Single photon nano-antennas

Learning objectives:

- *scatterering*
- *emission control*

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Resonant Nanophotonics

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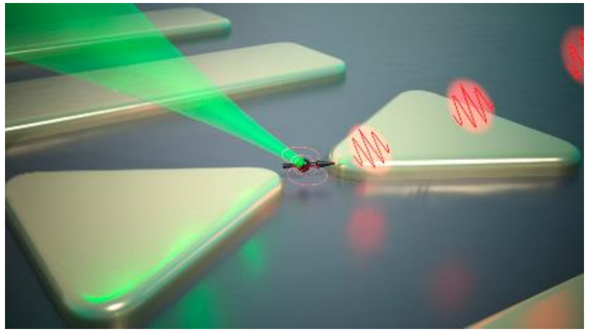
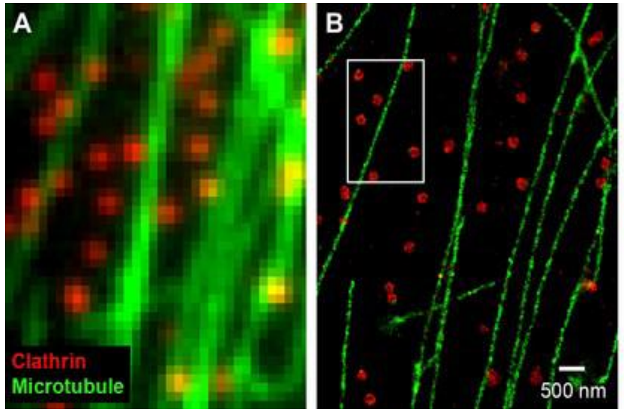
Motivation



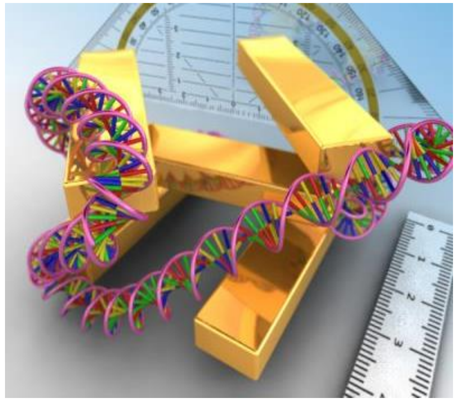
Nanophotonics to control emission, absorption, lasing…
- Directionality, polarization and phasefront of emission
- Efficiency, brightness and Purcell factor
- Spatial and temporal coherence



Bates & Zhuang [PALM, STORM]

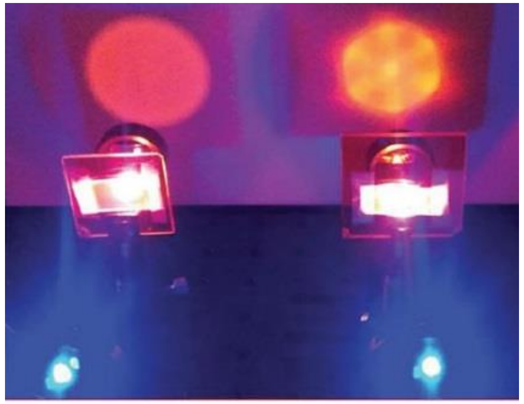
Optical microscopy below $\lambda/2$ limit

Single photon sources
Quantum information



Liu & Alivisatos

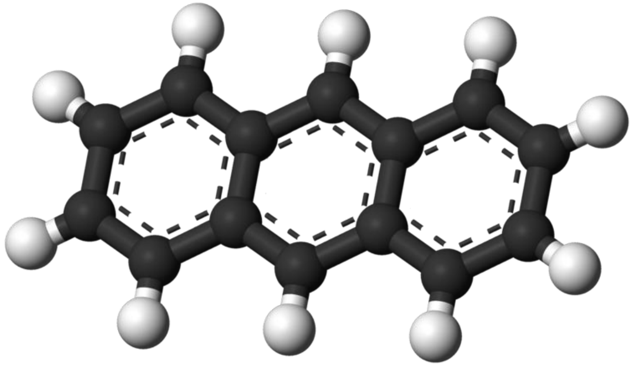
Spectroscopy

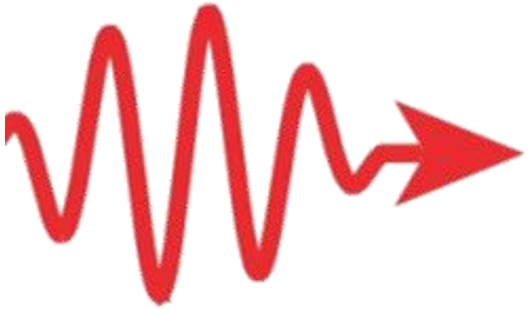


Lozano, Verschuuren, Rivas

LED lighting & phosphors

Making a light source





Space

- Whereto does the photon go ?
- With what polarization ?

Time

- How *long* does it take for the photon to appear ?

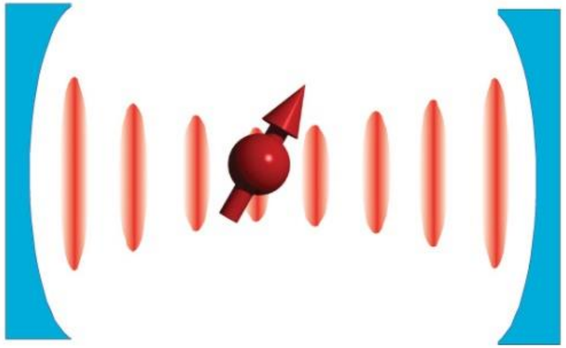
Matter

- Selection rules – what color comes out?

Engineering light

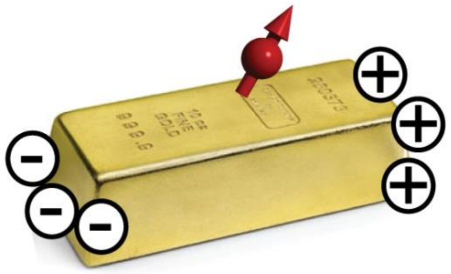
Engineering wave functions

Basic scenarios



Microcavity - barely leaks light

- high Q [stores light ~ 10^5 optical cycles]
- volume encloses a standing wave (λ^3)

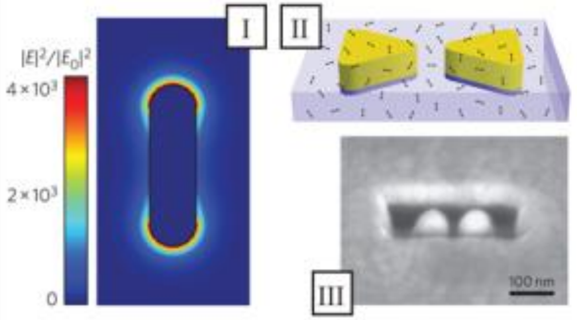


Antenna - excellent radiator

- low Q [looses light in 10 optical cycles]
- `antenna gain & directivity'

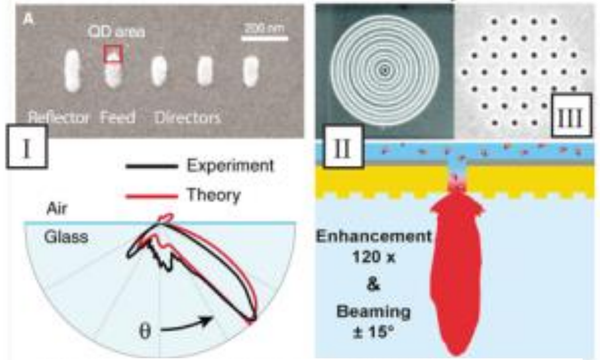
Antenna achievement chart

(a) Dipole resonators



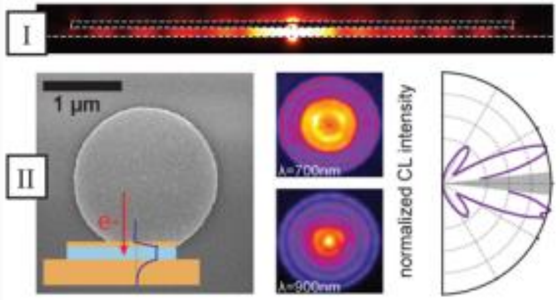
Orrit, Moerner, Wenger

(b) Phased arrays



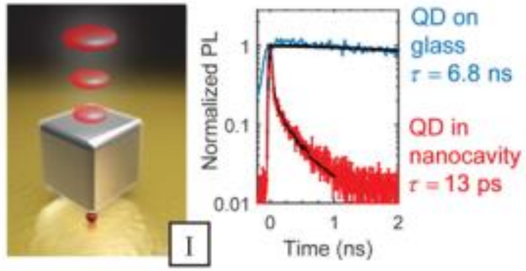
van Hulst, Wenger, AMOLF

(c) Patch / MIM-based



Greffet, AMOLF

(d) Nano-patch antenna



Mikkelsen, Baumberg

Brighter per fluorophore

1000x brighter

[total counts /sec on detector]

Directional sources

From 4π sr to 15º beams

[counts /sr / sec on detector]

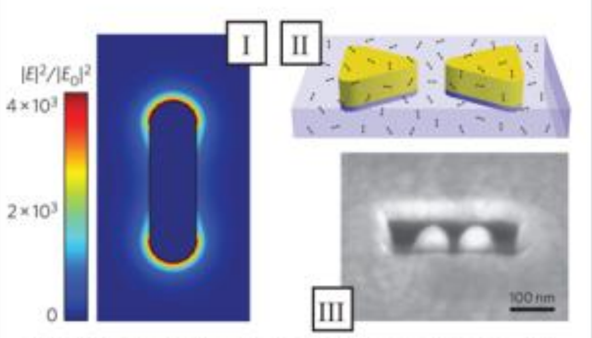
Directional & faster

1000x *faster* and brighter

[fluorescence decay rate]

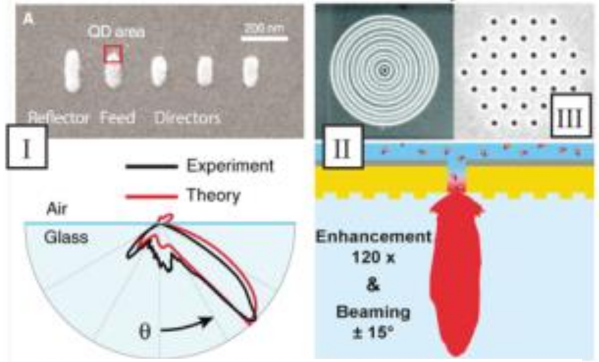
Antenna achievement chart

(a) Dipole resonators



Orrit, Moerner, Wenger

(b) Phased arrays



van Hulst, Wenger, AMOLF

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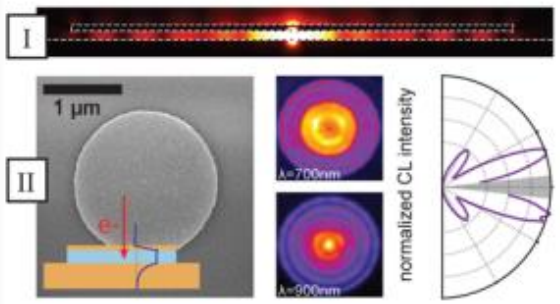
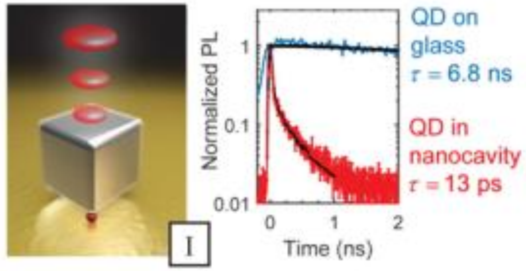


Greffet

(d) Nano-patch antenna



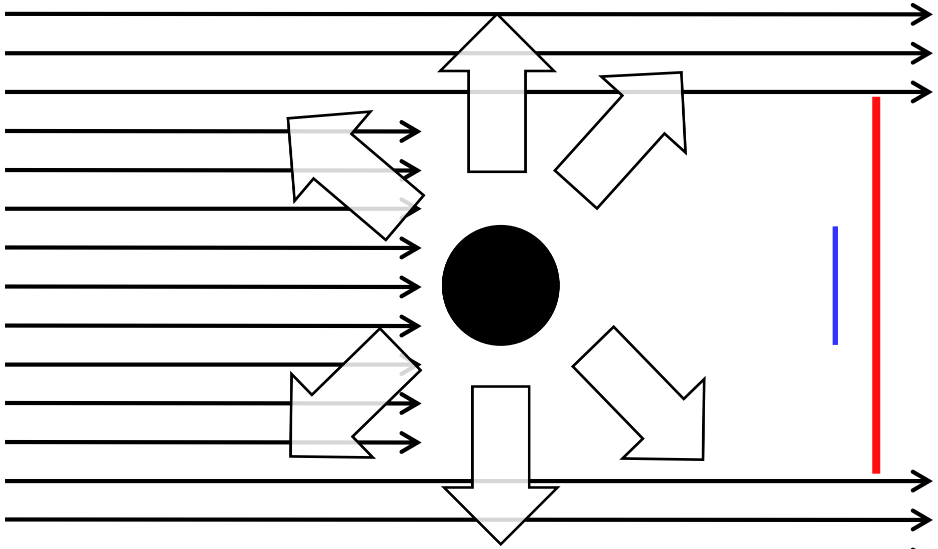
Mikkelsen, Baumberg

Why are metal particles extremely strong scatterers?
What is an `extremely strong' scatterer anyway?

