Probing the LDOS of plasmonic metamaterials

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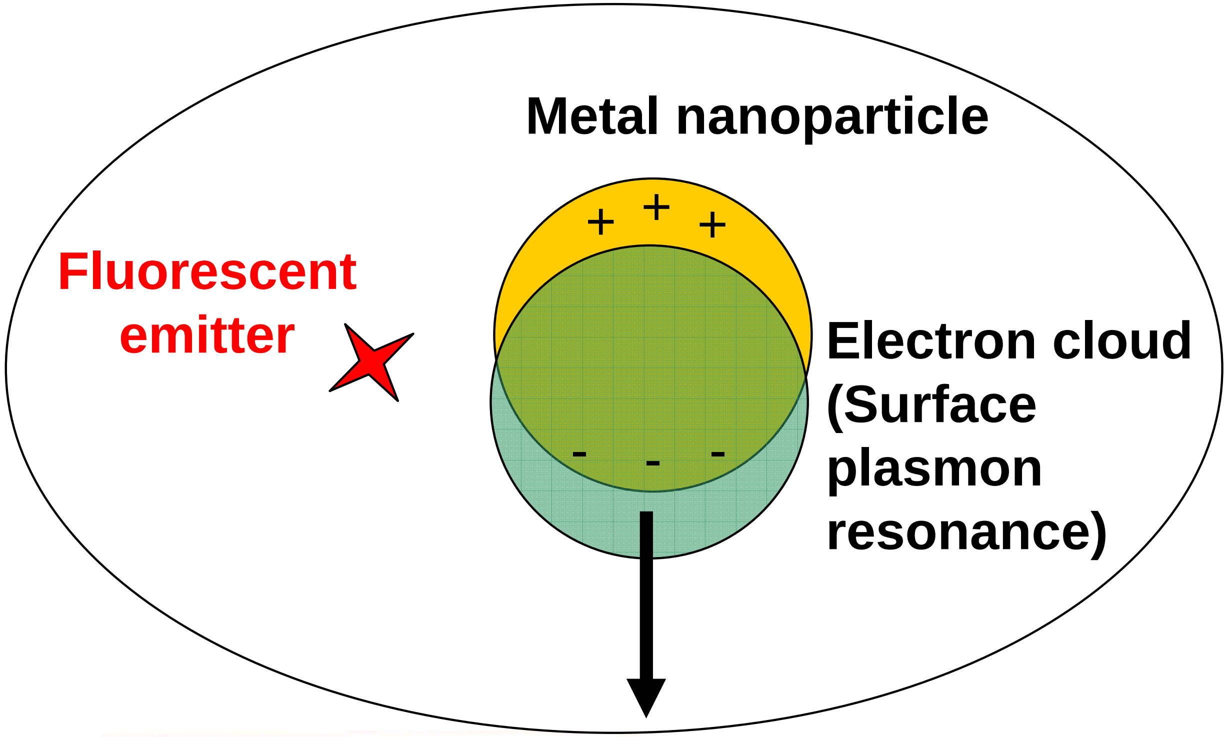
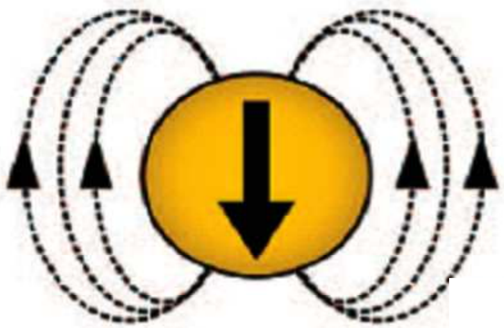
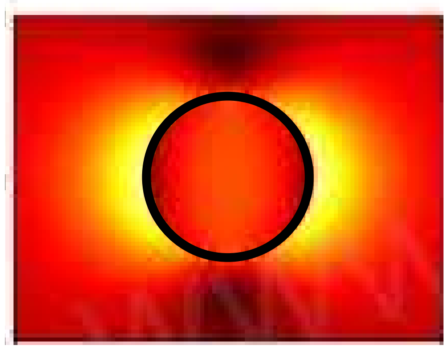
Outline

- Photonic LDOS and optical antennas
- Superradiant emitters coupled to optical antennas
- From nano-antennas to metamaterials
- Conclusion

Intro: Optical nano-antennas



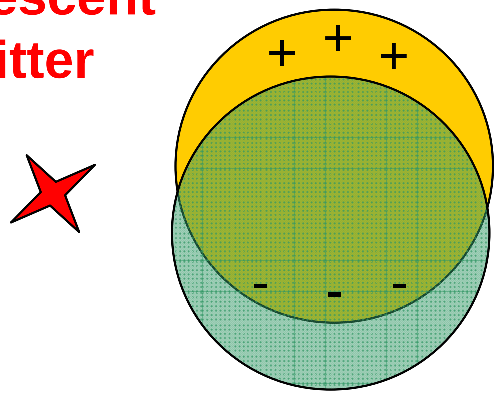




Optical nano-antennas used as emitters

Metal nanoparticle

Fluorescent emitter



When properly tuned, an optical antenna can enhance the emission rate of the emitter (Purcell effect).

Transition rate of a two-level quantum emitter located ar r_0 (Fermi's golden rule):

$$\Gamma = \frac{\pi\omega}{3\hbar\varepsilon_0}\left|\langle g \,|\, \hat{p} | e \rangle\right|^2 \rho_P(r_0,\omega)$$

Loc S)

The LDOS provides a link between the quantum and the classical descriptions

Electromagnetic power dissipated by a cla

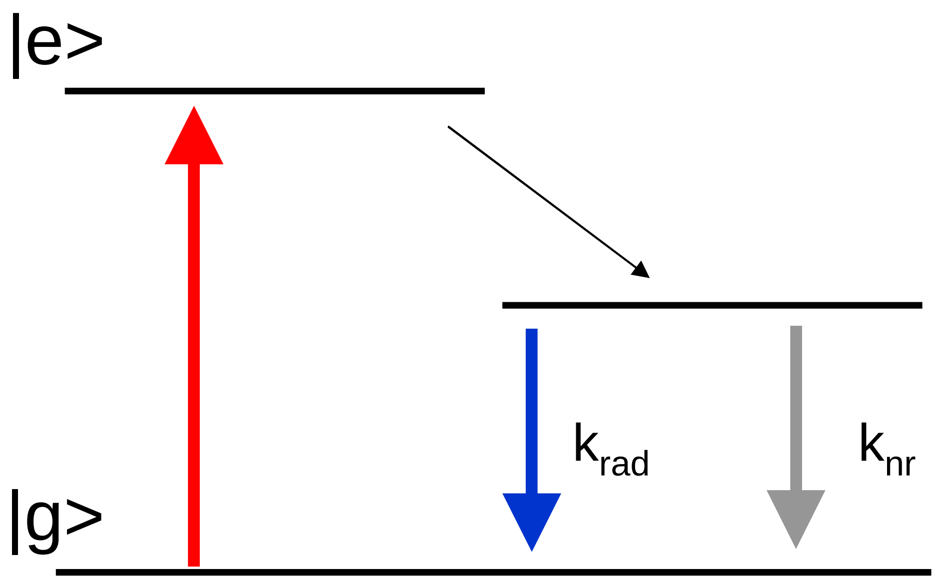
$$P = \frac{\pi\omega^2}{12\varepsilon_0}|p|^2 \rho_P(r_0,\omega)$$

Bharadwaj et al., Adv. Opt. Photon. (2009)

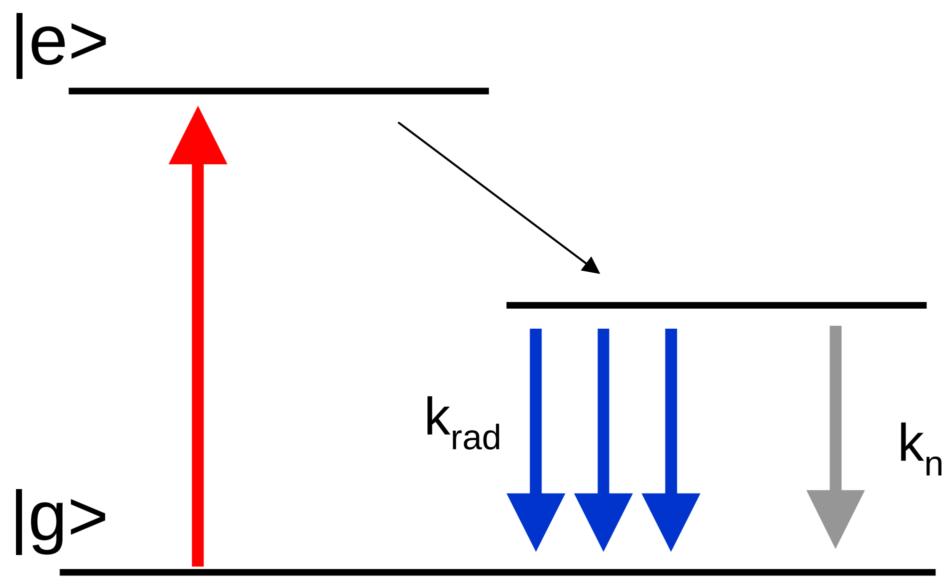
An intuitive description of the LDOS

-> The LDOS describes the number of radiative decay channels available for a given emitter, *at the emitter position*.

-> The luminescence properties of a quantum emitter are directly related to its environment.

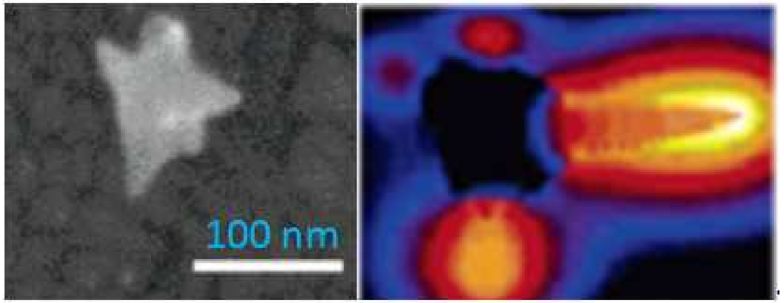


Emitter alone
(low LDOS)

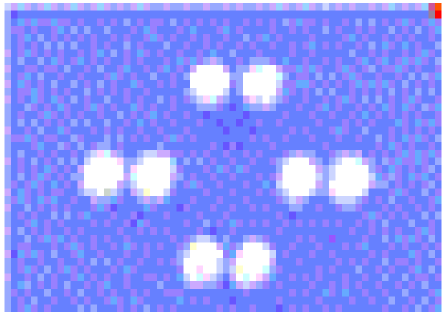


Emitter + Optical antenna
(high LDOS)

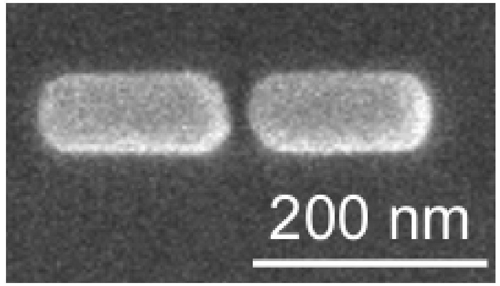
Some examples of nano-antennas



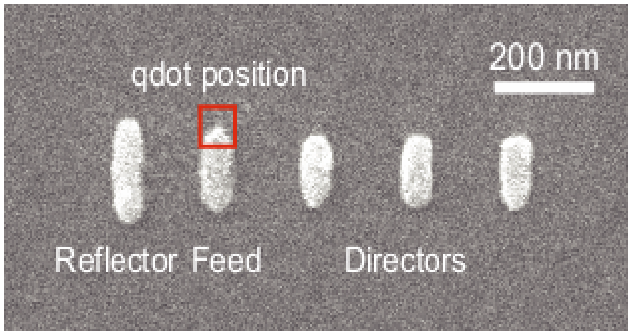
Nanostar, F. Hao *et al*,
Nano Letter **7**, 729 (2007)



