Single molecule controlled emission in planar plasmonic cavity

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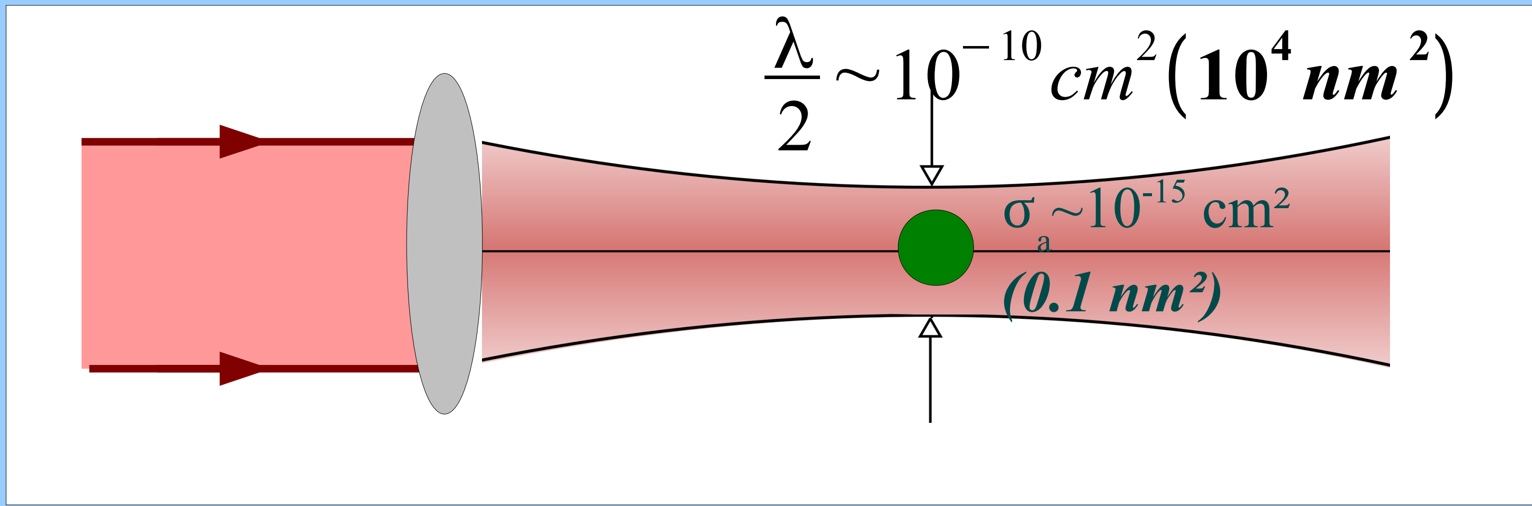
CNRS/Université de Bourgogne
Dijon - France

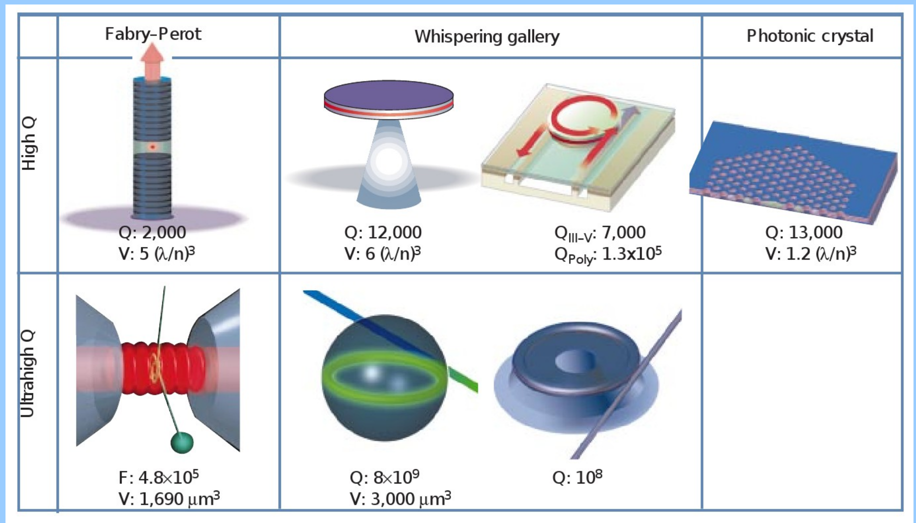
Nanolight
Benasque – March 2014

Light/matter interaction at the nanoscale



Strategies (at ambient T°C)

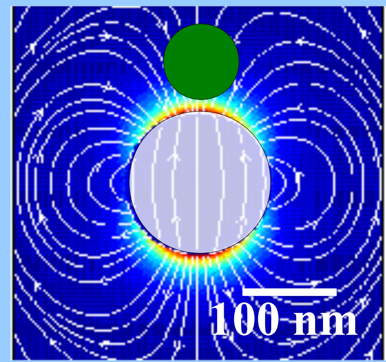
Cavity quantum electrodynamics (cQED)

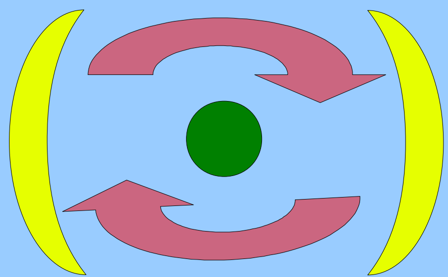


Purcell factor

$$F_p = \frac{\Gamma}{n_1 \Gamma_{tot}} = \frac{3}{4\pi^2}\left(\frac{\lambda}{n_1}\right)^3 \frac{Q}{V_{eff}}$$

Surface enhanced spectroscopies (SERS, SEF)

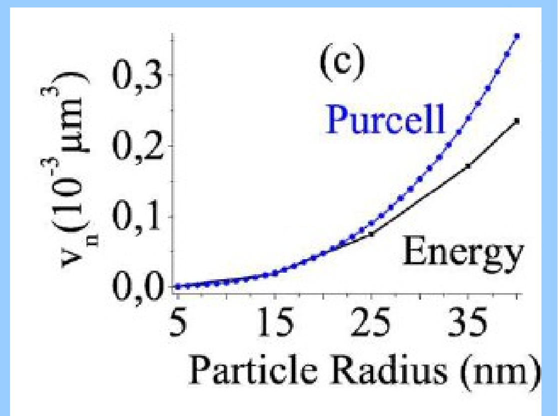




Interaction Duration (high Q)

Interaction volume

$$V_{SPP} \sim \frac{4}{3}\pi R^3 \ll \left(\frac{\lambda}{n_1}\right)^3$$



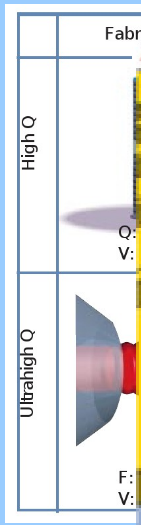
Optical microcavities (Review)
Vahala, Nature **424**, 839 (2003)

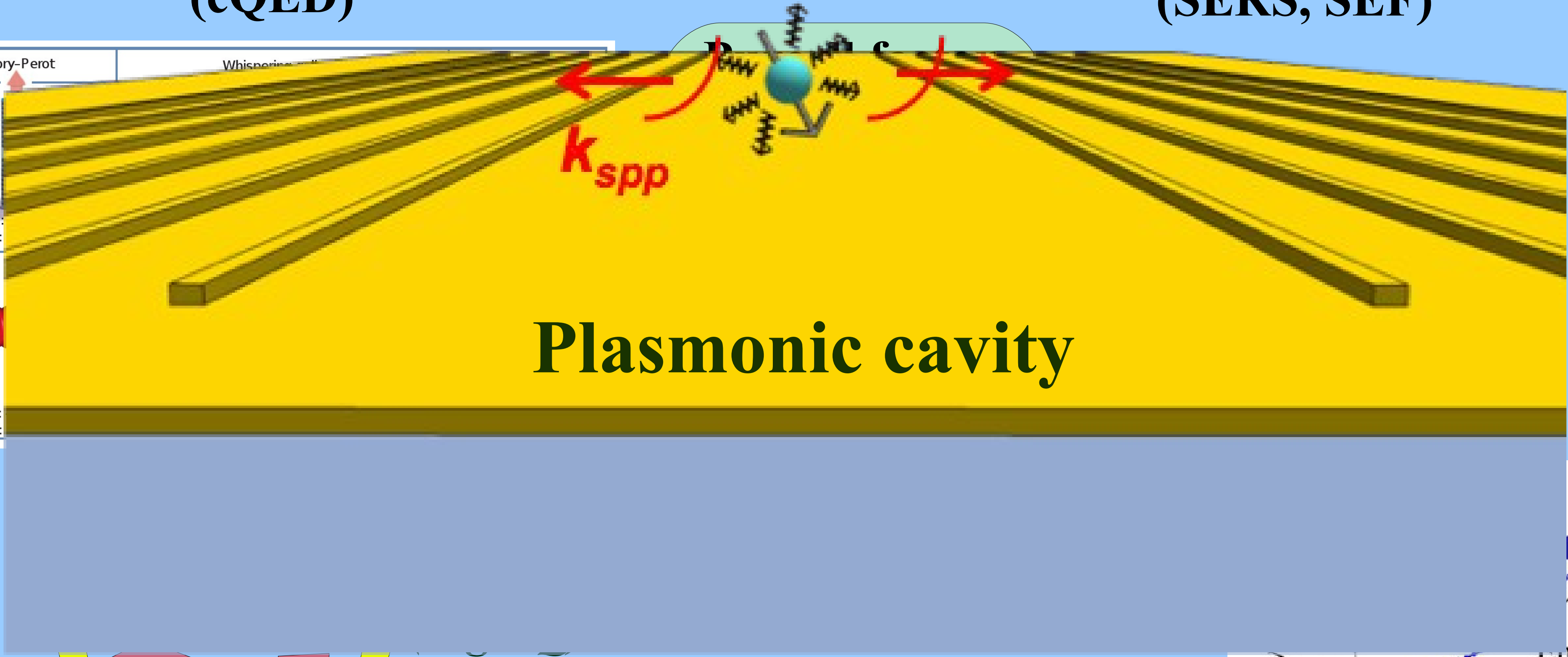
Resonance quality, radiative/ohmic losses and modal volume of Mie plasmons,
Derom *et al*, EPL **98**, 47008 (2012)

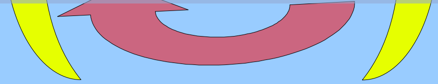
Strategies (at ambient T°C)

Cavity quantum electrodynamics (cQED)

Surface enhanced spectroscopies (SERS, SEF)







$$\ll \left(\frac{\lambda}{n_1}\right)^3$$

Optical microcavities (Review)
Vahala, Nature **424**, 839 (2003)

Resonance quality, radiative/ohmic losses and modal volume of Mie plasmons, Derom *et al*, EPL **98**, 47008 (2012)

Low threshold/thresholdless laser

$$\beta = \frac{\Gamma_{cav}}{\Gamma_{tot}} \approx \frac{F_p}{1 + F_p}$$

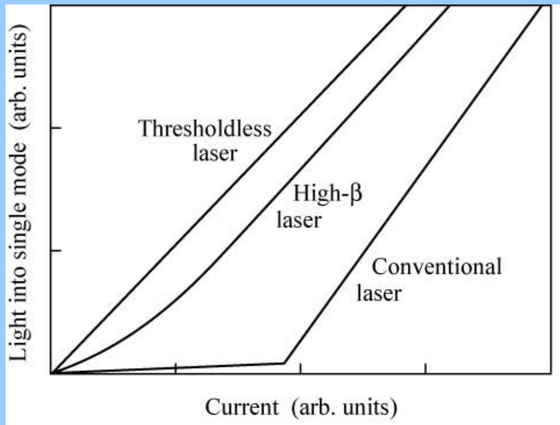
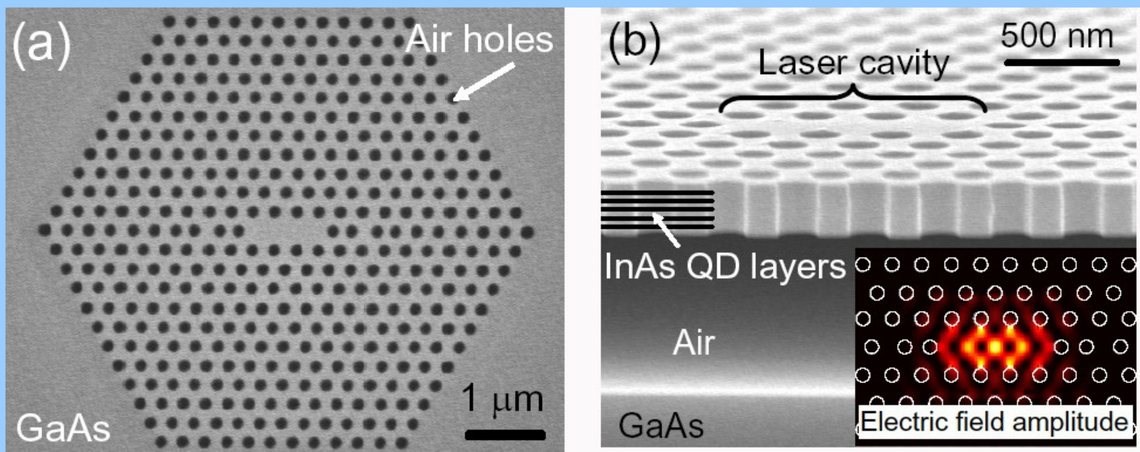
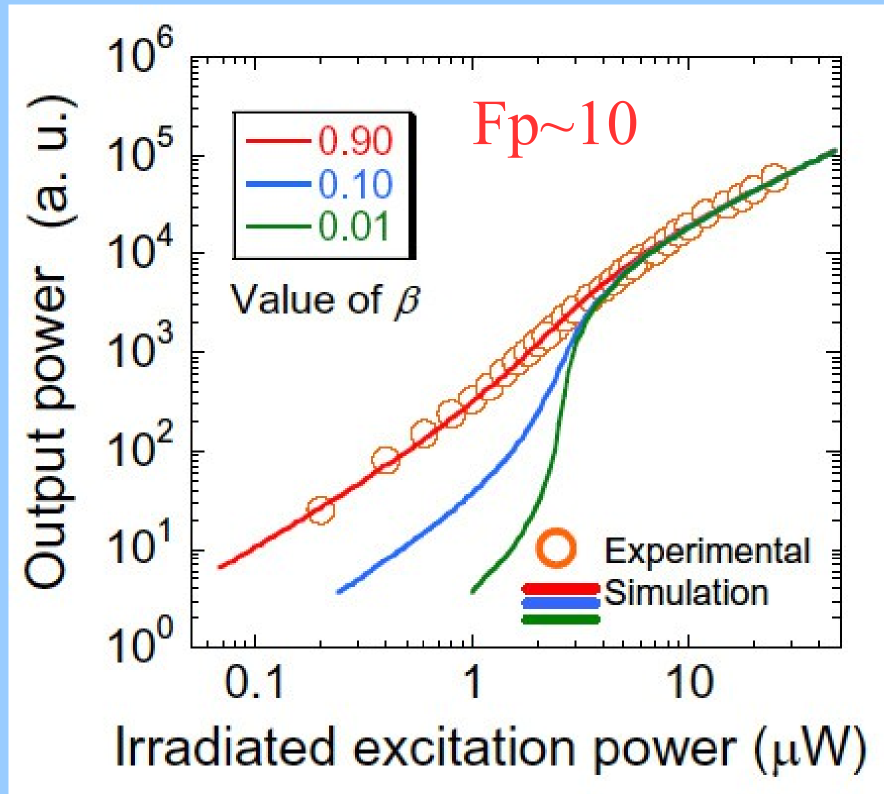


Fig. 15.14. Light-power-versus-current curves for single spatial-mode emission from a (*i*) conventional laser, (*ii*) a high β-factor laser, and (*iii*) a thresholdless laser. The conventional laser has a distinct current threshold. The high β-factor laser has a less distinct threshold. It would be noticeable in the spectrum and device modulation speed, however. A hypothetical thresholdless laser would have a β close to 1, and would somehow suppress all other lossy emission until the carrier density required for gain (or at least transparency) was achieved.

