

# Controlling photon emitters on nanometer & femtosecond scale

**Niek van Hulst**



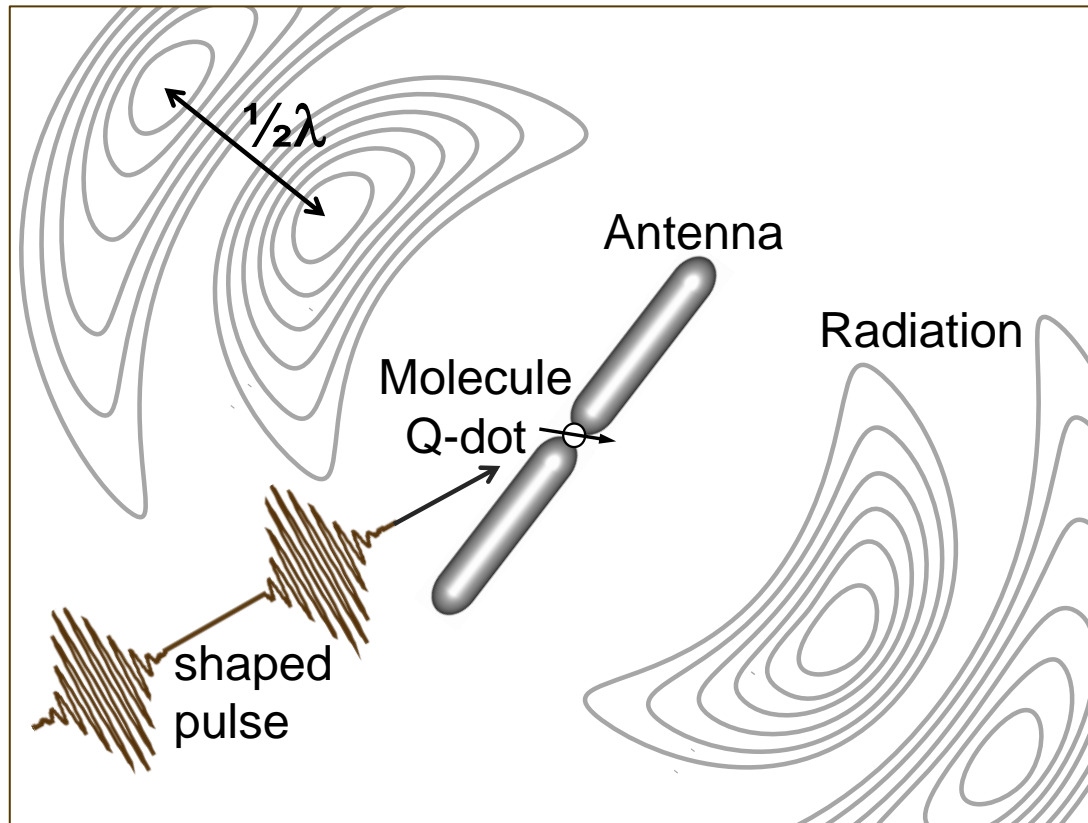
**NanoICT**  
**Donostia – San Sebastian 28 Oct 2009**

**ICFO**

Institute of Photonic Sciences  
Mediterranean Technology Park  
**Barcelona - Spain**

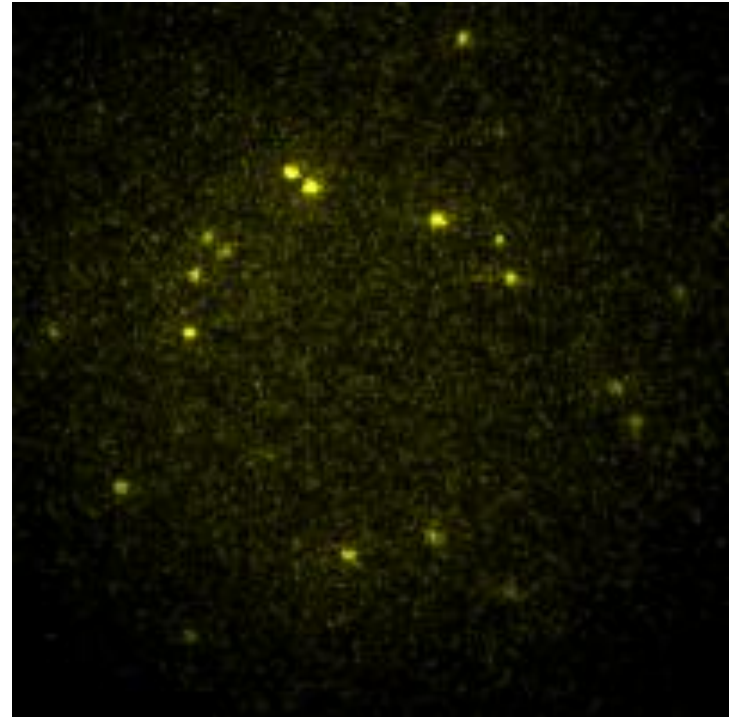
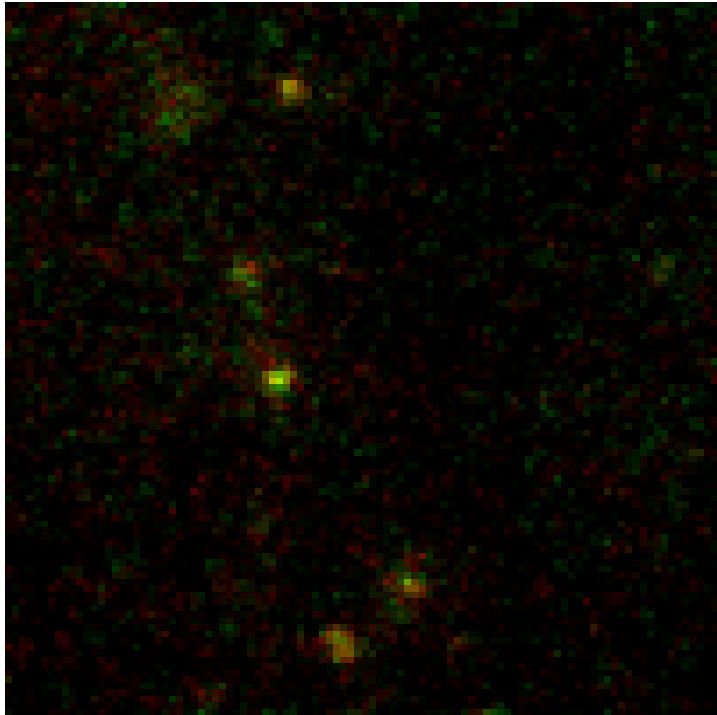
# NanoPhotonics: where “fs” and “nm” meet

[fs] phase controlled excitation of single molecules  
“Single Molecule Coherent Control”

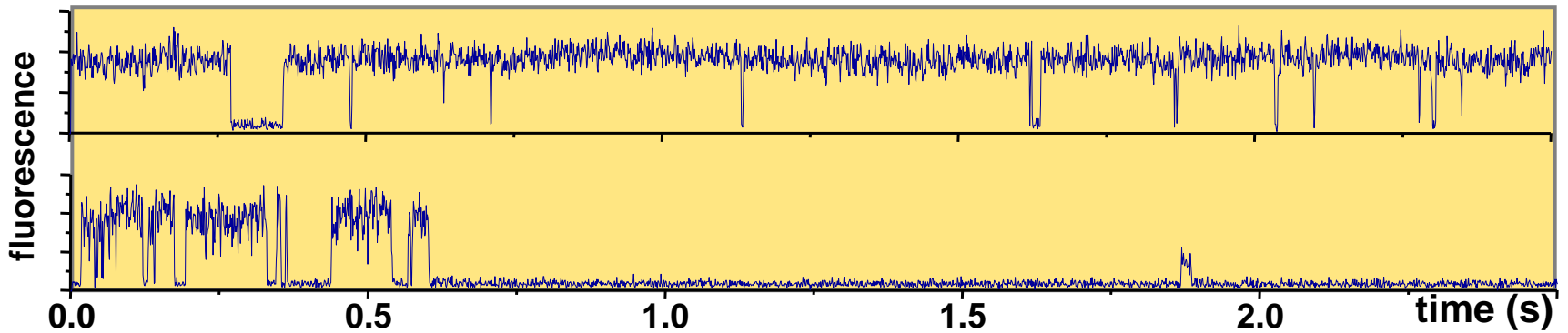


[nm] phase controlled driving of a nano-antenna  
“Coherent control of antennas”

# Detection of single molecule dynamics



30 x 30  $\mu\text{m}^2$ , 100 ms/frame



Diffusion, rotation, blinking, bleaching: hours, sec, ms,  $\mu\text{s}$ .....

