On the menu today

Decay rate engineering

- The electric dipole
- Green function
- Fields of electric dipole
- Power dissipated by an oscillating dipole
- The local density of optical states (LDOS)
- Decay rate of quantum emitters
- Decay rate engineering
- Example: Drexhage experiment
- Example: The Purcell effect
- Example: classical analogue of Drexhage experiment
- Example: optical antenna

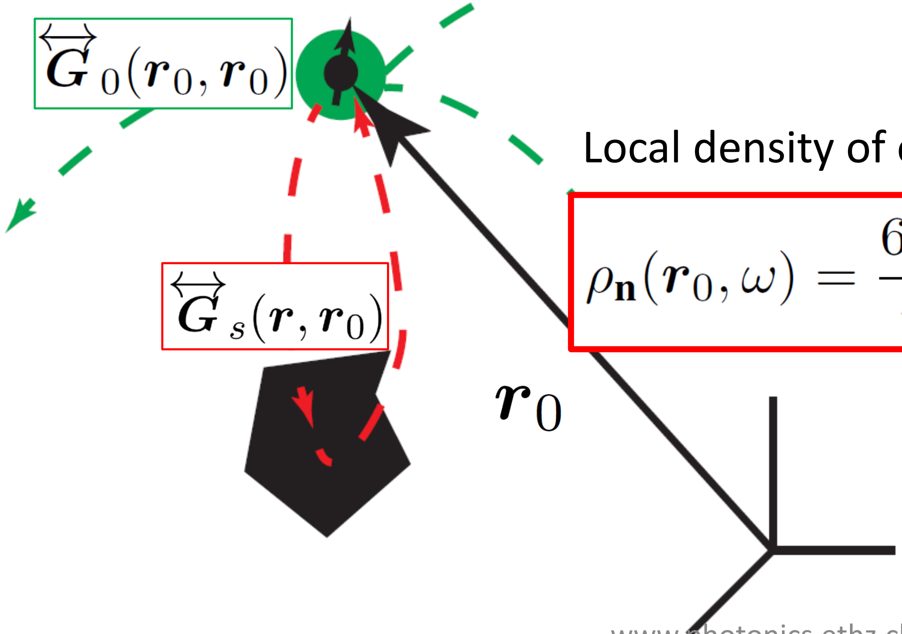
Optical antennas

- Dipolar scattering theory
- Radiation damping

Power radiated in inhomogeneous environment

- The power dissipated by a dipole depends on its environment and is proportional to the local density of optical states (LDOS).
- The LDOS is (besides prefactors) the imaginary part of the Green's function evaluated at the origin.
- Controlling the boundary conditions (and thereby the LDOS) allows us to control the power radiated by a dipole!

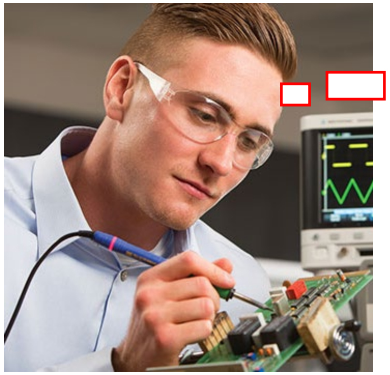
$$\langle P \rangle = \frac{\pi \omega^2}{12 \epsilon \epsilon_0} \left| \boldsymbol{p} \right|^2 \rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega)$$



Local density of optical states

$$\rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega) = \frac{6 \omega n^2}{\pi c^2} \left\{ \boldsymbol{n}_p^{\intercal} \, \mathrm{Im} \left[\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}_0, \boldsymbol{r}_0; \omega) \right] \boldsymbol{n}_p \right\}$$

Power radiated in inhomogeneous environment

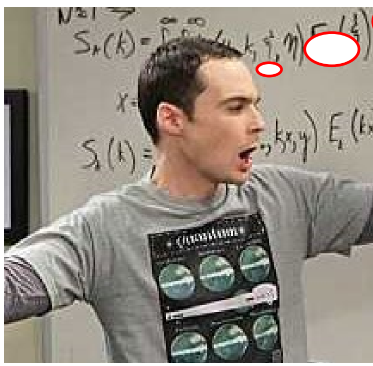


Radiation resistance!

Power dissipated in an electrical circuit:

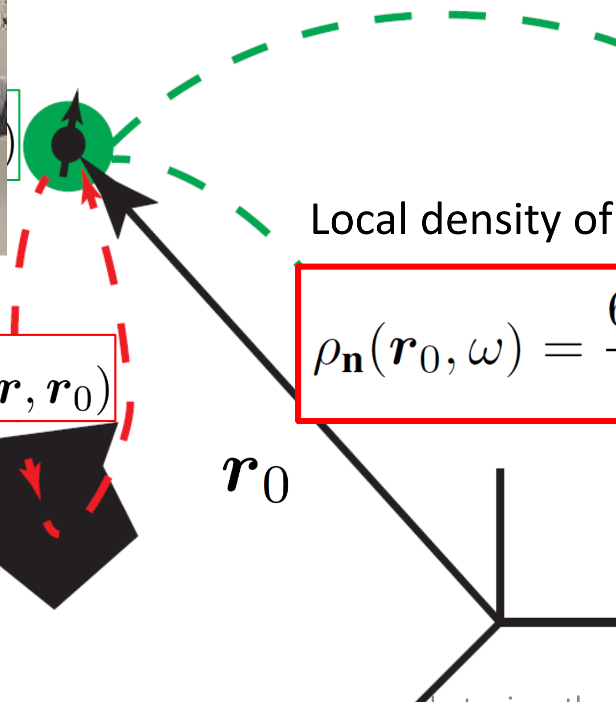
$$P \propto I^2 \cdot R$$

current — resistance



LDOS!

$$\langle P \rangle = \frac{\pi \omega^2}{12 \epsilon \epsilon_0} |\boldsymbol{p}|^2 \; \rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega)$$



Local density of optical states

$$\rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega) = \frac{6 \omega n^2}{\pi c^2} \left\{ \boldsymbol{n}_p^{\intercal} \, \mathrm{Im} \left[\overleftrightarrow{G}(\boldsymbol{r}_0, \boldsymbol{r}_0; \omega) \right] \boldsymbol{n}_p \right\}$$

Power radiated in inhomogeneous environment

Via the local density of states (LDOS)
the power radiated by a dipole depends on

- location of source within its environment
- frequency of source
- orientation of source

The LDOS can be interpreted as a <u>radiation resistance</u>

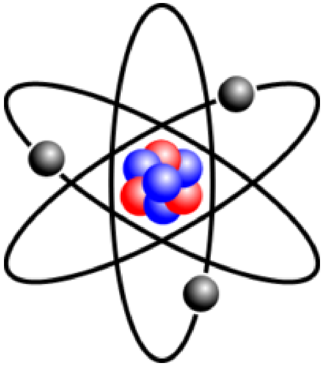
$$\langle P \rangle = \frac{\pi \omega^2}{12 \epsilon \epsilon_0} |\boldsymbol{p}|^2 \, \rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega)$$

$$\rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega) = \frac{6\omega n^2}{\pi c^2} \left\{ \boldsymbol{n}_p^{\intercal} \, \mathrm{Im}\left[\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}_0, \boldsymbol{r}_0; \omega) \right] \boldsymbol{n}_p \right\}$$

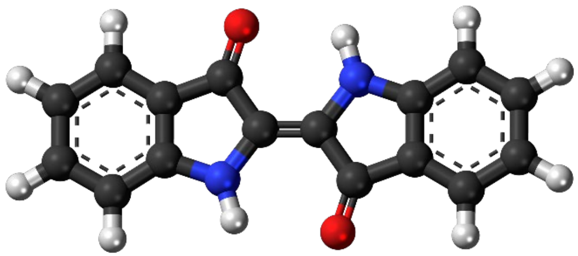
In analogy with $P = I^2 \cdot R$

Quantum emitters

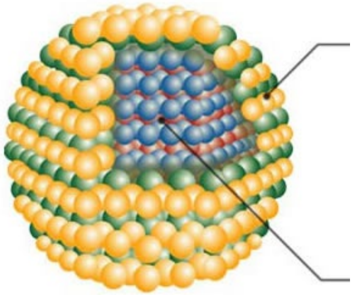
Radiating sources at 1000 THz :



