Heterostructure Epitaxy for fondamental research

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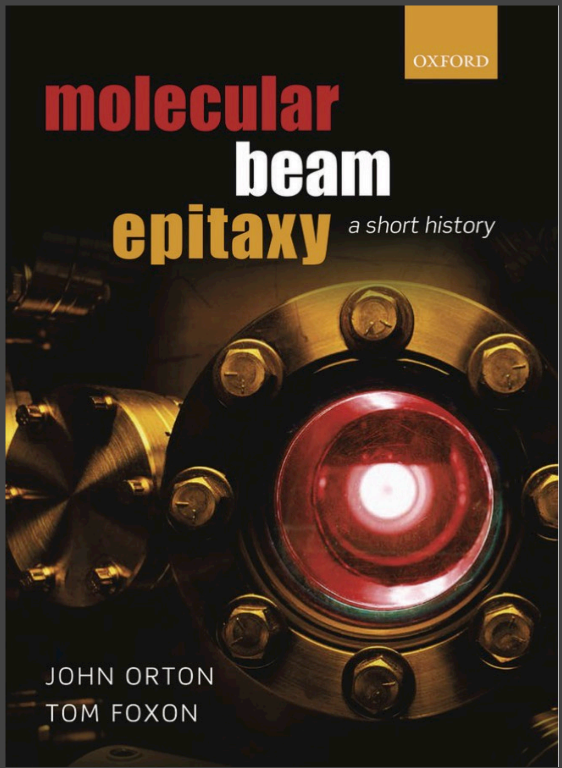
Outline

1. A bit of history
   Electron mean-free path
2. A few experiments on mesosopic transport
   Phase coherence
3. Light-Matter interaction
   Exciton, polariton, quantum light

A bit of History

a "Chicken or the Egg" question !

Fundamental research drove MBE development
or
MBE development drove new fundamental research?

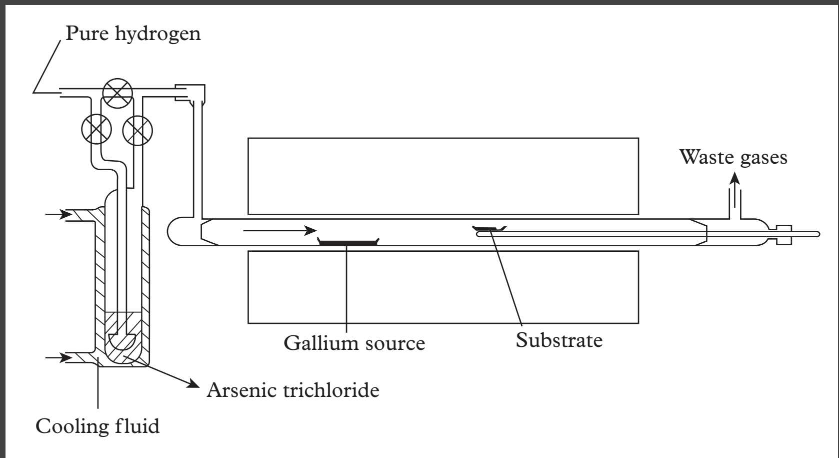


Early epitaxial techniques

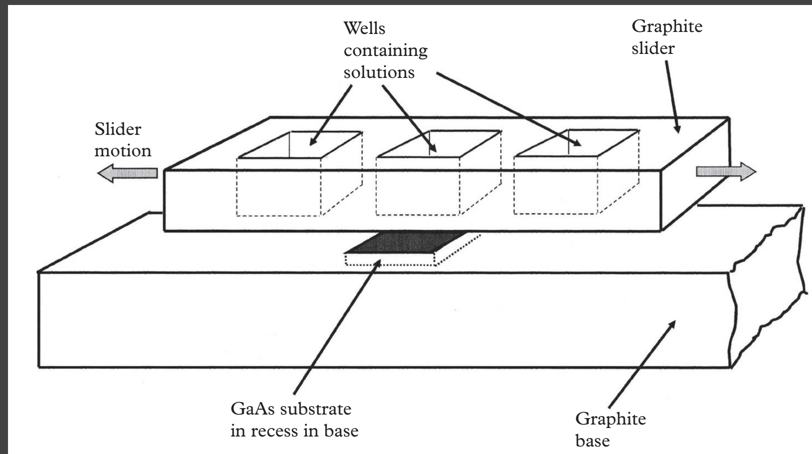
50's: tremendous improvement in single crystal quality

60's: first epitaxial techniques

VPE



LPE



VPE : mostly homoepitaxy, with dopant control (transistors, Gunn diode)
LPE : alloy epitaxy (AlGaAs), and then first heterostructures (lasers)

Very poor control of the layer thickness (~µm)

The Esaki and Tsu proposal in 1970

Superlattice and Negative Differential Conductivity in Semiconductors*

Abstract: We consider a one-dimensional periodic potential, or "superlattice," in monocrystalline semiconductors formed by a periodic variation of alloy composition or of impurity density introduced during epitaxial growth. If the period of a superlattice, of the order of 100Å, is shorter than the electron mean free path, a series of narrow allowed and forbidden bands is expected due to the subdivision of the Brillouin zone into a series of minizones. If the scattering time of electrons meets a threshold condition, the combined effect of the narrow energy band and the narrow wave-vector zone makes it possible for electrons to be excited with moderate electric fields to an energy and momentum beyond an inflection point in the *E*-*k* relation; this results in a negative differential conductance in the direction of the superlattice. The study of superlattices and observations of quantum mechanical effects on a new physical scale may provide a valuable area of investigation in the field of semiconductors.

IBM J Res Dev 14, 61 (70)

The Esaki and Tsu proposal

Entering the quantum regime

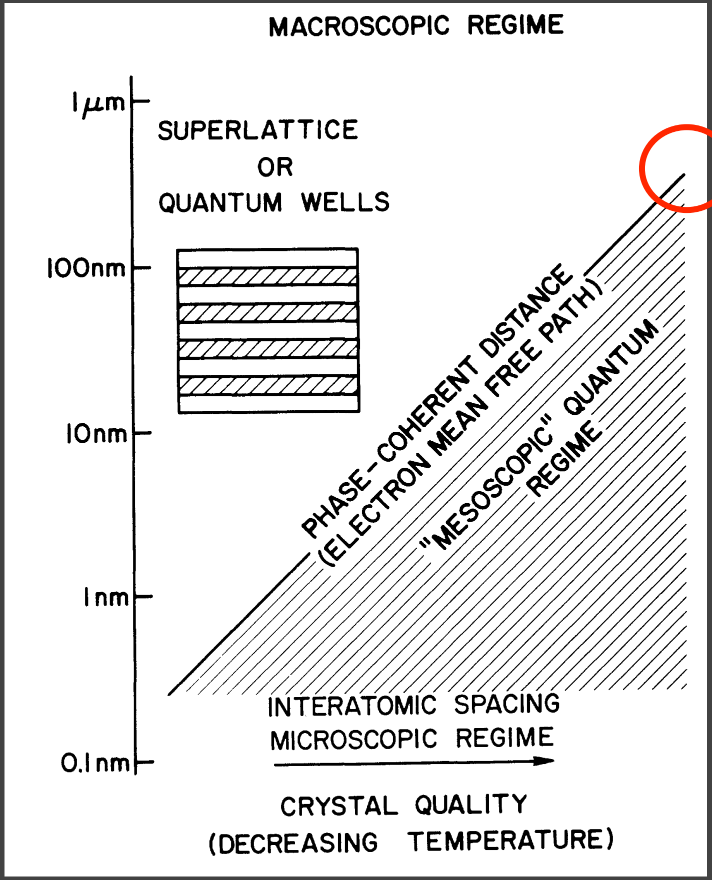
Building heterostructures with characteristic size lower than the electron mean-free path λ

$$\lambda = \tau v$$

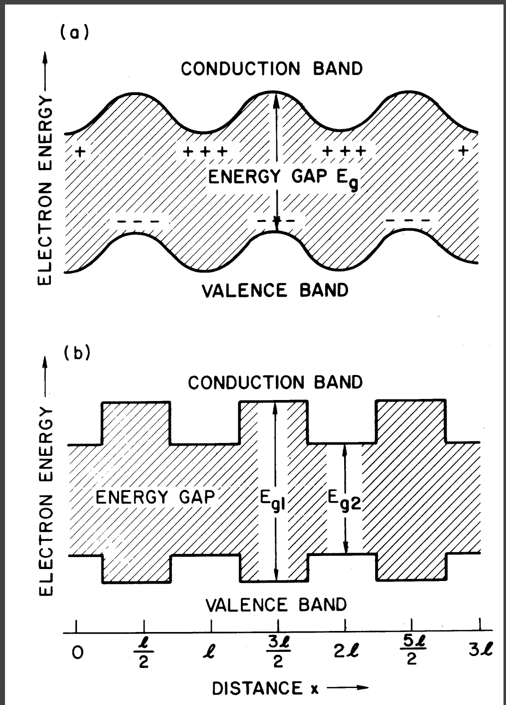
τ : scattering time
v : (thermal or Fermi) velocity

The wavefunction phase is well-defined:

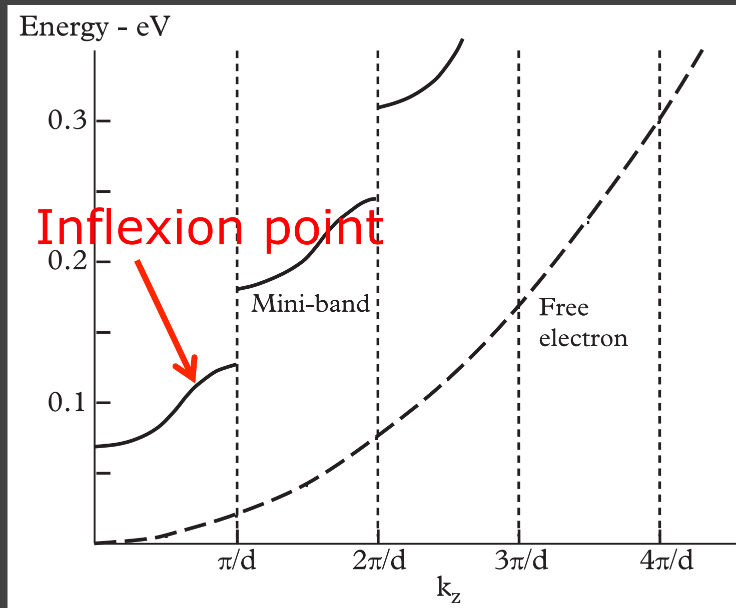
- For times shorter than τ
- For lengths shorter than λ



Electron transport in superlattices



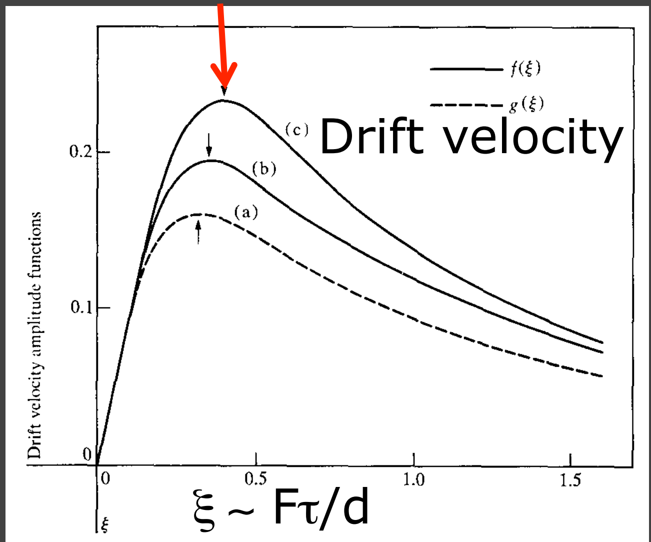
Energy dispersion



At room T
For μ=2500 cm²/Vs
λ ~ 50 nm

F electric field

$$\hbar \dot{k}_z = eF$$



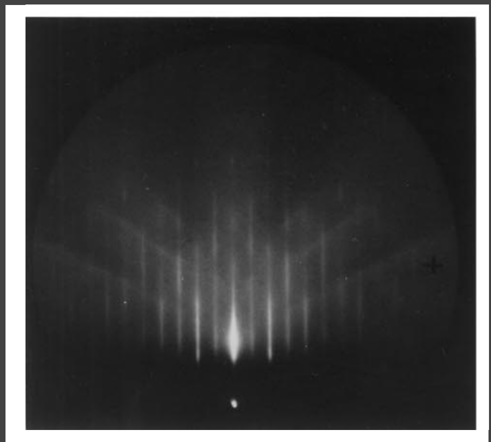
Materials

The achievement of a well-defined superlattice structure with a period of, say, 100Å will require considerable effort even with the use of the most advanced epitaxial thin-film technologies. The materials should be well-known semiconductors and their alloys; for examples, Ge, Si, Ge-Si alloys, III–V compounds and their alloys, II–VI com-

At that time…

Surface Physics

- Atomic adsorption studies
- HEED/LEED
- UHV
- Substrate preparation

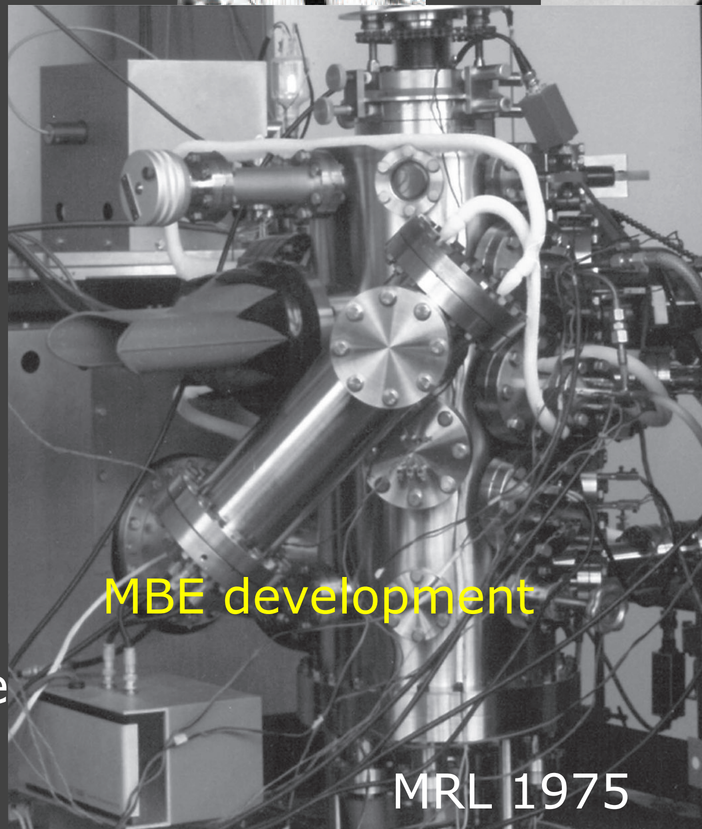


HEED GaAs substrate

Spatial technologies

- Development of effusion cells

for experiments on ion propulsion



MBE development

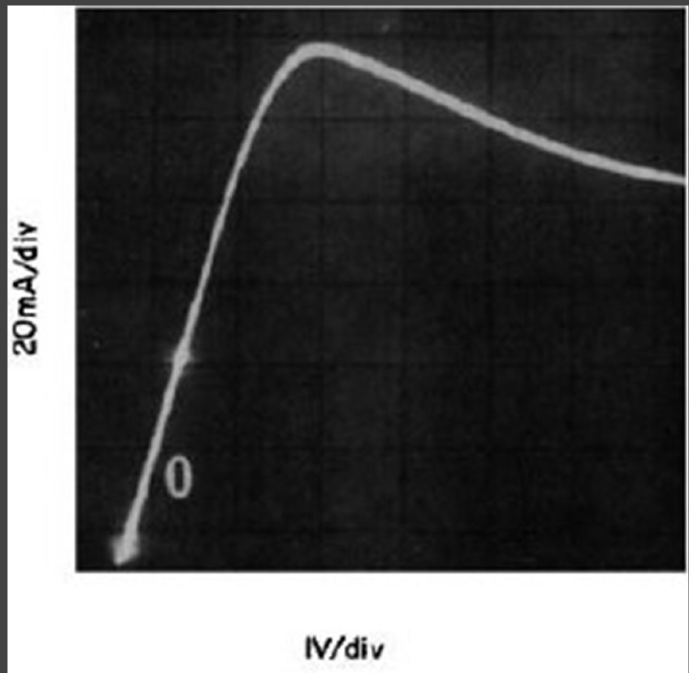
MRL 1975

yoshroud
mperature controller
ock

1972: Esaki and Chang

Esaki and Chang built their own system at IBM

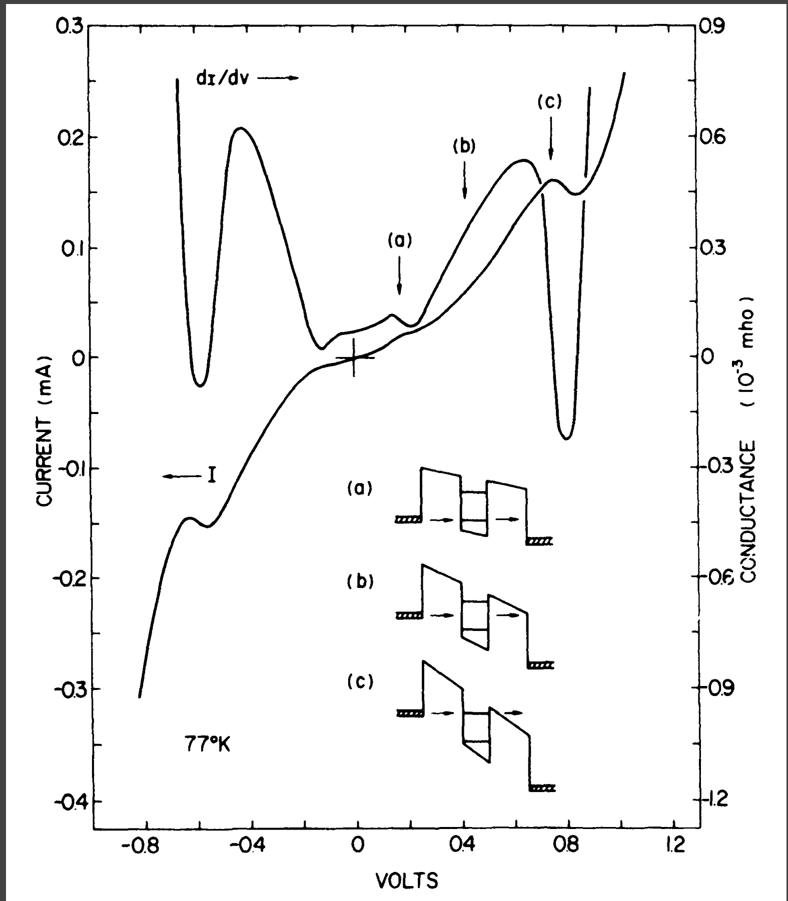
Measurement of negative resistance in AlGaAs/GaAs SL (70 Å period)



A measurable quantum effect !

1974 : Resonant tunneling in double-barriers

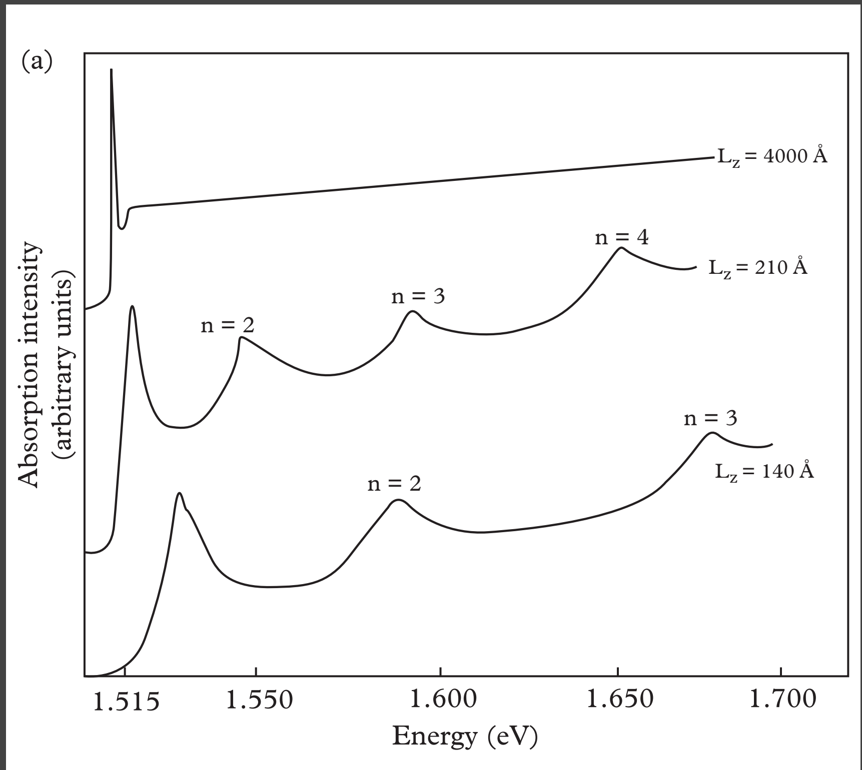
Chang, Esaki and Tsu



1974 Optical properties of QWs

At Bell Labs

Absorption AlGaAs/GaAs QW



Dingle et al. PRL

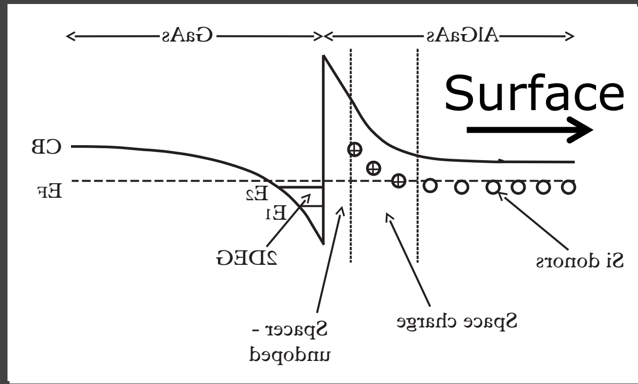
Direct visualization of quantum effect due to confinement

2D electron gas

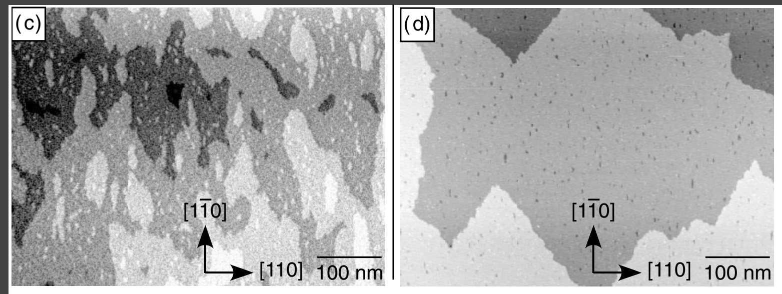
In 1978, μ ~10^4 cm^2/V.s

A long series of improvement

- High purity sources
- Remote doping (1981)
  spacer layer thickness



- Inverted interface (1981)



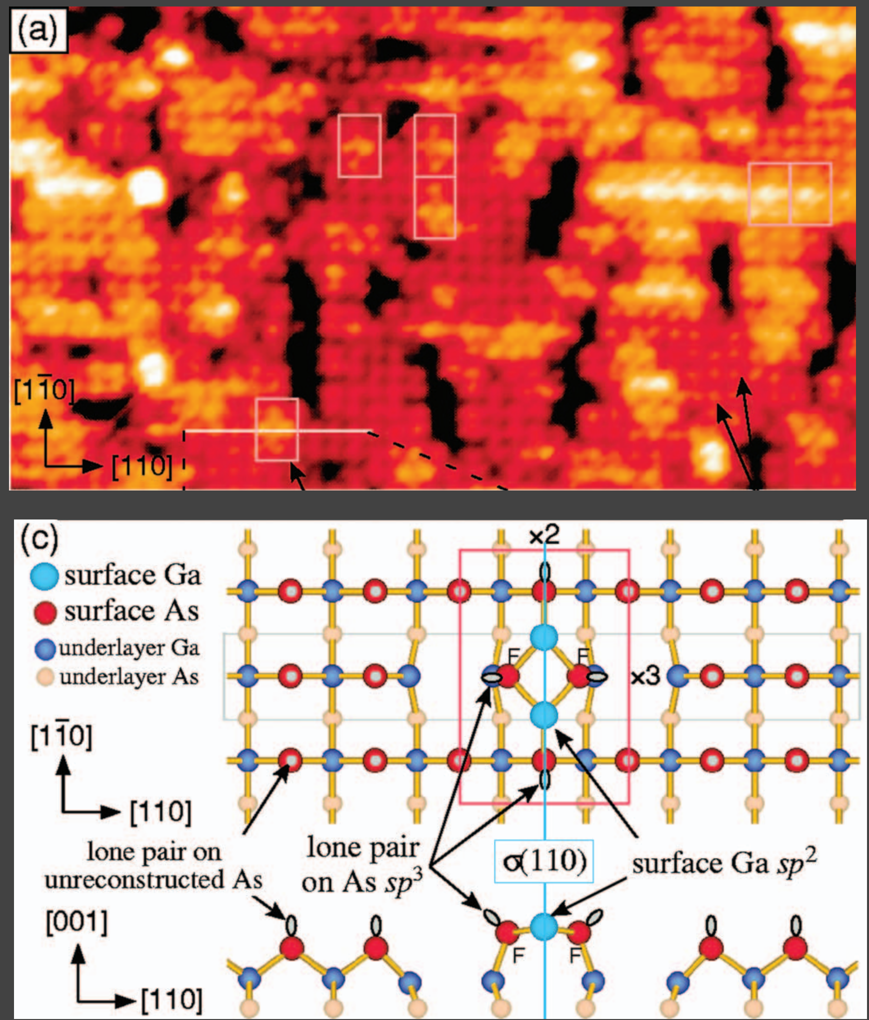
- Short GaAs/AlAs SL (“super réseau poubelle”) (1984)
- Planar doping (1980)
- Double planar doping (1987)
- Temperature and As pressure control (1985)

