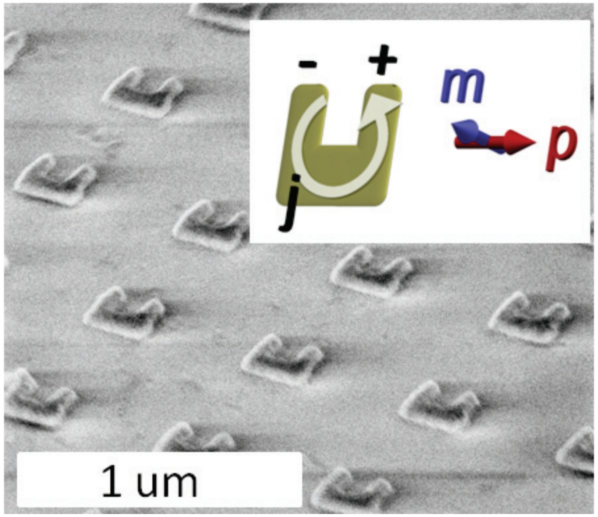
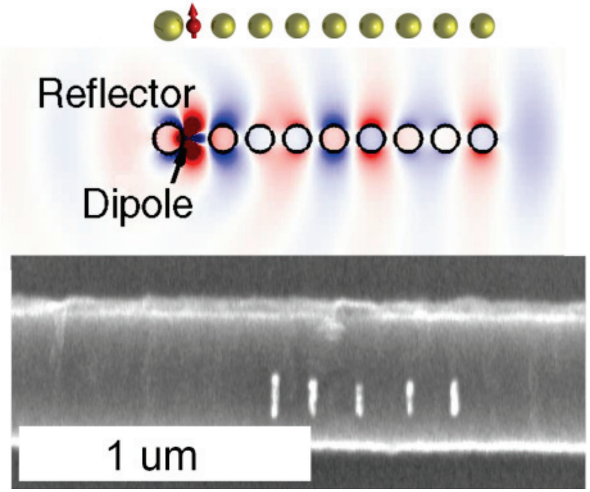
Controlling single photons and single molecules with nano-antennas

Ivana Sersic
Martin Frimmer
Andrej Kwadrin
Abbas Mohtashami
Felipe Bernal
Lutz Langguth
Hinke Schokker
Per Lunnemann

Femius Koenderink

Center for Nanophotonics
FOM Institute AMOLF, Amsterdam
www.amolf.nl





FOM Institute
AMOLF

Single photons from single emitters

photon

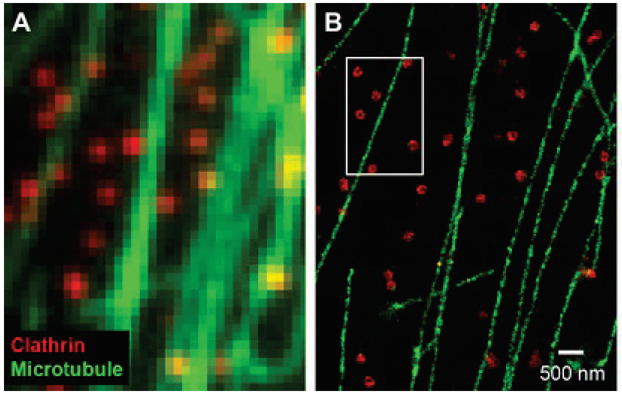
photon

Single emitter
Quantum dot, molecule

Challenges: (1) Surely catching each photon in a single beam
(2) Surely absorbing each photon from a beam

Addressing and seeing single molecules *with unit efficiency*

Motivation



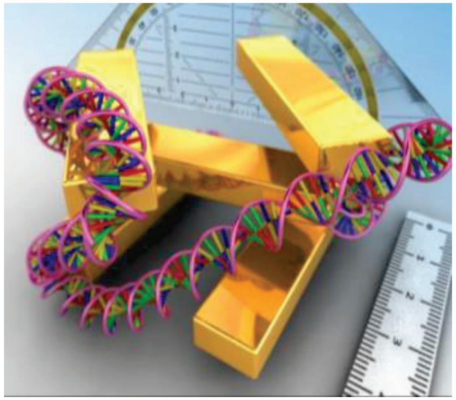
Bates & Zhuang [PALM, STORM]

