Light-Matter Interaction on the Nanoscale

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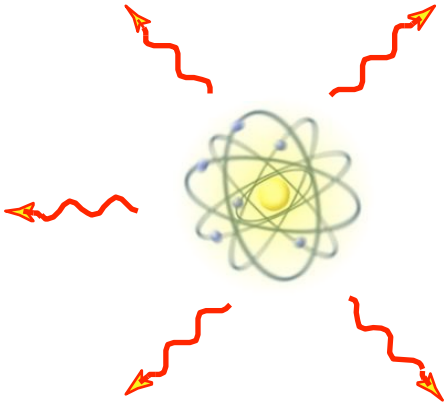
ETH Zurich



Outline

Classical dipoles

- Dipole radiation
- Power radiated by a classical dipole in an inhomogeneous environment
- The local density of optical states (LDOS)

Quantum emitters

- Lifetime of quantum emitters
- Fluorescence lifetime measurements
- Fermi's Golden Rule and decay rate engineering
- Examples: Microcavities, Optical antennas
- The second order correlation function

Part 1

CLASSICAL DIPOLES

Where does radiation come from?

- From the source terms in the inhomogeneous wave equation

$$\nabla \times \nabla \times \mathbf{E} + \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = -\mu_0 \frac{\partial}{\partial t}\left(\mathbf{j} + \frac{\partial \mathbf{P}}{\partial t} + \nabla \times \mathbf{M}\right)$$

- In the monochromatic case

$$\nabla \times \nabla \times \boldsymbol{E}(\boldsymbol{r}) - k^2 \boldsymbol{E}(\boldsymbol{r}) = \mathrm{i}\omega\mu_0\mu(\omega)\boldsymbol{j}_0(\boldsymbol{r})$$

For which source current distribution j(r) should we solve this equation?

The Green function

$$\nabla \times \nabla \times \boldsymbol{E}(\boldsymbol{r}) - k^2 \boldsymbol{E}(\boldsymbol{r}) = \mathrm{i}\omega\mu_0\mu(\omega)\boldsymbol{j}_0(\boldsymbol{r})$$

$$\mathcal{L}\,\mathbf{A}(\mathbf{r}) = \mathbf{B}(\mathbf{r})$$

Differential operator (given)

Field distribution (desired)

Source term (given)

$$\mathcal{L}\,\mathbf{G}_i(\mathbf{r},\mathbf{r}') = \mathbf{n}_i\,\delta(\mathbf{r}-\mathbf{r}') \qquad (i = x, y, z)$$

Green function solves operator $\mathcal{L}$ for δ-source

The Green function

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Green function solves operator $\mathcal{L}$ for δ-source

In matrix form:

$$\mathcal{L}\,\overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}') \;=\; \overleftrightarrow{\mathbf{I}}\;\delta(\mathbf{r}-\mathbf{r}')$$

What is so awesome about **G**?

$$\mathcal{L}\,\mathbf{A}(\mathbf{r}) = \mathbf{B}(\mathbf{r})$$

B is given, A is sought

Knowing **G**, we can calculate the field **A** for *any* source **B**!

$$\mathbf{A}(\mathbf{r}) = \int_V \vec{\mathbf{G}}\,(\mathbf{r},\mathbf{r}')\,\mathbf{B}(\mathbf{r}')\,dV' \qquad \mathbf{r} \notin V$$

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δ-function is mother of all source terms!
δ-source is the impulse, Green function the impulse response (in space)

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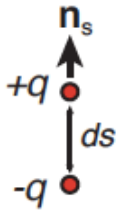
Back to the wave equation:

$$\nabla\times\nabla\times\boldsymbol{E}(\boldsymbol{r}) - k^2\boldsymbol{E}(\boldsymbol{r}) = \mathrm{i}\omega\mu_0\mu(\omega)\boldsymbol{j}_0$$

What is that?

So we need δ-current distribution here!

The oscillating dipole



$$\mathbf{p}(t) = q(t)\, d\mathbf{s} \quad \longrightarrow \quad \mathbf{j}(\mathbf{r},t) = \frac{\partial}{\partial t}\mathbf{p}(t)\,\delta(\mathbf{r}-\mathbf{r}_0)$$

Harmonic time dependence:

$$\mathbf{p}(t) = \mathrm{Re}\{\mathbf{p}\,\exp[-i\omega t]\} \quad \longrightarrow \quad \mathbf{j}(\mathbf{r}) = -i\omega\,\mathbf{p}\,\delta(\mathbf{r}-\mathbf{r}_0)$$

An oscillating dipole is a point-like time-harmonic current source.

The Green function of the wave equation

$$\nabla \times \nabla \times \overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}) - k^2 \overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}) = \mathrm{i}\omega\mu_0\mu(\omega)\underbrace{(-\mathrm{i}\omega p)\, \mathbb{1}\, \delta(\boldsymbol{r}-\boldsymbol{r}')}$$

$$\mathbf{j}(\mathbf{r}) = -i\omega\, \mathbf{p}\, \delta(\mathbf{r}-\mathbf{r}_0)$$

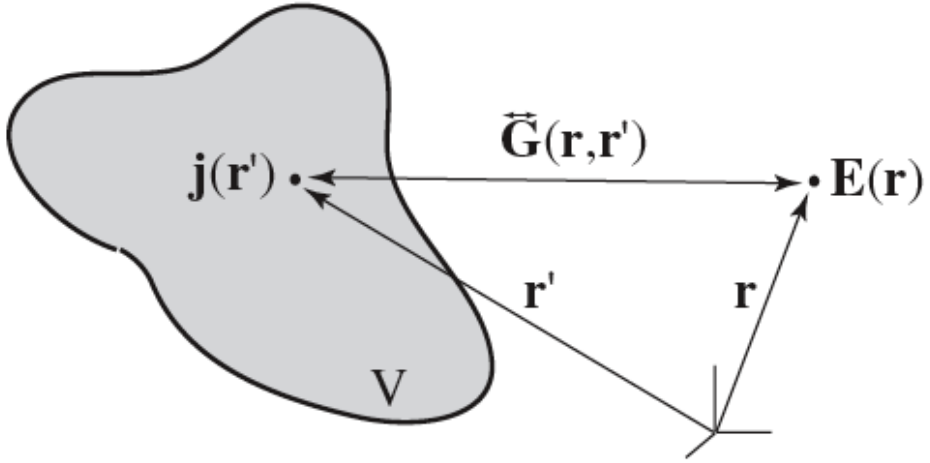
With **G** we can calculate the field distribution E of any current distribution j!

$$\mathbf{E}(\mathbf{r}) = i\,\omega\mu_0\mu \int_V \overleftrightarrow{\mathbf{G}}_0(\mathbf{r},\mathbf{r}')\,\mathbf{j}(\mathbf{r}')\,dV'$$

The Green function of the wave equation

$$\mathbf{E}(\mathbf{r}) = i\,\omega\mu_0\mu \int_V \overleftrightarrow{\mathbf{G}}_0(\mathbf{r},\mathbf{r}')\,\mathbf{j}(\mathbf{r}')\,dV' \qquad \mathbf{r}\notin V$$

For dipole: $\mathbf{j}(\mathbf{r}) = -i\omega\,\mathbf{p}\,\delta(\mathbf{r}-\mathbf{r}_0)$ $\longrightarrow$ $\mathbf{E}(\mathbf{r}) = \omega^2\mu_0\mu\,\overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}_0)\,\mathbf{p}$



The Green function for free space

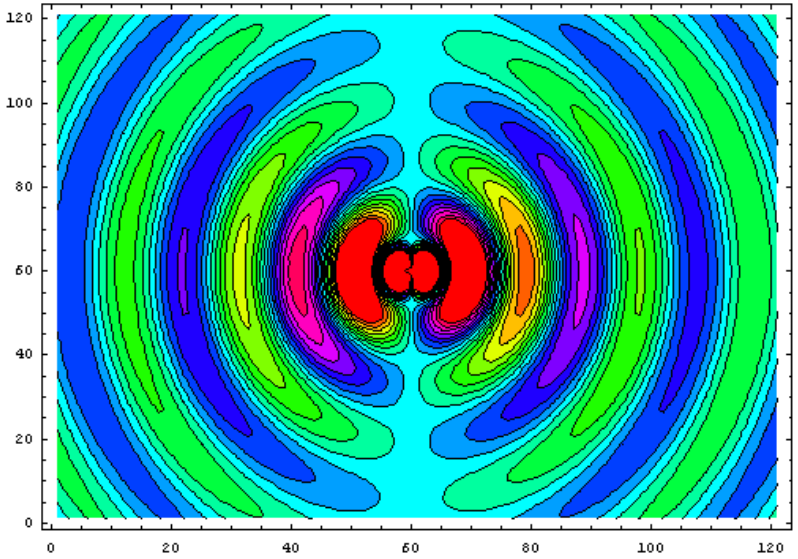
$$\nabla \times \nabla \times \overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}) - k^2 \overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}) = \mathrm{i}\omega\mu_0\mu(\omega)(-\mathrm{i}\omega p)\, \mathbb{1}\, \delta(\boldsymbol{r} - \boldsymbol{r}')$$

In cartesian coordinates and in a linear, homogeneous and isotropic medium:

$$\overleftrightarrow{\mathbf{G}}(\mathbf{r}, \mathbf{r}_o) = \frac{\exp(ikR)}{4\pi R}\left[\left(1 + \frac{ikR - 1}{k^2R^2}\right)\overleftrightarrow{\mathbf{I}} + \frac{3 - 3ikR - k^2R^2}{k^2R^2}\frac{\mathbf{RR}}{R^2}\right]$$

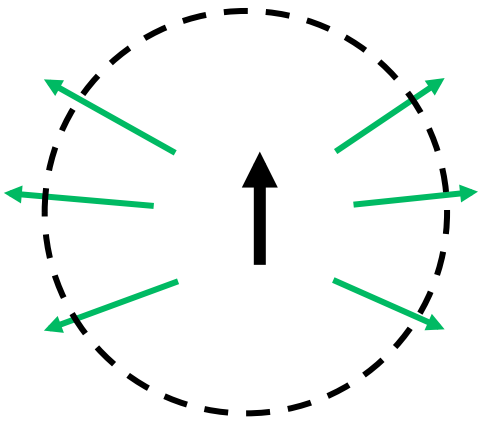
with $R = |\mathbf{r} - \mathbf{r}'|$

Dipole fields



Wikipedia.org

Power radiated by a dipole in free space

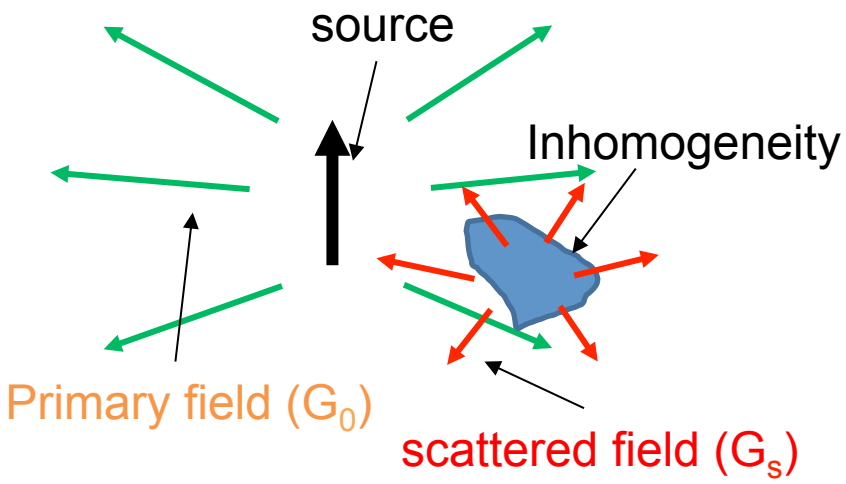


We calculated the power radiated by a dipole in free space by integrating the Poynting vector flux through a large sphere

$$\langle \boldsymbol{S}(\boldsymbol{r}) \rangle = \frac{1}{2} \mathrm{Re}\left[\boldsymbol{E}(\boldsymbol{r}) \times \boldsymbol{H}^*(\boldsymbol{r}) \right]$$

$$P = \int_{\partial S} r^2 \sin\theta \mathrm{d}\theta \mathrm{d}\phi \, \boldsymbol{n}_r \, \langle \boldsymbol{S}(\boldsymbol{r}) \rangle = \frac{|p|^2 \, \omega k^3}{12\pi\varepsilon_0 \varepsilon}$$

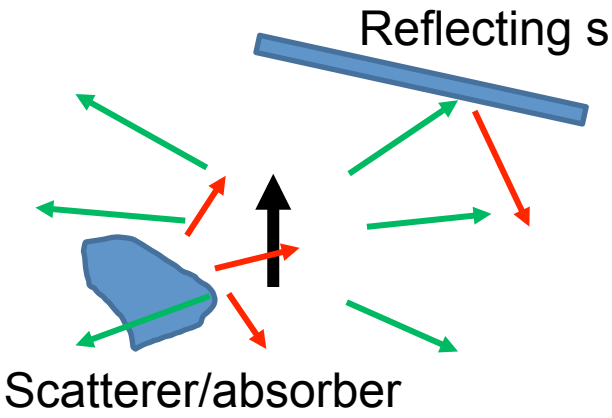
The Green function in an inhomogeneous environment



Split Green function of a complex photonic system into the “free-space part” and a “scattered part”.

$$\overleftrightarrow{\boldsymbol{G}} = \overleftrightarrow{\boldsymbol{G}}_0 + \overleftrightarrow{\boldsymbol{G}}_s$$

Power radiated in an inhomogeneous environment



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$$P = \int_{\partial S} r^2 \sin\theta \mathrm{d}\theta \mathrm{d}\phi \, \boldsymbol{n}_r \, \langle \boldsymbol{S}(\boldsymbol{r}) \rangle = \quad ?$$

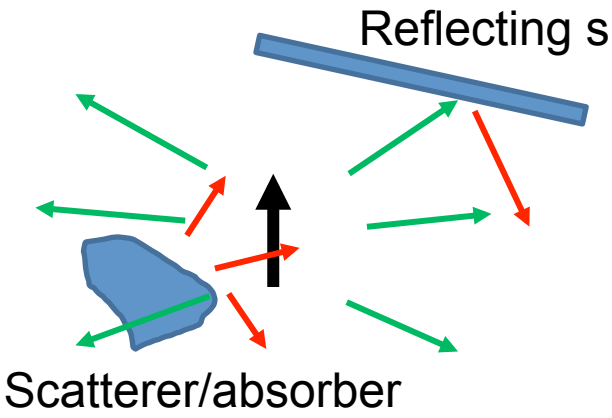
We could

- Make a huge sphere enclosing everything and integrate Poynting vector
- Make a very small sphere enclosing only the dipole and calculate the net Poynting flux

Both approaches are costly since we

- Need to perform integrations
- Might not be able to enclose the entire system

Power radiated in an inhomogeneous environment



$$\langle \boldsymbol{S}(\boldsymbol{r}) \rangle = \frac{1}{2} \mathrm{Re}\left[\boldsymbol{E}(\boldsymbol{r}) \times \boldsymbol{H}^*(\boldsymbol{r}) \right]$$

$$P = \int_{\partial S} r^2 \sin\theta \mathrm{d}\theta \mathrm{d}\phi \, \boldsymbol{n}_r \, \langle \boldsymbol{S}(\boldsymbol{r}) \rangle = \quad ?$$

We could

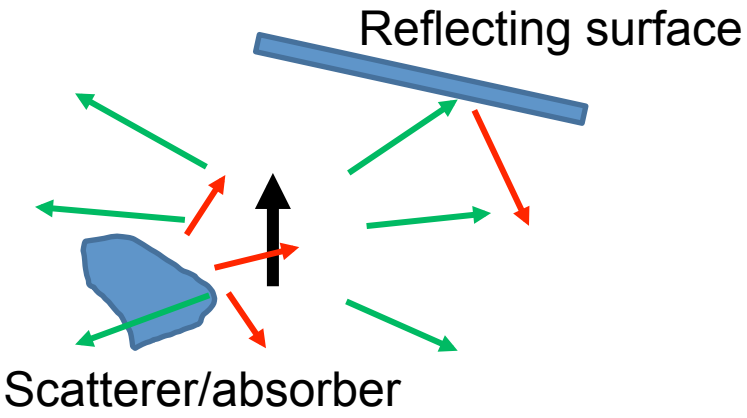
- Make a huge sphere enclosing everything and integrate Poynting vector
- Make [illegible] and calcu[illegible]

Is there an easier way?

