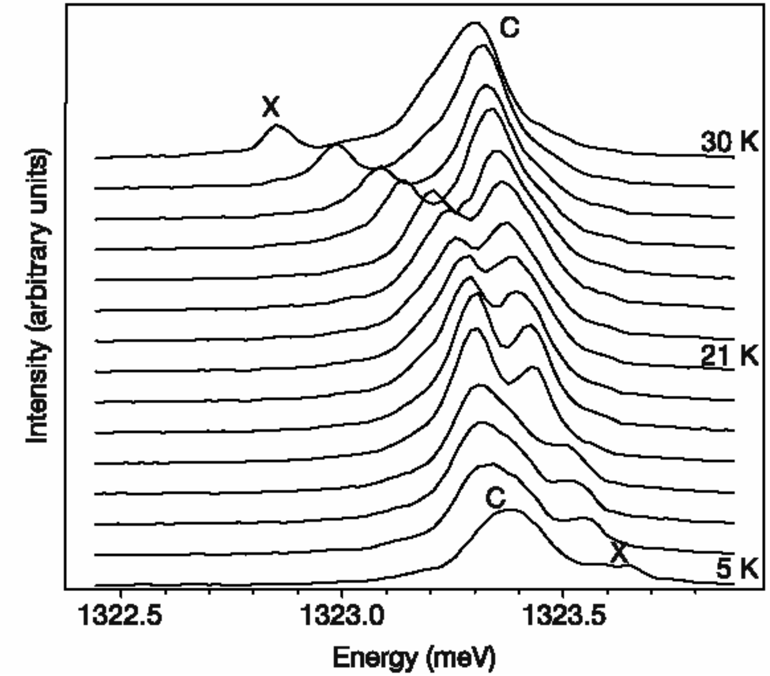


System: InAs quantum dots in a pillar cavity

V_{mode}: 0.04 μm^3

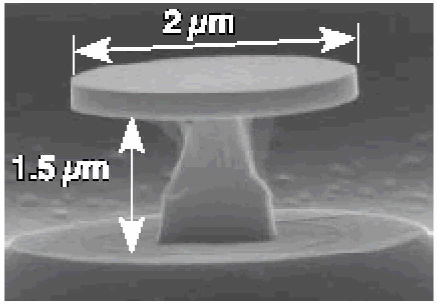
Q: 8,800

right:
tuning of the qdot through resonance



[Reithmaier et al., Nature 432, 197 (2004)]