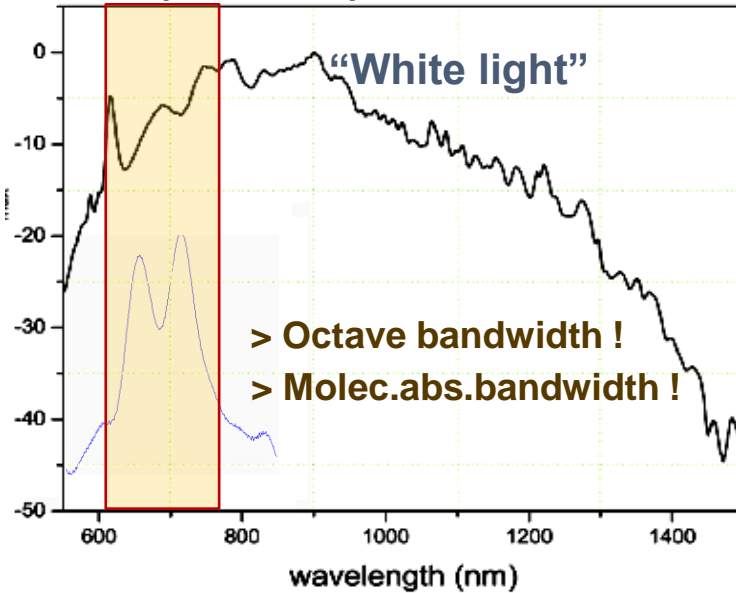
# Tools for ultrafast (fs) phase controlled excitation

Ultrafast laser  
7 fs pulse length

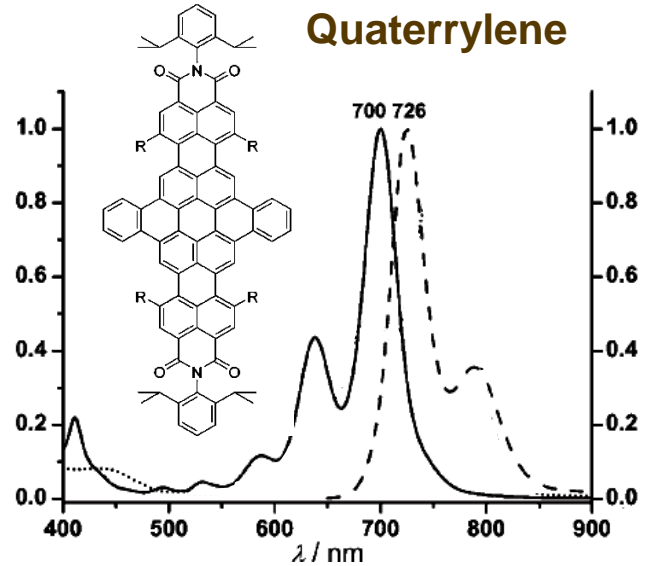


Menlo systems

Broad (coherent) excitation bandwidth

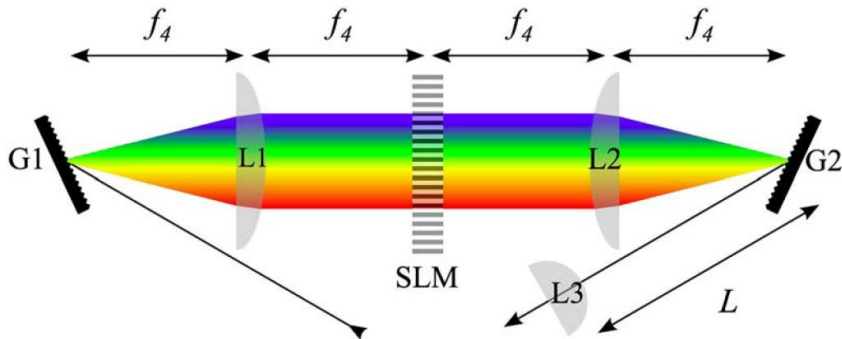


Molecule  
Quaterrylene



Yuri Avlasevich, Klaus Mullen,  
*Chem. Eur. J.* **13**, 6555 – 6561 (2007).

