QDs and optical microcavities: tailoring spontaneous emission

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Spontaneous emission control: why? (as of ~1982)

Only a tiny fraction of spontaneous emission is useful! 
\[ \beta \sim 10^{-5} \]

100000 times decrease of \( I_s \) expected for \( \beta \sim 1 \)!
Two strategies toward improved SE control

Optical microcavity = photon confinement on the wavelength scale discrete photon modes

If the emitter is coupled to a single mode, $\beta=1$
β=1 « Thresholdless »-laser

Ideal conversion of electrical signals into optical signals
- 1 e- -> 1 photon into the (single) mode
- Linear input/output characteristic curve
- No additional noise
- High cut-off frequency

Proposal:
Kobayashi et al, 1982

Yokoyama, Science 256, 62 (1992)

Björk et al, IEEE-JQE 27, 2386 (91)
CQED with QDs: historical background

CQED (80->...)
SE can actually be tailored to a large extent for atoms in electromagnetic cavities

optical microcavities (90->...)
it is possible to confine optical photons on the wavelength scale in 2D, 1D, 0D

self-assembled QDs (~94->...)
single QD optical spectroscopy reveals atom-like behavior

CQED with QDs (96->...)

Equipe nanophysique et semiconducteurs
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LABORATOIRE DE PHOTONIQUE ET DE NANOSTRUCTURES
Outline

1) Some basic microcavity effects
2) QDs as probes of photonic microstructures
3) CQED effects on QDs
4) Some application prospects:
   single-mode photon sources
   microlasers

For a recent review see:

*Solid state CQED with self-assembled QDs, in*
Topics in Applied Physics 30, 269 (2003), P. Michler ed. Springer
SE angular redistribution in planar microcavities

A. Kastler, Appl. Optics 1,17 (1962)

Application to light emitting diodes:
- directional emission + high efficiency $\eta \sim 25\%$
- $\eta \sim 2\%$

(Blondelle et al, Electron. Lett 31, 1286, 1995)
Tailoring the spontaneous emission rate

Fermi golden rule: exponential decay
\[ \frac{1}{\tau} \propto \text{density of modes per unit volume} \]

One can tailor spontaneous emission dynamics

- Dimensionality, refractive index...
- Coupling to a single mode (0D cavity, or optical selection rule)

E.g.: strong coupling for QWs in planar cavities (Weisbuch et al, PRL 69, 3314, 1992)
Emitter in a planar microcavity

Ex: Brorson et al, IEEE JQE 26, 1492 (1990)

ideal metallic mirrors

Both SE rate enhancement and inhibition expected

Thickness of the cavity layer $nL/\lambda$
InAs QD array in a planar microcavity

Very weak effect on QD spontaneous emission dynamics!

*Solomon et al, PRL 86, 3903 (2001)*

This is expected since the effective cavity length is around $4 \lambda/n$
Strong and weak coupling regimes in 0D cavities

Perfect cavity
=> reversible spontaneous emission

\[ h\Omega = \left| \mathbf{d} \cdot \mathbf{E}(r) \right| \]

Emitter dipole field per photon

Energy

\[ |g, 1\rangle \rightarrow |e, 0\rangle \]

=> Entangled eigenstates
Strong coupling for a single cesium atom in an optical microcavity

Strong coupling first observed for atoms in the microwave spectral range
(supercconductors are IDEAL mirrors!)
More recently observed in the optical spectral range

Hood et al, PRL 98
Strong and weak coupling regimes in 0D cavities

Ω >> 1/τ_{decoherence} \Rightarrow \text{damped Rabi oscillation}

Ω < 1/τ_{decoherence} \Rightarrow \text{exponential decay « weak coupling regime »}

\[ \tau \sim f(Q, V, \ldots) \]
Two important figures of merit

Ability to confine the e.m. field spatially:

\[ V_{\text{eff}} : \quad \varepsilon_{\text{max}} = \sqrt{\frac{\hbar \omega}{2 \varepsilon_0 n^2 V_{\text{eff}}}} \]

Actual cavity

Equivalent cavity volume \( V_{\text{eff}} \)

