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WINTER COLLEGE ON OPTICS ON OPTICS AND PHOTONICS IN NANOSCIENCE AND NANOTECHNOLOGY

(7 - 18 February 2005)

"Optical Response of Semiconductor Quantum Dots"

presented by:

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Optical response of semiconductor quantum dots

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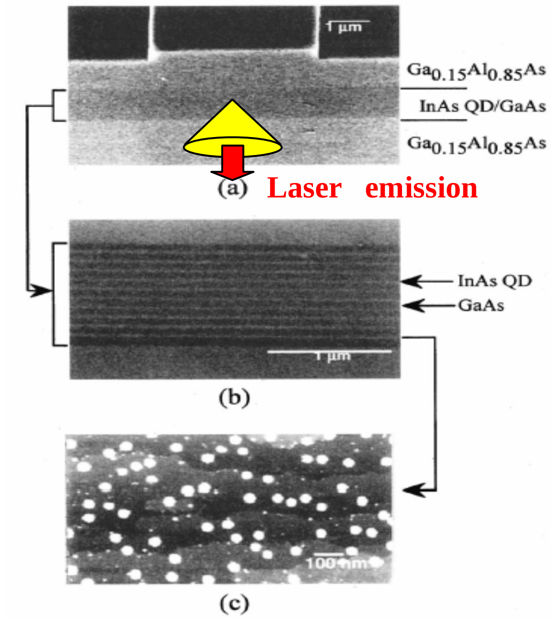
Outline

- fundamental optical properties
- single-photon emission
- spontaneous emission control

Doing optics with quantum dots



Nakamura
J. Appl. Phys. **94**, 1184 (2003)

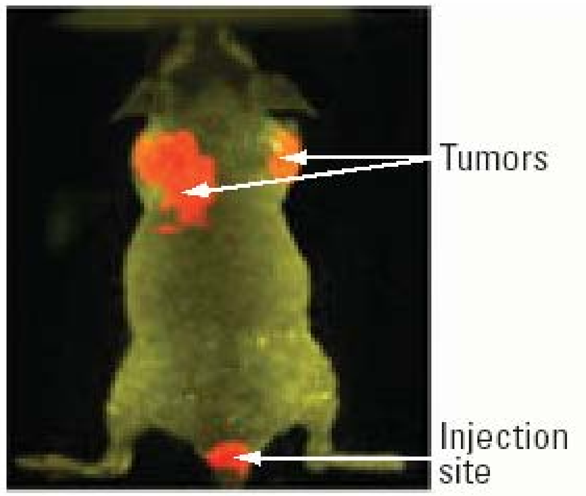
QD laser

Opto-electronics

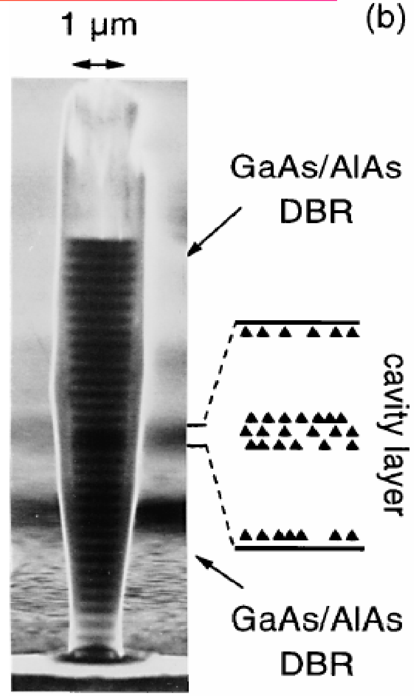
Bio-physics

Biological sensors

In vivo imaging of cancer cells in a living mouse



Gao, Nature Biot. **22**, 969 (2004)

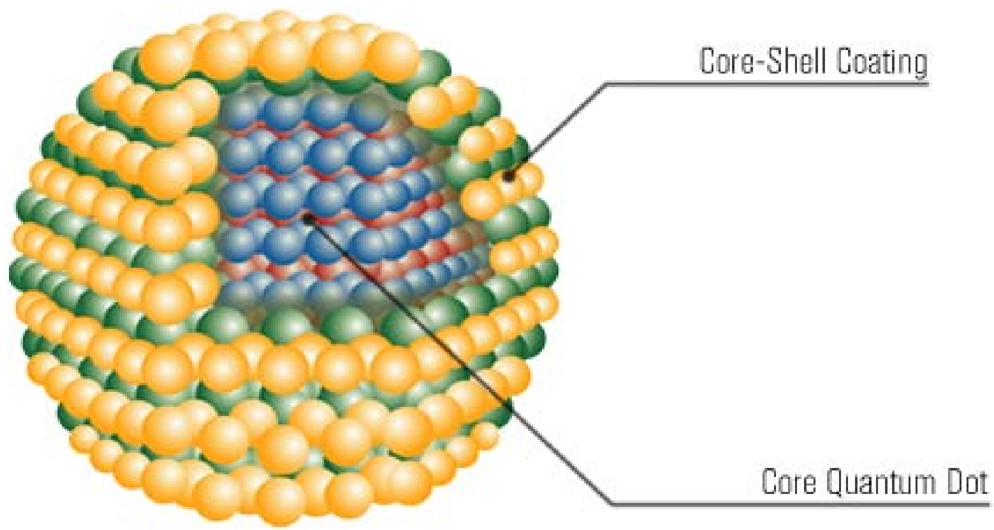


Gérard, PRL **81**, 1110 (1998)

Single photon source

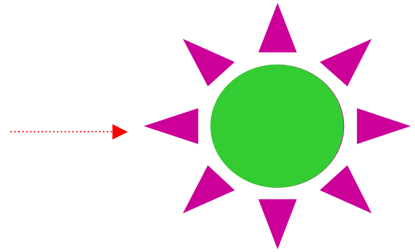
Quantum optics

Colloidal QD



- Chemically synthesized quantum dots in colloids (charged particles, inverse micelle)

surfactant molecule

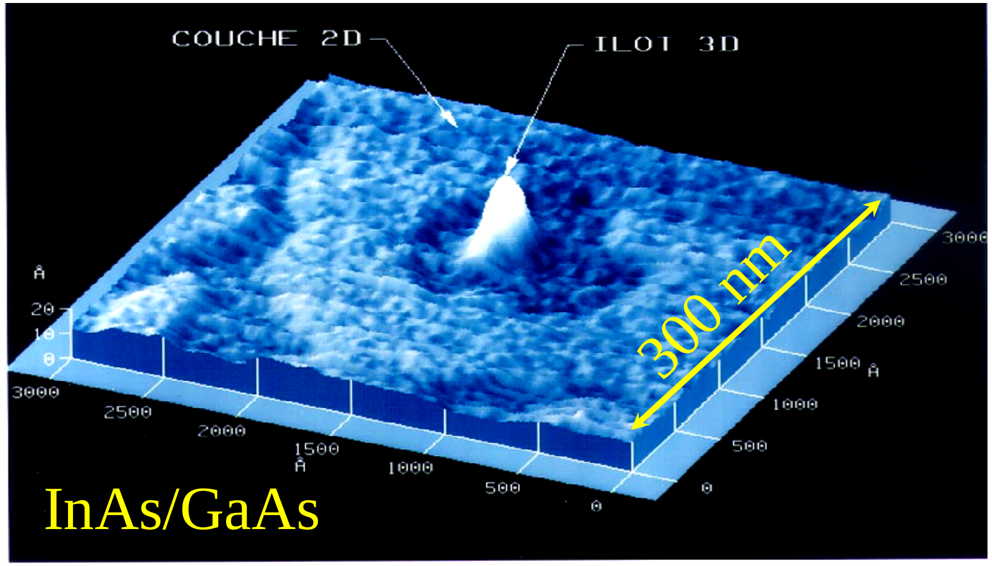


- Core material : CdS, CdSe, CdTe

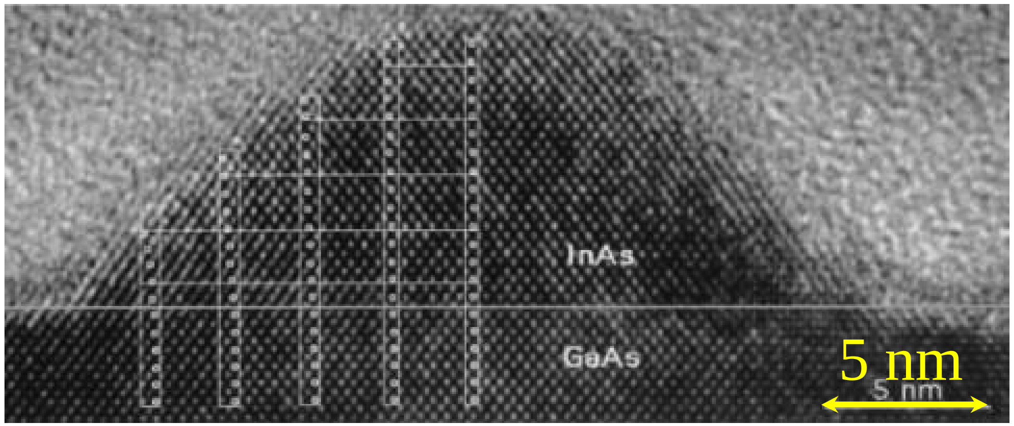
- Core-Shell structure
  - surface passivation
  - suppression of non-radiative decay channels

- Bio-conjugation of QD with adequate coating

Epitaxially grown QD



J. M. Gérard *et al.*, J. Cryst. Growth **150**, 351 (1995)



http://www.wsi.tum.de/E24/research/nanostructures/

- Molecular Beam Epitaxy

- Self-assembled quantum dots
  with areal density < 10^3 μm^{-2}

- Stranski-Krastanow growth mode
  - strain accumulation because of lattice mismatch
  - critical thickness of wetting layer ~1.7 monolayer for InAs
  - spontaneous formation of pyramids

- various systems : InAs/GaAs, InAs/InP, Ge/Si, GaN/AlN, CdTe/MnTe, CdSe/ZnSe …

Fundamental optical properties

Outline

- light-matter interaction
- interband and intraband transitions
- single QD spectroscopy
- specific solid-state features

Light-matter interaction

• Single particle Hamiltonian of an electron in a crystal reads

$$H_0 = \frac{\vec{p}^{\,2}}{2m} + V(\vec{r})$$

• Interaction of an electron with an electromagnetic field is obtained from generalized impulsion

$$\vec{p} = m\vec{v} - e\vec{A}$$

(Lagrangian mechanics)

• Single particle Hamiltonian becomes

$$\begin{aligned} H &= \frac{(\vec{p} + e\vec{A})^2}{2m} + V(\vec{r}) \\ &= \underbrace{\frac{\vec{p}^{\,2}}{2m} + V(\vec{r})}_{H_0} + \underbrace{\frac{e}{2m}(\vec{p}.\vec{A} + \vec{A}.\vec{p})}_{\text{dipole interaction}} + \underbrace{\frac{e\vec{A}^2}{2m}}_{\text{diamagnetic shift}} \end{aligned}$$

H_0 dipole interaction diamagnetic shift

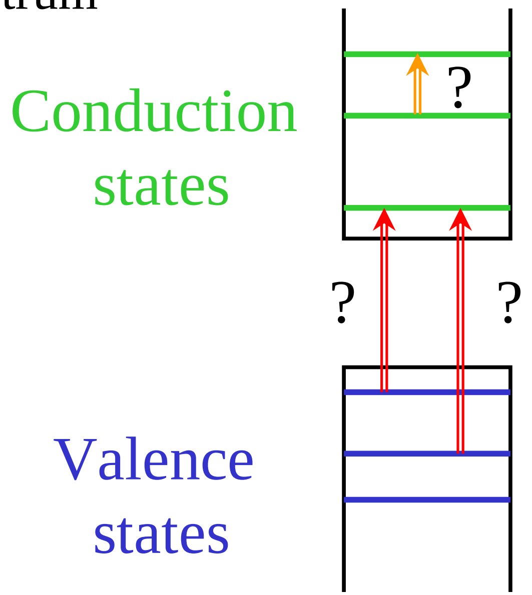
the second term simplifies because $\vec{p}.\vec{A} - \vec{A}.\vec{p} = \frac{\hbar}{i} div\,\vec{A} = 0$ (Coulomb gauge)

Dipole interaction

• Optical transitions between electronic states $|i\rangle \longrightarrow |f\rangle$ e induced by the dipole interaction term $H_I = \frac{e}{m}(\overrightarrow{A}.\overrightarrow{p})$

• Their strength depends on the matrix element $\langle f|H_I|i\rangle$

• In semiconductor QDs, the 3D confinement leads to a discrete electronic spectrum



One expects an absorption spectrum formed by a set of discrete lines

What are the selection rules ?

$$\langle f|H_I|i\rangle \neq 0$$

Envelope function approximation

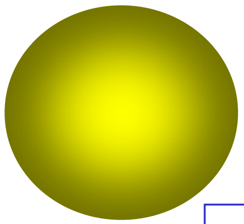
- In the envelope function approximation, electron wave-functions are given by

$$\psi_\alpha(\vec{r}) = \phi_\alpha(\vec{r})\, u_\alpha(\vec{r})$$

slowly varying envelope function

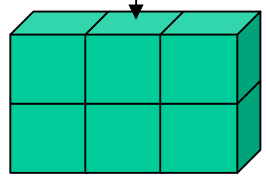
periodic rapidly varying atomic function

quantum dot



D ~ few nms

unit cell Ω_0



d ~ few Å

- Matrix element

$$\langle f|H_I|i\rangle \sim \frac{e}{m}\vec{A}.\langle f|\vec{p}|i\rangle$$ (Long wavelength approx.)

$$\sim \frac{e}{m}\vec{A}.\int d\vec{r}\,\phi_f^*(\vec{r})u_f^*(\vec{r})\,\vec{p}\,(\phi_i(\vec{r})u_i(\vec{r}))$$

$$\vec{p} = \frac{\hbar}{i}\vec{\nabla}$$

2 terms

$$\sim \frac{e}{m}\vec{A}.\int d\vec{r}\,\phi_f^*(\vec{r})u_f^*(\vec{r})\,u_i(\vec{r})\,\vec{p}\,(\phi_i(\vec{r}))$$

$$+ \frac{e}{m}\vec{A}.\int d\vec{r}\,\phi_f^*(\vec{r})u_f^*(\vec{r})\,\phi_i(\vec{r})\,\vec{p}\,(u_i(\vec{r}))$$

Intraband and interband transitions

$$\langle f|H_I|i\rangle \sim \frac{e}{m}\overrightarrow{A}.\int d\overrightarrow{r}\,\phi_f^*(\overrightarrow{r})\overrightarrow{p}(\phi_i(\overrightarrow{r})).\frac{1}{\Omega_0}\int_{\Omega_0} d\overrightarrow{r}\,u_f^*(\overrightarrow{r})u_i(\overrightarrow{r}) \quad \textbf{= (1)}$$

$$+ \frac{e}{m}\overrightarrow{A}.\int d\overrightarrow{r}\,\phi_f^*(\overrightarrow{r})\phi_i(\overrightarrow{r}).\frac{1}{\Omega_0}\int_{\Omega_0} d\overrightarrow{r}\,u_f^*(\overrightarrow{r})\overrightarrow{p}(u_i(\overrightarrow{r})) \quad \textbf{= (2)}$$

- These two terms can be rewritten as:

$$\langle f|H_I|i\rangle \sim \frac{e}{m}\overrightarrow{A}.\langle\phi_f|\overrightarrow{p}|\phi_i\rangle.\langle u_f|u_i\rangle \quad \textbf{= (1)}$$

$$+ \frac{e}{m}\overrightarrow{A}.\langle\phi_f|\phi_i\rangle.\langle u_f|\overrightarrow{p}|u_i\rangle \quad \textbf{= (2)}$$

- The first term **(1)** accounts for INTRABAND transitions

if $\quad u_f = u_i = u_c \quad \textbf{or} \quad u_f = u_i = u_v \quad \textbf{then} \quad \langle u_f|\overrightarrow{p}|u_i\rangle = 0$

- The second term **(2)** accounts for INTERBAND transitions

if $\quad u_f \neq u_i \quad \textbf{then} \quad \langle u_f|u_i\rangle = 0$

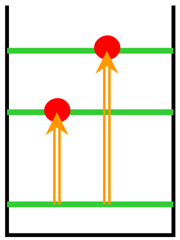
Intraband transitions

$$P_{|i\rangle\longrightarrow|f\rangle} = \frac{2\pi}{\hbar}\left|\frac{e}{m}\overrightarrow{A}.\langle\phi_f|\overrightarrow{p}|\phi_i\rangle\right|^2\delta(E_f - E_i - \hbar\omega)$$

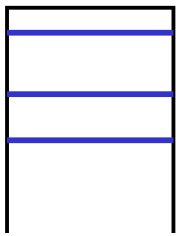
Polarization of incident electromagnetic field

Energy conservation

Conduction states



Valence states



Only electrons

or Only holes

- Absorption in { energy range of 10-100 meV
  wavelength range of 10-100 µm

Mid-Long IR

- Assumption of initial state occupied

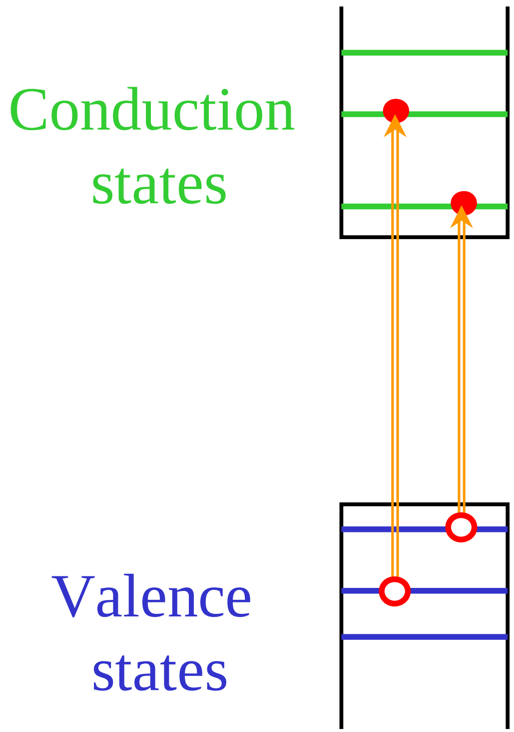
  and final state empty

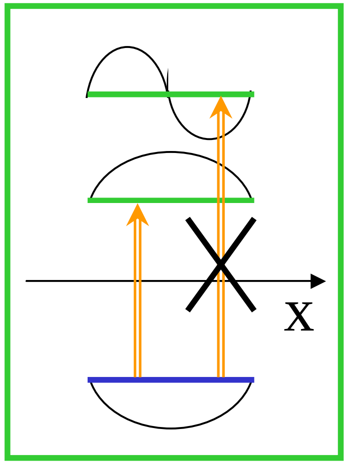
→ QDs must be doped

Interband transitions

$$P_{|i\rangle \longrightarrow |f\rangle} = \frac{2\pi}{\hbar} \left| \frac{e}{m} . \langle \phi_f | \phi_i \rangle . \overrightarrow{A} . \langle u_f | \overrightarrow{p} | u_i \rangle \right|^2 \delta(E_f - E_i - \hbar\omega)$$

Parity selection rule — Polarization selection rule — Energy conservation





Circularly polarized light propagating along (Oz), couples electronic states

$\Delta J_z = +/- 1$

In zinc-blende type semiconductors,

$$\begin{aligned} |electron\rangle &= |J = 1/2, J_z = \pm 1/2, ...\rangle \\ |heavy\,hole\rangle &= |J = 3/2, J_z = \pm 3/2, ...\rangle \\ |light\,hole\rangle &= |J = 3/2, J_z = \pm 1/2, ...\rangle \end{aligned}$$

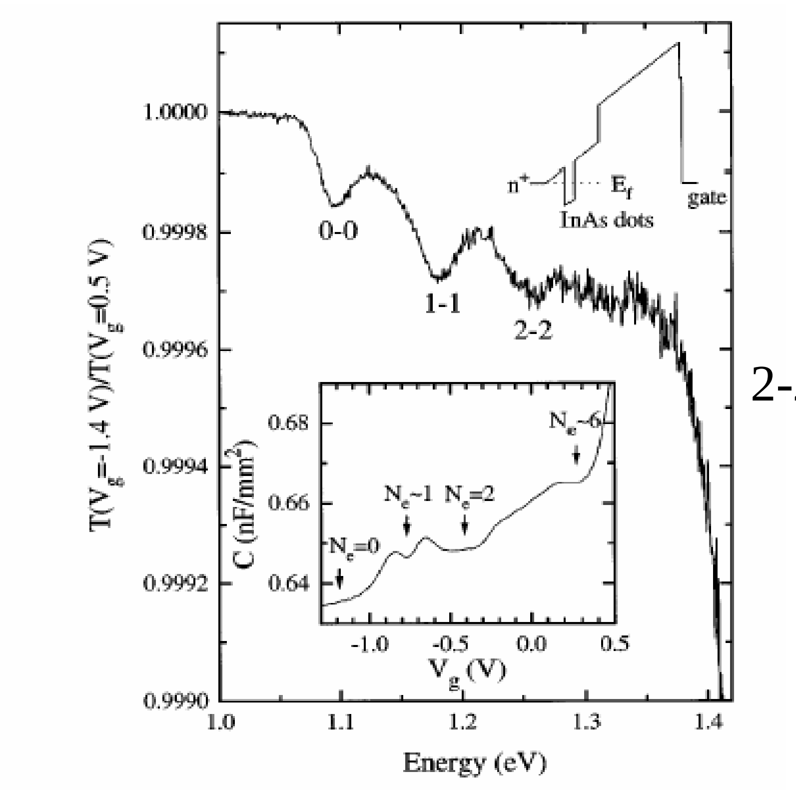
- Absorption in
  - energy range of 1- 2.5 eV
  - wavelength range of 0.4 -1 µm

Visible -NIR

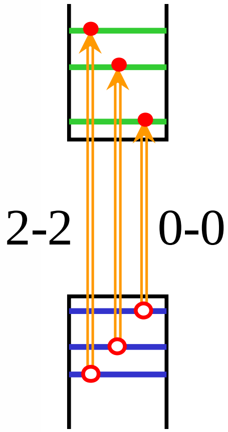
Electron AND hole

Interband absorption spectroscopy

Transmission experiment
in an ensemble of InAs QDs



Warburton, PRL **79**, 5282 (1997)



- Observation of three interband transitions (0-0, 1-1, and 2-2) in absorption spectrum

absorption spectrum
= set of discrete lines ??