3x1 RHEED surface reconstruction

Obtained for As/Ga ratio close to 1

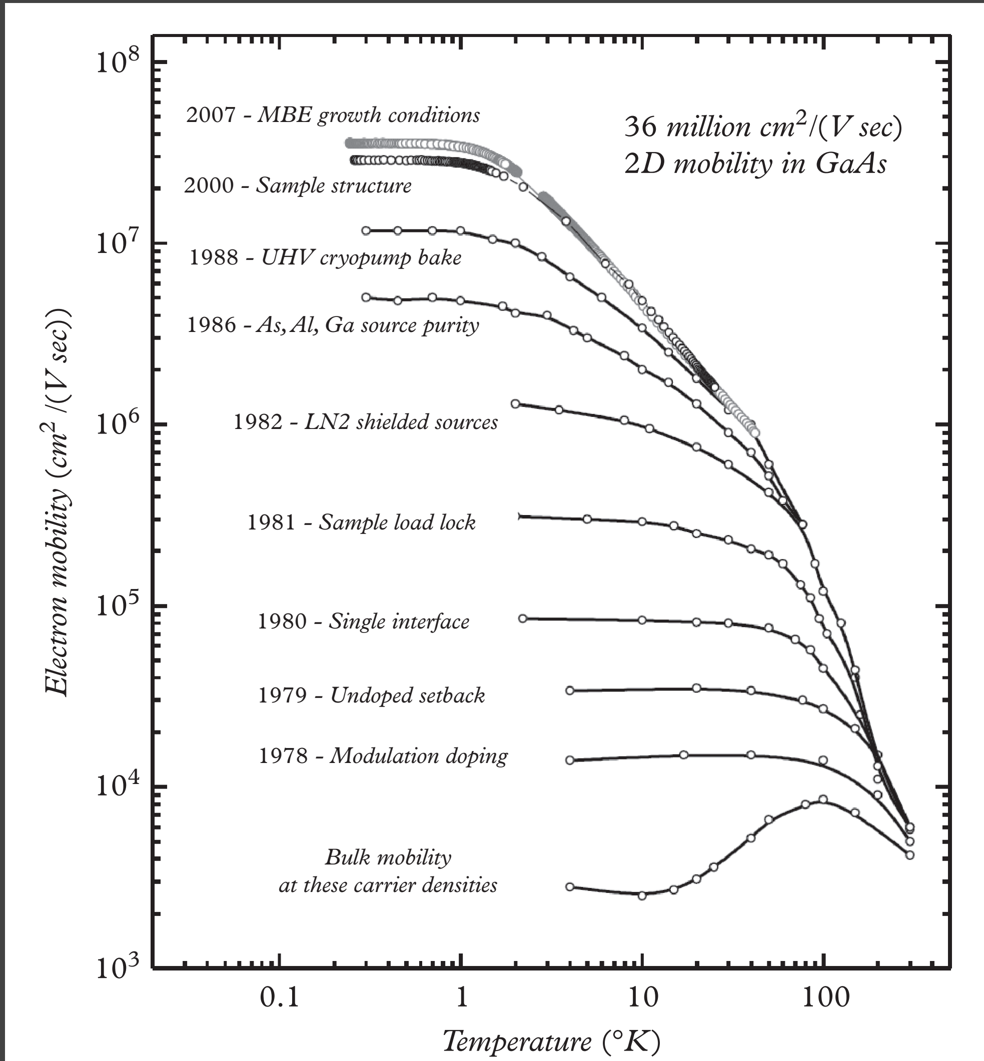
Martrou PRB 2005

Mixed surface:
(1x1) cells 45%
(3x2) cells 39%
others



27% decrease of the sp^2 reactive Ga sites compared to β2(2x4)

Less impurity incorporation



Schlom and Pfeiffer 2010

10^7 m^2/V.s

(Mobility) mean free path

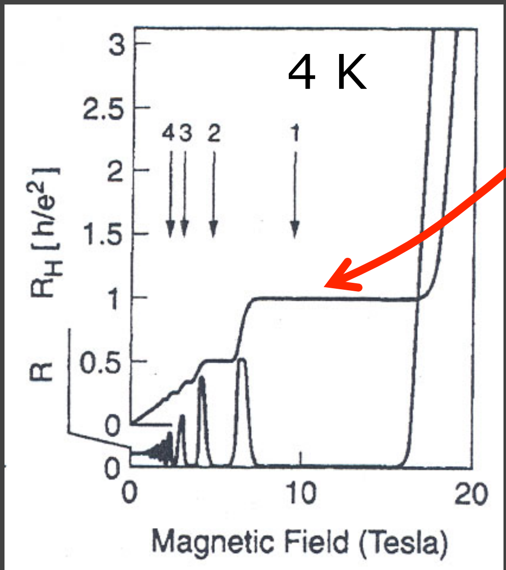
λ ~150 µm !

Mesoscopic experiments

- in-plane transport
- over very large distances

The fractional quantum Hall effect

Integer quantum Hall effect: a single particle effect

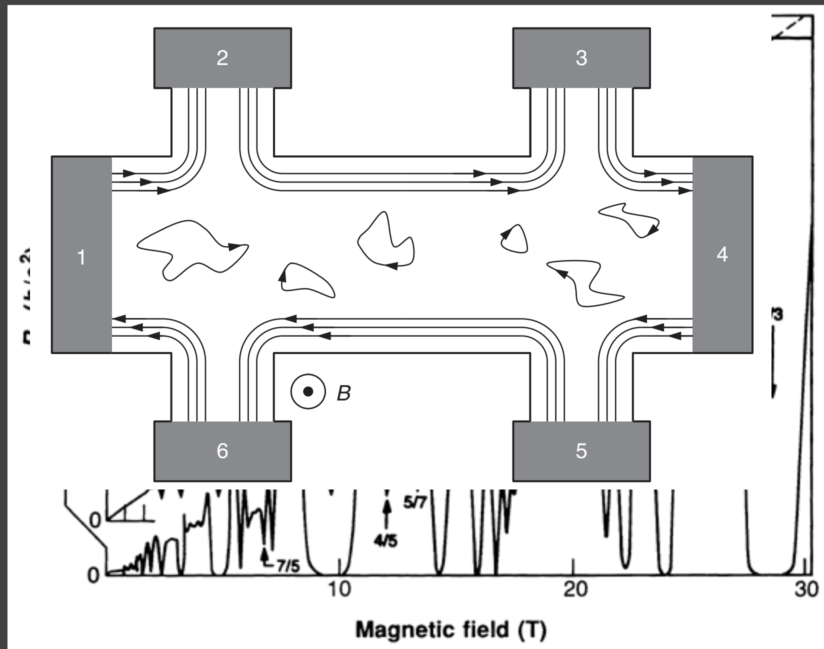


Klitzing et al (1980)
Si/SiO_2 MOSFET
(rough interface)

Hall plateaus
- Quantized resistance $R_H = h/e^2 \sim 25 k\Omega$
- Landau levels
- Edge modes
- involve impurities

The one that "only provided the sample"

Störmer, Tsui, Gossard (1982)
AlGaAs/GaAs high mobility 2DEG



FQHE : a collective effect

New composite particles with fractional charge: e/3, e/5, e/7…

1 electron + 2, 3,… magnetic flux quanta

Mesoscopic transport

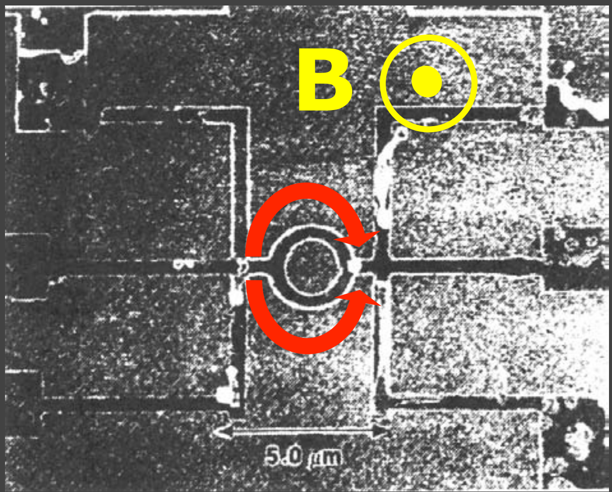
Aharonov-Bohm effect

Mankiewich et al. (88)

Interference effect

Phase difference between the two paths

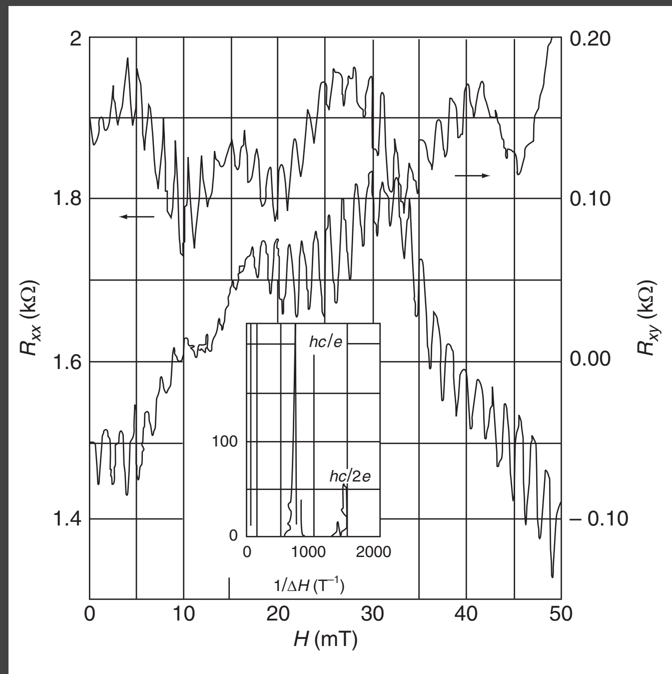
$$\delta\varphi = \frac{e}{\hbar}\int_0^{2\pi} \mathbf{A}\cdot\mathbf{a}_\vartheta r d\vartheta = \frac{e}{\hbar}\int_{ring} \mathbf{B}\cdot\mathbf{n}dS = 2\pi\frac{\Phi}{\Phi_0},$$



AlGaAs/GaAs ring

$$\Phi_0 = h/e$$ Quantum flux

Magneto-resistance oscillations



B induces a magnetic flux Φ through the loop.

Electrons pick up a phase

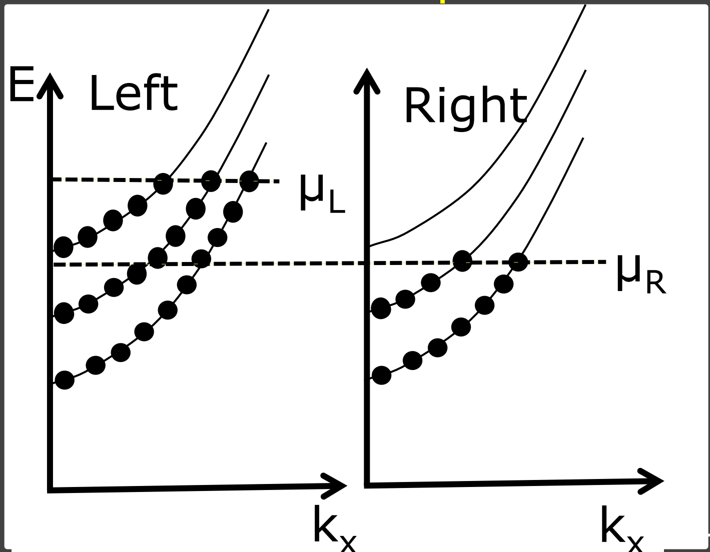
$$\varphi = \varphi_0 + \frac{1}{\hbar}(\mathbf{p} + e\mathbf{A})\cdot\mathbf{r},$$

A: potential vector

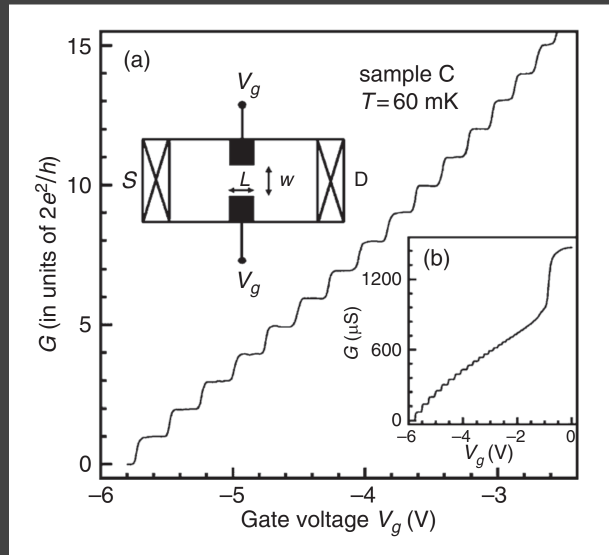
Quantum point contact

Van Wees et al. 1988
Wharam et al. 1988

Ballistic transport in 1D



Thomas et al. PRL 96



Constriction

1D modes (or channels)

$$E_n = \left[n + \frac{1}{2}\right]\hbar\omega_0 + \frac{\hbar^2 k_x^2}{2m^*}, \quad n = 1, 2, 3, \ldots$$

1D State density
$$D_n(E) = \frac{2}{\pi}\frac{\partial k_x}{\partial E}$$

Group velocity
$$v_n(E) = \frac{1}{\hbar}\frac{\partial E}{\partial k_x}$$

For a perfect conductor (T=1)

$$I = e \times \sum_{n \text{ occ}} \int_{\mu_D}^{\mu_S} v_n(E) D_n(E)\, dE$$

$$= e \times \sum_{n \text{ occ}} \frac{2}{h} \int_{\mu_D}^{\mu_S} dE$$

$$= e \times n \times \frac{2}{h} \times eV$$

At T=0K

$$G = n\frac{2e^2}{h} \approx n \times \frac{2}{25.8\,\mathrm{k}\Omega}$$

Light-matter interaction in semiconductor heterostructures

The dielectric constant of semiconductors

$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$

D: electric displacement field
P: polarization density or electric dipole moment per unit volume

In a linear, homogeneous, isotropic dielectric medium

$\mathbf{P} = \varepsilon_0 \chi \mathbf{E}$ χ: Electric susceptibility

$\mathbf{D} = \varepsilon_0 (1+\chi) \mathbf{E} = \varepsilon_0 \varepsilon_r \mathbf{E}$ ε_r : relative permittivity or dielectric constant

$\tilde{n} = \sqrt{\varepsilon_r}$ Complex refractive index

To interact with matter, light (frequency ω) has to couple to oscillators

$$\varepsilon_r(\omega) = 1 + \sum_j \frac{f_j}{\omega_{0j}^2 - \omega^2 - i\omega\gamma_j}$$

Oscillator *j*

f_j : oscillator strength
ω_{0j}: frequency
γ_j: damping constant

Oscillators in semiconductors that couple to e.m. field

Excitons

Optical properties close to the gap
NIR, visible, UV

Phonons

- Optical properties (optical phonon) in the IR range
- Raman (optical phonon) and
  Brillouin (acoustic phonon) scattering

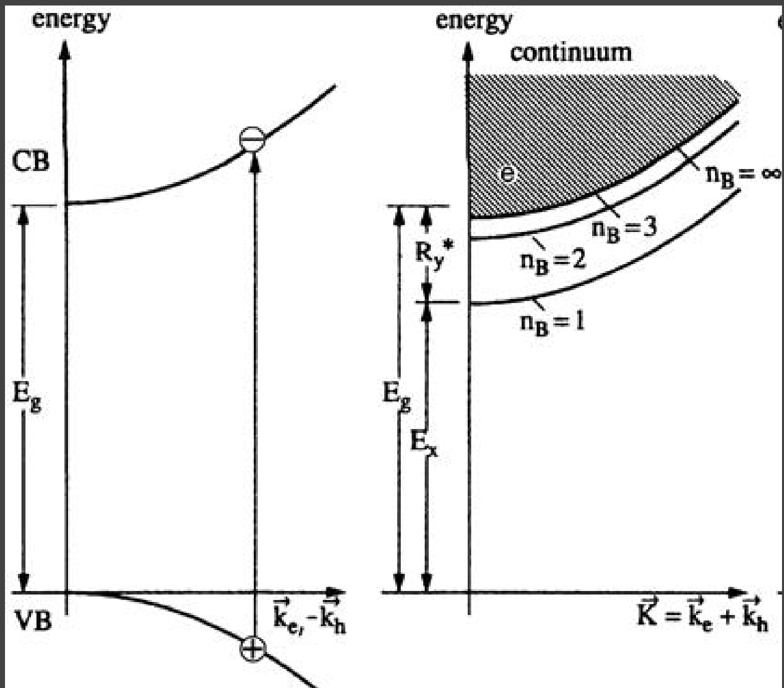
Plasmons

In highly doped structures
in the IR range (semiconductors)

Exciton

Hydrogen-like problem

Coulomb interaction between
an electron and a hole



$$E_{\text{ex}}(n_{\text{B}}, \boldsymbol{K}) = E_{\text{g}} - \text{Ry}^* \frac{1}{n_{\text{B}}^2} + \frac{\hbar^2 \boldsymbol{K}^2}{2M}$$

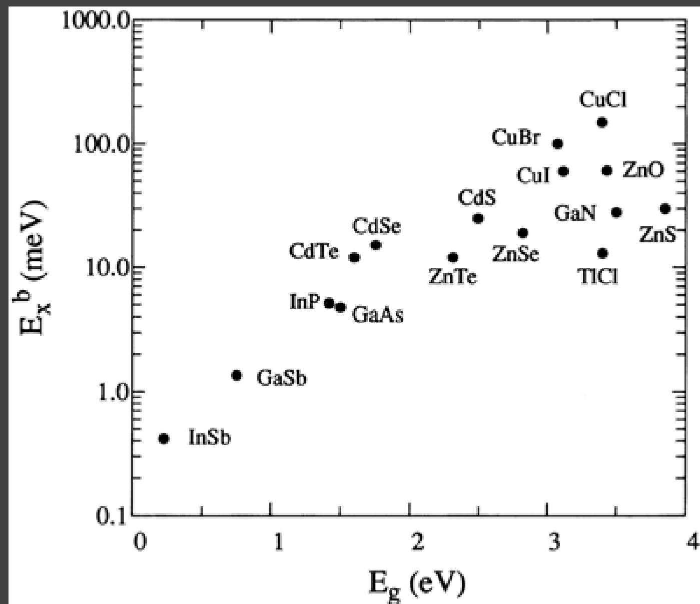
Exciton translational mass
and wave vector

$$M = m_{\text{e}} + m_{\text{h}}, \quad \boldsymbol{K} = \boldsymbol{k}_{\text{e}} + \boldsymbol{k}_{\text{h}}$$

Exciton binding energy

$$\text{Ry}^* = 13.6\,\text{eV} \frac{\mu}{m_0} \frac{1}{\varepsilon^2}$$

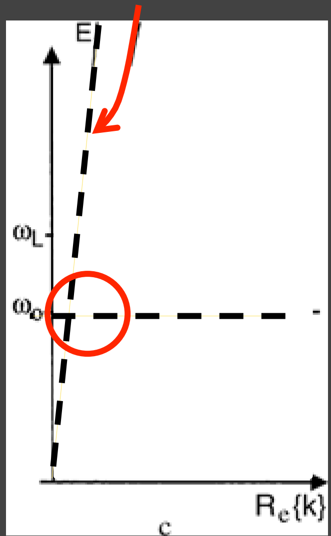
$$\mu = \frac{m_{\text{e}}\, m_{\text{h}}}{m_{\text{e}} + m_{\text{h}}}$$



Polariton : mixed light matter particle

Propagating light ($\hbar\omega$, **k**) close to an oscillator resonance (ω_0, **K**)

Light cone



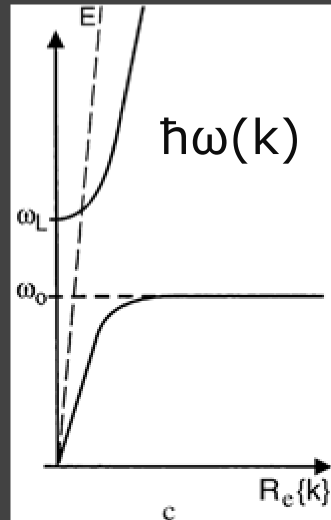
Oscillator

$$\varepsilon_r(\omega) = \varepsilon_b + \frac{f}{\omega_0^2 - \omega^2 - i\omega\gamma}$$

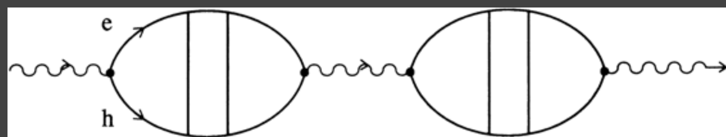
$$k^2 = [n(\omega)]^2 \left(\frac{\omega}{c}\right)^2 = \varepsilon_r(\omega)\left(\frac{\omega}{c}\right)^2$$

$$\frac{c^2k^2}{\omega^2} = \varepsilon_\mathrm{b} + \frac{f}{\omega_0^2 - \omega^2 + \mathrm{i}\omega\gamma}.$$

Polariton dispersion



Strong coupling regime: mixed photon-matter state

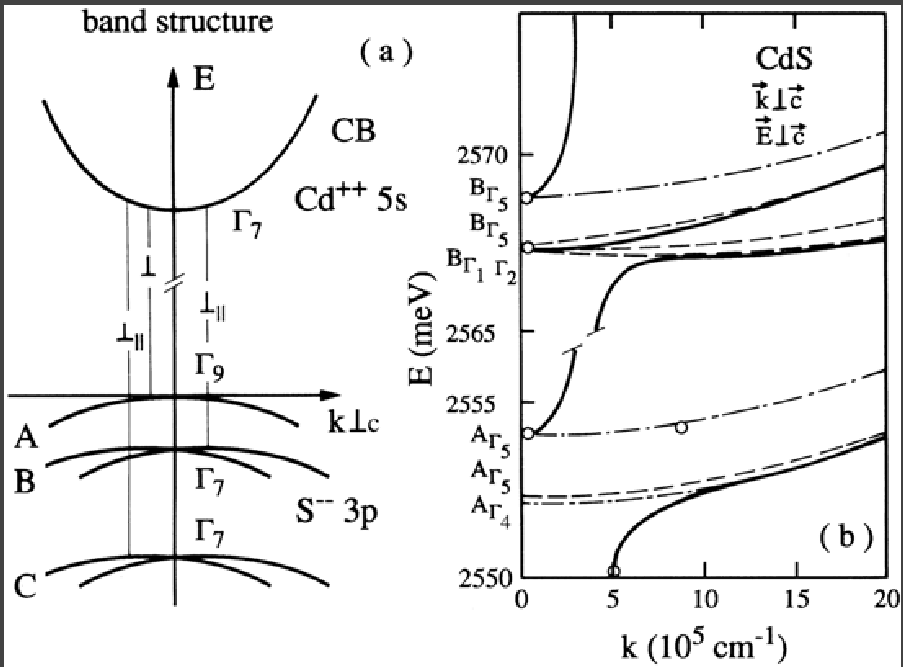


Not just an academic problem !