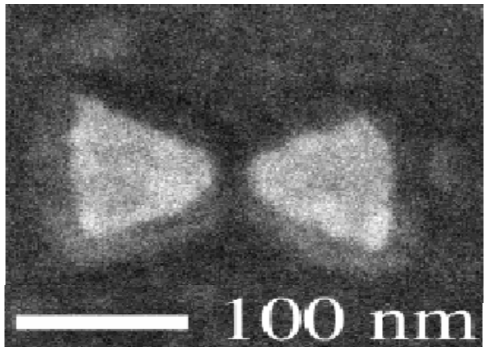
Dimer, Muskens, O.L., et al.,
Nano Letter **7**, 2871(2007)



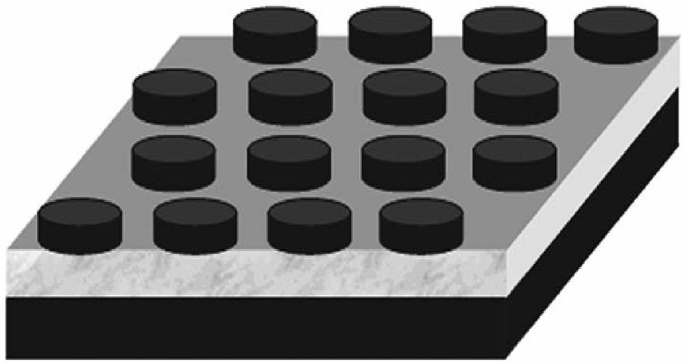
Dipole, Huang J-S *et al*,
Nat Comm **1**, 150 (2010)



Yagi-Uda, A. G. Curto,
Science **329**, 930 (2010)



Bowtie, Fromm, D.P. et al,
Nano Letter 4, 957(2004)



Patch, Esteban et al.,
PRL 2010

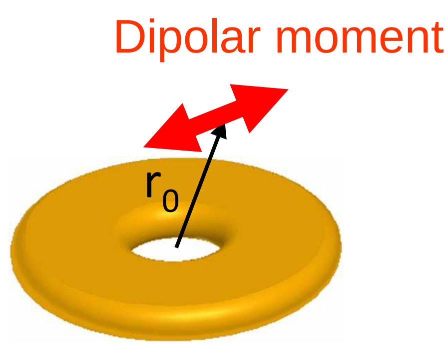
Optical properties of Au ring antennas

• We use the **boundary element method** to analyze the interaction between a dipolar emitter and a Au ring antenna.

• We assess the performances of the ring antenna based on:

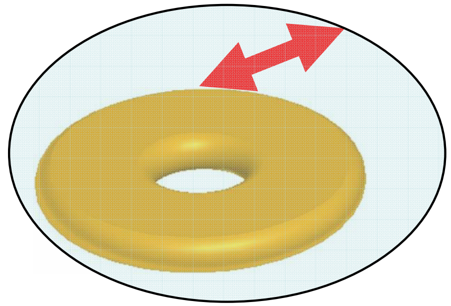
- The total power extracted from the emitter:

$$\boxed{P} = P_0\left[1+\frac{6\pi\varepsilon_0}{|p|^2 k^3}\,\mathrm{Im}\{p^*.E_s(r_0)\}\right]$$



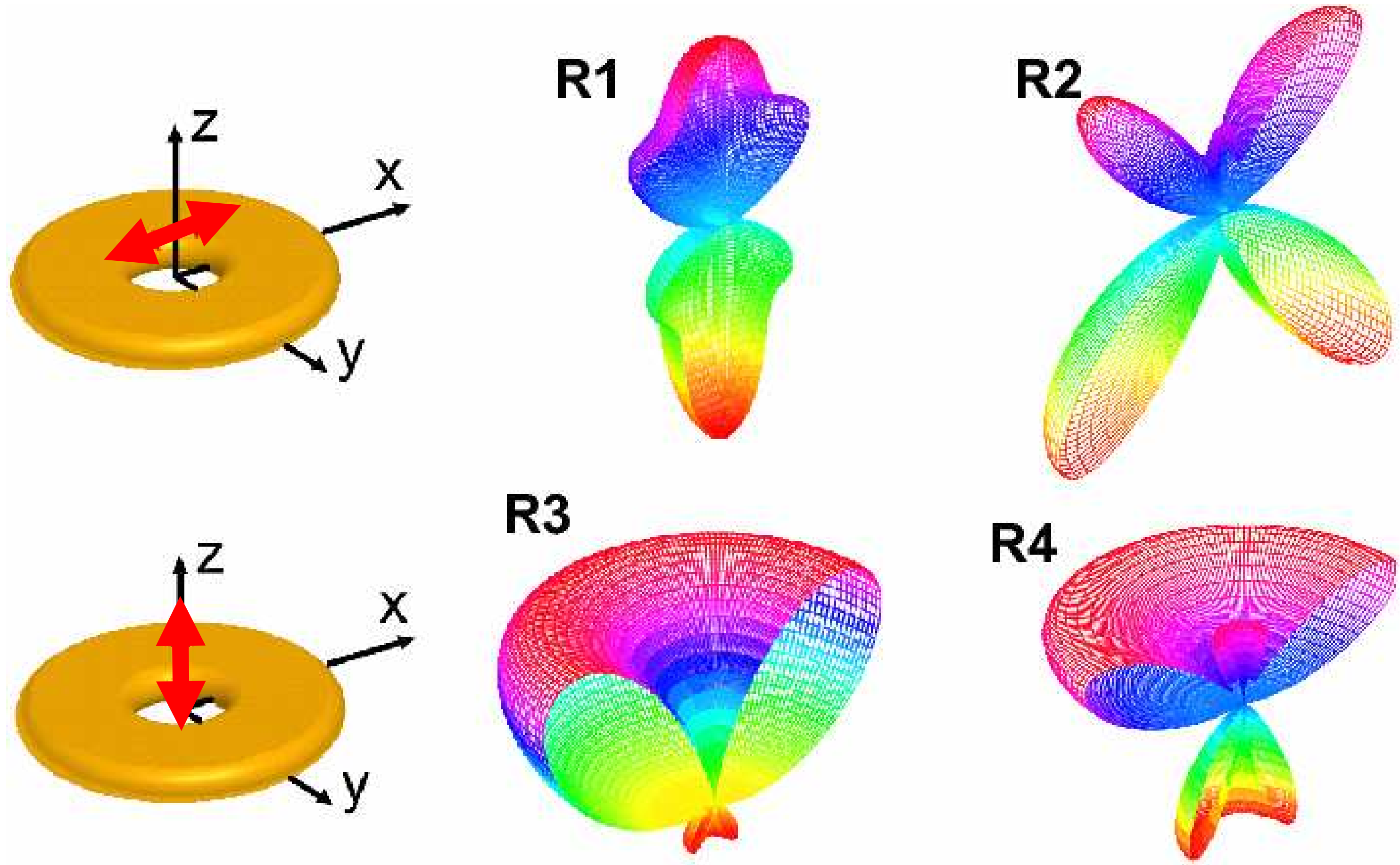
- The power reemitted as light:

$$\boxed{P_{rad}} = \int_{Surface} \Pi dS$$



Bharadwaj et al., Advances in Optics and Photonics ***1****, 438–483 (2009)*

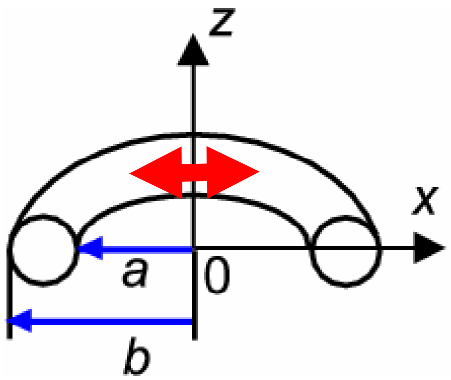
Ring with size comparable to λ: High directivity



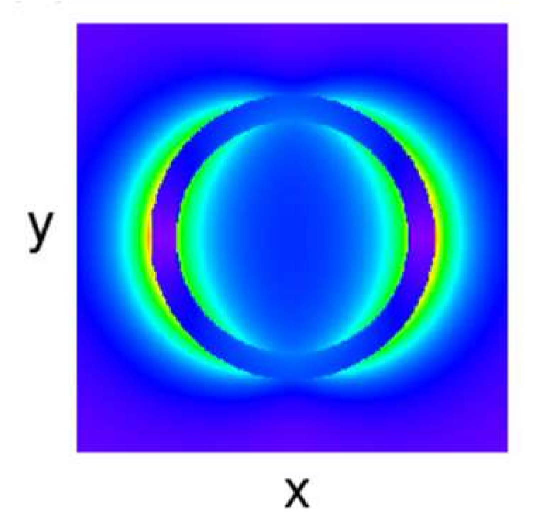
Directivity: $D = 4\pi\, p(\theta, \phi) / P_{rad}$

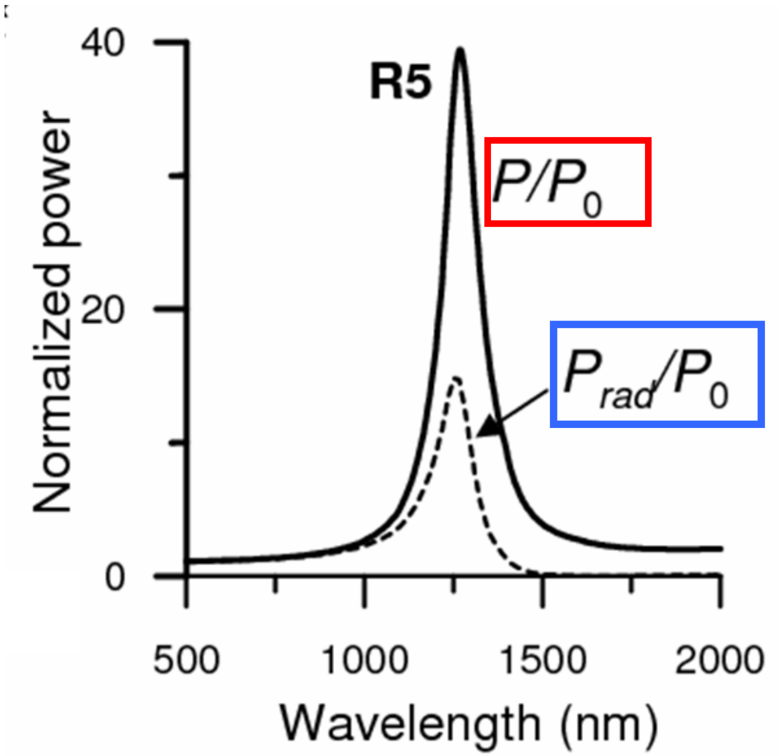
Teperik and Degiron, PRB 2011

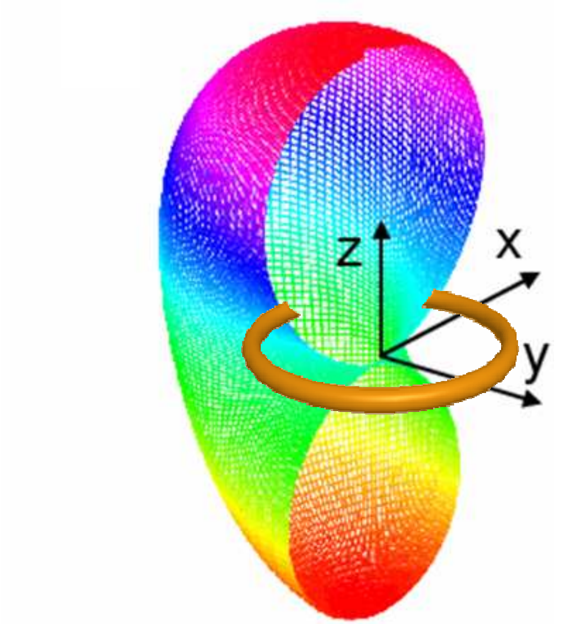
Ring much smaller than λ : dipolar SP resonance



