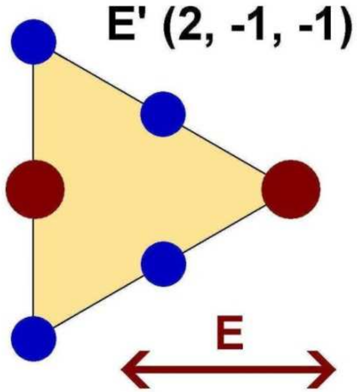
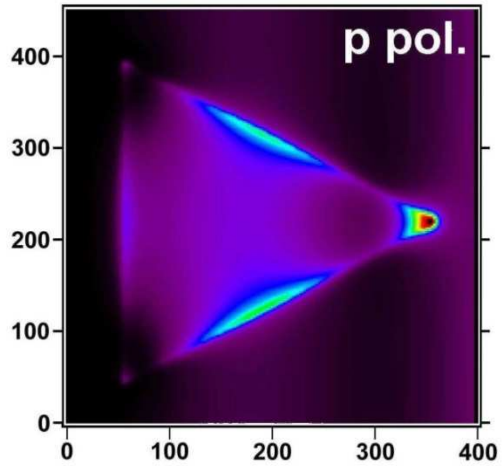
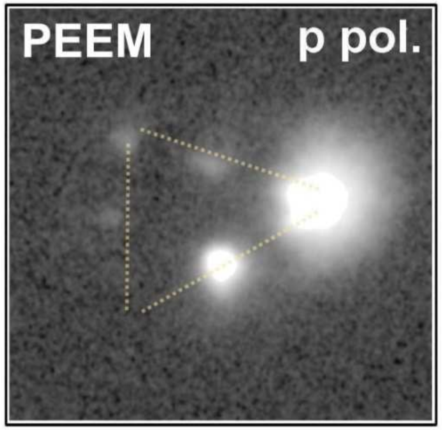
Plasmonics of single nanometric objects
Near field mapping by photoemission electron microscopy







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What is plasmonics ?

- "***A way to confine electromagnetic fields over dimensions on the order or smaller than the wavelength $\lambda_{h\nu}$***"

S. Maier in Plasmonics: Fundamentals and Applications (2007) Springer

- Interaction processes between electromagnetic radiation and conduction electrons at metallic interfaces = **coherent collective charge oscillations**

- **Basic ingredients**

  (i) *Surface plasmons-polaritons* SPP - Metal / dielectric interface - Hybrid wave between a photon and a plasma oscillations - Propagative mode along the interface, evanescent in perpendicular direction

  (ii) *Localised surface plasmons* LSP - Sub wavelength object - Non propagative mode

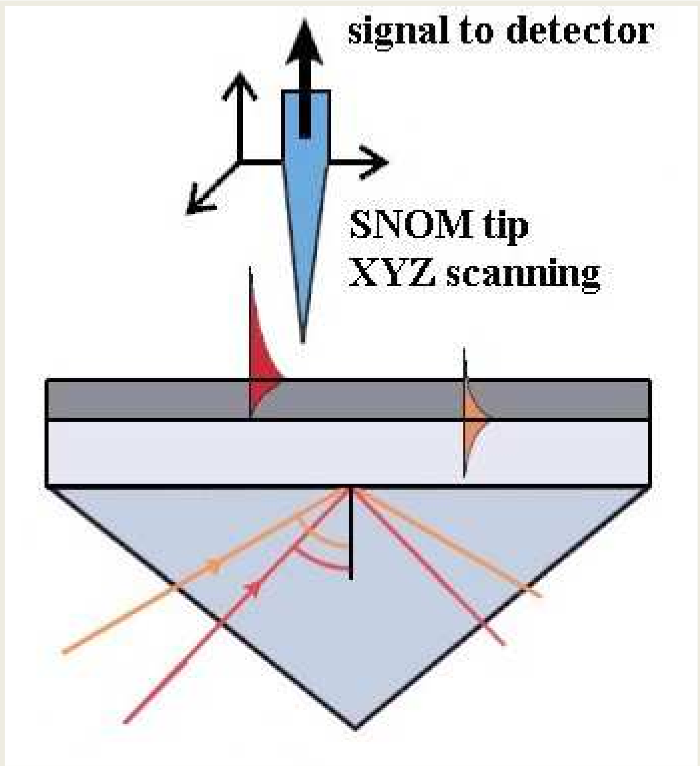
- **Plasmonics promises**

  (i) high spatial integration $\lambda_{h\nu}$ /10 ~ 60 nm (visible)

  (ii) high working frequencies $\nu \sim 10^{14}$ Hz = 10^5 GHz (visible)

Mapping the evanescent field at the nanometre

SNOM - *scanning near-field optical microscopy* (1984…)



- **Basic principle**

.scanning probe microscopy (SPM)

.introduction of a tip (glass fiber, metal …) in the near field of the object to be studied – **intrusive technique**

.routine resolution 50 - 100 nm

.many variants (illumination and collection modes)

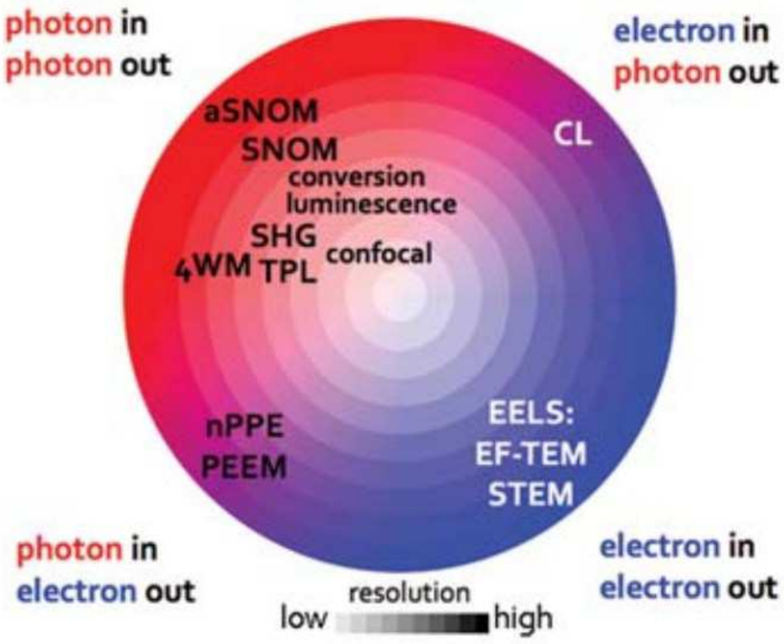
- **Drawbacks**

.possible perturbation of the evanescent field due to LSP excitation and lightning-rod effect at the tip-surface junction

.low reproducibility of tips

Mapping the evanescent field at the nanometre

Alternative non intrusive methods (2012)



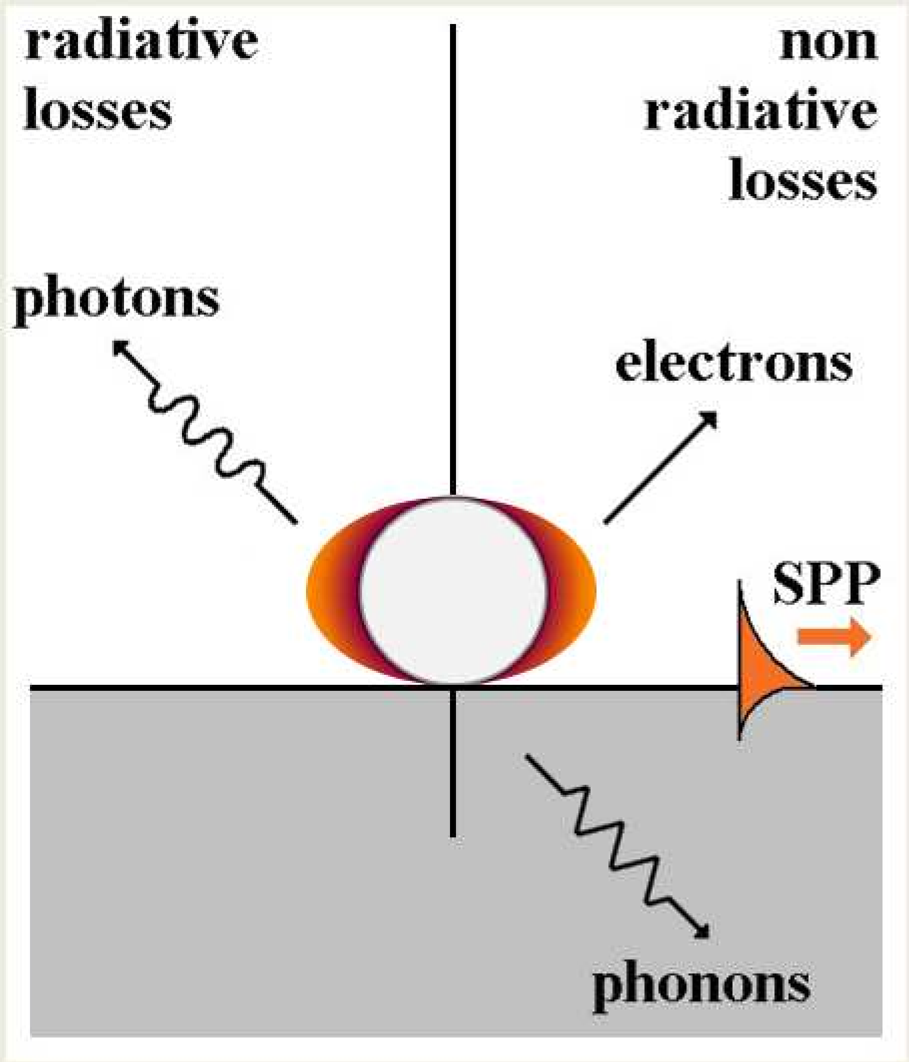
Process	Acronym	Measured quantity
	**4WM** – four-wave mixing **TPL** – 2-photon luminescence **SHG** – 2nd-harmonic generation **SNOM** – scanning nearfield optical microscopy **aSNOM** – apertureless SNOM	E, $\|E\|^2$, $\|E\|^4$
	**nPPE** – n-photon photoemission **PEEM** – photoemission electron microscopy	$\|E\|^{2n}$
	**EELS** – electron energy loss spectroscopy **TEM** – transmission electron microscopy **EF-TEM** – energy-filtered TEM **STEM** – scanning TEM	projected LDOS
	**CL** – cathodoluminescence	projected LDOS (partial)