Optical microscopy
Below λ/2 limit

Single molecules
Information from
fluctuations

Single photon sources
Quantum information
Quantum communication

Quantum information
in 1 photon can
not be eavesdropped



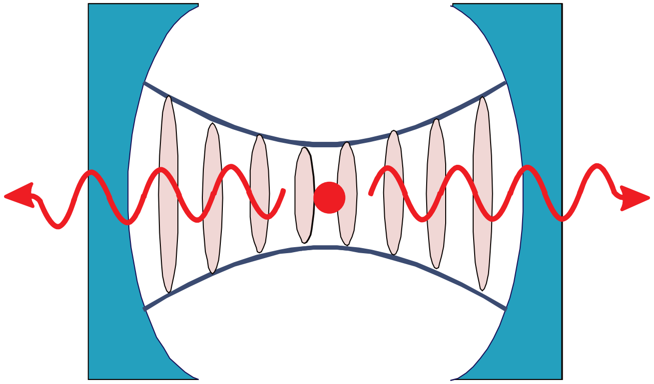
Liu & Alivisatos

Spectroscopy of
molecules

Distance ruler,
vibrations
THz, IR and VIS

Enhancing photon-emitter interaction

Cavity resonances



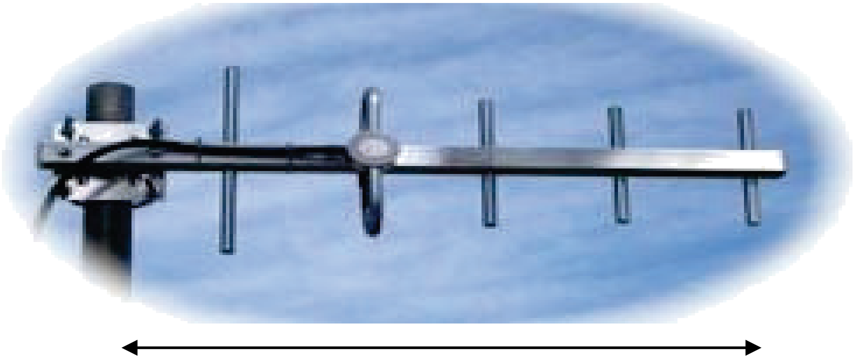
Enhanced interaction time $\propto$ Q

Enhanced $|E|^2$ per photon $\propto 1/V$

Limit on V $> (\lambda/2)^3$

Hence Q $> 10^4$

Antennas



$\lambda \approx 1$ *meter*

Very broadband: Q $\approx$ 5

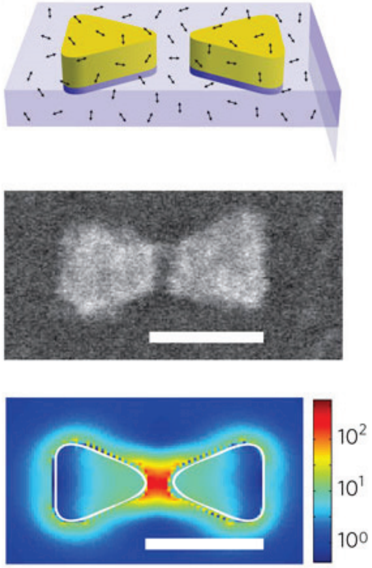
Strongly scattering, open system

Strong local field due to metal

$V \approx (\lambda/50)^3$

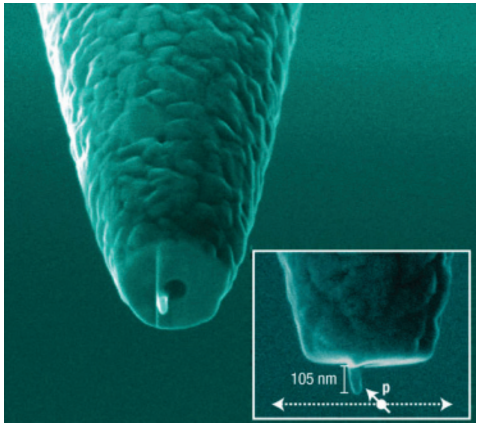
Famous reported optical antennas

'Bow-tie' antenna



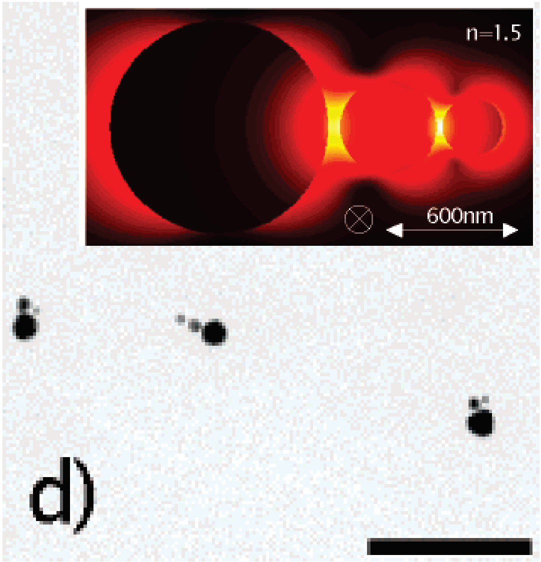
W. E. Moerner 2009
Nature Photonics

'λ/4' antenna

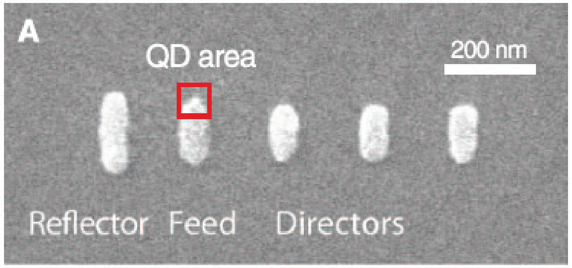


Van Hulst 2008
Nature Photonics

Self-similar trimer



Bidault & Polman
2009, JACS



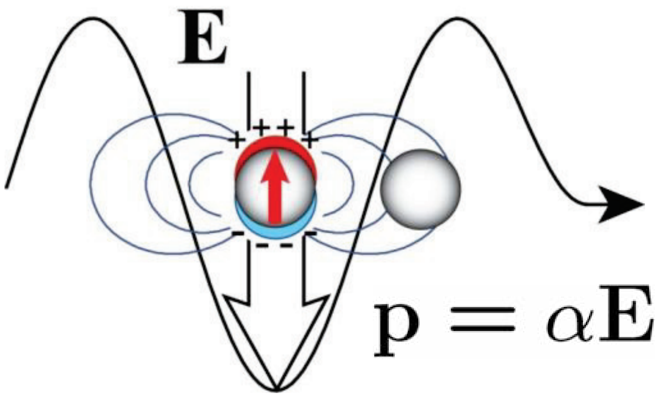
Yagi-Uda antenna

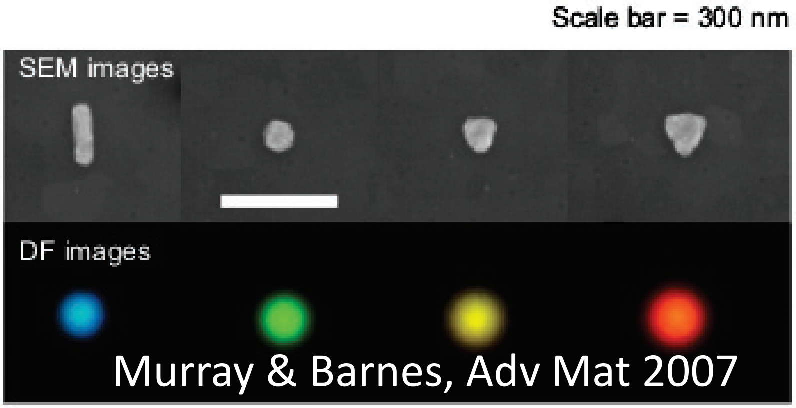
Van Hulst/Quidant 2010
Nature Photonics

- E-beam lithography
- Focused Ion Beam milling
- DNA-aided selfassembly

Scattering resonance

Plasmon resonance





Murray & Barnes, Adv Mat 2007

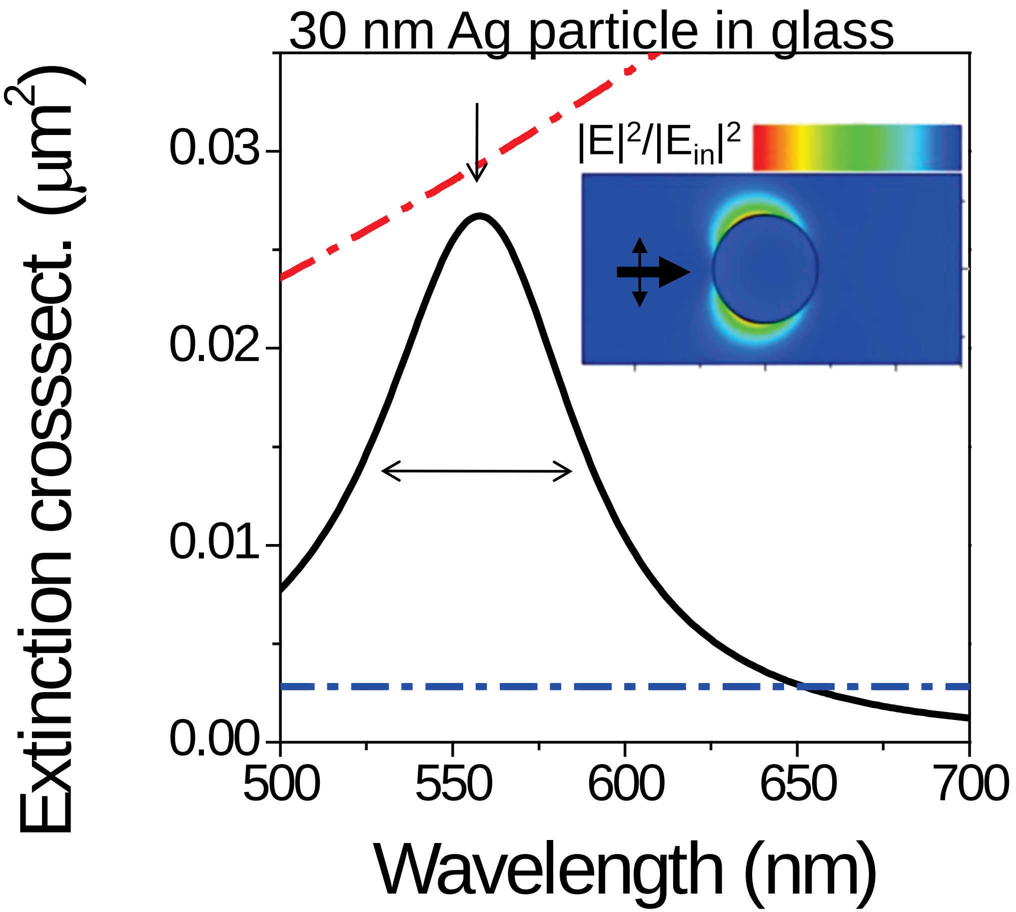
Circa 10^3-10^4 free electrons

Incident field separates e^- from ionic backbone

Linear restoring force implies a resonance

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

General properties



- Color tunable in visible
- Cross section ~ 10x πr^2
- Strong dipolar near field
- Q ~5 means 95% of loss is radiation into free space
- σ and α at upper bound:

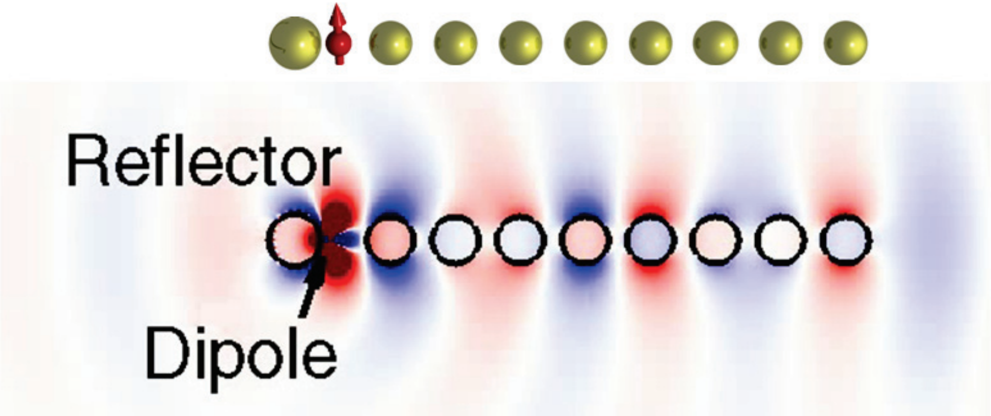
unitary limit (Chu limit) $\sigma \approx \frac{3}{2\pi}\lambda^2$

Plasmon particle is a solid state `strongest point-scatterer'

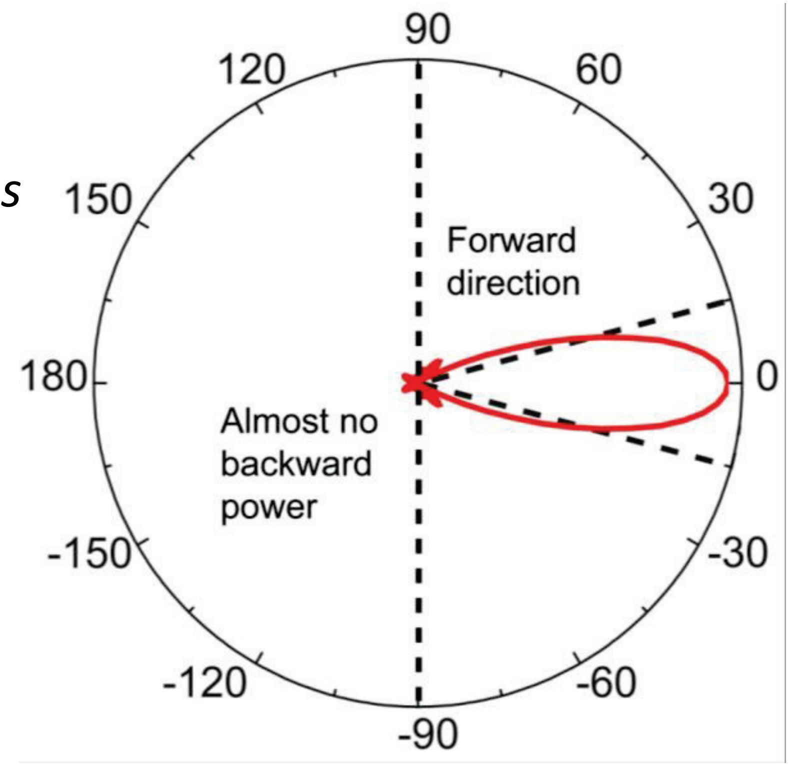
Yagi-Uda

Quantum emitter
λ = 650 nm

100 nm silver nanospheres



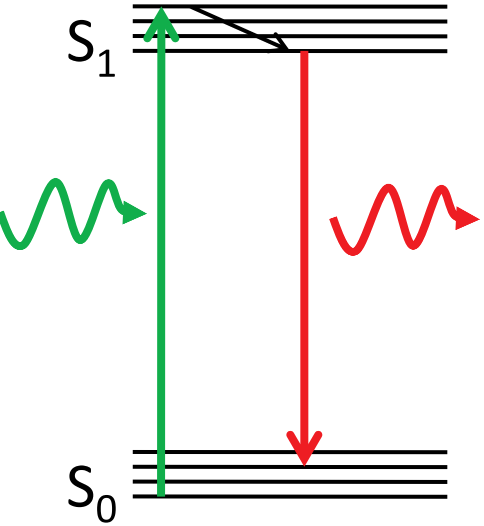
Snap shot of electric field

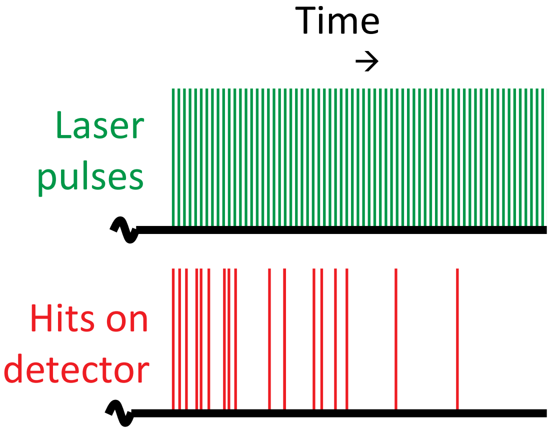


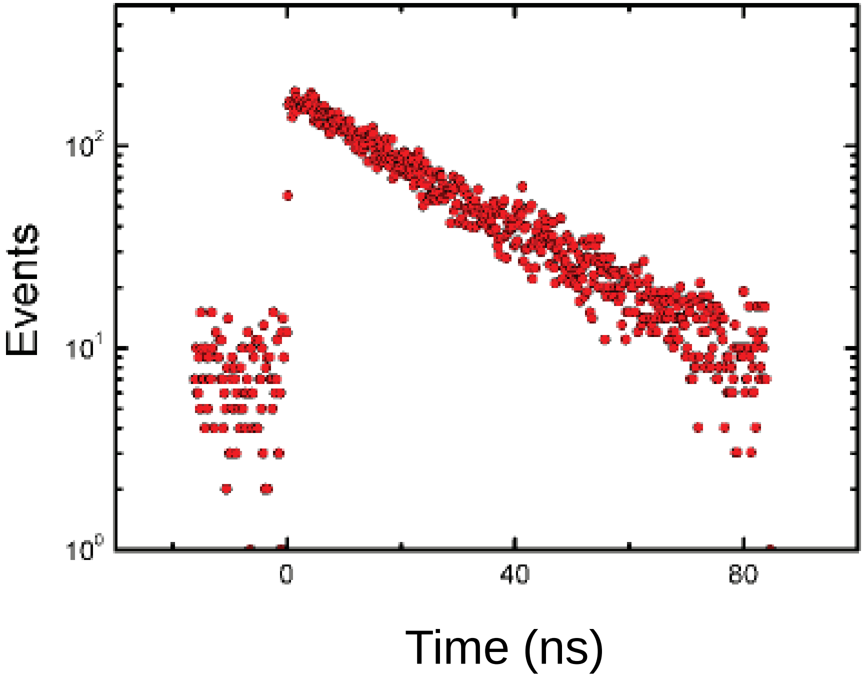
- Far-field: photon with 90% probability in a narrow beam
- Broadband (> 300 nm bandwidth in the visible)
- 90% chance that the photon is not lost to heat

de Waele Nano Lett 2007 / Koenderink Nano Lett 2009 / Curto Science 2010, Coenen Nano Lett. 2011

Single quantum emitter







- After one excitation, emits just one quantum of light
- Probabilistic timing of *when* emission occurs
- Spatial and temporal coherence of single photon wavepackets
- No such thing as setting up multiple *coherent* active elements

Fermi’s Golden Rule

1) Dipole antenna
2) Ground plane

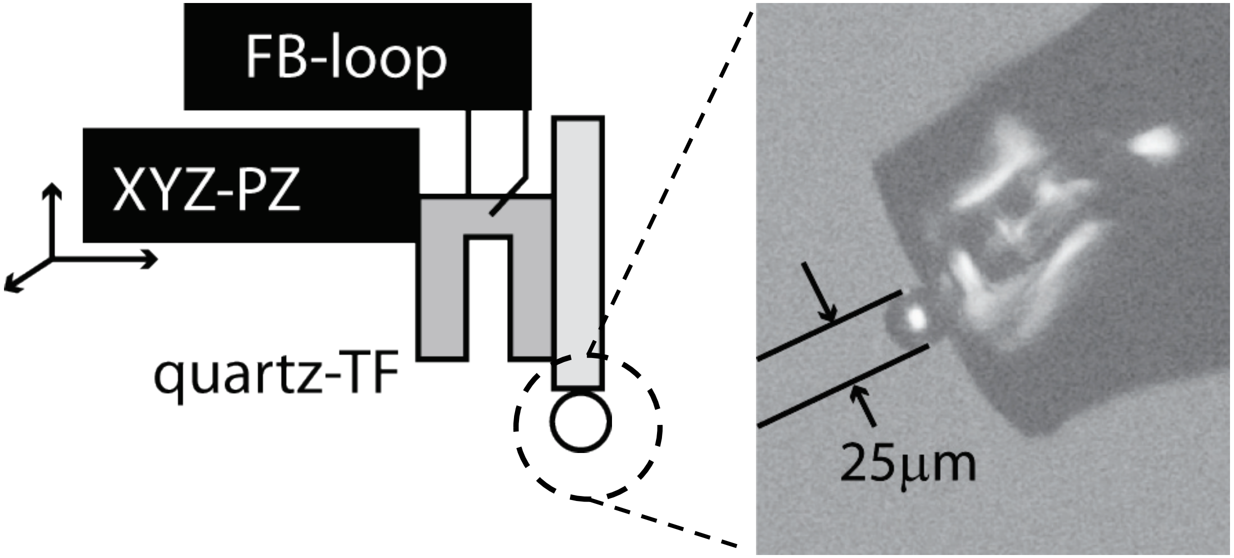
Optics:

