Plasmonique moléculaire : du facteur de Purcell à la plasmonique quantique

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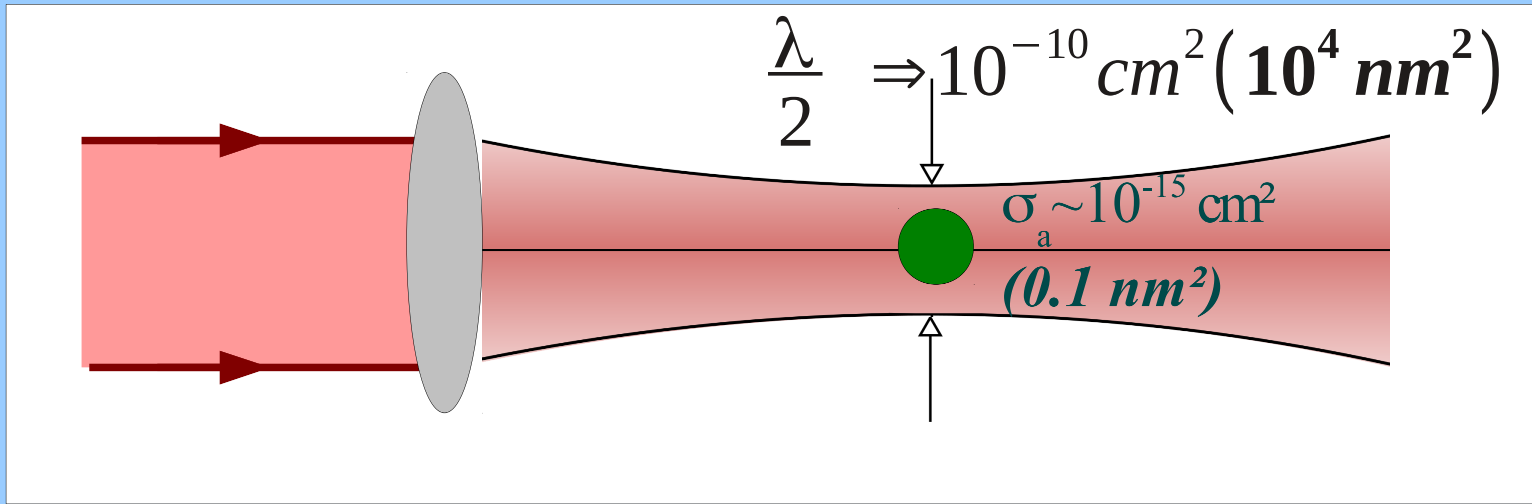
Dijon - France

UMR 6303 ICB

Light/matter interaction at the nanoscale

Light/matter interaction at the nanoscale



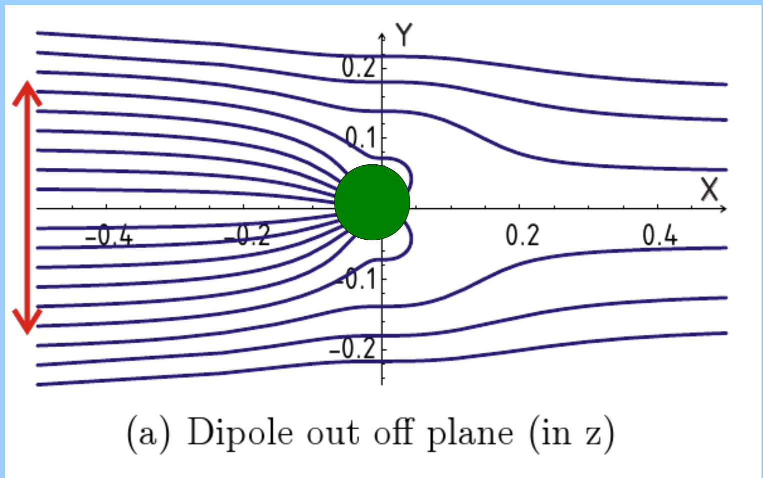
Gaussian beam $A_{eff} = \pi w_0^2$

Absorption cross-section σ_a

cooperativity $C_0 = \frac{\sigma_a}{A_{eff}} \sim 10^{-3}$

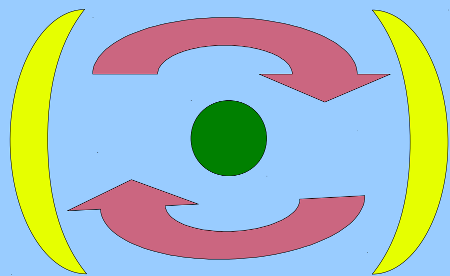
Strategies

Low T°C (<10K)
$\sigma_a \sim 3\lambda^2/2\pi \sim 10^{-10}$ cm²
(ϕ ~100x molecule size!)



Paul, Fischer
Light absorption by a dipole, Societ Physics Uspekhi **26**, 923 (1983)

Cavity quantum electrodynamics (cQED)

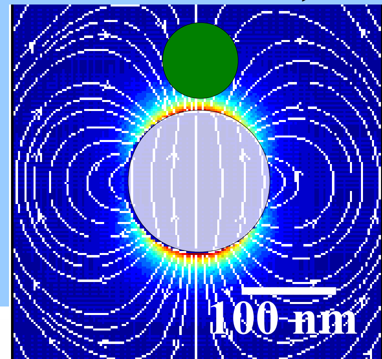


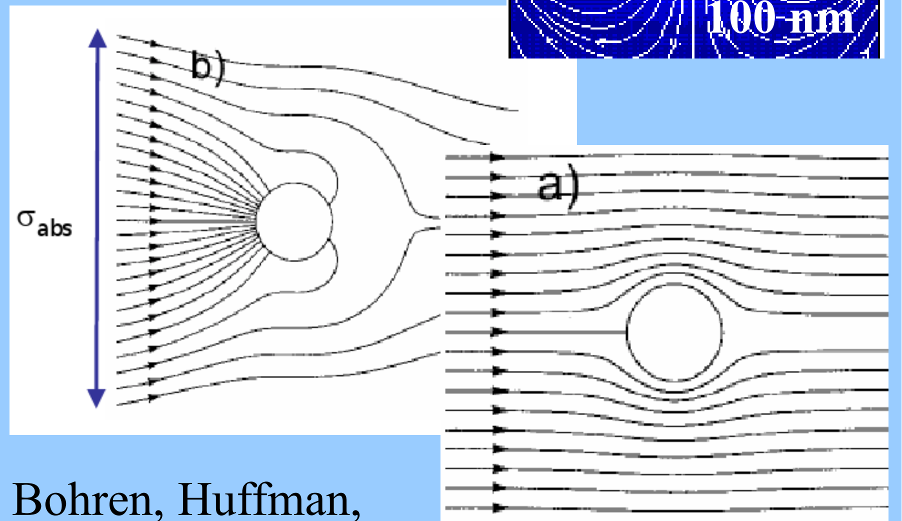
$$C = 4C_0 \frac{\mathcal{F}}{\pi}$$

Intensity enh.

Round trip

Surface enhanced spectroscopies (SERS, SEF)





Bohren, Huffman,
Absorption and Scattering of Light by Small Particles, Wiley, New York 1983

Purcell factor

Proceedings of the American Physical Society

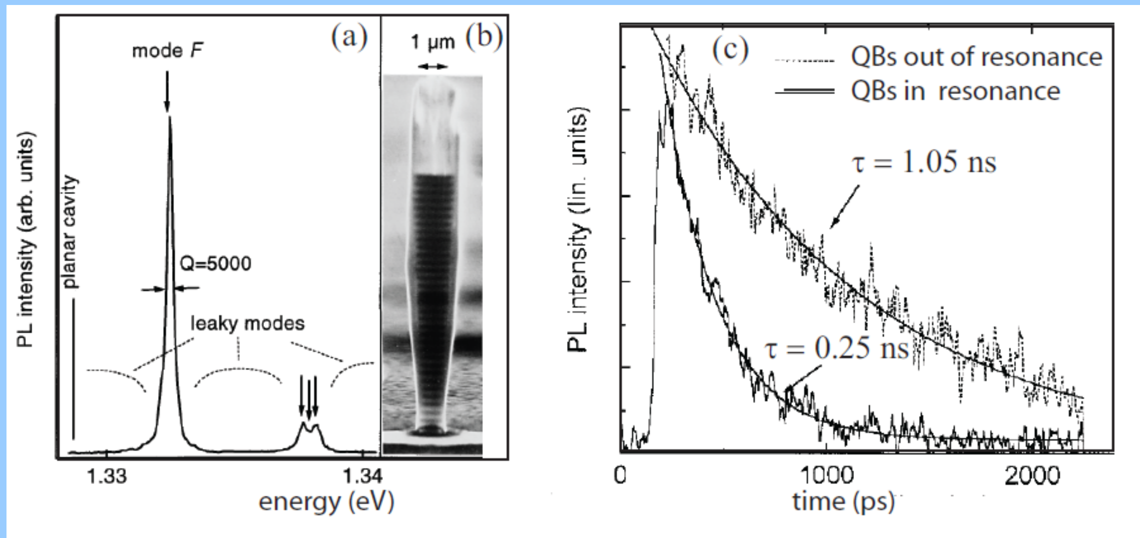
MINUTES OF THE SPRING MEETING AT CAMBRIDGE, APRIL 25–27, 1946

B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. PURCELL, *Harvard University*.—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

$$A_\nu = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2)\ \text{sec.}^{-1},$$

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu = 10^7$ sec.$^{-1}$, $\mu = 1$ nuclear magneton, the corresponding relaxation time would be 5×10^{21} seconds! However, for a system coupled to a resonant electrical circuit, the factor $8\pi\nu^2/c^3$ no longer gives correctly the number of radiation oscillators per unit volume, in unit frequency range, there being now *one* oscillator in the frequency range ν/Q associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor $f = 3Q\lambda^3/4\pi^2V$, where V is the volume of the resonator. If a is a dimension characteristic of the circuit so that $V \sim a^3$, and if δ is the skin-depth at frequency ν, $f \sim \lambda^3/a^2\delta$. For a non-resonant circuit $f \sim \lambda^3/a^3$, and for $a < \delta$ it can be shown that $f \sim \lambda^3/a\delta^2$. If small metallic particles, of diameter 10^{-3} cm are mixed with a nuclear-magnetic medium at room temperature, spontaneous emission should establish thermal equilibrium in a time of the order of minutes, for $\nu = 10^7$ sec.$^{-1}$.

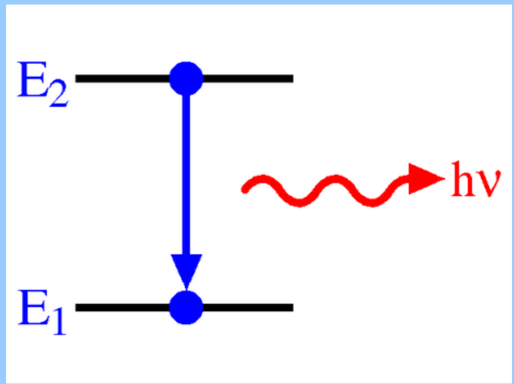
Purcell, 1946



Gérard, Gayral, J. Light. Tech. (1999)

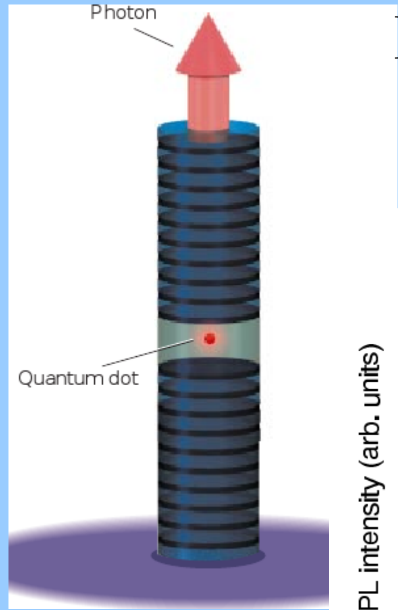
Control of the spontaneous emission – Purcell factor

Fermi's golden rule



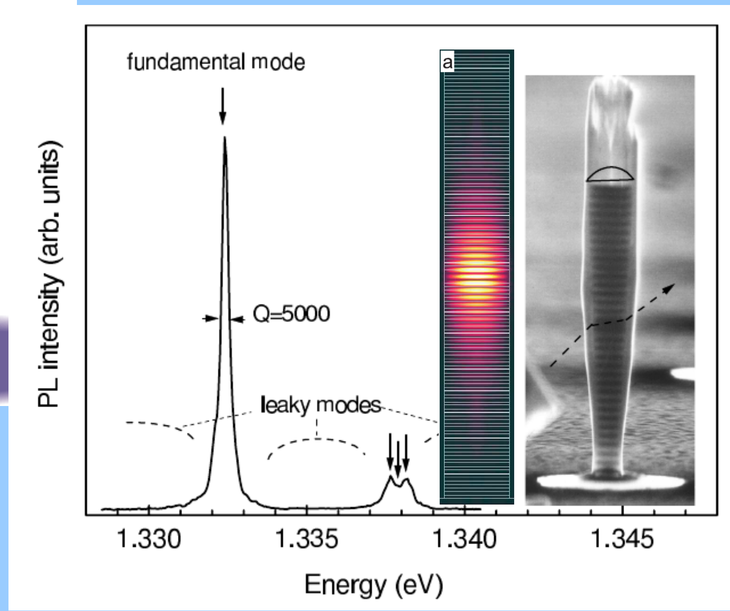
$$\Gamma = \frac{\pi\omega}{\hbar\epsilon_0}|\boldsymbol{p}|^2 \rho(\boldsymbol{r_m}, \omega)$$

Density of modes



J.-M. Gérard
CEA Grenoble

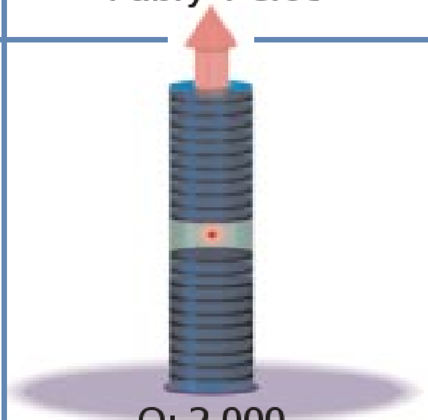
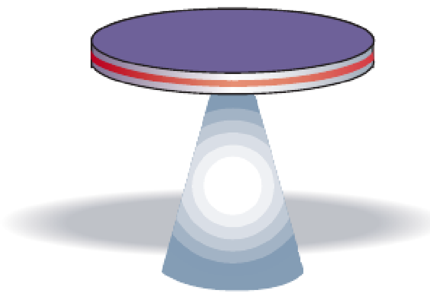
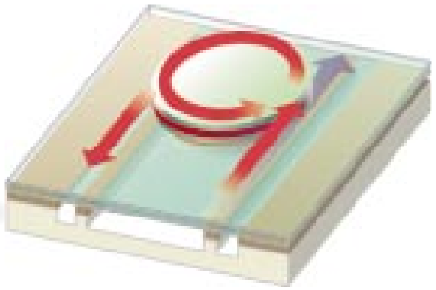
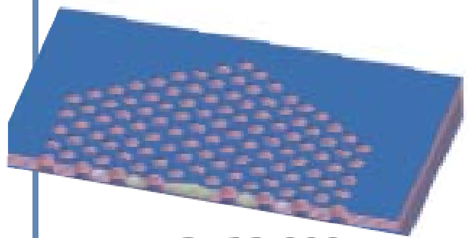
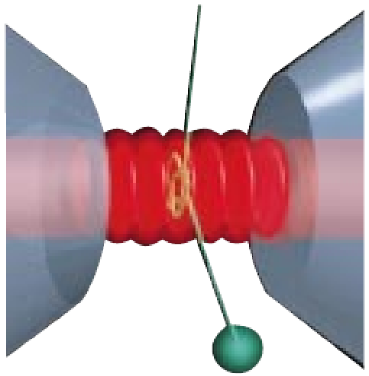
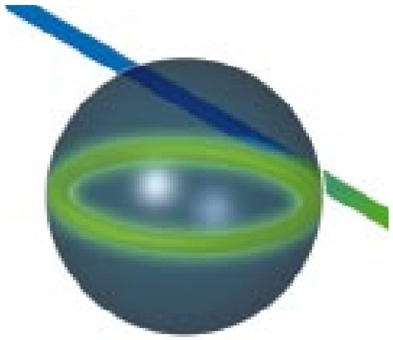
Purcell factor



Given cavity mode

$$F_p = \frac{\Gamma}{\Gamma_0} = \frac{3}{4\pi^2}\left(\frac{\lambda}{n}\right)^3 \frac{Q_{cav}}{V_{mode}}$$

Micro-optical cavities

	Fabry–Perot	Whispering gallery		Photonic crystal
High Q	 Q: 2,000 V: 5 $(\lambda/n)^3$	 Q: 12,000 V: 6 $(\lambda/n)^3$	 Q_{III-V}: 7,000 Q_{Poly}: 1.3×10^5	 Q: 13,000 V: 1.2 $(\lambda/n)^3$
Ultrahigh Q	 F: 4.8×10^5 V: 1,690 μm^3	 Q: 8×10^9 V: 3,000 μm^3	 Q: 10^8	*Vahala* Nature **424,**839(2003)

Low threshold/thresholdless laser

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

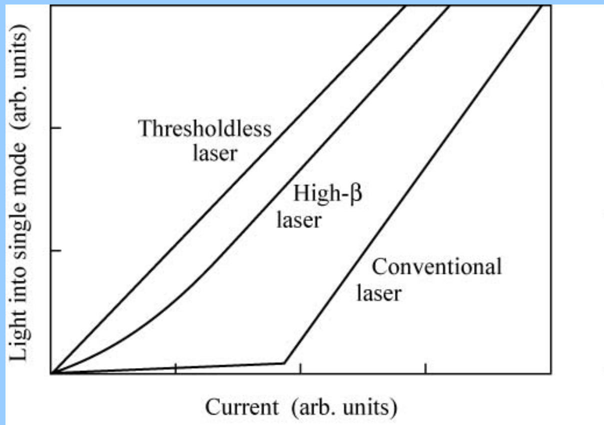
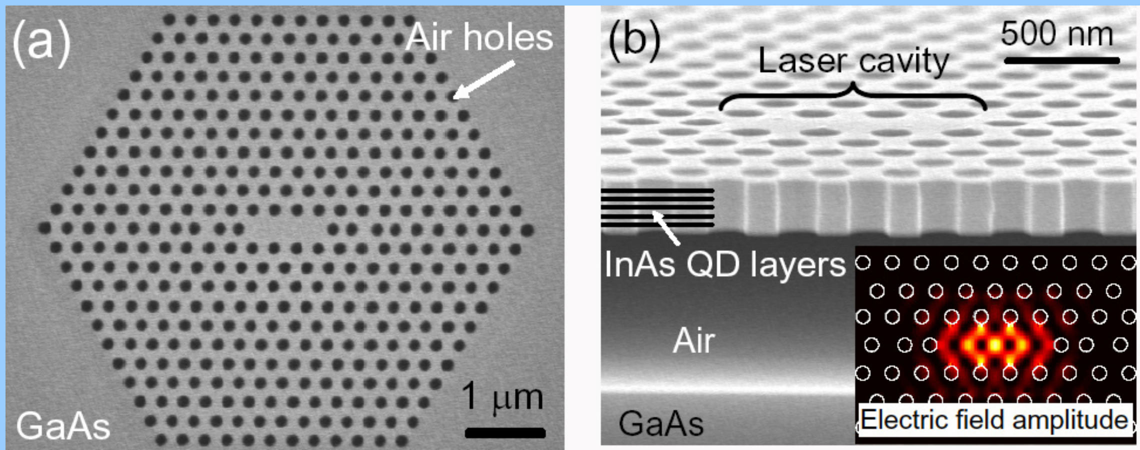
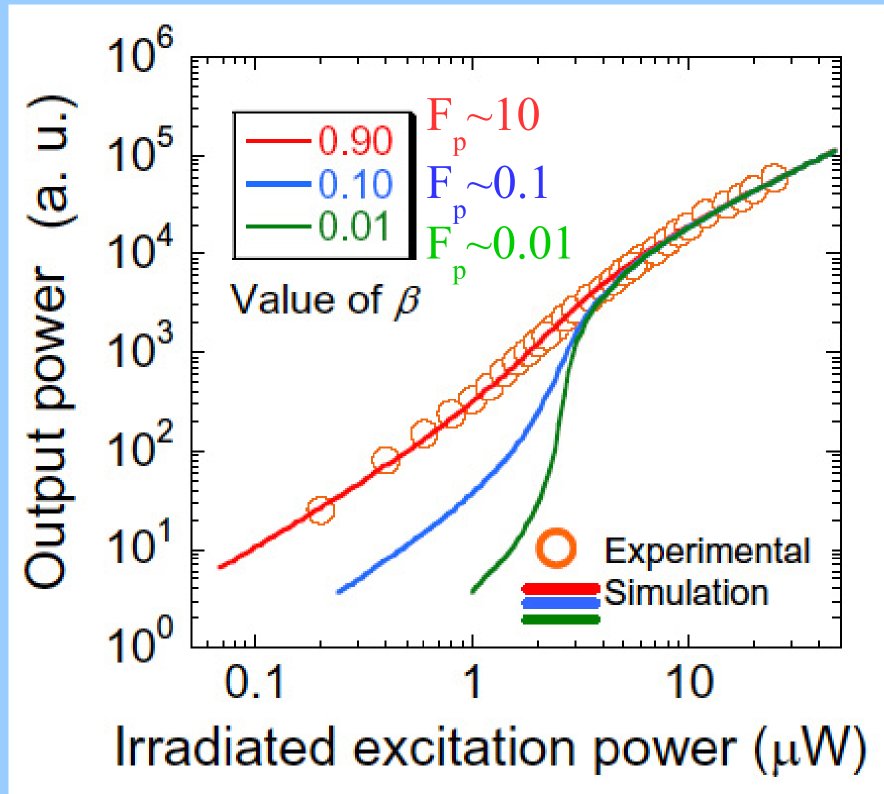


Fig. 15.14. Light-power-versus-current curves for single spatial-mode emission from a (*i*) conventional laser, (*ii*) a high β-factor laser, and (*iii*) a thresholdless laser. The conventional laser has a distinct current threshold. The high β-factor laser has a less distinct threshold. It would be 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.

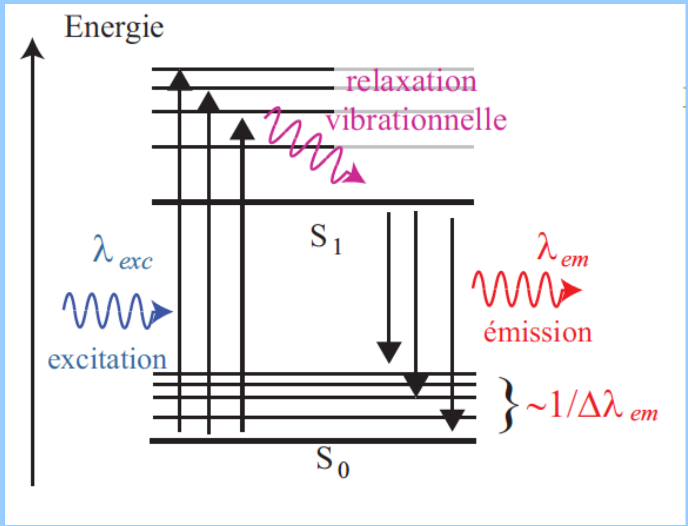
E. F. Schubert
Light-Emitting Diodes (Cambridge Univ. Press)
www.LightEmittingDiodes.org

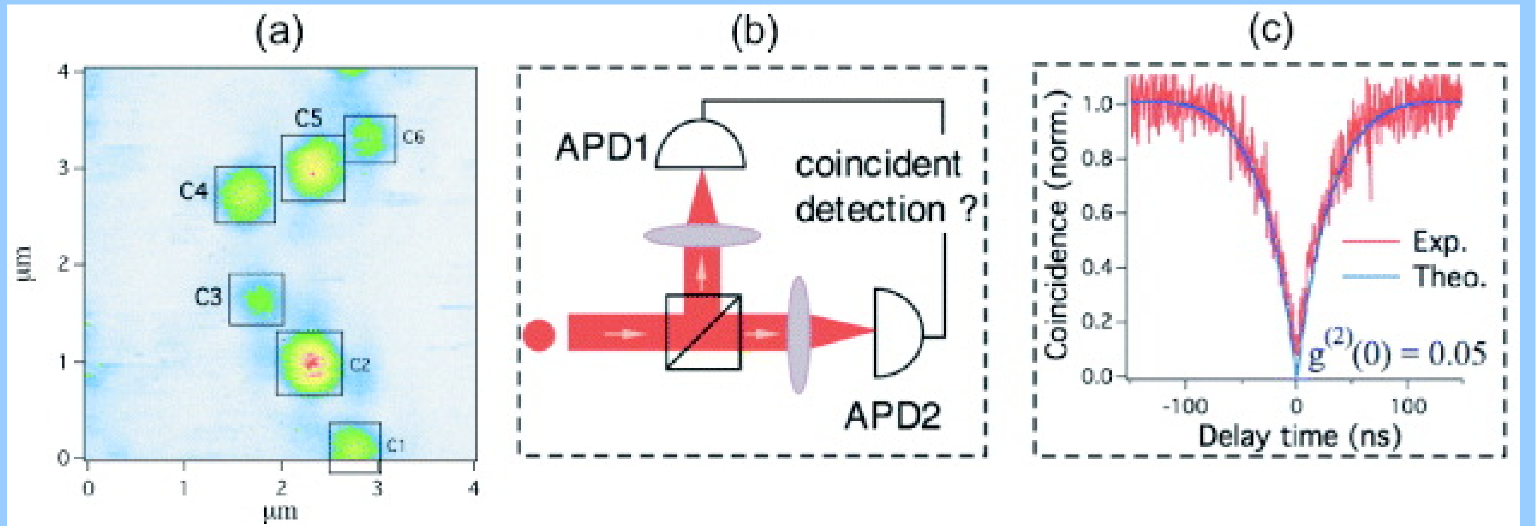




A photonic criystal nanocavity with ultralow threshold
Nomura, Iwatomoto,Arakawa, SPIE (2007)

Single photon source





Roch, Treussart (ENS Cachan)

Specifications

- Photon energy (wavelength)