9.3.3. Strong Coupling in a Micro-disc

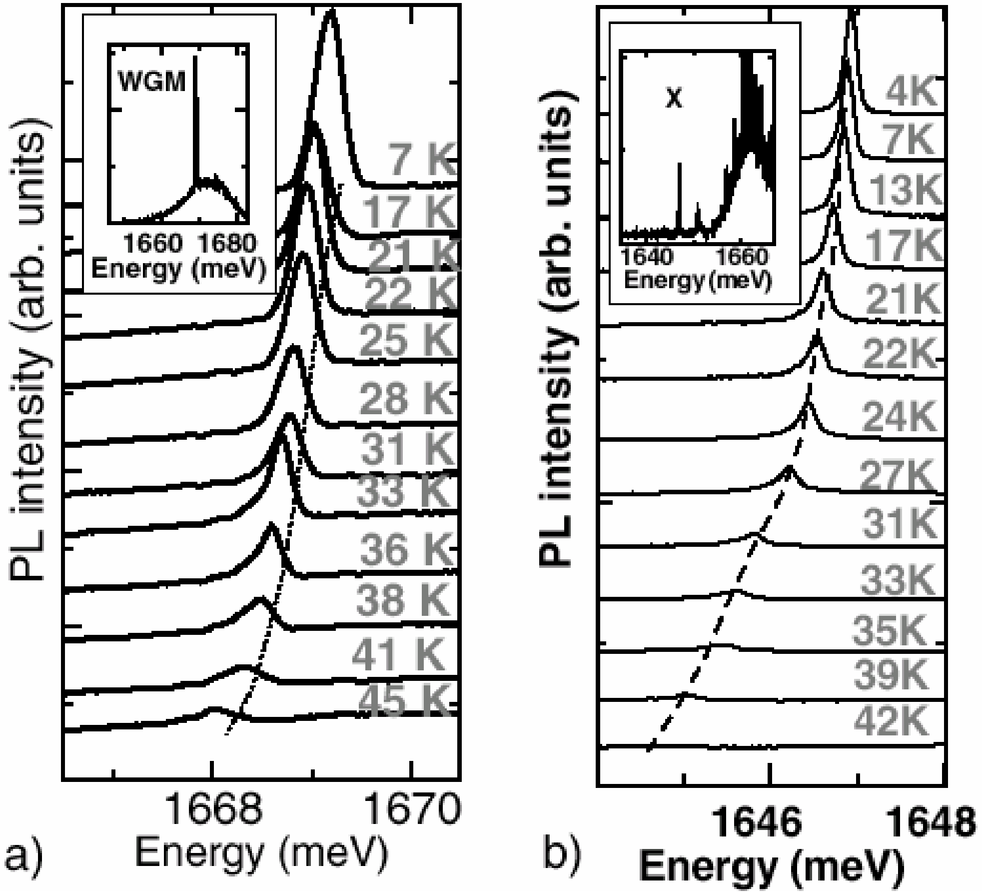


Microdisc cavity

System: GaAs quantum dots in a microdisc cavity

V_{mode}: 0.07 μm^3

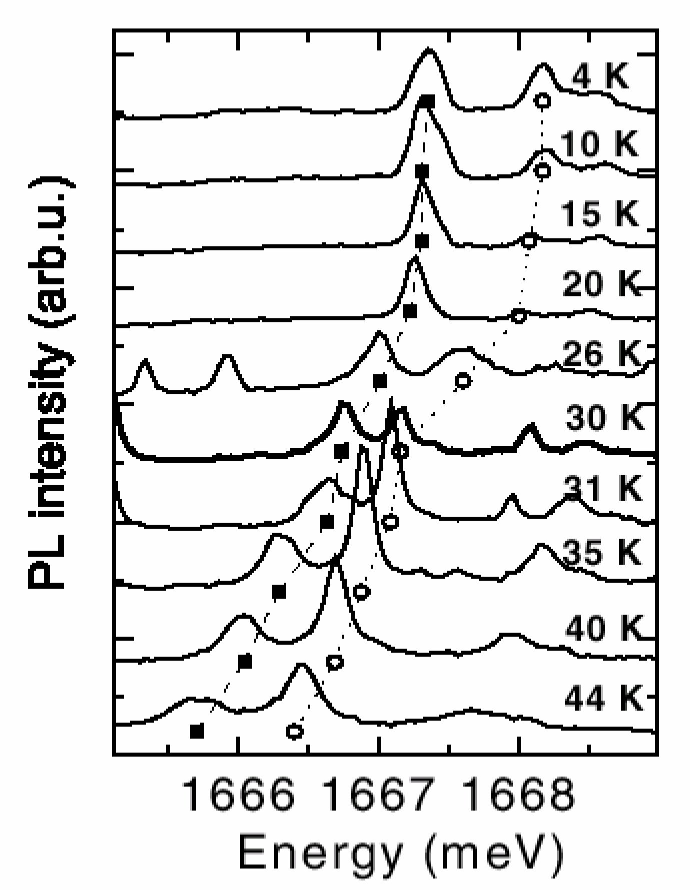
Q: 12,000

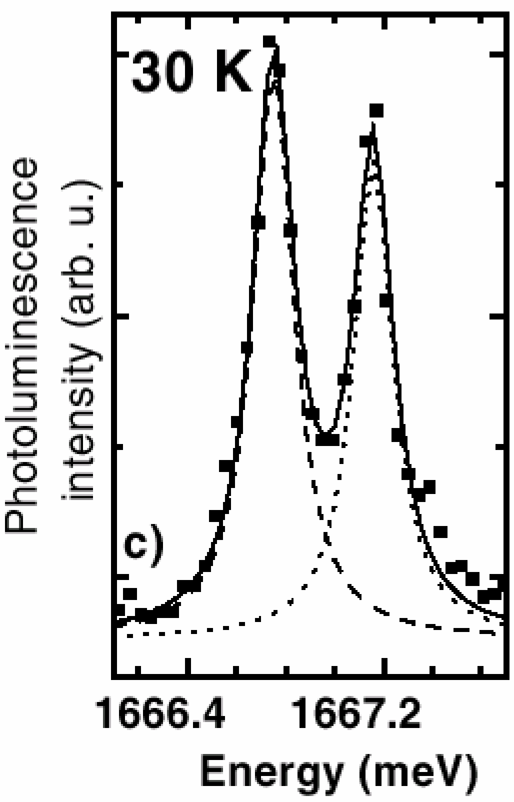


Temperature tuning of the quantum dot

[Peter et al., Phys. Rev. Lett. 95, 067401 (2005)]

Observation of Rabi splitting when the temperature is changed.





[Peter et al., Phys. Rev. Lett. 95, 067401 (2005)]

9.4. Hybrid Systems

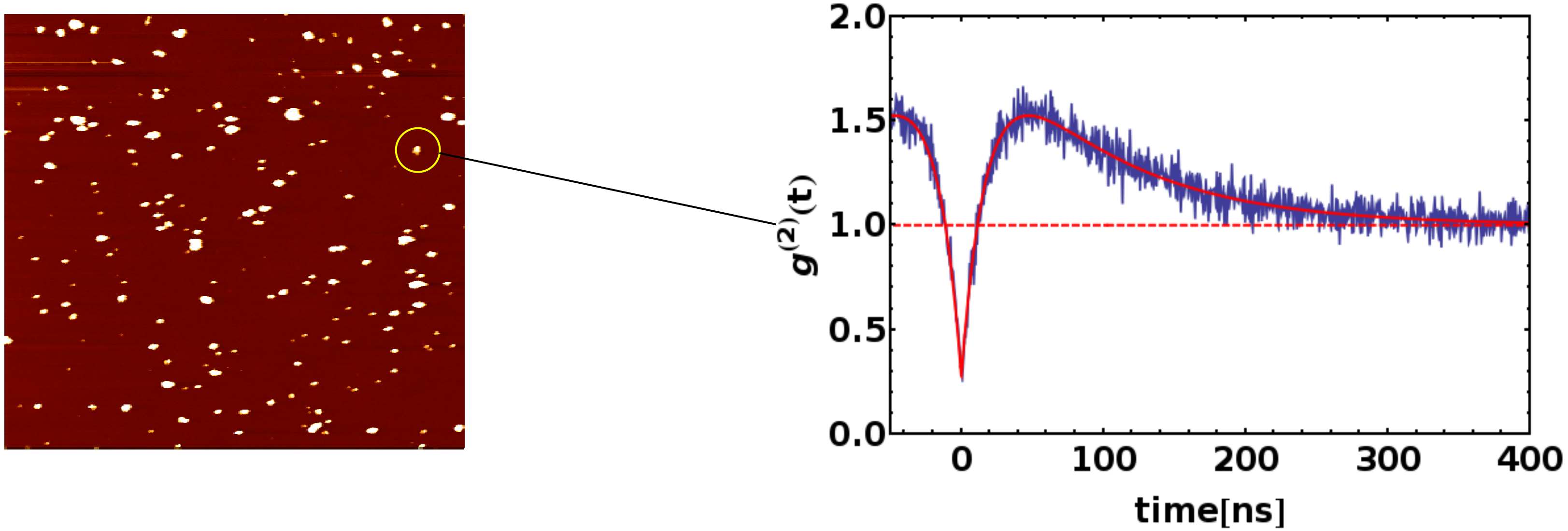
It is not yet possible to grow quantum dots at pre-defined positions and at the same time realize emission of Fourier-limited photons.

A **hybrid approach** utilizes optimized cavities and emitters from a different material system.

One possible hybrid system consist of small nano diamonds (40-90 nm) containing single defect center which are manipulated on photonic structures using scanning probe techniques.