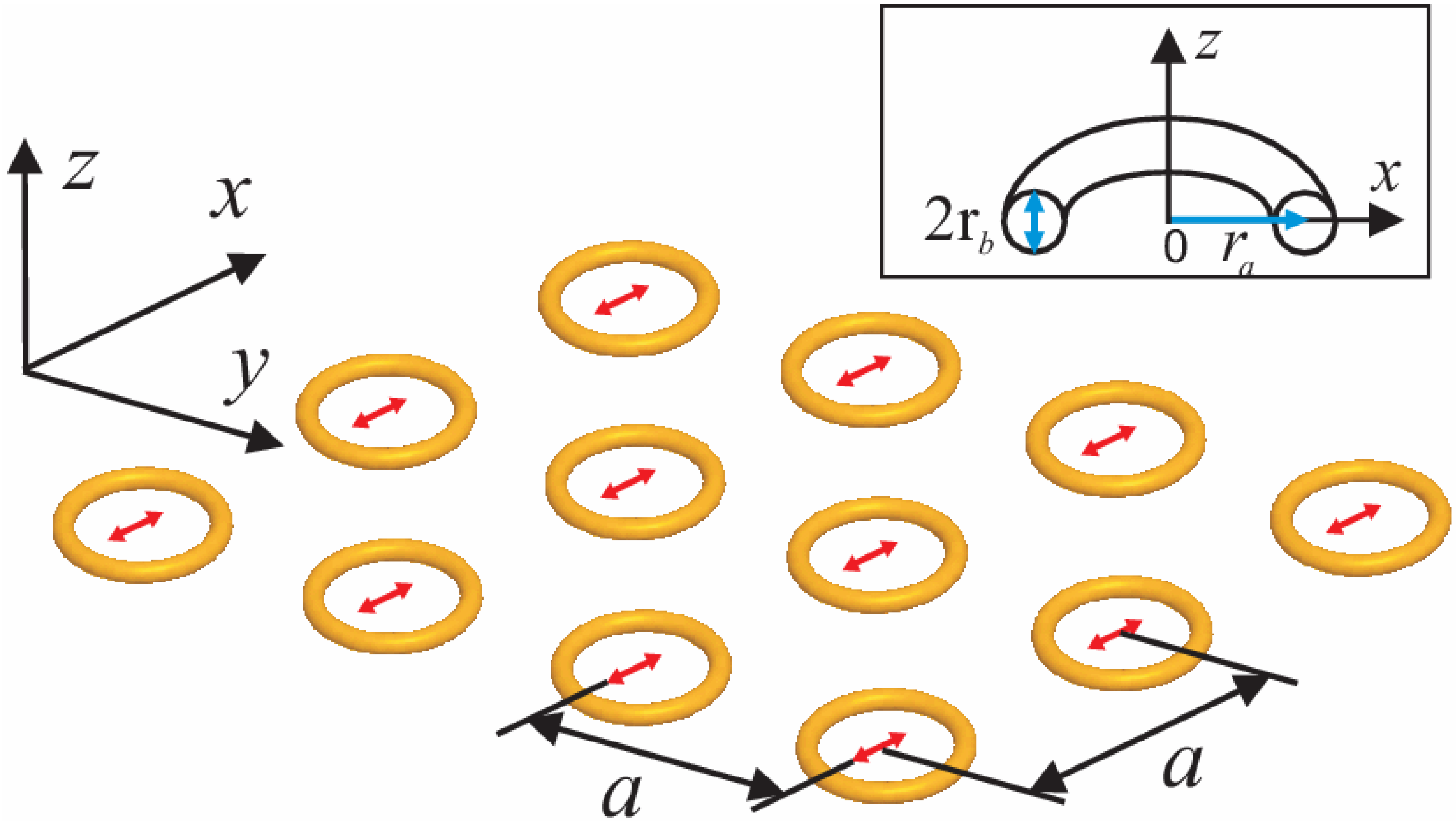
a = 80 nm
b = 100 nm







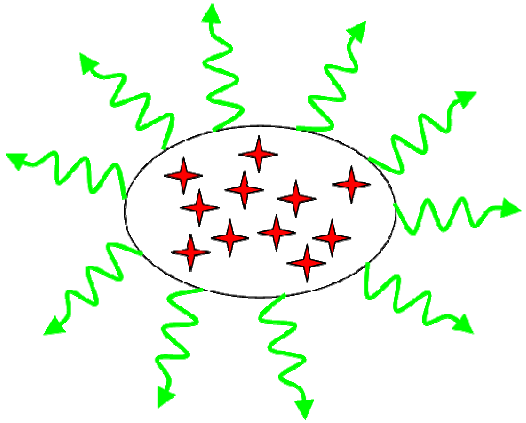
What if the emitters are periodically spaced and emit light in phase?



What if the emitters are periodically spaced and emit light in phase?

This condition arises when the emitters are superradiant

Spontaneous emission (e.g. fluorescence) of N emitters :

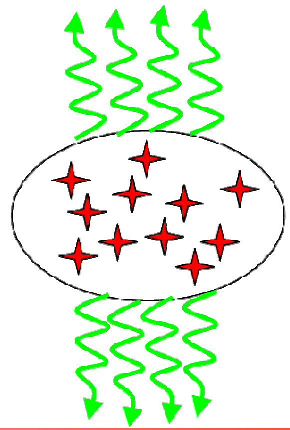


-**Isotropic emission**

-**Incoherent phase**

-**Total intensity ≈ N**

Superradiance of N emitters:

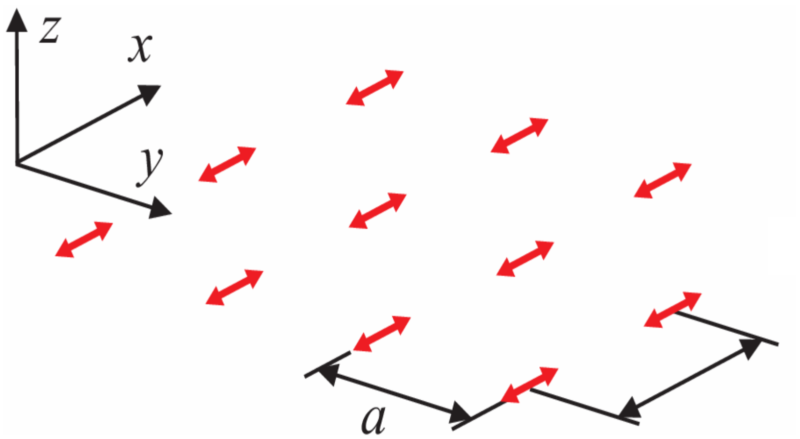


-**Anisotropic emission**

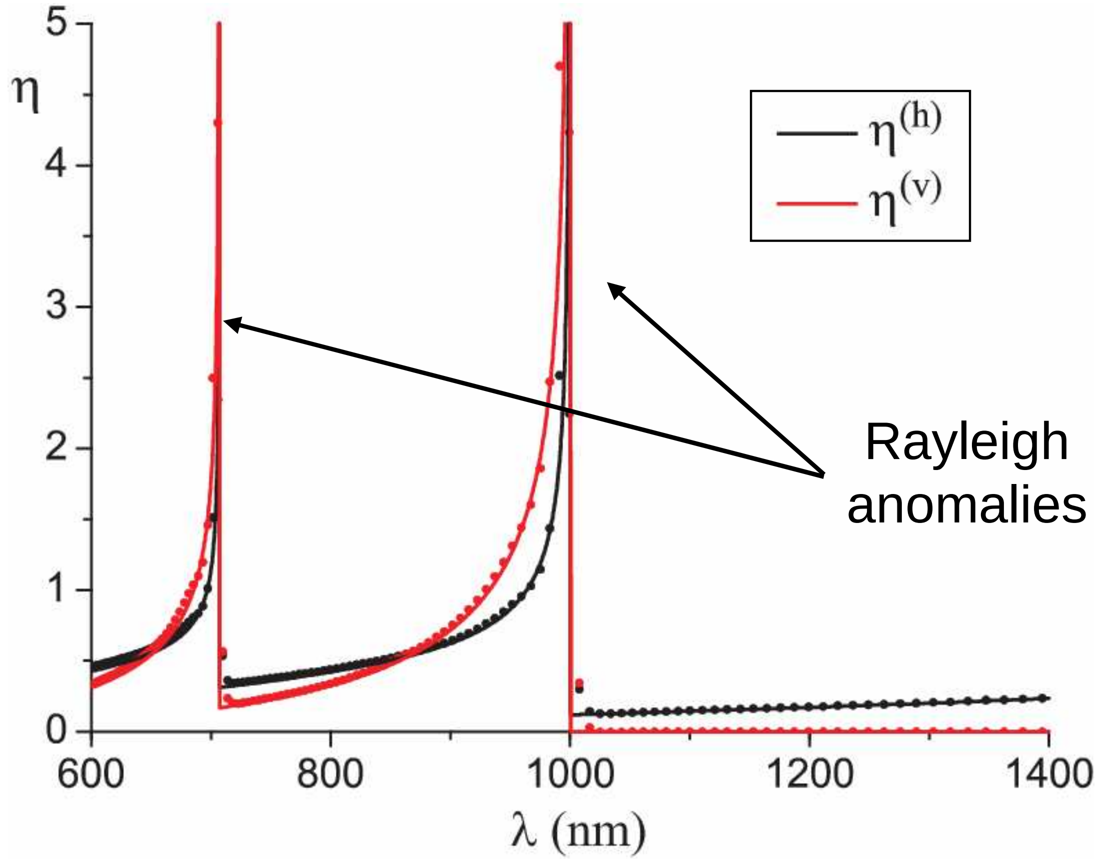
-**Emitters are in phase**

-**Total intensity ≈ N^2**

Lattice of coherent dipole emitters



Solid curves: analytical calculations
Points: numerical FEM calculations



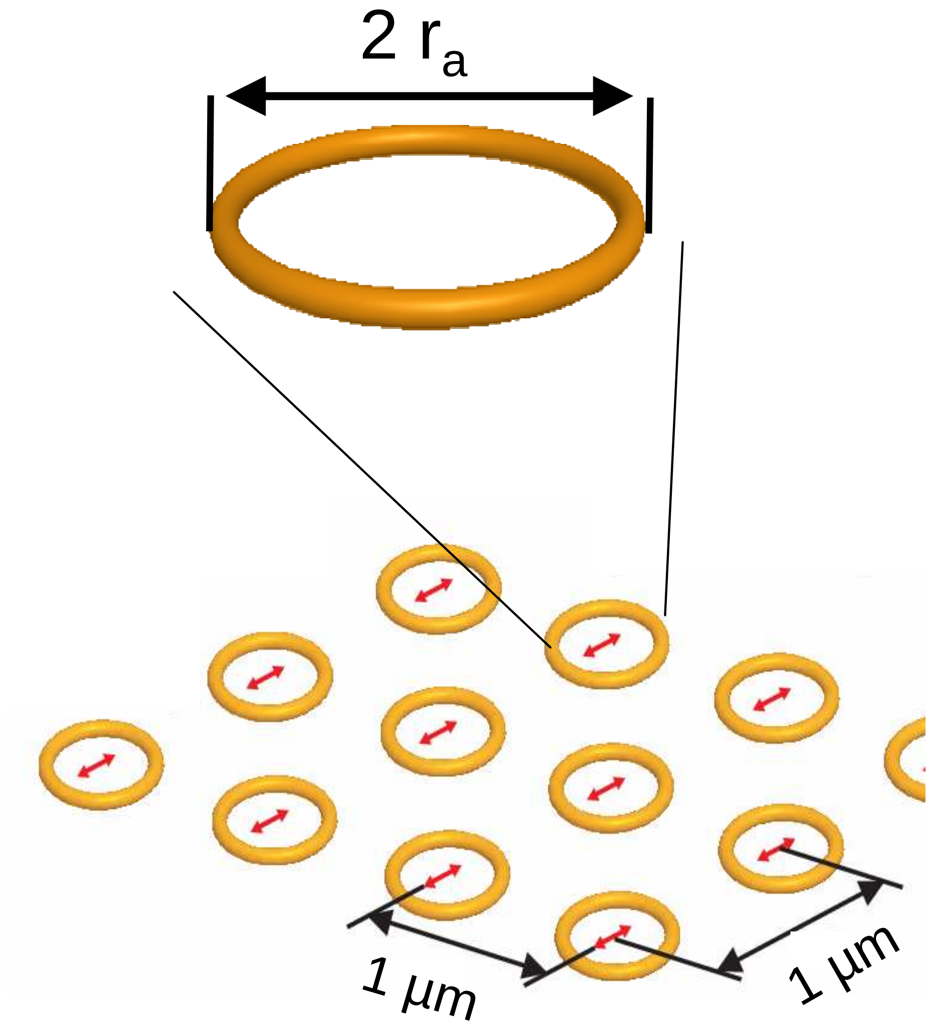
Power $\eta^{(h)}$ [$\eta^{(v)}$] radiated by a lattice of horizontal [vertical] dipoles:

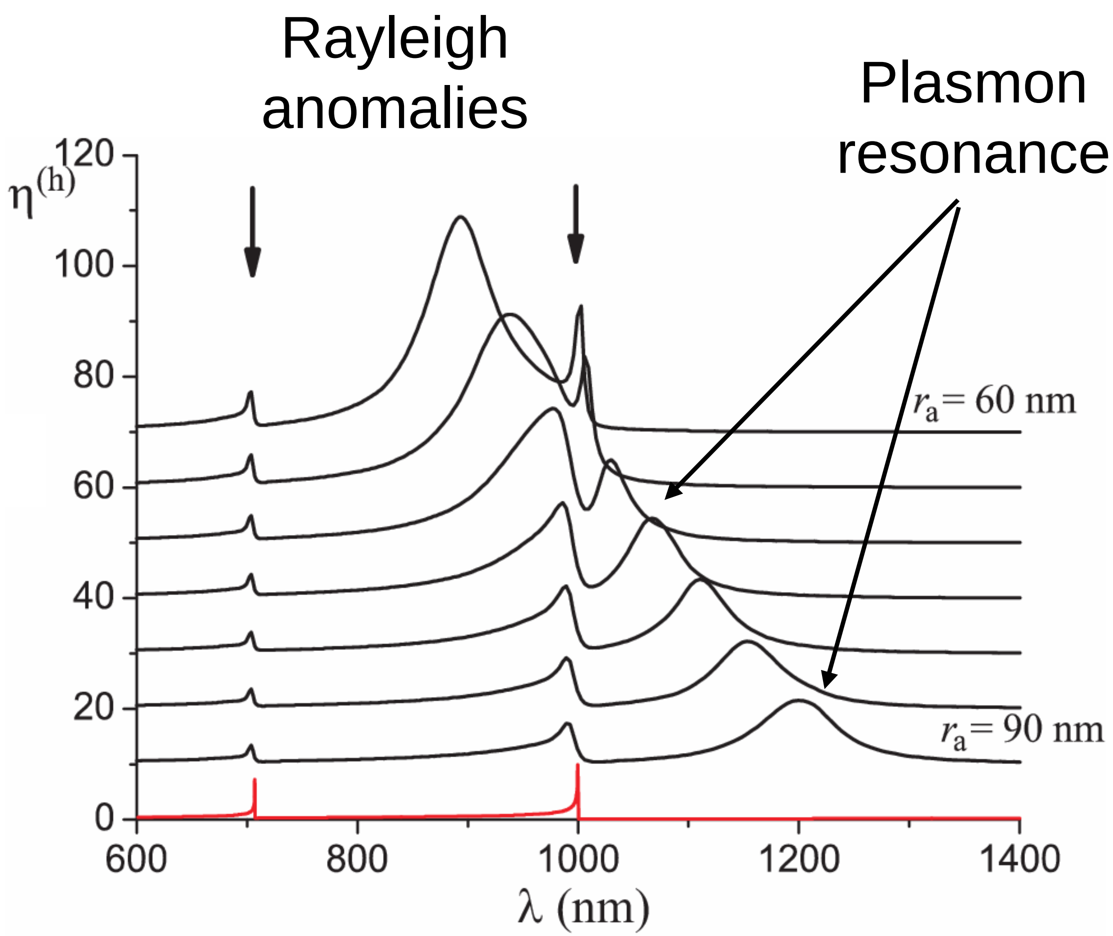
$$\eta^{(v)} = \frac{3\pi}{2k^3a^2}\sum_{\mathbf{g}} \frac{g_x^2 + g_y^2}{q_z}$$

$$\eta^{(v)} = \frac{3\pi}{2k^3a^2}\sum_{\mathbf{g}} \frac{k^2 - g_x^2}{q_z}$$

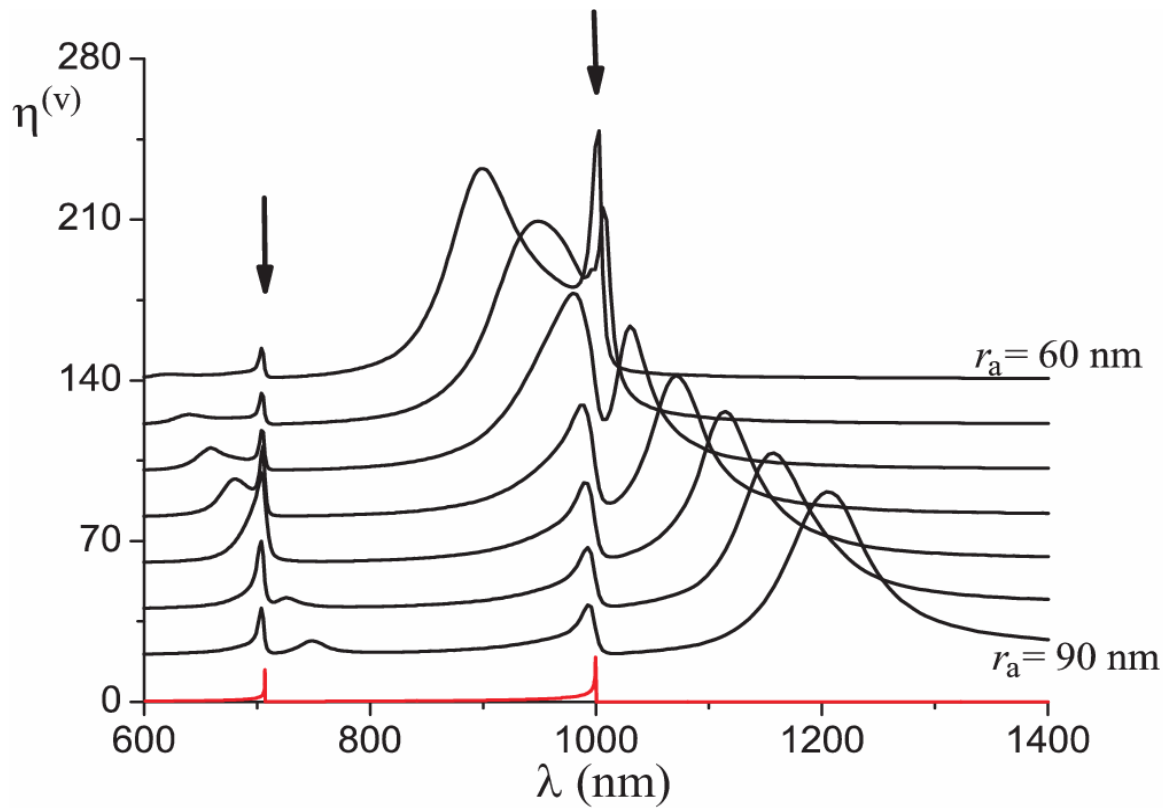
with $\mathbf{g} = \frac{2\pi}{a}(n\hat{\mathbf{x}} + m\hat{\mathbf{y}})$ and $q_z = \sqrt{k^2 - g_x^2 - g_y^2}$

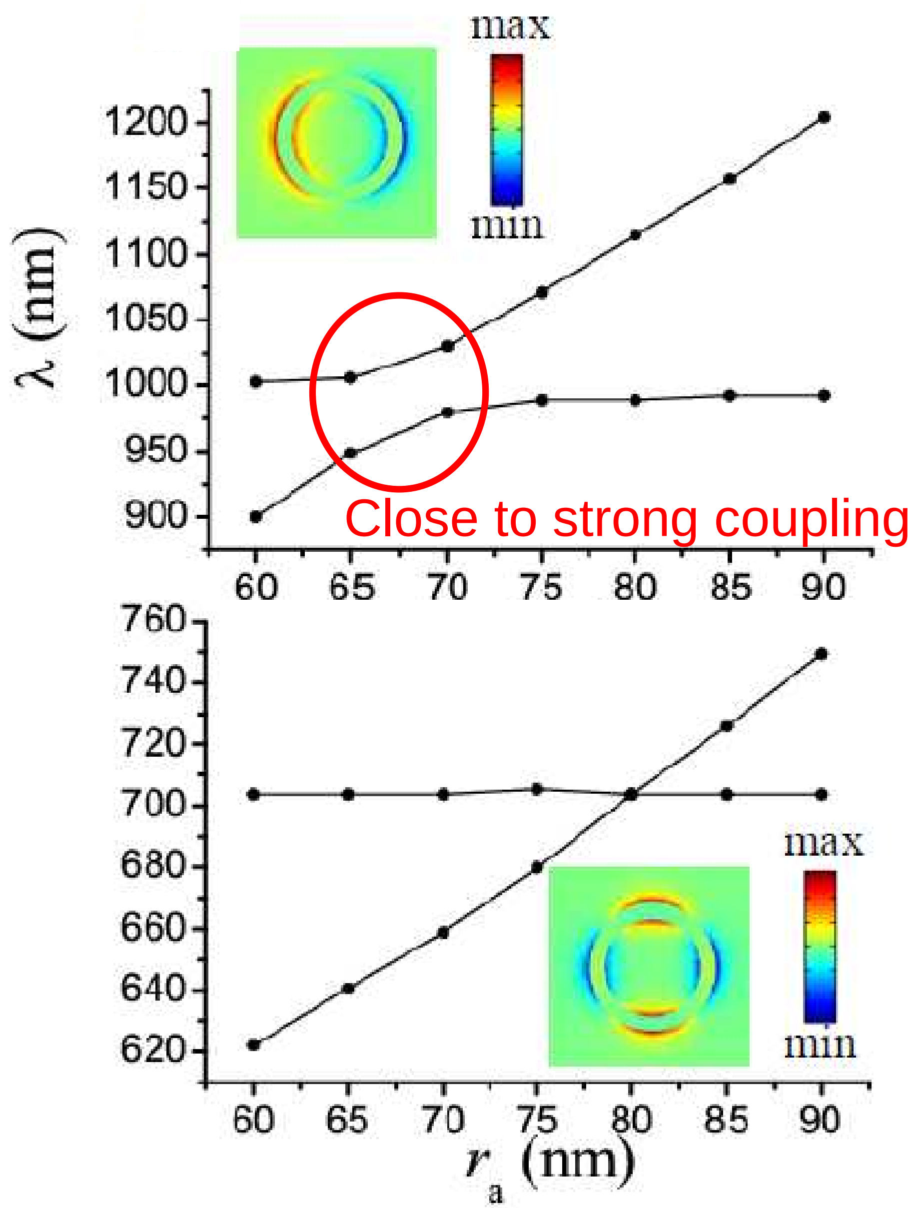
Horizontal dipole emitters coupled with Au rings