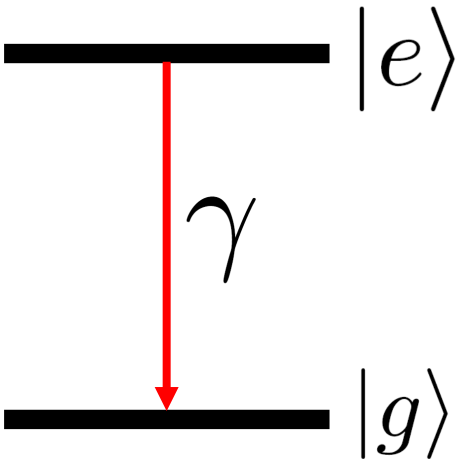
Atoms



Dye molecules

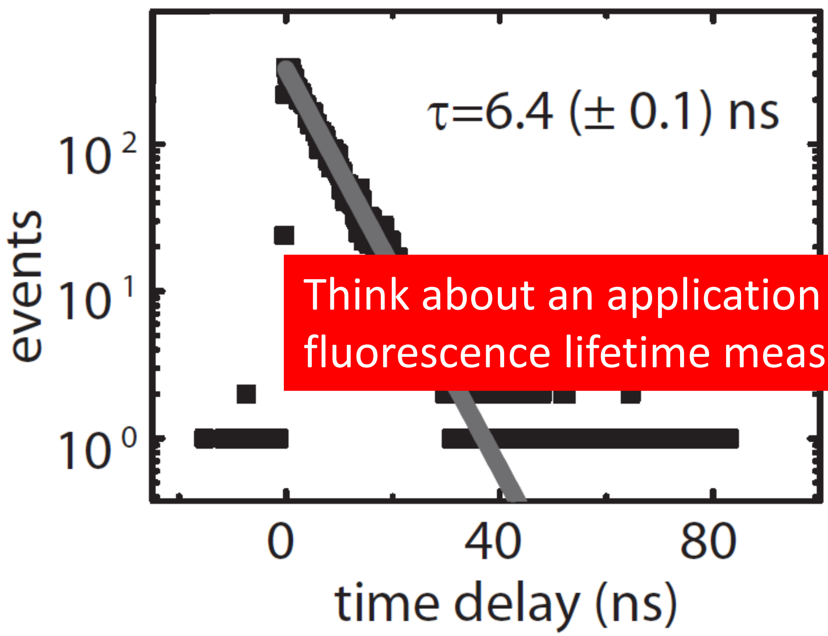


Quantum dots



Optical emitters have discrete level scheme (in the visible)
Let’s focus on the two lowest levels
How long will the system remain in its excited state?

Fluorescence lifetime measurements



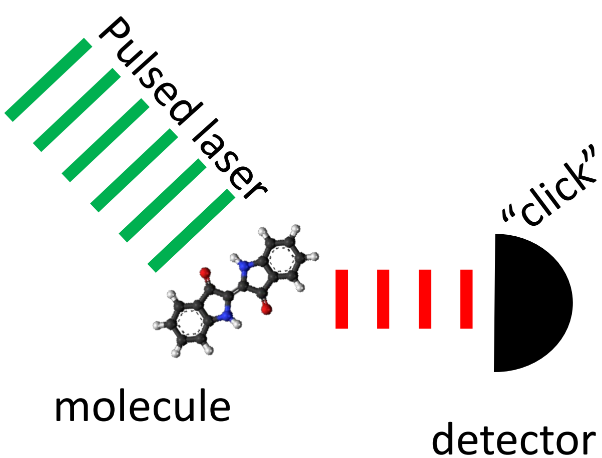
decay rate → $\gamma = 1/\tau$ ← lifetime

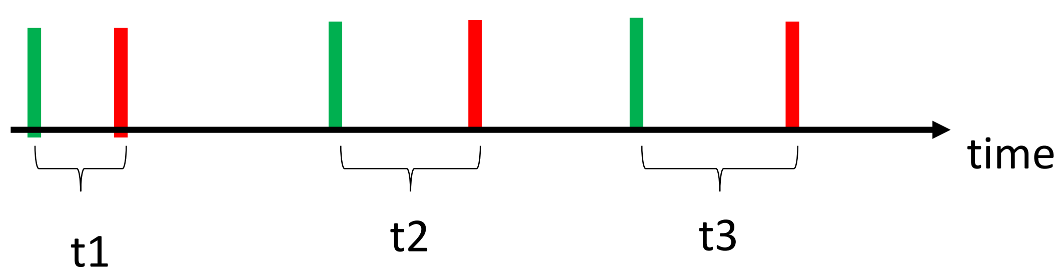
$$p_e(t) = p_e(0)\ \exp\left[-\gamma t\right]$$

Think about an application (beyond academic interest) for fluorescence lifetime measurements!

oton at

1. Prepare system in excited state with light pulse at t=0
2. Record time delay t1
3. Repeat experiment many times
4. Histogram arrival time delays





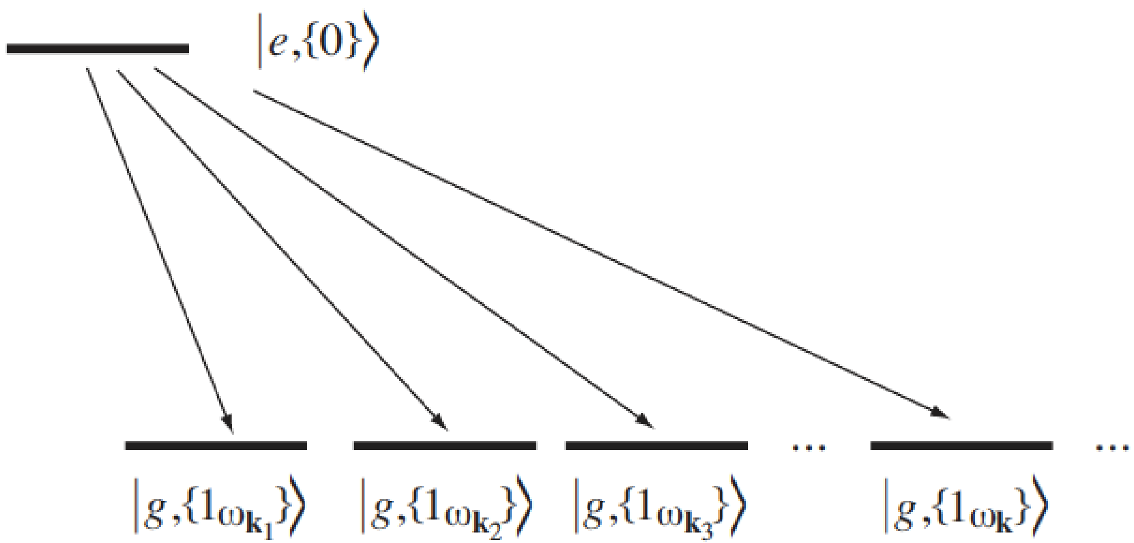
Calculation of decay rate γ

Fermi's Golden Rule:

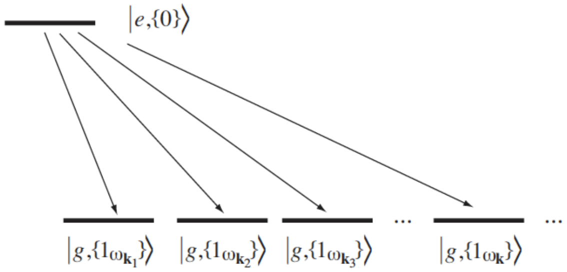
$$\gamma = \sum_f \frac{2\pi}{\hbar} \left|\langle f | \hat{\mathscr{H}} | i \rangle\right|^2 \delta(E_i - E_f)$$

Initial state (excited atom, no photon): $|i\rangle = |e, 0\rangle$

Final state (de-excited atom, 1 photon in state k at frequency omega): $|f\rangle = |g, 1_{\omega_{\mathbf{k}}}\rangle$



The Wigner-Weisskopf approximation



Calculation of decay rate γ

Fermi's Golden Rule:

$$\gamma = \sum_f \frac{2\pi}{\hbar} \left| \langle f | \hat{\mathscr{H}} | i \rangle \right|^2 \delta(E_i - E_f)$$

