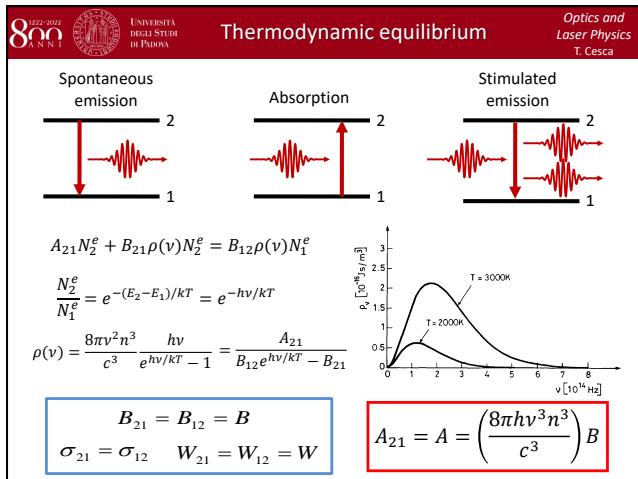


1 Lecture 8

Slide 1



We have also a relation between the spontaneous emission A and the absorption (or stimulated emission) B . n is the refractive index of the medium. We have a term v^3 : the spontaneous emission will dominate for very high frequency. At UV every material become fluorescent.

These results are valid for non degenerate levels.

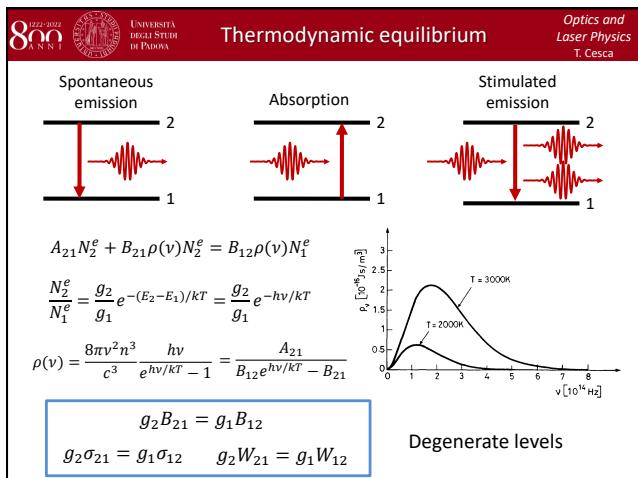
There is an elegant demonstration by Einstein whose exploit thermodynamic to calculate the three decay rates for the three processes.

We have a material embedded in a black body. At thermodynamic equilibrium, we have a balance between spontaneous emission, absorption and stimulated emission. The first two term on the lhs are the one of spontaneous and stimulated emission. The term on the rhs is the one off absorption.

At thermodynamic equilibrium, we have also the Boltzmann statistic for the population of the two levels.

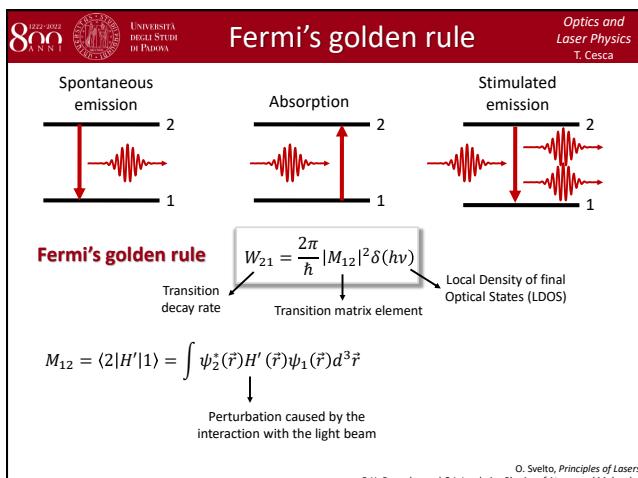
It is easy to obtain the energy density as this expression. In order to satisfy this equality, we have that the Einstein coefficient for stimulated emission should be equal to absorption and so on.