- On demand single photon, repetition rate

- Indistinguishable photons

Indistinguishable single photons

$$\Gamma = \frac{1}{T_1}$$

Population decay rate (inverse of the measure lifetime)

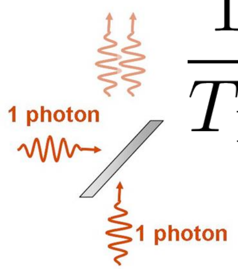
$$\Gamma_{coh} = \frac{1}{T_2}$$

Coherence rate (dipole lifetime)

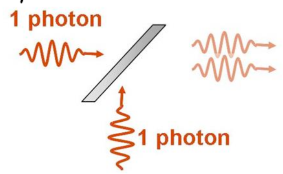
$$\frac{1}{T_2} = \frac{1}{2T_1} + \gamma^*$$

Indistinguishability

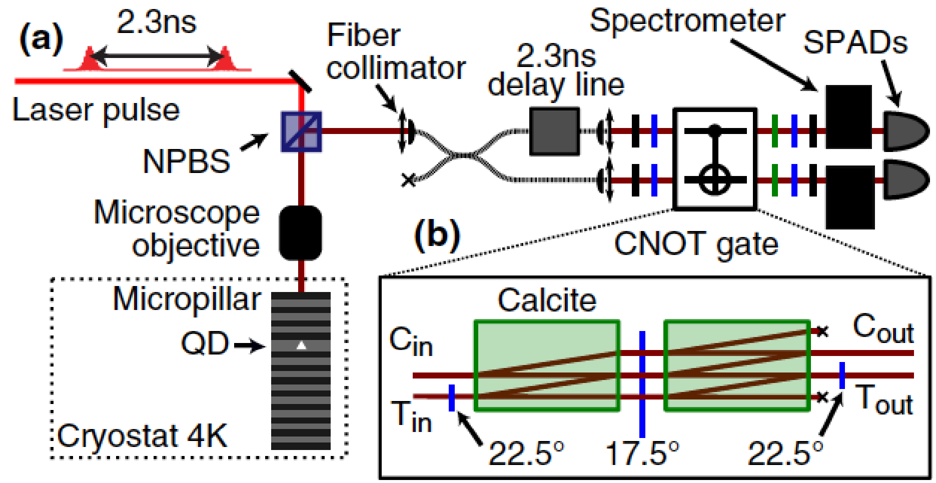
$$\frac{T_2}{2T_1}$$



$$\frac{1}{T_1} \gg \gamma^{\star}$$

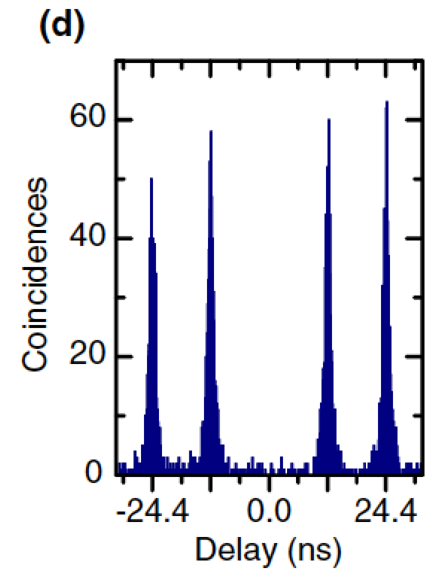


Coalescence effect



Fp~4

C-NOT gate
- purely quantum
- universal gate



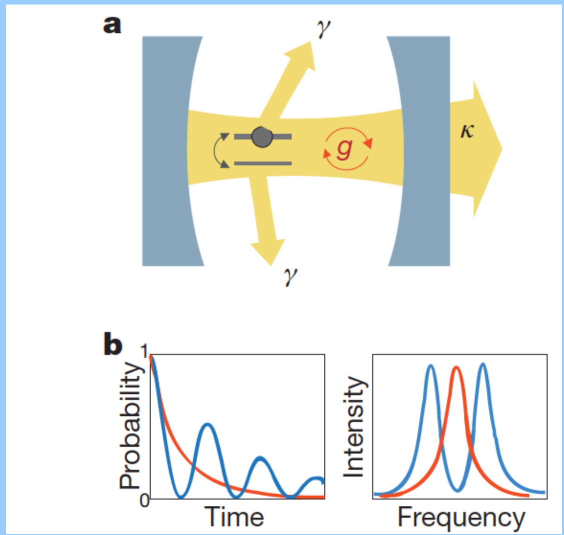
Senellart, PRL **110**, 250501 (2013)

Weak and strong coupling regimes

Weak coupling

$$F_p = \frac{\Gamma_{cav}}{n_1 \Gamma_0}$$

$$= \frac{3}{4\pi^2} \left(\frac{\lambda}{n_1} \right)^3 \frac{Q_{cav}}{V_{mode}}$$



Strong coupling

Coupling strength (g)

>>

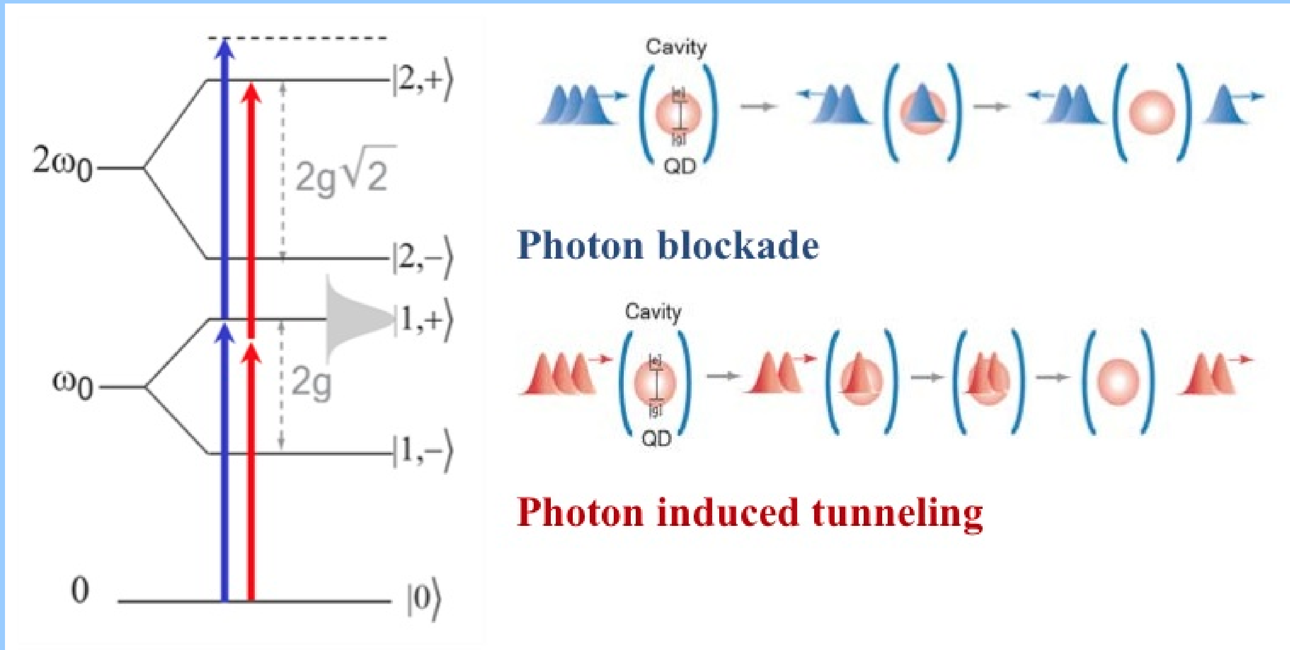
losses (Γ_0,K_{cav})

$$C = \frac{g^2}{2\kappa n_1 \Gamma_0} \gg 1$$

Note : Fabry-Perot cavity

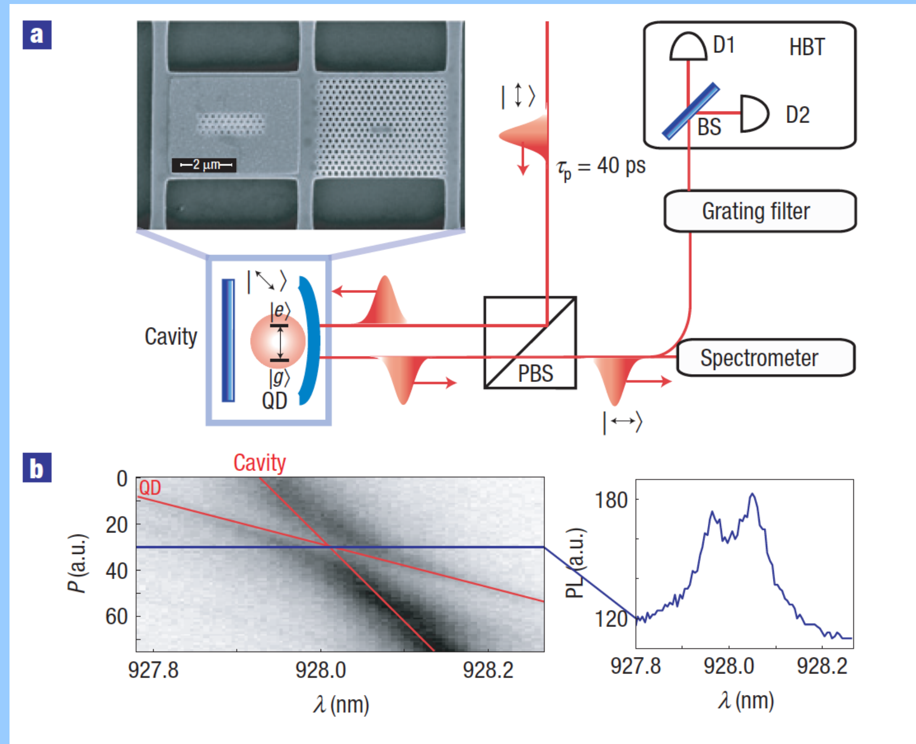
$$C = 4C_0 \frac{\mathcal{F}}{\pi} \qquad F_p = 2C$$

Strong coupling regime

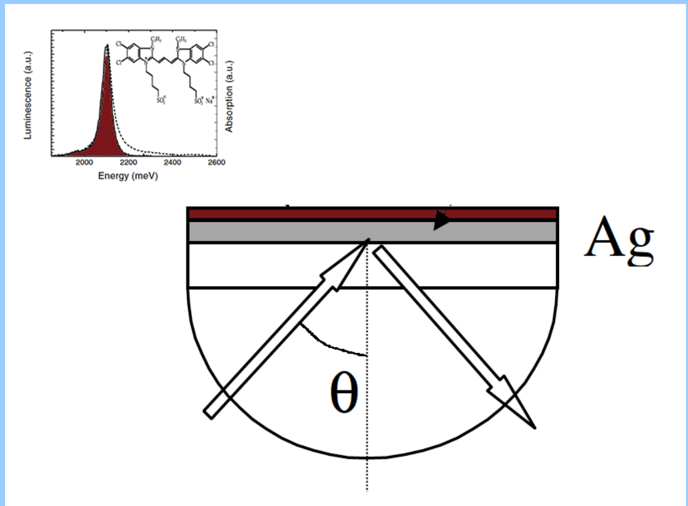


Vuckovic, Nature Physics **4**, 859 (2008)

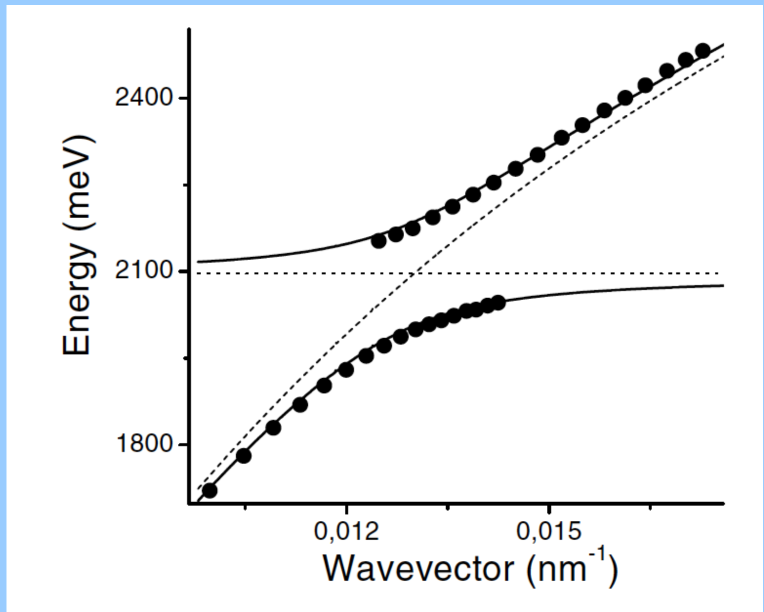
Generation of
non classical photon states



Strong coupling regime in plasmonics

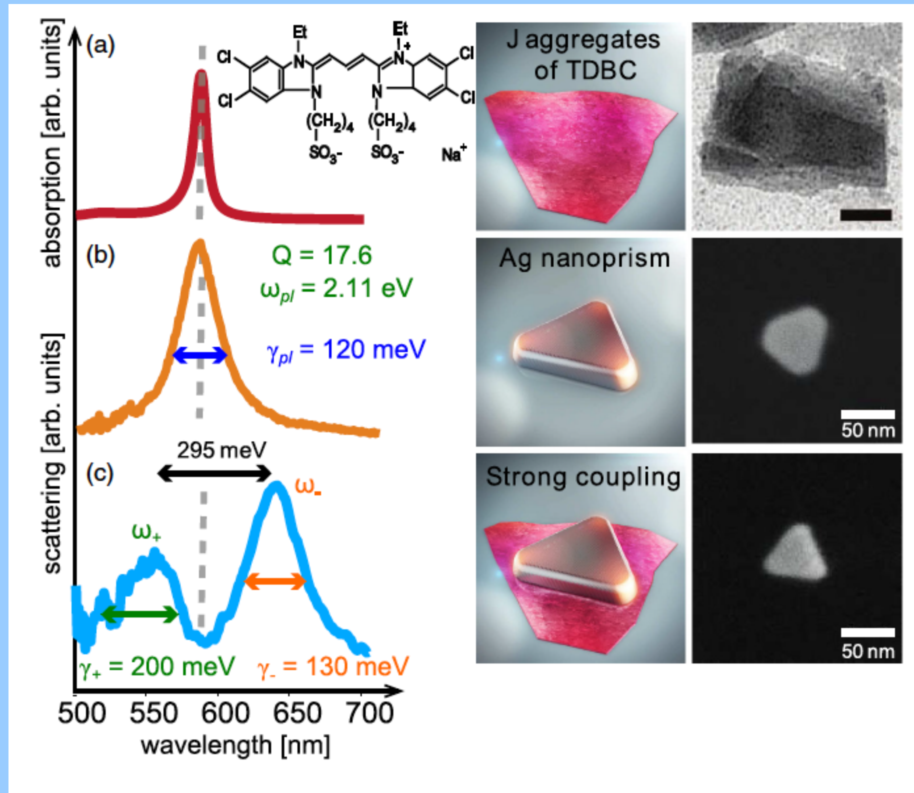


Delocalized SPP



Belessa *et al,* PRL **93**, 0364041 (2004)

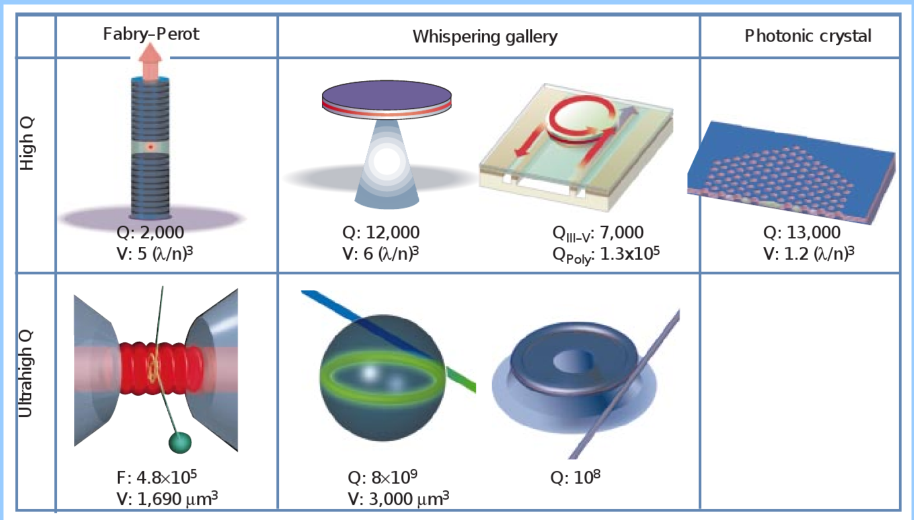
Localized SPP



Zengi *et al,* PRL **114**, 157401 (2015)

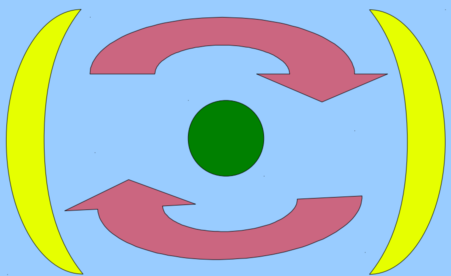
Towards integrated quantum nanophotonics

Cavity quantum electrodynamics (cQED)



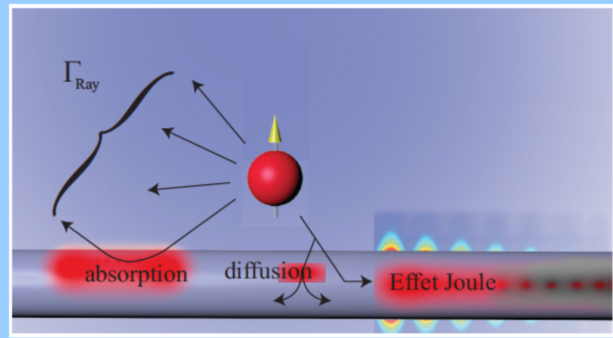
Purcell factor

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

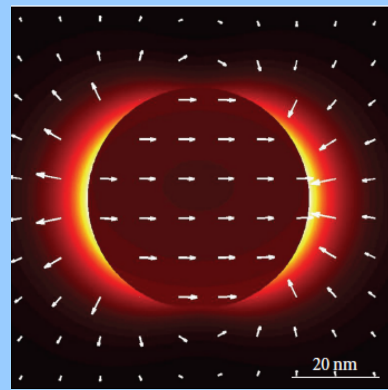


Duration of interaction (high Q)

Quantum plasmonics (cavity*less QED*)



Volume of interaction (sub-λ)



Derivation of the Purcell factor

Fermi rule
(weak coupling regime)

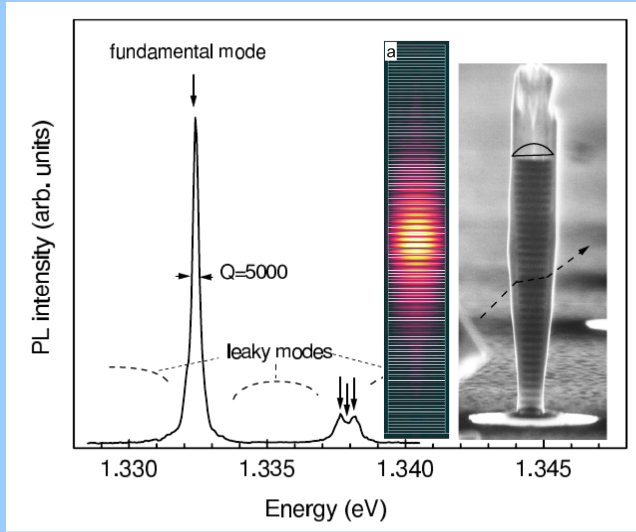
$$\Gamma(\mathbf{r}) = \frac{2\pi}{\hbar^2} \sum_{\mathbf{k}_n} |\langle a, \mathbf{k}_n | H_I | b, 0\rangle|^2 \delta(\omega_{em} - \omega_{\mathbf{kn}})$$

$$H_I = -\hat{\mathbf{p}} \cdot \hat{\mathbf{E}}(\mathbf{r})$$

$$\mathbf{E}_{cav}(\mathbf{r}, t) = i\sqrt{\frac{\hbar\omega_c}{2\varepsilon_0\varepsilon_1 V}}\mathbf{f}(\mathbf{r})e^{-i\omega_c t} + c.c$$

$$\delta(\omega - \omega_c) \rightarrow N(\omega) = \frac{1}{\pi}\frac{\kappa_{cav}/2}{(\omega - \omega_c)^2 + \kappa_{cav}^2/4}$$

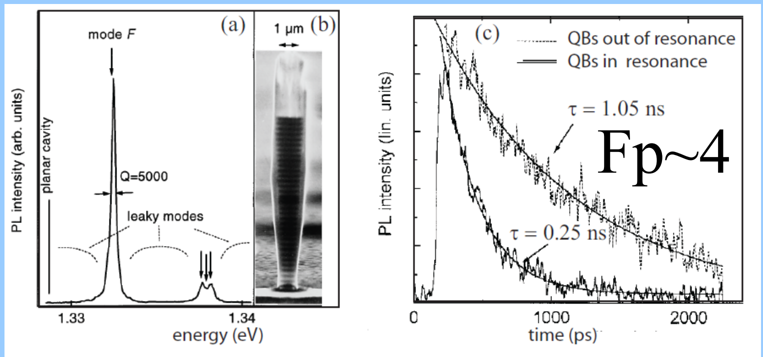
$$N(\omega) = \frac{2Q}{\pi\omega_c} \frac{1}{1 + 4Q^2(\frac{\omega - \omega_c}{\omega_c})^2}$$



$$\frac{\Gamma_{cav}}{n_1\Gamma_0} = \frac{3}{4\pi^2}\left(\frac{\lambda_{em}}{n_1}\right)^3 \frac{Q}{V} \frac{|\mathbf{u} \cdot \mathbf{f}(\mathbf{r})|^2}{1 + 4Q^2(\frac{\omega_{em} - \omega_c}{\omega_c})^2}$$

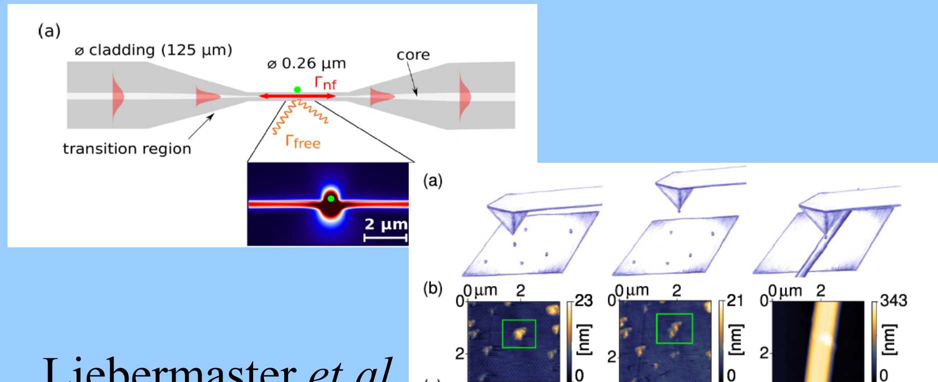
1D, 2D and 3D Purcell factor

Optical μ-cavity (3D confinement)



$$F_p = \frac{\Gamma_{cav}}{n_1 \Gamma_0} = \frac{3}{4\pi^2}\left(\frac{\lambda_{em}}{n_1}\right)^3 \frac{Q}{V_{eff}}$$

Nanofiber (2D confinement)

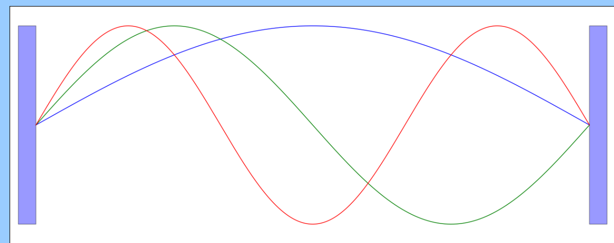


Liebermaster *et al*, Appl. Phys. Lett. **104**, 031101 (2014)

$$F_p = \frac{\Gamma_{guided}}{n_1 \Gamma_0} = \frac{3}{4\pi}\frac{(\lambda_{em}/n_1)^2}{A_{eff}}\frac{n_g}{n_1}$$

Coupling efficiency ~ 10 %
(ideally 36%)

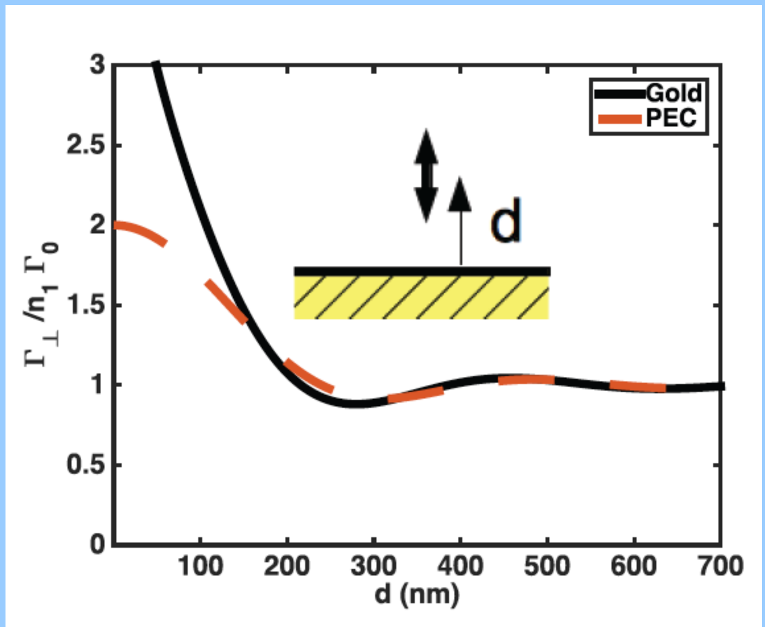
Fabry-Perot cavity (1D confinement)

