On the menu today

Decay rate engineering

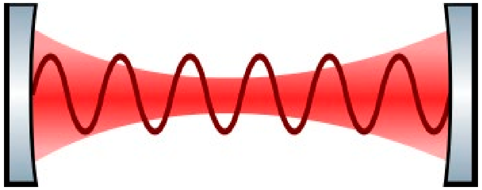
- The local density of optical states (LDOS)
- Decay rate of quantum emitters
- Decay rate engineering
- Example: Drexhage experiment
- Example: The Purcell effect
- Example: optical antenna
  - Simple picture: dipole moment booster
  - Dipolar scattering theory and radiation damping
  - More detailed picture: LDOS of a dipolar scatterer

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

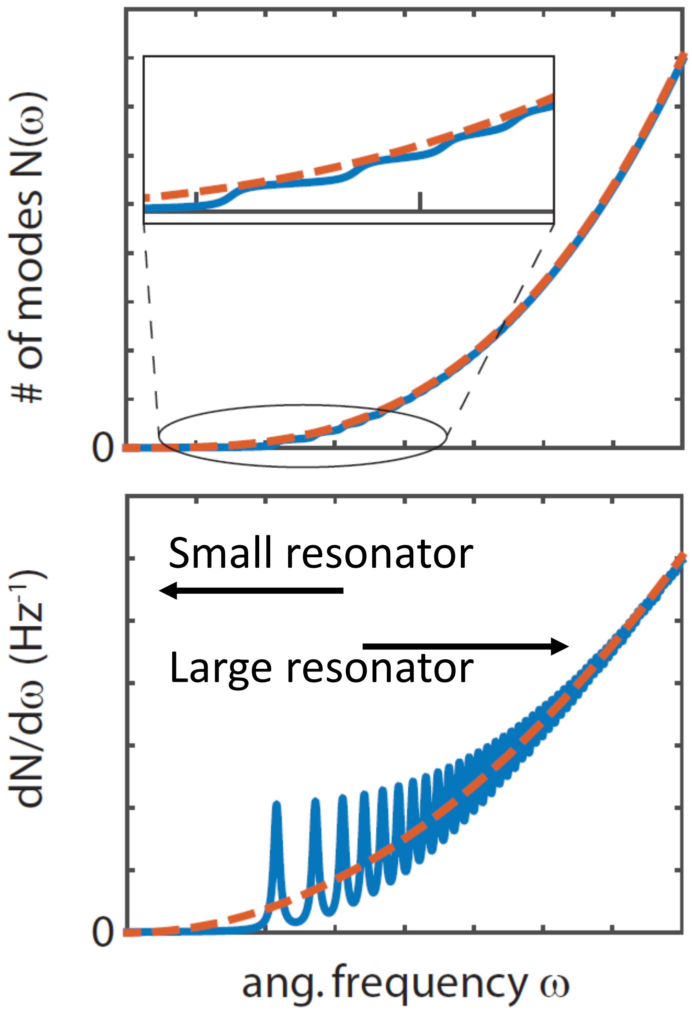
1. Located at the field maximum of the mode
2. Spectrally matched exactly to the mode
3. Oriented along the field direction of the mode



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

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):

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

Micro-cavities in the 21st century

How to squeeze more light out of a source:

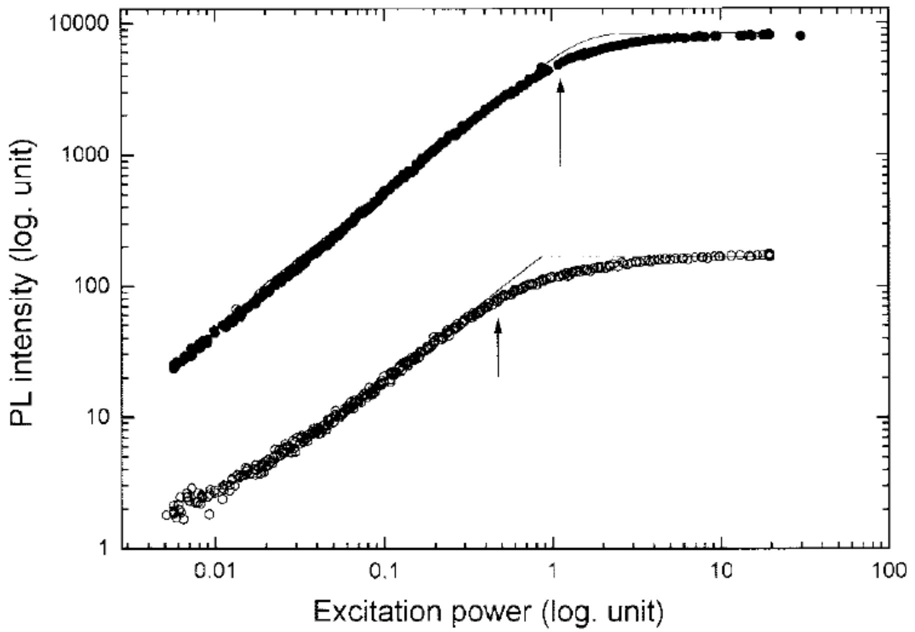
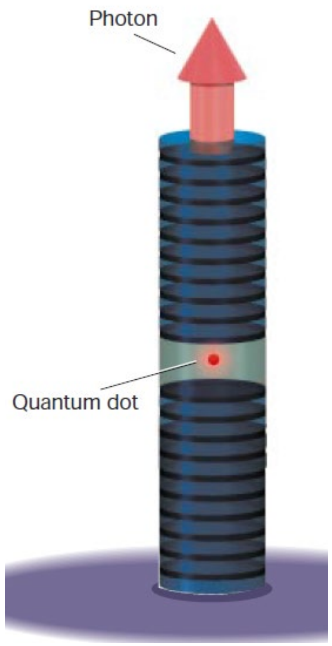