The case of vertical dipole emitters



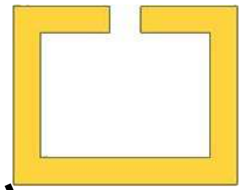


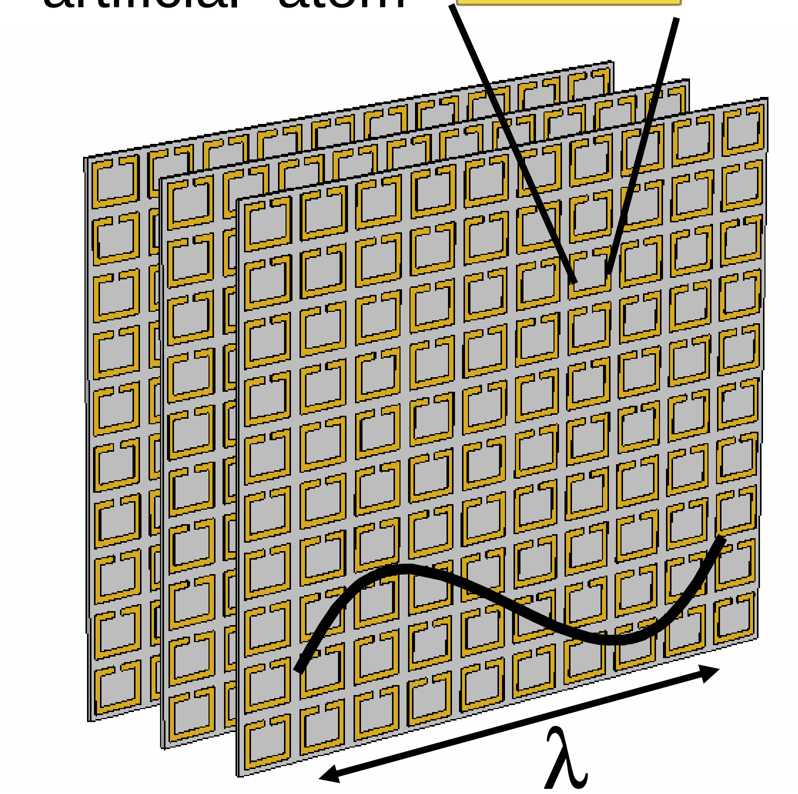
Teperik and Degiron, PRL **108** 147401 2012
Teperik and Degiron, PRB **86**, 245425 2012

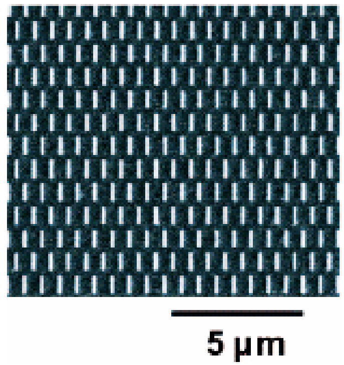
From optical antennas to metamaterials

Metamaterials are artificially structured composites that mimic the behavior of homogeneous media.

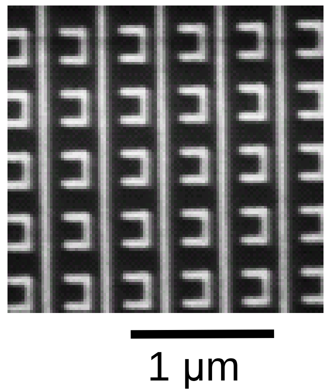
Metal nanoparticle: artificial “atom”



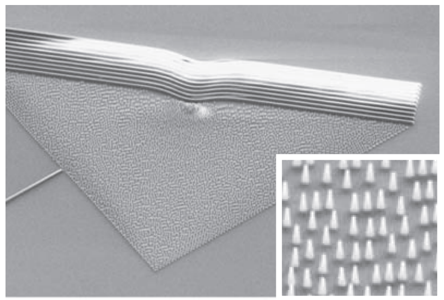




Negative index (Purdue 2005)



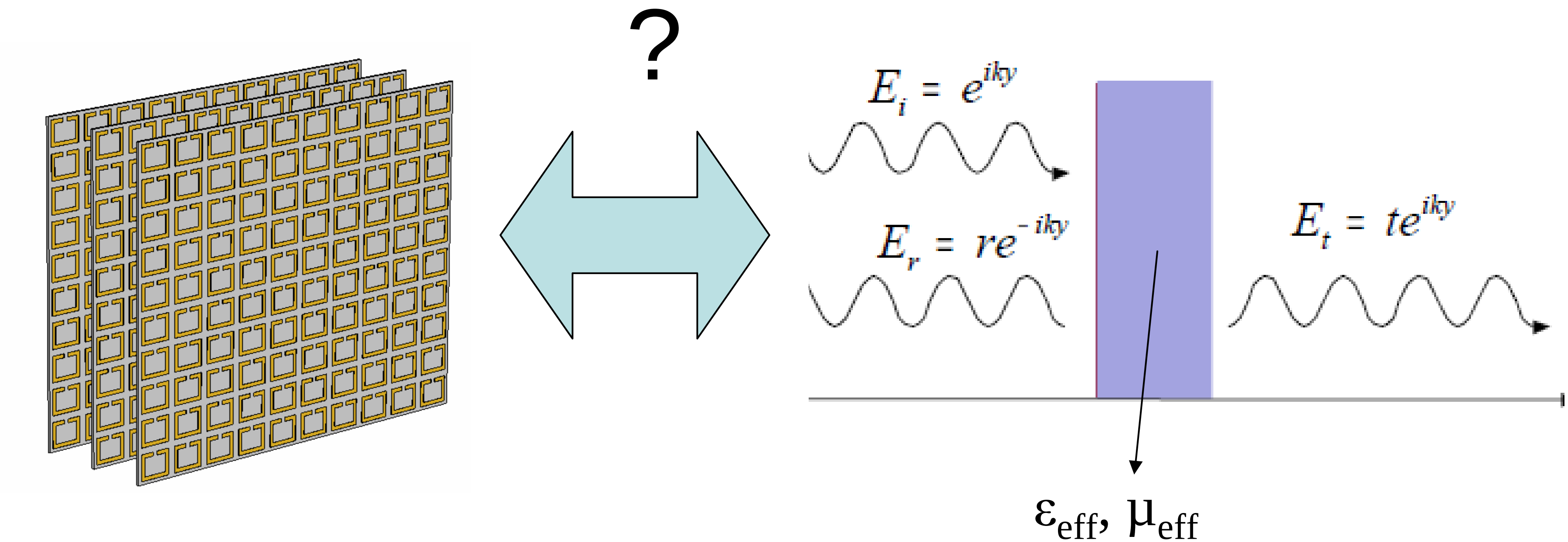
Artificial magnetism (IEF/LPN 2009)



“carpet” invisibility cloak (Cornell 2009)

The homogenization problem

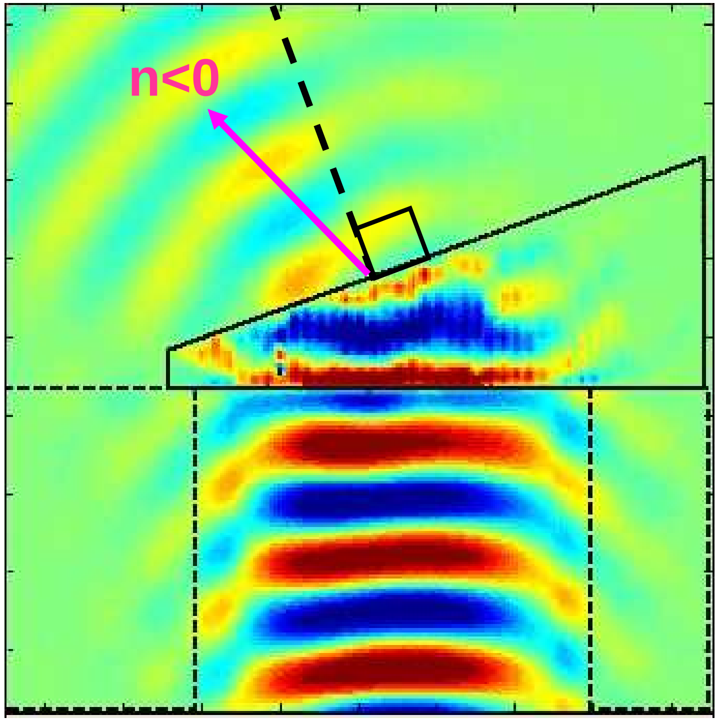
How can we tell that a given structure really acts as an effective homogeneous medium?



The homogenization problem

How can we tell that a given structure really acts as an effective homogeneous medium?

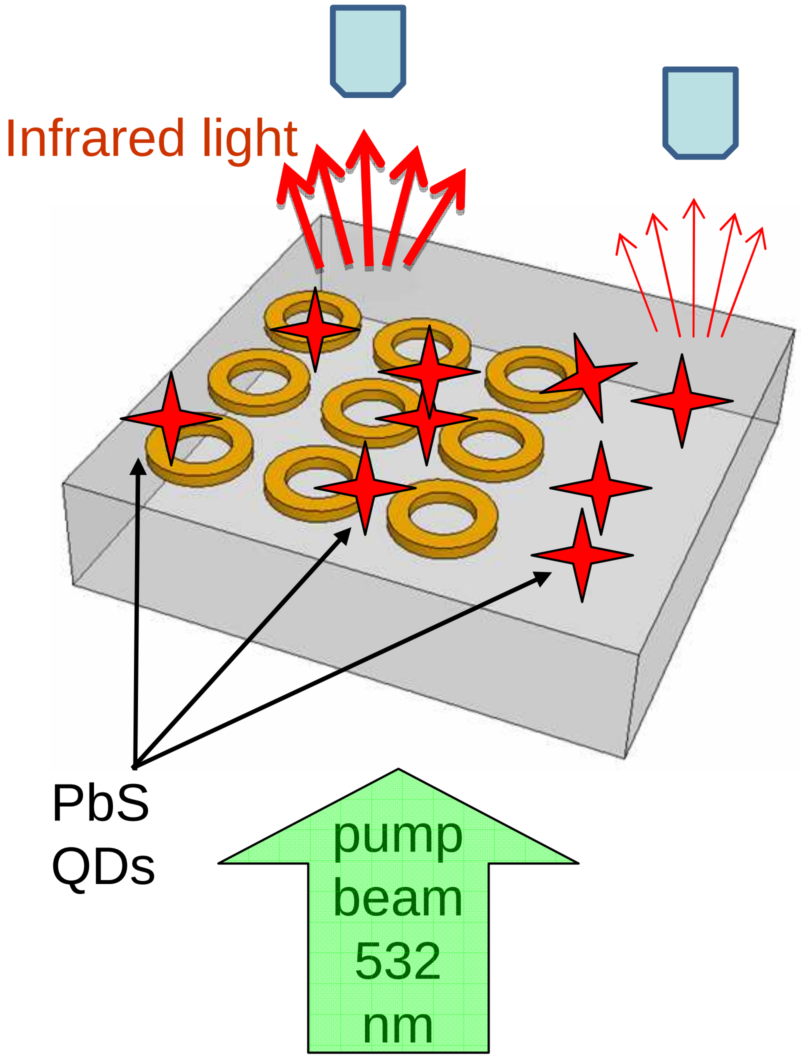
Experiments at 10 GHz



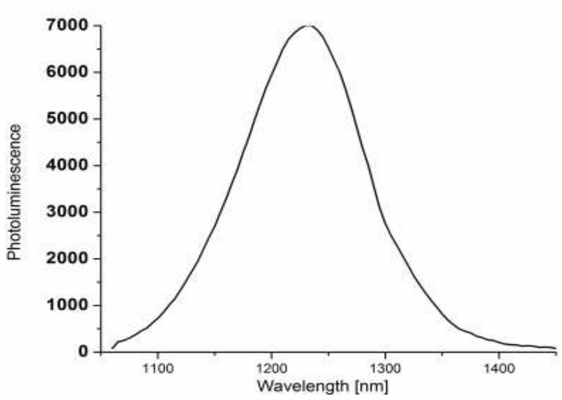
Justice, et al. Opt. Express **14,** 8694 (2006).

- Easy at microwave frequencies (direct visualization tools)
- What can we do in the optical regime?