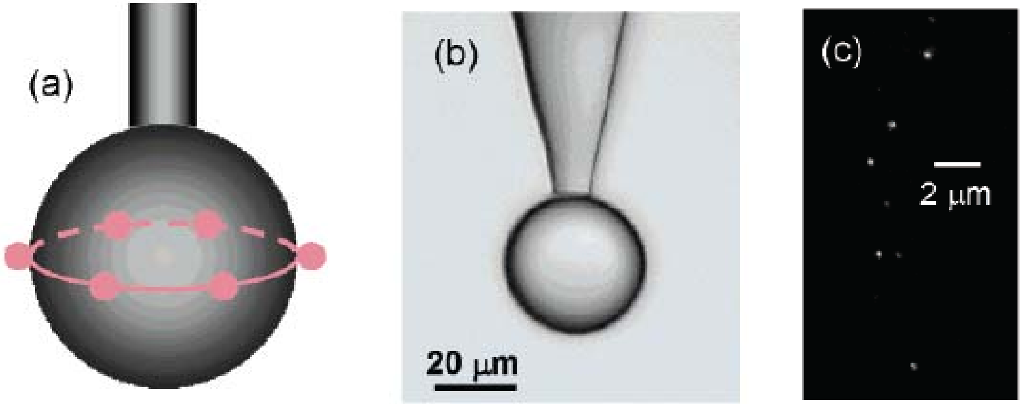
dispersed nano diamonds on a glass substrate

anti-bunching measurement from a single NV center:



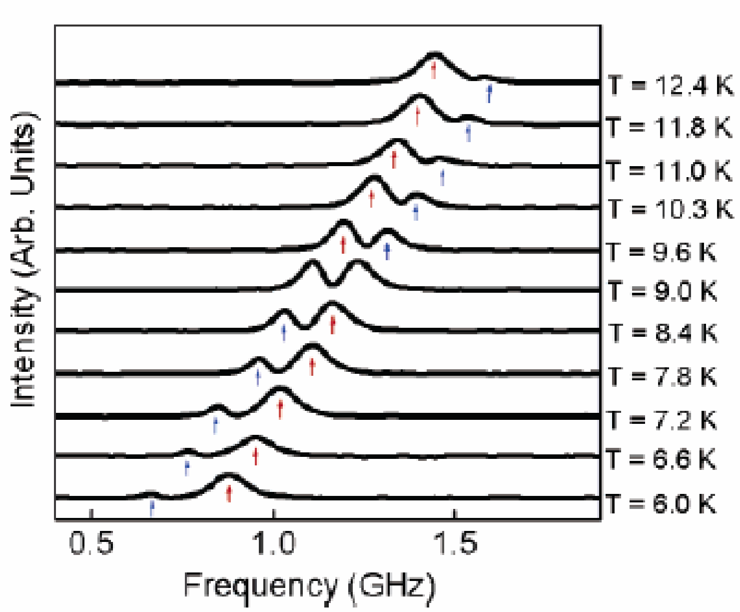
→ AFM manipulation allows to position single nano diamonds at will.

9.4.1. Microspherical Resonators



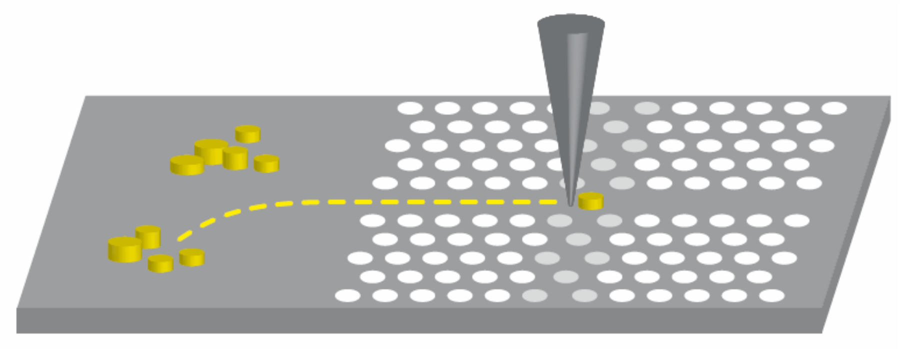
Schematics of a microsphere resonator (a), optical image (b) and microscope image of individual nano-diamonds coated on the microsphere (c).
[Park et al., Nano Lett. 6, 2075 (2006)]

Right:
Characteristic anti-crossing of cavity-line (red arrow) and NV- fluorescence (blue arrow) when the temperature of the microsphere is changed.



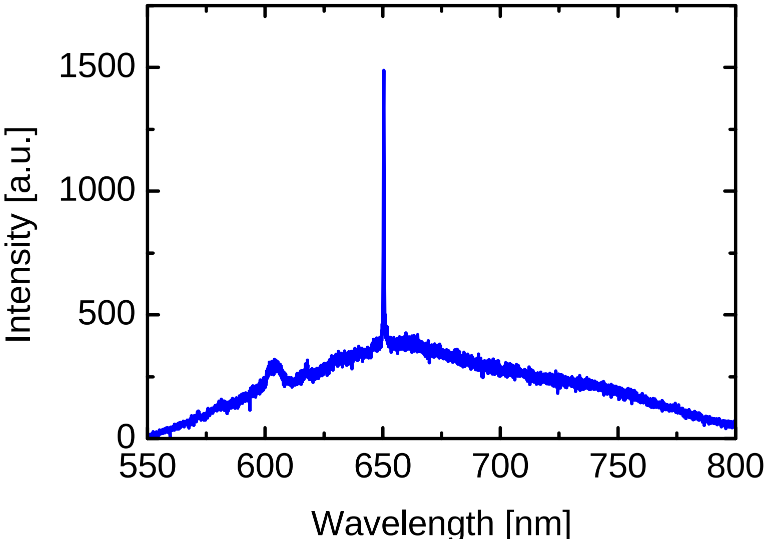
9.4.2. Photonic Crystal Cavities

The following shows an approach to position single defect centers on a photonic crystal structure (SiN membrane):

