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## Sonder les modes optiques avec des nanosources fluorescentes

Remi Carminati

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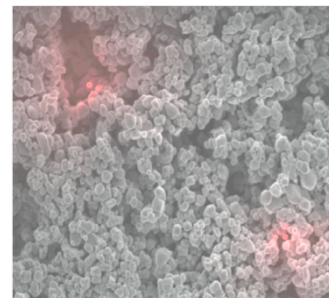
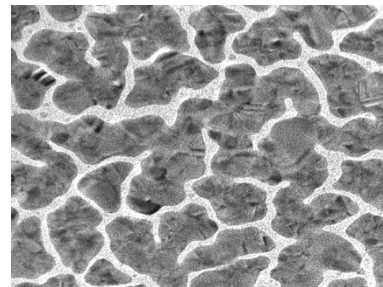
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# Sonder les modes optiques avec des nanosources fluorescentes

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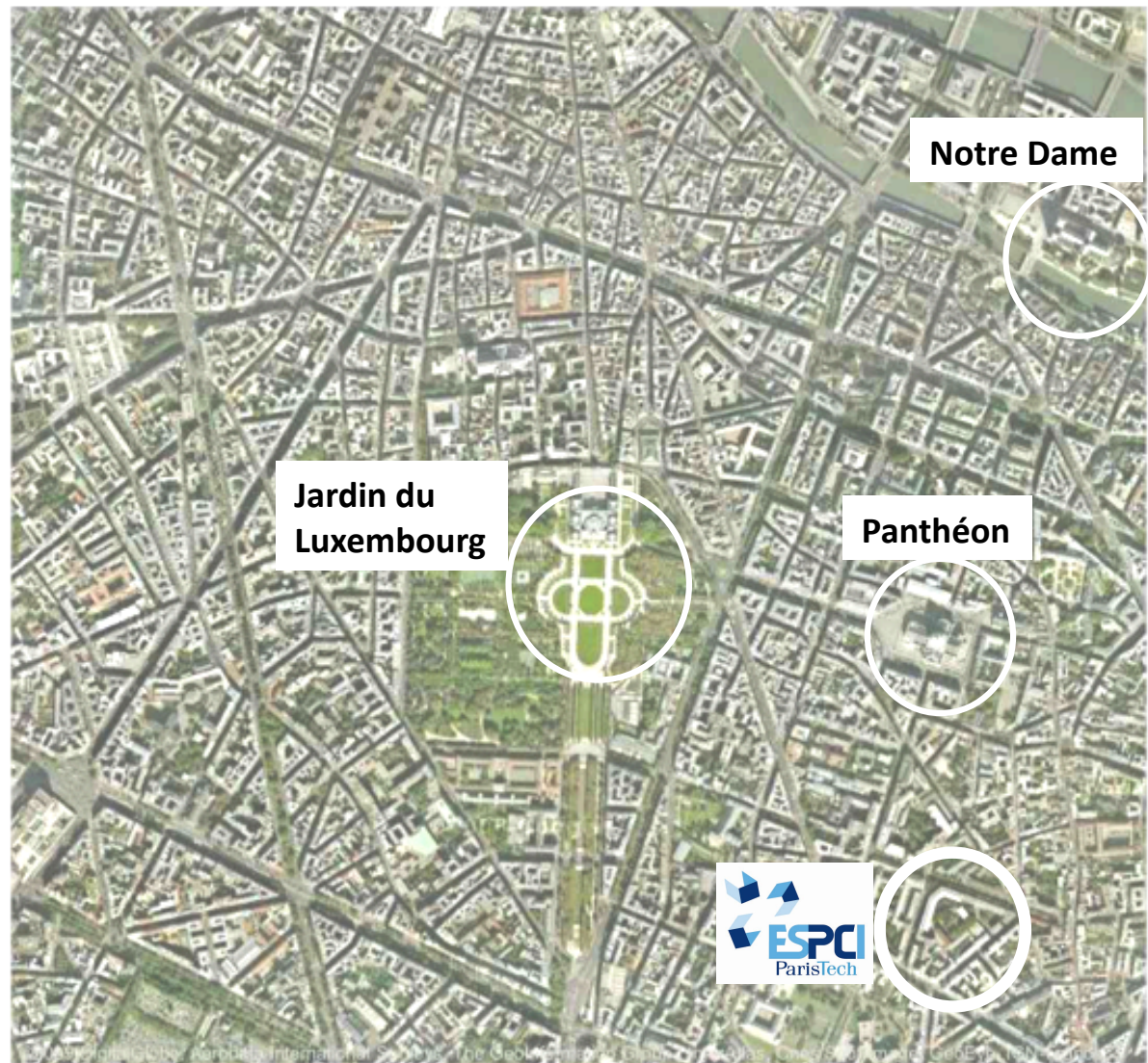
*remi.carminati@espci.fr*



# Institut Langevin @ ESPCI ParisTech



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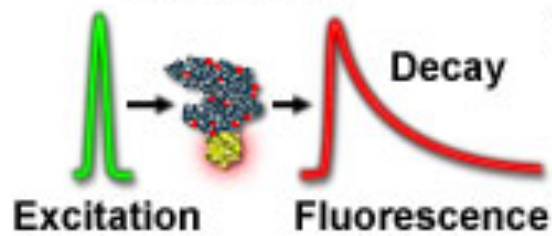


Spontaneous emission in nanostructured environments

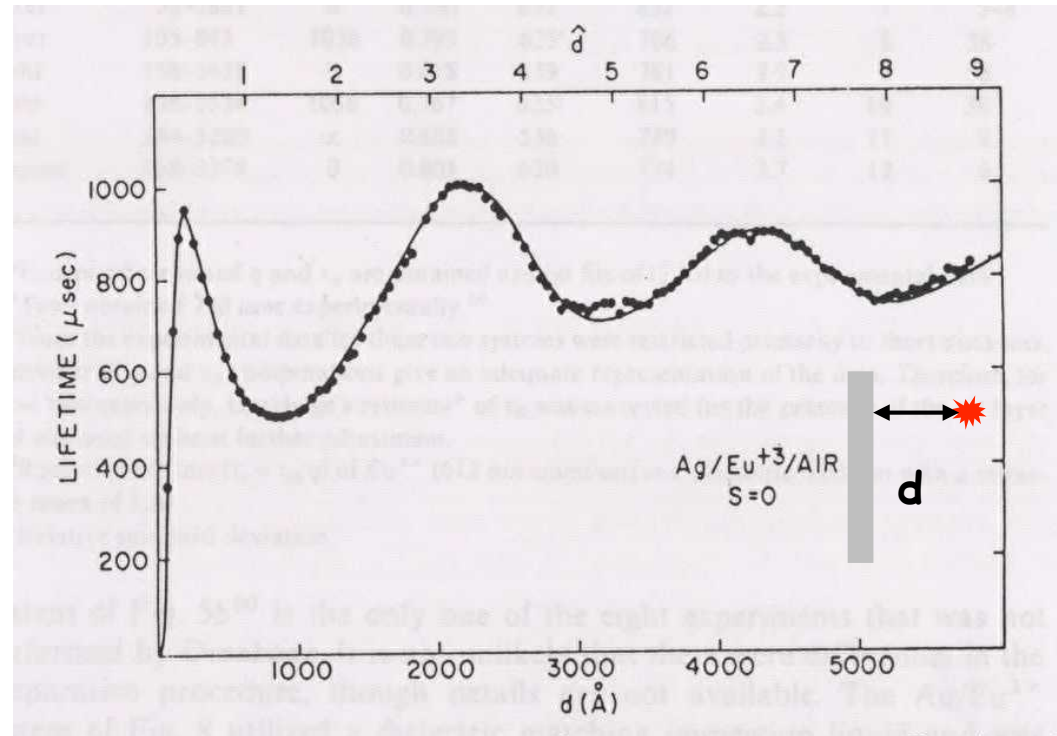
Probing localized plasmons on disordered metallic films