Both approaches are costly since we

- Need to perform integrations
- Might not be able to enclose the entire system

Power radiated by a dipole



$$P = \frac{\mathrm{d}W}{\mathrm{d}t} = \boldsymbol{j} \cdot \boldsymbol{E}$$

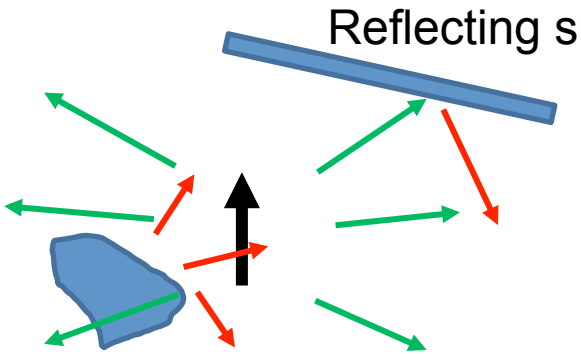
$$\langle P \rangle = \frac{\omega}{2} \mathrm{Im}(\boldsymbol{p}^* \cdot \boldsymbol{E})$$

$$\langle P \rangle = \frac{\omega}{2} |\boldsymbol{p}|^2 \left\{ \boldsymbol{n}_p^{\intercal} \, \mathrm{Im} \left[\omega^2 \mu_0 \mu \overleftrightarrow{\boldsymbol{G}} (\boldsymbol{r}_0, \boldsymbol{r}_0; \omega) \right] \boldsymbol{n}_p \right\}$$

$$\overleftrightarrow{\boldsymbol{G}} = \overleftrightarrow{\boldsymbol{G}}_0 + \overleftrightarrow{\boldsymbol{G}}_s$$

The energy radiated by a dipole equals the work done by the dipole's own field on the dipole itself!

Power radiated by a dipole



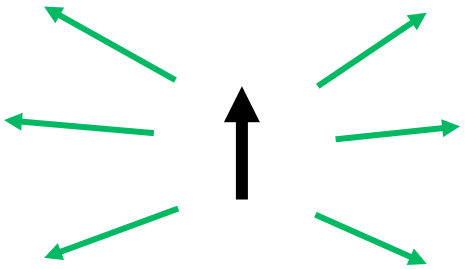
$$\langle P \rangle = \frac{\omega}{2} |\boldsymbol{p}|^2 \left\{ \boldsymbol{n}_p^{\mathsf{T}} \operatorname{Im} \left[\omega^2 \mu_0 \mu \overleftrightarrow{\boldsymbol{G}} (\boldsymbol{r}_0, \boldsymbol{r}_0; \omega) \right] \boldsymbol{n}_p \right\}$$

Radiated power is proportional to the local density of states (LDOS)

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

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

Power radiated by a dipole in free space



Same result as by integration of Poynting vector

In homogeneous medium:

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

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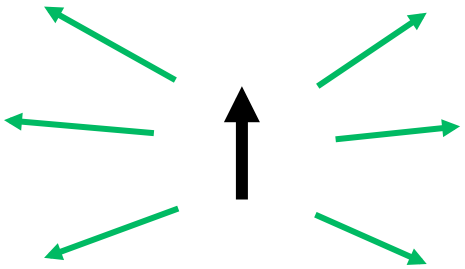

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Same result as by integration of Poynting vector

$$\langle P_0\rangle = \frac{\omega k^3}{12\pi\epsilon\epsilon_0}\,|\boldsymbol{p}|^2$$

Radiated power is proportional to the local density of states (LDOS)

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Power radiated by a dipole in an inhomogeneous environment

Via the local density of states (LDOS)

- Radiated power depends on location of source within its environment
- Radiated power depends on frequency of source
- Radiated power depends on orientation of source

The LDOS can be interpreted as a radiation resistance

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

Power enhancement provided by a photonic system

Normalize emitted power to power emitted in free space:

$$\frac{P}{P_0} = \frac{\boldsymbol{n}_p^\intercal \operatorname{Im}\left[\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}_0, \boldsymbol{r}_0; \omega)\right] \boldsymbol{n}_p}{\boldsymbol{n}_p^\intercal \operatorname{Im}\left[\overleftrightarrow{\boldsymbol{G}}_0(\boldsymbol{r}_0, \boldsymbol{r}_0; \omega)\right] \boldsymbol{n}_p} = 1 + \frac{\boldsymbol{n}_p^\intercal \operatorname{Im} \overleftrightarrow{\boldsymbol{G}}_s \boldsymbol{n}_p}{\operatorname{Im} G_0}$$

Depending on the sign (phase) of the scattered field returning to the dipole, it enhances or suppresses power dissipation.

Power enhancement provided by a photonic system

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Depending on the sign (phase) of the scattered field returning to the dipole, it enhances or suppresses power dissipation.

Warning: The term LDOS (enhancement) is used sloppily to refer to

$\boldsymbol{n}_p^\intercal \,\mathrm{Im}\left[\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}_0, \boldsymbol{r}_0; \omega)\right] \boldsymbol{n}_p$

$\dfrac{\boldsymbol{n}_p^\intercal \,\mathrm{Im}\,\overleftrightarrow{\boldsymbol{G}}_s \boldsymbol{n}_p}{\mathrm{Im}\, G_0}$

and more

$\dfrac{P}{P_0}$

$\mathrm{Im}\left[\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}_0, \boldsymbol{r}_0; \omega)\right]$

$\boldsymbol{n}_p^\intercal \,\mathrm{Im}\,\overleftrightarrow{\boldsymbol{G}}_s \boldsymbol{n}_p$

Radiation-matter interaction

Classical dipoles

- Dipole radiation
- Power radiated by a classical dipole in an inhomogeneous environment
- The local density of optical states (LDOS)

Quantum emitters

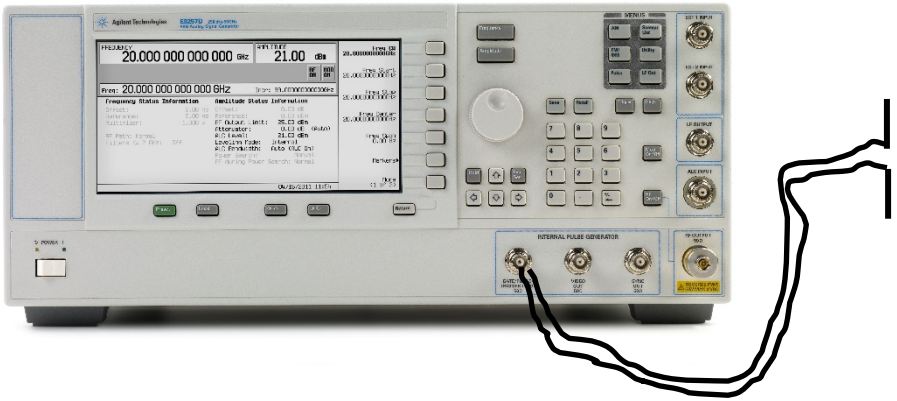
- Lifetime of quantum emitters
- Fluorescence lifetime measurements
- Fermi's Golden Rule and decay rate engineering
- Examples: Microcavities, Optical antennas
- The second order correlation function
- Resonant energy transfer

Part 2

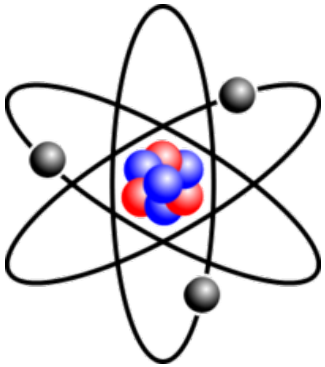
QUANTUM EMITTERS

Quantum emitters

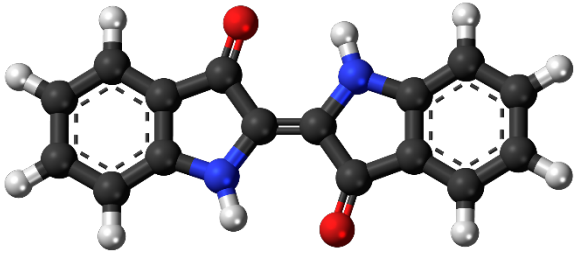
Radiating source up to GHz:



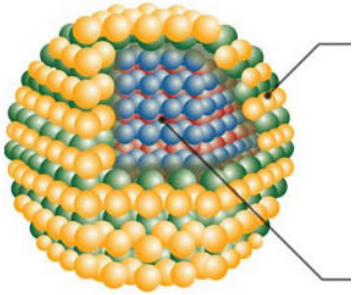
Radiating sources at 1000 THz (visible):



Atoms



Dye molecules



Quantum dots

Wikimedia; Emory.edu

Quantum emitters

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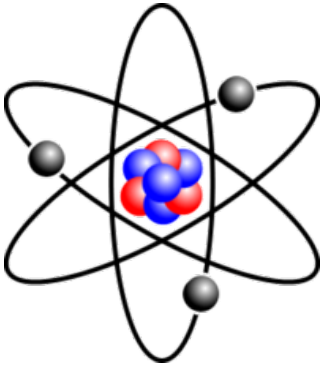
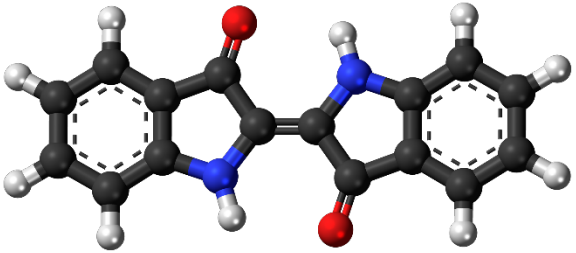
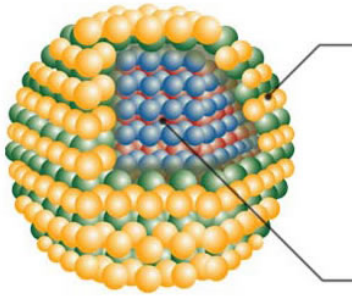


Atoms

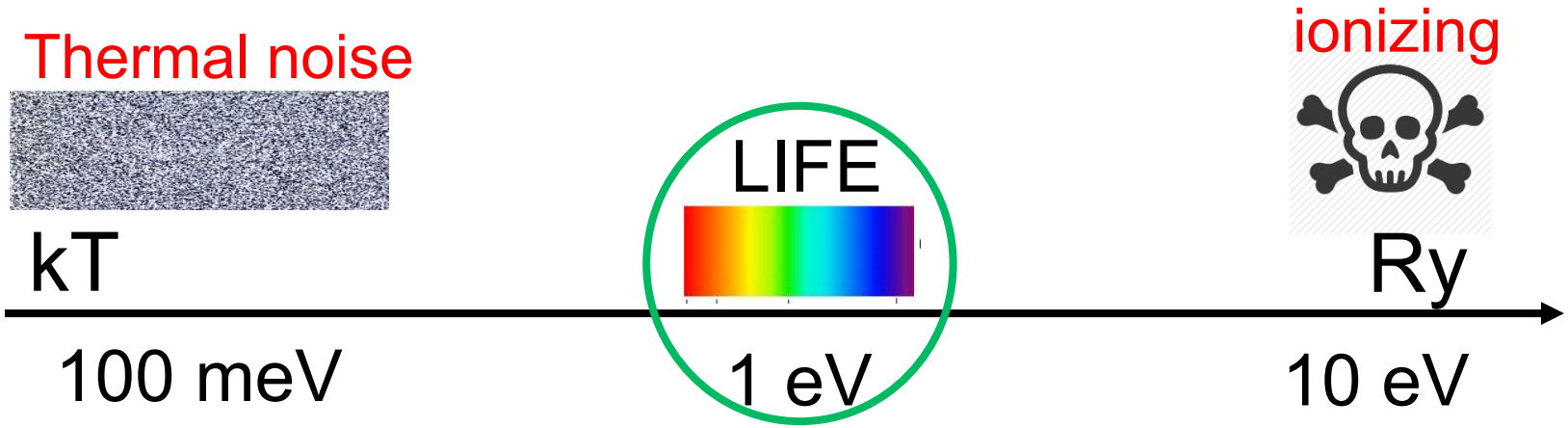


Dye molecules



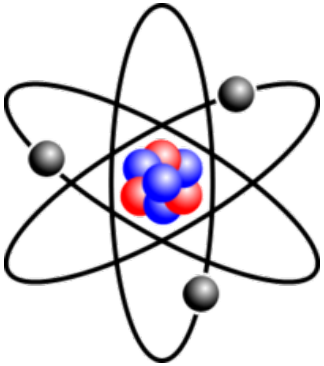
Quantum dots

"Visible optics" → E ~ 1 eV → λ ~ 1μm, "sub-λ optics" is "Nano-Optics"



Quantum emitters

Radiating sources at 1000 THz :



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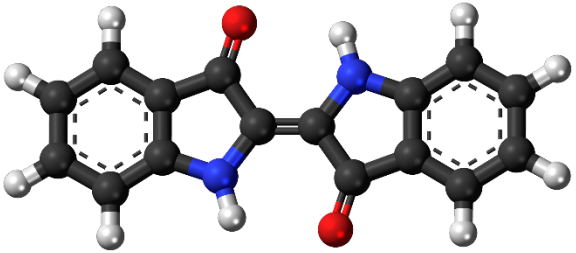

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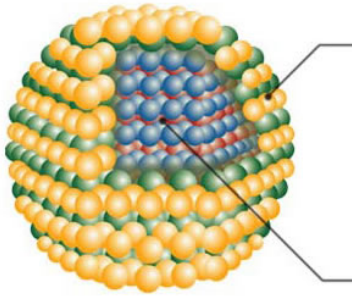

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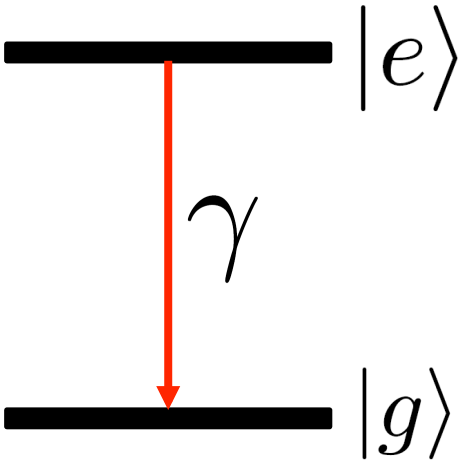

Why spontaneous emission?

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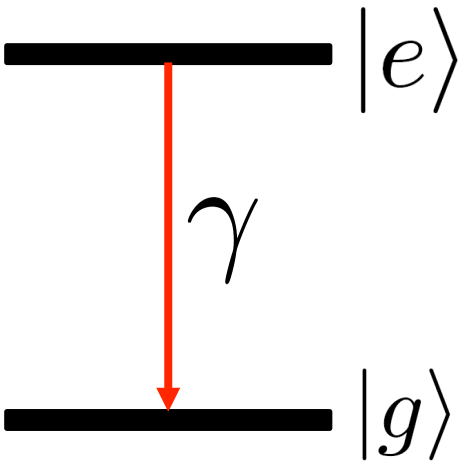
Department of Physics, University of Arkansas, Fayetteville, Arkansas 72701

(Received 3 March 1983; accepted for publication 24 June 1983)

This paper is a discussion of that perennial question, "Why does an excited atom radiate?" A satisfactory physical picture emerges when proper account is taken of the interplay between radiation reaction and the (quantum-mechanical) zero-point fluctuations of the radiation field. The fluctuation-dissipation connection between these two effects is therefore emphasized.