How do you generate directivity from a point source?
How does emission get accelerated?
What determines brightness?
What is a good *measurement* for performance?

Scattering & observables

AMOLF physics of functional complex matter



Extinction cross section [m^2]

$$\frac{\text{Power removed from beam}}{\text{Incident intensity}}$$

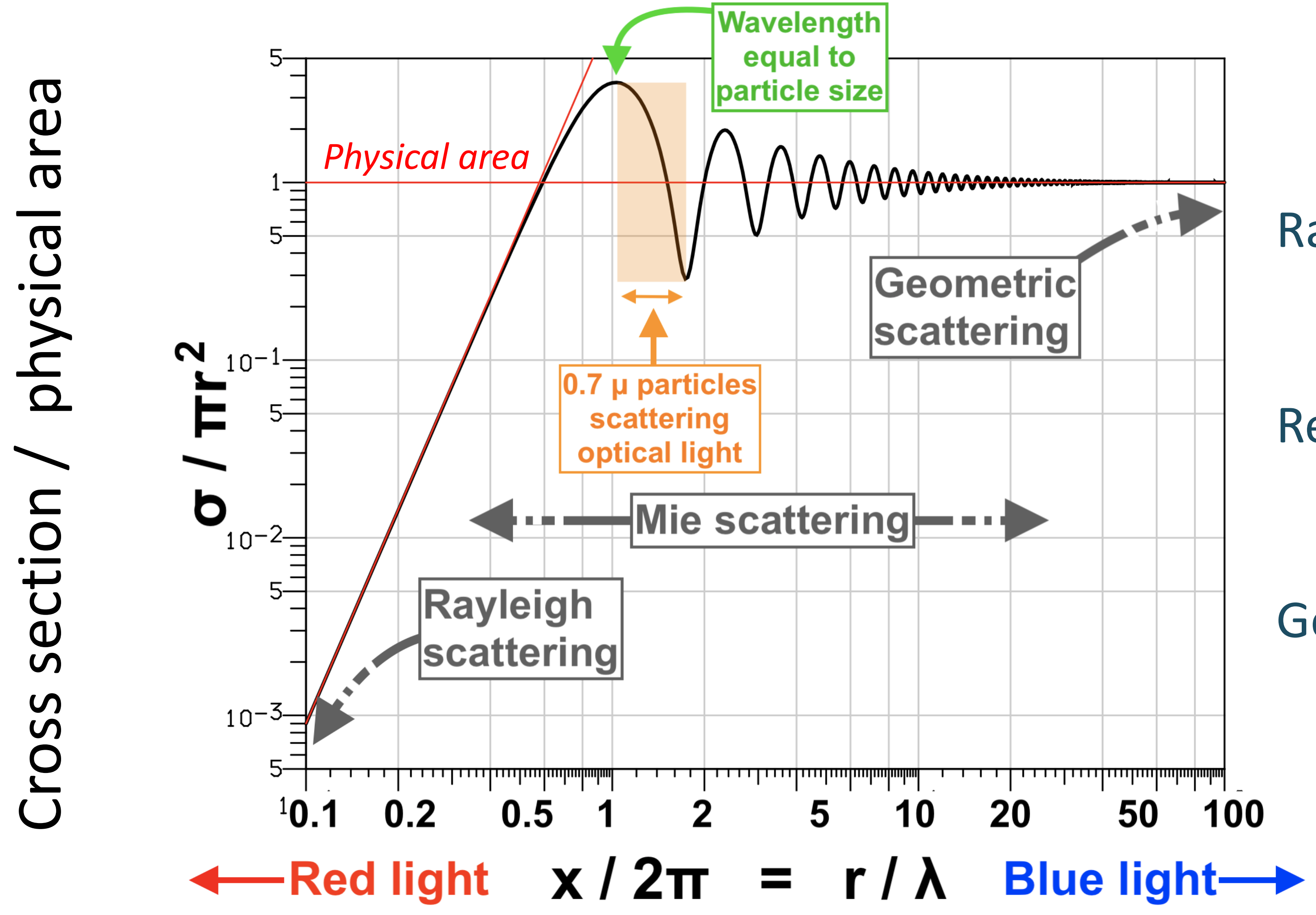
Extinction = scattering + absorption

removed from the beam

Re-radiated into all angles

Lost as heat in the scatterer

Scattering figures of merit



Rayleigh regime: $(r/\lambda)^4$

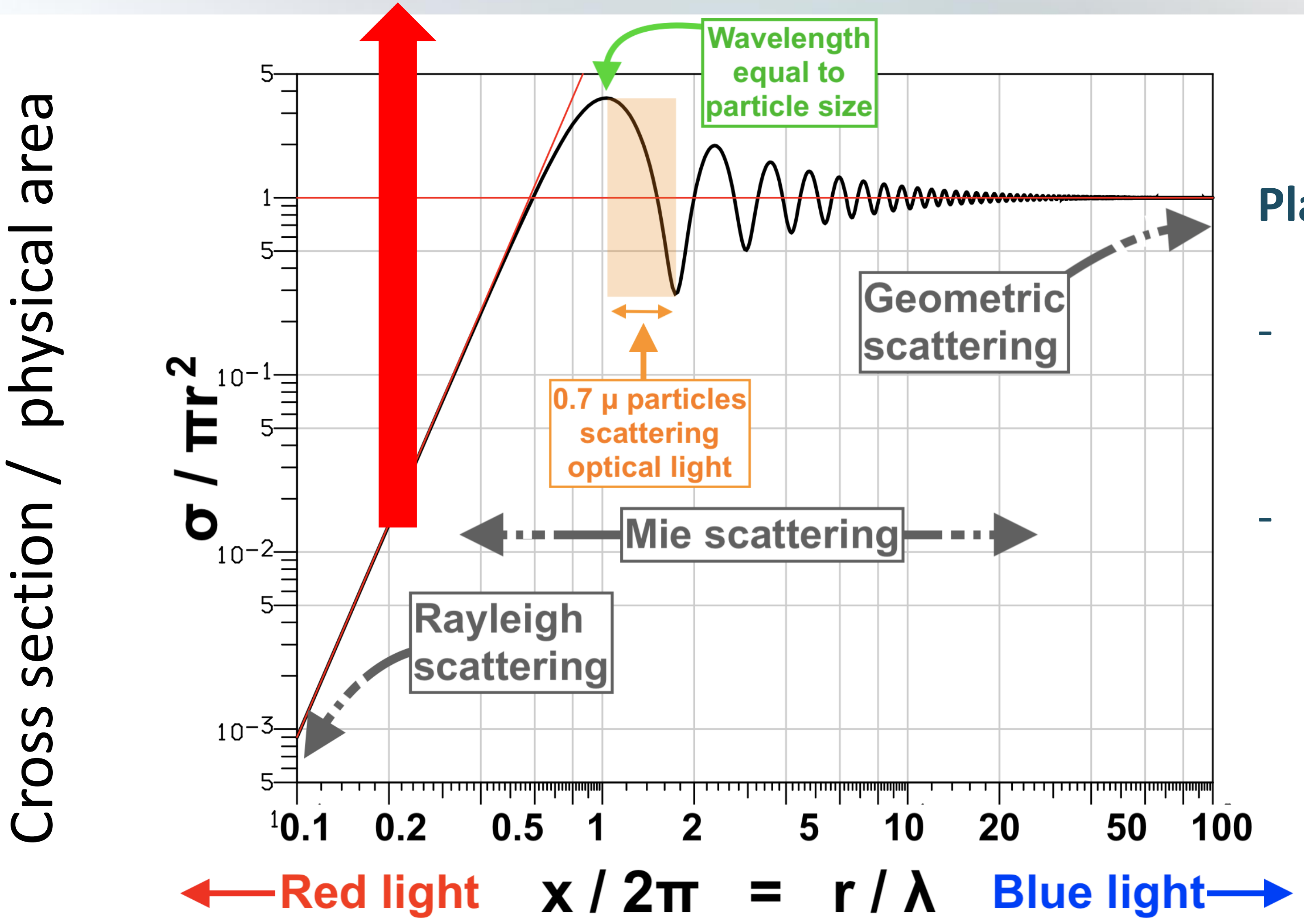
Resonances create $\sigma > \pi r^2$

Geometric resonances require size

Diagram: astronomy.stackexchange.com - actual theory: G. Mie, H. C. van de Hulst

The diagram is not quite correct – in the geometric limit, you actually find *twice* the physical cross section (Bohren & Huffman)

Scattering figures of merit



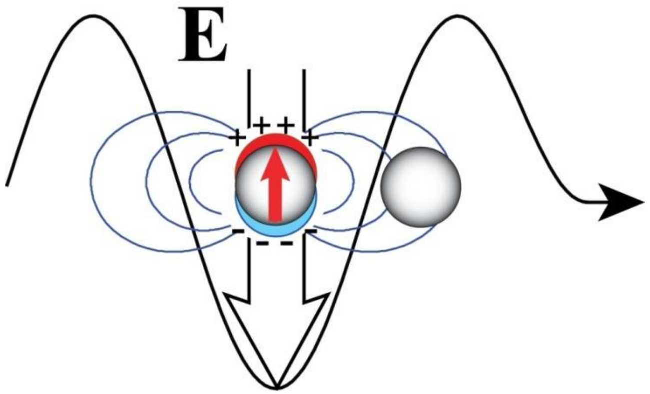
Plasmonics

- Deep sub-λ size, no geometry-only resonances
- Material resonance ensures large cross section

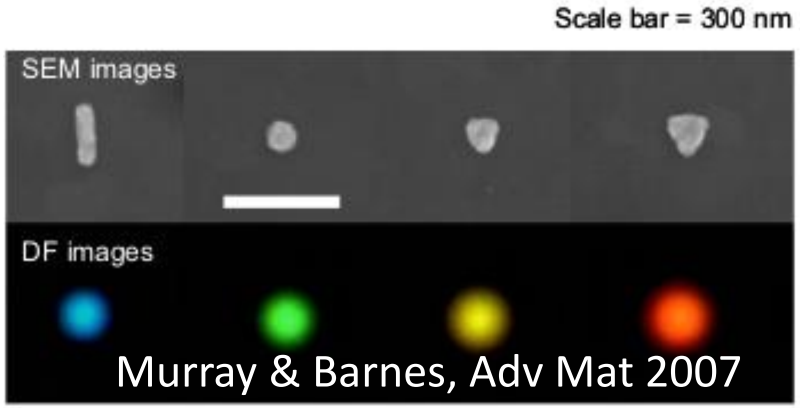
Diagram: astronomy.stackexchange.com - actual theory: G. Mie, H. C. van de Hulst