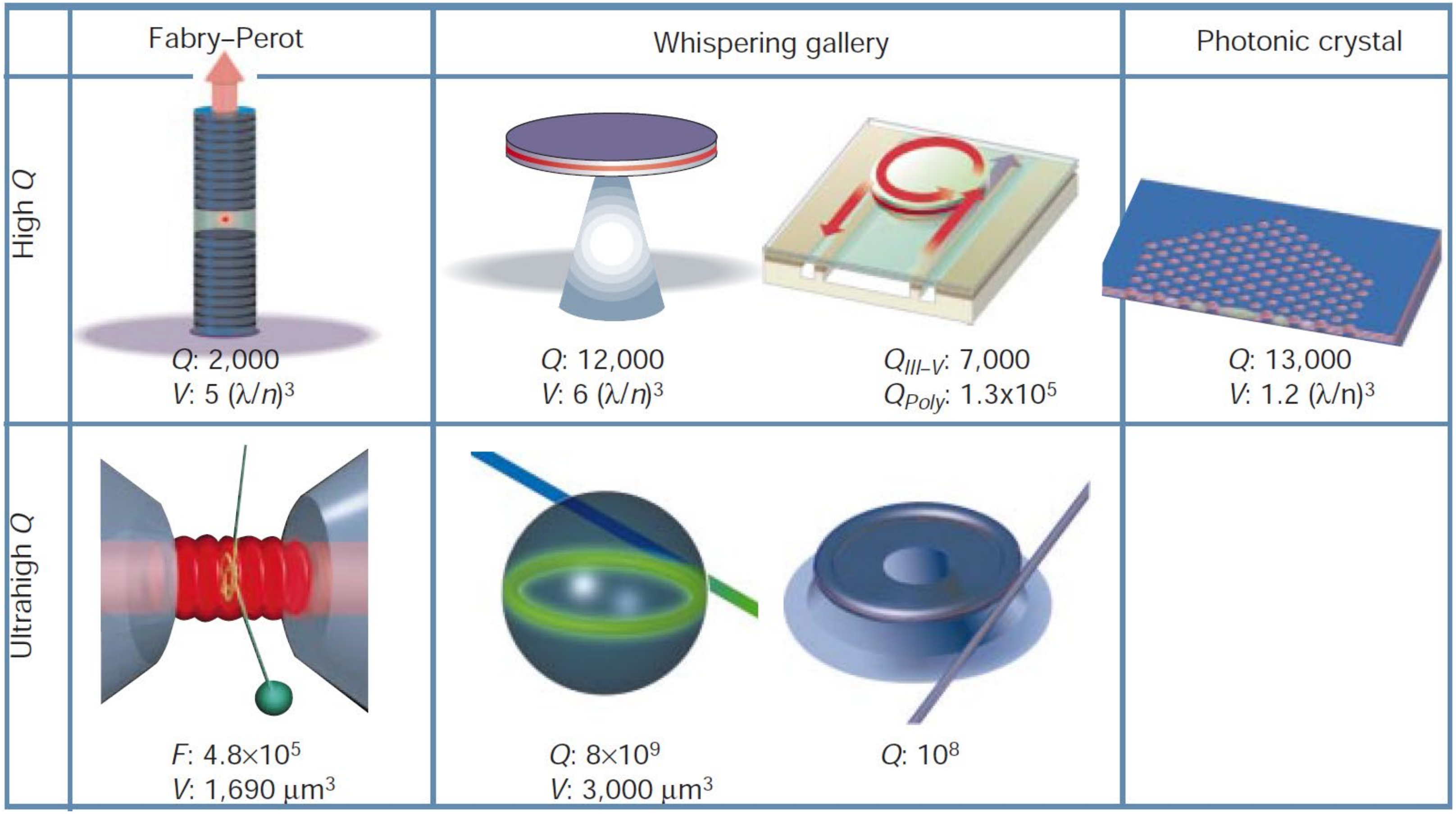
Fig. 3. PL intensity versus excitation power for a 1.8-μm diameter pillar, ($Q = 3000$, $F_p = 15$). The saturation of the PL signal is observed both for on-resonance, (●) and off-resonance (○) QB's. The beginning of the saturation,

www.photonics.ethz.ch



Vahala, Nature 424, 839

Micro-cavities in the 21st century



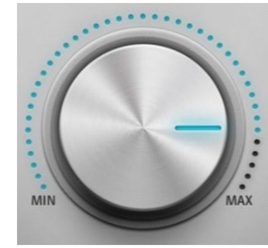
Vahala, Nature 424, 839

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

Digression: Q-factor vs. finesse

Decay rate engineering

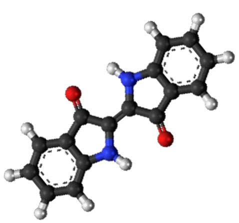
$$\gamma = \frac{\pi\omega}{3\hbar\epsilon_0} |\hat{\boldsymbol{p}}|^2 \rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega)$$