Ability to store the e.m. energy:

\[ Q : \quad Q = \frac{\omega}{\Delta \omega_{\text{mode}}} \quad \leftrightarrow \quad \tau_{\text{photon}} = \frac{1}{\Delta \omega_{\text{mode}}} \]

Around 1 eV, \( Q=1000 \) \( \leftrightarrow \) \( \tau_{\text{photon}} = 0.6 \) ps
The « Purcell effect »

Decoherence mostly due to photon escape (low Q) => \( \Delta \omega_{\text{mode}} \gg \Delta \omega_{\text{em}} \)

\[ \text{weak coupling regime} \quad \frac{1}{\tau} \propto \rho(\omega) \left| \langle \vec{d}, \vec{E} \rangle f \rangle \right|^2 \]

Purcell (1946)

\[ \frac{\tau_{\text{free}}}{\tau_{\text{cav}}} = \frac{3}{4\pi^2} \frac{Q(\lambda/n)^3}{V} \]

\[ F_p = \text{magnitude for an ideal emitter} \]

- quasi-monochromatic
- spatial matching
- spectral matching
Spontaneous emission rate for a « non-ideal » emitter

\[ \frac{\tau_{free}}{\tau_{cav}} = F_p \frac{\Delta \omega_{mode}^2}{4(\omega_{mode} - \omega_{em})^2 + \Delta \omega_{mode}^2} \frac{\left| E(\vec{r}) \right|^2}{E_{max}} \frac{\left| E(\vec{r}).\vec{d} \right|^2}{\left| E(\vec{r}) \right|^2 \left| \vec{d} \right|^2} \]

Spectral detuning \quad \text{spatial detuning} \quad \text{dipole misorientation}

If the emitter linewidth is not negligible, « natural » emitter line

\[ \rho(\omega_{em}) \rightarrow \int_\omega \rho(\omega)L(\omega)d\omega \]

for a broad emitter \( Q \rightarrow Q_{em} \) and the Purcell effect is washed out
How to confine light in all 3 dimensions with dielectrics?

metallic mirrors are too lossy at optical frequencies!

distributed Bragg reflection (DBR, photonic crystal)

total internal reflection (e.g. optical fibers)
Some 0D solid-state microcavities

\[ V = 6 \left( \frac{\lambda}{n} \right)^3 \]

\[ V < \left( \frac{\lambda}{n} \right)^3 \]

3D confined modes
but not perfect « photonic dots » (leaky modes)!

Q ?
Probing cavity modes with QDs

Systematic probing of cavity modes (no selection rule)

QD internal light source widely used to study microdisks or PC cavities see e.g. D. Labilloy et al, APL 73, 1314 (1998)
Probing cavity modes with one (few) QDs

Very strong homogeneous broadening of the single QD emission under high excitation conditions

=> broadband light source!
Probing cavity modes with QDs (2)

Resonance condition:

planar: \( L = \frac{\lambda}{n} \)

pillar : \( L = \frac{\lambda}{n_{\text{eff}}} \)

\[ \Delta E (\text{meV}) \]

\[ \text{Pillar Radius (\( \mu \text{m} \))} \]

Large diameter micropillars (d>4\(\lambda/n\))

Same profile for guided mode in GaAs and AlAs layers

\[\Rightarrow\text{No mode coupling at interfaces}\]
\[\Rightarrow\text{Fundamental pillar mode built using}\]

\[
\frac{n_{\text{eff}}(\text{GaAs})}{n_{\text{eff}}(\text{AlAs})} = \frac{n(\text{GaAs})}{n(\text{AlAs})}
\]

\[\Rightarrow\text{the structure is still a « } \lambda\text{-cavity » for the novel resonant wavelength } \lambda = L \cdot n_{\text{eff}}(\text{GaAs})\]

\[\Rightarrow\text{Cavity Q unchanged}\]
Probing cavity $Q$'s

Very weak absorption of the QD array => we probe the « empty » cavity $Q$
(as long as $Q<\sim 10000$)

![Graph showing $Q_{\text{planar}}$ vs. diameter (µm)]