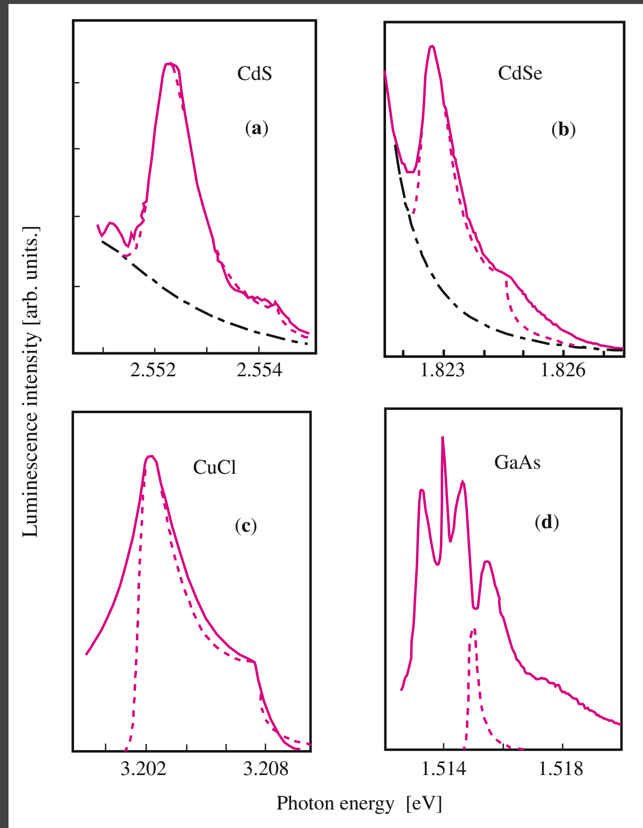
It occurs in all interaction processes between light and matter

Exciton polariton in bulk

CdS bulk
Wurzite structure
3 types of excitons



Bulk Luminescence



Nothing like a
Lorentzian shape! ⇒ Polariton emission

Polariton radiative lifetime

Strong coupling:

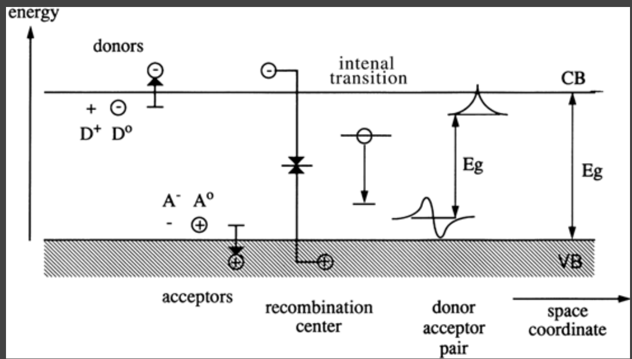
Polaritons are the system eigenstates

They should have an infinite radiative lifetime:

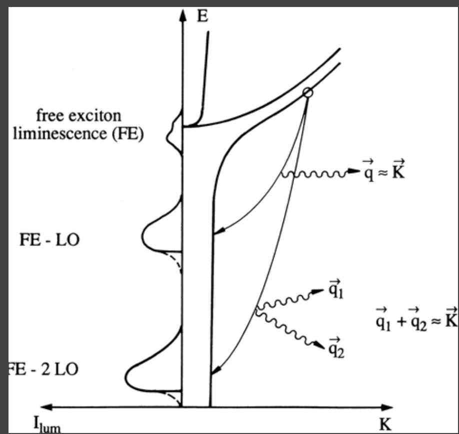
no light out!?!

Dissipative coupling or dephasing

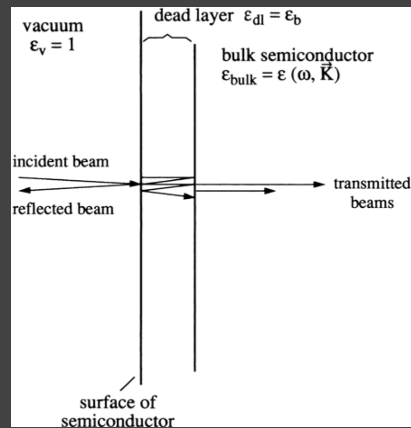
Defects



Phonons



Air/dielectric interface



In bulk long radiative lifetimes, up to several tens of ns

2D heterostructures like QWs

Breakdown of the polariton exciton picture

No more translational symetry for the exciton states along z

A 2D exciton can couple to many optical modes with different k_z

Weak coupling regime

Fermi golden-rule

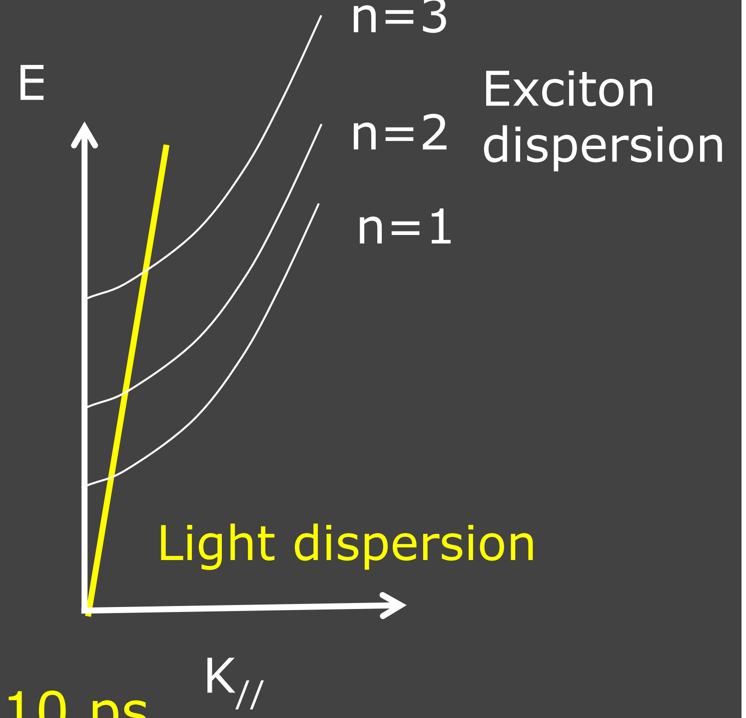
Decay rate

$$\Gamma_{|f\rangle\to|i\rangle} = \frac{\pi}{2\hbar}\left|\langle f|H_{int}|i\rangle\right|^2 \rho(\hbar\omega - E_i + E_f)$$

|i> = |1 exc, 0 ph>

|f> = |0 exc, 1 ph>

ρ density of optical modes



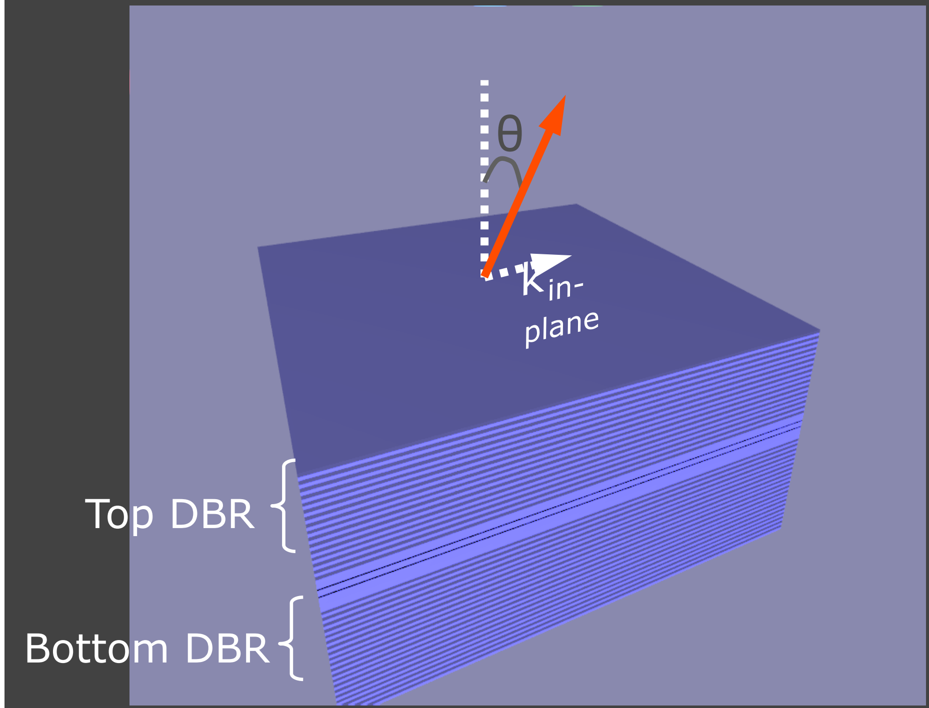
Radiative lifetime ~10 ps

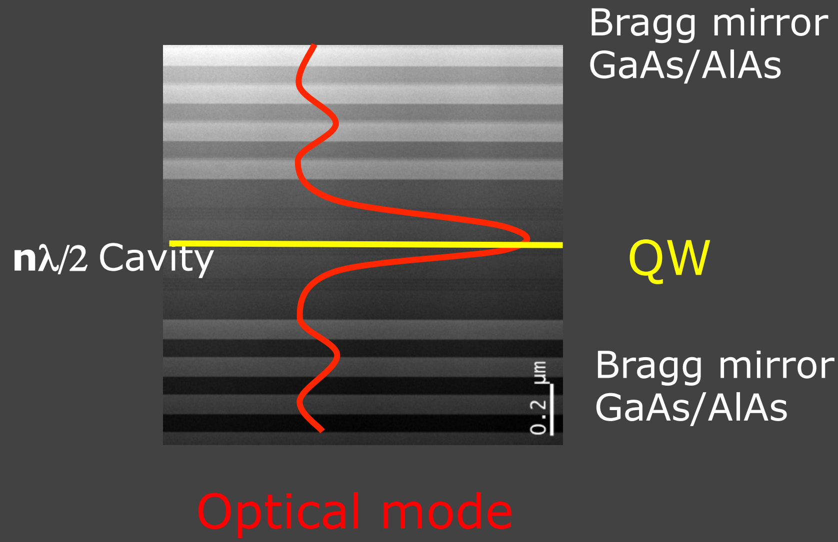
Recovering the strong coupling regime

Couple a QW exciton to a single optical mode
- discretized along z, dispersive along x and y
- localized close to the QW plane

QW inside a 2D optical cavity

GaAs nλ/2 cavity between 2 Bragg mirrors

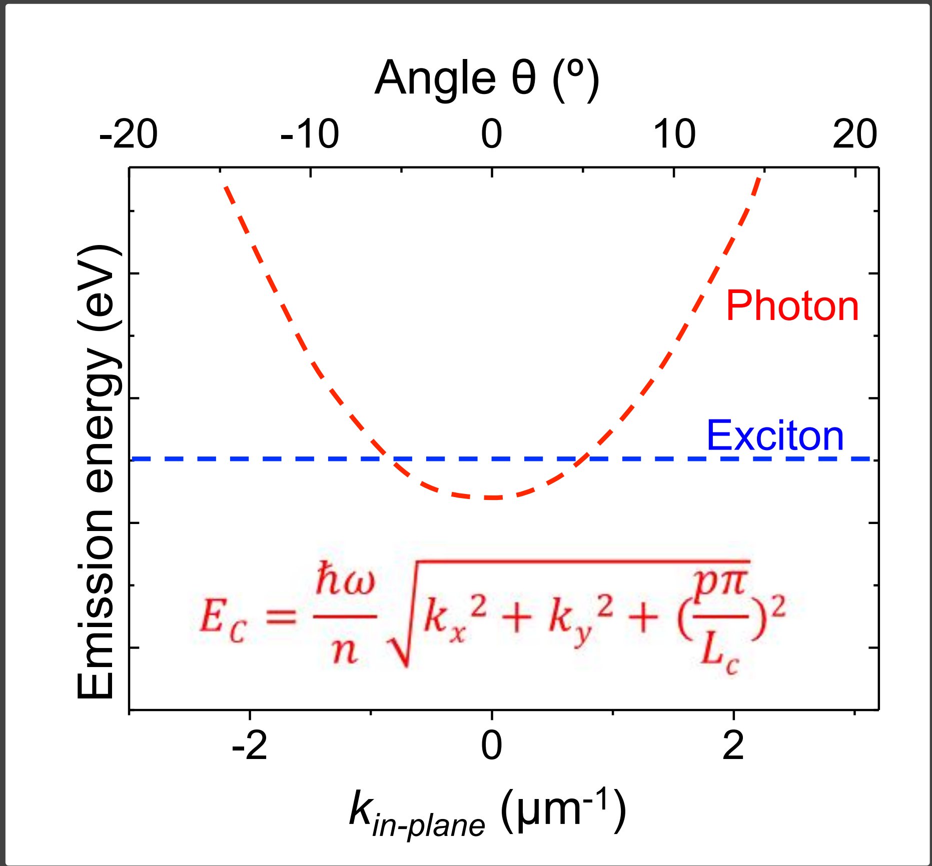




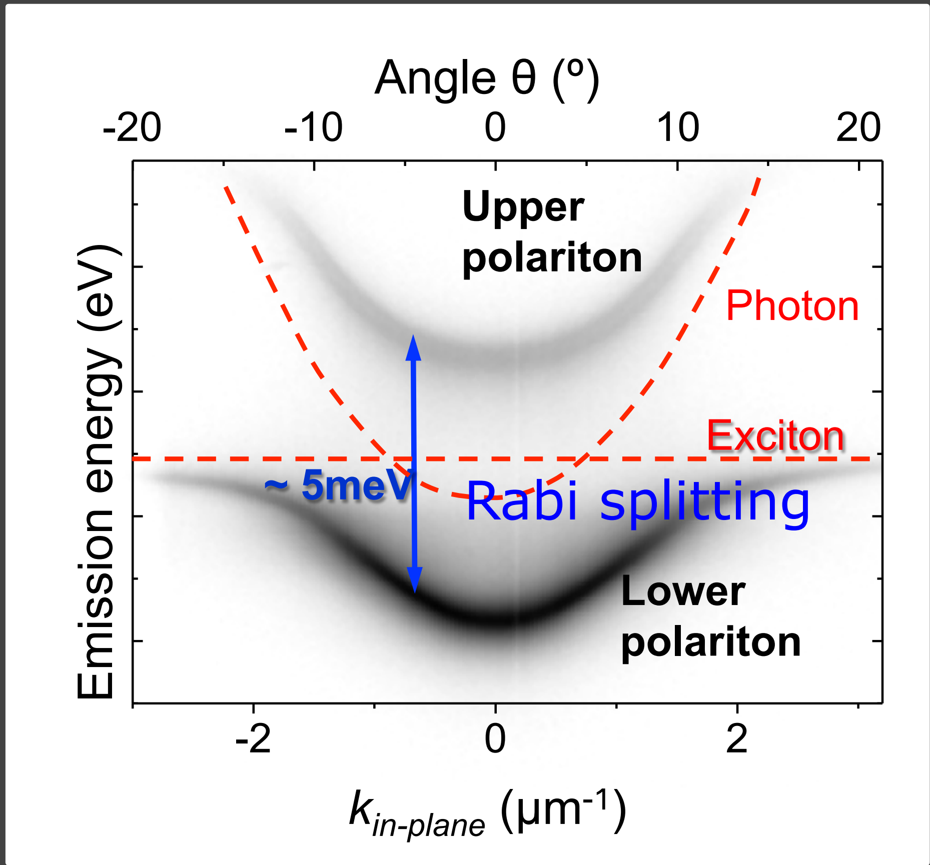
Coupling a 2D exciton to a 2D microcavity

Empty cavity

A uncoupled QW exciton



Turning on the light-matter coupling

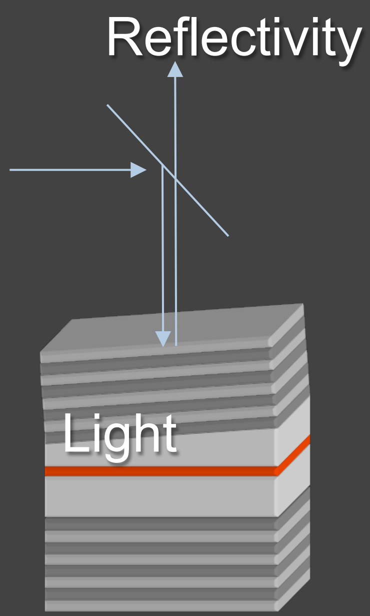


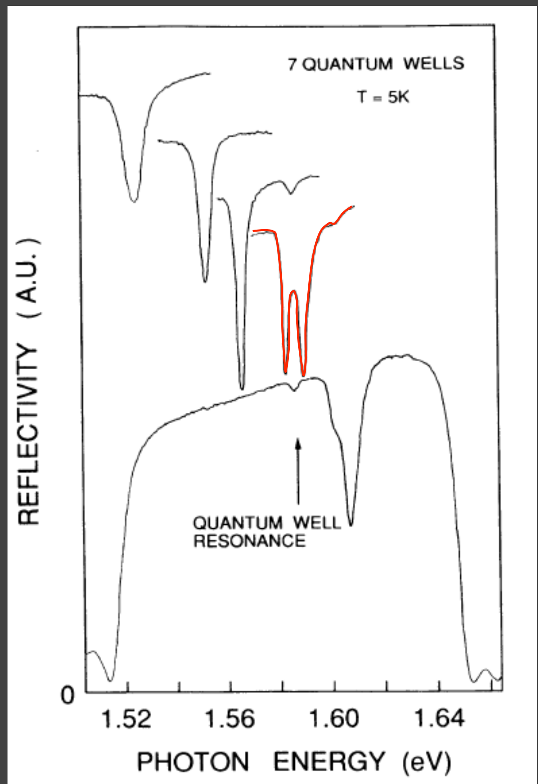
First Observation of the strong coupling

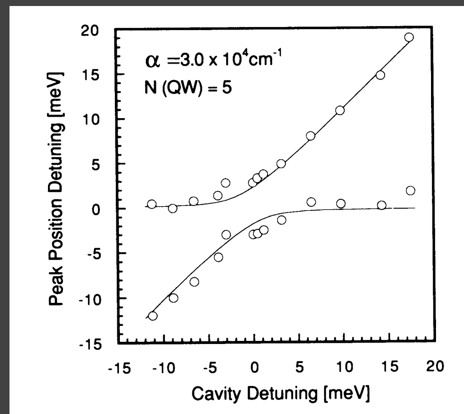
VOLUME 69, NUMBER 23 PHYSICAL REVIEW LETTERS 7 DECEMBER 1992

Observation of the Coupled Exciton-Photon Mode Splitting in a Semiconductor Quantum Microcavity

C. Weisbuch,[a] M. Nishioka,[b] A. Ishikawa, and Y. Arakawa
Research Center for Advanced Science and Technology, University of Tokyo, 4-6-1 Meguro-ku, Tokyo 153, Japan
(Received 12 May 1992)

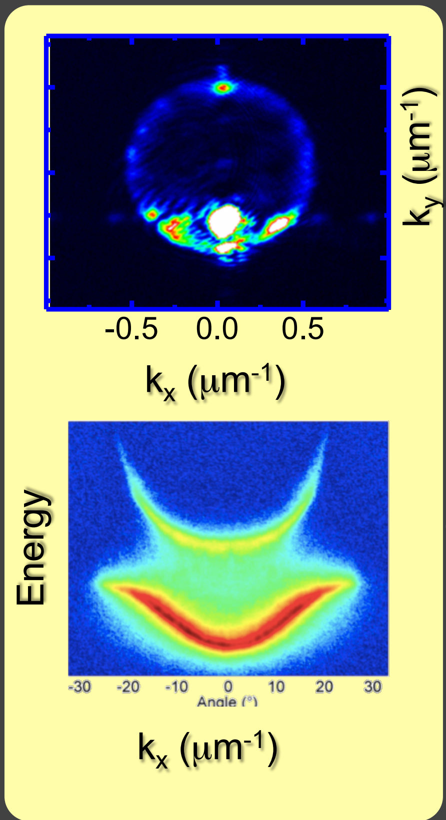




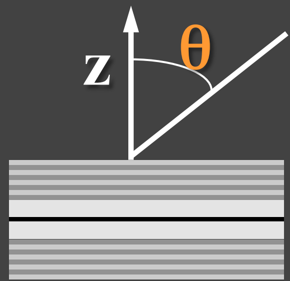


Microcavity polariton emission

Far field pattern: Direct visualization of the polariton dispersion

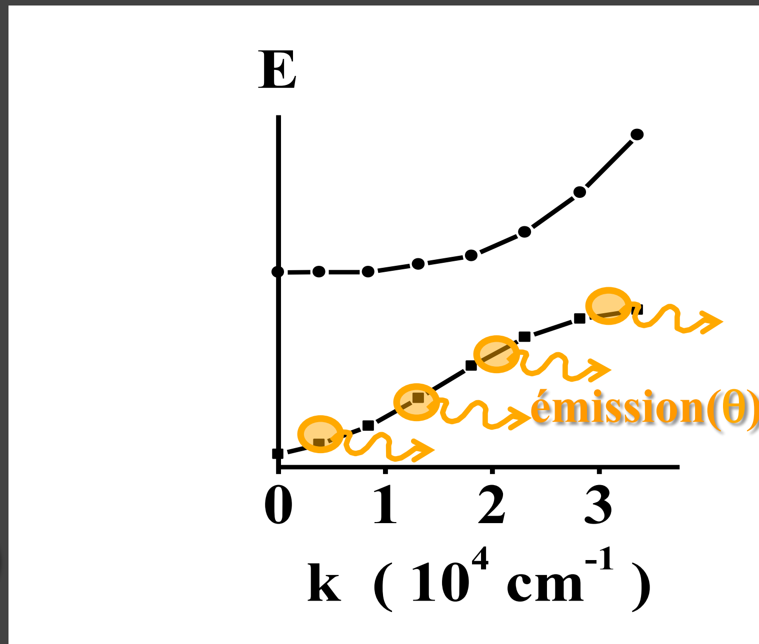


(d)