Pulse shaper



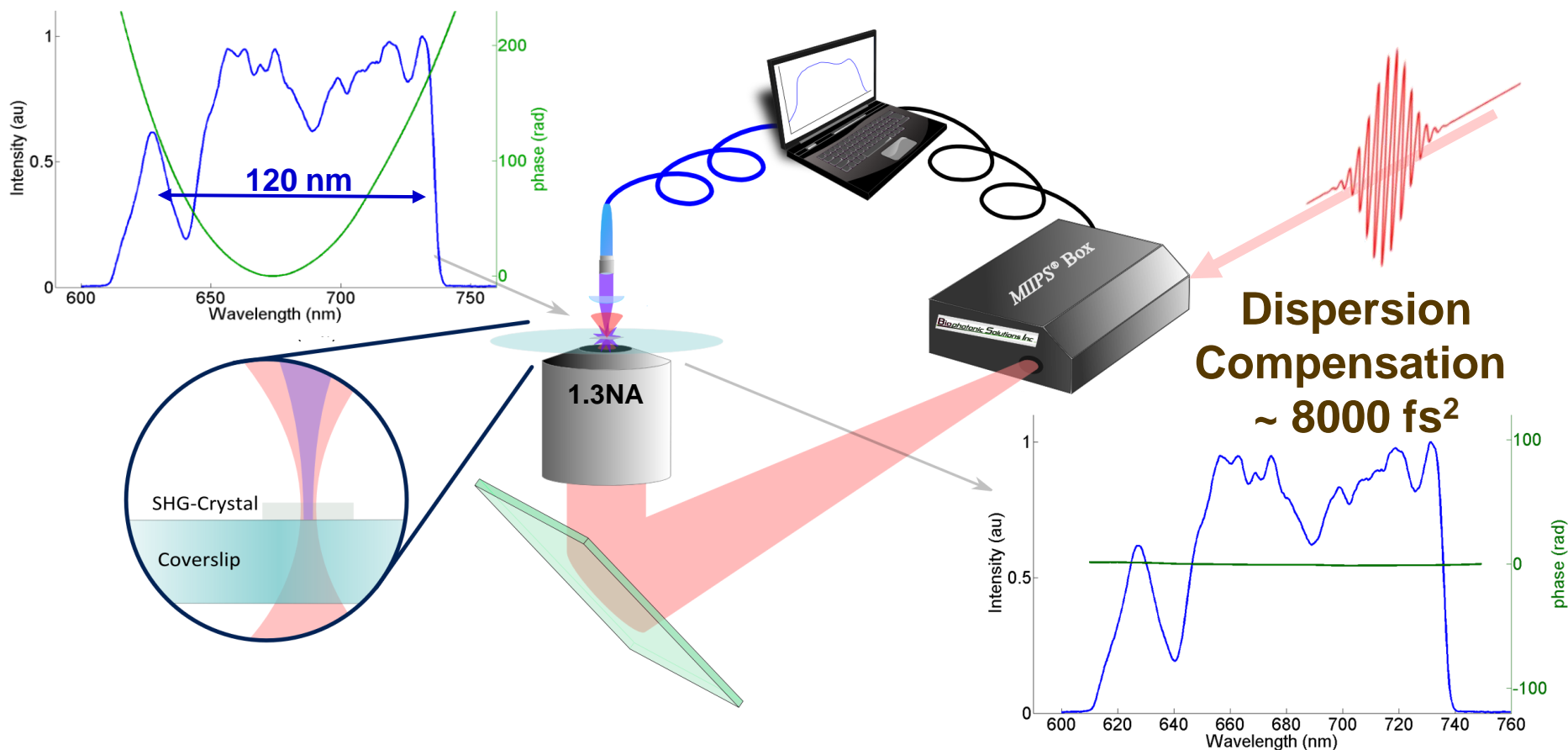
Single Molecule  
microscope



# In-focus spectral phase control and compensation

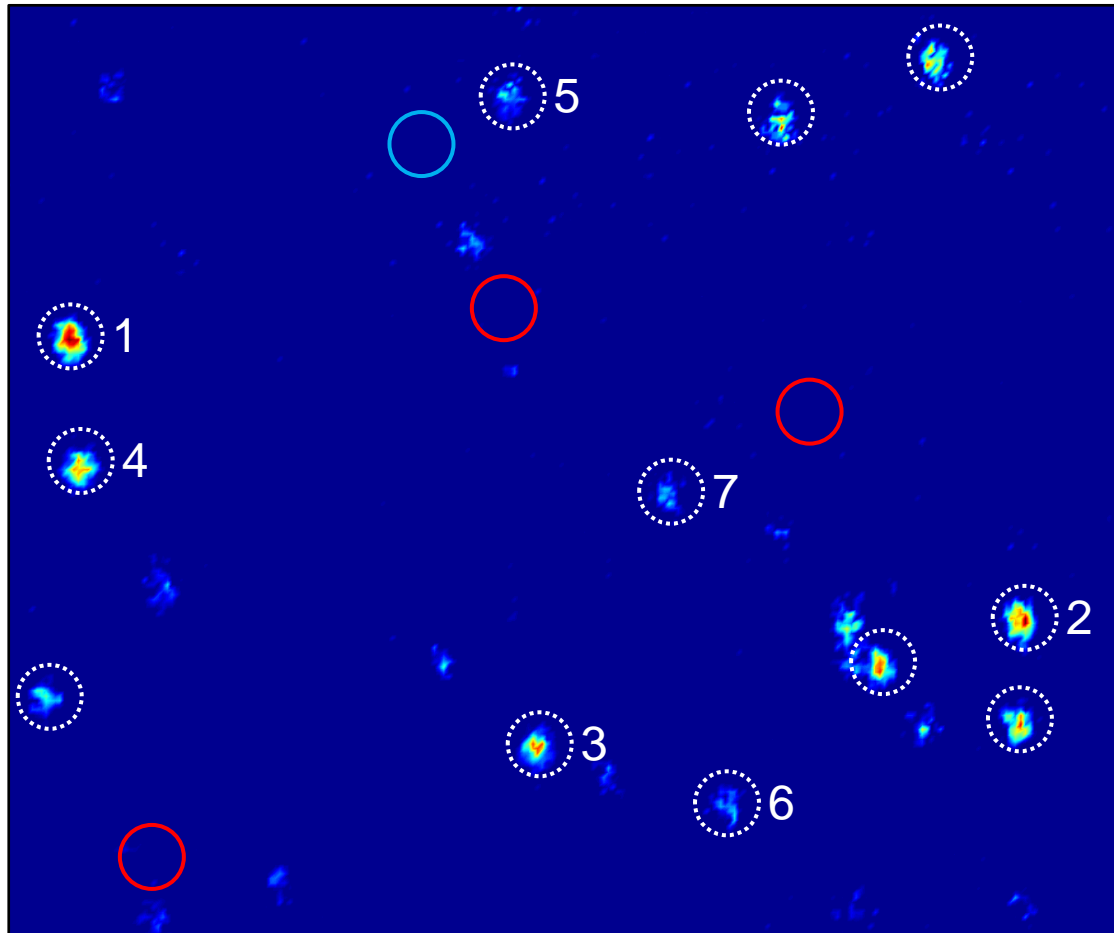
Broad band excitation in visible: 610 – 730 nm, 14 fs

Diffraction limited, 1.3NA objective

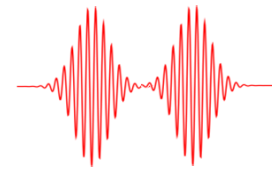


**MIIPS - Multiphoton Intrapulse Interference Phase Scan**

# Beyond the ensemble



Delay  
21 fs



**bleaching**

**blinking**

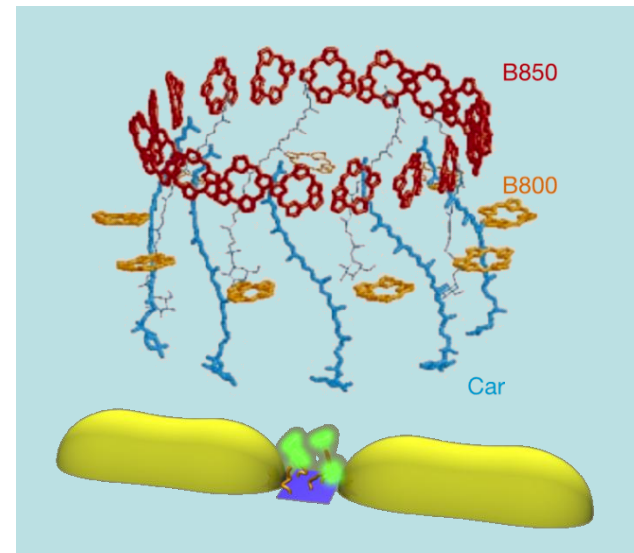
**heterogeneity**

**3  $\mu\text{m}$**

- **First phase controlled excitation of a Single Molecule**
- **At room temperature, on large molecules**
- **Single Molecular vibrational wavepacket, ~ 35 fs period**
- **Single Molecular wavepacket group velocity**
- **Starting phase differs between molecules: heterogeneity**
- **Decoherence time 20 - 50 fs**

## Outlook

- **Multiphoton excitation**
- **Coupled systems, energy transfer**
- **Biosystems....**



# What's NANO about antennas ?

Near-field, sub- $\lambda$   $\leftarrow \lambda \rightarrow$  Far field, many  $\lambda$

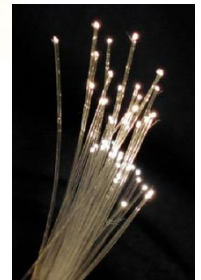
Optics, visible  
500 THz ( $10^{14}$  Hz)  
 $\lambda = 400 - 800$  nm  
period  $\sim 2$  fs

Optics on nanoscale  
atoms, molecules  $\ll \lambda$

Lenses, mirrors, fibers,...