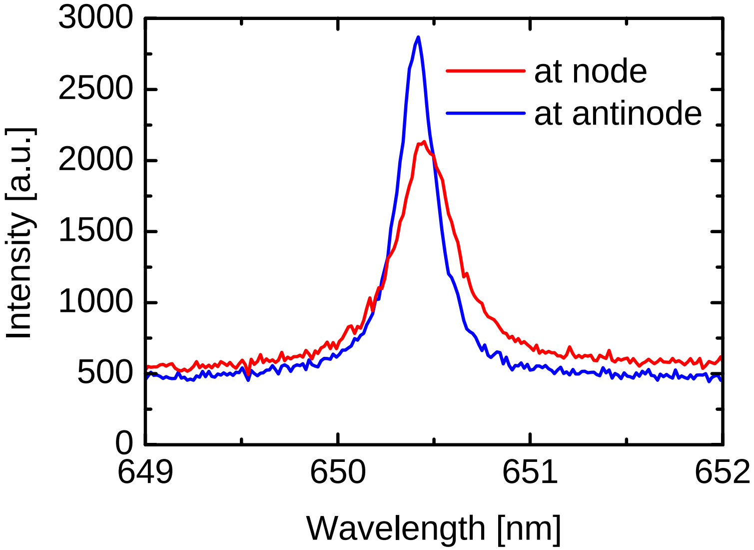


M. Barth, et al., Opt. Lett. 34, 1108 (2009)

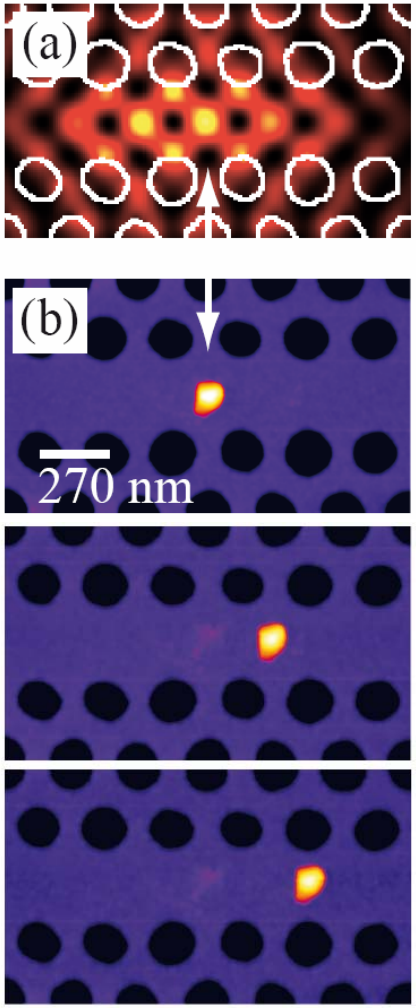
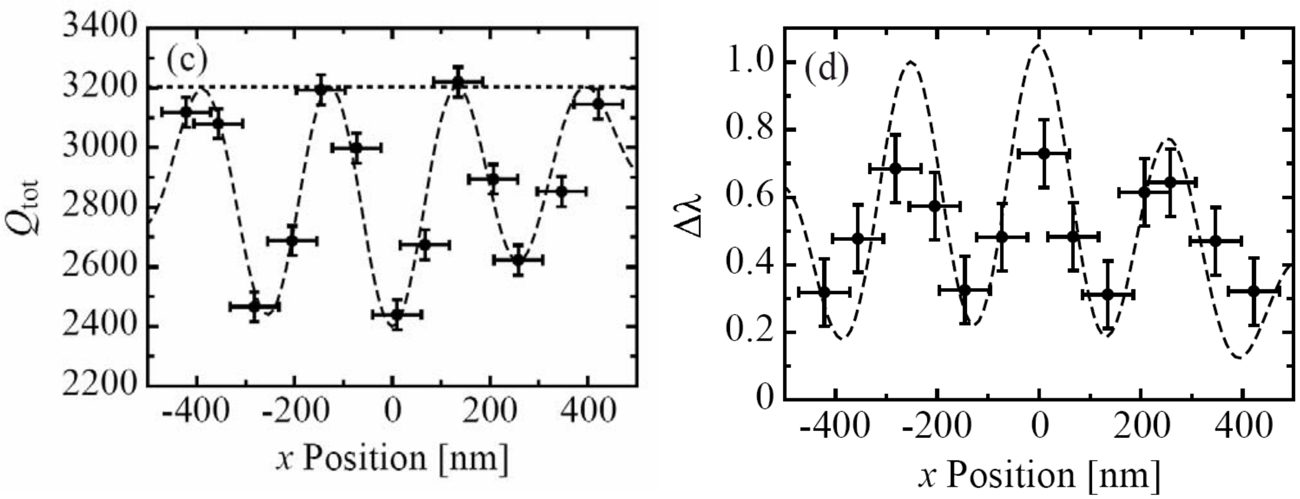
fluorescence



line shift & Q degradation

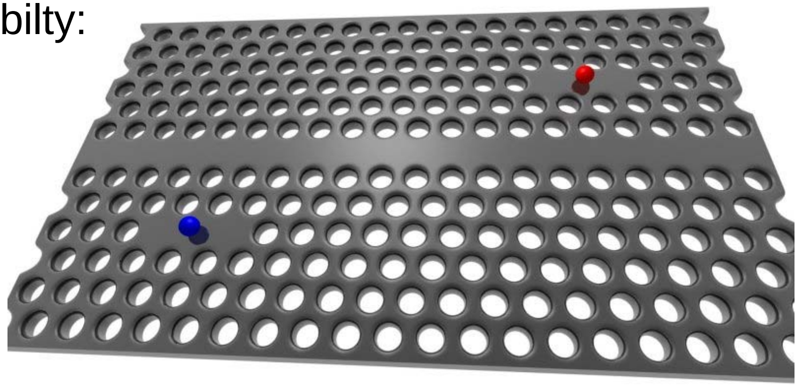


A degradation of the Q factor as well as a line shift is detected.

M. Barth, et al.,
Opt. Lett. 34, 1108 (2009)

Vision of scalabilty:



9.5. Applications of Solid State Cavity QED

Cavity QED allows to modify optical properties of light emitters in a unique way:

→ controlling the „natural“ lifetime and spatial emission pattern — weak coupling regime

→ changing the dynamics from incoherent to coherent — strong coupling regime

→ non-linear interaction at low intensities — strong coupling regime

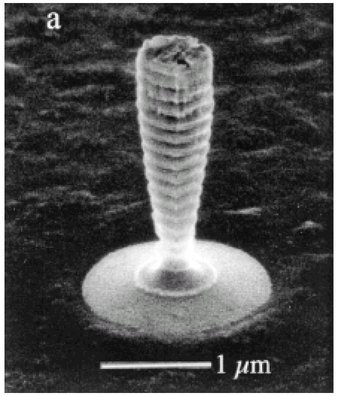
Possible applications are:

1. Realization of single mode single photon sources
2. Coherent interfaces between mater and light (stationary and flying qubits)
3. Ultra-low threshold lasers
4. Light standards based on quantum effects
5. All-optical (single photon) switches

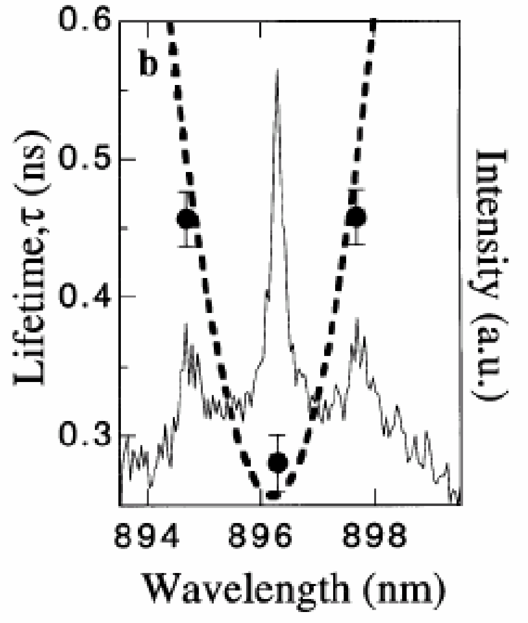
A few recent developments are discussed on the final slides.