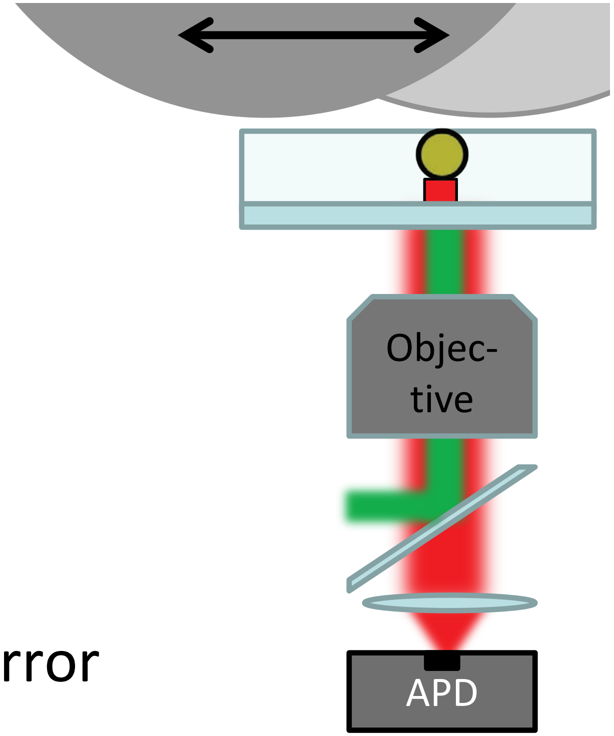
“Drexhage experiment”

Emitter in front of mirror

(Drexhage, 1968)

Scanning mirror ‘Drexhage experiment’

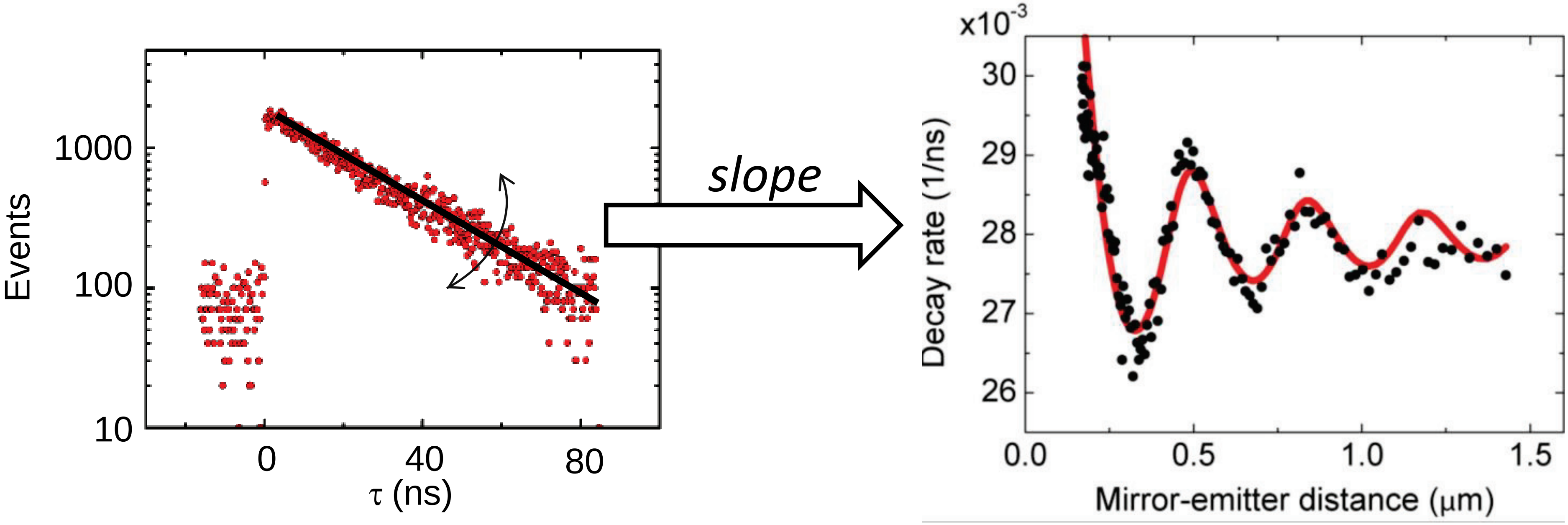




- 25µm PS bead covered with 400nm Ag as mirror
- PS bead glued to cleaved fiber
- Lateral scanning in shear force varies emitter-mirror distance

Method pioneered by B.C. Buchler et al. Phys. Rev. Lett. **95,** 063003 (2005)

Calibration example – single NV center



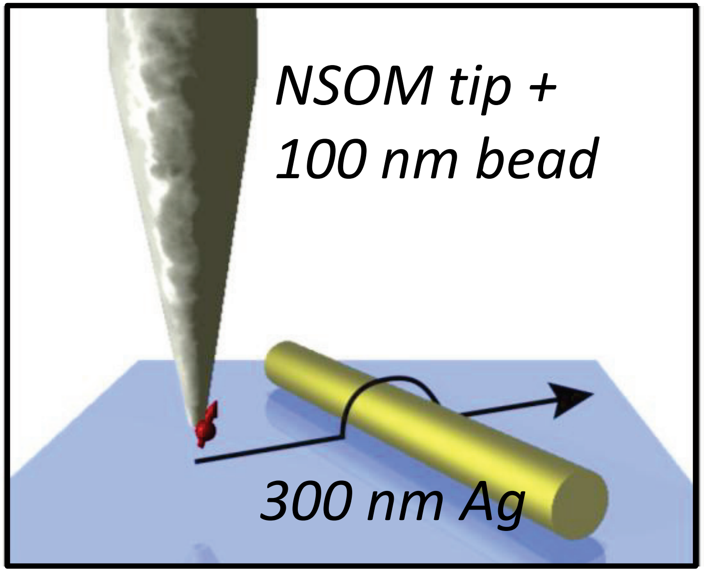
- *Single* NV center in a 100 nm nanodiamond (MicroDiamant AG)
- Decay rate varies with distance to the mirror

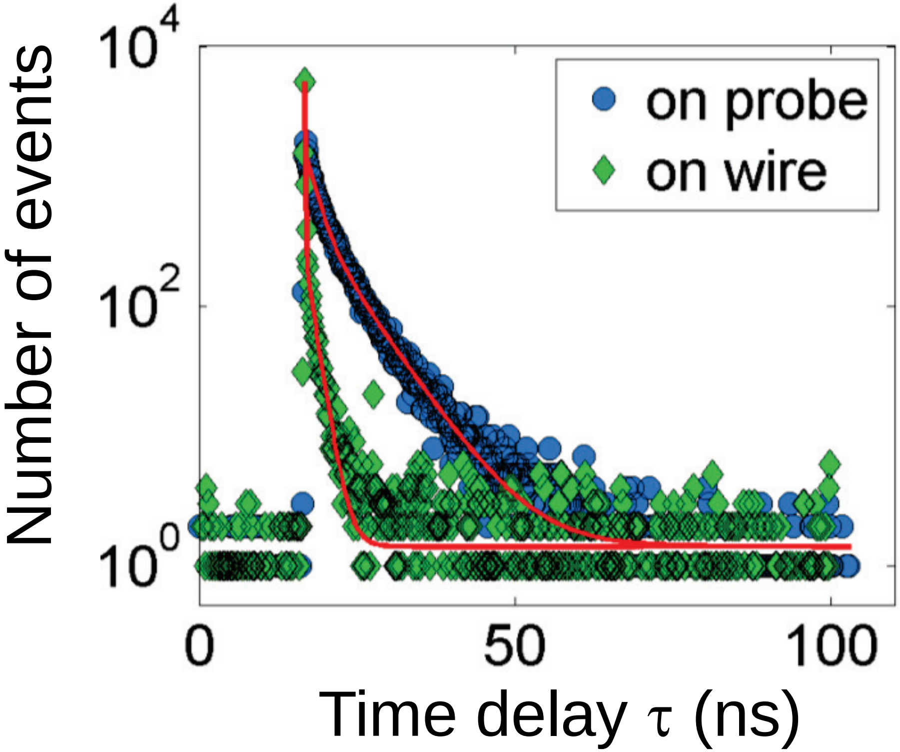
radiative decay rate ⇔ radiative impedance

nonradiative decay rate ⇔ Ohmic resistance

A. Mohtashami & M. Frimmer APL, New J Phys (2013)

Scanning LDOS microscope



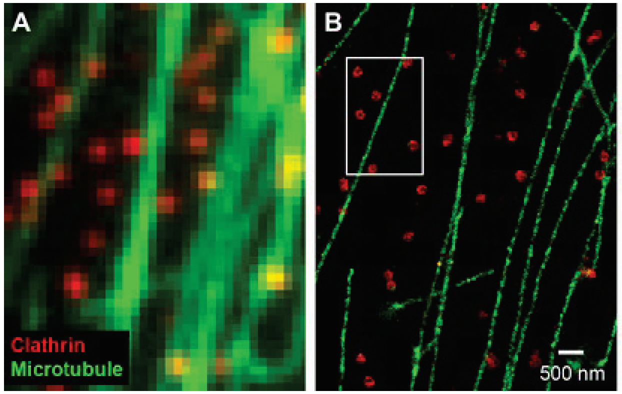


Reversible control of light-matter coupling for *any* nano-structure

Nanoscale imaging LDOS changes in Fermi's Golden Rule

Frimmer et al., Phys. Rev. Lett. **107** 123602 (2011)

Motivation



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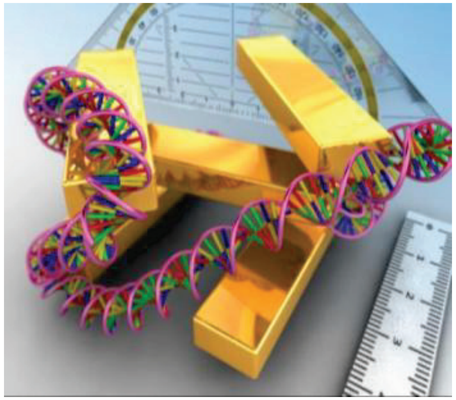

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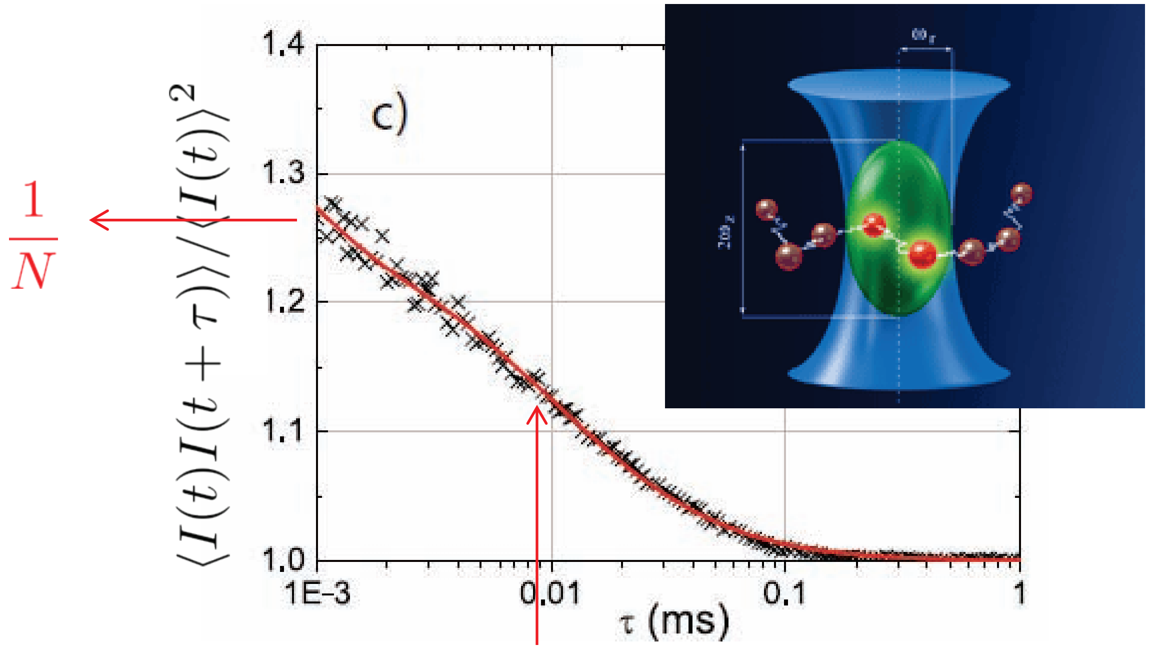
Microscopy & antennas