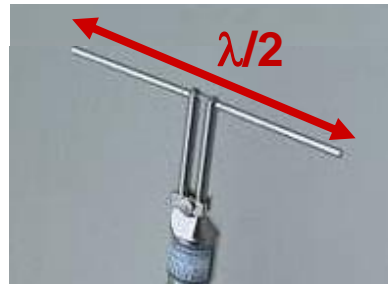
$\gg \lambda$

**¿ Optical NanoAntennas ?**



$10^8 \times$

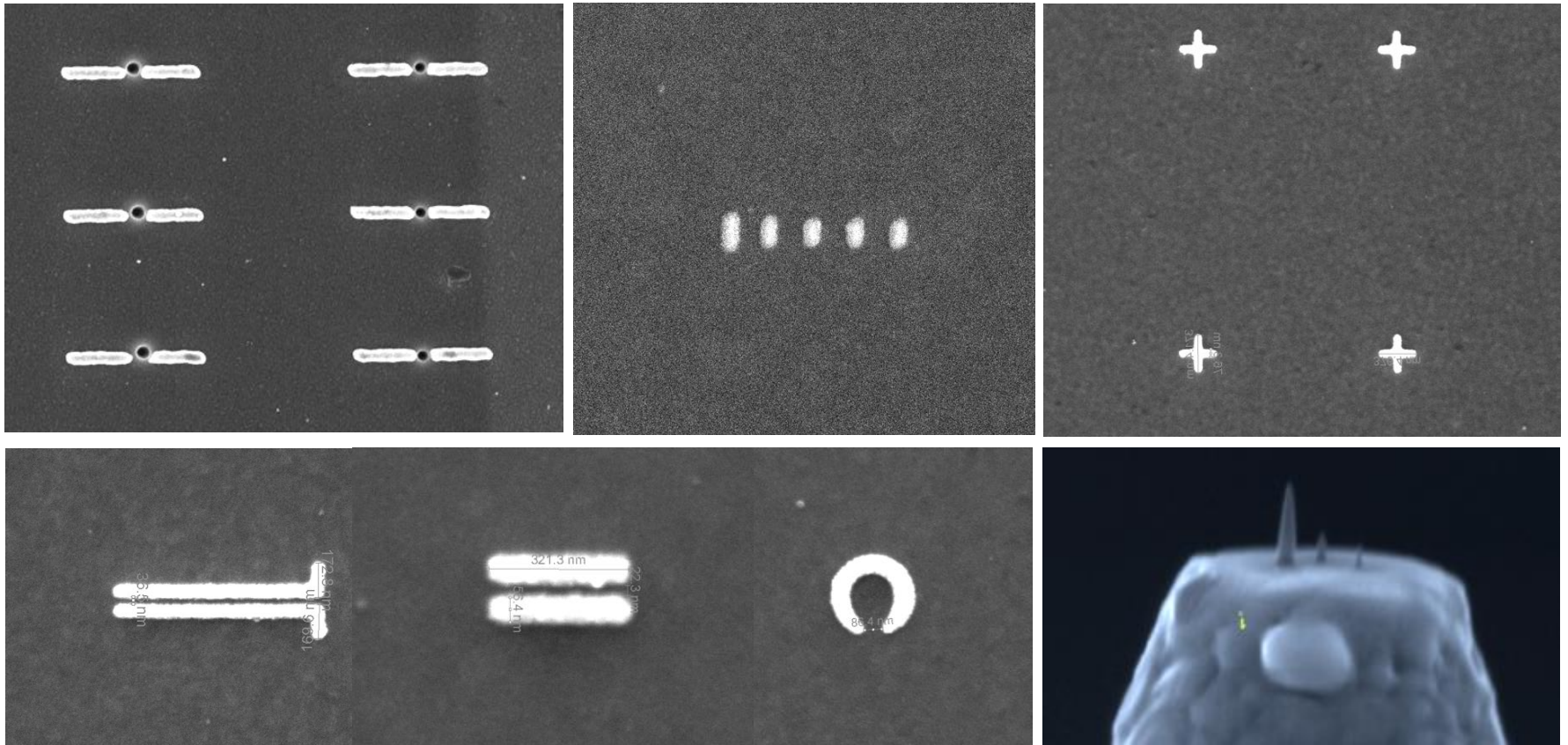
Radio frequency  
kHz-MHz  
 $\lambda = 1$  m @ 300MHz



Transducer near – far field

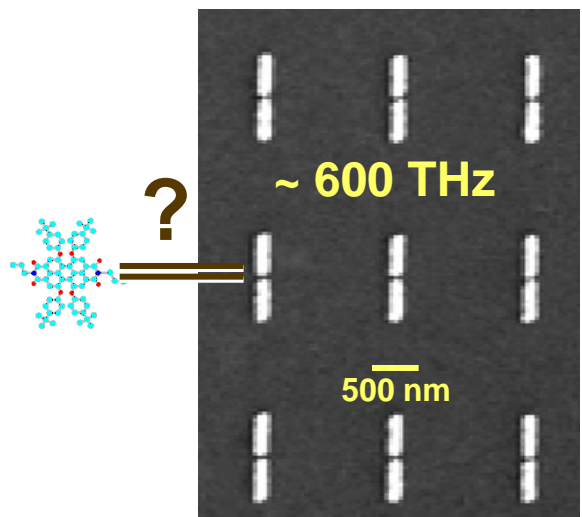
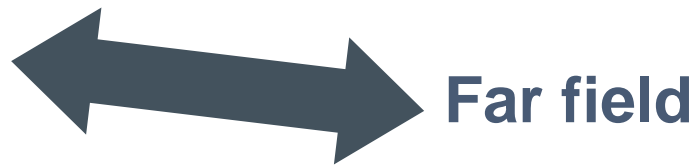
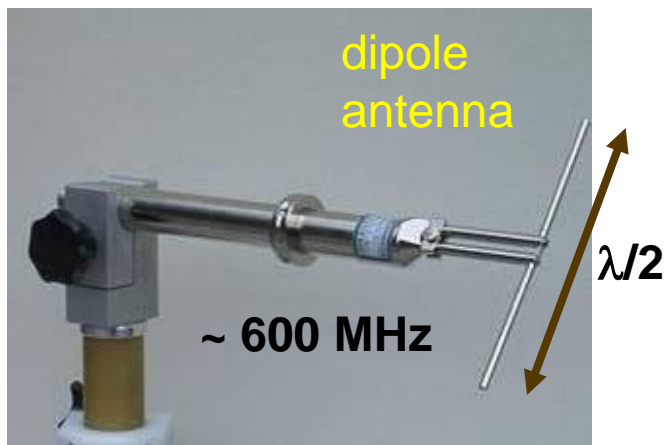


# Optical Nano-antennas

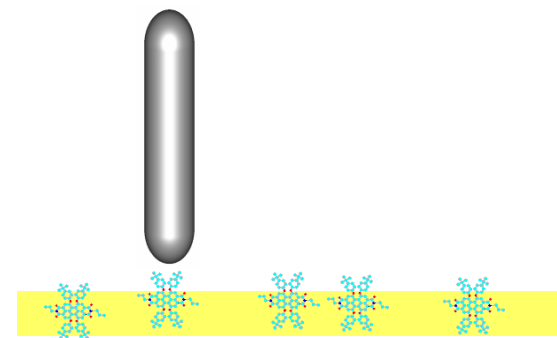
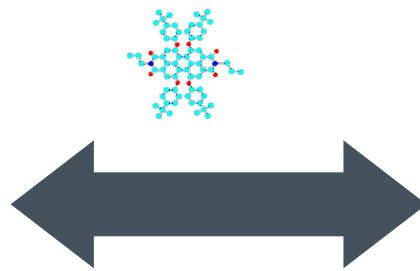


**E-beam lithography, focussed ion beam milling**

# Driving Antennas at optical frequencies ?

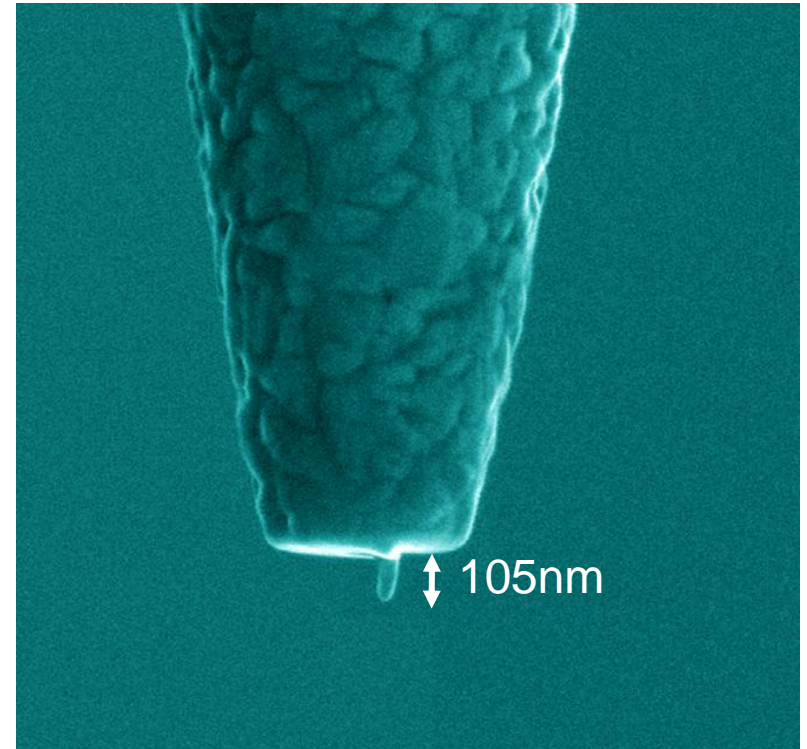
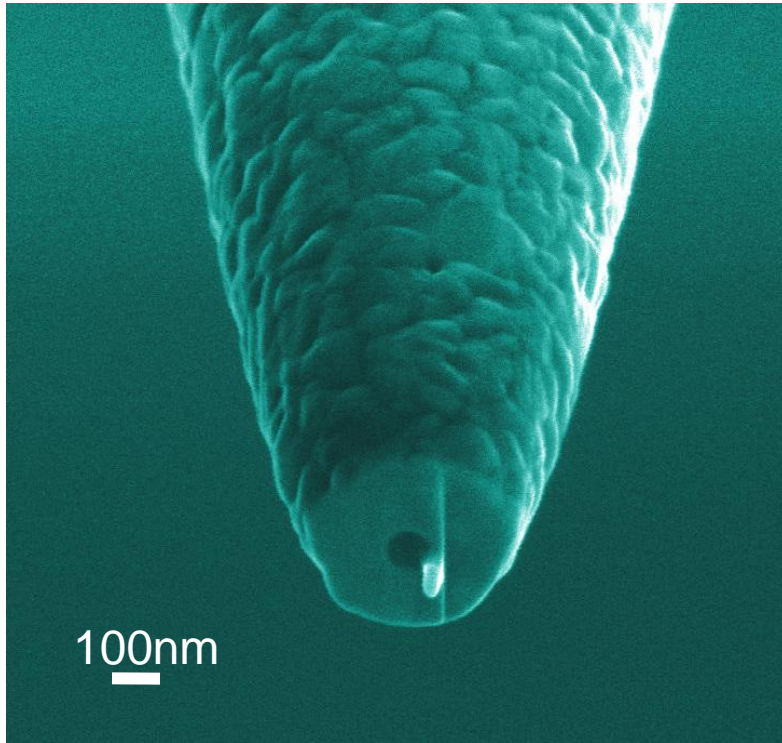