- Large spectral width of the lines

$\Delta E \sim 50$ meV

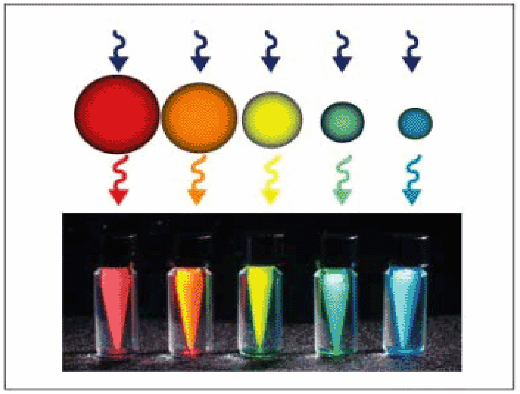
- Coupling to radiation leads to a natural linewidth of

$\Gamma \sim 1\ \mu$eV

→ size fluctuations

Inhomogeneous broadening

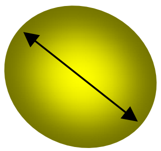
- Optical transition energy

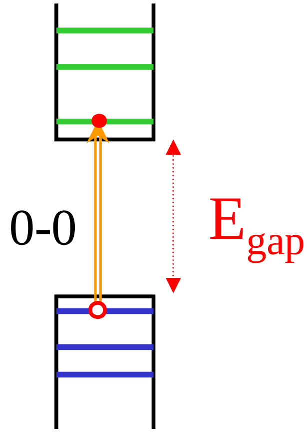


$$E_{n-n} \sim E_{gap} + E_{confinement}(n,n)$$

scales like $1/D^2$

quantum dot

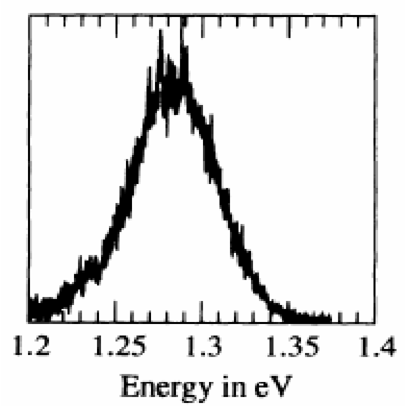




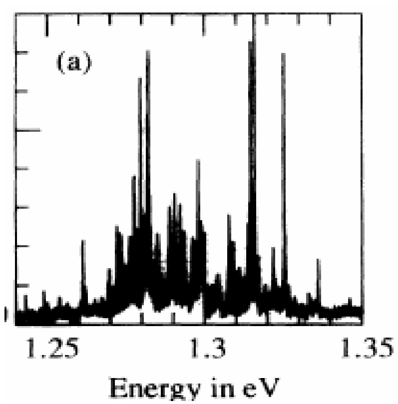
size fluctuations ⟹ inhomogeneous broadening

- From ensemble measurements to single QD spectroscopy

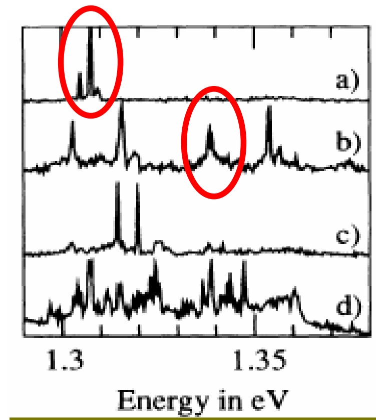
Marzin, PRL **73**, 716 (1994)



1000 QDs



10-100 QDs

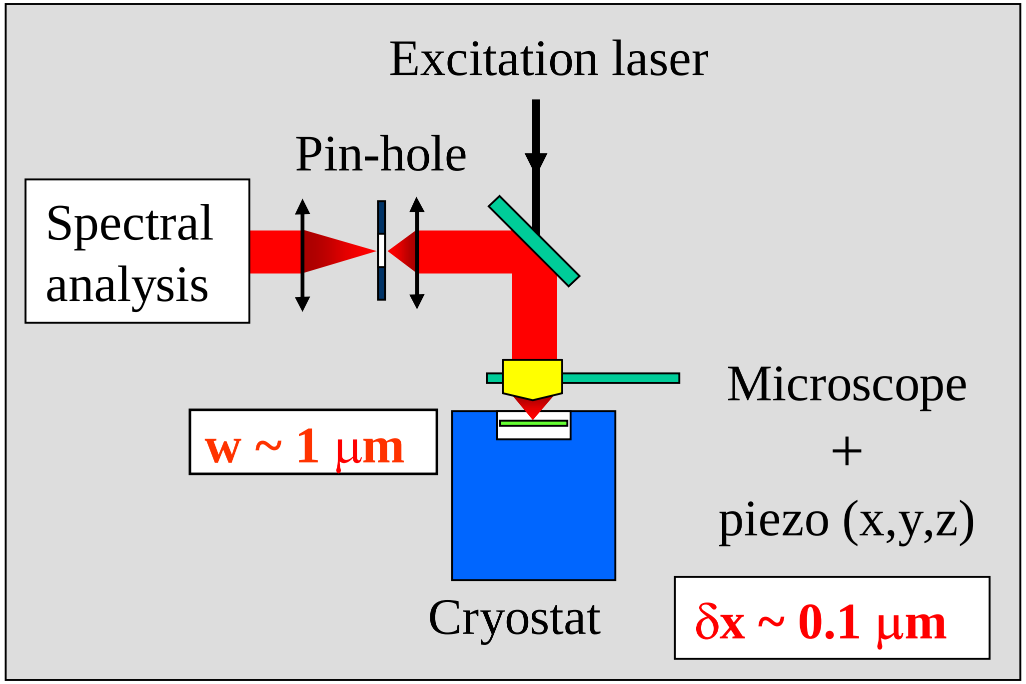


isolated QDs

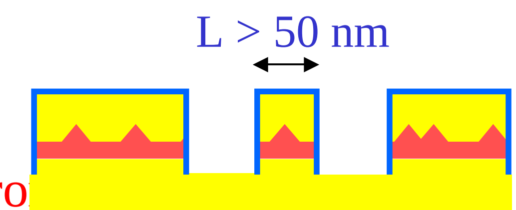
Single QD spectroscopy

- Near-field optical spectroscopy (see previous lectures)
- Far-field optical spectroscopy

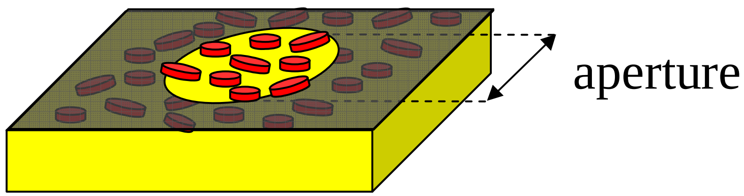
Microscope in confocal geometry ⟷ Sample preparation



- Dilute QD arrays
  - -dispersed solutions of colloidal QDs
  - -specific epitaxial growth modes
- Mesa structures

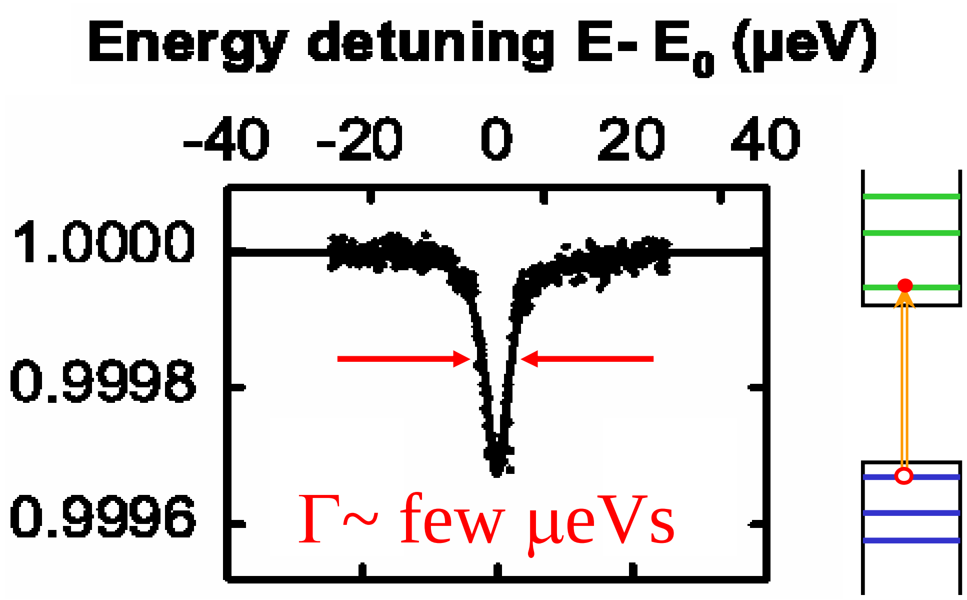


- Sub-micro



Absorption of a single QD

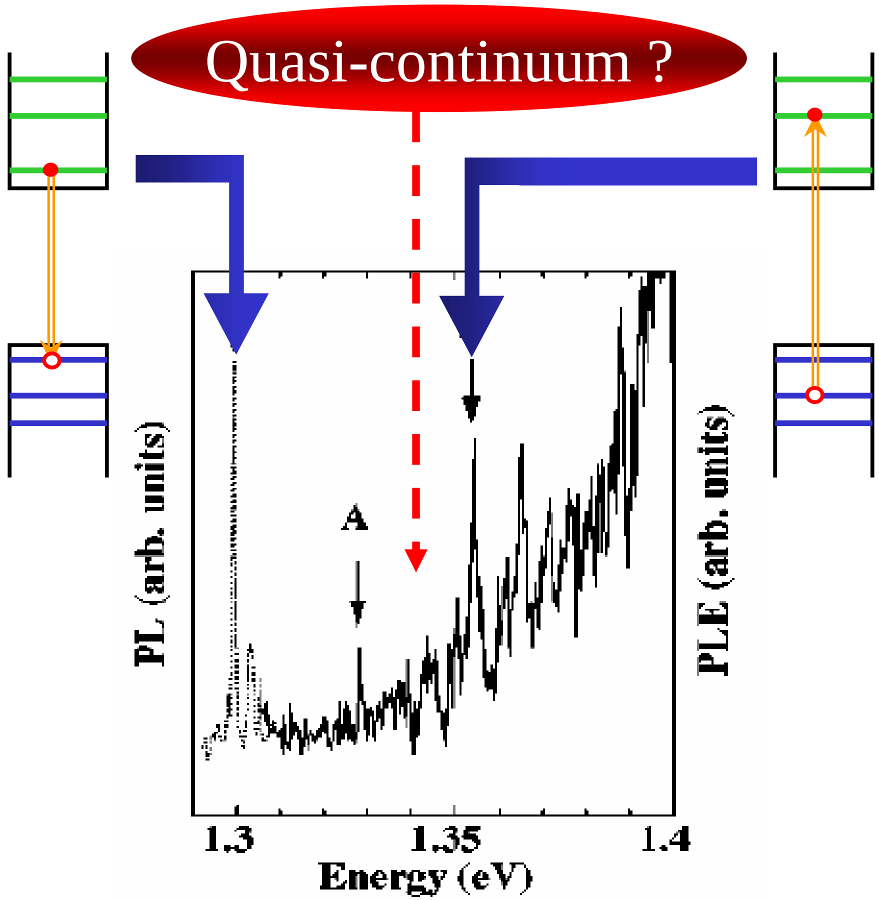
• Absorption spectroscopy



Högele, PRL **93**, 217401 (2004)

- Fundamental transition = sharp line
- Homogeneous linewidth ~ natural linewidth

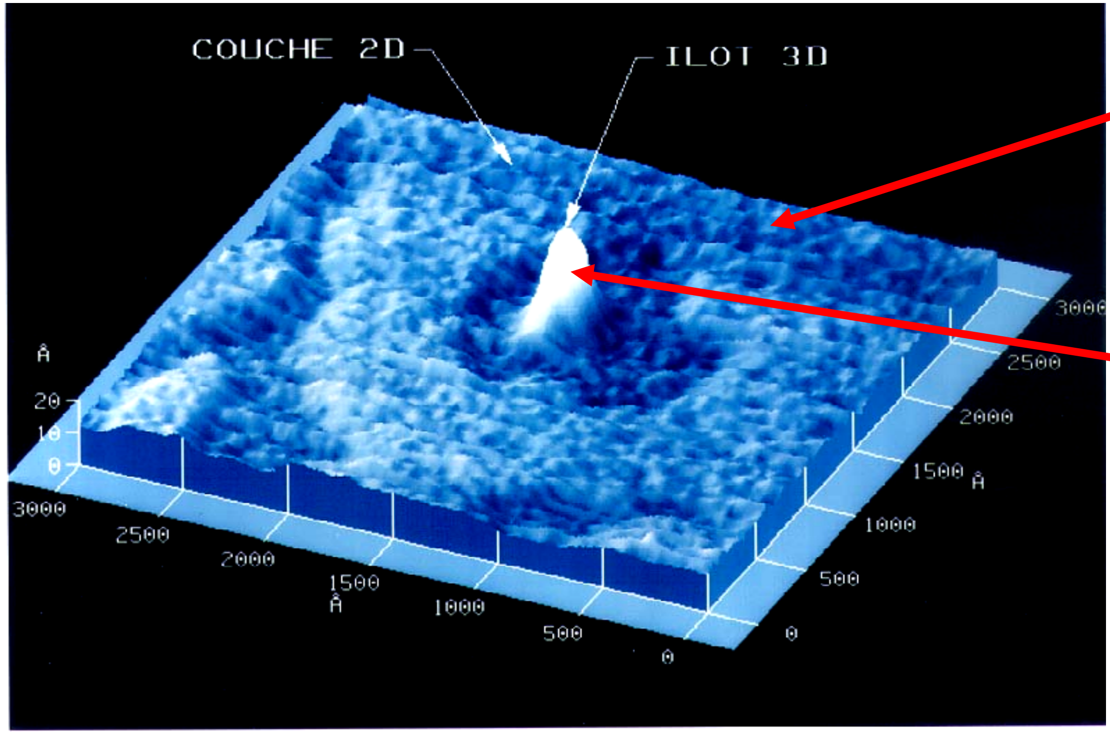
• Excitation spectroscopy



Kammerer, PRB **65**, 033313 (2001)

InAs/GaAs self-assembled QD

• Morphology of an epitaxially grown QD



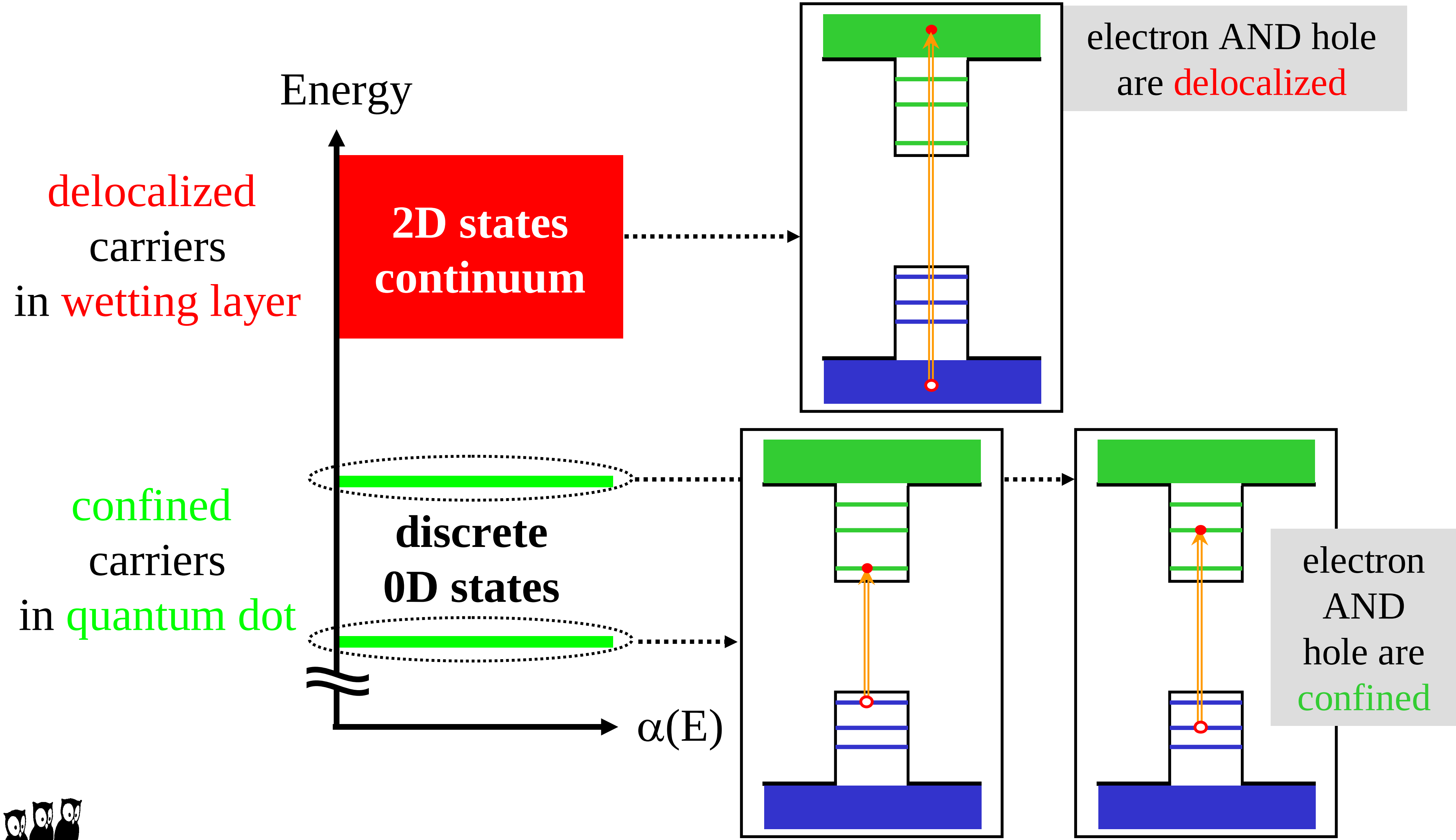
J. M. Gérard *et al*., J. Cryst. Growth **150**, 351 (1995)

InAs wetting layer
• thickness ~ 1 nm
(1.7 ML)

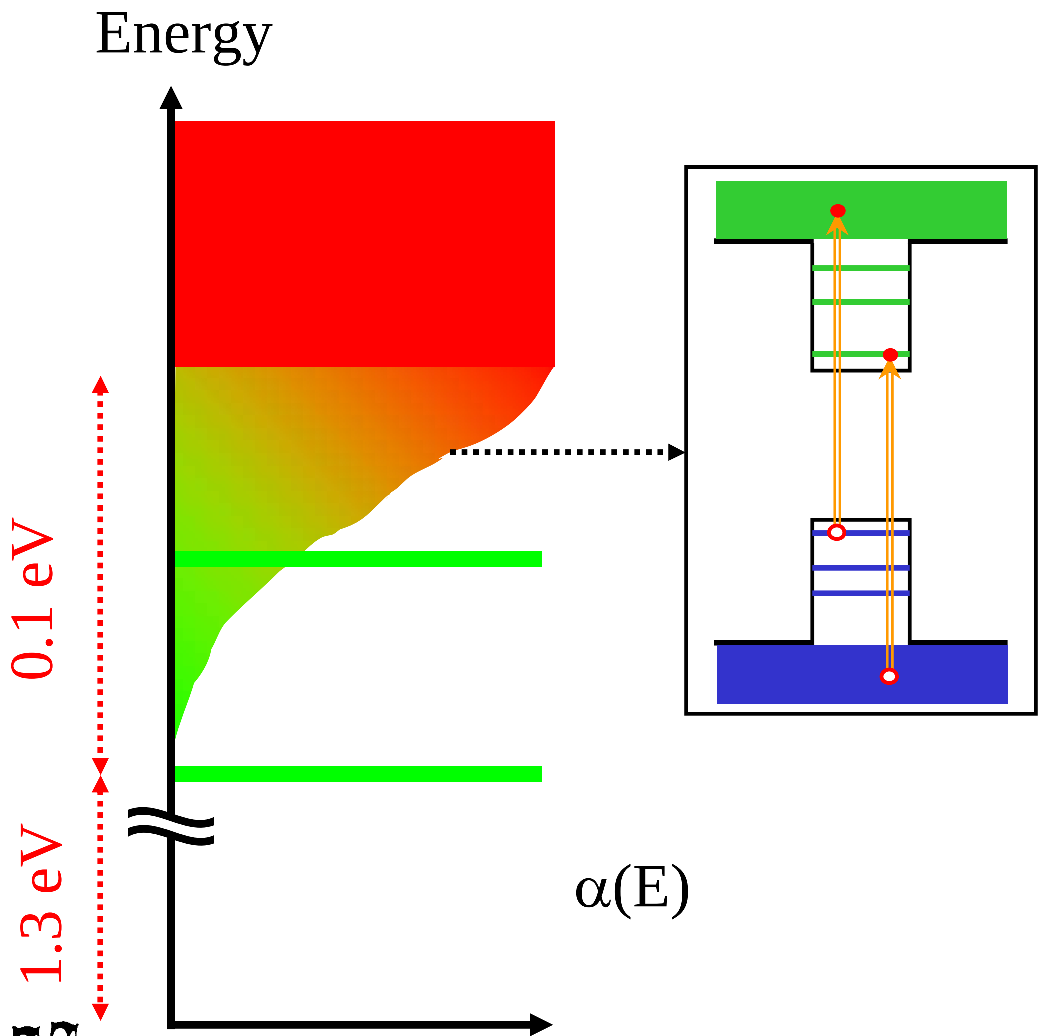
InAs quantum dot
• diameter ~ 20 nm
• height ~ 3 nm

3D confinement of electrons and holes

2D versus 0D electronic states



2D-0D interband transitions



• Interband transitions involve (electron-hole) pairs

one shall not miss the cases

where { hole confined / electron delocalized }

and

{ hole delocalized / electron confined }

• Weak transition $|\langle \phi_{2D} | \phi_{0D} \rangle| \ll 1$

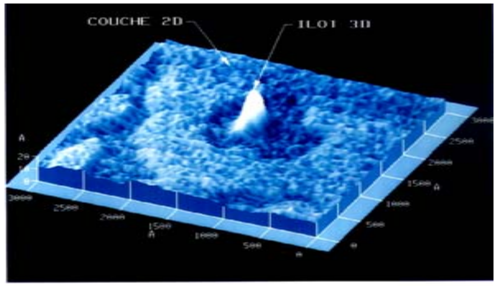
• But one carrier belongs to a quasi-continuum of states

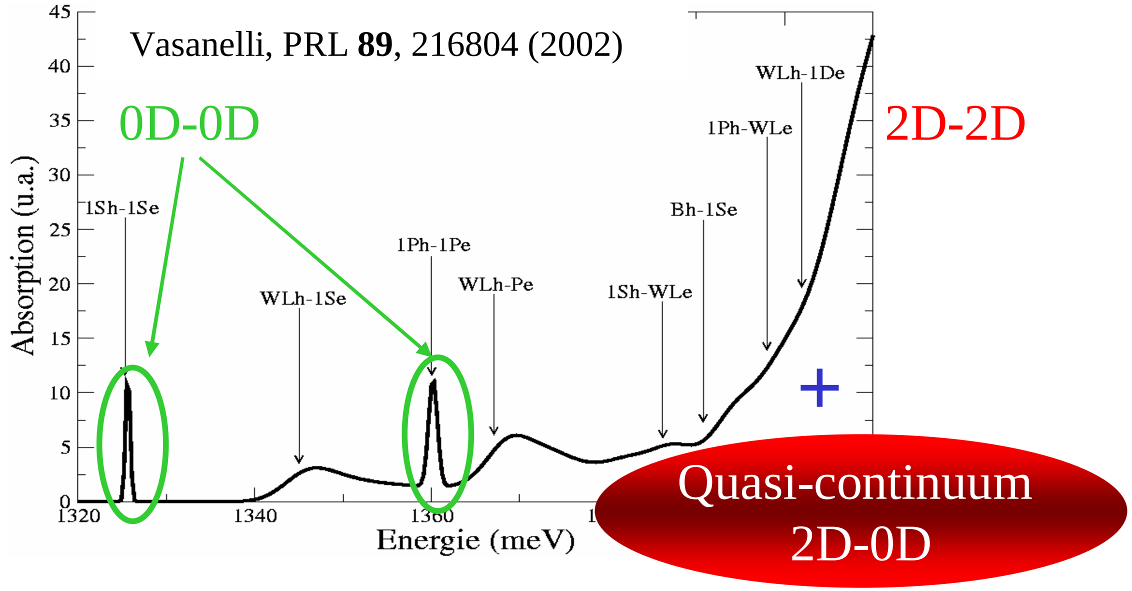
density of crossed states $\rho_{2D/0D}(E)$

INTRINSIC EFFECT !

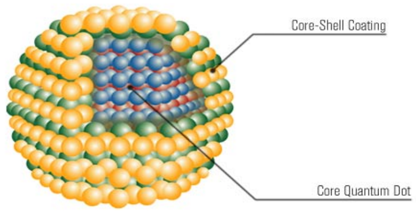
QD = system with discrete lines ?

