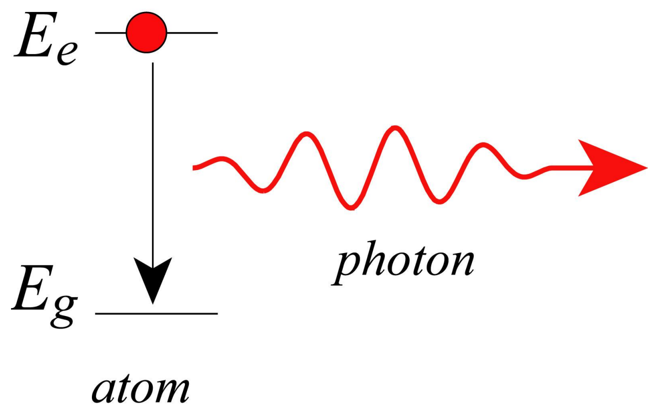
Nanophotonics

Class 4

Density of states

Outline

Spontaneous emission: an exited 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:

$$V = -\boldsymbol{\mu} \cdot \mathbf{E}$$

light

Dipole operator

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

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

Understanding Fermi's Golden Rule

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

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

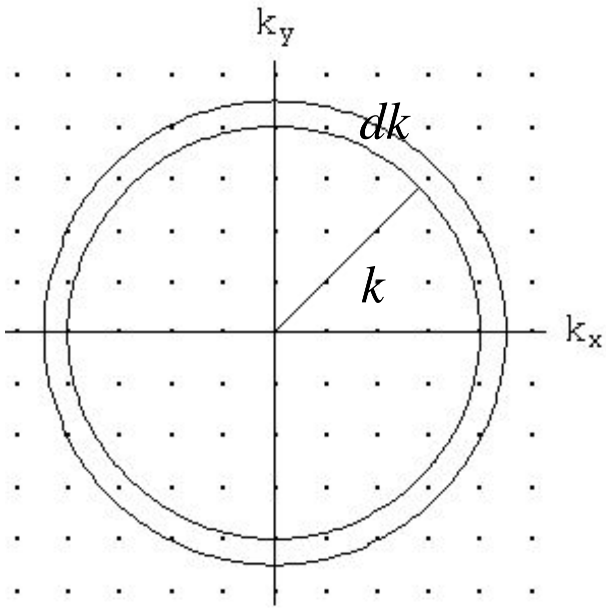
Question: how many states are there for the photon ???
(constraint: photon energy = atomic energy level difference)

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

States in an LxLxL 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



Picture from http://britneyspears.ac

Number of states with |**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

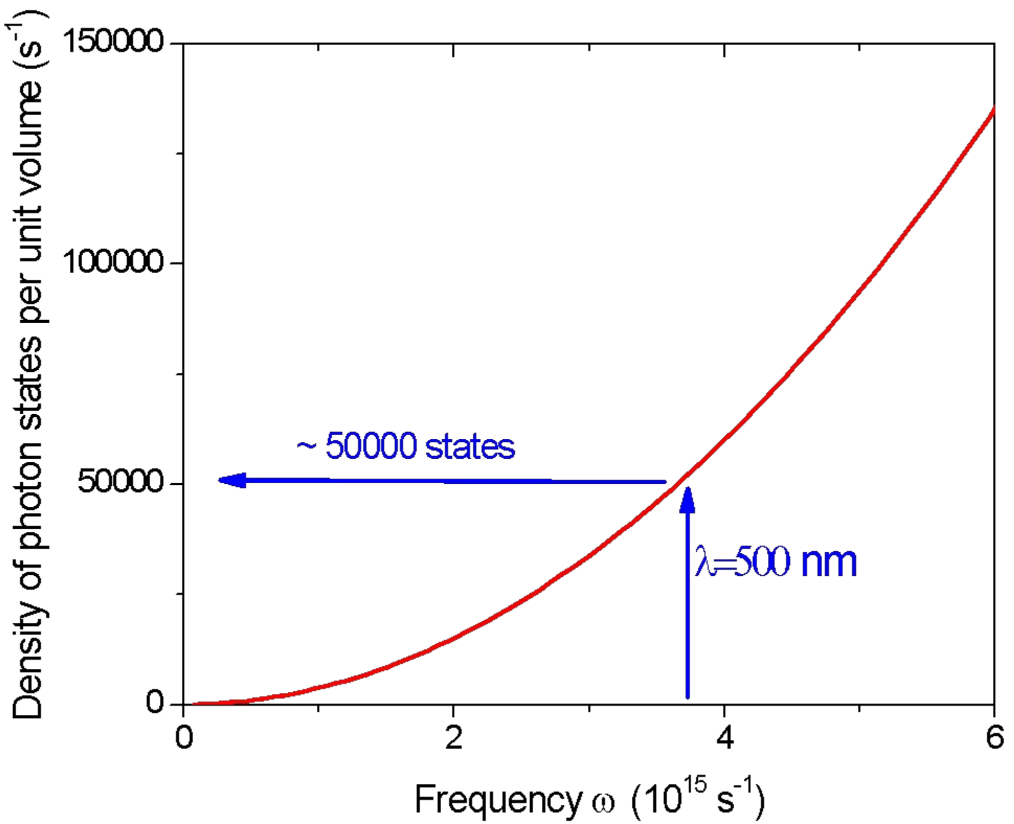
As a function of frequency ω ($=ck$):