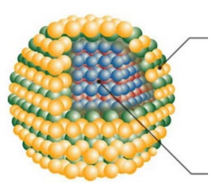


Emitter

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

Chemistry, material science

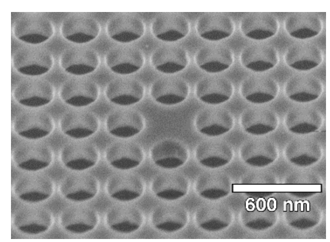


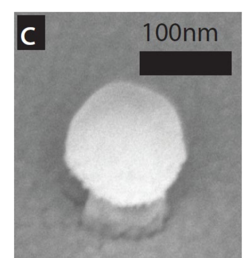


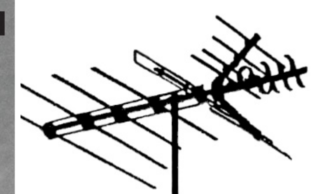
Environment

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

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

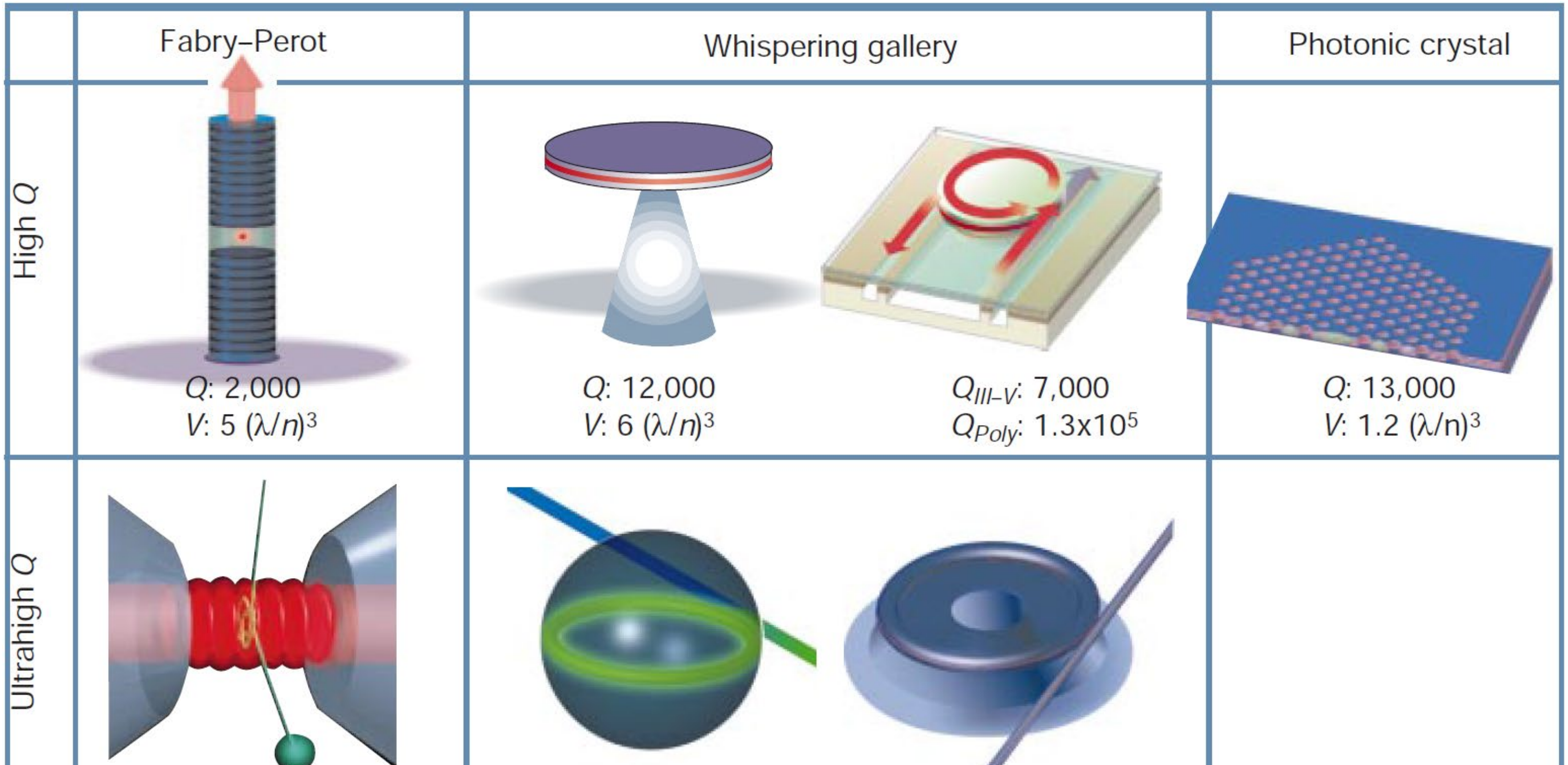
$$\langle P \rangle = \frac{\pi\omega^2}{12\epsilon\epsilon_0} |\boldsymbol{p}|^2 \ \rho_{\mathbf{n}}(\boldsymbol{r}_0, \omega)$$

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.

Micro-cavities in the 21st century

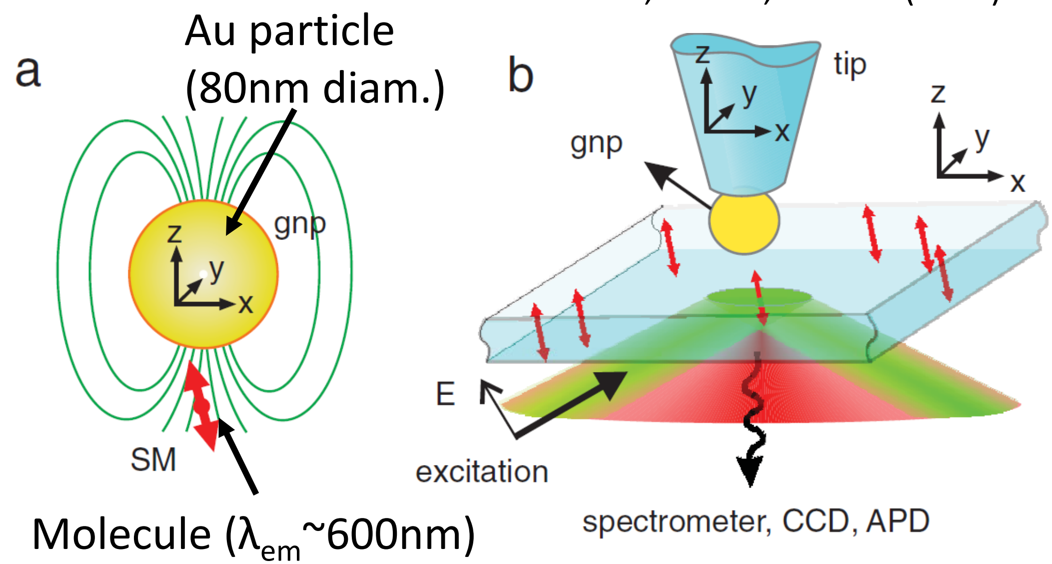


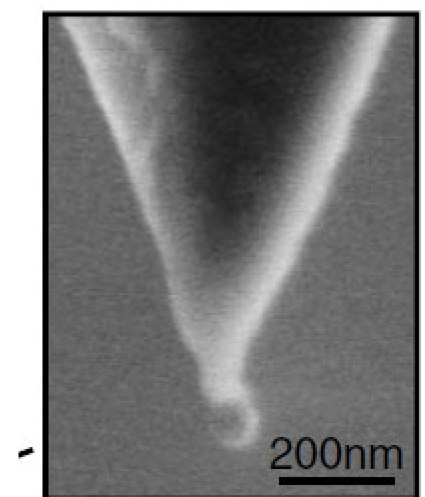
24, 839

These structures modulate LDOS on a length scale given by the wavelength.
How can you modulate LDOS on a sub-wavelength scale?

Optical antennas for LDOS engineering

Kühn et al., PRL 97, 017402 (2006)