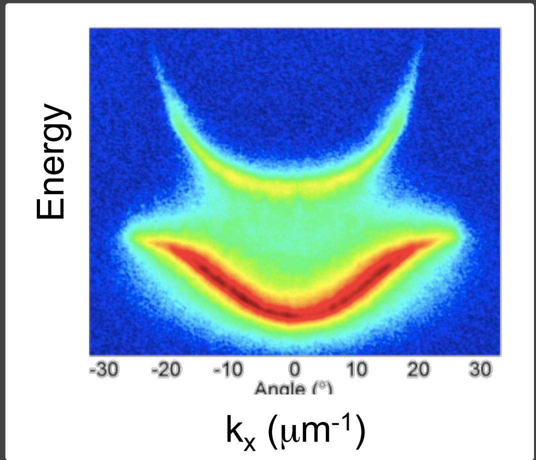
$\theta \Longleftrightarrow k_{//}$

$$k_{//} = \omega/c \sin(\theta)$$



R. Houdré et al., Phys. Rev. Lett. **73**, 2043 (1994)

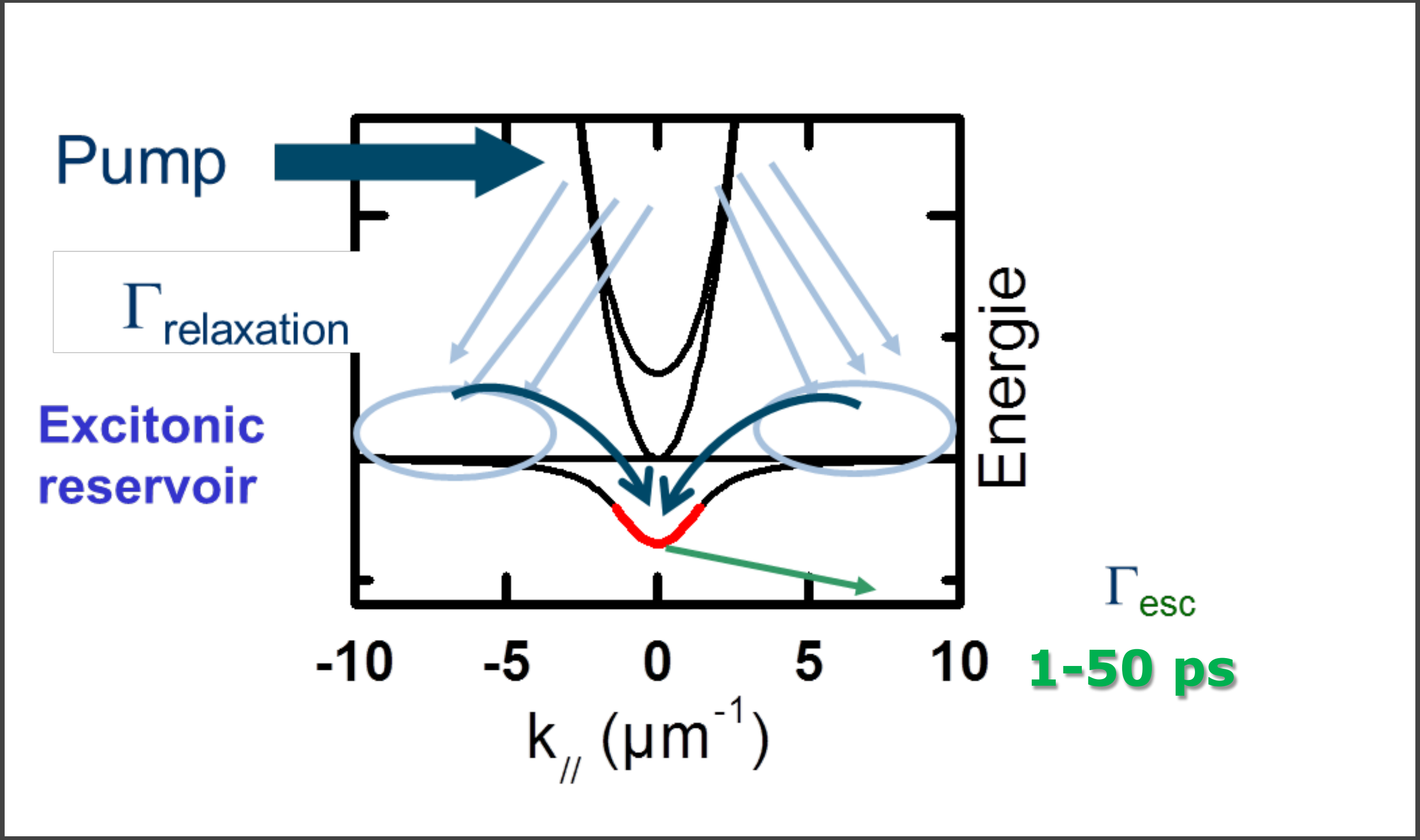
Some interesting properties



Properties

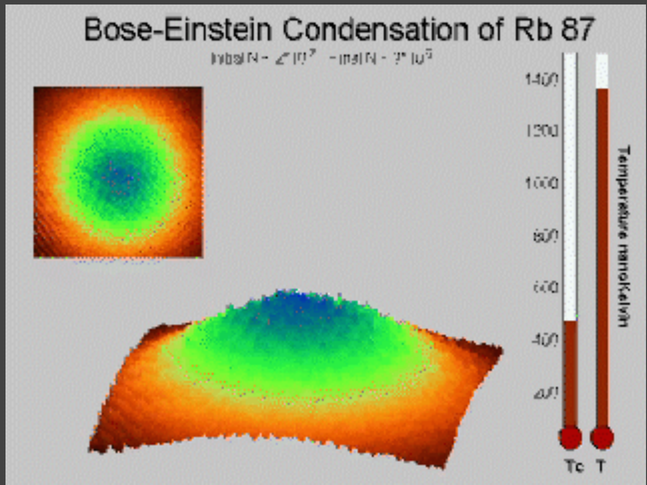
- Optical excitation and access to the polariton energy, momentum and space distribution
- Short lifetime (~1-30 ps) ⇒ **escape out of the cavity**
- Excitonic component ⇒ **strong interactions**
- Photonic component ⇒ **low mass (10^{-5} m_e)**
- Bosons

Polariton accumulation under non resonant condition



Relaxation is possible thanks to the excitonic part of the polariton

Bose Einstein Condensation in atoms

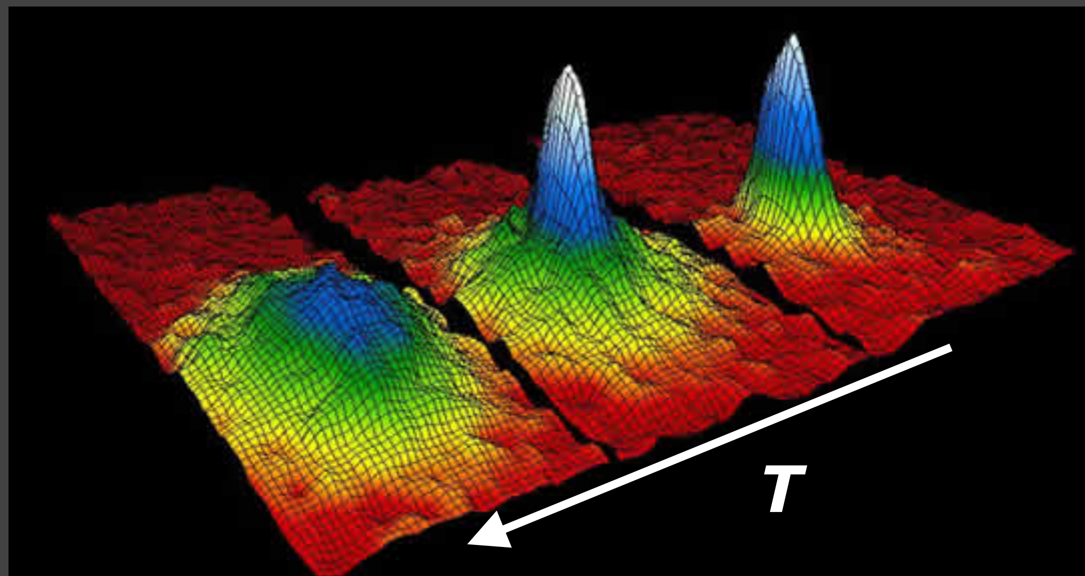


n: boson density

$$T_c = \left(\frac{n}{\zeta(3/2)}\right)^{2/3} \frac{2\pi\hbar^2}{mk_B} \approx 3.3125\ \frac{\hbar^2 n^{2/3}}{mk_B}$$

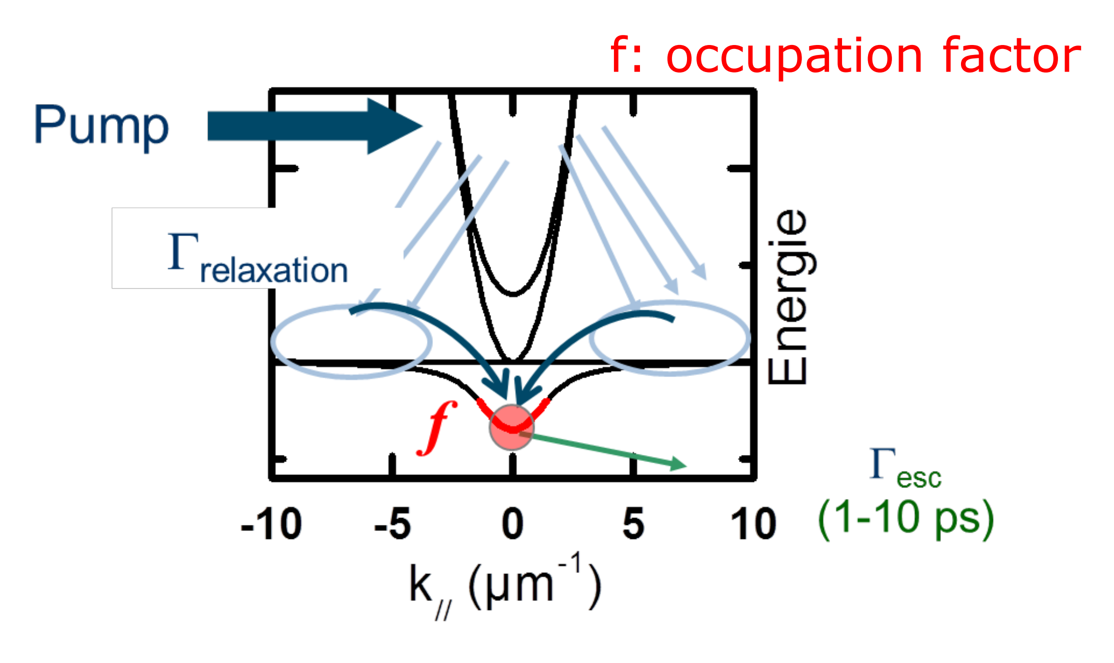
Cornell and Wieman's groups condensation of Rb atoms (1995)

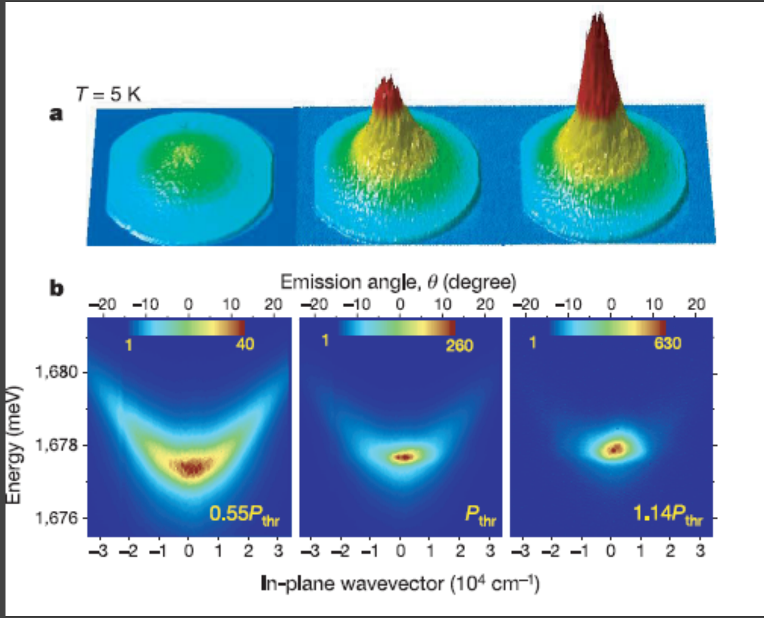
- $m = 10^4\ m_e$
- $T_c = 200$ nK



http://jilawww.colorado.edu/bec/

Polariton condensation





Nature 443, 409 (2006)

CdTe/CdMgTe cavity

When f exceeds unity : $\Gamma_{relaxation} \;\; \alpha \; (f+1)$ Bosonic stimulation

Massive occupation of the lowest energy states

Also observed in GaAs (10K), GaN, ZnO (room temperature)…

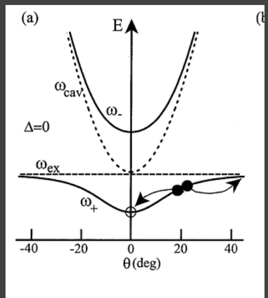
Playing with polaritons

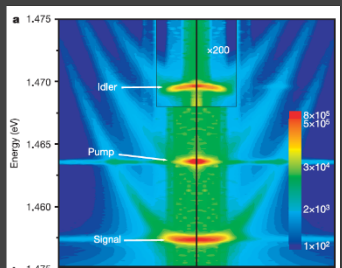
Polariton propagation and manipulation

thanks to their photonic part polaritons have large coherence length

thanks to their excitonic part, polaritons interact

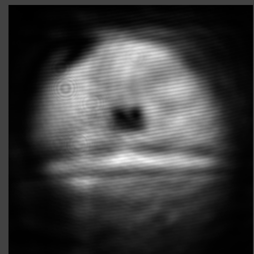
Optical Parametric Oscillator/Amplifier





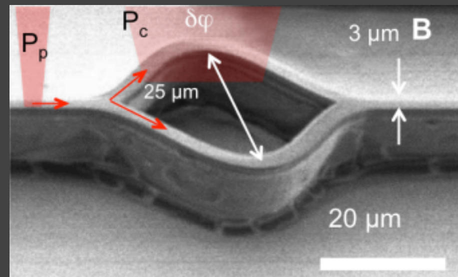
R. Stevenson, *et al*, PRL **85**, 3680 (2000)
P. Savidis, *et al*, PRL **84**, 1547 (2000)
C. Diederichs, *et al,* Nature **440**, 904 (2006)

Superfluidity



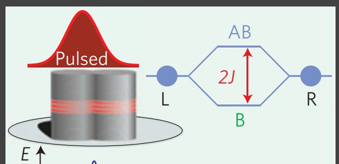
AA, Lefrère *et al.,* Nature Phys. **5**, 805 (2009)

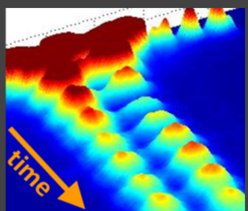
Polariton interferometer



C. Sturm, *et al*.
Nat. Comm 5, 3278 (2014)

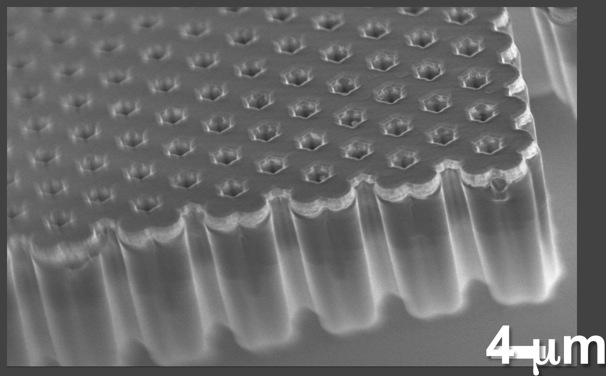
Polaritonic molecules

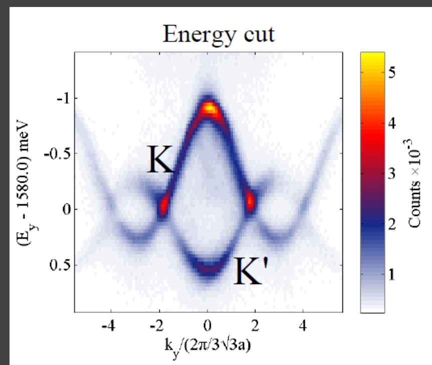




M. Abbarchi et al

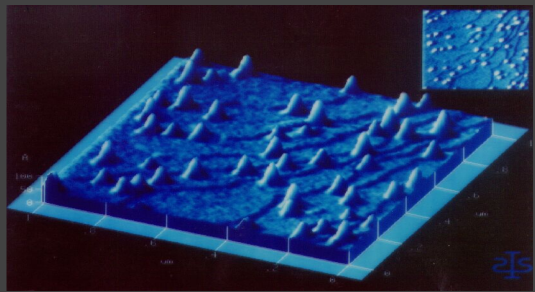
Polaritonic honeycomb lattice



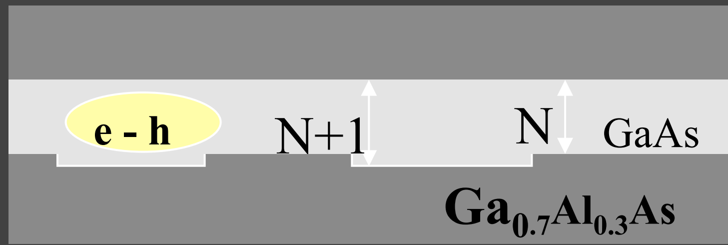


Light-matter interaction with 0D objects

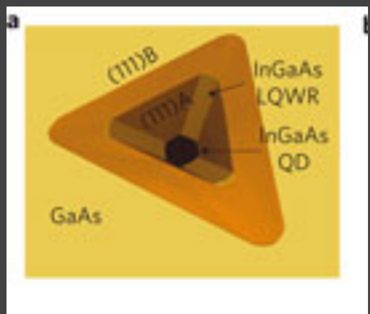
Light emission from single epitaxial emitters



Self-assembled QDs
(Stransky-Krastanov, dropplets)

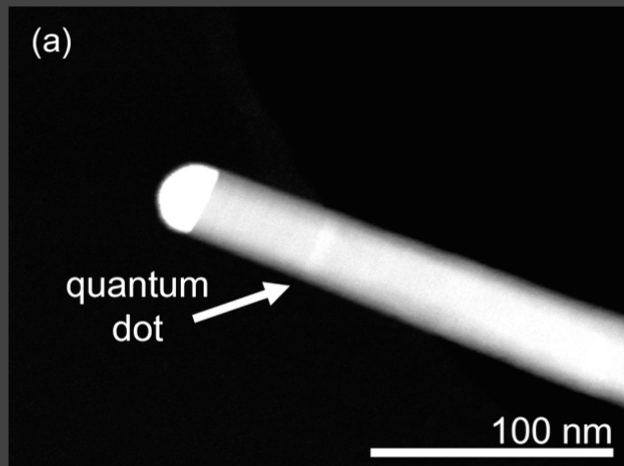


Interface fluctuations



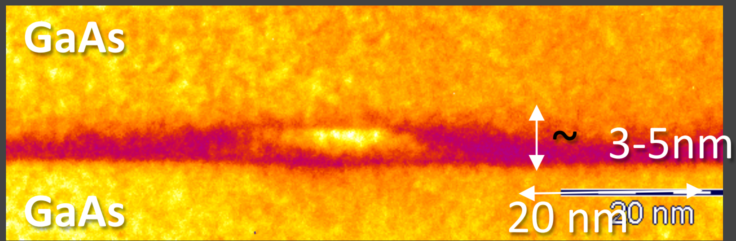
Kapon's group

Localized growth



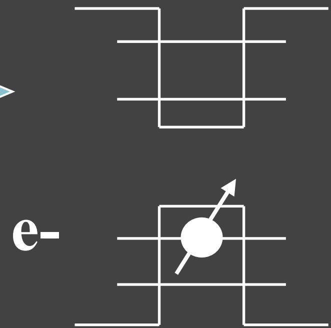
Dot in a nanowire

Artificial atoms



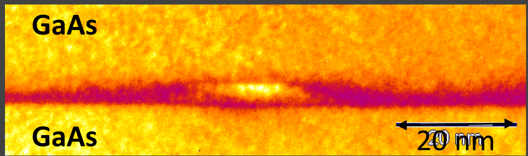
InAs/GaAs quantum dot

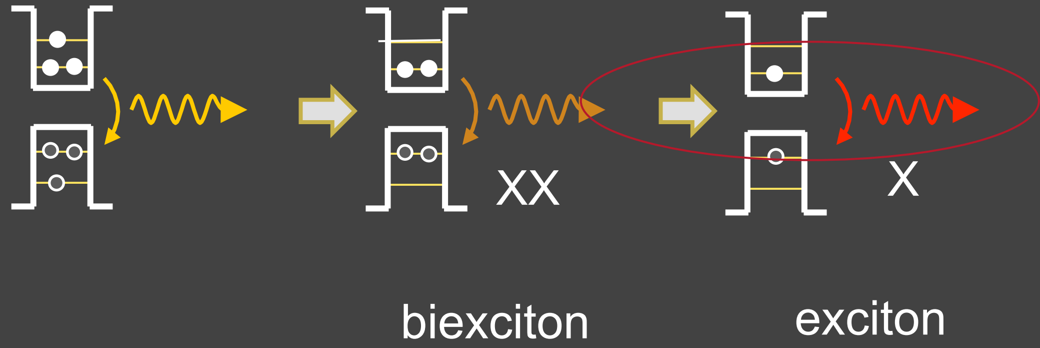
BQ

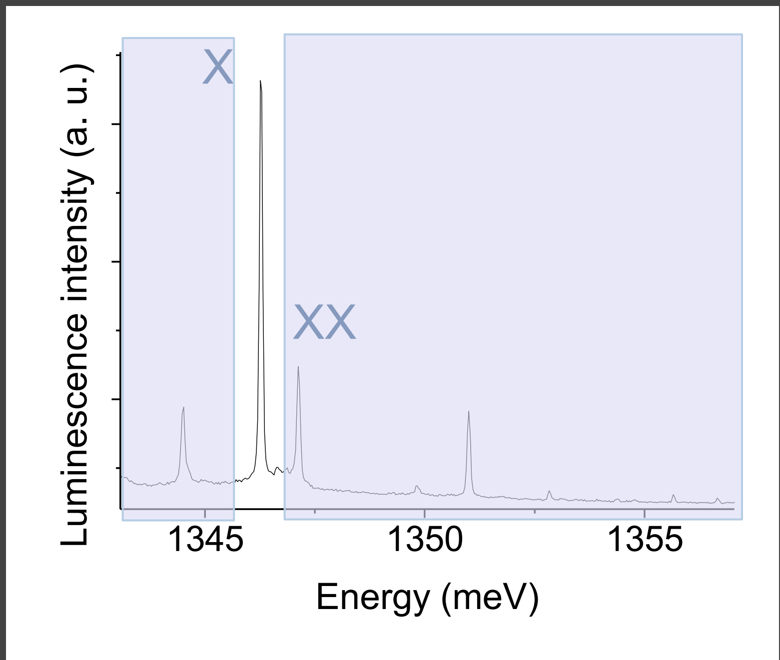


Discrete states

Solid state source of quantum light







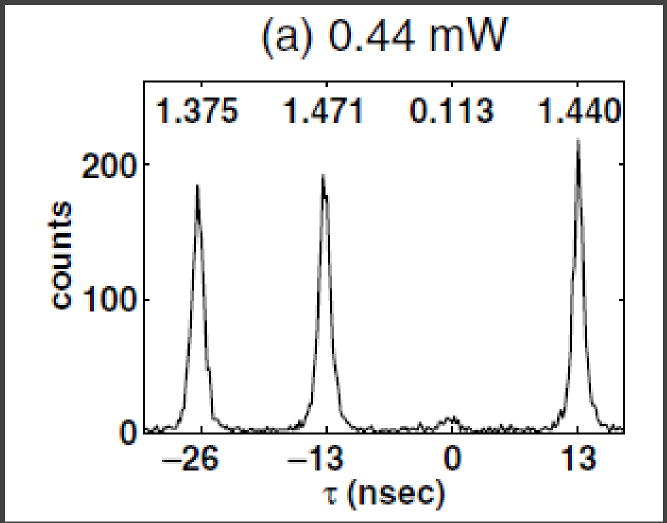
Creation of carriers in the QD (laser, bias)

Poisson statistic

Single photon emitted at the X energy

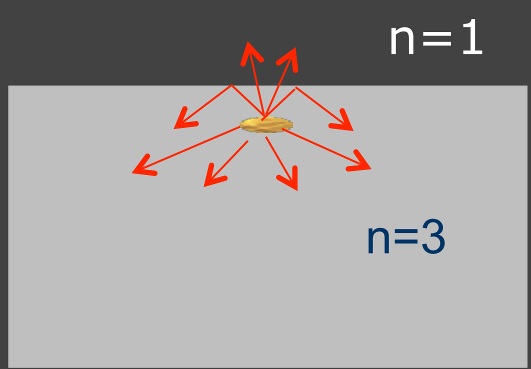
Generation of a single photon state

Michler et al, Science 290, 2282 (2000)