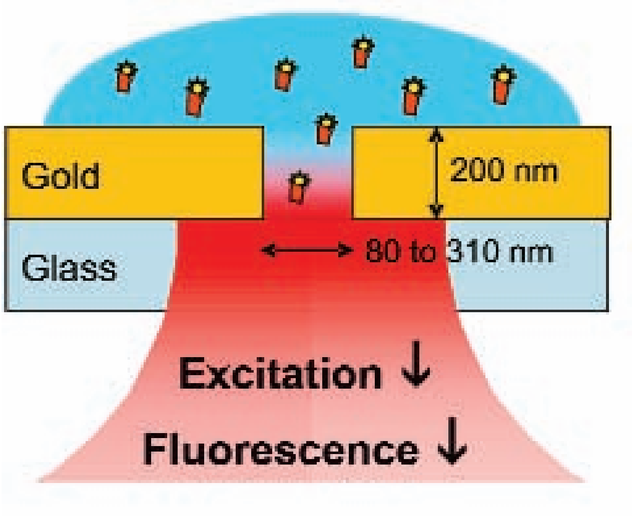


Transit time through focus

“Fluorescence correlation spectroscopy”
Photon correlations reveal density and diffusion constant of an analyte

Problems: requires < *1 molecule per focal volume*

Microscopy & antennas



A single nano-aperture confines geometrically for FCS

Plasmonic enhancements increase count rates and brightness

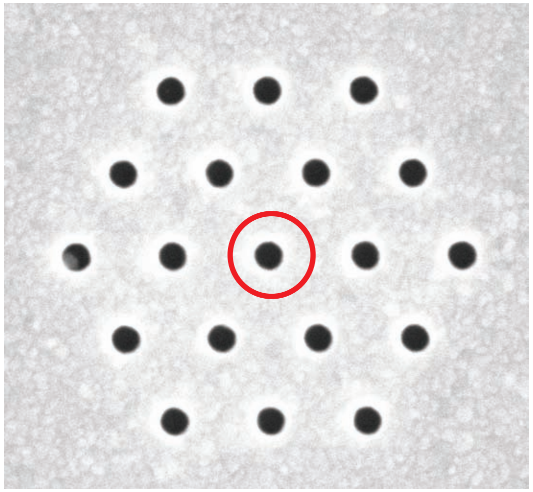
Pioneered by
Levene et al. [2003]
Wenger et al. [2005]

“Flu

Pho nalyte

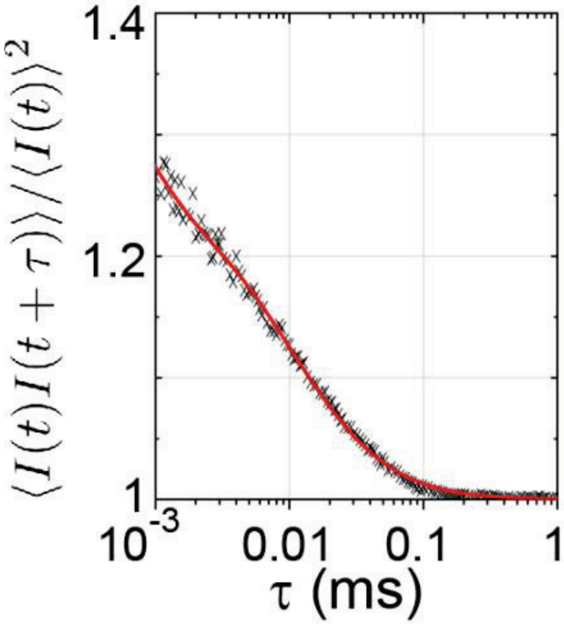
Problems: requires *< 1 molecule per focal volume*

Phased array



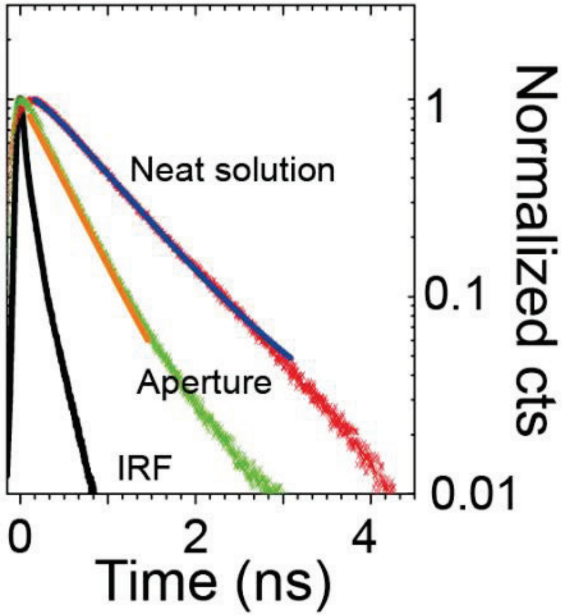
140 nm holes/150 nm Au film/440 nm pitch

We only pump molecules in the central hole



FCS correlation 1.3
3 molecules/focus

$V \sim \lambda^3/20$

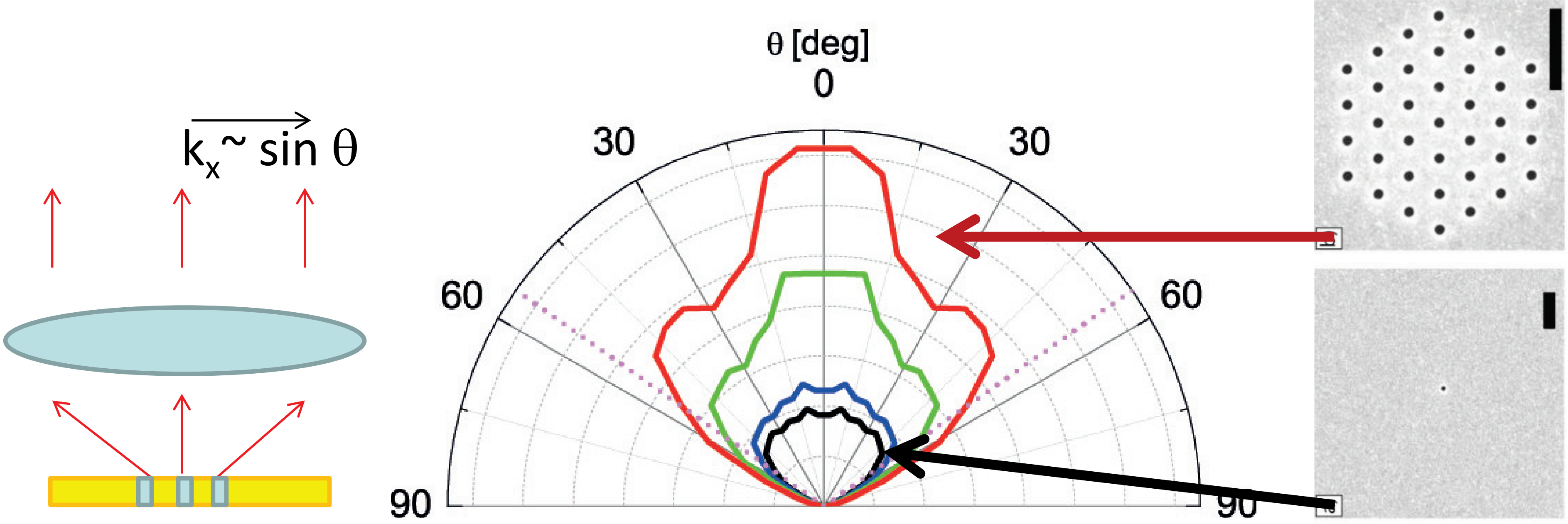


Fluorescence lifetime

Circa 2-fold enhanced

Collaboration with Institut Fresnel, Jérôme Wenger - Alexa 647 dye , 1 uM in water

Fourier images – angular distribution



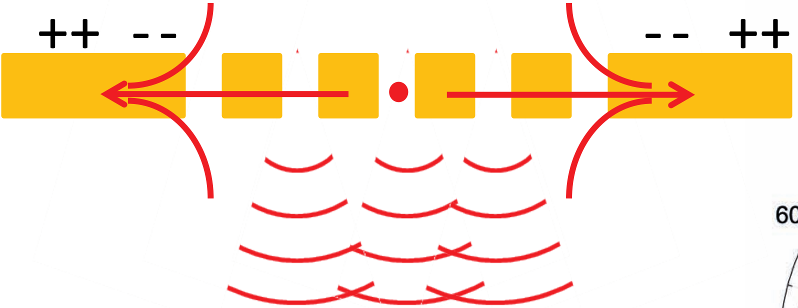
Emission *only* comes from the central hole

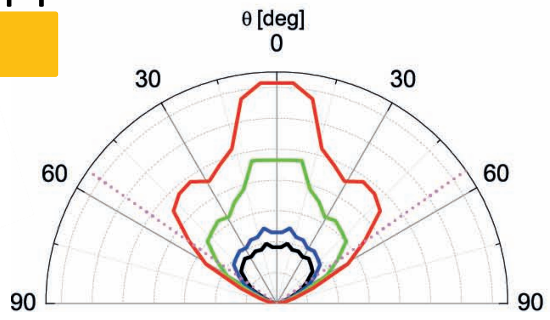
Yet, enhanced directivity and total strength (4 to 5 times)

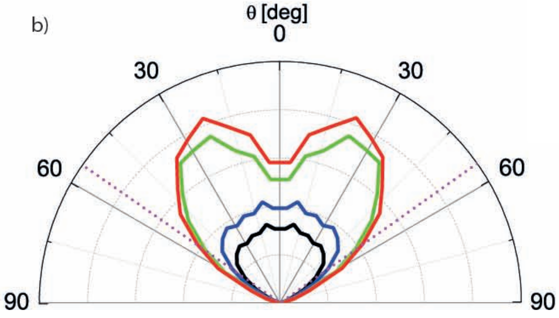
Plasmonic-crystal band edge phased array antenna

Langguth, Punj, Wenger, Koenderink, submitted (2013)

Phased array physics



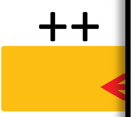

Hexagonal lattices

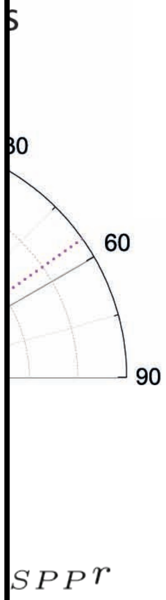

Square lattices

The gold film supports a surface plasmon guided moded [k_{SPP}]
Direct photon + amplitude scattered from each hole with phase $e^{ik_{SPP}r}$

Radiation pattern can be controlled via lattice parameter
Around 2nd order plasmon diffraction (near dispersion band edge)

Phased array physics





Plasmon phased arrays improve single molecule brightness and directivity, as well as emission rate

Also:

- Yagi Uda + single quantum dot [Curto et al., Science 2010]
- 1D chain antenna receiver [de Waele et al., Nano Lett. 2007]
- 2D infinite lattices to improve LED extraction [G. Vecchi et al., PRL 2007, LED's]
- Bull's eyes around holes [Wenger et al., Nano Lett. 2012, FCS]