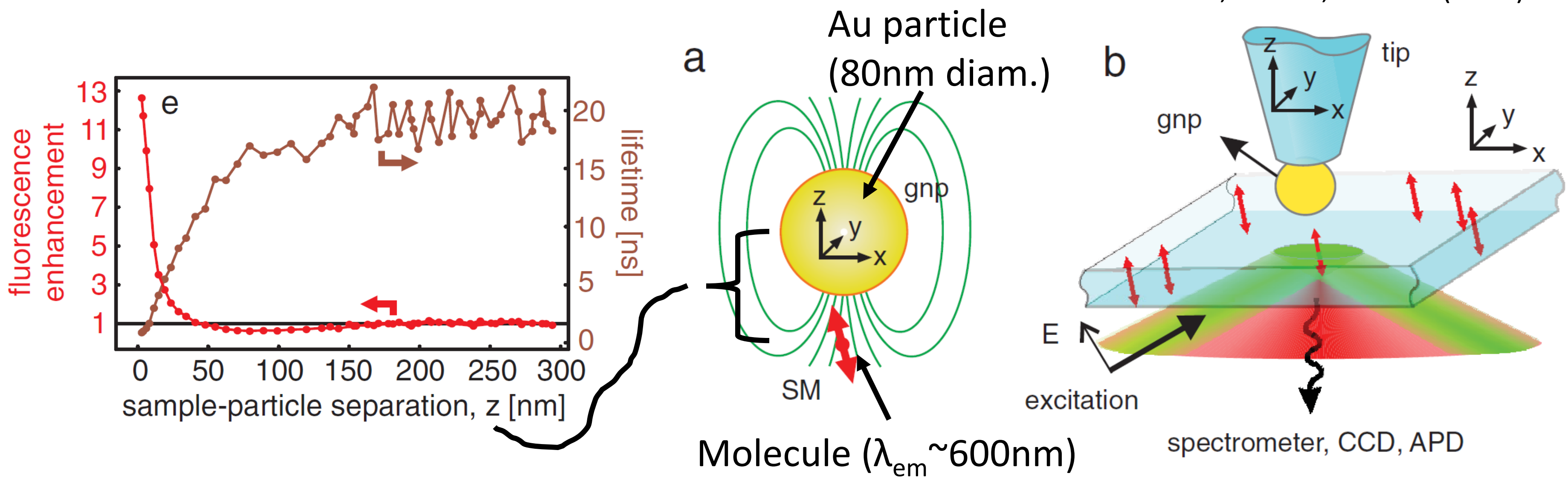




Anger et al., PRL 96, 113002 (2006)

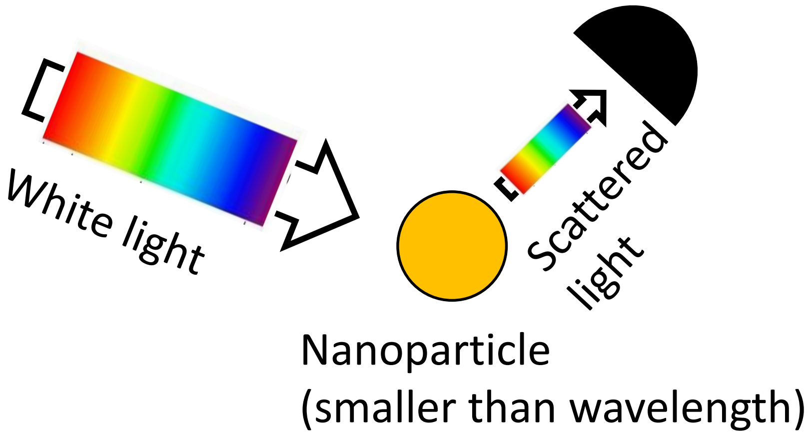
Optical antennas for LDOS engineering

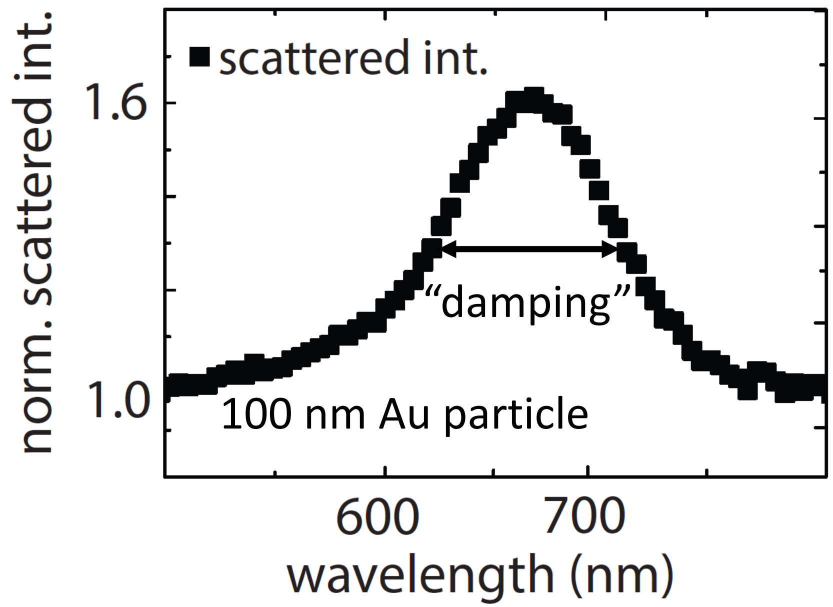
Kühn et al., PRL 97, 017402 (2006)



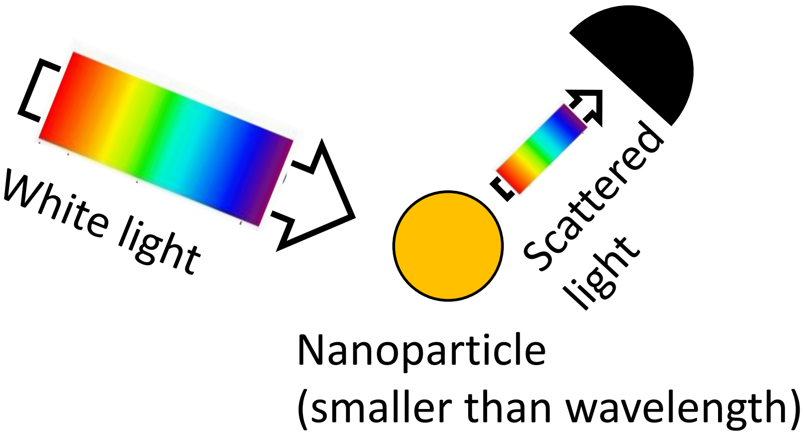
- Metallic nanoparticles can act as “antennas” and boost decay rate of quantum emitters in their close proximity
- Effect confined to length scale of order λ/10

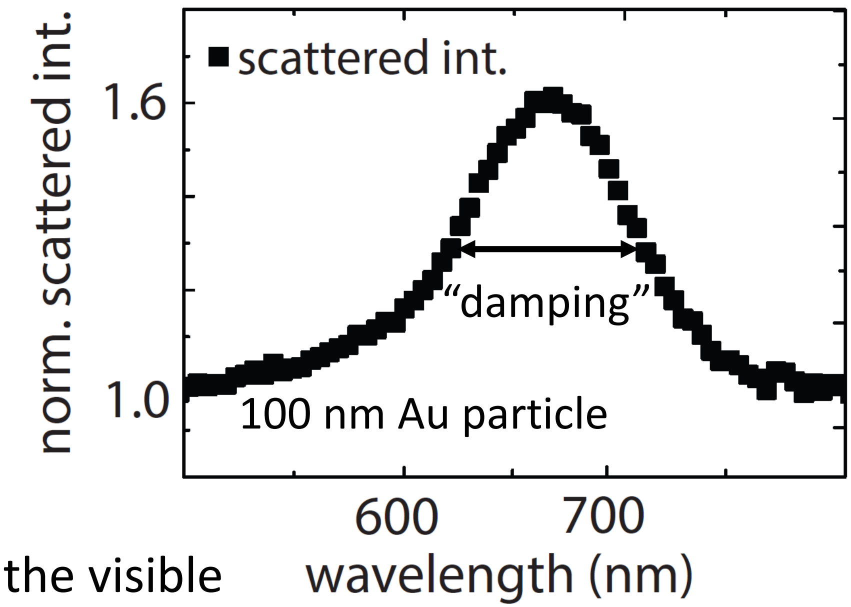
Nanoparticles: resonators at optical frequencies