- Epitaxially grown QD





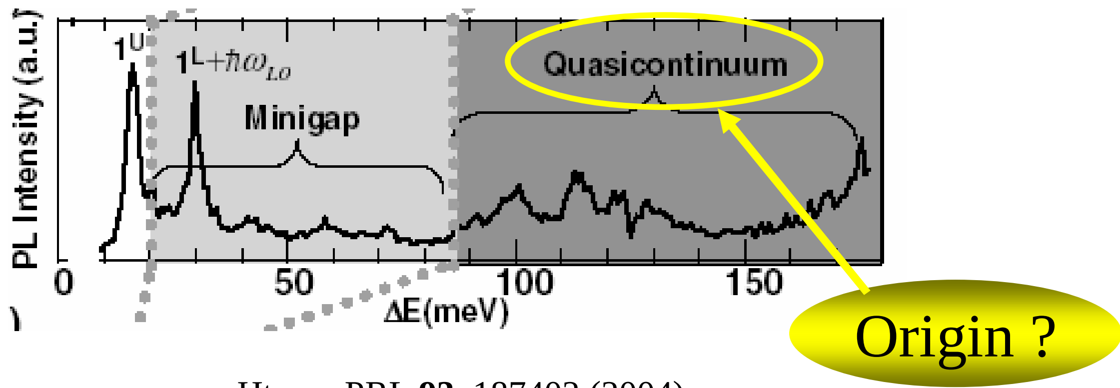
- Colloidal QD



no wetting layer

but an insulating matrix

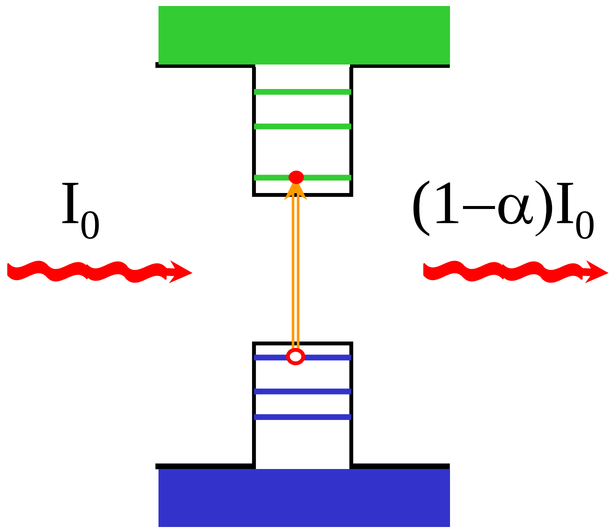
⇒ no 2D-0D quasi-continuum

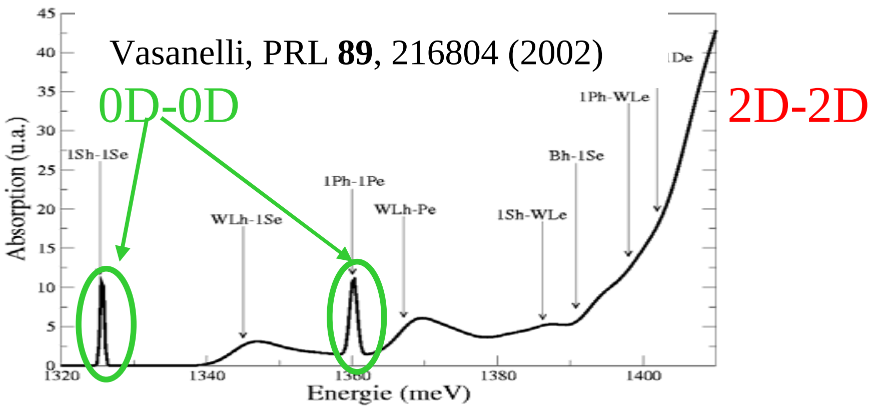


Htoon, PRL **93**, 187402 (2004)

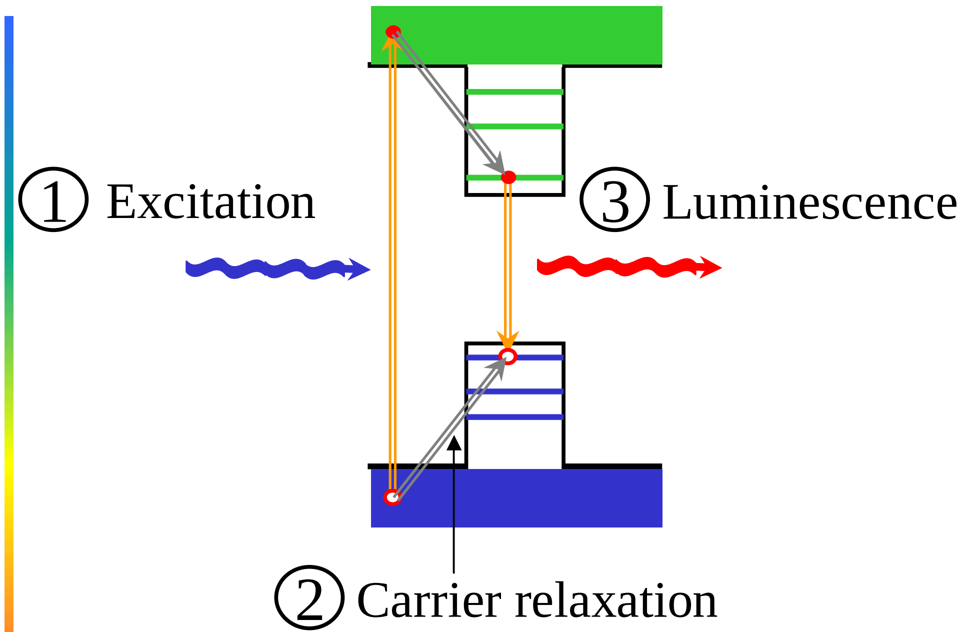
From absorption to emission

- Absorption





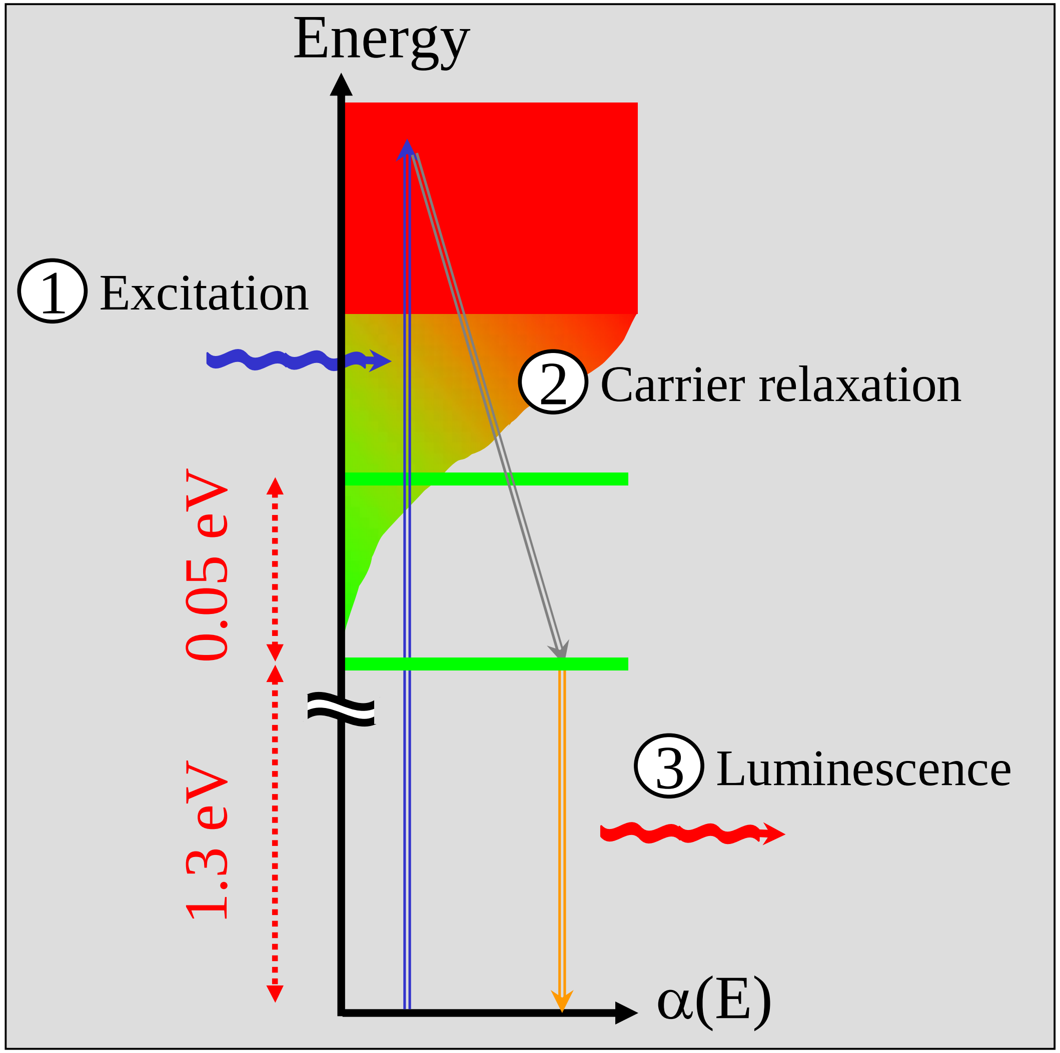
- Emission



ps range → $\tau_{②} \ll \tau_{③}$ ← ns range

Emission comes
<u>only</u> from the lowest transition

Two-level system approximation

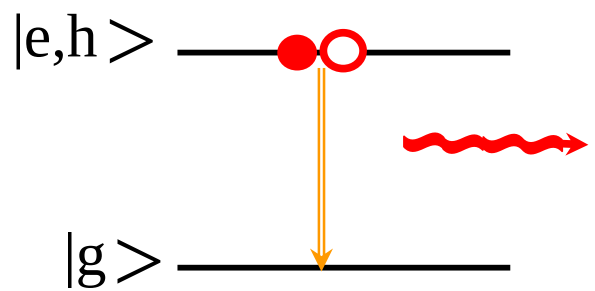


- QD = complex system in solid state physics

BUT

the efficient carrier relaxation shortcuts the dimensionality transition in the QD absorption spectrum

- Two-level system = first-order approximation for the description of the QD optical response in emission



Fundamental transition fine structure

- Fundamental interband transition corresponds to (e,h) pair state with a heavy-like hole

$$|electron\rangle = |J = 1/2, J_z = \pm 1/2, ...\rangle$$
$$|heavy\,hole\rangle = |J = 3/2, J_z = \pm 3/2, ...\rangle$$

- Four-fold degeneracy

$|e,h\rangle = |J = 2, J_z = +1, ...\rangle$	emits σ_+ light	bright states
$= |J = 2, J_z = -1, ...\rangle$	emits σ_- light	
$= |J = 2, J_z = +2, ...\rangle$	no emission	dark states
$= |J = 2, J_z = -2, ...\rangle$	no emission	

- Coulomb interaction between pair states

$$\langle \psi_{c'}(1), \psi_{v'}(2) | V_c | \psi_c(1), \psi_v(2) \rangle$$ direct interaction term

$$\langle \psi_{c'}(1), \psi_{v'}(2) | V_c | \psi_v(1), \psi_c(2) \rangle$$ indirect or exchange interaction term

Dark and bright states

0.5 meV (InAs) – 4 meV (CdSe)

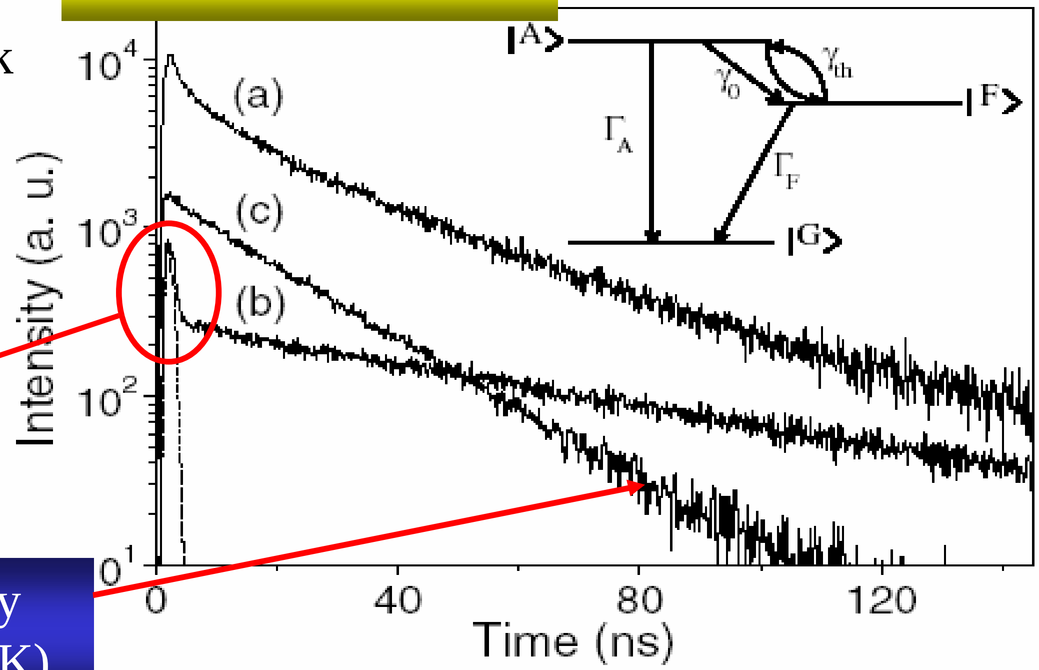
J_z=+/- 1 bright

J_z=+/- 2 dark

• Short range part of exchange interaction lifts degeneracy of bright and dark states

• No luminescence from dark states
BUT
interplay between the bright and dark states during population relaxation dynamics

Pulsed excitation



Bi-exponential decay at low temperature (16K)

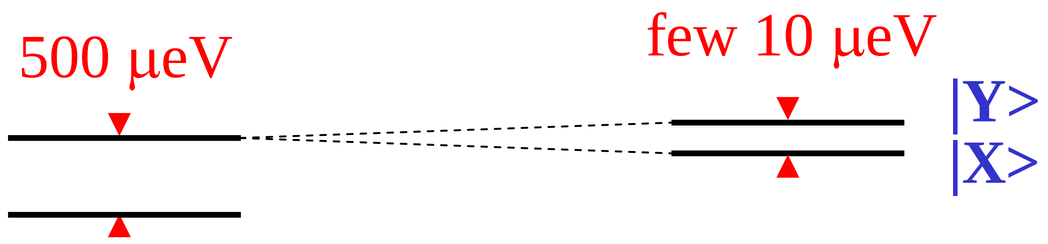
Mono-exponential decay at high temperature (140K)

Labeau, PRL **90**, 257404 (2003)

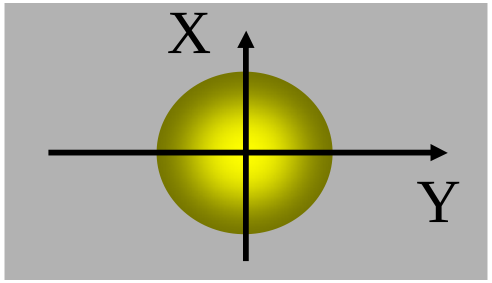
Rotational symmetry breaking

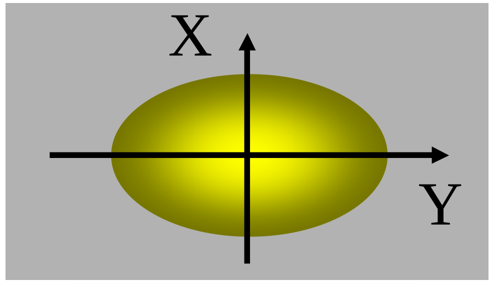
bright J_z=+/- 1

dark J_z=+/- 2



short range exchange interaction



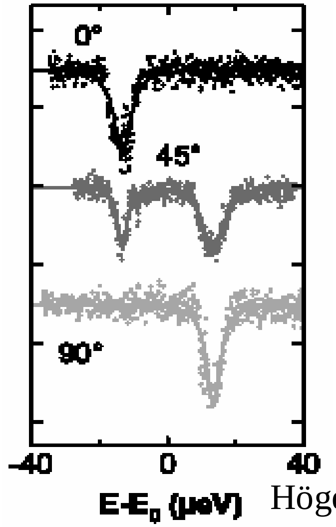


short range + long range exchange interaction

- In a QD of broken symmetry the new bright states emit linearly polarized light

$$|X\rangle = \frac{|+1\rangle + |-1\rangle}{\sqrt{2}}$$

$$|Y\rangle = \frac{|+1\rangle - |-1\rangle}{i\sqrt{2}}$$



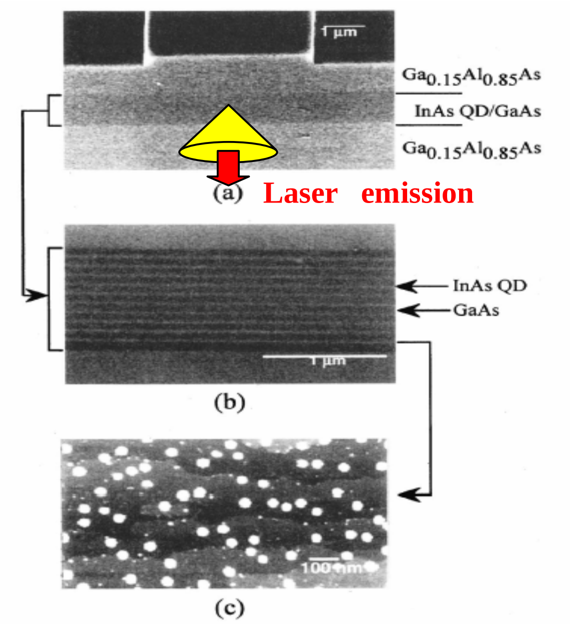
Analyzer // (Ox)

Analyzer // (Oy)

Högele, PRL **93**, 217401 (2004)

From fundamental optical properties to optical applications

QD laser



Nakamura
J. Appl. Phys. **94**, 1184 (2003)

QD laser

Opto-electronics

• Gain and threshold current density of ideal QD laser (no inhomogeneous broadening)

Arakawa and Sakaki APL **40**, 939 (1982)

- higher maximum gain due to narrow gain spectrum (0D density of states)
- no temperature dependence of threshold current

• size fluctuations in real QD laser > inhomogenous broadening to be reduced

• existence of intrinsic limit = homogeneous linewidth due to phonon coupling (Γ~10 meV at 300K)

• no quenching of carrier escape

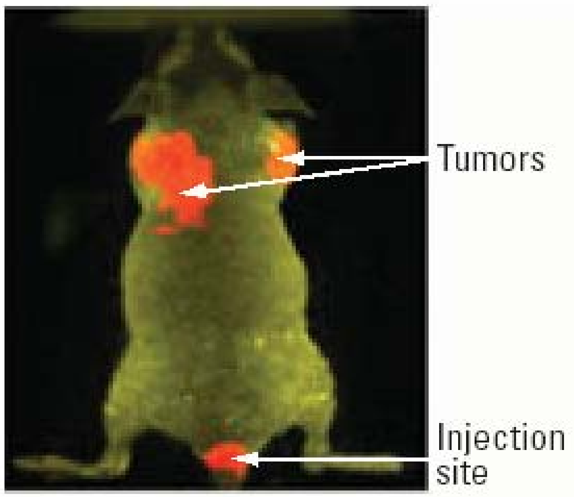
QW versus QD lasers = ?

Biological sensors

Bio-physics

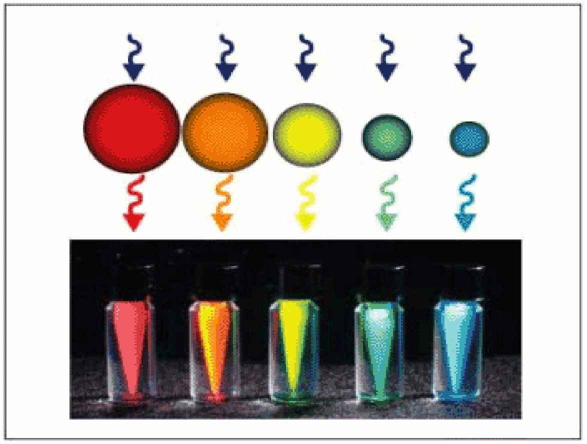
Biological sensors

In vivo imaging of cancer cells in a living mouse



Gao, Nature Biot. **22**, 969 (2004)

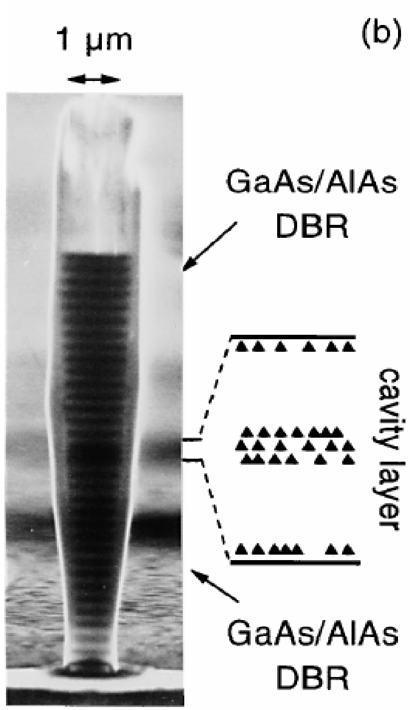
- colloidal QDs
- broadly tunable in the visible spectrum by size variation



- high quantum efficiency
- bio-conjugating colloidal QDs (cell labeling, cell tracking, *in vivo* imaging, DNA detection, multiplexed beads…)

(see Alivisatos, Nature Biot. **22**, 47 (2004) and previous lectures)

Single photon source



Gérard, PRL **81**, 1110 (1998)

Single photon source

Quantum optics

- single QD generates single photon state
- non-classical source of radiation : sub-Poissonian statistics
- quantum optics in solid state
- quantum information processing

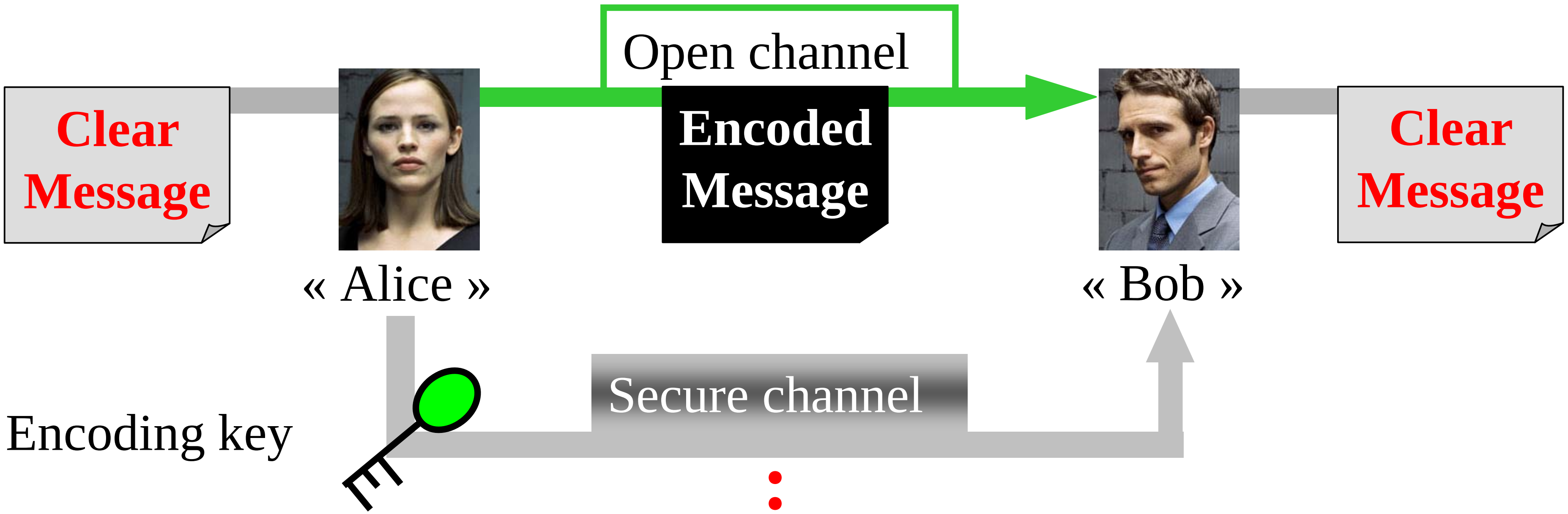
Quantum optics in solid state with a non-classical source of radiation

