Probing confined photons in nanoscale disordered media from inside

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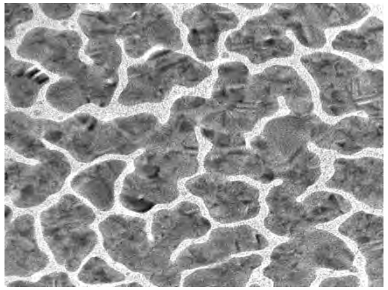

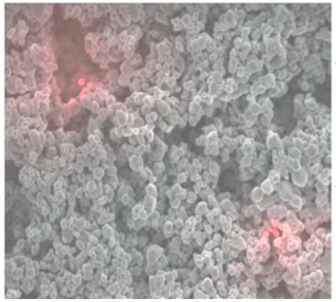

People involved

"Physical optics and wave theory" group (ESPCI)

Romain PIERRAT
CNRS researcher

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PhD student

Alexandre CAZE
PhD student

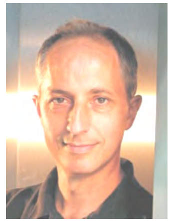
Valentina
KRACHMALNICOFF
Post-doc

Mohamed ELABED
Associate researcher

Rémi VINCENT
Post-doc
(until July 2011)

Collaborations

Yannick DE WILDE
(ESPCI)

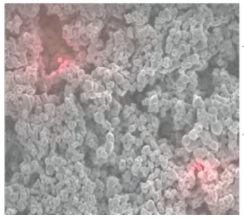


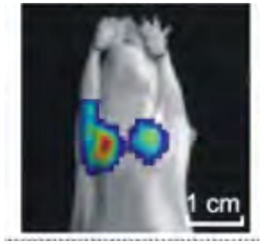
Riccardo SAPIENZA
Niek van HULST
(ICFO Barcelona, Spain)

ESPCI ParisTech — MAIRIE DE PARIS — cnrs — ANR — NanoSciences ILE-DE-FRANCE — nanomagma — molecular imaging — FONDATION PIERRE-GILLES DE GENNES POUR LA RECHERCHE — Quidd Smart Molecular Imaging

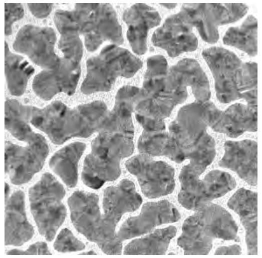
Coupling spontaneous emission with disorder

Fluorescence of nanosources in disordered media (photonic materials, imaging)

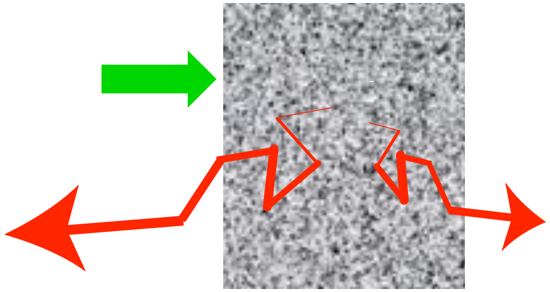




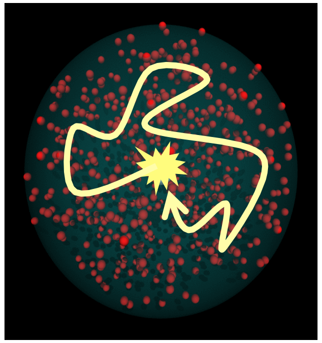
Nanophotonics - Light concentration on the nanoscale ("hot spots")



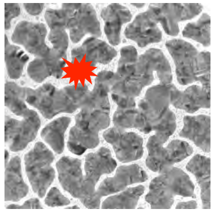
Novel light sources (e.g. random lasers)



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



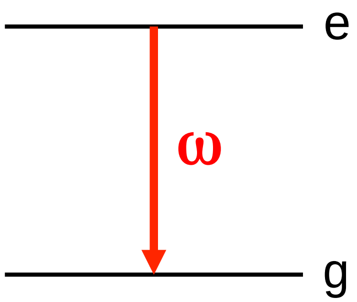
Outline



Spontaneous emission and plasmonics:
From nano-antennas to disordered systems

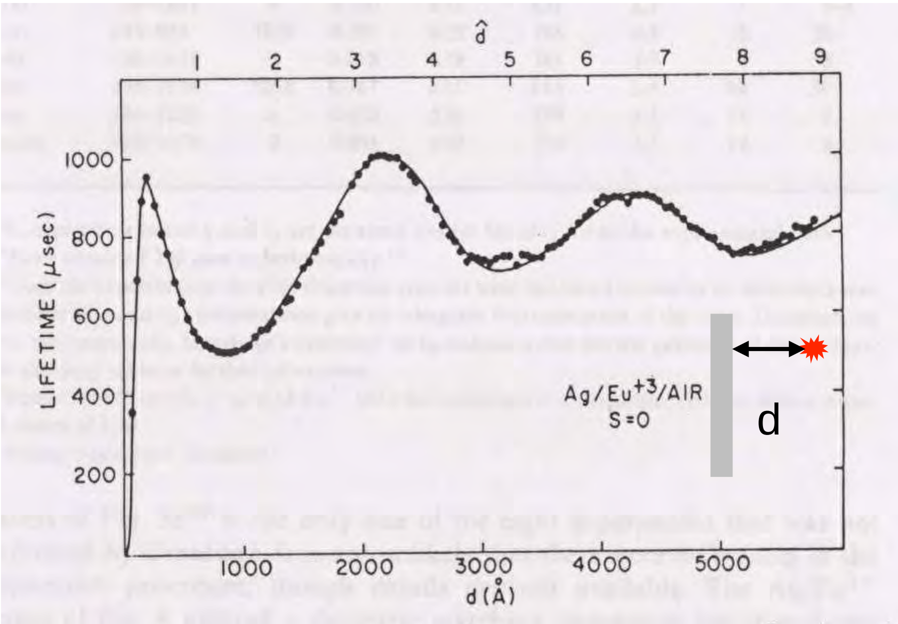
Probing near-field interactions in volume disordered systems

Spontaneous decay rate



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

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



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

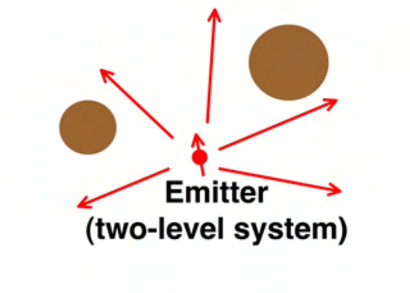
- The spontaneous decay rate depends on the environment

- Perturbation theory:

$$\Gamma = \frac{2}{\hbar}\mu_0\,\omega_{ge}^2\,|\mathbf{p}_{ge}|^2\,\mathrm{Im}\left[\mathbf{u}\cdot\mathbf{G}(\mathbf{r}_0,\mathbf{r}_0,\omega_{ge})\mathbf{u}\right]$$

Wiley and Sipe, Phys. Rev. A 30, 1185 (1984)

Decay rate and LDOS



$$\Gamma = \frac{2}{\hbar}\mu_0\,\omega^2 \left|\mathbf{p}_{ge}\right|^2 \mathrm{Im}\left[\mathbf{u}\cdot\mathbf{G}\left(\mathbf{r}_0,\mathbf{r}_0,\omega\right)\mathbf{u}\right]$$

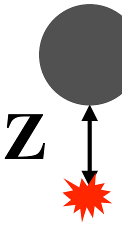
is also very often written as
(Fermi golden rule)

$$\Gamma = \frac{\pi\omega}{3\varepsilon_0\hbar}\left|\mathbf{p}_{ge}\right|^2 \rho_{\mathbf{u}}\left(\mathbf{r}_0,\omega\right)$$

← Local Density of States (LDOS)