Probing near-field interactions in volume disordered systems (white powders)

# Fluorescence depends on environment

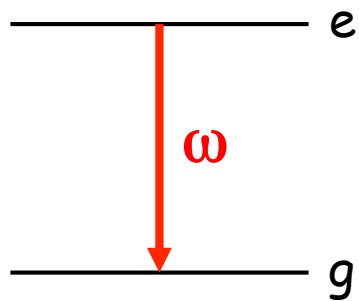


## Lifetime close to a silver mirror



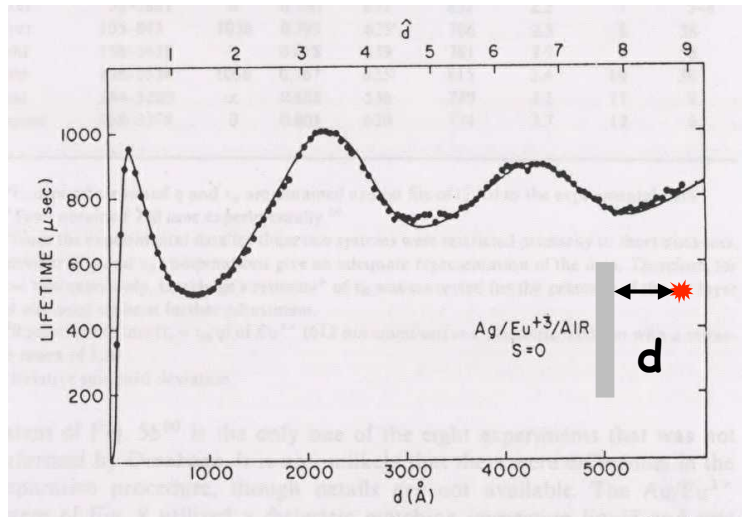
Drexhage (1970)  
Chance, Prock, Silbey (1978)

# How to describe the change in lifetime ?



Probability of being excited at time  $t$   $P(t) \propto \exp(-\Gamma t)$

Lifetime of excited state  $\tau = 1/\Gamma$



Drexhage (1970)  
Chance, Prock, Silbey (1978)

Perturbation theory  
(Fermi golden rule)

$$\Gamma = \frac{\pi \omega}{3\epsilon_0 \hbar} |\mathbf{p}_{ge}|^2 \rho_u(\mathbf{r}_0, \omega)$$

← Local Density of States (LDOS)

$$\frac{\Gamma}{\Gamma_0} = \text{change in the LDOS}$$

# Local Density Of States (LDOS)

- Density Of States (DOS)
  - Counts modes at a given frequency

$$\rho(\omega) = \sum_n \delta(\omega - \omega_n)$$

- Local Density Of States (LDOS)
  - Counts modes at a given frequency weighted by their contribution at point  $\mathbf{r}$

$$\rho(\omega, \mathbf{r}) = \sum_n |\mathbf{E}_n(\mathbf{r})|^2 \delta(\omega - \omega_n)$$

# Purcell effect

The change in the LDOS describes the Purcell effect

E. M. Purcell "Spontaneous emission probabilities at radio frequencies" *Phys. Rev.* **69**, 681 (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

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

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for  $\nu = 10^7 \text{ sec.}^{-1}$ ,  $\mu = 1$  nuclear magneton, the corresponding relaxation time would be  $5 \times 10^{21}$  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  $\nu/Q$  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  $\delta$  is the skin-depth at frequency  $\nu$ ,  $f \sim \lambda^3/a^2\delta$ . 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  $\nu = 10^7 \text{ sec.}^{-1}$ .

For a single mode cavity

$$\frac{\Gamma}{\Gamma_0} = \text{change in LDOS}$$

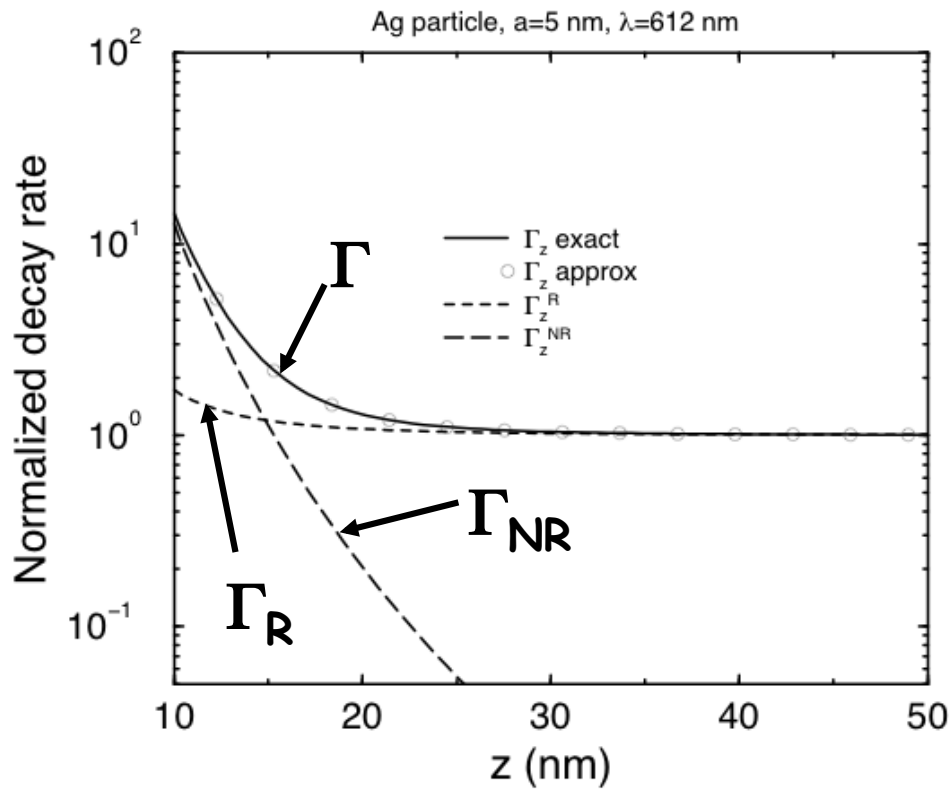
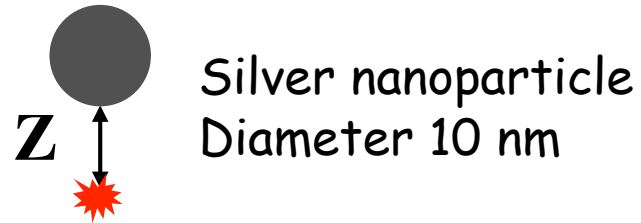


$$\frac{\Gamma}{\Gamma_0} = \frac{3}{4\pi^2} \lambda^3 \frac{Q}{V}$$

(Purcell factor)