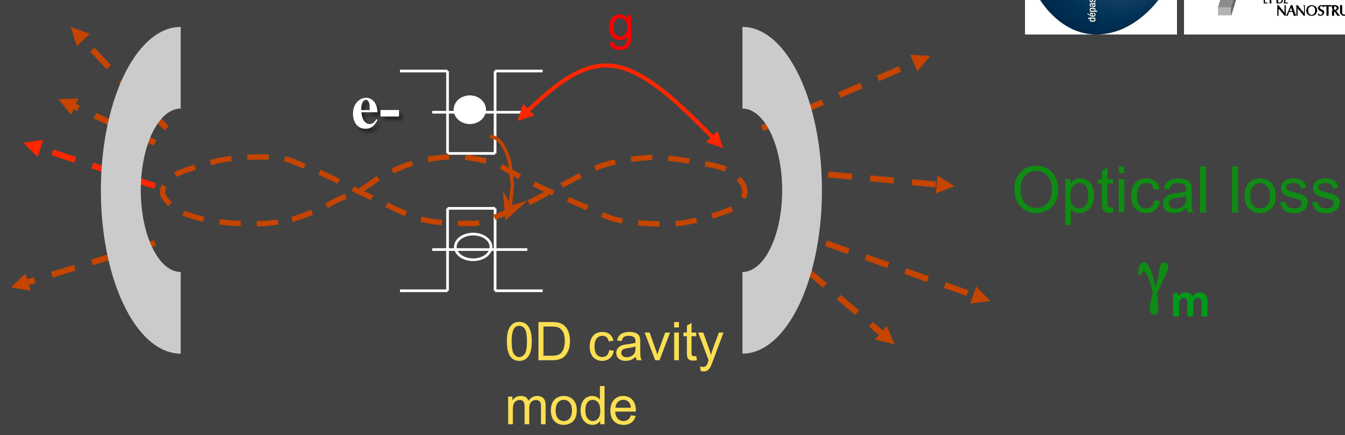
Santori et al, PRL 2001

Great source of single photons

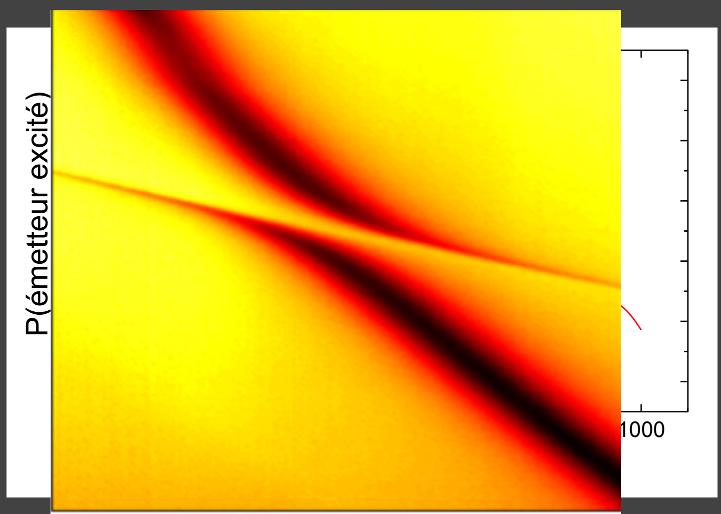


However limited extraction efficiency 3%

Coupling to a 0D cavity mode

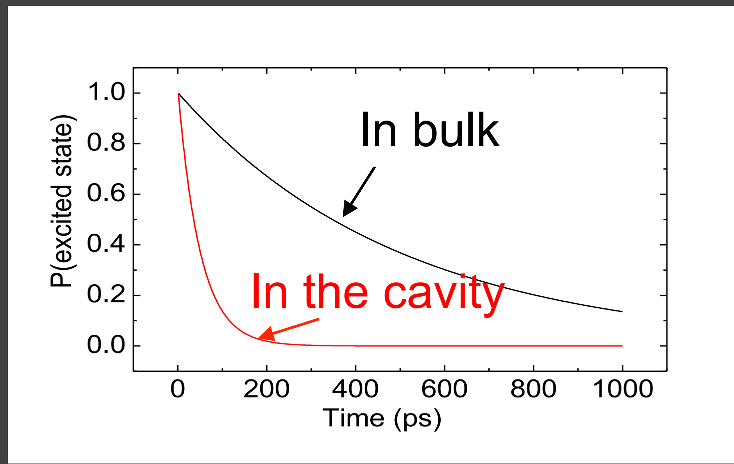


$g >> \gamma_m$



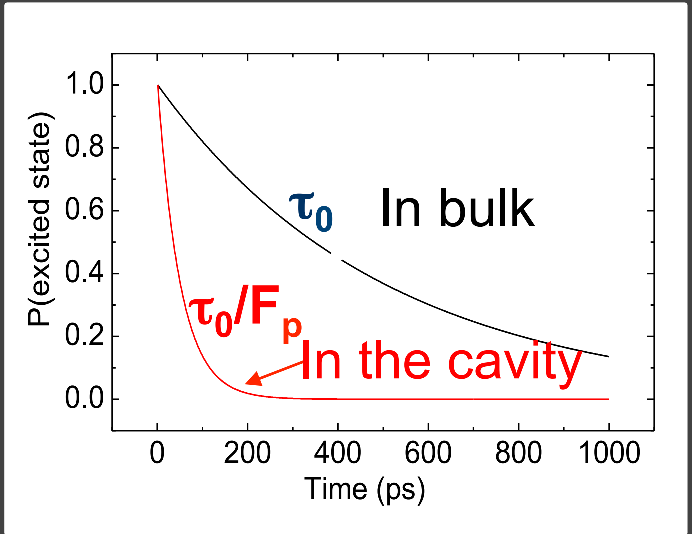
Strong coupling regime
Light-matter entangled states

$g << \gamma_m$



Weak coupling regime
Enhanced spontaneous emission
Purcell Effect

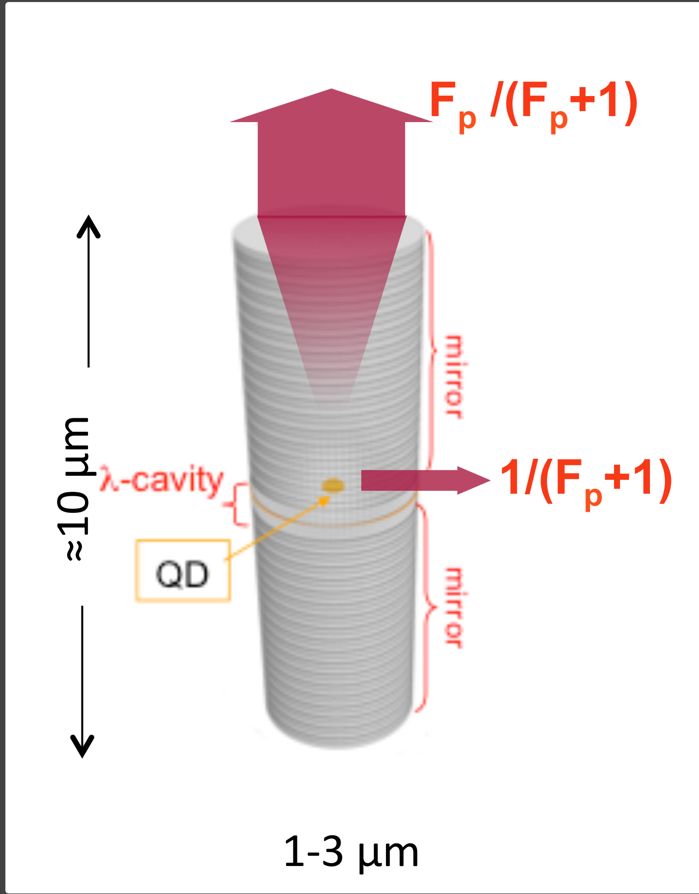
Weak coupling regime : the Purcell effect



Gérard et al, PRL 1998, Gayral et Gérard, Journal of Light. Tech. 2000

$$F_P = \frac{3}{4\pi^2}\frac{Q}{V/(\lambda_0/n)^3}$$

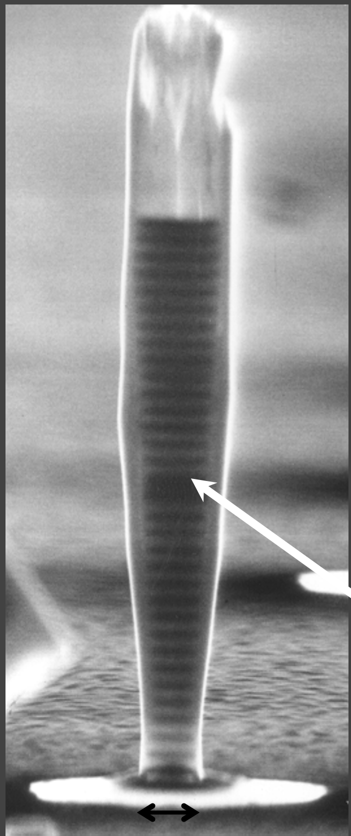
Q: cavity quality factor
V: modal volume



80% of emission coupled to the mode for F_p=4

Efficient and fast photon collection !

First demonstrations

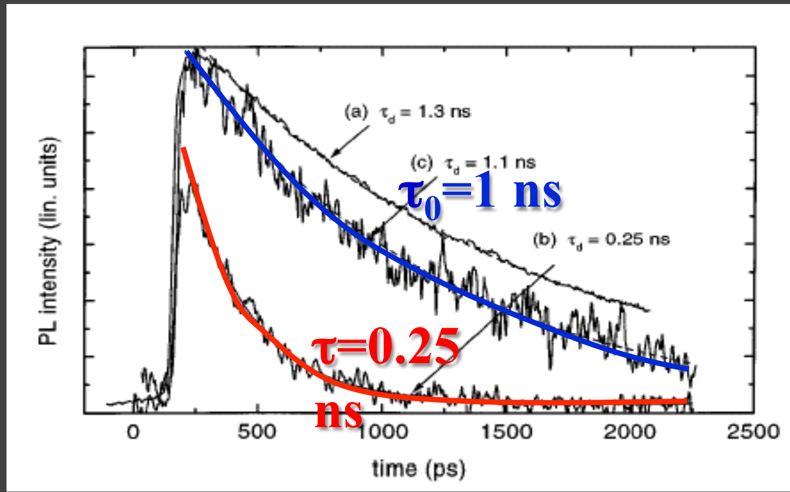


1 µm

Many InAs QDs

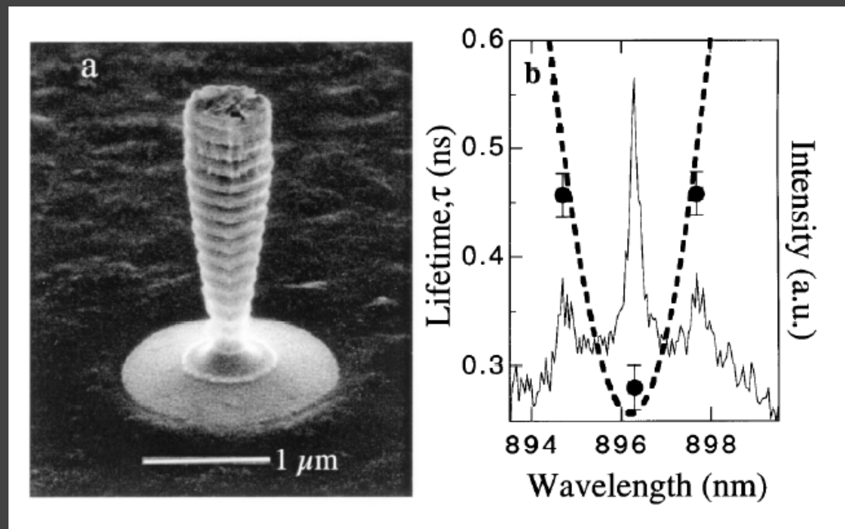
AlAs/GaAs Micropillar:
0D optical confinement

Large ensemble of QDs



Gérard et al (1998)

Single QD



G. Solomon et al , PRL 2000

Maximizing the Purcell effect

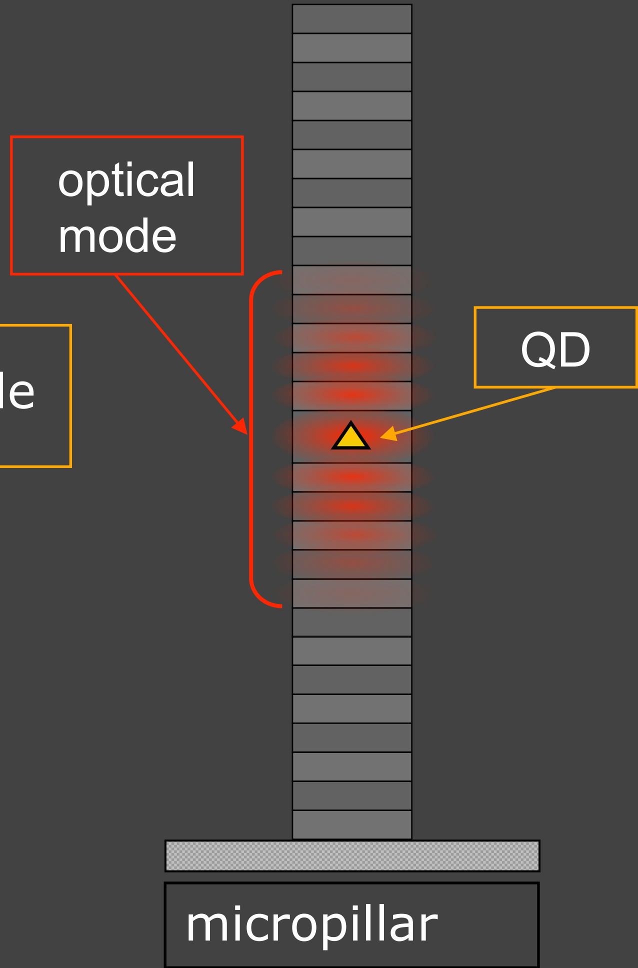
Spatial matching :

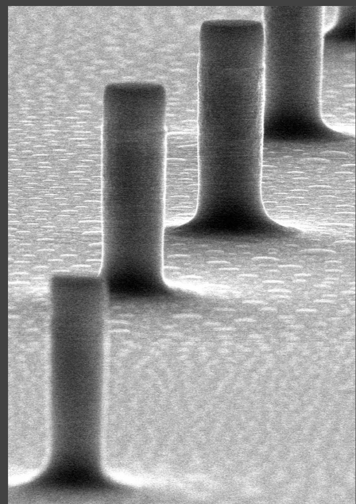
QD at the maximum of the field intensity

&

Spectral matching :