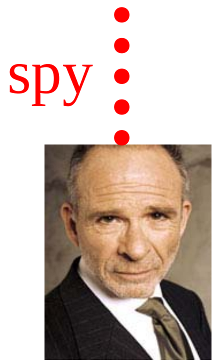
Outline

- quantum cryptography
- single-photon source
- carrier quantum cascade

Secret key cryptography

• Goal : transmission of secret message

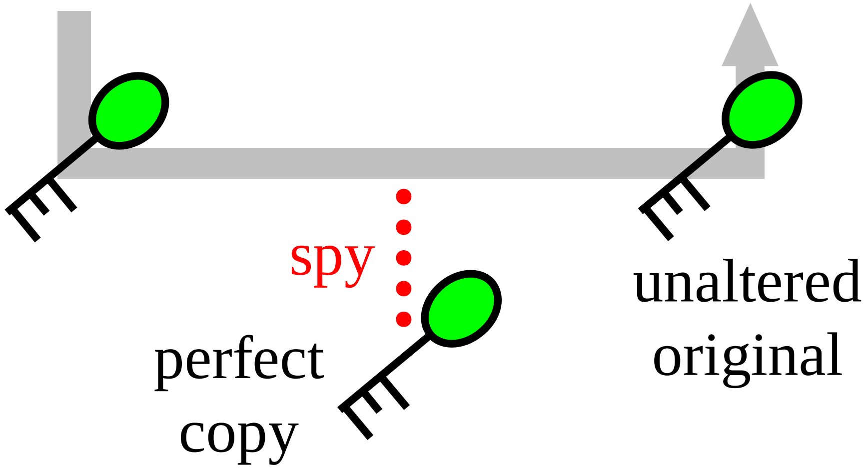




Secret key cryptography requires secure channel for key distribution

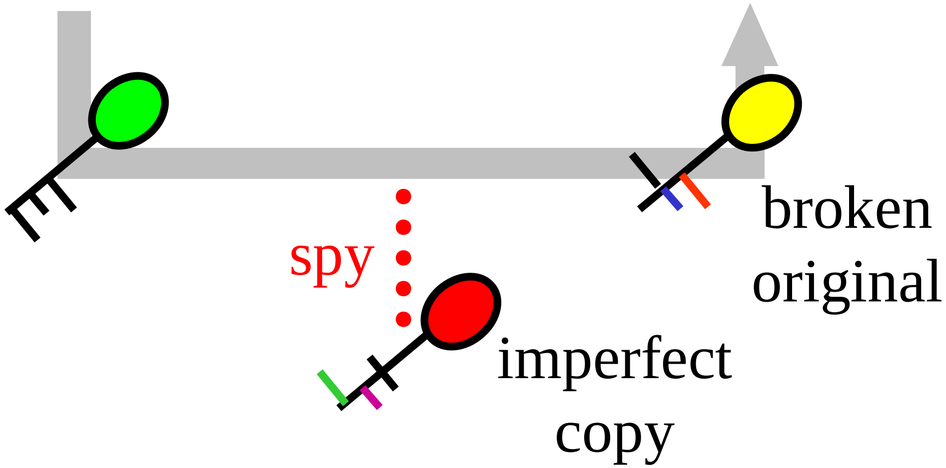
Classical versus quantum information

• Classical information



in principle every classical channel can be monitored passively

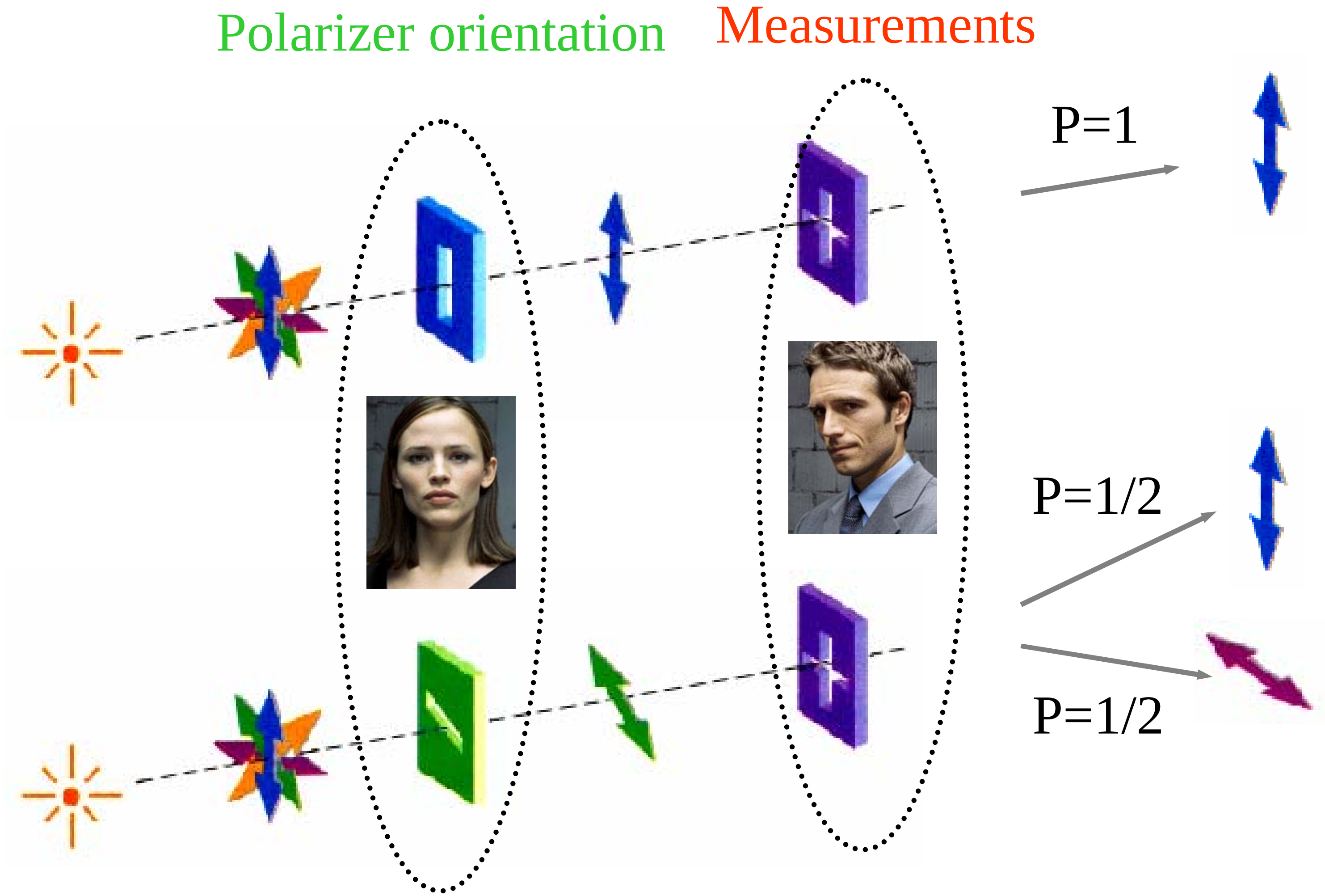
• Quantum information



• quantum cryptography distributes the key by transmitting quantum photon states in an open channel

• this open channel is intrinsically secure because a spy destroys the encoding key

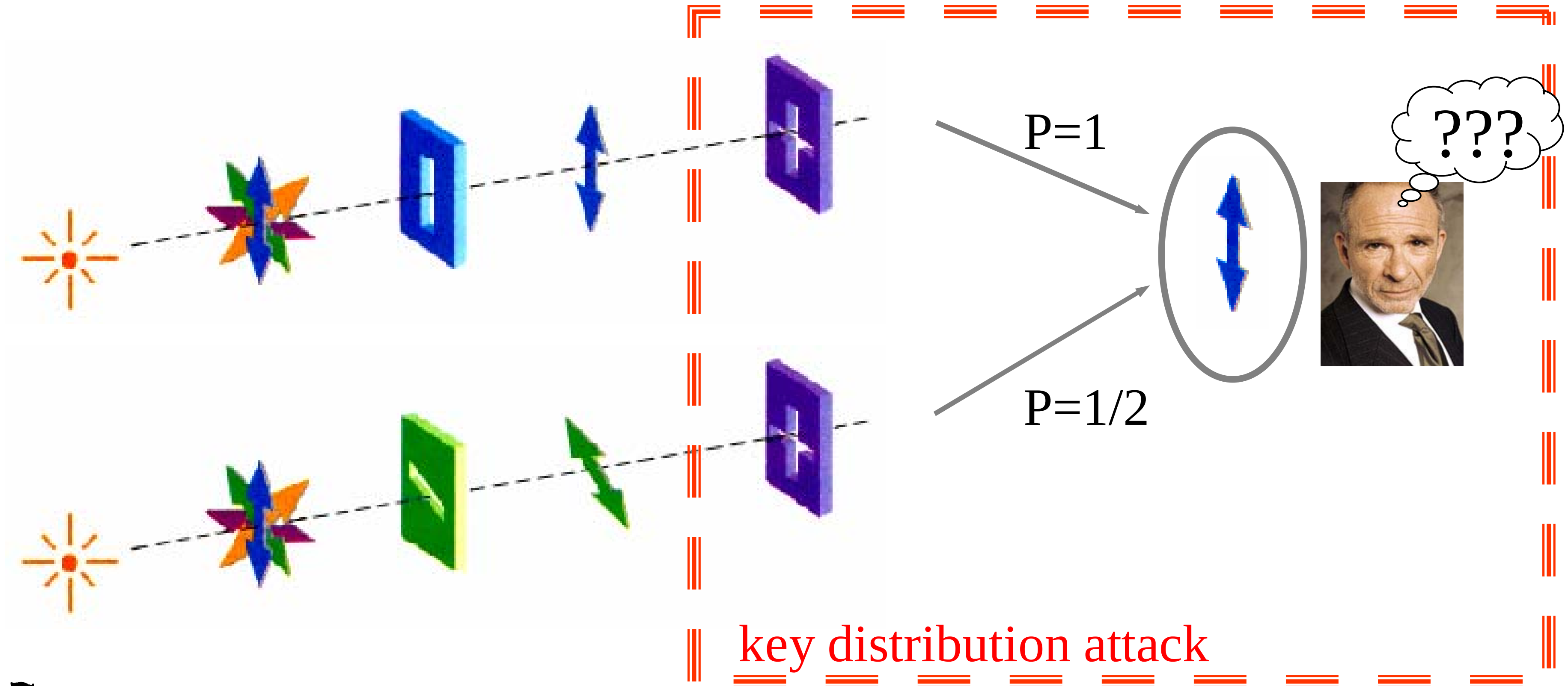
Polarization state of a single photon



Spy measurements

• Importance of having two polarization basis to avoid perfect copy of single photon state

→ the spy will generate errors in the key distribution



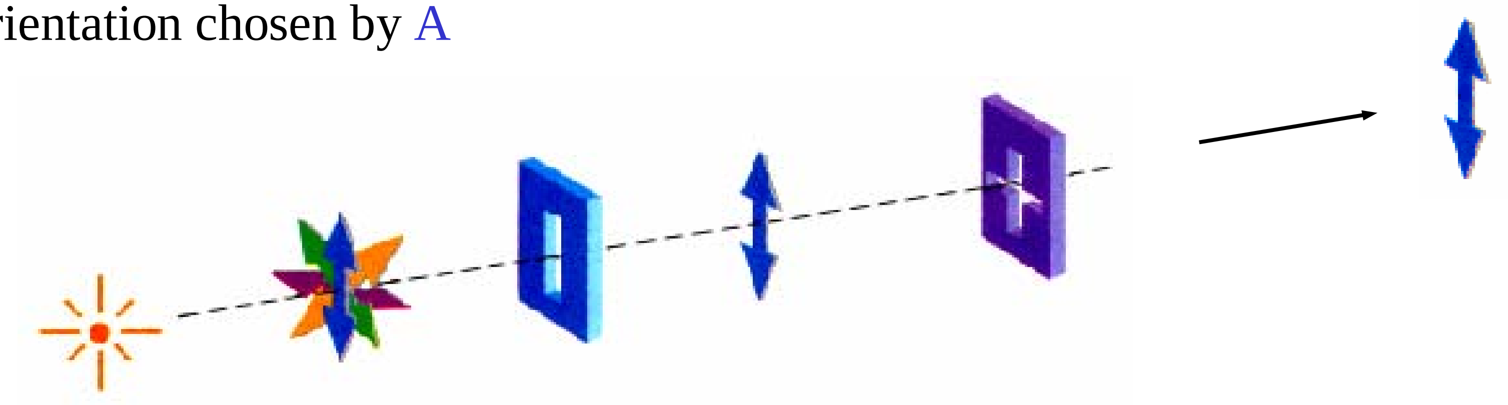
Quantum key distribution

ALICE's bit sequence	1	0	1	1	0	0	1	1	0	0	1	1	1	0
BOB's detection basis	+	×	+	+	×	×	+	+	×	+	×	×	+	+
BOB's measurements	1	0	0	1	0	0	1	1	0	0	0	1	0	0
retained bit sequence	1	–	–	1	0	0	–	1	0	0	–	1	–	0

Key distribution analysis

• B sends to A by open channel his set of detection basis and half of his measurements

• A selects the cases where the detection basis corresponds to the polarizer orientation chosen by A



in that cases B must have found the same result as A

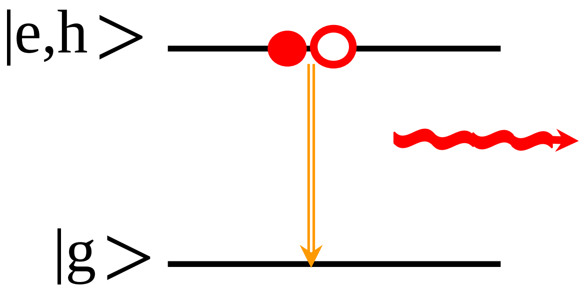
if not, this means there is a spy which has generated a single photon with an incorrect polarization orientation !

• among the retained bit sequence, A chooses a subset of bits to compose the encoding key and sends the location of these bits in the total message to A by open channel

Single QD as single-photon source

Solid-state single-photon source

- single QD

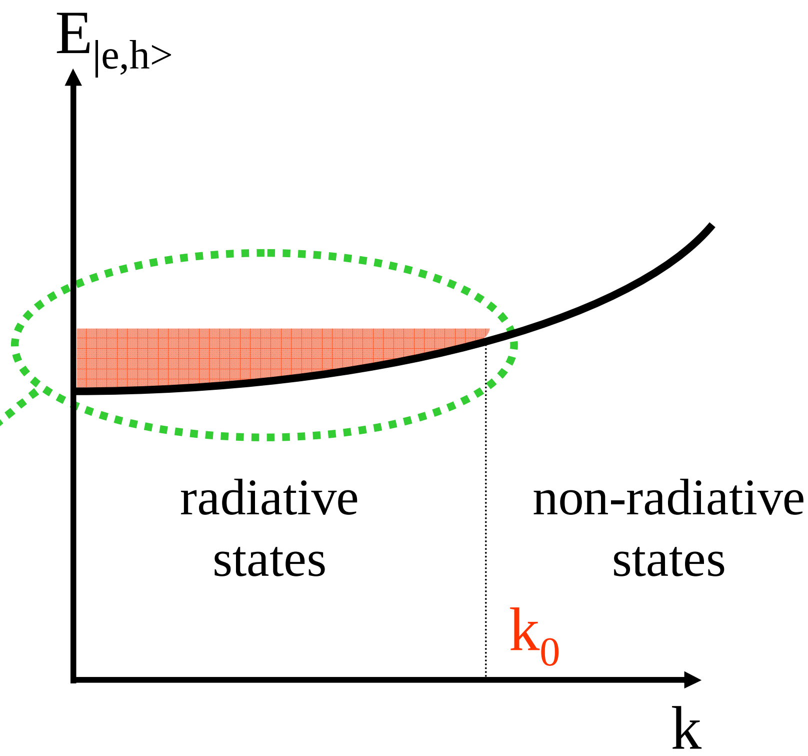


- radiative recombination of single (e,h) pair generates single photon

- this picture breaks down for a QDs ensemble

- quantum wire (1D), quantum well (2D)

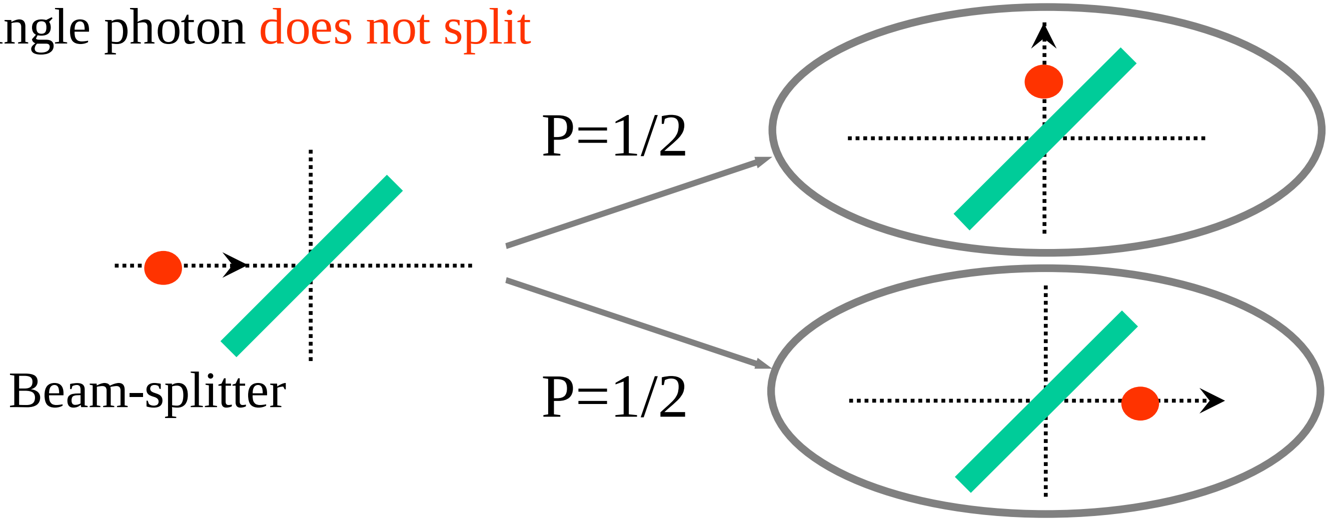
continuum of electronic states



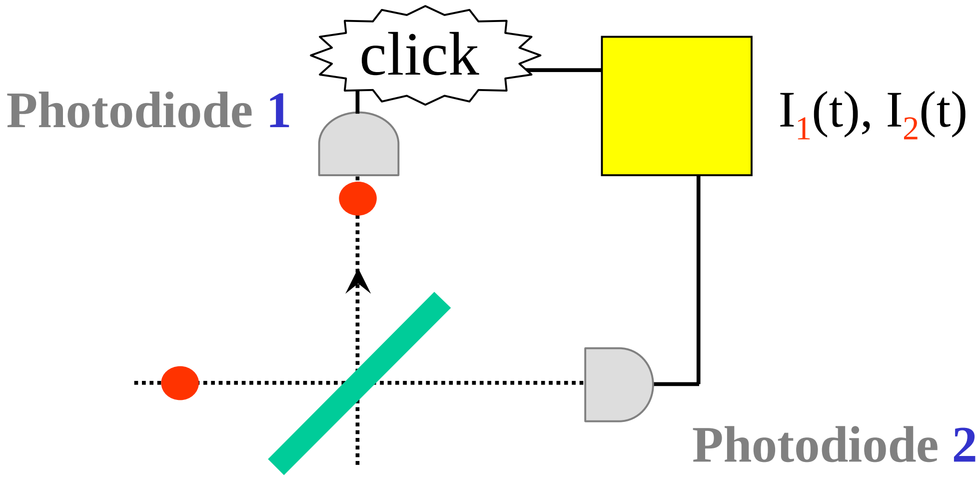
translational quantum number

Coincidence detection

• single photon does not split

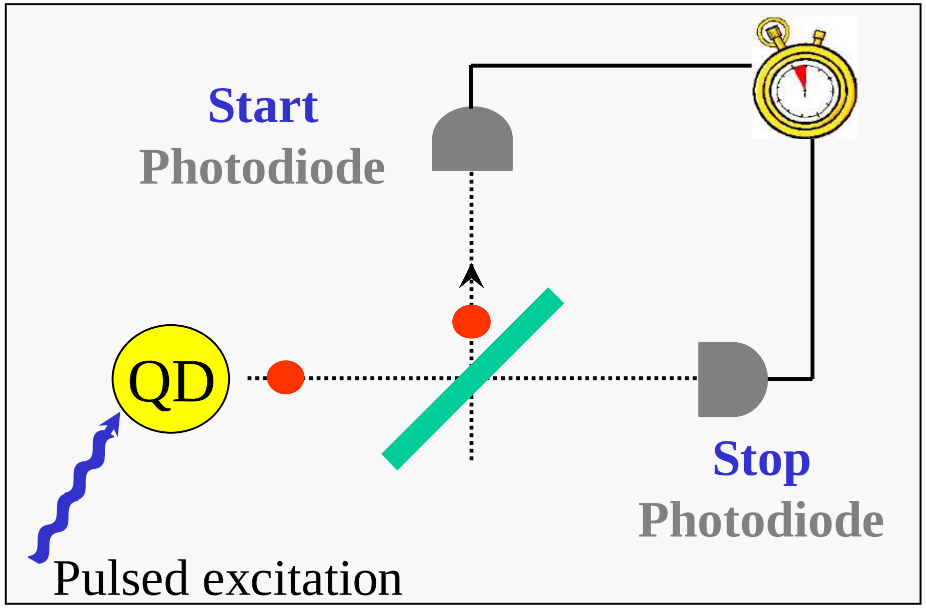


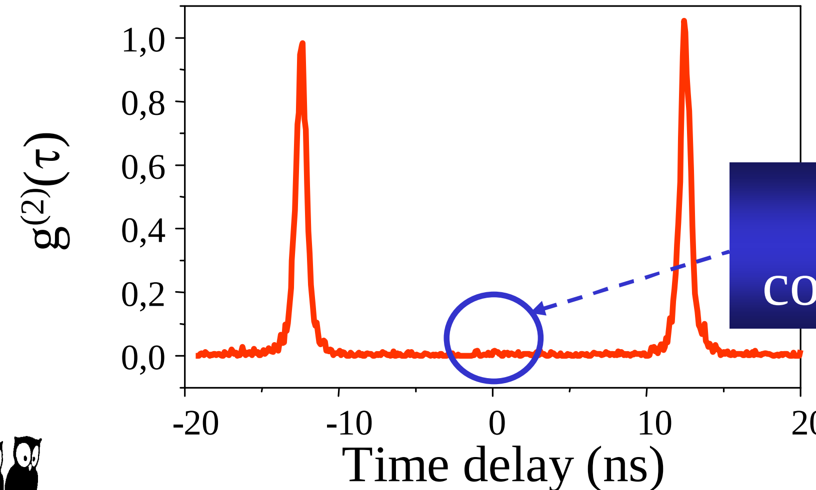
• intensity correlation experiments



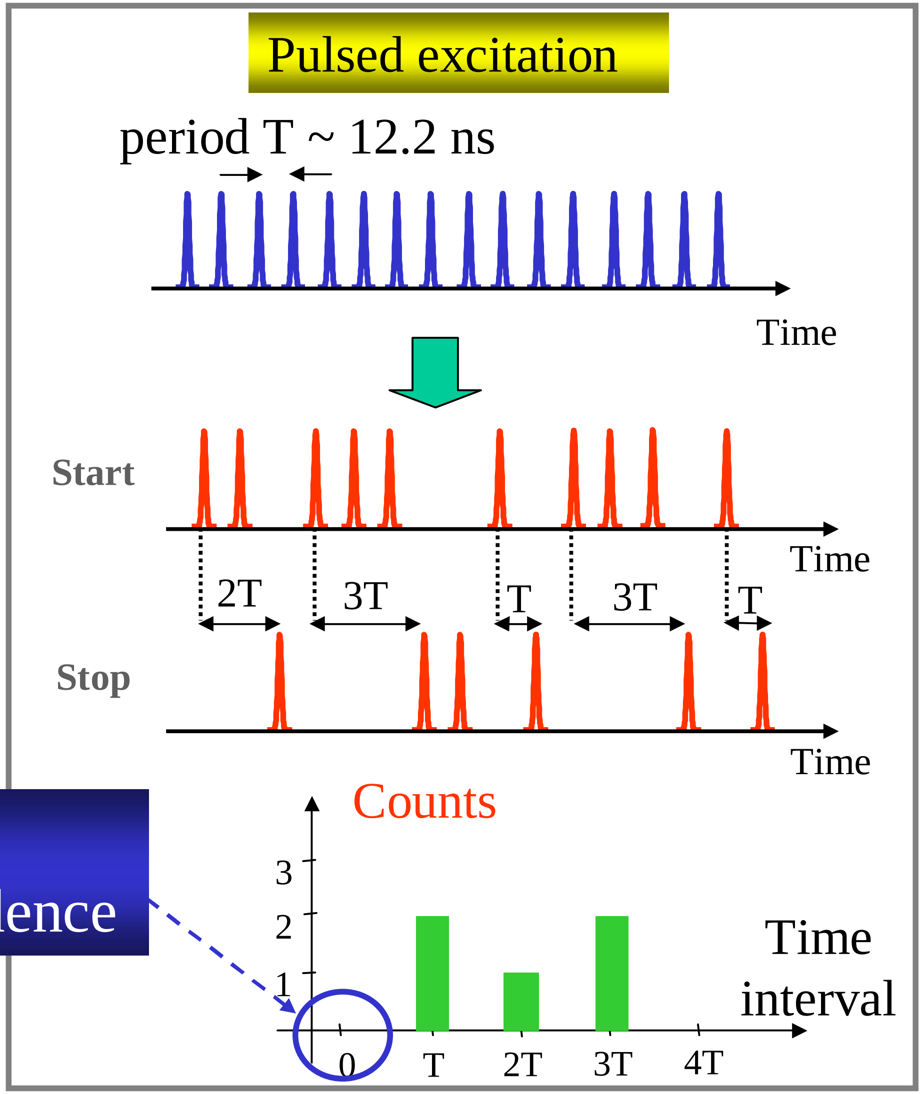
no simultaneous events on both photodiodes