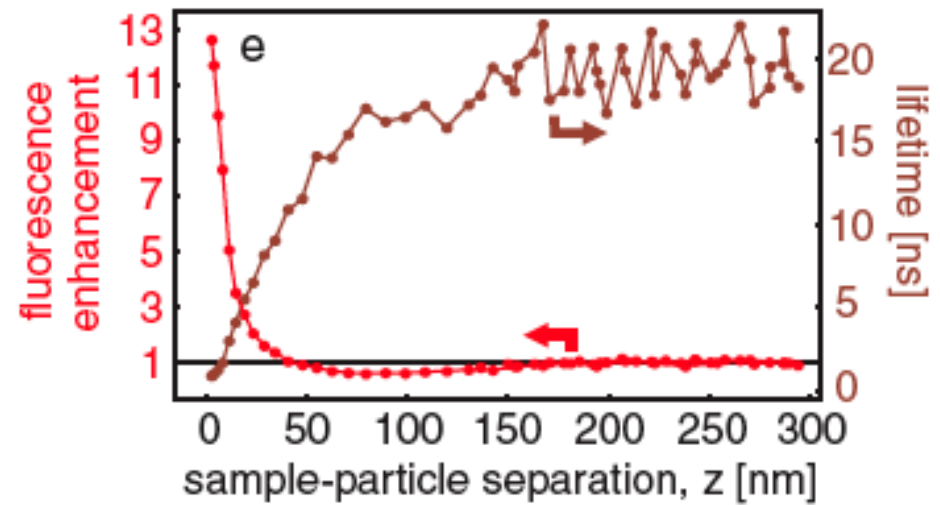
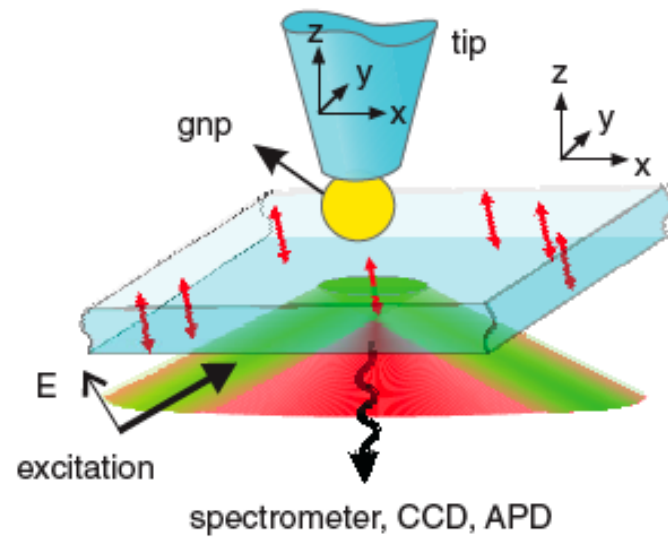
# Interaction with a single nanoparticle



$$\Gamma = \Gamma_R + \Gamma_{NR}$$

Photon emission      Absorption

# Nanoscale controlled experiments on single emitter

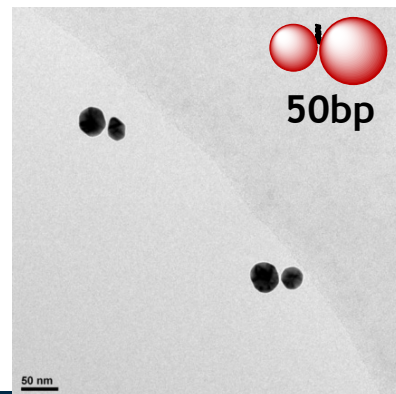
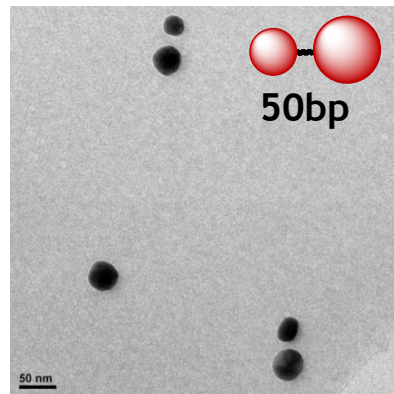
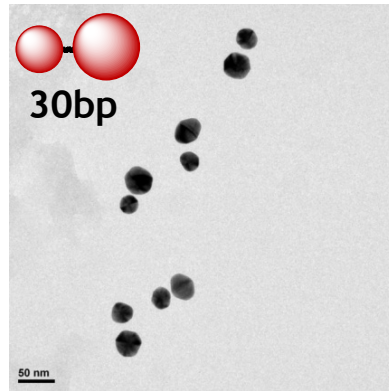


S. Kühn *et al.*, PRL 97, 017402 (2006)

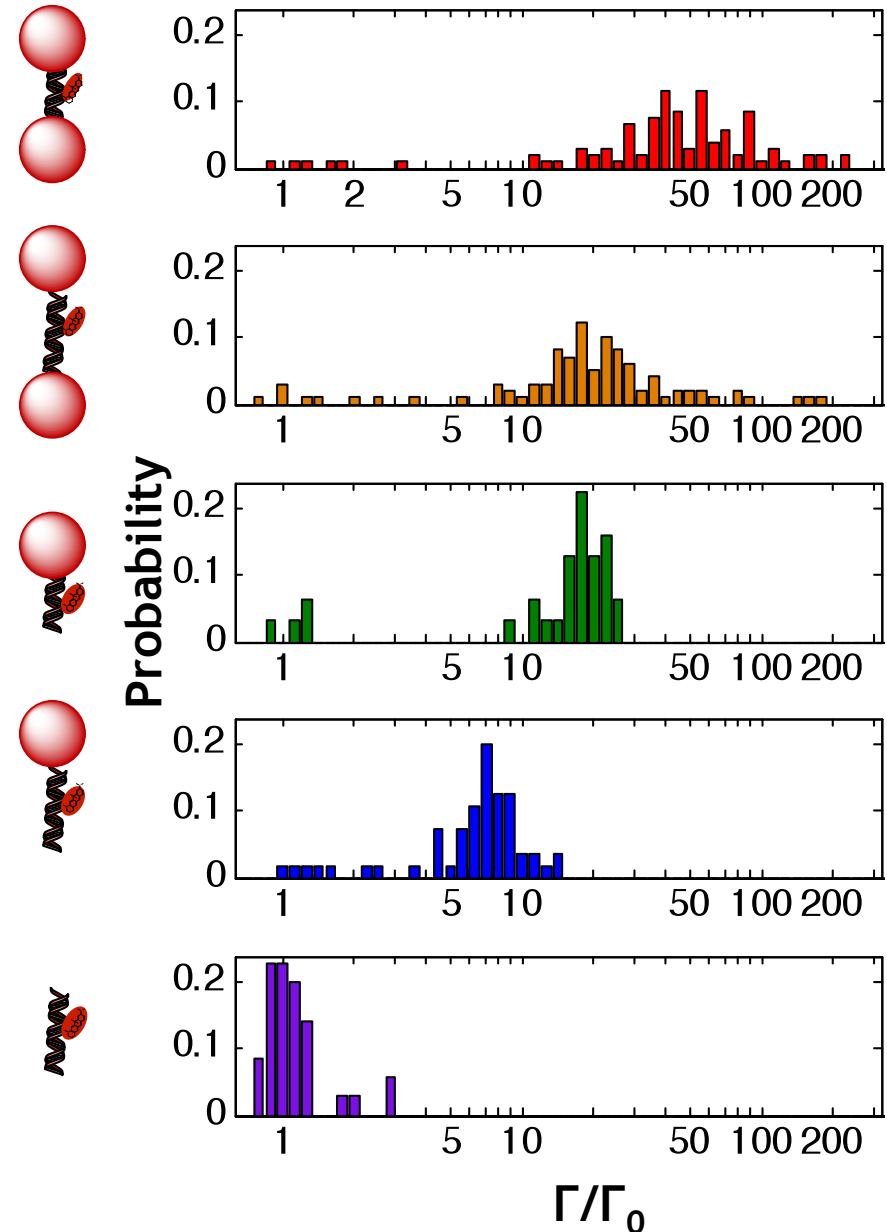
# Nanoparticle dimers as optical antennas