9.5.1. Single Mode Single Photon Sources

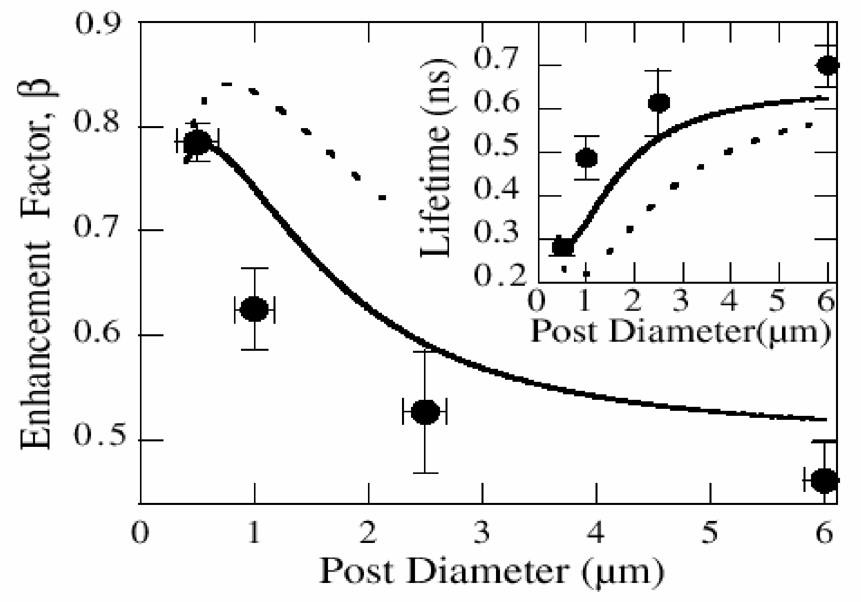
Solomon et al. [Phys. Rev. Lett. 86, 3903 (2001)] used a micropillar cavity to enhance spontaneous emission from a single quantum dot into a single cavity mode.



$$\beta\text{-factor} = \frac{\text{spont. emission rate into cavity mode}}{\text{total spont. emission rate}}$$

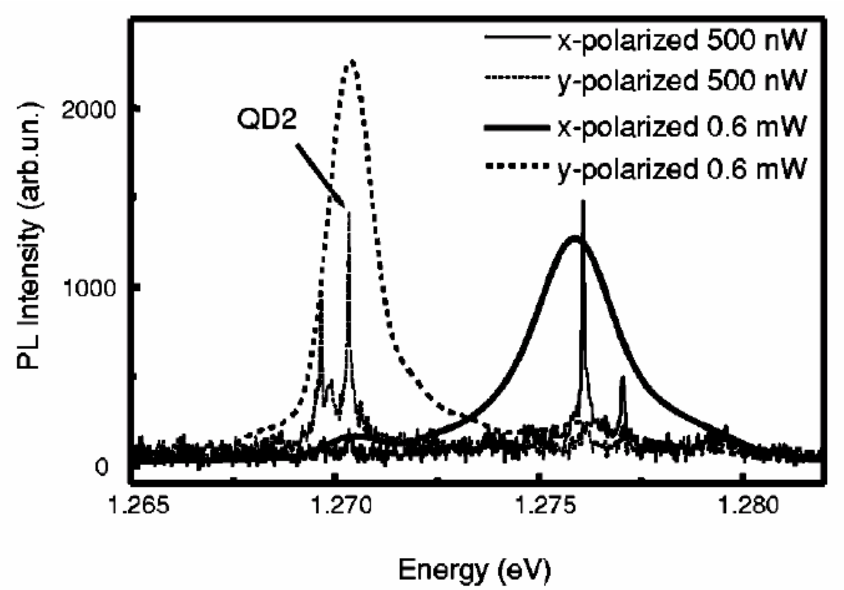


enhancement of emission rate
for dot in resonance with cavity

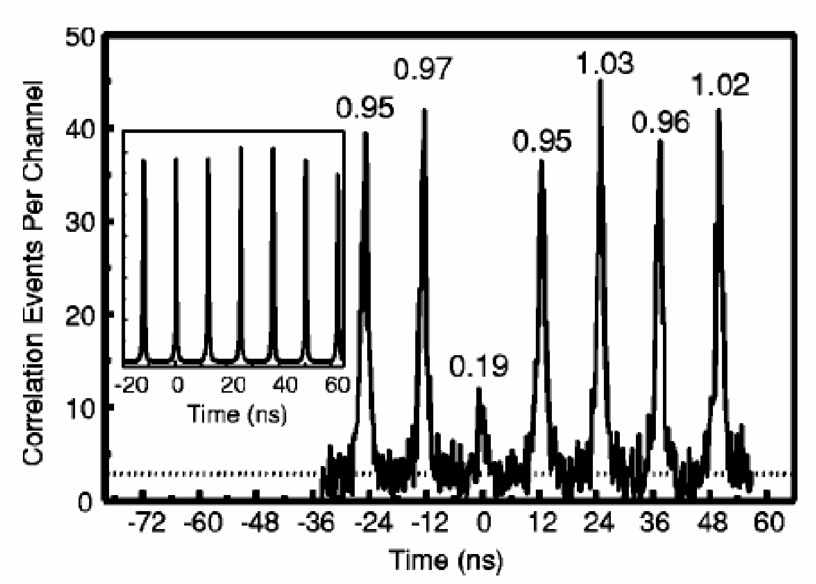


enhancement of β-factor with increasing
Purcell-factor

Moreau et al. [Appl. Phys. Lett. 79, 2865 (2001)] obtained nearly 70% of single photon emission into a single mode of an elliptical (to select one linearly polarized mode) pillar microcavity.



emission of a single quantum dot into a single mode of an elliptical cavity



intensity correlation measurement ($g^{(2)}(t)$) under pulsed excitation of a single quantum dot

9.5.2. Ultra-low Threshold Lasers

The ultimate limit of a solid state laser is a laser with only a single quantum dot as the active material.