Initial state (excited atom, no photon): $|i\rangle = |e, 0\rangle$

Final state (de-excited atom, 1 photon in state k at frequency ω): $|f\rangle = |g, 1_{\omega_{\mathbf{k}}}\rangle$

Interaction Hamiltonian: $\hat{\mathcal{H}} = -\hat{\boldsymbol{p}} \cdot \hat{\boldsymbol{E}}$

Sum over final states is sum over photon states (**k**) at transition frequency ω.

$$\gamma = \frac{\pi\omega}{3\hbar\epsilon_0} |\hat{\boldsymbol{p}}|^2 \rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega)$$

Atomic part:
transition dipole moment (quantum)

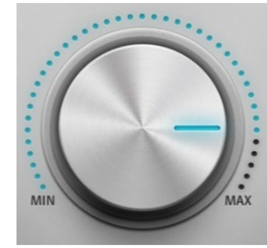
$$|\hat{\boldsymbol{p}}|^2 = |\langle g | \hat{\boldsymbol{p}} | e \rangle|^2$$

Field part:
Local density of states (classical)

$$\rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega) = \frac{6\omega}{\pi c^2} \left\{ \boldsymbol{n}_p^{\intercal} \operatorname{Im} \left[\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}_0, \boldsymbol{r}_0; \omega) \right] \boldsymbol{n}_p \right\}$$

Decay rate engineering

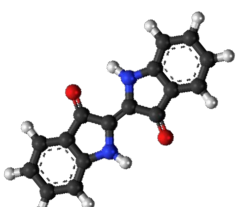
$$\gamma = \frac{\pi\omega}{3\hbar\epsilon_0}|\hat{\boldsymbol{p}}|^2 \rho_{\mathbf{n}}(\boldsymbol{r}_0,\omega)$$

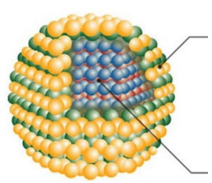


Emitter

Transition dipole moment: Wave function engineering by synthesizing molecules, and quantum dots

Chemistry, material science

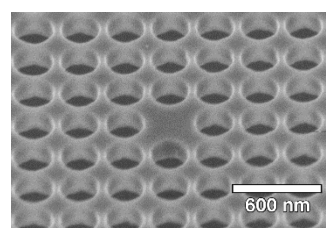


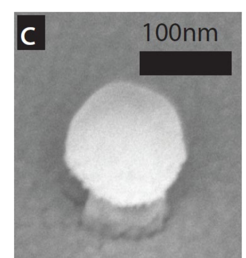


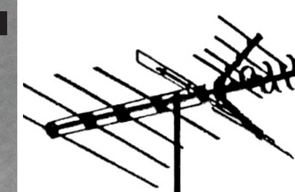
Environment

LDOS: Electromagnetic mode engineering by shaping boundary conditions for Maxwell’s equations

Physics, electrical engineering







antennaking.com, Wikimedia, emory.edu

Rate enhancement – quantum vs. classical

$$\gamma = \frac{\pi\omega}{3\hbar\epsilon_0} \left|\hat{\boldsymbol{p}}\right|^2 \rho_{\mathbf{n}}(\boldsymbol{r}_0,\omega)$$

$$\langle P\rangle = \frac{\pi\omega^2}{12\epsilon\epsilon_0} \left|\boldsymbol{p}\right|^2 \rho_{\mathbf{n}}(\boldsymbol{r}_0,\omega)$$

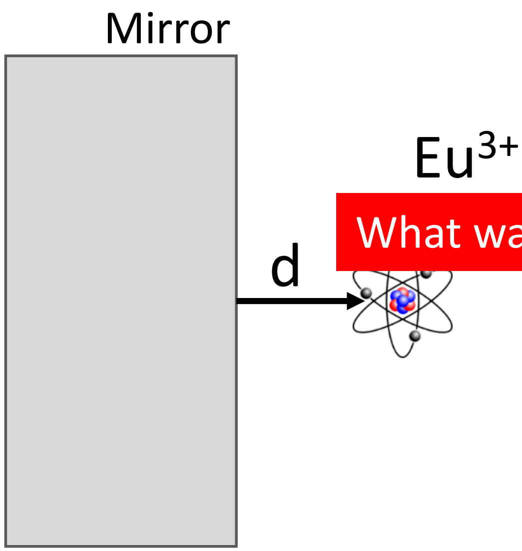
Transition dipole moment is NOT classical dipole moment, but

$$\frac{\gamma}{\gamma_0} = \frac{P}{P_0}$$

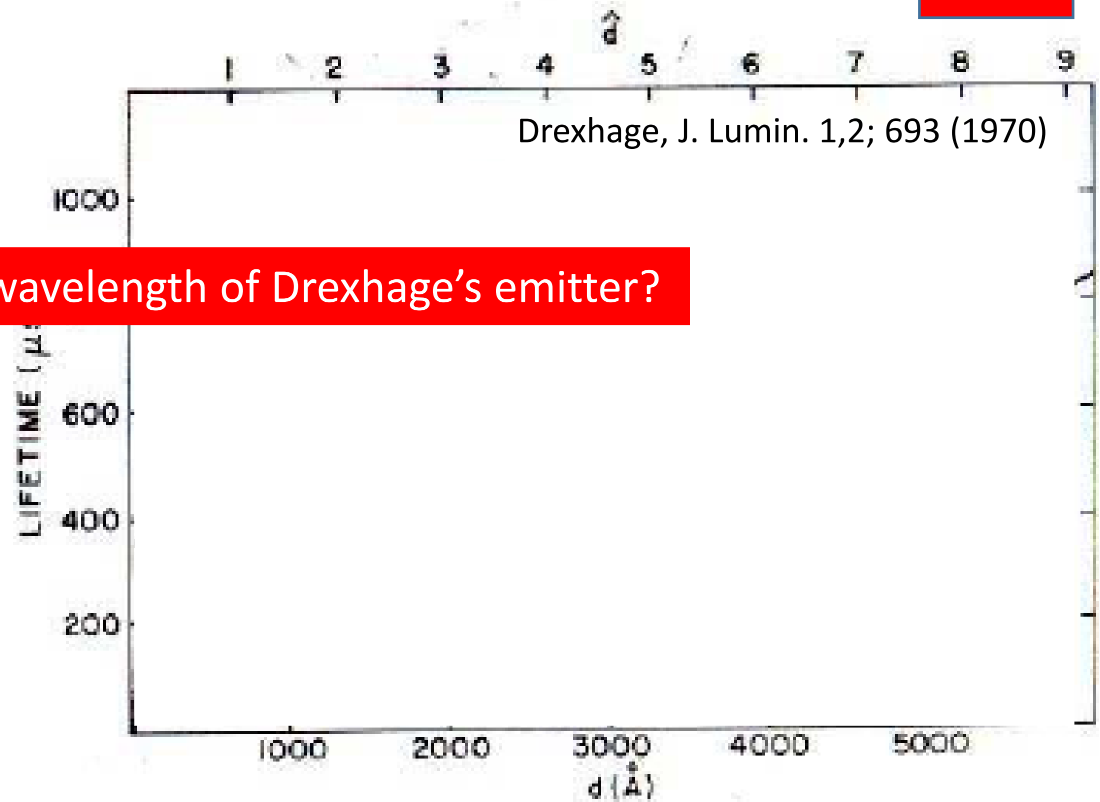
Classical electromagnetism CANNOT make a statement about the absolute decay rate of a quantum emitter.
BUT: Classical electromagnetism CAN predict the decay rate *enhancement* provided by a photonic system as compared to a reference system.

Drexhage's experiment (late 1960s)

HW3



What was the wavelength of Drexhage's emitter?