Sébastien BIDAULT  
(CNRS - ESPCI)

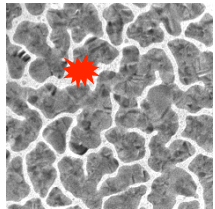


M. P. Busson *et al*,  
Nano Lett. (2011)  
10.1021/nl2032052





Spontaneous emission in nanostructured environments

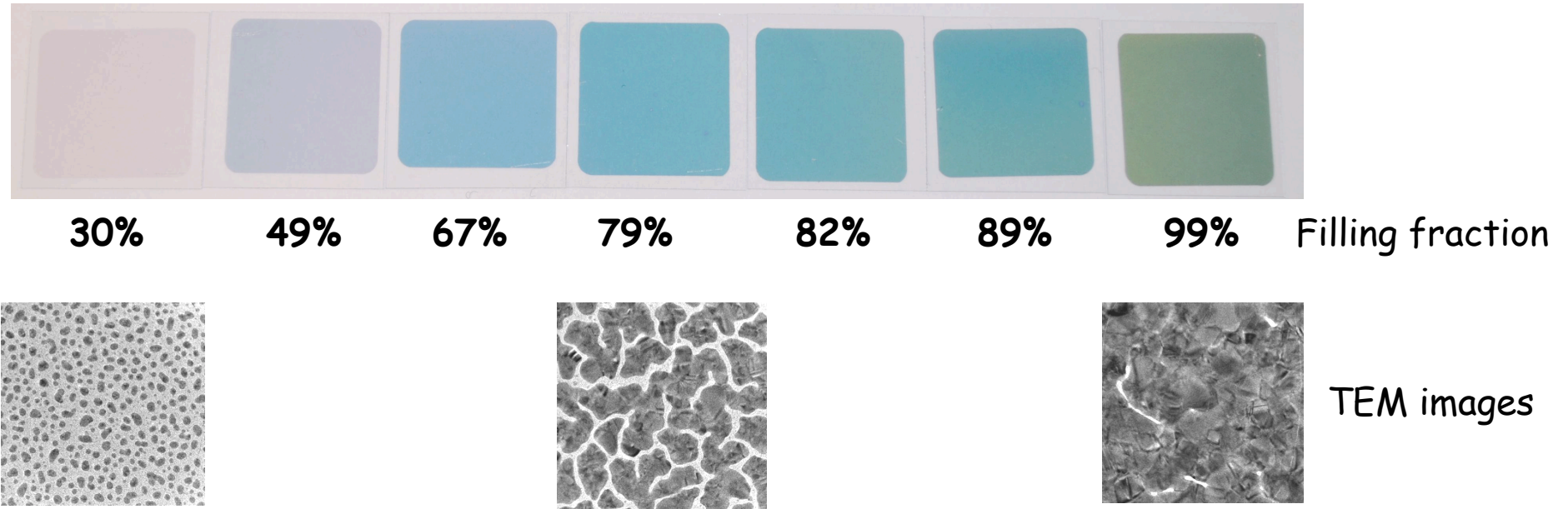


Probing localized plasmons on disordered metallic films

Probing near-field interactions in volume disordered systems (white powders)

# Peculiar optical properties of disordered metal films

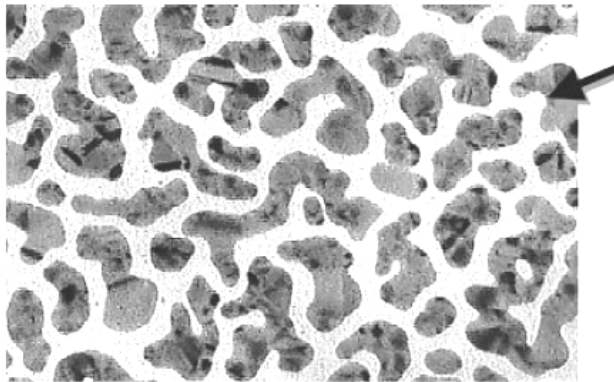
Semi-continuous gold films on a glass substrate



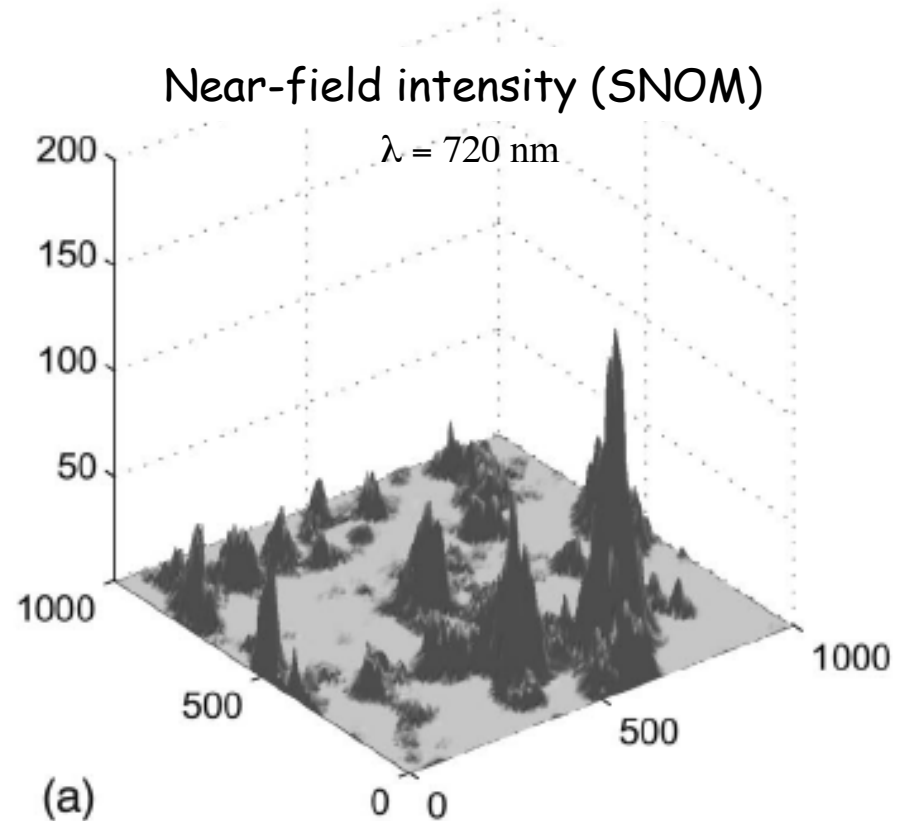
P. Gadenne *et al.*, *J. Appl. Phys.* **66**, 3019 (1989)