Nano-antennas  
on surface

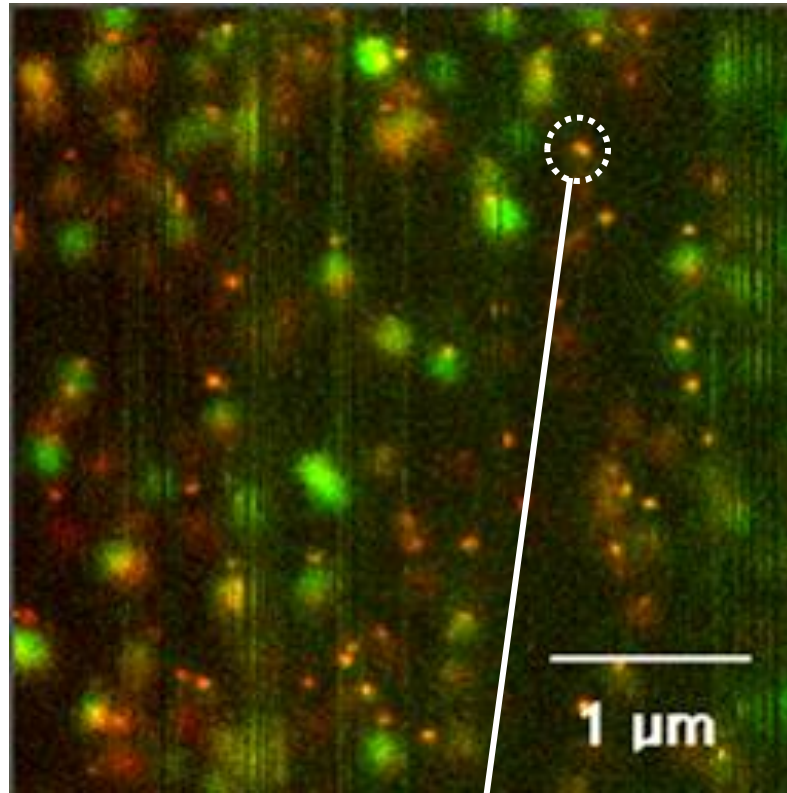
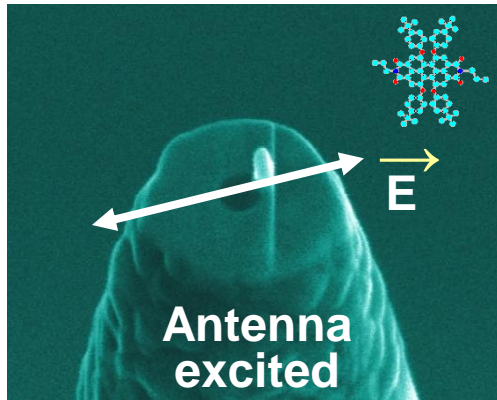
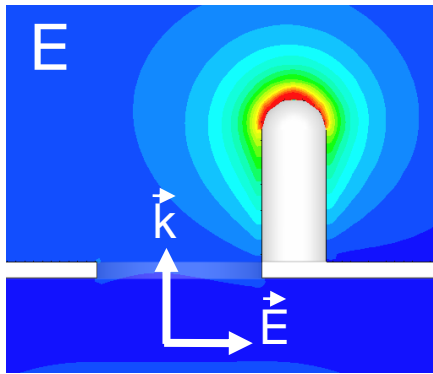


Antenna probe

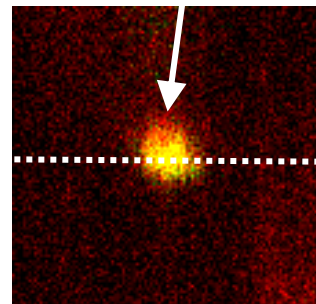
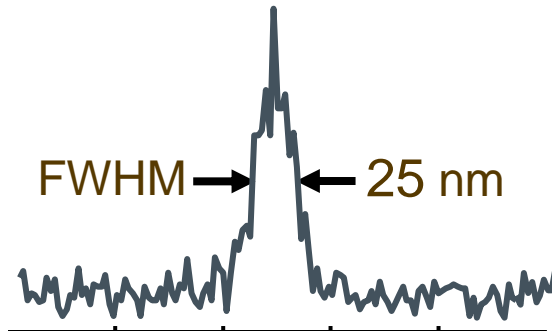
# A resonant nano-antenna light source



