



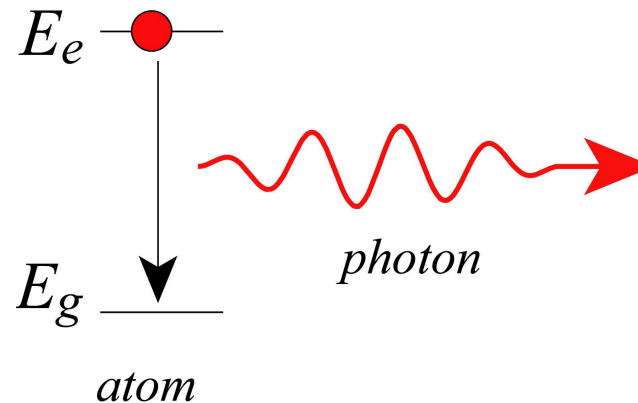
Nanophotonics

Class 4

Density of states

Outline

Spontaneous emission: an excited atom/molecule/.. decays to the ground state and emits a photon



- Emission rates are set by Fermi's Golden Rule
- Fermi's Golden Rule & the number of available photon states (LDOS)
- Experiments demonstrating emission rate control via LDOS
- Conclusion

Fermi's Golden Rule

- Consider an atom, molecule or quantum dot with eigenstates ψ .
- Suppose the system is perturbed, e.g. by incident light.
Perturbing term in hamiltonian:

$$V = \underbrace{-\boldsymbol{\mu}}_{\text{Dipole operator}} \cdot \underbrace{\mathbf{E}}_{\text{light}}$$

The coupling can take the atom in initial state ψ_i to another state ψ_f

Fermi's Golden Rule: rate of decay of the initial state ψ_i

$$\Gamma = \frac{2\pi}{\hbar^2} \sum_{\text{all final states } f} \left| \langle \psi_f | V | \psi_i \rangle \right|^2 \delta(E_f - E_i)$$

Understanding Fermi's Golden Rule

$$\Gamma = \frac{2\pi}{\hbar^2} \sum_{\text{all final states } f} \left| \langle \psi_f | V | \psi_i \rangle \right|^2 \delta(E_f - E_i)$$

Matrix elements:
Transition strength
Selection rules

Energy conservation

Spontaneous emission of a two-level atom:

Initial state: excited atom + 0 photons.

Final state: ground state atom + 1 photon in some photon state

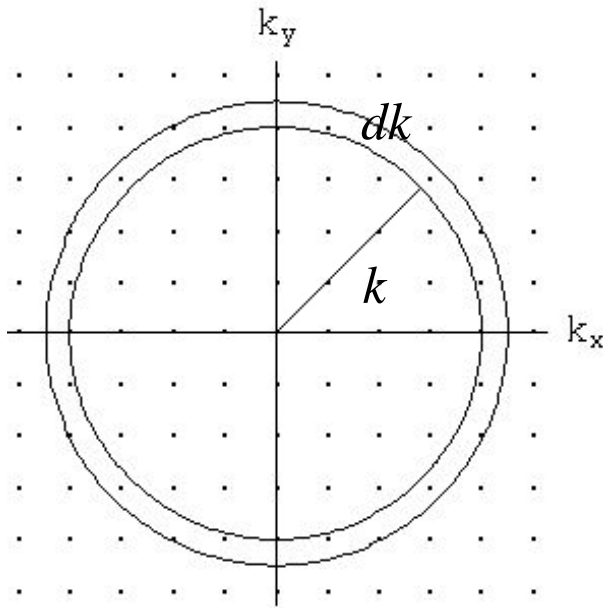
Question: how many states are there for the photon ???
(constraint: photon energy = atomic energy level difference)

How many photon states are there in a box of vacuum ?