V.M. Shalaev, *Nonlinear Optics of Random Media* (Springer, 2000)

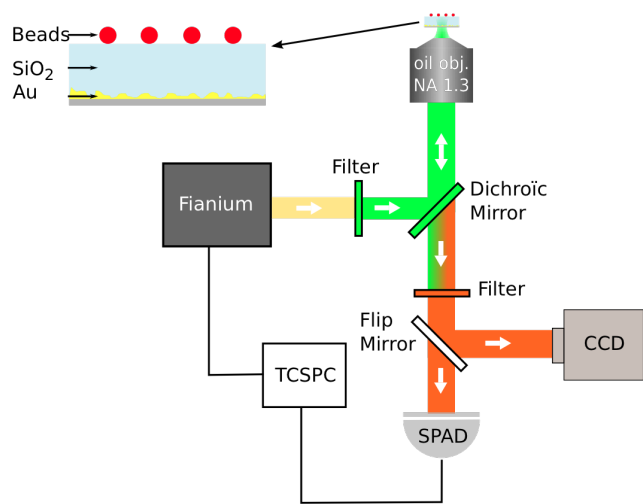
# Near-field intensity distribution - « hot spots »



Surface (TEM image)  
Gold on glass substrate



# LDOS distributions on disordered metal films



V. KRACHMALNICOFF  
Post-doc

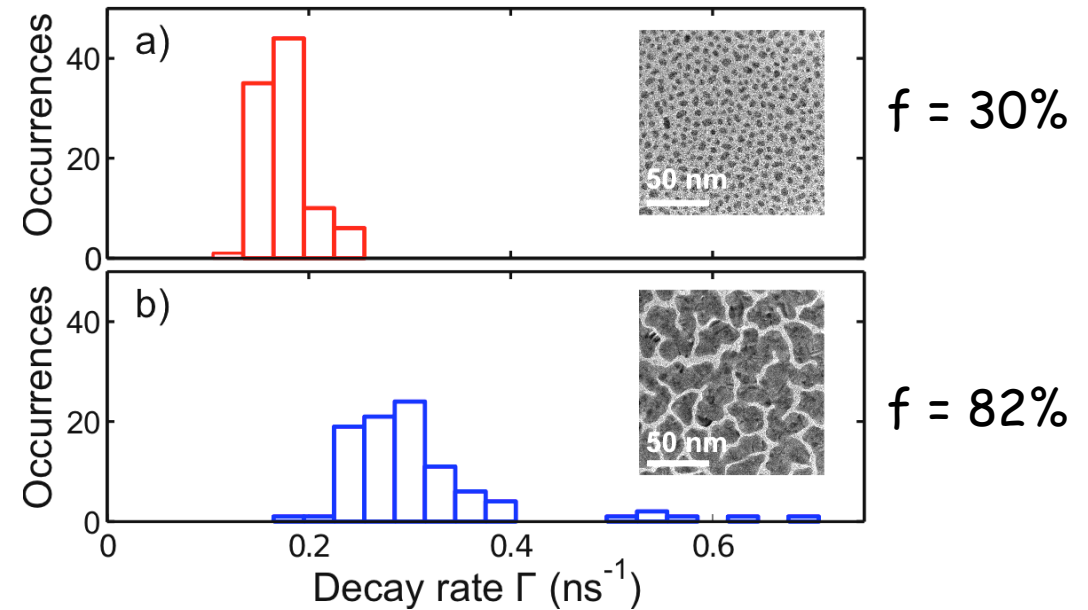
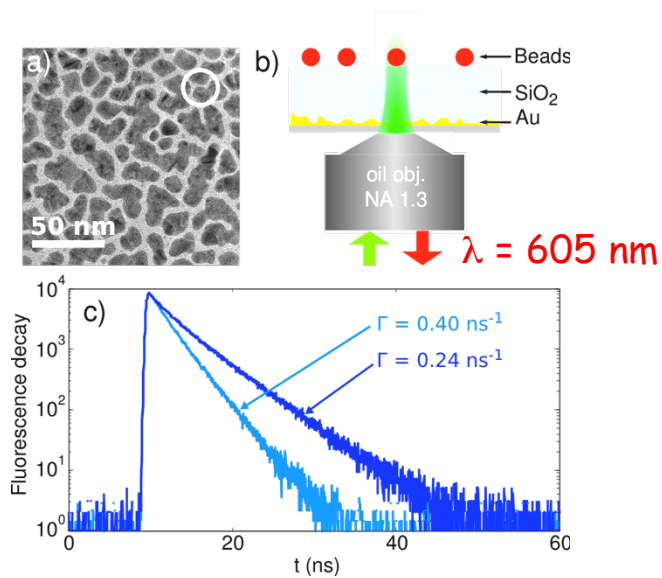