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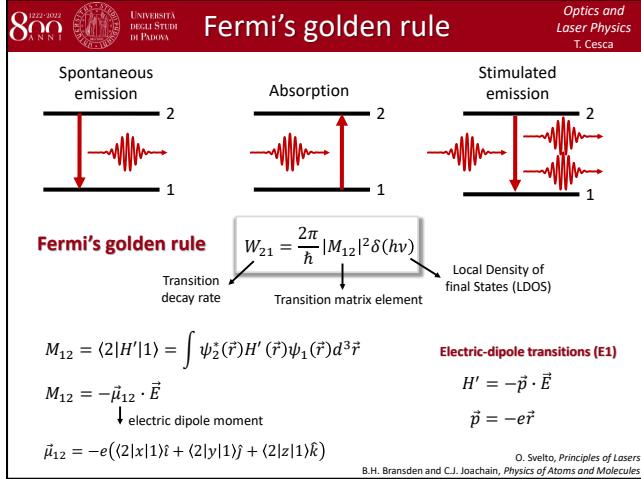
In the degenerate case, we have to take into account the degeneracy g_1 and g_2 of level 1 and 2. However, the physics of the process does not change.

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Until now, we have considered a semi-classical approach. If you want to adopt a more formal quantum mechanical approach, we have to introduce **Fermi's golden rule** in order to describe the decay rate for one of these processes. The transition rate can be written as a function of the **local density of final optical states (LDOS)**. The transition matrix should be written as the integral of the Hamiltonian.

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We can use different Hamiltonian and so we have different kind of transitions.

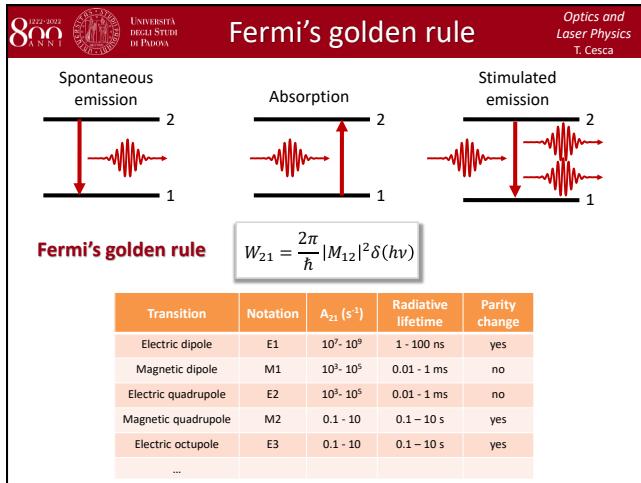
The most probable transition is the **Electric-dipole transition (E1)**: the Hamiltonian is written as

$$H' = -\vec{p} \cdot \vec{E}$$

We think the two level system as a dipole. We have the interaction of the dipole and the electric field of the beam.

The term μ_{12} take into account the characteristic of the level we are considering and E is the electric field of the beam.

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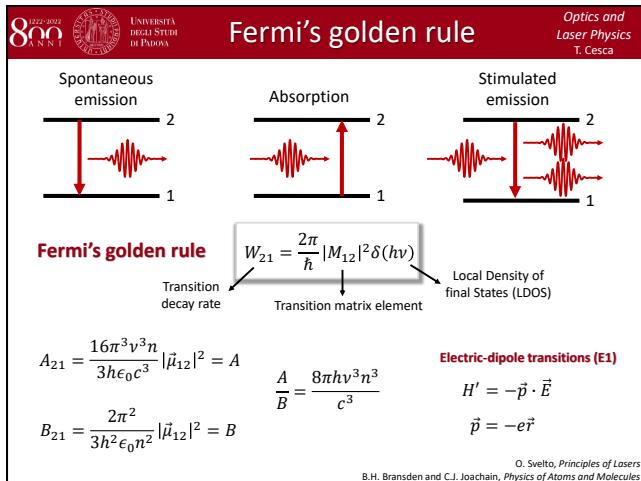


The probability of electric-dipole transition is strongest wrt others. However, we have also other transitions.