Estimation of Threshold:

The average spontaneous emission rate of a quantum dot is N_A/τ_{sp}, where N_A is the average probability over time that the QD contains an exciton.

Of all emitted photons, a fraction β will be captured by the optical cavity.

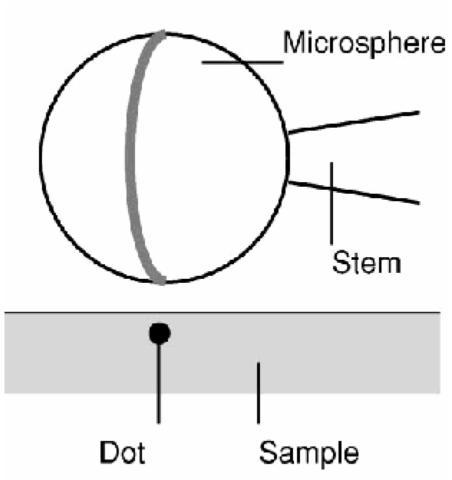
Captured photons will remain in the cavity for an average storage time $\tau_{ph} = Q/\omega$ before leaking out.

Combining all the above factors, we obtain:

$$\boxed{n_{sp} \approx \frac{\beta \tau_{ph} N_A}{\tau_{sp}} \geq 1}$$

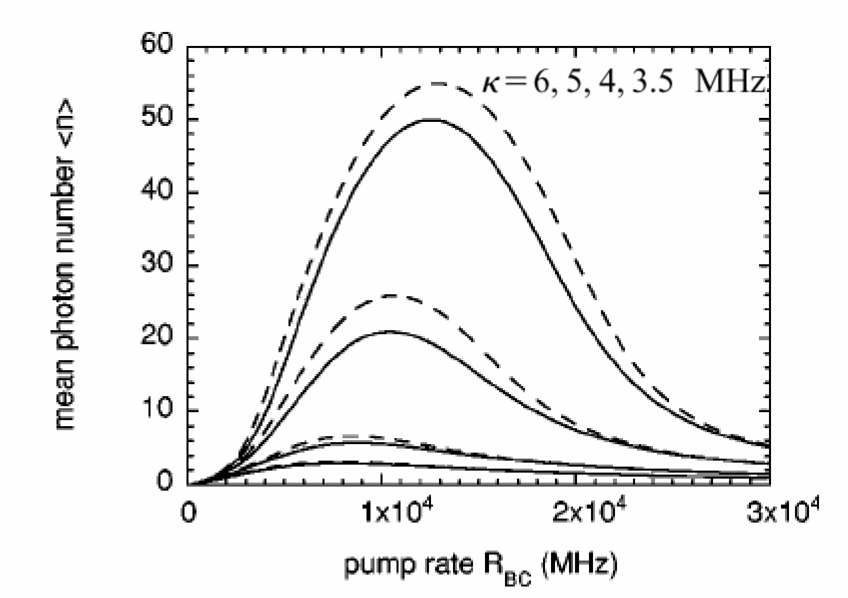
This condition means that the cavity *Q* must be high in order to reach threshold.

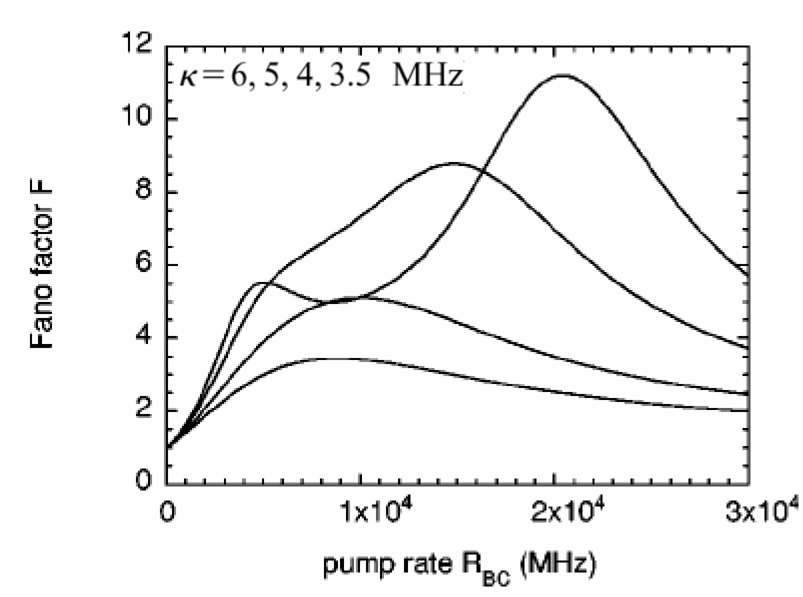
The highest Q-factors are provided by Whispering-Gallery mode cavities.
Microspheres can provide *Q >*10^9



Proposal: Coupling of a single quantum dot to a microsphere
[Pelton & Yamamoto, Phys. Rev. A 59, 2418 (1999)]