E. CASTANIE  
PhD student

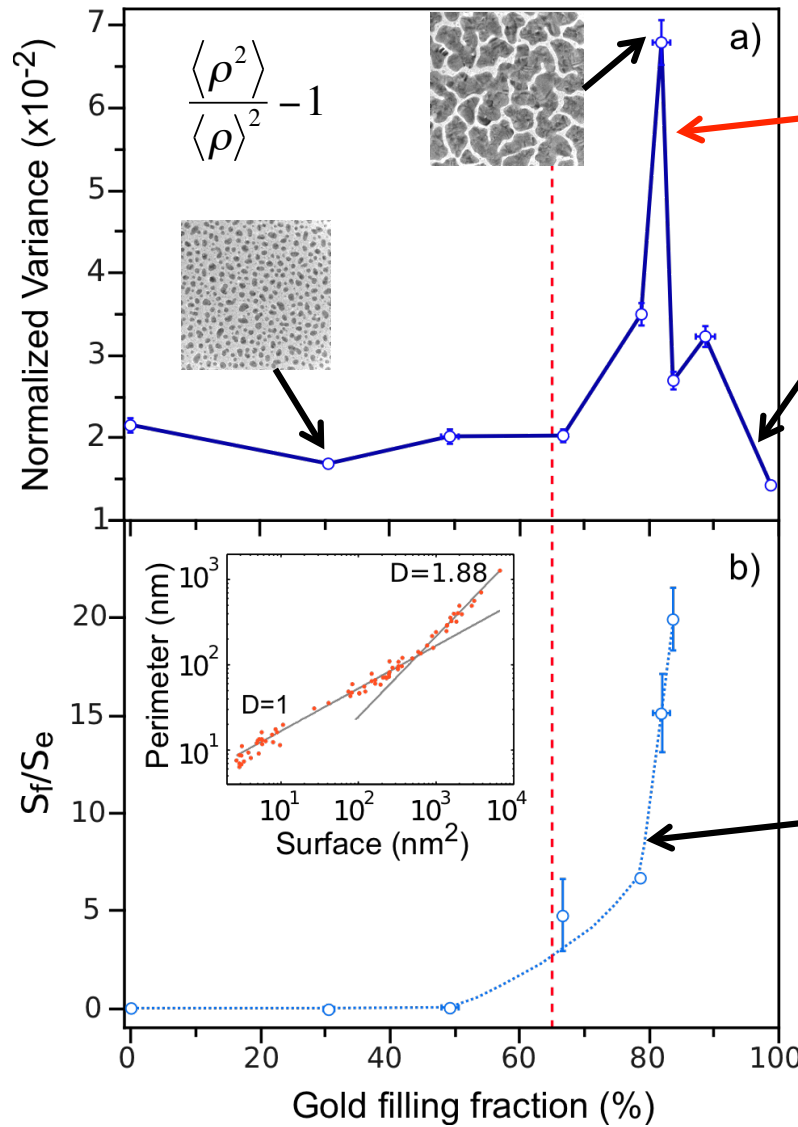


Y. DE WILDE  
(CNRS - ESPCI)

## Statistical distributions of $\Gamma$ (LDOS)



# LDOS fluctuations reveals mode localization



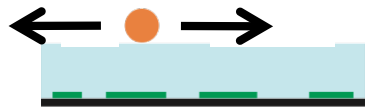
The peak in the LDOS fluctuations is the signature of localized plasmon modes

The disordered film is dominated by fractal clusters

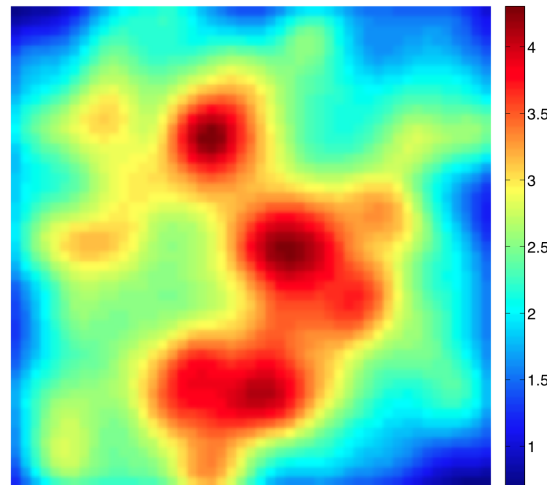


# Numerical simulation provides additional information

$$\Gamma = \Gamma_R + \Gamma_{NR}$$

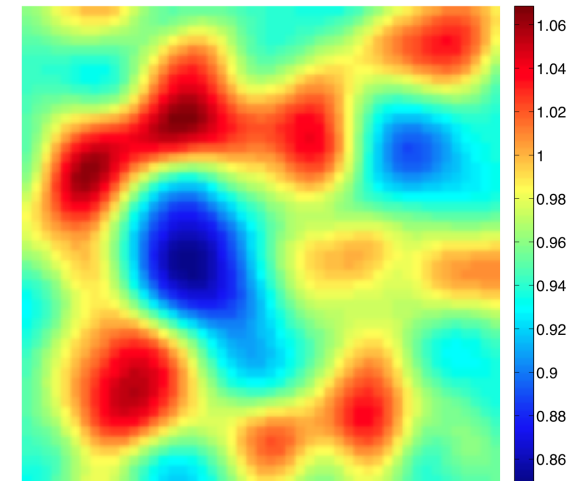


$\Gamma^{NR}/\Gamma_0$

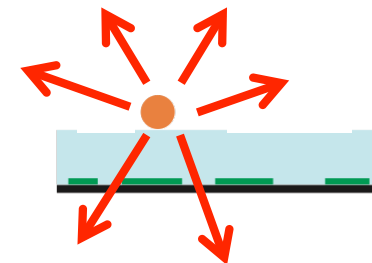


Non-radiative modes

$\Gamma^R/\Gamma_0$

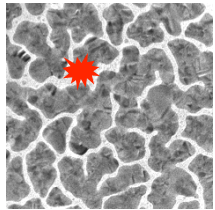


Radiative modes

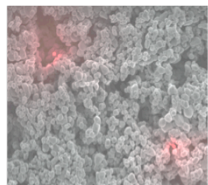




Spontaneous emission in nanostructured environments

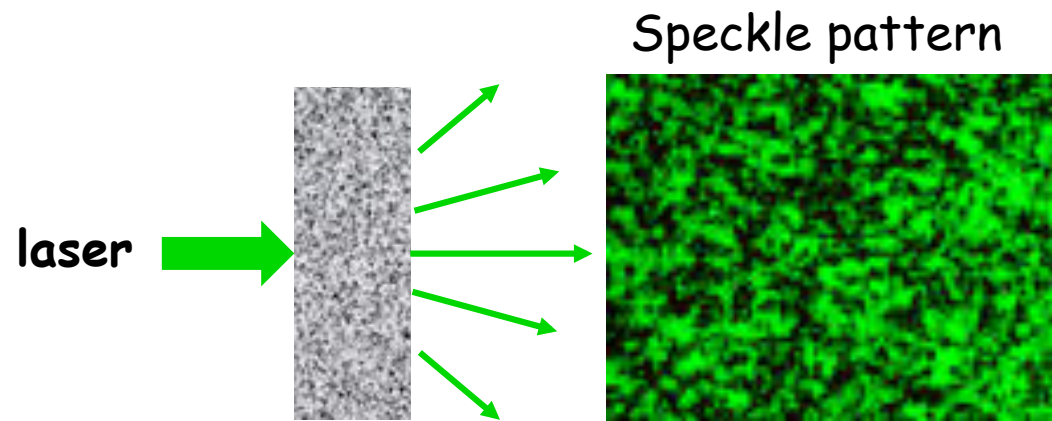


Probing localized plasmons on disordered metallic films



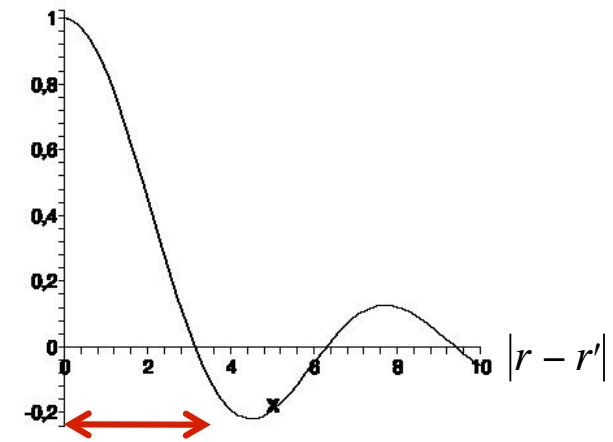
Probing near-field interactions in volume disordered systems (white powders)