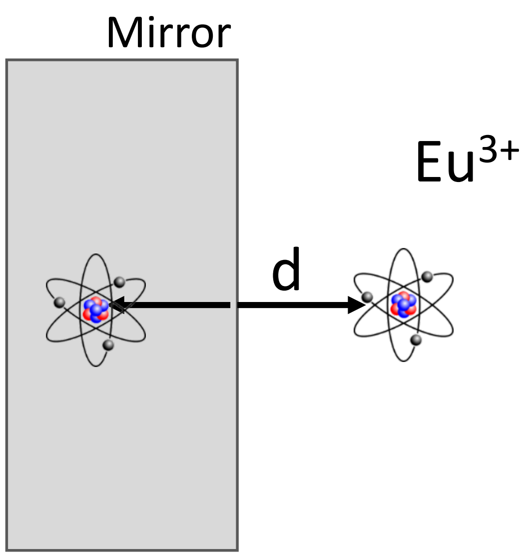


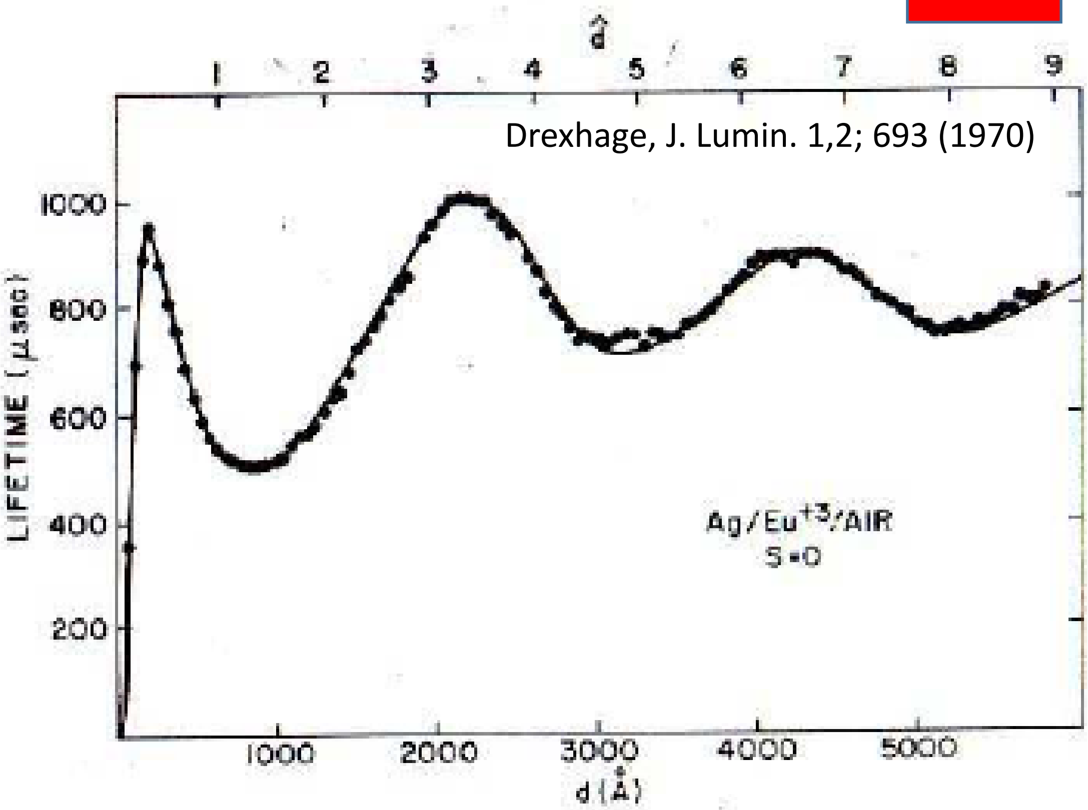
Drexhage, J. Lumin. 1,2; 693 (1970)

First observation of the local (!) character of the DOS!

Drexhage's experiment (late 1960s)

HW3



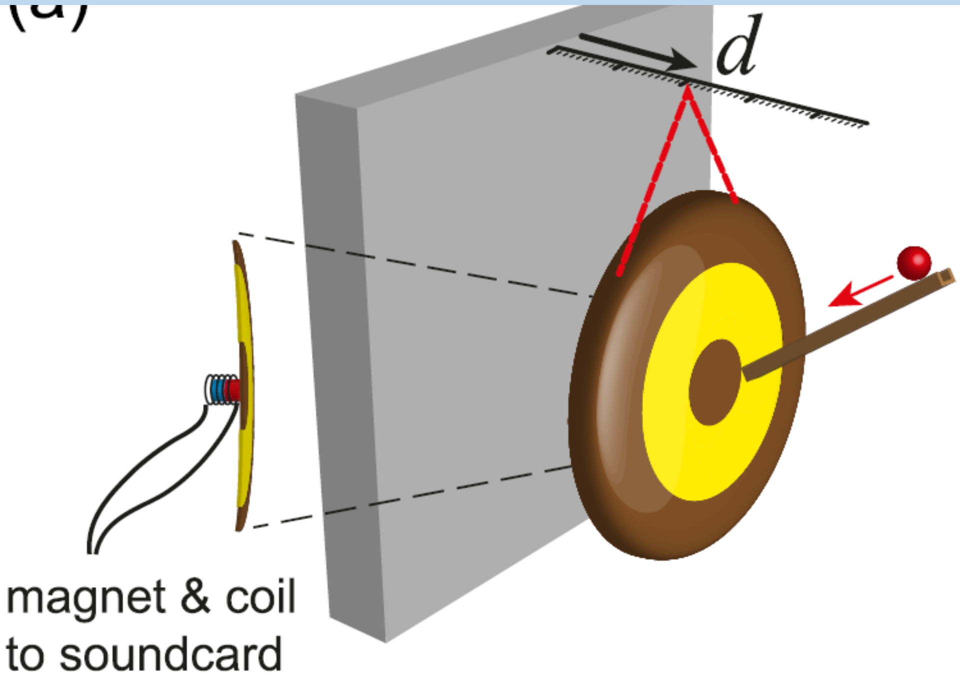


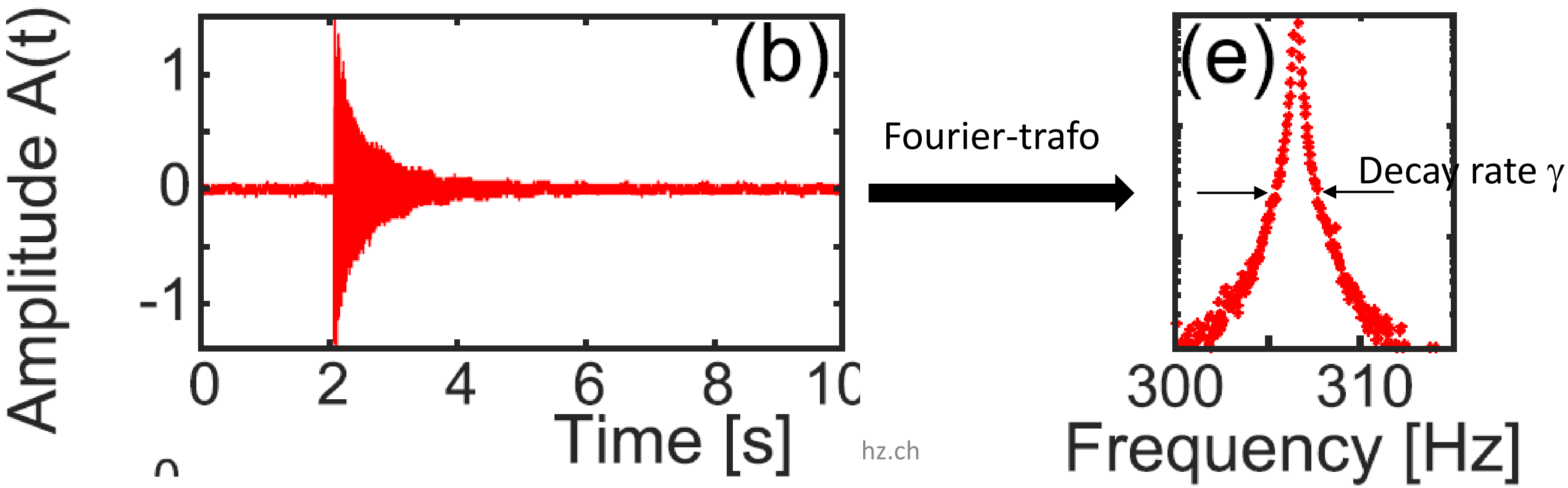
Emitter sees its own mirror image.

G_s is given as the field generated by the mirror dipole.