Quantum emitters

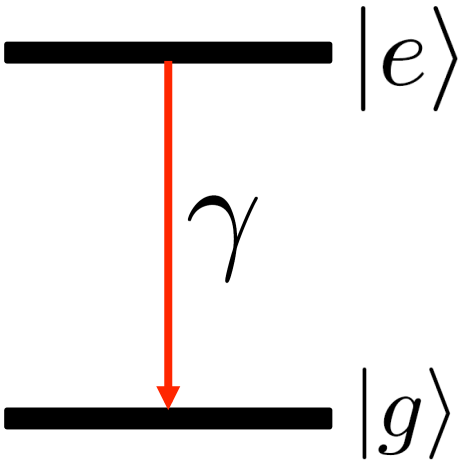


$$p_e(t) = p(0)\ \exp\left[-\gamma t\right]$$

Probability to find the system in the excited state decays exponentially with rate γ.

How can we measure the population of the excited state?

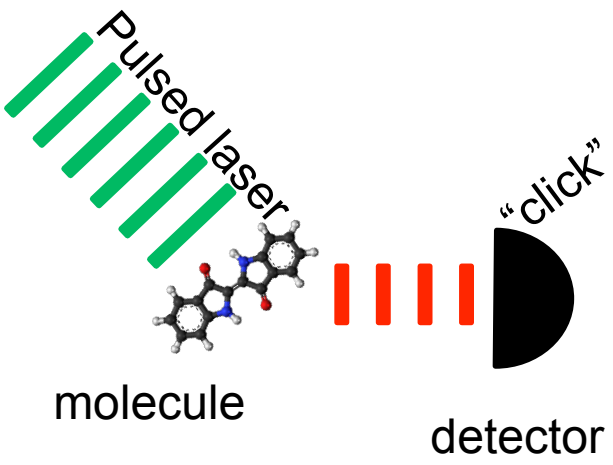
Fluorescence lifetime measurements

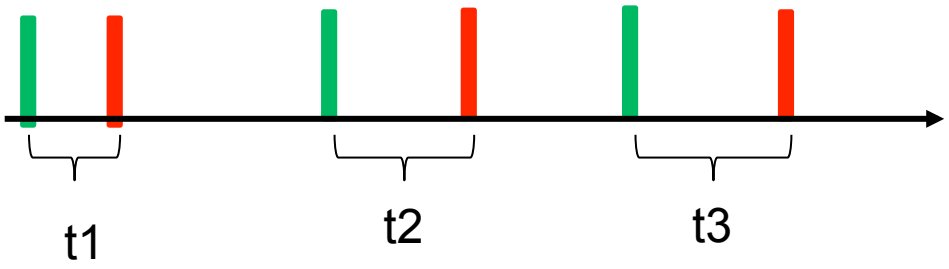


$$p_e(t) = p_e(0)\ \exp\left[-\gamma t\right]$$

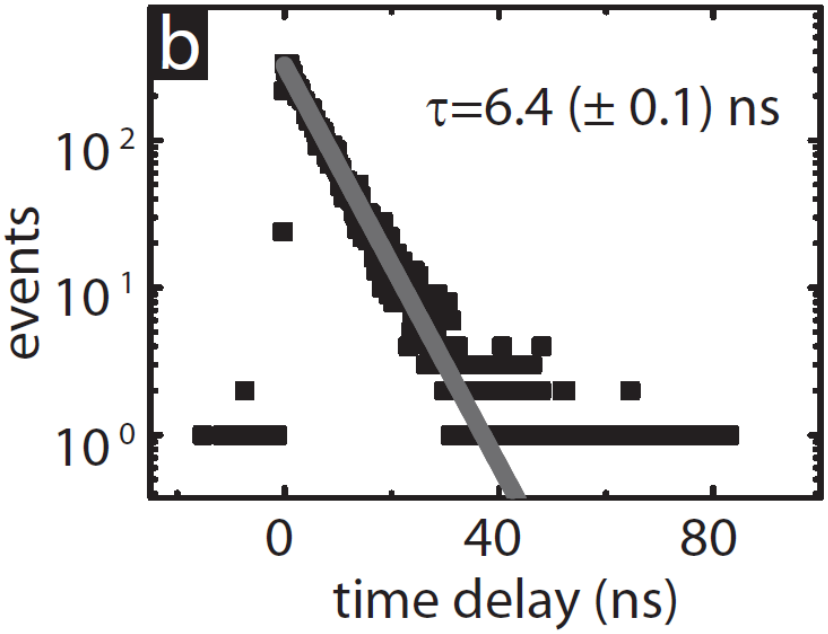
The probability to detect a photon at time t is proportional to p(t)!

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





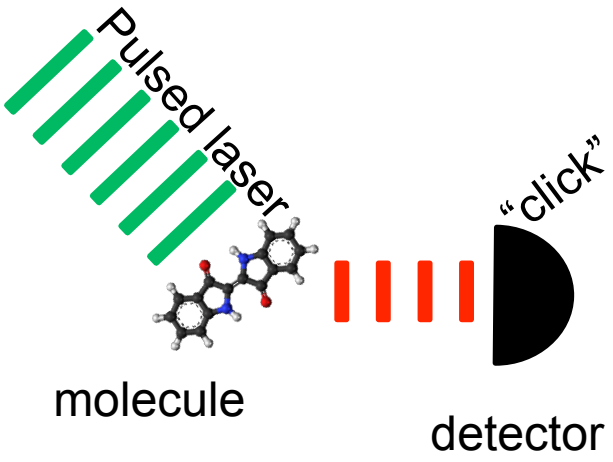
Fluorescence lifetime measurements

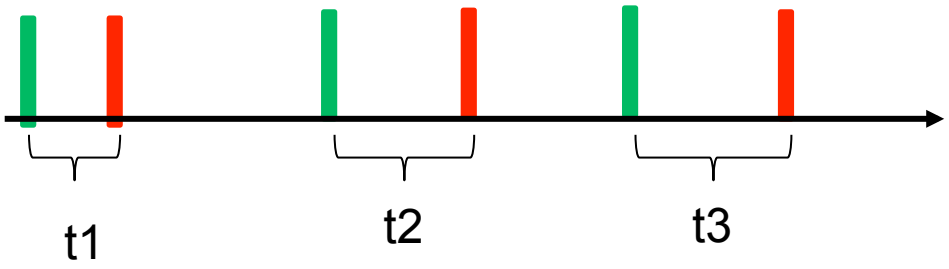


$$p_e(t) = p_e(0)\ \exp\left[-\gamma t\right]$$

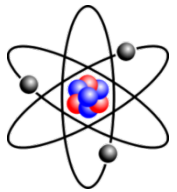
The probability to detect a photon at time t is proportional to p(t)!

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





(Digression) Fluorescence decay *cannot* be strictly exponential



Why not?

Because **exponential** decay arises from a **Lorentzian** lineshape.

$$p(t) = \int e^{-(iEt/\hbar)}\, \omega(E)\, dE$$

But the spectrum of a real system is *never* truly Lorentzian for all E.
⇒ Decay turns into a power law for long times.

(Digression) Fluorescence decay *cannot* be strictly exponential

SOVIET PHYSICS JETP VOLUME 6 (33), NUMBER 6 JUNE, 1958

CONTRIBUTION TO THE DECAY THEORY OF A QUASI-STATIONARY STATE

L. A. KHALFIN

Submitted to JETP editor April 23, 1957

J. Exptl. Theoret. Phys. (U.S.S.R.) 33, 1371-1382 (December, 1957)

Some general problems relating to the decay theory of a quasi-stationary state are considered. Dispersion relations are derived on the basis of the semi-finiteness of the energy distribution density $\omega(E)$. A criterion of physical feasibility in decay theory is formulated and studied on the basis of the Paily and Wiener theorem. It is shown that an exponential decay law cannot hold for all $\Gamma t/\hbar$. Corrections to the exponential decay law are computed under the simplest assumptions. Dispersion relations between the modulus and the phase of the function $p(t)$ are derived and investigated. On the basis of a knowledge of the decay law, these allow us to determine the energy distribution density $\omega(E)$ analytically. The results obtained are derived from the general laws of quantum mechanics and do not depend on the model of the decaying system.

Thus, we finally get for values of t that satisfy (3.17)

$$p(t) = \exp\{-iE_0 t/\hbar - \Gamma t/\hbar\} - \frac{i}{\pi}\frac{\Gamma\hbar}{(E_0^2+\Gamma^2)\,t}. \qquad (3.20)$$

Some remarks relative to the experimental proof. As is easily understood from the conclusions reached above, it is necessary to observe the decay, beginning with the initial instant of time $t = 0$. For this purpose we could use an experiment with the formation of an artificial, short-lived isotope, fixing the initial moment of time $t = 0$ by having a "pulse" of particles bombard a stable nucleus, a product of which is an artificial short-lived isotope. In this same experiment, we could observe the departure of the decay law from the exponential.

PRL 96, 163601 (2006) PHYSICAL REVIEW LETTERS week ending 28 APRIL 2006

Violation of the Exponential-Decay Law at Long Times

C. Rothe, S. I. Hintschich, and A. P. Monkman

Department of Physics, University of Durham, Durham, DH1 3LE, United Kingdom

(Received 4 July 2005; published 26 April 2006)

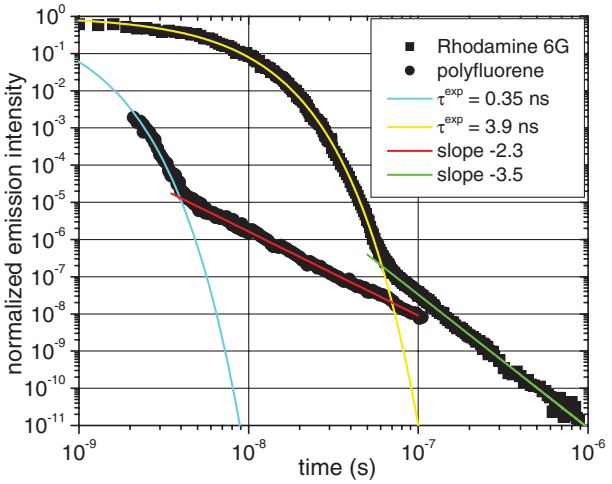
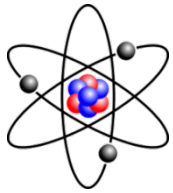


FIG. 2 (color). Corresponding double logarithmic fluorescence decays of the emissions shown in Fig. 1. Exponential and power law regions are indicated by solid lines and the emission intensity at time zero has been normalized.

Calculation of decay rate γ



Fermi's Golden Rule:

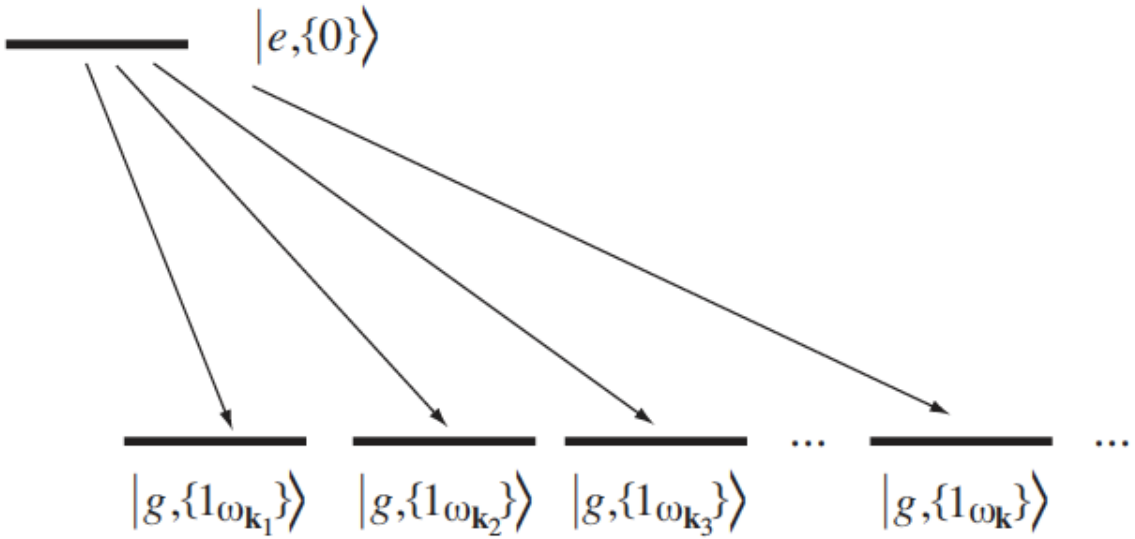
$$\gamma = \sum_f \frac{2\pi}{\hbar} \left| \langle f | \hat{\mathscr{H}} | i \rangle \right|^2 \delta(E_i - E_f)$$

Initial state (excited atom, no photon):

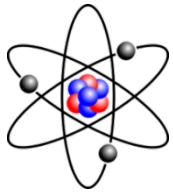
$$|i\rangle = |e, 0\rangle$$

Final state (de-excited atom, 1 photon in state k at frequency omega):

$$|f\rangle = |g, 1_{\omega_{\mathbf{k}}}\rangle$$



Calculation of decay rate γ



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$$|f\rangle = |g, 1_{\omega_{\mathbf{k}}}\rangle$$

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

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

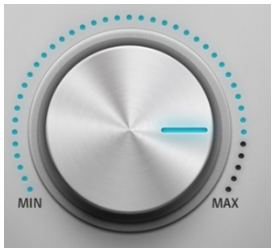
Atomic part:
transition dipole moment (quantum)

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

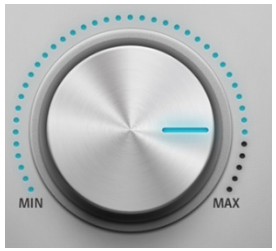
Field part:
Local density of states (classical)

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

Decay rate engineering



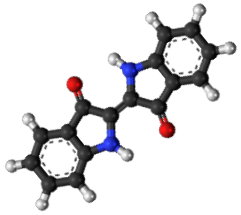
$$\gamma = \frac{\pi\omega}{3\hbar\epsilon_0}\left|\hat{\boldsymbol{p}}\right|^2 \rho_{\mathbf{n}}(\boldsymbol{r}_0,\omega)$$

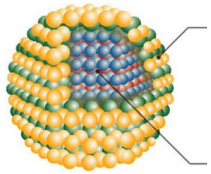


Emitter

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

Chemistry, material science

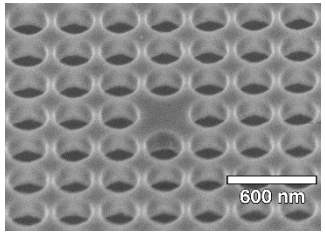


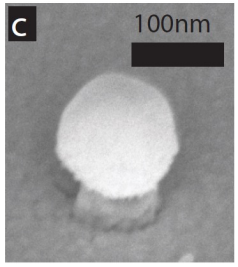


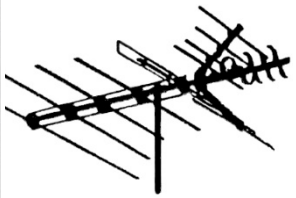
Environment

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

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

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

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.

Spontaneous emission control – Why?

1. Because it is awesome!

2. Some people like **bright** sources. Increase photon production rate of emitter by LDOS enhancement.

3. Some people like **efficient** sources. Increase quantum efficiency of emitter by LDOS enhancement.

4. Some people like to investigate the excited states of quantum emitters. Increase lifetime of excited state by LDOS *reduction*.