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

Interaction with a single nanoparticle

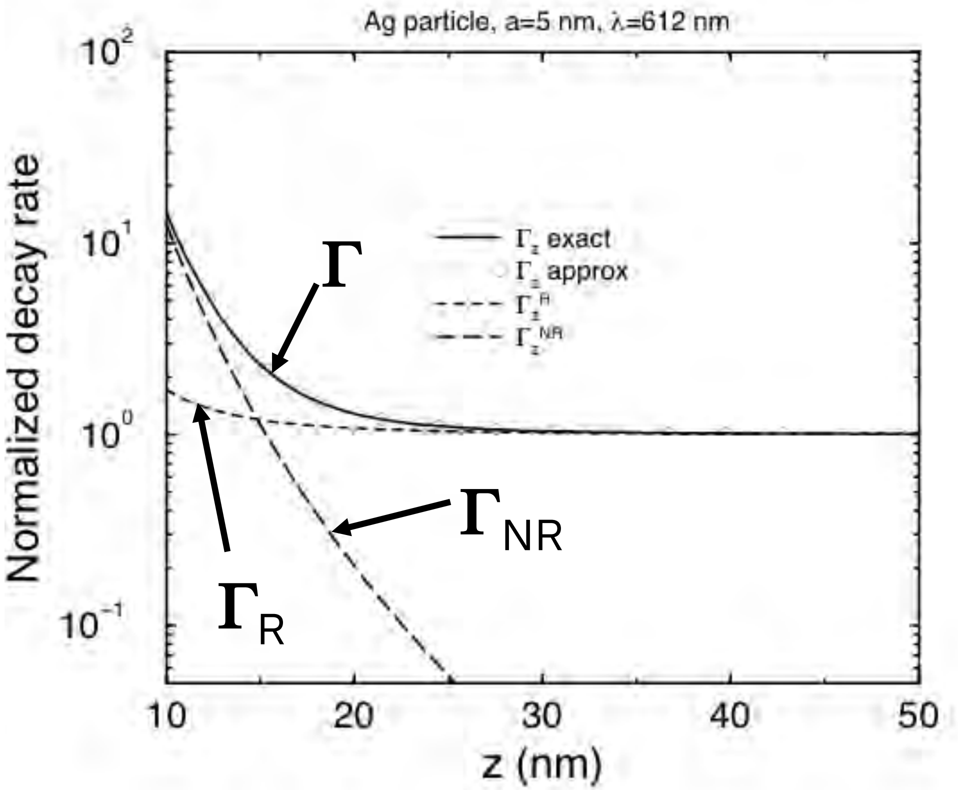


Silver nanoparticle
Diameter 10 nm

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

Photon emission (Γ_R)

Absorption (Γ_{NR})

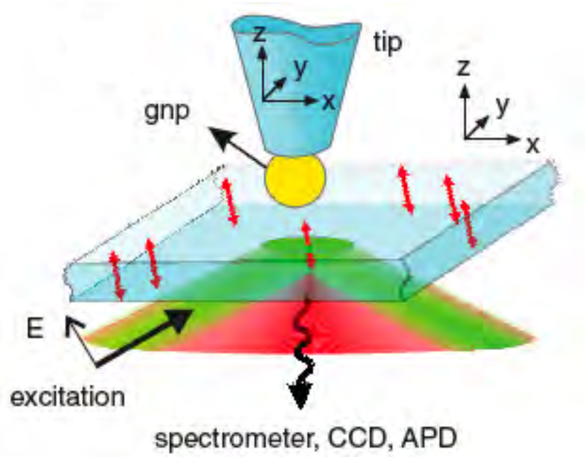


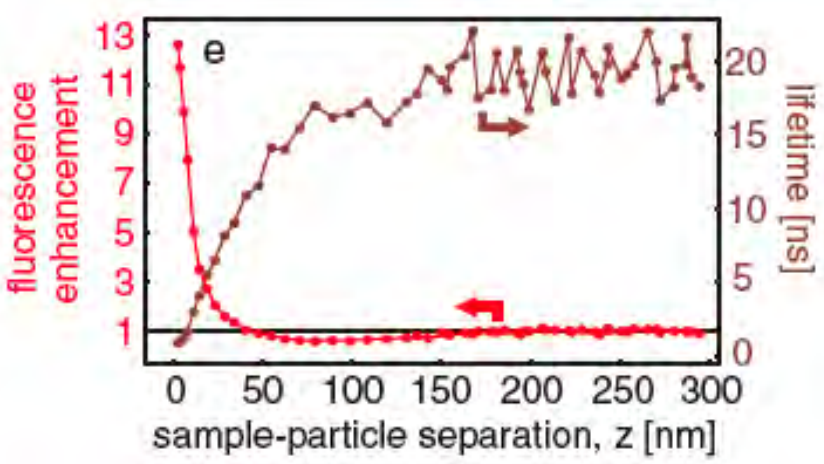
Leading contributions
at short distance

$$\Gamma_R \propto \frac{1}{(kz)^3}$$

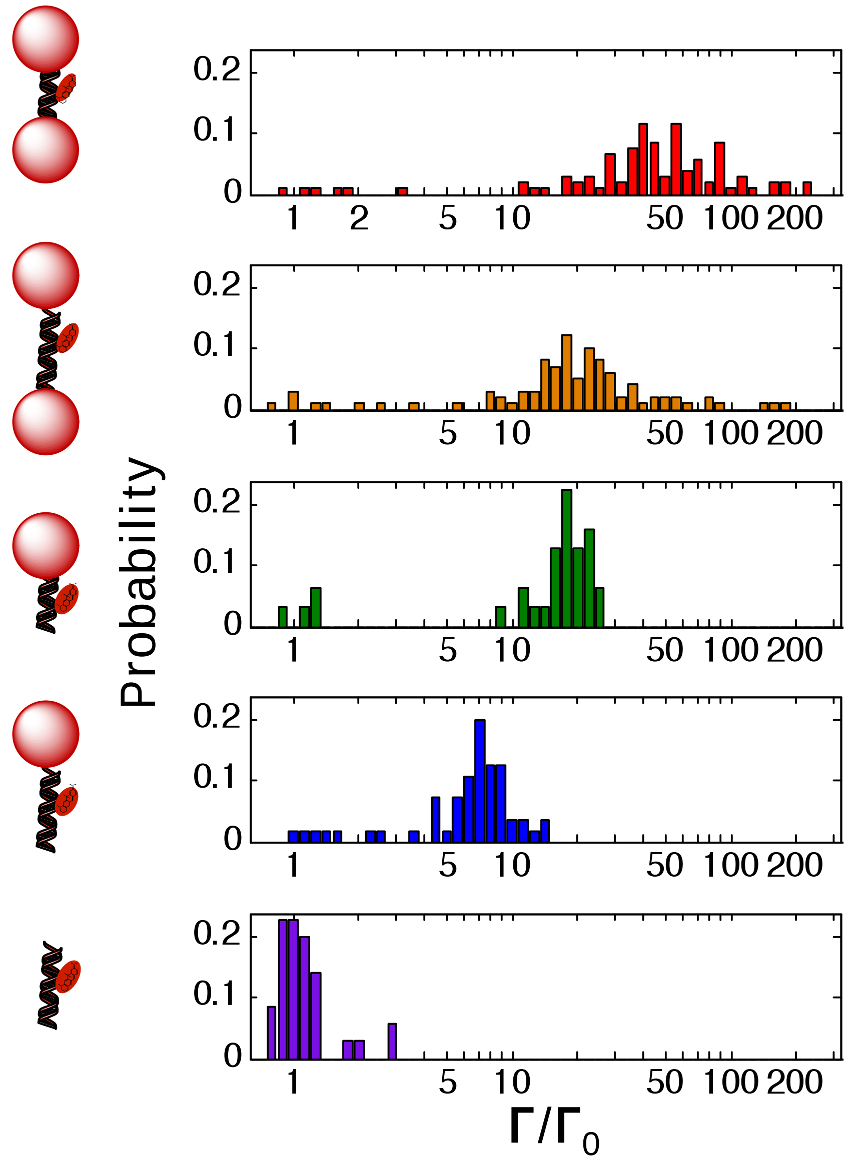
$$\Gamma_{NR} \propto \frac{1}{(kz)^6}$$