A classical analogy for Drexhage’s experiment

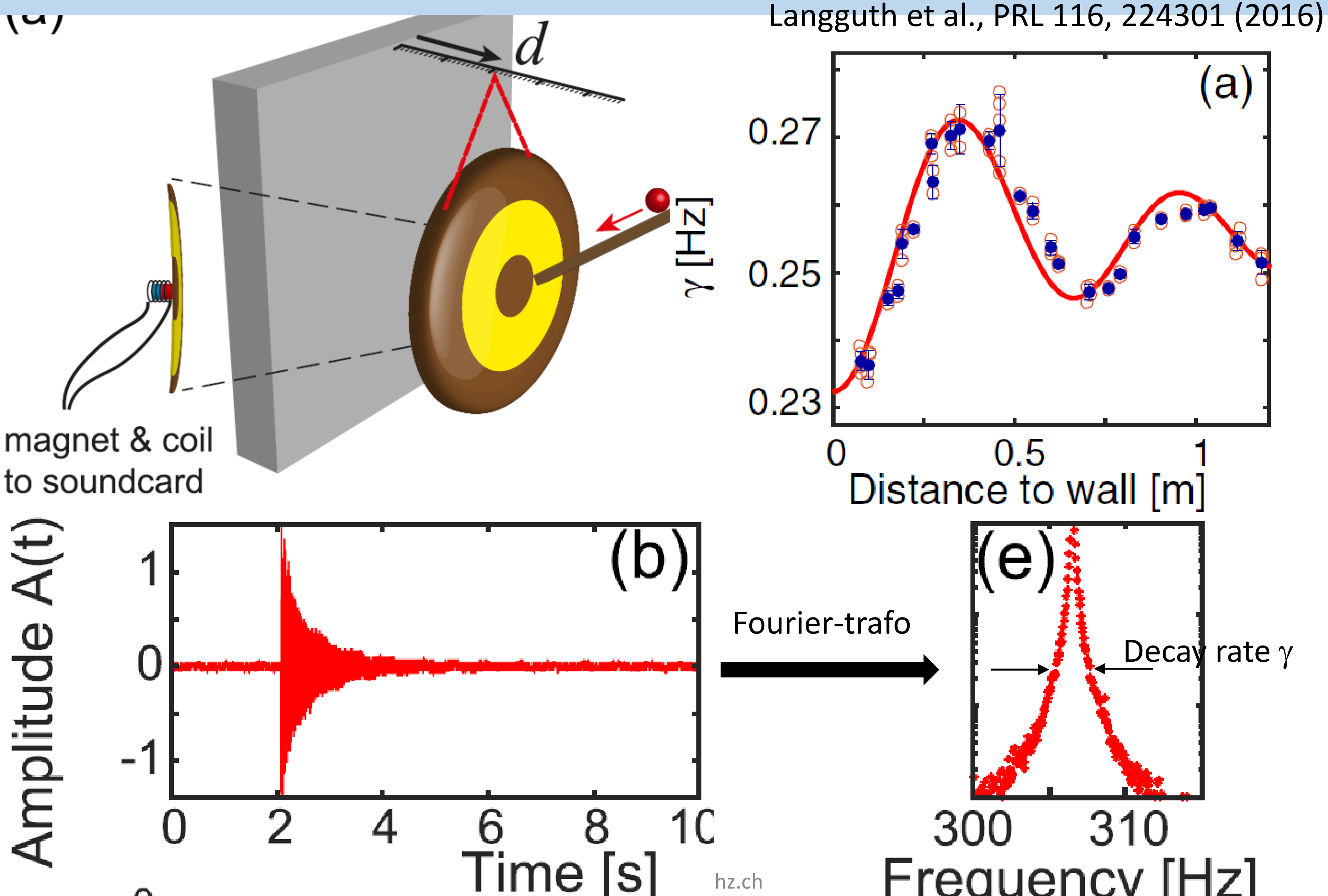
Langguth et al., PRL 116, 224301 (2016)





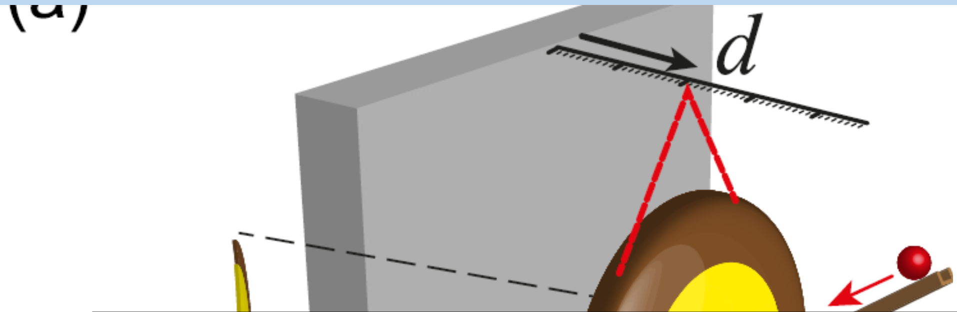
A classical analogy for Drexhage's experiment

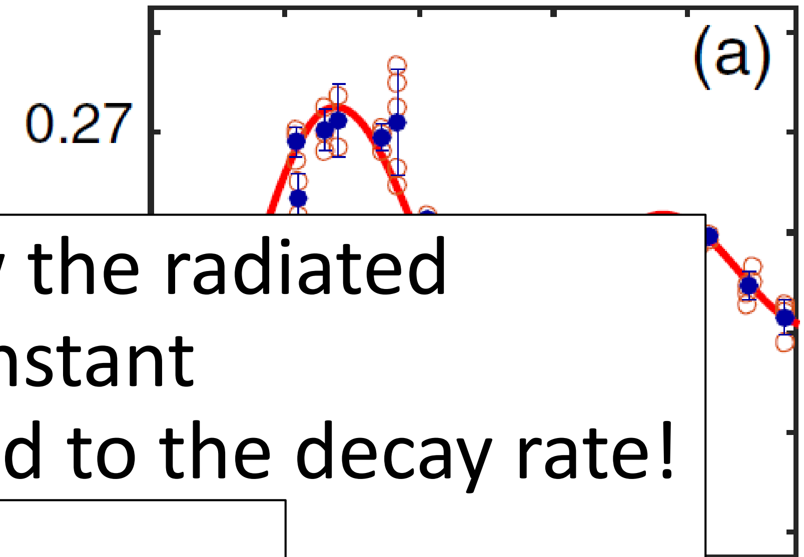
Langguth et al., PRL 116, 224301 (2016)



A classical analogy for Drexhage's experiment

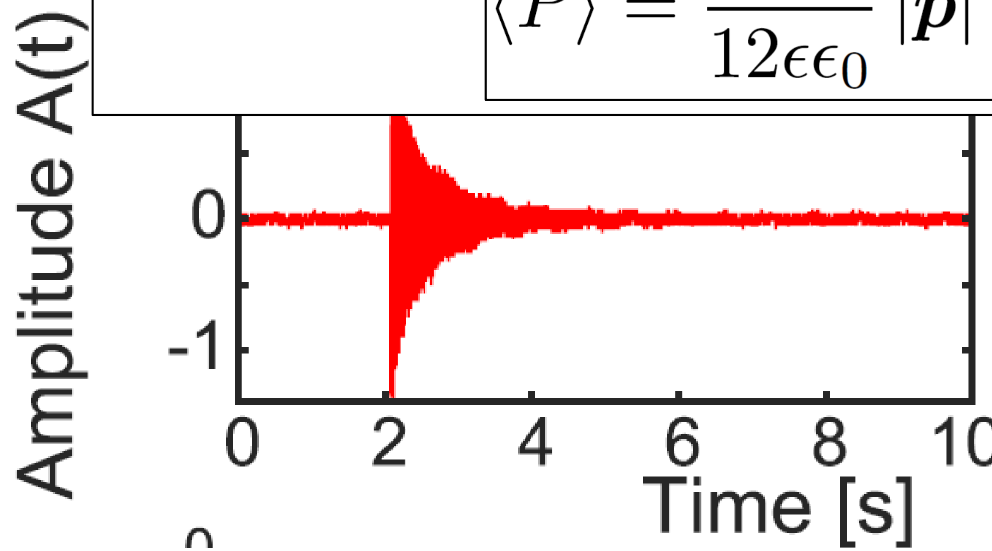
Langguth et al., PRL 116, 224301 (2016)