- STEM EELS *scanning transmission microscopy in electron energy loss spectro. mode* (electron in, electron out) = Mapping of an electron energy loss signature
- CL *cathodoluminescence* (electron in, photon out) related to EELS
- PEEM *photoemission electron microscopy* (photon in, electron out), other acronym = n-PPE *n photon photoemission*

R. Vogelgesang *et al. Analyst* **135** (2010) 1175

Photoemission electron microscopy, a tool for plasmonics

Basic principle – plasmon decay channels



➤ **Plasmon decay channels**

.free-space radiative losses
(scattering, luminescence),

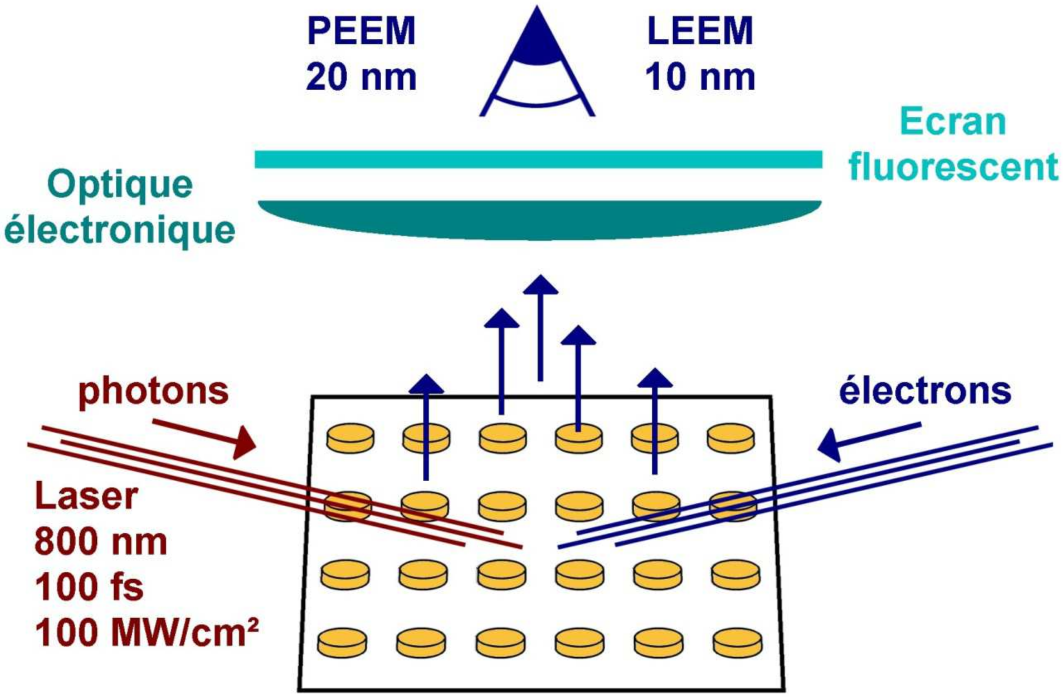
.non radiative losses
ohmic losses (phonons),
secondary SPP excitation,
electron emission,
...

➤ **Mapping of the near–field through electron emission.**

L. Douillard, C. Charra, *J. Phys. D: Appl. Phys.* **44** (2011) 464002

Mapping the evanescent field at the nanometre

LEEM/PEEM instrument (CEA Saclay)



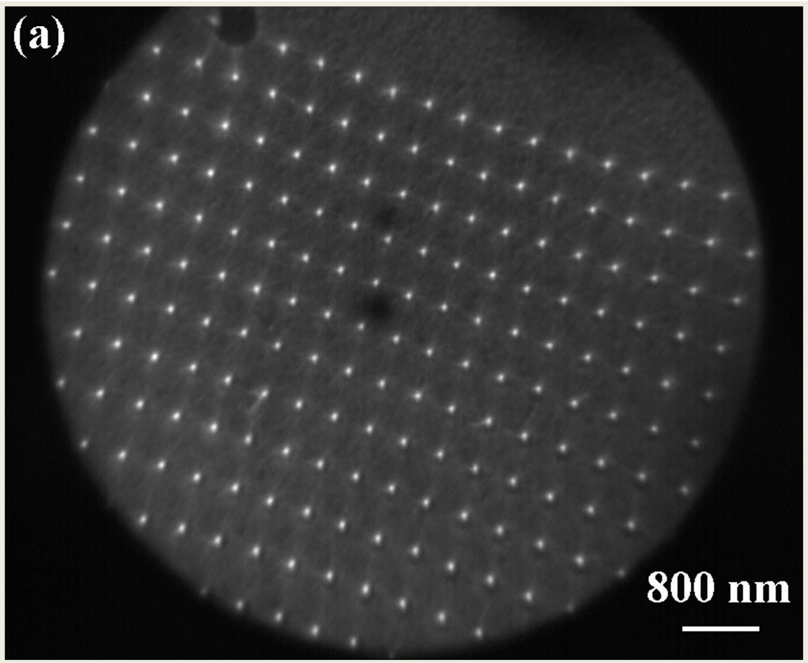
- Conductive sample (no charging effect) of low roughness

- LASER source Ti:Al_2O_3, 100 fs, wavelength ranges [690 nm, 1100 nm] (IR) + [525 nm, 680 nm] (visible, OPO). Incidence angles 0° & 75° / $\perp$ = (normal, grazing)

- LEEM / PEEM III Elmitec, www.elmitec-gmbh.com
  PEEM field of view 1.5 - 150 µm, lateral resolution (16/84) 25 nm
  LEEM field of view 1.5 - 80 µm, lateral resolution (16/84) 10 nm
  PEEM spectral resolution 10 meV (100 fs laser pulse width)

Mapping the evanescent field at the nanometre - PEEM

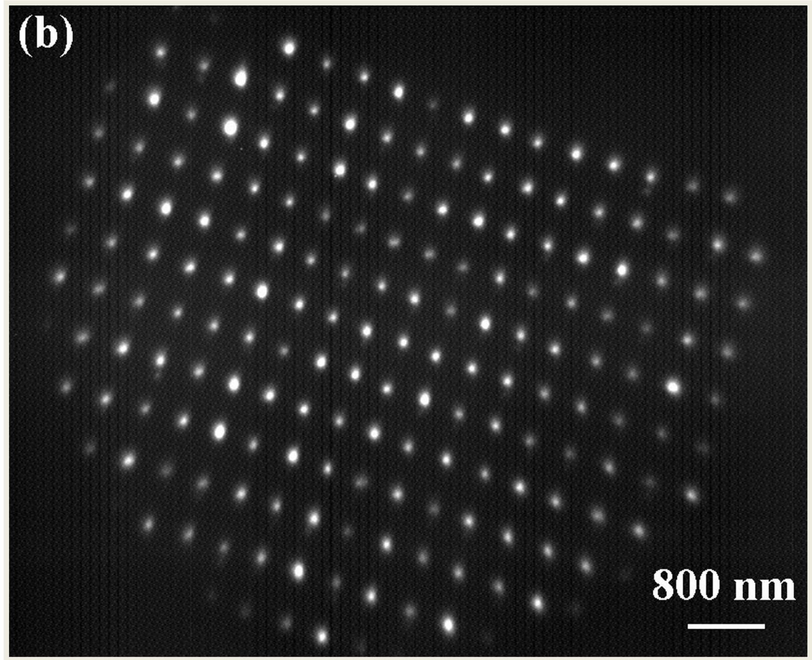
Nano-particle assemblies - LSP



Au disks/ITO ∅120 nm, lattice spacing 400 nm

➢ **LEEM picture (topographic imaging mode)**

Excitation = electrons (LASER off),
Signal = backscattered electrons.