Drexhage's experiments (late 1960s)

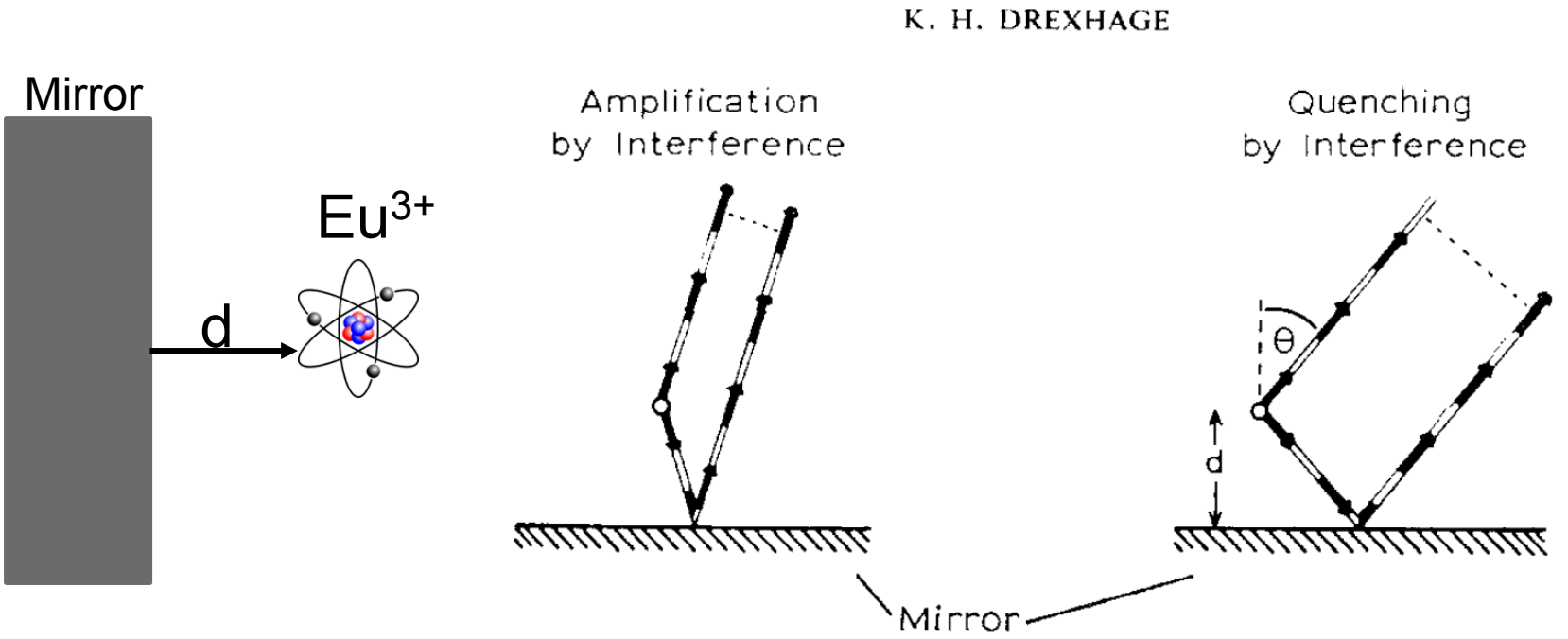
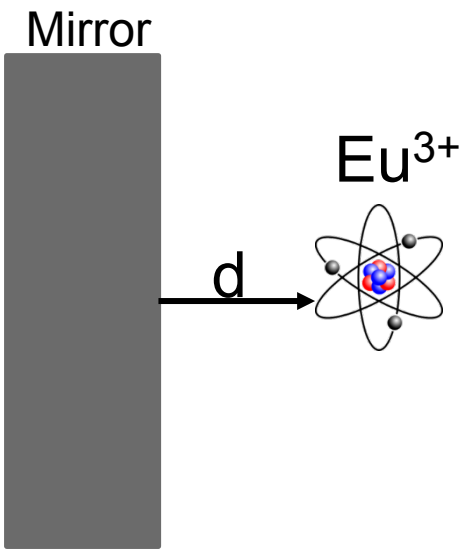
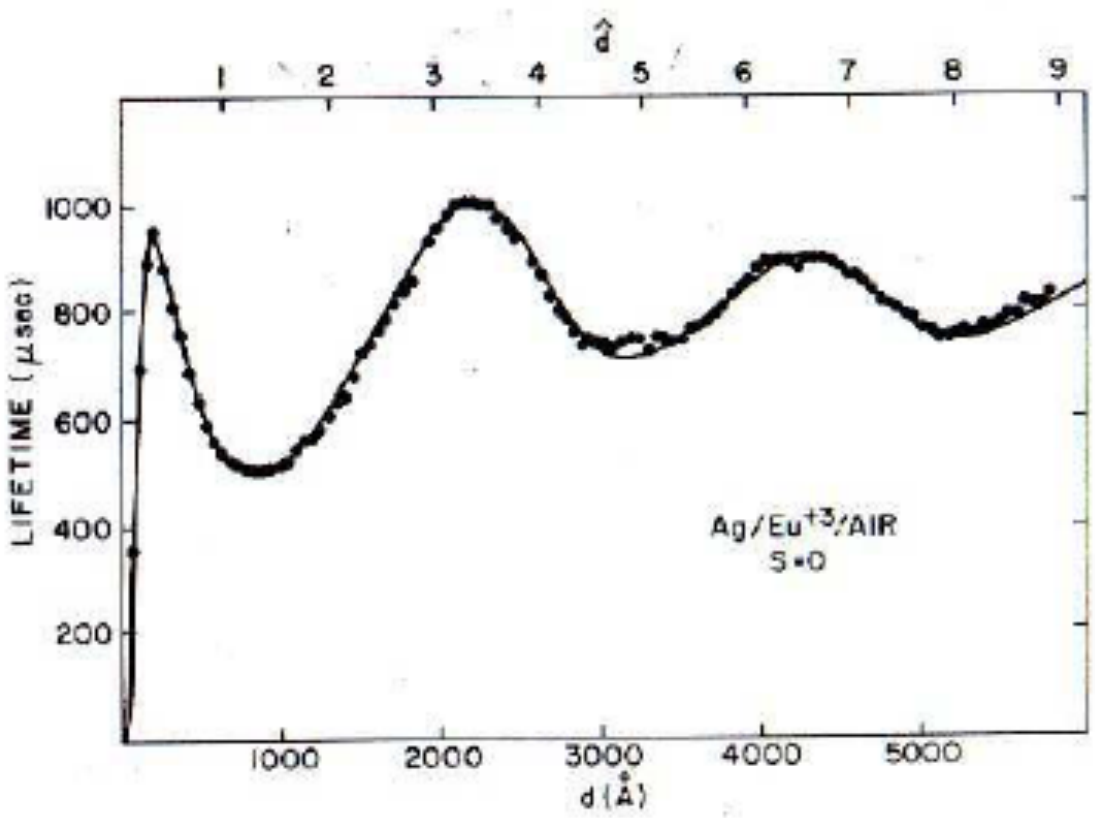


Fig. 1. Electric dipole radiation interacting with plane mirror.

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

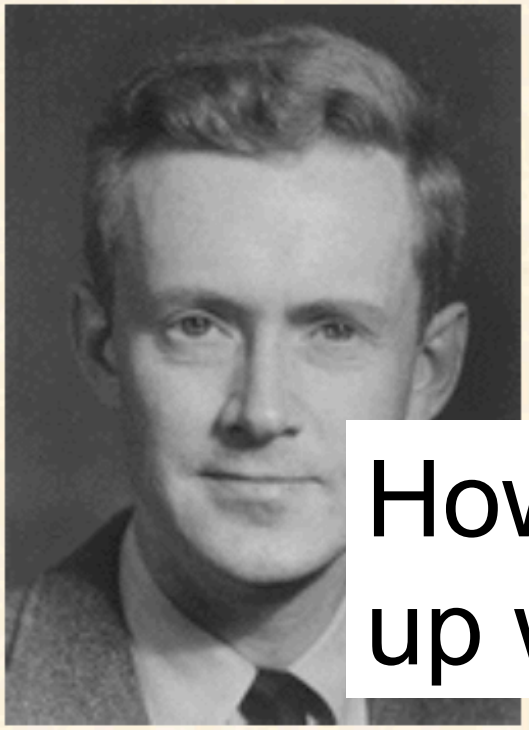
Drexhage's experiments (late 1960s)





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

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



Edward M. Purcell
1912-1997

How did he come up with that?

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

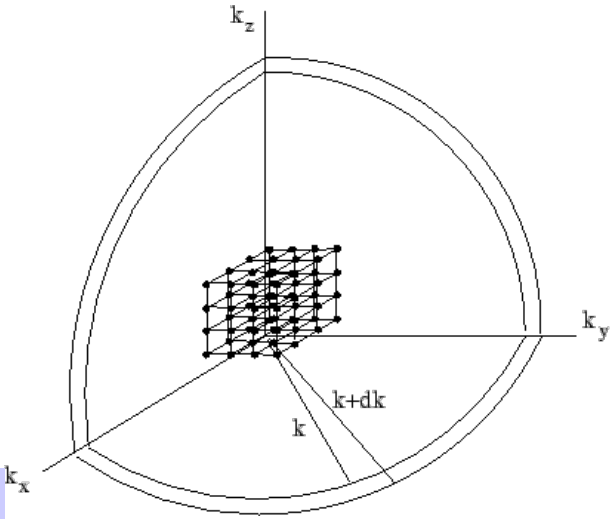
$$A_{\nu} = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2) \text{ sec.}^{-1},$$

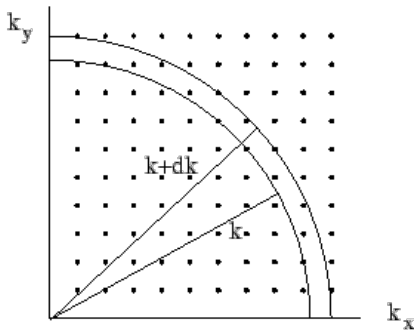
is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu = 10^7$ sec.$^{-1}$, $\mu = 1$ nuclear magneton, the corresponding relaxation time would be 5×10^{21} seconds! [illegible] to a resonant electrical [illegible] longer gives correctly the [illegible] per unit volume, in unit [illegible] now *one* oscillator in the [illegible] 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 equilib[illegible] in a time of the order of minutes, for $\nu = 10^7$ sec.$^{-1}$.

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

The Purcell effect

Look at modes in rectangular box with perfectly reflecting walls. Let box size go to infinity to approximate free space.





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

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

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

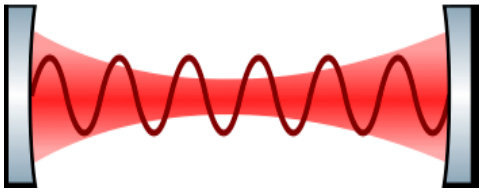
$$Q = \frac{\omega}{\Delta\omega}$$

The Purcell effect

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

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 $\eta_{cav} = 3Q\lambda^3/4\pi^2 v$ 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

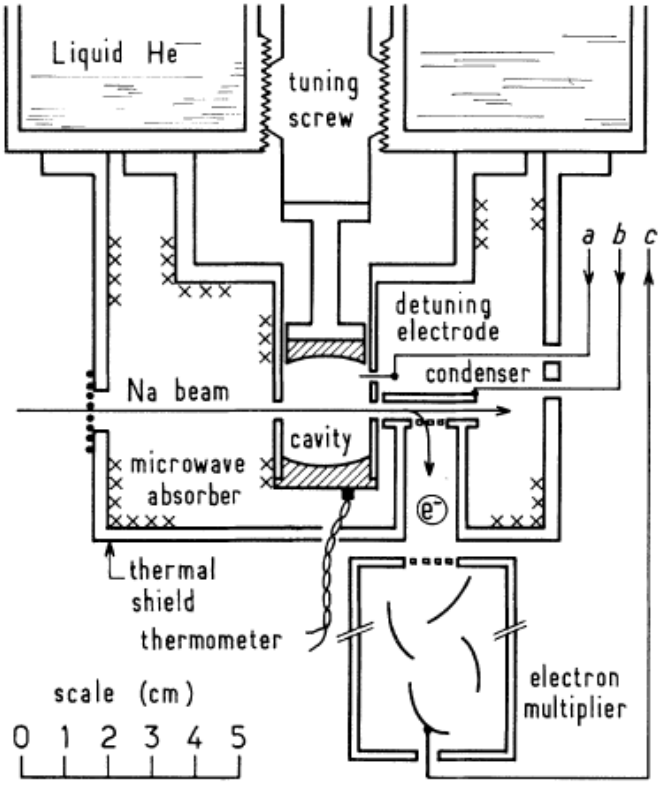


FIG. 1. Experimental arrangement.

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.

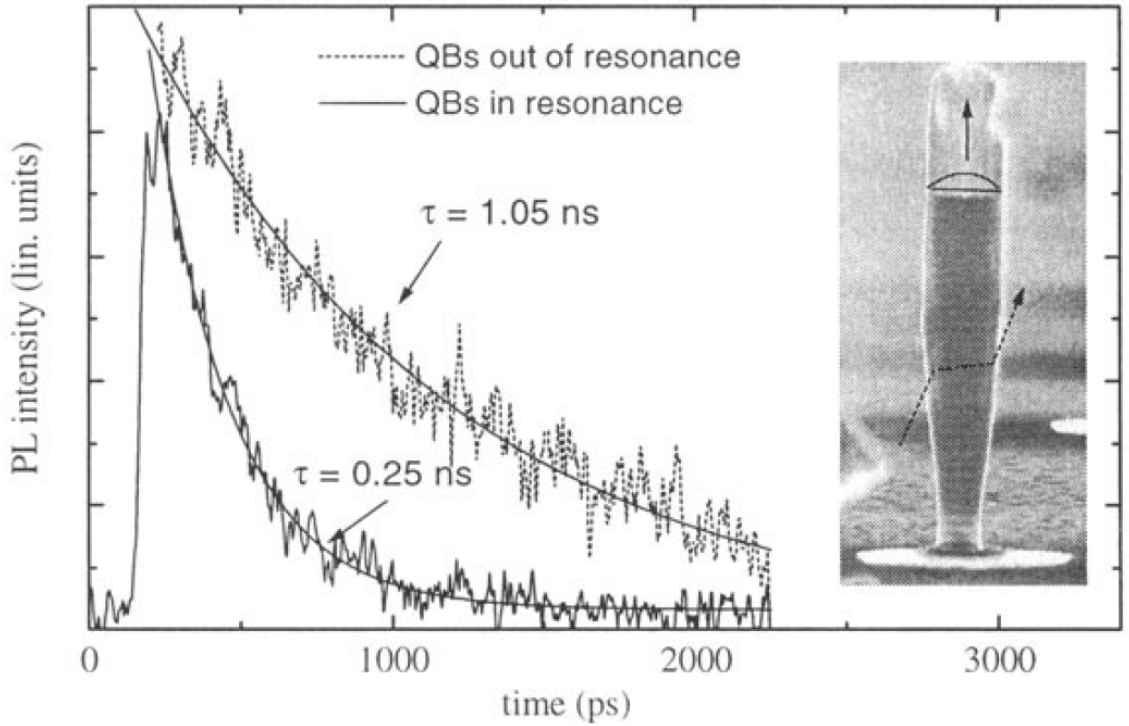
Spontaneous emission is often regarded as an unavoidable consequence of the coupling between matter and space. However, as one of the authors has pointed out,[1,2] by surrounding the atom with a cavity which has no modes at the transition frequency, spontaneous emission can be inhibited or "turned off." 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