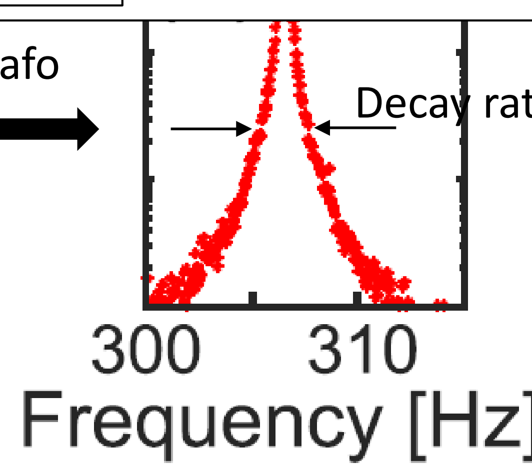


Make clear to yourself how the radiated power for a dipole with constant current/amplitude is related to the decay rate!

$$\langle P \rangle = \frac{\pi \omega^2}{12 \epsilon \epsilon_0} \left| \boldsymbol{p} \right|^2 \rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega)$$

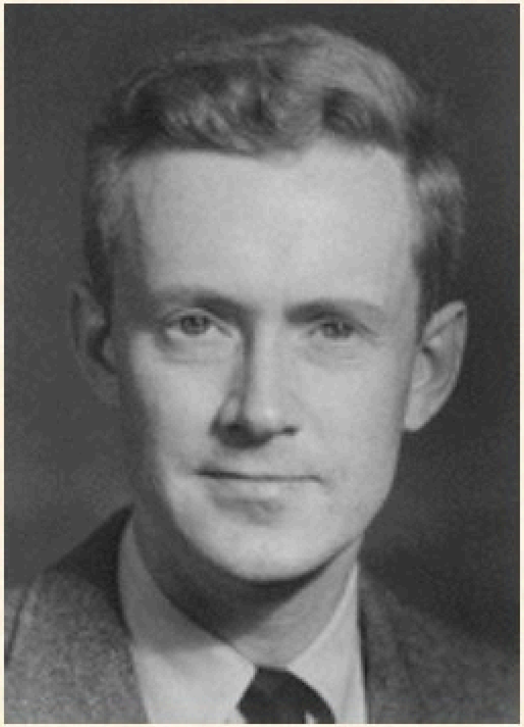


Fourier-trafo



hz.ch

1946 - E. M. Purcell predicts modification of spontaneous emission rates in complex media



Edward M. Purcell
1912-1997

B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. Purcell, *Harvard University*.—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

$$A_\nu = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2)\ \text{sec.}^{-1},$$

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu = 10^7$ sec.$^{-1}$, $\mu = 1$ nuclear magneton, the corresponding relaxation time would be 5×10^{21} seconds! However, for a system coupled to a resonant electrical circuit, the factor $8\pi\nu^2/c^3$ no longer gives correctly the number of radiation oscillators per unit volume, in unit frequency range, there being now *one* oscillator in the frequency range ν/Q associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor $f = 3Q\lambda^3/4\pi^2V$, where V is the volume of the resonator. If a is a dimension characteristic of the circuit so that $V\sim a^3$, and if δ is the skin-depth at frequency ν, $f\sim\lambda^3/a^2\delta$. For a non-resonant circuit $f\sim\lambda^3/a^3$, and for $a<\delta$ it can be shown that $f\sim\lambda^3/a\delta^2$. If small metallic particles, of diameter 10^{-3} cm are mixed with a nuclear-magnetic medium at room temperature, spontaneous emission should establish thermal equilibrium in a time of the order of minutes, for $\nu = 10^7$ sec.$^{-1}$.

Phys. Rev. **69**, 681 (1946)

1946 - E. M. Purcell predicts modification of spontaneous emission rates in complex media

Edward M. Purcell
1912-1997

B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. PURCELL, *Harvard University.*—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

$$A_\nu = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2) \text{ sec.}^{-1},$$

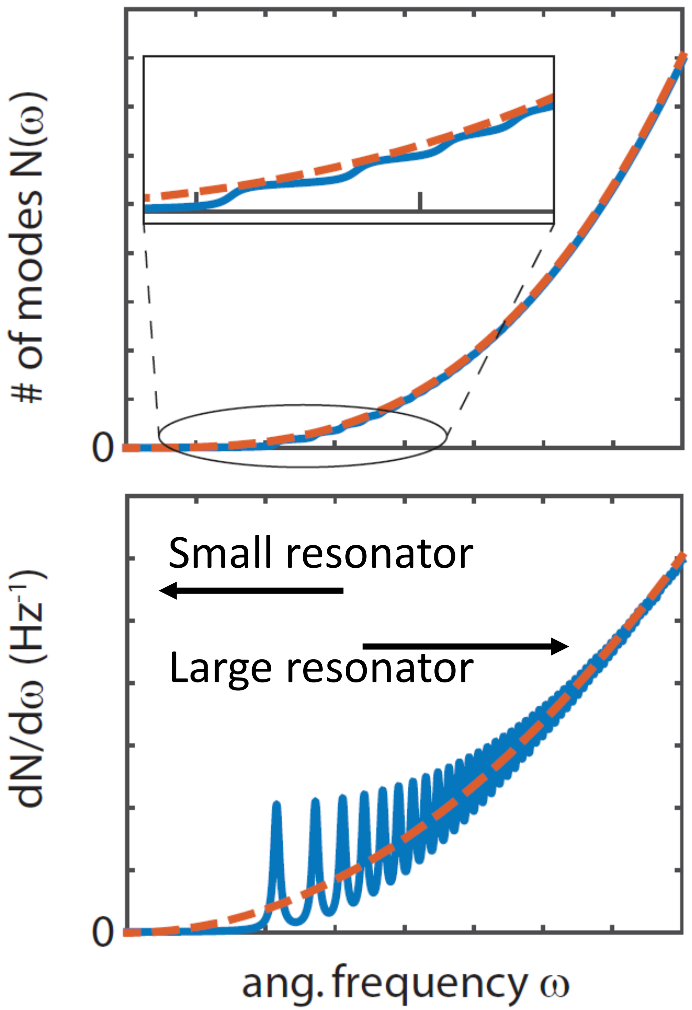
is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu=10^7$ sec.$^{-1}$, $\mu=1$ nuclear magneton, the corresponding relaxation time would be 5×10^{21} seconds! ...d to a resonant electrical ...longer gives correctly the ... per unit volume, in unit ...now *one* oscillator in the ...d with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor $f=3Q\lambda^3/4\pi^2V$, where V is the volume of the resonator. If a is a dimension characteristic of the circuit so that $V\sim a^3$, and if δ is the skin-depth at frequency ν, $f\sim\lambda^3/a^2\delta$. For a non-resonant circuit $f\sim\lambda^3/a^3$, and for $a<\delta$ it can be shown that $f\sim\lambda^3/a\delta^2$. If small metallic particles, of diameter 10^{-3} cm are mixed with a nuclear-magnetic medium at room temperature, spontaneous emission should establish thermal equilibrium in a time of the order of minutes, for $\nu=10^7$ sec.$^{-1}$.

How did he come up with that?

Phys. Rev. **69**, 681 (1946)

Density of states in a realistic resonator

How many modes in frequency band [ω, ω+Δω] and resonator volume V?



