CONTROLLING FLUORESCENCE EMISSION WITH NANO-PHOTONICS AND –PLASMONICS.

ANALOGIES WITH CONTROLLING BLACKBODY RADIATION ?

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PhD thesis (2012) available on http://tel.archives-ouvertes.fr

Context & Summary

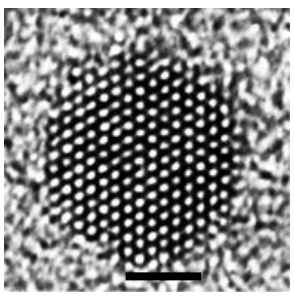
Aim of the group (INSP) **:** Achieving a higher degree of control over the spontaneous emission of fluorescence in the visible range *(Control of intensity, life-time, direction and polarization of emission).*

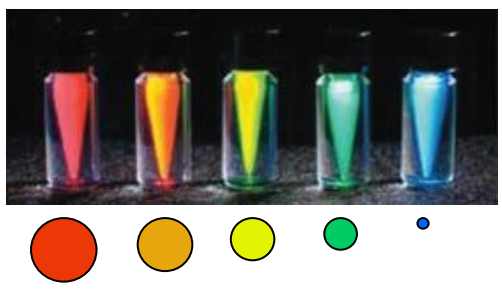
Systems studied : Semiconductor Nanocrystals coupled to photonic and plasmonic crystals. Utilization of self-assembly methods to obtain the structures.

My contributions : optical caracterisation and modelisation of opals and 2D plasmonic crystals, study of nanocrystals emission enhancement by 2D plasmonic crystals.