Nanoscale controlled experiments on single emitter





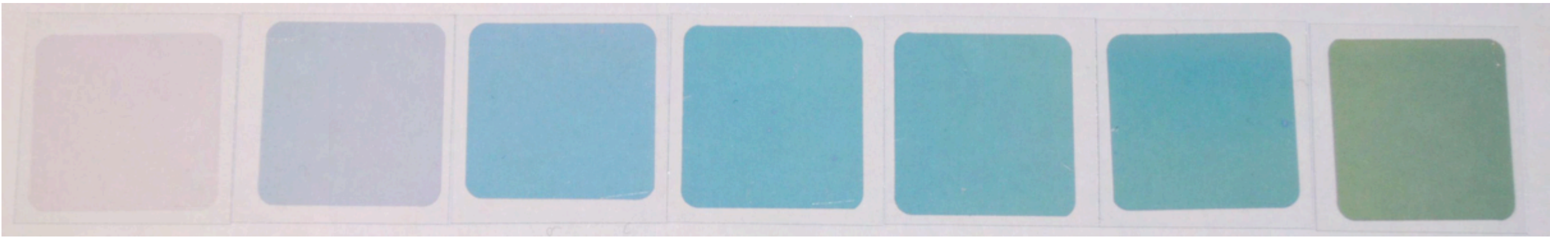
S. Kühn *et al.*, PRL 97, 017402 (2006)



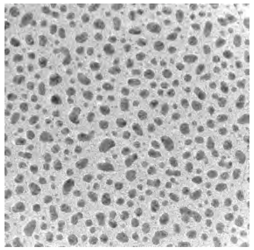
M. Busson, S. Bidault *et al.* (2011)

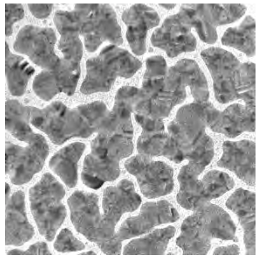
Peculiar optical properties of disordered metal films

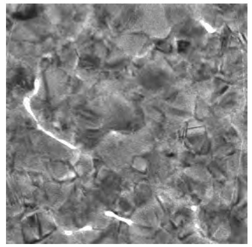
Semi-continuous gold films on a glass substrate



30% 49% 67% 79% 82% 89% 99% Filling fraction





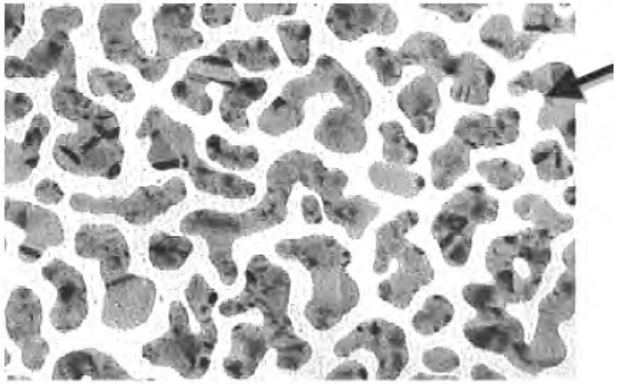


TEM images

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

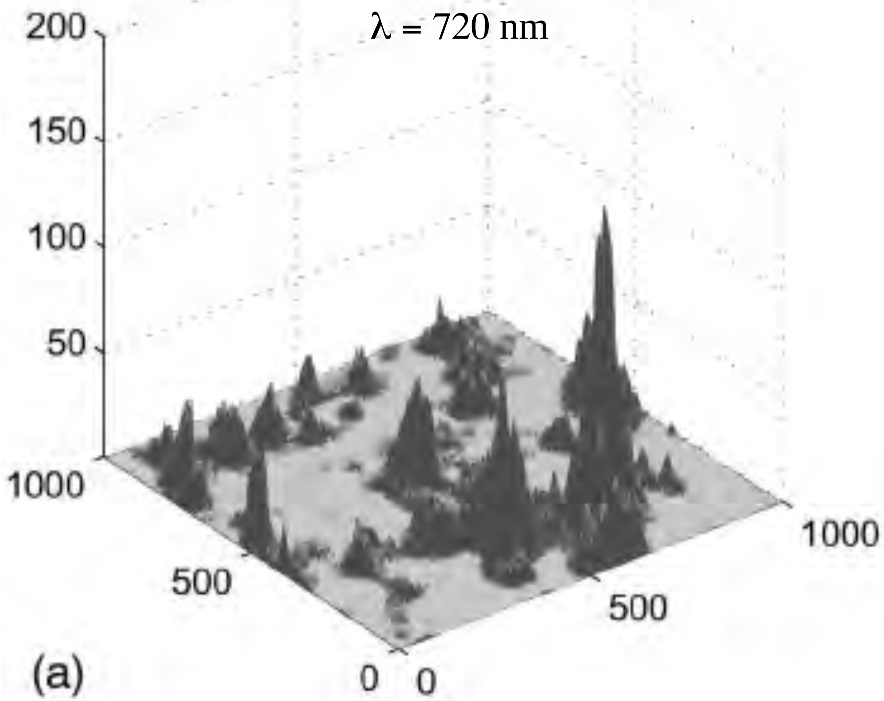
V.M. Shalaev, *Nonlinear Optics of Random Media* (Springer, 2000)

Near-field intensity distribution – « hot spots »



Surface (TEM image)
Gold on glass substrate

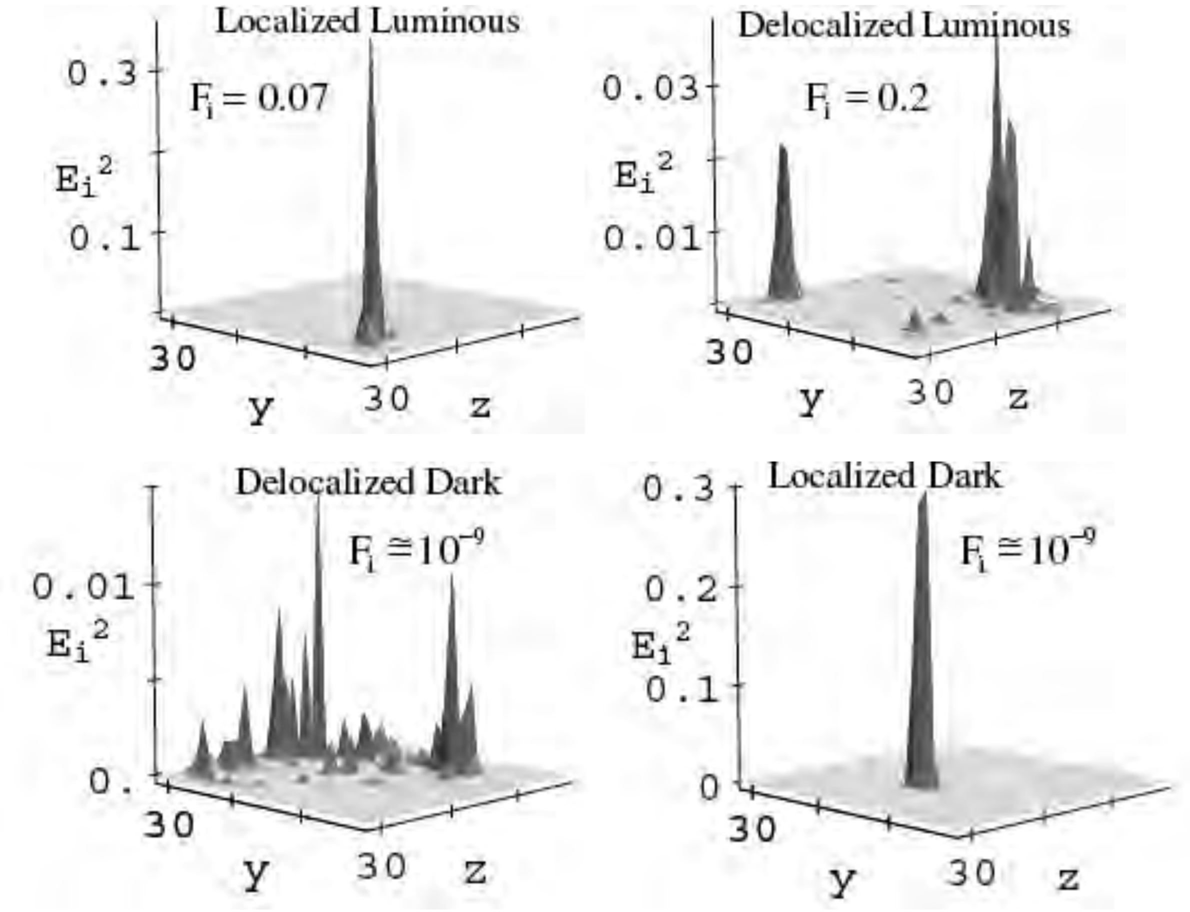
Near-field intensity (SNOM)