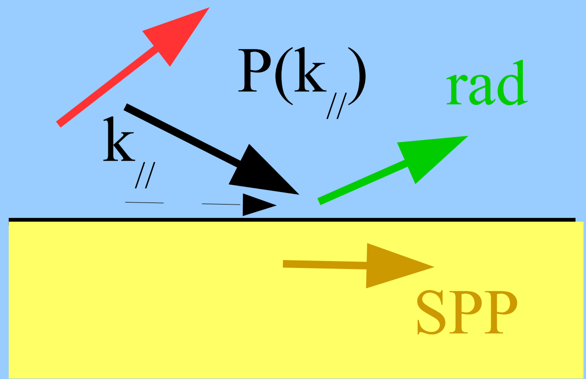


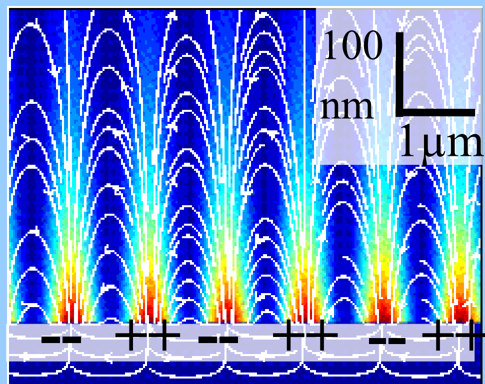
$$F_p = \frac{\Gamma_{guided}}{n_1 \Gamma_0} = \frac{3}{4}\frac{(\lambda_{em}/n_1)}{L_{eff}}\frac{n_{eff} n_g}{n_1^2}$$

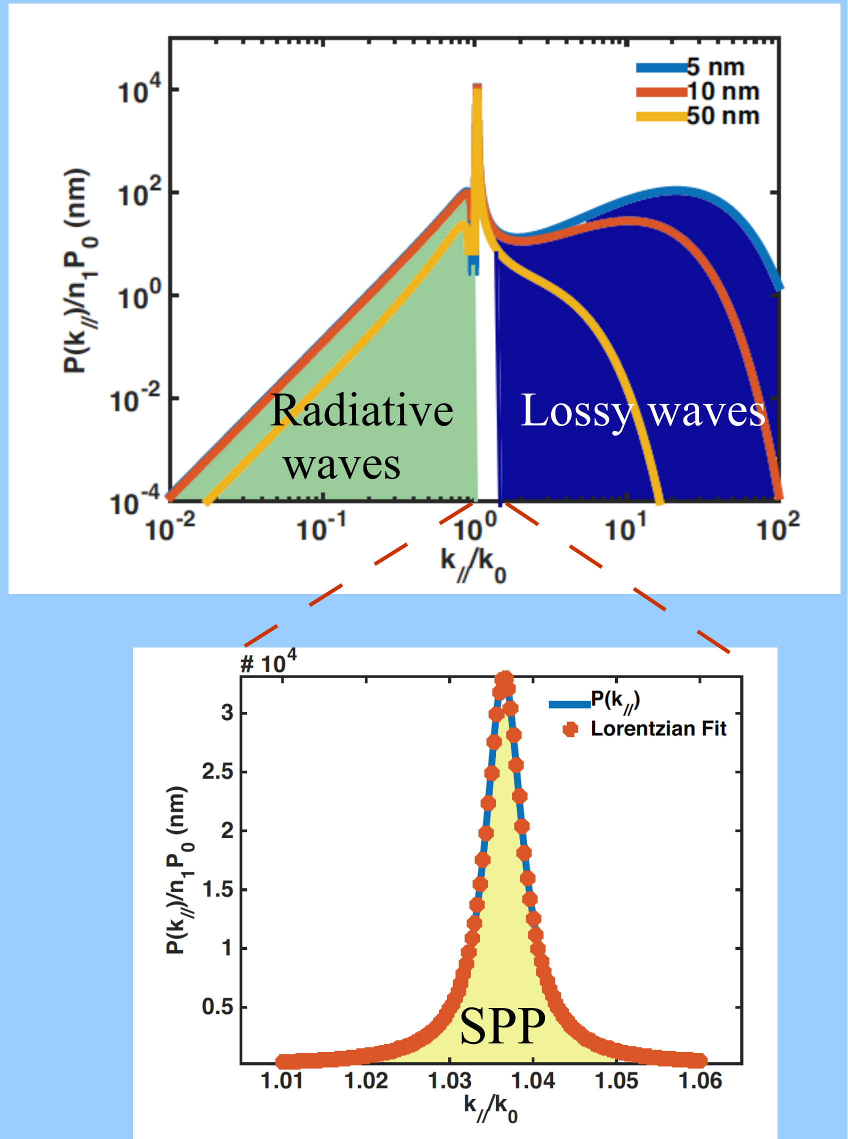
The planar film revisited

$$\frac{\Gamma_u(d)}{n_1\Gamma_0} = \int_0^\infty \mathcal{P}(k_\parallel)dk_\parallel$$



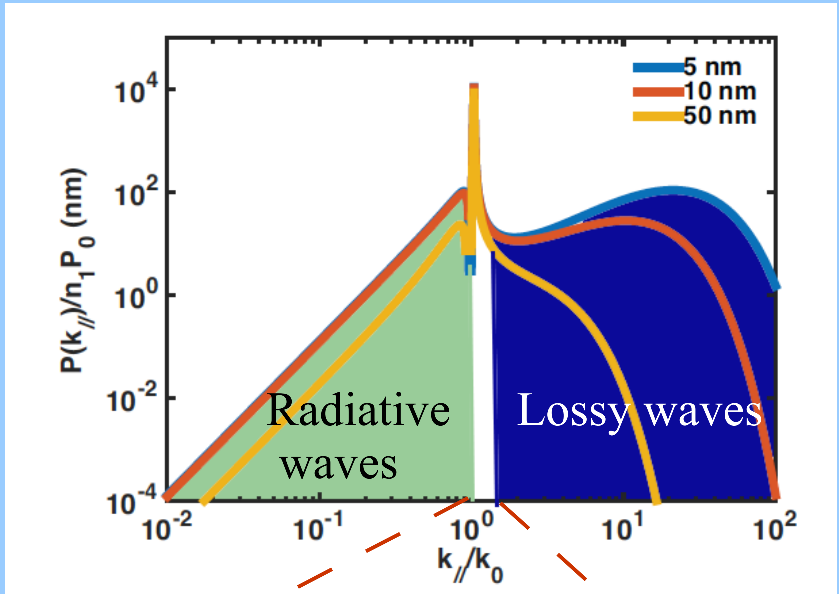


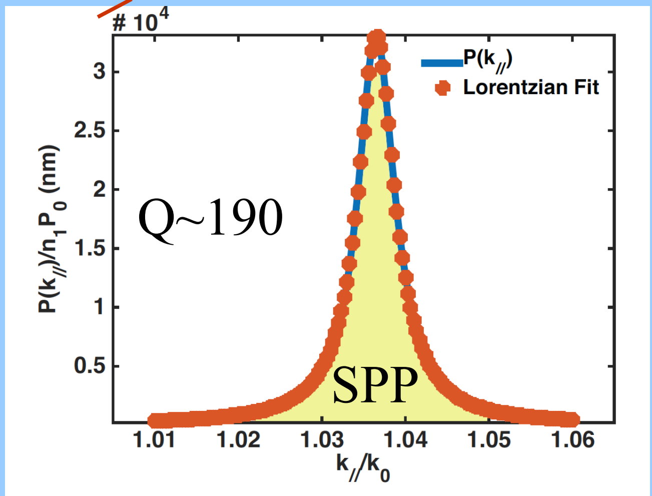


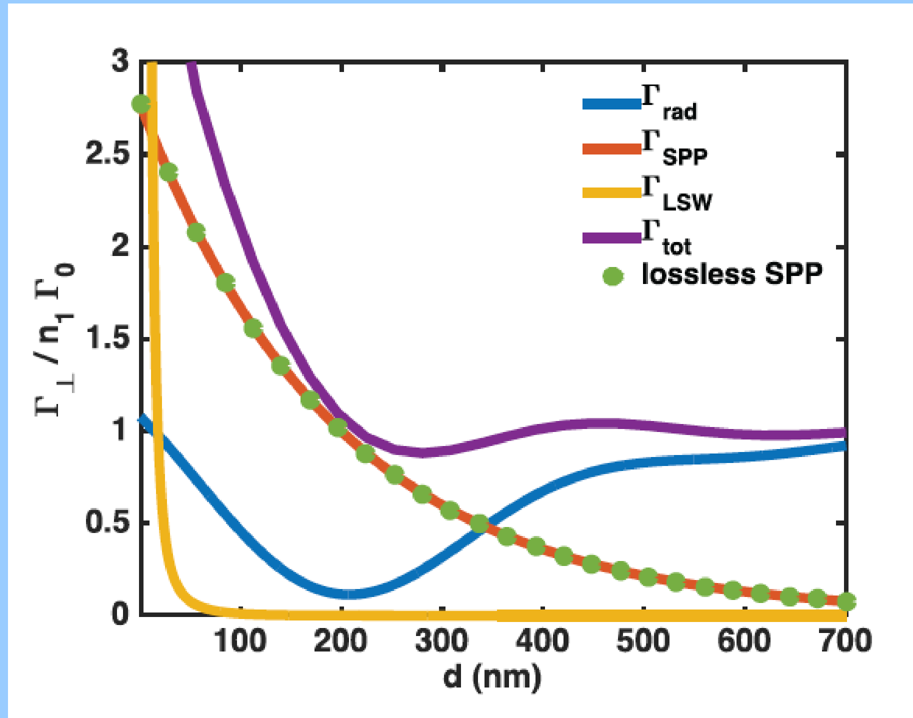


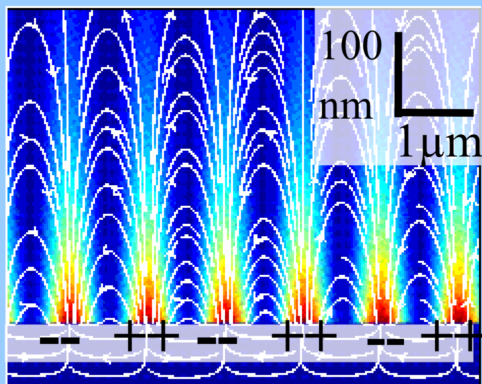
Adapted from
Barnes, J. Modern Optics (1998)

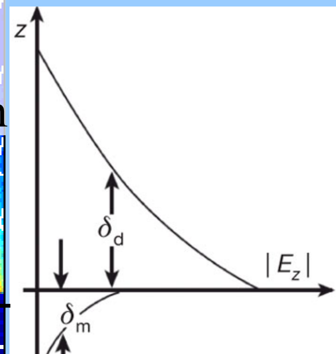
Decay channels











$$L_{eff} = \frac{\int |\mathbf{E}(z)|^2 dz}{Max\,[|\mathbf{E}(z)|^2]} = \int_0^\infty e^{-2z/\delta} dz = \frac{\delta}{2}$$

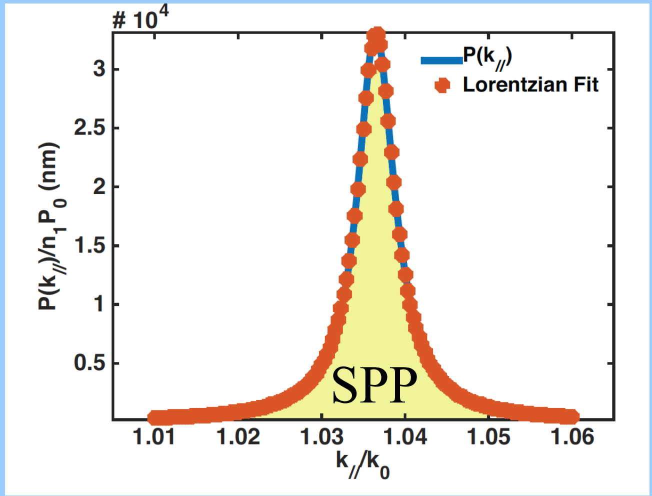
L_{eff}~penetration depth in air

$$\sim 0.5 \frac{\lambda_{SPP}}{2}$$

Plasmonic Purcell factor and coupling efficiency to surface plasmons. Implications for addressing and controlling optical nanosources

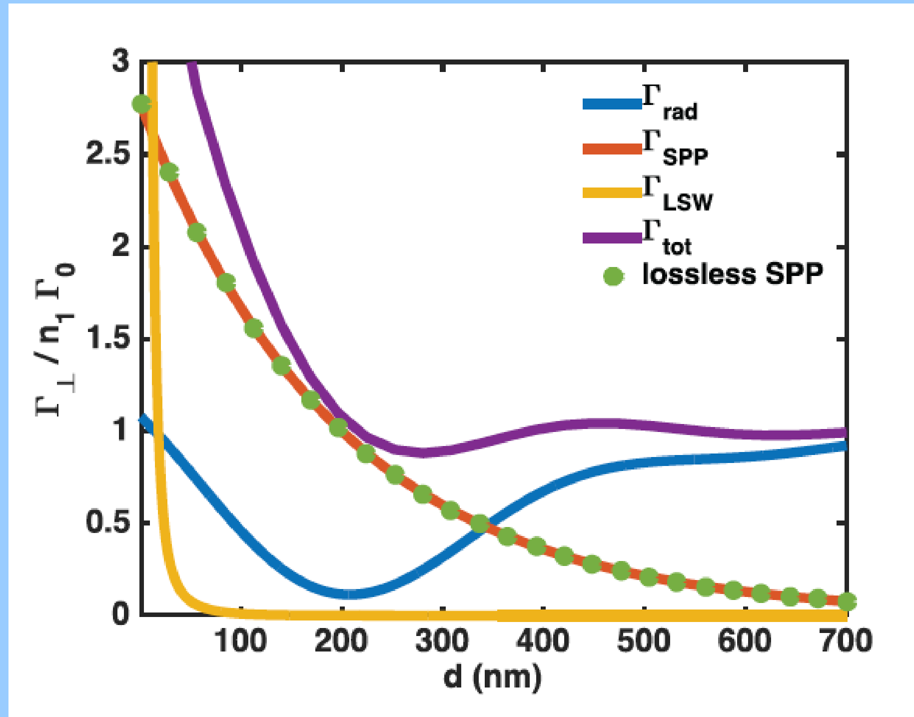
G. Colas des Francs *et al, J. Opt.* (accepted, 2016)

Losses



Lorentzian =>
(lossy metal)

$$\frac{\Gamma_{SPP}}{n_1\Gamma_0} = \frac{\pi}{2}\frac{\mathcal{P}(k_{SPP})}{L_{SPP}}$$



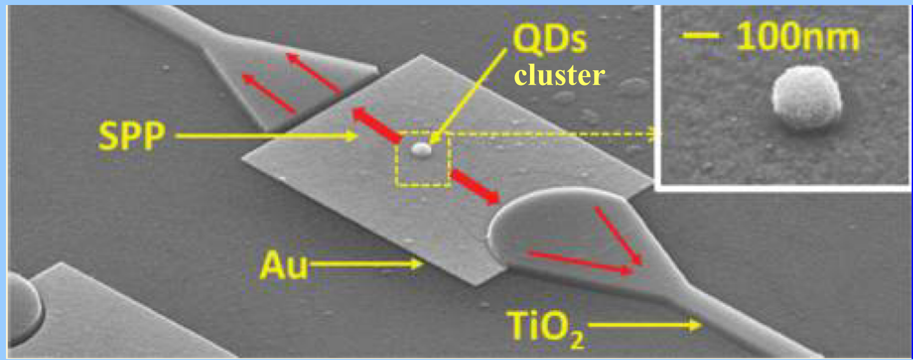
Lossless ideal metal

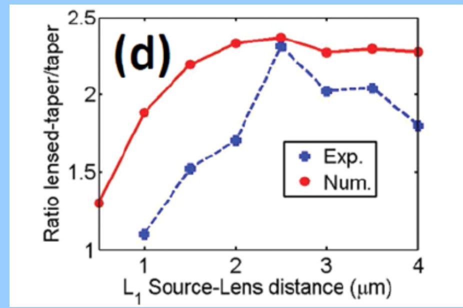
$$\frac{\Gamma_{\perp}(d)}{n_1\Gamma_0} = \frac{3\pi}{n_1^3}\frac{n_{SPP}^5}{\varepsilon_1 - \varepsilon_2}|\varepsilon_2|^{1/2}e^{-2(\varepsilon_1/|\varepsilon_2|)^{1/2}k_{SPP}d}$$

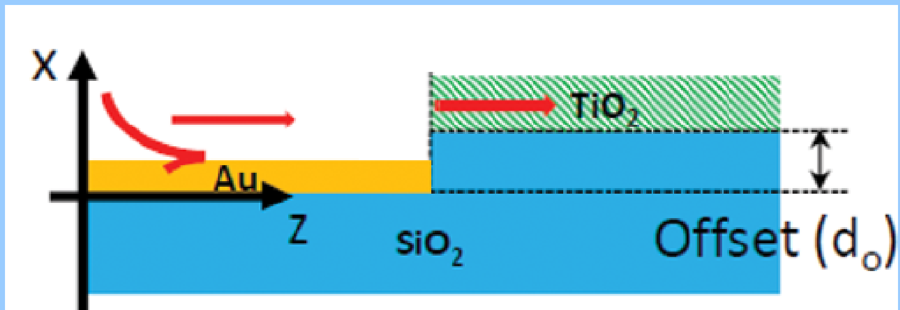
Γ_{SPP} does not depend on losses (coupling to a mode then propagates with or without losses)

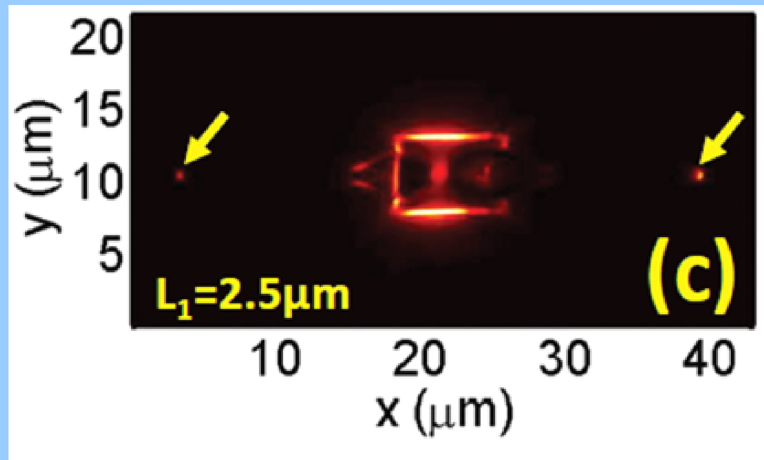
High Purcell factor

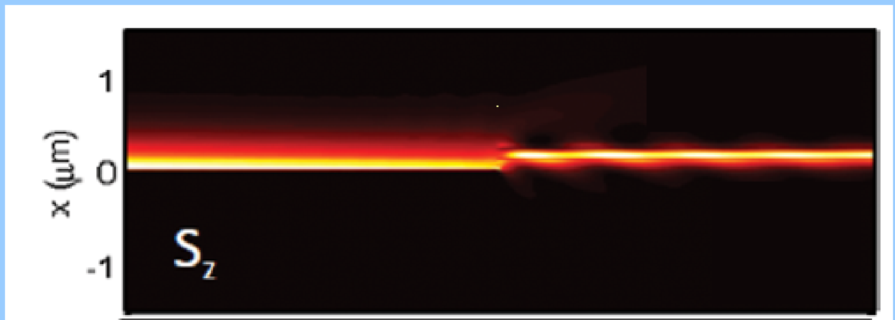
Overcoming losses – hybrid plateform





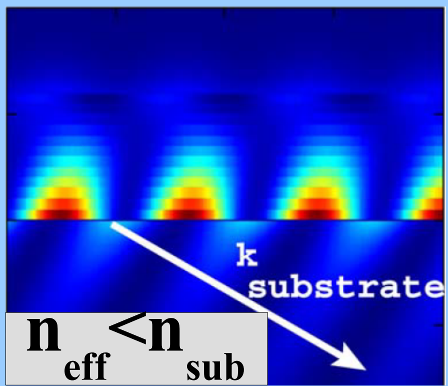


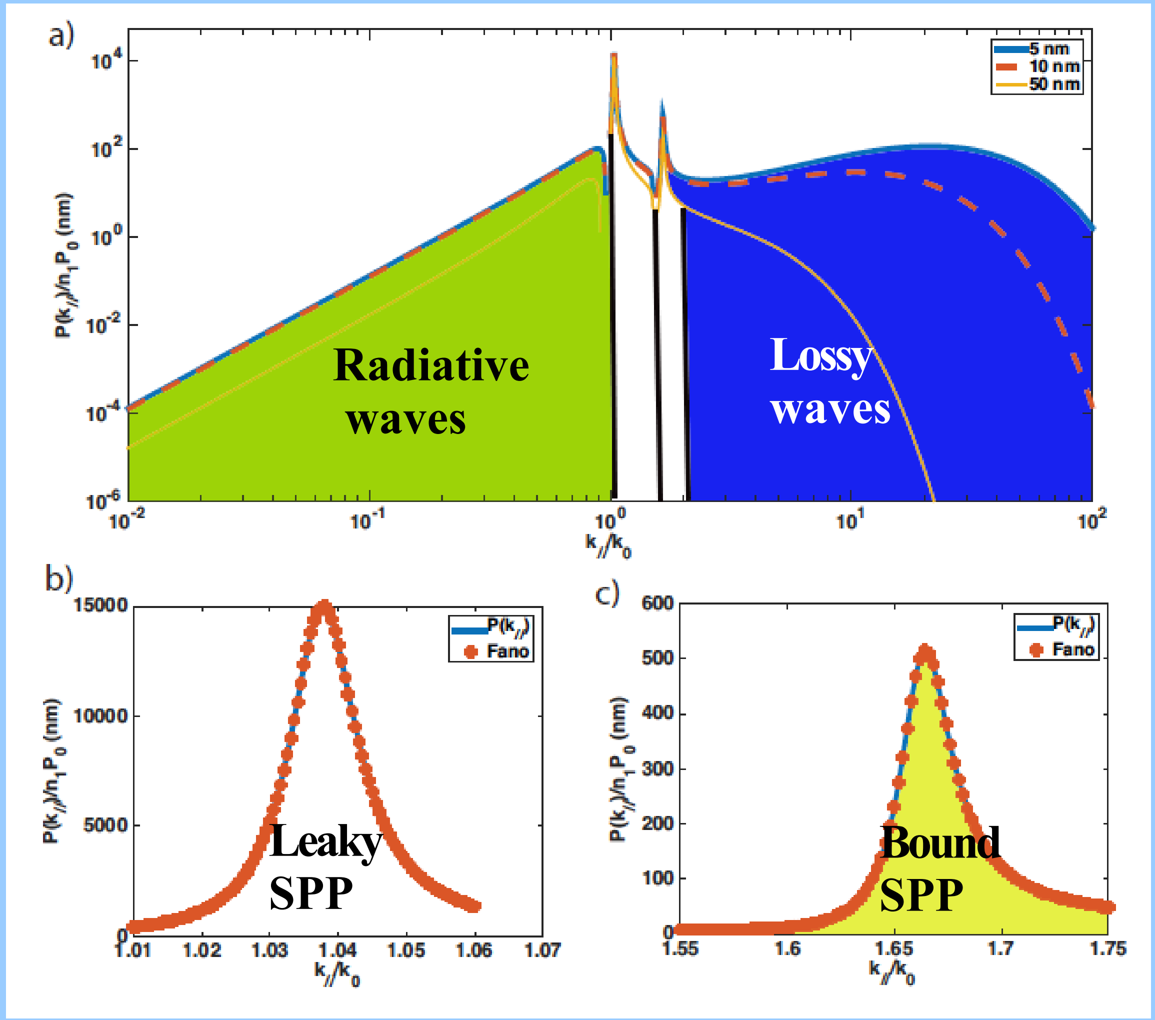




Colloidal Quantum Dot Integrated Light Sources for Plasmon Mediated Photonic Waveguide Excitation
Weeber *et al*, ACS Phot. **3**, 844 (2016)

Thin film – leaky SPP

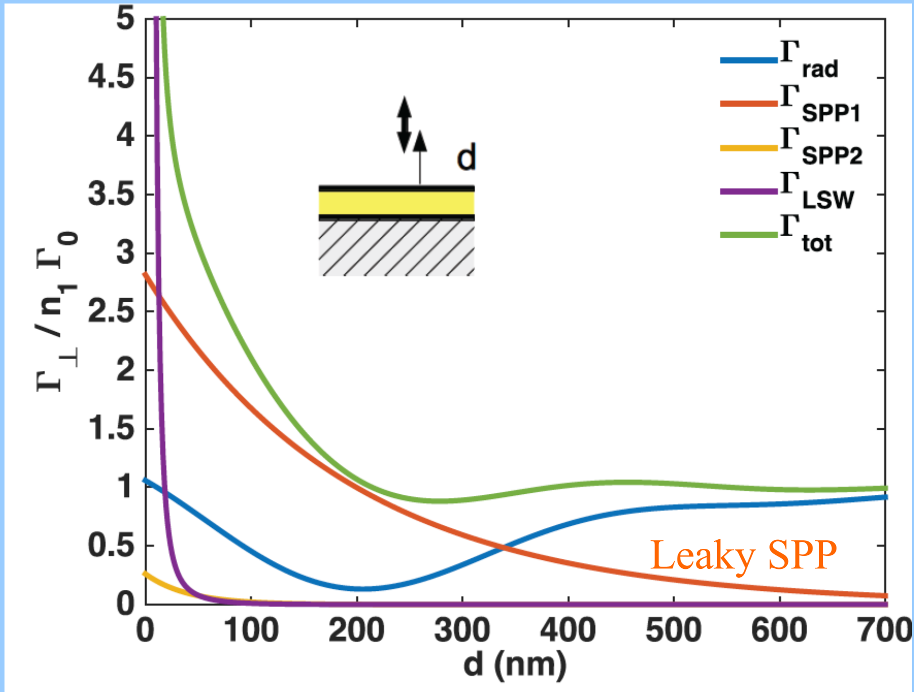




(Fano profile)