Au disks/ITO grazing incidence p pol. 150 MW/cm²

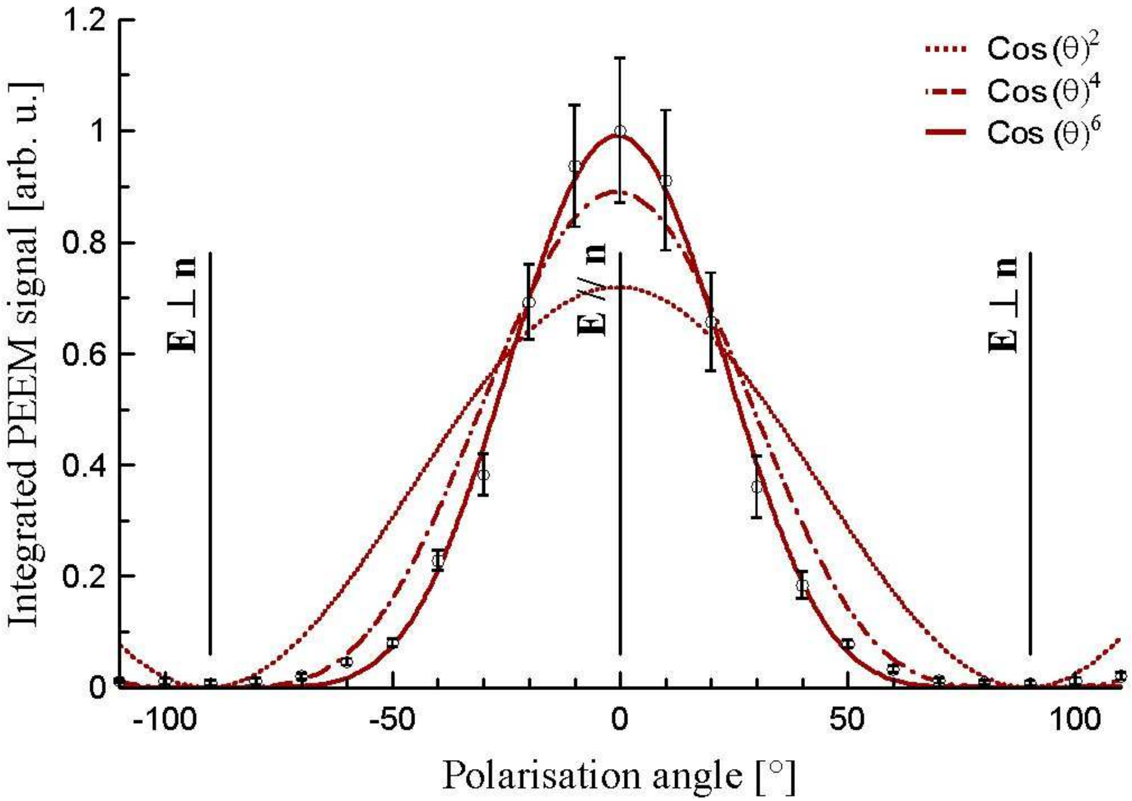
➢ **PEEM picture under LASER illumination**

Excitation = 766 nm photons,
hν (1.62 eV) < Φ_{Au} (4.6 - 5.1 eV),
Signal = photoelectrons !

L. Douillard *et al. J. of Appl. Phys.* **101** (2007) 083518

Non linear photoemission – PEEM

Nano-particle assemblies – Off plane polarisation dependence



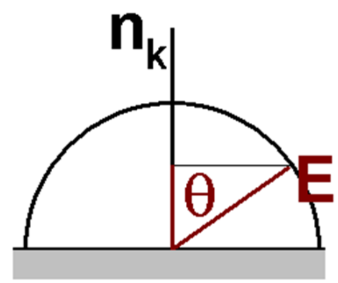
Au disks/ITO ∅120 nm

grazing incidence 17°

photon 880 nm (1.41 eV)

power dens. 140 MW/cm²

Keldysh factor 36 >> 1

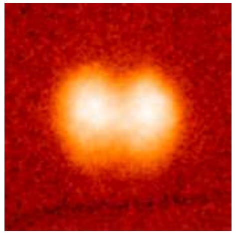


➢ Dependence of the photoemission yield with the polarisation angle θ at grazing incidence ⇔ PEEM integrated signal scales as $\cos(\theta)^6$

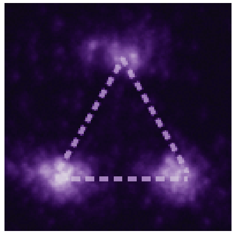
➢ **3 photon photoemission process** (3*1.41 eV = 4.23 eV, $\Phi_{Au} \approx 4.6 – 5.1$ eV). Probable electronic temperature assisted 3-PPE mechanism.

L. Douillard *et al. J. of Appl. Phys.* **101** (2007) 083518

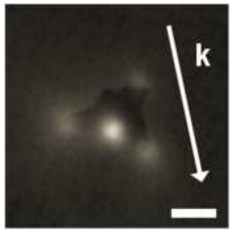
Plasmonics of single nanometric objects
Near field mapping by photoemission electron microscopy



➢ Nanometric objects = Wire & Rod

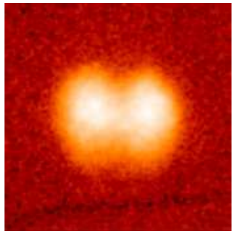


➢ Nanometric object = Triangle



➢ Nanometric object = Star

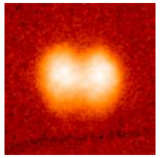
Plasmonics of single nanometric objects
Near field mapping by photoemission electron microscopy



- **Nanometric objects = Wire & Rod**
  Wire, short-range SPP waveguide
  Rod, optical nanoantenna

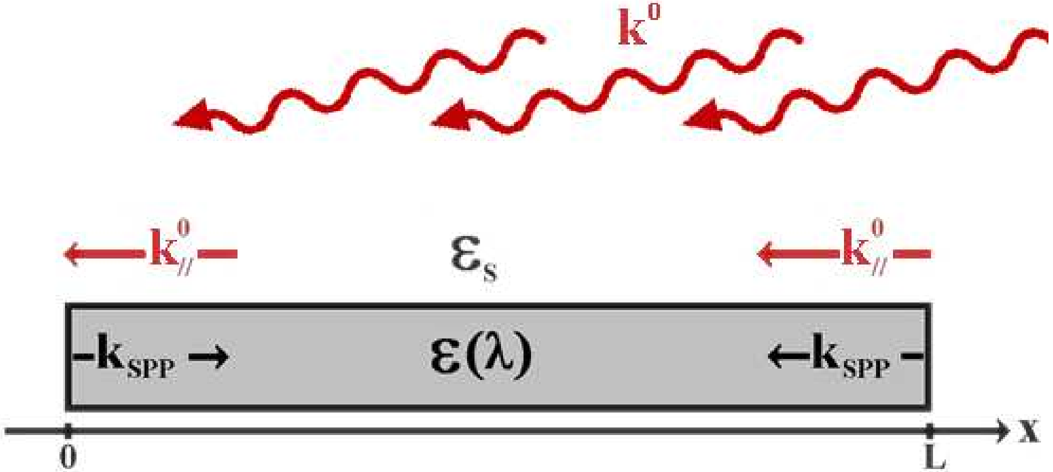
- **Objectives**
  Short-range SPP investigation

- **Degrees of freedom**
  Light polarization & wavelength



Surface plasmon-polariton propagation – PEEM

Nanowire – 1D SPP wave equation



$$\partial_t^2 F = V_\varphi^2 \partial_x^2 F + \frac{1}{\tau}\partial_t F + \frac{eE_0}{m_e} e^{i(\omega t - k_{//} x)}$$

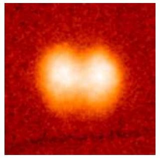
Damping Excitation

- **NanoWire** (length, radius) = (L, R), R ~ skin depth of the SPP field
- **1D SPP Wave equation**

  F amplitude of the charge displacement from equilibrium [m]