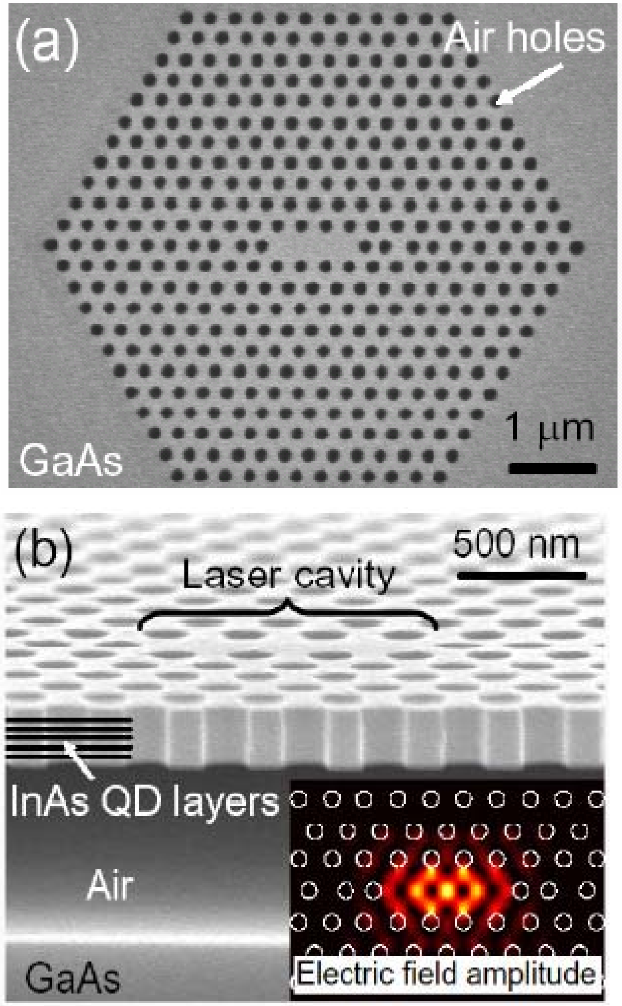
Single quantum dot lasers have unusual properties, such as self-quenching and sub-Poissonian fluctuations.
[Benson & Yamamoto, Phys. Rev. A 59, 4756 (1999)]





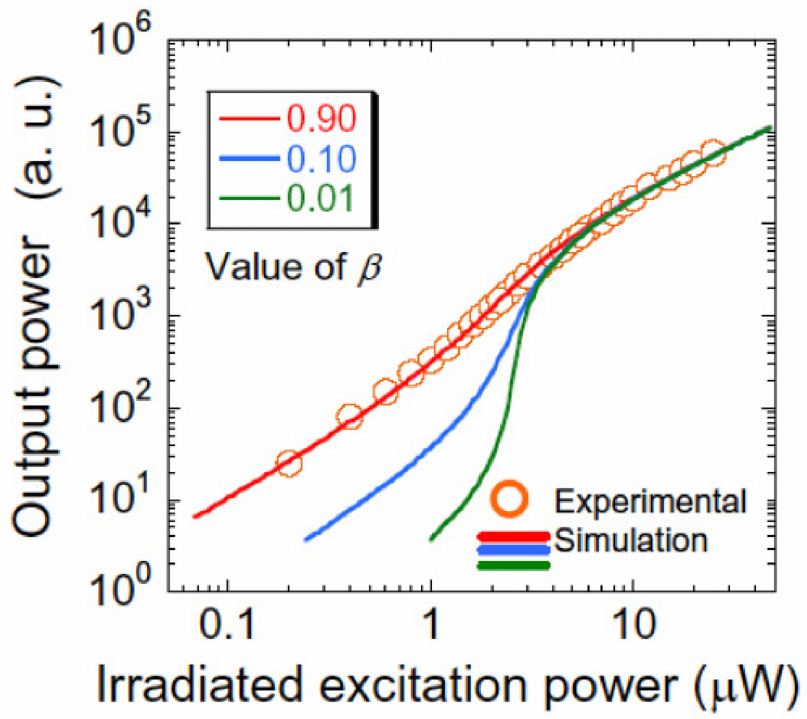
At a very large β the characteristic threshold of a laser in the input/output power curve vanishes.

There is a rather continuous transition from spontaneous to stimulated emission.



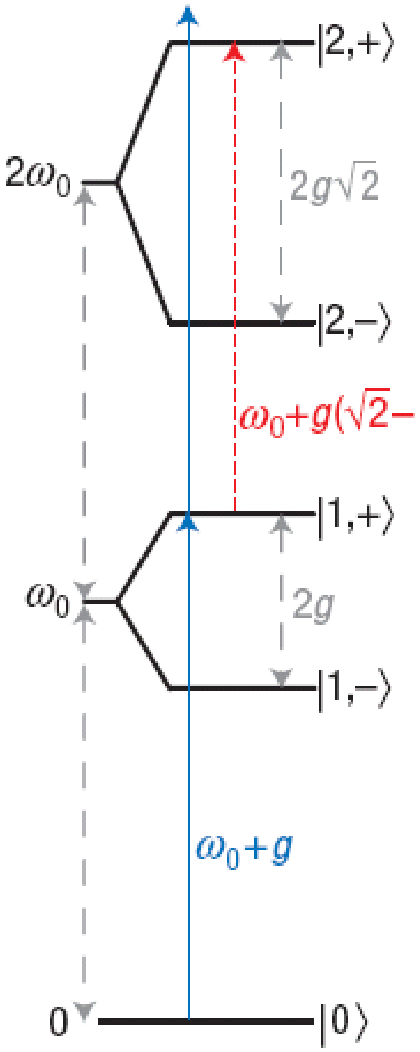
Left: design of an ultra-low thresold laser

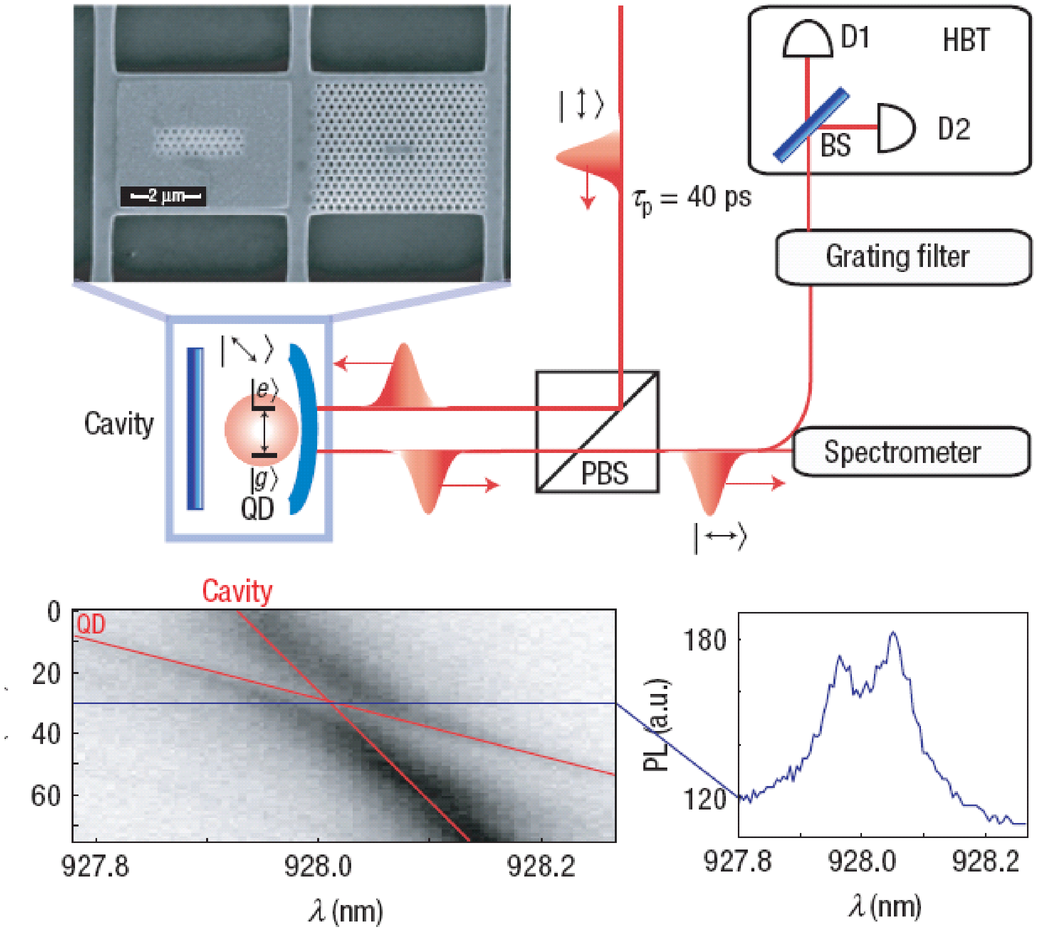
Bottom: measured input/output characteristics
[Nomura et al., SPIE 10.1117/2.1200711.0915]