Decay channels

Relaxation channels



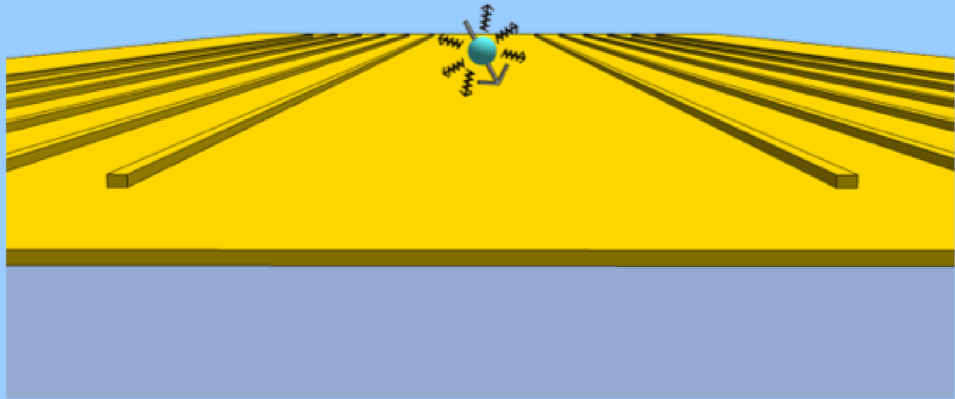
	Q	L_{eff} (nm)	$\delta/2$ (nm)	n_g	F_p
leaky	85	159	193	0.92	3.02
bound	57	27	76	0.57	5.1

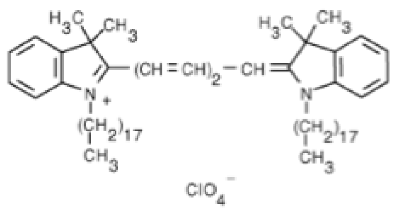
$$L_{SPP} = \frac{1}{\alpha_{abs}} + \frac{1}{\alpha_{leak}}$$

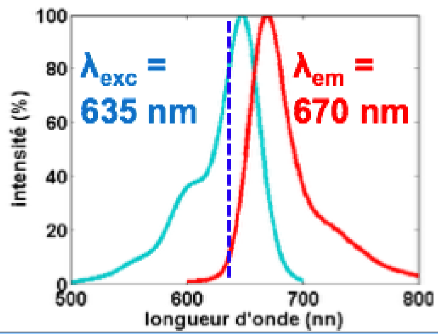
t_{Au}~50 nm=> 50 % leakage

Detectable using high NA (SPCE)

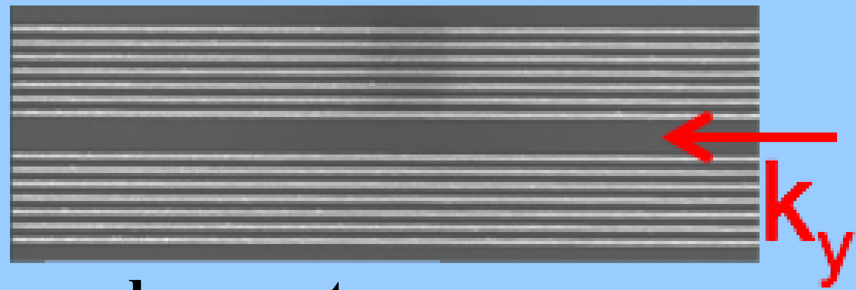
In plane SPP cavity







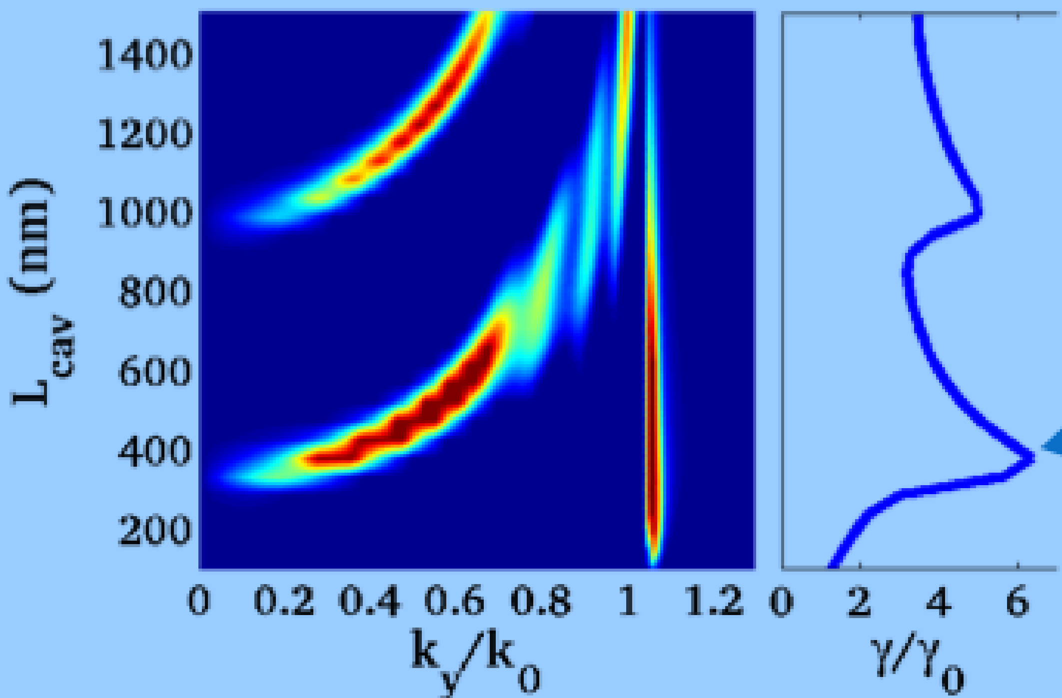
In plane SPP cavity

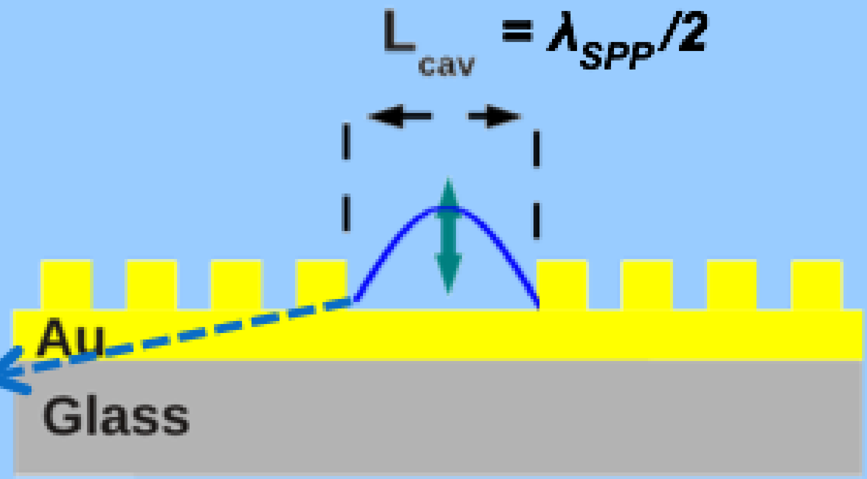


emitted power
P(ky)

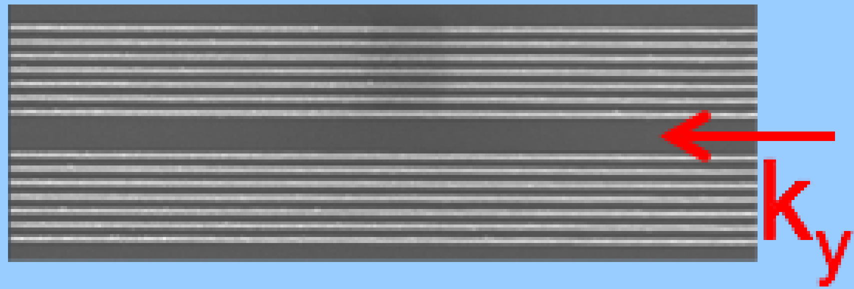
decay rate

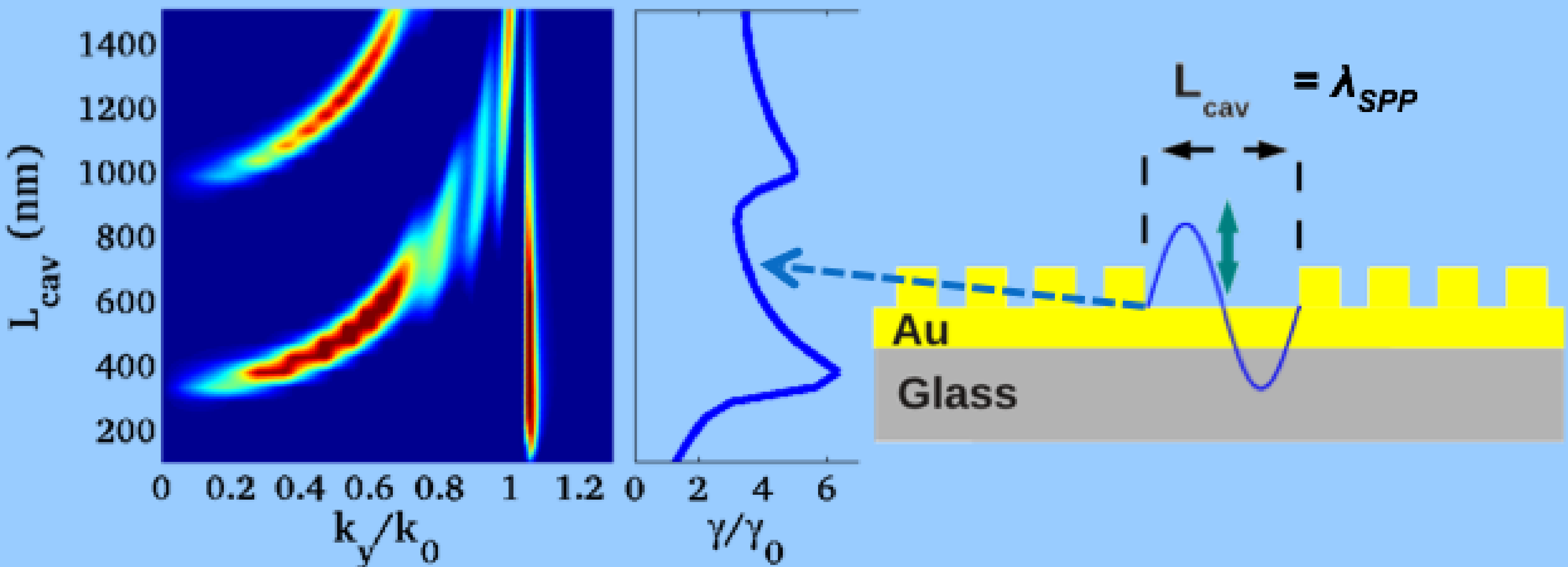
$$\gamma \propto \int P(k_y)\, dk_y$$



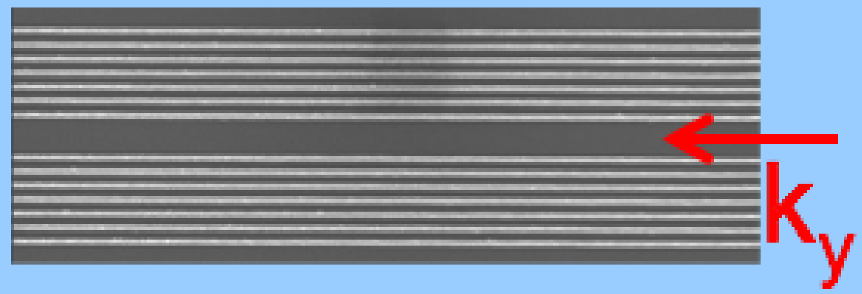


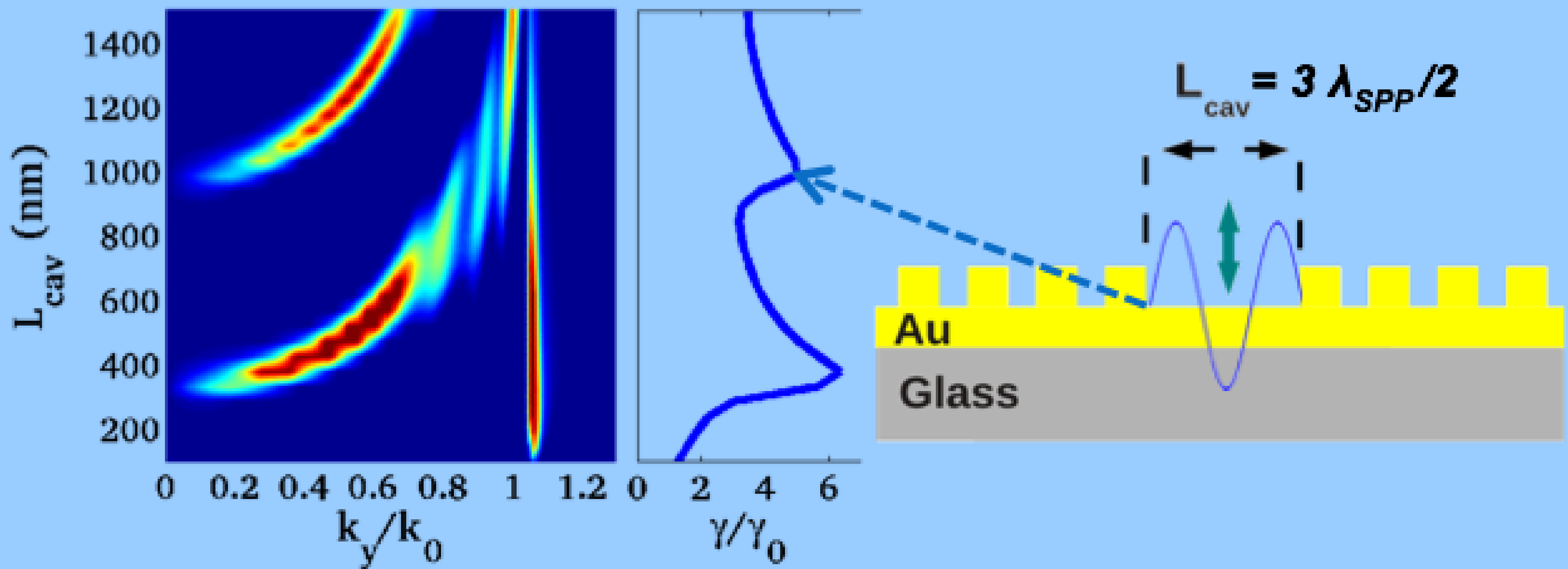
In plane SPP cavity



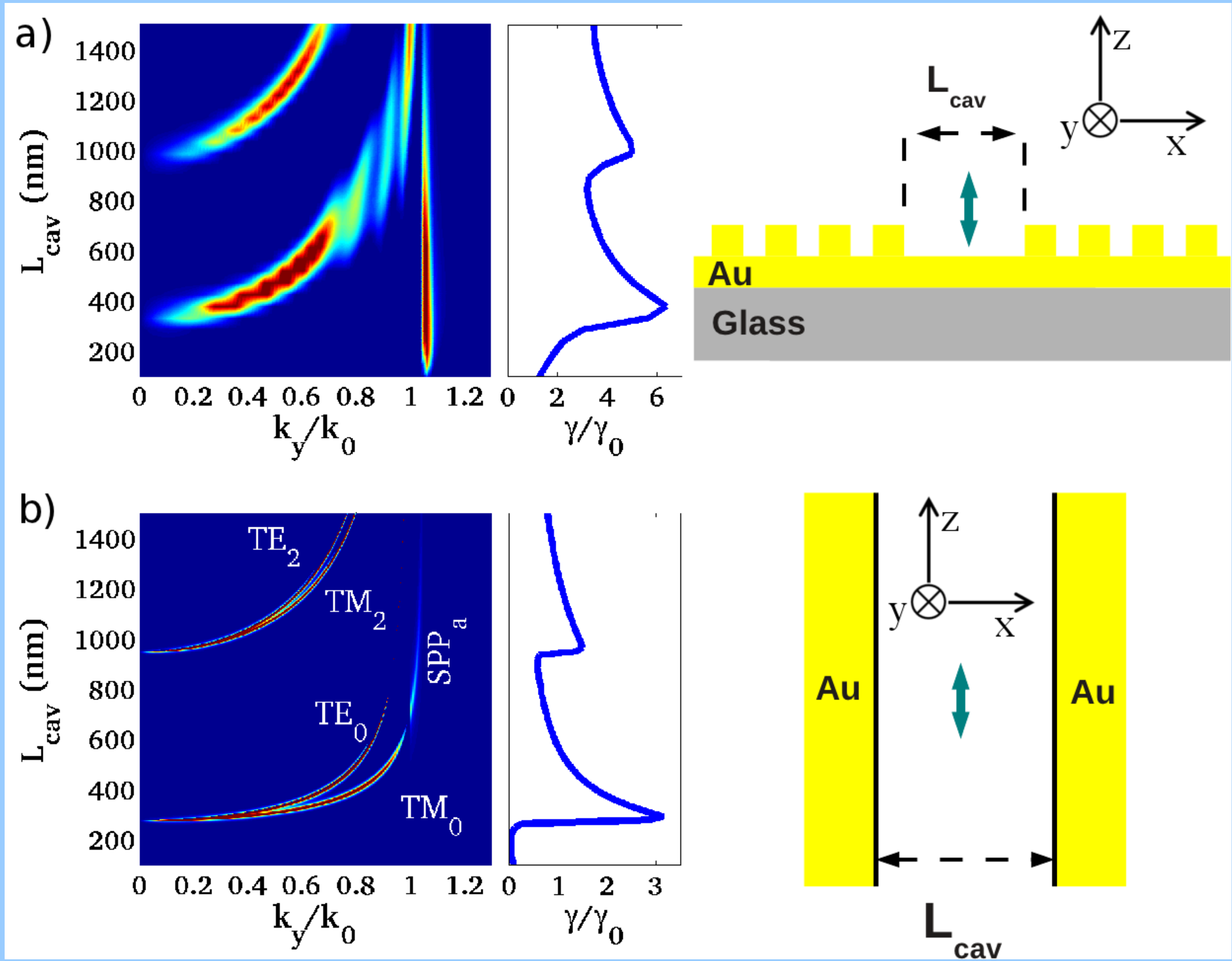


In plane SPP cavity

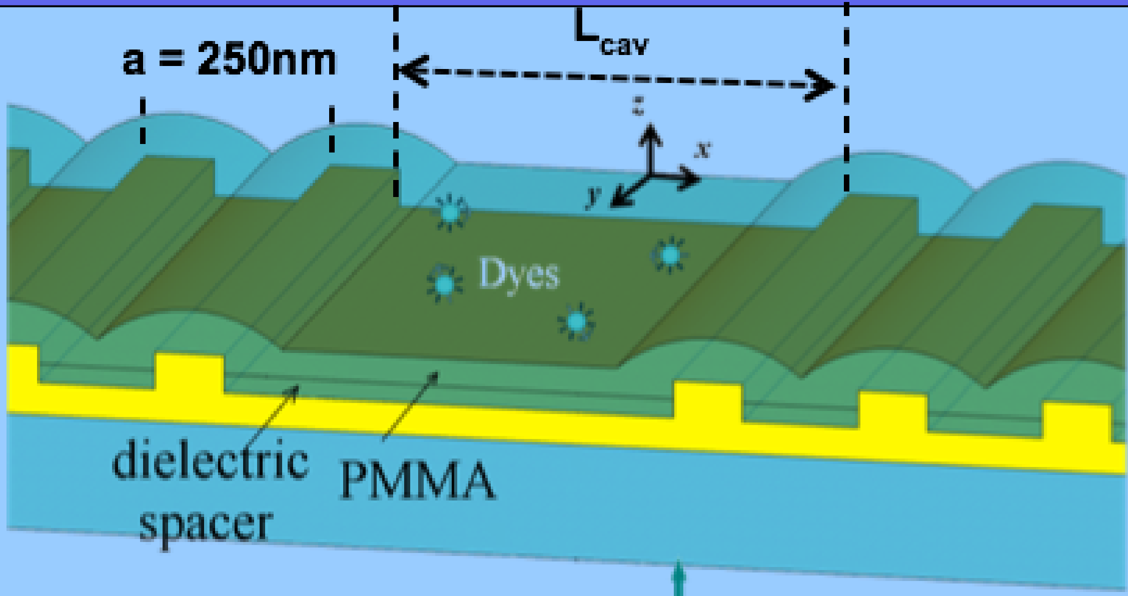




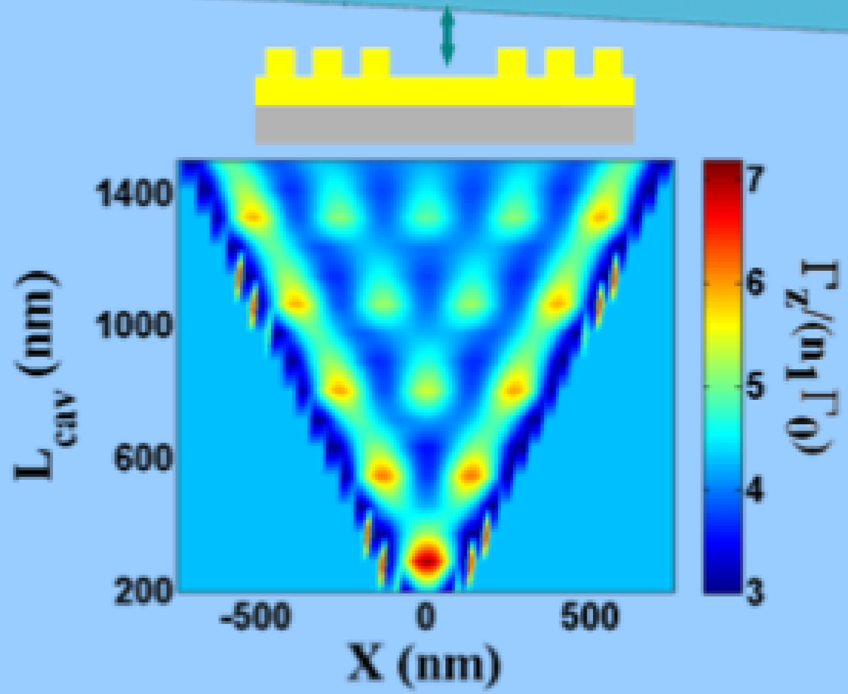
Comparison between planar and bulk cavities

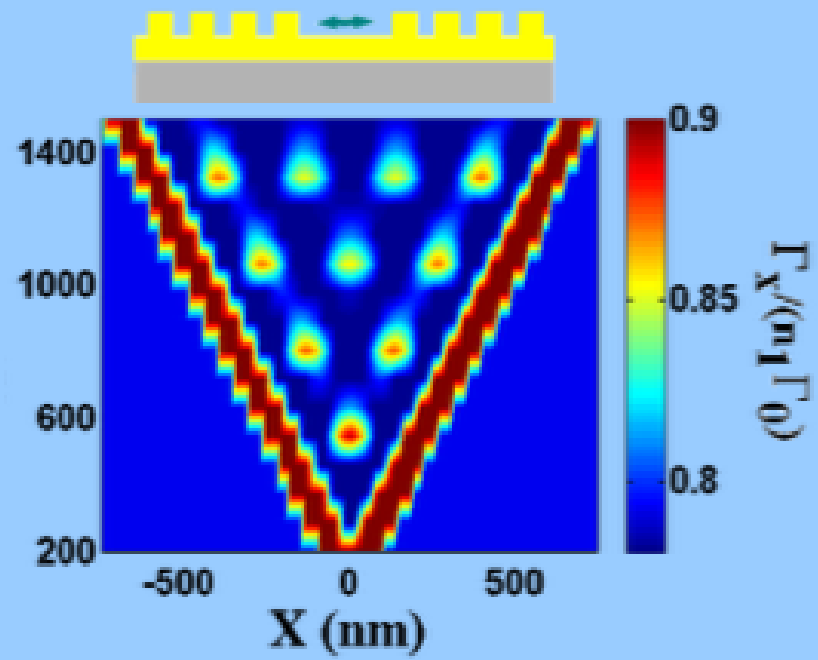


In plane SPP cavity

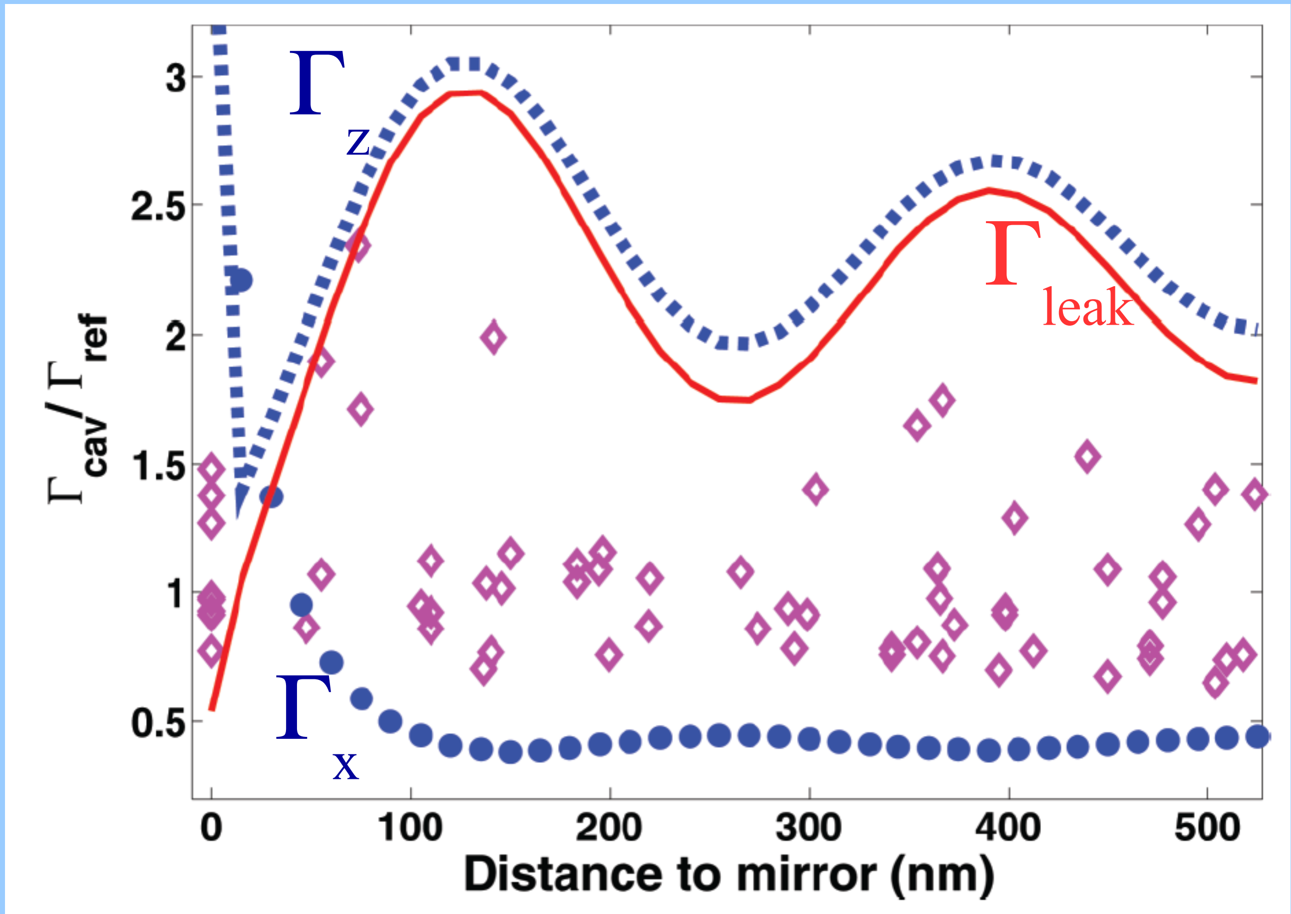


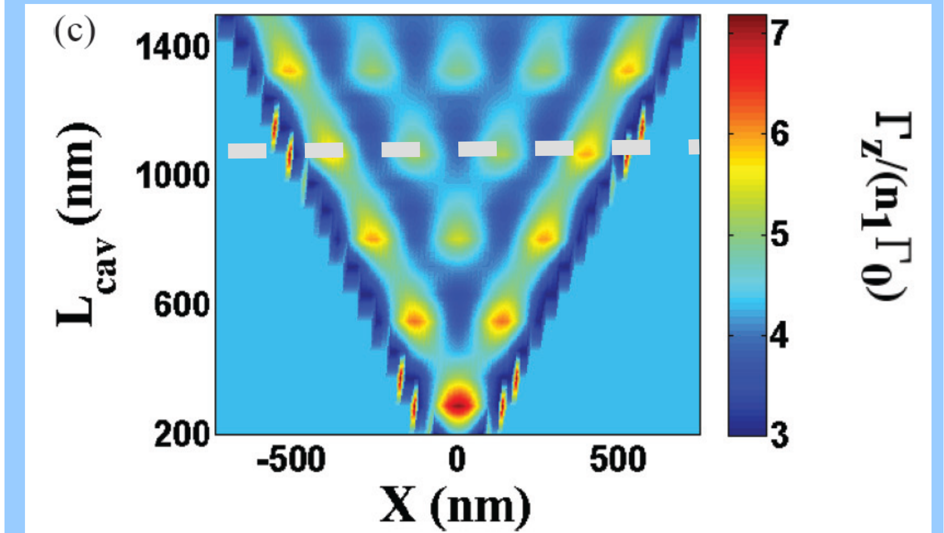
Decay rate depends on
- the cavity length
- the molecule position
- the molecule orientation