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Dire

Radi
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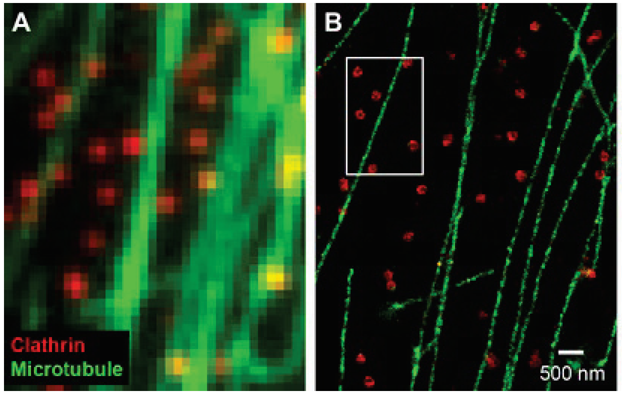
Motivation



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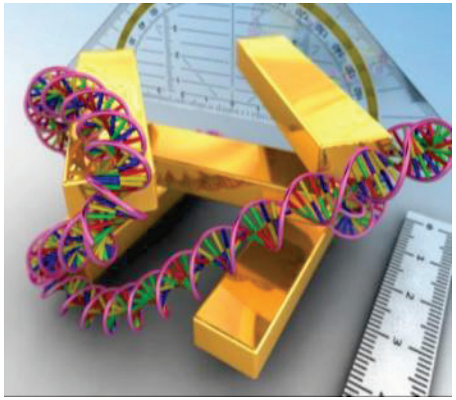

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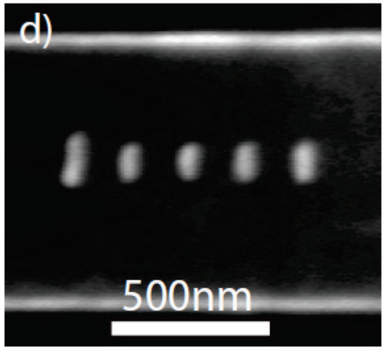
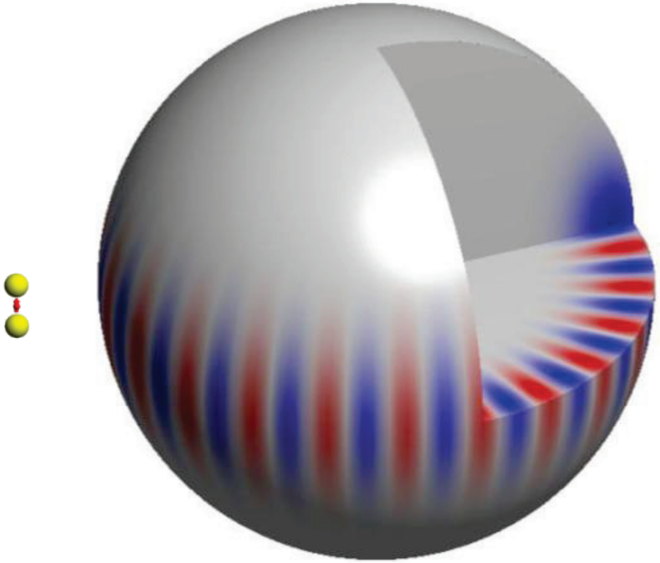


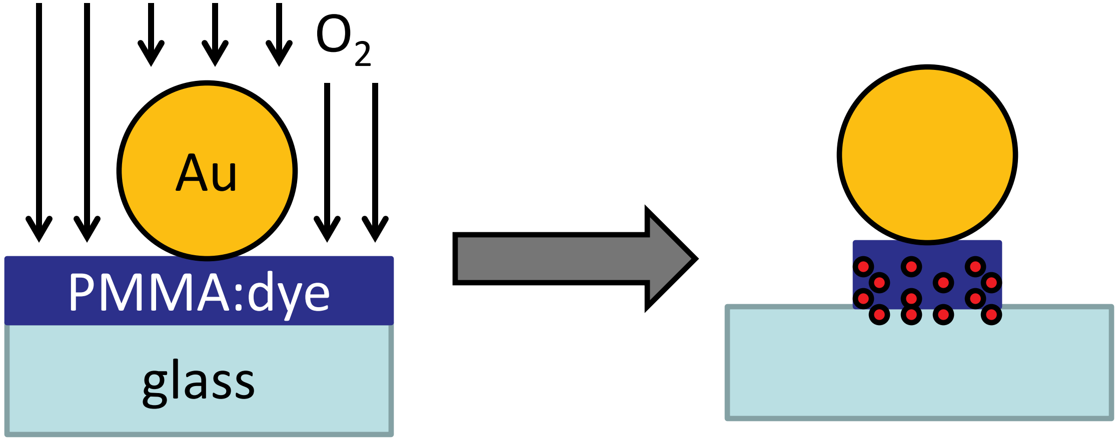


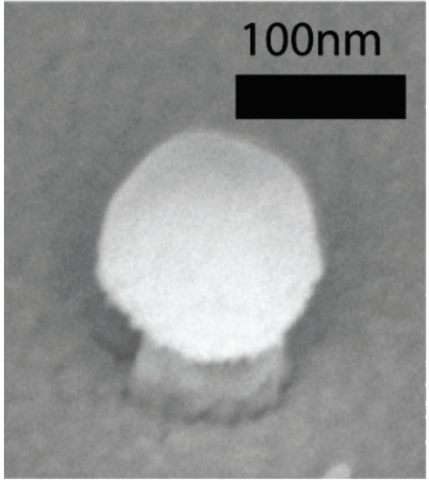
Yagi – Uda *ON* a waveguide
Full scattering study:
Bernal, ACS Nano **6** 10156 (2012)

Cavity-assisted plasmonics

Fundamental question: antenna in a nontrivial mode bath ?
Key parameter: decay rate enhancement (Purcell factor)

Simplest possible “dipole”antenna

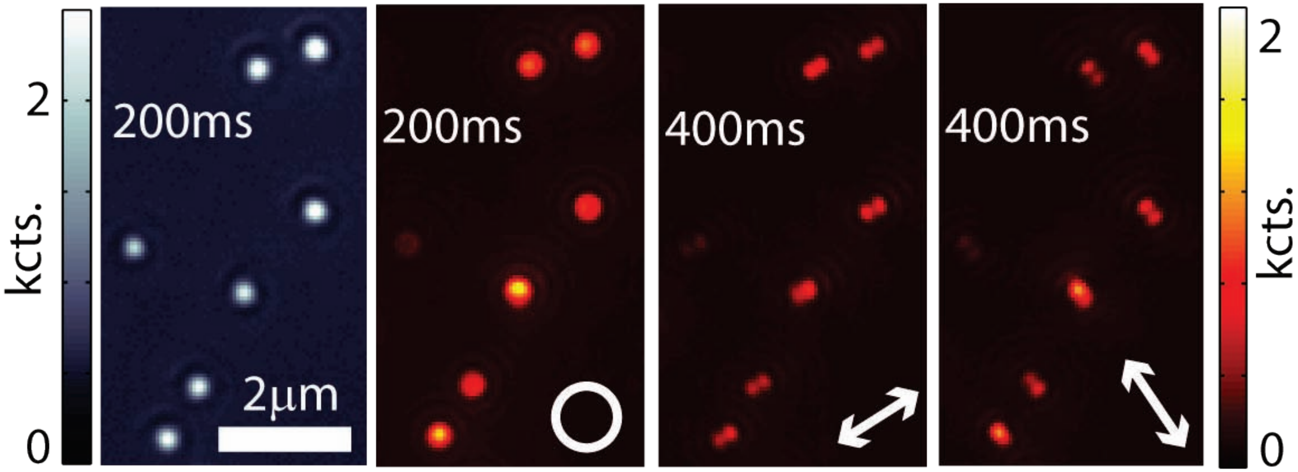




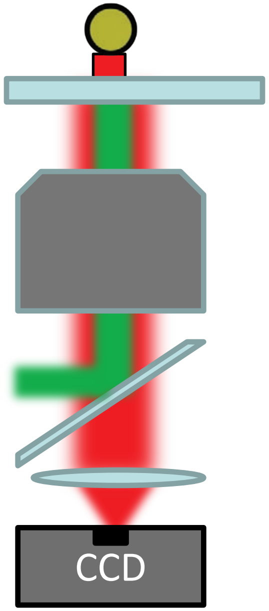
- Au colloid on dye doped PMMA layer on glass
- O_2 plasma removes dyes layer except under colloid

Similar method to: Sorger et al., Nano Lett. **11**, 4907 (2011)

Superemitter - polarization



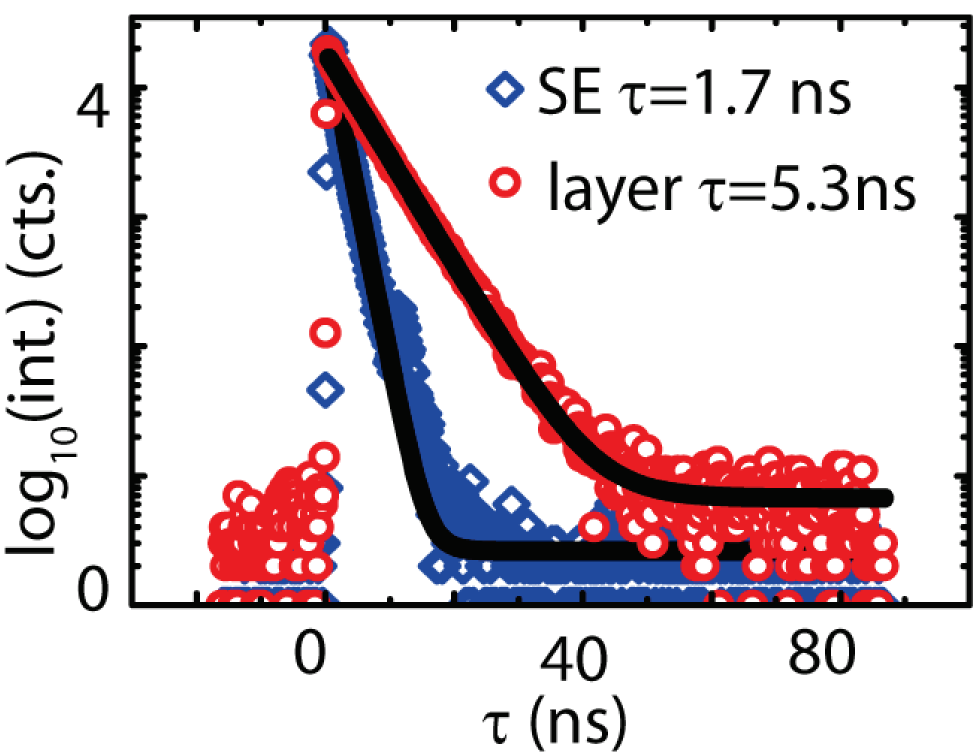
scattering fluorescence

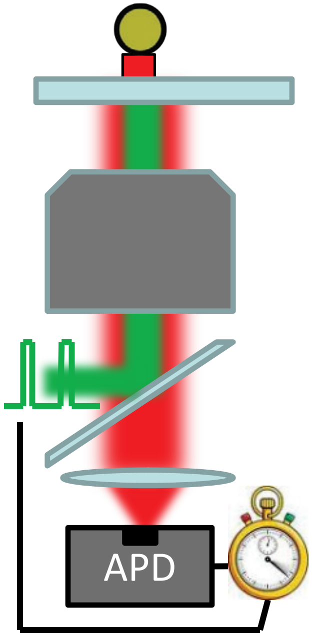


- Unpolarized: donut-shaped image
- Polarization analyzed: double lobes

→ Superemitter dipole moment along optical axis

Superemitter - lifetime

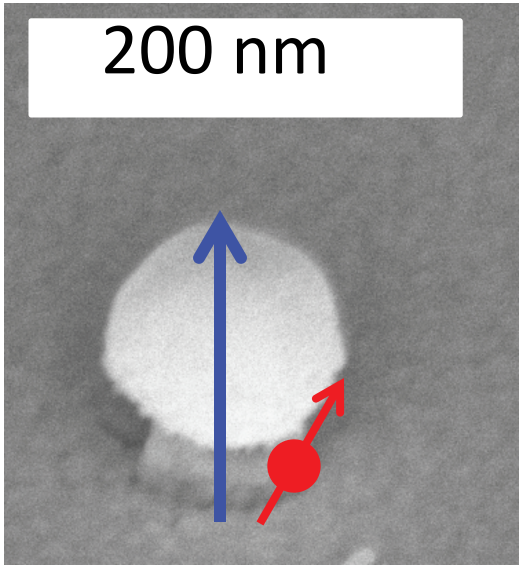




- TCSPC-FLIM measurement (pump 532 nm, 10 MHz)

