Probing confined photons in nanoscale disordered media from inside

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People involved

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Collaborations

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(ESPCI)

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Niek van HULST
(ICFO Barcelona, Spain)
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 $t$: $P(t) \propto \exp(-\Gamma t)$
- Lifetime of excited state: $\tau = 1/\Gamma$

- The spontaneous decay rate depends on the environment
- Perturbation theory:

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

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

Decay rate and LDOS

\[ \Gamma = \frac{2}{\hbar} \mu_0 \omega^2 |p_{ge}|^2 \text{Im}[u \cdot G(r_0, r_0, \omega) u] \]

is also very often written as (Fermi golden rule)

\[ \Gamma = \frac{\pi \omega}{3 \varepsilon_0 \hbar} |p_{ge}|^2 \rho_u(r_0, \omega) \]

\[ \frac{\Gamma}{\Gamma_0} = \text{change in the LDOS} \]
Interaction with a single nanoparticle

Silver nanoparticle
Diameter 10 nm

\[ \Gamma = \Gamma_R + \Gamma_{NR} \]

Photon emission
Absorption

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

<table>
<thead>
<tr>
<th>Filling fraction</th>
<th>TEM images</th>
</tr>
</thead>
<tbody>
<tr>
<td>30%</td>
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<tr>
<td>99%</td>
<td>![TEM image]</td>
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</tbody>
</table>


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)

$\lambda = 720 \text{ nm}$

Localized and delocalized modes

Hot-spots modes on a fractal disordered film

Localized Luminous

Delocalized Luminous

Delocalized Dark

Localized Dark

« Inhomogeneous localization »

Stockman, Faleev, Bergman, PRL 87, 167401 (2001)
LDOS distributions on disordered metal films

Statistical distributions of $\Gamma$ (LDOS)

- $f = 30\%$
- $f = 82\%$

$\lambda = 605\text{ nm}$
LDOS fluctuations

\[ \frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} - 1 \]

Fractal and Euclidian clusters

\[ f = 82\% \]

The peak reveals modes localization

\[ \frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} - 1 \]

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

Mode localization length (inverse participation ratio)

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

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

[1/2 \langle \rho^2 \rangle \approx \frac{1}{\xi^2}]
Numerical simulations

**Experiment**

![Image of experimental setup](image1)

<table>
<thead>
<tr>
<th>Occurrences</th>
<th>$\Gamma/\Gamma_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Référence : billes sur verre</td>
<td></td>
</tr>
<tr>
<td>$\text{SiO}_2 : 80 \text{ nm}$</td>
<td></td>
</tr>
<tr>
<td>$\text{SiO}_2 : 40 \text{ nm}$</td>
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<tr>
<td>$\text{SiO}_2 : 20 \text{ nm}$</td>
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</tbody>
</table>

**Numerical simulations**

(volume integral equation + moment method)

![Image of numerical simulation results](image2)

Castanié, Krachmalnicoff, Cazé, Pierrat, De Wilde, Carminati (2011)
Radiative and non-radiative decays can be separated

\[ \frac{\Gamma}{\Gamma_0} \]

\[ \frac{\Gamma_{NR}}{\Gamma_0} \]

\[ \frac{\Gamma^R}{\Gamma_0} \]

Castanié, Krachmalnicoff, Cazé, Pierrat, De Wilde, Carminati (2011)
Mapping radiative and non-radiative contributions

\[ \frac{\Gamma}{\Gamma_0} \]

\[ \frac{\Gamma_{NR}}{\Gamma_0} \]

Non-radiative modes

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 »

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

Statistical distribution of decay rate $\Gamma$ (LDOS)
Long tail: Near-field interactions

Multiple scattering and collective interactions

\[ P(\Gamma/\Gamma_0) \]

\[ \Gamma/\Gamma_0 \]

Near-field interaction with one scatterer

\[ \propto \Gamma^{-3/2} \]

Near-field interaction with more than one scatterer

One-scatterer cut-off

Broad - asymmetric distribution of decay rates (LDOS)

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

ZnO powder
Polydisperse particles
(140 ± 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)

Sapienza, Bondareff, Habert, Pierrat, Carminati, van Hulst, PRL 106, 163902 (2011)
Long tail controlled by near-field interactions

Tail results from near-field interactions

High Purcell factors (rare events)

\[
\frac{\Gamma_{\text{max}}}{\Gamma_{\text{peak}}} \approx 9 \quad \frac{\Gamma_{\text{max}}}{\Gamma_0} \approx 15
\]
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*