We can consider also higher order transitions.

Actually, the higher is the order, the lower is the transition rate. That is why these system from a quantum mechanic point of view are described by dipoles.

Slide 6



Making the exact calculation for the transition E1, it is possible to obtain the exact value for the Einstein coefficient A_{21} (spontaneous emission) and B_{21} (stimulated emission equal to absorption if we are not considering degenerate levels).

Einstein could determine the ratio

$$\frac{A}{B} = \frac{8\pi h\nu^3 n^3}{c^3}$$

well in advance wrt a quantum mechanical approach.

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Fermi's golden rule

Fermi's golden rule

$$W_{21} = \frac{2\pi}{\hbar} |M_{12}|^2 \delta(\hbar\nu)$$

Transition decay rate Transition matrix element Local Density of final States (LDOS)

$$A_{ji} = \frac{16\pi^3 v_{ji}^3 n e^2}{3h\epsilon_0 c^3} \sum_{m_j m_i} |\langle j, m_j | i, m_i \rangle|^2$$

↓ degeneracy of the upper state

$$f_{ji} = \frac{2m(2\pi)^2 v_{ji}}{3he^2} |\vec{\mu}_{ij}|^2$$

Oscillator's strength

B.H. Bransden and C.J. Joachain, *Physics of Atoms and Molecules*

Just for completeness, by considering the degeneracy of the levels, we have to consider the different quantum number for the level.

The **oscillator's strength** take into account the amplitude of the different transition. It is related to the different strength of the process of the transition. Indeed, the transition are not the same in terms of amplitude.

Slide 8

Fermi's golden rule

Fermi's golden rule

$$W_{21} = \frac{2\pi}{\hbar} |M_{12}|^2 \delta(\hbar\nu)$$

Transition decay rate Transition matrix element Local Density of final States (LDOS)

Selection rules

- (1) The parity of the wave function must change
- (2) $\Delta l = \pm 1$ for the changing electron
- (3) $\Delta L = 0, \pm 1$ but $L = 0 \rightarrow 0$ is forbidden
- (4) $\Delta J = 0, \pm 1$ but $J = 0 \rightarrow 0$ is forbidden
- (5) $\Delta S = 0$

Electric-dipole transitions (E1)

$$H' = -\vec{p} \cdot \vec{E}$$

$$\vec{p} = -e\vec{r}$$

B.H. Bransden and C.J. Joachain, *Physics of Atoms and Molecules*

The different transitions have to obey to different **selection rules**: we have allowed and forbidden transition. If you have a *forbidden transition* does not mean that you never get that transition, but it means that (you have to think in term of probability) they have a very low probability to occur.

The oscillator strength depends also on the selection rules and on the fact that the transition is allowed or forbidden.

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Radiative relaxation control: emitter near a planar interface

K.H. Drexhage, Influence of a dielectric interface on fluorescence decay time *J. Lumin.* 1 693-701 (1970)

Fermi's golden rule

$$W_{fi} = \frac{2\pi}{\hbar} |M_{if}|^2 \delta_f$$

Transition decay rate Interaction matrix element Local density of final states (LDOS)

Emitters-containing monolayer

An interface in close proximity ($d < \lambda_{em}$) of an emitter changes the photonic local density of states.

Local density of final states (LDOS)

Lifetime (ms)

Emitter: Eu³⁺-complex
Spacer: fatty acid

Distance (nm)