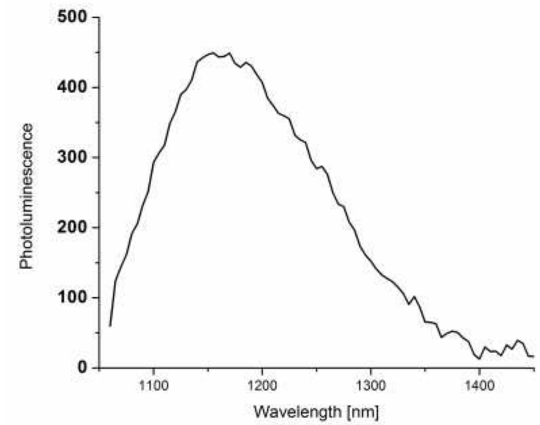
Experimental evidence: probing the LDOS with Photoluminescence (PL) measurements



PL above rings



PL above substrate



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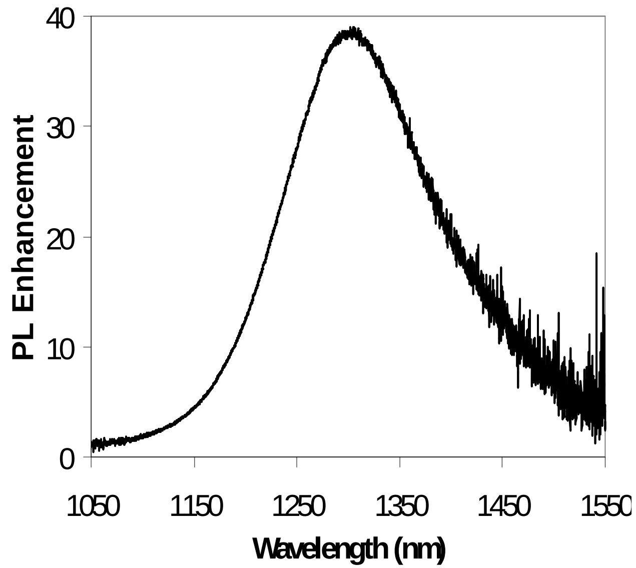
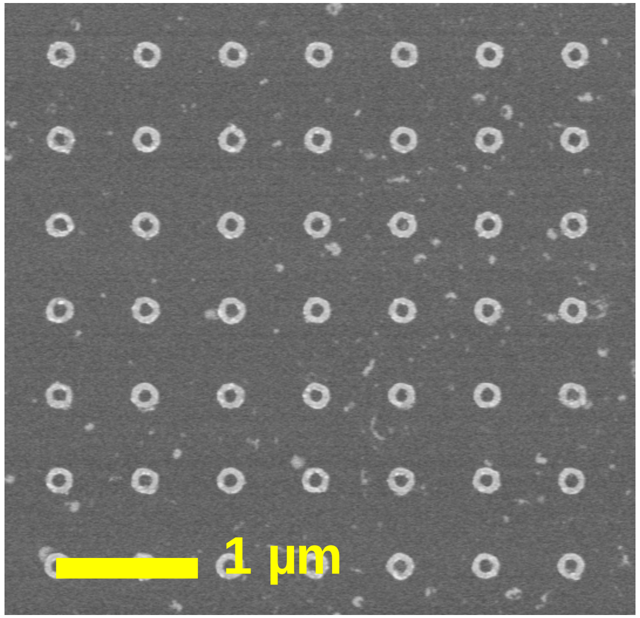
PL enhancement (apparent quantum yield)



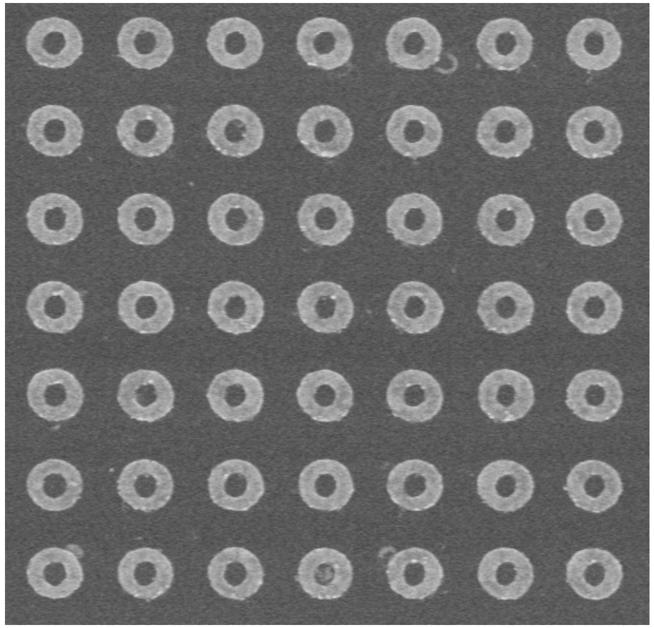
Structures under investigation

From small rings…
(radius ~80 nm)



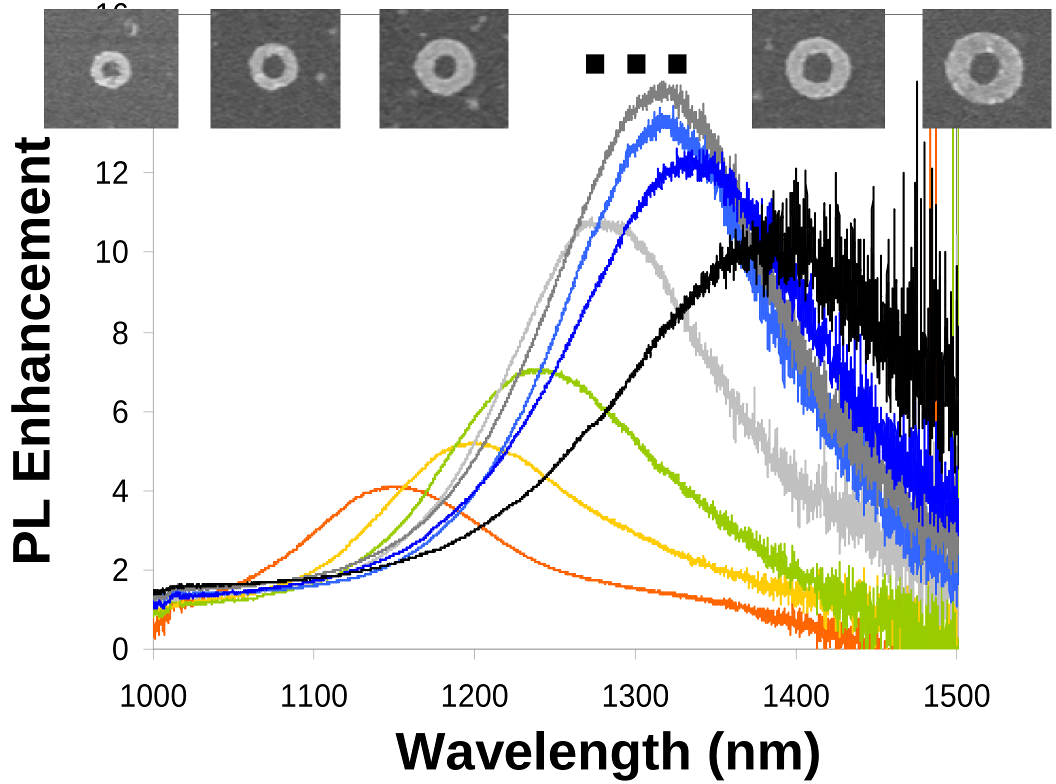
8 intermediate structures
between these 2

… to large rings
(radius ~120 nm)

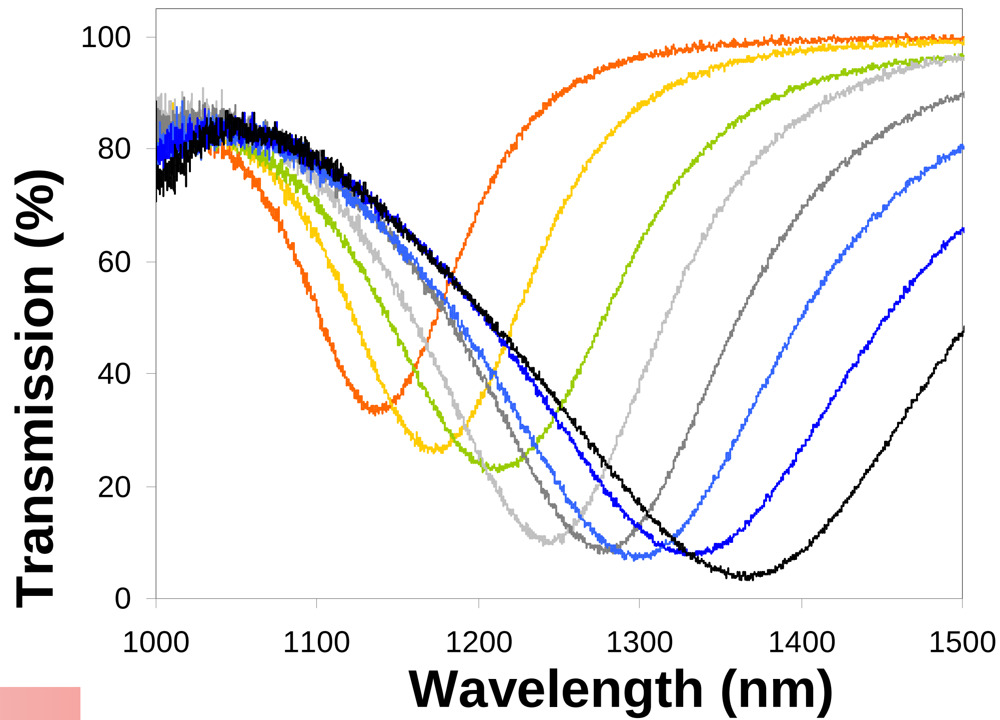


ALL STRUCTURES HAVE THE SAME PERIOD (600 nm)

PL Enhancement spectra

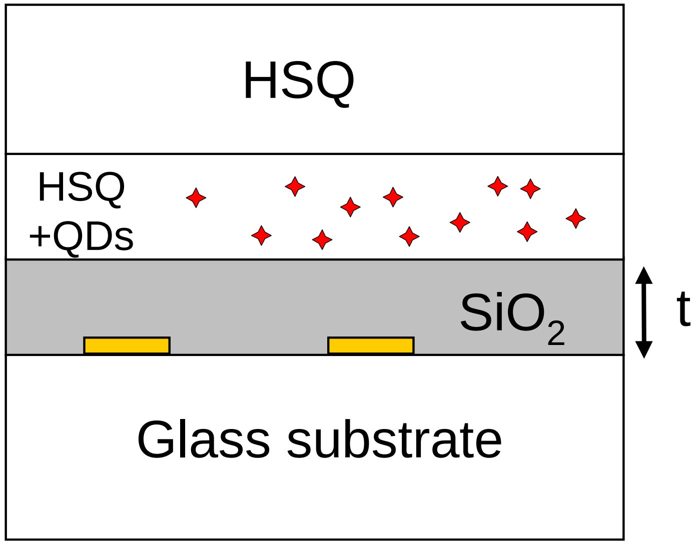


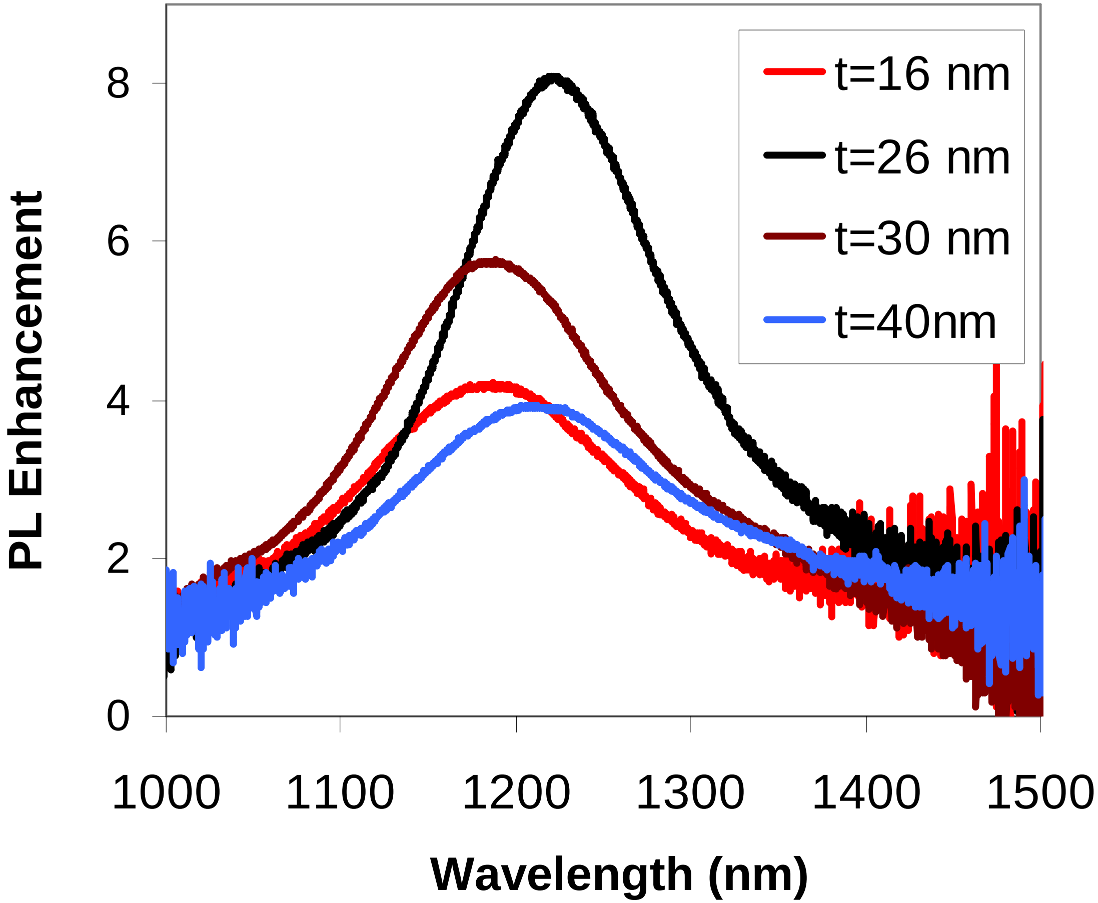
Passive transmission spectra
(FTIR measurements):