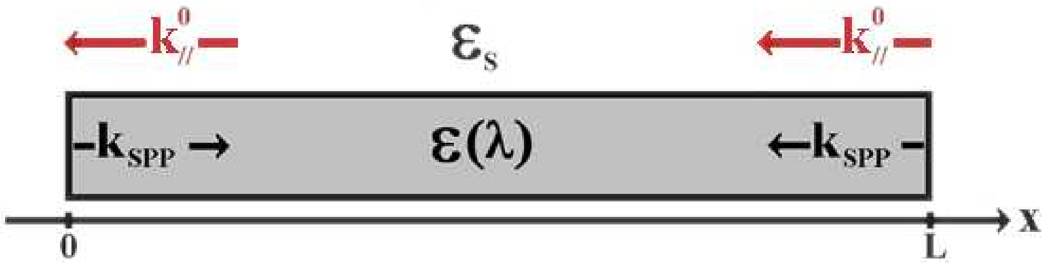
  V_φ phase velocity of the longitudinal plasma wave [m/s]

  τ SPP lifetime [s]



Surface plasmon-polariton propagation – PEEM

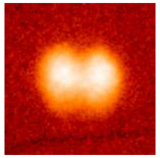
Nanowire – 1D SPP wave equation



$$F(x,t) = (A_p e^{+iKx} + A_m e^{-iKx} - \frac{eE_0 / m_e}{V_\varphi{}^2 (K^2 - k_{//}^2)} e^{-ik_{//}x}) e^{i\omega t}$$

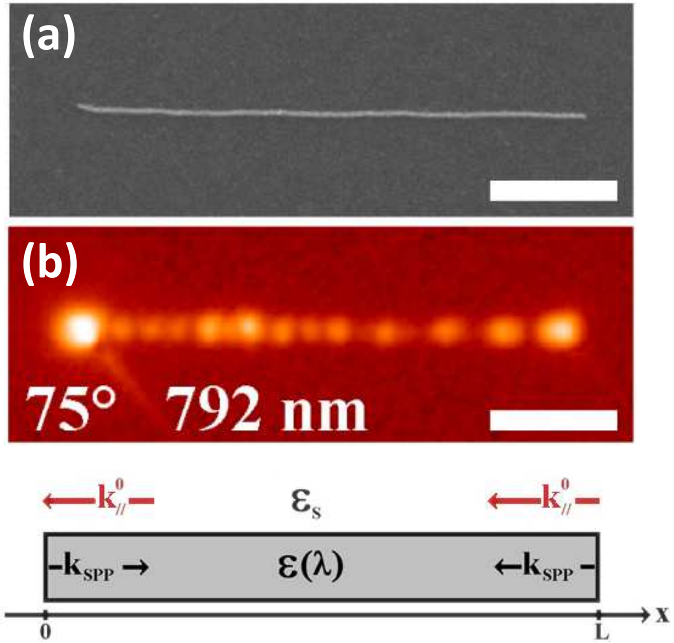
- **General solutions**
  (1) Two counterpropagative SPP waves of wavevectors $\pm$K,
  (2) A force wave of wavevector $k_{//}$

- **Beating stationary pattern with two distinct periods.**
  Long period $2\pi/(K_{SPP} - k_{//})$ at leading edge
  Short period $2\pi/(K_{SPP} + k_{//})$ at opposite edge



Surface plasmon-polariton propagation – PEEM

Polycrystalline nanowire – 1D symmetry



Au polycrystalline wire (L, R) = (4 µm, 15 nm), EBL Fabrication (UT Troyes, Fr)

photon wavelength $\lambda_{h\nu}$ 792 nm
grazing incidence angle 15°
p polarisation
irradiance ~ 150 MW/cm²

SEM (a) & PEEM (b) microscopies
white scale bar 1 µm

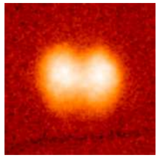
➢ **Surface plasmon-polariton propagation along a Au nanowire (waveguide)**

Short-range SPPs ⇔ R ~ SPP field skin depth ≈ 25 nm

SPP wavelength $\lambda_{SR\text{-}SPP}$ = 335 nm, attenuation length $L_{SR\text{-}SPP}$ = 3300 nm

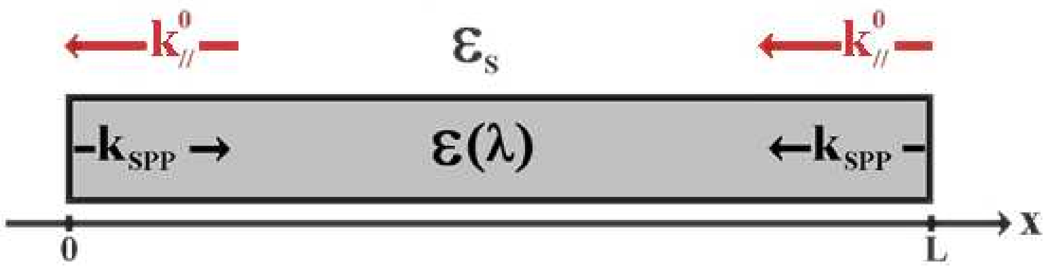
Shrinking of the light wavelength at optical frequency, λ : 800 nm → 335 nm

L. Douillard *et al. Nano Lett.* **8** (2008) 935



Near-field mapping of single objects – PEEM

NanoRod - Plasmonic resonators



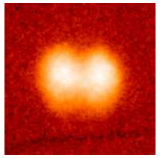
- Solution with boundary conditions F(-L/2) = F(+L/2) = 0

$$F(x,t) \propto \left(\frac{\cos(k_{//}L/2)}{\cos(KL/2)}\cos(Kx) + i\frac{\sin(k_{//}L/2)}{\sin(KL/2)}\sin(-Kx) - e^{-ik_{//}x}\right)e^{i\omega t}$$

Odd m modes Even m modes

- For L >> $L_{SR\text{-}SPP}$
  NanoWire = SPP wave guide

- For L << $L_{SR\text{-}SPP}$
  NanoRod = ***Plasmonic resonator (optical antenna)***
  Resonance modes ***$K_{SR\text{-}SPP}L = m\pi$, m integer***



Near-field mapping of single objects – PEEM
NanoRod - Plasmonic resonators

➢ **Resonance modes, simple analytical model**

(1) Resonance modes, m order

$K_{SR\text{-}SPP}L = m\pi$, $K_{SR\text{-}SPP} = 2\pi / \lambda_{SR\text{-}SPP}$

(2) For an infinite cylinder of radius R (L. Novotny *PRL* **98** (2007) 266802)

$$\lambda_{SR-SPP} = 2\pi R(a_1 + a_2 \frac{\lambda_{h\upsilon}}{\lambda^p})$$

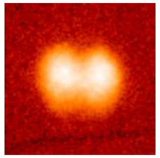
a_1, a_2 numerical coefficients = f(ε_{Metal}, $\varepsilon_{Sur.\ media}$)