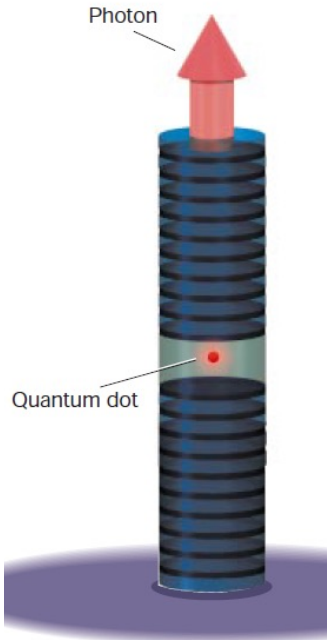
Micro-cavities in the 21st century - micropillars

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

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

Jean-Michel Gérard and Bruno Gayral





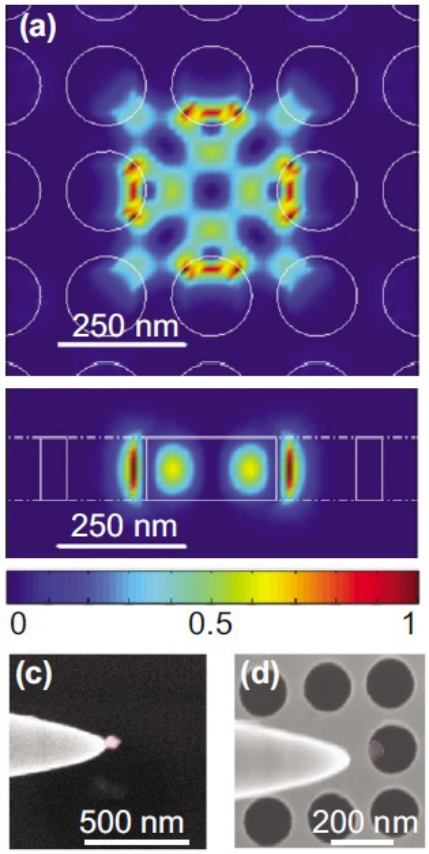
Vahala, Nature 424, 839

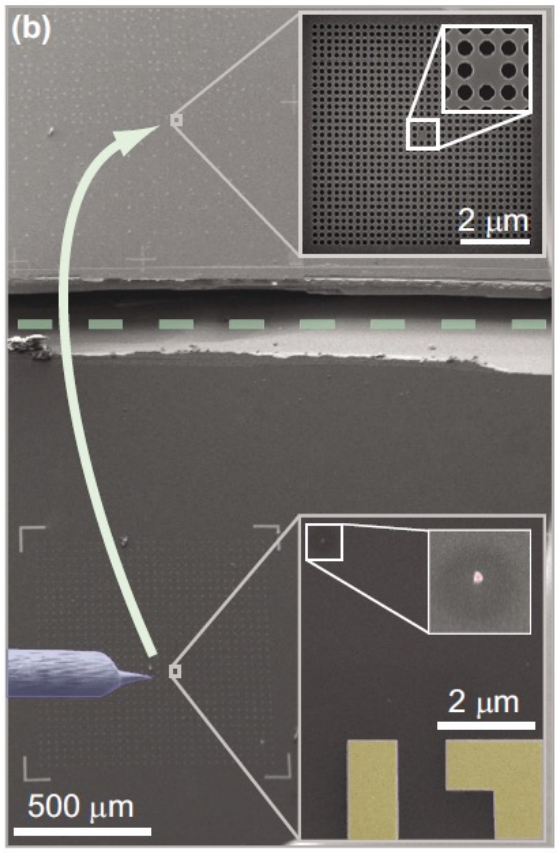
Micro-cavities in the 21st century – photonic crystals

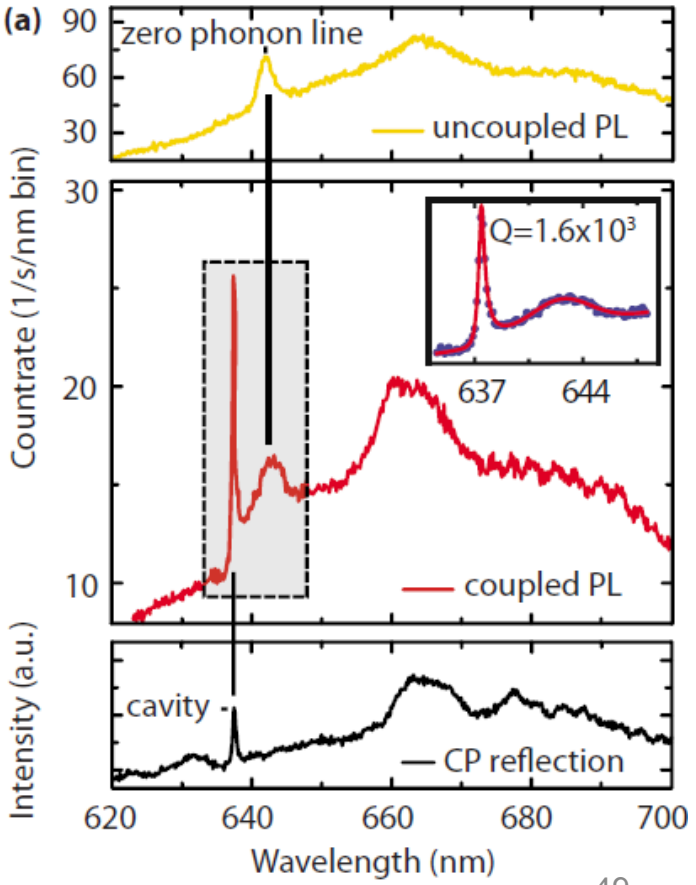
APPLIED PHYSICS LETTERS **98**, 193103 (2011)

Deterministic nanoassembly of a coupled quantum emitter–photonic crystal cavity system

T. van der Sar,[1,a)] J. Hagemeier,[2] W. Pfaff,[1] E. C. Heeres,[3] S. M. Thon,[2] H. Kim,[2,b)]
P. M. Petroff,[4] T. H. Oosterkamp,[3] D. Bouwmeester,[2,3] and R. Hanson[1]







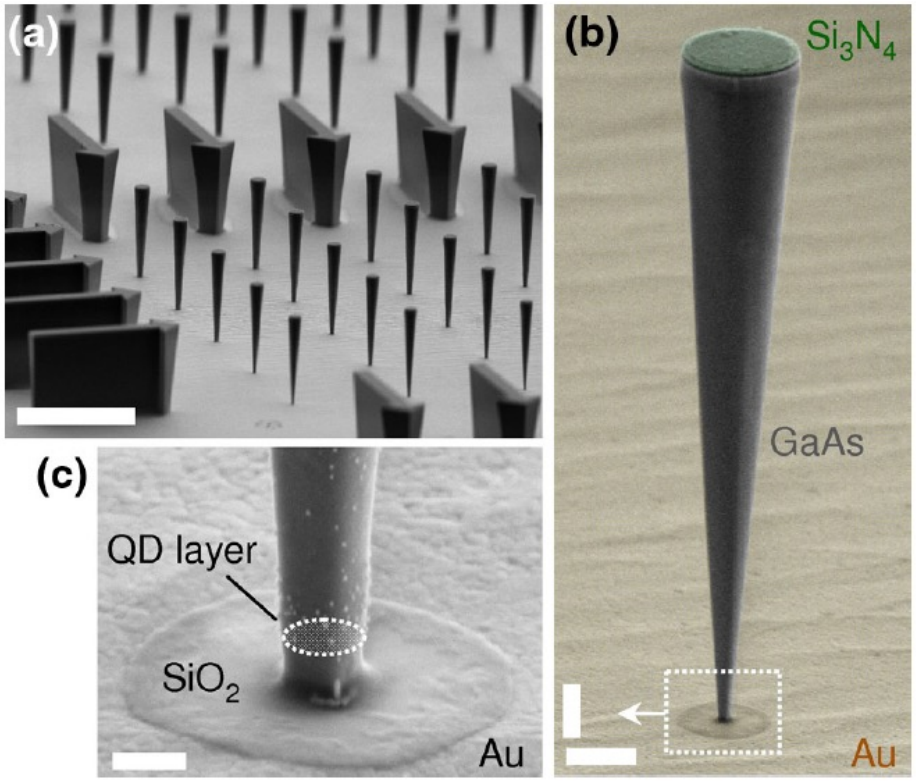
Antennas – resonators with engineered radiation loss

PRL **110,** 177402 (2013) PHYSICAL REVIEW LETTERS week ending 26 APRIL 2013

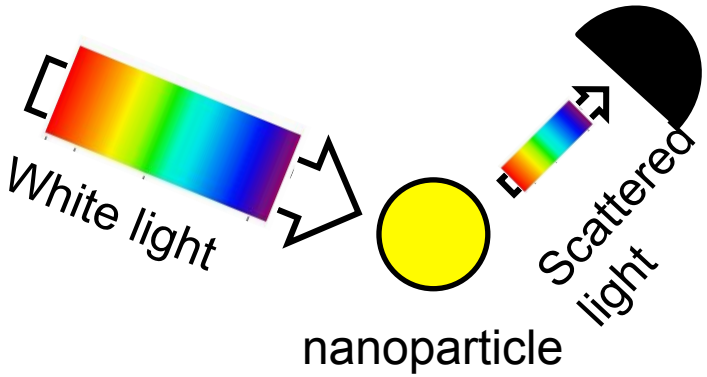
Dielectric GaAs Antenna Ensuring an Efficient Broadband Coupling between an InAs Quantum Dot and a Gaussian Optical Beam

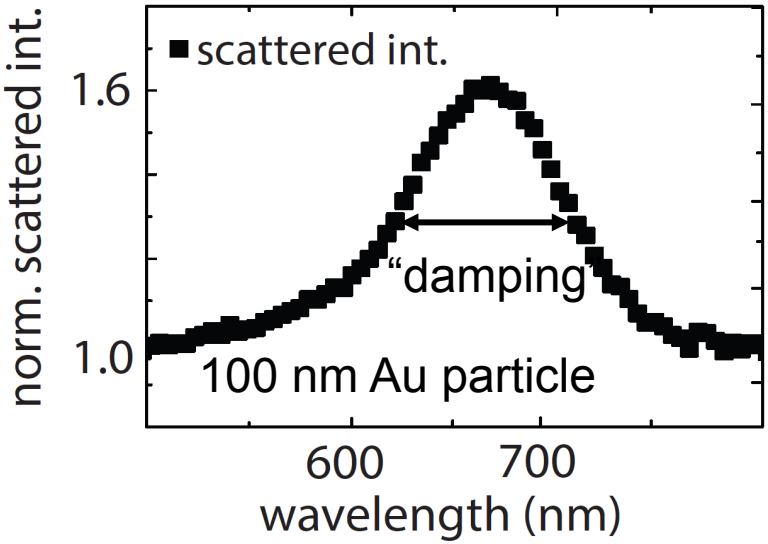
Mathieu Munsch, Nitin S. Malik, Emmanuel Dupuy, Adrien Delga, Joël Bleuse, Jean-Michel Gérard, and Julien Claudon*
CEA-CNRS-UJF Group, Nanophysique et Semiconducteurs, CEA, INAC, SP2M, F-38054 Grenoble, France

Niels Gregersen and Jesper Mørk
Department of Photonics Engineering, DTU Fotonik, Technical University of Denmark, Building 343, 2800 Kongens Lyngby, Denmark



Nanoparticles: resonators at optical frequencies





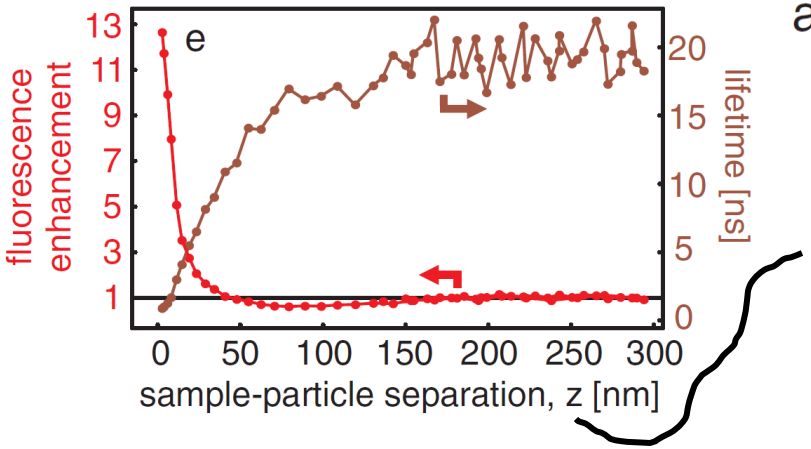
- Metal nano-particles show resonances in the visible

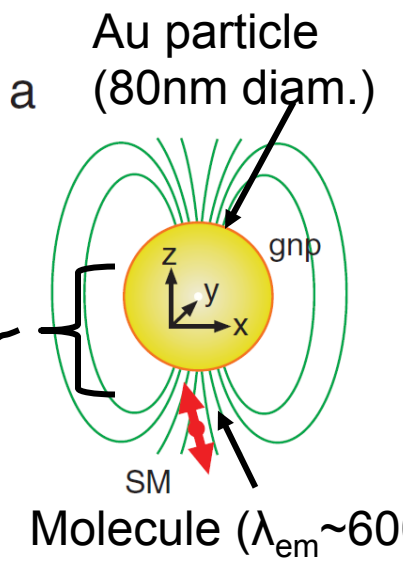
Lycurgus Cup (glass with metal nano-particles):
Green when front lit→
← Red when back lit

How does that work?

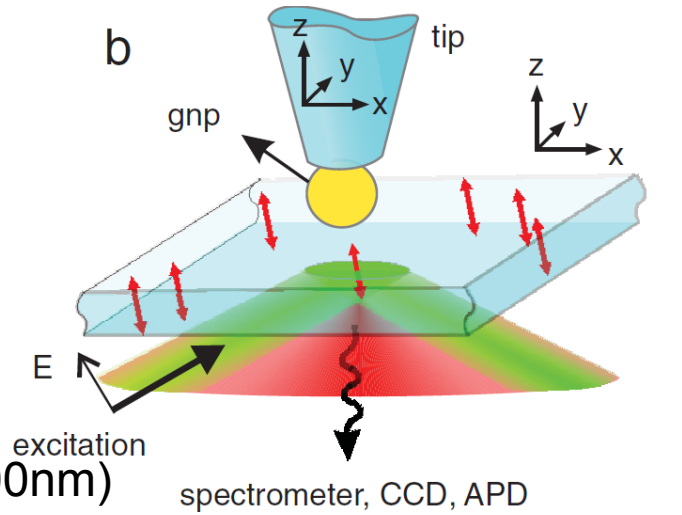
Wikipedia.org

Optical antennas

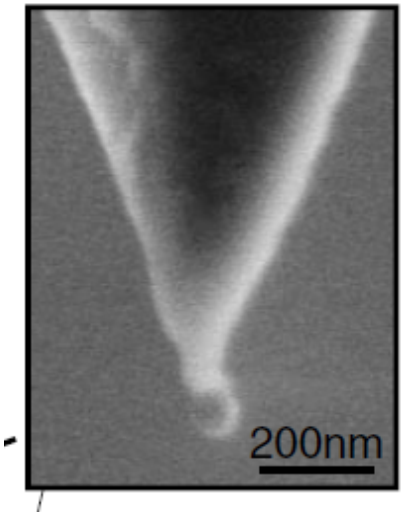




Kühn et al., PRL 97, 017402 (2006)



- Metallic nanoparticles can act as “antennas” and boost decay rate of quantum emitters in their close proximity
- Effect confined to length scale of order λ/10

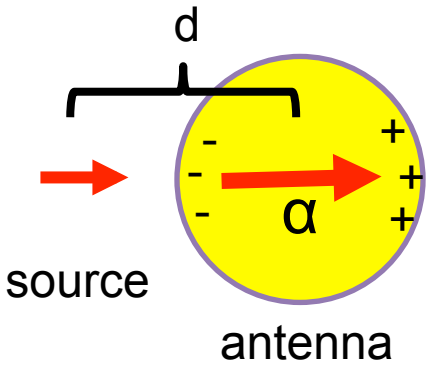


Anger et al., PRL 96, 113002 (2006)

Optical antennas – an intuitive approach

$$\boldsymbol{p}_{\text{ind}} = \alpha \boldsymbol{E}_s(\boldsymbol{r}_{\text{ant}})$$

$$\boldsymbol{E}_s \propto 1/d^3$$



$$P \propto |\boldsymbol{p}_{\text{ind}}|^2 \propto \frac{|\alpha|^2}{d^6}$$

- Assume an oscillating dipole close to a polarizable particle
- Assume that particle is small enough to be described as dipole
- Assume distance d<<λ, near field of source polarizes particle
- If polarizability α is large, antenna dipole largely exceeds source dipole
- Radiated power dominated by antenna dipole moment

Optical antenna is a dipole moment booster!