Same spectral signature (far-field response in both cases)

Influence of spacer thickness





Very different behavior than for a single antenna

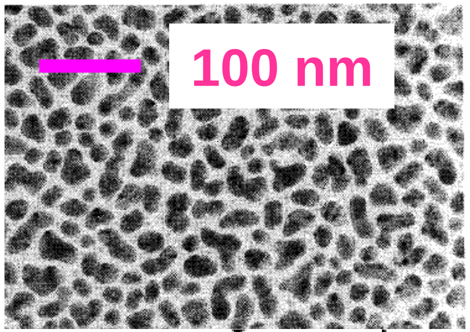
Looking at the literature

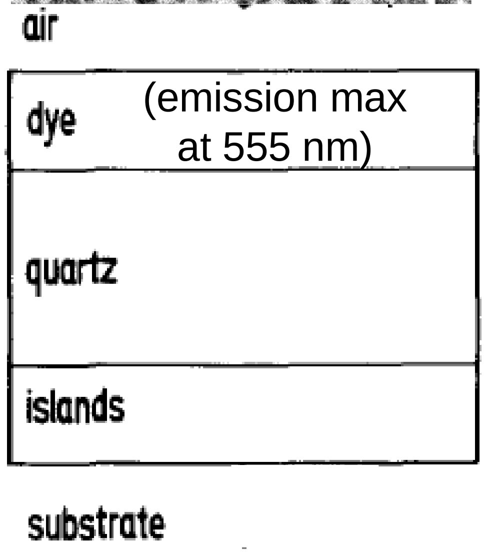
MOLECULAR PHYSICS, 1993, VOL. 80, NO. 5, 1031–1046

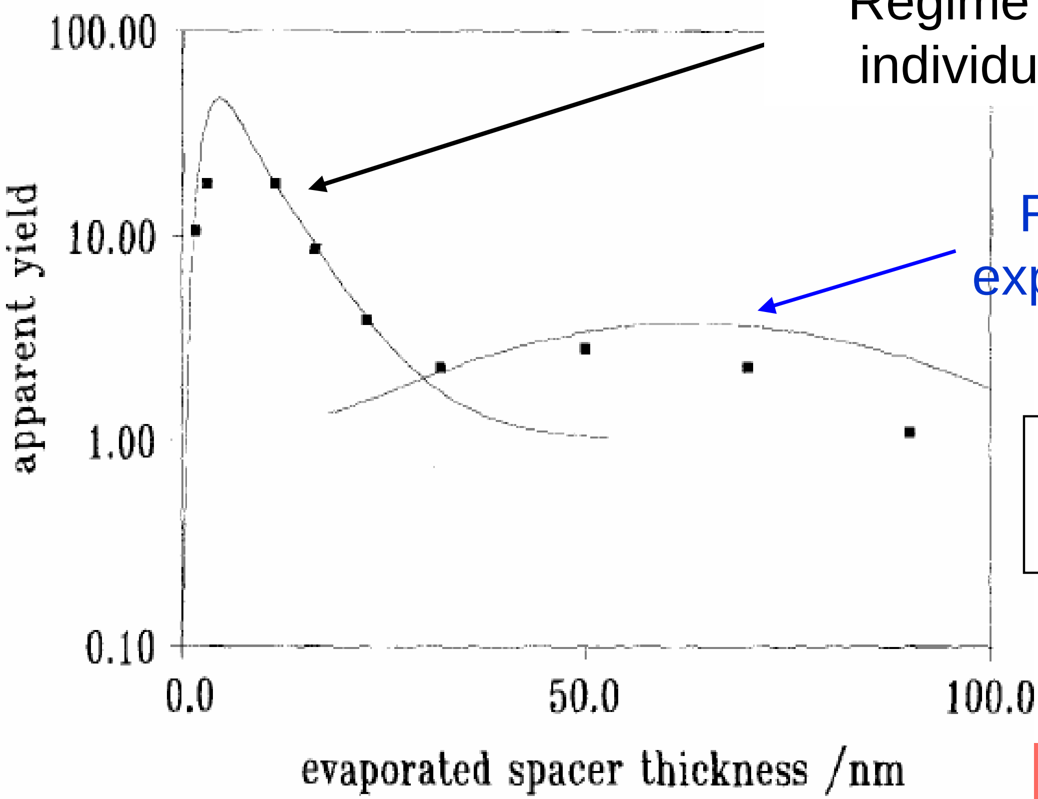
Enhanced dye fluorescence over silver island films: analysis of the distance dependence

By J. KÜMMERLEN†, A. LEITNER‡, H. BRUNNER‡, F. R. AUSSENEGG‡, and A. WOKAUN†

† Physical Chemistry II, University of Bayreuth, D-95440 Bayreuth, Germany
‡ Institute of Experimental Physics, Karl–Franzens–University, A-8010 Graz, Austria





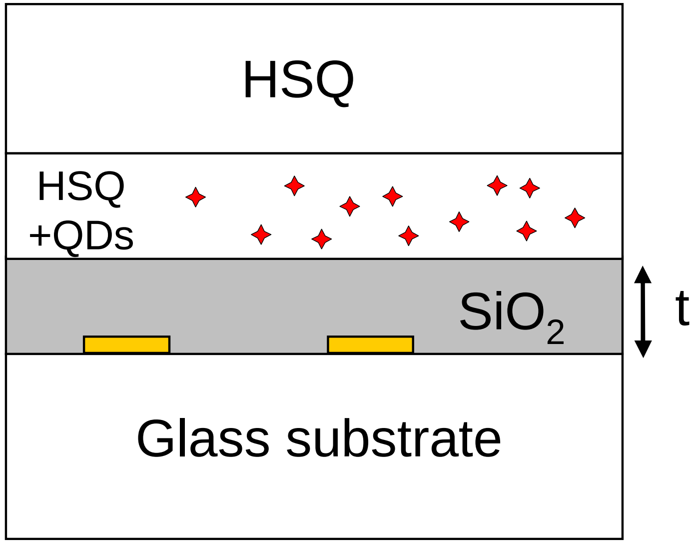


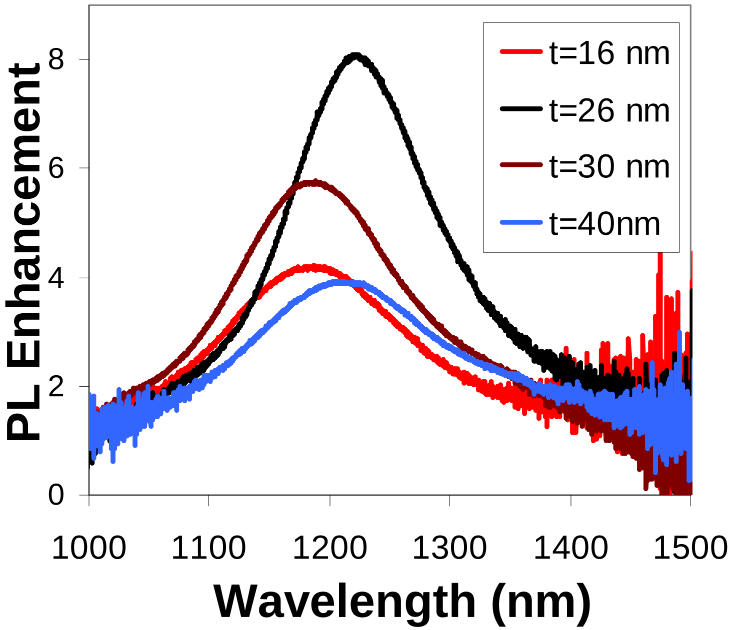
Regime 1: dominated by individual nanoparticles

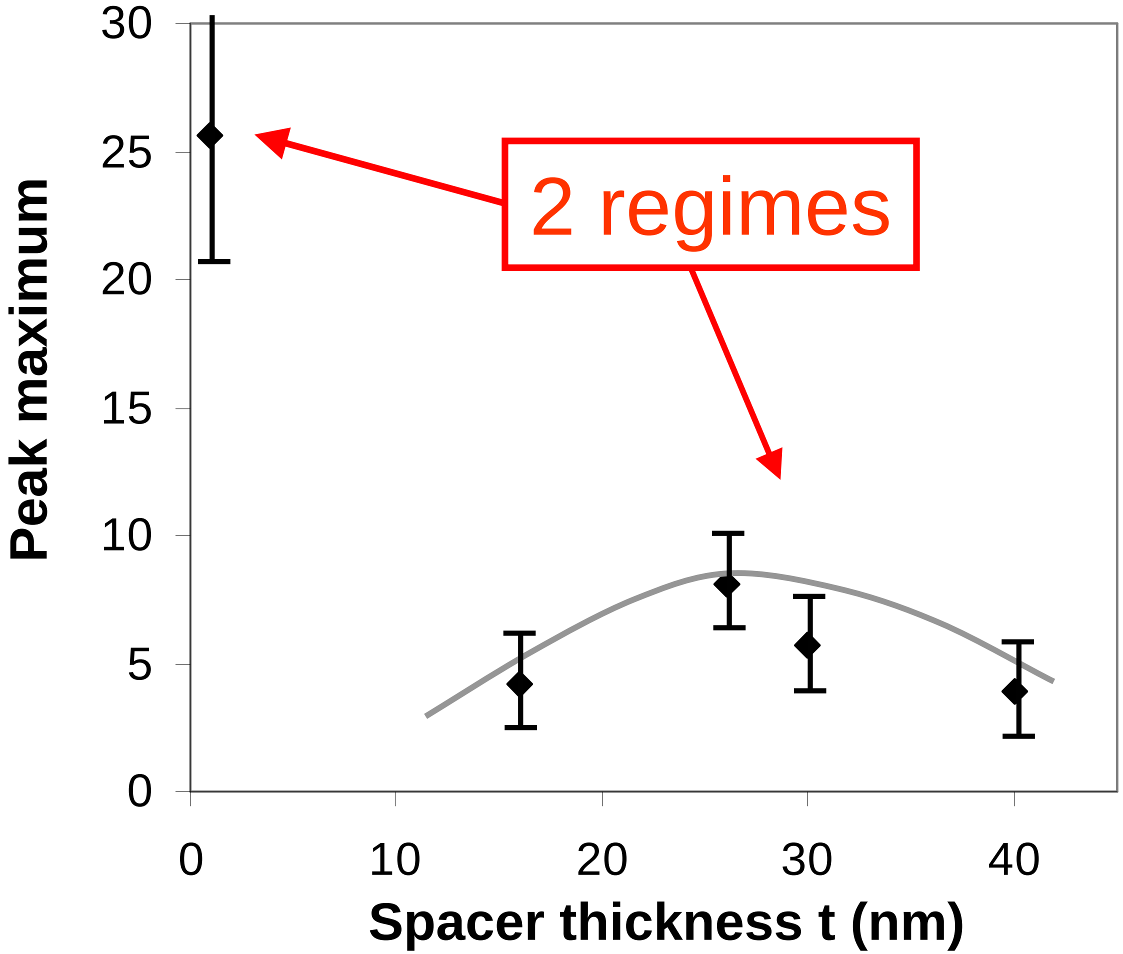
Regime 2: can be explained by effective medium theory