States in an $L \times L \times L$ box:

$$E(x,t) = Ae^{i\omega t} \sin(\mathbf{k} \cdot \mathbf{r}) \quad \text{with} \quad \mathbf{k} = \frac{\pi}{L}(l,m,n)$$

l,m,n positive integers



Number of states with $|\mathbf{k}|$ between k and $k+dk$:

$$N(k)dk = \frac{4}{8} \pi k^2 dk \left(\frac{L}{\pi} \right)^3 \cdot 2$$

$l,m,n > 0$ fill one octant

fudge 2 for polarization

As a function of frequency ω ($=ck$):

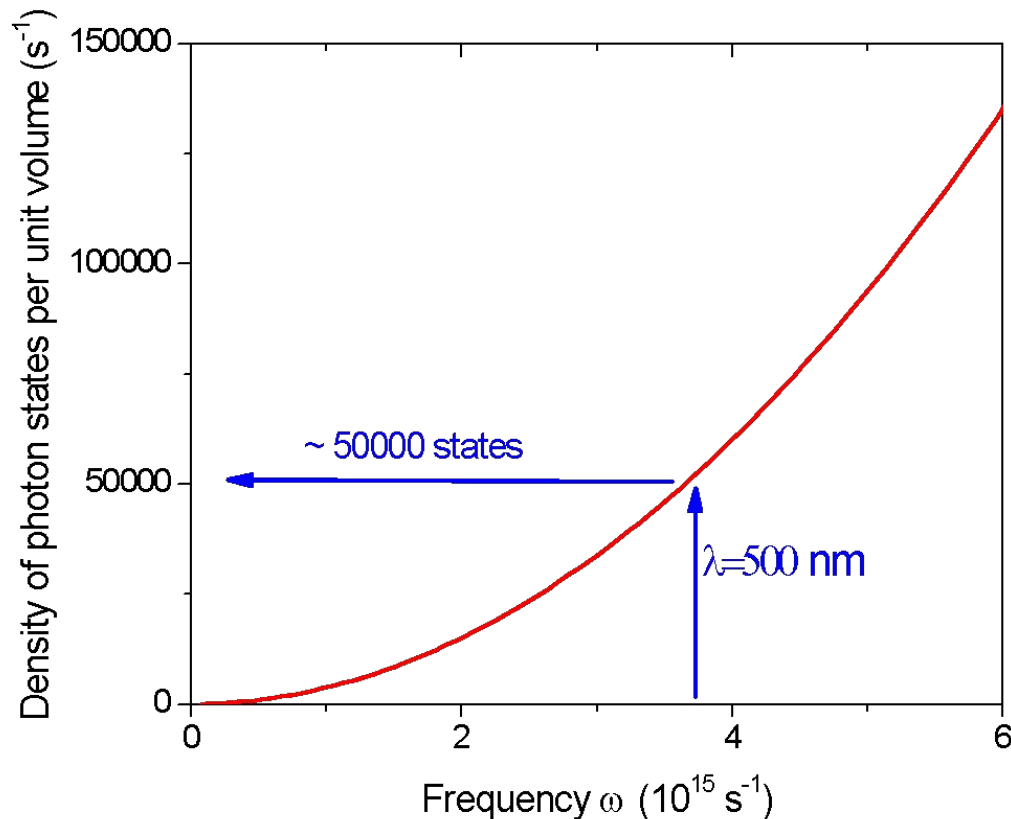
$$N(\omega)d\omega = L^3 \frac{\omega^2}{\pi^2 c^2} \frac{dk}{d\omega} d\omega = L^3 \frac{\omega^2}{\pi^2 c^3} d\omega$$