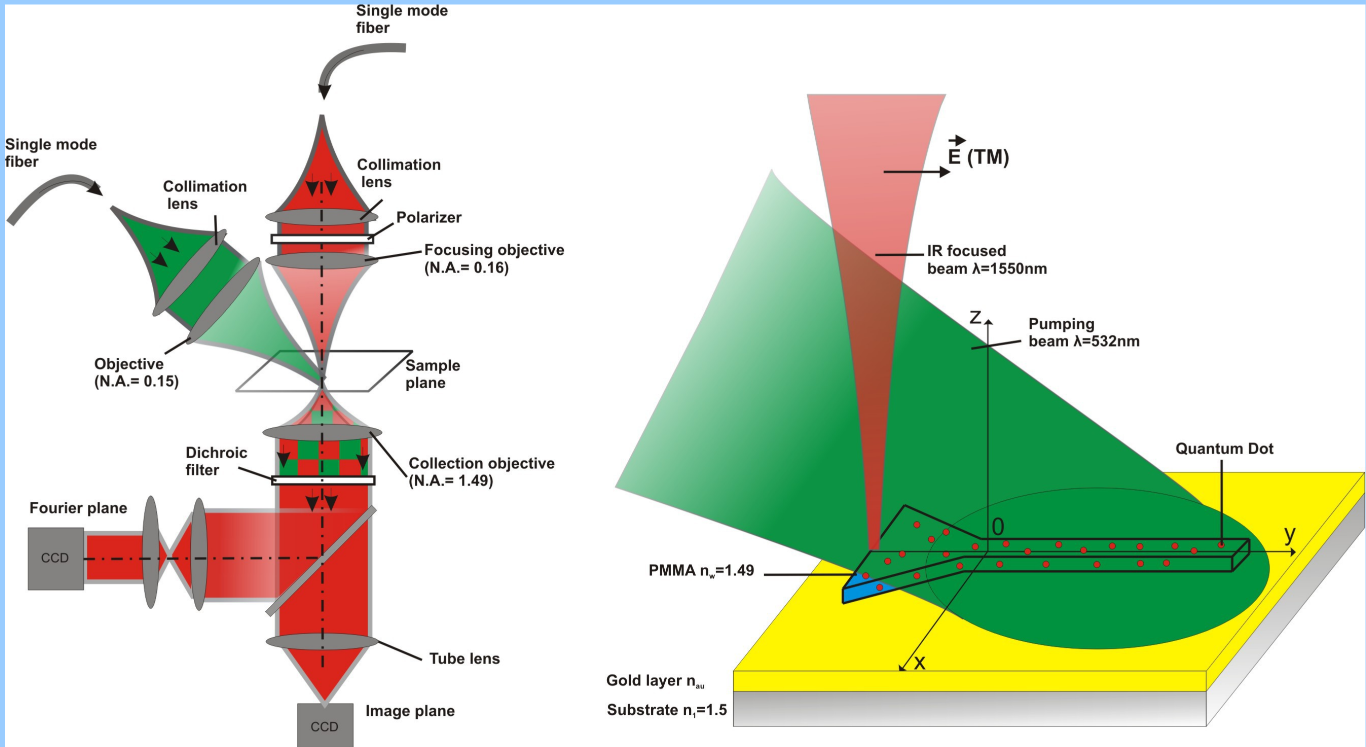
E. F. Schubert
Light-Emitting Diodes (Cambridge Univ. Press)
www.LightEmittingDiodes.org

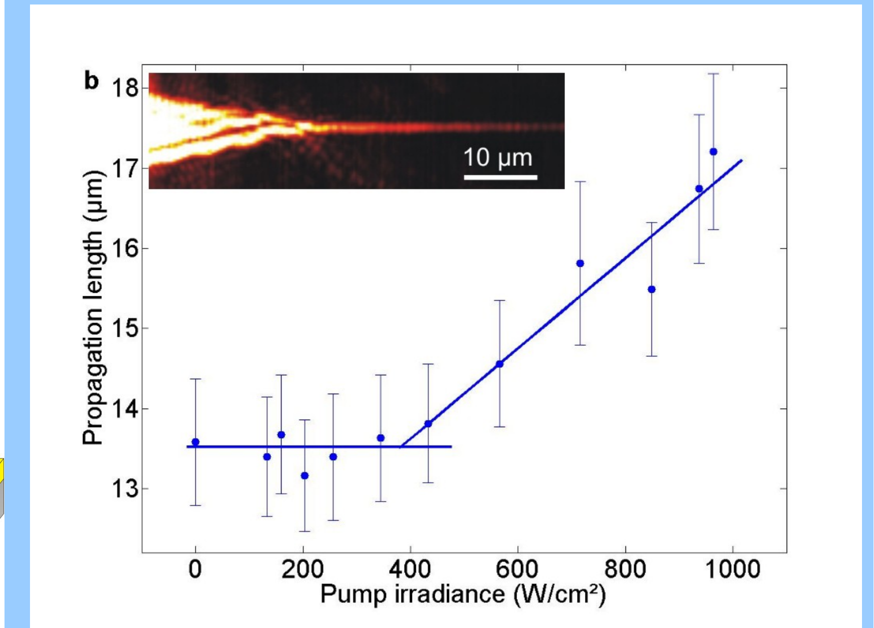


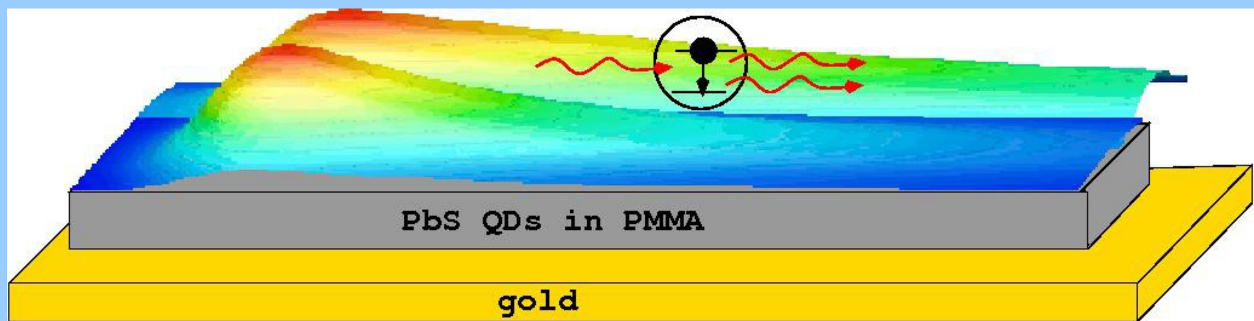


A photonic criystal nanocavity with ultralow threshold
Nomura, Iwatomoto,Arakawa, SPIE (2007)

Gain-assisted propagation

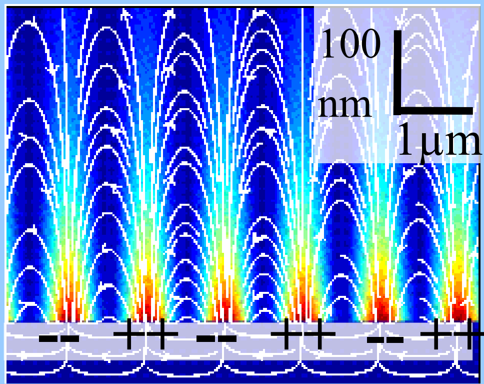


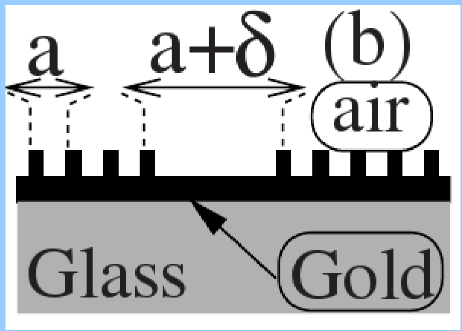


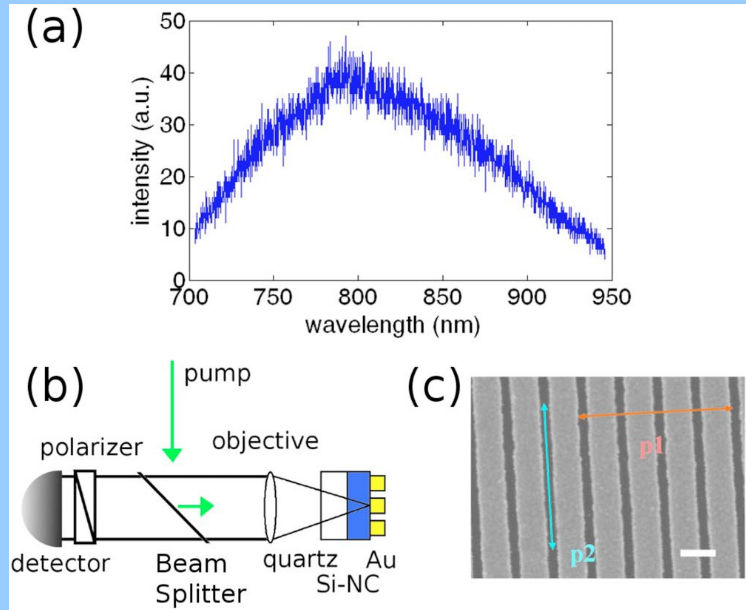


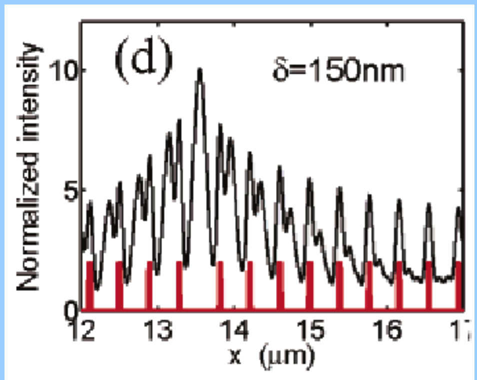
Gain-Assisted Propagation in a Plasmonic Waveguide at Telecom Wavelength
Grandidier *et al*, Nano Letters **9**, 2935 (2009)

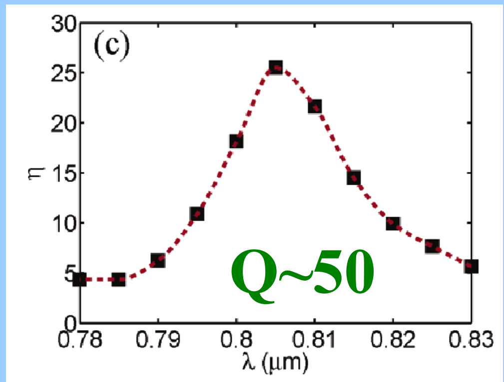
In-plane plasmonic cavity

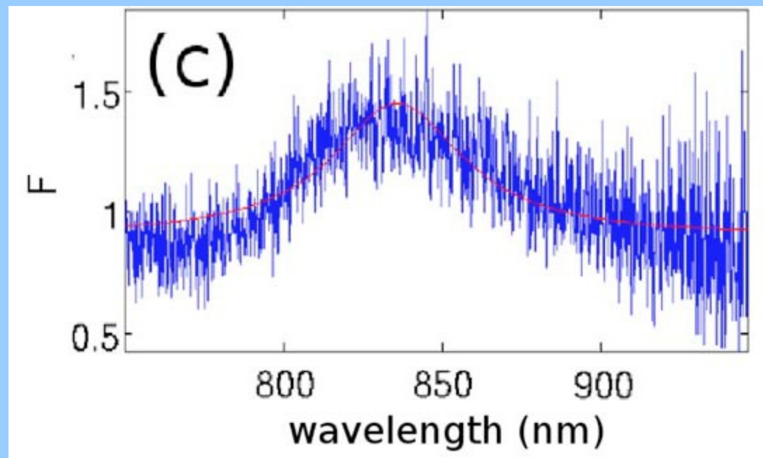












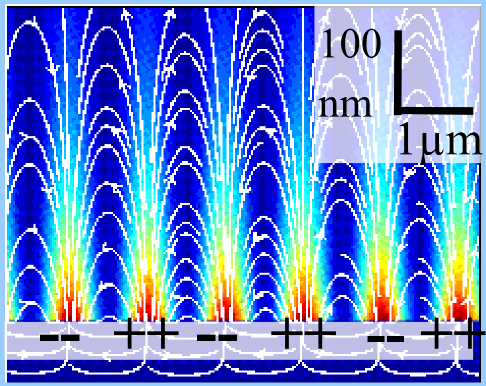
Mode confinement
Light extraction (LED, ...)

Submicrometer In-Plane Integrated Plasmon Cavities
Weeber *et al*, Nano Lett. **7**, 1352 (2007)

Gong *et al*, APL **94**, 013106 (2009)

Plasmonic Purcell factor

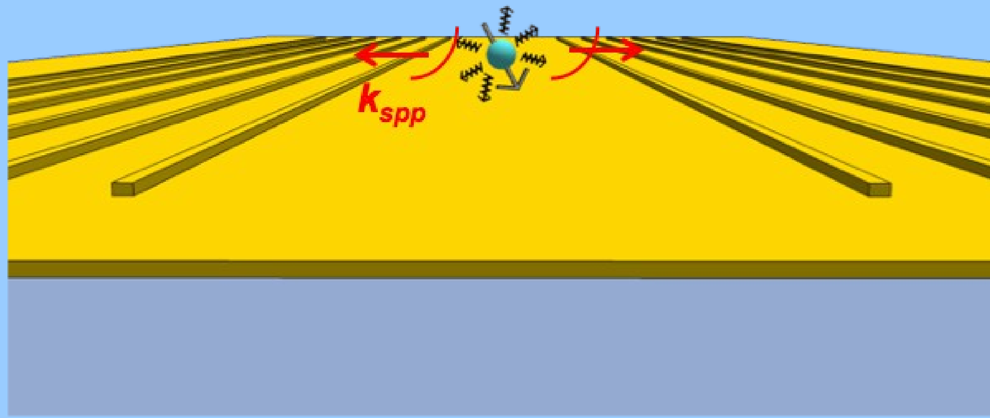
Flat film



$$Q = \frac{k_{SPP}}{\Delta k_{SPP}} = k_{SPP} L_{SPP}$$
$$\approx 100$$

$$V_{SPP} \approx \delta L_{SPP}^2 \approx 30 \left(\frac{\lambda}{n}\right)^3$$

Single mode cavity

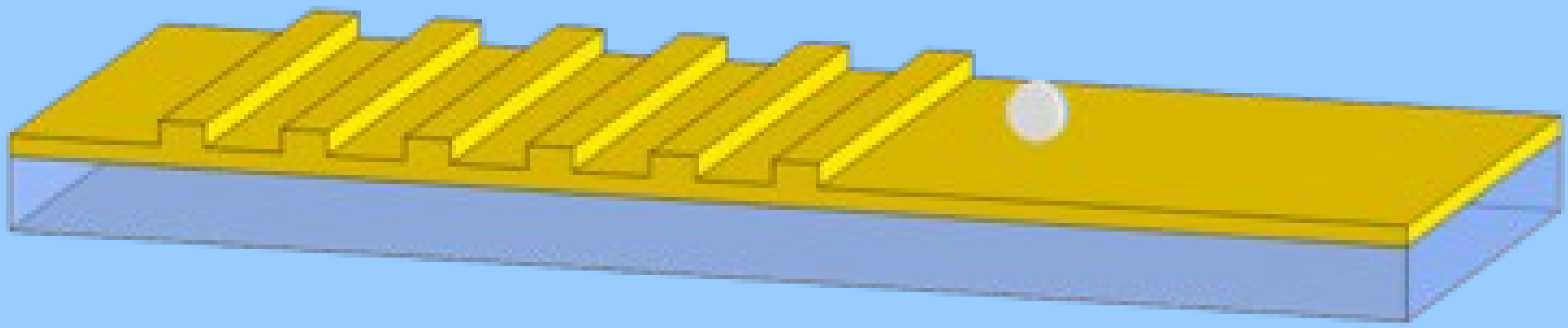


$$V_{SPP} \approx \delta L_{SPP} L_{cav}$$
$$\approx 0.5 \left(\frac{\lambda}{n}\right)^3$$

