



Sonder les modes optiques avec des nanosources fluorescentes

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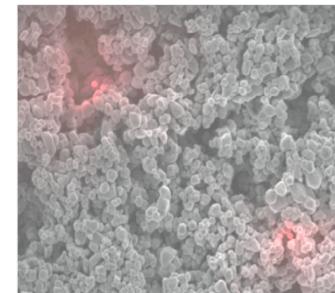
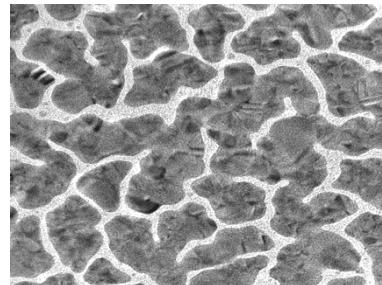
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Sonder les modes optiques avec des nanosources fluorescentes

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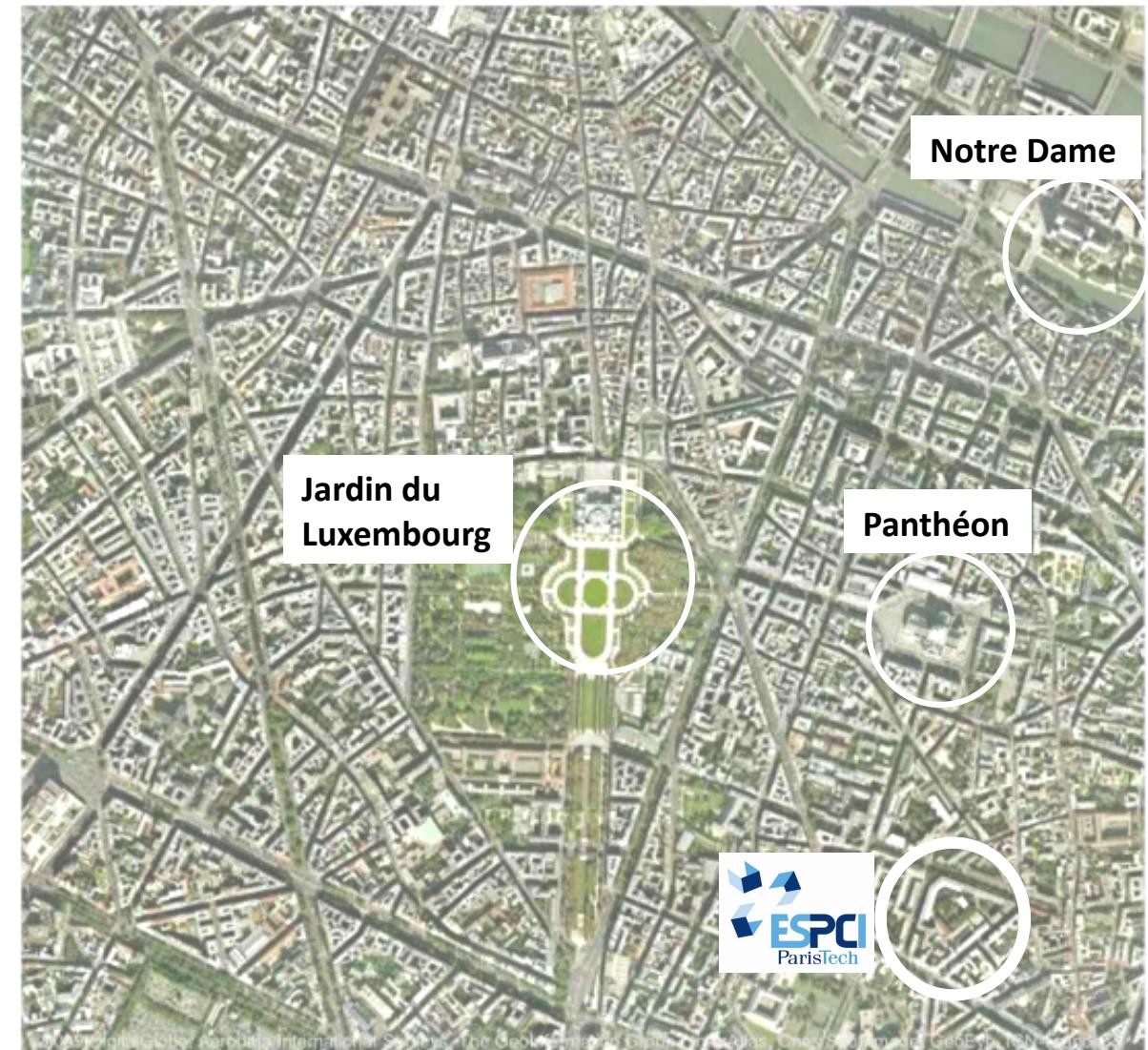
remi.carminati@espci.fr



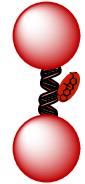
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Outline

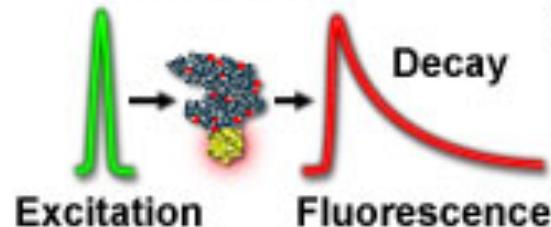


Spontaneous emission in nanostructured environments

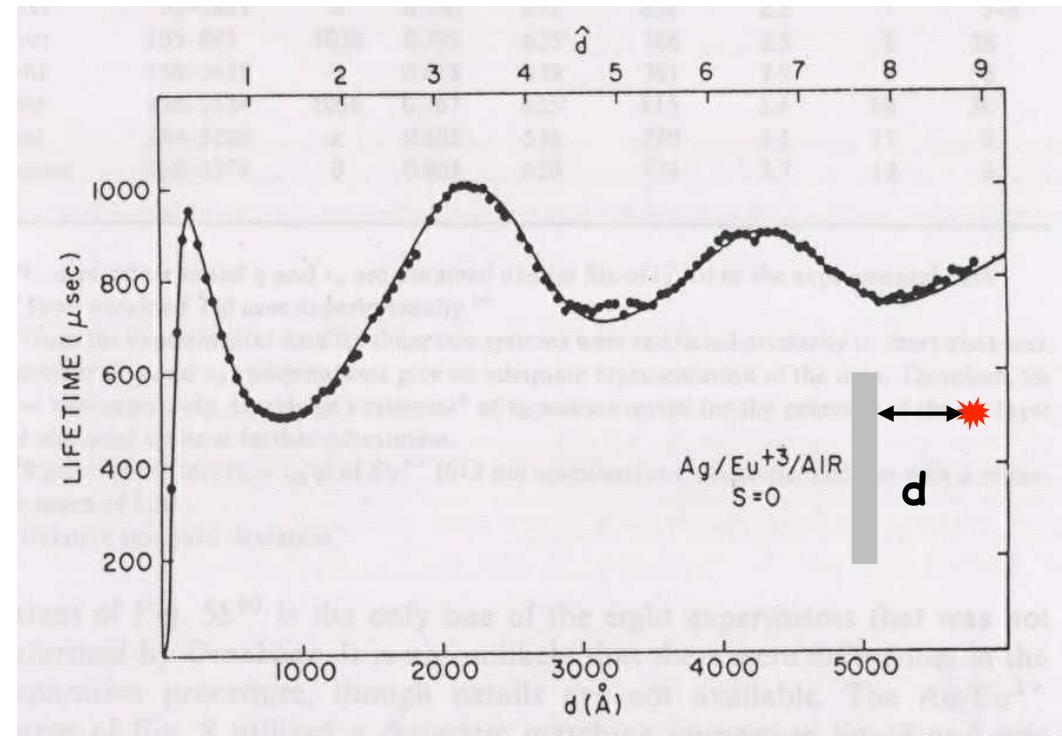
Probing localized plasmons on disordered metallic films