Picture from
<http://britneyspears.ac>

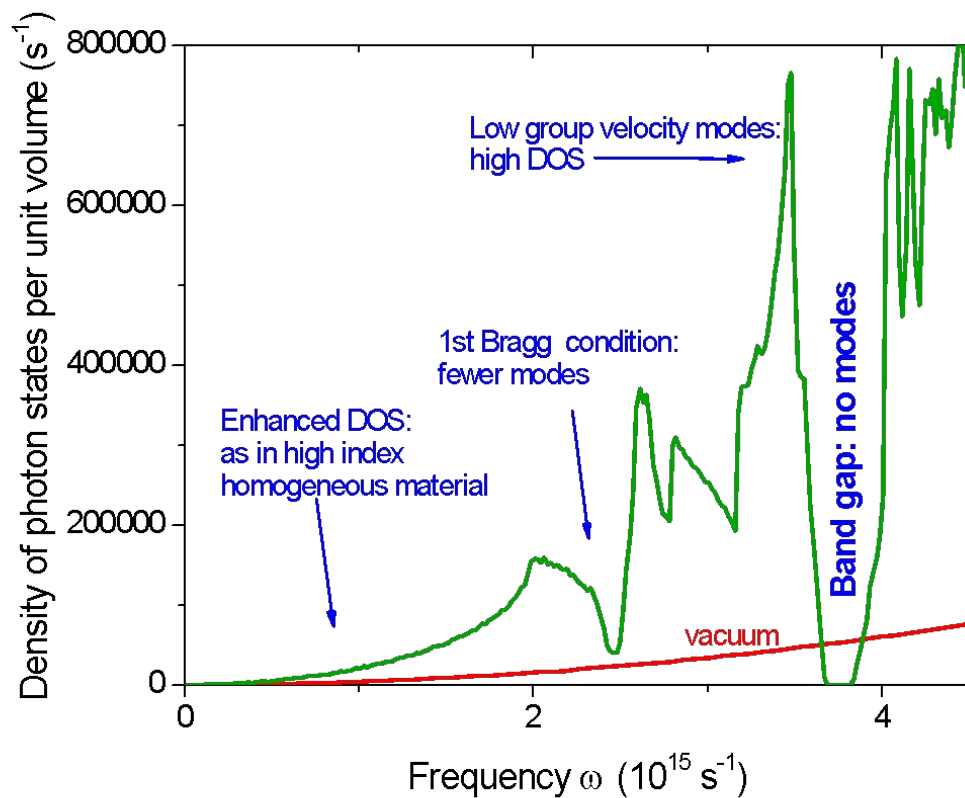
Density of states in vacuum

$$N(\omega)d\omega = L^3 \frac{\omega^2}{\pi^2 c^2} \frac{dk}{d\omega} d\omega = L^3 \frac{\omega^2}{\pi^2 c^3} d\omega$$

Example: ~50000 photon states per m³ of vacuum per 1 Hz @ $\lambda=500$ nm

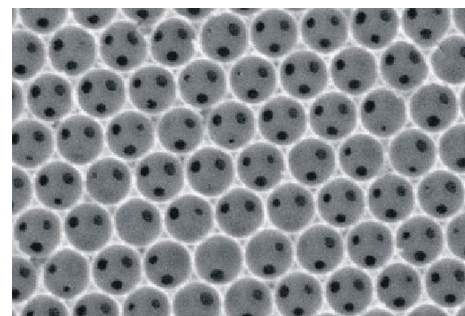


Controlling the DOS



Photonic band gap material

Example:
fcc close-packed
air spheres in $n=3.5$
Lattice spacing 400 nm



Photonic band gap: no states = no spontaneous emission

Enhanced DOS: faster spontaneous emission according to Fermi G. Rule

Local DOS

An emitter doesn't just count modes (as in DOS)

It also feels *local mode strength* $|E|^2$.

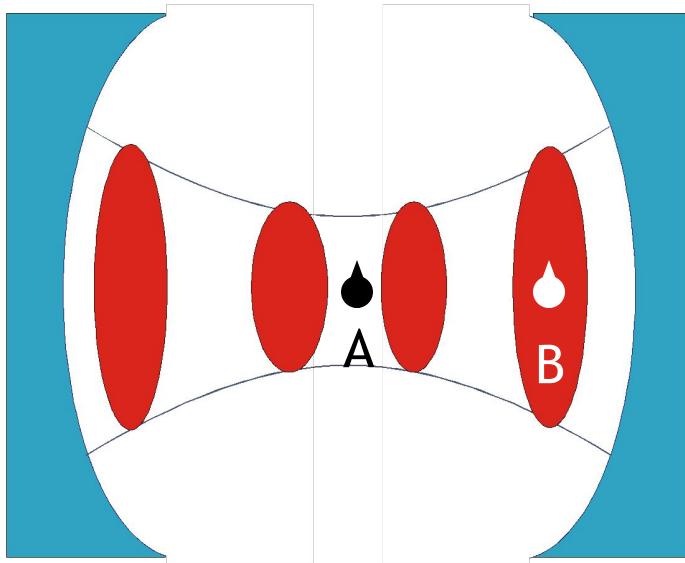
It can only emit into a mode if the mode is not zero at the emitter