Semiconductor Nanocrystals

fluorescent particles (CdSe/CdS) in the visible range

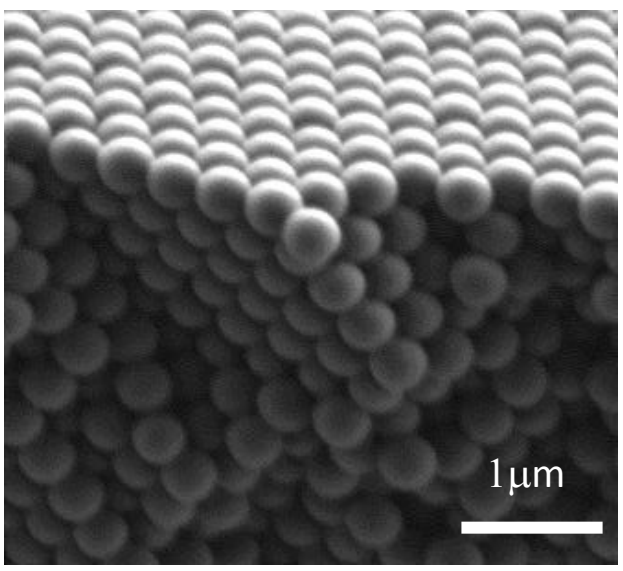




Photonic Crystals

periodically modulated dielectric material

3D

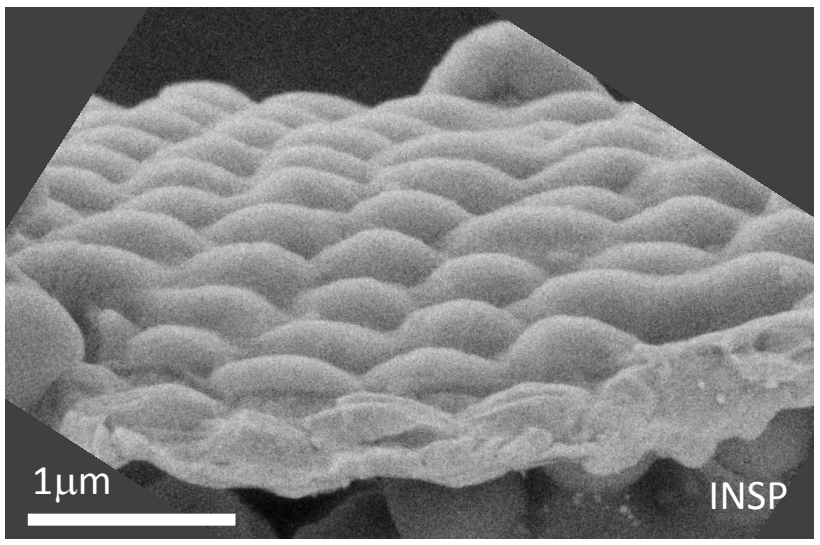


Opals

Plasmonic Crystals

peridodically modulated plasmonic surface

2D

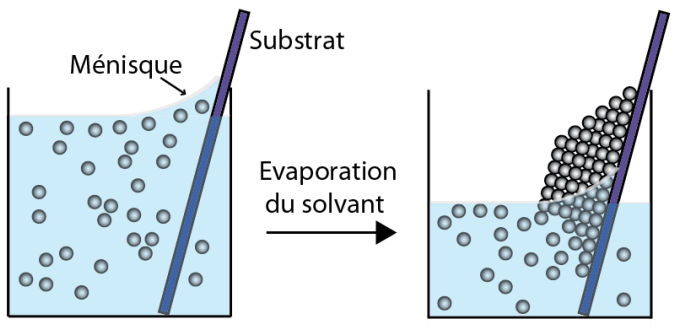


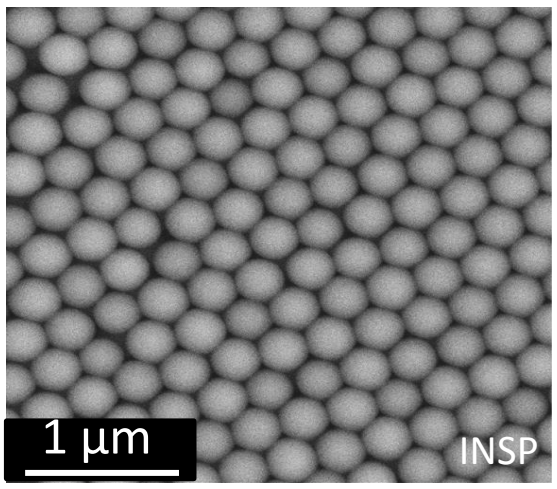
Self-assembled Gold Grating

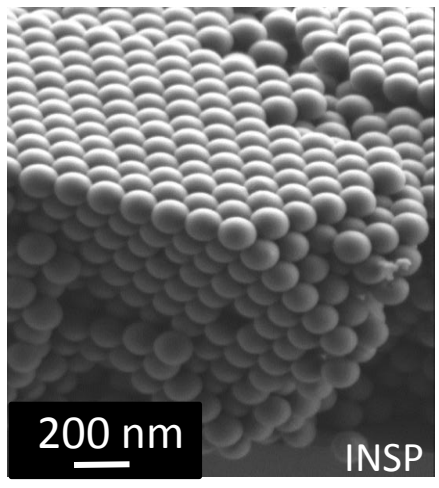
3D Self-assembled Photonic crystals

▪ Sample fabrication

Phan Ngoc Hong (INSP)