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

Fluorescence depends on environment

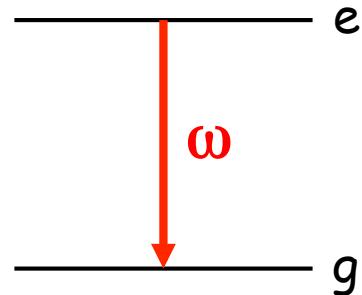


Lifetime close to a silver mirror



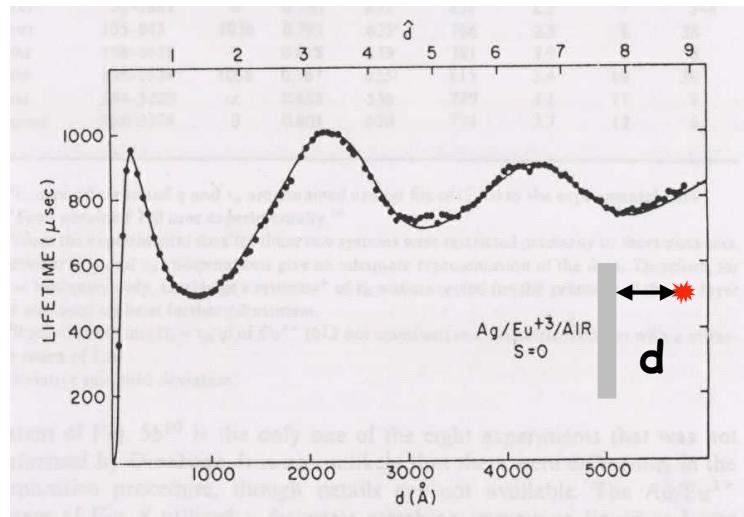
Drexhage (1970)
Chance, Prock, Silbey (1978)

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

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

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu = 10^7 \text{ sec.}^{-1}$, $\mu = 1$ 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 $\nu = 10^7 \text{ sec.}^{-1}$.

For a single mode cavity

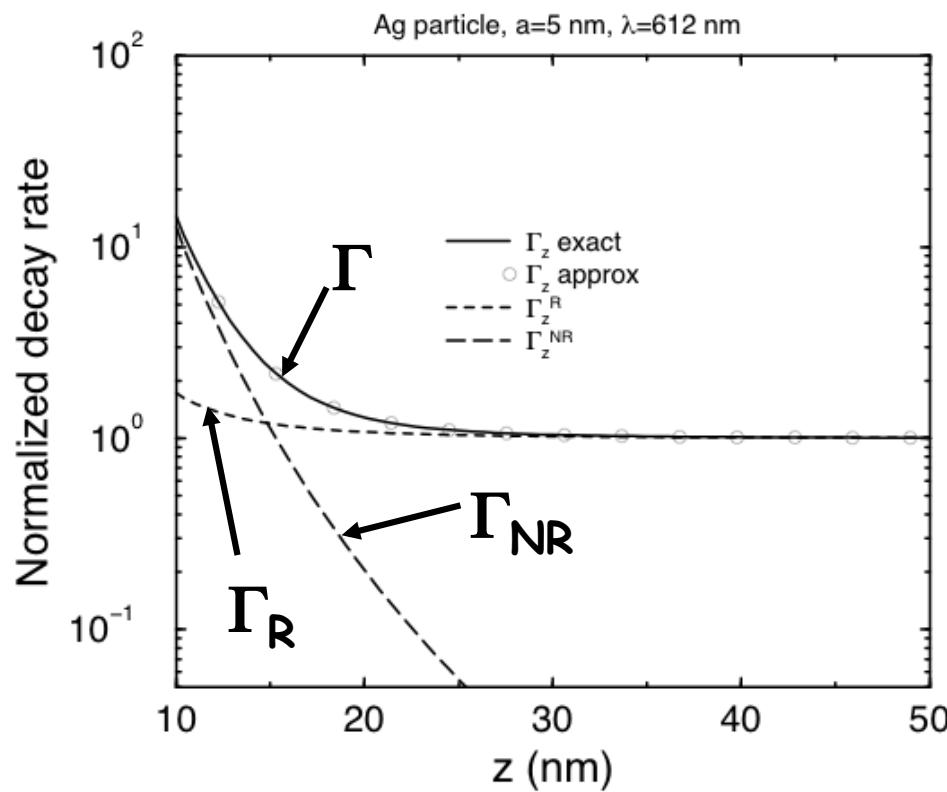
$$\frac{\Gamma}{\Gamma_0} = \text{change in LDOS}$$



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

(Purcell factor)

Interaction with a single nanoparticle

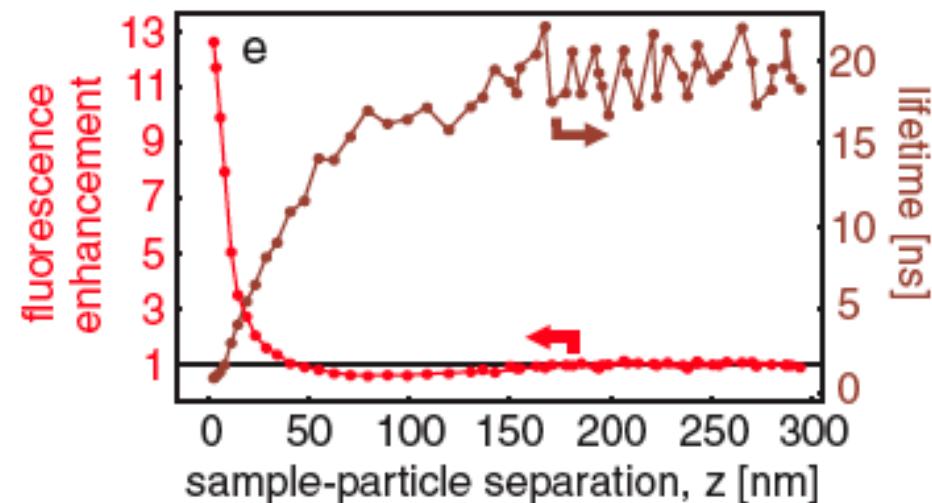
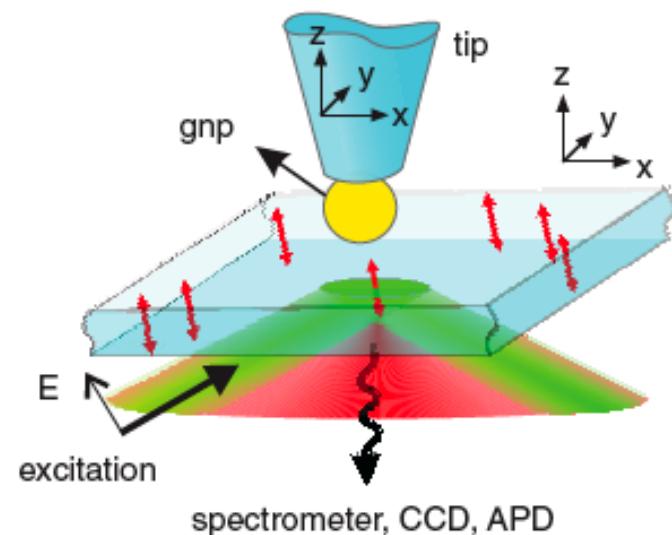