λ^p free-space wavelength at plasma frequency, $\lambda^p(Au)$ = 138 nm

Resonant vacuum wavelength of the mode m

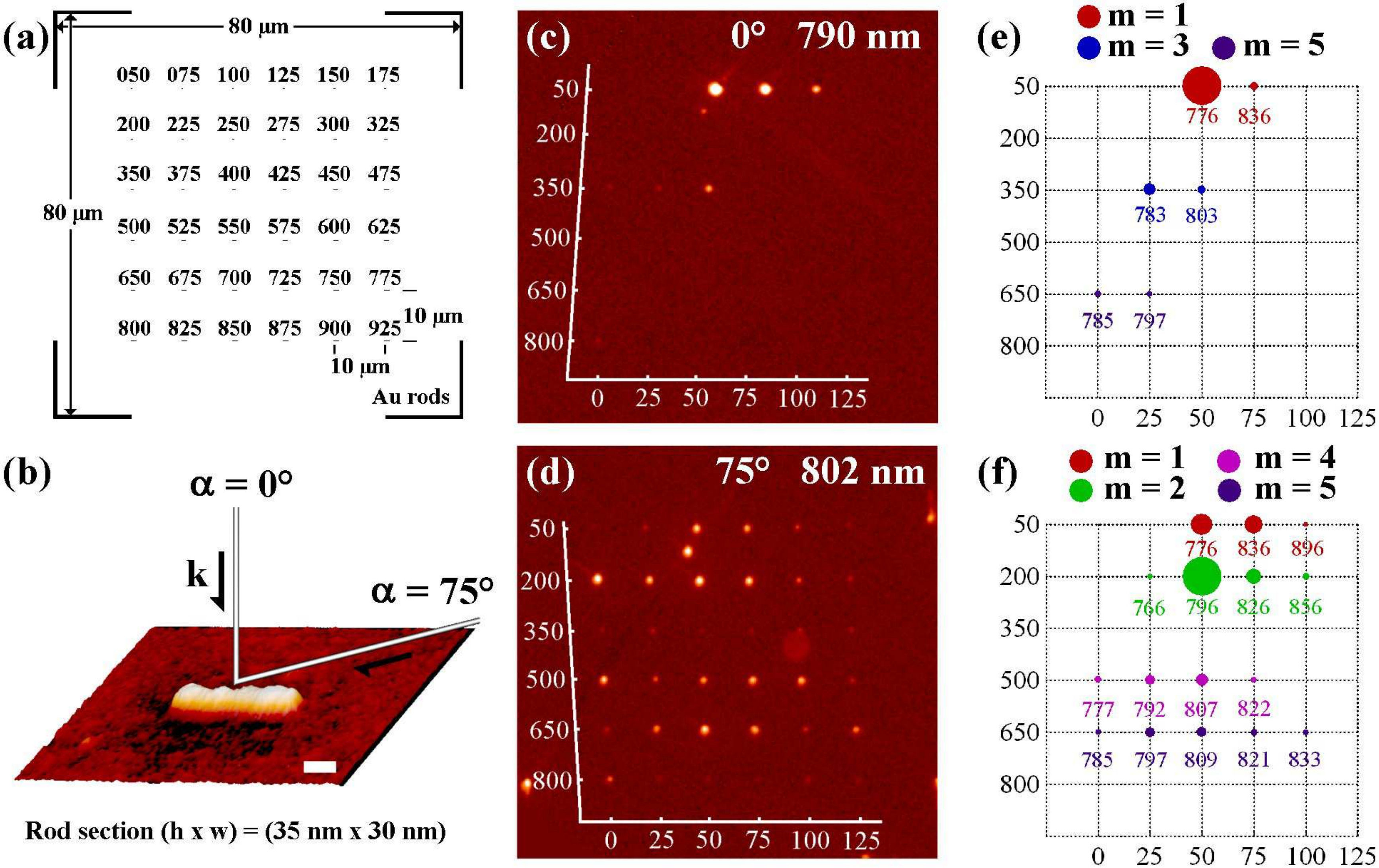
$$\lambda_{h\upsilon}^{Res.}(m) = \frac{\lambda^p}{a_2}(\frac{1}{m\pi}\frac{L+2R}{R} - a_1)$$

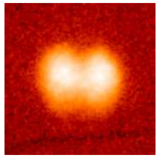
Phase retardation at end reflection L $\rightarrow$ L + 2R



Near-field mapping of single objects – PEEM

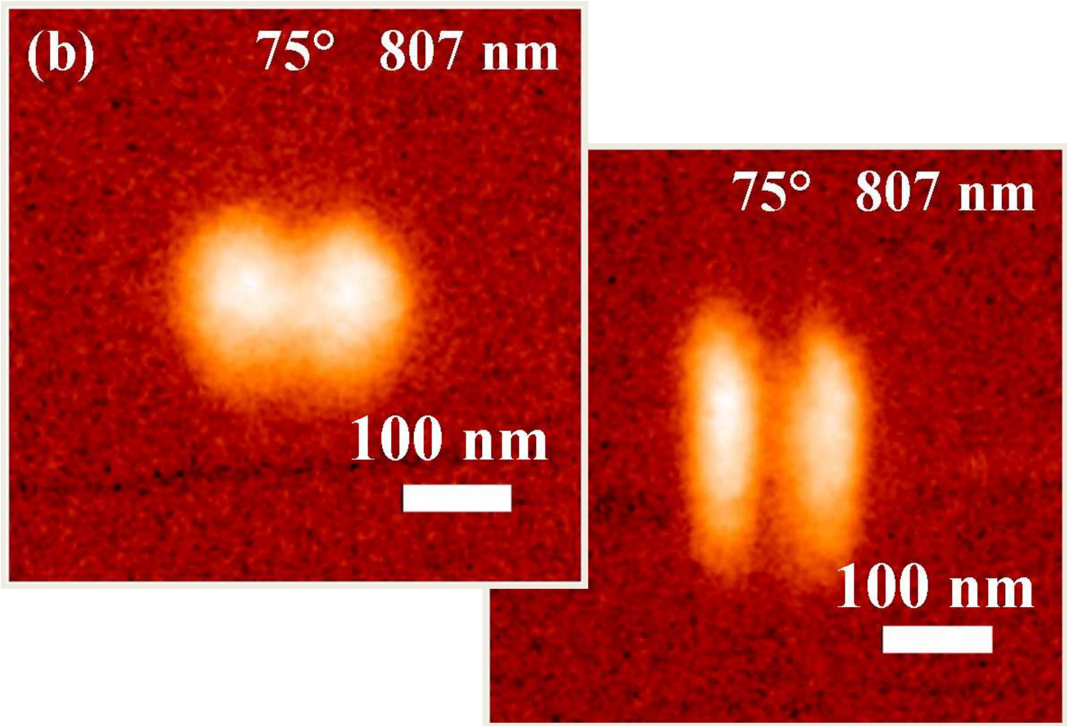
NanoRod - Plasmonic resonators

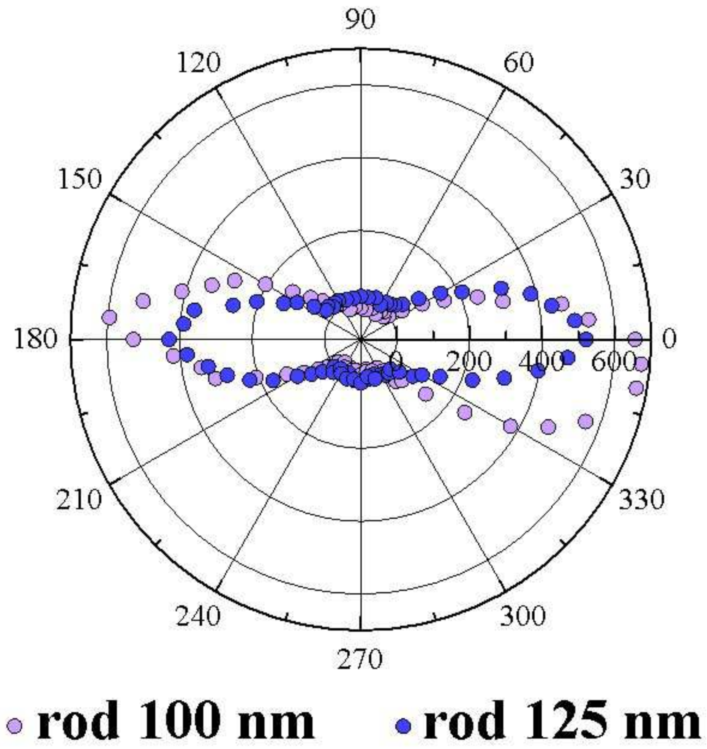




Near-field mapping of single objects – PEEM

NanoRod antenna – Dipolar mode, m = 1

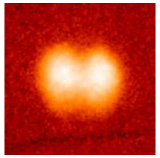




- PEEM imaging of one single polycrystalline Au rod ($100 \times 35 \times 30$ nm^3)
  lateral resolutions 40 & 21 nm
  photon wavelength $\lambda_{h\nu}$= 807 nm
  excitation field // rod axis

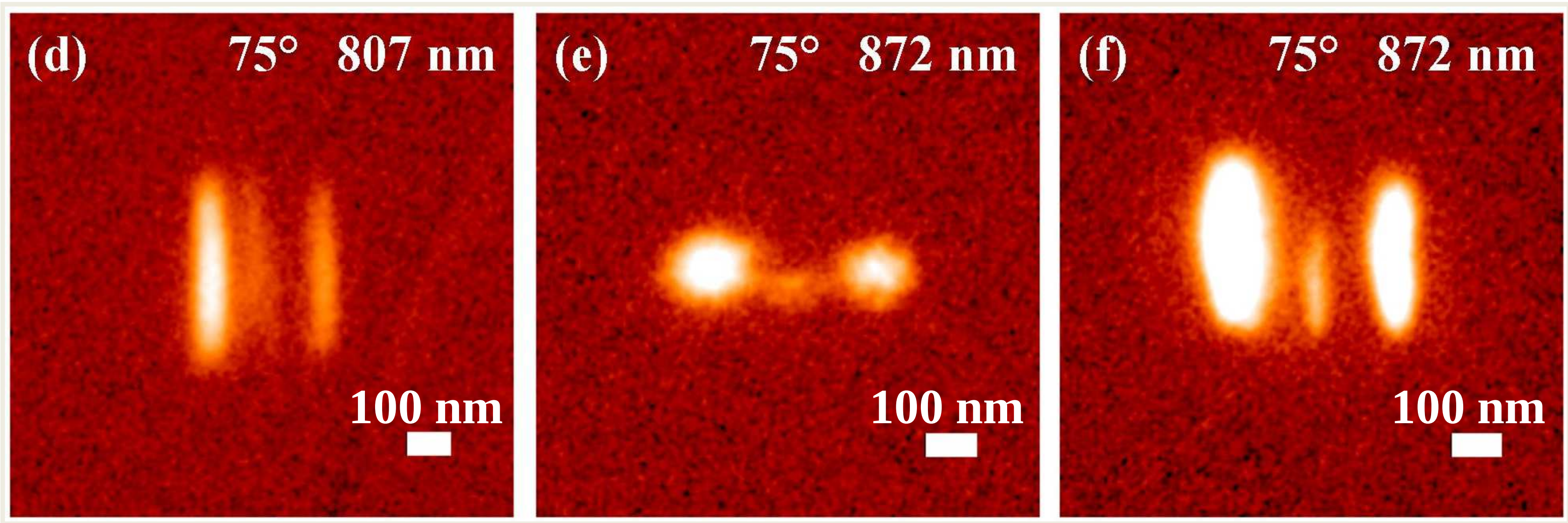
- Polar diagram of the photoelectron yield of one single rod