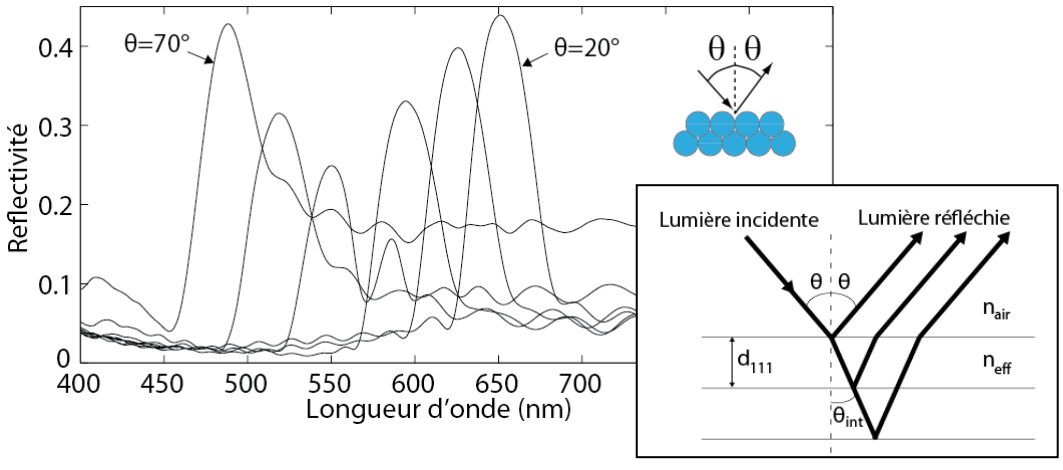




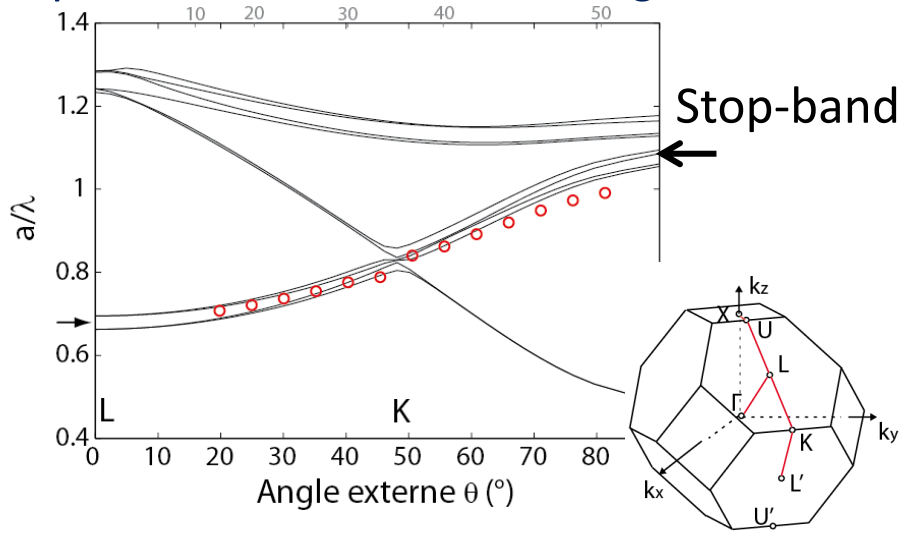
➢ **FCC structure of silica spheres** (200 to 450 nm) : obtained by soft chemistry methods. Samples of several cm² with a unique orientation of lattice.

▪ Optical properties of opals, Stop-bands.

Specular reflectivity measurement



Dispersion relation – band diagram



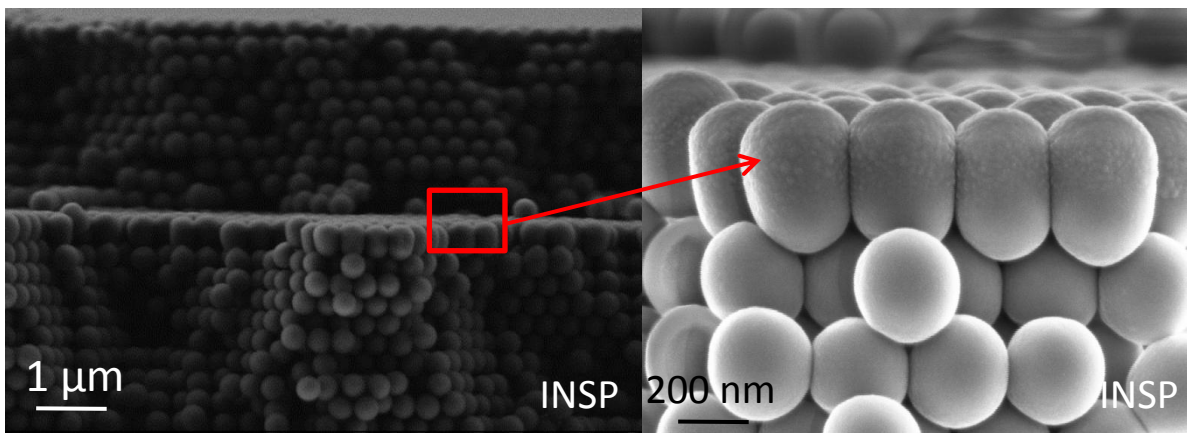
➢ **Stop bands** are created in the dispersion relation of light as a result of destructive interferences in the photonic crystal.

Avoine, PRB 86, 165432 (2012)

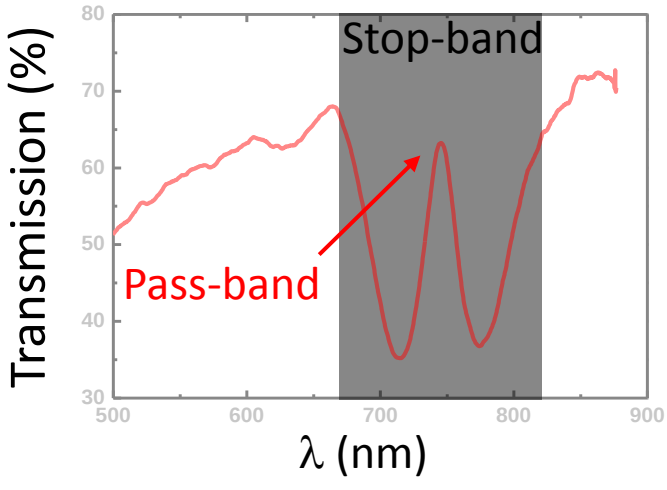
▪ Emission modification by in a defect layer acting like a photonic cavity