$$\Gamma = \Gamma_R + \Gamma_{NR}$$

Photon emission

Absorption

Nanoscale controlled experiments on single emitter

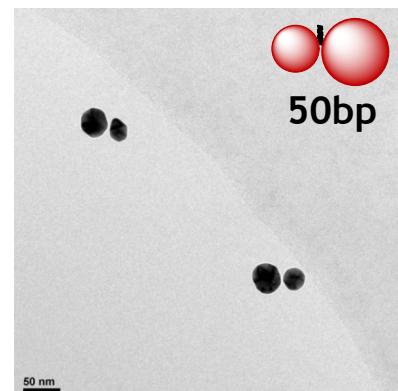
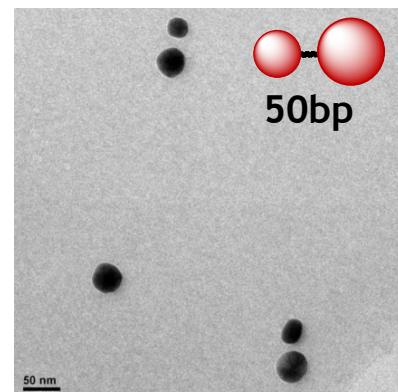
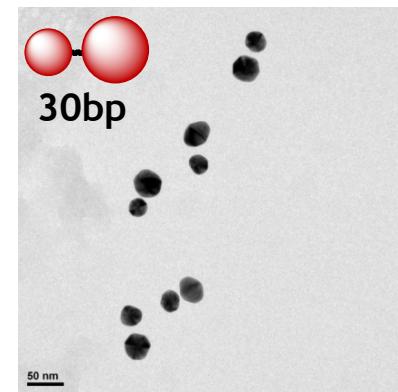


S. Kühn *et al.*, PRL 97, 017402 (2006)

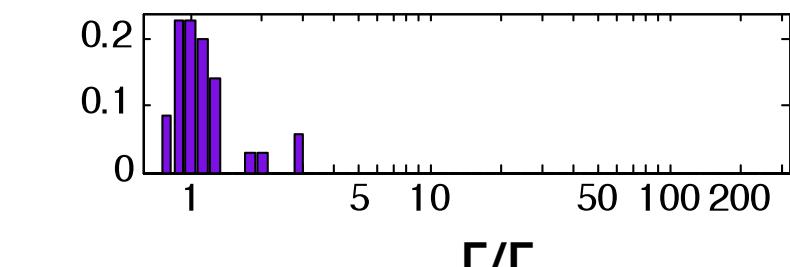
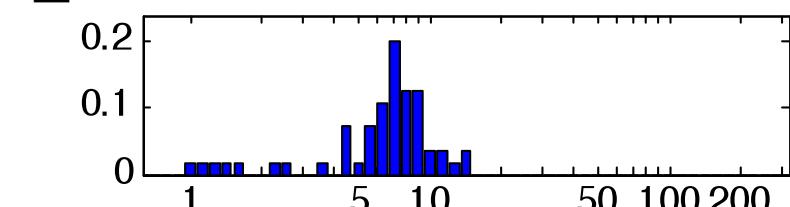
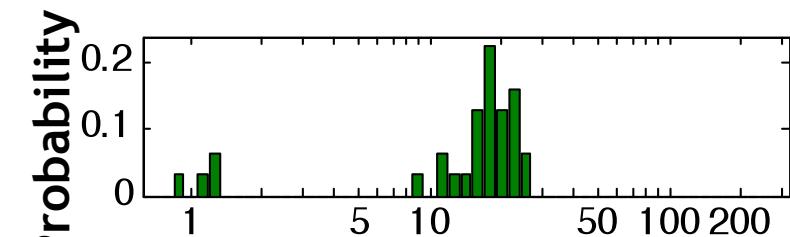
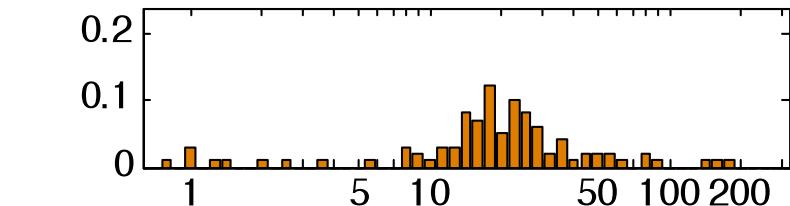
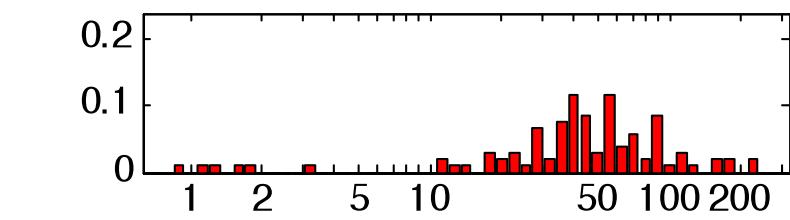
Nanoparticle dimers as optical antennas

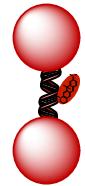


Sébastien BIDAULT
(CNRS - ESPCI)