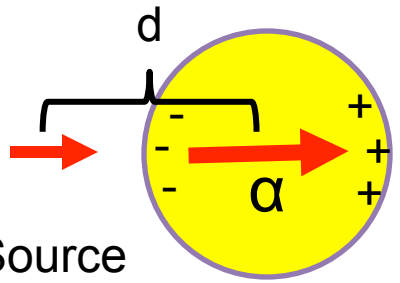
Optical antennas – a cleaner derivation

Calculate rate enhancement via power enhancement

$$\langle P \rangle = \frac{\omega}{2} \mathrm{Im}\left[\boldsymbol{p}^* \cdot \boldsymbol{E}(\boldsymbol{r}_0)\right]$$

$$\boldsymbol{E}(\boldsymbol{r}_0) = \overleftrightarrow{\underline{G}}_0(\boldsymbol{r}_0, \boldsymbol{r}_0)\boldsymbol{p}_0 + \overleftrightarrow{\underline{G}}_0(\boldsymbol{r}_0, \boldsymbol{r}_{\mathrm{ant}})\alpha\, \overleftrightarrow{\underline{G}}_0(\boldsymbol{r}_{\mathrm{ant}}, \boldsymbol{r}_0)\boldsymbol{p}_0$$

$\overleftrightarrow{\boldsymbol{G}}_s$



$$\mathrm{Re}\underline{G}_0^{ii}(\boldsymbol{r} \to \boldsymbol{r}_0) \to \frac{1}{2\pi k^2 d^3}$$

$$\mathrm{Im}\,\underline{G}_0^{ii}(\boldsymbol{r} \to \boldsymbol{r}_0) \to \frac{\omega^2 \mu_0 \mu k}{6\pi}$$

$$\mathrm{Im}\left[\boldsymbol{p}^* \cdot \overleftrightarrow{\underline{G}}_0(\boldsymbol{r}_0, \boldsymbol{r}_{\mathrm{ant}})\alpha\, \overleftrightarrow{\underline{G}}_0(\boldsymbol{r}_{\mathrm{ant}}, \boldsymbol{r}_0)\boldsymbol{p}_0\right] \longrightarrow |\boldsymbol{p}|^2\, \mathrm{Re}\underline{G}_0^{ii}(d)\, \mathrm{Im}\,\alpha\, \mathrm{Re}\underline{G}_0^{ii}(d)$$

$$\frac{P}{P_0} = 1 + \frac{A}{d^6}\frac{\mathrm{Im}\,\alpha}{\mathrm{Im}\,\underline{G}_0}$$

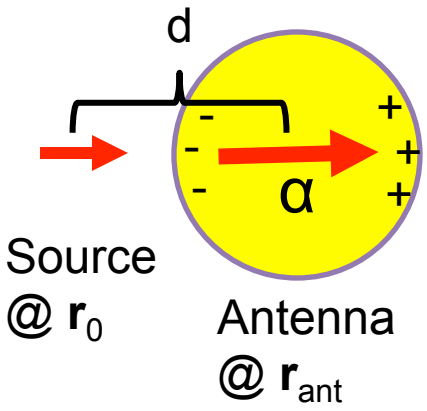
A=const.

- Field at source is primary field + field generated by induced antenna dipole
- Assume source is close to particle (near-field terms dominate)
- Close to source ReG along dipole axis diverges as $1/d^3$, ImG is constant

Optical antennas – a cleaner derivation

Calculated rate enhancement (equals power enhancement):

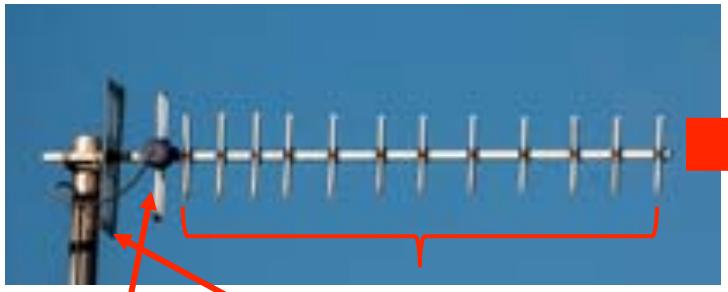
$$\frac{P}{P_0} = 1 + \frac{A}{d^6} \frac{\mathrm{Im}\,\alpha}{\mathrm{Im}\,\underline{G}_0}$$



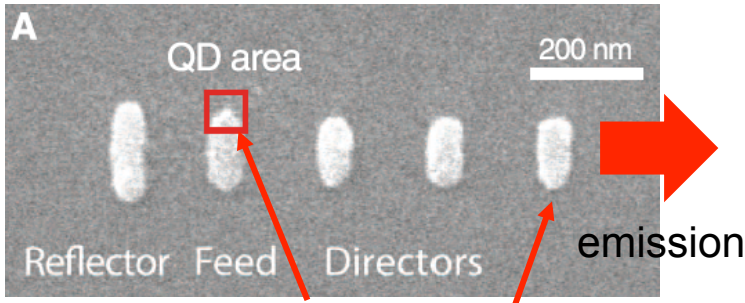
- Rate enhancement goes with the imaginary part of polarizability
- Rate enhancement goes with inverse source-antenna distance d^{-6}

Optical antennas for directional photon emission

Yagi and Uda (1920s)



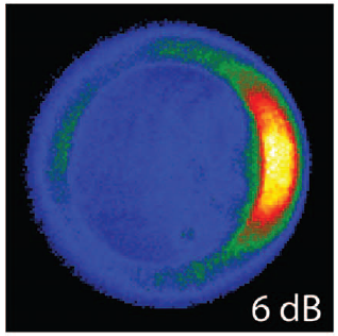
Curto et al., Science 329, 930 (2010)

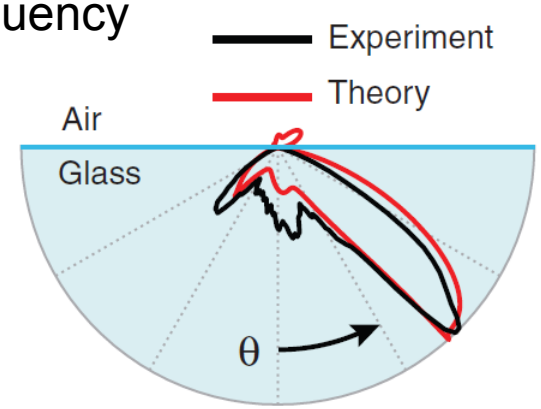


10^6

Scale down size, scale up frequency

- Optical antennas allow control of directionality of light emission for quantum emitters
- Antenna is photonic environment that offers a large density of states with specific k-vector





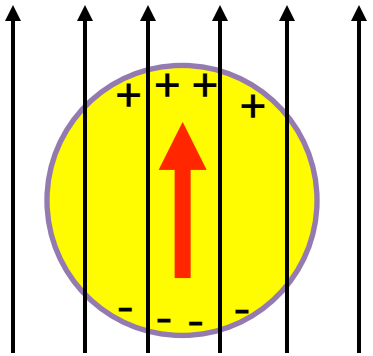
The electrodynamic polarizability

$$\boldsymbol{p}_{\text{stat}} = \boldsymbol{\alpha}_0 \boldsymbol{E}_{\text{stat}}(\boldsymbol{r}_0) \qquad \boldsymbol{p} = \boldsymbol{\alpha}_{\text{eff}} \boldsymbol{E}_{\text{ext}}$$

$$\boldsymbol{p} = \boldsymbol{\alpha}_0 \left[\boldsymbol{E}_{\text{ext}}(\boldsymbol{r}_0) + \overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}_0, \boldsymbol{r}_0)\boldsymbol{p} \right]$$

$$\boldsymbol{\alpha}_{\text{eff}}^{-1} = \boldsymbol{\alpha}_0^{-1} - \underline{\overleftrightarrow{\boldsymbol{G}}}(\boldsymbol{r}_0, \boldsymbol{r}_0)$$

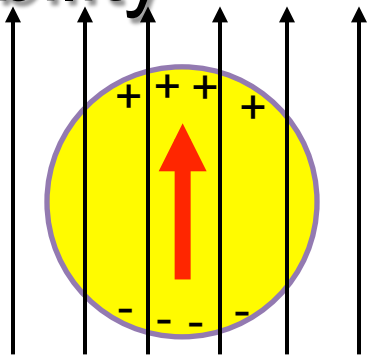
$$\underline{\overleftrightarrow{\boldsymbol{G}}} = \cancel{\text{Re}\,\underline{\overleftrightarrow{\boldsymbol{G}}}_0} + \text{Re}\,\underline{\overleftrightarrow{\boldsymbol{G}}}_s + \text{i}\left[\text{Im}\,\underline{\overleftrightarrow{\boldsymbol{G}}}_0 + \text{Im}\,\underline{\overleftrightarrow{\boldsymbol{G}}}_s \right]$$



- Static polarizability: induced dipole moment due to static E-field
- Dynamic case: additional field generated by induced dipole moment
- Define effective electrodynamic polarizability “dressed” with Green function
- ReG_0 diverges at origin! Fact that we describe the scatterer as a mathematical point backfires. Need to fit experimentally found resonance frequency.
- ReG_s shifts resonance frequency depending on environment.
- ImG represents radiation damping term: essential for energy conservation

The electrodynamic polarizability

$$\boldsymbol{\alpha}_{\mathrm{eff}}^{-1} = \boldsymbol{\alpha}_0^{-1} - \mathrm{i}\,\mathrm{Im}\,\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}_0, \boldsymbol{r}_0)$$

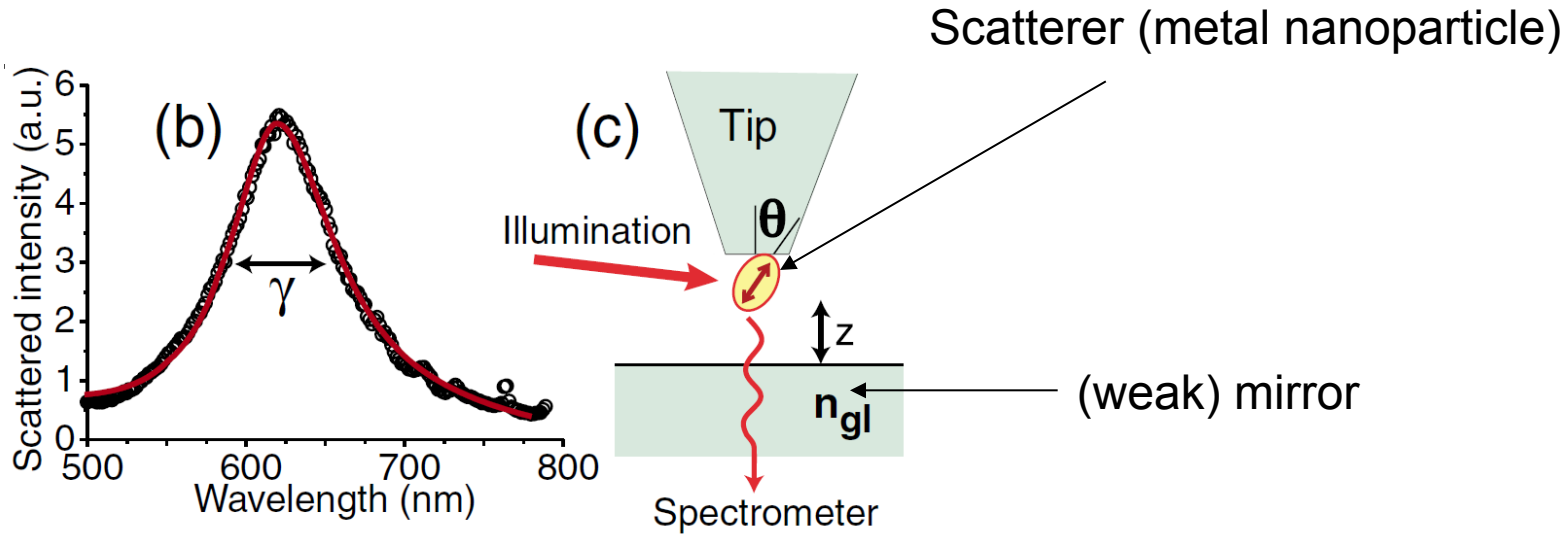


- Above formula is a recipe to amend any electrostatic polarizability α_0 with a radiation damping term to ensure energy conservation
- Electrodynamic polarizability depends on position within photonic system
- Radiation correction small for weak scatterers (small α_0)
- Radiation correction significant for strong scatterers (large α_0)
- Limit of maximally possible scattering strength

$$\mathrm{Im}\,\boldsymbol{\alpha}_{\mathrm{eff}} \xrightarrow{\boldsymbol{\alpha}_0 \text{ large}} \left[\mathrm{Im}\,\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}_0, \boldsymbol{r}_0)\right]^{-1}$$

Drexhage's experiment with a scatterer

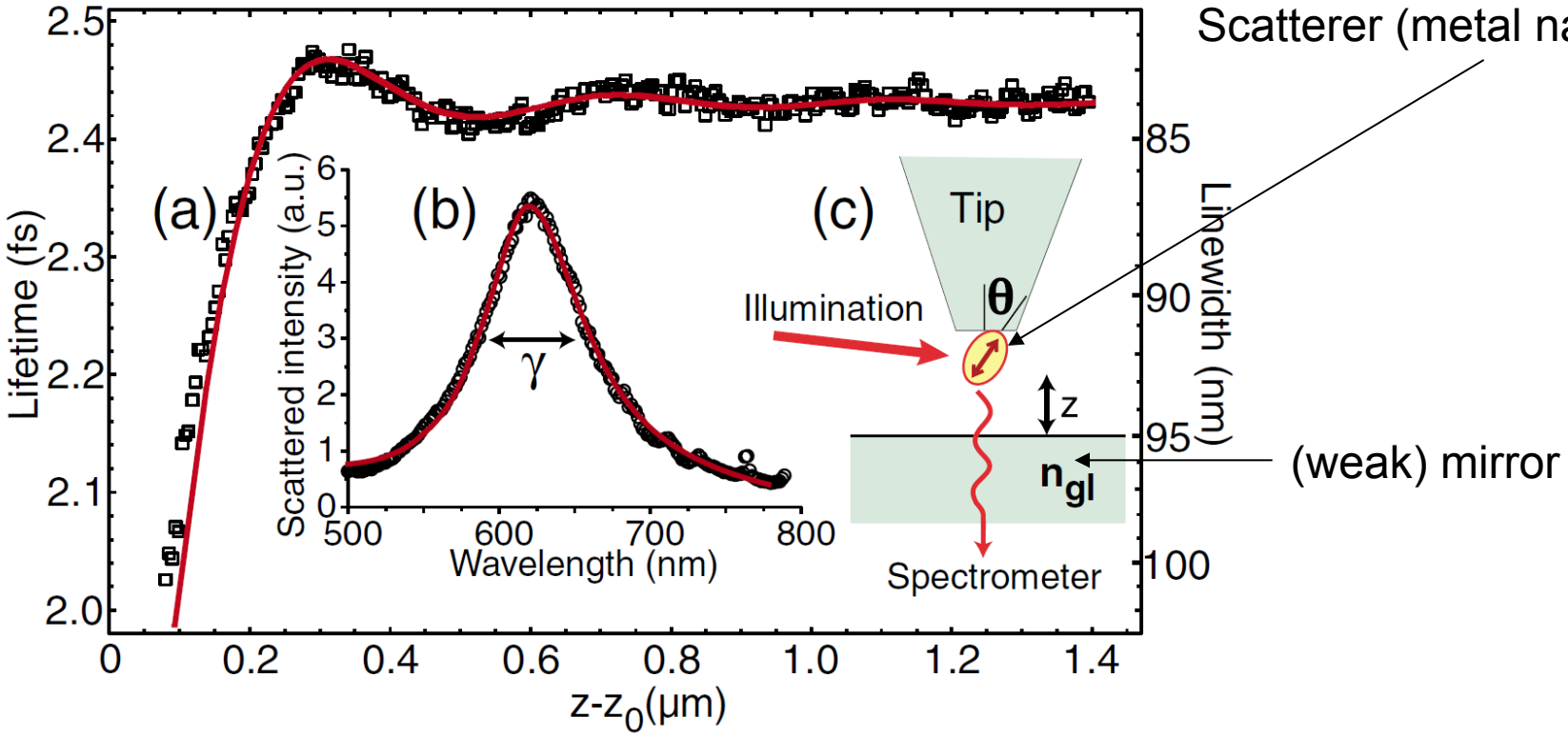
Buchler et al., PRL 95, 063003 (2005)