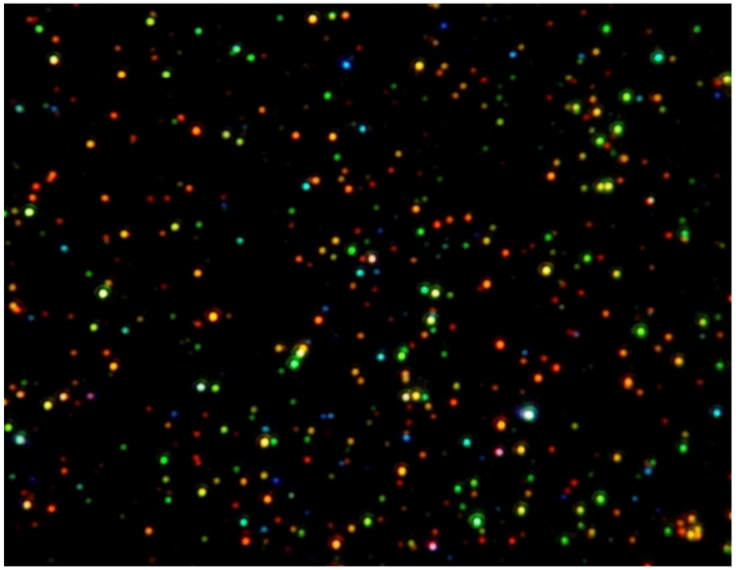


Nanoparticles: resonators at optical frequencies



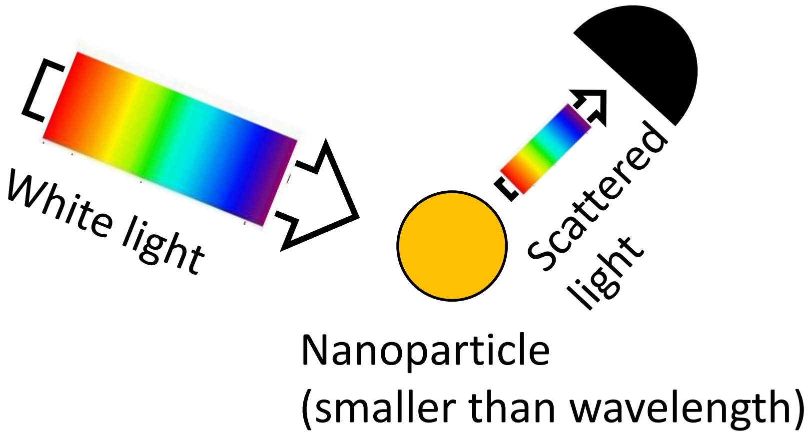


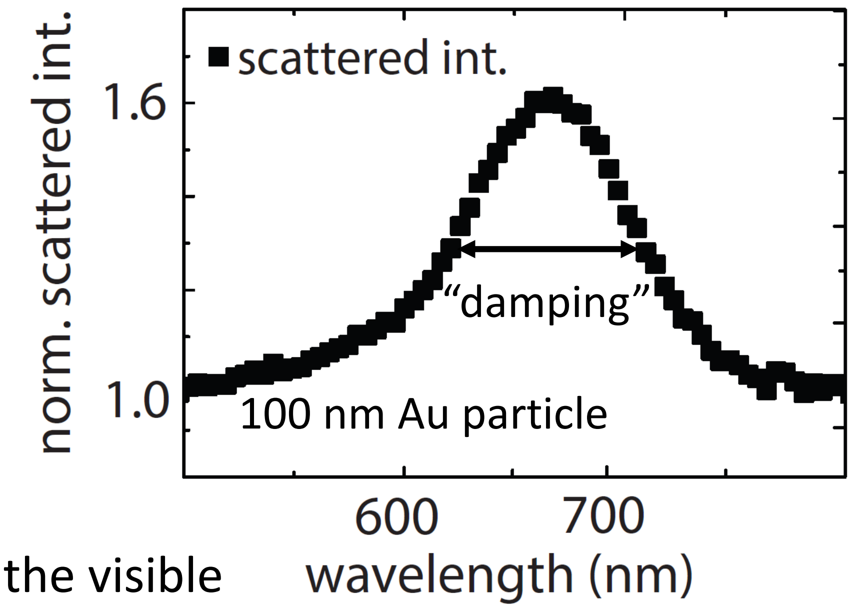
- Metal nano-particles show resonances in the visible



Nanoparticle.com

Nanoparticles: resonators at optical frequencies





- Metal nano-particles show resonances in the visible

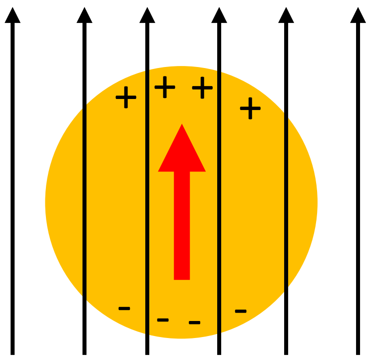
Lycurgus Cup (glass with metal nano-particles):
Green when front lit→
← Red when back lit

How does that work?

Wikipedia.org

The electrostatic polarizability

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

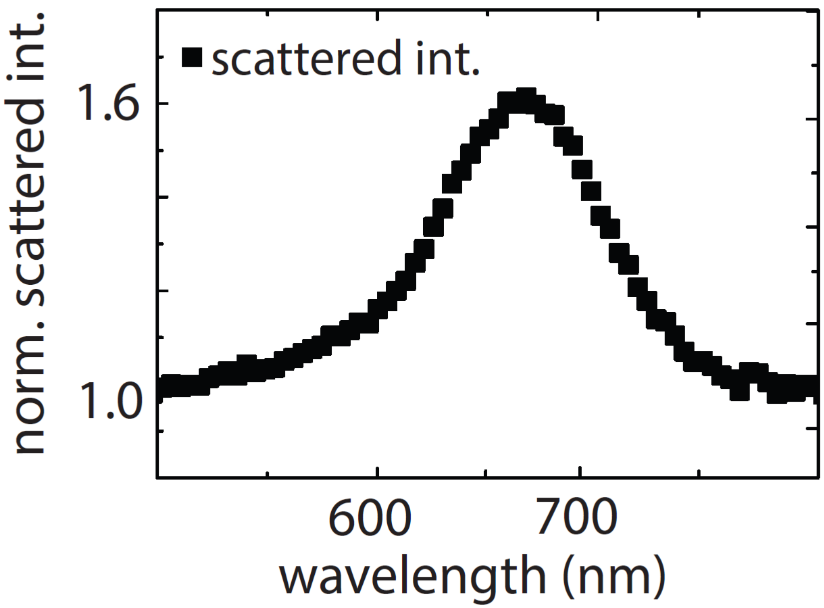


- Static polarizability: induced dipole moment due to static E-field
- For a sphere in vacuum: $$\alpha_0 = 4\pi\epsilon_0 a^3 \frac{\epsilon-1}{\epsilon+2}$$
- For a Drude metal, we find a Lorentzian polarizability