# Speckle patterns



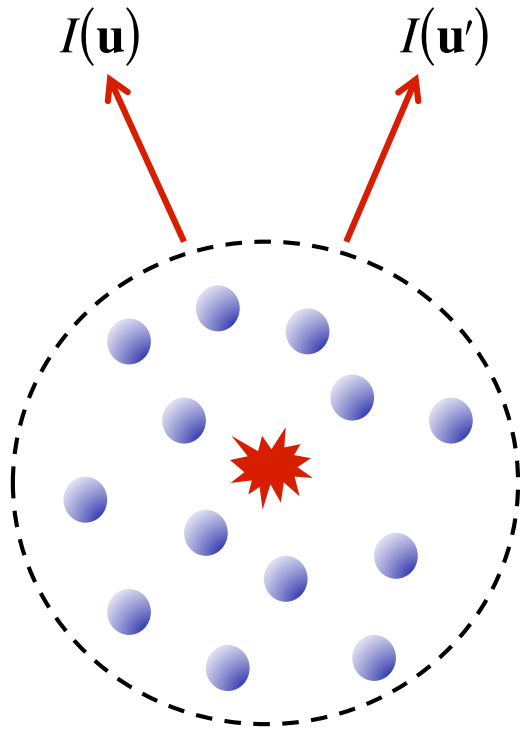
## Intensity-intensity correlation

$$\langle I(r)I(r') \rangle$$



Size of speckle spot

# Speckle produced by a source inside a disordered medium



Infinite-range  $C_0$  speckle correlation

$$C(\mathbf{u}, \mathbf{u}') = C_0 + F(\mathbf{u}, \mathbf{u}')$$

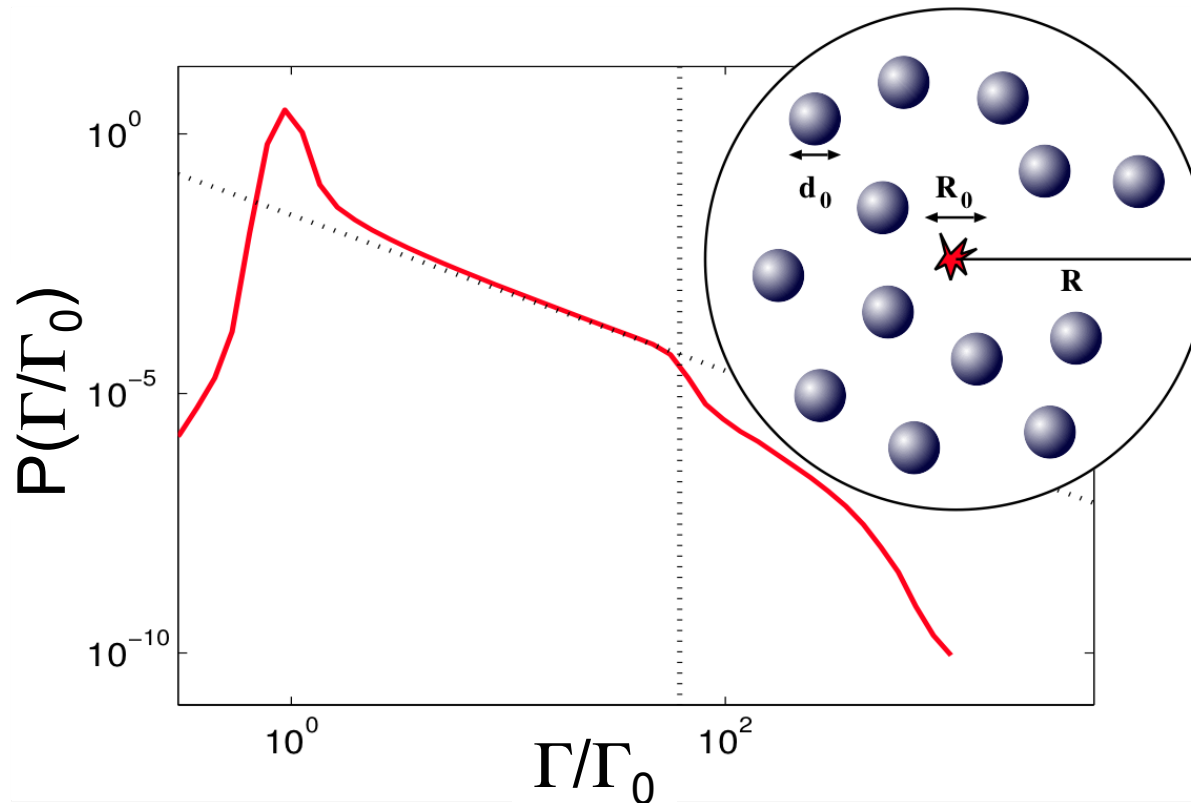
$C_0 =$  LDOS fluctuations

Shapiro, Phys. Rev. Lett. **83**, 4733 (1999)

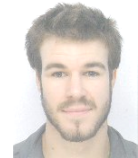
van Tiggelen, Skipetrov, Phys. Rev. E **73**, 045601(R) (2006)

# Typical « numerical experiment »

## Statistical distribution of decay rate $\Gamma$ (LDOS)



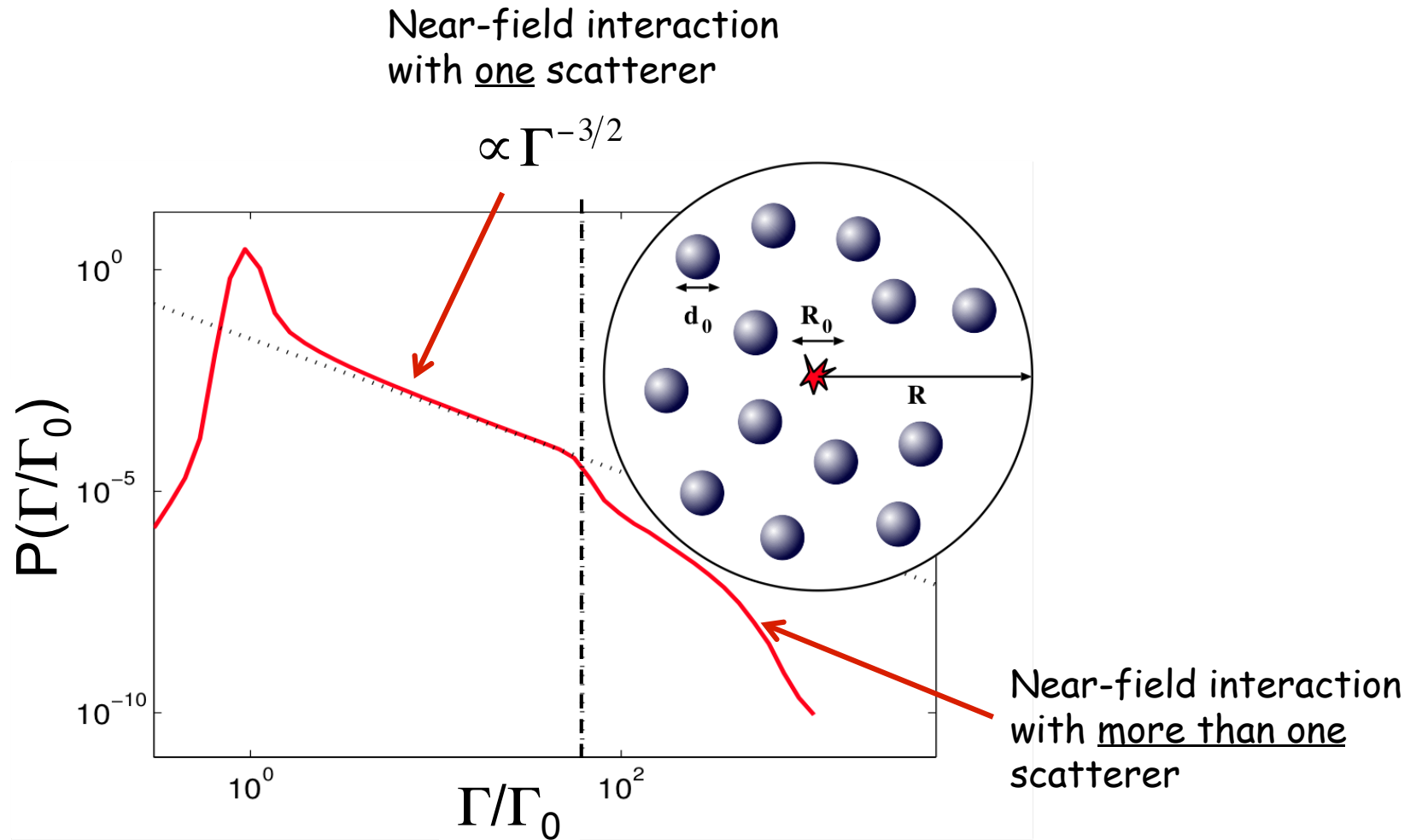
Romain PIERRAT  
(CNRS - ESPCI)