# The antenna field is localized within 25 nm

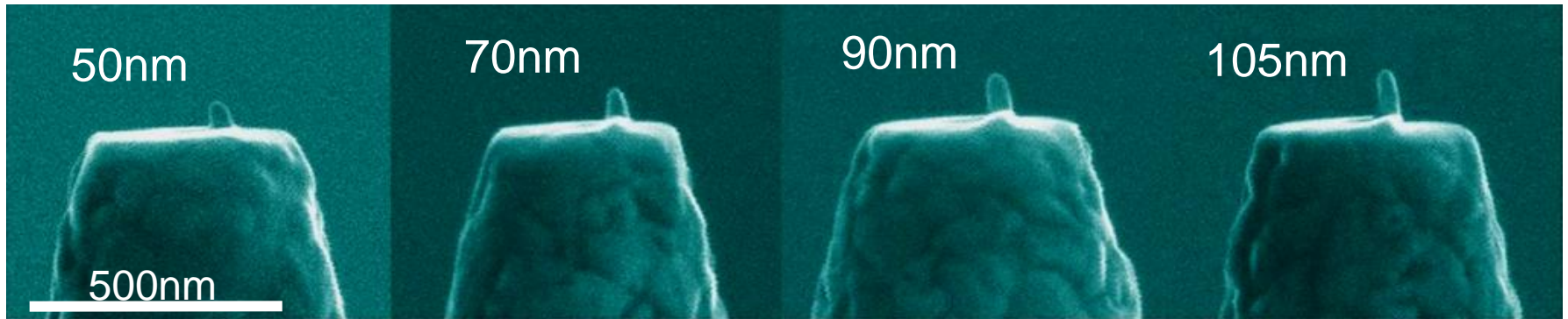


single  
fluorescent  
molecules

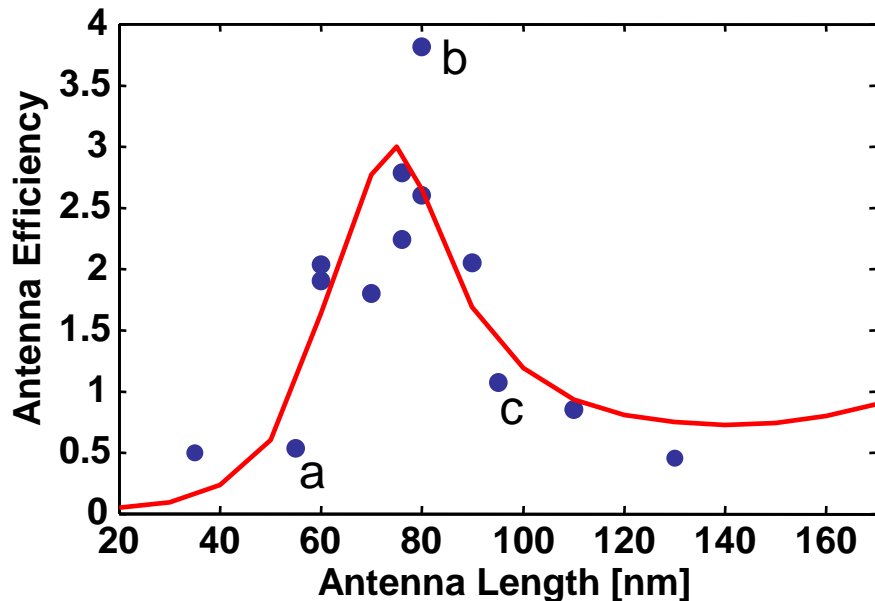


**~10% of  
diffraction limit**

# Tweaking the antenna resonance



**Fabricated antenna lengths 30 to 170 nm, radius ~20 nm**



**Antenna “Efficiency”:**

Antenna signal / Aperture signal

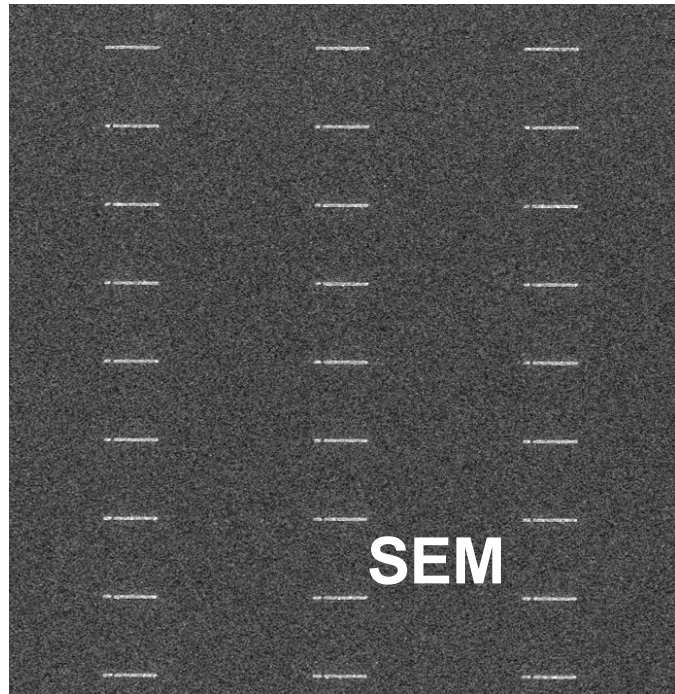
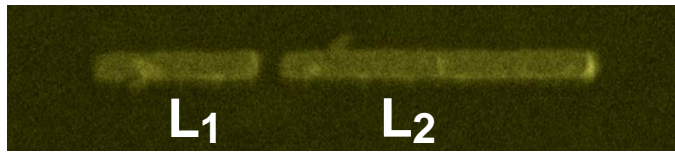
**Monopole resonance for**

**~ 80 nm length Al ( $\lambda=514$  nm)**

**Limited quality factor ~4**

**Sharper antennas ??**

# $\lambda/2$ dipole resonance and antenna length



Au on glass, 50 nm height, 50 nm width

$L_1$

0 nm

100 nm

110 nm

120 nm

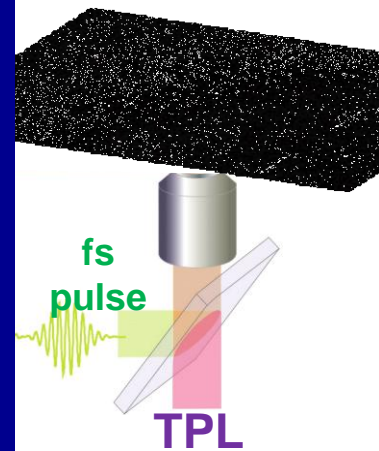
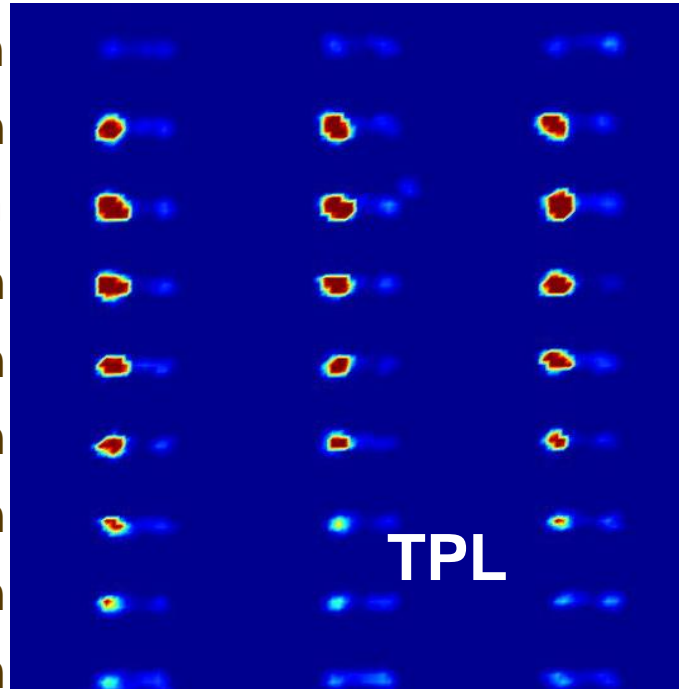
130 nm