The diagram is not quite correct – in the geometric limit, you actually find *twice* the physical cross section (Bohren & Huffman)

Scattering resonance



$$\mathbf{p} = \alpha \mathbf{E}$$



Murray & Barnes, Adv Mat 2007

Circa 10^3-10^4 free electrons

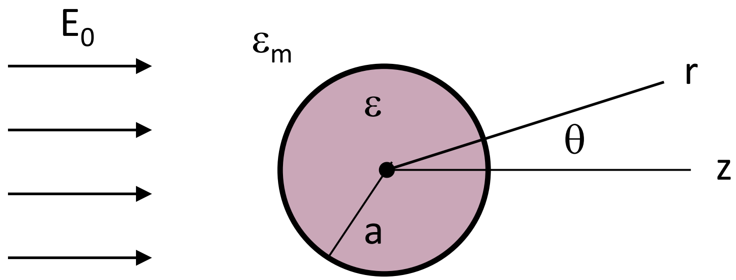
Incident field separates e- from ionic backbone

Linear restoring force implies a resonance

Resonant dipole scatterers
λ ~300-1000 nm, Q ~ 5-30

Induced dipoles

$$\mathbf{p} = \int_V \epsilon_0 \chi \mathbf{E}_{in} dV = \epsilon_0 V(\epsilon - 1)\mathbf{E}_{\text{in}}$$



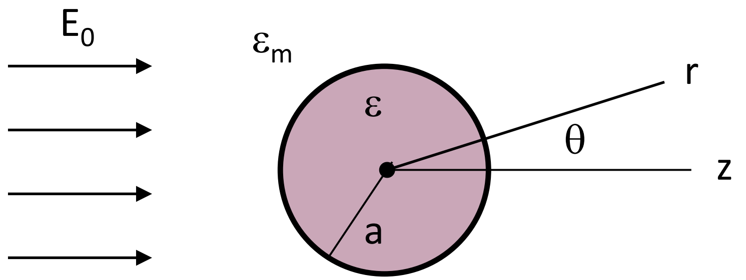
Qualitatively:

Incident field induces local polarization density in the medium $\mathbf{P} = \epsilon_0 \chi \mathbf{E}_{\text{in}}$

If the object is so small that E_{in} hardly varies over it ($a << \lambda$), the local induced response adds up to an induced dipole

Electrostatic sphere

Electrostatic treatment (frequency = 0)
Sphere in a homogeneous electric field



In the ball:

$$\Phi_1 = -E_0 r\cos\theta + \left(\frac{\varepsilon-\varepsilon_m}{\varepsilon+2\varepsilon_m}\right)E_0 r\cos\theta = -\left(\frac{3\varepsilon_m}{\varepsilon+2\varepsilon_m}\right)E_0 r\cos\theta$$

Outside:

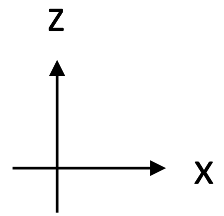
$$\Phi_2 = -E_0 r\cos\theta + a^3\left(\frac{\varepsilon-\varepsilon_m}{\varepsilon+2\varepsilon_m}\right)E_0\frac{\cos\theta}{r^2} = -E_0 r\cos\theta + \frac{p\cos\theta}{4\pi\varepsilon_m r^2}$$

Incident plane wave E_z — Scattered, dipole

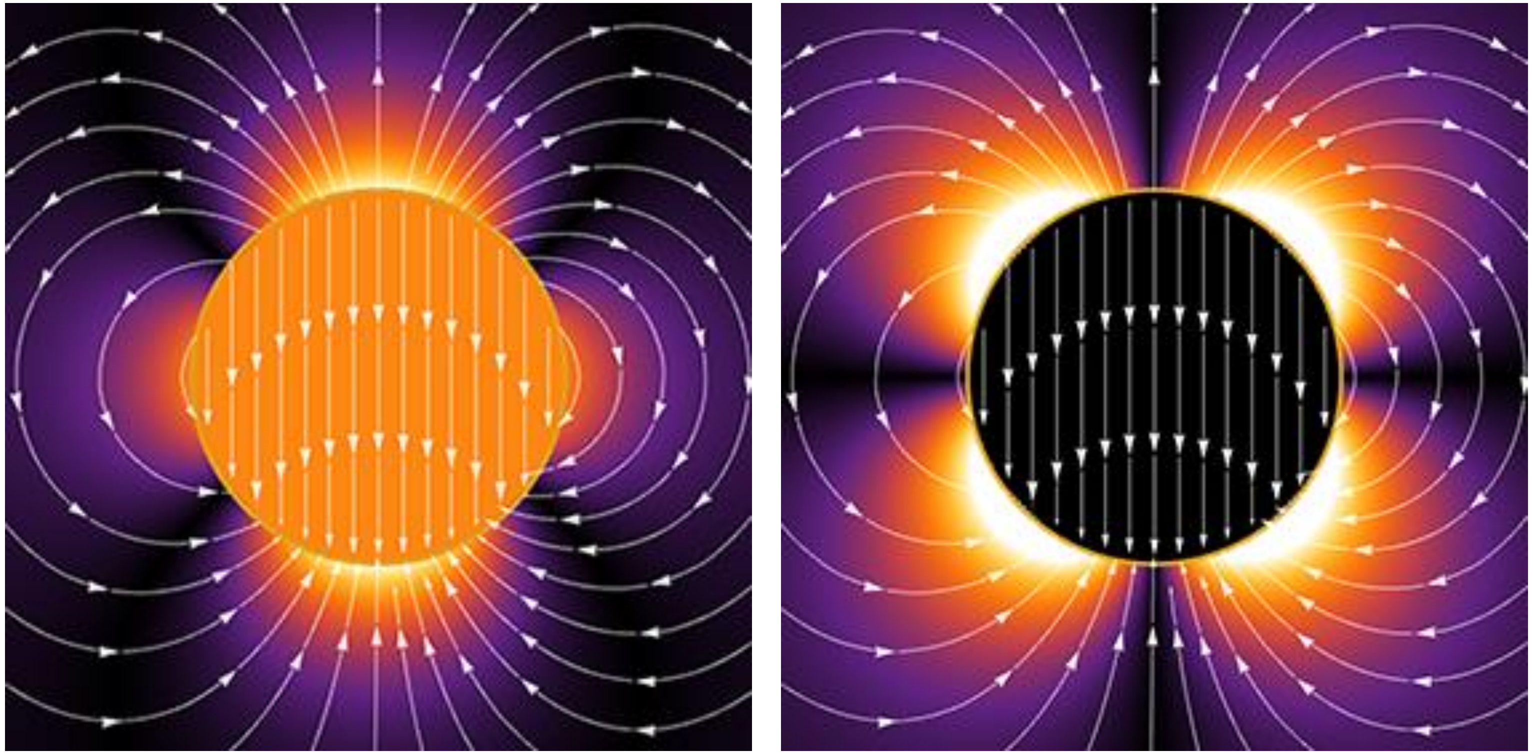
Inside sphere: homogeneous field along incident field (E_z)
Outside sphere: background field plus **field of a dipole**

Check: (1) this potential is itself *continuous,* yet (2) its derivative jumps by ε, since Div D=0

Electrostatic solution – scattered field



$|E_z|$ $|E_x|$

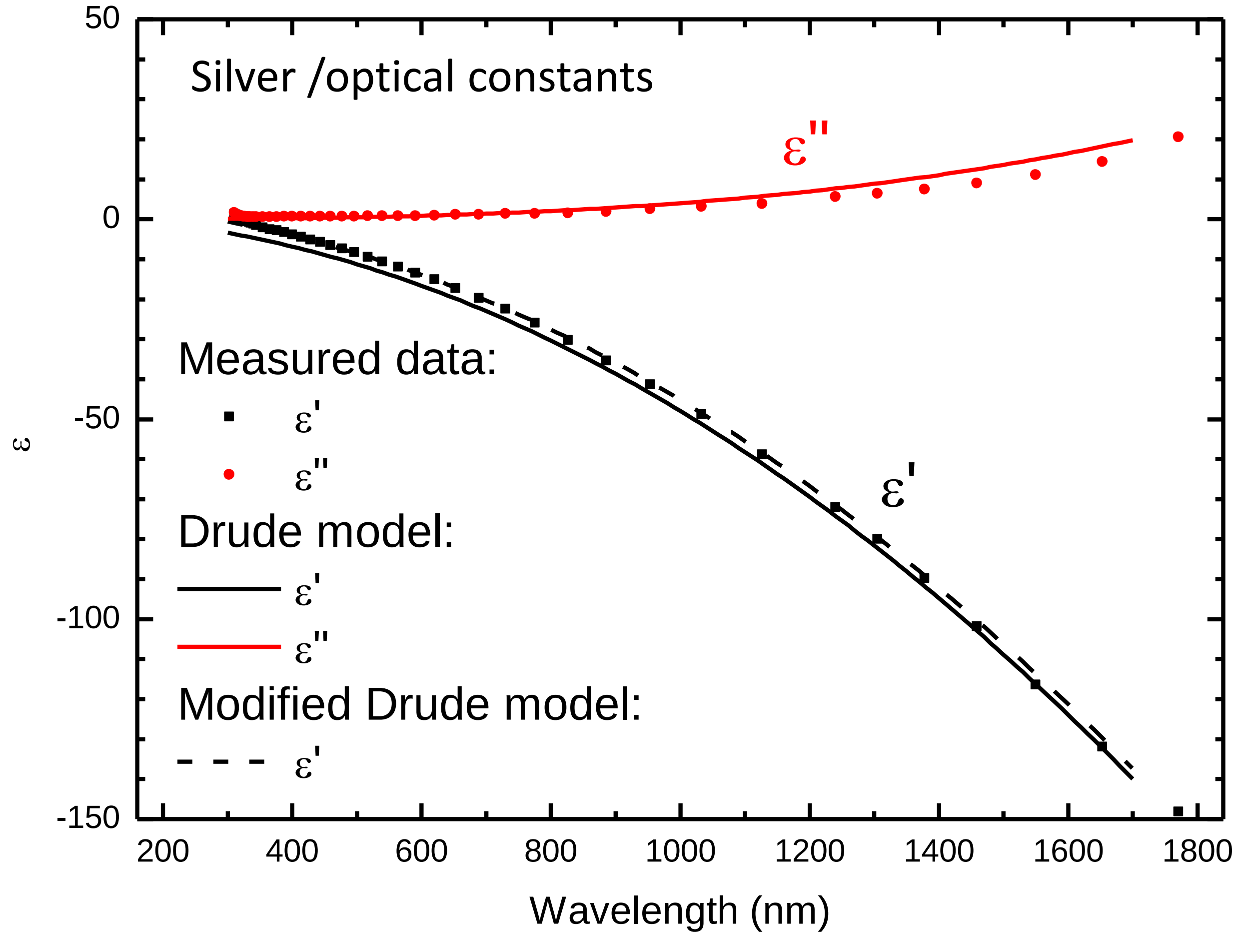


http://people.ee.duke.edu/~drsmith/plasmonics/enhancement.htm

$$\vec{p} = \alpha_{SI} \vec{E}_0 \quad \text{with} \quad \alpha_{SI} = 4\pi\varepsilon_0\varepsilon_m a^3 \left(\frac{\varepsilon - \varepsilon_m}{\varepsilon + 2\varepsilon_m} \right)$$

Drude model

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



Electrostatics: free electrons shield all fields

$\varepsilon = -\infty$ at $\omega = 0$

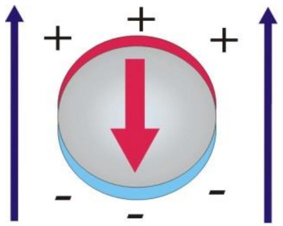
Natural response “plasma” frequency in UV

ε flips sign, will go through $-2\varepsilon_m$

Deep UV: electrons can not keep up

$\varepsilon = 1$ (transparency) for $\omega > \omega_p$

Metal sphere



$$\vec{p} = 4\pi\epsilon_0\alpha_{\text{easy}} \quad \text{with} \quad \alpha_{\text{easy}} = a^3\left(\frac{\epsilon-1}{\epsilon+2}\right)$$