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega+\mathrm{i}\gamma)}$$

plasma frequency

Ohmic damping rate



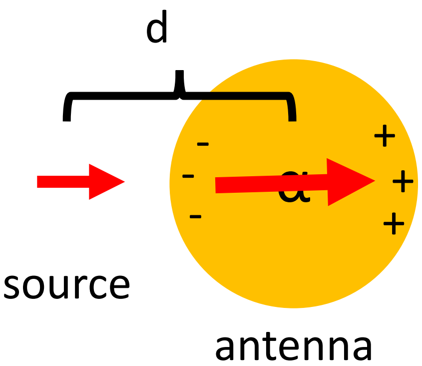
$$\alpha_0 \propto \frac{1}{\omega_0^2 - \omega^2 - \mathrm{i}\gamma\omega}$$

Optical antennas – an intuitive approach

Optical antennas – an intuitive approach

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

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



LDOS enhancement:

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

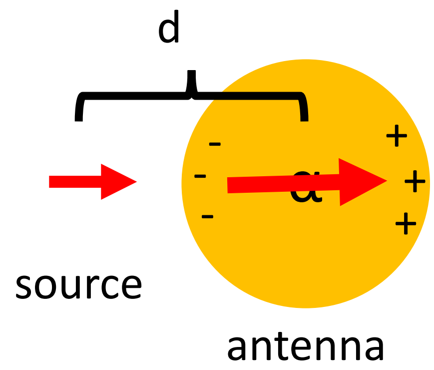
- assume an oscillating dipole close to a polarizable particle
- Assume that particle is small enough to be described as dipole
- Assume distance d<<λ, near field of source polarizes particle
- If polarizability α is large, antenna dipole largely exceeds source dipole
- Radiated power dominated by antenna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

Optical antenna is a dipole moment booster!

Optical antennas – an intuitive approach

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

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



LDOS enhancement:

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

According to this, the only limit to the LDOS enhancement provided by an optical antenna are material properties (such as Ohmic loss rate γ).

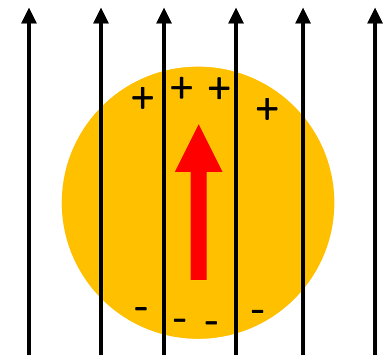
Are there other damping mechanisms?

The electrodynamic polarizability

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

Remember we used the expression for the <u>static</u> polarizability for α_0!

$$\alpha_0 = 4\pi\epsilon_0 a^3 \frac{\epsilon-1}{\epsilon+2}$$



The electrodynamic polarizability

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

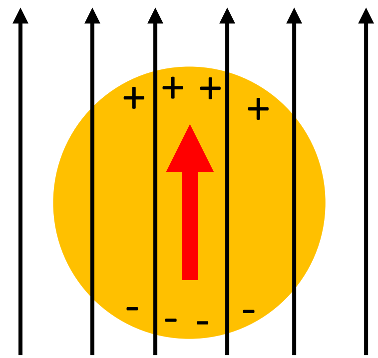
$$\boldsymbol{p} = \boldsymbol{\alpha}_{\text{eff}} \boldsymbol{E}_{\text{ext}}$$

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

$$\underline{\overleftrightarrow{G}} = \omega^2 \mu \mu_0 \overleftrightarrow{G}$$

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

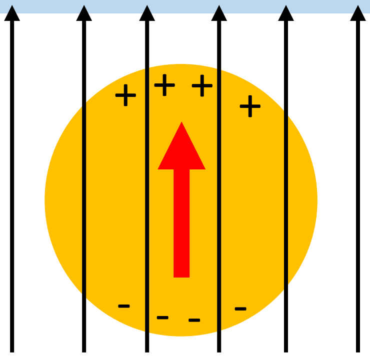
$$\underline{\overleftrightarrow{G}} = \text{Re}\,\underline{\overleftrightarrow{G}}_0 + \text{Re}\,\underline{\overleftrightarrow{G}}_s + \text{i}\left[\text{Im}\,\underline{\overleftrightarrow{G}}_0 + \text{Im}\,\underline{\overleftrightarrow{G}}_s\right]$$



- 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. Choose 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

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



- This 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 is small for weak scatterers (small α_0)
- Radiation correction is significant for strong scatterers (large α_0)
- Limit of maximally possible scattering strength:

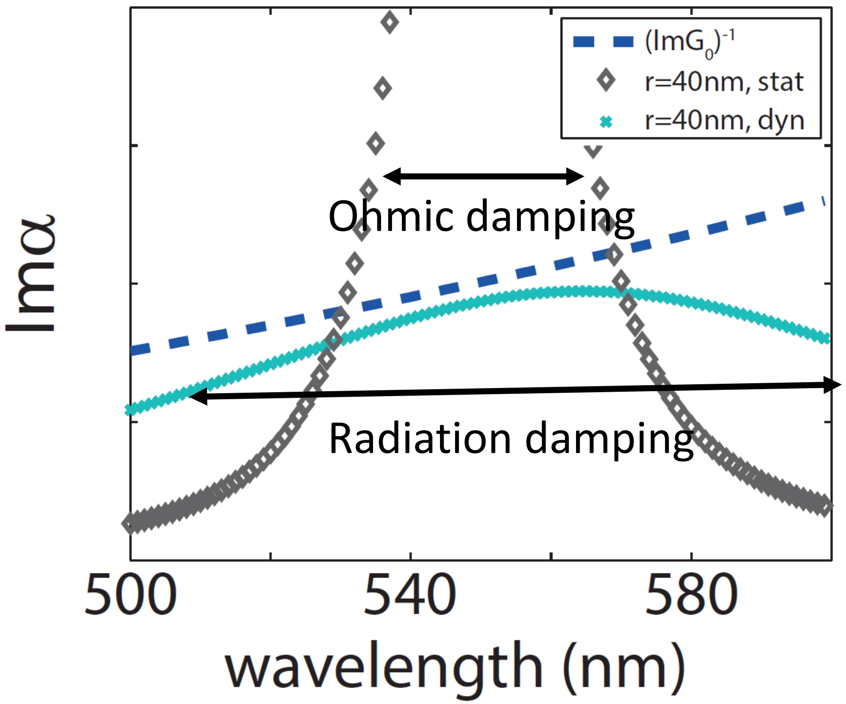
$$\mathrm{Im}\,\boldsymbol{\alpha}_{\mathrm{eff}} \xrightarrow{\boldsymbol{\alpha}_0\ \mathrm{large}} \left[\mathrm{Im}\,\overleftrightarrow{\underline{\boldsymbol{G}}}(\boldsymbol{r}_0, \boldsymbol{r}_0)\right]^{-1}$$