Phan Ngoc Hong (INSP)

Introduction of a defect layer between two opals



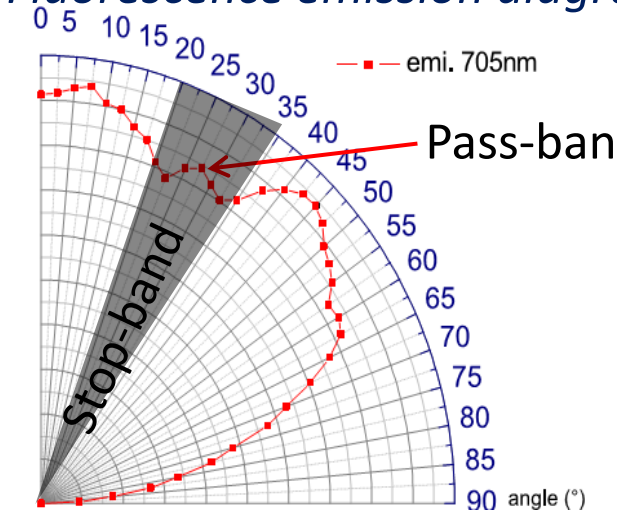
Opal with defect layer : Transmission



➢ **A cavity is created** by introducing a defect layer between two opals (sputtering of silica). **A pass-band** opens in the photonic stop-band.

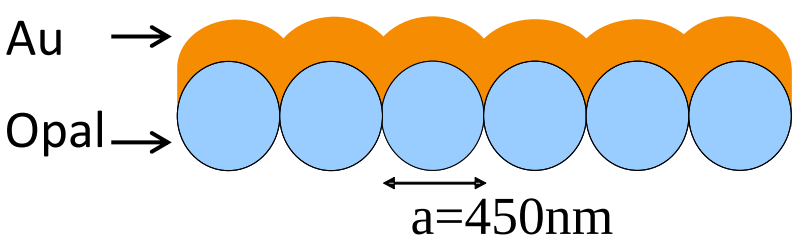
➢ **Nanocrystals deposited in the defect layer** undergo an inhibition of their fluorescence in the stop-band and an exaltation in the pass-band.