Units of volume [m^3]

Drude model for a metal: Lorentzian `plasmon resonance’

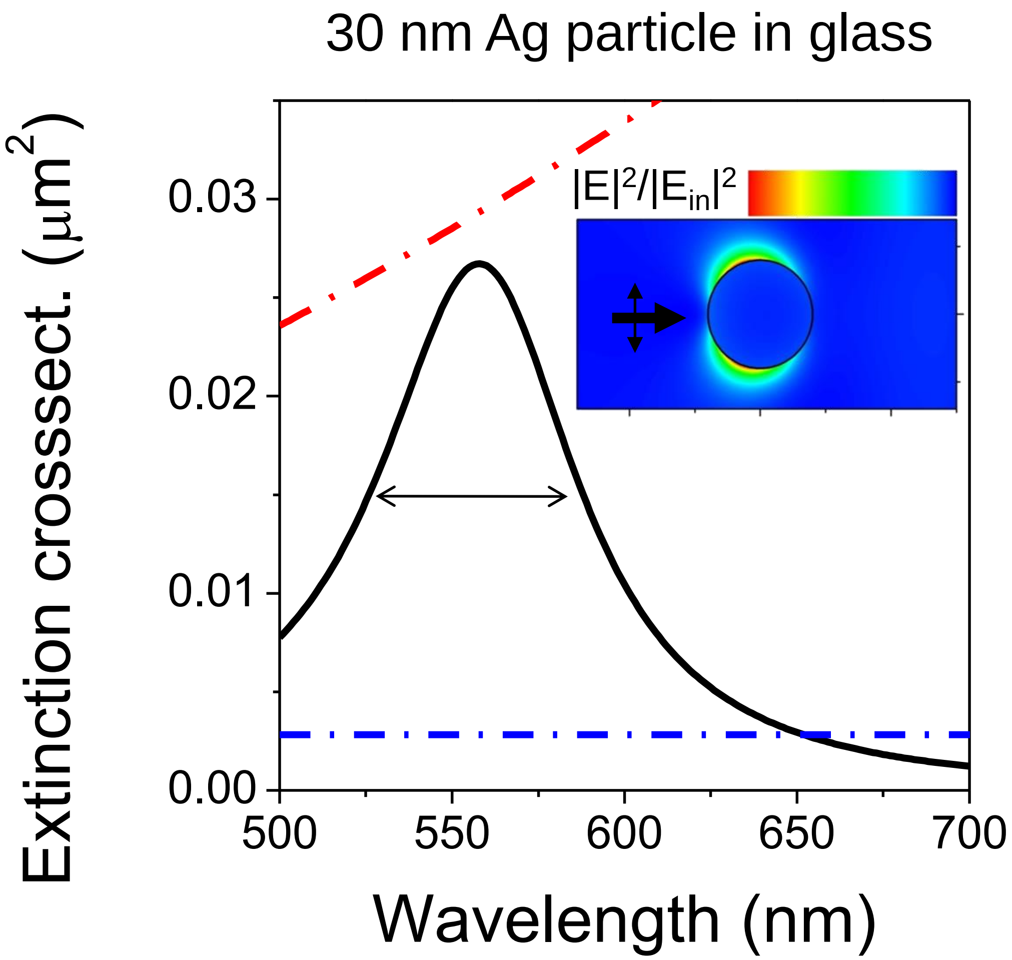
$$\varepsilon = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \quad \text{means} \quad \alpha_{easy} = a^3\left(\frac{\omega_0^2}{\omega_0^2 - \omega^2 + i\omega\gamma}\right)$$

$$\omega_0 = \omega_p/\sqrt{3}$$

Note: Now I chose the medium to be vacuum ε_m=1. Otherwise you still get a resonance, but not quite Lorentzian

General properties

AMOLF physics of functional complex matter



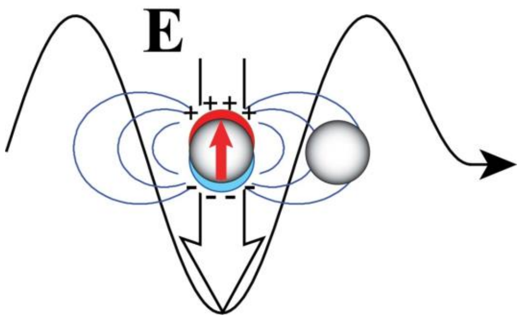
- Color tunable in visible
  $\varepsilon+2\varepsilon_m$, shape effects
- Cross section ~ 10x πr^2
- Strong dipolar near field

$$E_{\text{dipole}} = \frac{p}{4\pi\epsilon_0 r^3} \propto \left(\frac{\epsilon-1}{\epsilon+2}\right)\frac{a^3}{r^3}E_{\text{in}}$$

$$|E_{\text{dipole}}/E_{\text{in}}| \propto |\text{Re}[\epsilon-1]/\text{Im}[\epsilon]| = 40$$

Optical theorem

AMOLF physics of functional complex matter



Extinction = scattering + absorption

Extinction		Scattering
$W \propto \left\langle E_{in} \cdot \frac{dp}{dt} \right\rangle \propto \mathrm{Im}\,\alpha$	$\geq$	$P \propto \lvert p \rvert^2 \propto \lvert \alpha \rvert^2$
Work done to drive p		*Radiated power P*

Proportional to volume V *Proportional to V^2*

In these units: extinction: σ_{ext} = $4\pi k\ \mathrm{Im}\alpha$ and $\sigma_{scat} = 8\pi/3k^4\ \lvert\alpha\rvert^2$ - see for units Phys. Rev. B **83**, 245102, (2011).

Optical theorem

Dimensional analysis

- Rayleighs' law: scattering cross section [m^2] scales as $V^2 \sim r^6$ $\sigma_{scat} \sim r^2 (r/\lambda)^4$

- Absorption or scattering? extinction [m^2] scales as $V \sim r^3$ $\sigma_{ext} \sim r^2(r/\lambda)$

  small particles only absorb

- Since extinction > scattering, polarizability is limited in magnitude

$$\mathrm{Im}\,\alpha \geq \frac{2}{3} k^3 |\alpha|^2 \qquad k = 2\pi/\lambda$$

More polarizability means more radiative loss - limiting polarizability

Note: Since clearly $|\mathrm{Im}\,\alpha| < |\alpha|$, you immediately find $|\alpha|$ is bound to be no greater than $3/2\ k^{-3}$