- Losses broaden delta-spike into Lorentzian
- Area under Lorentzian is unity
- The lower the loss, the higher the density of states on mode resonance
- Density of states on resonance exceeds that of free space

In free space (large resonator):

$$\rho(\omega) = \frac{\omega^2 n^3(\omega)}{\pi^2 c^3}$$

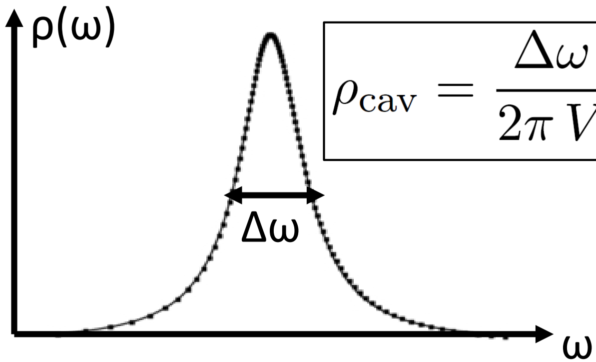
The Purcell effect

Free space: Density of states via Green function.
Alternatively, count states in large box (see EM course).

$$\rho_0(\omega) = \frac{\omega^2 n^3}{\pi^2 c^3}$$

$$\rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega) = \frac{6\omega n^2}{\pi c^2} \left\{ \boldsymbol{n}_p^{\intercal} \operatorname{Im}\left[\overleftrightarrow{\boldsymbol{G}}_0(\boldsymbol{r}_0, \boldsymbol{r}_0) \right] \boldsymbol{n}_p \right\}$$

In cavity: Lorentzian with essentially one mode per Δω and cavity volume V



$$\rho_{\text{cav}} = \frac{\Delta\omega}{2\pi\, V} \frac{1}{(\omega - \omega_0)^2 + (\Delta\omega/2)^2}$$

$$\mathcal{F}_P = \frac{\rho_{\text{cav}}(\omega_0)}{\rho_0(\omega_0)} = \frac{3}{4\pi^2} \left(\frac{\lambda}{n} \right)^3 \frac{Q}{V}$$

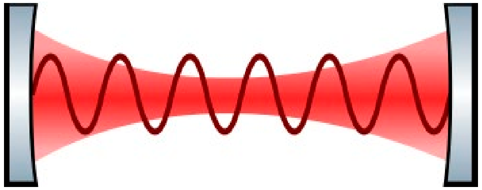
$$Q = \frac{\omega}{\Delta\omega}$$

The Purcell effect

$$\mathcal{F}_P = \frac{\rho_{\rm cav}(\omega_0)}{\rho_0(\omega_0)} = \frac{3}{4\pi^2}\left(\frac{\lambda}{n}\right)^3 \frac{Q}{V}$$

The Purcell factor is the maximum rate enhancement provided by a cavity given that the source is

1. Located at the field maximum of the mode
2. Spectrally matched exactly to the mode
3. Oriented along the field direction of the mode



Caution: Purcell factor is only defined for a cavity. The concept of the LDOS is much more general and holds for any photonic system.

Observation of Cavity-Enhanced Single-Atom Spontaneous Emission

P. Goy, J. M. Raimond, M. Gross, and S. Haroche
Laboratoire de Physique de l'Ecole Normale Supérieure, F-75231 Paris Cedex 05, France

(Received 1 April 1983)

It has been observed that the spontaneous-emission lifetime of Rydberg atoms is shortened by a large ratio when these atoms are crossing a high-Q superconducting cavity tuned to resonance with a millimeter-wave transition between adjacent Rydberg states.

Spontaneous atomic emission inside an electromagnetic cavity is expected to occur at a rate different from the same process in free space.[1-4] If the cavity is resonant with a transition between two atomic levels, the partial spontaneous emission rate associated with the transition is multiplied by $\eta_{\text{cav}} = 3Q\lambda^3/4\pi^2 v$ where Q is the cavity quality factor, v its volume, and λ the transition wavelength. This effect, first discussed in the context of radio frequencies by Purcell in 1946,[1] is due to the change of the number of radiator modes per unit volume and unit frequency induced by the presence of the cavity. It can equivalently be understood as resulting from the interaction between the atom and its electric images reflected in the cavity mirrors. This effect has never

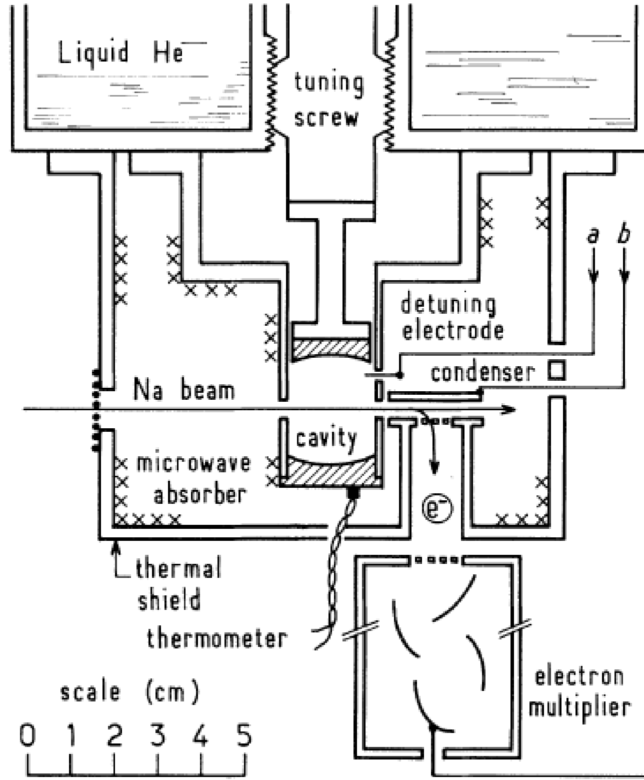


FIG. 1. Experimental arrangement.

Inhibited Spontaneous Emission by a Rydberg Atom

Randall G. Hulet,[a] Eric S. Hilfer, and Daniel Kleppner
Research Laboratory of Electronics and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 29 July 1985)

Spontaneous radiation by an atom in a Rydberg state has been inhibited by use of parallel conducting planes to eliminate the vacuum modes at the transition frequency. Spontaneous emission is observed to "turn off" abruptly at the cutoff frequency of the waveguidelike structure and the natural lifetime is measured to increase by a factor of at least 20.

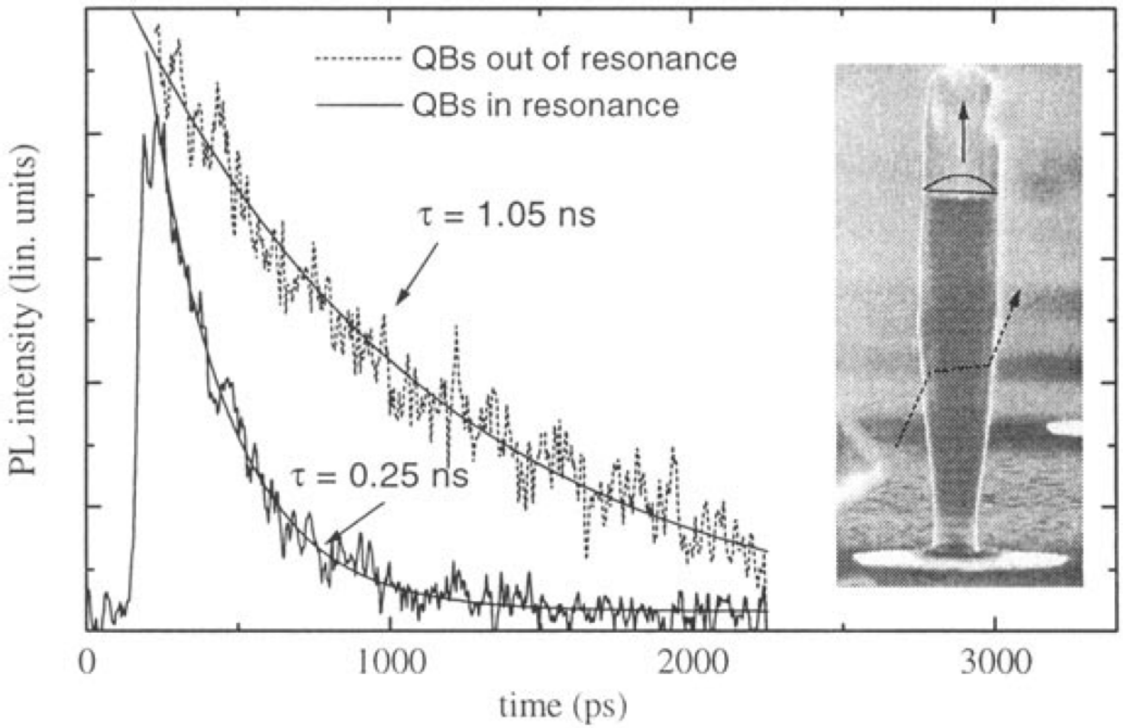
Spontaneous emission is often regarded as an unavoidable consequence of the coupling between matter and space. However, as one of the authors has pointed out,[1,2] by surrounding the atom with a cavity which has no modes at the transition frequency, spontaneous emission can be inhibited or "turned off." Drexhage, in studies of fluorescence by dye molecules deposited on a dielectric film over a conducting plane, observed a decrease of up to 25% in the fluorescent decay rate due to cavitylike effects.[3] Rydberg atoms provide the