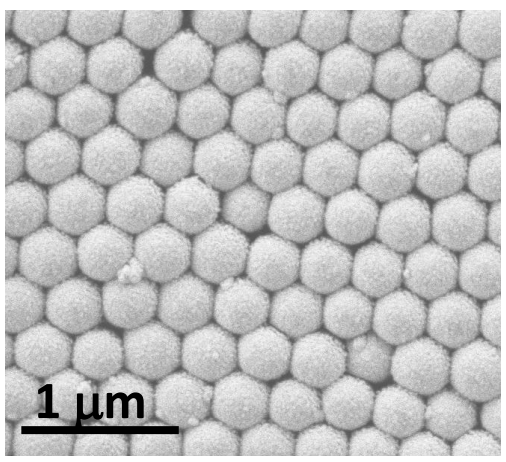
Fluorescence emission diagram

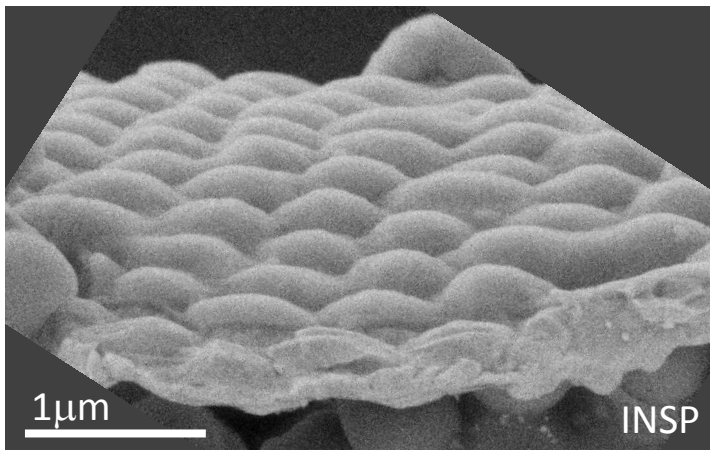


Surface Plasmon Polaritons-mediated fluorescence on a gold plasmonic crystal

▪ Sample fabrication

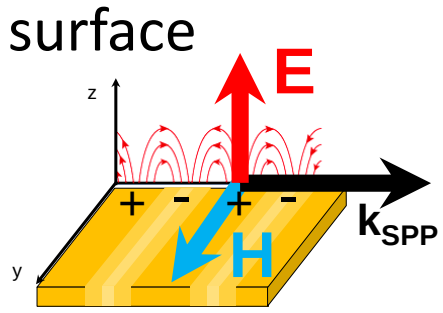






➢ **Surface Plasmon Polaritons (SPP) :** propagation on the gold surface

➢ **Gold 2D grating :** period close to λ_{SPP}



Surface Plasmon Polaritons (p-polarized on a planar surface)

➢ **Self-assembly method** : large area of structured surface (several cm^2)

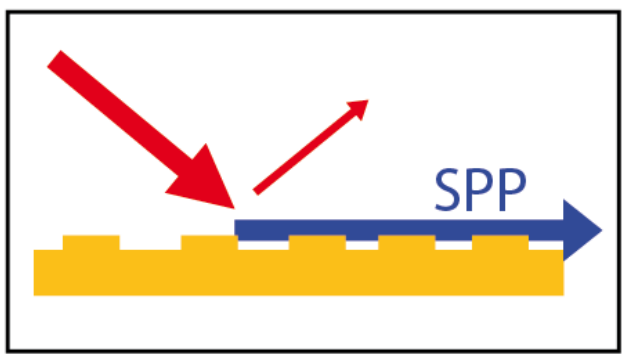
▪ Grating induced Light – SPP coupling

➢ **SPP scattering** by the grating (Introduction of a periodicity in the SPP dispersion relation)

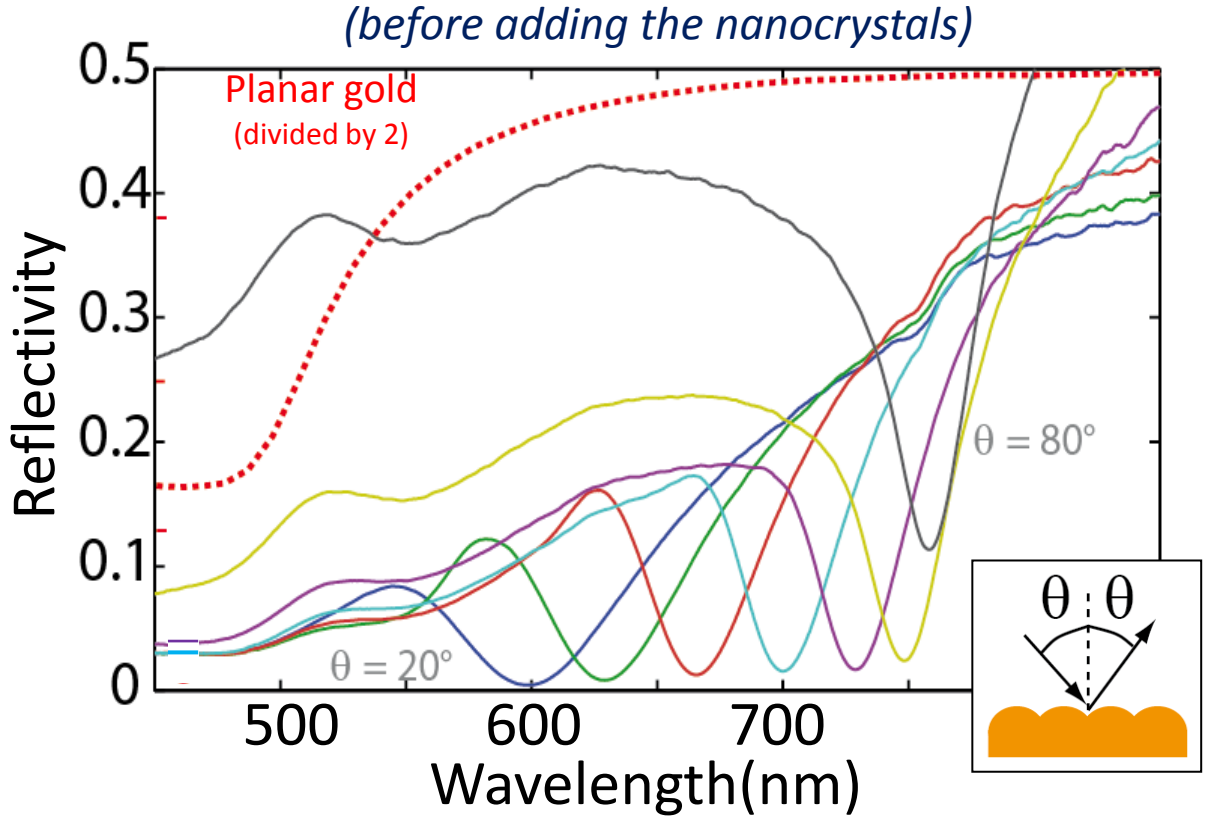
➢ **Interaction with light:** intense coupling (zero-reflectivity) observed between 600 and 700 nm)