Ag island film = metamaterial

Back to our measurements



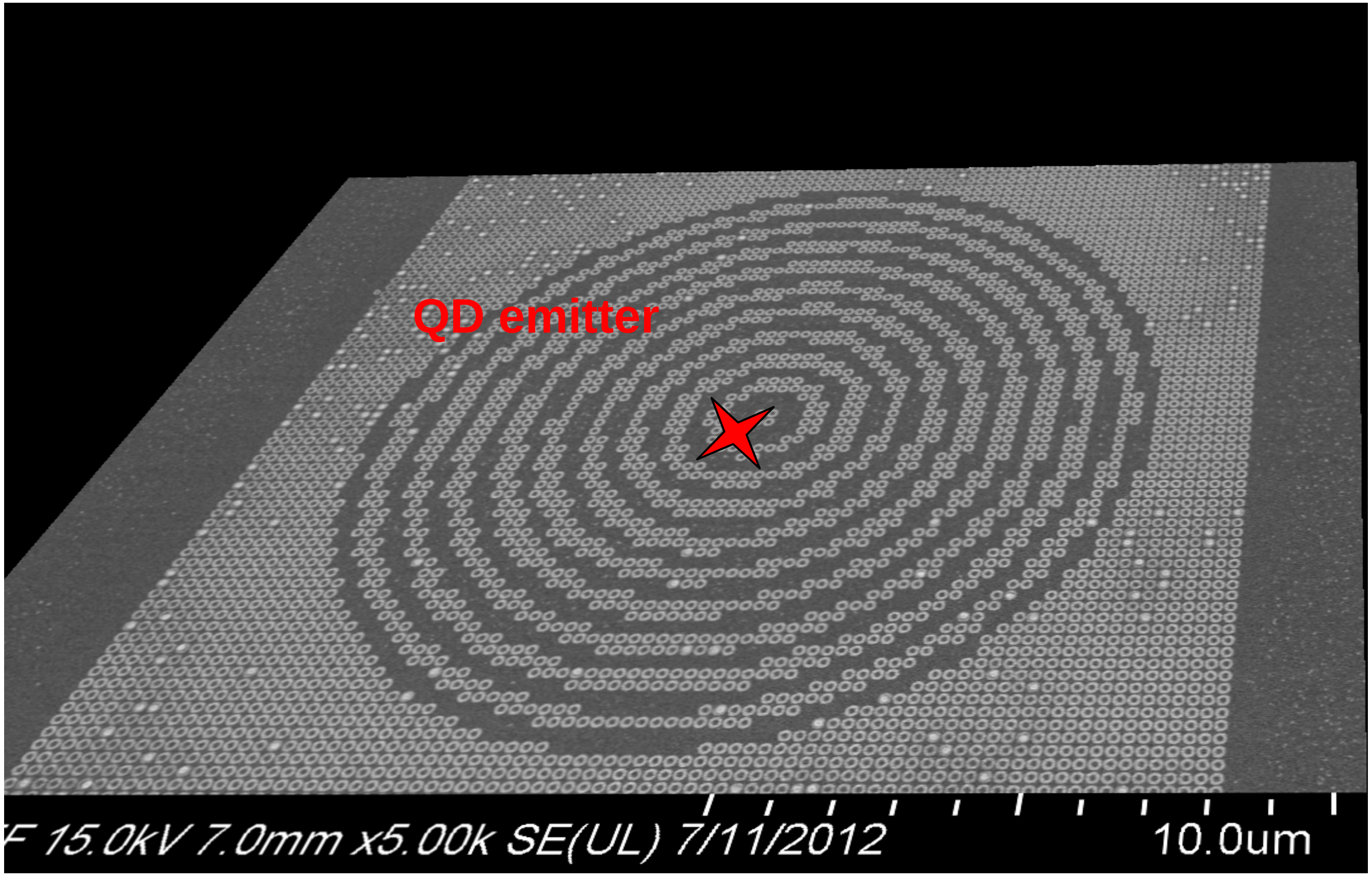




To act as a an effective medium, the plasmonic array must be used in the intermediate/far field regime.

Perspectives

Tailoring the LDOS with patterned metamaterial arrays



(In preparation)

Conclusion

Arrays of optical antennas are very different from single antennas

-*in the near-field*: strong non-local effects (not treated here)

-*in the intermediate and far-field*: the properties of the array are dictated by its effective macroscopic properties (metamaterial regime)

Also: opportunities for superradiance.

Acknowledgments

Quynh Le Van (graduate student, fabrication, experiments)

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Benjamin Habert, (graduate student, first PL experiments)

Prof. Jean-Jacques Greffet, (theoretical and material support)

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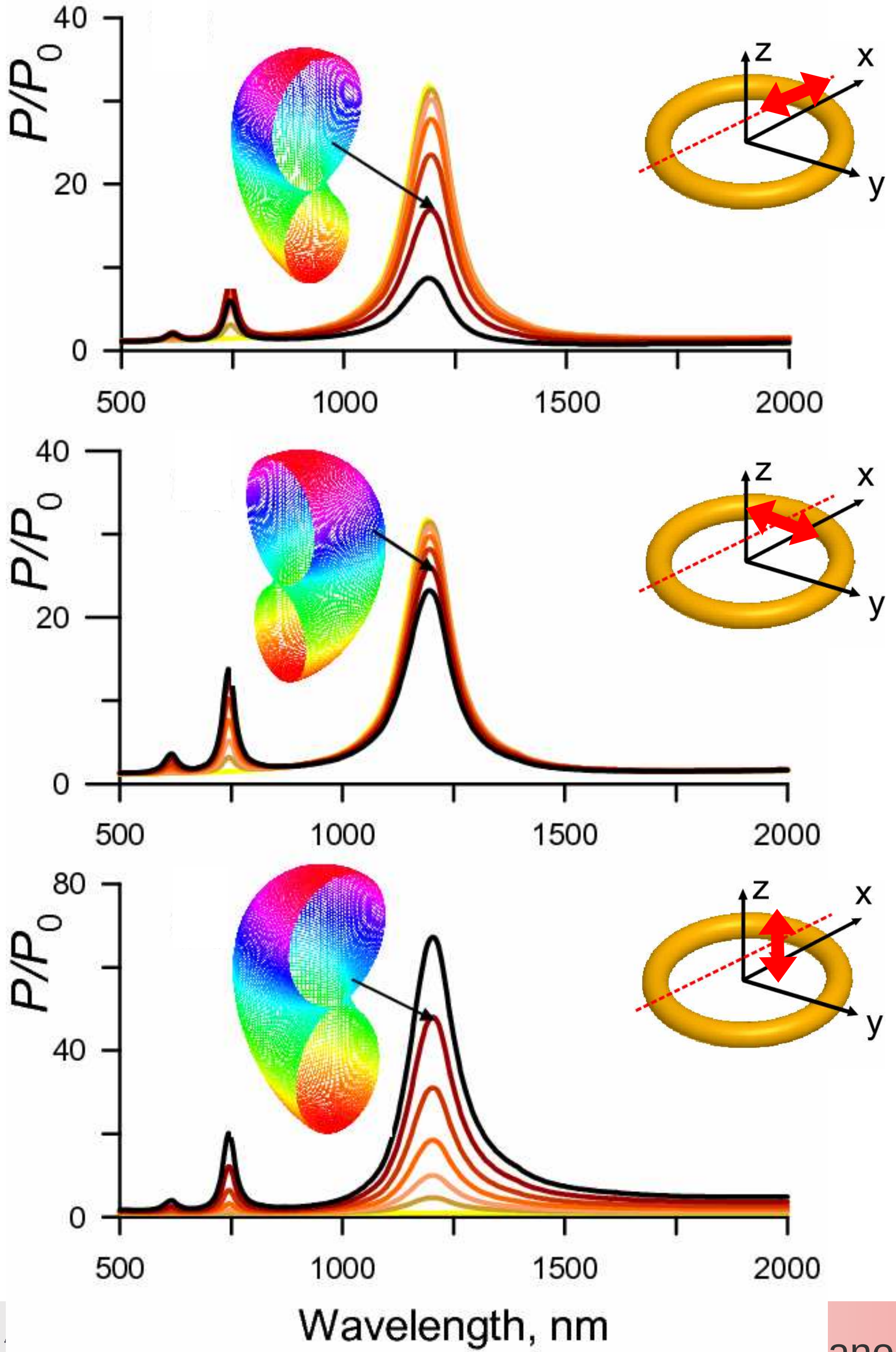
RTRA Triangle de la Physique

University Paris-Sud

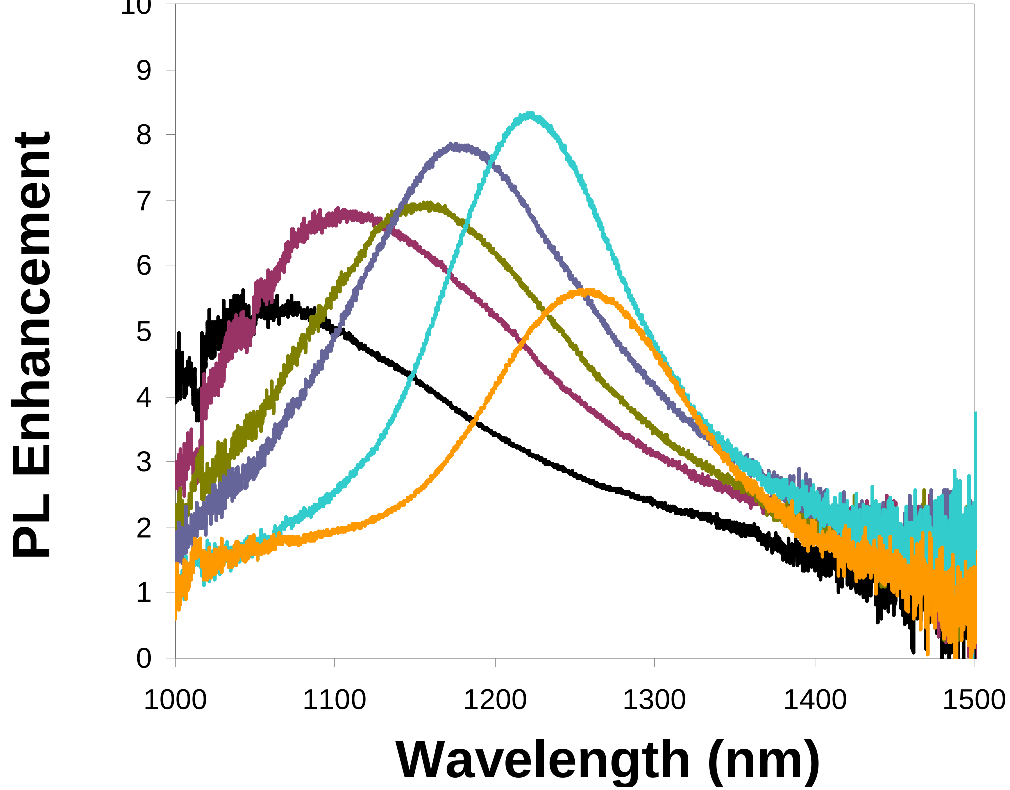
Properties of the dipolar resonance



The ring's response is independent from the position of the emitter and the orientation of its dipolar moment.

Teperik and Degiron, PRB 2011

PL Enhancement spectra



Passive transmission spectra (FTIR measurements):

Same spectral signature