Grésillon *et al.*, Phys. Rev. Lett. 85, 4520 (1999) ; Phys. Rev. B 64, 165403 (2001)

Localized and delocalized modes

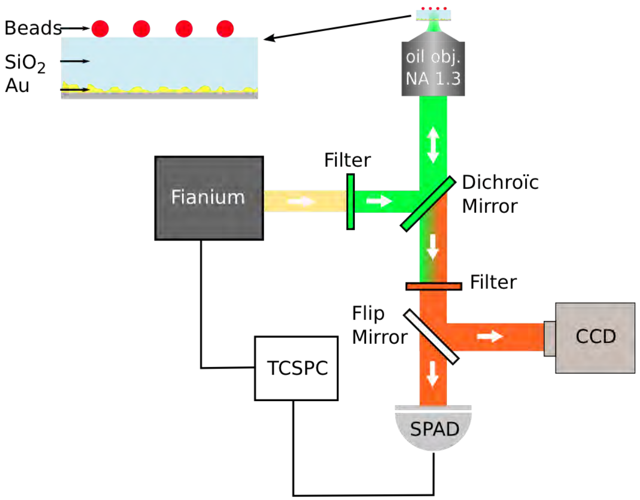
Hot-spots modes on a fractal disordered film

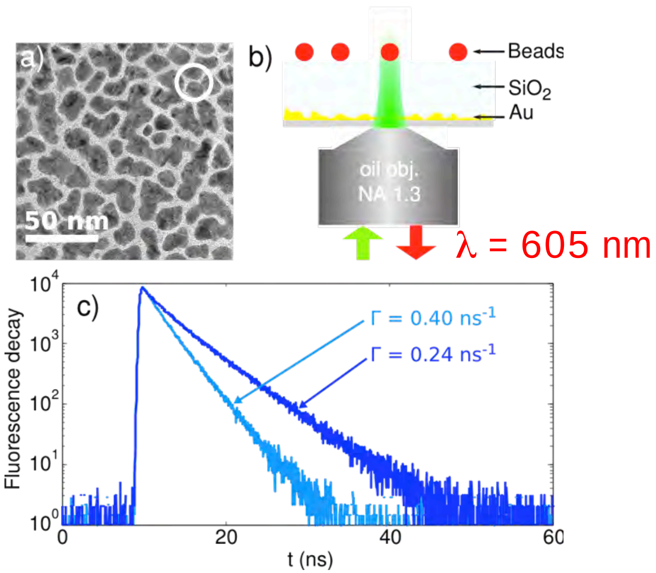


« Inhomogeneous localization »

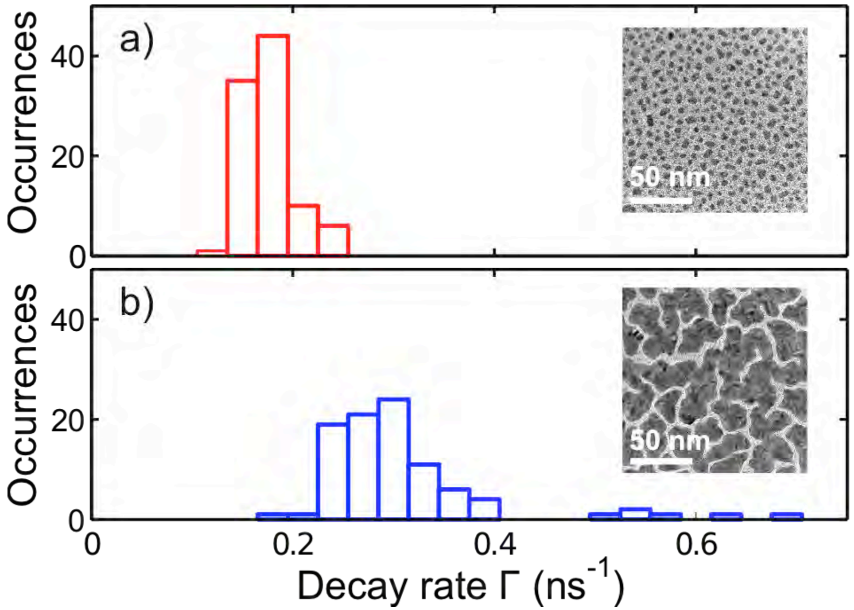
Stockman, Faleev, Bergman, PRL 87, 167401 (2001)

LDOS distributions on disordered metal films





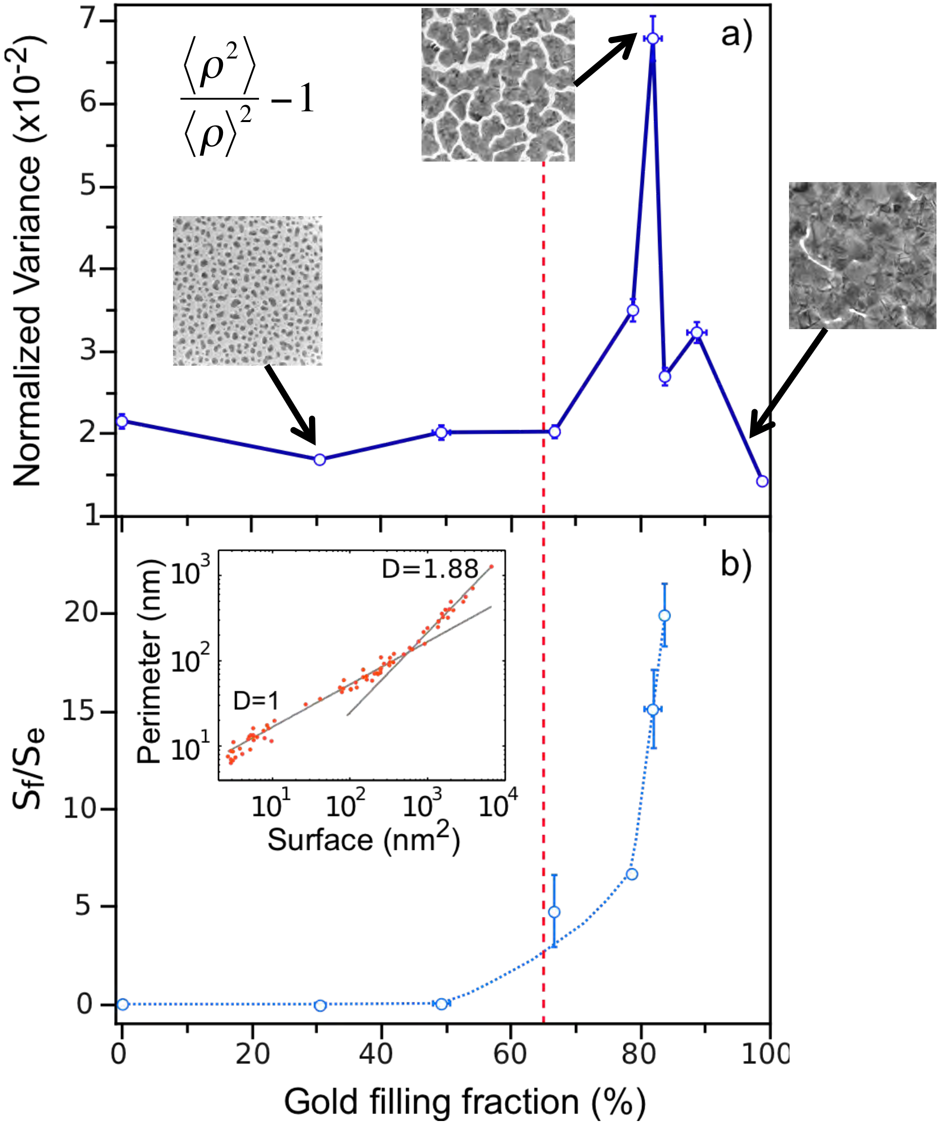
Statistical distributions of Γ (LDOS)