The electrodynamic polarizability

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

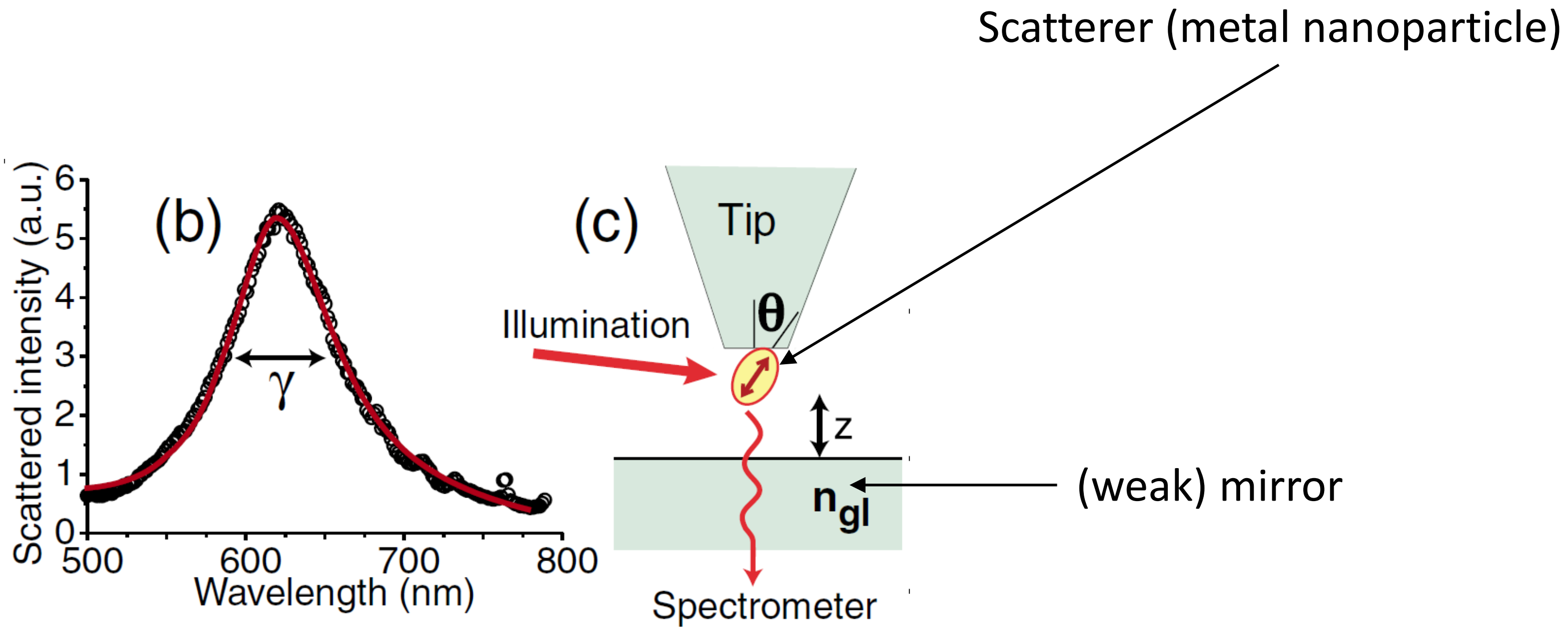
- Compare static and dynamic α
- Static α_0 may be huge, dynamic α_{eff} is always bounded by inverse LDOS
- Radiation damping is a loss channel and dampens resonance
- Radiation damping is given by the LDOS at the scatterer's position

Metallic particle (Drude model for ε)



Drexhage's experiment with a scatterer

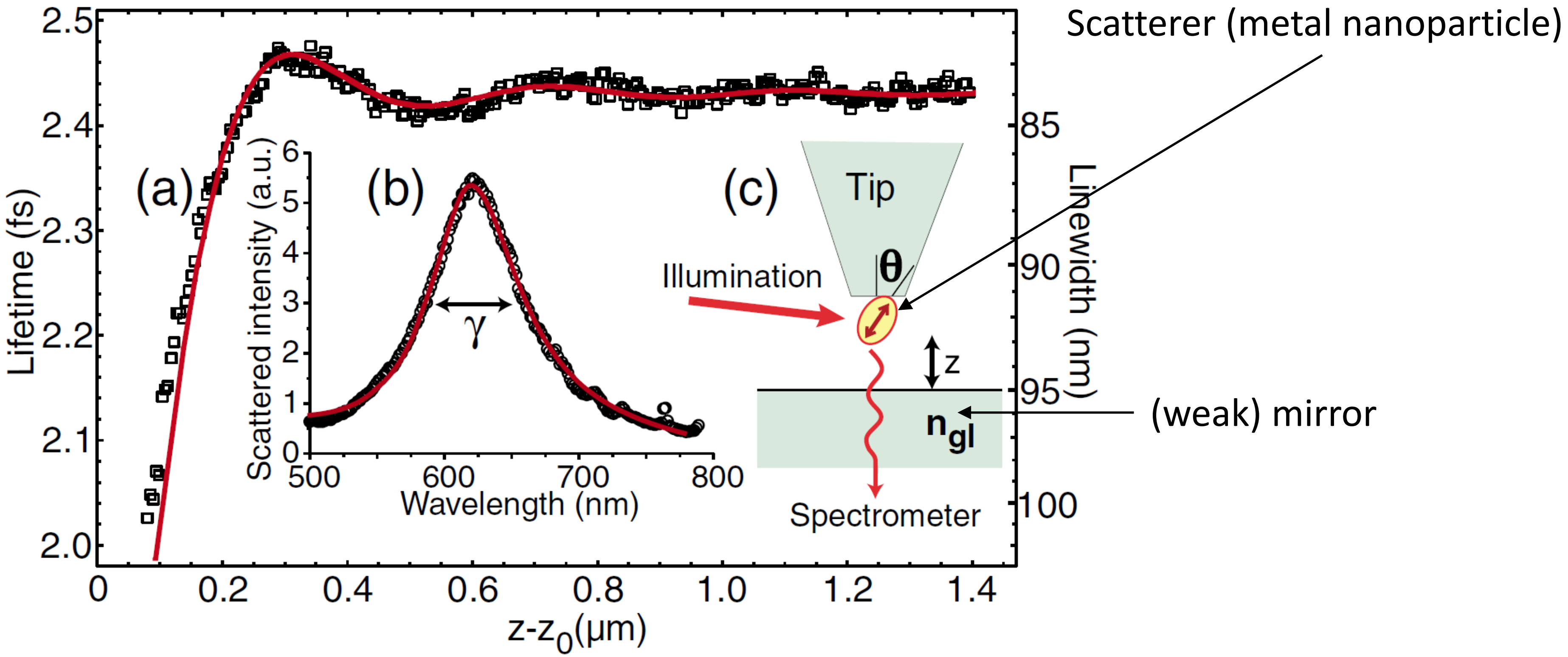
Buchler et al., PRL 95, 063003 (2005)