Example: simple spheres

Calculated exact cross section of Au spheres r=10 -50 nm

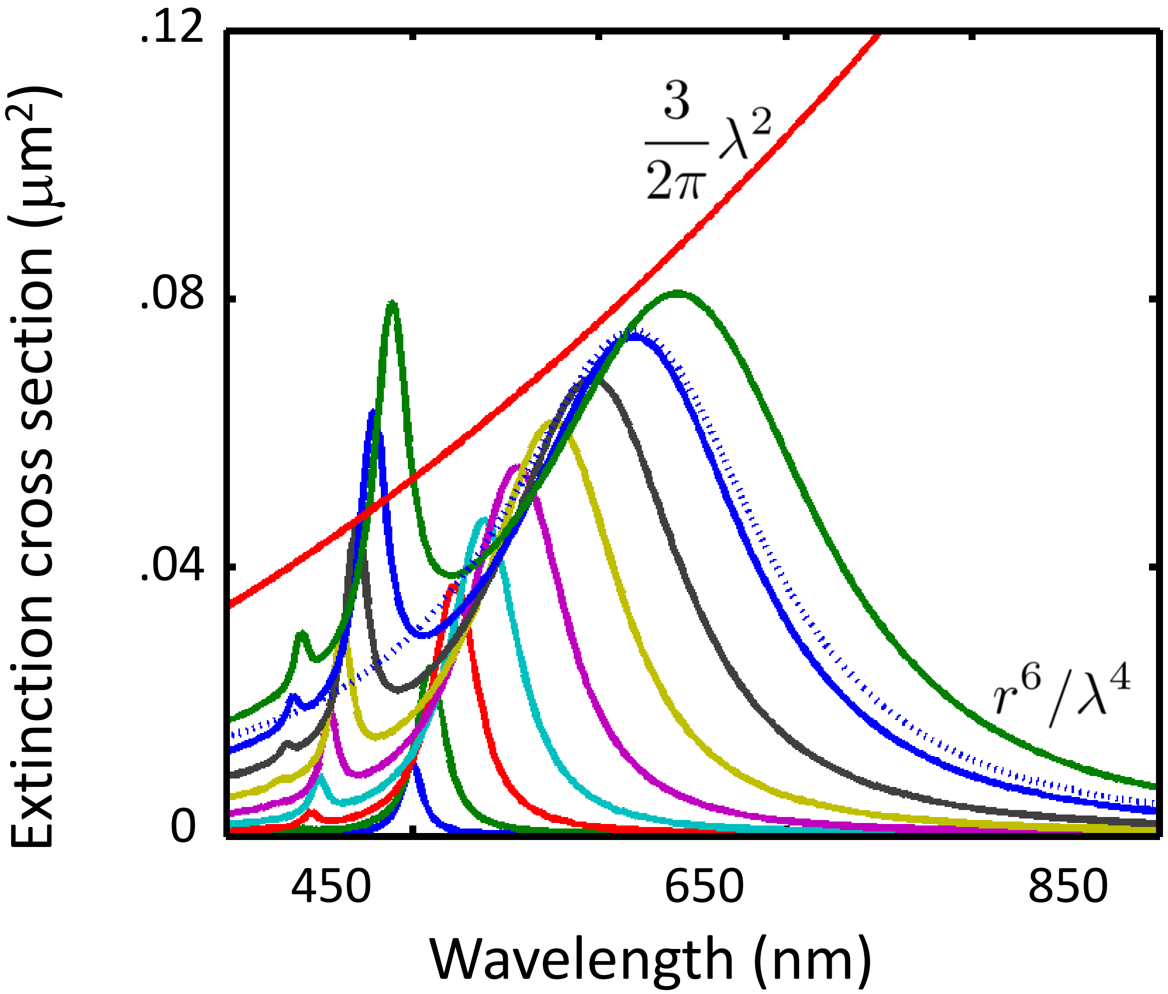
(1) Long wavelength - Rayleigh

(2) Increase in σ with volume

(3) Until σ approaches $\frac{3}{2\pi}\lambda^2$

(4) Radiation damping lowers antenna Q

(5) Absorption bounds Q, radiation limits σ

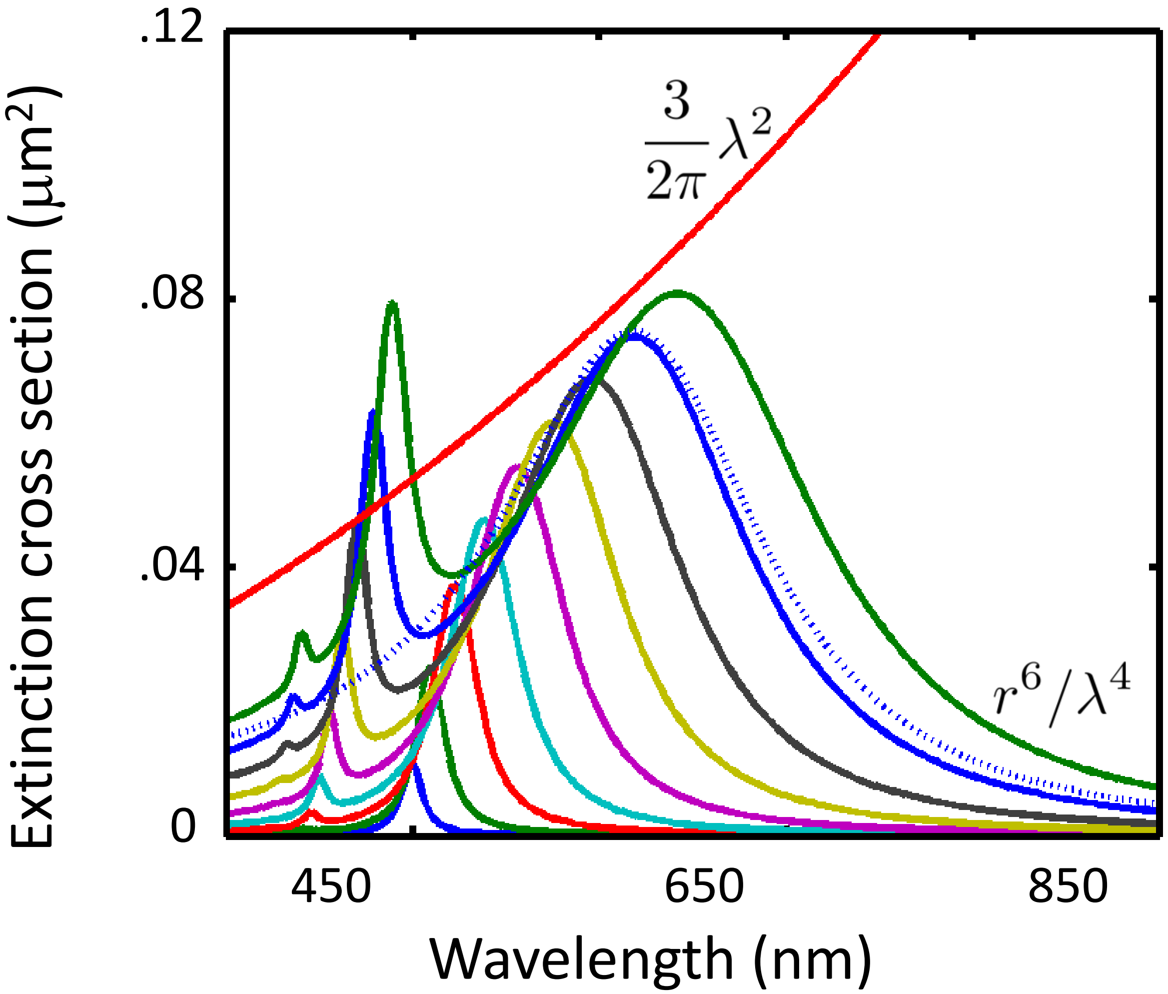


Example: simple spheres

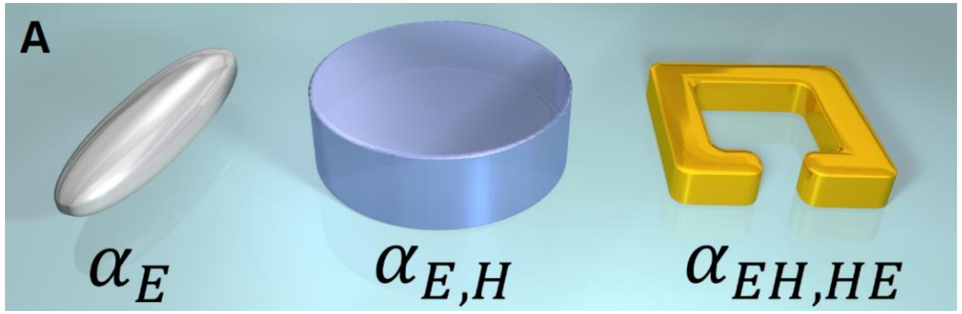
$$\alpha_{\text{electrostatic}} = a^3 \left(\frac{\omega_0^2}{\omega_0^2 - \omega^2 - i\omega\gamma} \right)$$

$$\frac{1}{\alpha} = \frac{1}{\alpha_{\text{electrostatic}}} - i\frac{2}{3}k^3$$

Addition of "radiation damping"



Beyond point polarizability



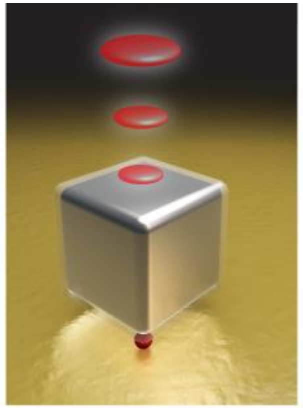
Polarizability intuition extends to:

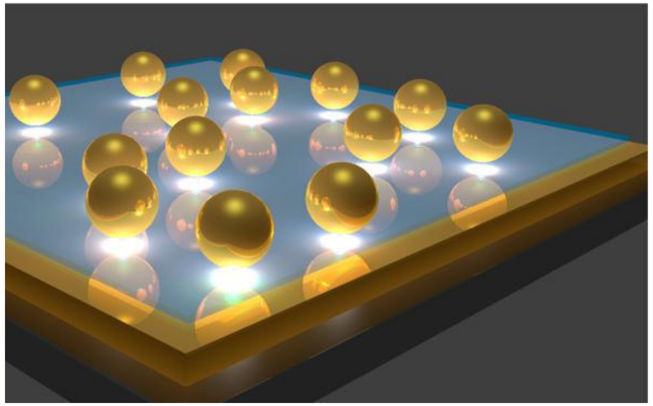
Magnetic scatterers

Magneto-electric, chiral scatterers

Magnetic modes in Mie scatterers

Isabelle Staude





Similar cross sections but far larger local |E|^2

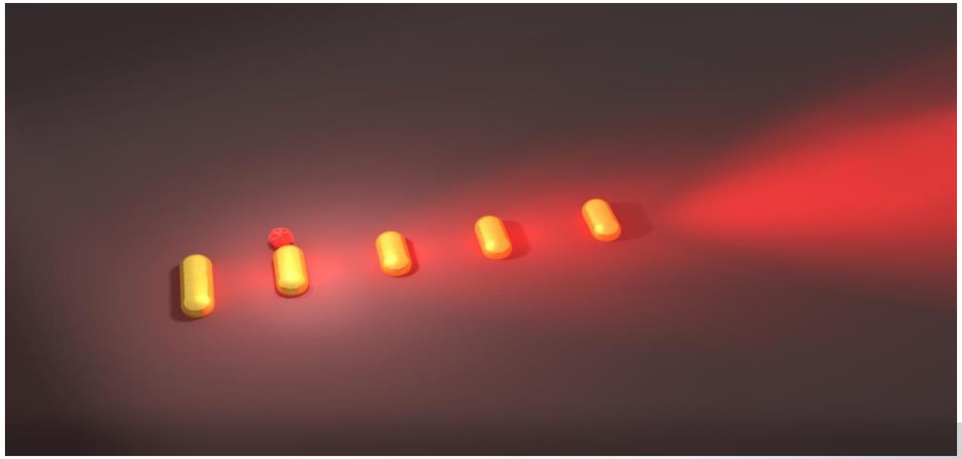
Gap-plasmons

Nanocube patch antennas

Nanosphere on mirror antennas

Javier Aizpurua

Mikkelsen, Baumberg



Oligomers & arrays

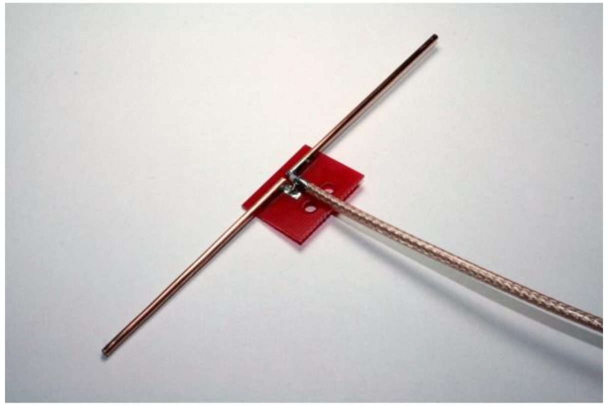
Hybrid, higher Q multipolar modes

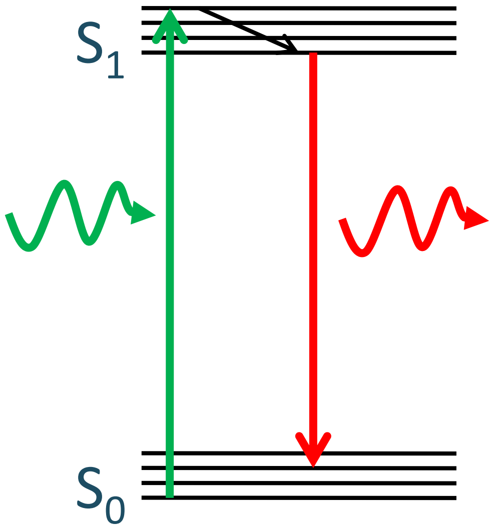
Phased arrays, metasurfaces

Curto, Giessen

Engheta, AMOLF

What is a source ?





Antenna – driven dipole
Fixed *current* source

Spontaneous emitter
$\hbar\omega$ at a time

Three distinct effects

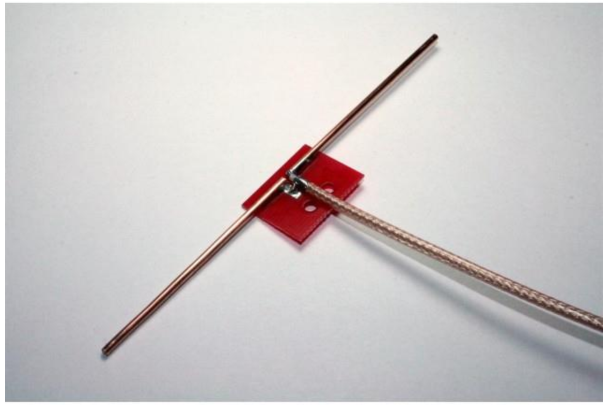
- Source brightness [counts/sec]   is enhanced because pump field is *concentrated* at the emitter

- Given that the source has been excited and emits a photon, antenna effects redirect the light into a narrow beam

- Fluorescence decay rates are enhanced - Purcell effects on rate and IQE

Radiation provides resistance

$$\nabla \times \mathbf{H} = \mathbf{J}_{\mathrm{f}} + \frac{\partial \mathbf{D}}{\partial t}$$

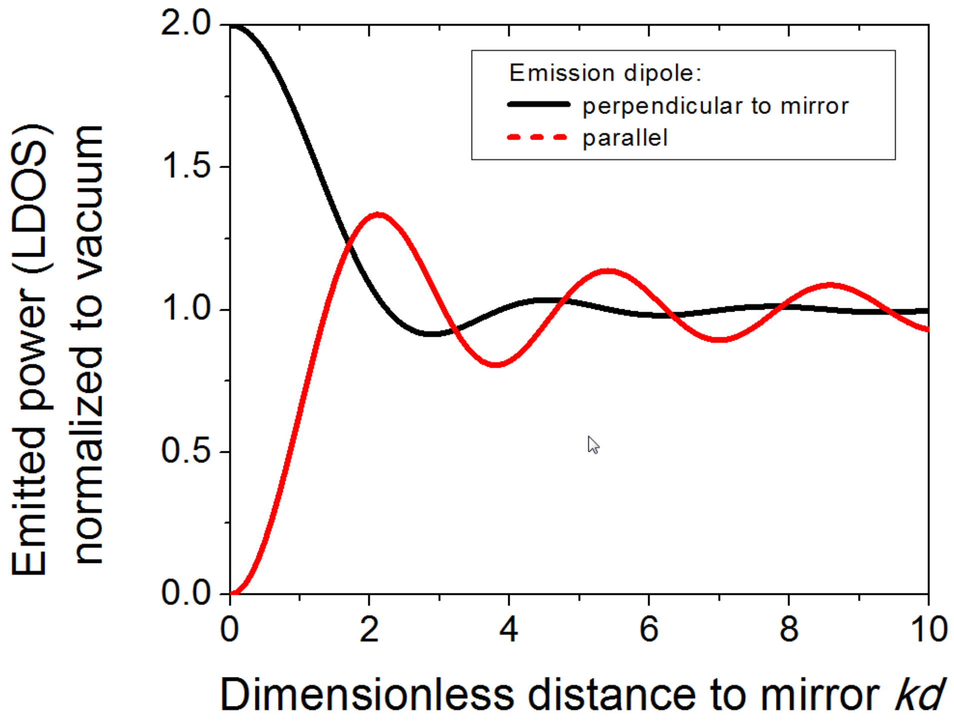


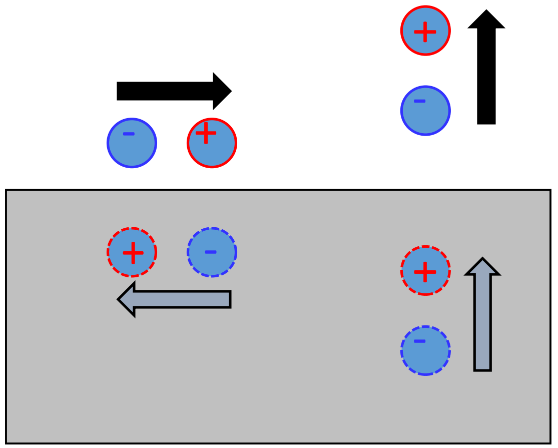
Radiation resistance – environment sets power to current ratio