Single cavity ($L_{cav}=2\ \lambda_{SPP}=1{,}1\mu m$)





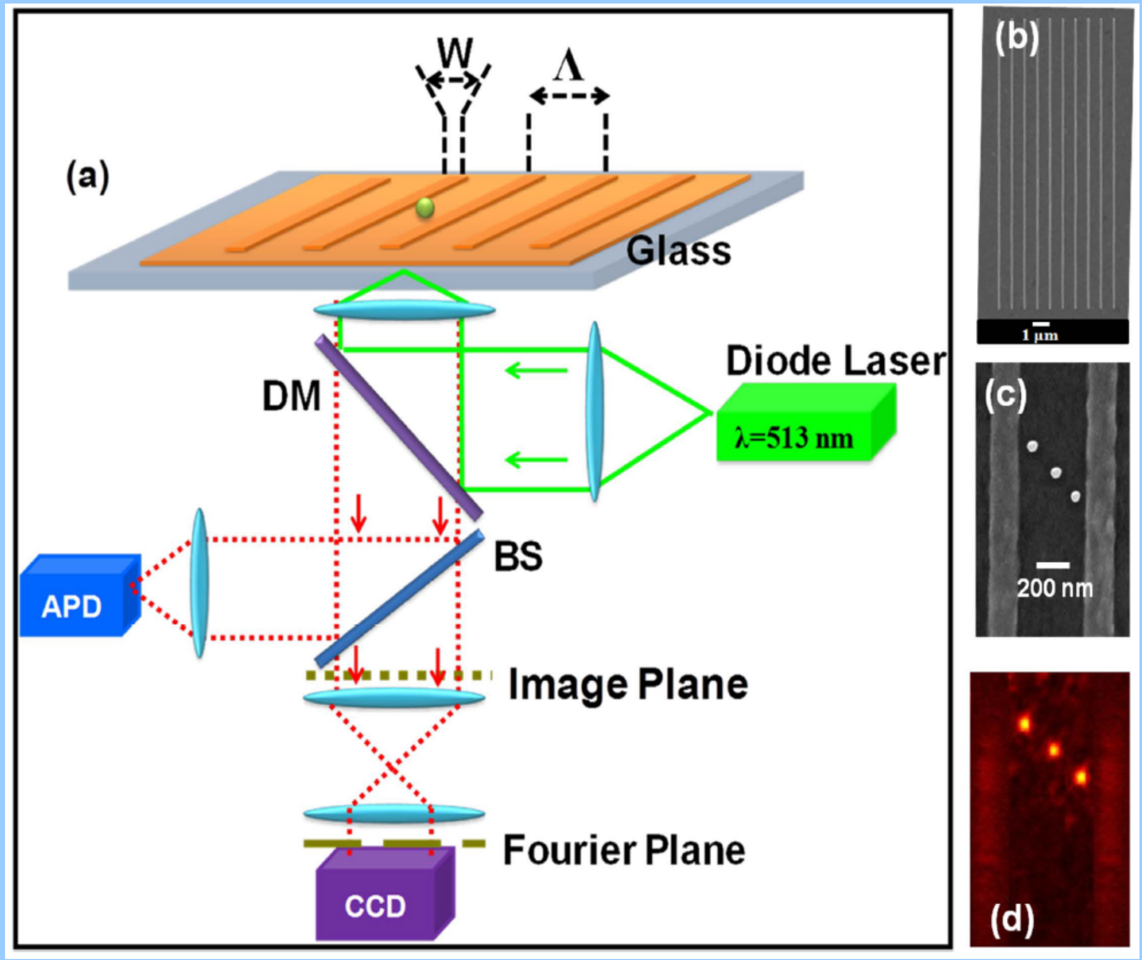
Fp ~7 ($\frac{\lambda_{SPP}}{2}$ cavity)

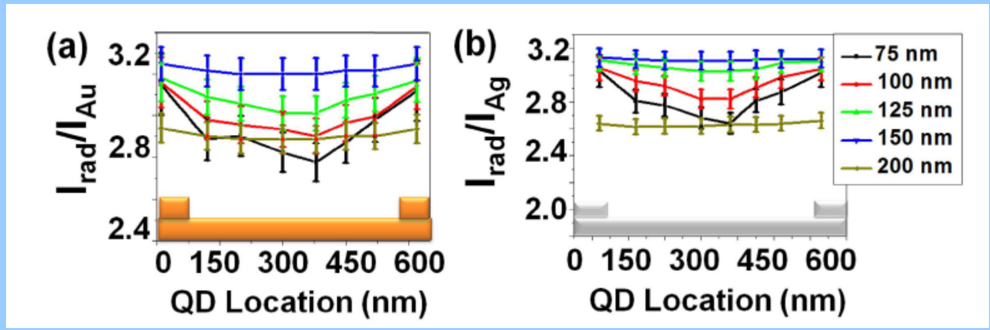
$$\frac{\Gamma_{leak}}{\Gamma_{tot}} \approx 0.8$$

Extraction efficiency (leakage into the substrate)

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

Grating decoupler

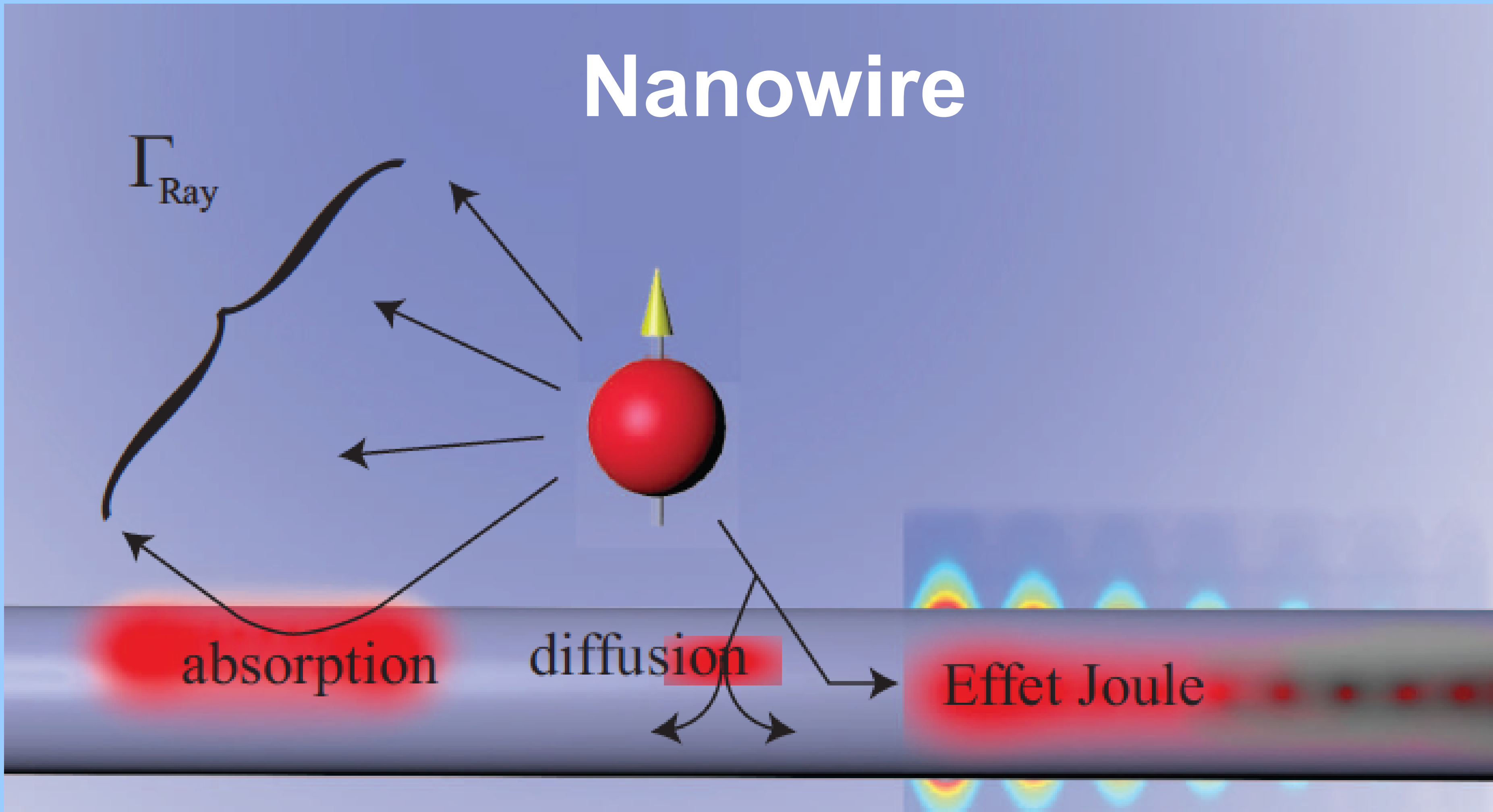




Fluorescence enhancement
(independent of position)

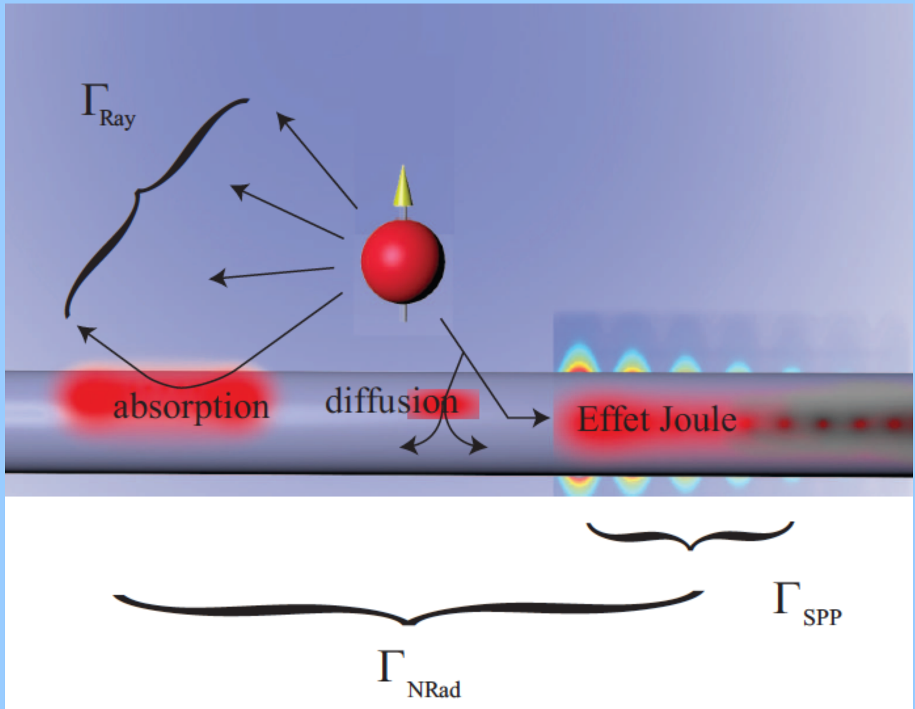
Extraction efficiency

Spatially uniform enhancement of single quantum dot emission
using plasmonic grating decoupler
Kumar *et al*, Sci. Rep. **5**, 16796 (2015)



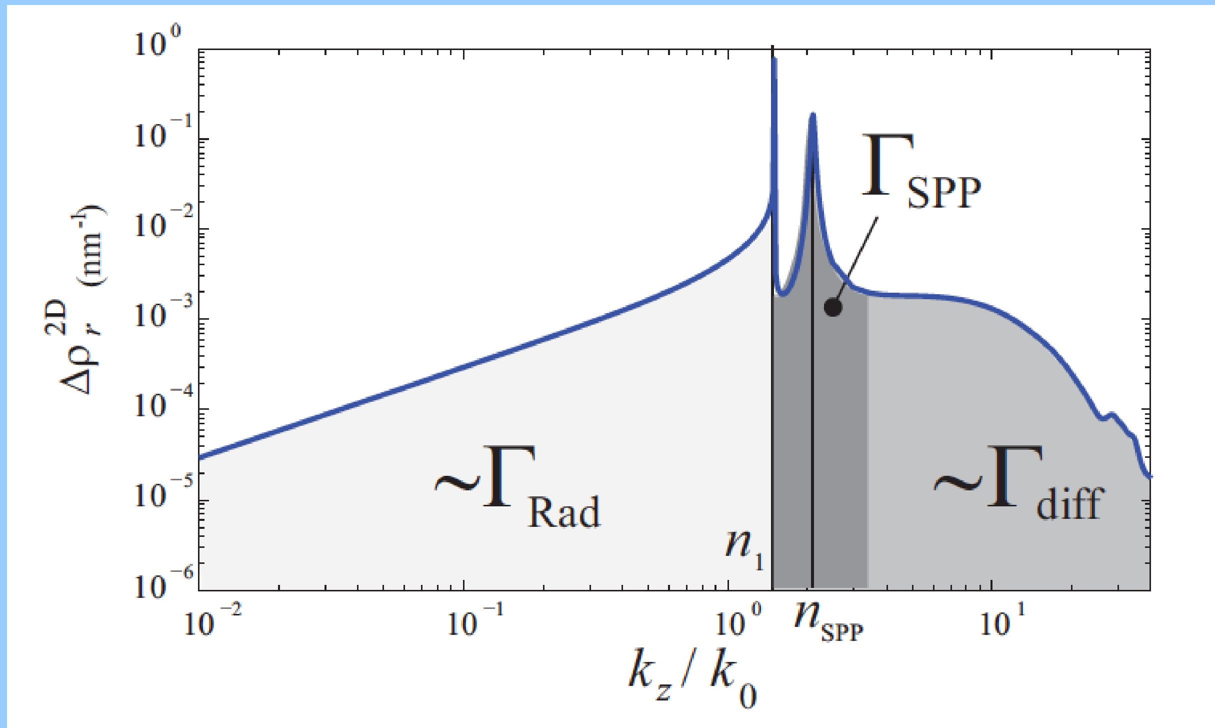
Nanowire
Γ_{Ray}
absorption
diffusion
Effet Joule

Decay channels



2D−DOS

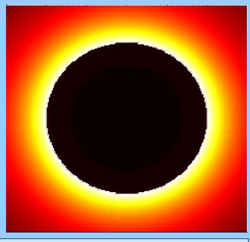
$$\rho^{2D}(k_z) = \frac{-2k_z}{\pi} Im Tr[\epsilon \mathbf{G}^{2D}(k_z)]$$

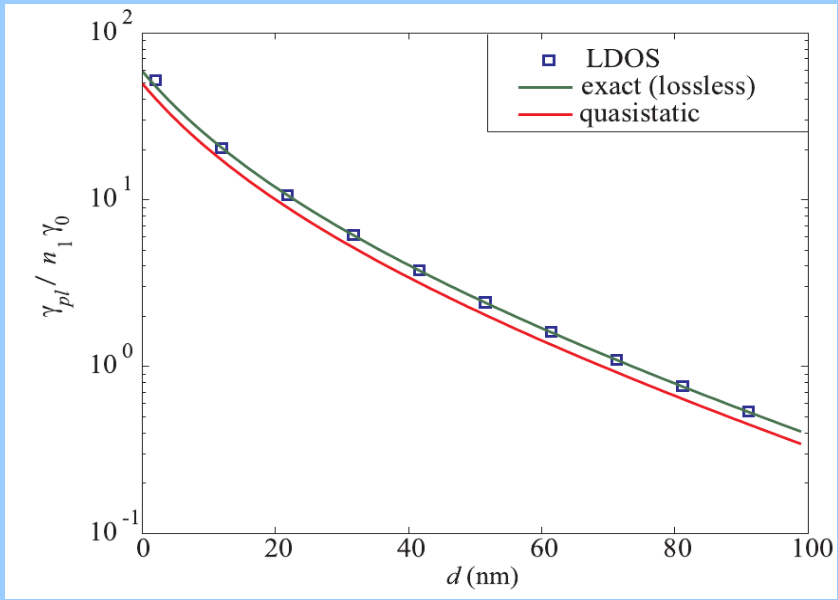


Purcell factor for point-like dipolar emitter coupling to 2D plasmonic waveguides
Barthes *et al*, Physical Review B **84** 073403 (2011)

Purcell and β factors

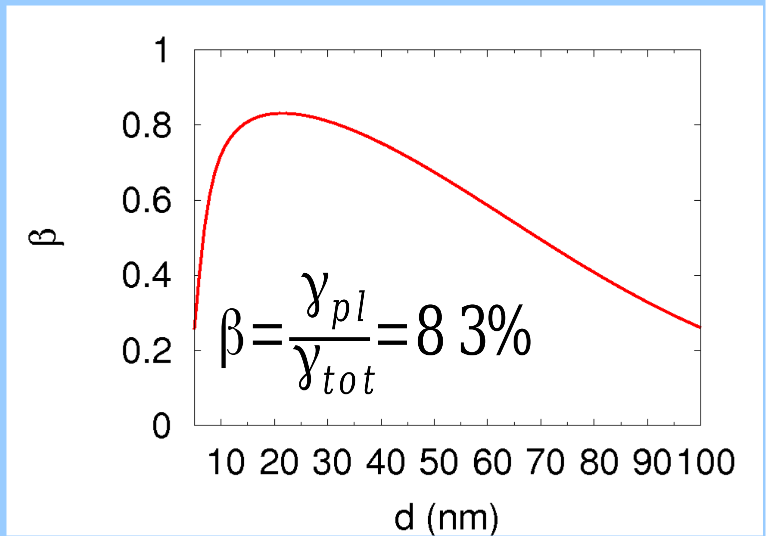
Overlap with the guided mode





$$\frac{\Gamma_{spp}}{\Gamma_0} = \frac{3}{8\, n_{eff}} \left(\frac{\lambda}{n}\right)^2 \frac{\rho^{2D}(k_{spp})}{L_{spp}}$$

propagation in the 3rd direction



High coupling efficiency (β-factor)

Purcell factor for point-like dipolar emitter coupling to 2D plasmonic waveguides
Barthes *et al*, Physical Review B **84** 073403 (2011)

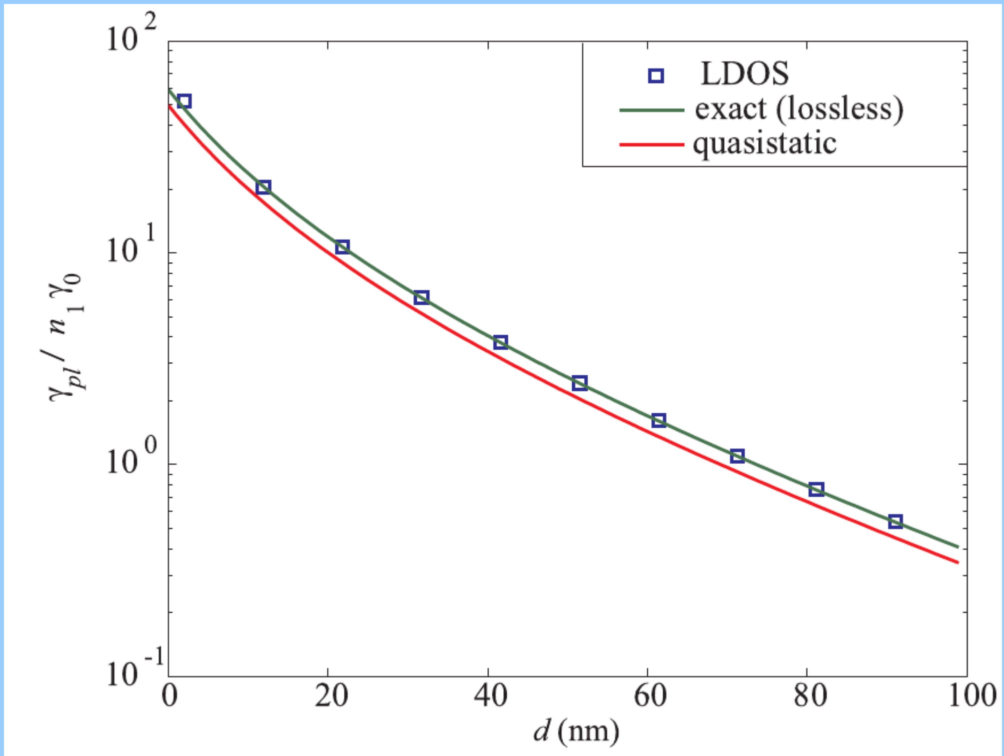
Effect of Joule losses

Exact *lossless* case

$$\frac{\Gamma_{spp}}{\Gamma_0}=\frac{3\pi c}{k_0^2}\frac{E_u(d)E_u^*(d)}{\int_{A\infty}(\mathbf{E}\times\mathbf{H}^*).\mathbf{z}\,dA}$$

2D-LDOS (*lossy*)

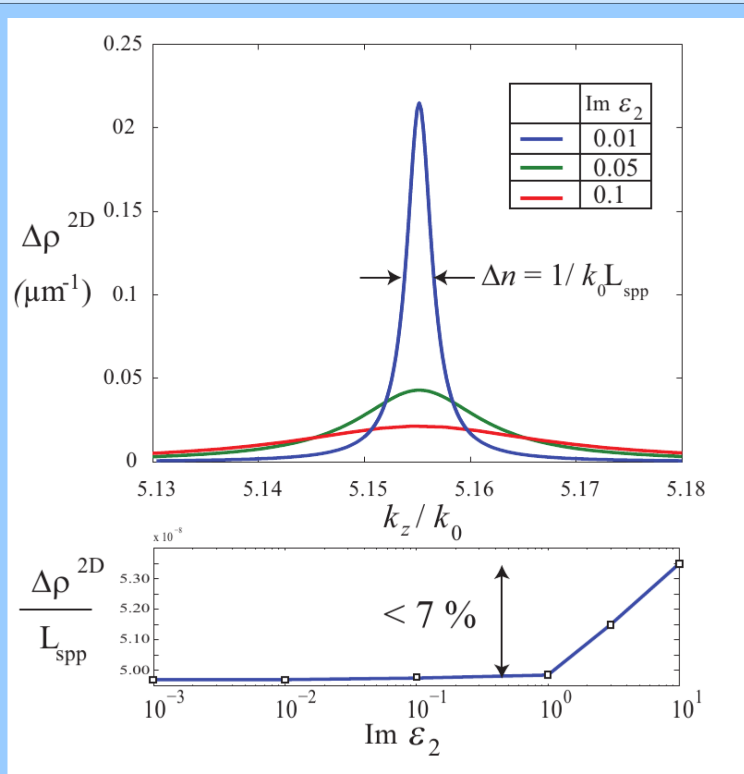
$$\frac{\Gamma_{spp}}{\Gamma_0}=\frac{3}{8n_{eff}}\left(\frac{\lambda}{n}\right)^2\frac{\rho^{2D}(k_{spp})}{L_{spp}}$$