E_X = E mode





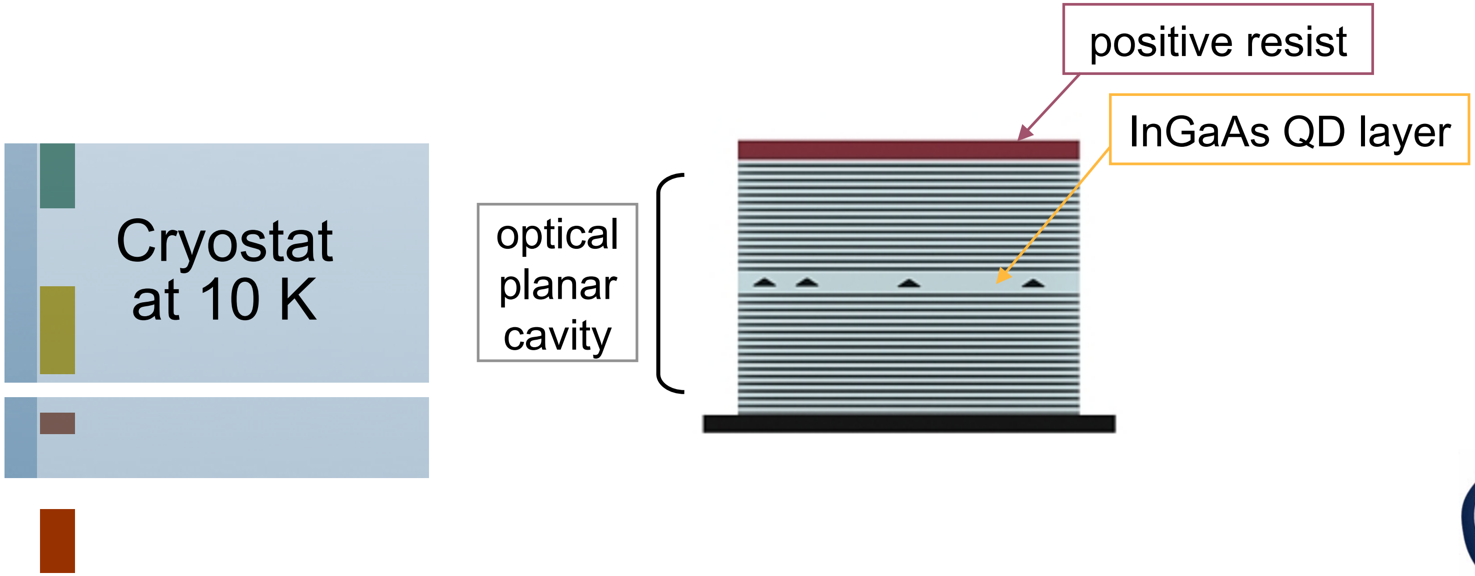
Pascale Senellart's group at LPN

An all optical technique : Low temperature in-situ lithography

- Single step spectral and spatial matching

- Scalability

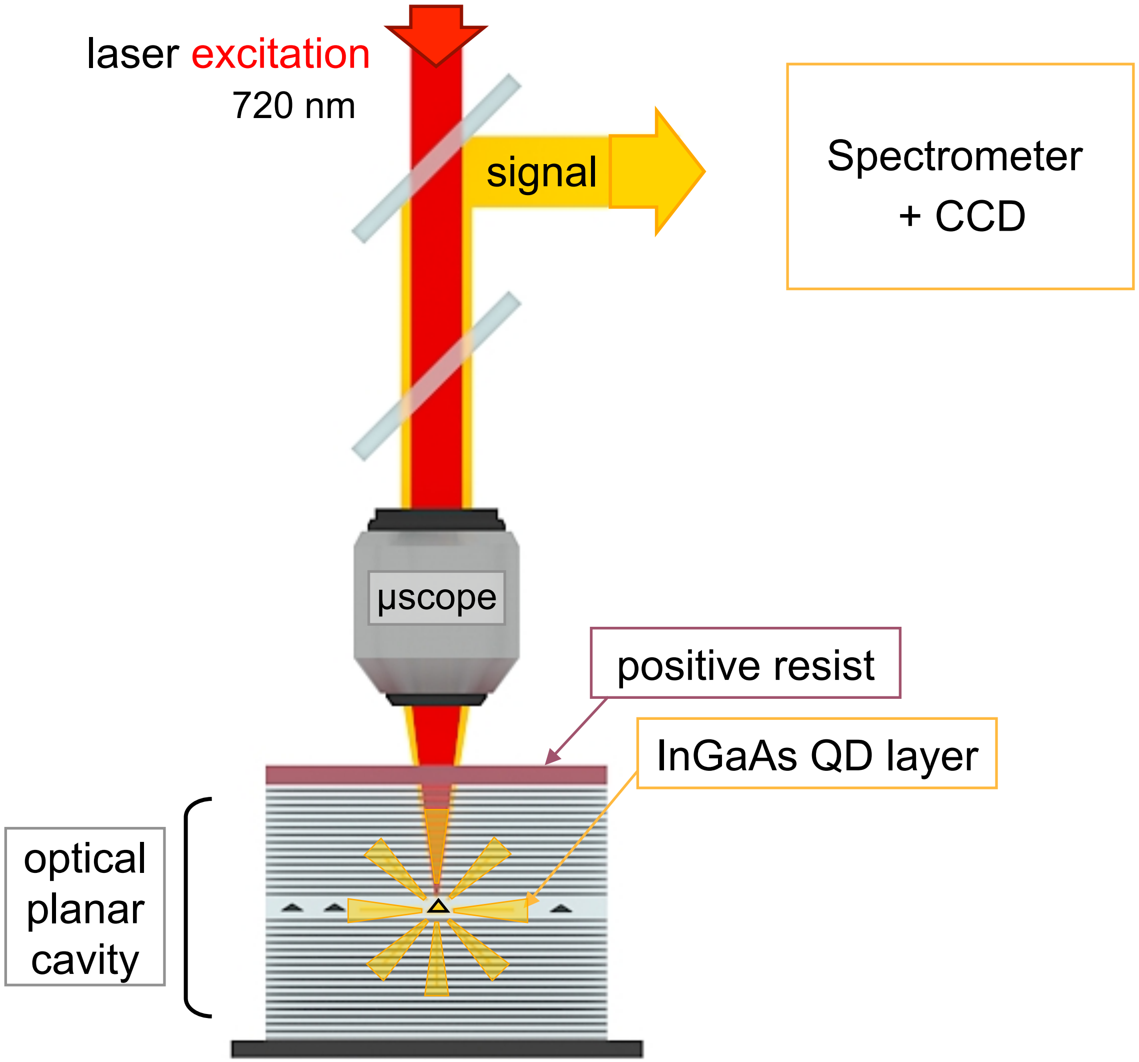
In situ lithography



In situ lithography

optical lithography + µPL

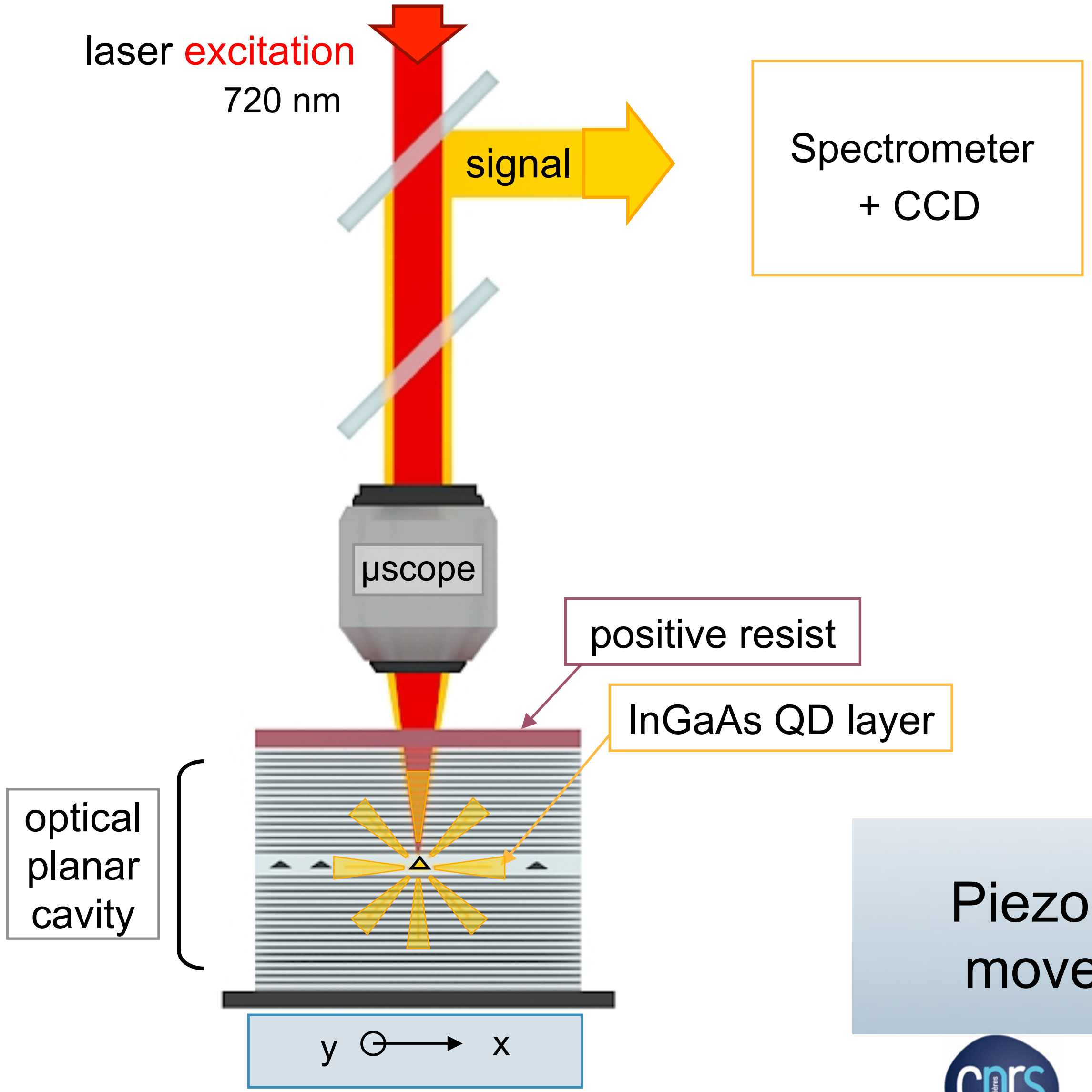
Cryostat at 10 K



In situ lithography

optical lithography
+ µPL

Cryostat at 10 K



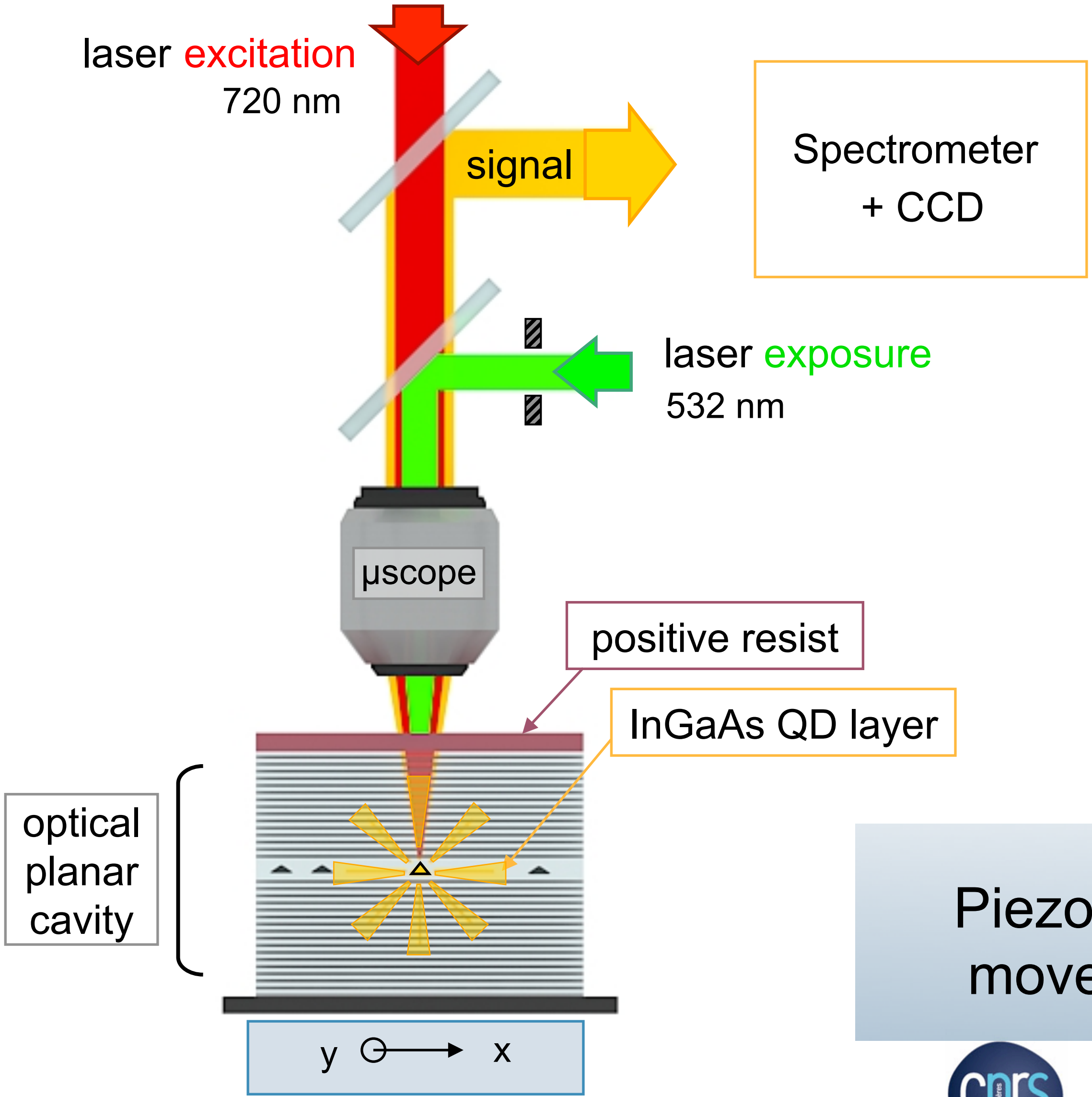
Piezoelectric movements

In situ lithography

optical lithography + µPL

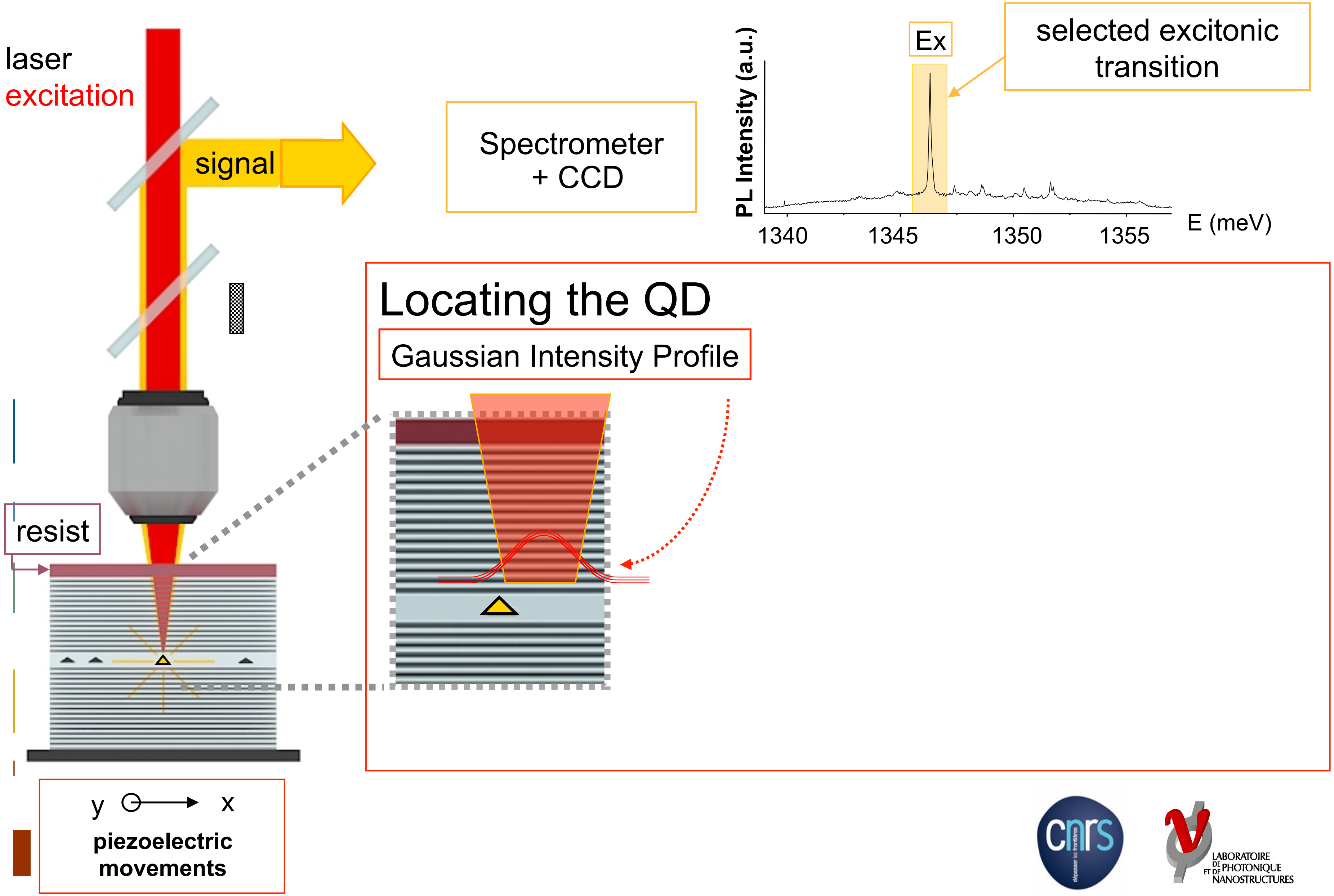
2 aligned lasers

Cryostat at 10 K

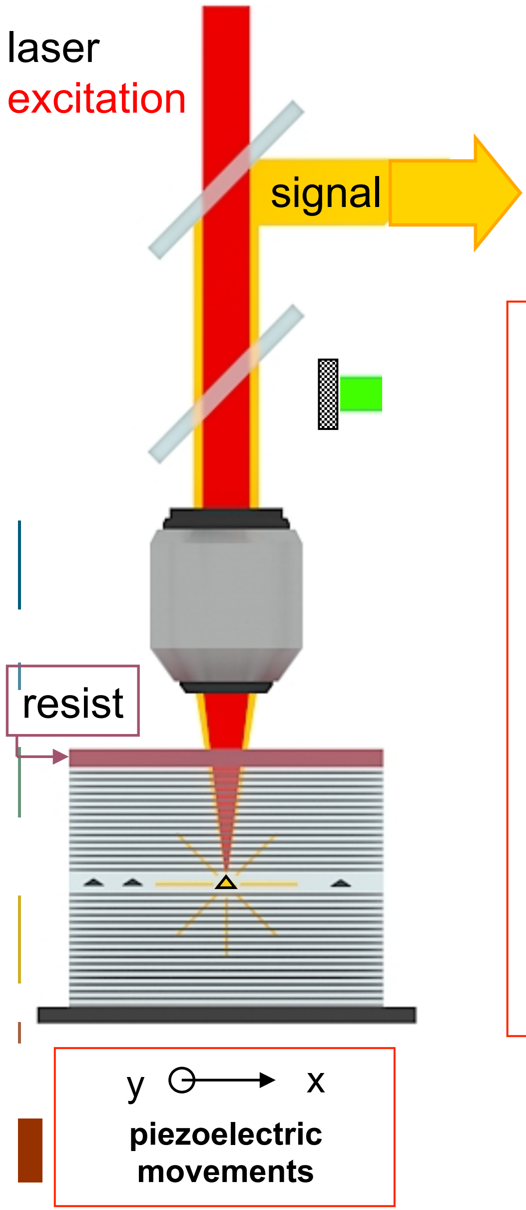


Piezoelectric movements

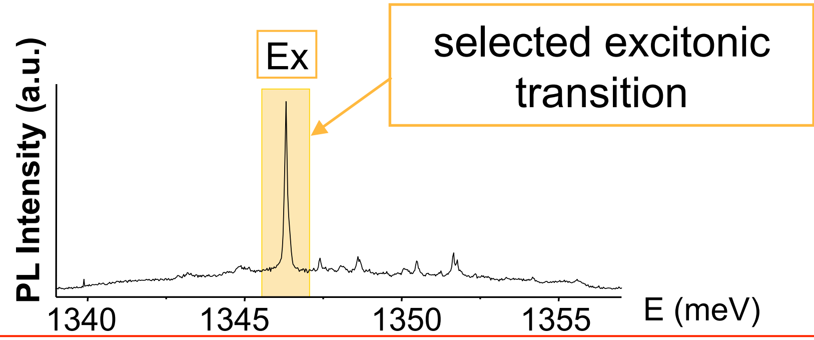
Spatial Matching



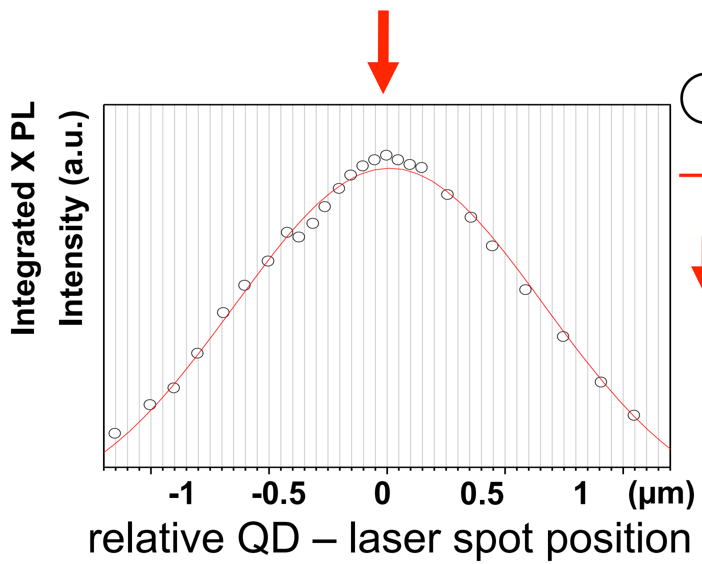
Spatial matching



Spectrometer
+ CCD



Locating the QD

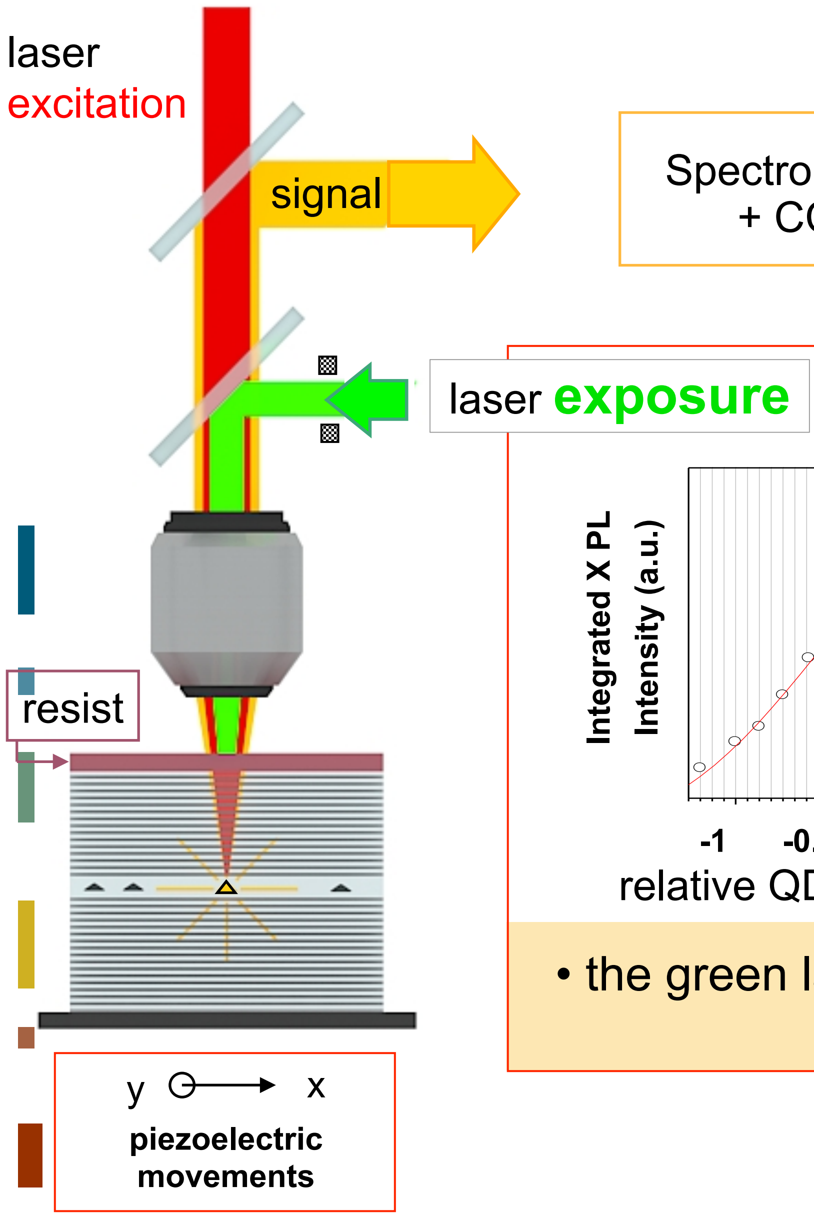


PL signal intensity

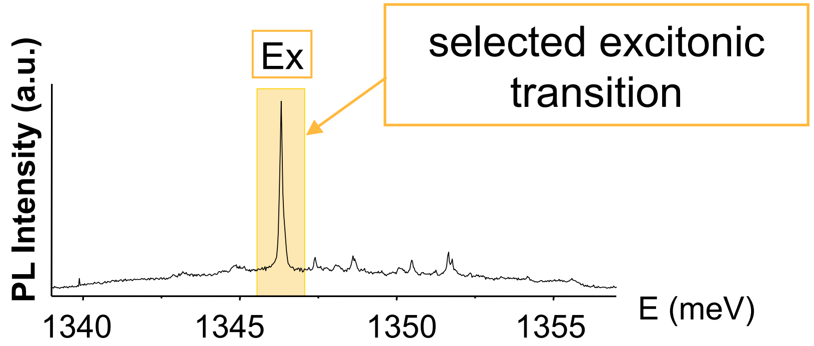
Gaussian fit

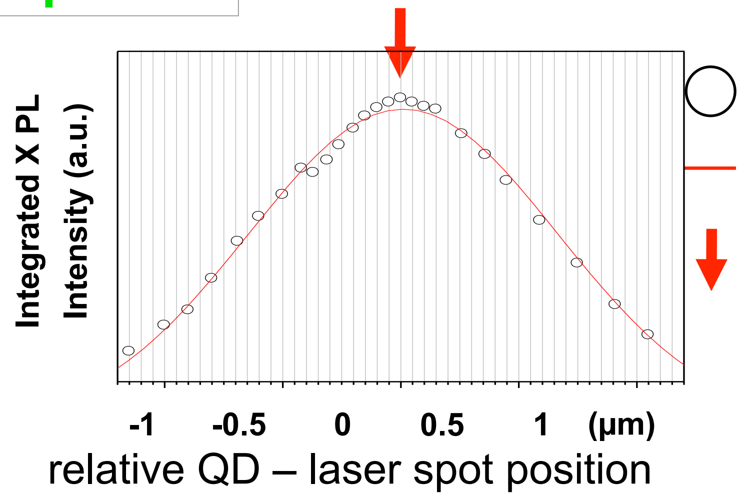
Centered position of the QD relative to the laser spot with **50 nm** accuracy

Spatial matching



Spectrometer
+ CCD





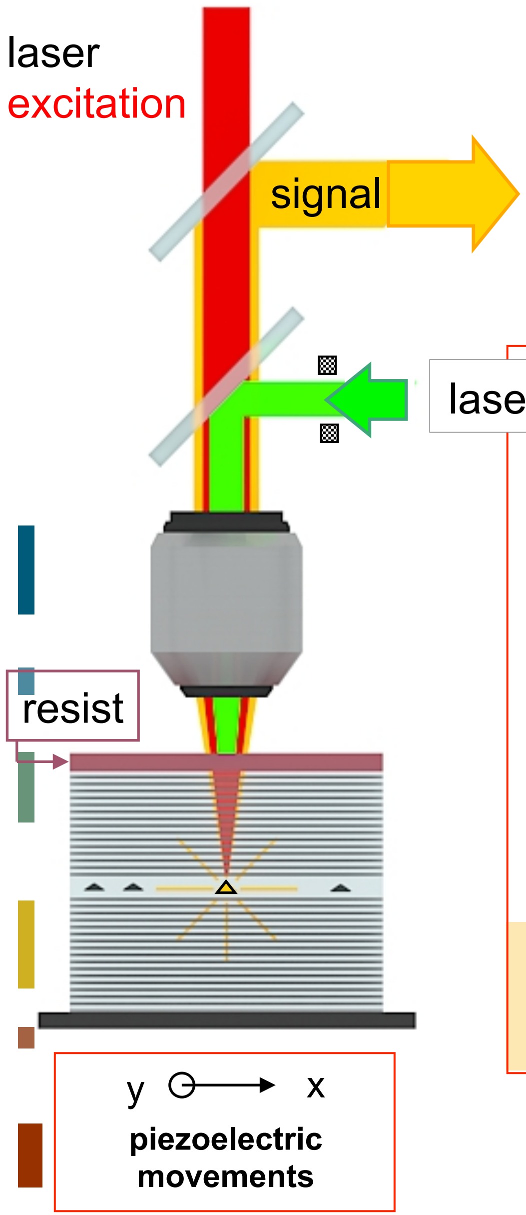
PL signal intensity

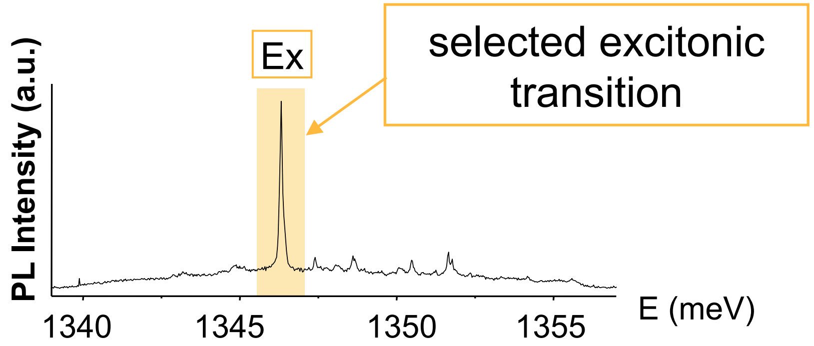
Gaussian fit

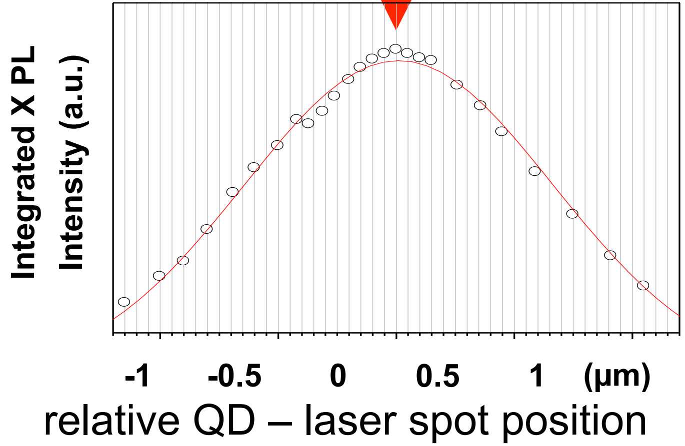
Centered position of the QD relative to the laser spot with 50nm accuracy

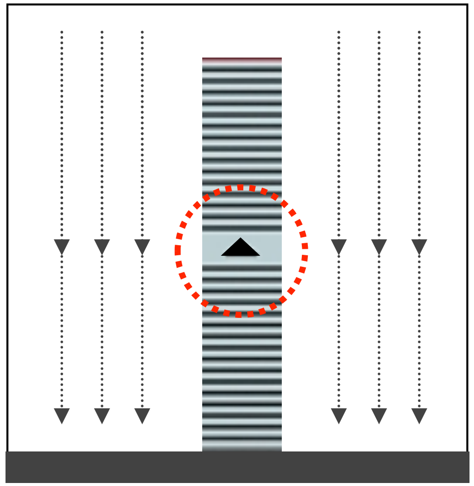
- the green laser exposes the resist

Spatial matching



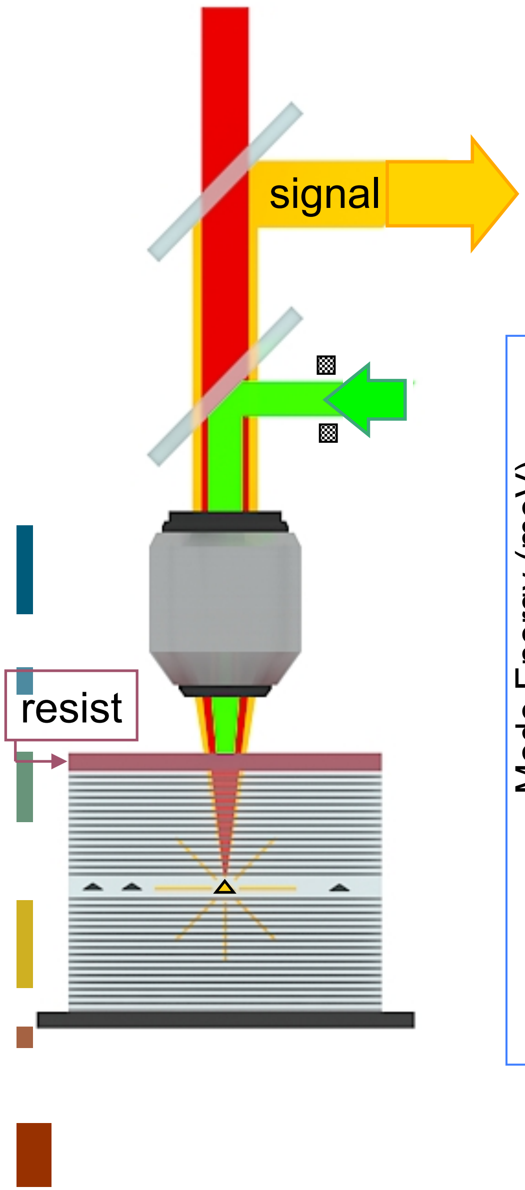




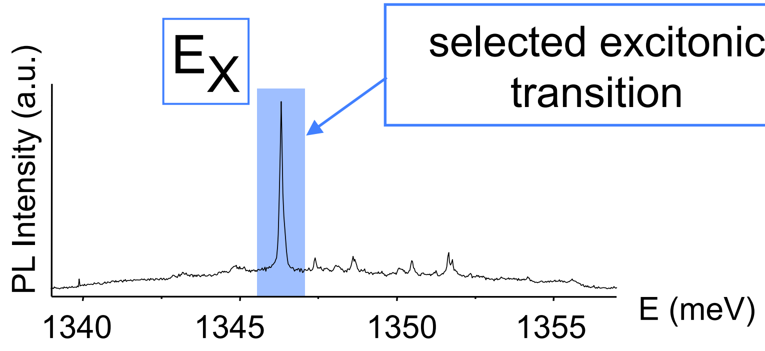


- the green laser exposes the resist
- the QD will be centered in the micropillar after etching