Start-stop correlation experiments





No coincidence

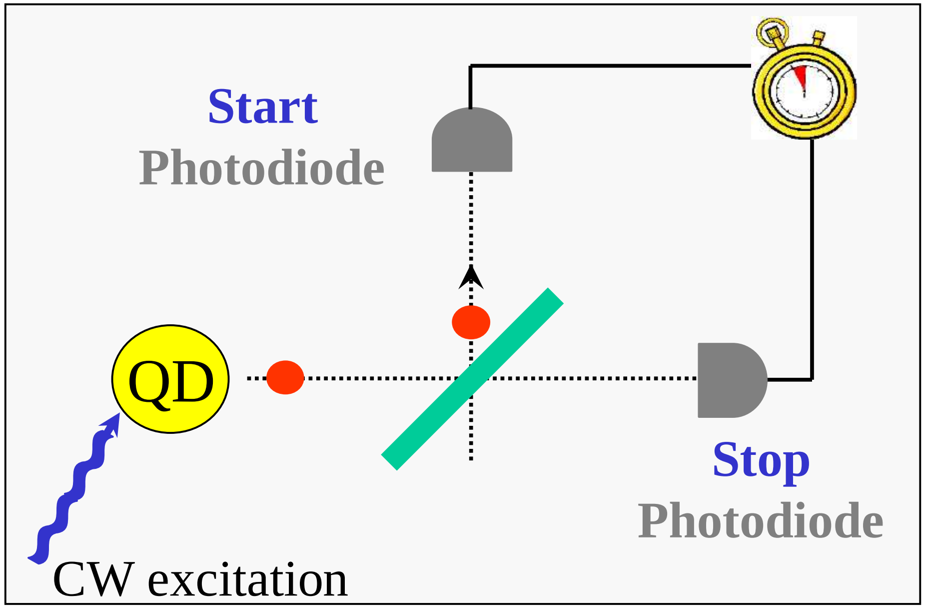


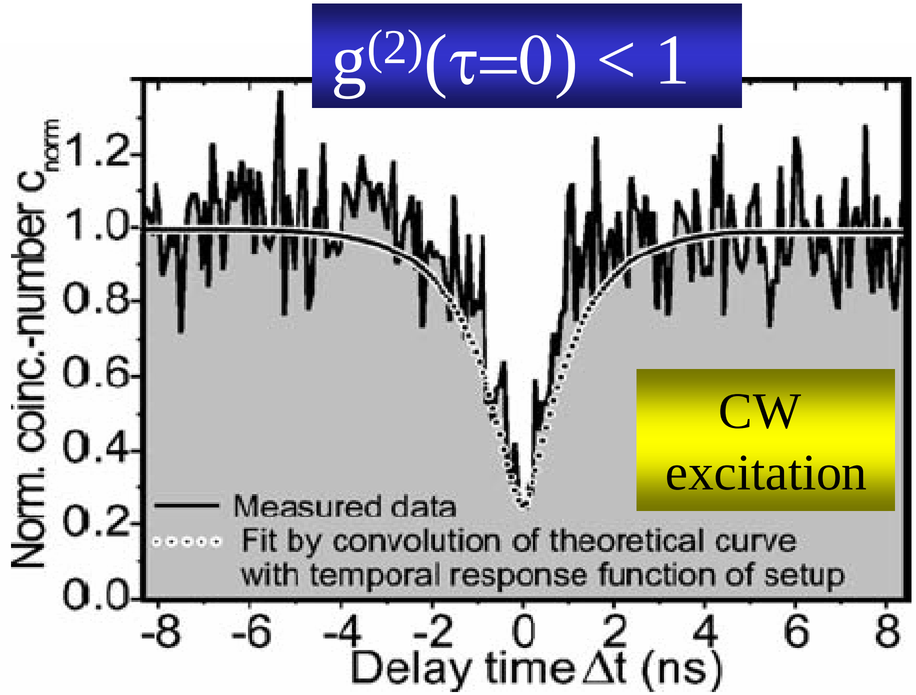
Photon anti-bunching

• normalized intensity correlation function $g^{(2)}(\tau)$

$$g^{(2)}(\tau) = \frac{\langle I_1(t) I_2(t+\tau) \rangle}{\langle I_1(t) \rangle \langle I_2(t) \rangle}$$

• start-stop correlation experiments under continuous-wave excitation

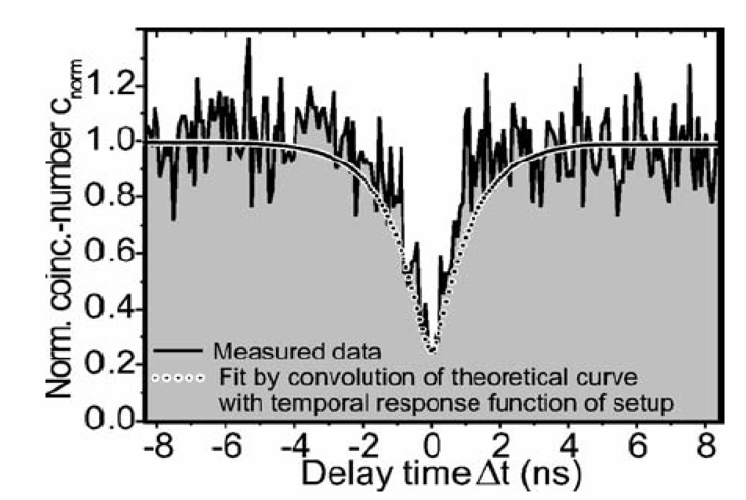


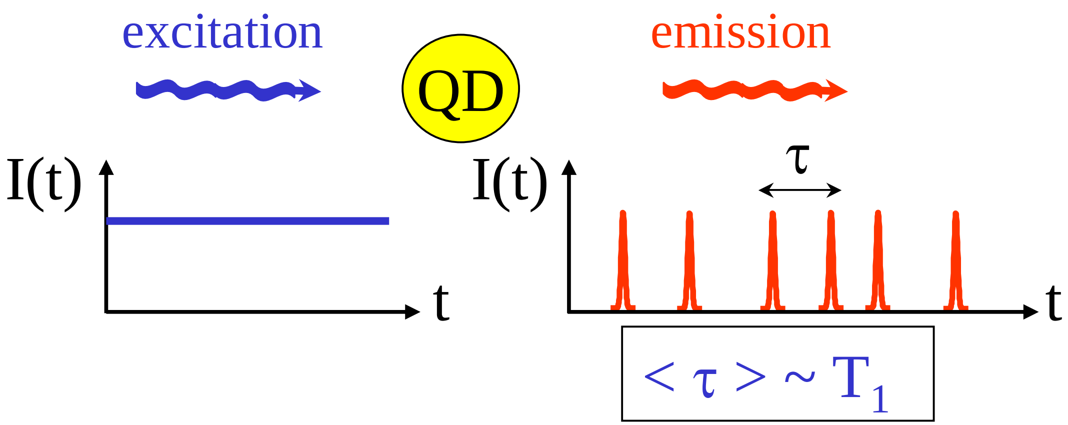


Baier, APL **84**, 648 (2004)

Sub-poissonian statistics

- $g^{(2)}(\tau=0) < 1$ reveals non-classical character of light source
- sub-poissonian photon statistics

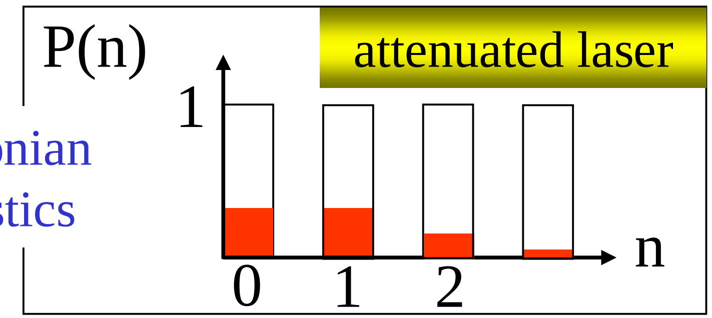


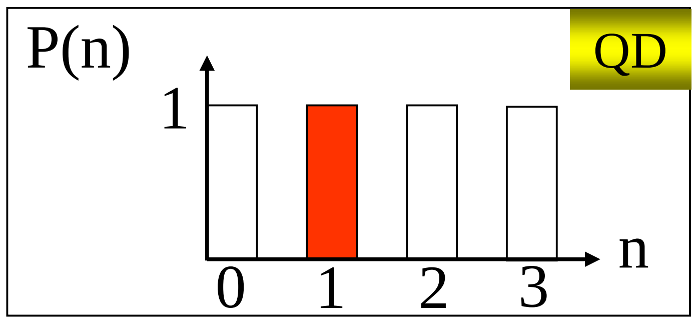


T_1 = population relaxation time

- attenuated laser versus single QD when $< n > = 1$

Poissonian statistics



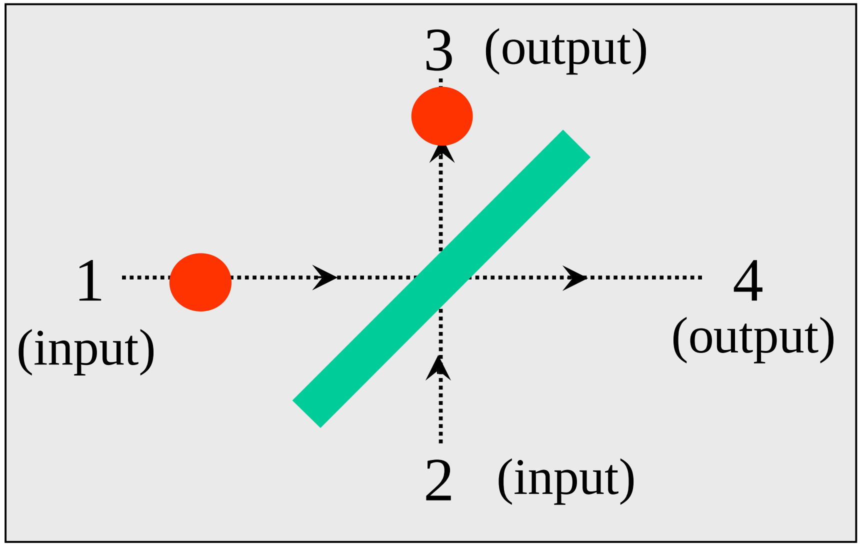


Photon coalescence

- One photon impinges on beam-splitter

$$|1_1\rangle\,|0_2\rangle \longrightarrow r\,|1_3\rangle\,|0_4\rangle + t\,|0_3\rangle\,|1_4\rangle$$

$$|0_1\rangle\,|1_2\rangle \longrightarrow t\,|1_3\rangle\,|0_4\rangle - r\,|0_3\rangle\,|1_4\rangle$$



- Two photons impinge on beam-splitter

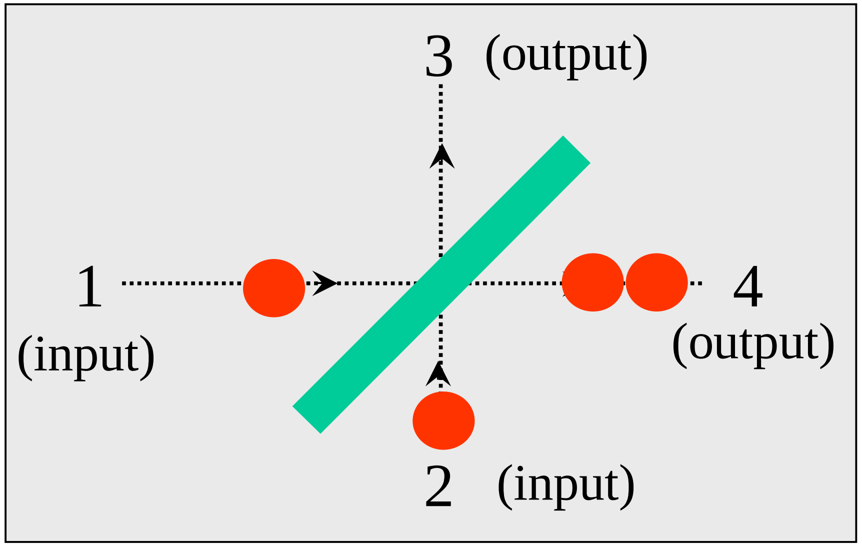
$$|1_1\rangle\,|1_2\rangle$$

$$\downarrow$$

$$t^2\,|1_3\rangle\,|1_4\rangle - r^2\,|1_3\rangle\,|1_4\rangle$$

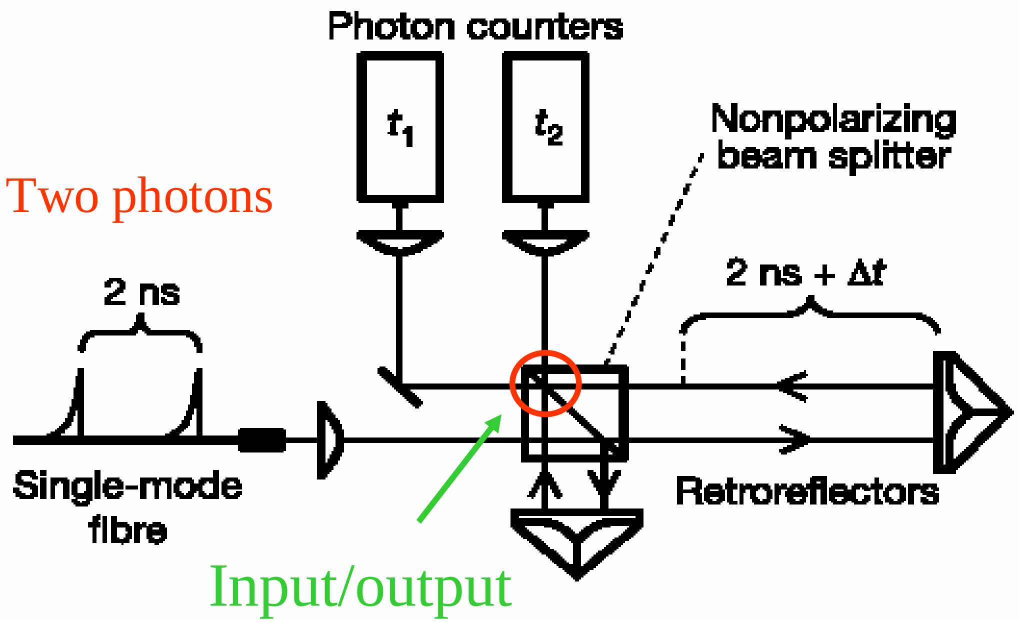
$$+\,rt\,|2_3\rangle\,|0_4\rangle - rt\,|0_3\rangle\,|2_4\rangle$$

if r = t, only two-photon states



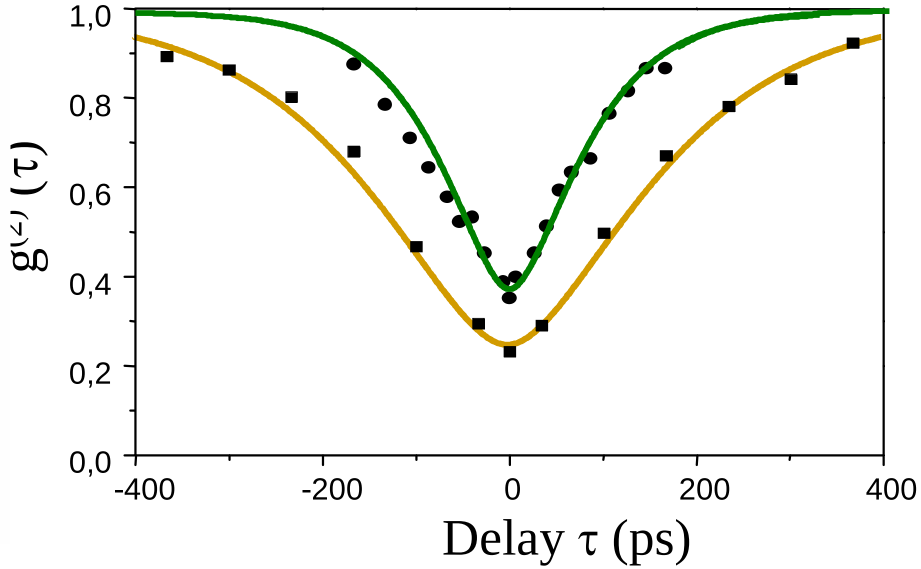
Indistinguishable photons

• Coalescence requires generation of two indistinguishable photons



Santori, Nature **419**, 594 (2002)

I. Robert-Philip, private communication



Sequential emission from the same QD

No coincidence :
coalescence of the two photons

Single-mode single-photon emission

AND ALSO

- Quantum teleportation Fattal, PRL **92**, 037904 (2004)
- Bell's inequality violation Fattal, PRL **92**, 037903 (2004)

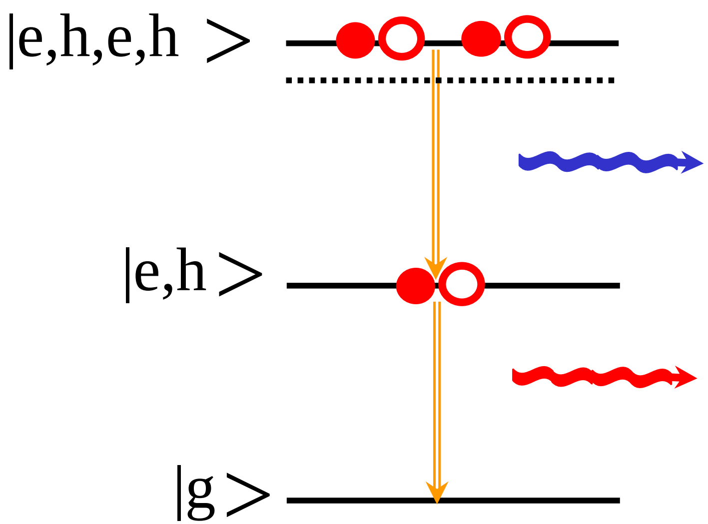
Demonstration of single-mode single-photon emission (polarization, space, spectrum) from a single QD

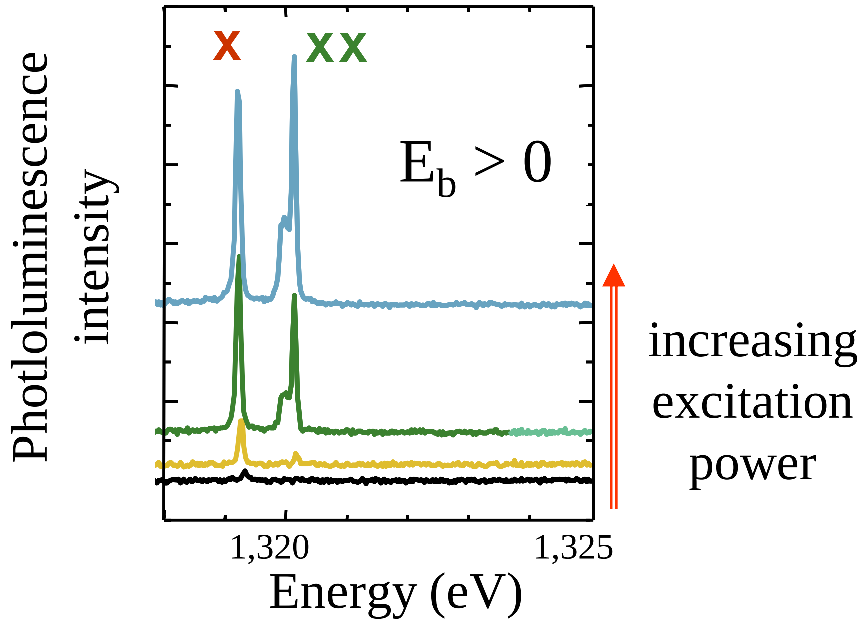
see also Moreau, APL **79**, 2865 (2001)

Single QD as two-photon source

Exciton and biexciton

• two (e,h) pairs are photogenerated in a single QD





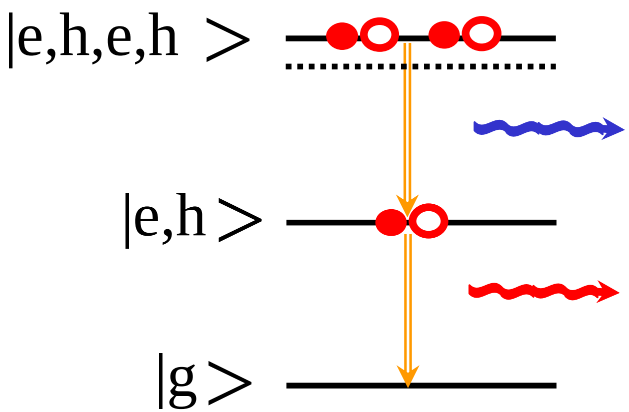
• Coulomb correlated state with 1 (e,h) pair = exciton (X)