Nb of modes

$$N\propto\int dk_z\rho^{2D}(k_z)\propto\frac{\rho^{2D}(k_{spp})}{L_{spp}}$$

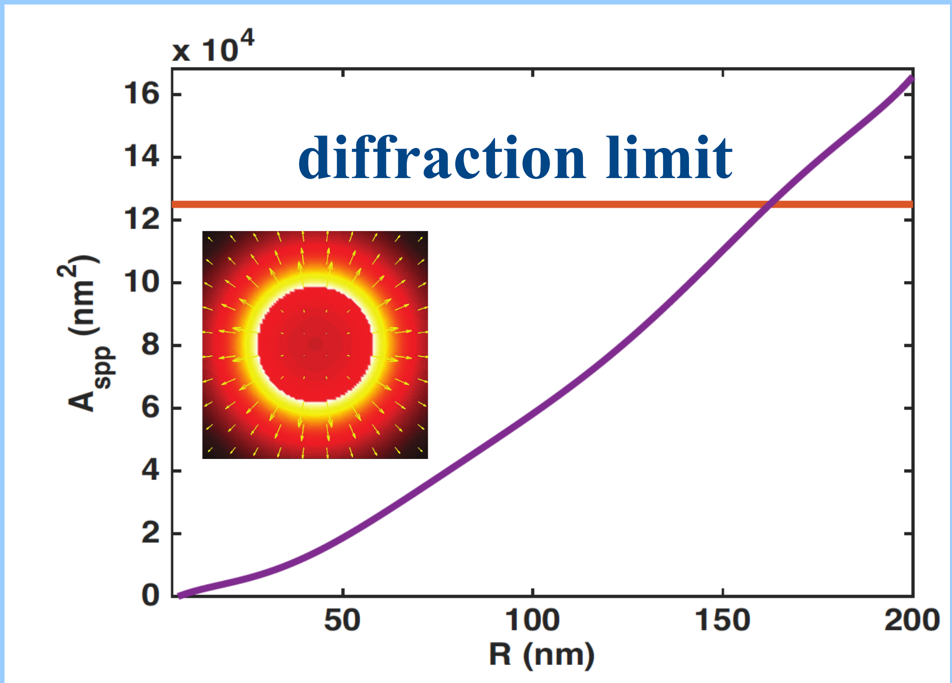
independent on losses



Purcell factor for point-like dipolar emitter coupling to 2D plasmonic waveguides
Barthes *et al*, Physical Review B **84** 073403 (2011)

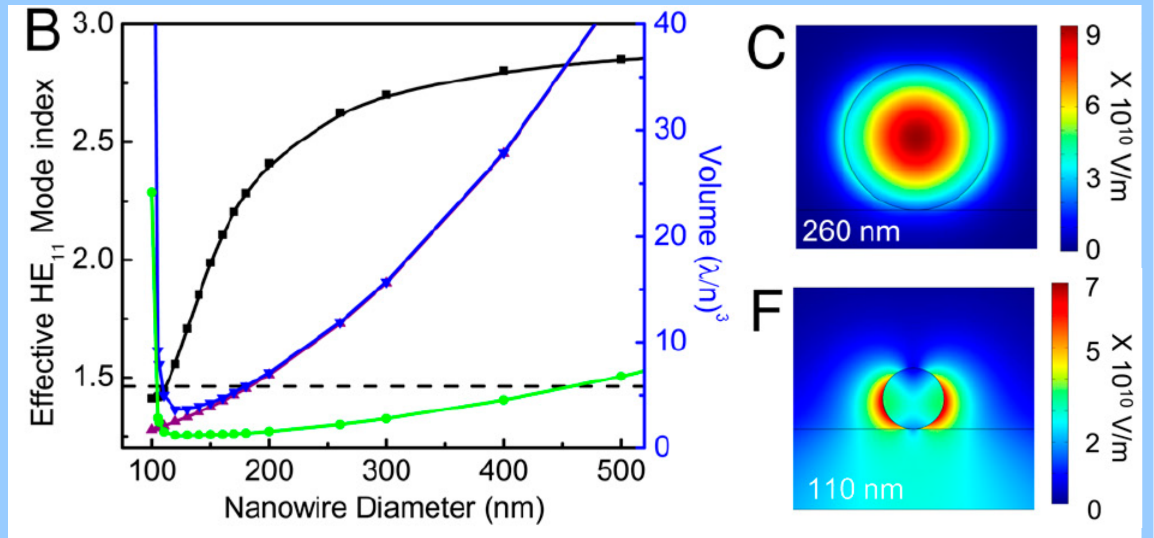
Mode confinement

SPP nanowire



$$\frac{\Gamma_{spp}}{\Gamma_0} = \frac{3}{8 n_{eff}} \left(\frac{\lambda}{n}\right)^2 \frac{\rho^{2D}(k_{spp})}{L_{spp}}$$

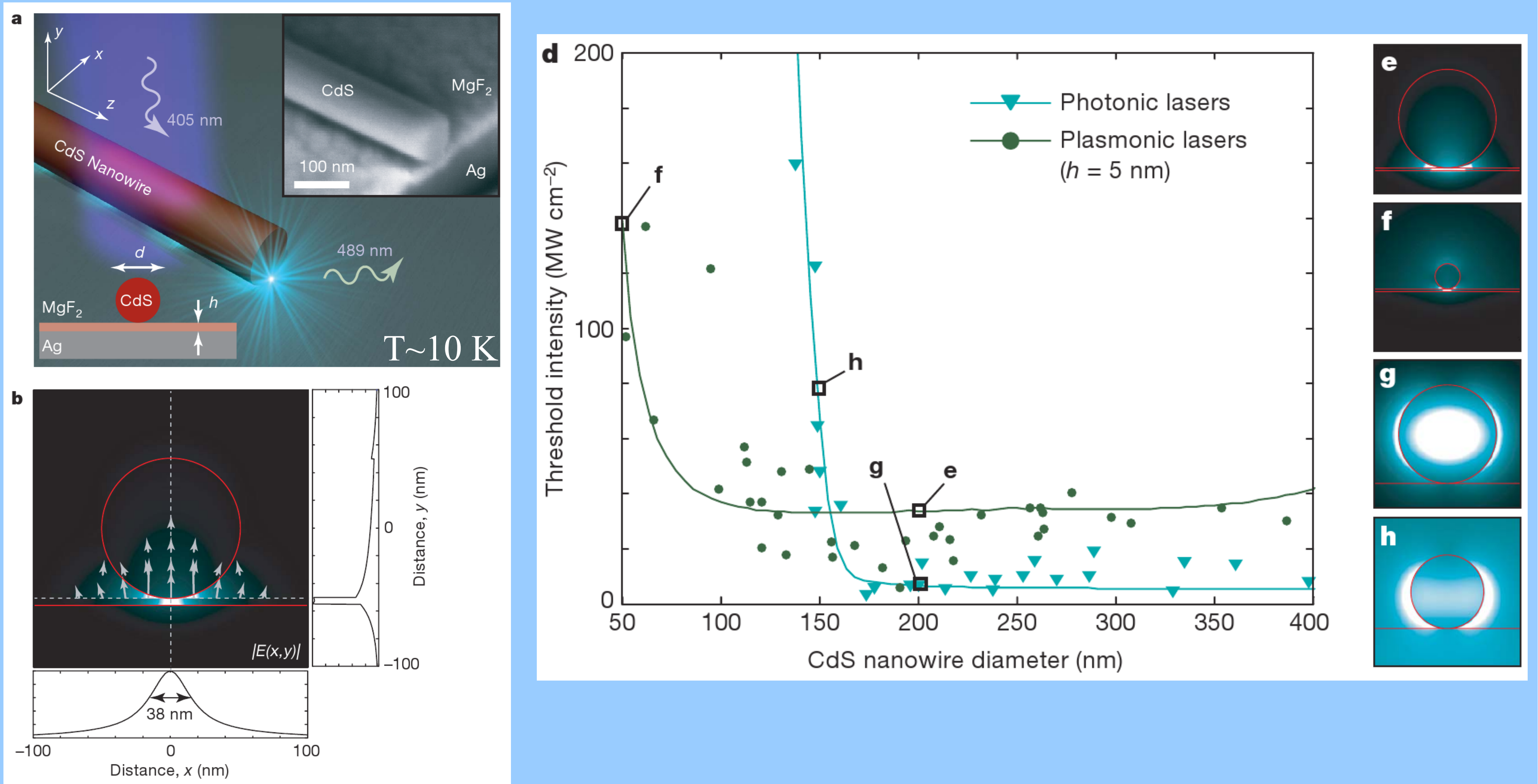
Photonic waveguide



Agarwal *et al*, PNAS **108**, 10050 (2011)

$$\frac{\Gamma_{guided}}{n_1 \Gamma_0} = \frac{3 n_g}{4\pi n_1} \frac{(\lambda / n_1)^2}{A_{eff}}$$

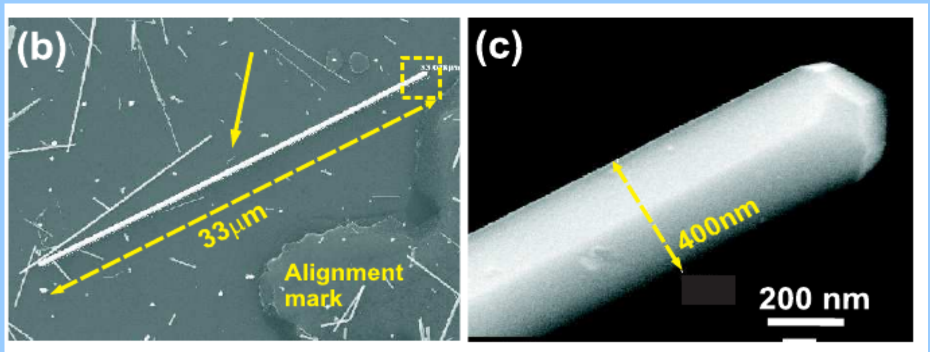
Plasmonic laser

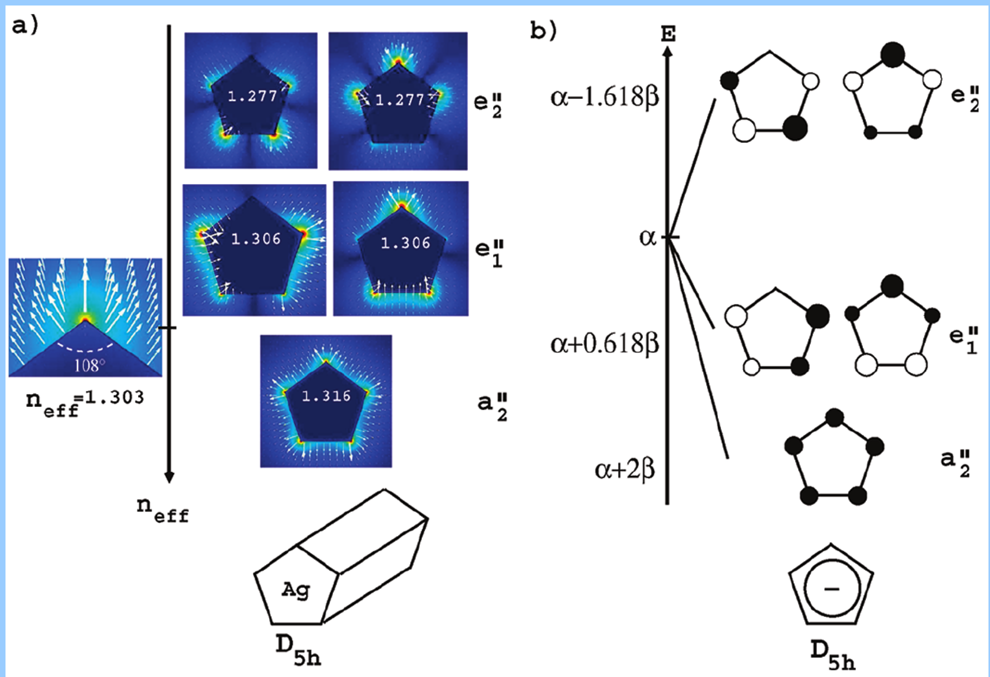


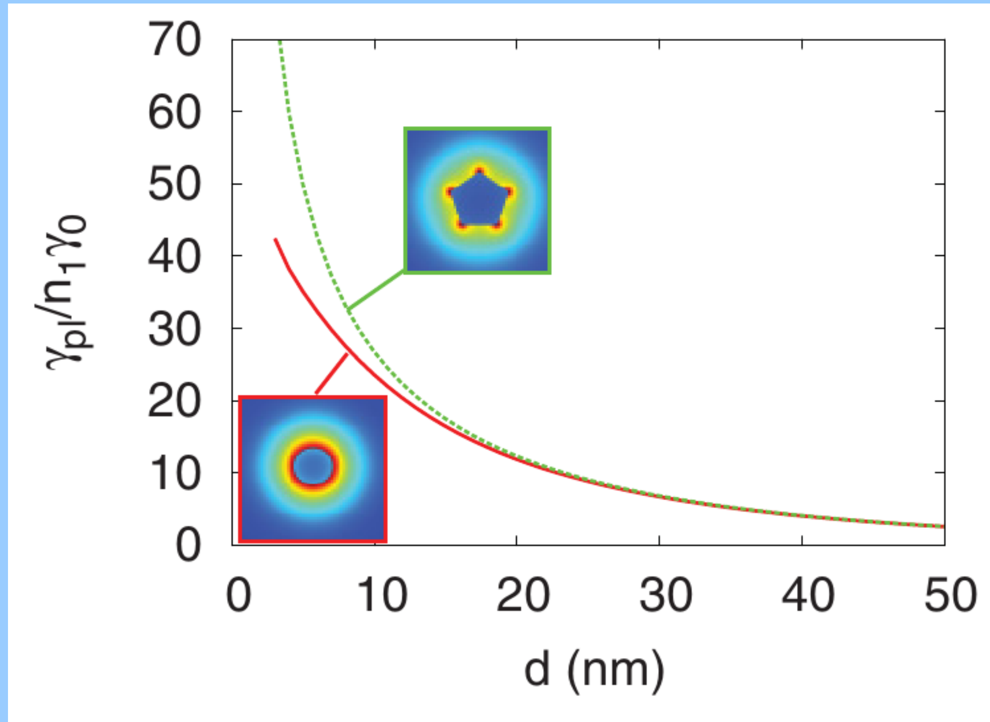
Plasmon laser at deep subwavelength scale

Oulton *et al,* Nature **461**(2009) 629 ,

Crystalline Ag wire







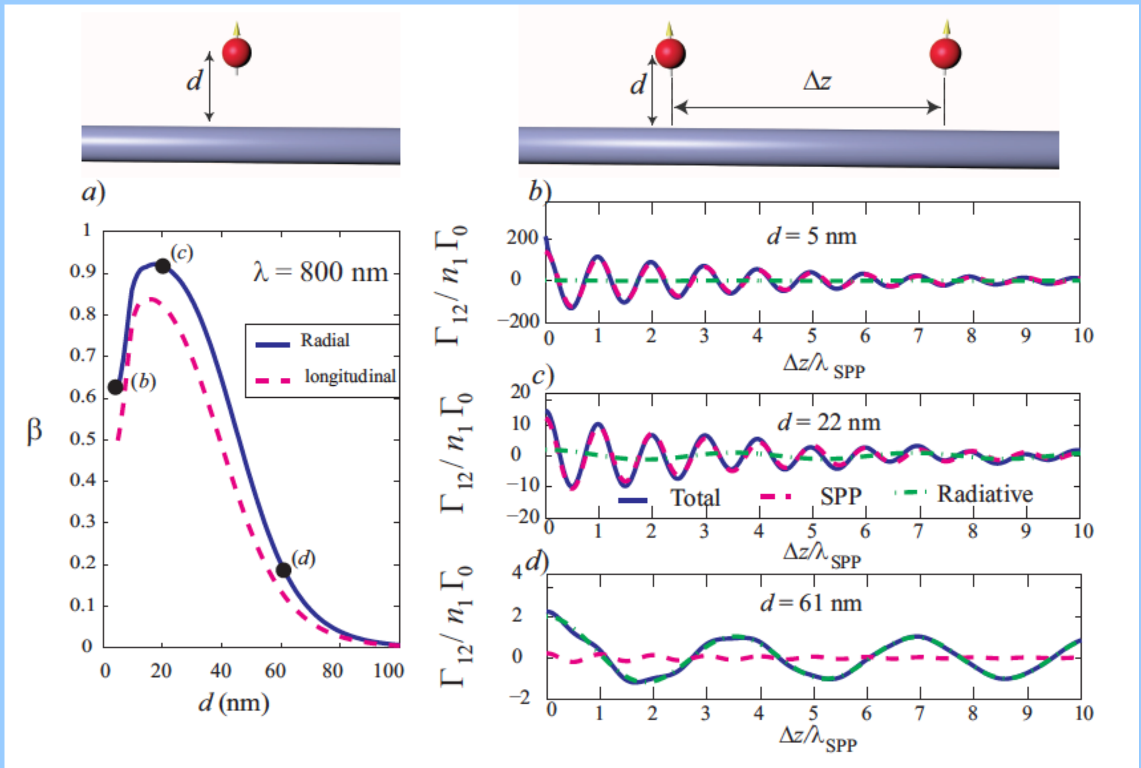
High modal confinement in the corners
=> high Purcell factor

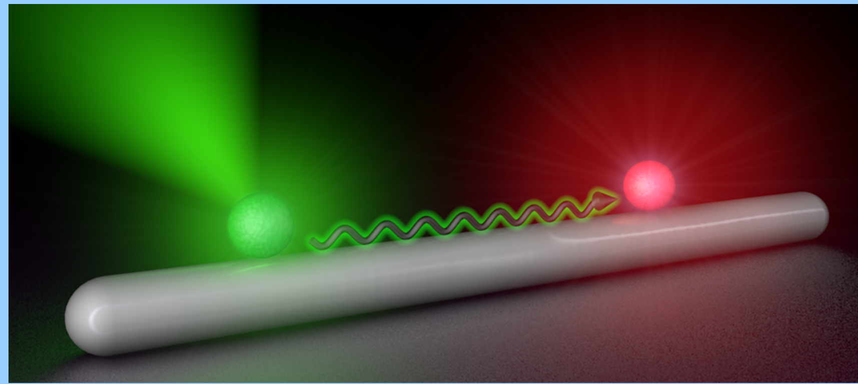
Lower losses (crystalline/amorphous metal)
=> high β factor

Imaging plasmonmodes in penta-twinned crystalline Ag nanowires
Song *et al*, ACS Nano **5**, 5874 (2011)

Purcell factor for dipolar emitter coupling to 2D plasmonic waveguides, Barthes *et al*, Physical Review B **84** 073403 (2011)

Resonant energy transfer

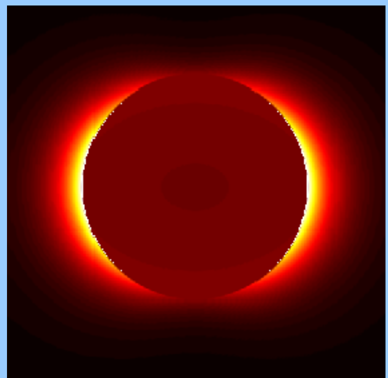


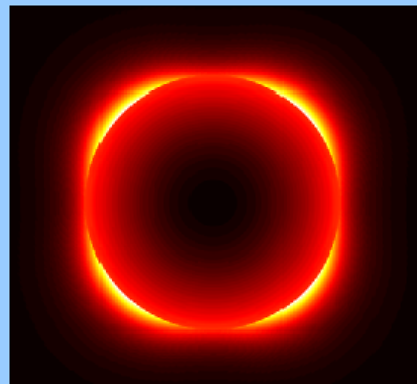


de Torres, Wenger *et al*,
ACS Phot. 10, 3968 (2016)

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

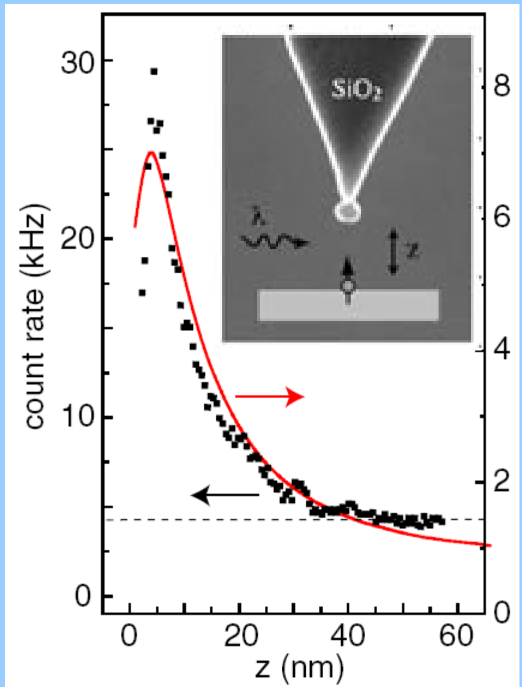
Localized plasmon (3D)-Purcell factor



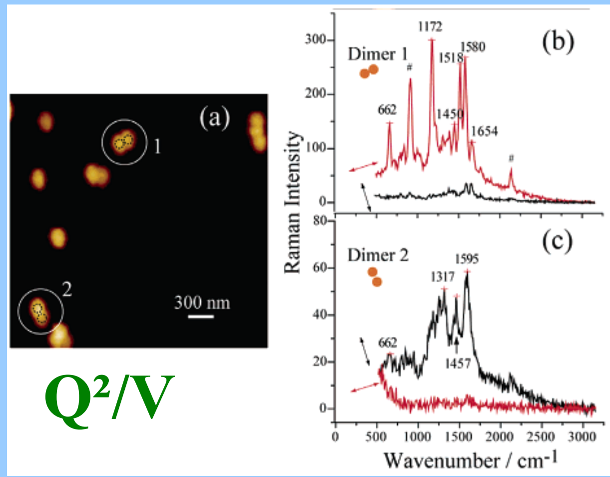


Surface enhanced spectroscopies

Surface enhanced spectroscopie
Raman (SERS), Fluorescence



P. Anger, L. Novotny
*PRL **96**, 113002 (2006)*

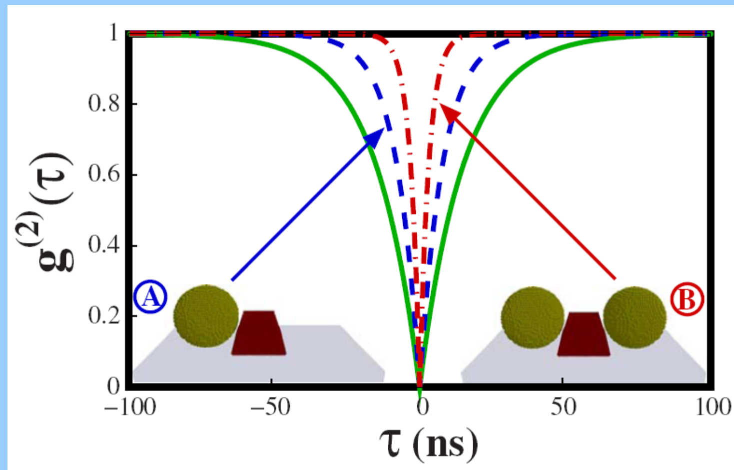


Imura et. al.
*Nano Lett., **6** 2006)*

Maier
Opt. Exp 4, 1957 (2006)

Coupling
Q/V

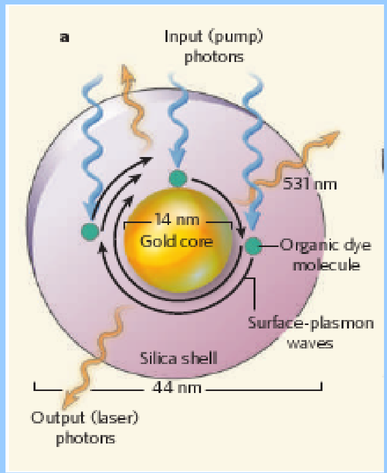
Control of photons antibunching:

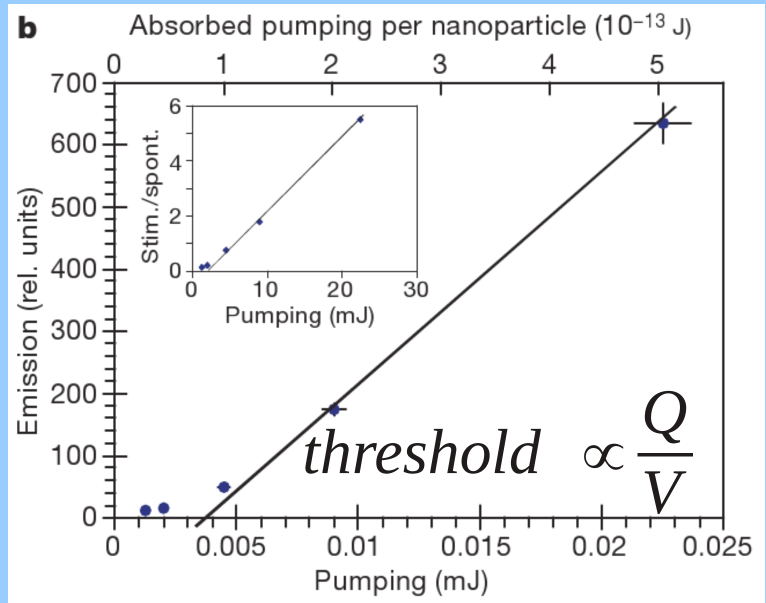


S. Schietinger, O. Benson
*Nano Lett. **9** , 1694 (2009)*

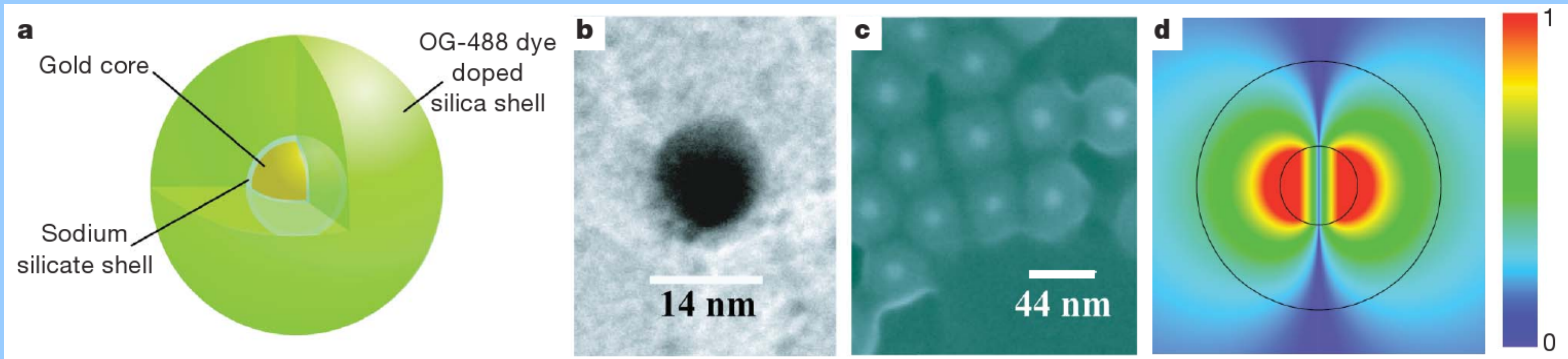
Marty,Girard, Colas des Francs
*PRB **82**, 081403R(2010)*