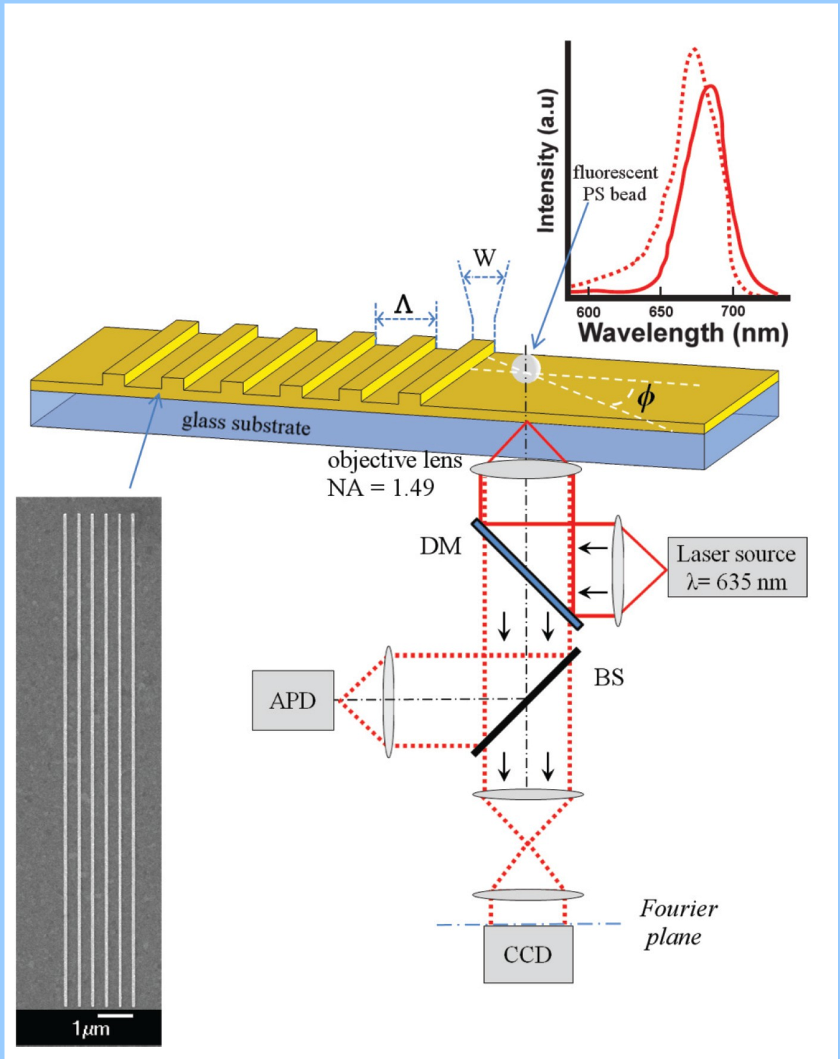
$$F_p \approx \frac{\Gamma_{SPP}}{n_1 \Gamma} = \frac{3}{4\pi^2} \left(\frac{\lambda}{n_1}\right)^3 \frac{Q}{V_{SPP}}$$
$$\approx 15$$

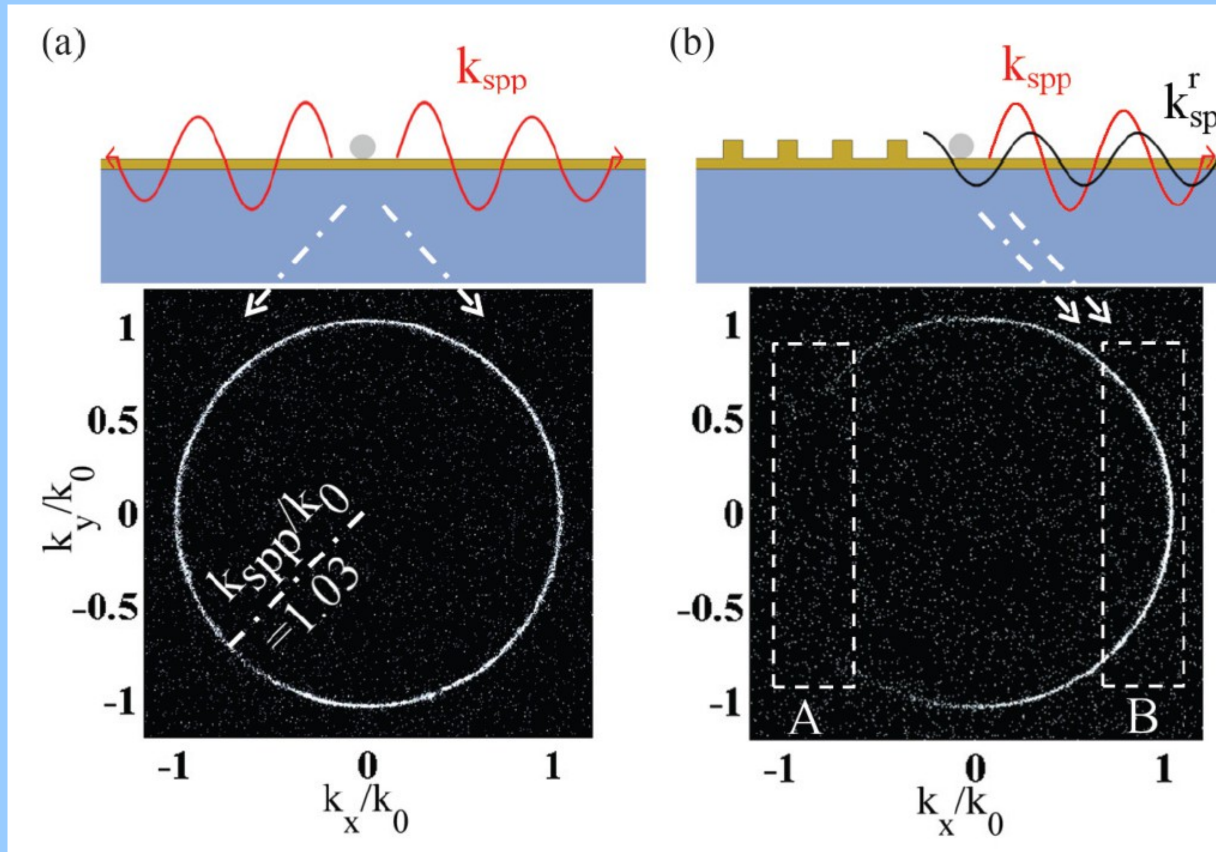
see also
Coupling of a dipolar emitter into one-dimensional surface plasmon
Barthes *et al,* Sci. Rep. **3**, 2734 (2013)

Plasmonic Bragg mirror



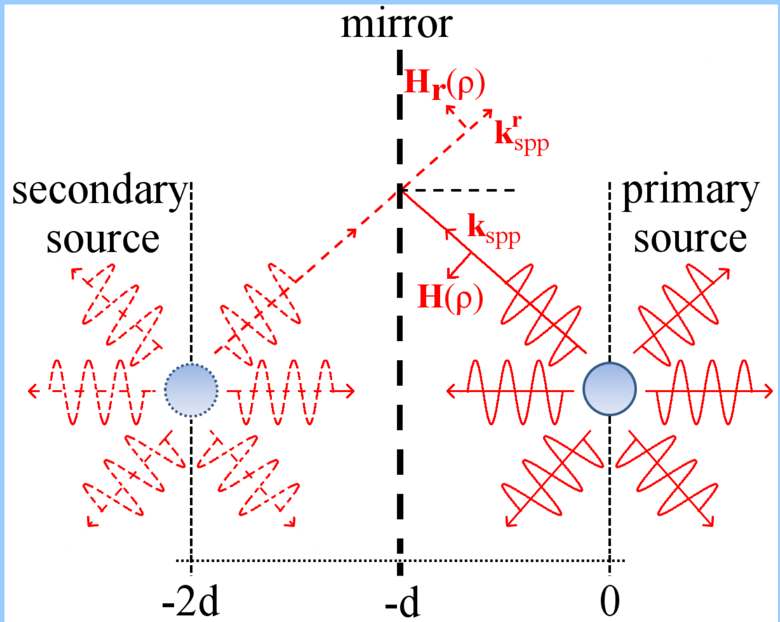
Surface plasmon coupled emission near a Bragg mirror

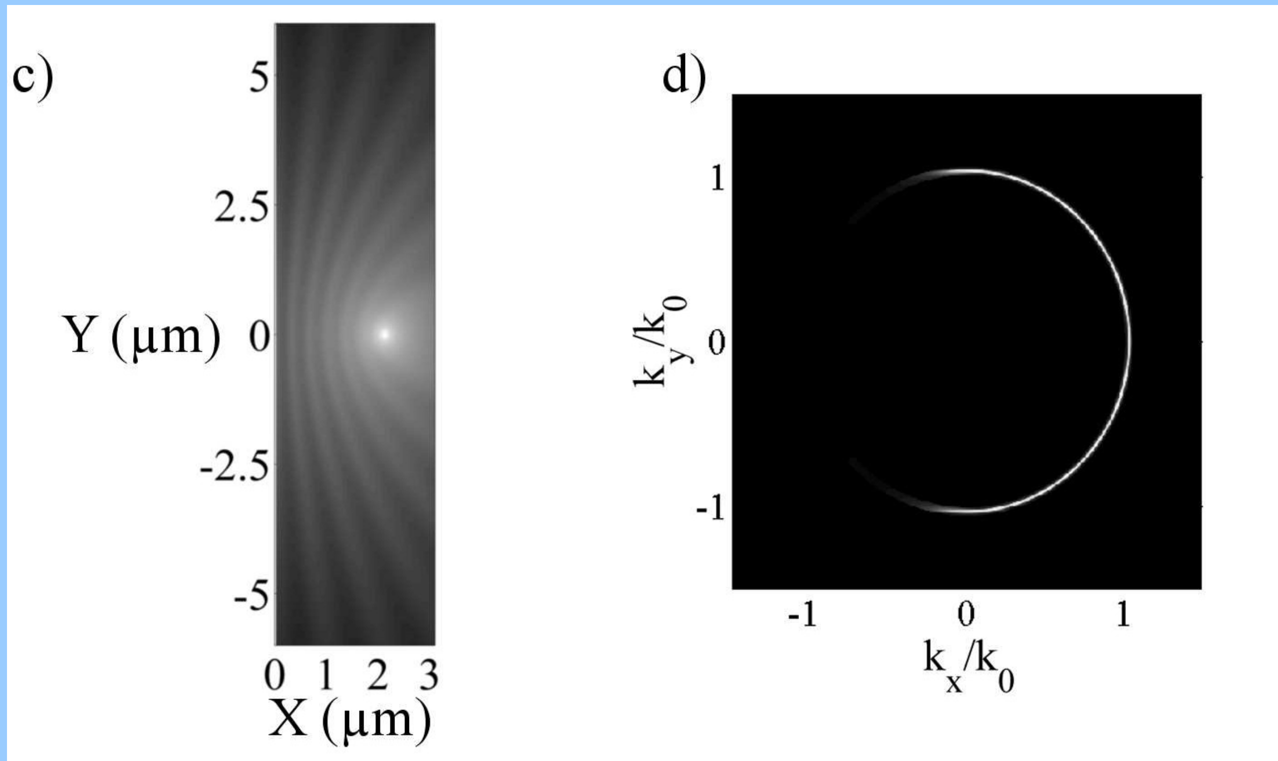




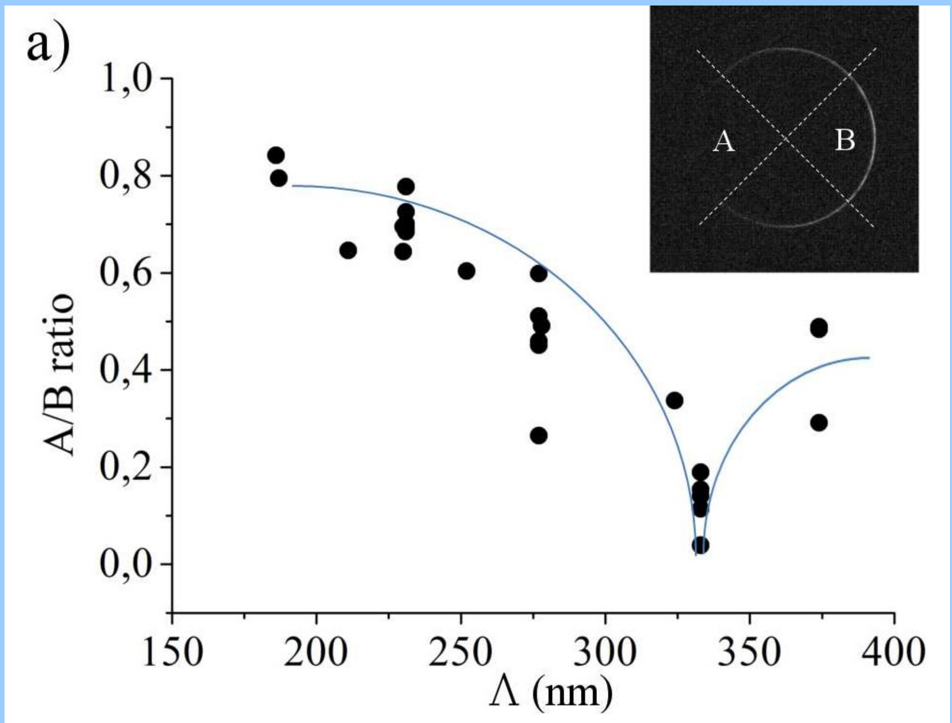
Single-molecule controlled emission in planar plasmonic cavities
Derom *et al*, Phys. Rev . B **89**, 035401 (2014)