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

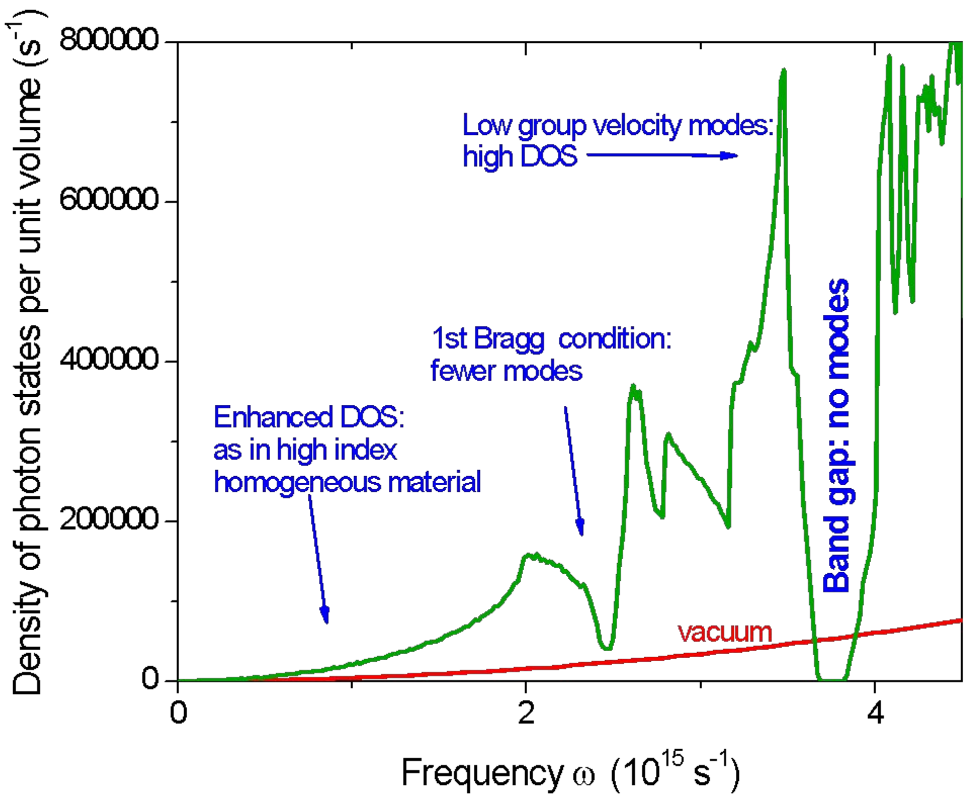
Density of states in vacuum

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

Example: ~50000 photon states per m^3 of vacuum per 1 Hz @ λ=500 nm

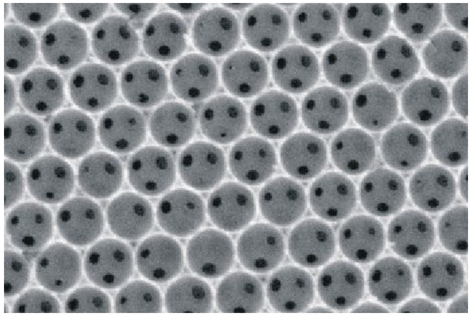


Controlling the DOS



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

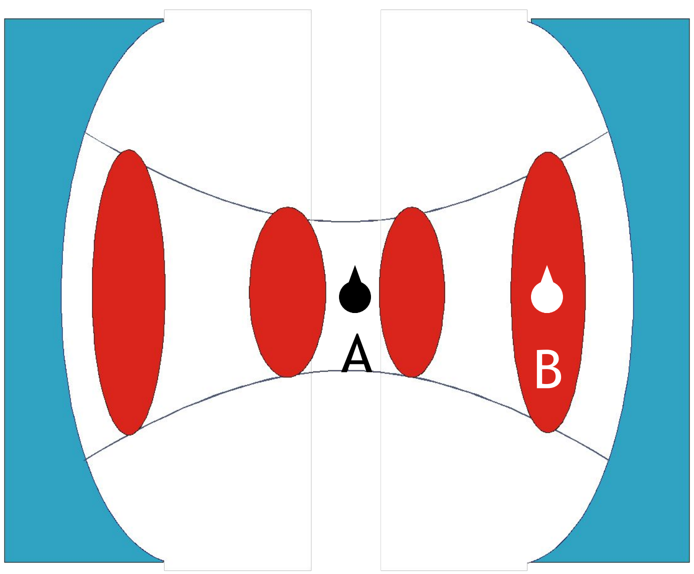
Local DOS

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

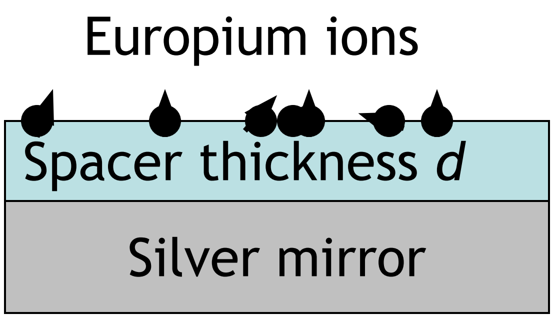
DOS: just count states	*Local DOS*
$N(\omega) = \sum_{\text{all modes } m} \delta(\omega_m - \omega)$	$N(\mathbf{r},\mathbf{d},\omega) = \sum_{\text{all modes}} \lvert \mathbf{d} \cdot \mathbf{E}_m(\mathbf{r}) \rvert^2 \, \delta(\omega_m - \omega)$