L. Douillard *et al. Nano Lett.* **8** (2008) 935



Near-field mapping of single objects – PEEM

NanoRod antenna – Quadrupolar mode, m = 2



- PEEM imaging of single polycrystalline Au rods.
  rod dimensions (d) 250 x 35 x 30 nm^3 and (e, f) 325 x 35 x 30 nm^3
  lateral resolutions 40 & 21 nm
  excitation field // rod axis

L. Douillard *et al. Nano Lett.* **8** (2008) 935

Plasmonics of single nanometric objects
Near field mapping by photoemission electron microscopy

Nanometric objects

Regular triangles of D_{3h} symmetry

Optical antennas

Objectives

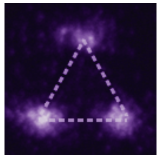
Manipulate light on the nanoscale

Active control of the radiation pattern of individual nanoantennas

Degree of freedom

Laser light polarization

Selective excitation of plasmon resonances of single Au Triangles using polarization dependent light excitation



Near-field mapping of single objects – PEEM

Regular D_{3h} triangles

- **Regular triangles**

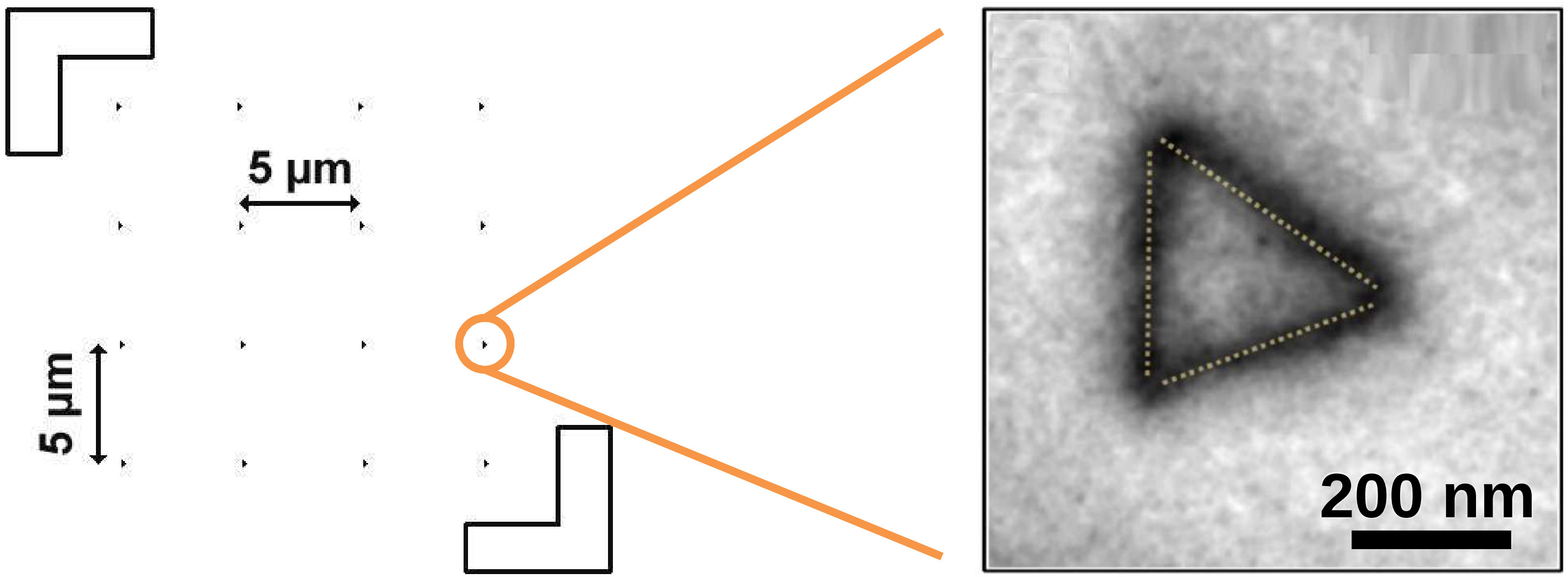
  Nanofabrication technique = electron beam lithography (UT Troyes, Fr)

  In-plane dimensions (altitudes) spanning {100, 140, 200, 250, 300 nm}

  Metal = Au deposited on a 5 nm TiO_2 adhesion layer

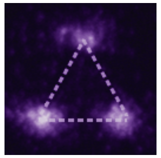
  Metal thickness = 50 nm

  Float glass substrate coated with 40 nm indium tin oxide (ITO)



4 x 4 matrix of 16 identical objects, 5 µm distance apart

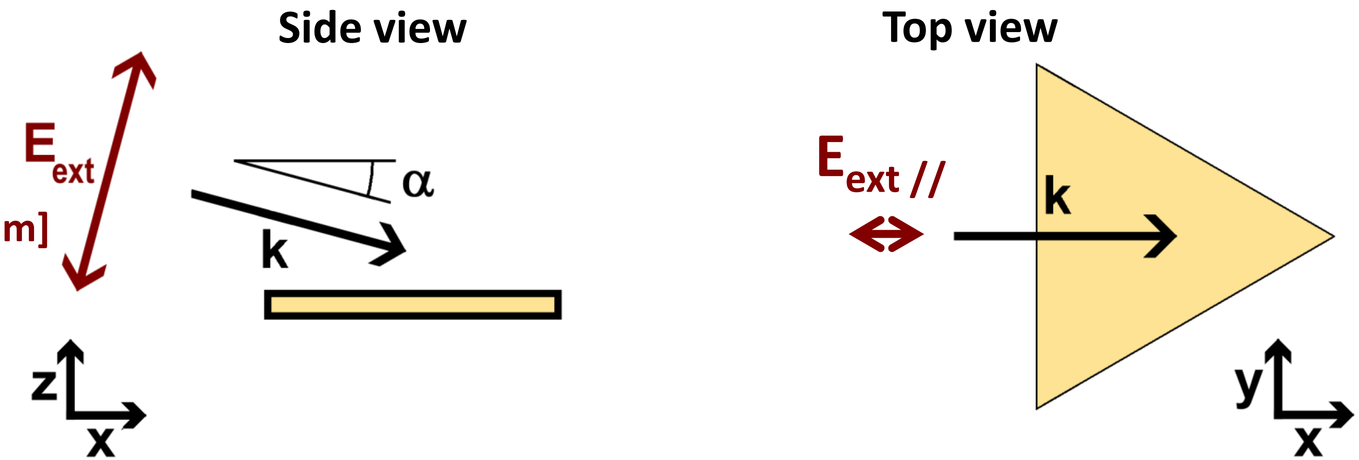
Triangle 300 x 50 nm, LEEM



Plasmonics of Regular D_{3h} NanoTriangles

Group theory & Vectorial selection rules for dipolar resonances

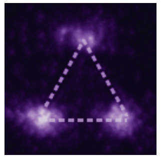
Laser
$Ti:Al_2O_3$
100 fs
[690 - 1100 nm]



➢ Selection rules applied to the interaction between the LSPRs of (i) a regular Triangle of D_{3h} symmetry and (ii) an external field E_{ext} yields to 3 optically active eigenmodes of irreducible representations (irreps) E’ and A''_2.