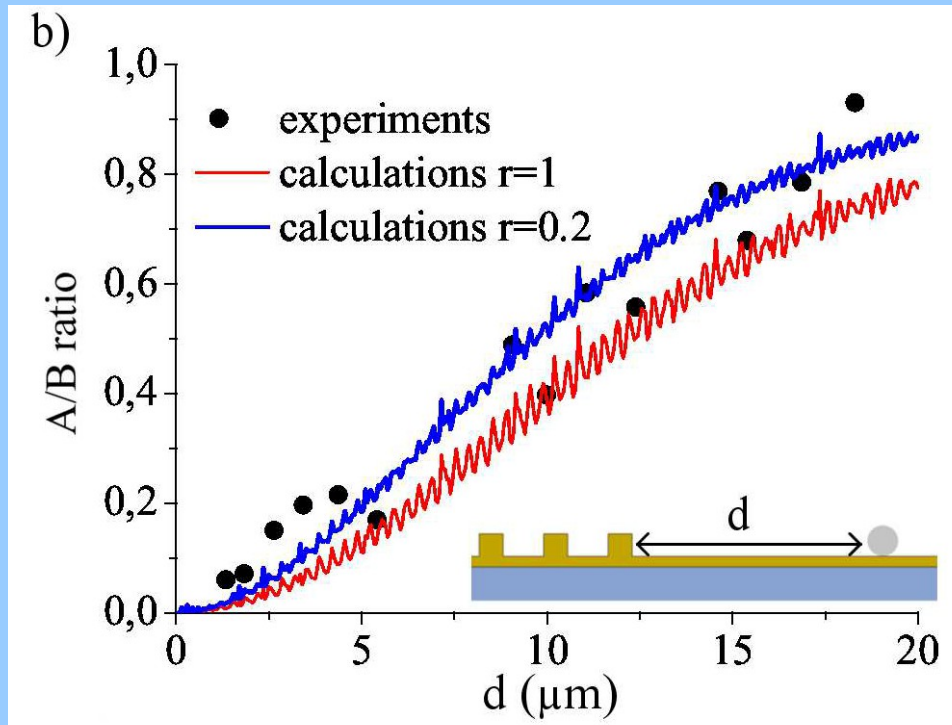
Mirror efficiency



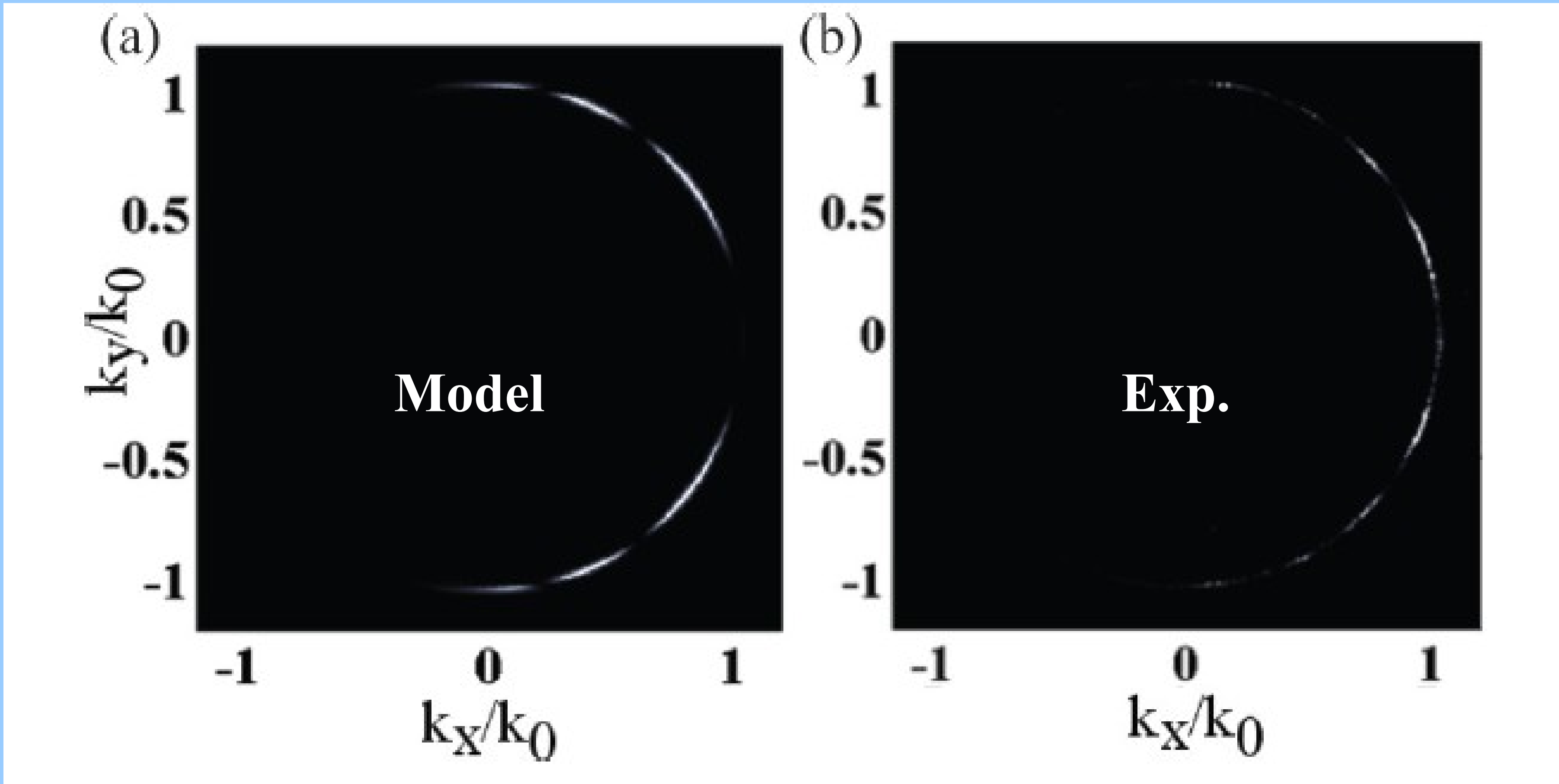


Grating period



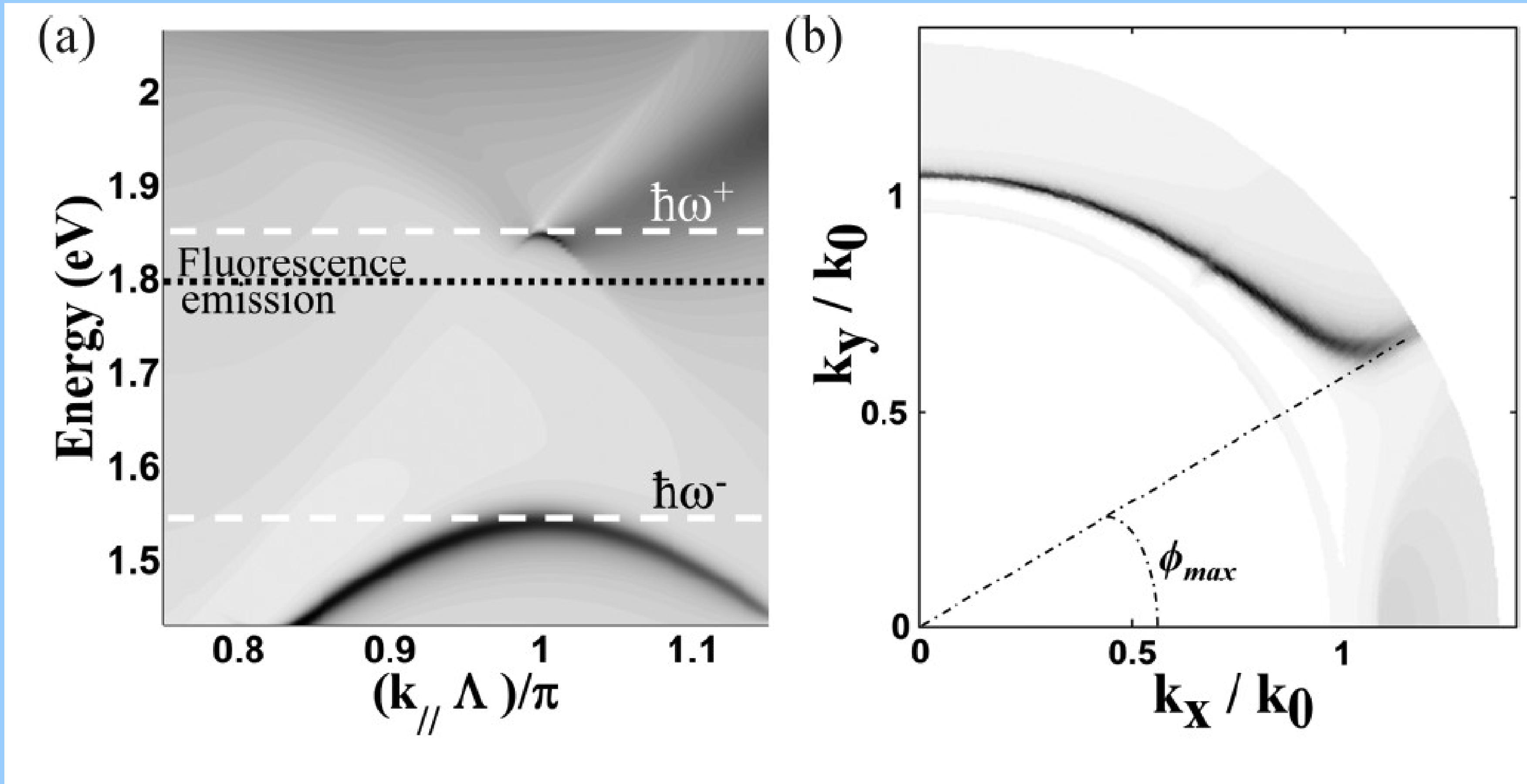


Surface plasmon coupled emission near a Bragg mirror



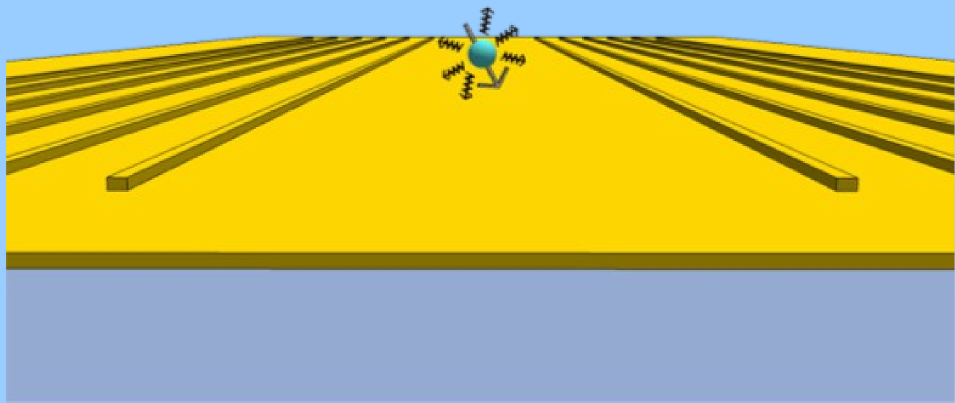
Spatial coherence of the localized nanosource

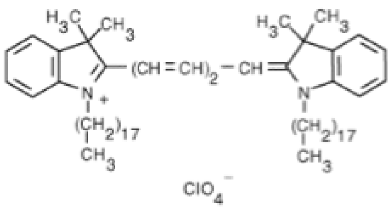
Mirror bandgap

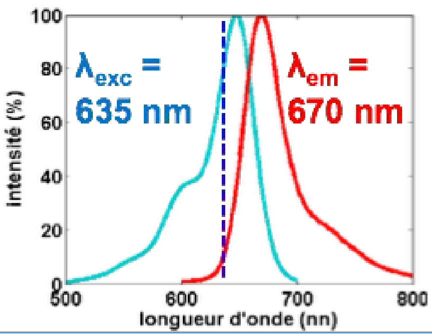


Large bandwidth system (675 nm < λ < 790 nm)

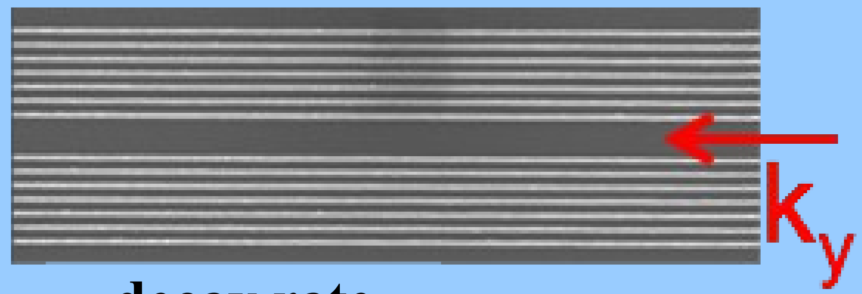
In plane SPP cavity





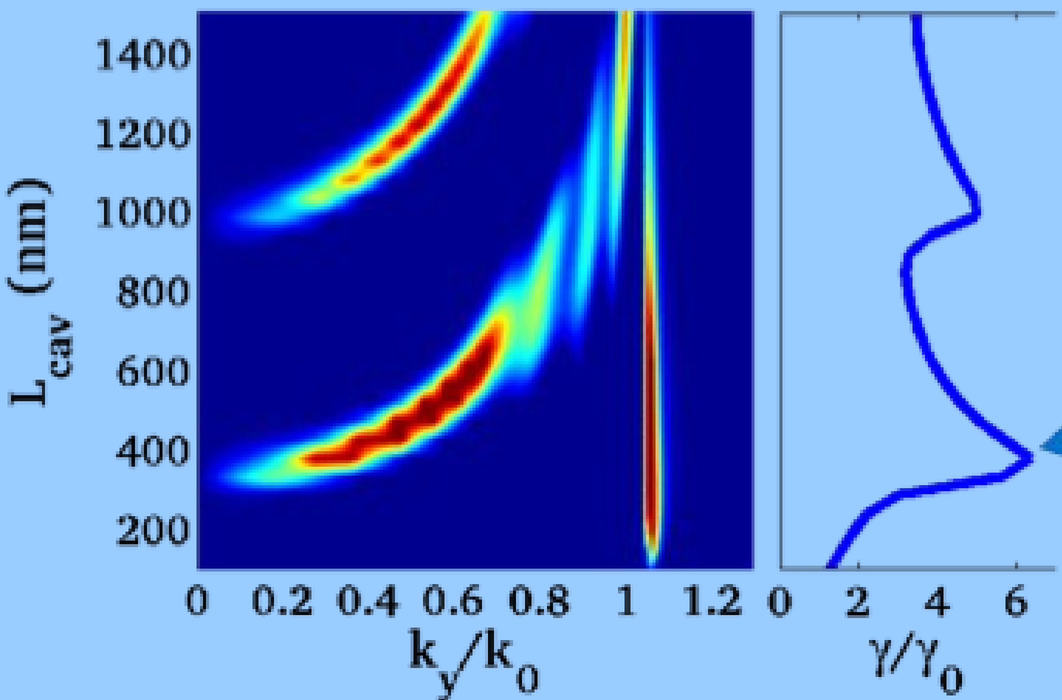


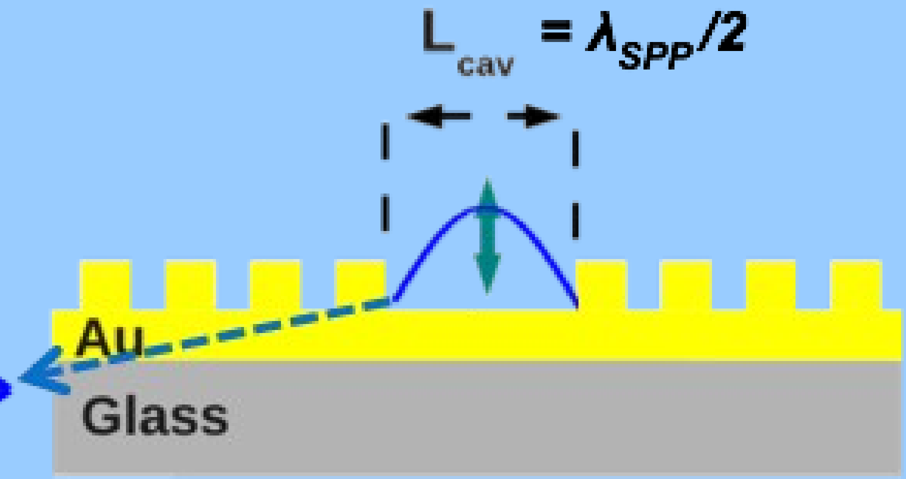
In plane SPP cavity



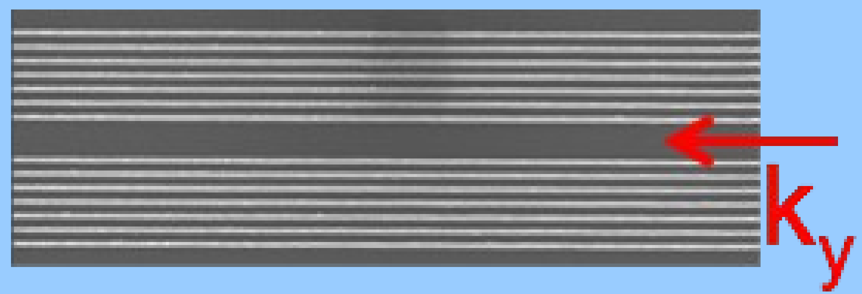
emitted power
P(ky)