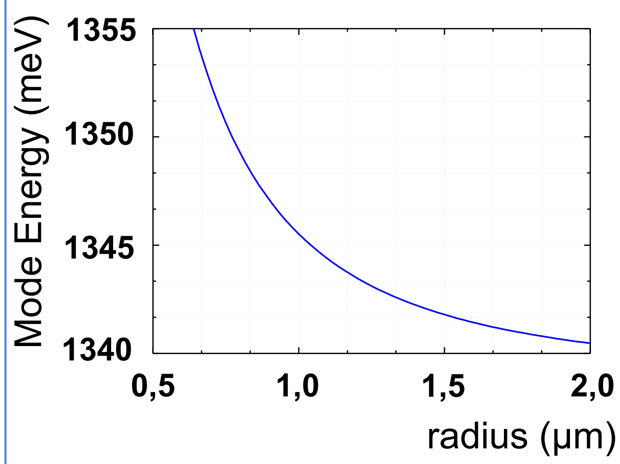
Spectral matching



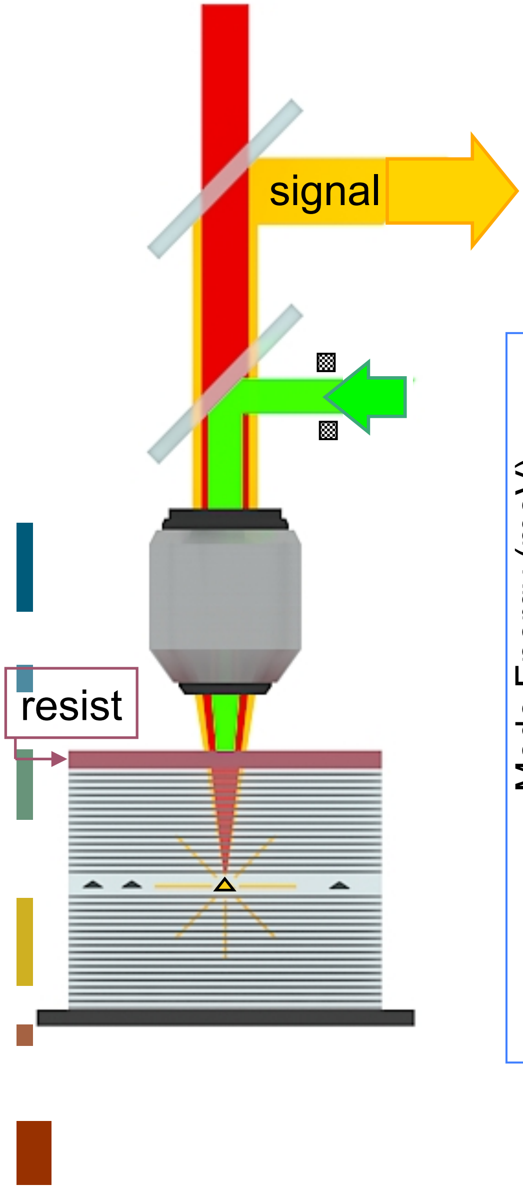
Spectrometer
+ CCD



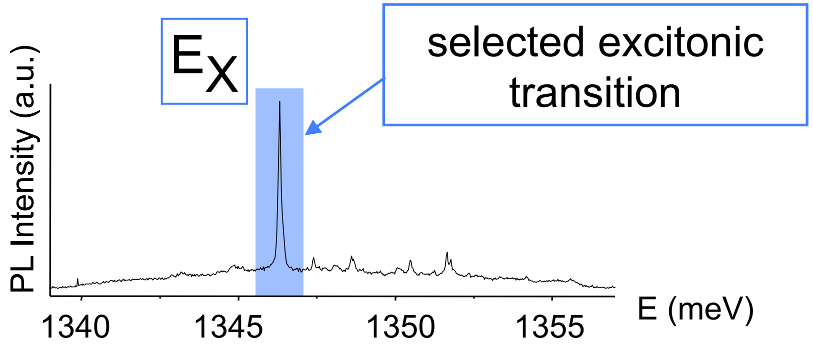
Selecting the appropriate radius



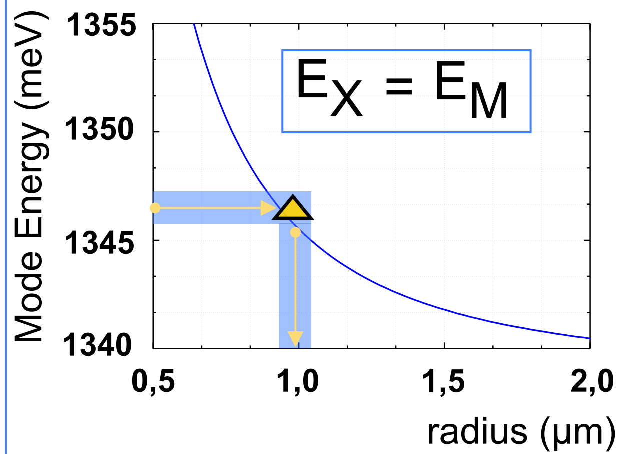
Spectral matching



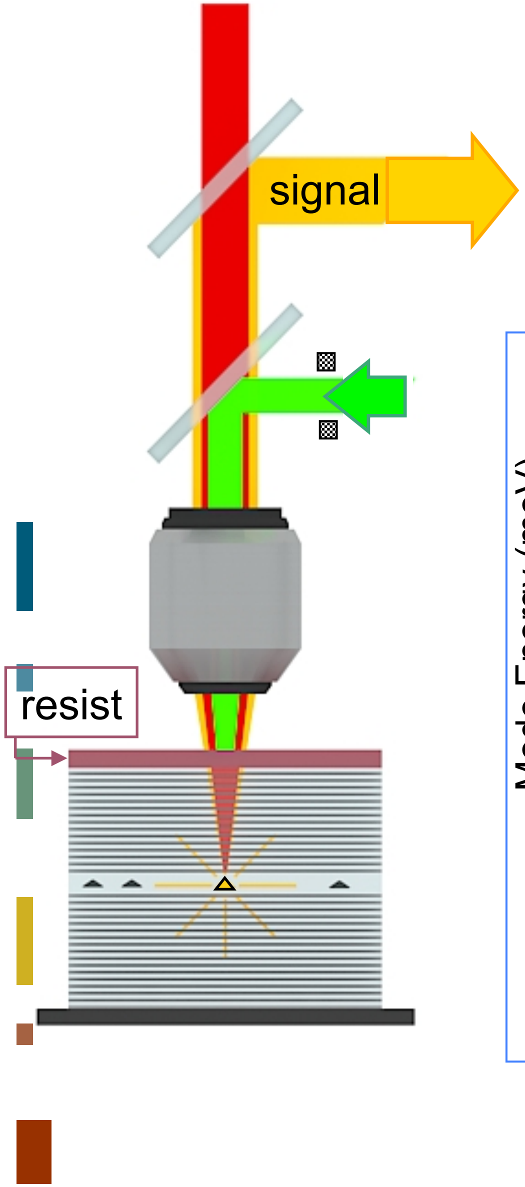
Spectrometer
+ CCD



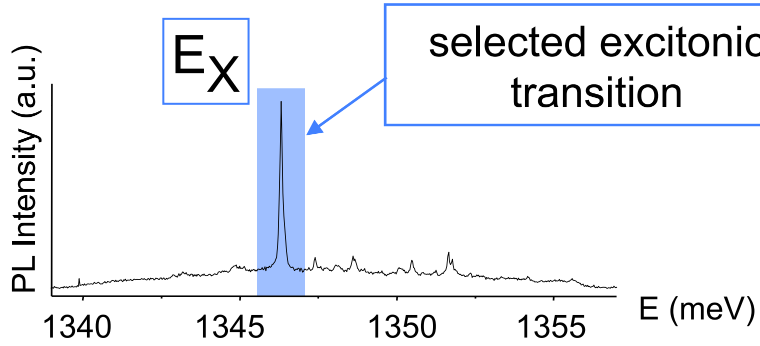
Selecting the appropriate radius



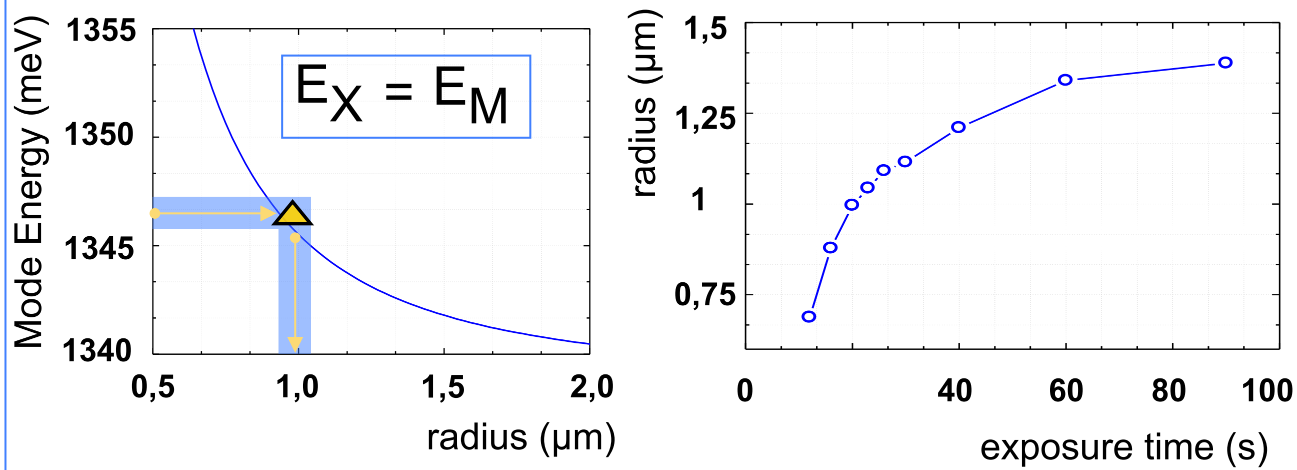
Spectral matching



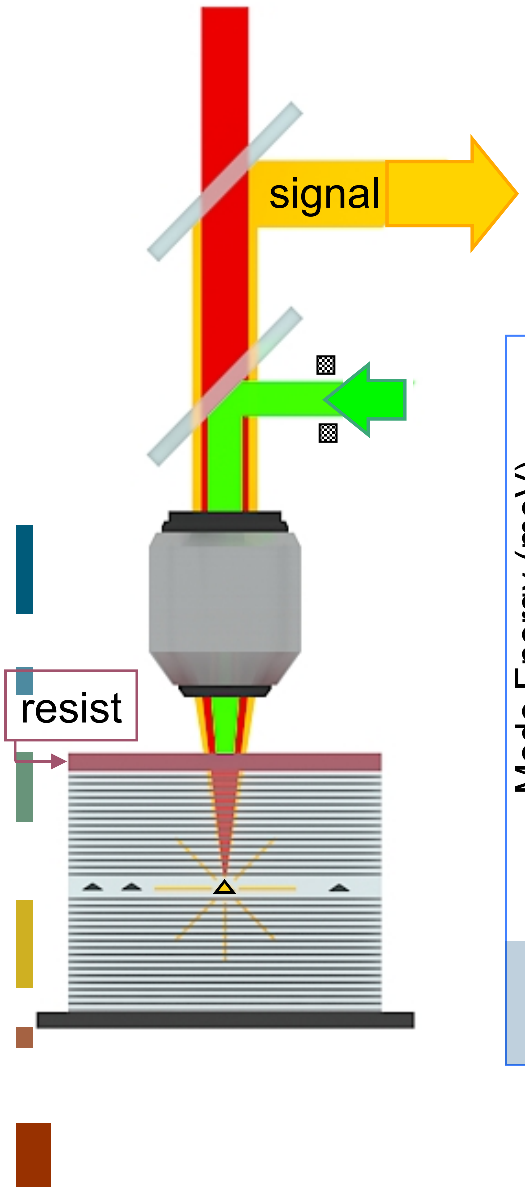
Spectrometer
+ CCD



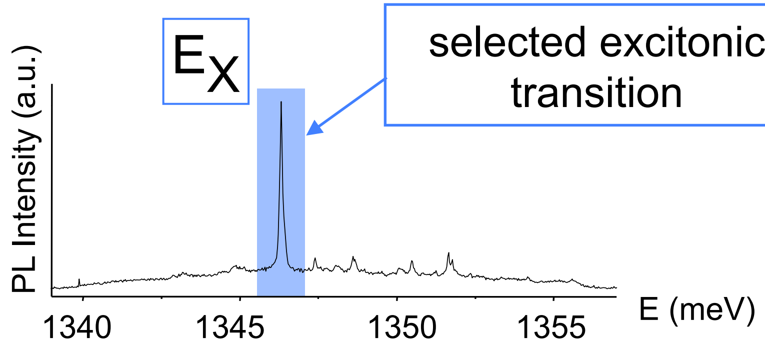
Selecting the appropriate radius



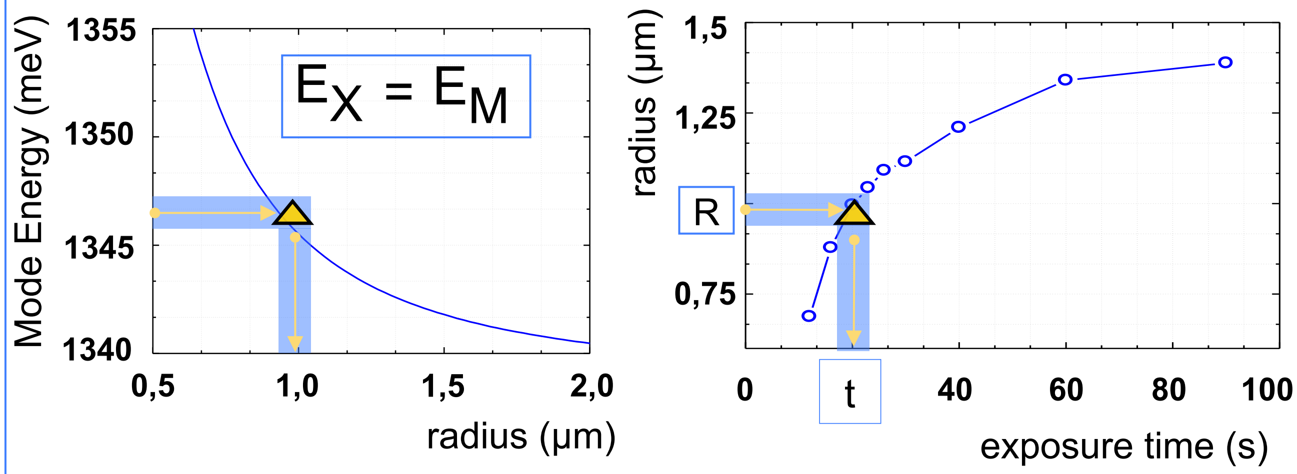
Spectral matching



Spectrometer
+ CCD

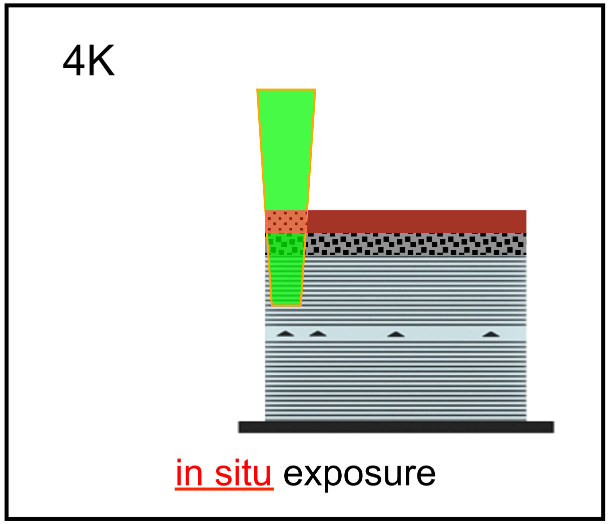


Selecting the appropriate radius

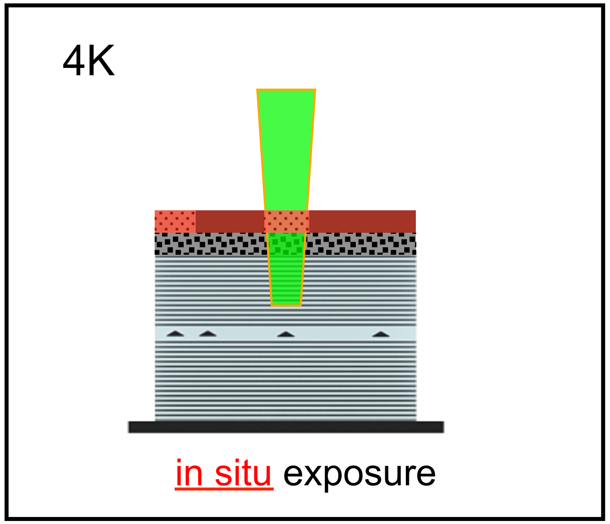


- non linear exposure dose → • pillar radius → • mode energy

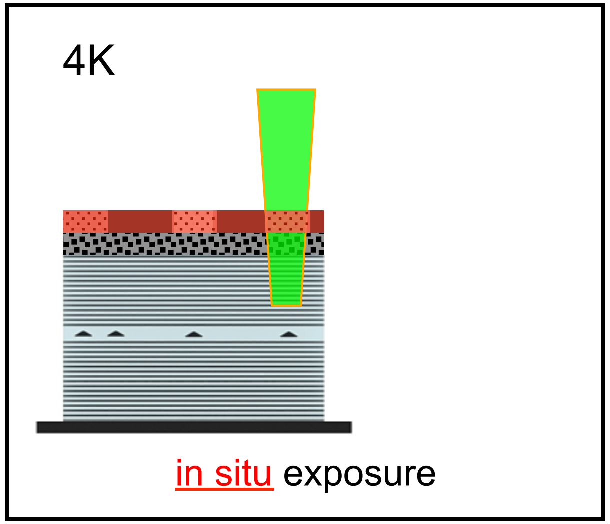
Post processing



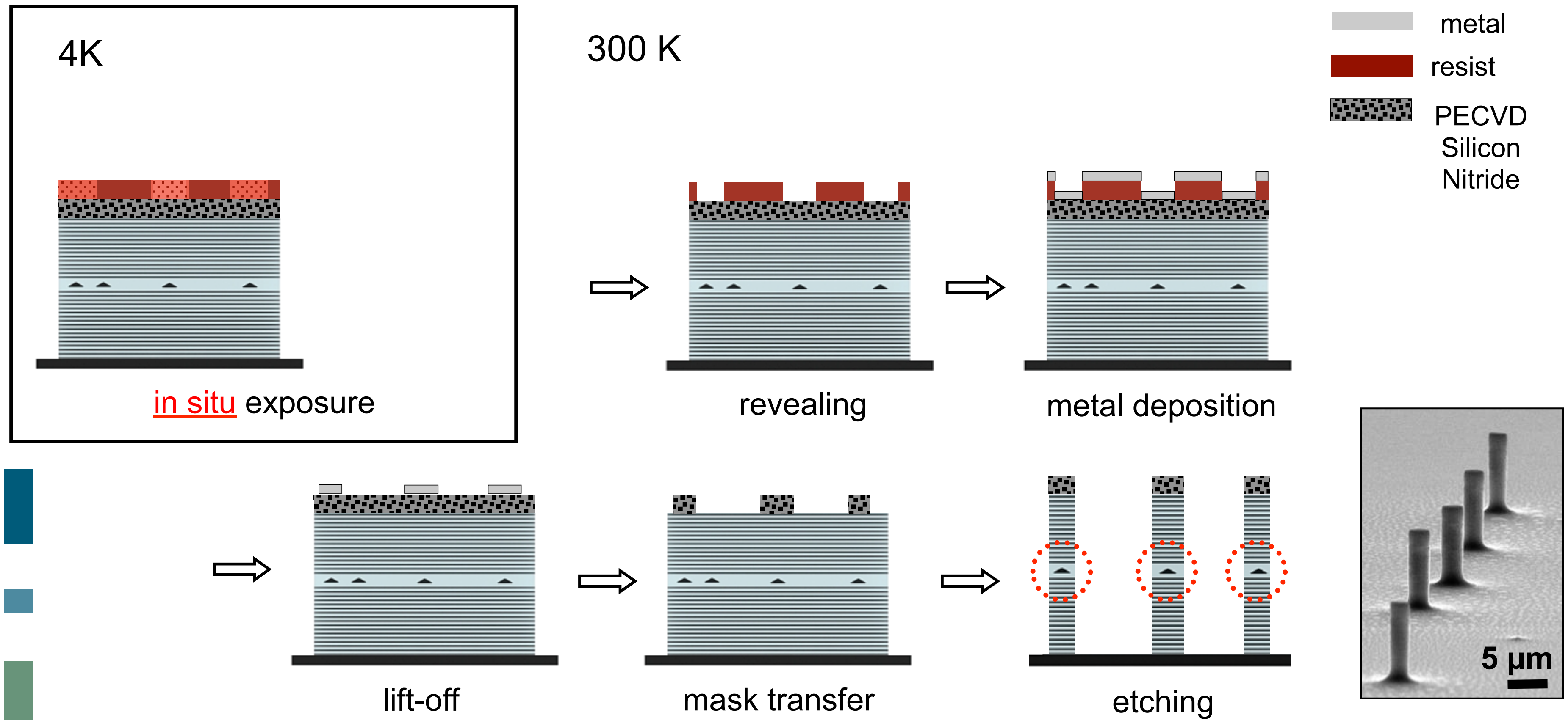
Post processing



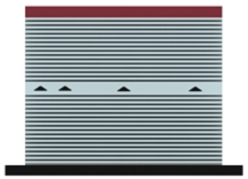
Post processing



Post processing



On demand weak coupling regime

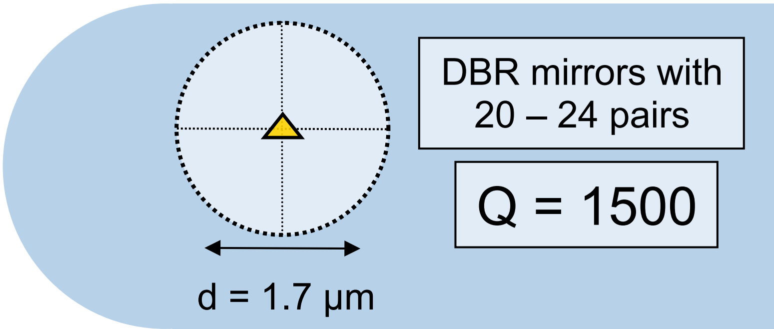


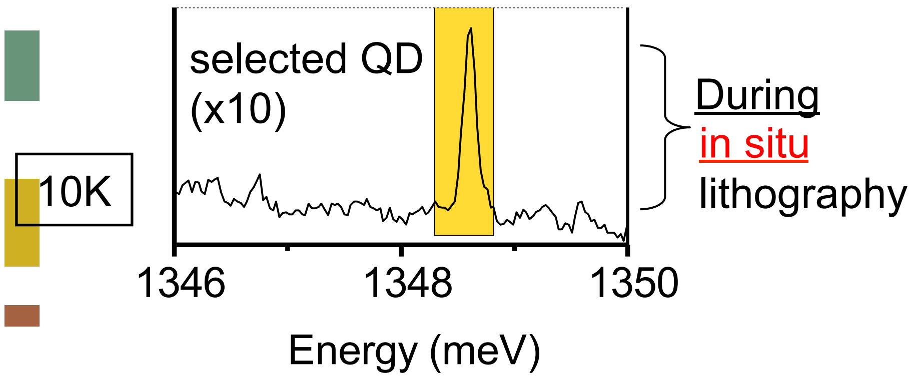
DBR mirrors with 20-24 pairs

Planar Cavity factor : Q = 5000

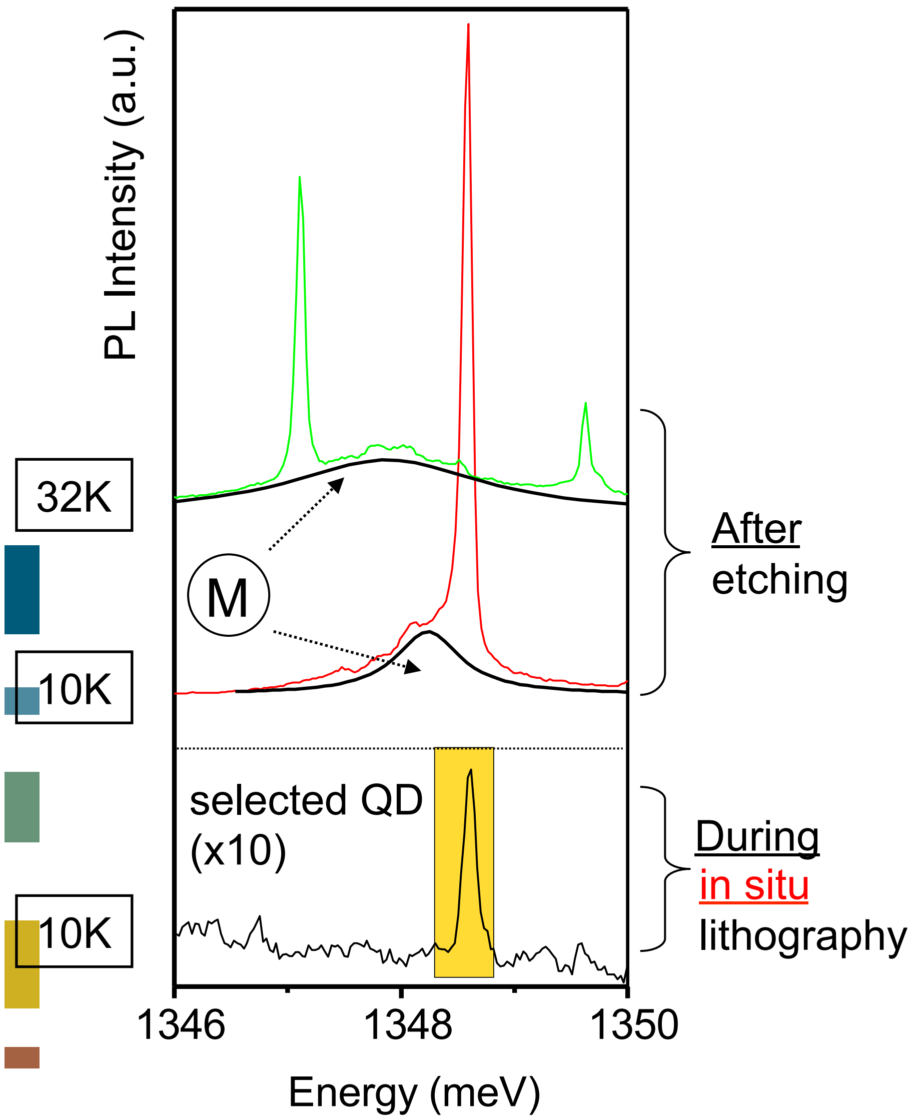
A. Dousse, et al., **Phys. Rev. Lett. 101, 267404 (2008)**
Research Highlight, **Nature Materials 8, 86 (2009)**