DOS: just count states

$$N(\omega) = \sum_{\text{all modes } m} \delta(\omega_m - \omega)$$

Local DOS

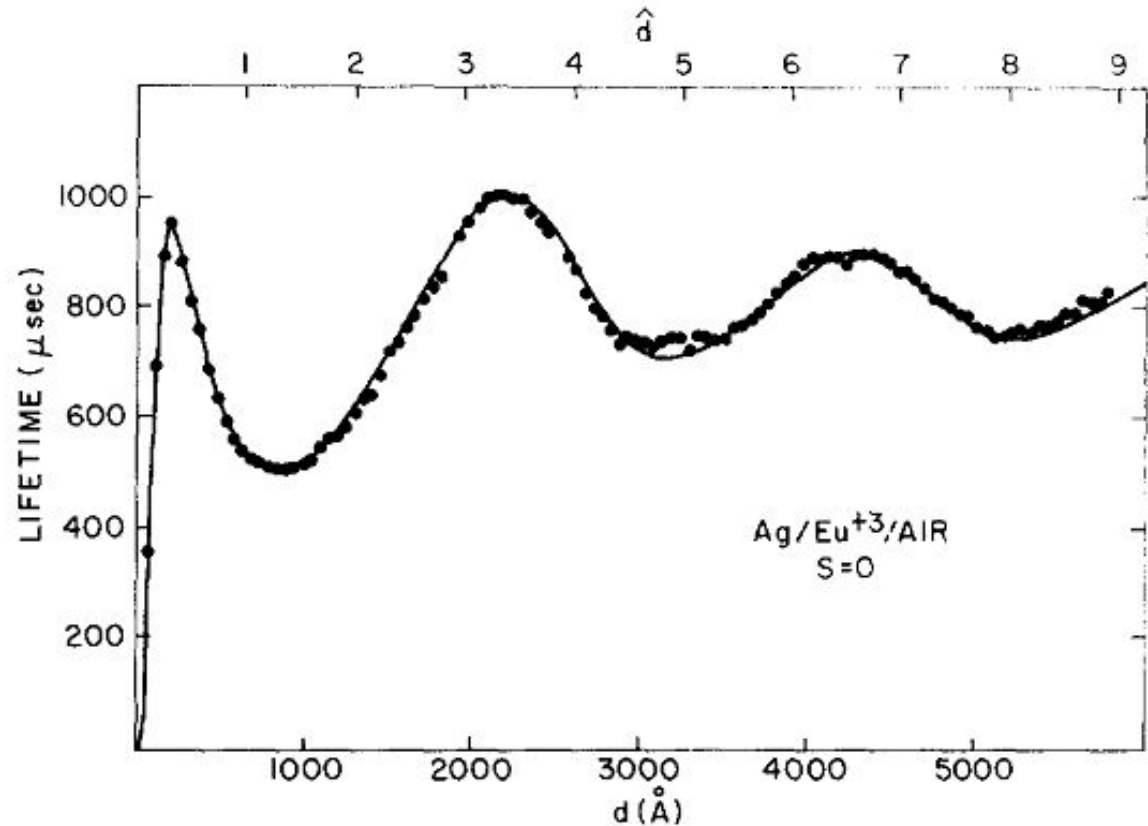
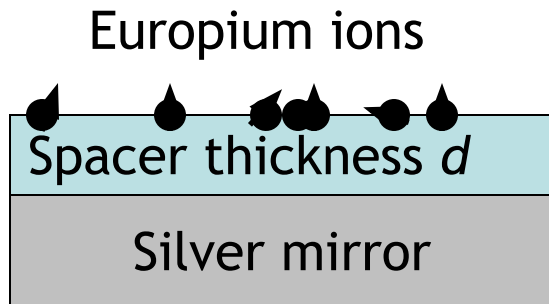
$$N(\mathbf{r}, \mathbf{d}, \omega) = \sum_{\text{all modes}} |\mathbf{d} \cdot \mathbf{E}_m(\mathbf{r})|^2 \delta(\omega_m - \omega)$$



Atom at position A can not emit into cavity mode.

Atom at position B can emit into cavity mode.