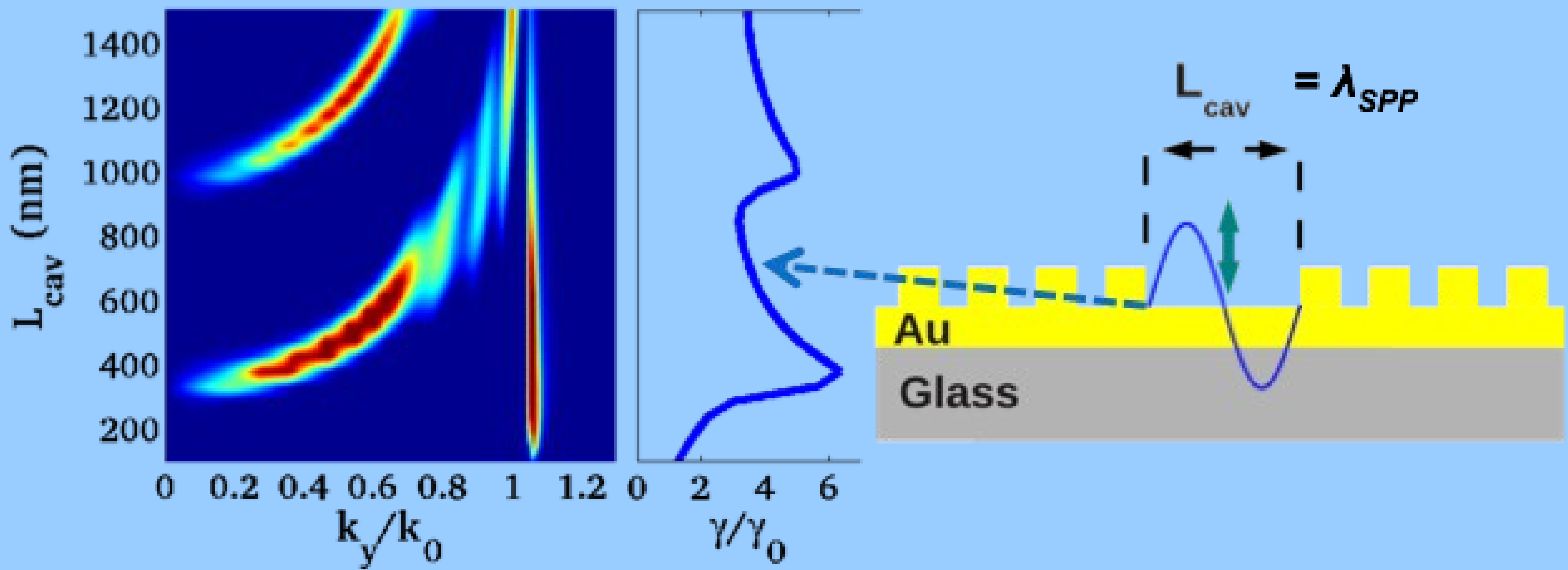
decay rate
$\gamma \propto \int P(k_y)\,dk_y$