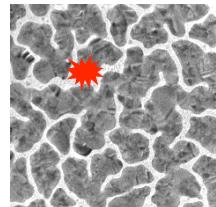


M. P. Busson *et al.*,
Nano Lett. (2011)
10.1021/nl2032052





Spontaneous emission in nanostructured environments

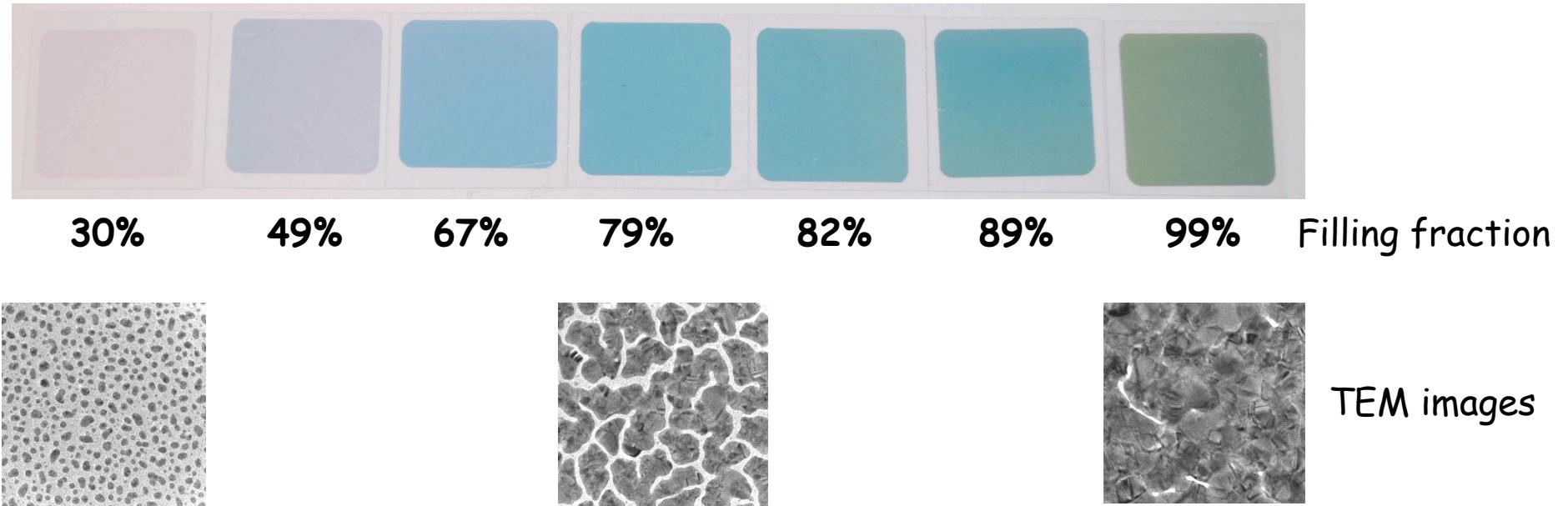


Probing localized plasmons on disordered metallic films

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

Peculiar optical properties of disordered metal films

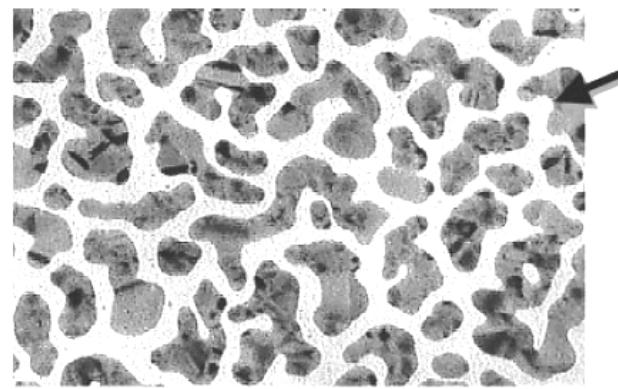
Semi-continuous gold films on a glass substrate



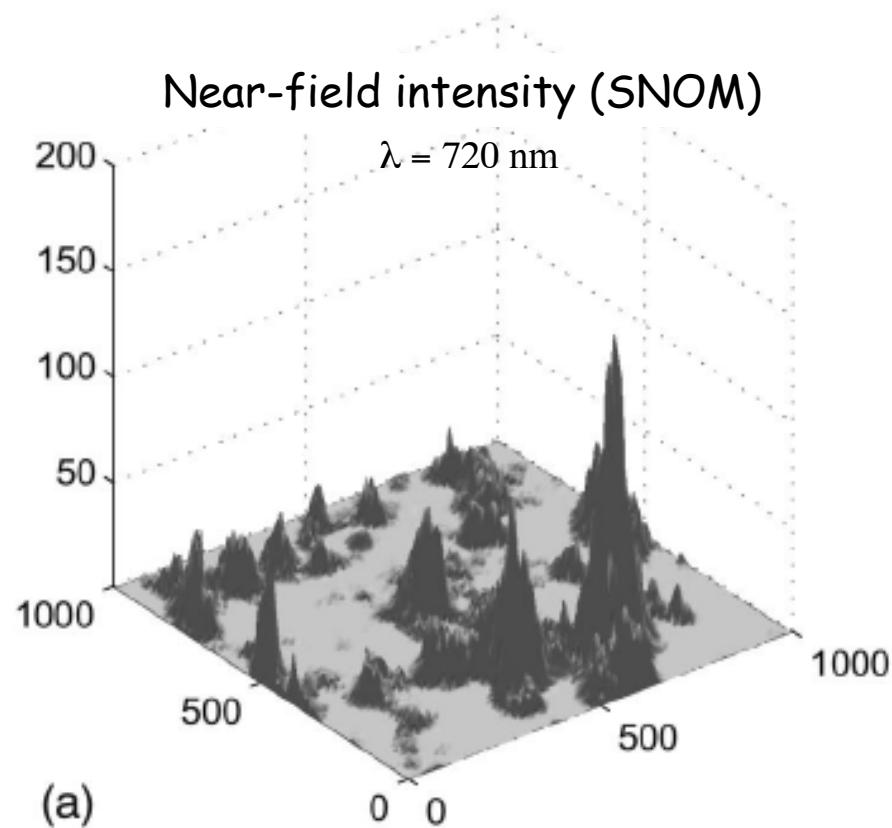
P. Gadenne *et al.*, J. Appl. Phys. **66**, 3019 (1989)

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

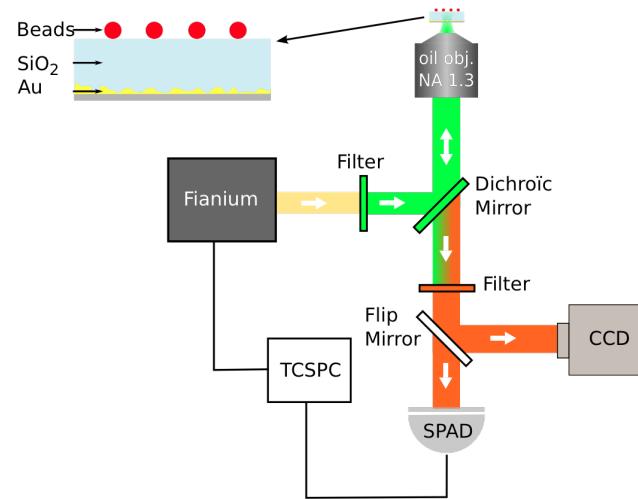
Near-field intensity distribution - « hot spots »