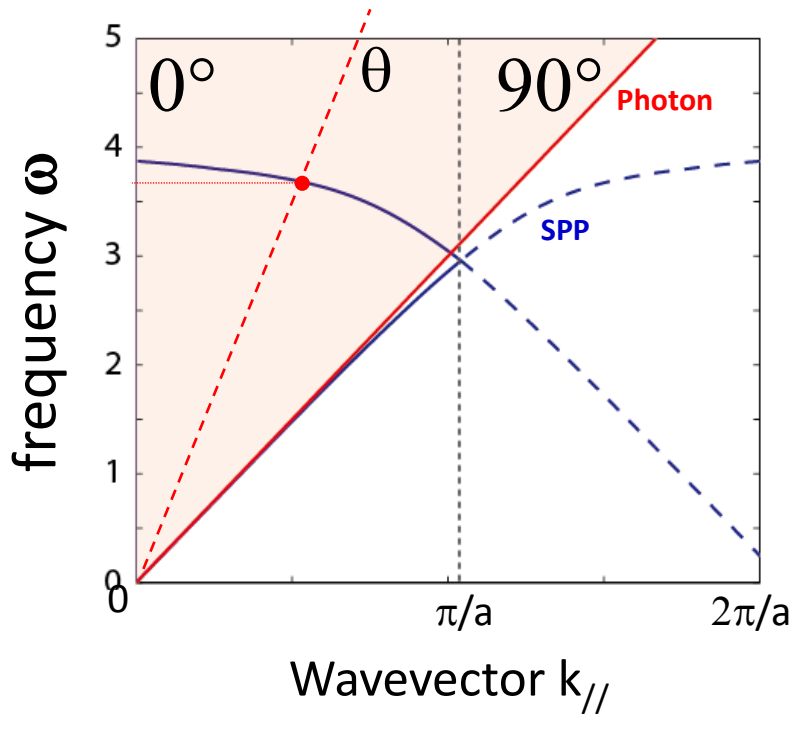
➢ **Broadband coupling :** mostly due to self-assembly

Principle of SPP-light couling by a grating



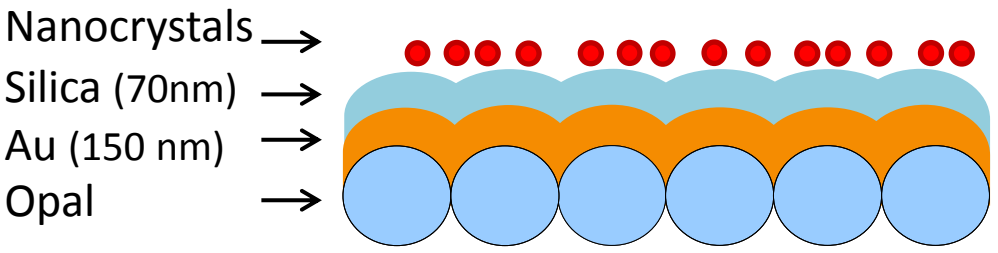
Specular reflectivity in polarization p
(before adding the nanocrystals)