• Coulomb correlated state with 2 (e,h) pairs = biexciton (XX)

$$E_{XX} = 2E_X + E_b$$

binding energy

Carrier quantum cascade

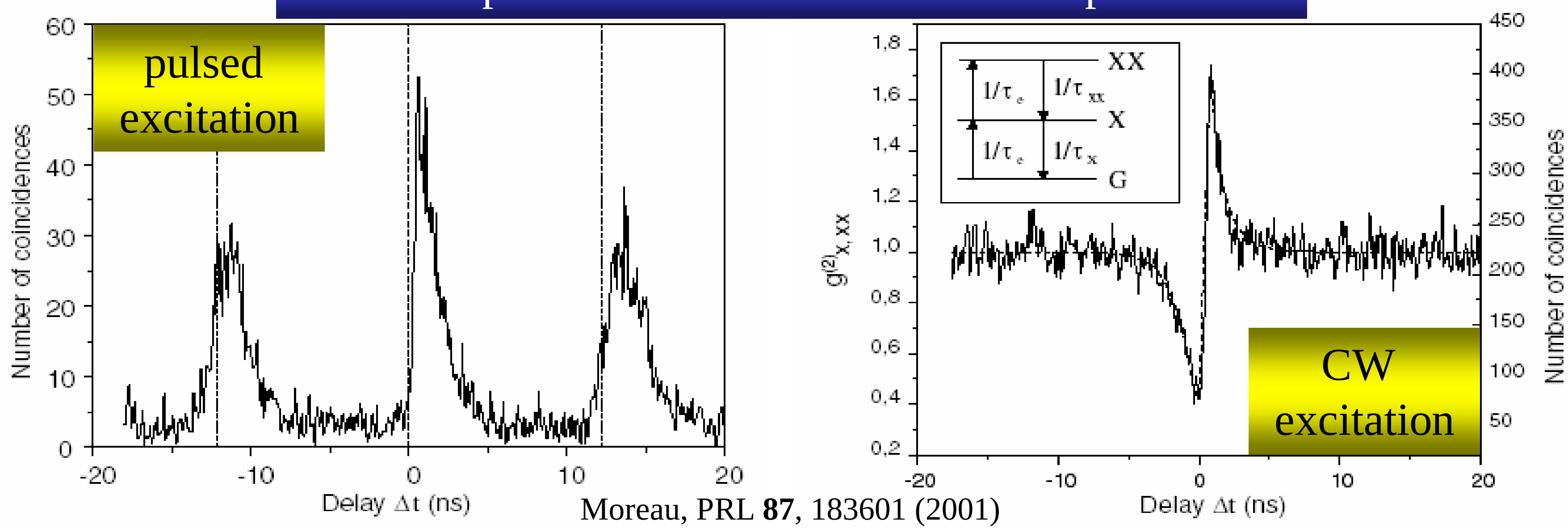


Two-color correlation experiments

XX photon on Start photodiode

X photon on Stop photodiode

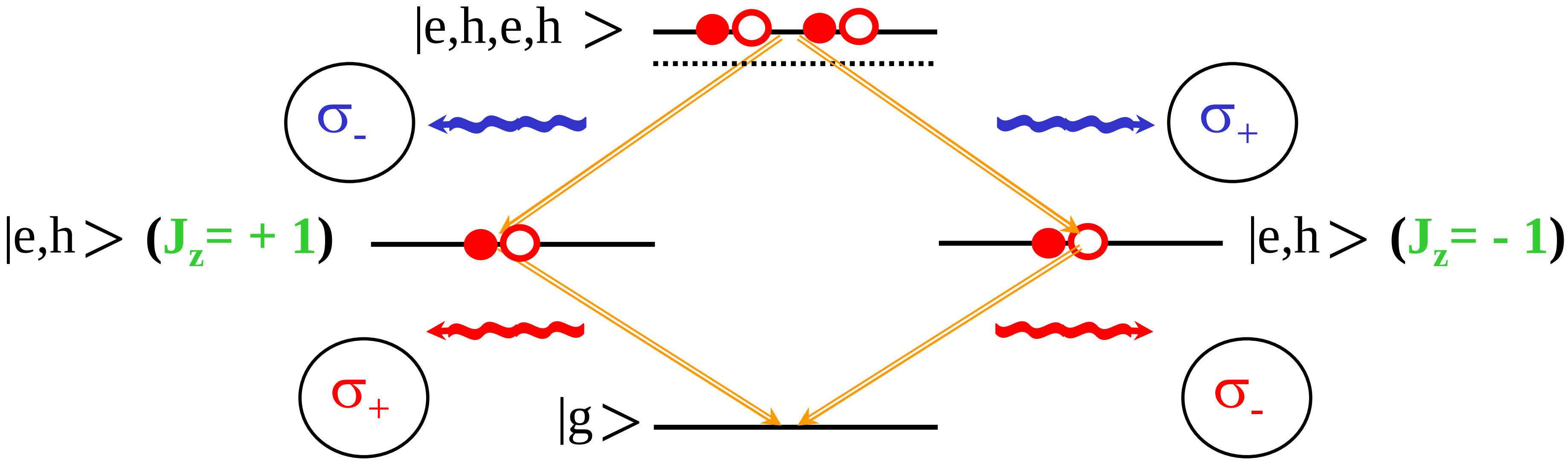
Biexciton photon is emitted before exciton photon



Moreau, PRL **87**, 183601 (2001)

Bell states generation

- Fundamental transition degeneracy



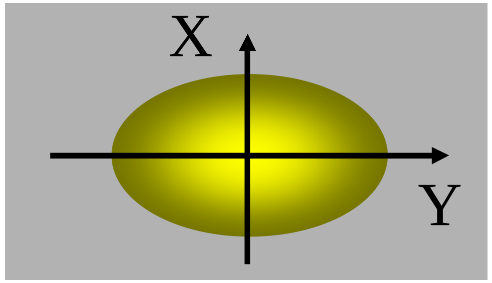
- Two parallel paths ➡ possible generation of entangled photon states

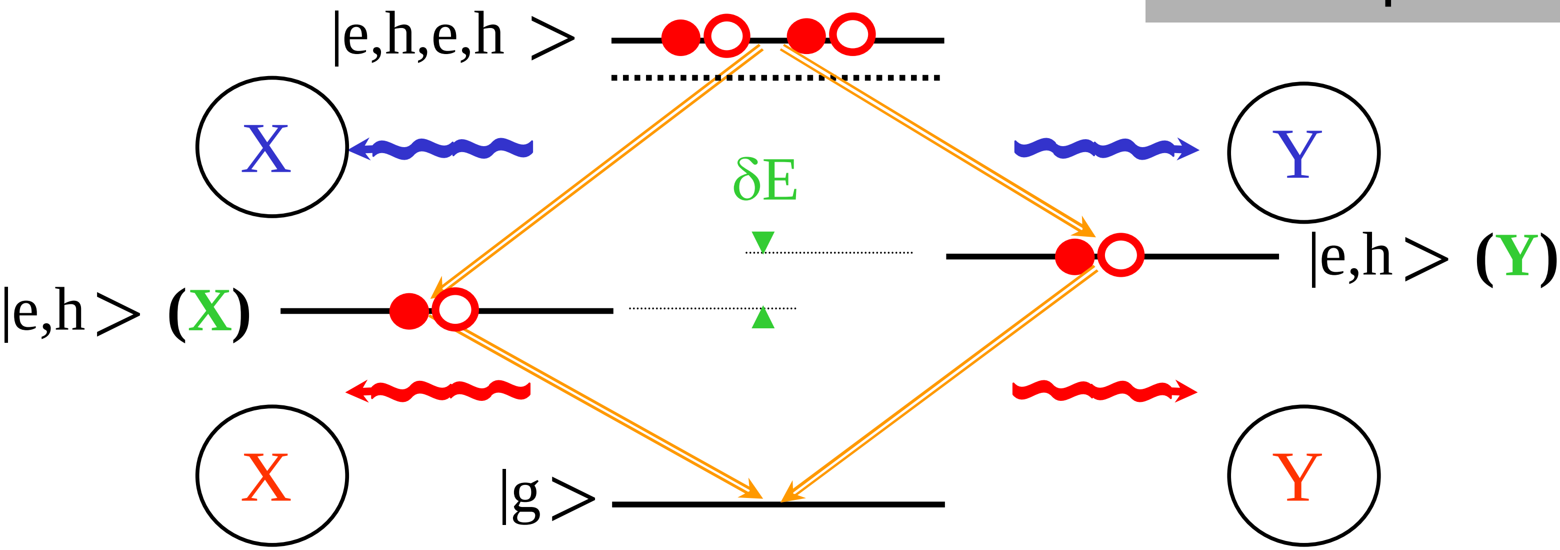
$$|\psi\rangle = \frac{|\sigma_-\rangle_{xx}|\sigma_+\rangle_x + |\sigma_+\rangle_{xx}|\sigma_-\rangle_x}{\sqrt{2}} \neq |\phi\rangle_{xx}|\phi\rangle_x$$

Benson, PRL **84**, 2513 (2000)

Rotational symmetry breaking

• Degeneracy lifted in asymmetric QDs





• Observation of polarization correlations but not entanglement

if $\delta E \gg \Gamma_X , \Gamma_Y$

Santori, PRB **66**, 045308 (2002)

Towards efficient single photon sources

All these experiments are performed at low temperature (T~5-10K) to avoid phonon-assisted dephasing processes

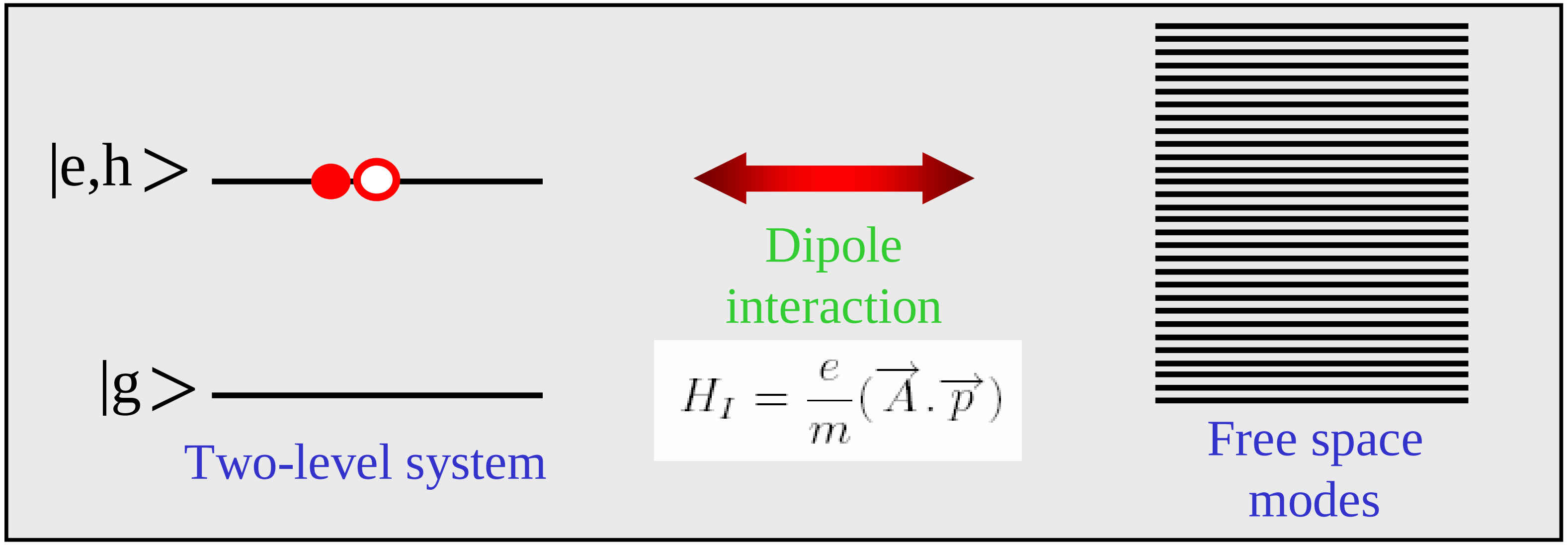
All these experiments use single QDs embedded in micropillar to control spontaneous emission

Spontaneous emission control

Outline

- Fermi's golden rule
- Purcell effect
- from weak to strong coupling

Fermi's golden rule



$$H_I = \frac{e}{m}(\vec{A}.\vec{p})$$

• Time domain analysis

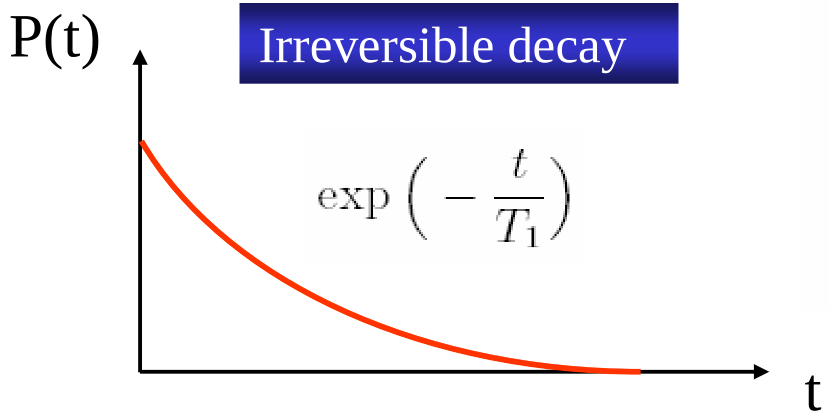
$$|i\rangle = |e,h\rangle|0_\omega\rangle \qquad |f\rangle = |g\rangle|1_\omega\rangle$$

$$P_{|i\rangle}(t) = \exp\left(-\frac{t}{T_1}\right)$$

where
$$\frac{1}{T_1} = \frac{2\pi}{\hbar}\sum_{\{|1_\omega\rangle\}} |\langle f|\widetilde{H_I}|i\rangle|^2 \delta(E_{|e,h\rangle} - \hbar\omega)$$

Natural linewidth

Irreversible decay



$$\exp\left(-\frac{t}{T_1}\right)$$

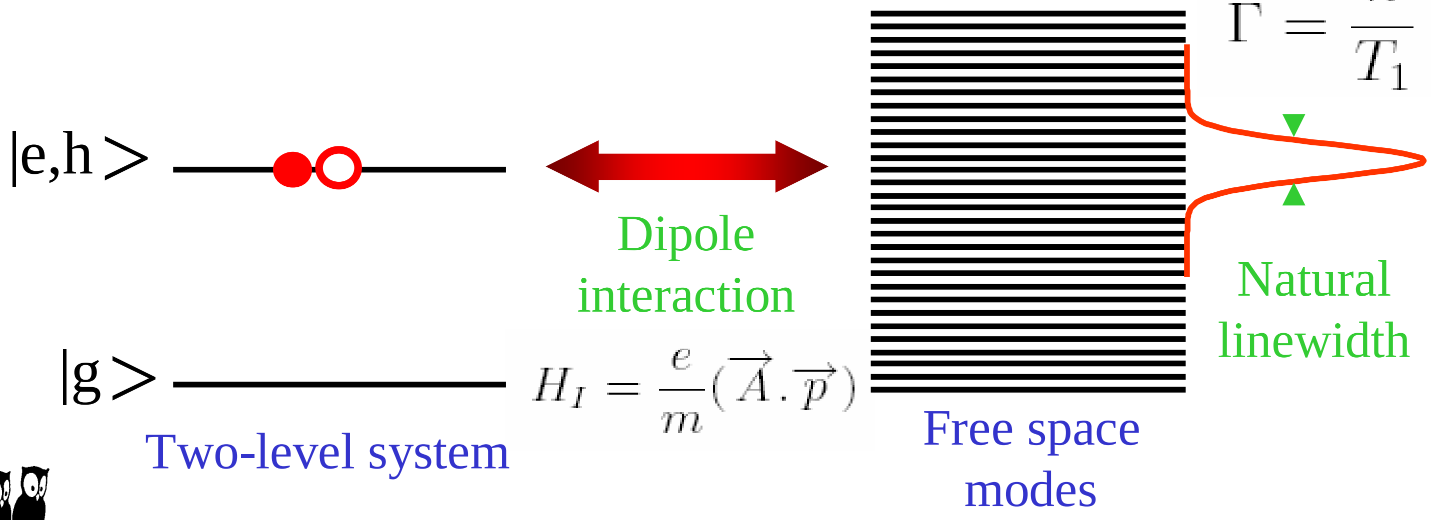
$$\frac{1}{T_1} \sim \frac{2\pi}{\hbar}|\langle f|\widetilde{H_I}|i\rangle|^2 \sum_{\{|1_\omega\rangle\}} \delta(E_{|e,h\rangle} - \hbar\omega)$$

$$\sim \frac{2\pi}{\hbar}|\langle f|\widetilde{H_I}|i\rangle|^2 \underbrace{\rho_{e.m.}(E_{|e,h\rangle})}$$

free-space density of electromagnetic modes

- Spectral domain analysis

$$\Gamma = \frac{\hbar}{T_1}$$ α Total emission



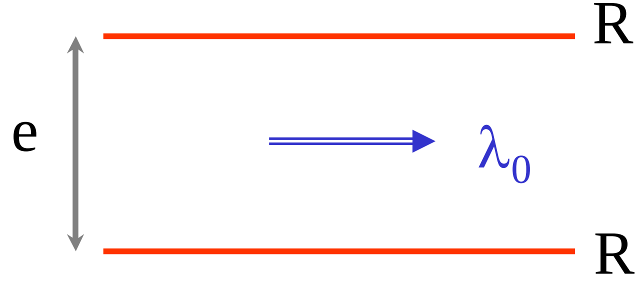
$$H_I = \frac{e}{m}(\vec{A}.\vec{p})$$

From free-space to cavities

Fabry-Pérot cavity

• Fabry-Pérot cavity with metallic mirrors

$e \gg \lambda_0/2$

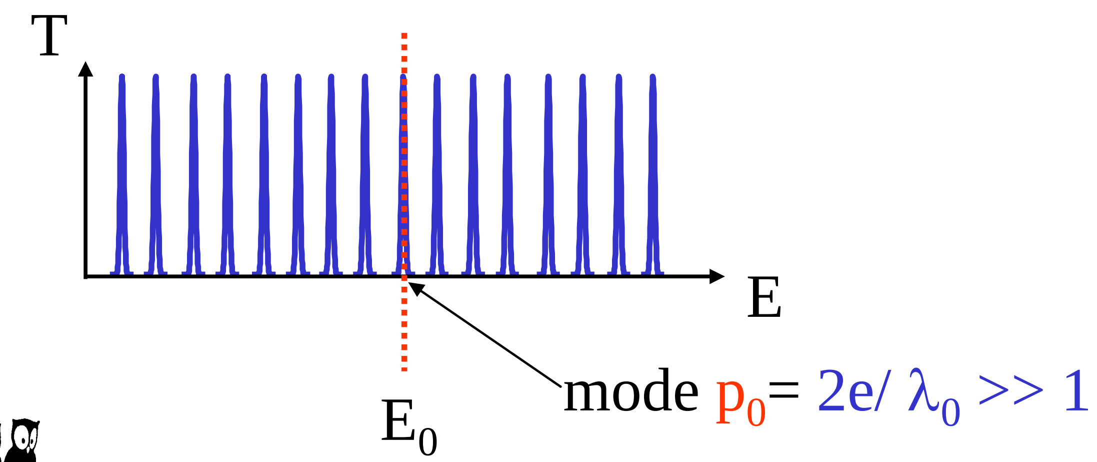


$$\nu_p = p\frac{c}{2e}\frac{1}{\cos(\theta_p)}$$

$$E_p = E_0\frac{\lambda_0/2}{e}\frac{p}{\cos(\theta_p)}$$

with $E_0 = \frac{hc}{\lambda_0}$

• Fabry-Pérot transmission at normal incidence



• Free spectral range

$$\frac{E_0}{p_0}$$

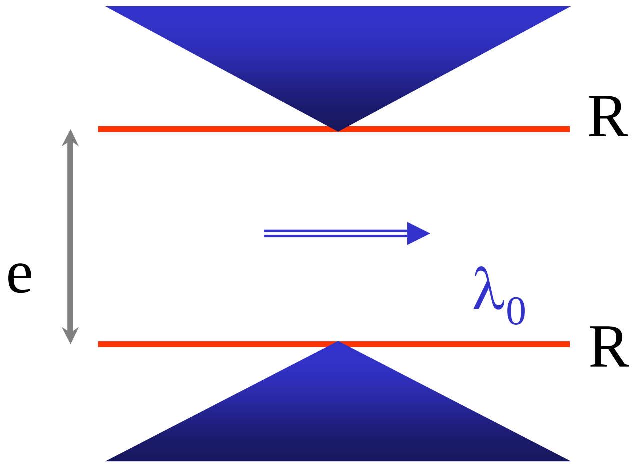
• for large cavities

$$\frac{E_0}{p_0} \ll E_0$$

Angular redistribution in planar cavities

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

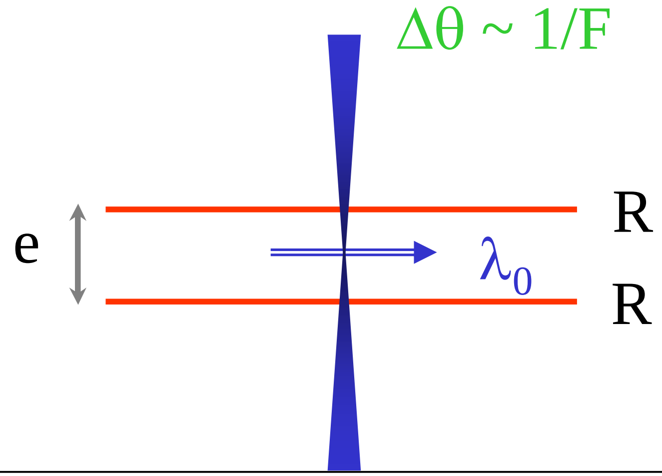
$e \gg \lambda_0/2\ ,\ p_0 \gg 1$



$$E_p = E_0 \frac{p/p_0}{\cos(\theta_p)}$$

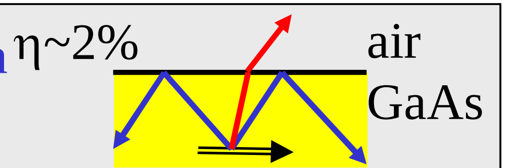
resonant interaction of emitter with a large number of modes at different emission angles

$e = \lambda_0/2\ ,\ p_0 = 1$



- resonant interaction of emitter with a single mode
- angular redistribution of the dipole emission
- enhanced extraction efficiency η~25 %

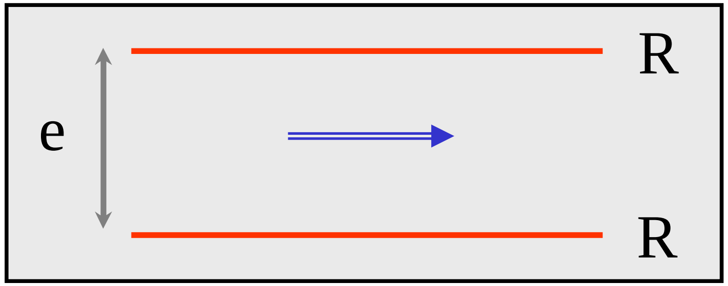
Total internal reflection for i > 13°

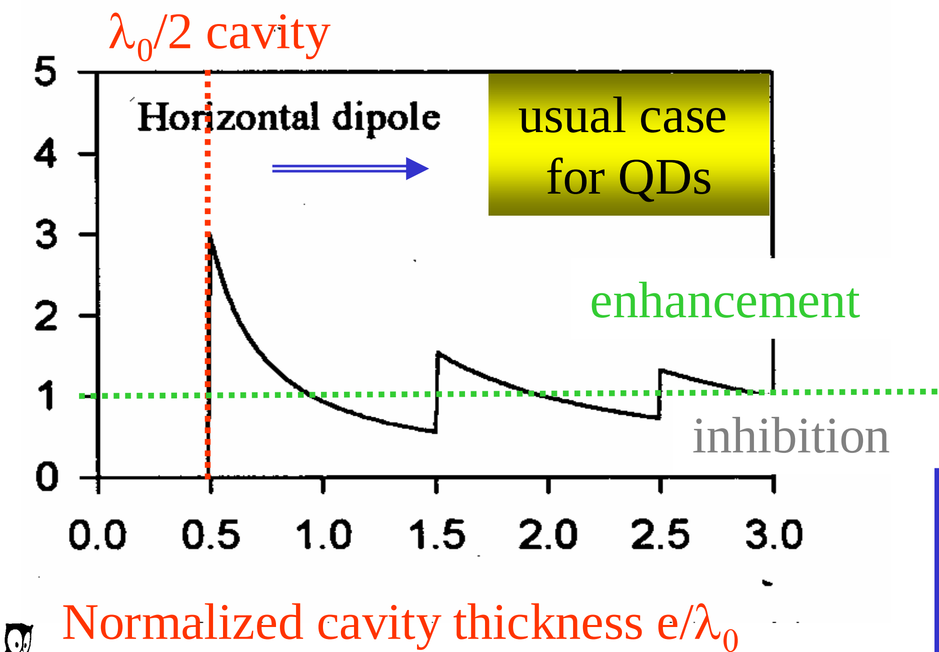


Enhancement and inhibition

Brorson, IEEE JQE **26**, 1492 (1990)

Horizontal dipole





$$\frac{1}{T_1} \sim \frac{2\pi}{\hbar} |\langle f|\widetilde{H_I}|i\rangle|^2 \rho_{e.m.}(E_{|e,h\rangle})$$

- free space $E = \hbar c k$

$$\rho_{3D}(E) = \frac{\Omega}{\pi^2} \frac{E^2}{(\hbar c)^3}$$

- $\lambda_0/2$ cavity

$$E = \hbar c \sqrt{\left(\frac{\pi}{e}\right)^2 + k_\parallel^2} \qquad [\, E_0 \,,\, 2E_0 \,]$$

$$\rho_{2D}(E) = \frac{2\Sigma}{\pi} \frac{E}{(\hbar c)^2}$$

for $\lambda_0/2$ cavity $\quad \dfrac{\rho_{2D}(E_0)/(e\Sigma)}{\rho_{3D}(E_0)/\Omega} = 2$??