- Antenna rate enhancement ca. 3x compared to bare dye layer

Zero order idea



$$\mathbf{P} = \mathbf{p}_0 [1 + \frac{\alpha(\omega)}{r^3}]$$

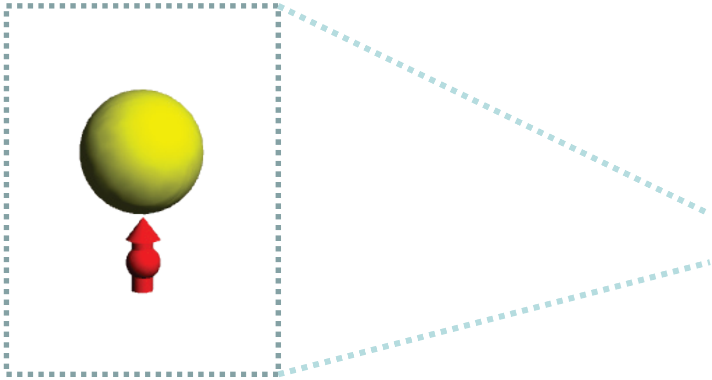
Antenna dominates molecular dipole

Classically, the radiated power of a dipole scales with $|p|^2$

Decay rate change = $1 / Z_{\text{antenna}} \sim \left|\alpha(\omega)/r^3\right|^2$

Hybrid systems – lumping LDOS

System 1: Super-emitter



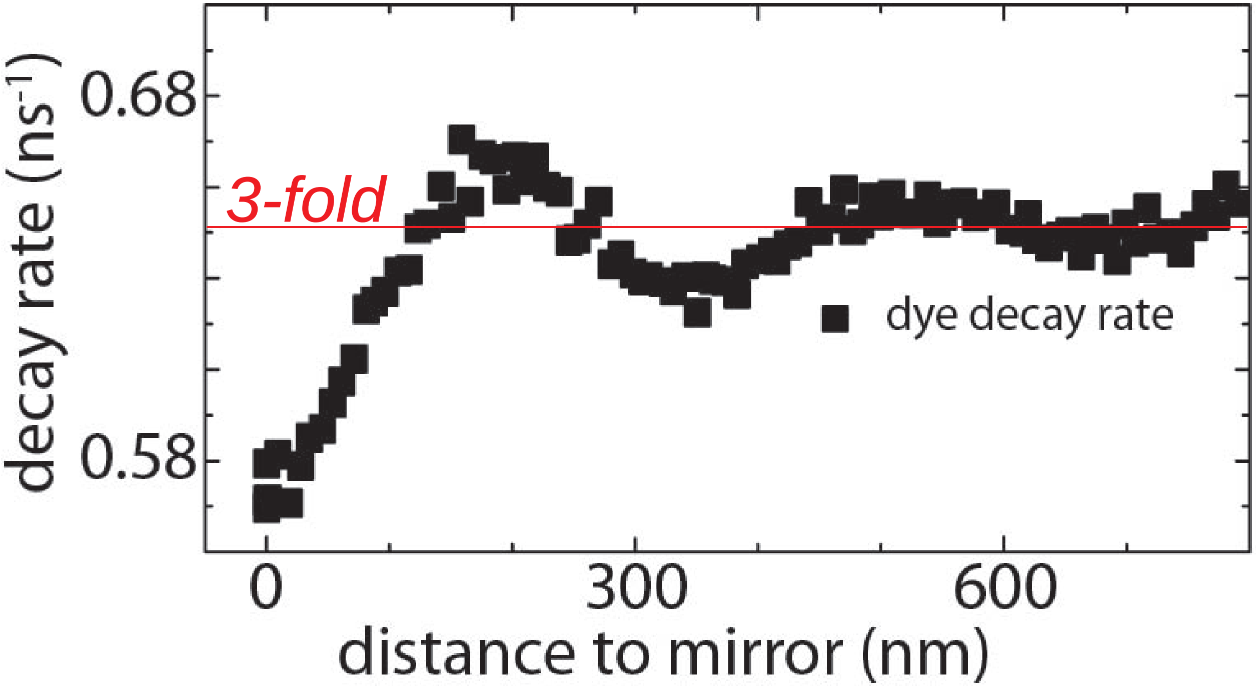
Polarizable nanoparticle
in near-field of molecule

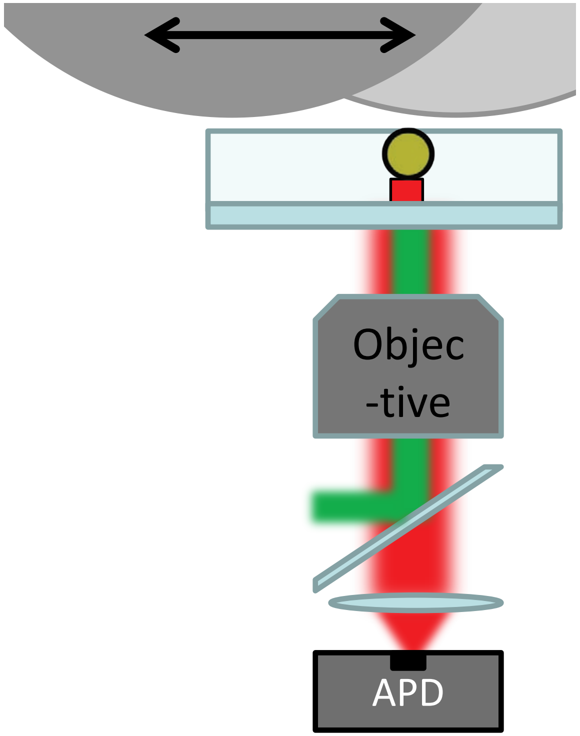
System 2: ground plane

What happens to the joint radiative impedance, given that I know the radiative impedance change offered by antenna and ground plane ?

Greffet et al. Phys. Rev. Lett. 105, 117701 (2010) - LDOS as impedance
Benson, Nature 480, 193 (2011)

Scanning mirror 'Drexhage experiment'



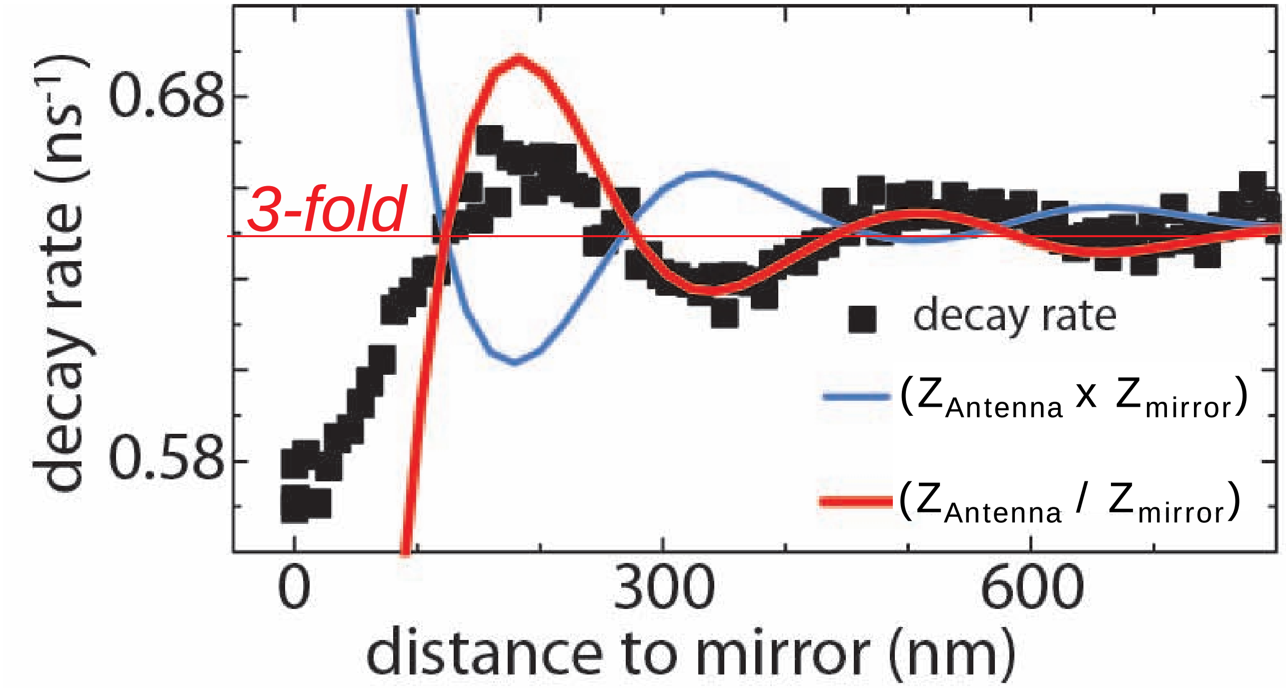


Decay rate at superemitter shows characteristic variations

Mirror LDOS modifies the already antenna-accelerated decay

Experiment: Frimmer, arxiv: 1212.6396 (under review Phys Rev)

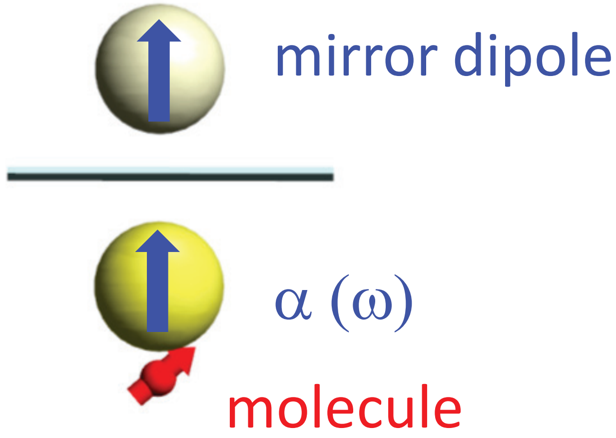
Scanning mirror ‘Drexhage experiment’

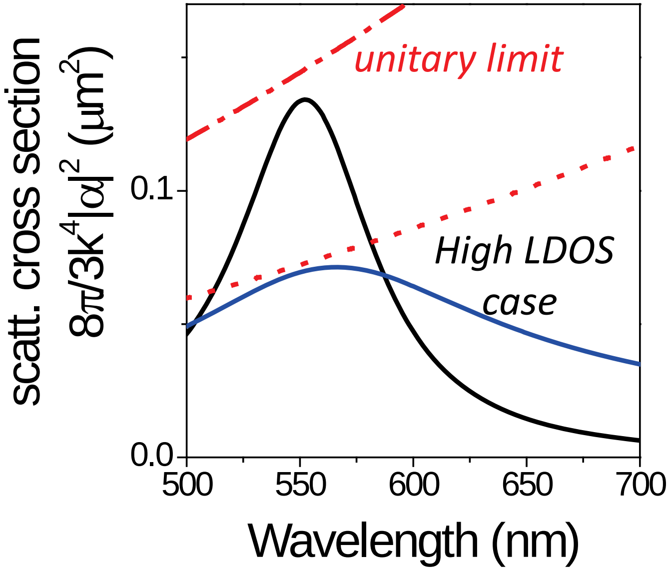


Lumped system Purcell factor is clearly not simply a multiplication

Inverse effect confirmed by *full* dyadic Green function calculation.