- 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

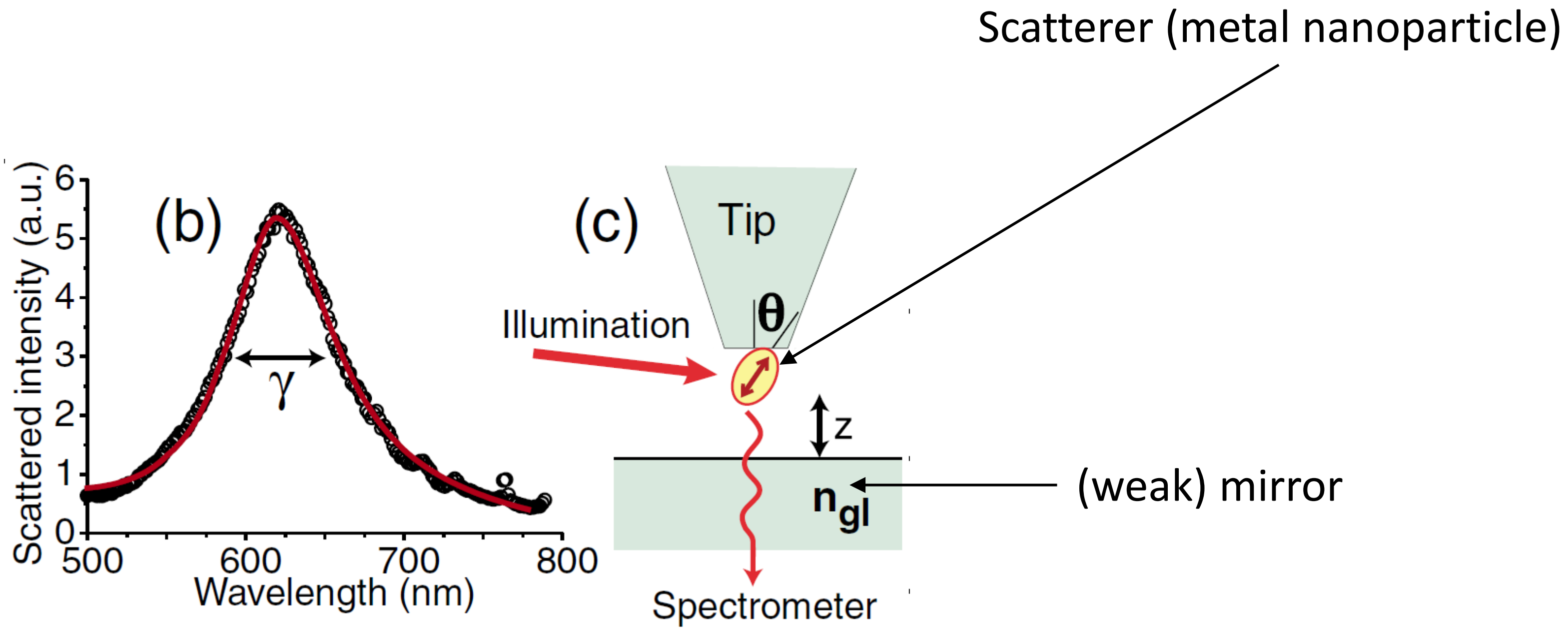
Buchler et al., PRL 95, 063003 (2005)



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

Drexhage's experiment with a scatterer

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

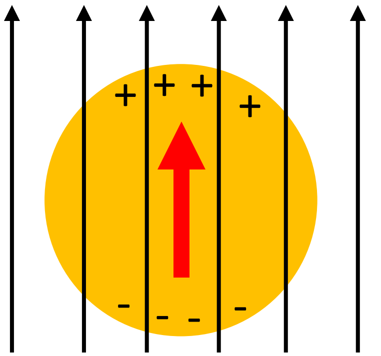


We can use material resonances to build resonant optical antennas of sub-wavelength size.

The Q-factor of strongly polarizable antennas is dominated by radiation loss.

The electrodynamic polarizability

The polarizability determines all optical properties of a dipolar scatterer:



Extinction cross section: $\sigma_{\mathrm{ext}} \propto \mathrm{Im}\left[\alpha\right]$

Scattering cross section: $\sigma_{\mathrm{scat}} \propto \left|\alpha\right|^2$

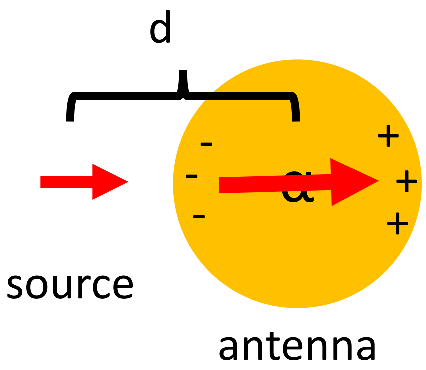
- There is an intimate relationship between $\mathrm{Im}\left[\alpha\right]$ and $\left|\alpha\right|^2$ (you derive this in your homework!)
- What is the absorption cross section? (be careful with textbooks!)

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

Optical antennas – revisited

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

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



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

Remember our (sloppy) derivation?

Optical antenna is a dipole moment booster!

Optical antennas – a cleaner derivation

Calculate rate enhancement via power enhancement

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

$$\overleftrightarrow{\underline{\boldsymbol{G}}} = \omega^2 \mu \mu_0 \overleftrightarrow{\boldsymbol{G}}$$