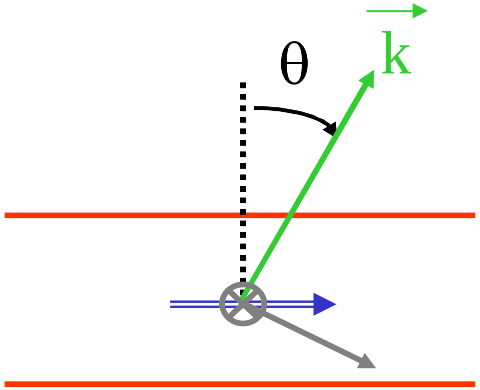
Dipole-field orientation

$$\frac{1}{T_1} = \frac{2\pi}{\hbar} \sum_{\{|1_\omega\rangle\}} |\langle f|\widetilde{H_I}|i\rangle|^2 \delta(E_{|e,h\rangle} - \hbar\omega)$$

mode-dependent
term in matrix element

$$\vec{A}.\langle u_c|\vec{p}\,|u_v\rangle = \vec{A}.\vec{P_{cv}} = AP_{cv}\cos\alpha$$

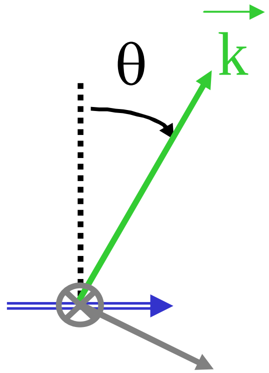
Fabry-Pérot cavity



given E → single angle θ

two modes { α= π/2, α= θ } **1/2**

Free space



given E → θ : [0, π]

two modes { α= π/2, α= θ } **1/2**

$$\int_0^\pi \cos^2\theta \sin\theta d\theta d\varphi = \frac{2}{3} d\varphi$$

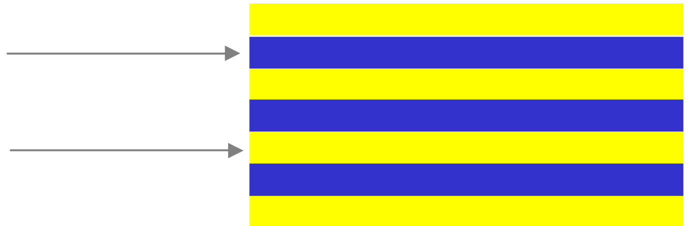
for $\lambda_0/2$ cavity $\quad \dfrac{\rho_{2D}(E_0)/(e\Sigma) * 1/2}{\rho_{3D}(E_0)/\Omega * 1/3} = 3$

Semiconductor microcavities

- metallic mirrors = lossy at optical frequencies
- epitaxially grown Distributed Bragg Reflectors

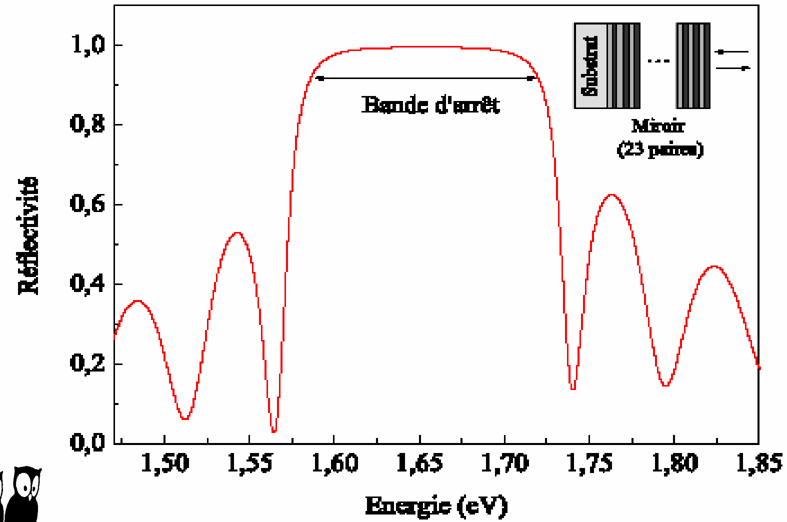
GaAs (n_1~3.54)

AlAs (n_2~2.95)



stacked pairs of GaAs/AlAs with $n_1e_1 = n_2e_2 = \lambda_0/4$

- reflectivity spectrum

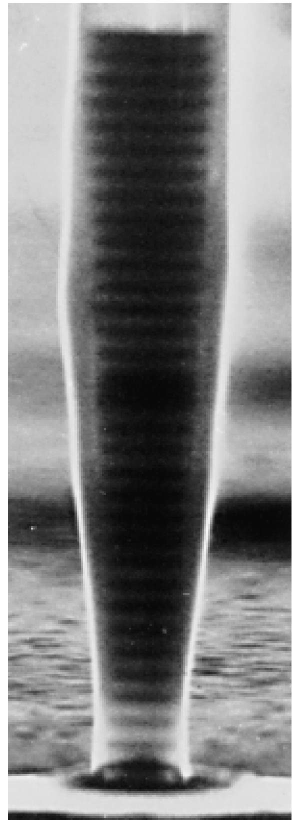


- High reflectivity

  R_{max} ~ 99.5%

  with 23 pairs
- Stop-band width ΔE

  $\Delta E/E_0 \sim (n_1-n_2)/\langle n \rangle$

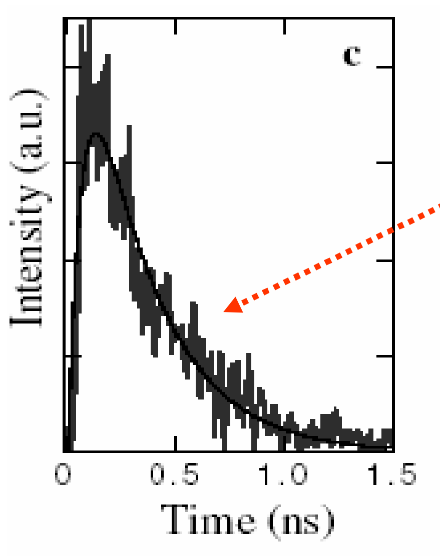


Gérard, PRL **81**, 1110 (1998)

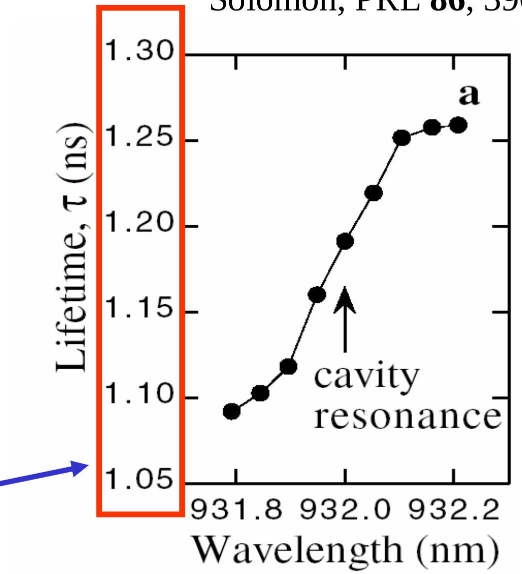
QDs in planar semiconductor microcavities

• time-resolved photoluminescence experiments

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



$$\exp\left(-\frac{t}{T_1}\right)$$

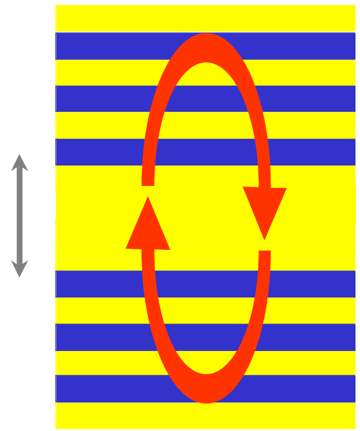


+/- 10%

• very weak effect on QD spontaneous emission dynamics !

• in DBR based-cavity, the effective cavity length ne is around $4\lambda_0$

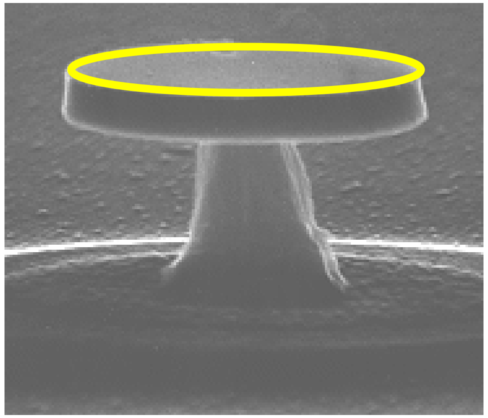
$ne=\lambda_0$



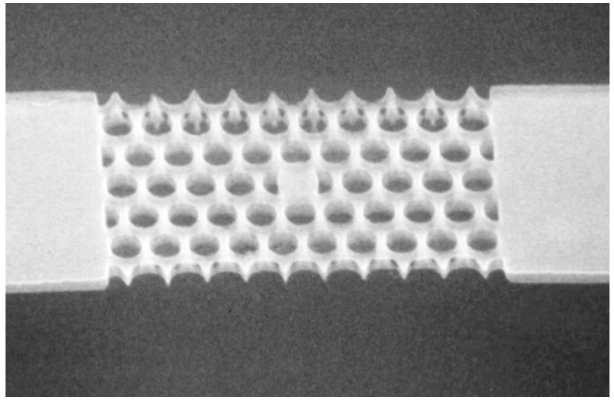
Some solid-state 0D microcavities

1 μm

2 μm



3 μm



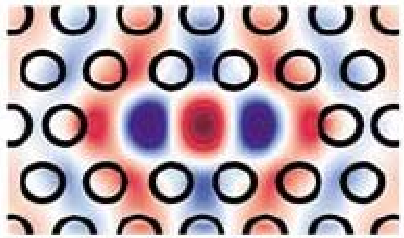
Micropillars

lateral etching of planar semiconductor DBRs

Microdisks

whispering gallery modes

Photonic crystals defects



Akahane, Nature **425**, 944 (2003)

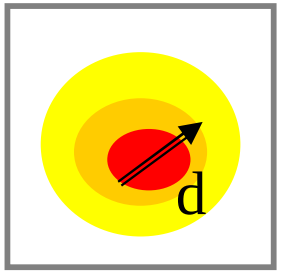
Two important figures of merit

• spatial confinement of electromagnetic field

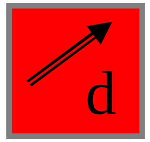
V_{eff} :

effective mode volume

$$\varepsilon_{\max} = \sqrt{\frac{\hbar\omega}{2\varepsilon_0 n^2 V_{eff}}}$$



Actual cavity



Equivalent cavity volume V_{eff}

• temporal confinement of electromagnetic field

Q :

mode quality factor

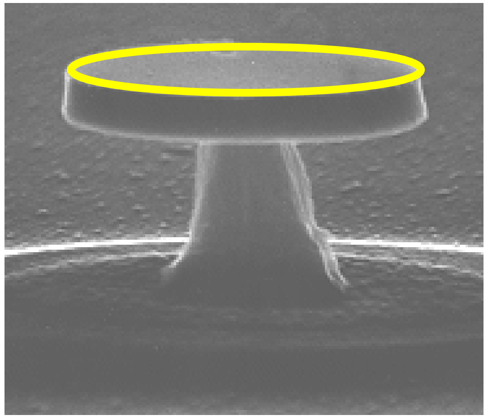
$$Q = \frac{E_0}{\Gamma_{cav}} \longrightarrow \tau_{photon} = \frac{\hbar}{\Gamma_{cav}}$$

around 1 eV, Q = 1000 ⟷ τ_{photon} =0.6 ps

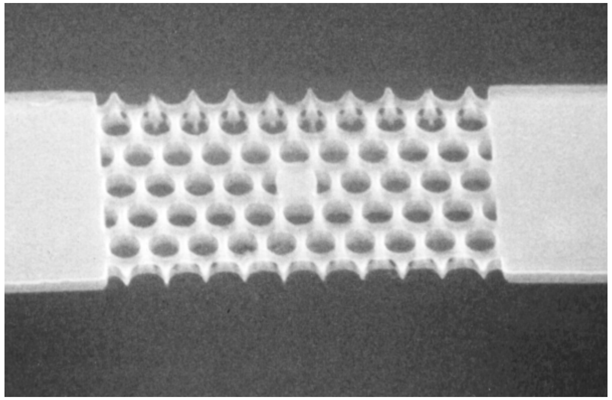
High-Q small-mode volume 0D cavities

1 µm

2 µm



3 µm

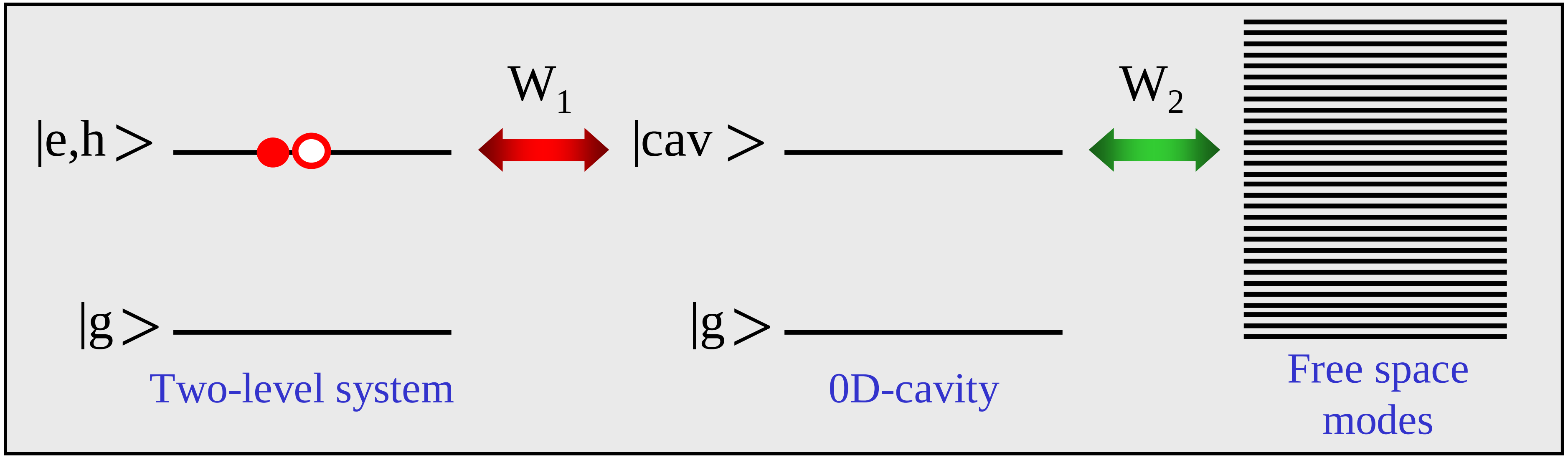


Micropillars	Microdisks	Photonic crystals defects
$V=5\ (\lambda/n)^3$	$V=6\ (\lambda/n)^3$	$V=1.2(\lambda/n)^3$
$Q \sim 7000$	$Q \sim 12000$	$Q \sim 20000$

Reithmaier, Nature **432**, 197 (2004)

Yoshie, Nature **432**, 200 (2004)

Quasi-mode approximation



• two cases : $W_1 > W_2$ strong coupling regime

$W_1 < W_2$ weak coupling regime

• in the weak coupling regime, QD is coupled to the electromagnetic field which mode density is given by :

$$\rho(E) = \frac{2Q}{\pi E_{cav}} \frac{E_{cav}^2}{4Q^2(E - E_{cav})^2 + E_{cav}^2}$$

damped 0D-cavity mode
= quasi-mode

Fano, Phys. Rev. **124**, 1866 (1961)

Purcell factor

$$\frac{1}{T_1} = \frac{2\pi}{\hbar} \sum_{\{|1_\omega\rangle\}} |\langle f|\widetilde{H_I}|i\rangle|^2 \delta(E_{|e,h\rangle} - \hbar\omega)$$

with $\vec{A}(\vec{r}) = i\sqrt{\frac{\hbar}{2\varepsilon_0 \omega V_{eff}}}(a\vec{f}^*(\vec{r}) + a^+\vec{f}(\vec{r}))$ where $\begin{cases} \left|\vec{f}(\vec{r})\right|_{max} = 1 \\ \int d^3\vec{r}\left|\vec{f}(\vec{r})\right|^2 = V_{eff} \end{cases}$

free space

$$\left(\frac{1}{T_1}\right)_{free\ space} = \frac{\Gamma}{\hbar} = \frac{1}{3}\frac{P_{cv}^2}{\varepsilon_0}\frac{E_0}{\pi\hbar^2 c^3}$$

0D cavity

- dipole at the field maximum
- dipole parallel to field
- $E_0 = E_{cav}$

$$\left(\frac{1}{T_1}\right)_{cav} = \frac{\Gamma_p}{\hbar} = 2\frac{P_{cv}^2}{\varepsilon_0}\frac{\hbar Q}{E_0^2 V_{eff}}$$

Purcell factor

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

$$F_p = \frac{\Gamma_p}{\Gamma}$$

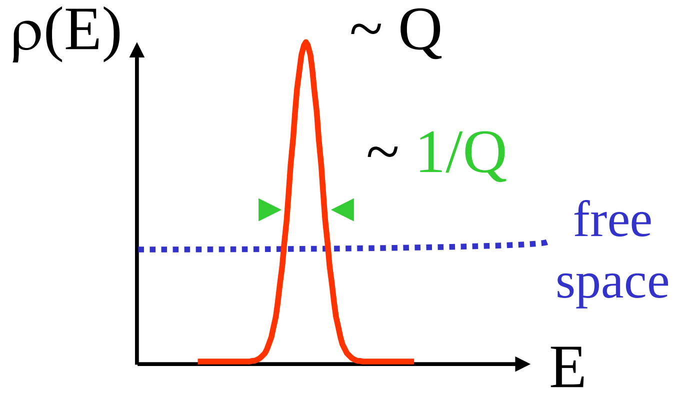
and we find

$$F_p = \frac{3}{4\pi^2}\frac{\lambda_0^3 Q}{V_{eff}}$$

Purcell effect in 0D-cavity

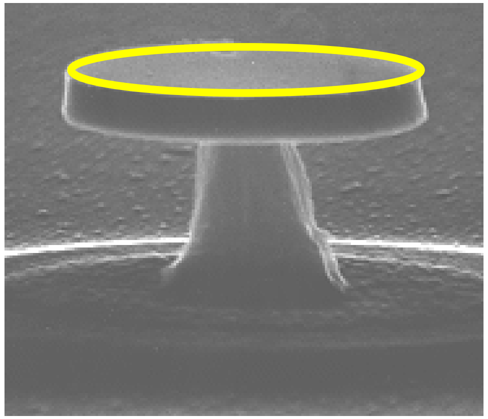
- QD = dipole in a dielectric 0D-cavity

$$F_p = \frac{3}{4\pi^2}\frac{(\lambda_0/n)^3 Q}{V_{eff}}$$

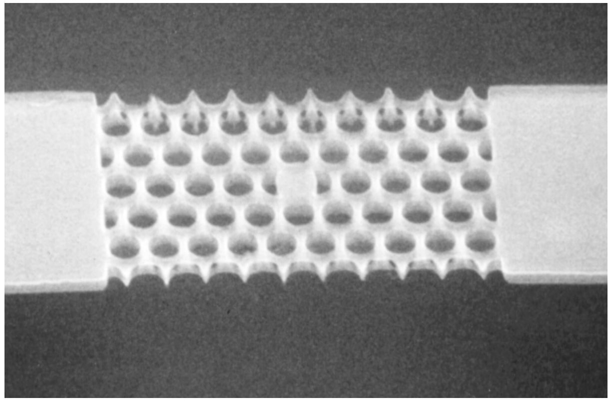


- Calculated Purcell factors

$V=5\ (\lambda_0/n)^3$
$Q \sim 7000$
$F_p \sim 100$



$V=6\ (\lambda_0/n)^3$
$Q \sim 12000$
$F_p \sim 150$



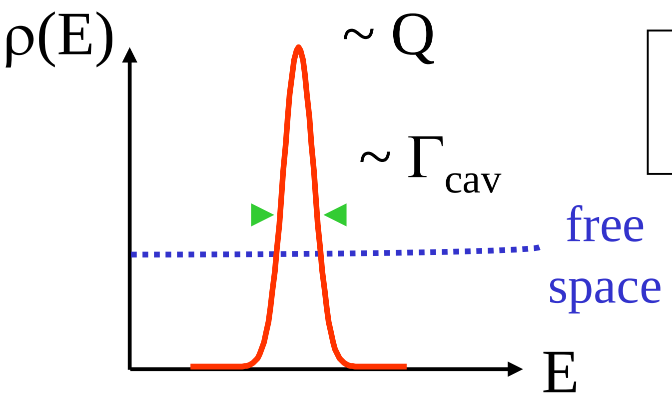
$V=1.2(\lambda_0/n)^3$
$Q \sim 20000$
$F_p \sim 800$

Purcell effect in a non-ideal case

$$F_p = F_p^0 \frac{E_{cav}^2}{4Q^2(E_0 - E_{cav})^2 + E_{cav}^2} \left| \vec{f}\,(\vec{r_0}) \right| \cos^2\alpha$$

Spectral detuning — Spatial detuning — Dipole misorientation

- broadening of the QD emission line



efficient dephasing

Γ_{QD} = QD linewidth

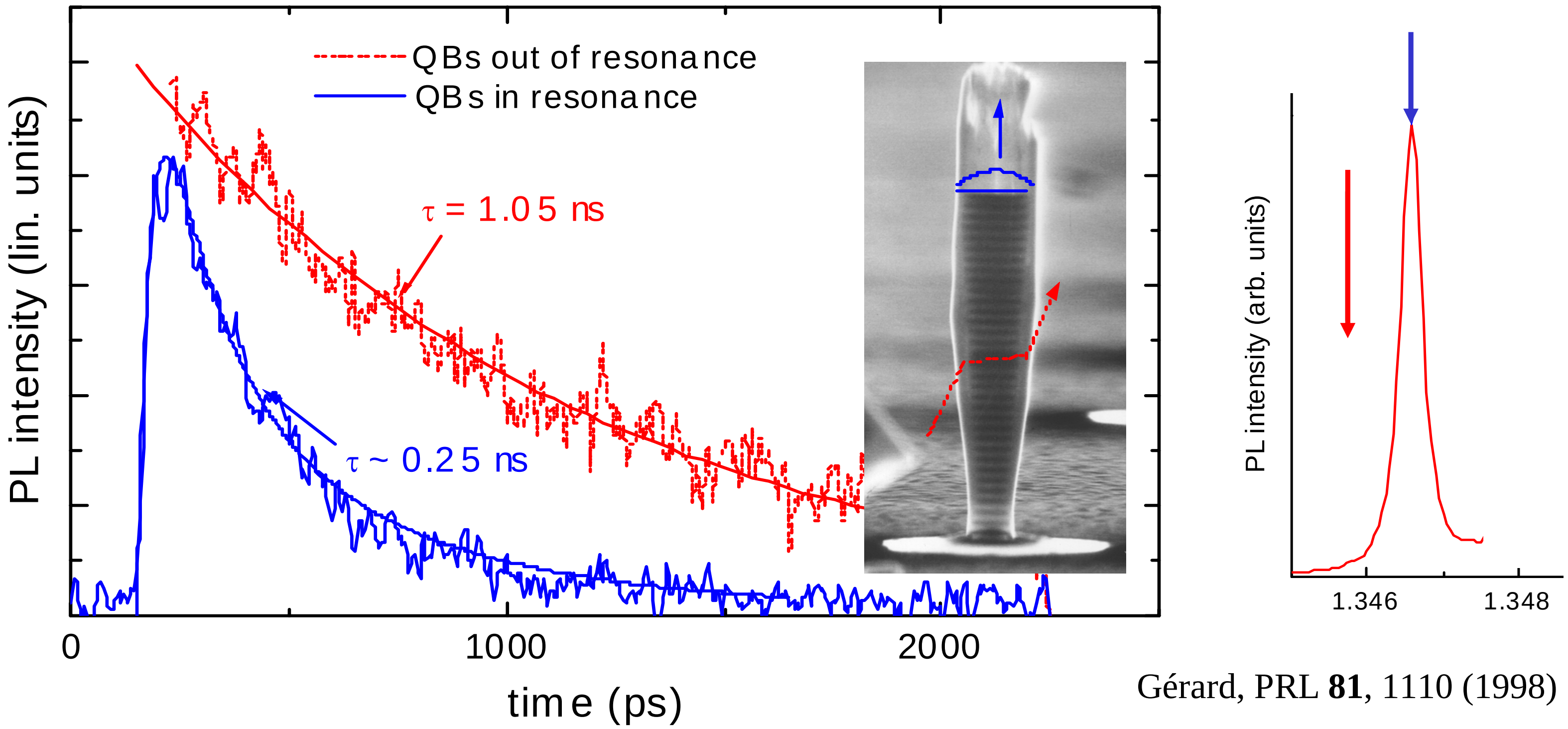
$$\rho(E) \longrightarrow \int du\,\rho(E-u)\,L_{\Gamma_{QD}}(u)$$

if $\Gamma_{QD} >> \Gamma_{cav}$,
the Purcell effect is washed out

no phonon-assisted dephasing

Purcell effect in InAs QDs

• time-resolved experiments in QD arrays : spatial + spectral averaging



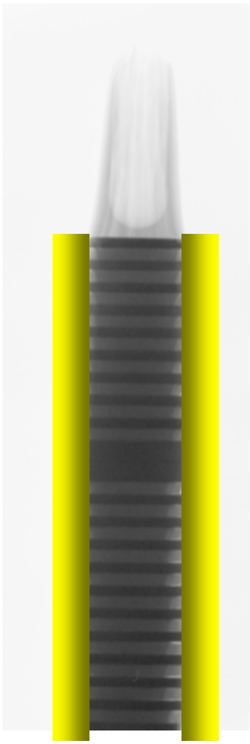
Gérard, PRL **81**, 1110 (1998)