How an antenna gets spoiled



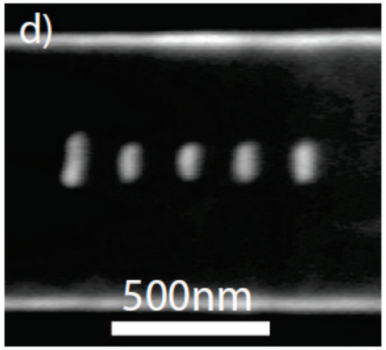


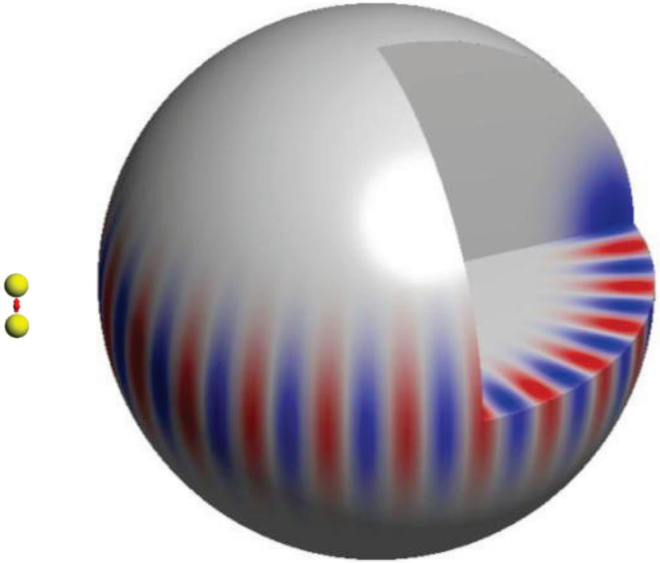
Fixed current sources: twice the dipole moment radiates twice as much
quantum emiter: doubled decay rate

Scatterer: at *twice* the radiative loss, the scatterer ***p*** is much weaker
polarizability is *spoiled* / Chu limit changed by the mirror

Theory: Frimmer PRB **86** 235428:1-6 (2012).

Complex world





Yagi – Uda *ON* a waveguide

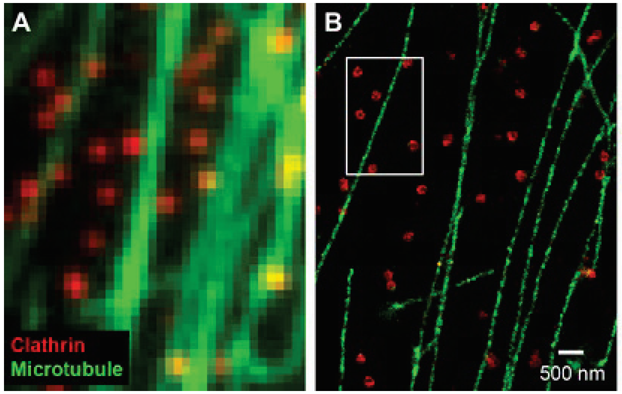
Cavity-assisted plasmonics

Current effort: *practical* Q, i.e., Q = 100, structures of antenna + cavity

Separate regimes where cavities aid or spoil antennas

Radiative impedance lumping in complex systems

Motivation



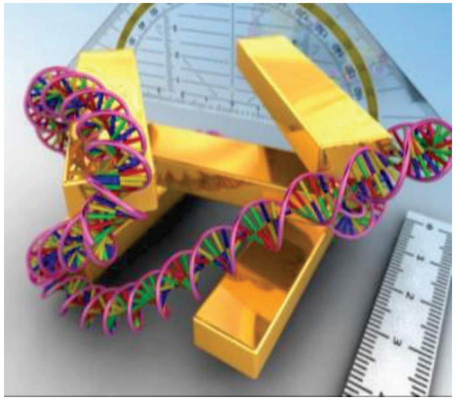
Bates & Zhuang [PALM, STORM]

Optical microscopy
Below $\lambda/2$ limit

Single molecules
Information from
fluctuations

Single photon sources
Quantum information
Quantum communication

Quantum information
in 1 photon can
not be eavesdropped

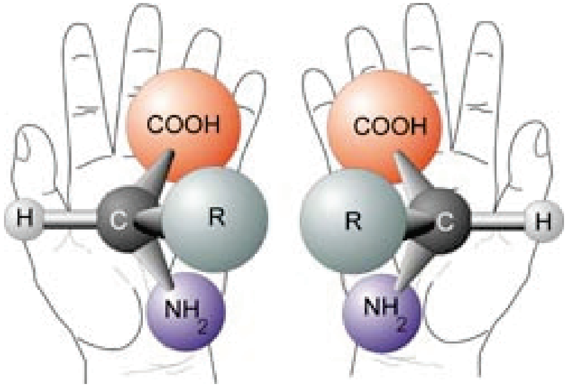


Liu & Alivisatos

Spectroscopy of
molecules

Distance ruler
Vibrations
THz, IR and VIS

Handedness



$$\begin{pmatrix} \mathbf{p} \\ \mathbf{m} \end{pmatrix} = \begin{pmatrix} \alpha_E & i\alpha_{EH} \\ -i\alpha_{HE} & \alpha_H \end{pmatrix} \begin{pmatrix} \mathbf{E} \\ \mathbf{H} \end{pmatrix}$$

$\alpha_E \sim 10^3 \alpha_{EH} \sim 10^6 \alpha_H$

Almost *only* electrically polarizable
Very weak magnetic moment, induced by electric driving

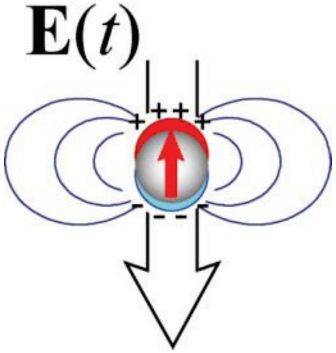
Perturbative effect: optical activity, circular dichroism, optical rotation

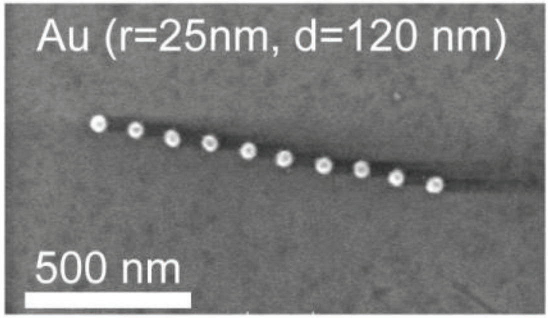
Ubiquitous & biochemically of huge impact - optically weak

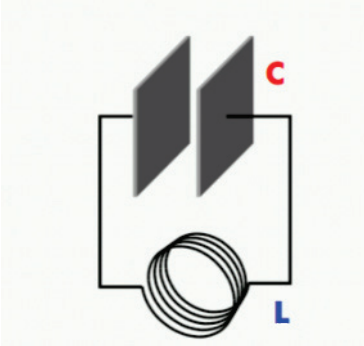
Resonant nano-scatterers

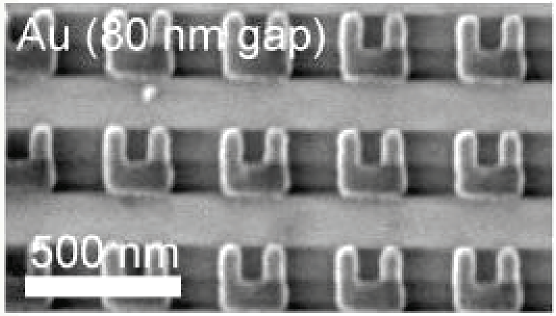
Resonant electric dipoles

Resonant magnetic dipoles









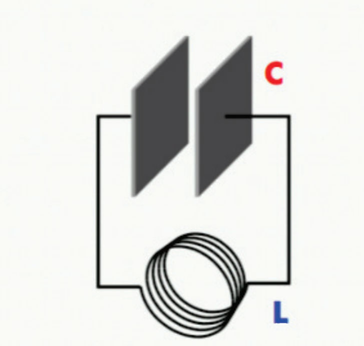
Charge separation: electric dipole moment
Current loop: magnetic dipole moment

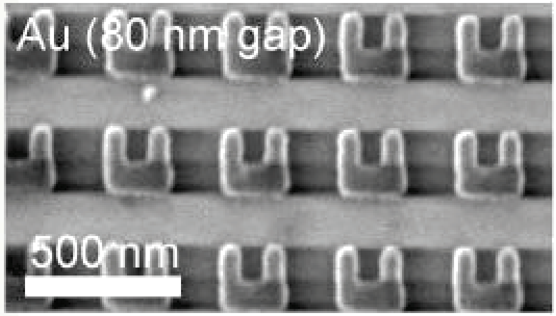
Noble metal U-shaped particles: LC resonators `split rings’
Pendry & Smith, Linden & Wegener, Giessen

Electron beam lithography down to 20 nm with <5 nm error

Resonant nano-scatterers

$$\begin{pmatrix}\mathbf{p}\\ \mathbf{m}\end{pmatrix}=\left(\begin{array}{c|c}\alpha_E & i\alpha_{EH}\\ \hline -i\alpha_{HE} & \alpha_H\end{array}\right)\begin{pmatrix}\mathbf{E}\\ \mathbf{H}\end{pmatrix}$$





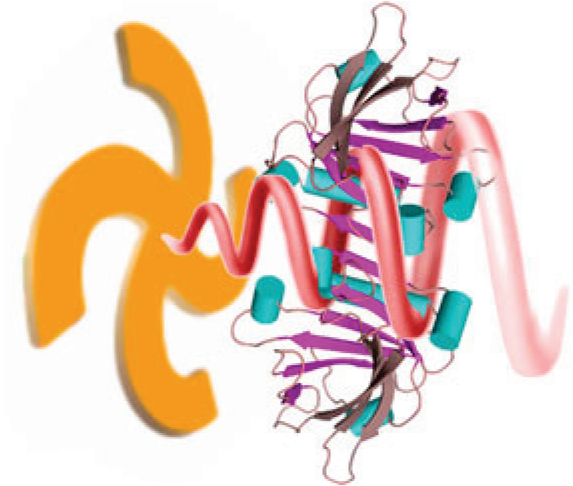
Split ring: E-field charges the split – with a quarter phase lag the split discharges, giving rise to a magnetic dipole

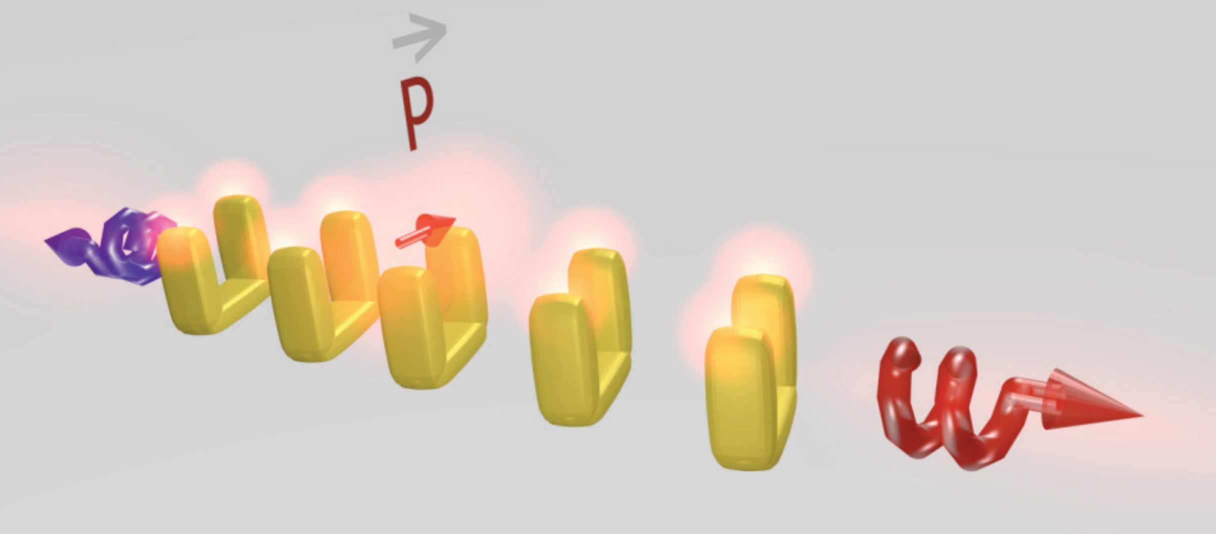
The same polarizability tensor as for chiral molecules, but with