9.5.3. Non-linearites with Ultra-low Intensities

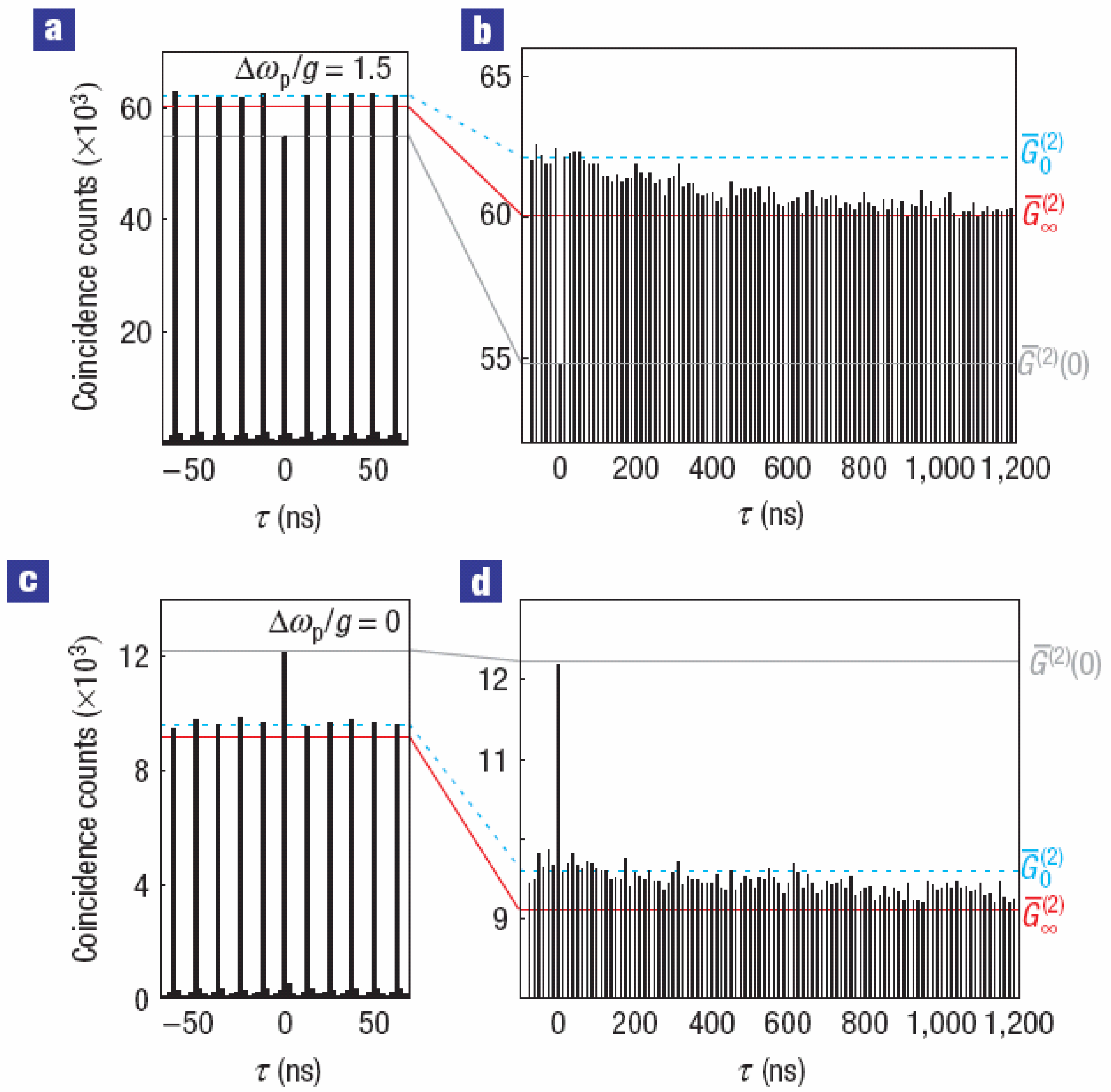
Photon blockade in a solid state system [Faraon et al., Nature Physics 4, 850 (2008)]:





Left: Schematic of Coulomb blockade in the strong coupling regime
Right: Setup of a solid state system consisting of a single quantum dot in a PC cavity

Experimental results of photon blockade and photon-induced tunneling [Faraon et al., Nature Physics 4, 850 (2008)]:



Summary of part 9

- Cavity QED effects can be observed in solid state systems using quantum dots and microcavities.

- Different cavities (pillar, Whispering-Gallery mode, photonic crystal cavities) have been developed to investigate the weak and the strong coupling regime.

- Solid state cavity QED has been applied to improve optical properties of single photon sources

- Various applications in classical optics (ultra-low threshold or thresholdless lasers) or quantum optics (interfaces between flying and stationary qubits, quantum gates) are expected in the near future.