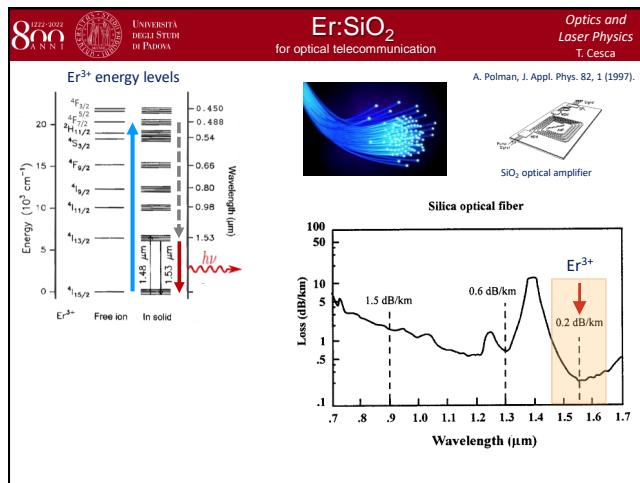
The transition rate (probability for a transition to occur) is not an intrinsic property of a material. The *interaction matrix element* depends only on intrinsic properties, however the transition rate **depends** also on the *local density of final states*.

So, by changing the local density, you can control the transition rate!

The plot show that it is possible to modulate the lifetime for spontaneous emission by modifying the local density of final state. In the paper a mono layer of a material containing emitters is considered. It is on top of a *spacer* such that it is at a distance from a mirror.

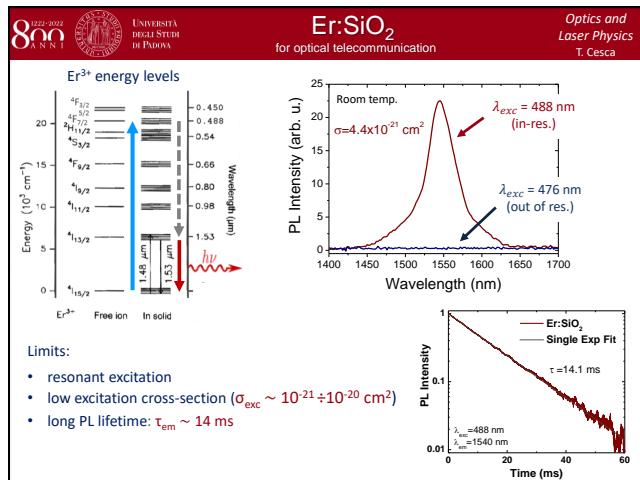
By changing the relative distance of the spacer, in this paper is shown that the lifetime (and a consequence the radiative decay rate) is changed. So, what is doing in this paper is changing the local density of final states, nothing else.

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The possibility to control the lifetime of an emitter is interesting for many applications. Let us consider Erbium in Silica. Erbium is used for optical communication (optical fiber). It can be excited with a light beam in the visible and it emits in a range which minimize the losses in optical fiber.

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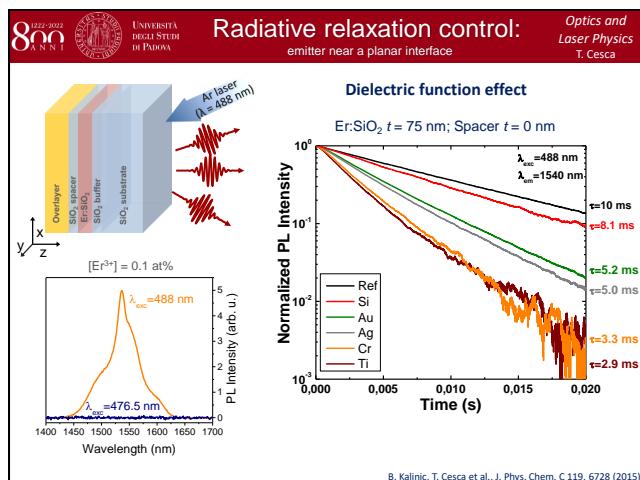


Drawbacks:

- they require resonant excitation (you have to pump at some specific absorption transition). You are forced to use only specific frequency to pump.
- low excitation cross-section.
- slow emitter: long luminescent time. It means that it could be prone to other processes as non radiative ones. So, you cannot get fast commutation when you are using emitters with such a long lifetime.