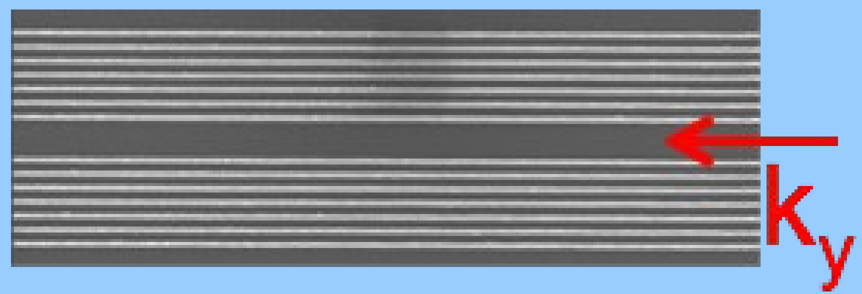


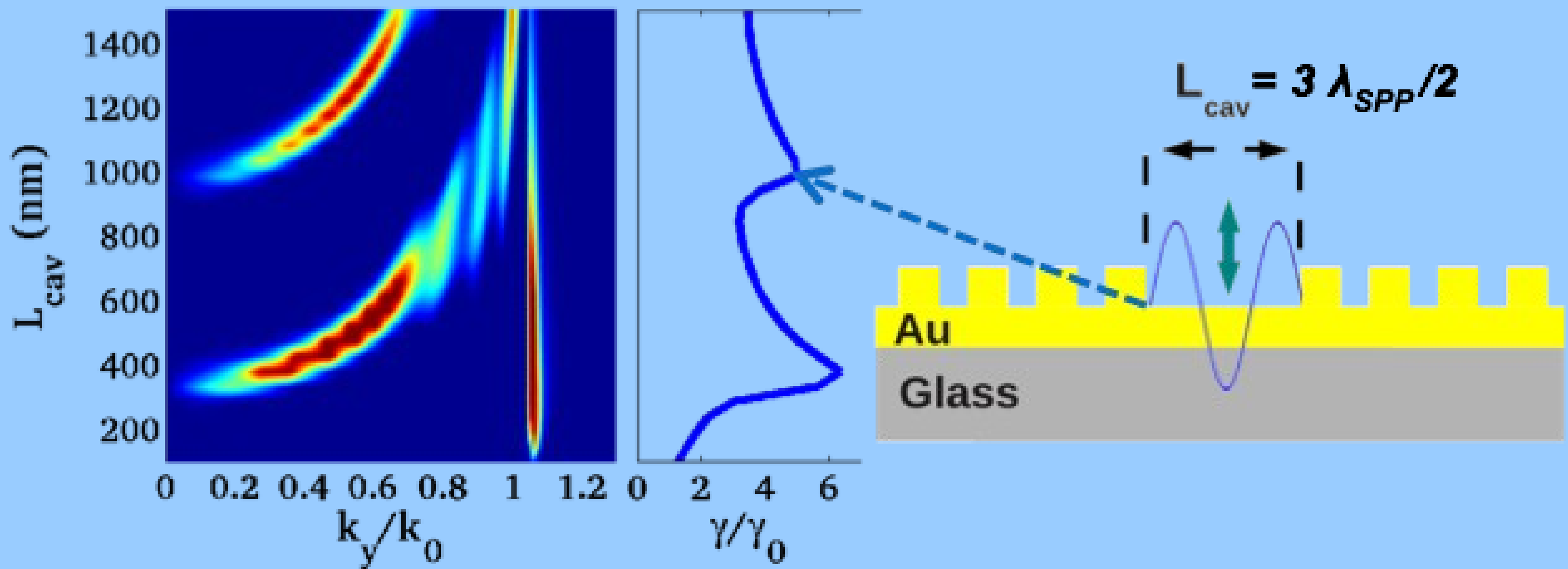
In plane SPP cavity



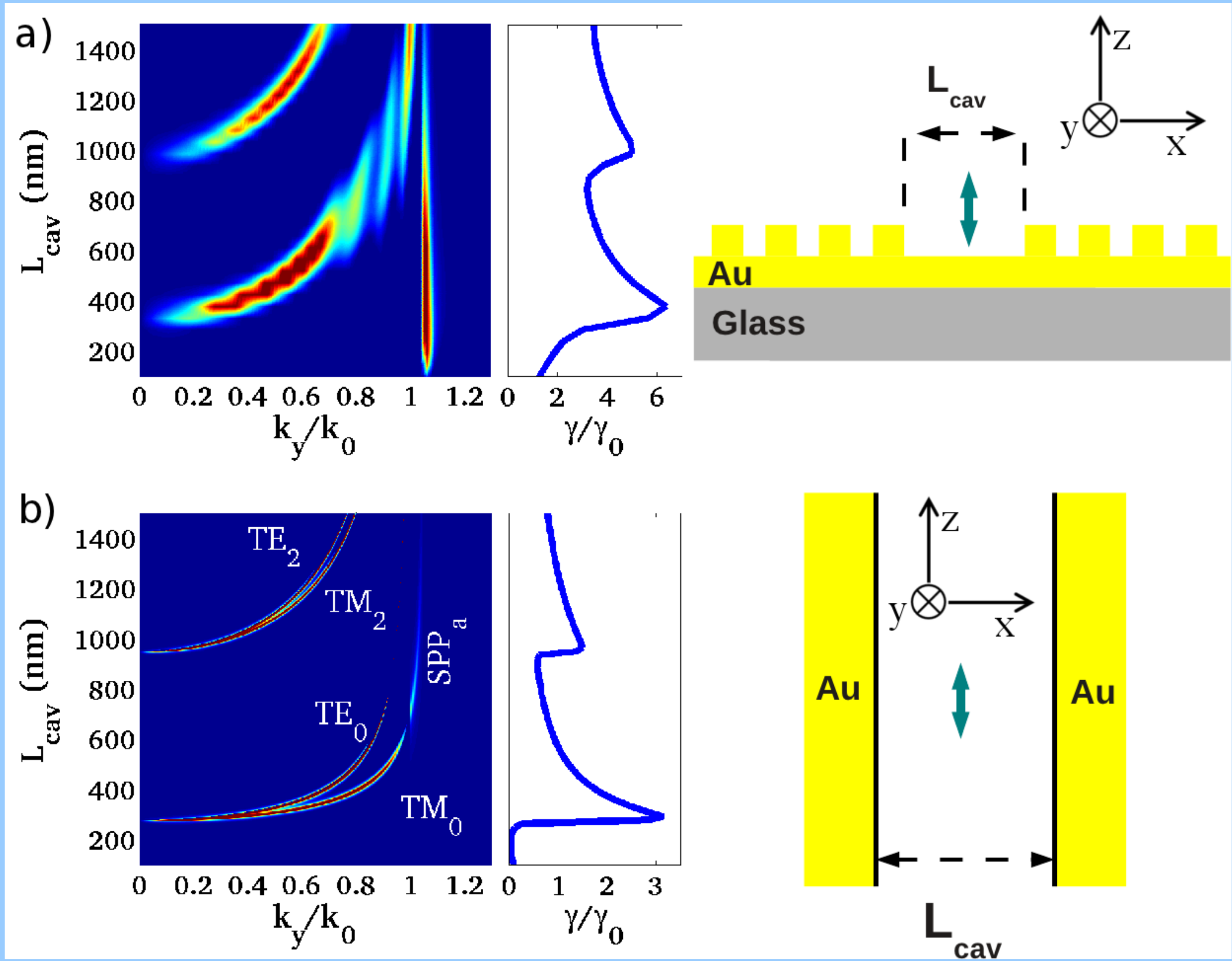


In plane SPP cavity

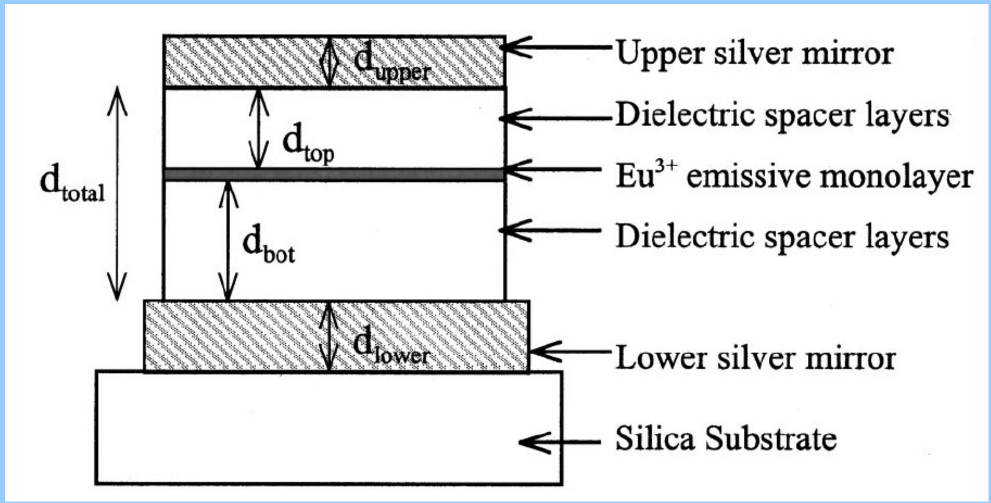


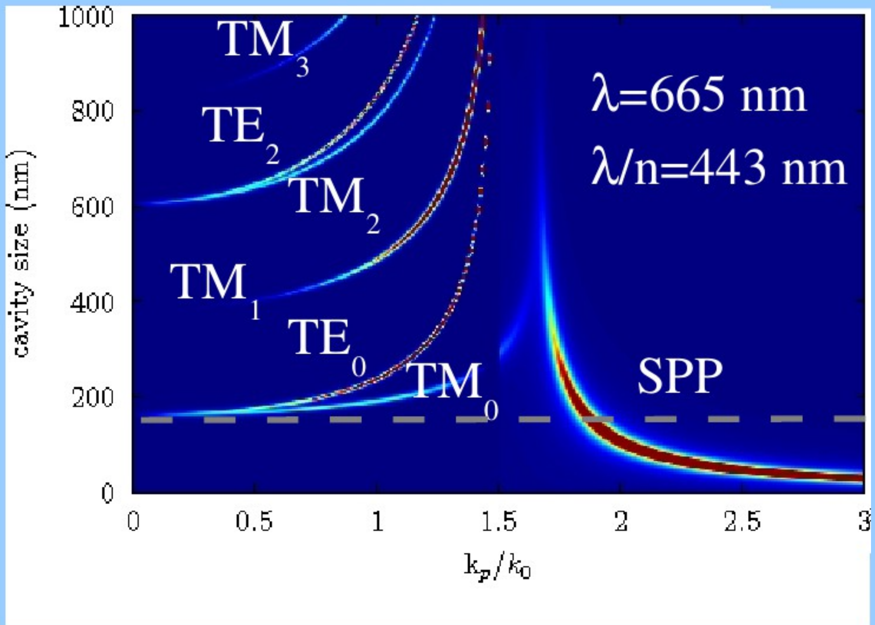


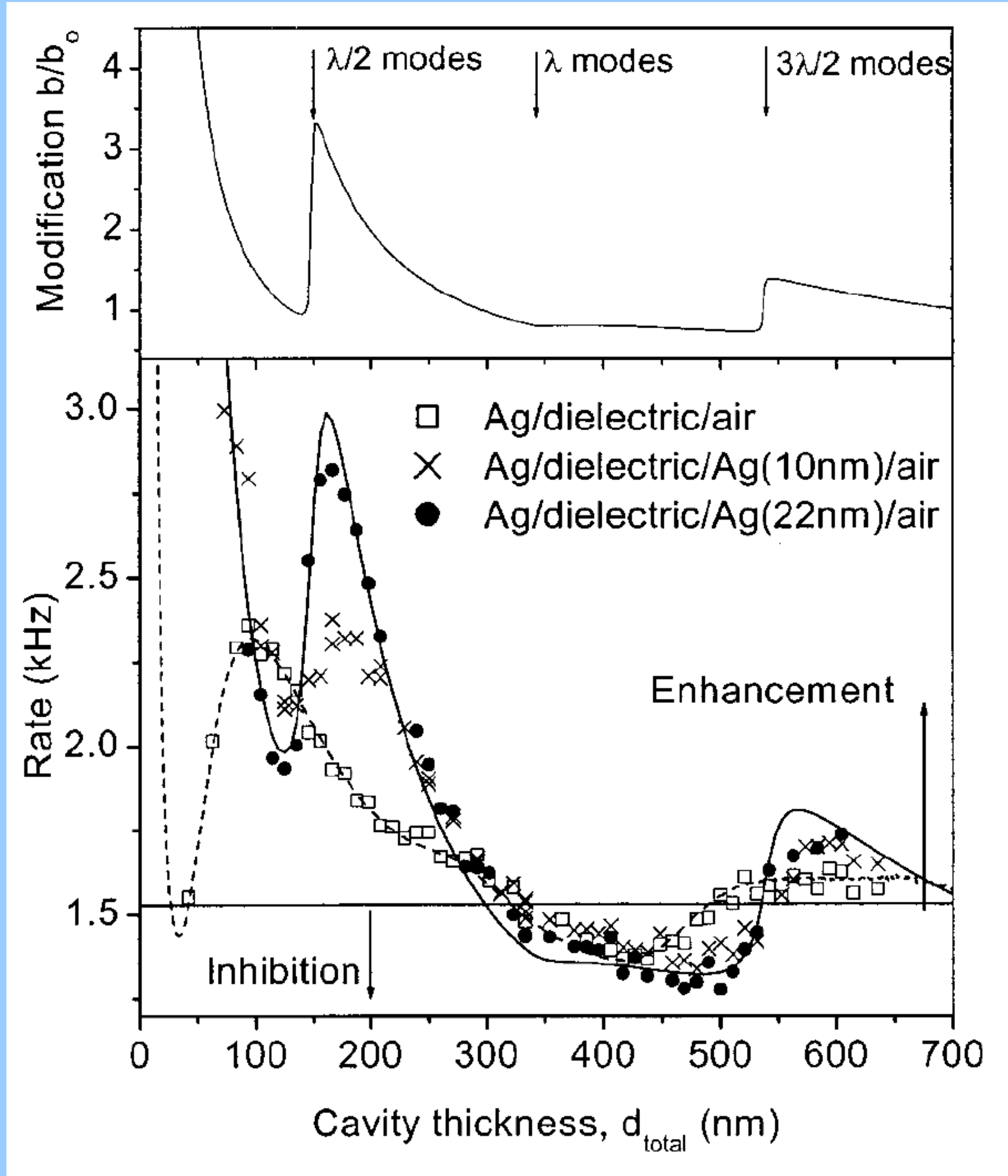
Comparison between planar and bulk cavities



Optical µcavity



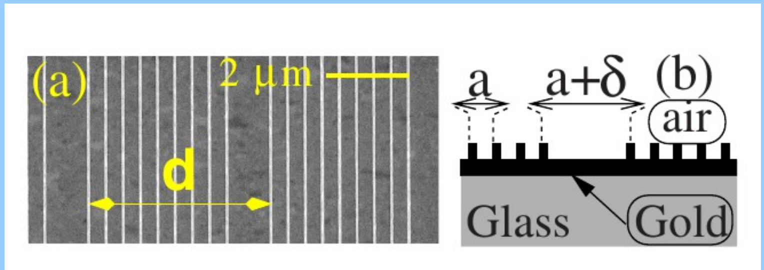




Rate an efficiency of spontaneous emission in metal-clad µcavities Worthing *et al*, J. App. Phys. **89** 615 (2001)

See also The Single Molecule Probe: Nanoscale Vectorial Mapping of Photonic Mode Density in a Metal Nanocavity Hoogenboom *et al*, Nano Letters **9**, 1189 (2009)

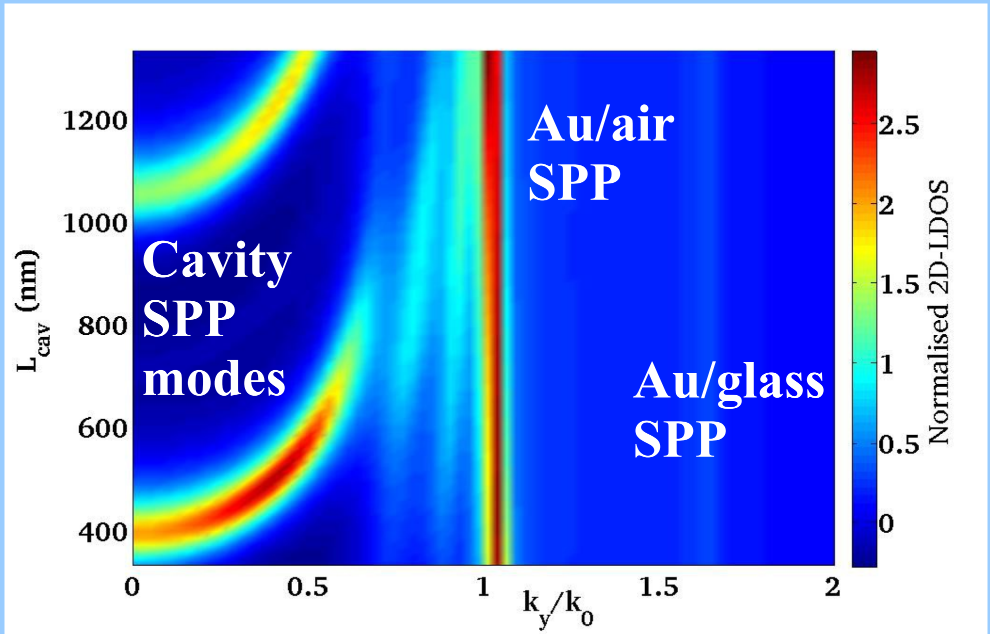
Plasmonic (planar) cavity

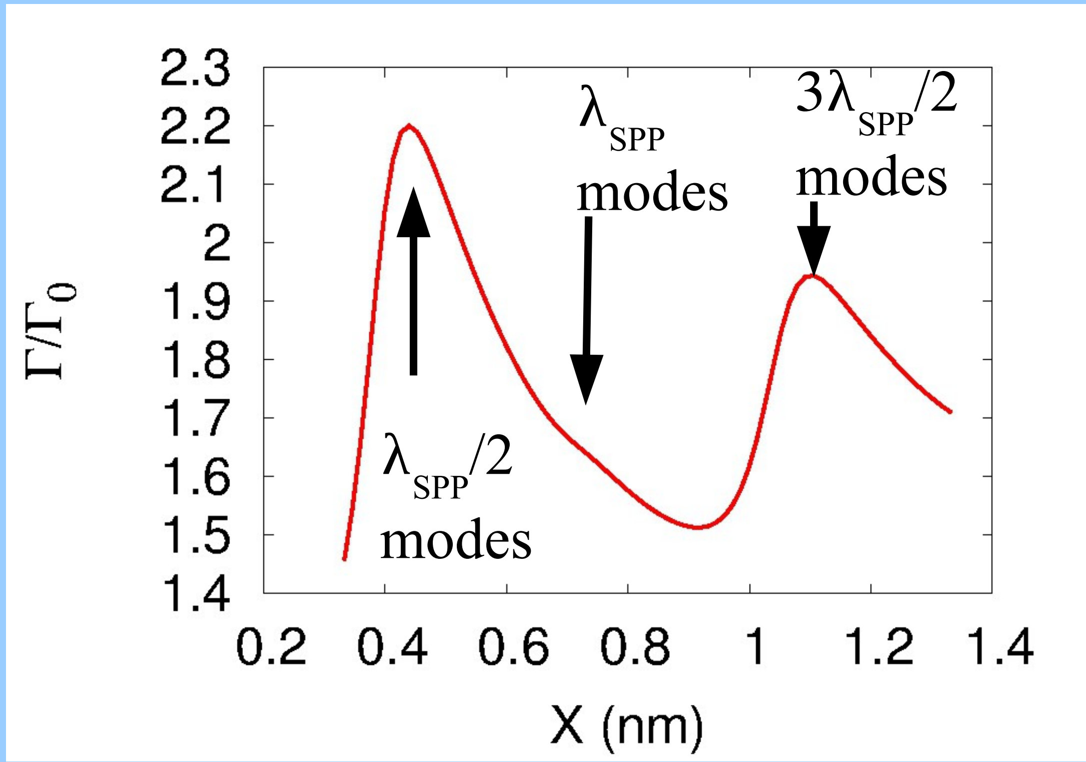


Weeber *et al*, Nano Let. **7**, 1352 (2007)

λ_0=690 nm

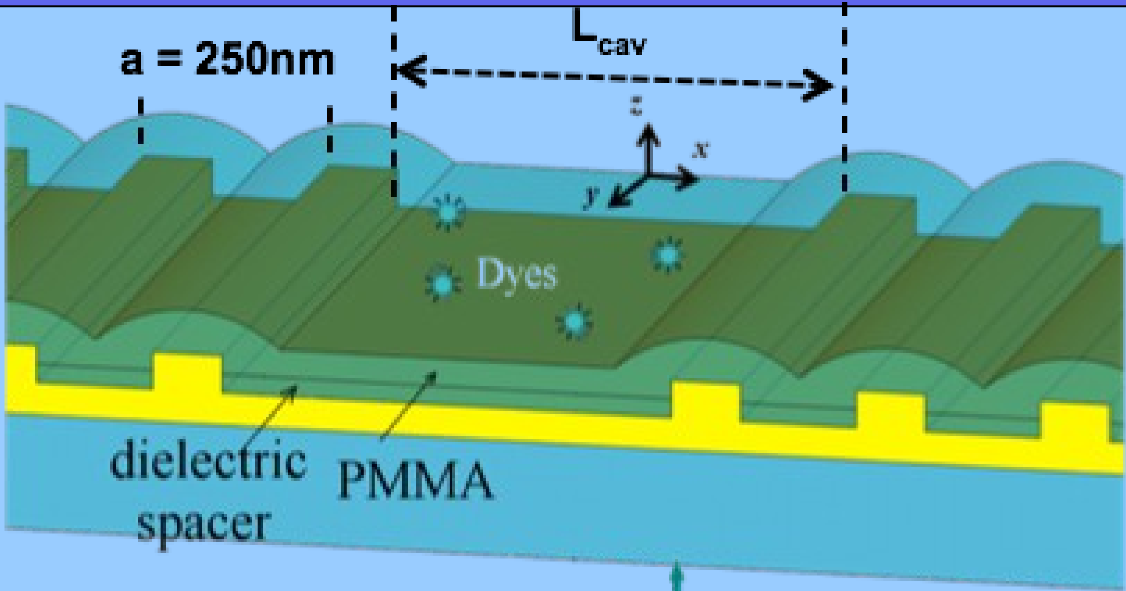
λ_{SPP}=665 nm (n_{eff}=1.035)



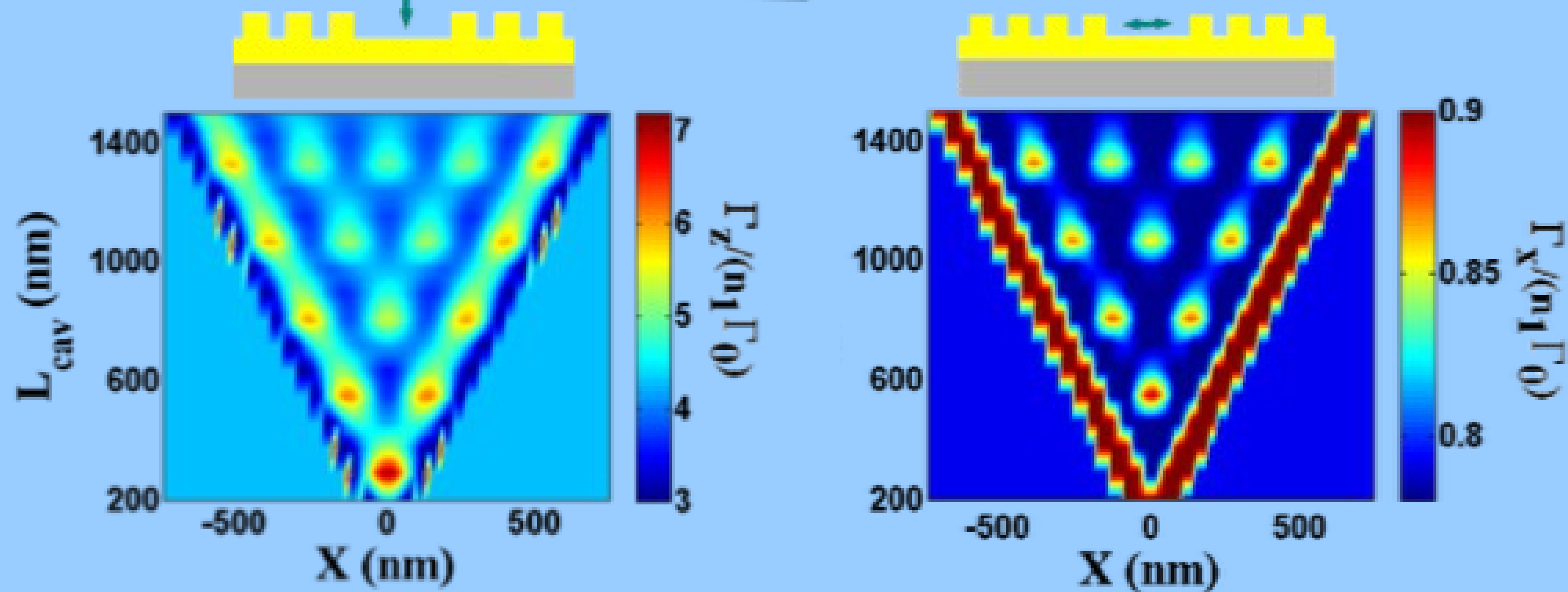


Single-molecule controlled emission in planar plasmonic cavities
Derom *et al*, Phys. Rev . B **89**, 035401 (2014)