High $Q$’s can be achieved ($Q=5000$, $\tau\sim 3$ps)

$Q$ drops at small sizes : intrinsic or extrinsic effect ?
Numerical study of the intrinsic Q of GaAs/AlAs micropillars

P. Lalanne et al, APL june 2004

Strong oscillations in the small diameter limit!
Small diameter micropillars ($d > 4\lambda/n$)

(Slightly) different mode profile in GaAs and AlAs layers

$\Rightarrow$ inter-mode coupling at interfaces

$\Rightarrow$ fundamental pillar mode built using

and
Blue shift of the DBR stopband as a result for the lateral confinement
Energy of the fundamental pillar mode

Resonant cavity mode = $\alpha + \beta$ with $|\alpha| >> |\beta|$

The component propagates freely through the DBRs

=> interferences and oscillations on $Q$ !
Numerical study of the intrinsic Q of GaAs/AlAs micropillars

P. Lalanne et al, APL June 2004

Behavior well understood in a two-mode transfer matrix model

\[
\begin{align*}
\sigma & = (\text{Red} + \text{Green})
\end{align*}
\]
High $Q$’s can be achieved ($Q=5000$, $\tau\sim3\text{ps}$)

$Q$ drops at small sizes: scattering by sidewall roughness

$F_p = 32$ achieved for $d=1\ \mu\text{m}$!
Some 0D solid-state microcavities

High Q, low volume, 3D confined modes
Some solid-state CQED experiments

performed with QDs
CQED : strong coupling for single QDs?

\[ |\text{ground, 1 photon}\rangle \quad (\hbar \Omega_{\text{rabi}}) \quad |\text{excited, 0 photon}\rangle \]

\[ \hbar \Omega = |\vec{d} \cdot \vec{E}_{\text{max}}| \]

InAs QD in solid-state cavity:
- large dipole (0.6 e.nm)
- huge vacuum field (10^5 V/cm)

Sizeable vacuum Rabi splitting expected: \( 2\hbar \Omega \sim 50 \mu\text{eV} \) for \( V \sim 10 (\lambda/n)^3 \)

VRS observable \( \Leftrightarrow 2\hbar \Omega > \frac{\Delta E_{\text{emitter}}}{\Delta E_{\text{cavity}}} \)

Here, negligible emitter linewidth
\( \Rightarrow \) the relevant criteria for strong coupling is:

\[ 4\hbar \Omega > \Delta E_{\text{cavity}} \]

Andreani et al, PRB 60, 13276 (1999)
Strong coupling for single QDs?  Gérard et al, Physica E9, 131 (2001)

For a single InAs QD, strong coupling already observable with microdisks and PC cavities
Strong coupling for single QDs?

QDs with giant oscillator strength are very attractive!
- strong coupling achievable also with micropillars
- weakly damped Rabi oscillation for QD in PC cavities

Gérard et al, Physica E9, 131 (2001)
Andreani et al, PRB 60, 13276 (1999)
Fresh news!

The strong coupling regime has been observed by two research groups:

. micropillars + big InGaAs QDs displaying a « giant » oscillator strength (Forchel et al, Würzburg)

. InAs QD in high Q photonic crystal microcavity (Scherer et al, Caltech)

(still unpublished)
Purcell effect on InAs QDs

1946: Purcell’s paper
1983: Observation for atoms in cavities (x 500! Haroche et al, ENS Paris)

1997: First observation of the Purcell effect in a solid-state microcavity!

QD array in micropillar x 5 J.M. Gérard et al, PRL 81, 1110 (1998)
QD array in microdisk x 18 B. Gayral et al, Physica E7, 641 (2000)
      x 12 B. Gayral et al, APL 78, 2828 (2001)

single QD in microdisk x 6 A. Kiraz et al, APL 78, 3932 (2001)
single QD in micropillar > x 3 E. Moreau et al, APL 79, 2865 (2001)
          x 4 G. Solomon et al, PRL 86, 3903 (2001)
          x 5 J. Vuckovic et al, APL 82, 3596 (2003)
Purcell effect for a collection of QDs ($d=1 \ \mu m$, $F_p=32$)