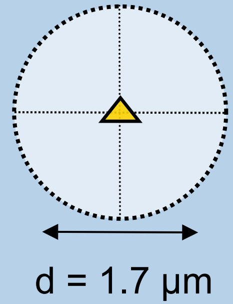
On demand weak coupling regime





On demand weak coupling regime

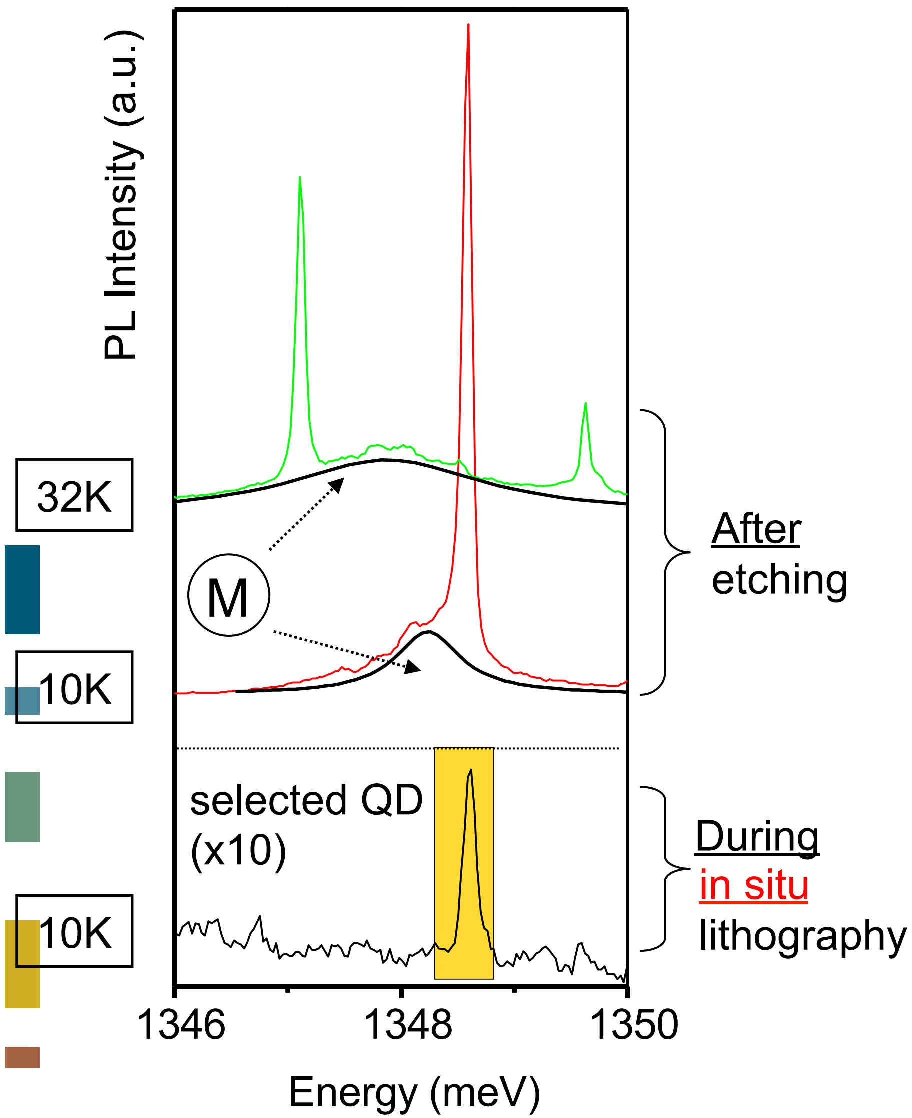


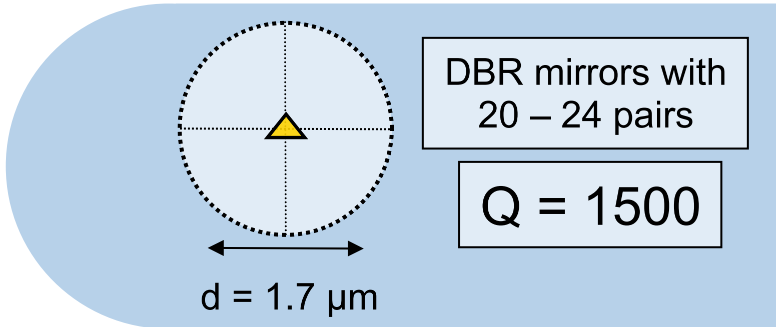


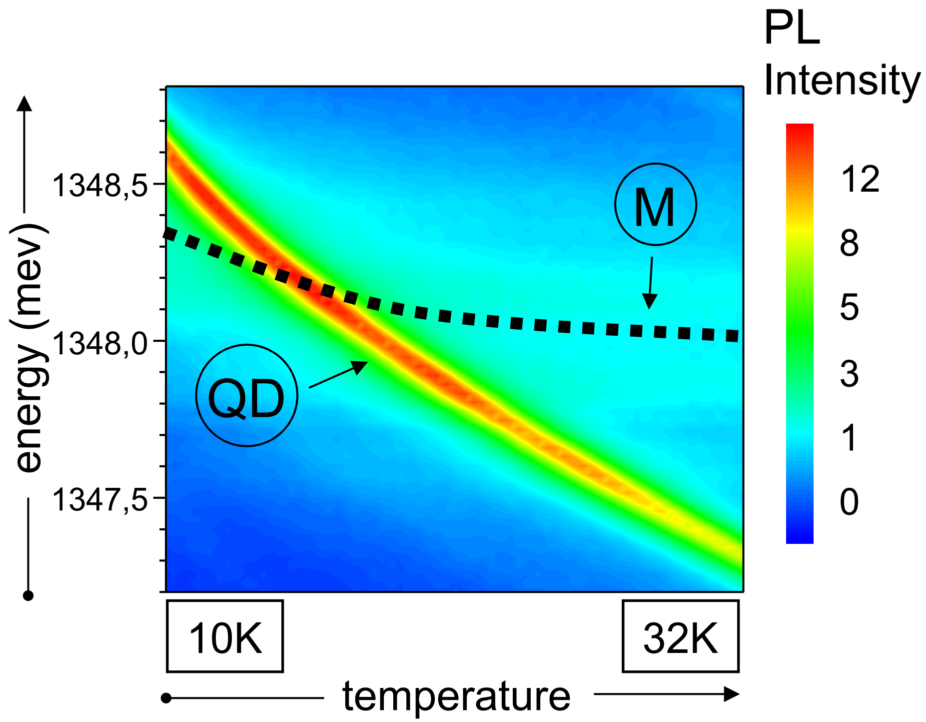
DBR mirrors with 20 – 24 pairs

Q = 1500

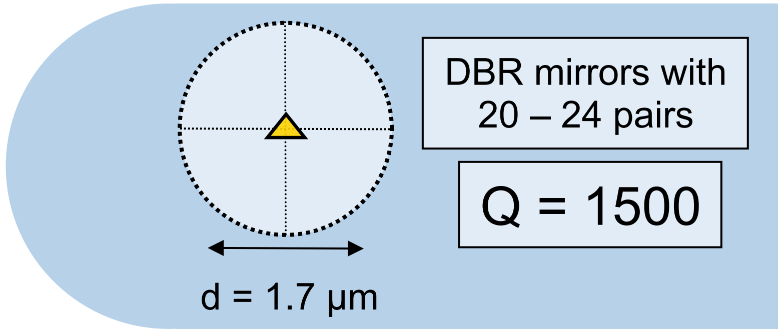
On demand weak coupling regime

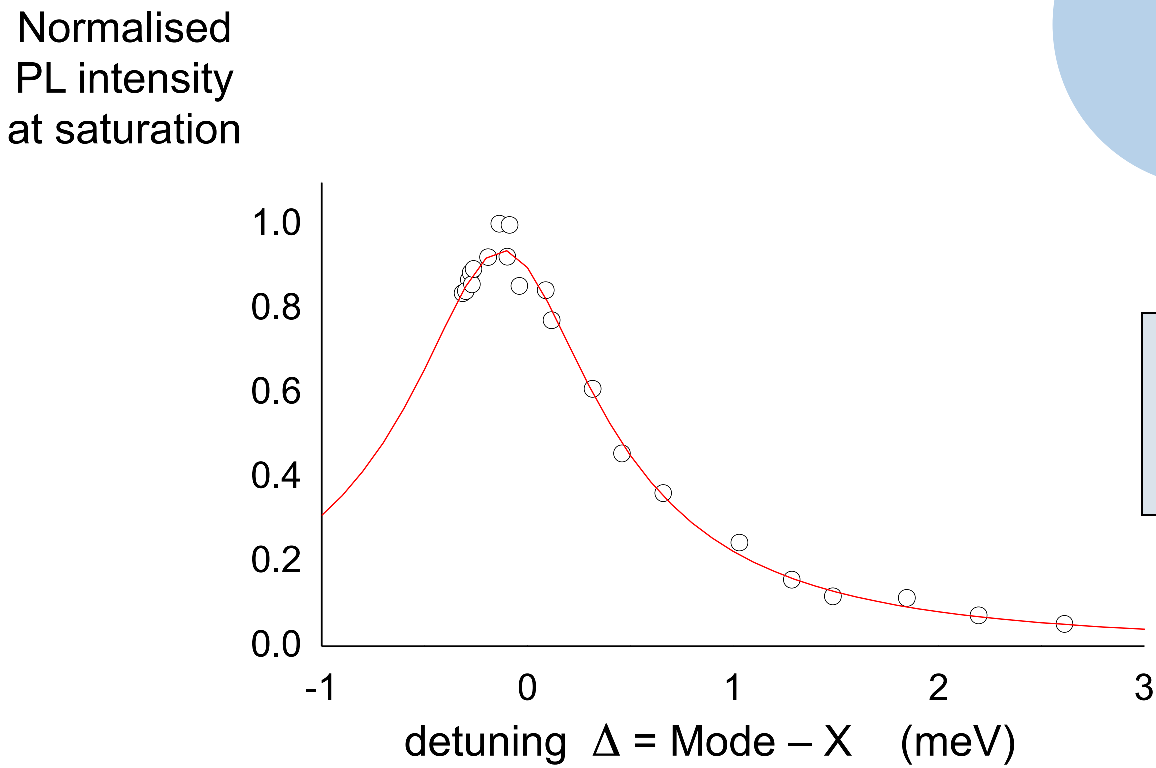






On demand weak coupling regime





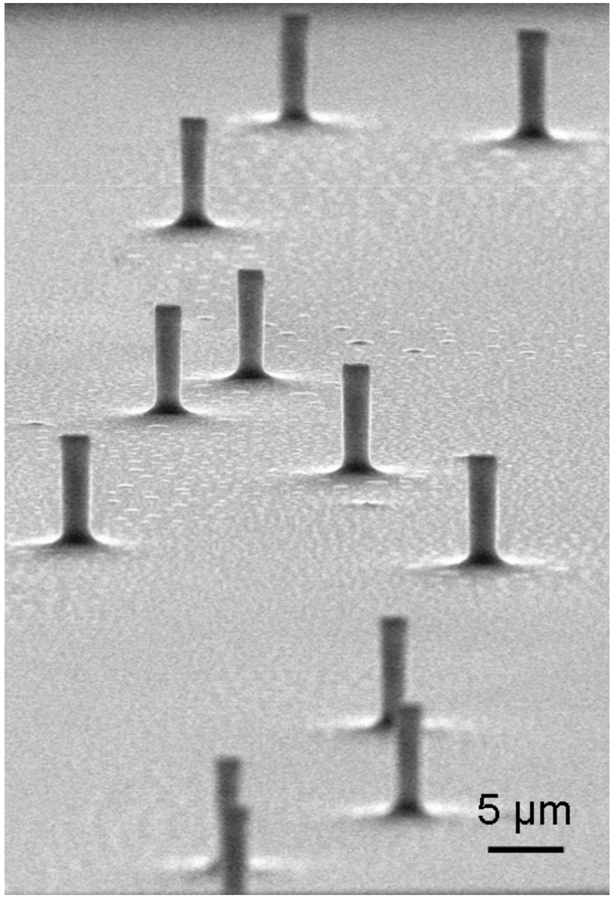
measured : $F_p = 9 \pm 3$

expected : $F_p = 9.5$

Good Spatial Matching !

H. Lohmeyer et al., Appl. Phys. Lett. 92, 011116 (2008).

Scalability

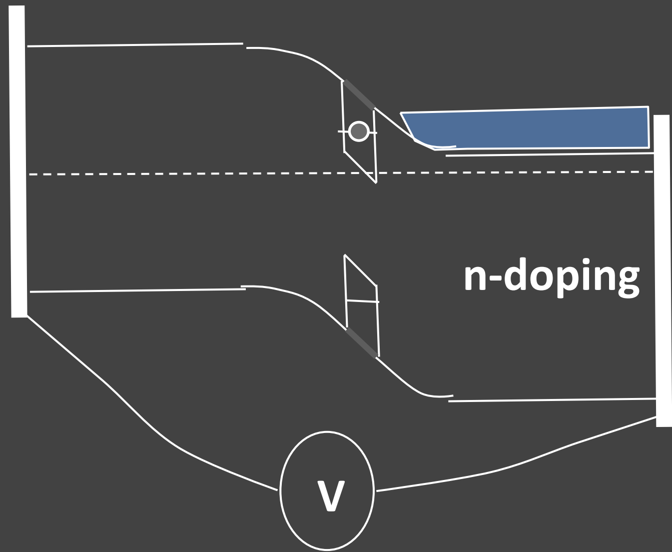


Around 40 pillars in one lithography process

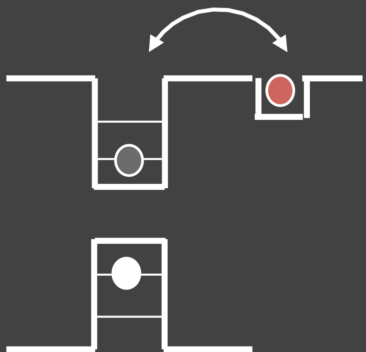
A. Dousse, et al., **Phys. Rev. Lett. 101, 267404 (2008)**
Research Highlight, **Nature Materials 8, 86 (2009)**

LABORATOIRE DE PHOTONIQUE ET DE NANOSTRUCTURES

Electric control



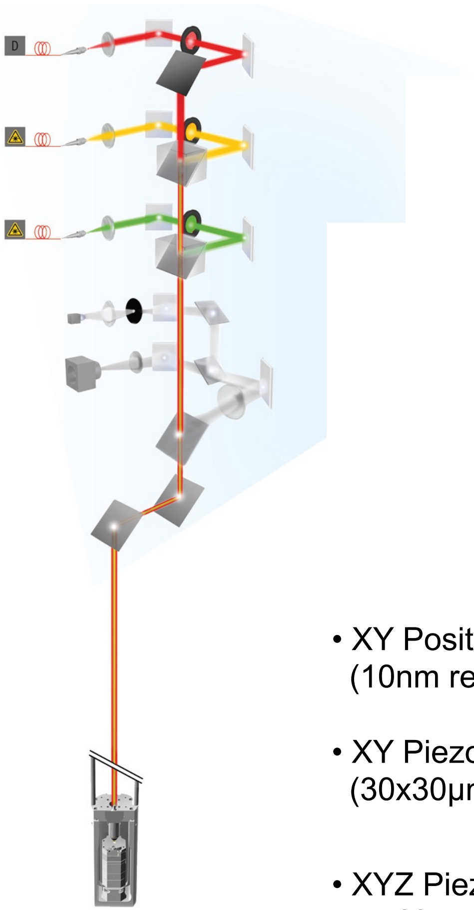
Reduce influence of
outside residual charge

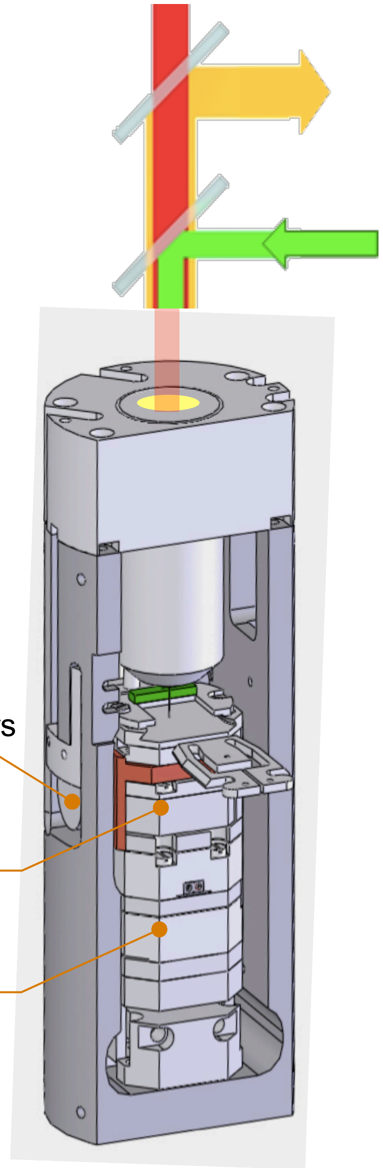


Control the charge
inside the dot

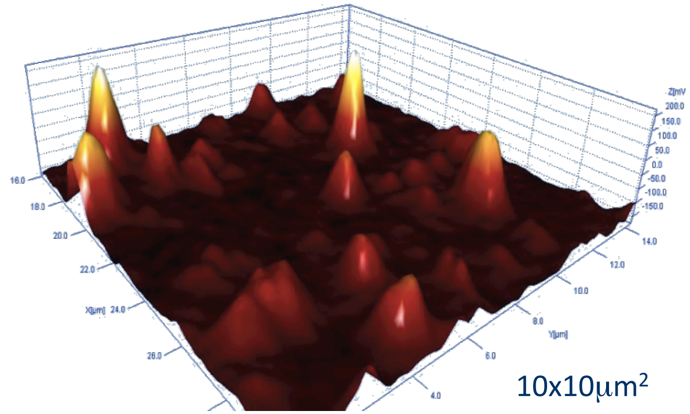
Tune the QD
emission energy

In situ optical lithography machine





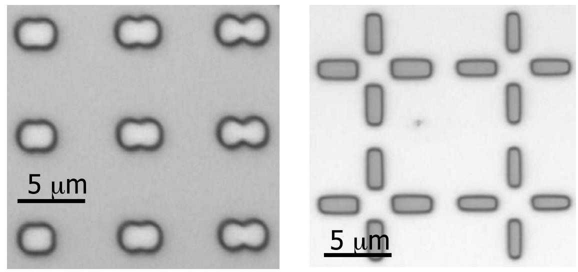
- XY Position sensors (10nm resolution)
- XY Piezo scanner (30x30µm at 4K)
- XYZ Piezo stepper positioners (5x5x5 mm)



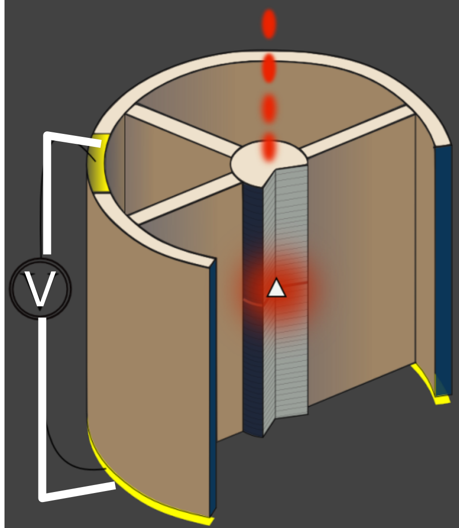
10x10μm^2

Absolute QD position
10 nm

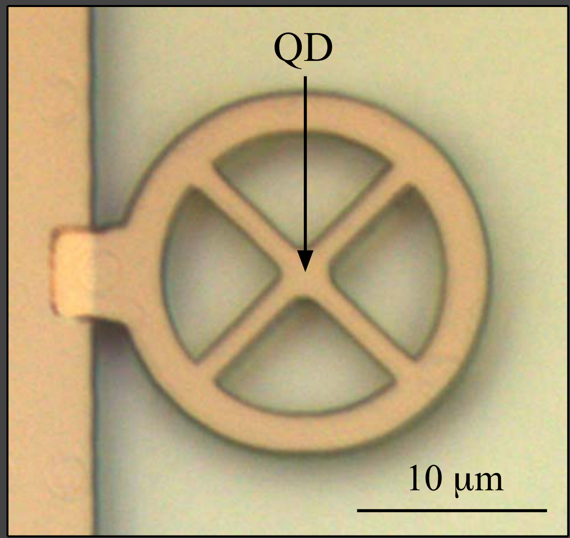
Lithography patterns



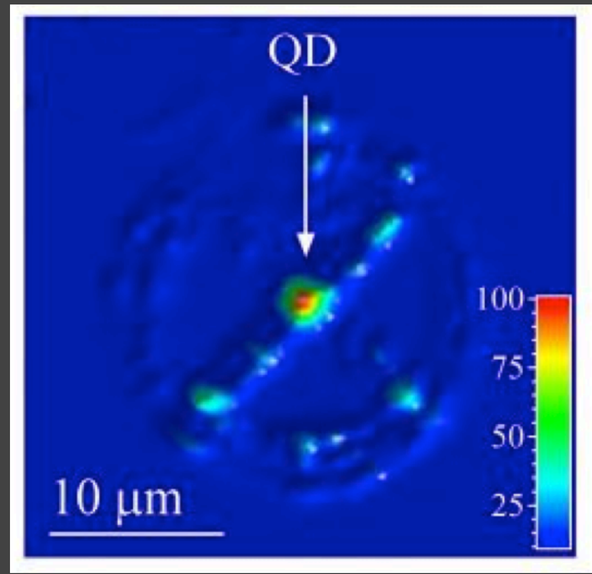
An electrical device



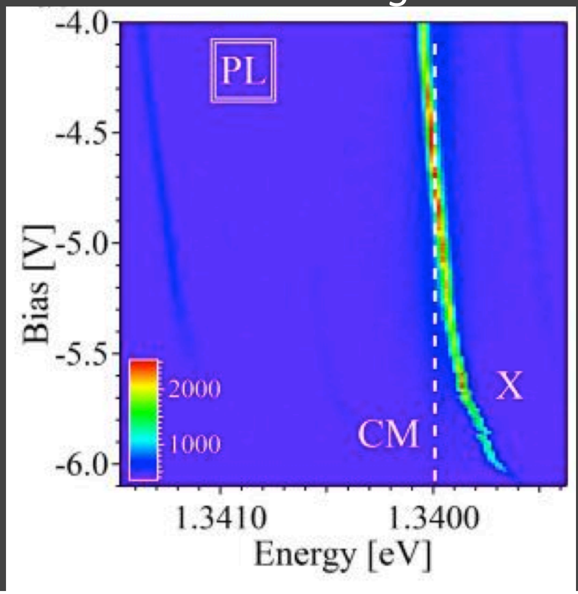
Optical microscope



Emission mapping



Emission tuning



Electrical control

Brightness > 55%

Nowak et al,
Nature Communication 2014

Conclusions

Material science does drive new fondamental studies