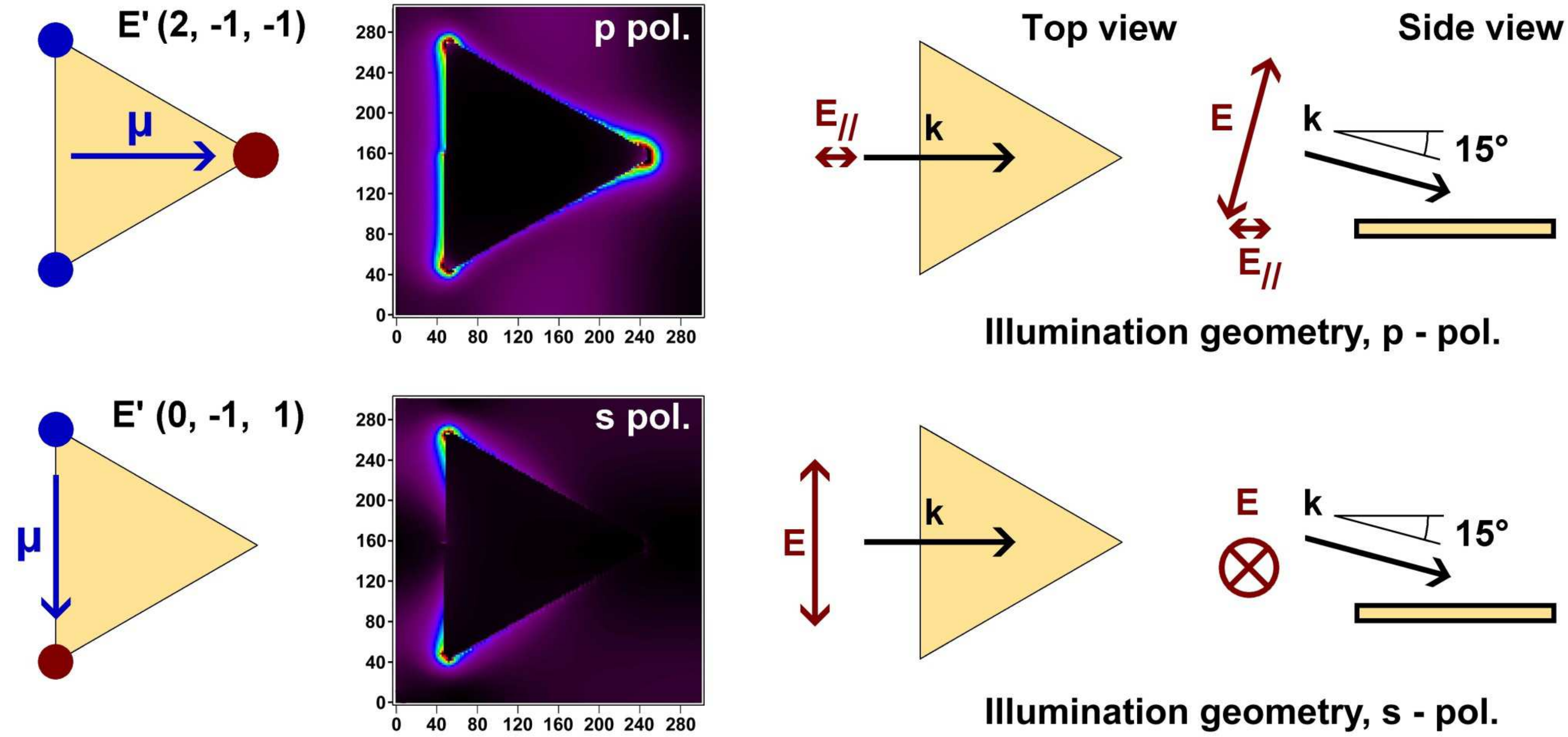
E’ irrep	Electric dipoles in the (x,y) plane
2D doubly deg.	In-plane coherent oscillation of electrons
	Low energy LSPRs ($\lambda_{res.} \sim 800$ nm)
A''_2 irrep	Electric dipole along the principal z axis
1D singly deg.	Transverse coherent charge oscillation along the thickness
	High energy LSPR ($\lambda_{res.} < 690$ nm)

W. Zhang, B. Gallinet, O. J. F. Martin, *Phys. Rev. B* **81** (2010) 233407

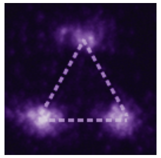


Plasmonics of Regular D_{3h} NanoTriangles

Group theory – In-plane E’ modes – Dipolar resonances

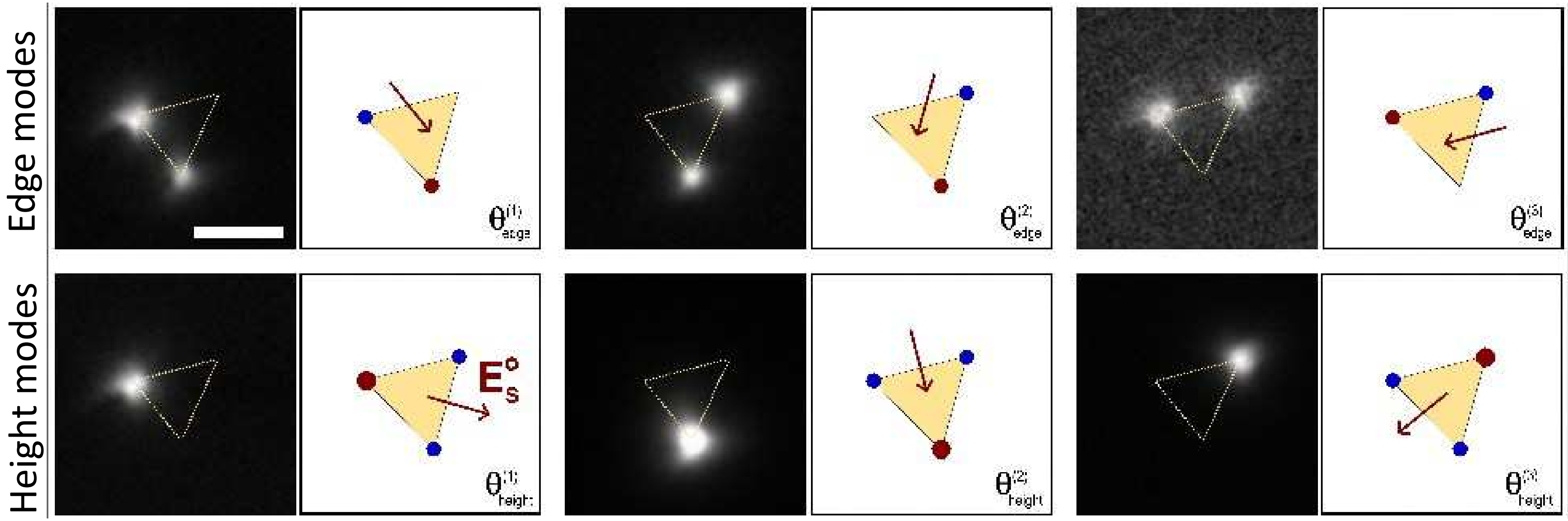


- ➢ Symmetry-adapted linear combination (SALCs) of the 2 dipolar eigenmodes (charge pattern basis) ⇔ 2 degenerate modes of perpendicular moments
- ➢ Selective excitation by proper choice of light polarisation ⇔ lifting of degeneracy & active control of the radiation pattern of a D_{3h} nanoantenna



Plasmonics of Regular D_{3h} Nanotriangles - PEEM

Selective excitation of dipolar E' modes

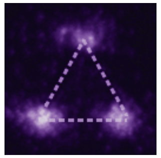


Au Triangle of in-plane height 200 nm – Dipolar mode, PEEM Imaging & Group theory

- Selective addressing of the E' dipolar plasmon resonances of a Au triangle using light polarization = active control of the radiation pattern of a D_{3h} nanoantenna

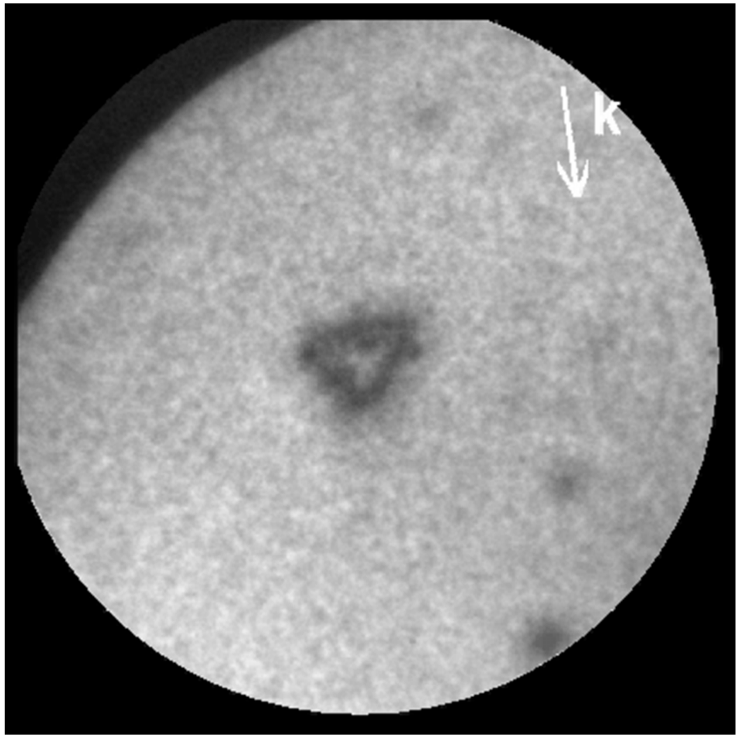
- Lifting of degeneracy of dipolar E' mode resonances