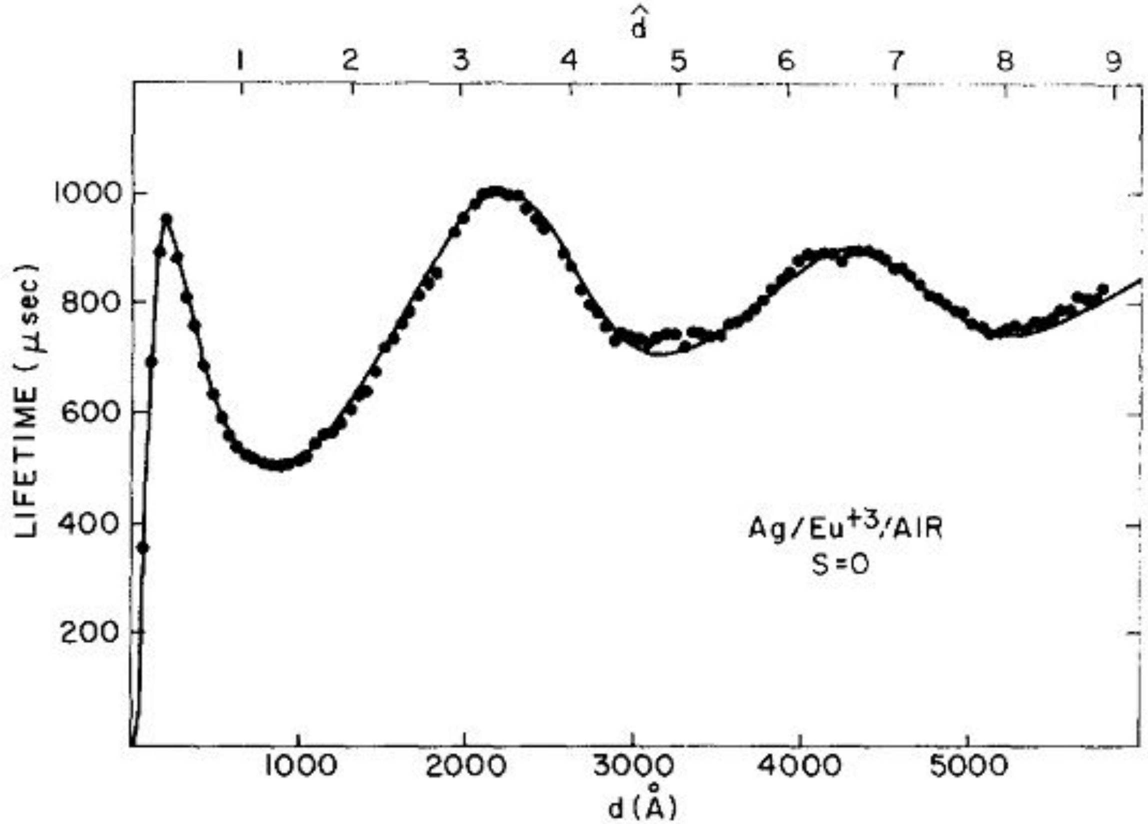


Atom at position A can not emit into cavity mode.

Atom at position B can emit into cavity mode.

LDOS: emission in front of a mirror