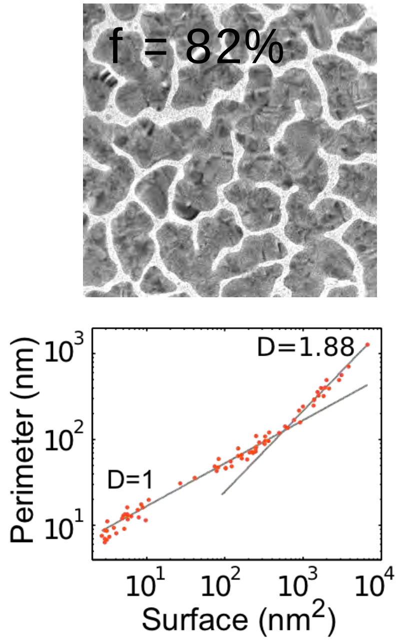
f = 30%

f = 82%

LDOS fluctuations

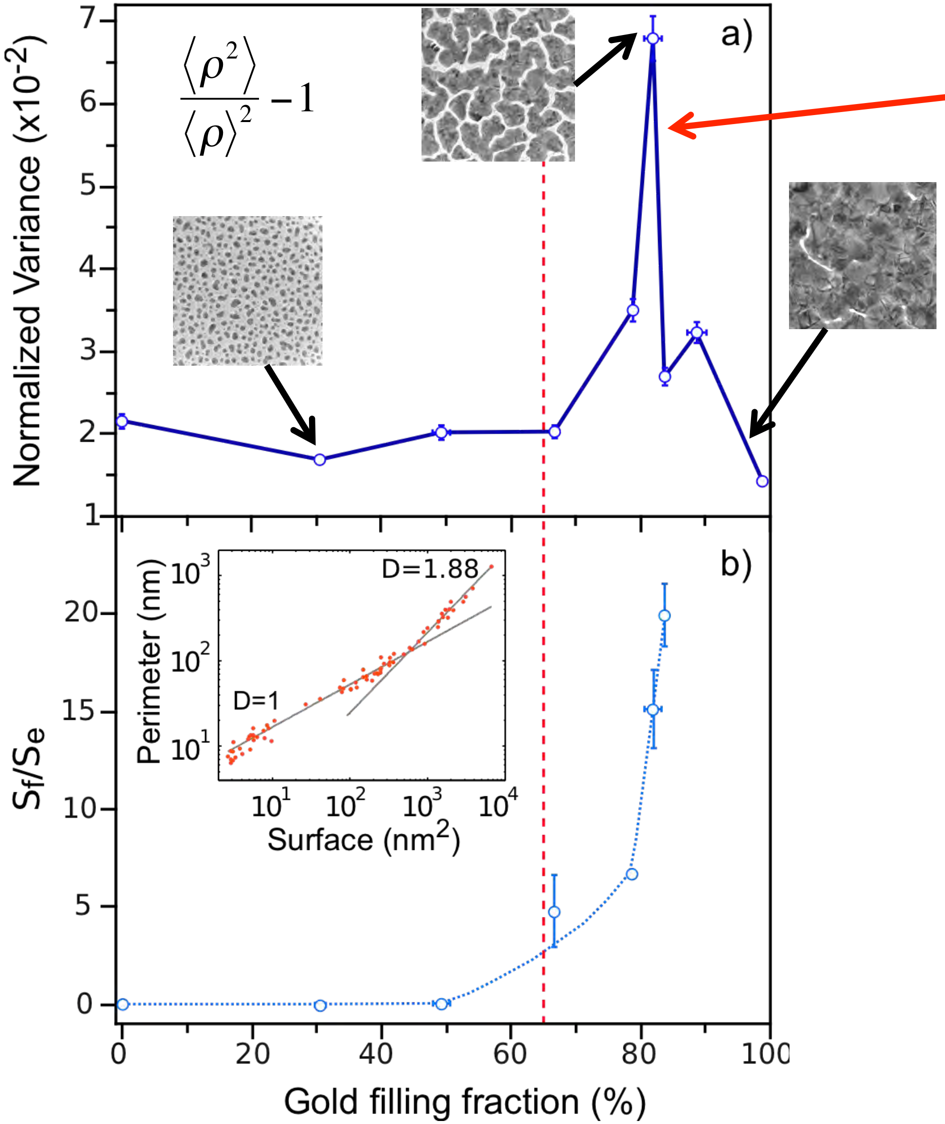


Fractal and Euclidian clusters



Krachmalnicoff, Castanié, De Wilde, Carminati, Phys. Rev. Lett. 105, 183901 (2010)

The peak reveals modes localization



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

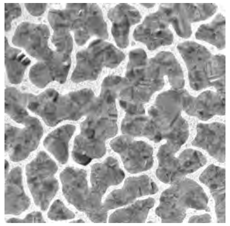
Mode localization length
(inverse participation ratio)

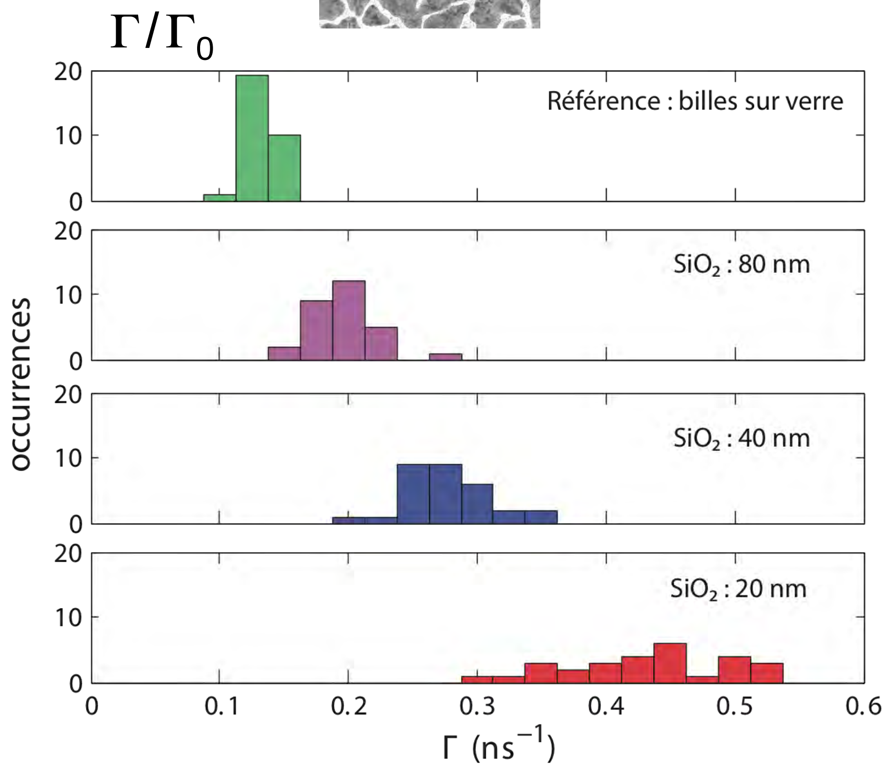
$$R_{IP} = \frac{\int |\mathbf{E}(\mathbf{r})|^4 \, d^2r}{\left[\int |\mathbf{E}(\mathbf{r})|^2 \, d^2r\right]^2} \approx \frac{1}{\xi^2}$$