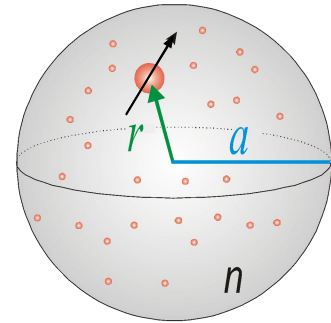
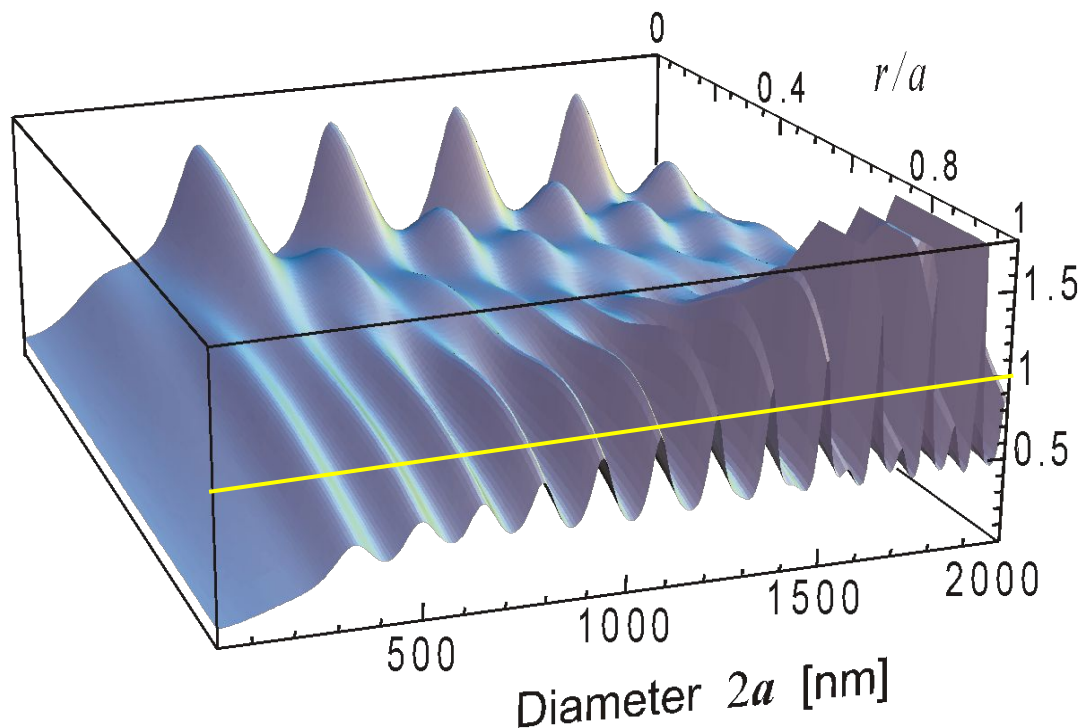
LDOS: emission in front of a mirror



Drexhage (1966): fluorescence lifetime of Europium ions depends on source position relative to a silver mirror ($\lambda=612 \text{ nm}$)

Example II: dielectric nano-sphere

Eu ions in 100 nm - 1 μm polystyrene spheres [1]
Er ions in 340 nm SiO₂ spheres [2]

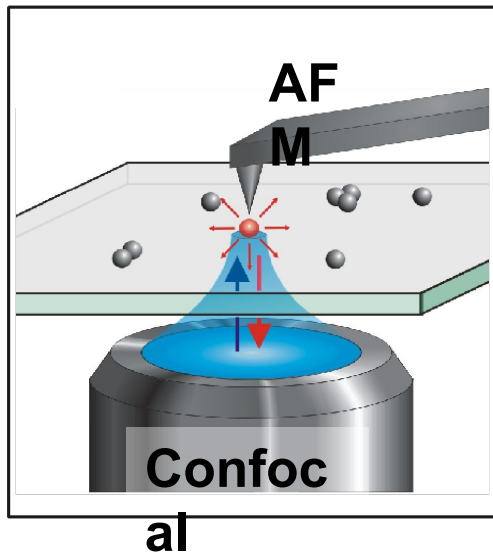


LDOS
normalized to
LDOS in SiO₂

[1] Schniepp & Sandoghdar, Phys. Rev. Lett **89** (2002)

[2] de Dood, Slooff, Polman, Moroz & van Blaaderen, Phys. Rev. A **64** (2001)