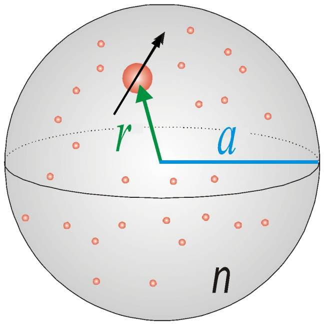


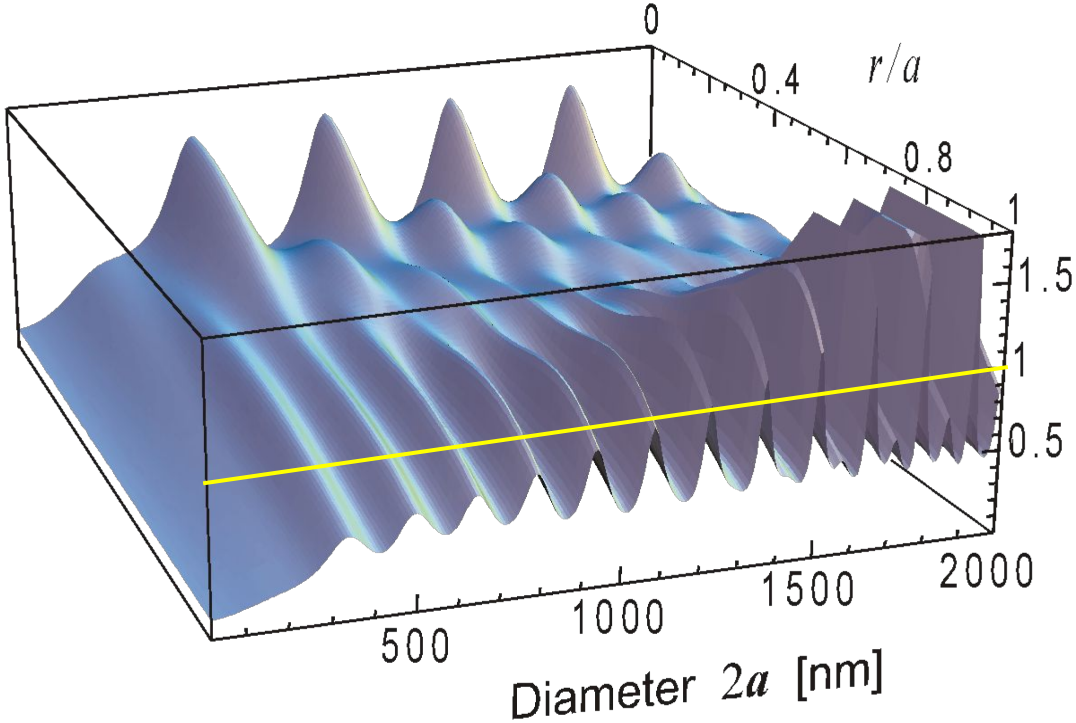


Drexhage (1966): fluorescence lifetime of Europium ions depends on source position relative to a silver mirror
(λ=612 nm)