- Metal nanoparticle on a scanning probe close to a reflecting substrate
- Measure width of scatterer's resonance as a function of distance to substrate

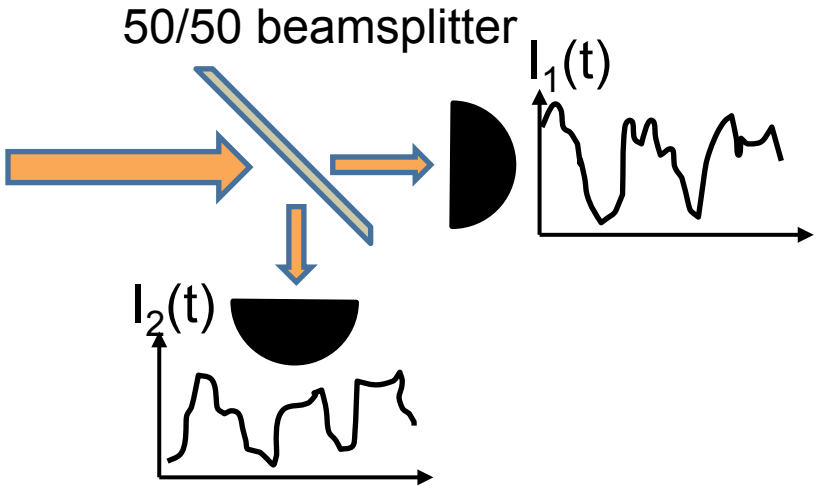
Drexhage's experiment with a scatterer

Buchler et al., PRL 95, 063003 (2005)



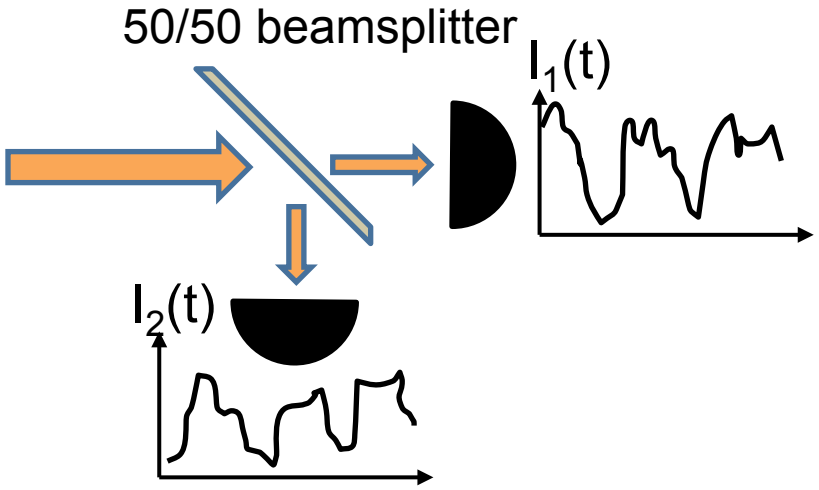
- Spectral width of scattering cross section (i.e. damping) can be tuned by changing scatterer-mirror distance
- LDOS determines damping rate of scatterer

The Hanbury Brown-Twiss experiment



- Beam of light impinging on a 50/50 beamsplitter (BS)
- Record intensity I(t) in each arm after BS
- Calculate normalized cross correlation between signals I_1 and I_2

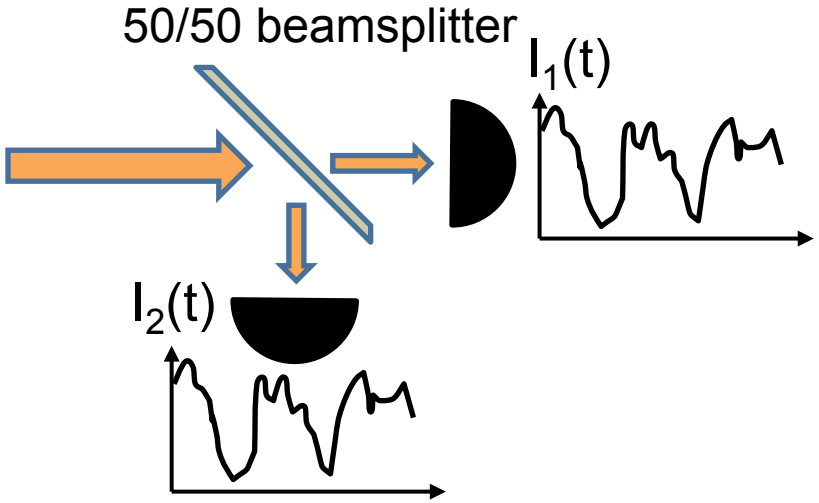
The second order correlation function



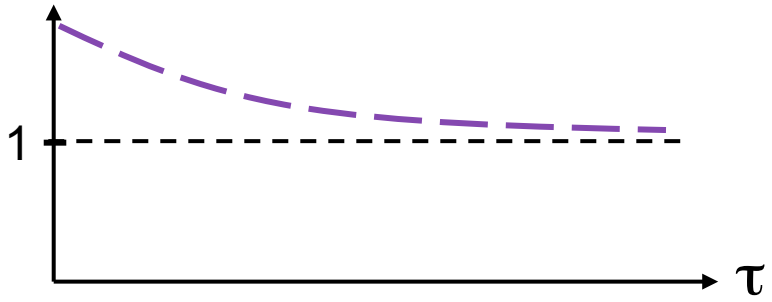
$$g^{(2)}(\tau) = \frac{\langle I_1(t) I_2(t+\tau) \rangle}{\langle I_1(t) \rangle \langle I_2(t) \rangle}$$

- Beam of light impinging on a 50/50 beamsplitter (BS)
- Record intensity I(t) in each arm after BS
- Calculate normalized cross correlation between signals I_1 and I_2

Intensity autocorrelation - the classical case

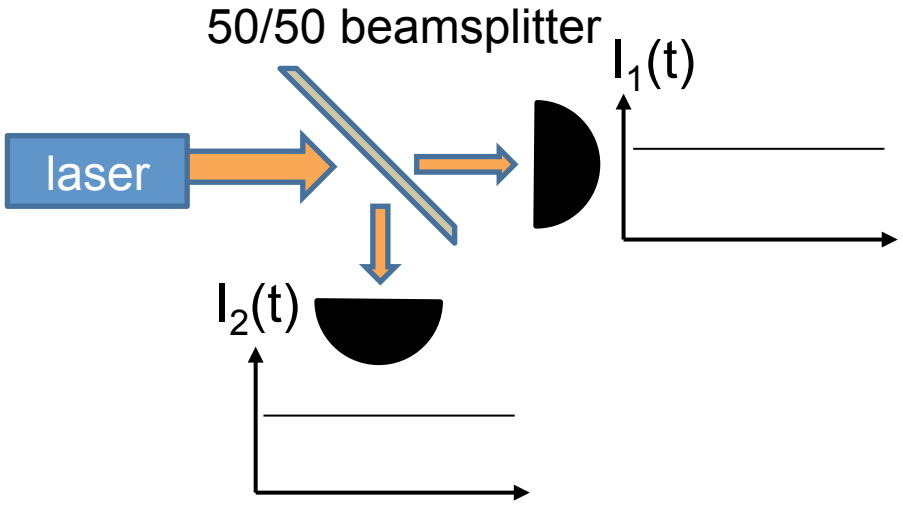


$$g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau)\rangle}{\langle I(t)\rangle^2}$$

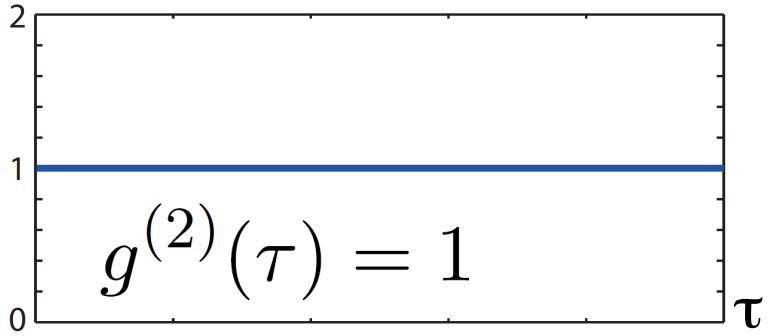


- Beam of light impinging on a 50/50 beamsplitter (BS)
- Record intensity I(t) in each arm after BS
- For a classical field $I_1(t) = I_2(t)$, so $g^{(2)}$ is intensity autocorrelation
- For long delay times $g^{(2)}(\tau \to \infty) = 1$
- Correlation at zero delay $g^{(2)}(\tau = 0) \geq 1$
- global maximum at zero delay $g^{(2)}(0) \geq g^{(2)}(\tau)$

Intensity autocorrelation - the coherent case

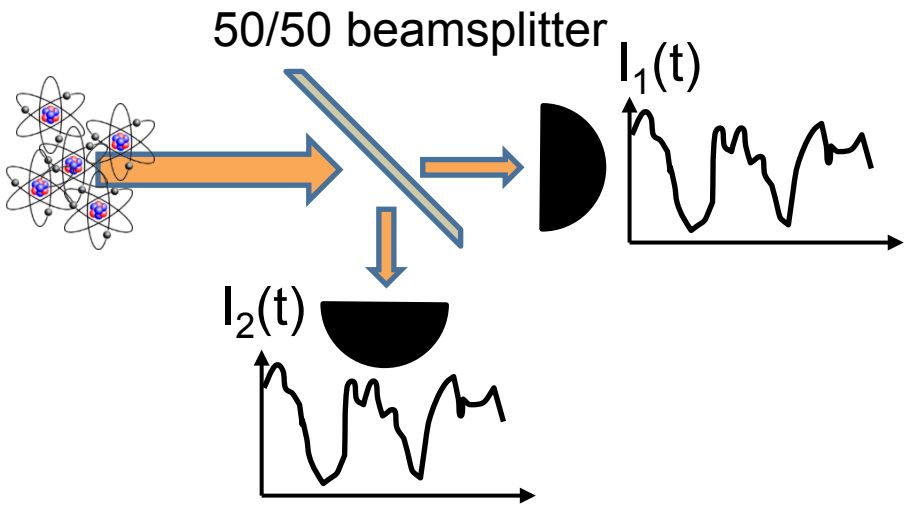


$$g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau)\rangle}{\langle I(t)\rangle^2}$$

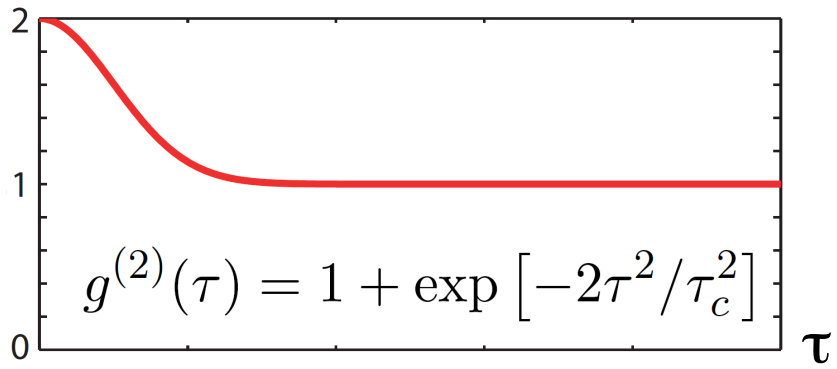


- Perfectly monochromatic field $E(t) \propto \cos(\omega t)$
- Intensity is therefore $I(t) = \text{const.}$

Intensity autocorrelation - the chaotic case



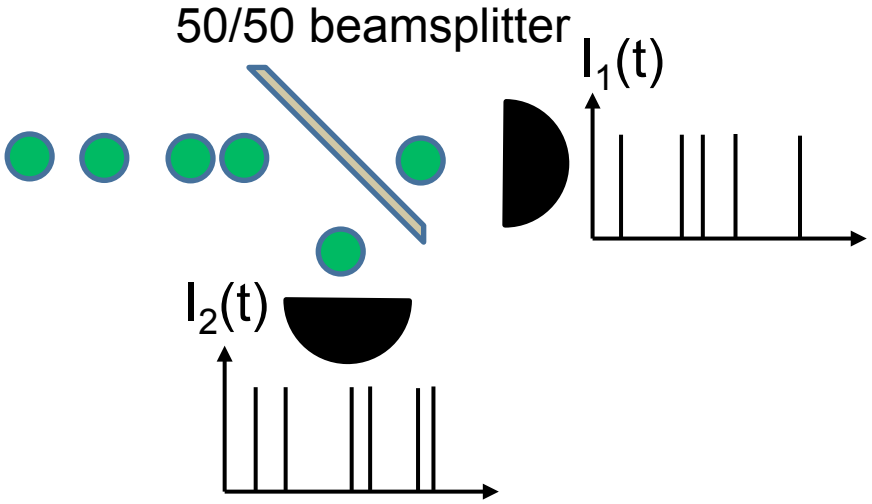
$$g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau)\rangle}{\langle I(t)\rangle^2}$$