Alexandre CAZE  
PhD student

- Resonant point scatterers (« atoms »)
- $\lambda \approx 630$  nm
- Cluster size  $R = 1.2$   $\mu\text{m}$
- Exclusion volume  $R_0 = 50$  nm

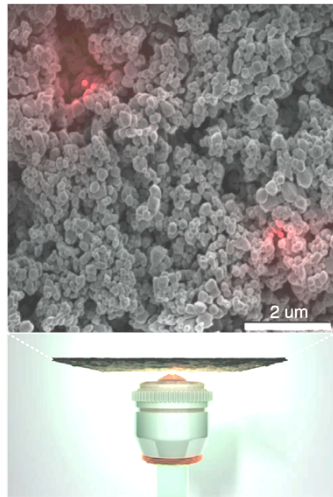
# Long tail: Near-field interactions



# Broad - asymmetric distribution of LDOS (Purcell factor)



Riccardo SAPIENZA  
Niek van HULST  
(ICFO Barcelona, Spain)



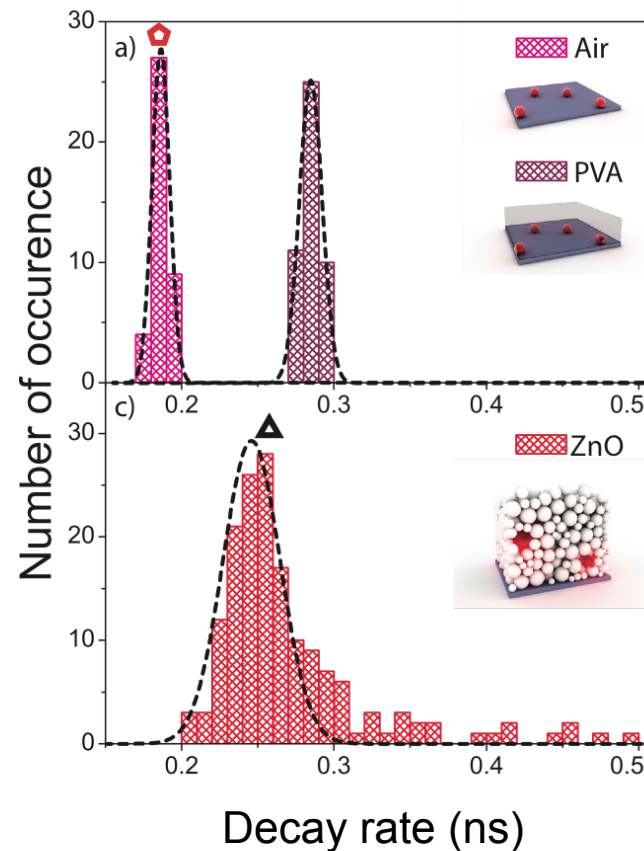
LDOS statistics probed by lifetime of nanosources (24 nm fluorescent beads)

ZnO powder  
Polydisperse particles  
( $140 \pm 50 \text{ nm}$ )

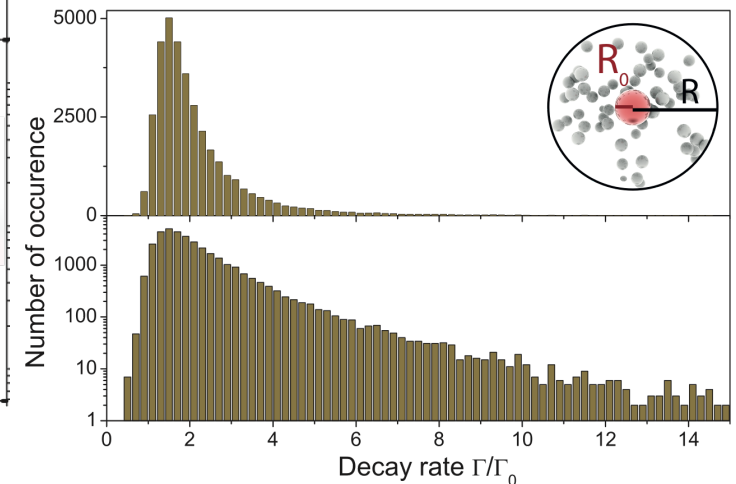
Photon mean free path

$$l = 0.9 \mu\text{m}$$

$$kl = 9.4$$

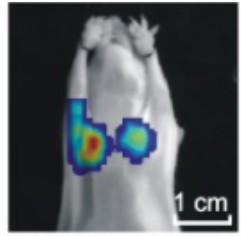


Theory

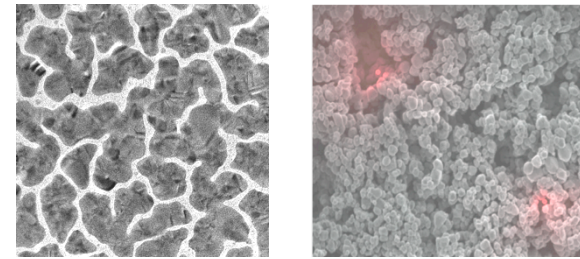


# Coupling spontaneous emission with disorder: Why ?

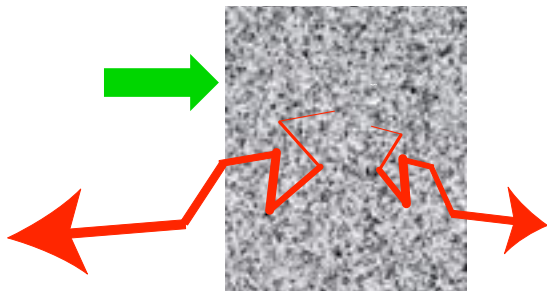
Fluorescence imaging  
in complex media



Nanophotonics - Novel materials



Novel light sources  
(e.g. random lasers)



Fundamental studies of light  
transport in scattering media  
(e.g. probing Anderson localization)