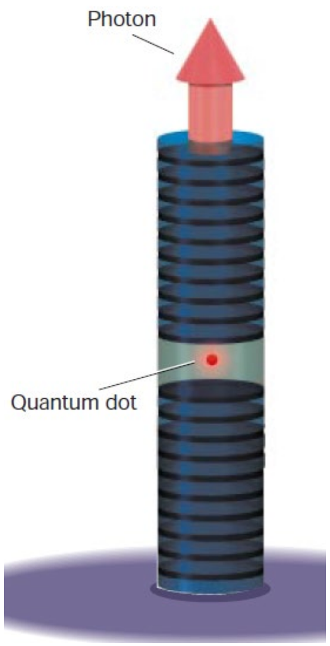
Micro-cavities in the 21st century

JOURNAL OF LIGHTWAVE TECHNOLOGY, VOL. 17, NO. 11, NOVEMBER 1999 2089

Strong Purcell Effect for InAs Quantum Boxes in Three-Dimensional Solid-State Microcavities

Jean-Michel Gérard and Bruno Gayral





Vahala, Nature 424, 839

Micro-cavities in the 21st century

How to squeeze more light out of a source:

HW3

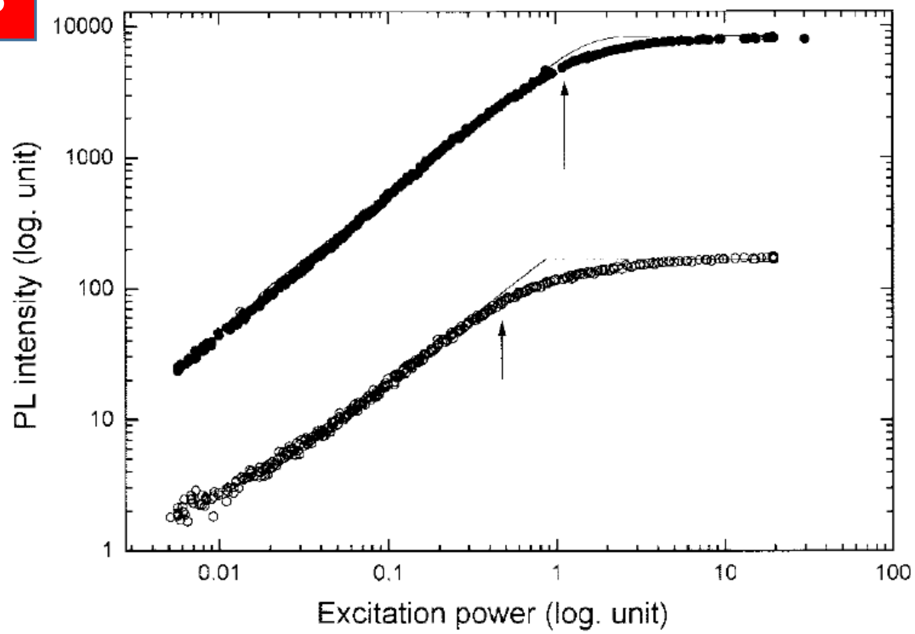
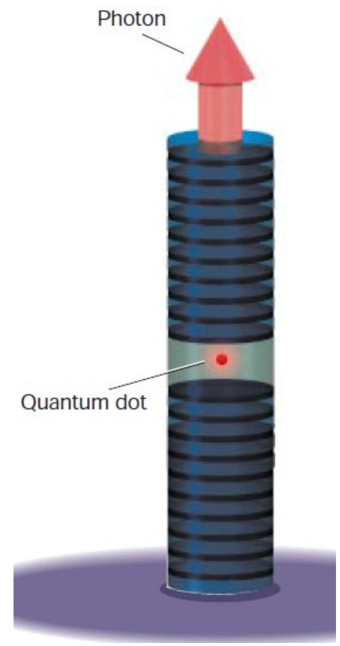


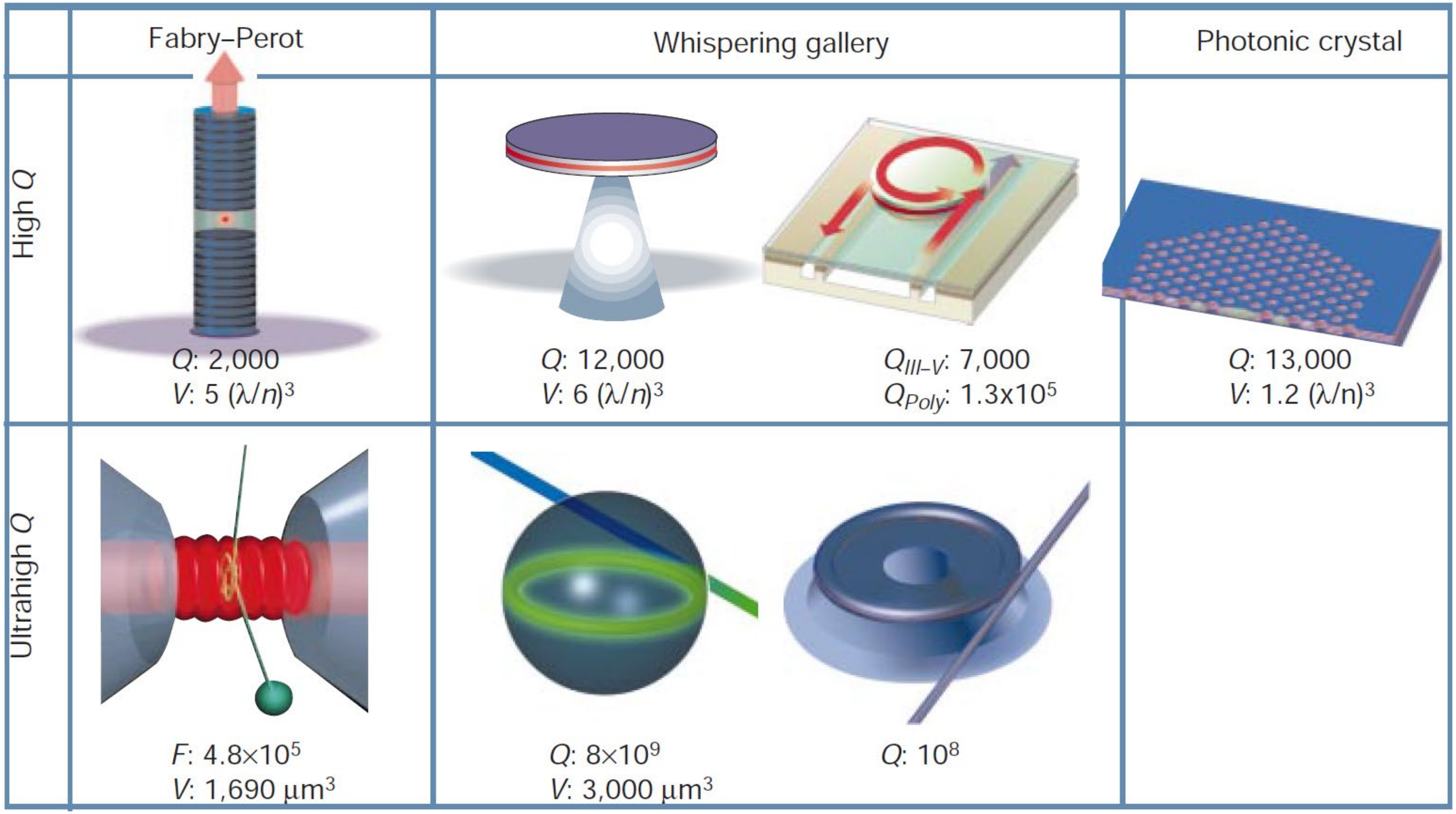
Fig. 3. PL intensity versus excitation power for a 1.8-μm diameter pillar, $(Q = 3000,\ F_p = 15)$. The saturation of the PL signal is observed both for on-resonance, (●) and off-resonance (○) QB's. The beginning of the saturation,

www.photonics.ethz.ch



Vahala, Nature 424, 839

Micro-cavities in the 21st century



Vahala, Nature 424, 839

A cavity is a tool to increase light-matter interaction.