140 nm

150 nm

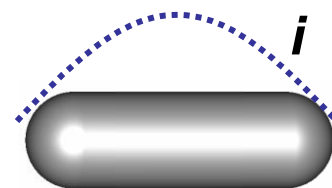
160 nm

170 nm

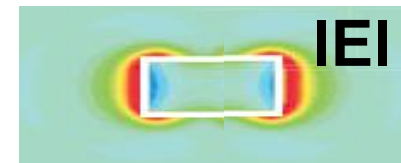


$10 \mu\text{m}$

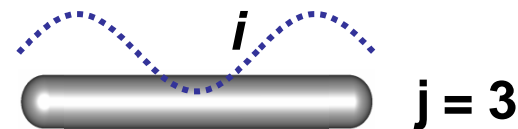
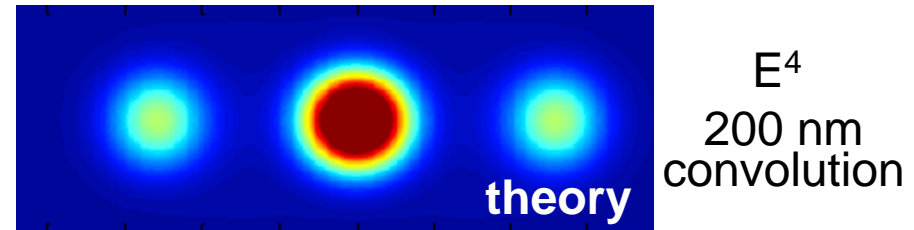
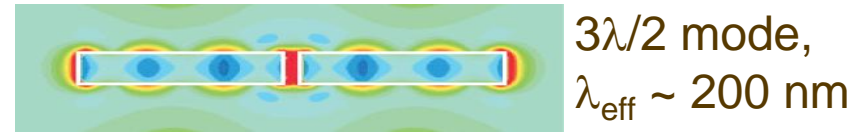
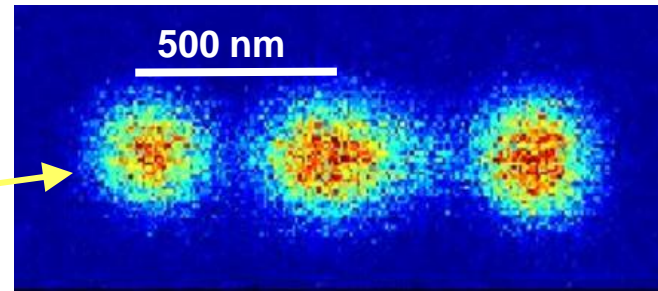
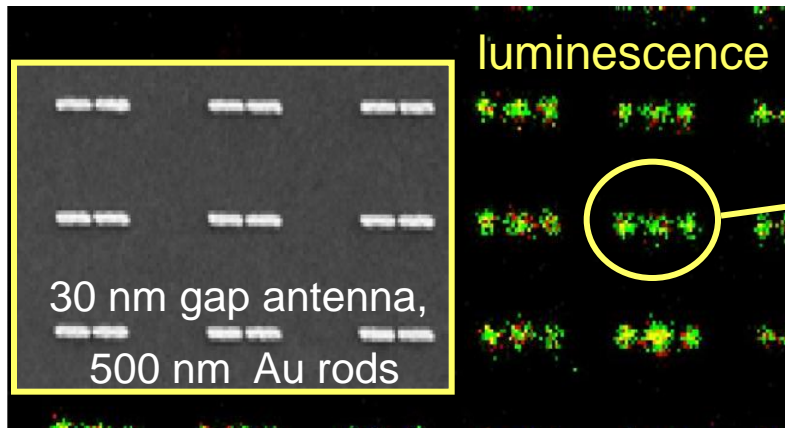
$\lambda/2$  dipole resonance for Au bar of  $L = 110 - 120 \text{ nm}$ , at  $\langle \lambda \rangle = 750 \text{ nm}$ , i.e.  $\lambda_{\text{eff}} \sim 225 \text{ nm}$



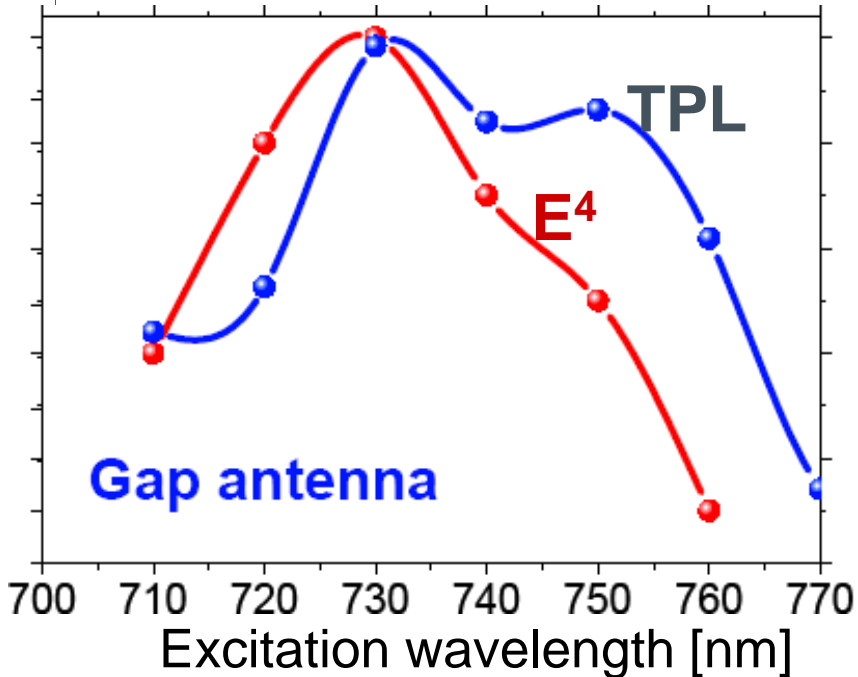
$j = 1$



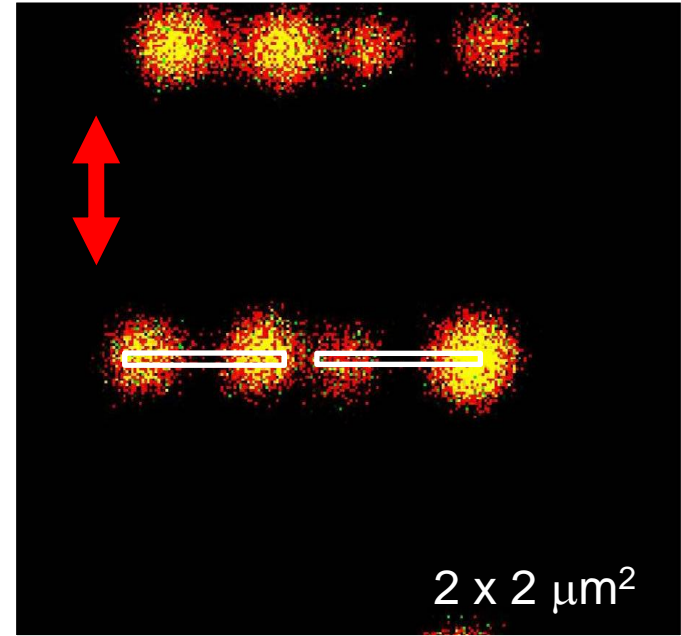
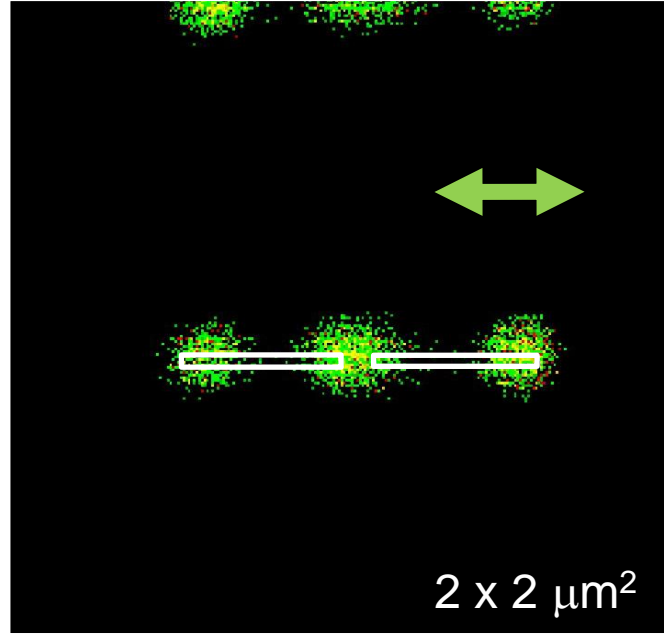
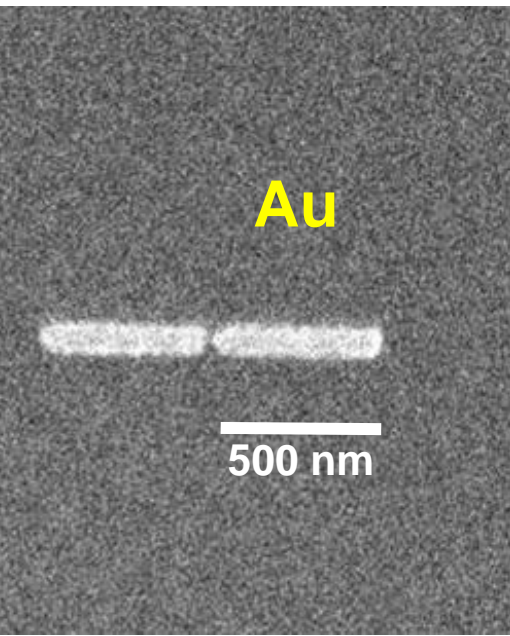
# Multipole and gap resonances of symmetric antenna