The *work* you need to do keep current **j** going depends on environment

Conversely, the *same* current can generate more power (or less)

Dipole above a mirror in classical terms

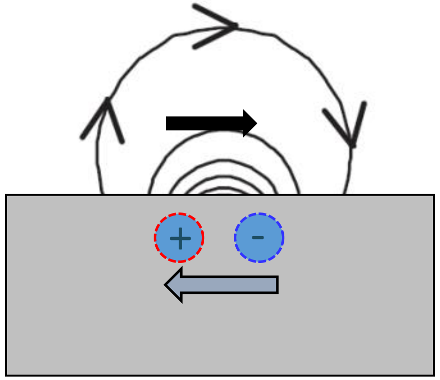




Interference of the dipole field with that of its mirror image

The same current radiates a *different* far field power

In order to *maintain* constant current
one does work against ones *own* field

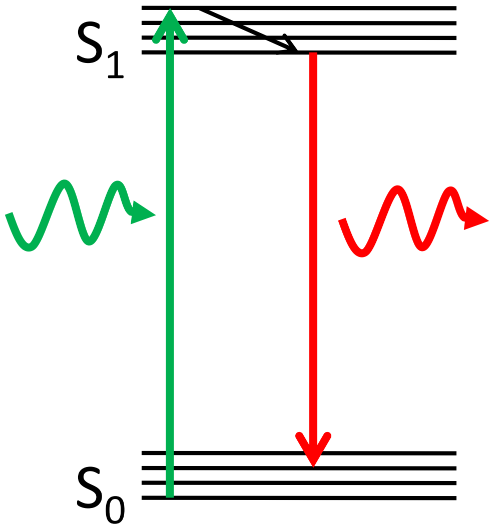


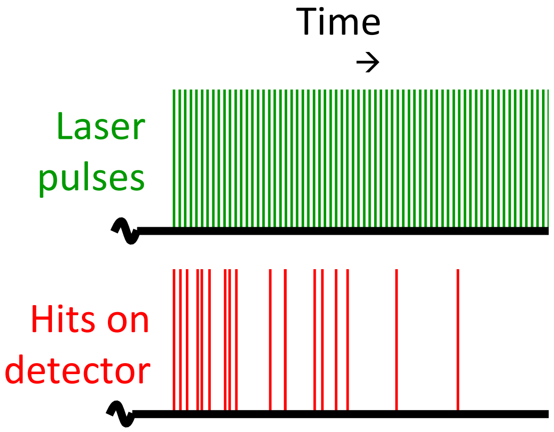
$$W = \langle \mathbf{F} \cdot \frac{d\mathbf{x}}{dt} \rangle = \langle \mathbf{E} \cdot \frac{d\mathbf{p}}{dt} \rangle$$
$$= \langle \mathrm{Re}\mathbf{E}e^{i\omega t} \cdot \mathrm{Re}(i\omega \mathbf{p}e^{i\omega t}) \rangle$$

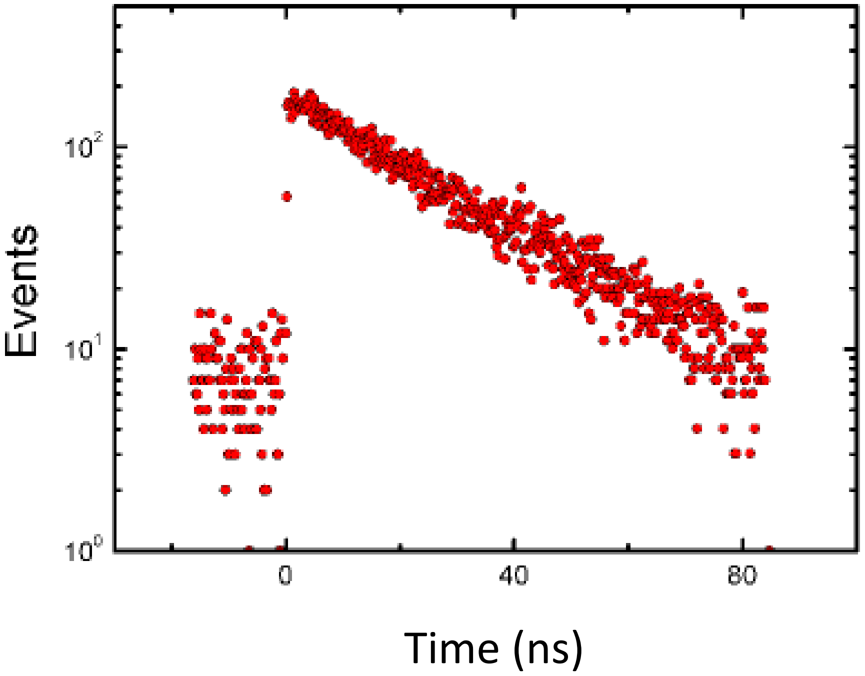
Suppose we call the dipole field $\mathbf{E}(\mathbf{r}) = \mathbf{G}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{p}$

Resistance is due to field **Im p^T.G(r',r').p** that comes back to the source
"Imaginary part of the Green function"

Single quantum emitter

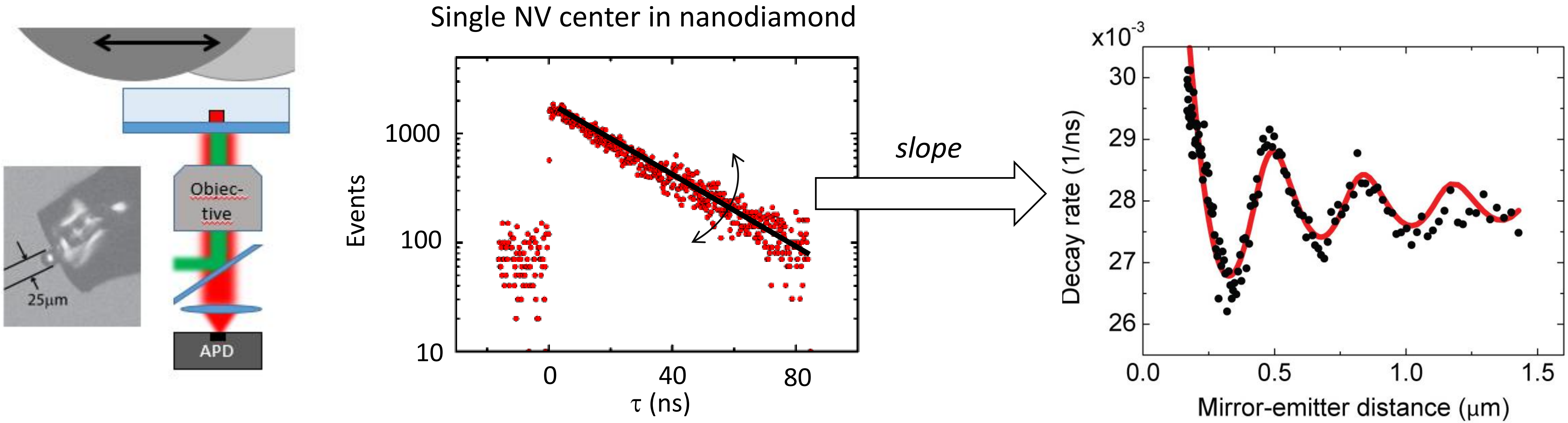






- After one excitation, emits just one quantum of light
- Probabilistic timing of *when* emission occurs
- Fermi's Golden rule: exponential decay, characteristic rate [inverse time]

Drexhage experiment on single quantum emitter



Note how: the *power* is not the variable of choice: one photon out per photon in (QE=1)
the *decay rate* varies with mirror-geometry

K.H. Drexhage and many since - ensembles of emitters (1966)
Single emitters: Buchler PRL (2005), Frimmer (2013), Huck (2016)

Understanding Fermi's Golden Rule

AMOLF physics of functional complex matter

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

Matrix elements:
Transition strength
Selection rules

Energy conservation

Spontaneous emission of a two-level atom:

Initial state: excited atom + 0 photons.

Final state: ground state atom + 1 photon in some photon state

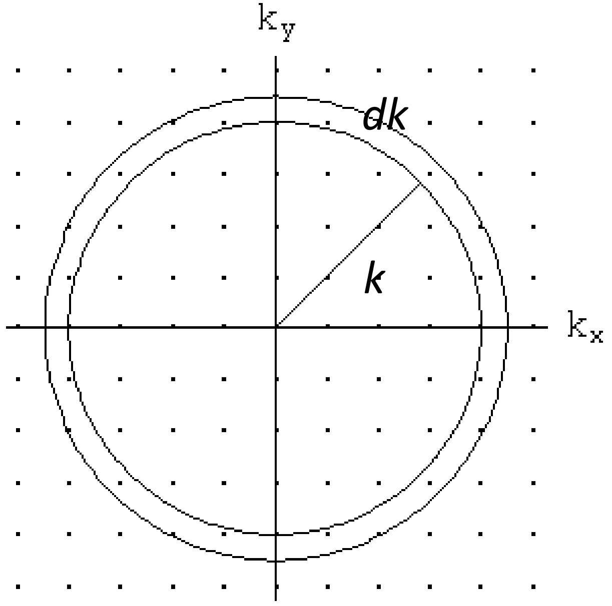
Question: how many states are there for the photon ???
(constraint: photon energy = atomic energy level difference)

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

States in an LxLxL box:

$$E(x,t) = Ae^{i\omega t}\sin(\mathbf{k}\cdot\mathbf{r}) \quad \text{with} \quad \mathbf{k} = \frac{\pi}{L}(l,m,n)$$

l,m,n positive integers



Picture from
http://britneyspears.ac

Number of states with |**k**| between *k* and *k+dk:*

$$N(k)dk = \frac{4}{8}\pi k^2 dk\left(\frac{L}{\pi}\right)^3 \cdot 2$$

l,m,n > 0
fill one octant

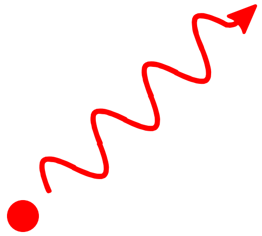
fudge 2 for
polarization

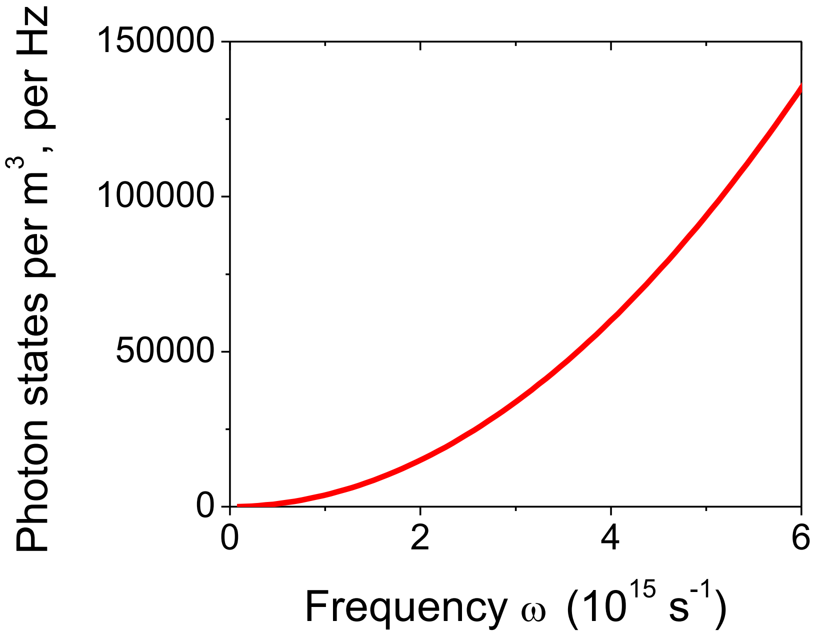
As a function of frequency ω ($=ck$):