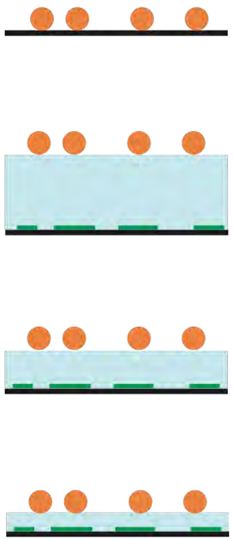
$$R_{IP} \approx \frac{1}{S} \frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2}$$

$$\frac{1}{S} \frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} \approx \frac{1}{\xi^2}$$

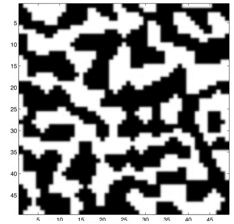
Experiment

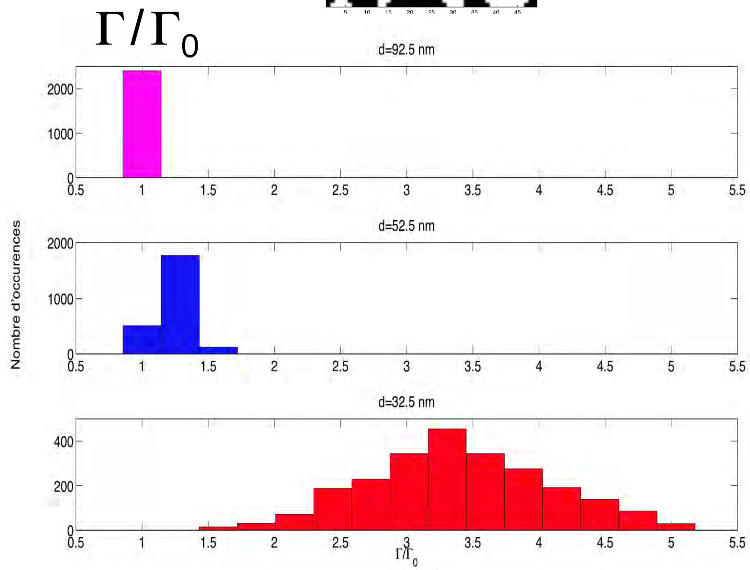




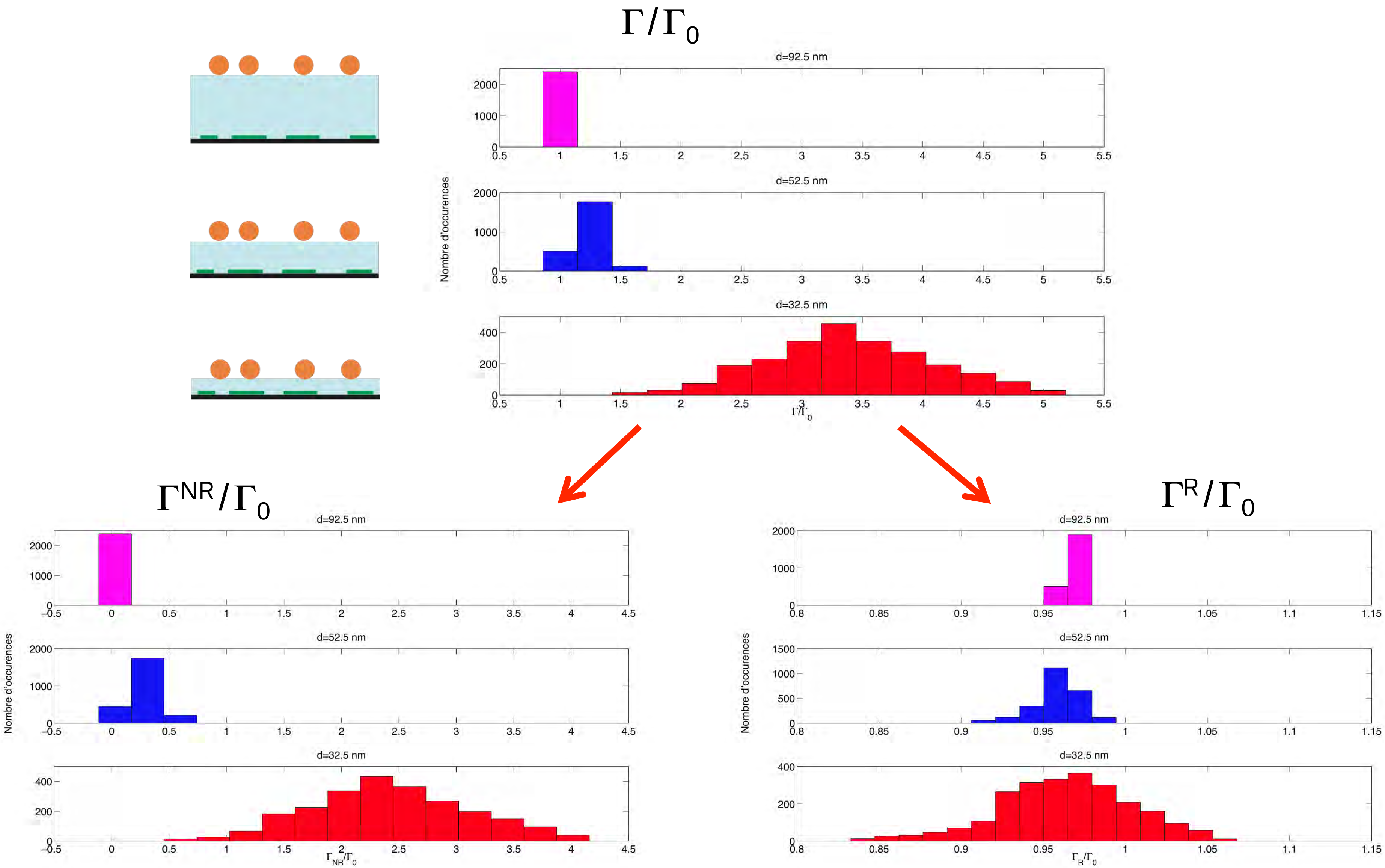


Numerical simulations
(volume integral equation + moment method)



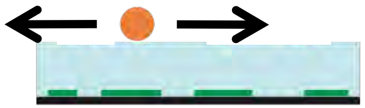


Radiative and non-radiative decays can be separated

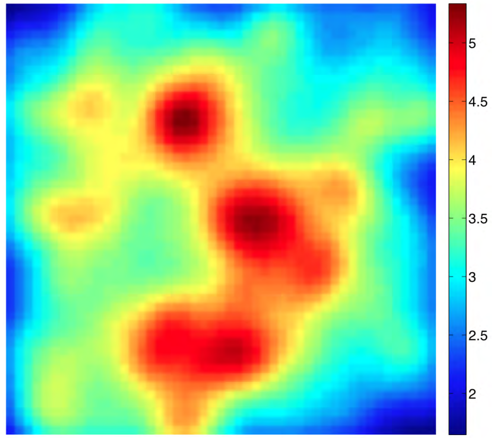


Castanié, Krachmalnicoff, Cazé, Pierrat, De Wilde, Carminati (2011)

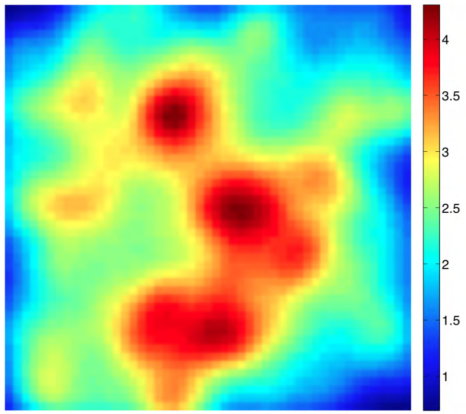
Mapping radiative and non-radiative contributions