Perspectives : plasmon nanolaser (SPASER)



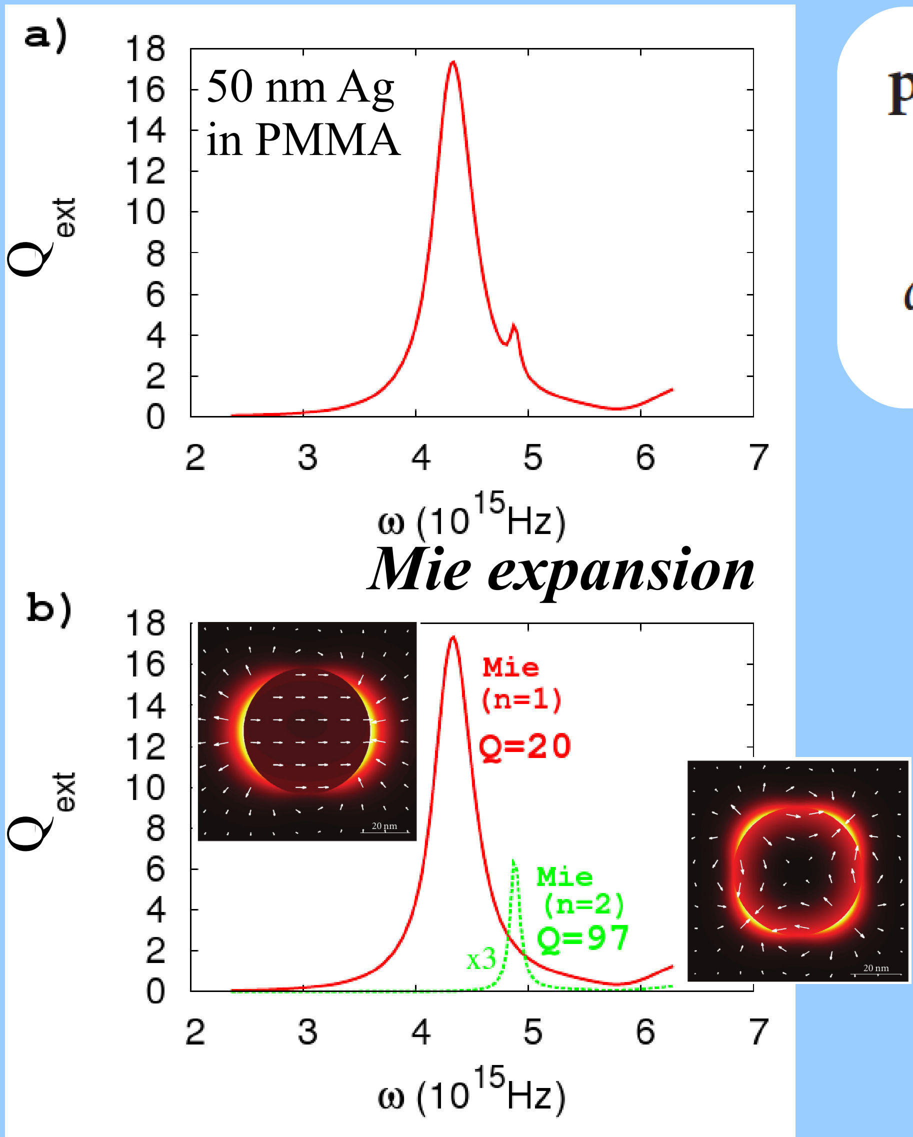


localized,
coherent
and ultra-fast
nanosource



Stockman, Nat. Phys. **2**, 327 (2008)
Noginov *et al*, Nature **460**, 1110 (2009)

Quality factor of localized SPP



$$\mathbf{p}^{(1)}(\omega) = 4\pi\epsilon_0\alpha_1(\omega)\mathbf{E}_0$$

$$\alpha_1(\omega) = \frac{\epsilon_m(\omega) - 1}{\epsilon_m(\omega) + 2}R^3$$

Quasi-static approx

$$\varepsilon_m = 1 - \frac{\omega_p^2}{\omega^2 + i\kappa_{abs}\omega}$$

$$\alpha_1^{eff}(\omega) \underset{\omega_1}{\sim} \frac{\omega_1}{2(\omega_1 - \omega) - i\kappa_1}R^3$$

$$\kappa_1 = \kappa_{abs} + \frac{2(k_1R)^3\omega_1}{3}$$

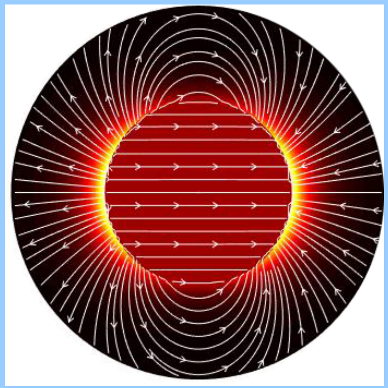
Joule losses *Radiative leakage*

Quality factor

$$Q_1 = \frac{\omega_1}{\mathrm{K}_1}$$

Mie plasmons: quality factors, effective volumes and coupling strength to a dipolar emitter
Colas des Francs, Derom, Vincent, Bouhelier, Dereux, Int. J. Optics **2012**, 175162 (2012)

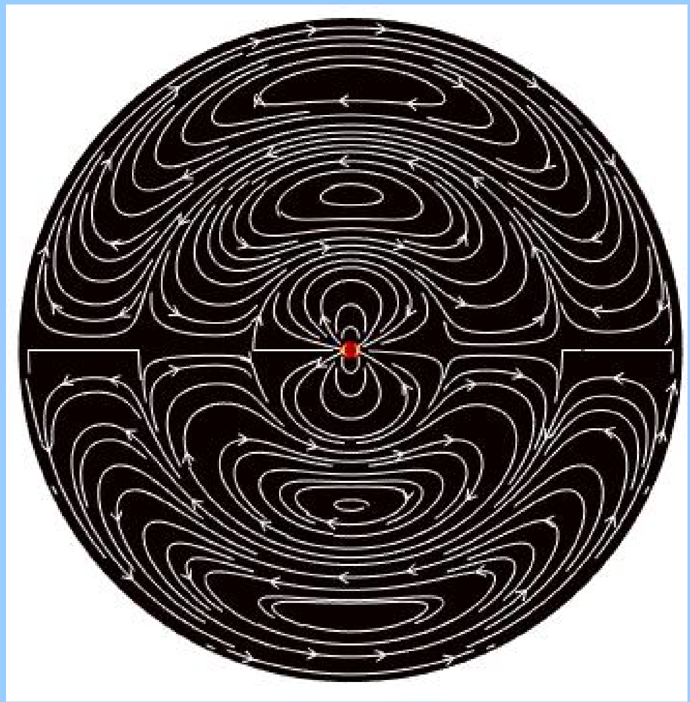
Mode volume definition

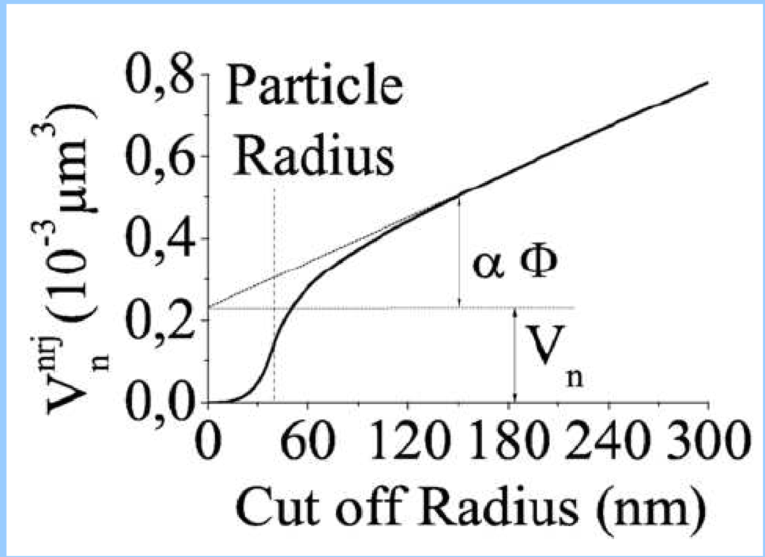


Near field confinement

$$V_n^{nrj} = \frac{\int U_n(\mathbf{r}) d\mathbf{r}}{max[\varepsilon_0 \varepsilon_1 \, |\mathbf{E_n}(\mathbf{r})|^2]} \, ,$$

$$U_n(\mathbf{r}) = \frac{\partial[\omega \varepsilon_0 \varepsilon(\mathbf{r}, \omega)]}{\partial \omega} \, |\mathbf{E_n}(\mathbf{r})|^2 + \mu_0 |\mathbf{H_n}(\mathbf{r})|^2$$





Far field scattering

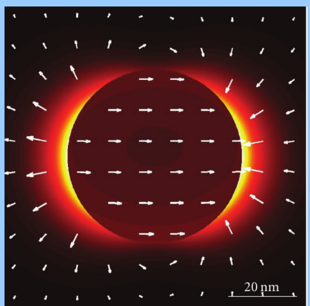
Mode volume - quasi-static approximation

Dipolar LSP response

$$\frac{\gamma_1^\perp}{\gamma_0} \sim \frac{6}{k^3 z_0^6}\,\mathrm{Im}(\alpha_1)$$

$$\frac{\langle\Gamma_1\rangle}{\Gamma_0} = \frac{\Gamma_1^\perp + 2\Gamma_1^\parallel}{3\Gamma_0} \sim \frac{3}{4\pi^2}\lambda_1^3\frac{1}{2\pi R^3}Q_1$$

$$= \frac{3}{4\pi^2}\lambda_1^3\frac{Q_1}{V_1}$$



$$V_1 = 1.5 V_{part}$$

$$V_{part} = \frac{4}{3}\pi R^3$$

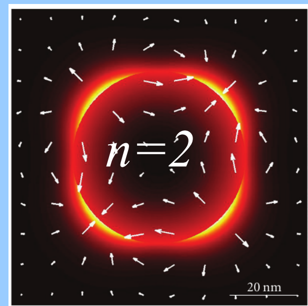
n^{th} LSP response

$$\mathbf{p}^{(n)} = \frac{4\pi\epsilon_0}{(2n-1)!!}\alpha_n \nabla^{n-1}\mathbf{E}_0$$

$$\alpha_n = \frac{n(\epsilon_m - 1)}{n\epsilon_m + (n+1)}R^{(2n+1)}$$

$$\frac{\Gamma_n}{\Gamma_0} = (2n+1)F_p$$

degenerescence



$$V_n = \frac{3}{n+1}V_{part}$$

Mie plasmons: quality factors, effective volumes and coupling strength to a dipolar emitter

Colas des Francs, Derom, Vincent, Bouhelier, Dereux, Int. J. Optics **2012**, 175162 (2012)

Mode confinement – cQED *extrapolation*

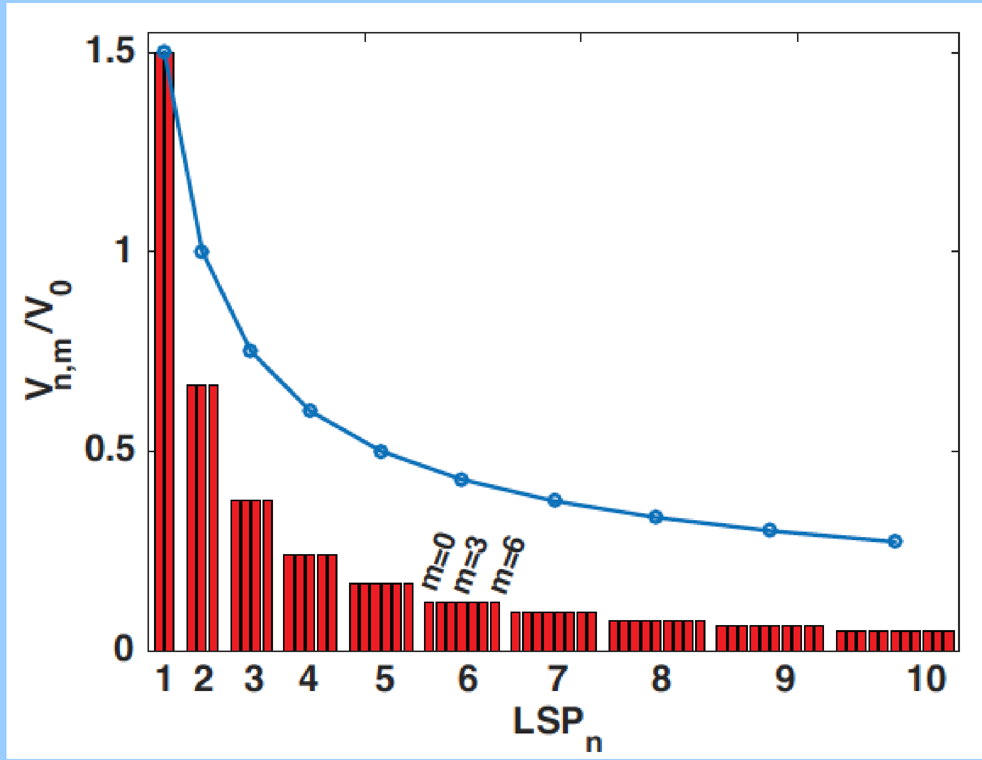
$$V_n^{nrj} = \frac{\int U_n(\mathbf{r}) d\mathbf{r}}{max[\varepsilon_0 \varepsilon_1 \ |\mathbf{E_n}(\mathbf{r})|^2]} \ ,$$

$$U_n(\mathbf{r}) = \frac{\partial[\omega \varepsilon_0 \varepsilon(\mathbf{r}, \omega)]}{\partial \omega} \ |\mathbf{E_n}(\mathbf{r})|^2 + \mu_0 |\mathbf{H_n}(\mathbf{r})|^2$$

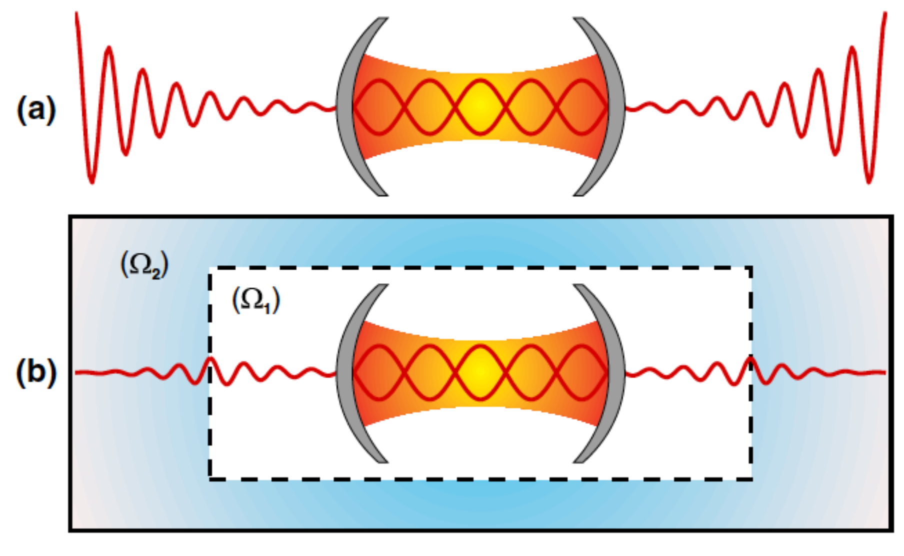
Quasi-static approx

$$V_n^{nrj} = \frac{6}{(n+1)^2} V_{part}$$

Khurgin and Sun
*JOSA B **26** , B83 (2009)*



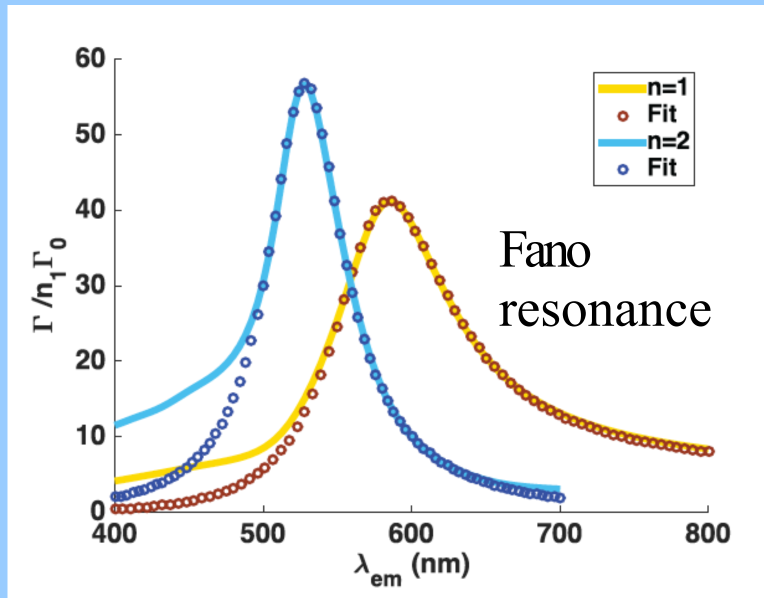
Complex mode volume – the reconcilation



Sauvan, Lalanne
*PRL **110** , 237401 (2013)*

$$V_n = \frac{\frac{1}{2}\int \tilde{\mathbf{E}}_\mathbf{n} \cdot \frac{\partial(\omega\varepsilon_0\varepsilon)}{\partial\omega} \cdot \tilde{\mathbf{E}}_\mathbf{n} - \mu_0 \tilde{\mathbf{H}}_\mathbf{n}^2 \mathbf{dr}}{max[\varepsilon_0\varepsilon_1\ [\tilde{\mathbf{E}}_\mathbf{n}(\mathbf{r})]^2]}$$

$$F_p = \frac{\Gamma_n}{n_1\ \Gamma_0} = \frac{3}{4\pi^2}\left(\frac{\lambda}{n_1}\right)^3\ Re\left(\frac{Q_n}{V_n}\right)$$

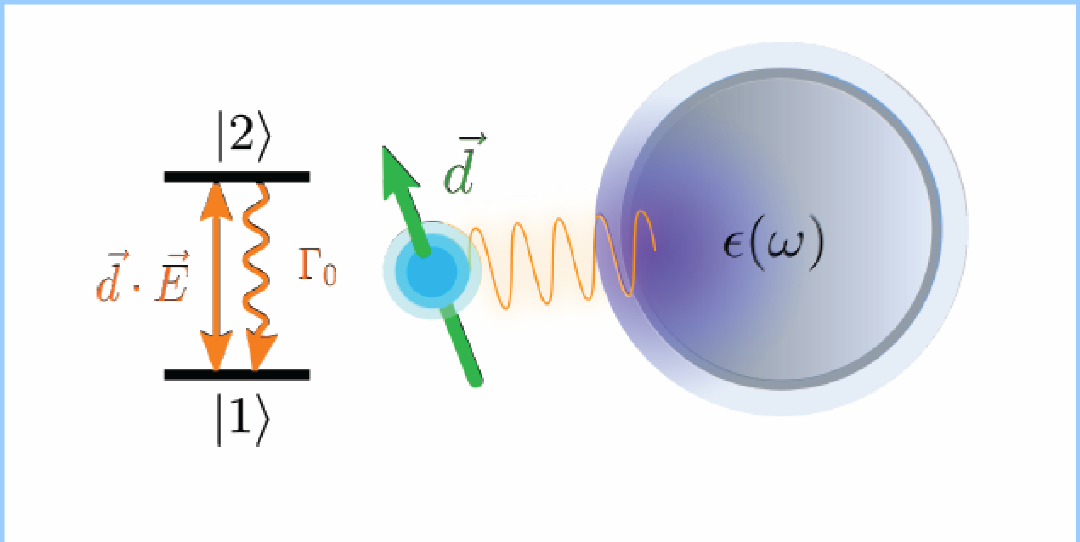


F_p 35
$Q \sim 10$
$Re(V) \sim 0.01 \left(\frac{\lambda}{n}\right)^3$
$Im(V) \sim 0.001 \left(\frac{\lambda}{n}\right)^3$

$$\frac{\langle\Gamma_n\rangle}{n_1\Gamma_0} = \frac{(2n+1)}{3} F_p \frac{(\omega_n/\omega_{em})^2}{1 + 4\ Q^2\delta\tilde{\omega}^2}\left[1 + 2Q\delta\tilde{\omega}\frac{Im(V_n)}{Re(V_n)}\right]$$

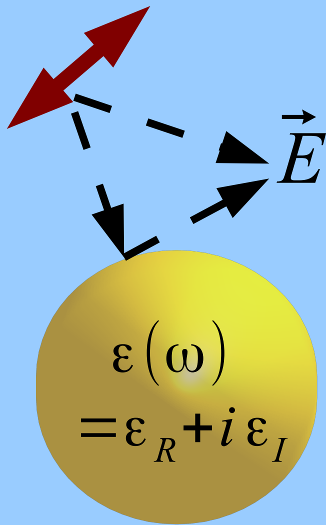
$$\delta\tilde{\omega} = \frac{\omega_{em} - \omega_n}{\omega_{em}}$$

Quantum plasmonics
cQED description



Towards plasmon quantization

Classical description – dipolar scattering



$$\vec{E}(\vec{r})=\frac{k_0^2}{\varepsilon_0}\bar{G}(\vec{r},\vec{r}_0,\omega)\cdot\vec{p}$$

Quantization in an absorbing/dissipative medium – electric-field operator

$$\hat{E}(\vec{r})=ik_0^2\sqrt{\frac{\hbar}{\pi\varepsilon_0}}\int d\vec{r}\,'\sqrt{\varepsilon_I}\,\bar{G}(\vec{r},\vec{r}\,',\omega)\hat{f}_\omega(\vec{r}\,')$$

Gruner, Welsch, PRA ***53****, 3 (1996)*

Plasmon resonances

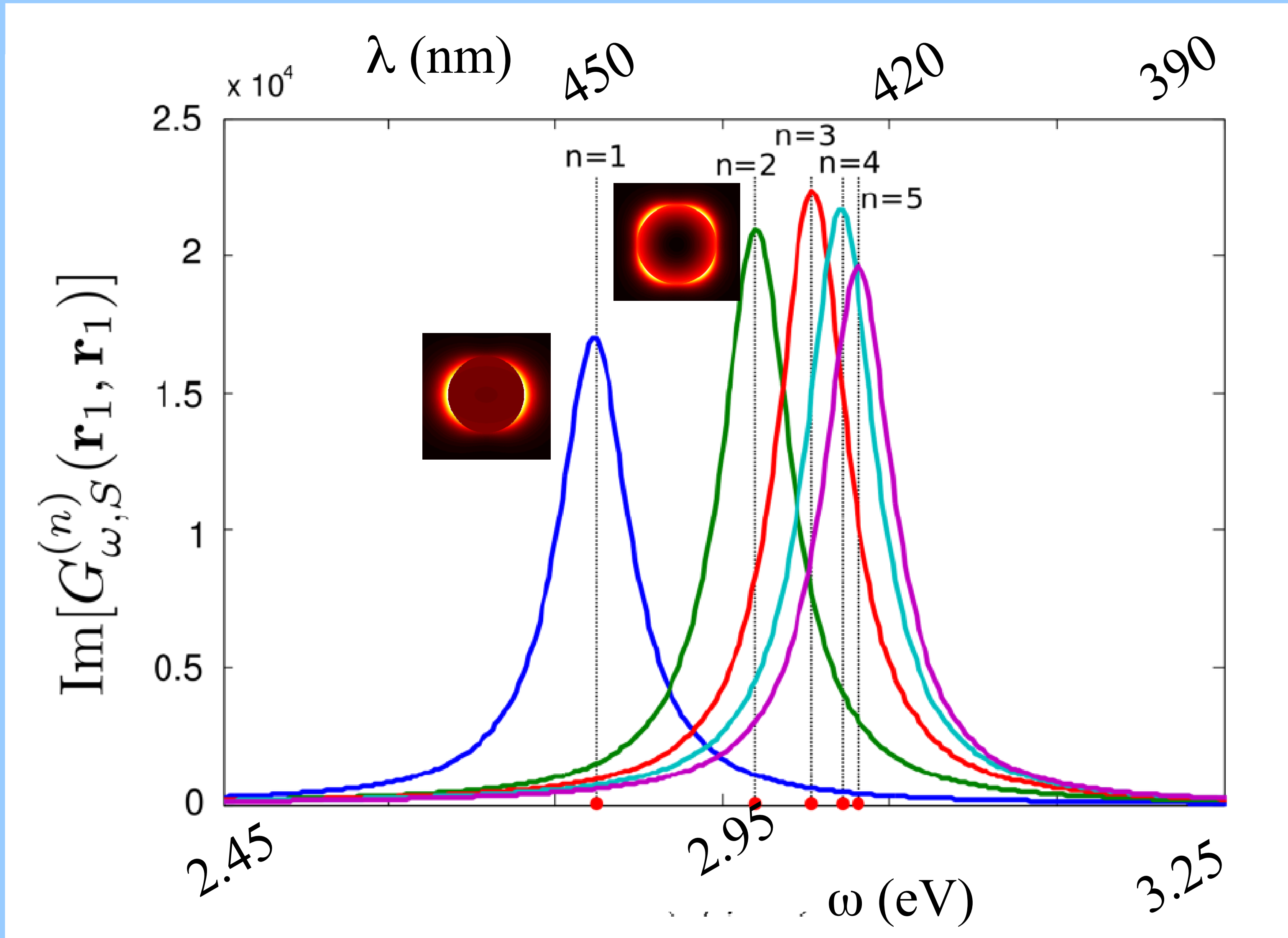
Modal expansion (Mie formalism)

$$G = G_0 + G_S$$

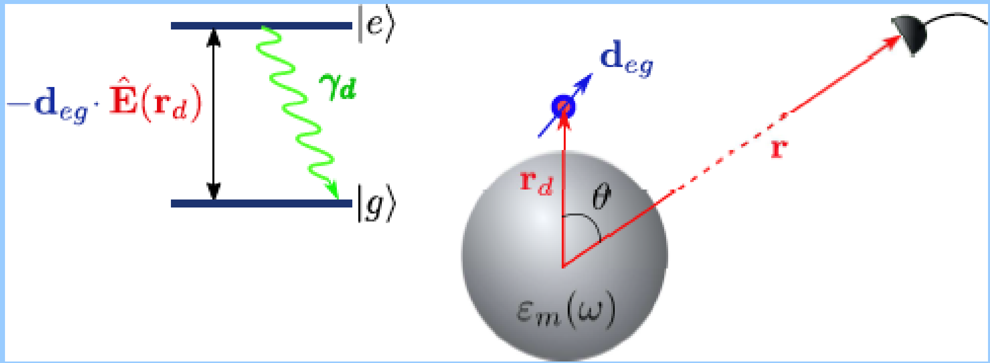
$$G_S = \sum G_S^{(n)}$$

Density of modes

$$LDOS \propto Im\, G$$



Quantum plasmonics



$$|\psi(t)\rangle = C_e(t)e^{-i\omega_e t}|e\rangle|\varnothing\rangle + \int d\mathbf{r}\int_0^\infty d\omega\, e^{-i(\omega+\omega_g)t}\mathbf{C}_g(\mathbf{r},\omega,t)\cdot|g\rangle|\mathbf{1}(\mathbf{r},\omega)\rangle$$

$$\hat{H}_{QD} = \sum_{i=1}^{2}\hbar\omega_{i1}\hat{\sigma}_{ii} - i\hbar\frac{\gamma_0}{2}\hat{\sigma}_{22}$$