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

Fluorescence decay rates

Fermi's Rule: Fluorescence rate ∝ number of photon states



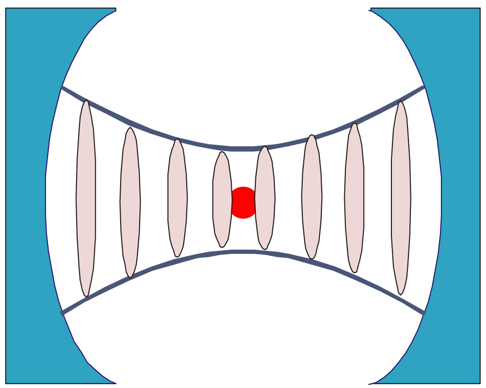


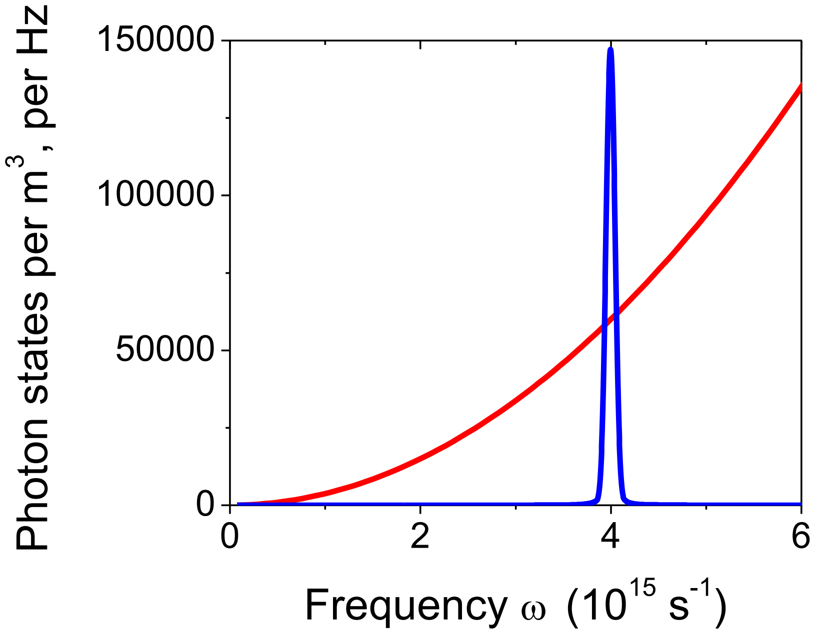
Visible light: ~10^5 photon states per Hz, per m^3 of vacuum

Loudon, The Quantum Theory of Light

Fluorescence in a cavity

Fermi's Rule: Fluorescence rate $\propto$ number of photon states

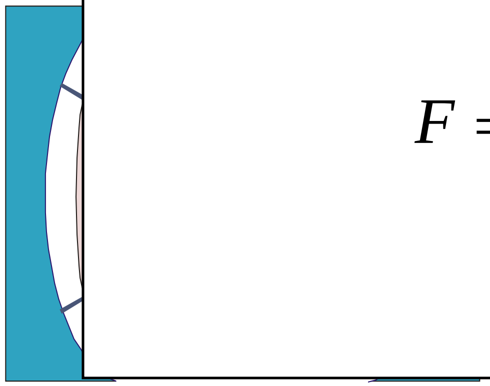




Microcavity: **Exactly** one extra state per $\Delta\omega=\omega/Q$ in a volume V

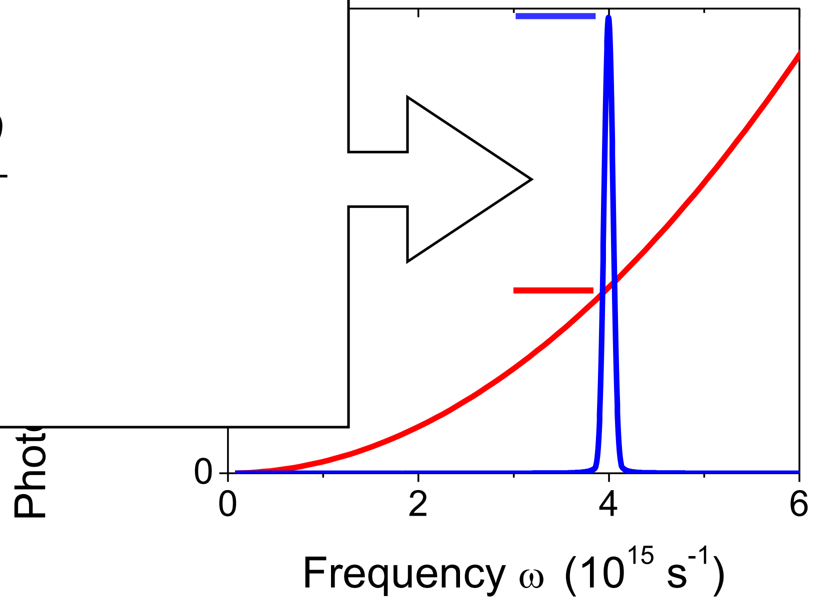
Fluorescence in a cavity

Fermi's Rule: Fluorescence rate $\propto$ number of photon states



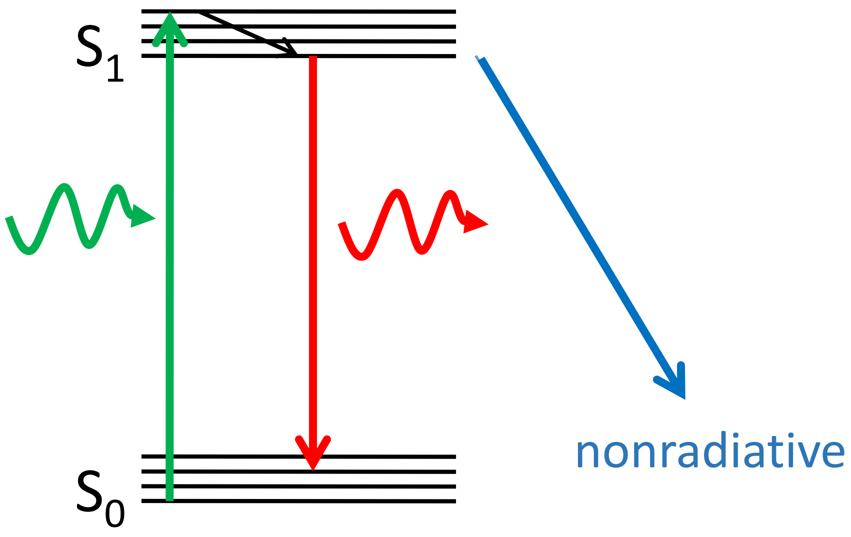
Purcell factor

$$F = \frac{3}{4\pi^2}\lambda^3\frac{Q}{V}$$



Microcavity: **Exactly** one extra state per Δω=ω/Q in a volume V

From LDOS to actual performance



$$\Gamma_{\text{total}} = \Gamma_{\text{nonrad}} + \Gamma_{\text{rad}} \times \text{LDOS}$$

$$\text{Q.E.} = \frac{\Gamma_{\text{rad}} \times \text{LDOS}}{\Gamma_{\text{nonrad}} + \Gamma_{\text{rad}} \times \text{LDOS}}$$

A unit efficiency emitter, emits exactly a single $\hbar\omega$ per pump photon

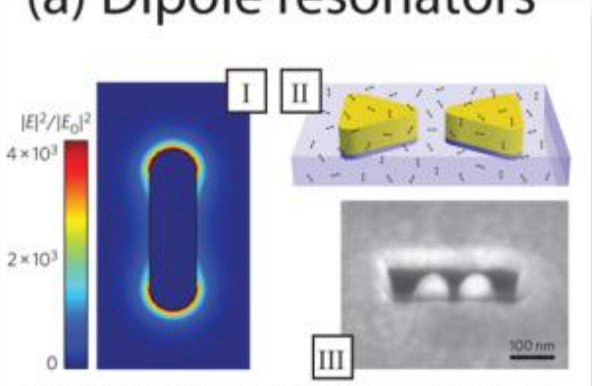
High LDOS will make the photon come out faster, but the source is not more bright

A low-efficiency sources gains efficiency by outcompeting nonradiative loss

LDOS itself may contain new loss channels - quenching

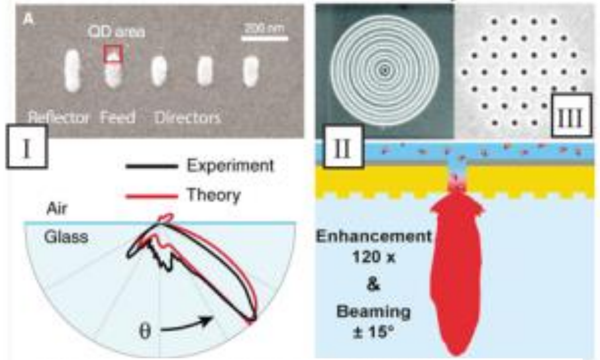
Antenna achievement chart

(a) Dipole resonators



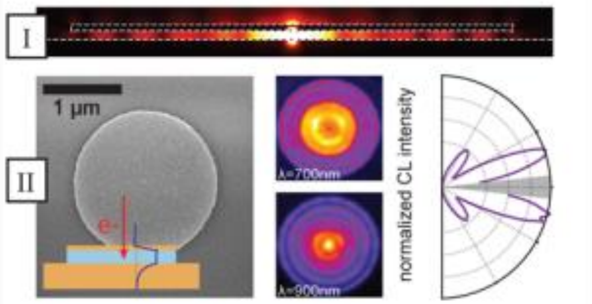
Orrit, Moerner, Wenger

(b) Phased arrays



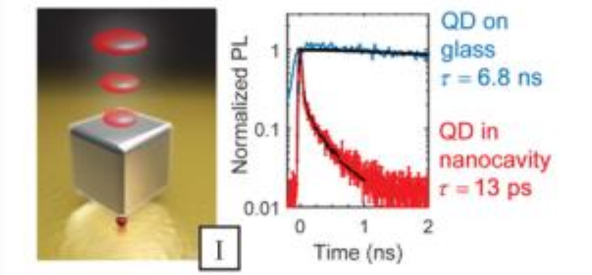
van Hulst, Wenger, AMOLF

(c) Patch / MIM-based



Greffet

(d) Nano-patch antenna



Mikkelsen, Baumberg

Brighter per fluorophore

1000x brighter

Intrinsically poor emitters

- **10x boost in QE**
- **100x boost in pump**

Brighter by beaming

Factor ~ 5

Directional & faster

Intrinsically good emitters

- **500 - 1000x *faster***
- **Brighter by pump boost**

Doing a good measurement is not easy

$$I(\mathbf{r}, \omega_{pump}, \omega_{em}) \propto P_{pump}(\mathbf{r}, \omega_{pump}) \cdot \varphi(\mathbf{r}, \omega_{em}) \cdot C_{NA}(\mathbf{r}, \omega_{em})$$