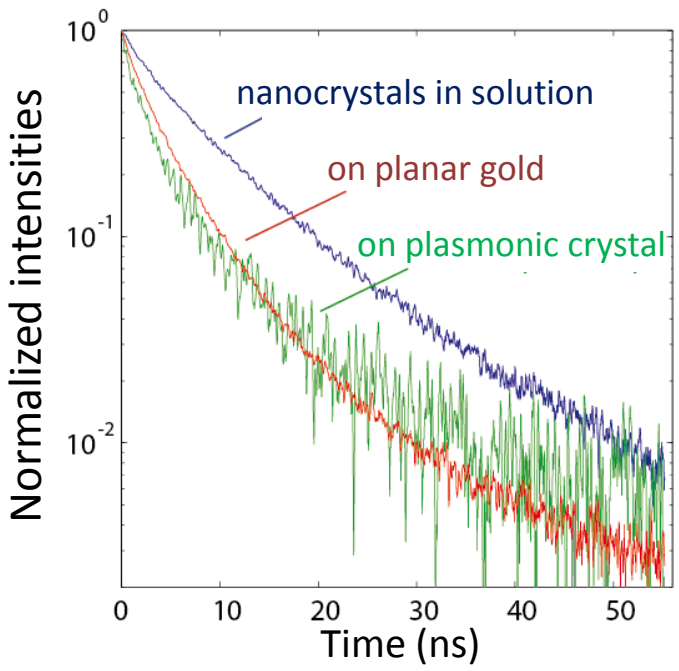
Frederich et al., Optics express, 19, 24, (2011)

▪ Near-field coupling of nanocrystals to SPP



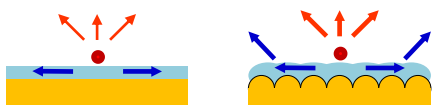
Nanocrystals: B.Dubertret, ESPCI, Paris

Fluorescence intensity decay



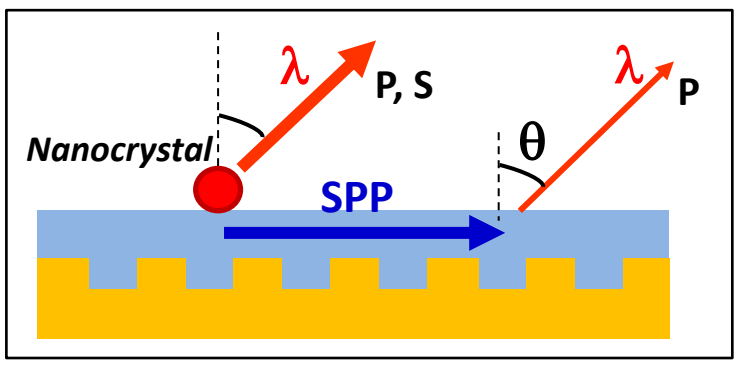
➢ **Nanocrystals emit SPP in near field.** Deposited in the vicinity of the gold surface, NCs can decay in SPP chanel.

$$\Gamma = \Gamma_{rad} + \Gamma_{SPP}$$



▪ SPP-mediated fluorescence enhancement

Principle of SPP grating re-emission

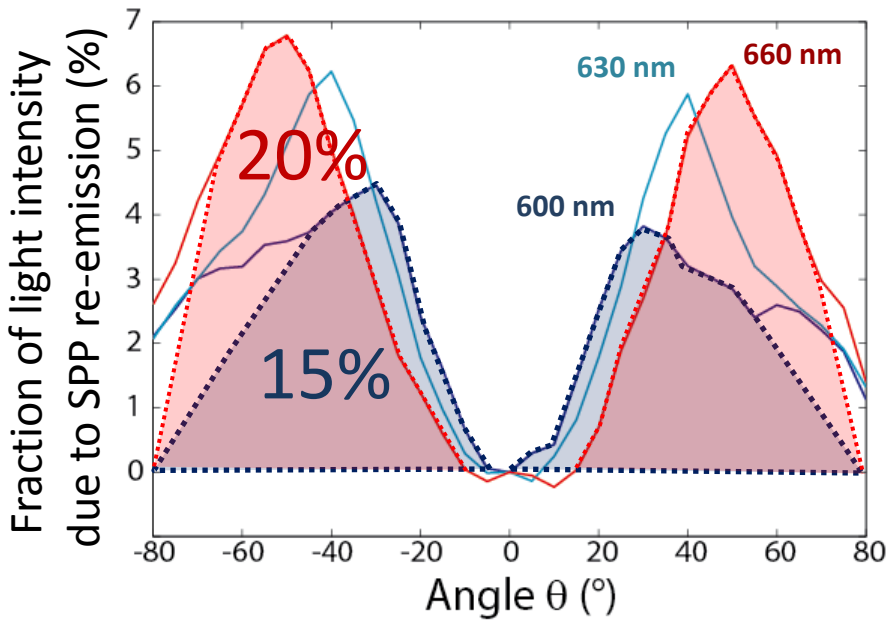


➢ **SPP are partially re-emitted in far field** by the grating (in p-polarization).