Γ/Γ_0

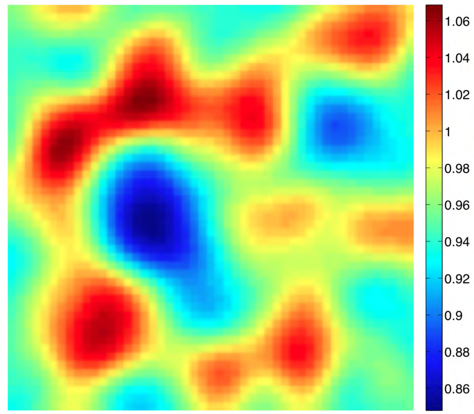


Γ^{NR}/Γ_0



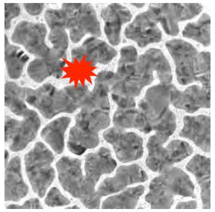
Non-radiative modes

Γ^{R}/Γ_0

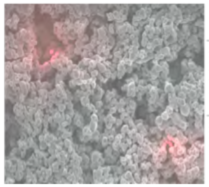


Radiative modes

Castanié, Krachmalnicoff, Cazé, Pierrat, De Wilde, Carminati (2011)



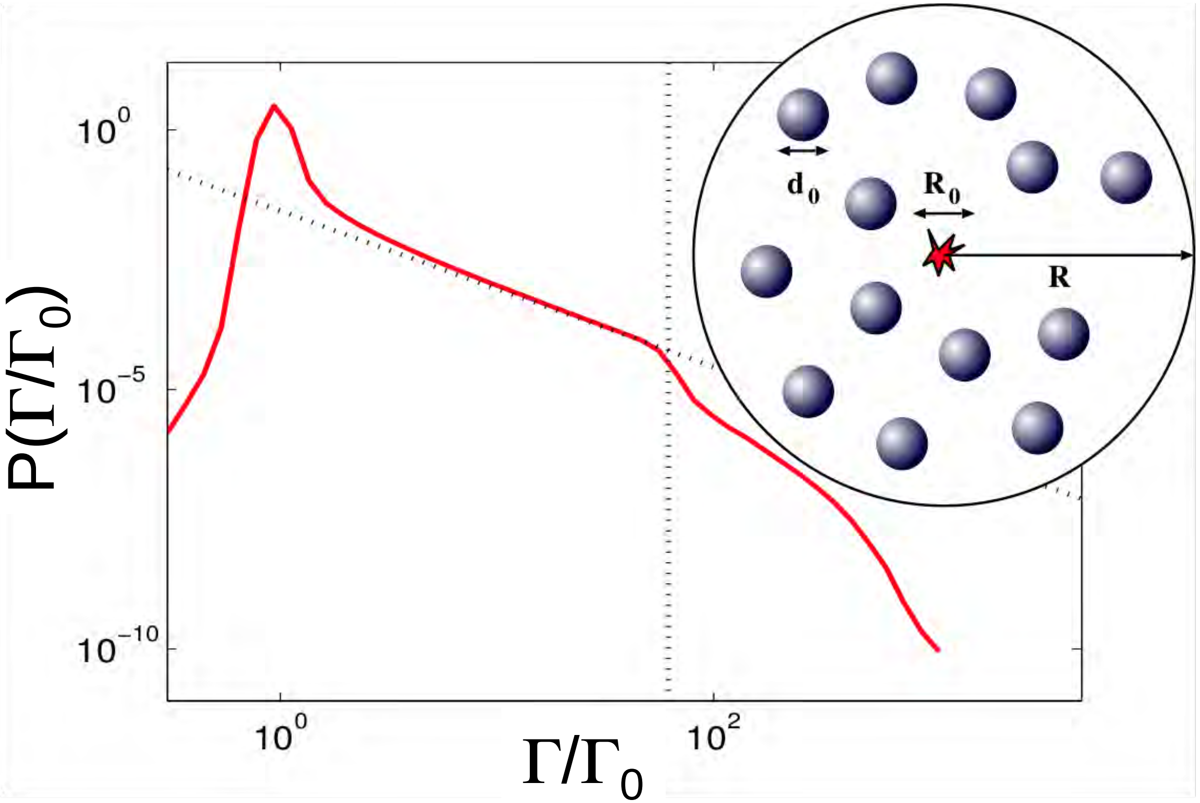
Spontaneous emission and plasmonics:
From nano-antennas to disordered systems



Probing near-field interactions in volume disordered systems

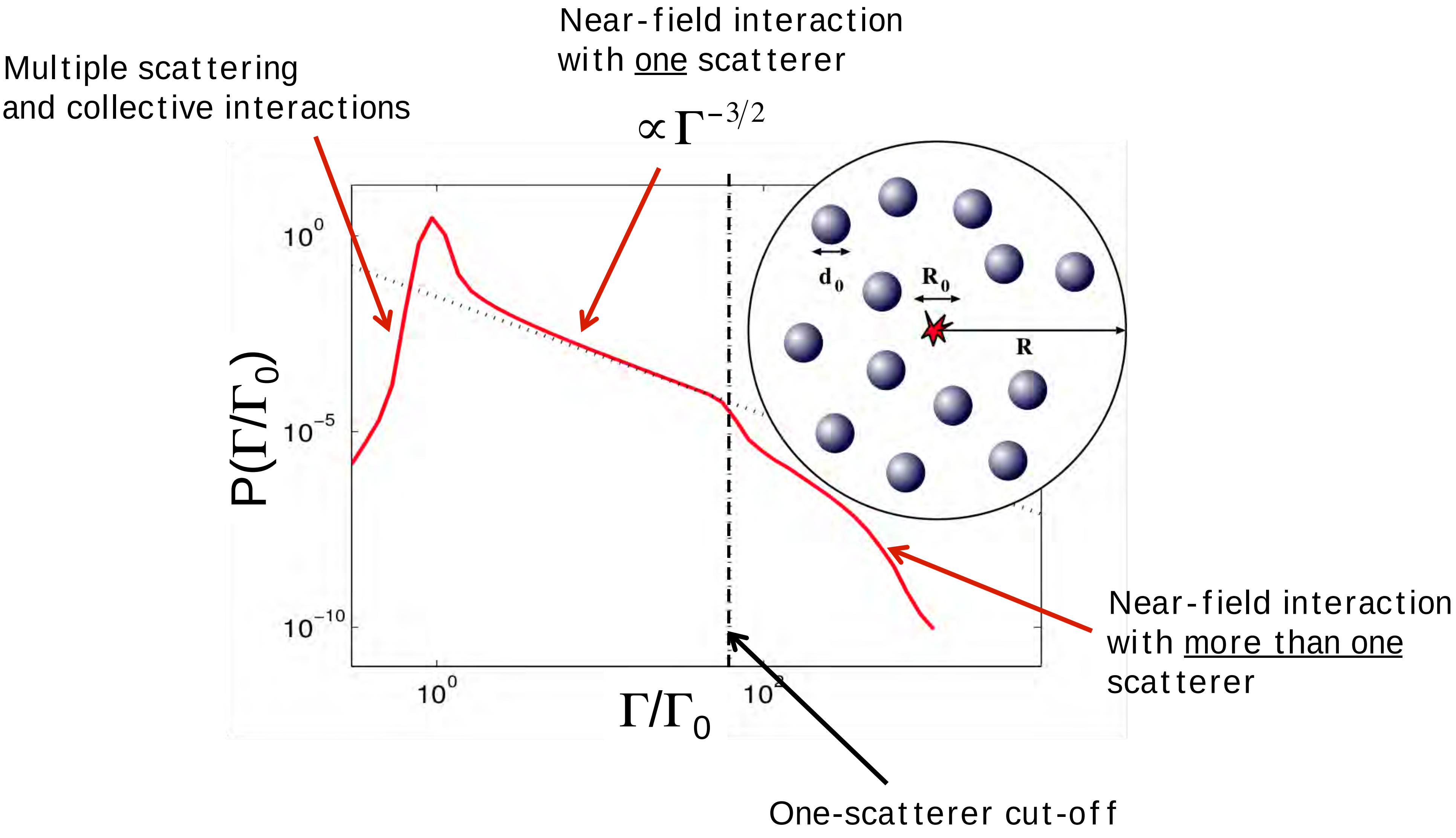
LDOS statistics from « numerical experiments »