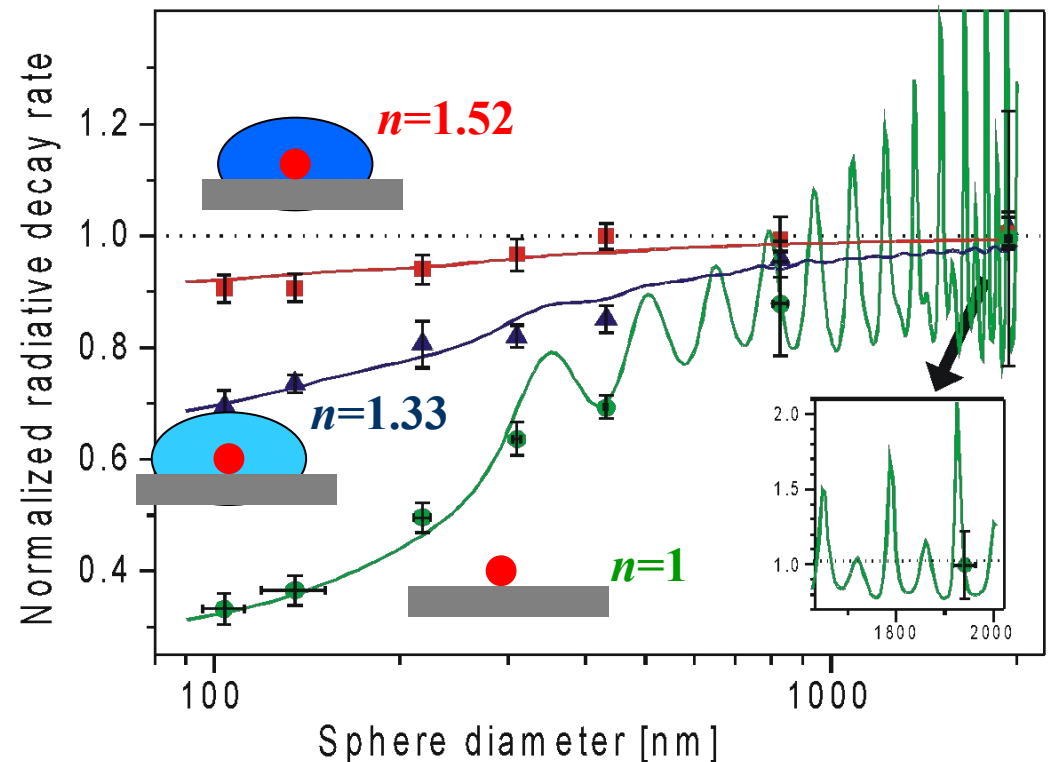
Dielectric nanosphere



AFM to check individual particle diameters
Confocal microscopy to collect luminescence

Index matching of sphere
with fluid droplets:

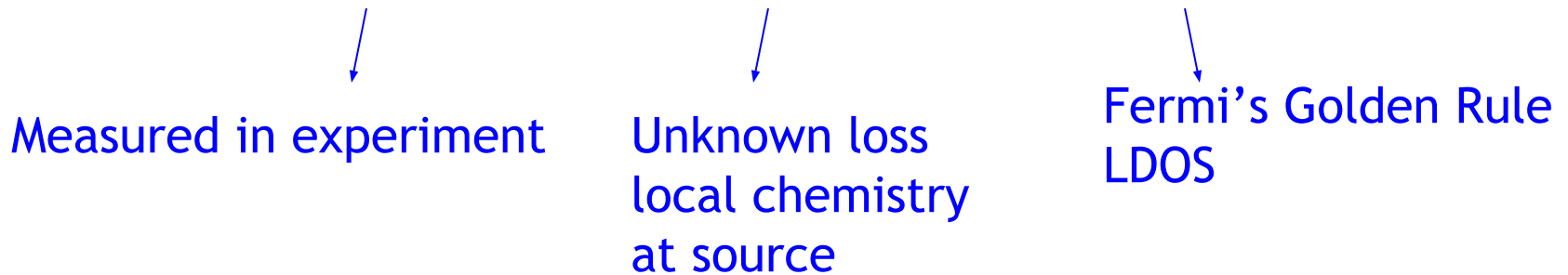
Emitter stays the same
Lifetime change disappears



LDOS & measuring nonradiative decay

A *real* emitter often also decays nonradiatively (no photons but heat)

$$\Gamma_{total} = \Gamma_{non-radiative} + \Gamma_{radiative}$$



Measurement technique: vary the nanophotonic configuration
vary LDOS and not the chemistry

Example

Emitter in sphere: index match sphere to vary $\Gamma_{radiative}$

Assignment: you can find $\Gamma_{non-radiative}$ by varying LDOS

Conclusions

- Spontaneous emission rates are controlled by nanophotonic structures
- Fermi's Golden Rule: transition rate depends on availability of final states
- Spontaneous emission: final states for photon ?
- Density of states (DOS): number of photon states depending on frequency
- Local density of states (LDOS): number of photon states available locally for spontaneous emission

Applications

- Enhance the efficiency of light sources
- Characterize non-radiative mechanisms