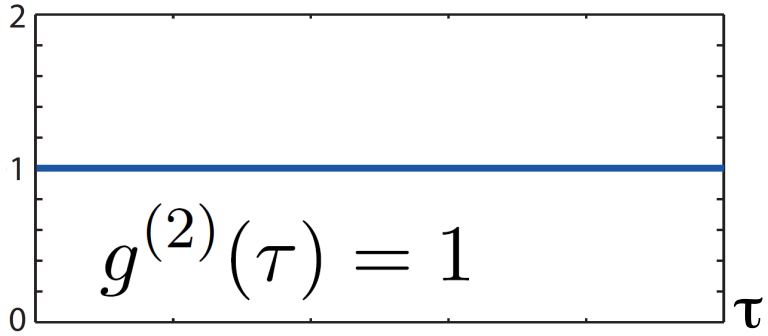
- Collection of sources $E(t) = E_0 \sum_{\text{atoms}} \exp\left[-i\Omega_a t - \phi_a\right]$
- Random phases ϕ_a
- Gaussian distribution of emission frequencies

$$P(\Omega_a) \propto \exp\left[-(\Omega_0 - \Omega_a)^2 \tau_c^2\right]$$

Counting photons – coherent case



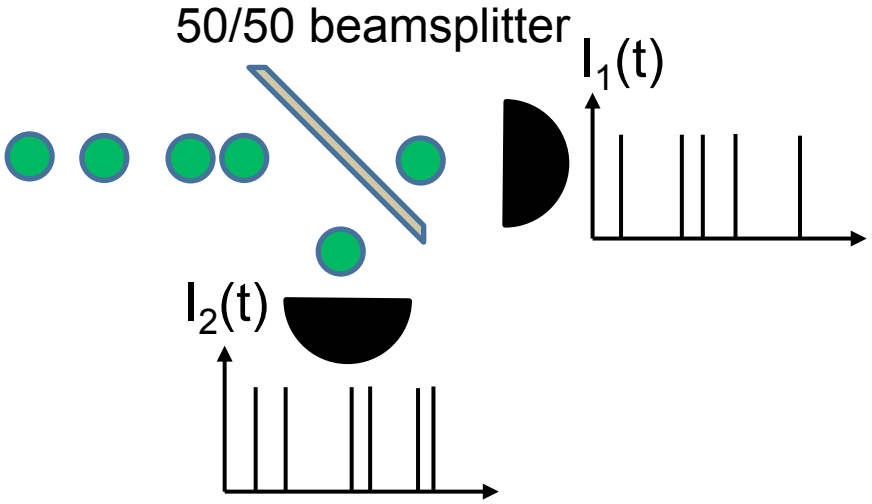
$$g^{(2)}(\tau) = \frac{\langle n_1(t) n_2(t+\tau) \rangle}{\langle n_1(t) \rangle \langle n_2(t) \rangle}$$



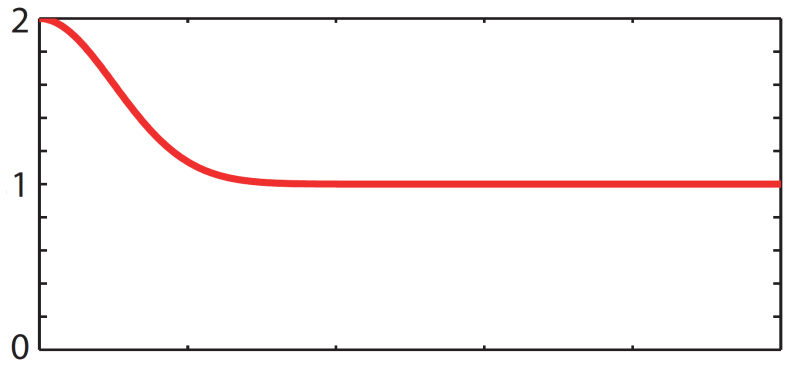
- $n_i(t)$ is the number of photons on detector i at time t
- Interpret $g^{(2)}(\tau)$ as the probability of detecting a photon on detector 2 at t= τ given that a photon was detected on detector 1 at t=0
- $g^{(2)}(\tau) = 1$ means that photons arrive with Poissonian distribution

$$P(n) = \frac{\langle n \rangle^n}{n!} \exp\left[-\langle n \rangle\right]$$

Counting photons – chaotic case

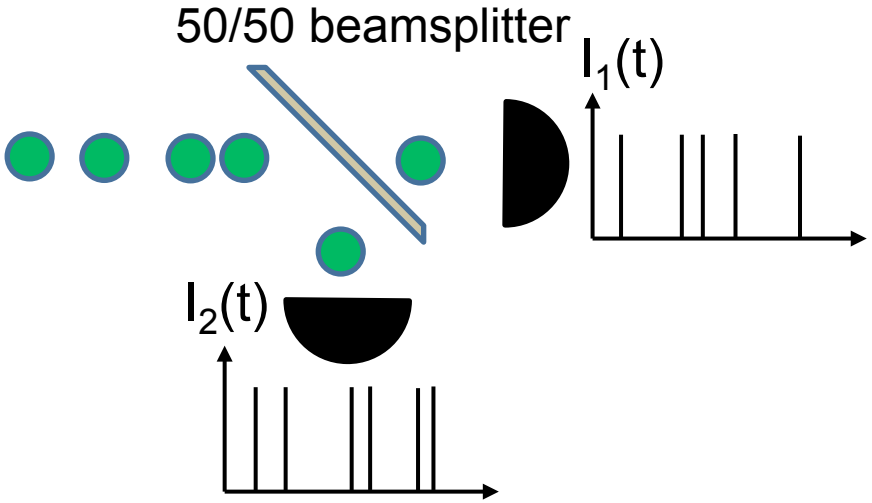


$$g^{(2)}(\tau) = \frac{\langle n_1(t) n_2(t+\tau) \rangle}{\langle n_1(t) \rangle \langle n_2(t) \rangle}$$

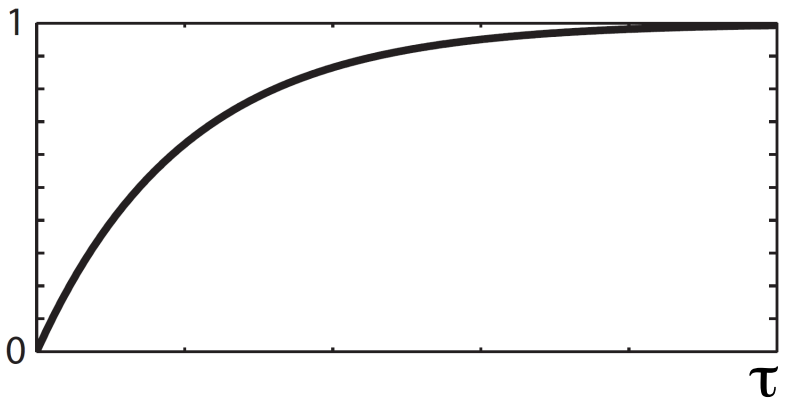


- $n_i(t)$ is the number of photons on detector i at time t
- Interpret $g^{(2)}(\tau)$ as the probability of detecting a photon on detector 2 at t= τ given that a photon was detected on detector 1 at t=0
- $g^{(2)}(\tau=0) > 0$ means that photons tend to arrive in bunches

Counting photons – a single quantum emitter

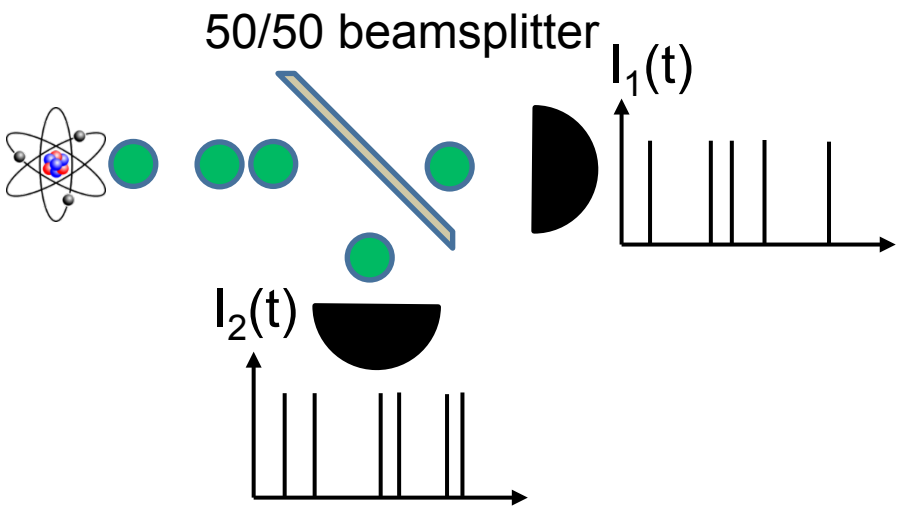


$$g^{(2)}(\tau) = \frac{\langle n_1(t) n_2(t+\tau) \rangle}{\langle n_1(t) \rangle \langle n_2(t) \rangle}$$

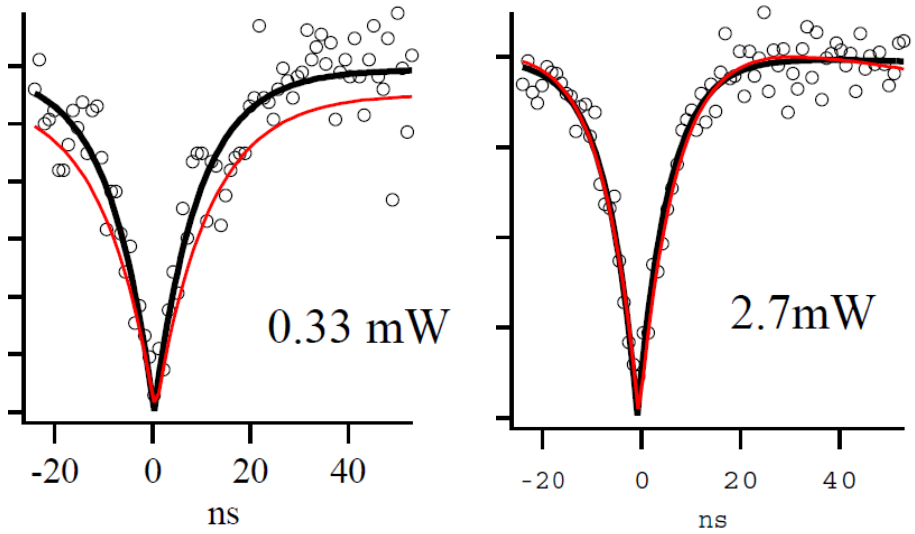


- Assume source is a single emitter
- Single emitter can only emit one photon at a time
- If there is a photon on D1 there cannot be a photon on D2 → antibunching
- Photon antibunching is at odds with classical electromagnetism
- $g^{(2)}(\tau=0) = 0$ is the signature of a single photon source
- What determines the rise time of $g^{(2)}(\tau)$?

Intensity correlations – counting single photons



Beveratos, PhD thesis, Univ. Paris Sud (2002)

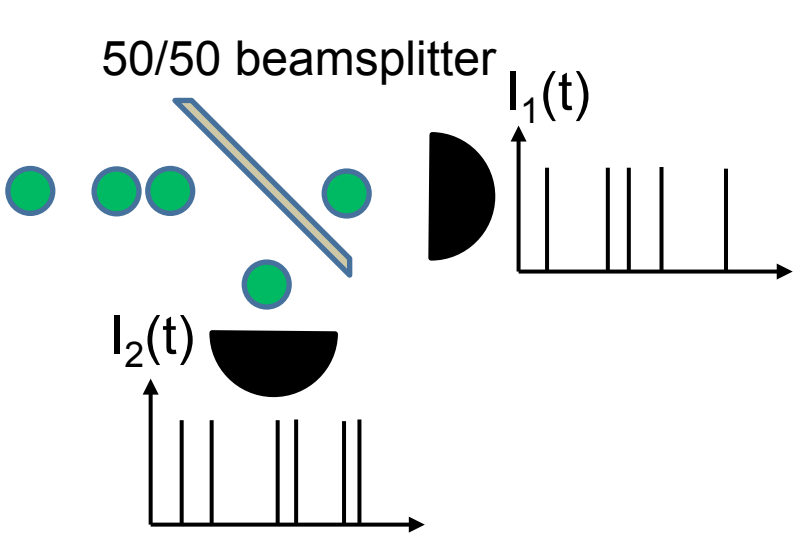


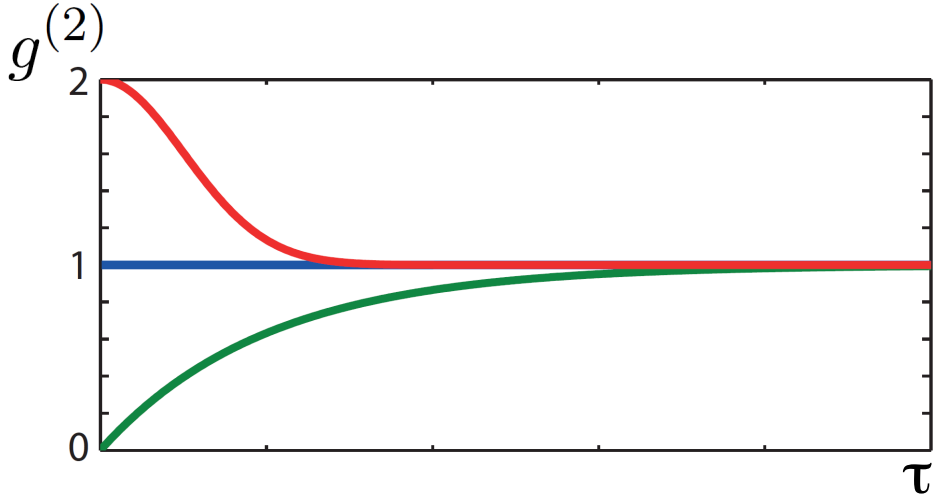
- How do you know your emitter is a single photon source?

  For n emitters: $g^2(0) = 1 - \frac{1}{n}$

- How does the lifetime show up in the correlation function? Rise time is lifetime in the case of weak pumping.

Intensity correlations – summary





- Second-order correlation function measures temporal intensity correlation
- Bunching: photons tend to “arrive together”, classically allowed/ expected
- Antibunching: photons tend to “arrive alone”, classically forbidden

$g^2(\tau = 0) \gg 2$ "Superbunching"

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Stokes–anti-Stokes correlations in diamond

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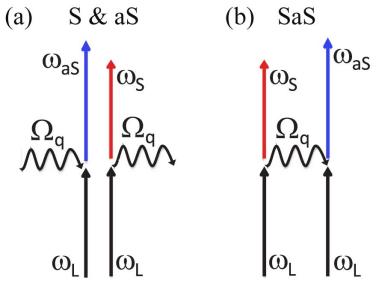
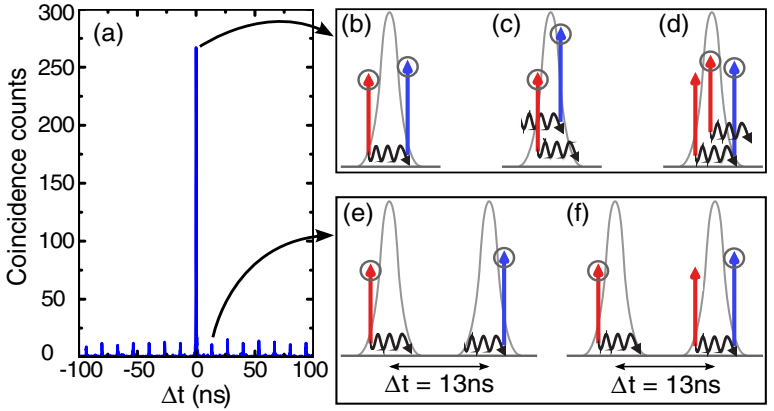


Fig. 1. Schematic representation of Stokes (S) and anti-Stokes (aS) Raman scattering. (a) Uncorrelated (i.e., spontaneous) S and aS processes (S & aS). The phonons responsible for the aS process are generated thermally. (b) Anti-Stokes photons can also be generated by the phonons created through the Stokes process. Stokes photons and Stokes-induced anti-Stokes (SaS) photons become correlated.



Take Home 1: Optical antennas …

- Modulate LDOS on sub-λ length scale
- Can boost decay rates of quantum emitters
- Can direct the emission of quantum emitters
- Rely on resonances in the polarizability of their constituents

The polarizability of strong dipolar scatterers …

- has to take radiation effects into account
- depends on position within photonic system

Take Home 2: The local density of optical states (LDOS) …

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

- Is the imaginary part of the Green function (besides constants)
- Governs light-matter interaction, e.g.
  - Determines the decay rate (enhancement) of quantum emitters
  - Determines the linewidth of dipolar scatterers
  - …