Averaging over many emitters is highly misleading

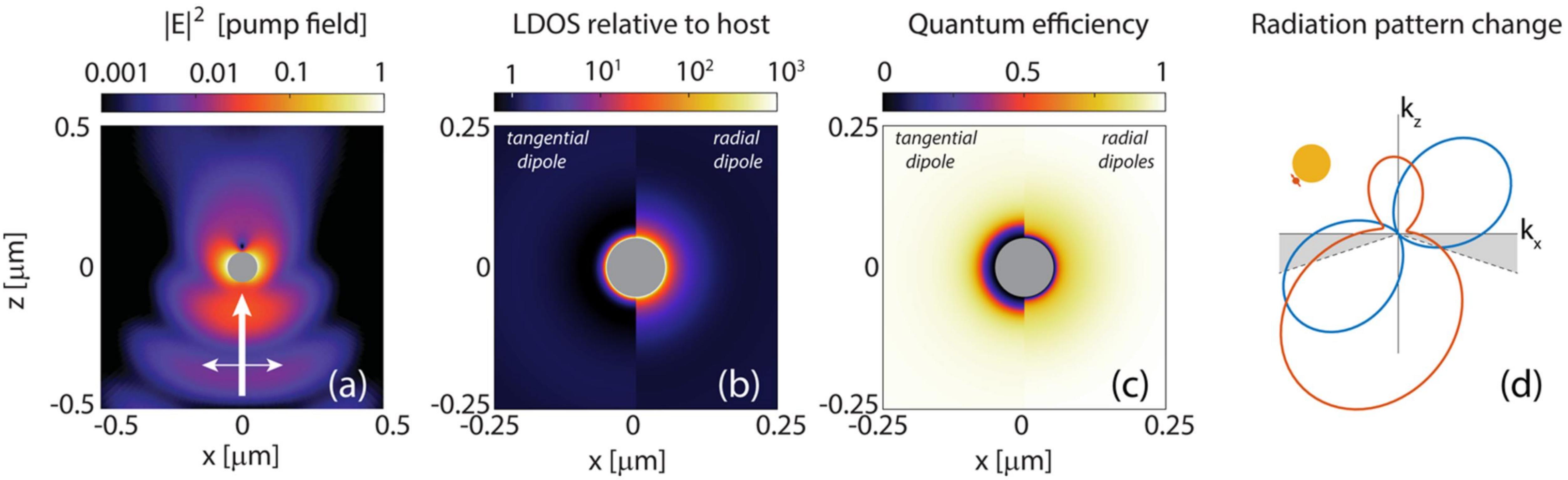
1. Average over a diffraction limit is useless - very far from peak numbers

2. Average is biased – fluorescence signals select on unquenched emitters

3. Product of averages *and* ***average of product*** *is very different*

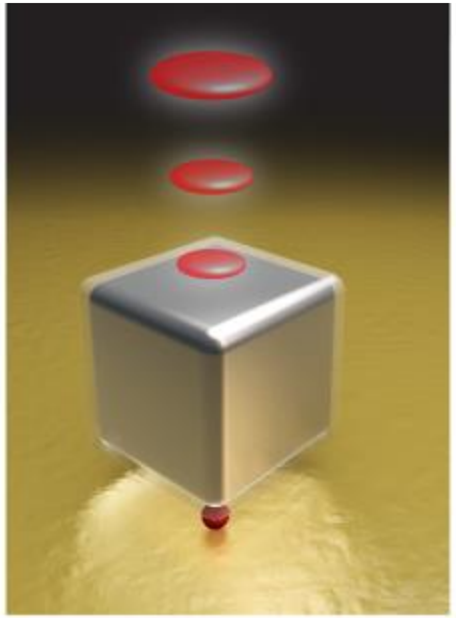
- Make sure you have a *single* emitter (at a time)
- Of known quantum efficiency
- At the right location
- Calibrate *collection efficiency, directivity, rate, and efficiency*
- *All that* in a limited photon budget [< 10^7 total detected photons usually]

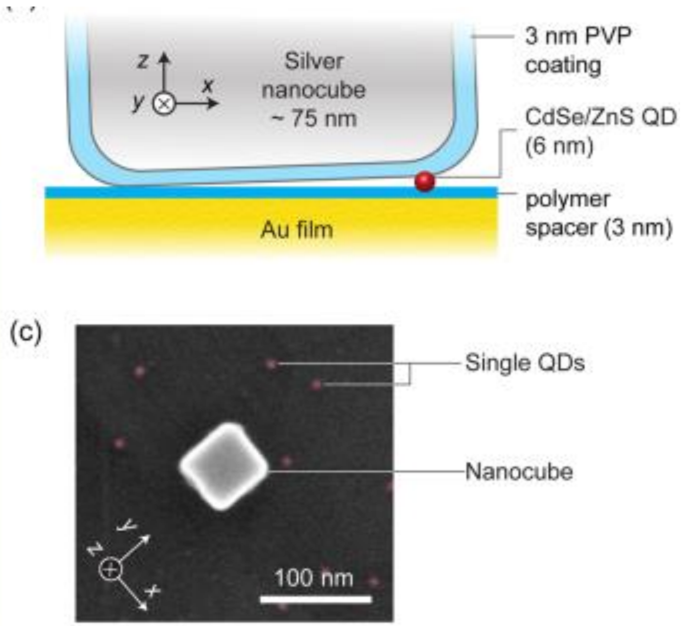
Actual calculation – Au sphere in a pump focus

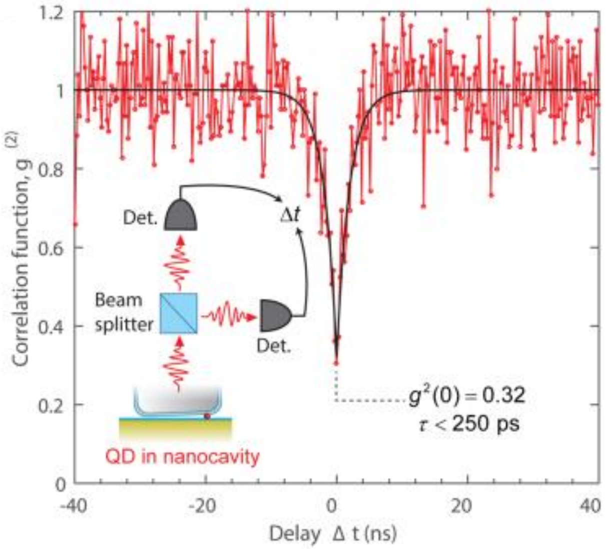


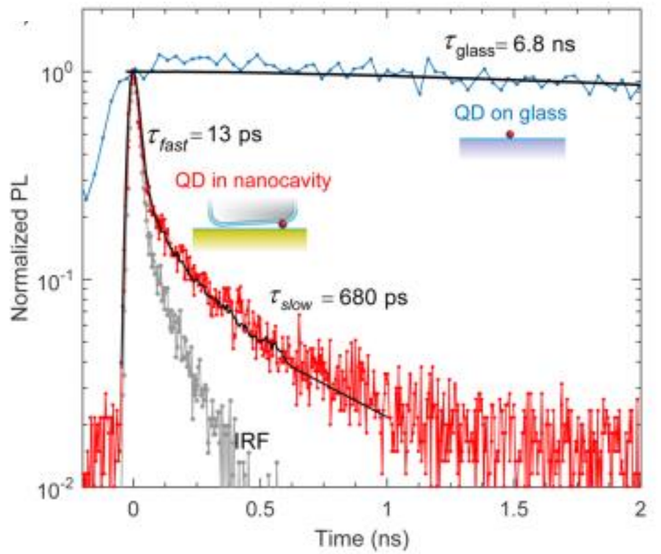
$$I(\mathbf{r}, \omega_{\mathrm{pump}}, \omega_{\mathrm{em}}) \propto P_{\mathrm{pump}}(\mathbf{r}, \omega_{\mathrm{pump}}) \cdot \varphi(\mathbf{r}, \omega_{\mathrm{em}}) \cdot C_{\mathrm{NA}}(\mathbf{r}, \omega_{\mathrm{em}})$$

Record reported antenna

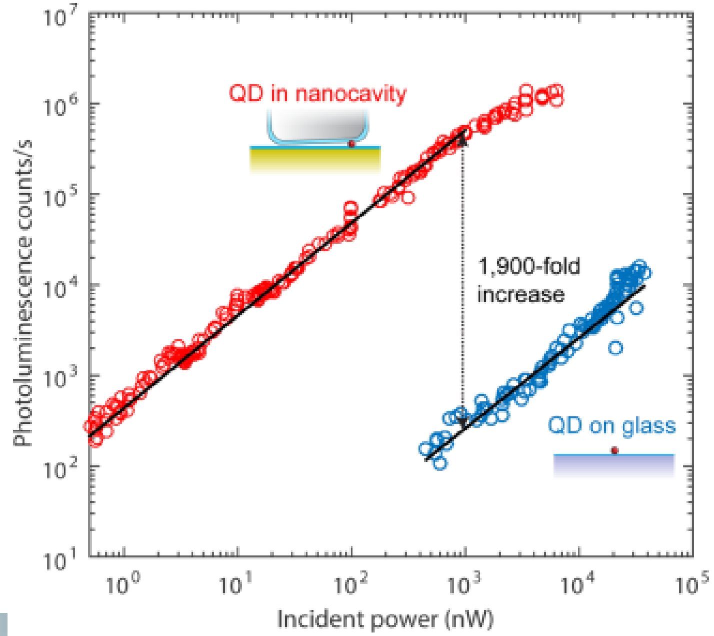








Hoang, Akselrod & Mikkelsen



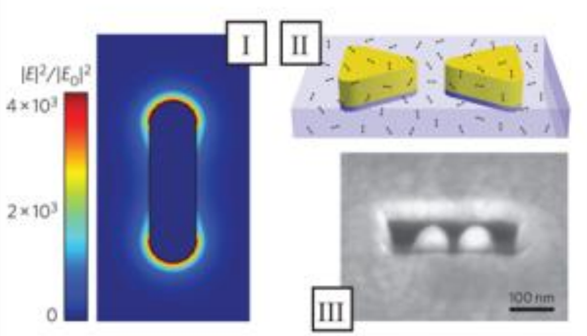
Ag nanopatch on template-stripped Au, CdSe/CdS quantum dots

- Lifetime suggests $F_p > 540$ – quantum efficiency presumed 20%
- 1900x overal brightness increase in cw excitation

Factorized as 225 [pump] x 2 [Q.E.] x 4 [collection]

Next steps

(a) Dipole resonators



Orrit, Moerner, Wenger

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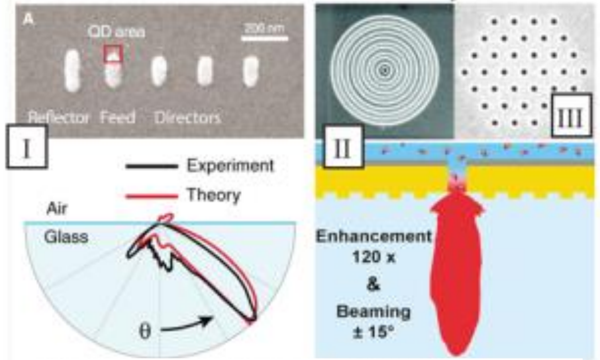

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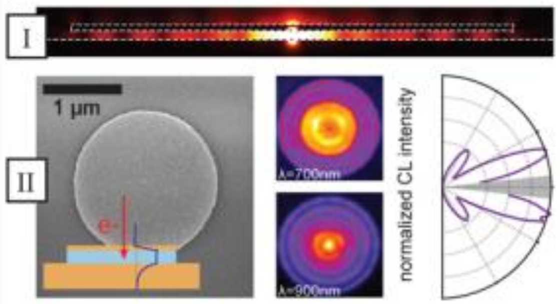

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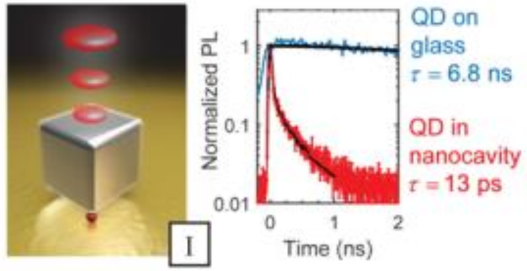

Mikkelsen, Baumberg

Better emission control
Quenching & efficiency control
Integration with optical circuits
Q-factor control & indistinguishable photons
Cooperative emission & lasing

Beyond emission control
Molecular optomechanics & SERS
Quantum optics with single molecules
Breaking selection rules
Extreme-gap nanophotonics

Nature Materials (2019), DOI: 10.1038/s41563-019-0290-y