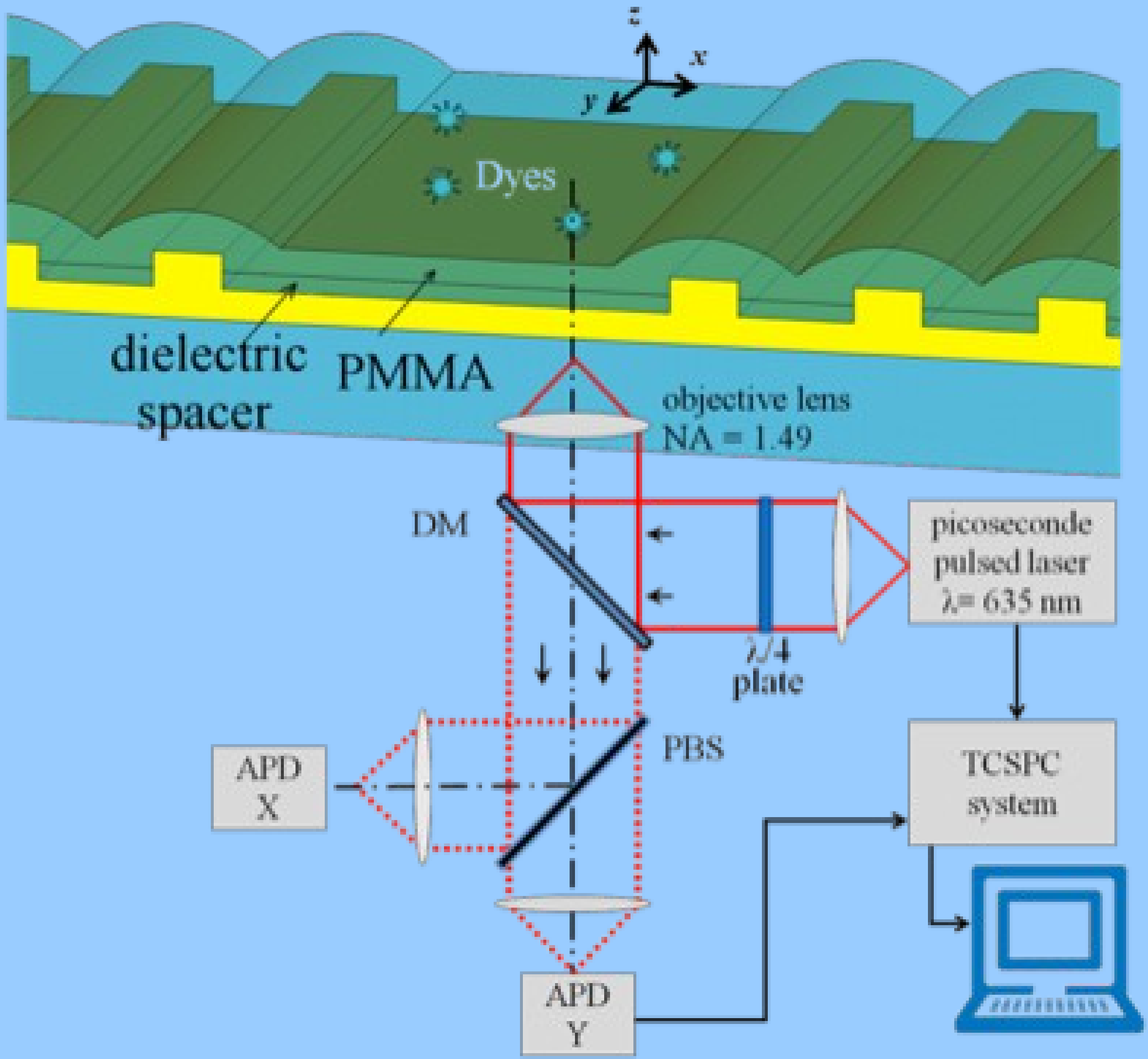
In plane SPP cavity



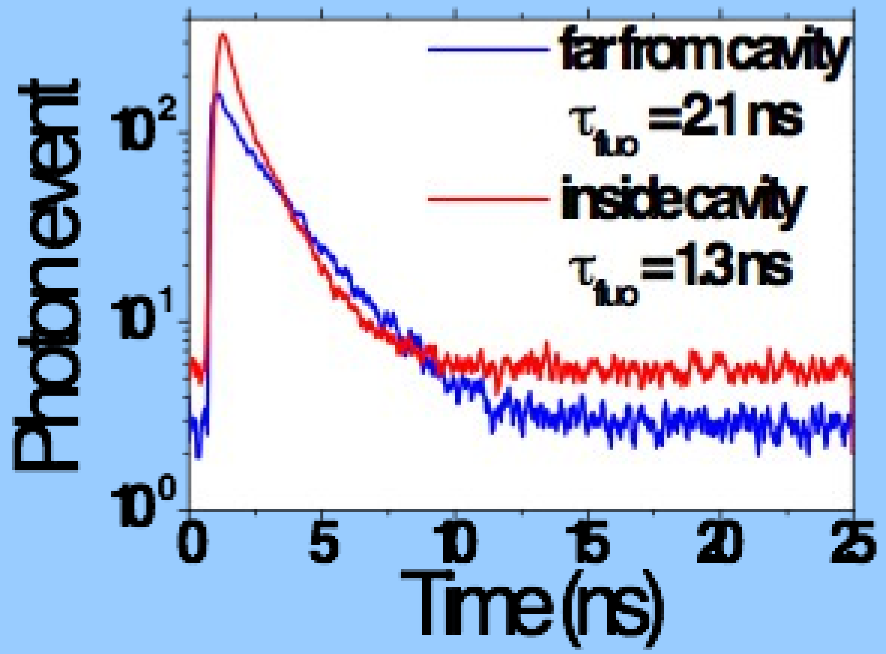
Decay rate depends on
- **the cavity length**
- **the molecule position**
- **the molecule orientation**



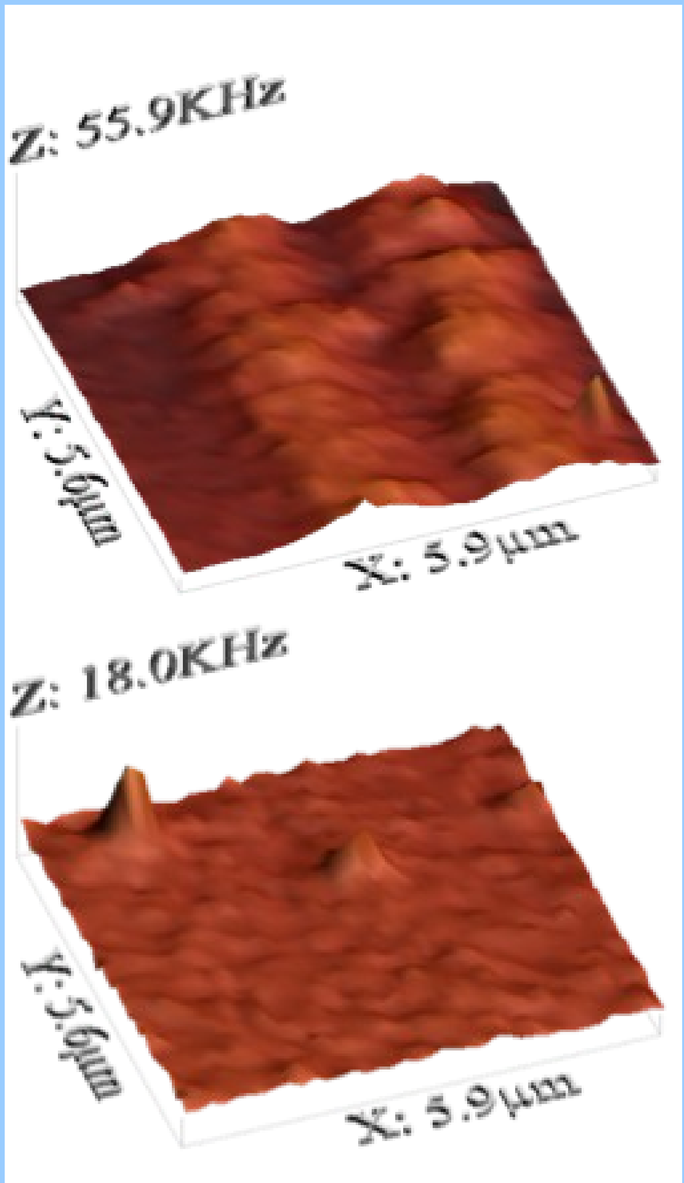
In plane SPP cavity

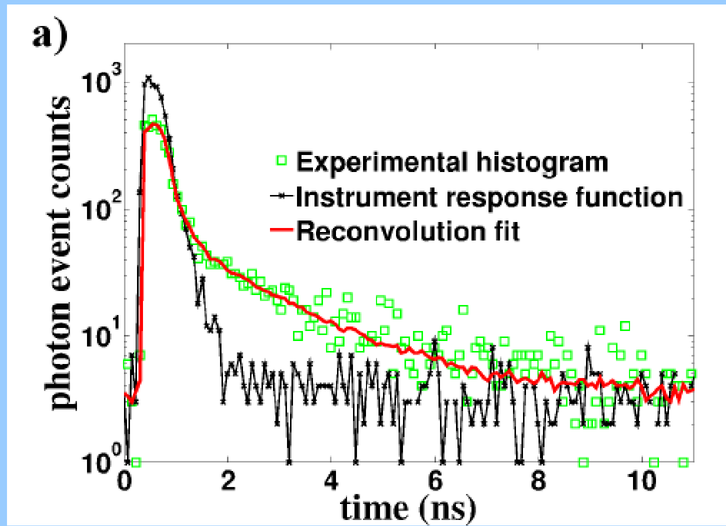


DiD molecule
λ emission = 670 nm



Lifetime measurement





137 molecules into the cavities

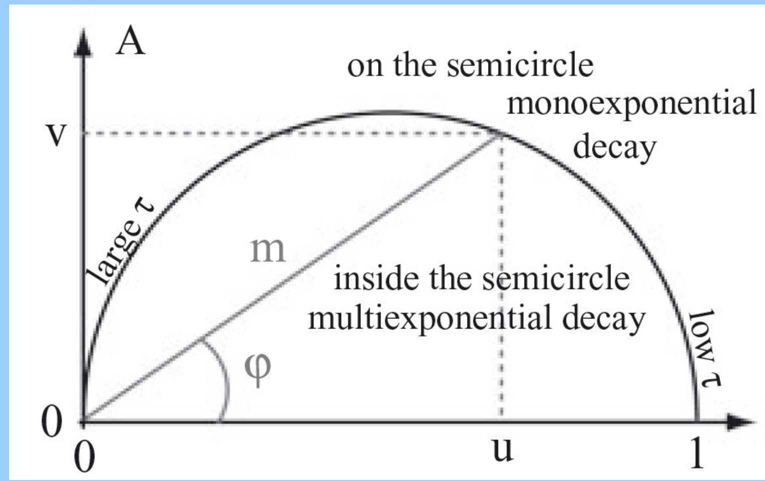
Reference :
75 molecules far from cavities

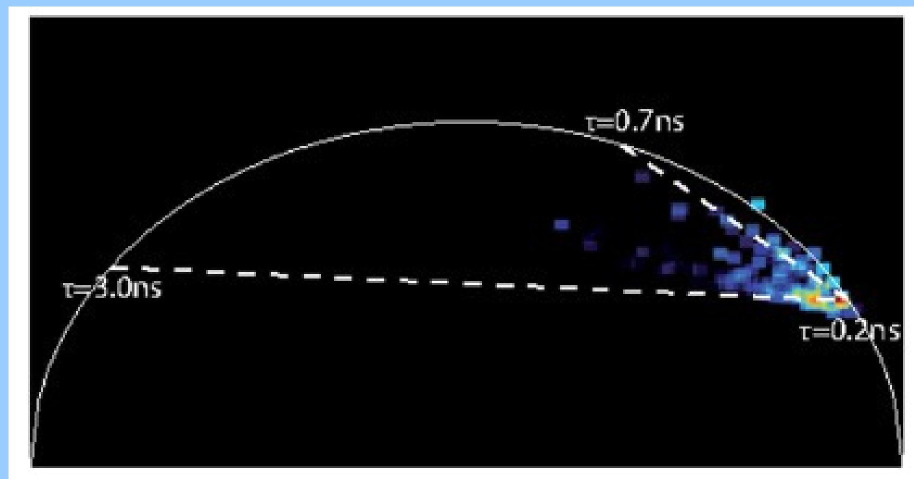
Bi exponential fitting
- short components (~10 ps)
  background signal (notably gold photoluminescence)
- single molecule fluorescence lifetime (~ ns)

Polar representation

$$u(\omega) = \frac{\int_0^T \cos(\omega t) I(t) dt}{\int_0^T I(t) dt}$$

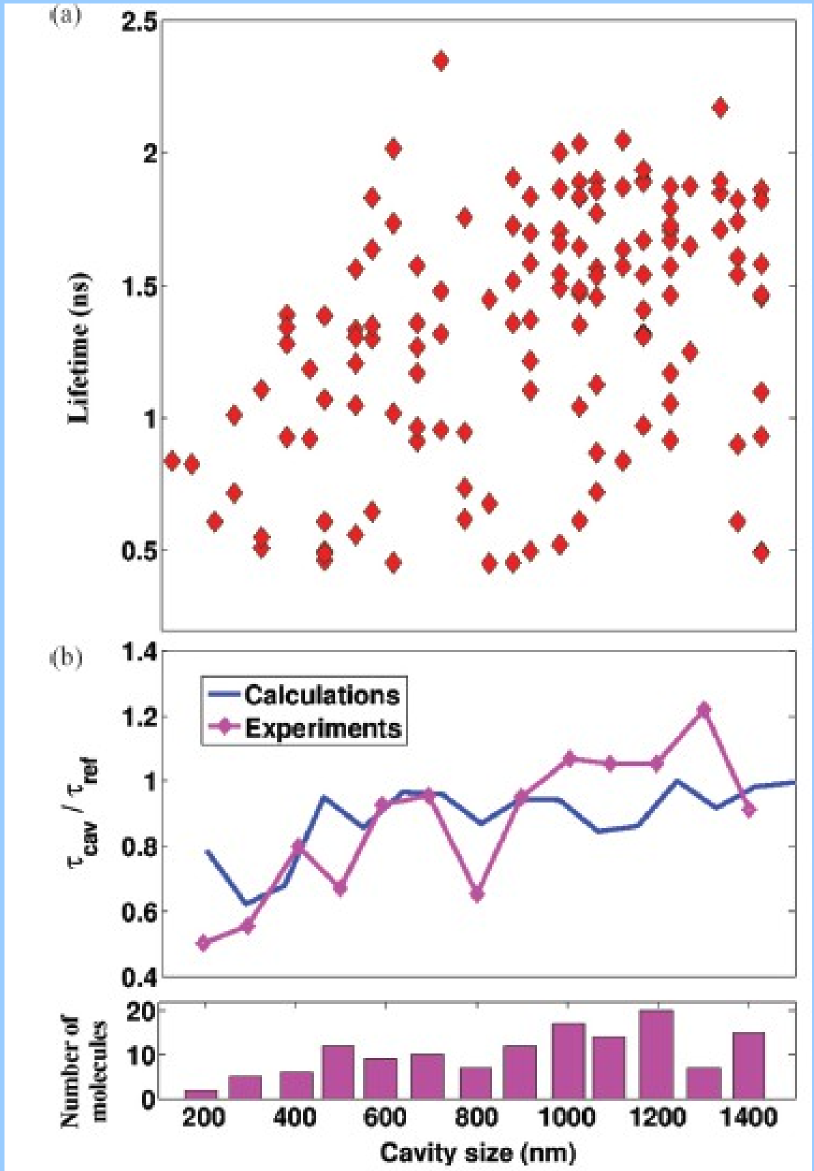
$$v(\omega) = \frac{\int_0^T \sin(\omega t) I(t) dt}{\int_0^T I(t) dt}$$