PL decay faster only for on-resonance QDs: intrinsic effect
SE rate enhancement (x5) much smaller than $F_p$

$PRL\ 81,\ 1110\ (1998)$
How to understand the magnitude of the Purcell effect

Random distribution of QDs
=> spatial + spectral averaging
$F_p$ : the relevant figure of merit

Enhancement factor depends as expected on $F_p$, not $d$ or $Q$

Good agreement with theory

*PRL 81, 1110 (1998)*
Dependence of SE rate enhancement on the spectral detuning

from
Solomon et al, PRL 86, 3903 (2001)

see also
Graham et al, APL 74, 3408 (1999)

Experiment in good agreement with the expected spectral dependence
SE inhibition in metal-coated micropillars

No inhibition in standard micropillars, due to leaky modes => gold-coated pillars

Efficient reduction of the density of non-resonant modes by the metallic coating
=> strong SE inhibition (/10)
for out-of-resonance QDs

M. Bayer et al,

PRL 86, 3168 (2001)
Purcell effect for QDs in microdisks

Up to $\times 12$ SE rate enhancement

B. Gayral et al
APL 78, 2828 (2001)
Purcell effect in microdisks

B. Gayral et al, APL 78, 2828 (2001)

Intensity PL (lin. scale)

Time (ps)

models:
- $F_p$ + spectral/spatial averaging
- + "jitter" due to relaxation time (30 ps)
Purcell effect in a c.w. experiment

Saturation due to filling of fundamental QD states
Purcell effect ($\times 18$ here!) $\Rightarrow$ saturation occurs for higher pumping power

Same behavior observed for QDs in PC cavities (Happ et al, PRB 2002)

B. Gayral et al
Physica E7, 641 (2001)
Few QDs in a micropillar
Purcell effect on isolated QDs in a micropillar
($Q_{2D}$=1000, $Q$=500, $F_p$=7)

Moreau et al,
APL 79, 2865 (2001)
Beware!

Unlike experiments on large collections of QDs, quantitative modelling is not possible since the QD location (and $\tau_{\text{free}}$) are not precisely known!

(see e.g. Gayral et al, PRL 90, 229701, 2003)

Coupling the QD in/out of resonance is a good way to highlight the Purcell effect and to estimate its magnitude
Purcell effect for a single QD in a microdisk

Kiraz et al, APL 78, 3932 (2001)
UCSB

\[ T = 4K, 1.75 \text{ns} \]
\[ T = 48K, 1.75 \text{ns} \]
\[ T = 31K, 850 \text{psec (resonance)} \]

\( F > 2 \)
Applications of the Purcell effect
(Nearly) single-mode spontaneous emission

Purcell effect =
selective enhancement of SE into the resonant mode

\[ \frac{\gamma}{\tau_{\text{free}}} \approx 1 \]

\[ \beta = \frac{F}{F + \gamma} \approx 1 \]

J.M. Gérard et al
PRL 81, 1110 (1998)

Very interesting for single photon sources and microlasers!
What is a single photon source?

Source able to emit single photons pulses on demand

N.B. : Non-classical state of light
⇒Impossible to generate it using a thermal source or a laser

Applications : quantum cryptography, metrology...
A single-mode single-photon source

Isolated InAs QD

GaAs/AlAs micropillar

20 nm

⇒ Single-photon emission

⇒ Efficient collection + single-mode behavior

proposal: J.M. Gérard et B. Gayral,
J Lightwave Technol. 17, 2089 (1999)
exp: E. Moreau et al, APL 79, 2865 (2001)
Some properties of this single photon source

Single photon collection efficiency

44\% \quad \text{Moreau et al, Physica E 13, 418 (2002)}
38\% \quad \text{Pelton et al, PRL 89, 233602 (2002)}

All photons prepared in the same mode (spatial+ polarization)

\sim \text{No two-photon pulses}

Key device for the practical implementation of quantum cryptography
SPS efficiency for GaAs/AlAs micropillars

scattering by sidewall roughness

\[ Q_{\text{scat}} \]

\[ \varepsilon < \beta \]

A careful optimisation of the intrinsic losses is necessary!