Statistical distribution of decay rate Γ (LDOS)



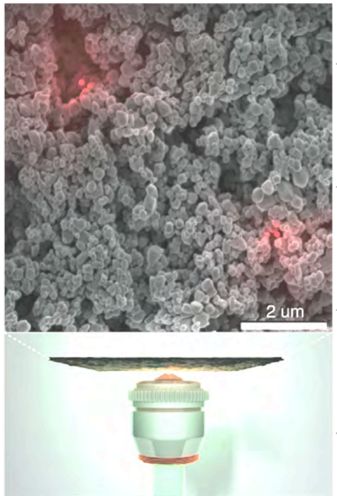
- Resonant point scatterers (« atoms »)
- $\lambda \approx 630$ nm
- Cluster size R = 1.2 μm
- Exclusion volume R_0 = 50 nm

Long tail: Near-field interactions



Broad - asymmetric distribution of decay rates (LDOS)

Experiments: Sapienza, Bondareff, Habert, van Hulst, ICFO (Barcelona, Spain)



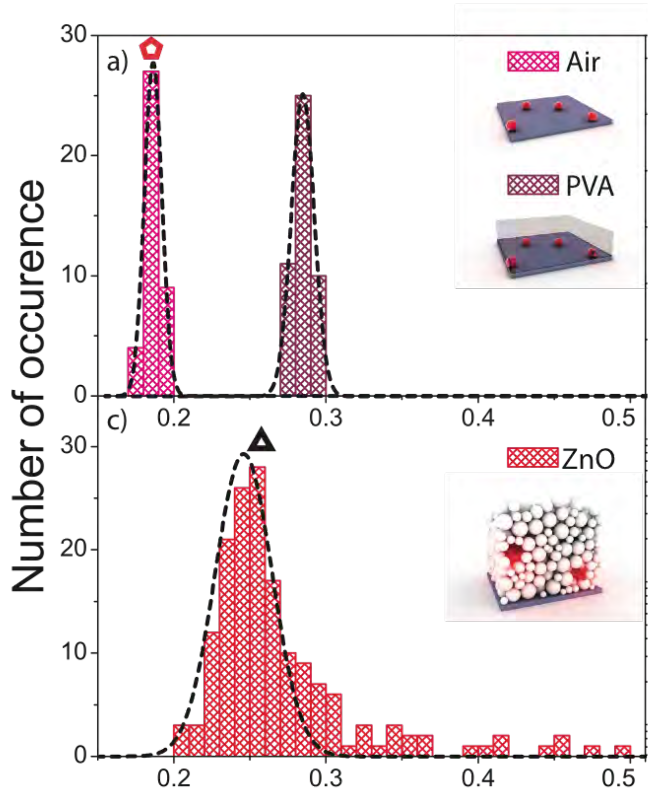
ZnO powder
Polydisperse particles
$(140 \pm 50\ nm)$

Photon mean free path

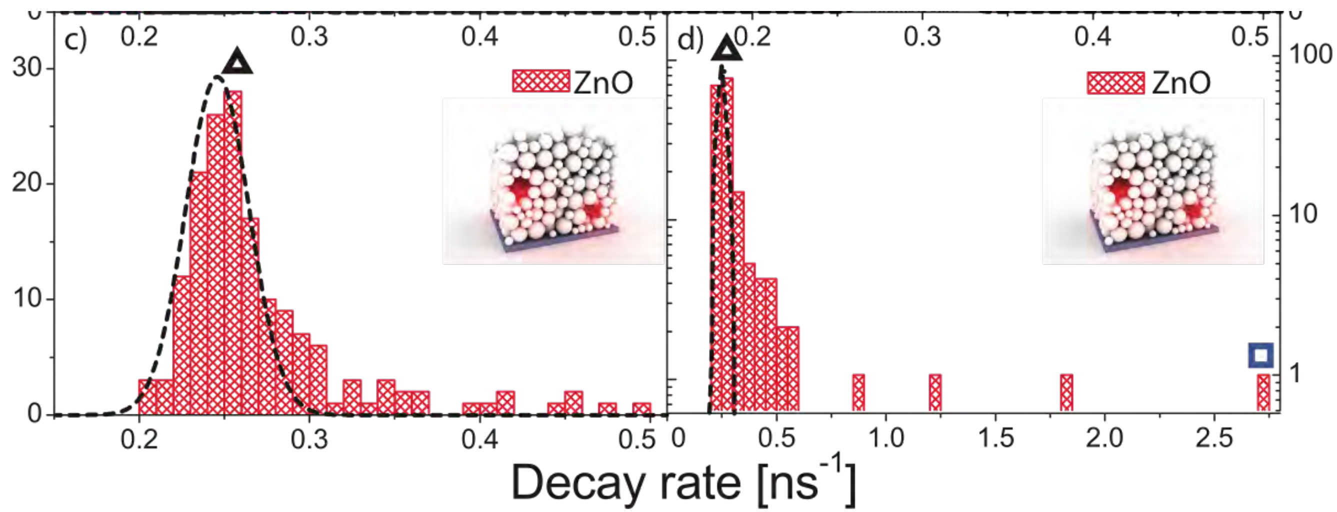
$$\ell = 0.9\ \mu m$$

$$k\ell = 9.4$$

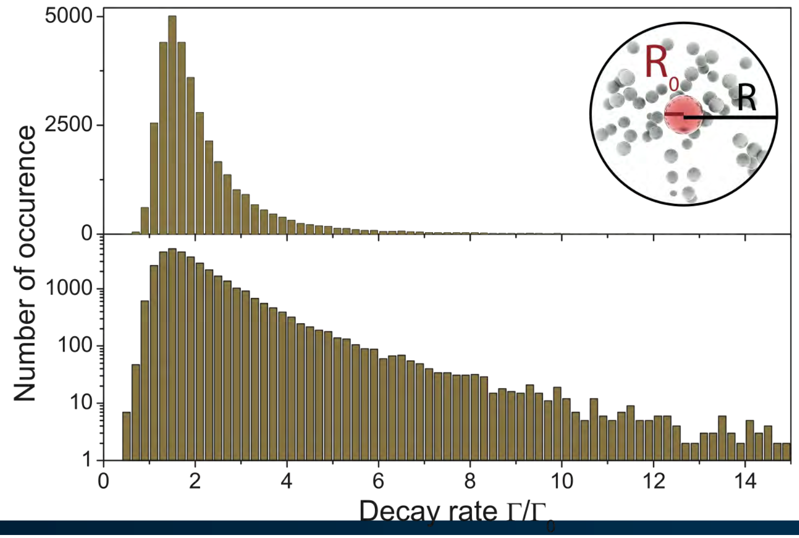
LDOS statistics probed by lifetime of nanosources (24 nm fluorescent beads)



Long tail controlled by near-field interactions



Theory



- Tail results from near-field interactions
- High Purcell factors (rare events)

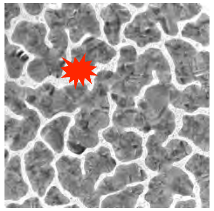
$$\frac{\Gamma_{max}}{\Gamma_{peak}} \approx 9 \qquad \frac{\Gamma_{max}}{\Gamma_0} \approx 15$$

Sapienza, Bondareff, Habert, Pierrat, Carminati, van Hulst, PRL 106, 163902 (2011)

Summary

- Photonic modes in complex systems can be probed with LDOS statistics

  Evidence of spatially localized modes
  Radiative versus non-radiative decay



- Disordered photonic materials can lead to substantial modifications of spontaneous emission

  Rare events can produce substantial changes
  Sensitive probe of nanoscale environment