1) *Polarization- and angle-resolved spectroscopy of the emission is performed.*
2) *The **polarization ratio** shows an angle- and wavelength-dependent surplus of p-polarized emission…*
3) *…that follows the SPP dispersion relation measured by reflectivity :*
signature of SPP re-emission.

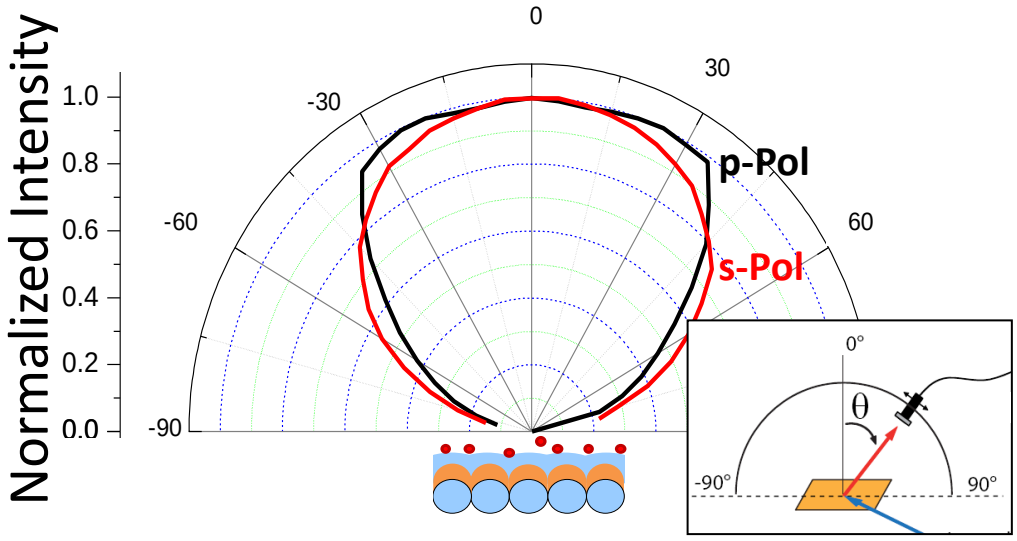
Quantifying SPP re-emission

by comparing emission diagrams with a planar reference

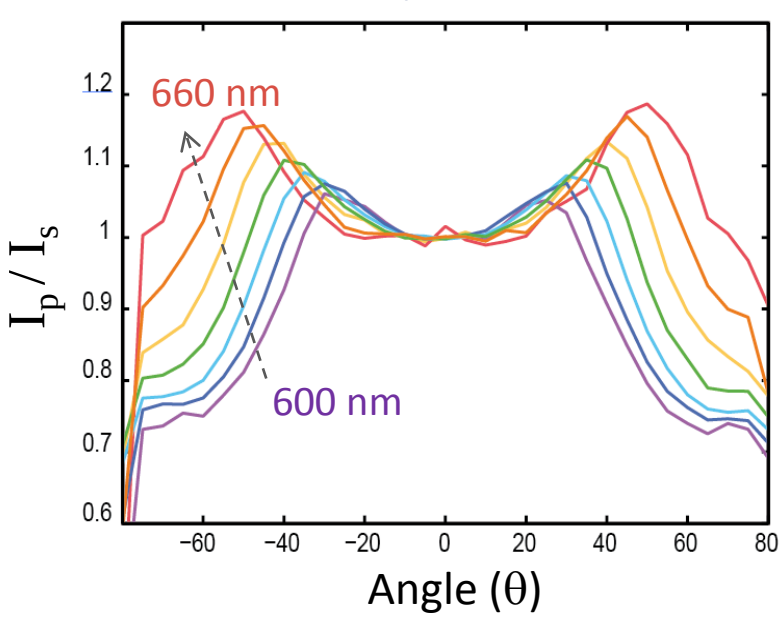


Frederich et al., Determination of the SPP extraction efficiency from a self-assembled plasmonic crystal, TO BE PUBLISHED IN 2013

1) Polarized emission diagrams



2) Fluorescence polarization ratio



3) Dispersion relation