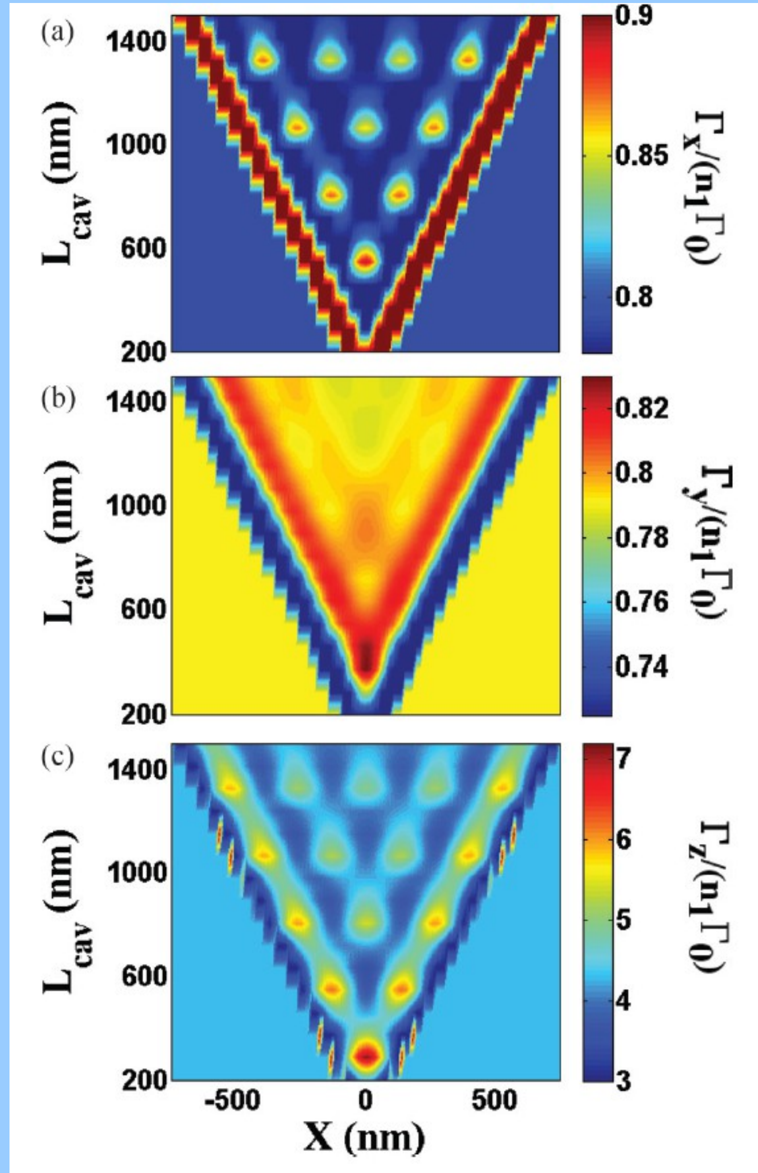
Generalization of the polar representation in time domain fluorescence lifetime imaging microscopy for biological applications: practical implementation
Leray *et al*, J. µscopy **248**, 66 (2012)

Effect of the cavity size

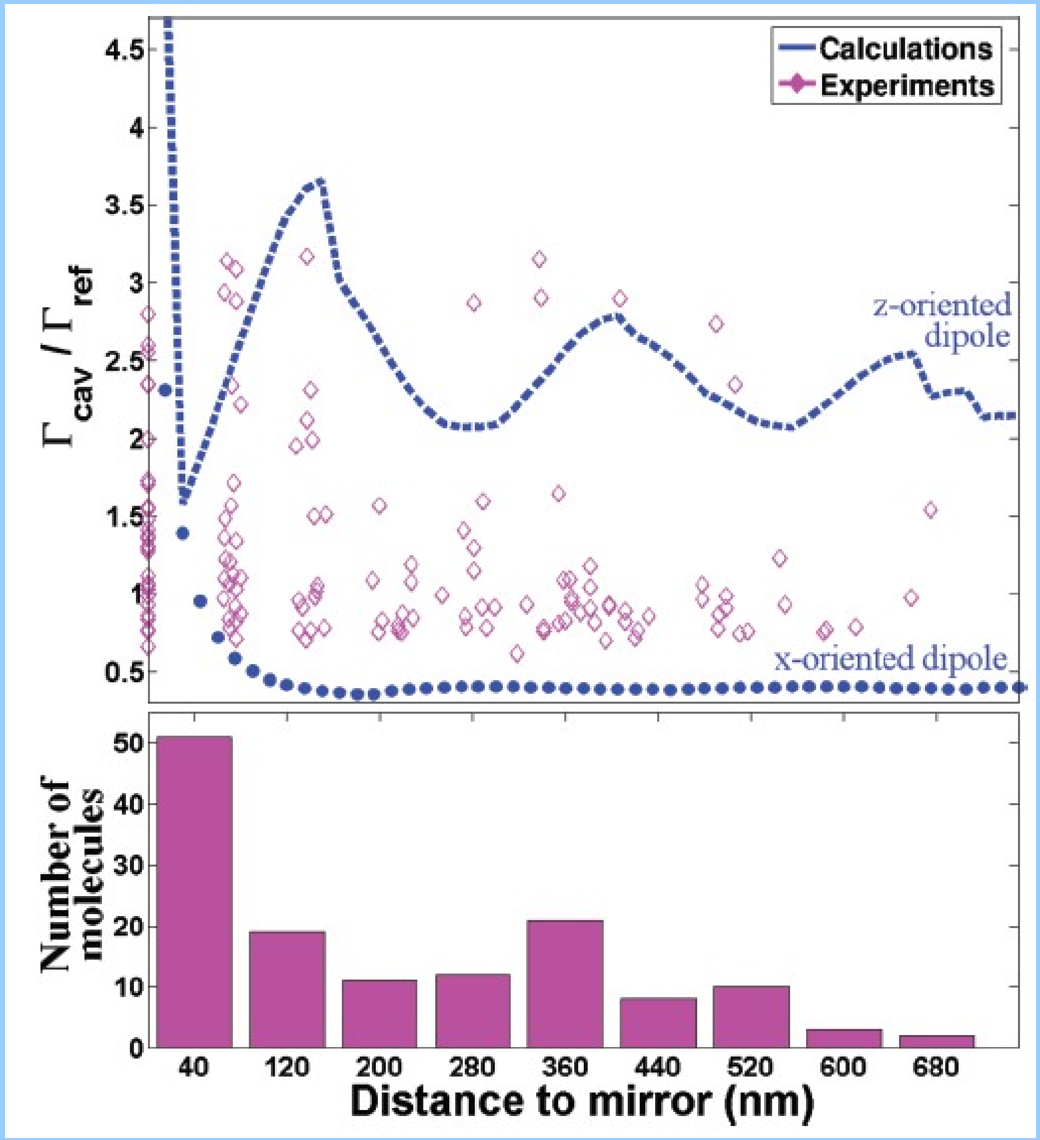


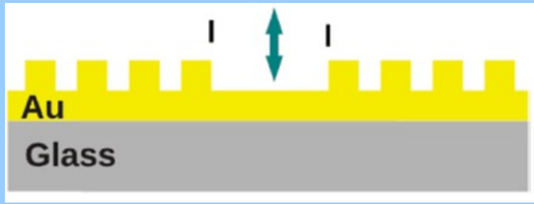
- strong dispersion at the single molecule level

- lifetime averaged over position for each cavity size : small variations related to coupling to the cavity modes



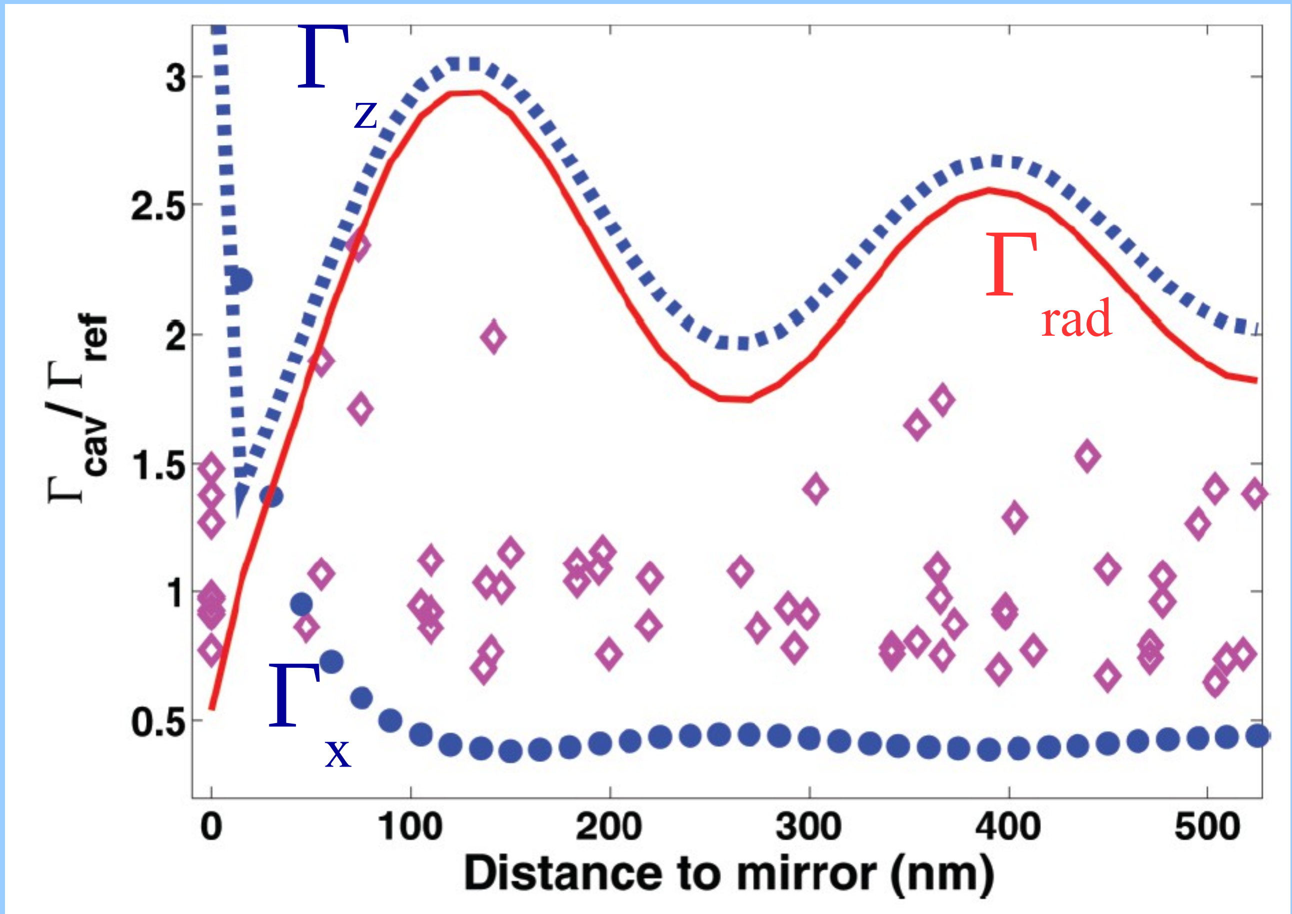
Molecule position

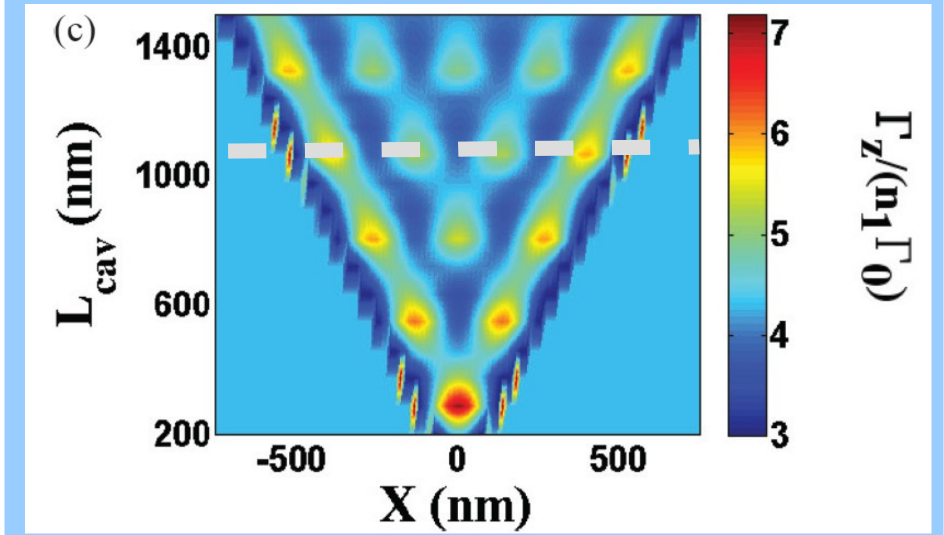




dispersion decay rate due to molecules orientation

Single cavity (L_{cav}=2 λ_{SPP}=1,1µm)

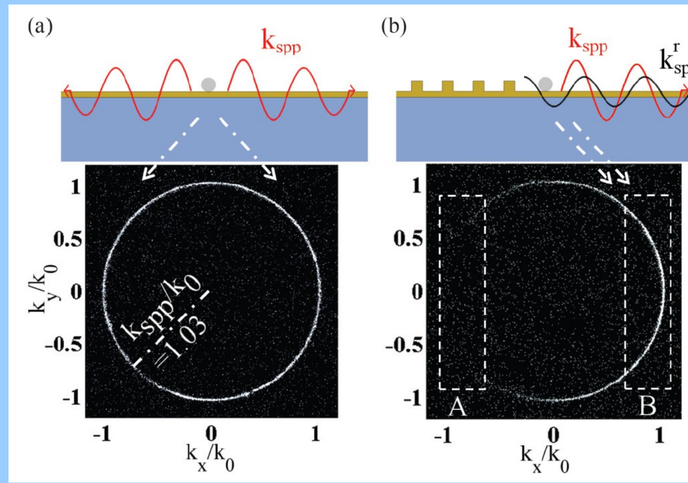




$$\frac{\Gamma_{rad}}{\Gamma_{tot}} \approx 0.9$$

Extraction efficiency (leakage into the substrate)

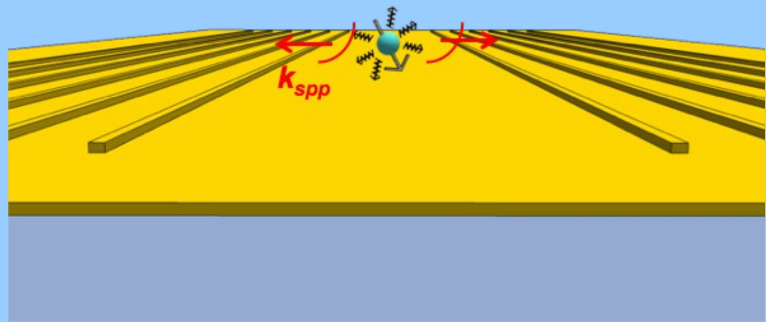
Summarize



Plasmonic Bragg mirror

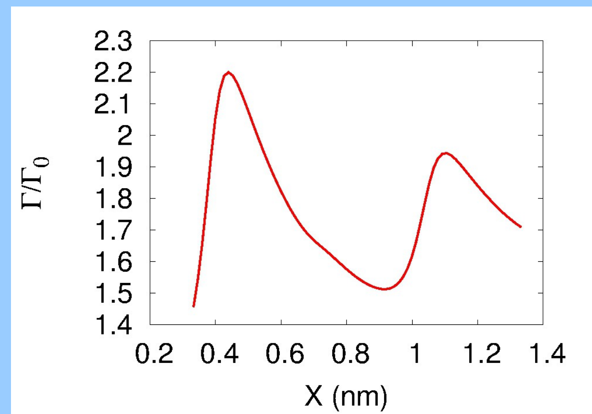
- efficient reflexion
  of locally excited SPP over ~ 40 °
- large bandwidth

=> control emission at room temperature



Planar plasmonic cavity

- surface wave confinement
- planar analogous of bulk optical µcavity
- F_p~7 (β ~ 85%)
- good extraction efficiency



Single-molecule controlled emission in planar plasmonic cavities
Derom et al, Phys. Rev . B **89**, 035401 (2014)

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