Example II: dielectric nano-sphere

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

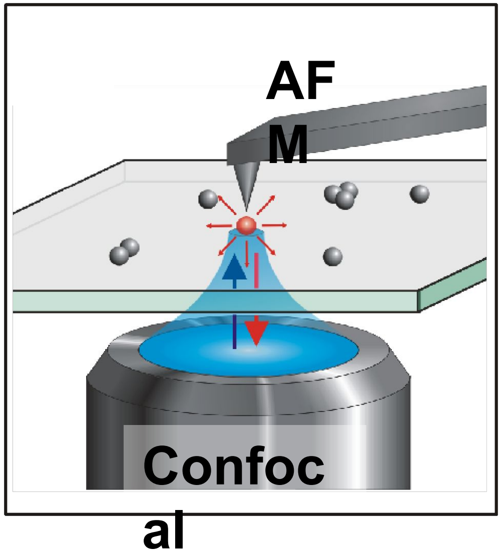




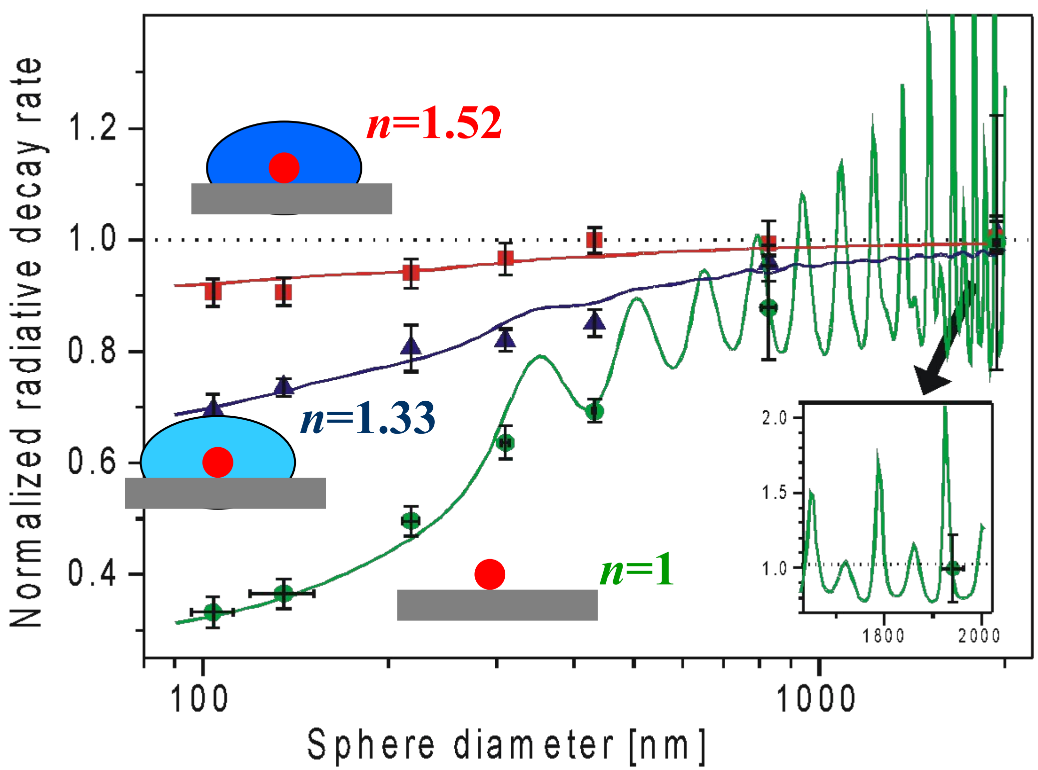
LDOS normalized to LDOS in SiO_2

[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

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

LDOS & measuring nonradiative decay

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

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

Measured in experiment

Unknown loss
local chemistry
at source

Fermi's Golden Rule
LDOS

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