Surface (TEM image)
Gold on glass substrate



LDOS distributions on disordered metal films



V. KRACHMALNICOFF
Post-doc

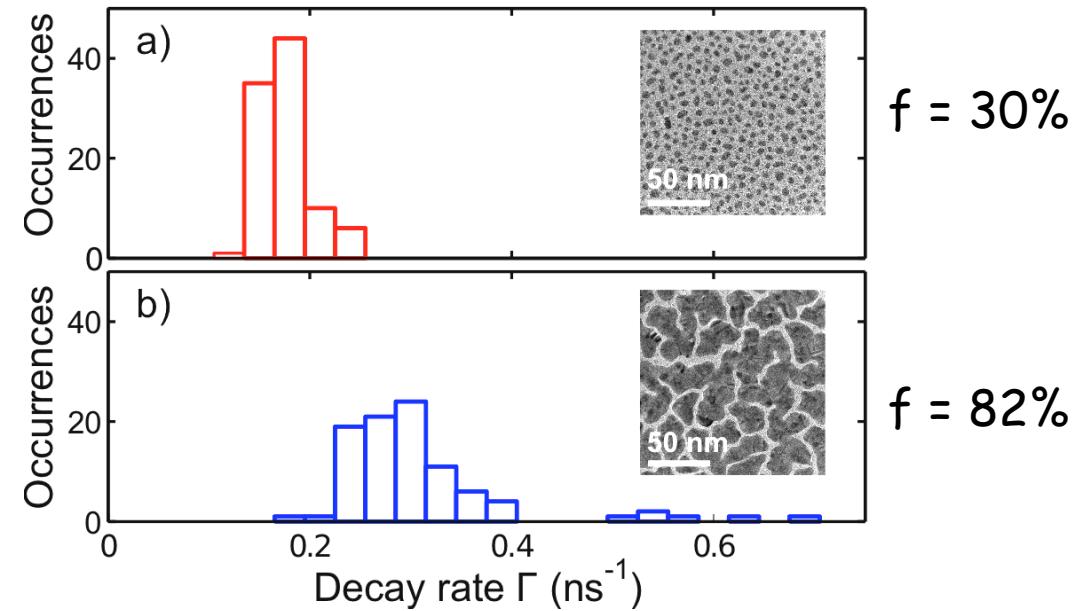
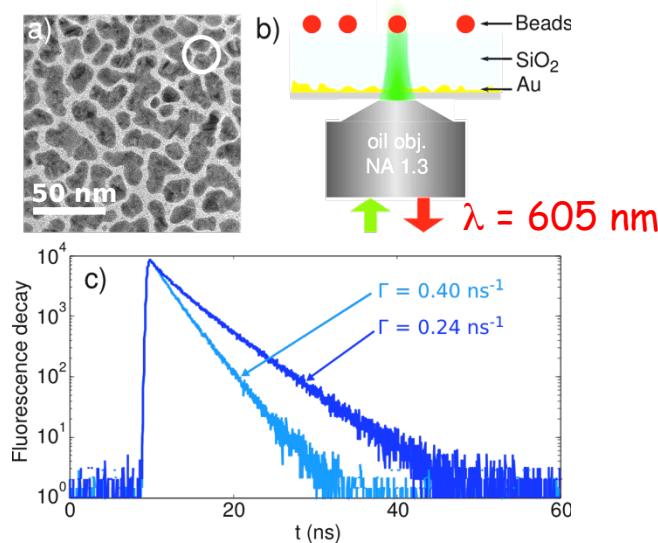


E. CASTANIE
PhD student

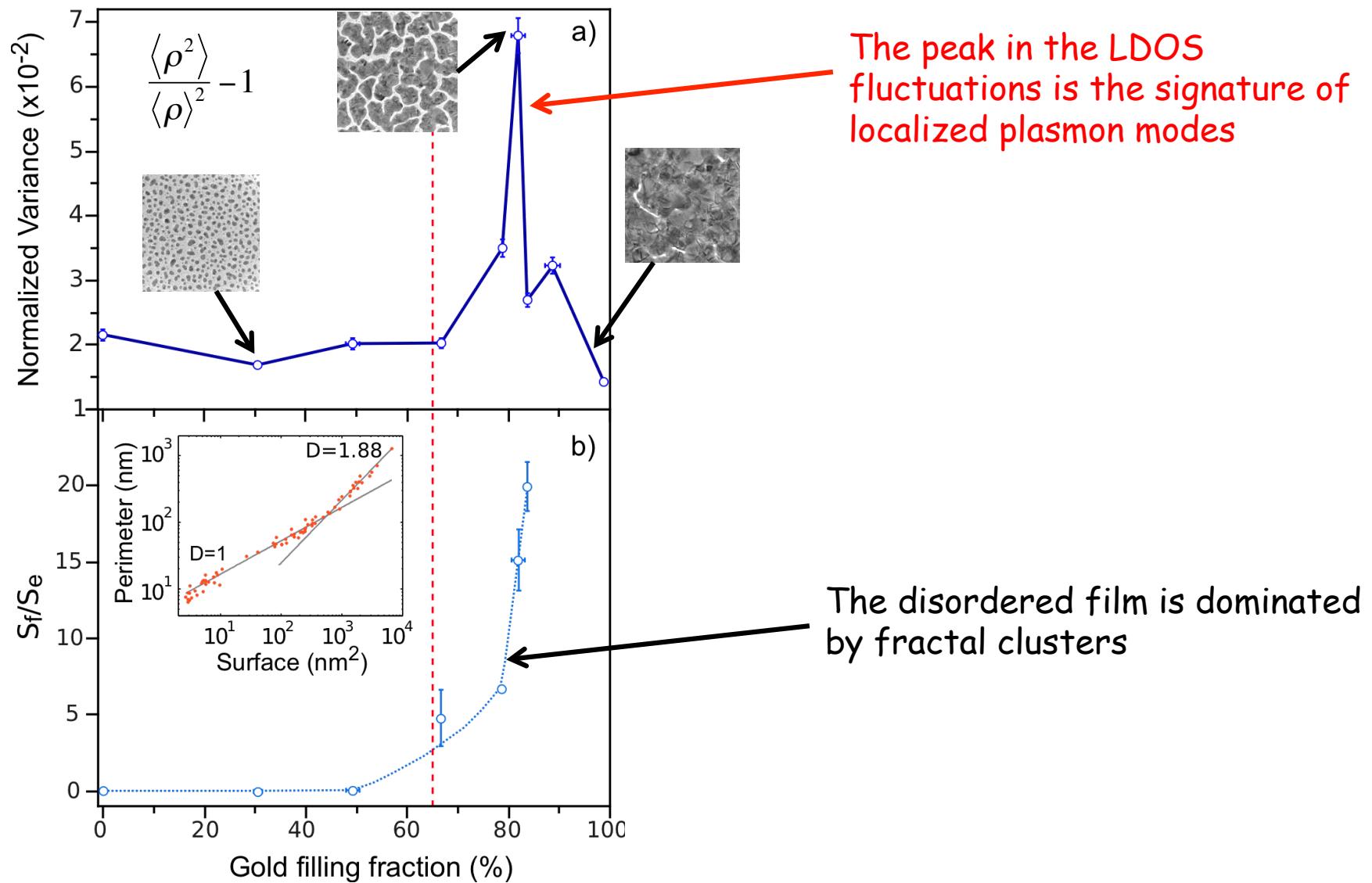


Y. DE WILDE
(CNRS - ESPCI)

Statistical distributions of Γ (LDOS)