Quantum plasmonics

cQED

$$\hat{H}_R = \int d\mathbf{r}\int_0^{+\infty} d\omega\ \hbar\omega\ \hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega)$$

$$\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)|\varnothing\rangle = |\mathbf{1}(\mathbf{r},\omega)\rangle \Longleftrightarrow \hat{a}^\dagger|\varnothing\rangle = |1\rangle$$

$$\hat{H}_I = \left[\hat{\sigma}_{21}\int_0^{+\infty} d\omega\mathbf{d}_{21}\ \hat{\mathbf{E}}(\mathbf{r}_d,\omega) + H.c.\right]$$

$$\hat{\mathbf{E}}(\mathbf{r},\omega) = i\sqrt{\frac{\hbar}{\pi\epsilon_0}\frac{\omega^2}{c^2}}\int d\mathbf{r}'\sqrt{\epsilon_I(\mathbf{r}',\omega)}\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\hat{\mathbf{f}}(\mathbf{r}',\omega)$$

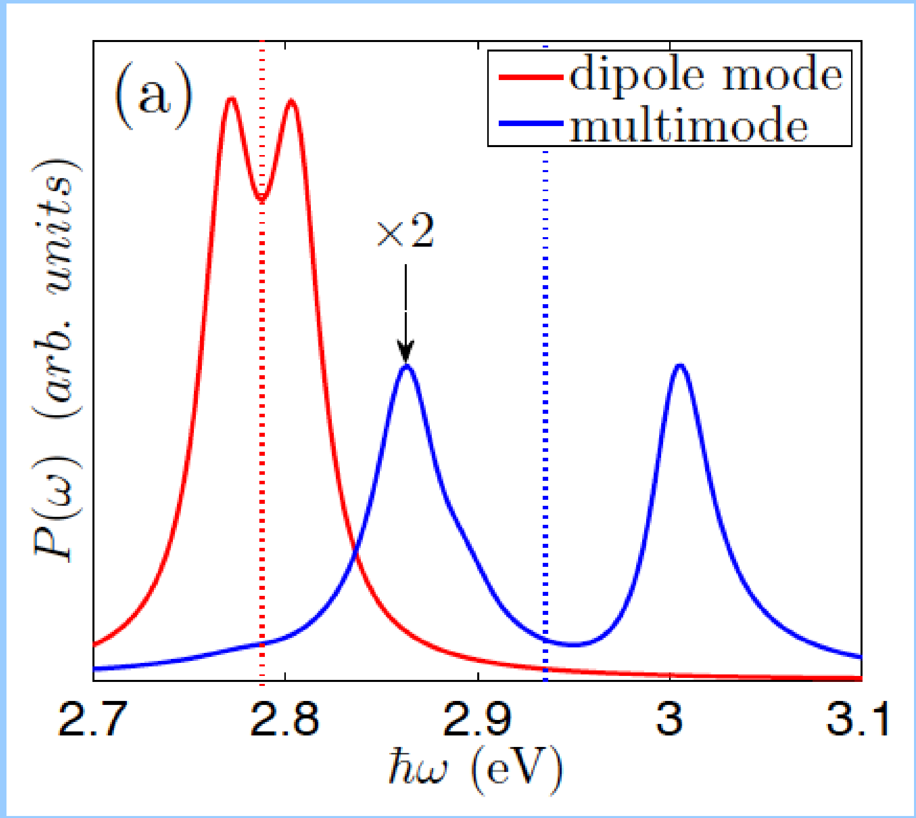
Mode-selective quantization and easy-to-handle effective cQED models with plasmonic resonance
D. Dzsotjan *et al, submitted* (2016)

Strong coupling

Polarization (near field) spectrum

$$P(\omega) = \langle \hat{\sigma}_{ge}^{\dagger}(\omega)\hat{\sigma}_{ge}(\omega)\rangle$$

$$P(\omega) = \left| \frac{1}{\omega_{eg} - \omega - i\frac{\gamma_d}{2} - \frac{k_0^2}{\hbar\epsilon_0} d_{eg}^2 G_{uu}^{scatt}(\mathbf{r}_d, \mathbf{r}_d, \omega)} \right|^2$$



Effective model – modal analysis

$$\hat{H}_I = \left[\hat{\sigma}_{21}\int_0^{+\infty} d\omega \mathbf{d}_{21}\cdot\hat{\mathbf{E}}(\mathbf{r}_d,\omega) + H.c.\right]$$

$$\hat{\mathbf{E}}(\mathbf{r},\omega) = i\sqrt{\frac{\hbar}{\pi\epsilon_0}\frac{\omega^2}{c^2}}\int d\mathbf{r}'\sqrt{\epsilon_I(\mathbf{r}',\omega)}\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\hat{\mathbf{f}}(\mathbf{r}',\omega)$$

$$\Rightarrow \quad \hat{H}_I = i\hbar\int_0^{+\infty} d\omega \sum_{n=1}^{N}\left(\kappa_n^*(\omega)\hat{b}^\dagger_{\omega,n}\hat{\sigma}_{12} - \kappa_n(\omega)\hat{b}_{\omega,n}\hat{\sigma}_{21}\right)$$

Structure of the coupling for each plasmon mode

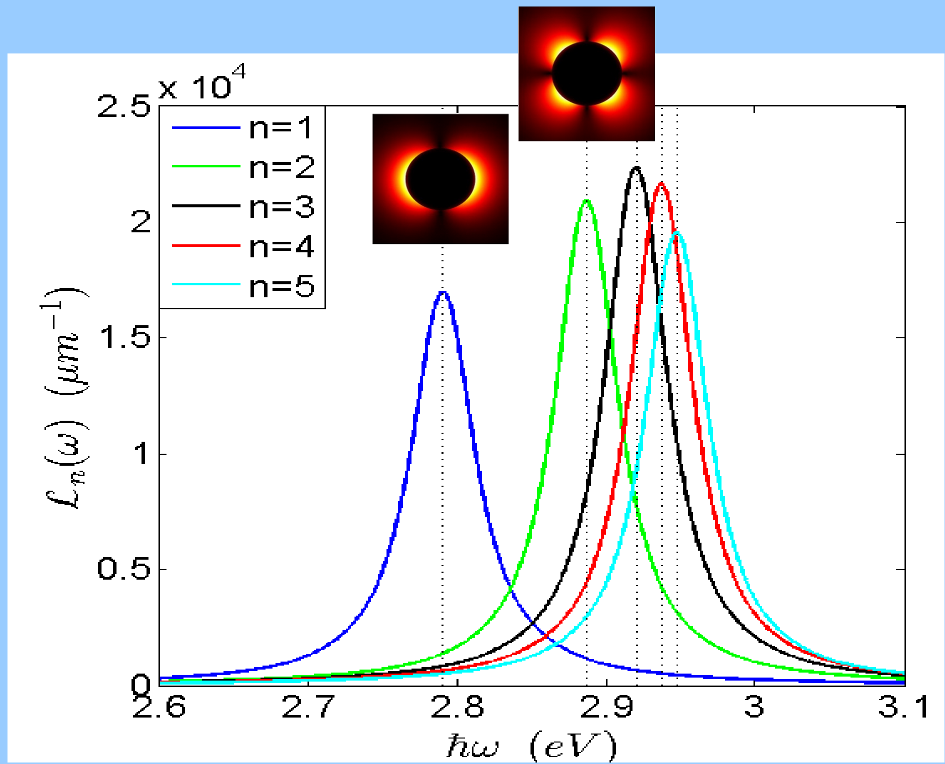
Structure of the coupling

Connection between effective model and Green's tensor :

$$\sum_{n=1}^{N} |\kappa_n(\omega)|^2 = \frac{1}{\hbar\pi\epsilon_0}\frac{\omega^2}{c^2}\mathbf{d}_{21}\cdot\left(Im[\mathbf{G}(\mathbf{r}_d,\mathbf{r}_d,\omega)]\mathbf{d}_{21}^*\right)$$

Lorentzian fitting of the plasmon modes

$$\kappa_n(\omega) = \sqrt{\frac{\gamma_n}{2\pi}}\frac{g_n}{\omega-\omega_n+i\frac{\gamma_n}{2}}$$



Mode-selective quantization and easy-to-handle effective cQED models with plasmonic resonance
D. Dzsotjan *et al, submitted* (2016)

Quantum plasmonics – Effective hamiltonian

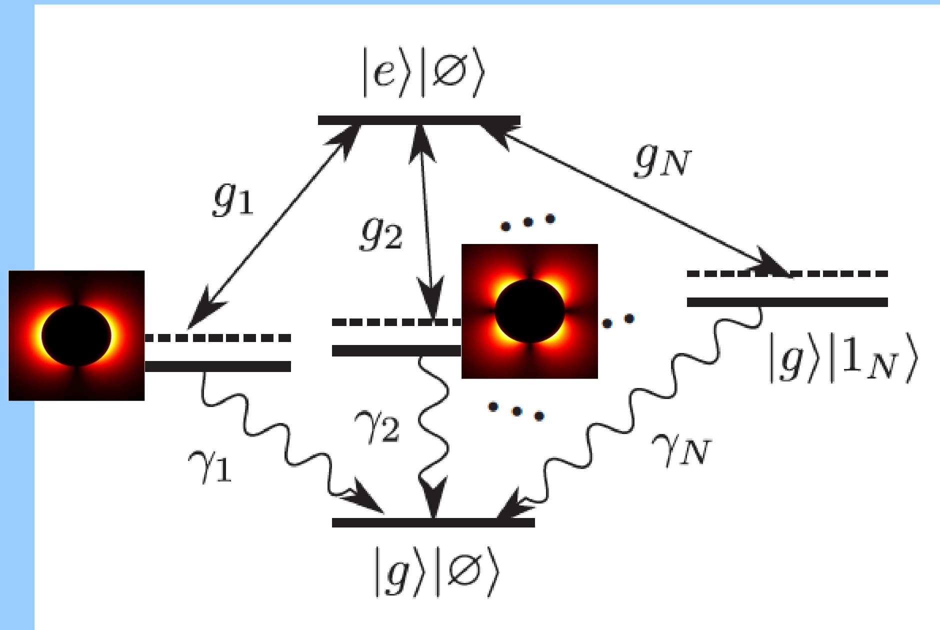
$$\hat{H}_I = i\hbar \int_0^{+\infty} d\omega \sum_{n=1}^{N} \left(\kappa_n^*(\omega)\hat{b}^\dagger_{\omega,n}\hat{\sigma}_{12} - \kappa_n(\omega)\hat{b}_{\omega,n}\hat{\sigma}_{21}\right)$$

$$\kappa_n(\omega) = \sqrt{\frac{\gamma_n}{2\pi}}\frac{g_n}{\omega - \omega_n + i\frac{\gamma_n}{2}}$$

Integral

$$H_{eff} = \hbar \begin{bmatrix} -i\frac{\gamma_0}{2} & ig_1 & ig_2 & \cdots & ig_N \\ -ig_1 & \Delta_1 - i\frac{\gamma_1}{2} & 0 & \cdots & 0 \\ -ig_2 & 0 & \Delta_2 - i\frac{\gamma_2}{2} & \ddots & \vdots \\ \vdots & \vdots & \ddots & \ddots & 0 \\ -ig_N & 0 & \cdots & 0 & \Delta_N - i\frac{\gamma_N}{2} \end{bmatrix}$$

Dressed states

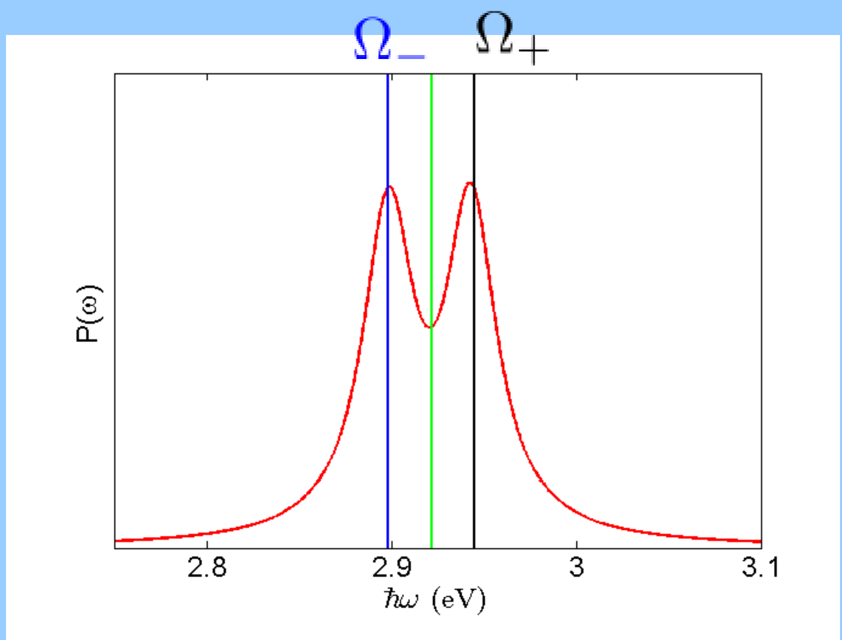


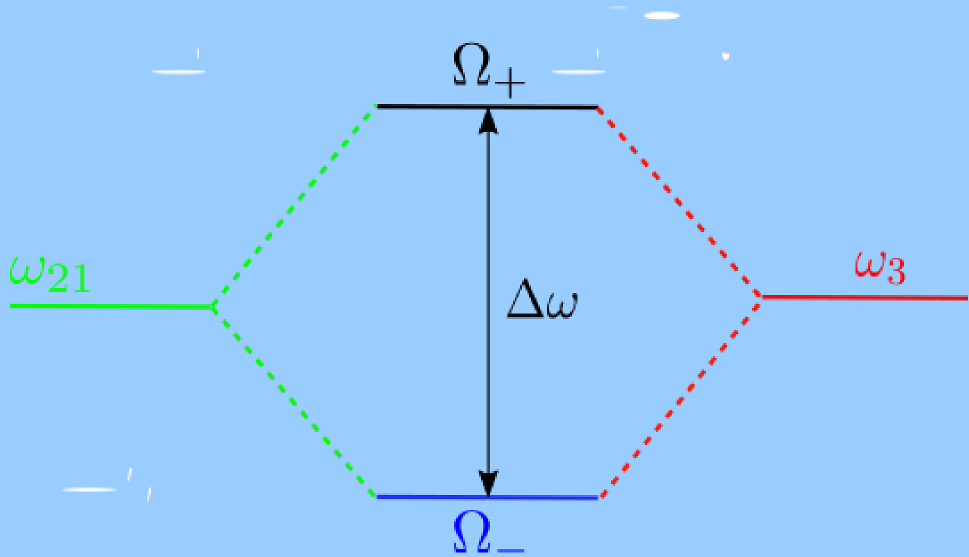
Adiabatic passage mediated by plasmons: a route towards a decoherence-free quantum plasmonic platform
B. Rousseaux *et al*, Phys. Rev. B 93, 045422 (2016)

Strong coupling regime

Monomode cavity (LSP_3)

$$H_{eff} = \hbar \begin{bmatrix} -i\frac{\gamma_0}{2} & ig_3 \\ -ig_3 & \Delta_3 - i\frac{\gamma_3}{2} \end{bmatrix}$$

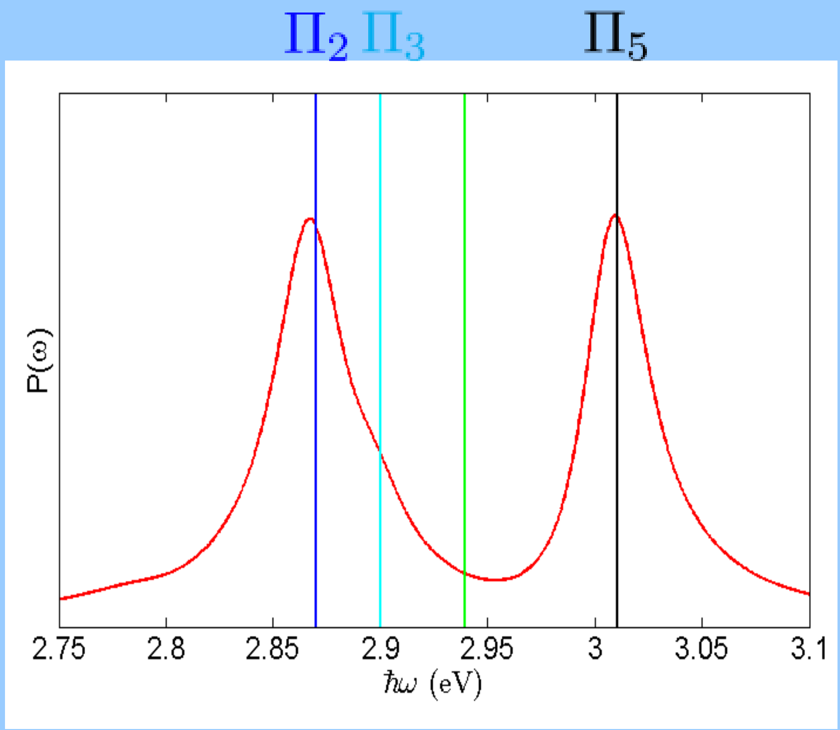


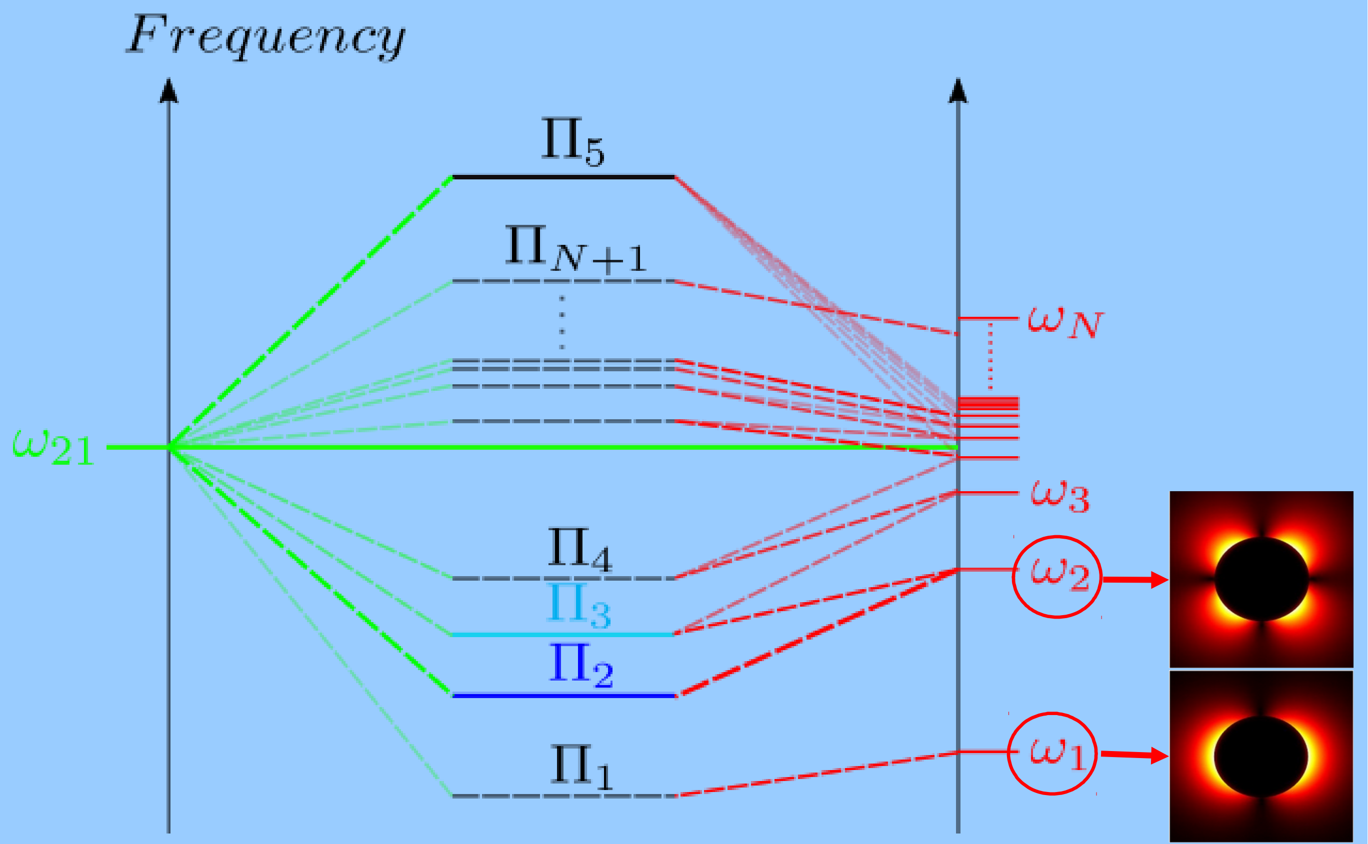


$$\Omega_{\pm} = \omega_{21} \pm \sqrt{g_3^2 - \left(\frac{\gamma_3 - \gamma_0}{4}\right)^2}$$

Strong coupling regime

Multimodal lossy cavity (LSPs)

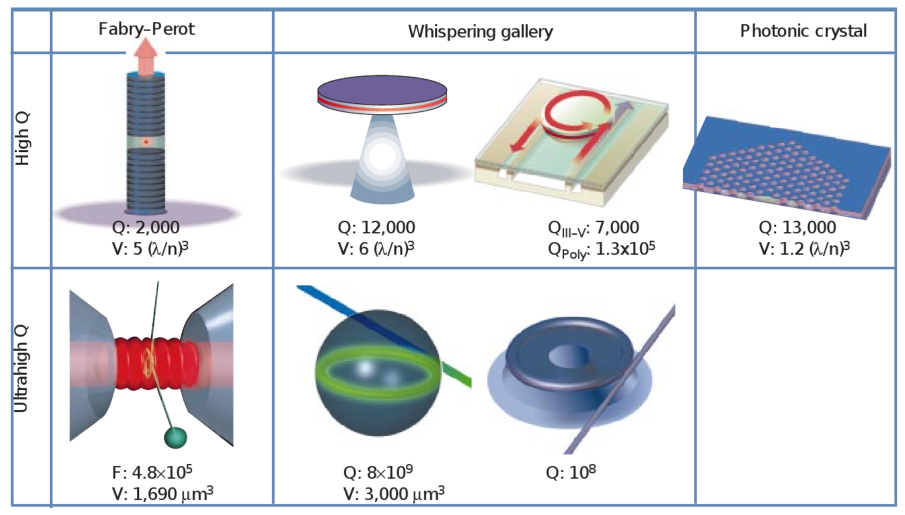




Dressed states of a quantum emitter strongly coupled to a metal nanoparticle
H. Varguet *et al, submitted* (2016)

Conclusion

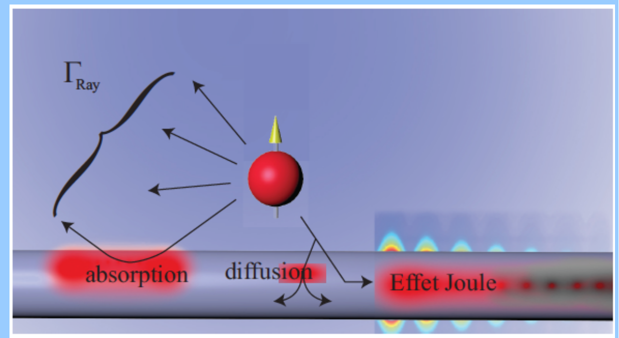
Cavity quantum electrodynamics (cQED)

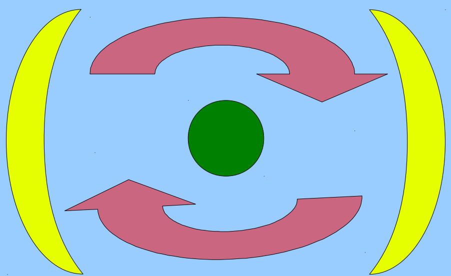


Purcell factor

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

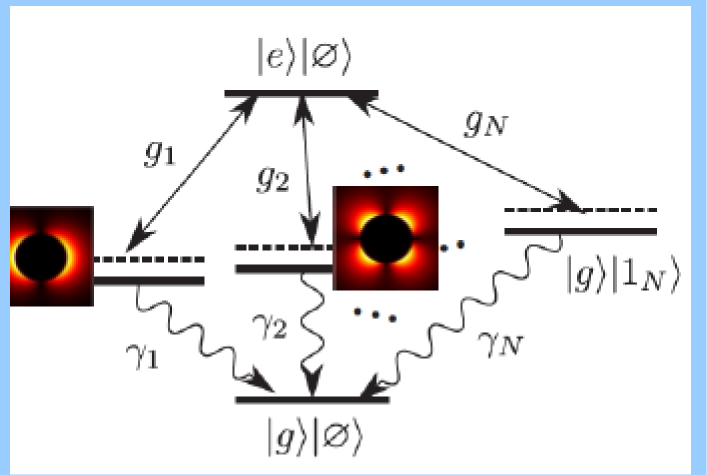
Quantum plasmonics (cavity*less QED)*





Duration of interaction (high Q)

Volume of interaction (sub-λ)



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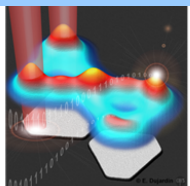
ICB
Quantum control

H. Jauslin
S. Guérin



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