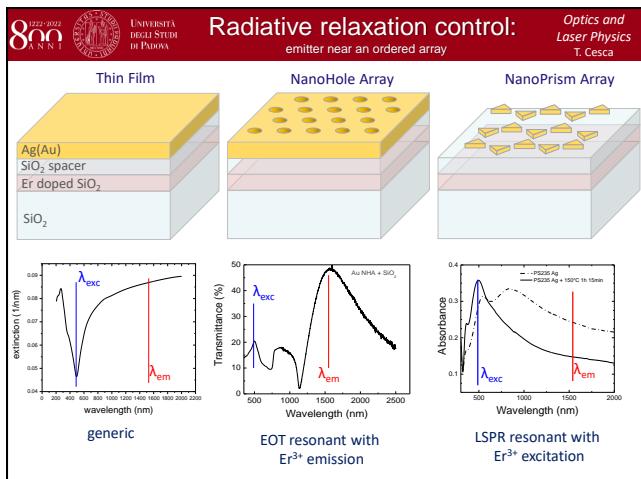
Is it possible to reduce the long lifetime? Yes, by controlling the local density of optical states for erbium emitter.

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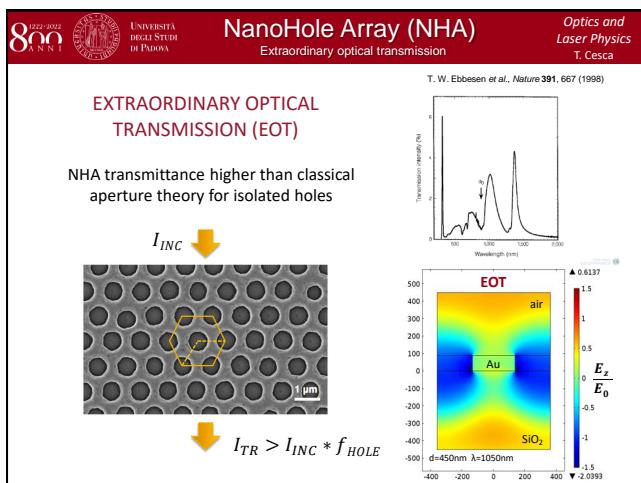
This is an example. We measure the temporal evolution of photo luminescence intensity at the Erbium emission (476.5 nm) by changing different materials used as overlayer on top. Changing the material is the simplest way to change the local density of optical states.

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This is a more complicated strategy: engineer the local density by using nanohole or nanoprism arrays.

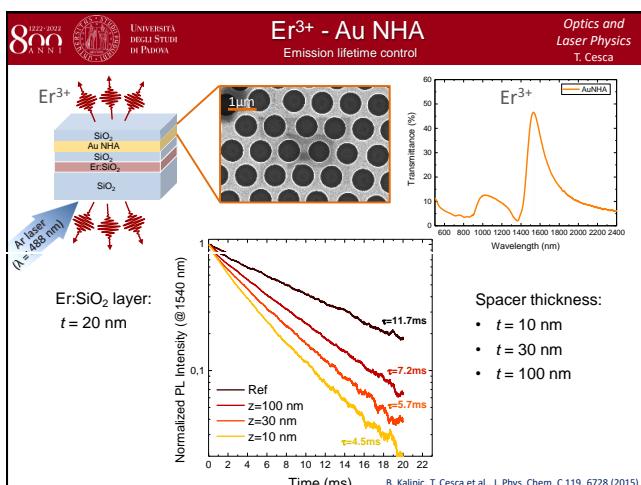
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For instance, we work with nanohole array (a layer in which you made holes with a diameter much smaller of the radiation light).

The intensity transmitted through this array of holes is larger than if you consider classical aperture theory for isolated holes. This is a phenomenon that occur because of surface plasmon at the nanoscale.

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The presence of a nanohole array is modifying the local optical density. The peak related to **extraordinary optical transition** was perfectly matching the Erbium emission. It is possible again to modulate as a function of the thickness (provided by spacer placed between Erbium containing layer and the nanohole array) the lifetime. The smaller is the spacer thickness, the smaller is the reduction on the lifetime.