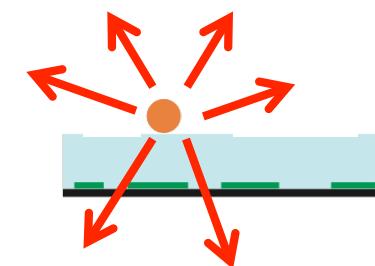
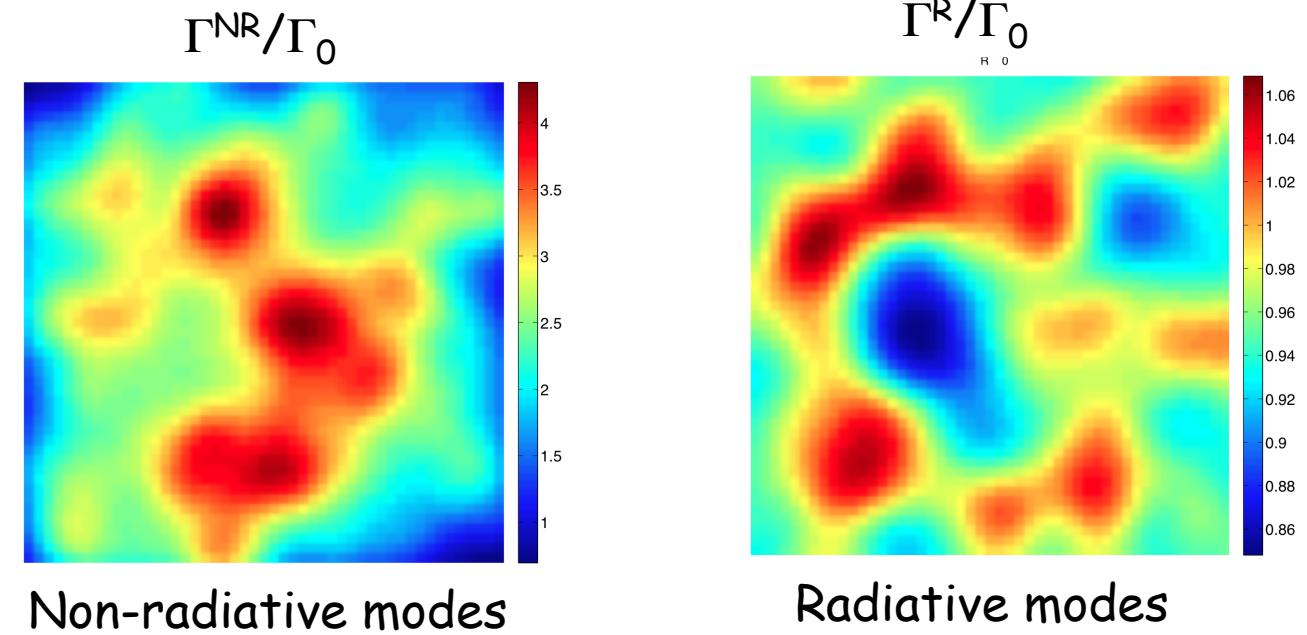
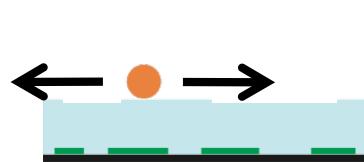


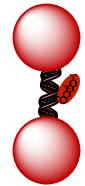
LDOS fluctuations reveals mode localization



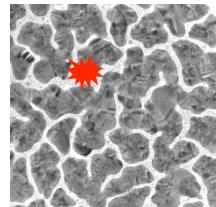
Numerical simulation provides additionnal information

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

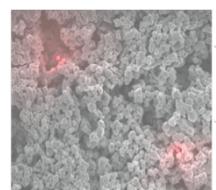




Spontaneous emission in nanostructured environments

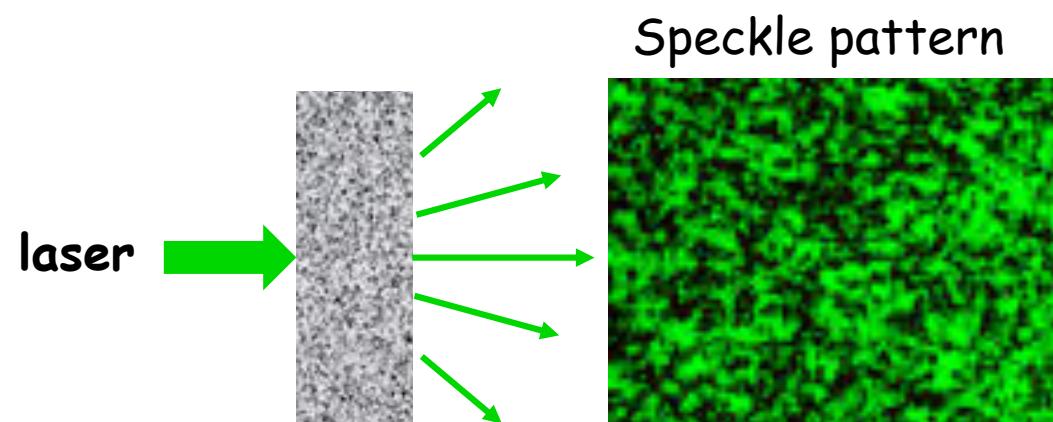


Probing localized plasmons on disordered metallic films



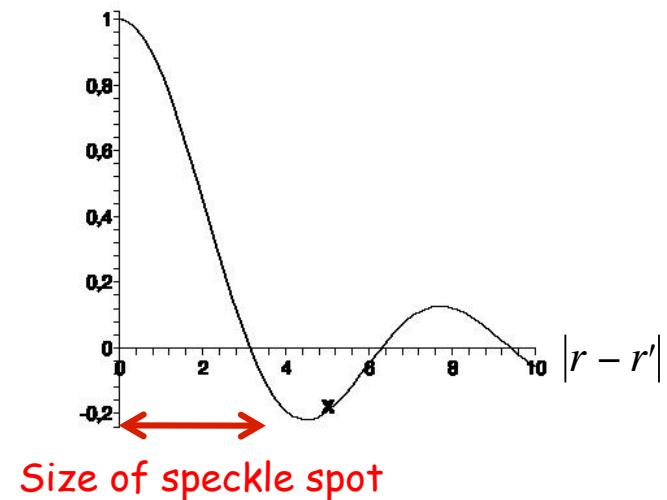
Probing near-field interactions in volume
disordered systems (white powders)