\[ \frac{1}{Q} = \frac{1}{Q_{\text{int}}} + \frac{1}{Q_{\text{scat}}} \]

\[ \varepsilon = \beta \frac{Q}{Q_{\text{int}}} \]

J.M. Gérard et al, quant-ph/0207115
Polarization control in single-mode micropillars

Elliptical cross section

1.4 µm

0.7 µm

Non degenerate fundamental mode

Gayral et al, APL 72, 1421, 1998

~ 90% linear polarisation degree of single QD emission

Moreau et al, APL 79, 2865, 2001

x-polarized

y-polarized

PL Intensity (lin.)

Energy (eV)
Indistinguishable single photons and the issue of decoherence

Indistinguishable single photons $\iff T_2 = 2 \, T_1$
InAs QD SPS: photon coalescence experiment


InAs QD in bulk GaAs: $T_2 \sim 600\text{ps} \ll 2\ T_1 \sim 2\ \text{ns}$

Here, emission of nearly-Fourier-transform limited single photons by the InAs QD, thanks to the Purcell effect
Toward « nanolasers »

$\beta \sim 10^{-5}$
$I_{th} = 5 \text{ mA}$

$\beta \sim 0.04$
$I_{th} = 36 \mu\text{A}$

*Huffaker et al,*
*APL 71, 1449 (1997)*

$\beta \sim 0.1$
$I_{th} = 40 \mu\text{A}$

*Fujita et al,*
*Electron. Lett. 36, 790 (2000)*

Sub $\mu\text{A}$ threshold current expected in the $\beta \sim 1$ limit!
The smallest PC cavities exhibit V’s as low as 0.2 \((\lambda/n)^3\)

Best QD arrays ~ 15 meV linewidth at 300K

Assuming \(Q_{\text{cav}} > Q_{\text{QDs}}\), the SE rate enhancement factor is:

\[
\frac{\tau_{\text{free}}}{\tau_{\text{cav}}} = \frac{3}{4\pi^2} \frac{Q_{\text{QDs}} (\lambda/n)^3}{V} \sim 30
\]

and \(\beta > 0.95\)!

Probably the best strategy to date for approaching the regime of « thresholdless » lasing at 300K
Conclusion

Using QDs has been the key for observing CQED effects

Solid-state CQED is a very lively domain

Still many basic effects to be observed!

Important potential applications in optoelectronics and quantum information science
What is beyond?

- moderate $\beta$, huge $Q$
  - 200 $\mu$W at 300K for InAs QDs
  - (ENS/LKB2003)

- moderate $Q$, high $\beta$
  - using the Purcell effect
  - ex: QDs in 2D PCs

$\beta$~1 in 3D PCs
Single quantum dot laser?

Low temperature required !!!

\[
\frac{\beta}{\tau} > \gamma \quad \Rightarrow \quad \Delta \omega_{em} > \Delta \omega_{cav}
\]

\(\Delta \omega_{em} - \omega_{em} < \hbar \Omega\)  
Strong coupling

\(\Delta \omega_{cav} < \hbar \Omega < \Delta \omega_{em}\)
Weak coupling and lasing

\(I_{th} \approx 10\) pA predicted
Pelton et Yamamoto,
PRA 59, 2418 (1999)
Semiconductor microdisks

2 µm

GaAs disk
GaAlAs pedestal

3D photon confinement using waveguiding + total internal reflexion (whispering gallery modes)

\[ V \sim 5 (\lambda/n)^3 \]
Probing WGMs with QDs

Observation of high $Q$ WGMs
Great sensitivity of Q on sidewall roughness
$Q > 10000$ for wet-etched microdisks

QD internal light source also commonly used to probe PC cavities

see e.g. D. Labilloy et al, APL 73, 1314 (1998)
An ideal dielectric microcavity

E. Yablonovitch, PRL, 2238 (1991)

photonic crystal + defect

$\beta = 1$

All photons are collected/prepared in a single mode!

Rem: spontaneous emission control = main initial motivation for developing photonic crystals
3D PCs and SPSs

To be done !!


Photonic defect +

SC or diamond nanoX, molecule