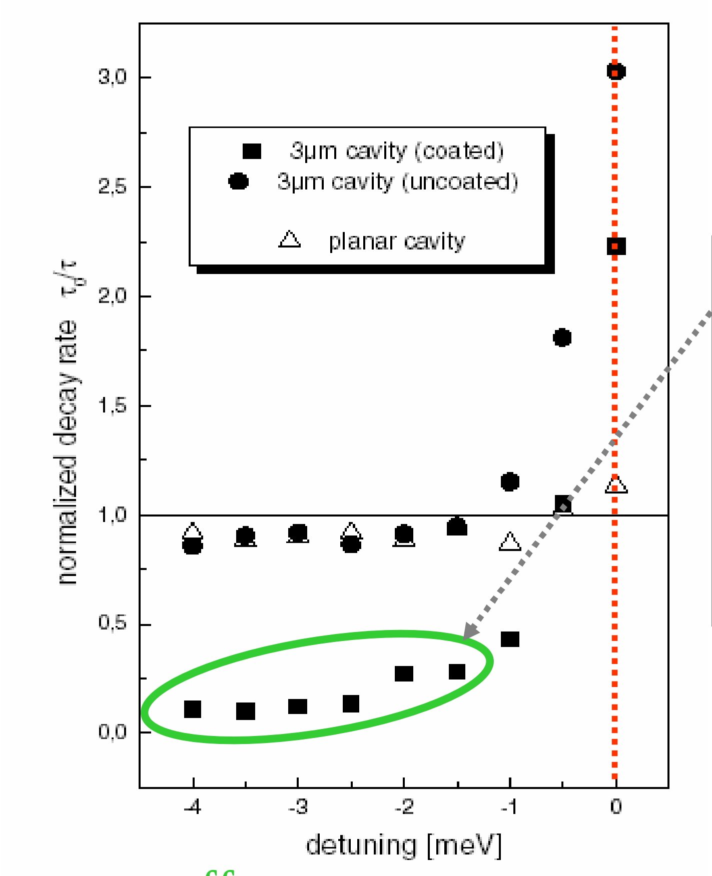
• PL decay faster only for on-resonance QDs : (x5) enhancement of spontaneous emission rate (much smaller than F_p)

• for off-resonance QDs, free-space like recombination dynamics : leaky modes

Spontaneous emission inhibition

• suppression of leaky modes in gold-coated micropillars





off-resonance QDs

on-resonance QDs

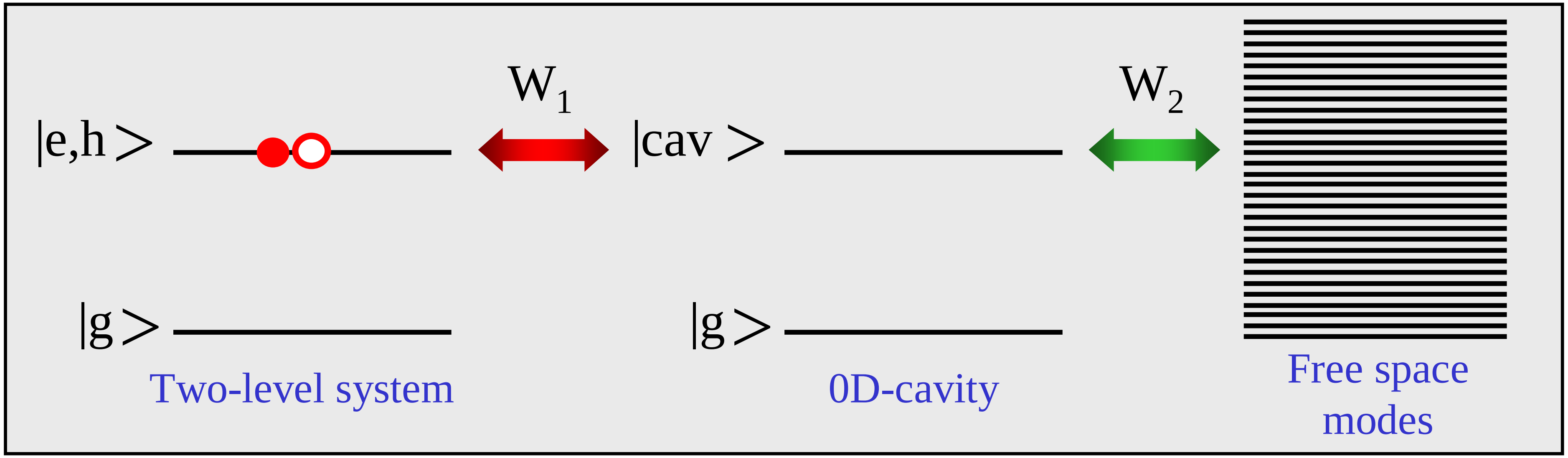
• strong inhibition (/10) of spontaneous emission rate

• efficient reduction of the density of non-resonant modes by the metallic coating

Bayer, PRL **86**, 3168 (2001)

Strong coupling regime

From weak to strong coupling



- two cases : $W_1 > W_2$ strong coupling regime
  $W_1 < W_2$ weak coupling regime

- in the strong coupling regime, the coupling to free space modes is a perturbation to (QD-cavity) sub-system

existence of mixed electron-photon states = microcavity polaritons

Microcavity polaritons

- two states of microcavity polariton

$$|-\rangle = \alpha|e,h\rangle + \beta|cav\rangle$$
$$|+\rangle = \beta|e,h\rangle - \alpha|cav\rangle$$

- anti-crossing

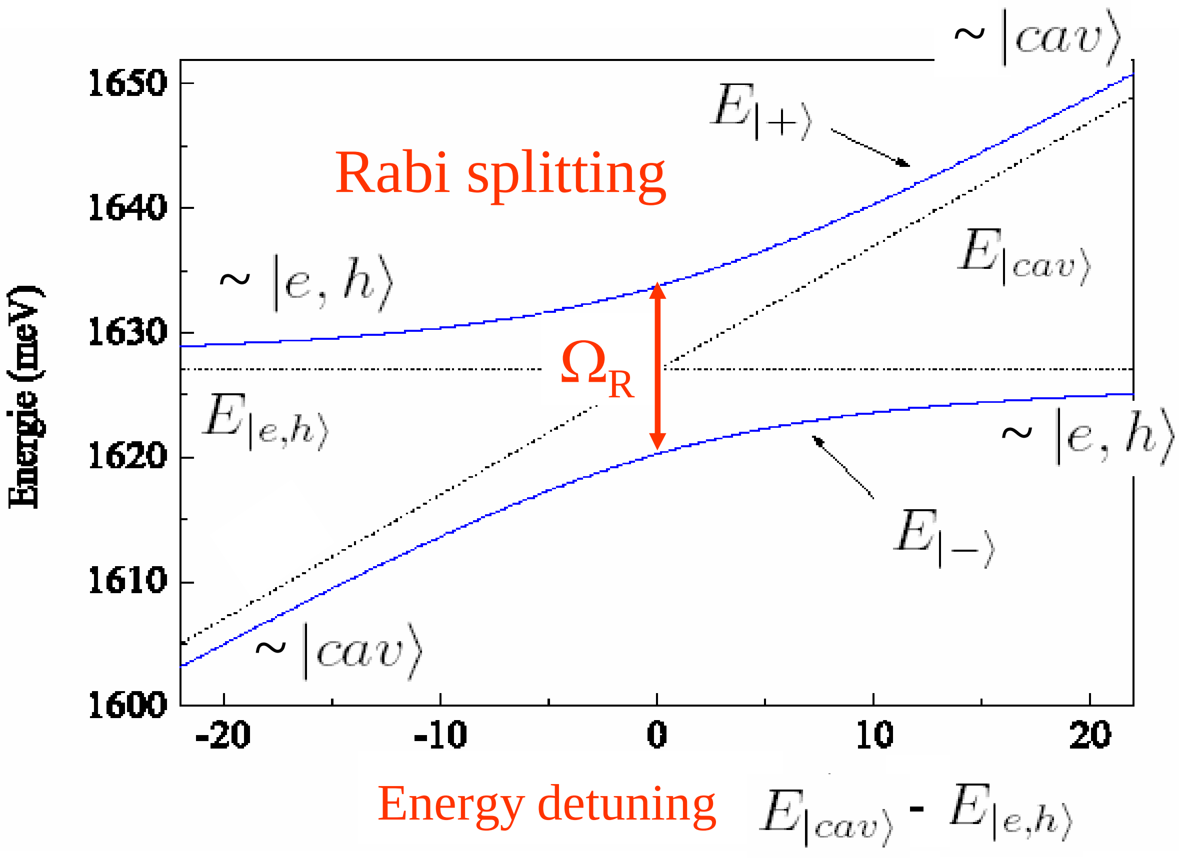
at zero detuning

$$E_{|+\rangle} - E_{|-\rangle} = \Omega_R$$

- at zero-detuning, the polaritons

are half electron
half photon

$$|\alpha|^2 = |\beta|^2 = \frac{1}{2}$$



Weak to strong coupling crossover

Andreani, PRB **60**, 13276 (1999)

$$\Omega_{\pm} = \omega_0 - \frac{i}{4}(\gamma_a + \gamma_{c,\mu}) \pm \sqrt{g^2 - \left(\frac{\gamma_a - \gamma_{c,\mu}}{4}\right)^2}$$

$$\hbar g \propto |\langle \vec{p}.\vec{A}\rangle| \propto \frac{1}{\sqrt{V_{eff}}}$$

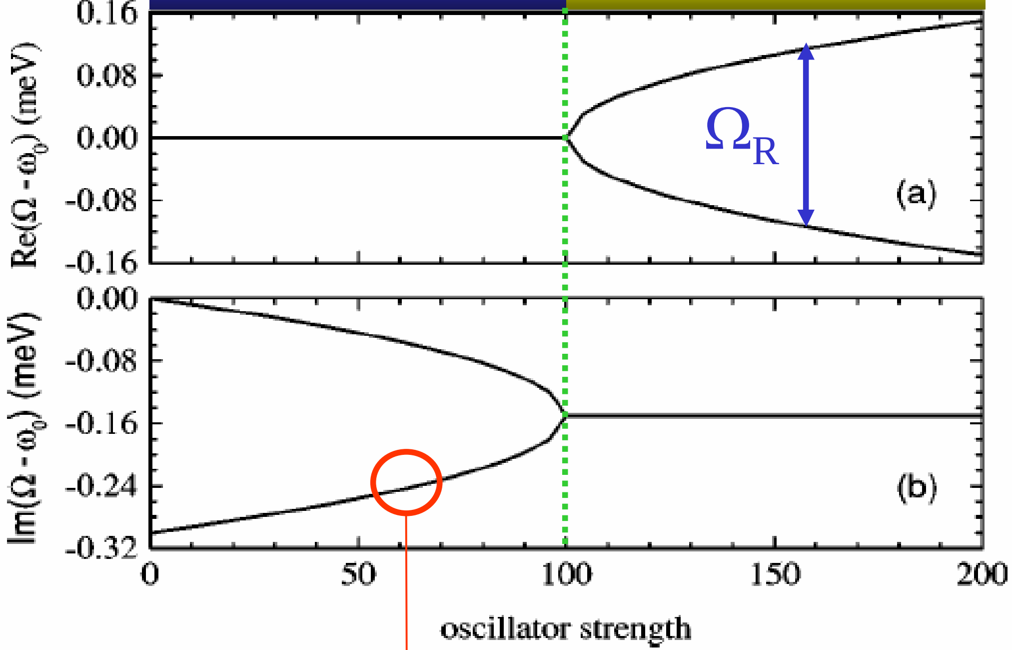
$$\hbar\gamma_a = \Gamma_{QD}$$

$$\hbar\gamma_{c,\mu} = \Gamma_{cav} \propto \frac{1}{Q}$$

weak coupling | strong coupling

energy position

linewidth



$\Gamma_p = F_p \Gamma$, modified spontaneous emission rate (Purcell effect)

weak coupling

$$g \ll |\gamma_a - \gamma_{c,\mu}|$$

$$g \ll \gamma_{c,\mu}$$

high V_{eff} small Q

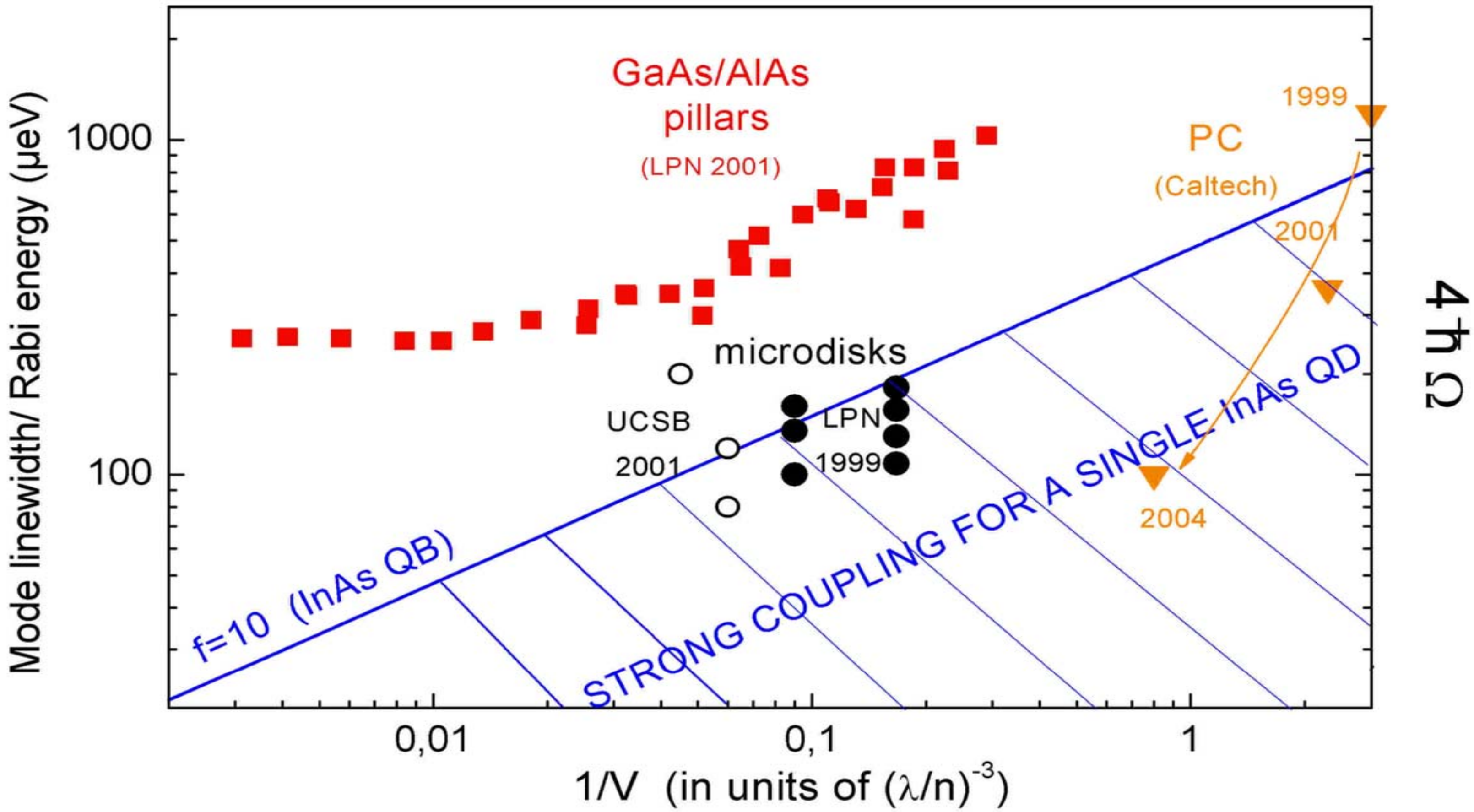
strong coupling

$$g \gg |\gamma_a - \gamma_{c,\mu}|$$

$$g \gg \gamma_{c,\mu}$$

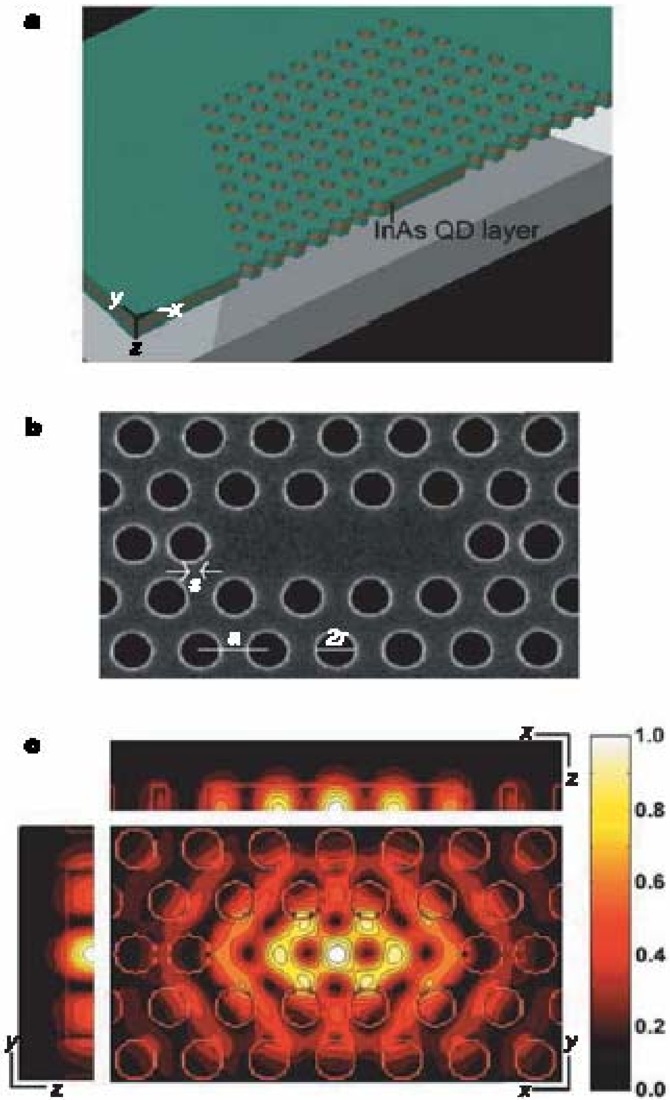
small V_{eff} high Q

Towards strong coupling for a single QD

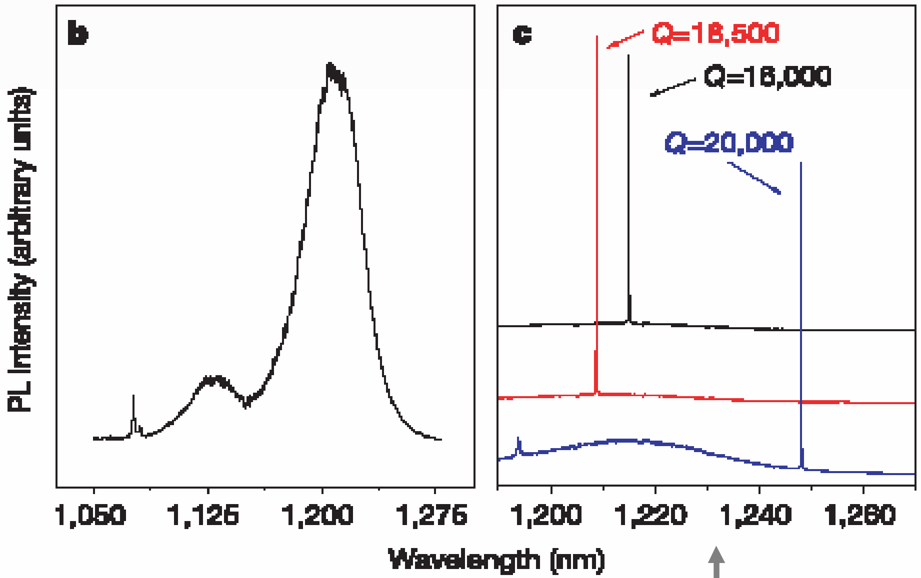


For a single InAs QD, strong coupling already observable with microdisks and PC cavities

Photonic crystals high-Q microcavities



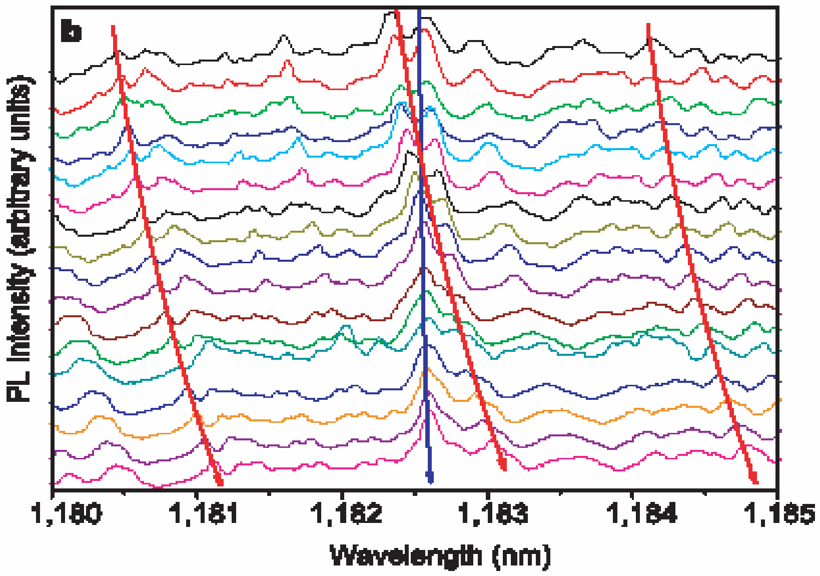
Yoshie, Nature **432**, 200 (2004)

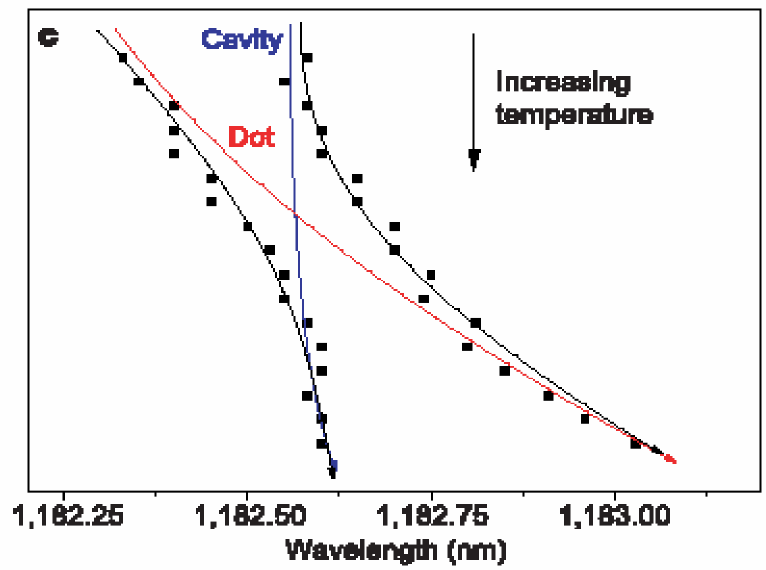


three different
cavities of high Q

Strong coupling for a single InAs QD

Yoshie, Nature **432**, 200 (2004)





- evidence for a single InAs QD in the strong coupling regime : Ω_R ~ 170 μeV

- strong coupling also obtained (simultaneously !!) for
  - a single InGaAs QD in micropillar — Reithmaier, Nature **432**, 197 (2004)
  - a single GaAs QD in microdisk — Peter (LPN-France), submitted to PRL