Speckle patterns

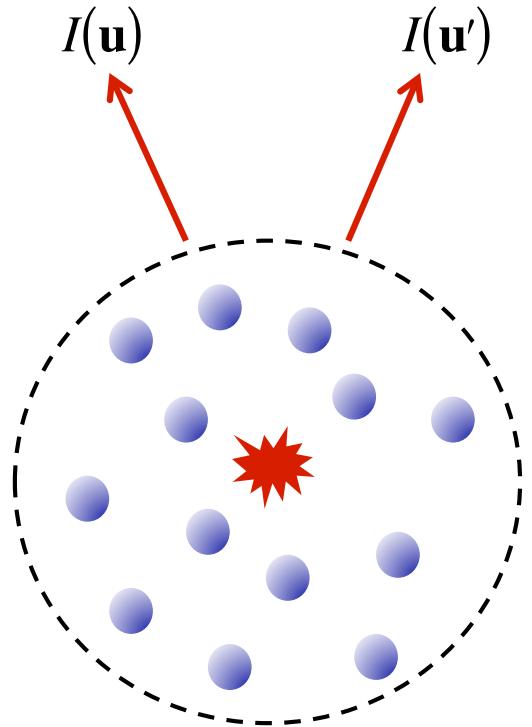


Intensity-intensity correlation

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



Speckle produced by a source inside a disordered medium



Infinite-range C_0 speckle correlation

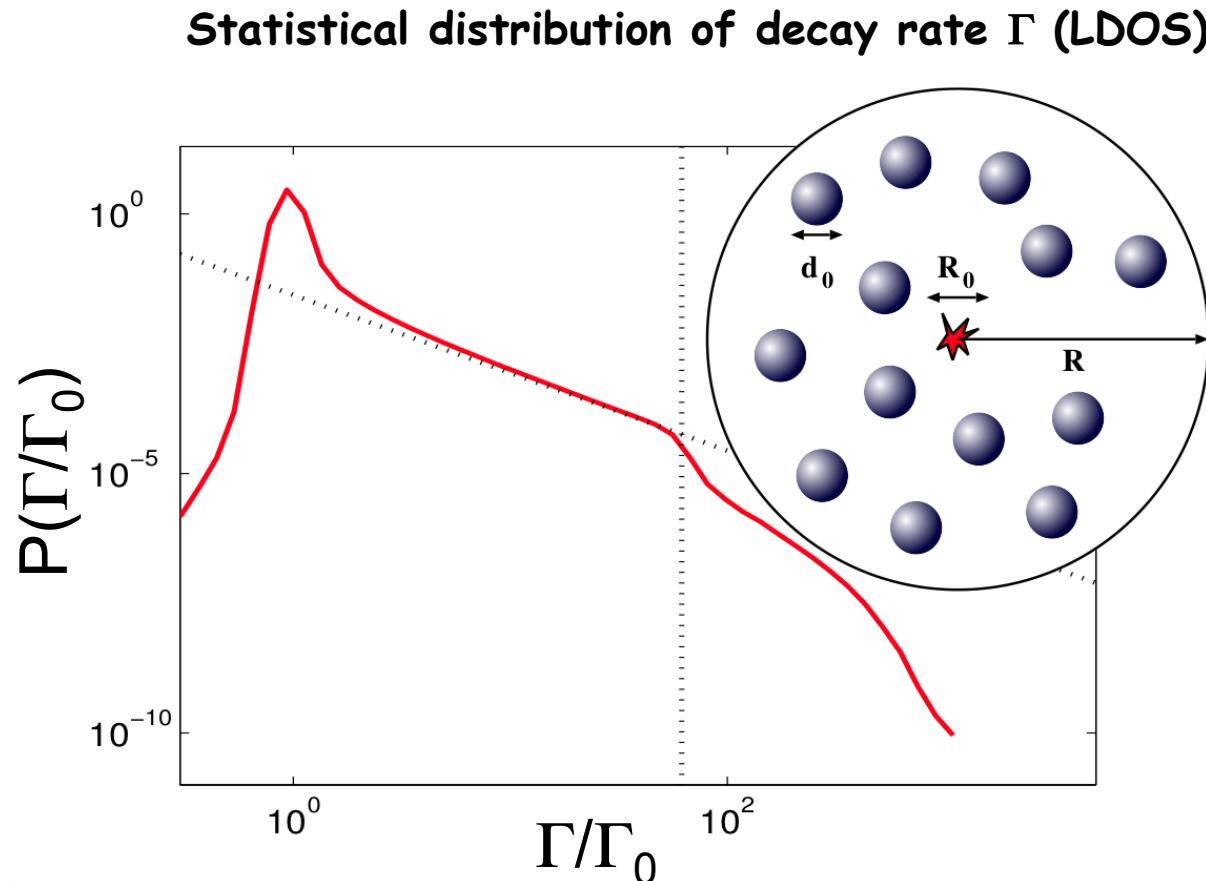
$$C(\mathbf{u}, \mathbf{u}') = C_0 + F(\mathbf{u}, \mathbf{u}')$$

C_0 = LDOS fluctuations

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

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

Typical « numerical experiment »



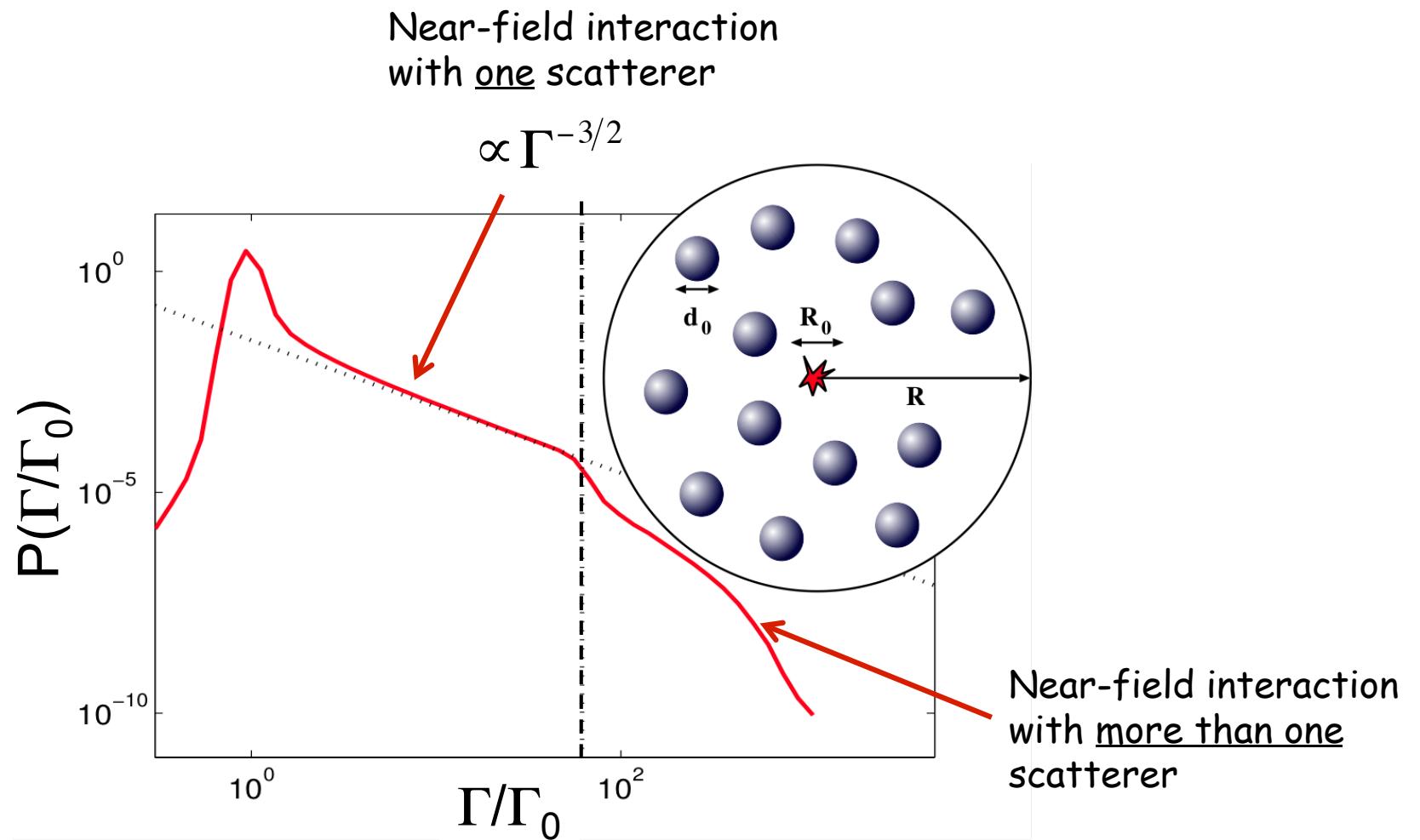
Romain PIERRAT
(CNRS - ESPCI)