$$\alpha_E \sim \alpha_{EH} \sim \alpha_H \sim V \sim \lambda^3$$

Electron beam lithography down to 20 nm with <5 nm error

Superchiral spectroscopy



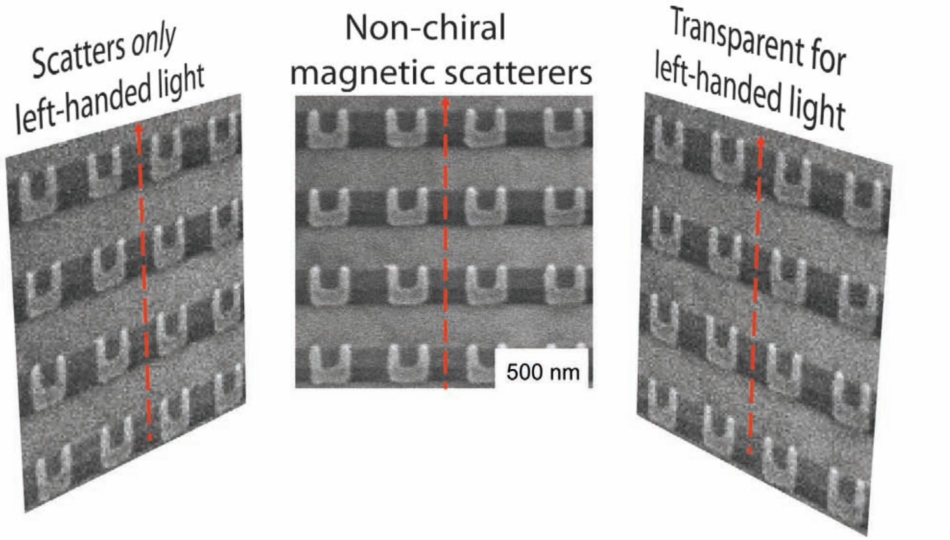


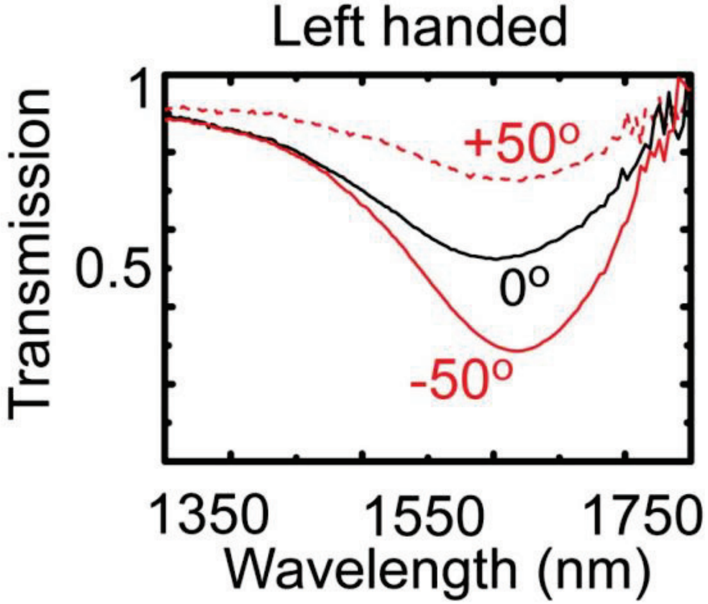
Hendry, Exeter
Tang & Cohen, Harvard
Schaferling & Giessen, Stuttgart

Single molecule enantioselective detection ‘superchiral’fields

Selectively exciting, and spoofing emitters with nontrivial selection rules – spin & orbital angular momentum antennas

Not chiral – pseudo chiral



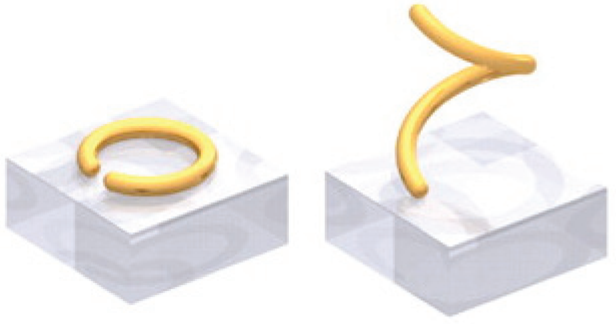


Pseudochirality

Huge optical activity – though split rings are not chiral at all

Sersic, PRL **108** , 223903:1-5 (2012)

How can an LC circuit / SRR be chiral ?



Geometrically a SRR is not 2D or 3D chiral

Obliquely, the SRR looks line one turn of a screw – COULD be optically active

---

Quantitative fit of α to many angle resolved spectra:

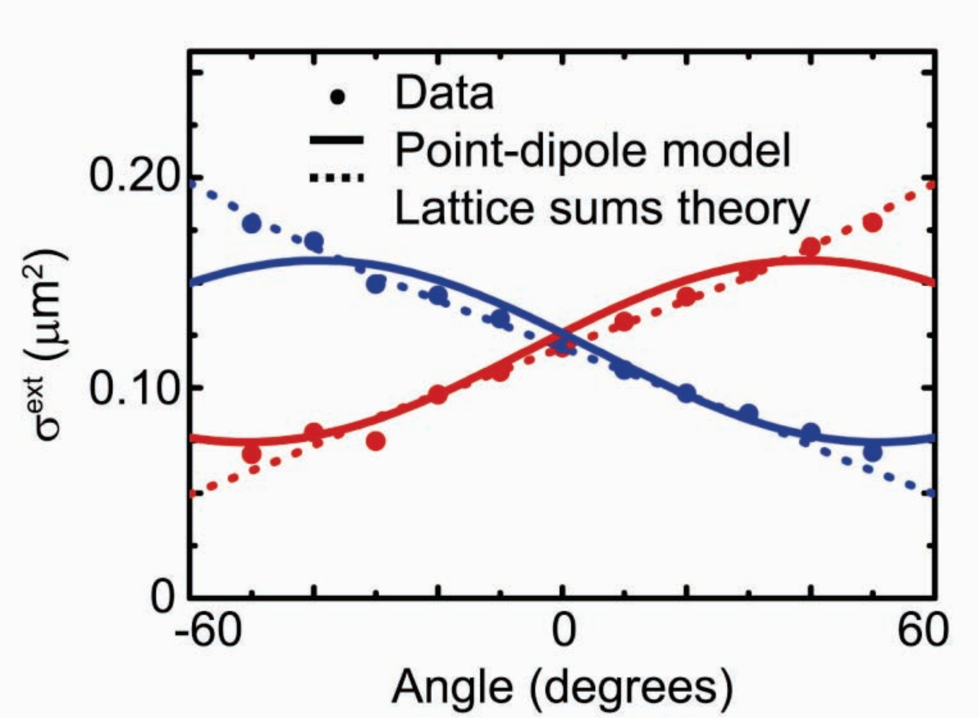
$$|\alpha_E| = 5.7V, |\alpha_H| = 2.3V, |\alpha_C| = 3.4V$$

$$|\alpha_C| \sim 0.88\sqrt{\alpha_E \alpha_H}$$

Strong scattering: α around 50% of unitary limit
Hugely optically active or “bi-anisotropic”

Existence of “pseudochirality”: Plum & Zheludev

Fit of polarizability



Quantitative fit of α to

$$|\alpha_E| = 5.7V, |\alpha_H| = 2.3V, |\alpha_C| = 3.4V$$

$$|\alpha_C| \sim 0.88\sqrt{\alpha_E \alpha_H}$$

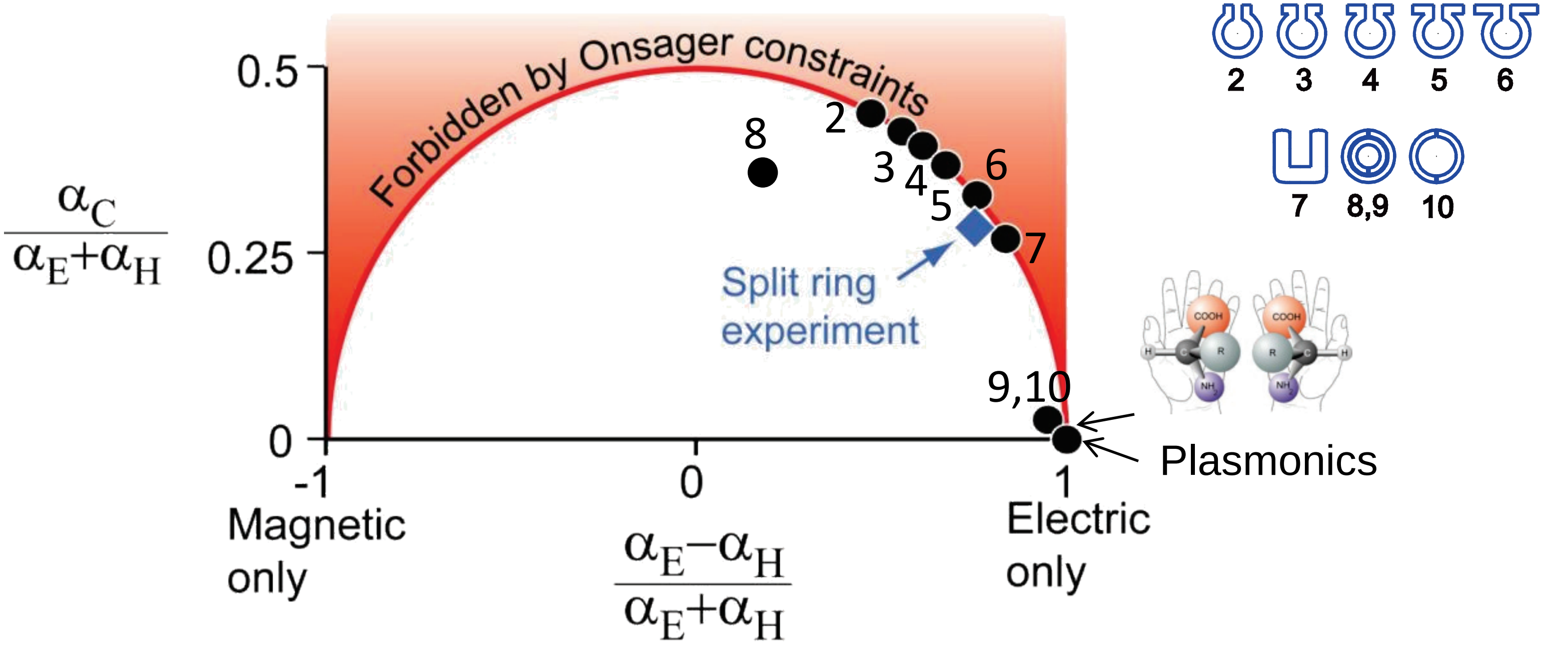
α around 50% of unitary limit

Strong magnetic scatterers

For one handedness, transmission resonance almost disappears entirely

Sersic, PRL **108** , 223903:1-5 (2012)

Phase diagram



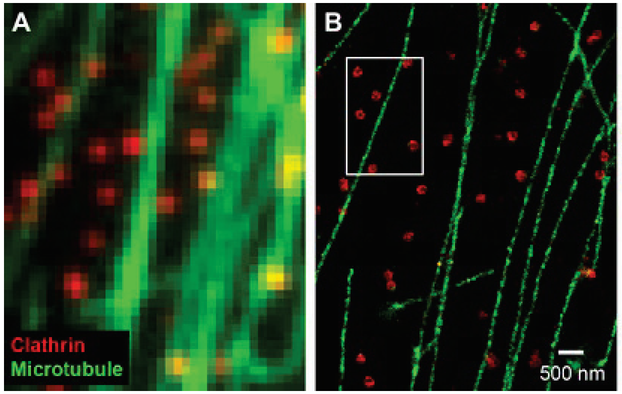
Almost all meta-scatterers we tested are maximally cross coupled

Reason: free charges generate **p** and **m** – bound by continuity

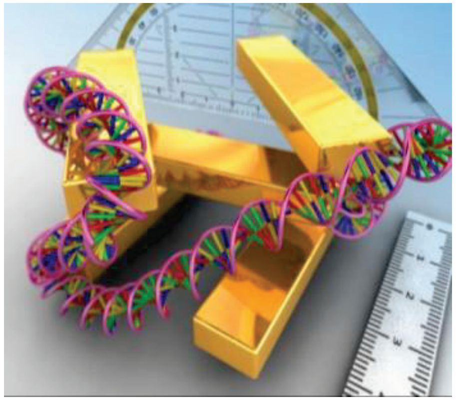
Method (1): α-retrieval: Mühlig et al., Metamaterials 5, 64 (2011).

(2): full-field: Kern & Martin , J. Opt Soc. Am. A vol. 26, p. 73 (2009)

Conclusions



Bates & Zhuang [PALM, STORM]



Liu & Alivisatos

Antenna physics at optical wavelengths:

- Plasmonics instead of perfect metals
- Quantum emitters & single photons instead of *I, V, Z*

Phased array physics + radiative impedance + spoof magnetism

Brighter microscopy, quantum optics, spectroscopy & LEDs

Thanks

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NWO

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