$3/2 \lambda$  resonance for Au bar of  
 $L = 500$  nm, at  $\lambda = 730$  nm

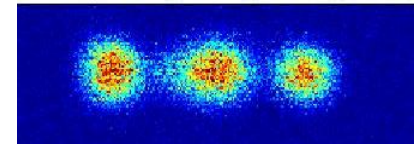
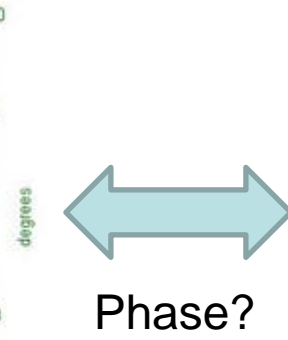
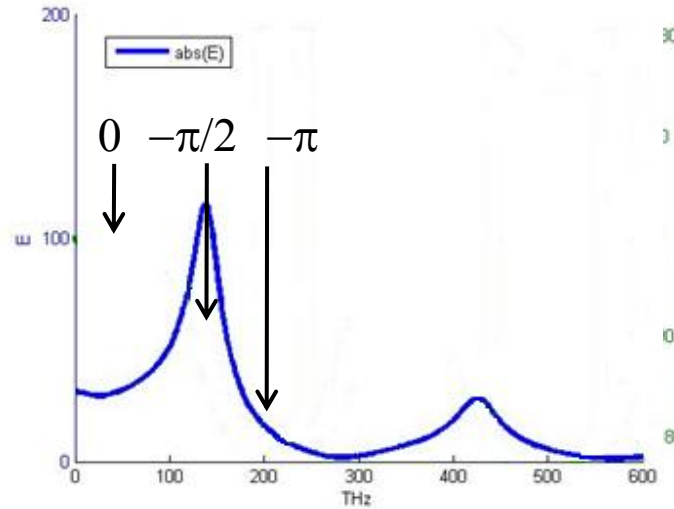
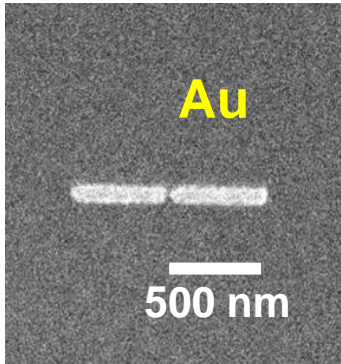


# The effect of polarization



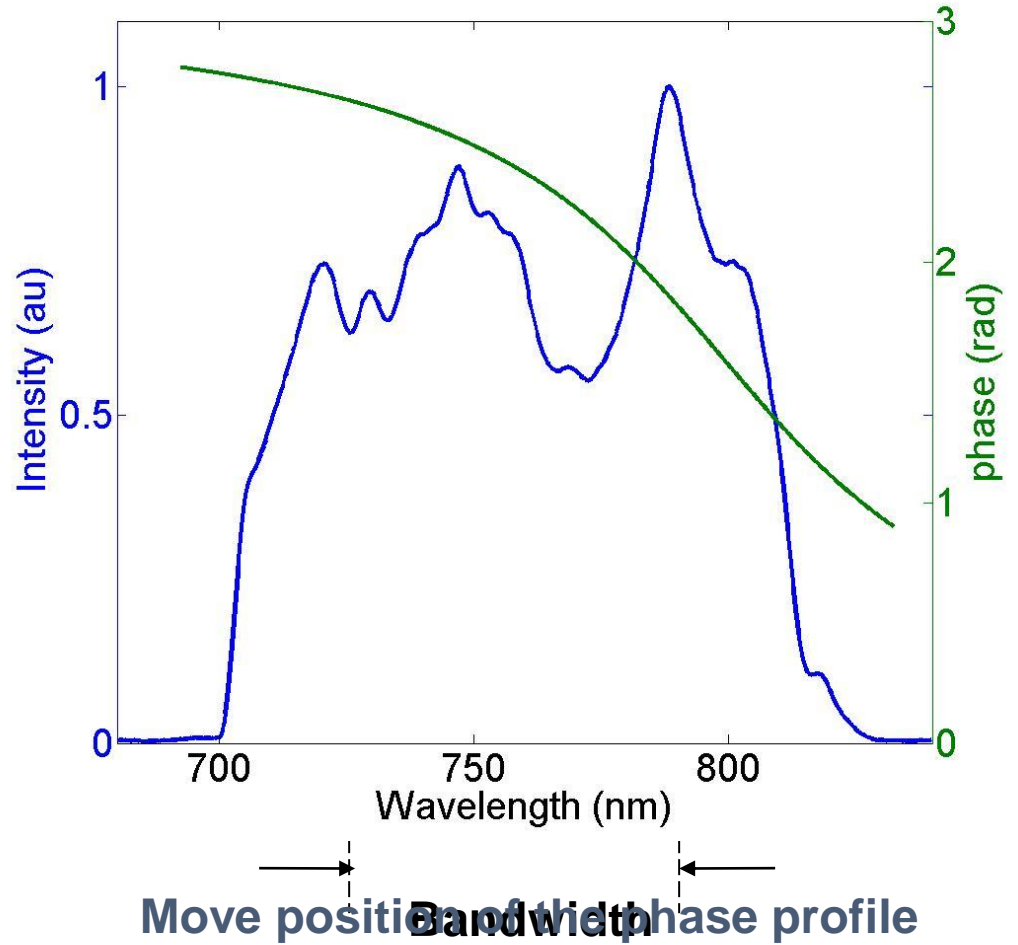
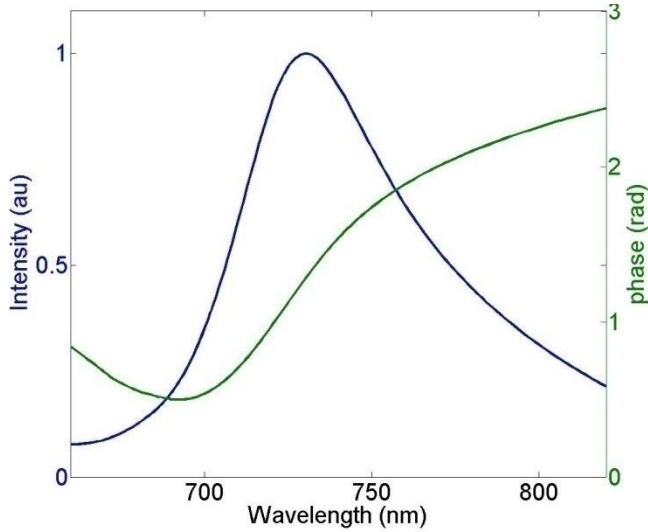
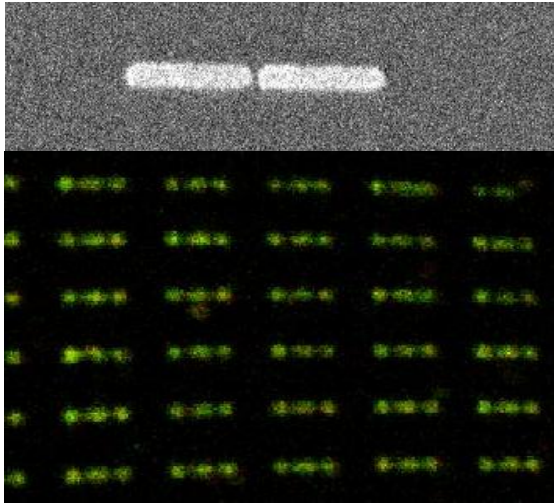


# The phase of the antenna resonance




**Phase controlled driving of nanoantenna?**  
**Spatial response?**

# Broad band excitation



- 
- **Pulse phase compensates antenna dispersion**
  - **Shifting phase profile maps antenna resonance**
  - **Phase controlled switching of antenna hotspots**

## **Outlook**

- **Antenna coupled to single quantum system**
  - **Controlled local excitation of molecules**
  - **Control of single photon emission (direction, decay time)**
- 

# Players at ICFO



Fernando Stefani



Daan Brinks



Tim Taminiau



Lars Neumann



Alberto Gonzalez-Curto



Martin Kuttge



Dominique Heinis



Florian Kulzer



Richard Hildner



Marta Castro-Lopez



Salvatore Minissale



Riccardo Sapienza



MICINN-Consolider-Ingenio



*Thank you*  
*Muchas gracias*  
*Mila esker*



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