Alexandre CAZE
PhD student

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

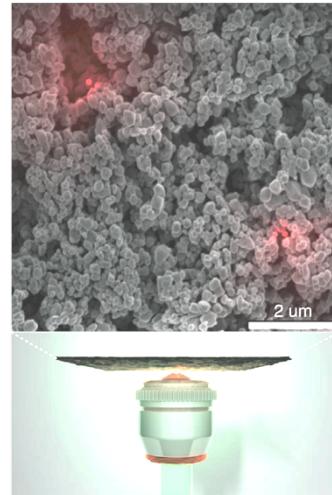
Long tail: Near-field interactions



Broad - asymmetric distribution of LDOS (Purcell factor)



Riccardo SAPIENZA
Niek van HULST
(ICFO Barcelona, Spain)



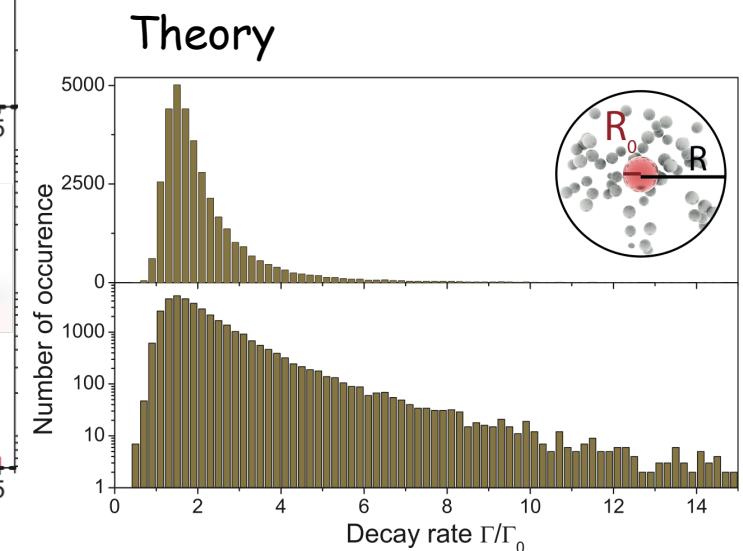
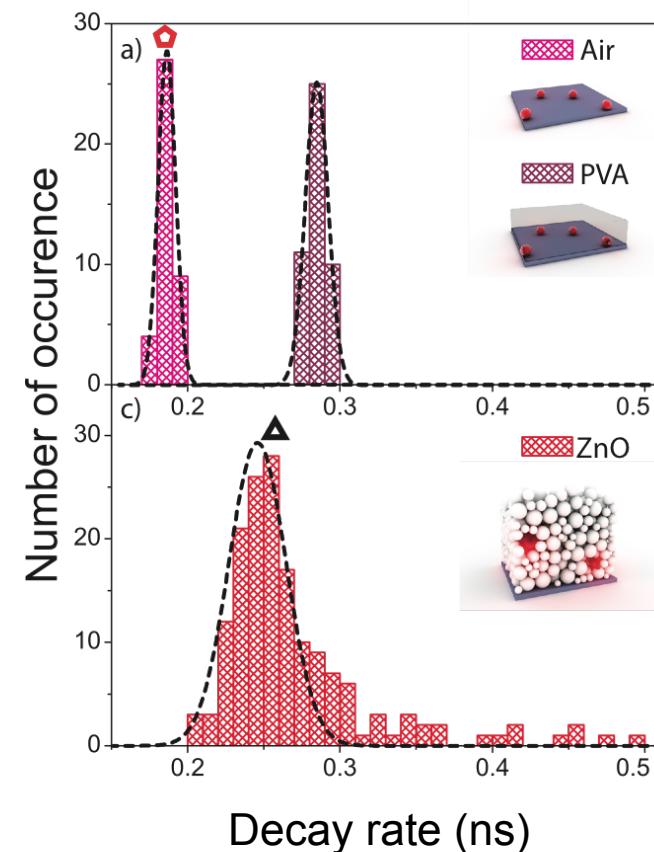
ZnO powder
Polydisperse particles
 $(140 \pm 50 \text{ nm})$

Photon mean free path

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

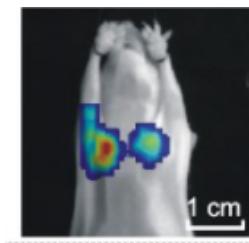
$$k\ell = 9.4$$

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

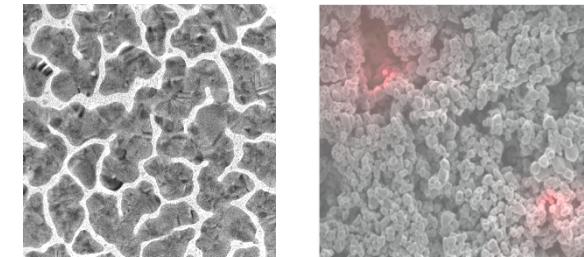


Coupling spontaneous emission with disorder: Why ?

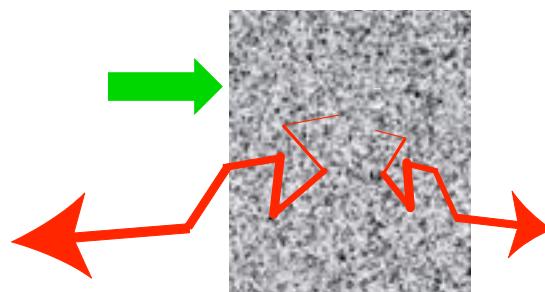
Fluorescence imaging
in complex media



Nanophotonics - Novel materials



Novel light sources
(e.g. random lasers)



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