C. Awada *et al.*, *J. Phys. Chem. C* **116** (2012) 14591

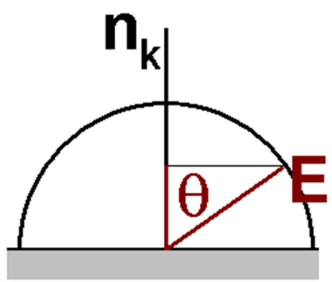


Plasmonics of Regular D_{3h} Nanotriangles - PEEM

Selective excitation of dipolar E' modes



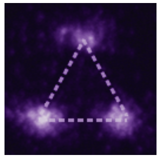
Au Triangle of in-plane height 200 nm
Dipolar mode
Wavelength 800 nm
Dependence on the polarisation angle θ at grazing incidence
PEEM imaging



➢ Selective addressing of the E' dipolar plasmon resonances of a Au triangle using light polarization = active control of the radiation pattern of a D_{3h} nanoantenna

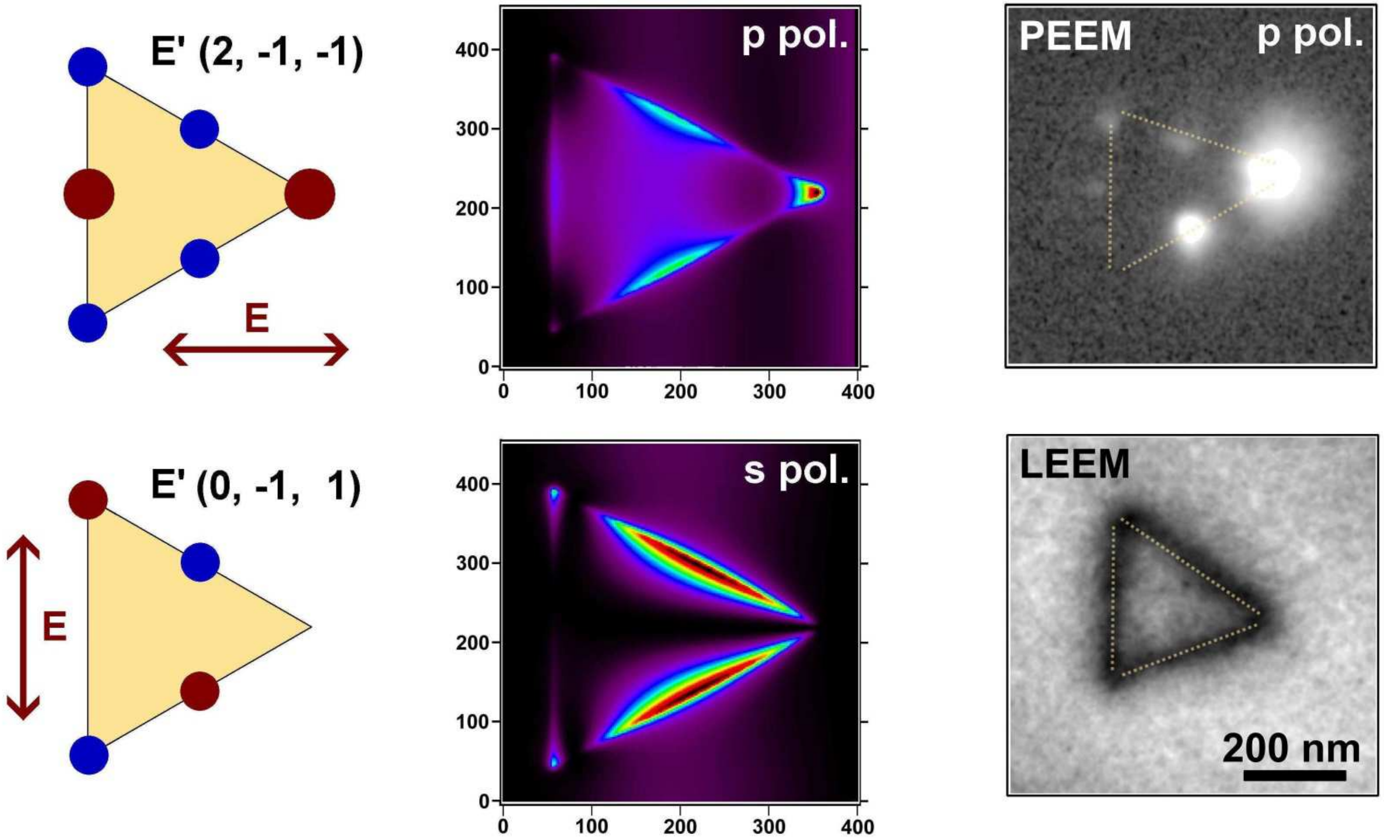
➢ Lifting of degeneracy of dipolar E' mode resonances

C. Awada *et al.*, *J. Phys. Chem. C* **116** (2012) 14591

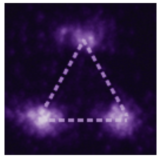


Plasmonics of Regular D_{3h} NanoTriangles

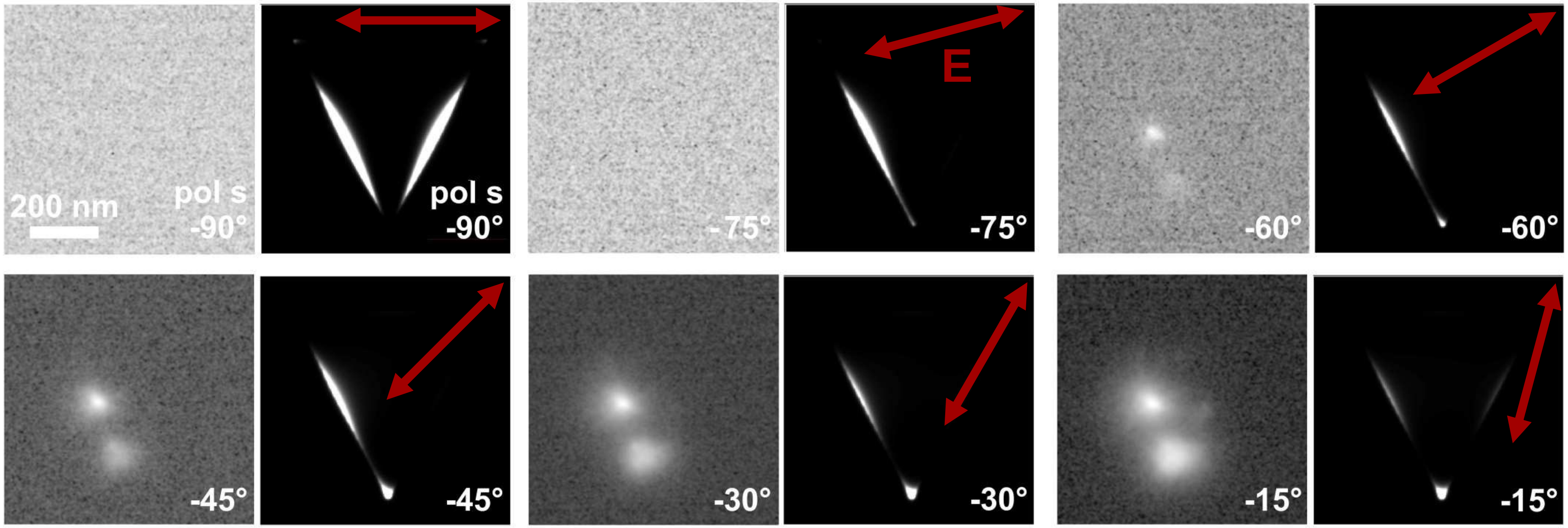
Group theory – In-plane E' modes – Quadrupolar resonances



- ➢ Symmetry-adapted linear combination (SALCs) of the 2 quadrupolar E' eigenmodes (charge pattern basis)
- ➢ Selective excitation by proper choice of light polarisation ⇔ active control of the radiation pattern of a D_{3h} nanoantenna, but <u>small azimuthal differences</u>



Plasmonics of Regular Nanotriangles
In-plane E' modes – Quadrupolar resonances

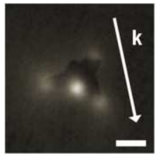


Au Triangle of in-plane height 300 nm – Quadrupolar E' mode
(1) PEEM Imaging & (2) FDTD calculations

➢ Selective excitation by proper choice of light polarisation ⇔ active control of the radiation pattern of a D_{3h} nanoantenna, but small azimuthal differences

C. Awada *et al.*, *J. Phys. Chem. C* **116** (2012) 14591

Plasmonics of single nanometric objects
Near field mapping by photoemission electron microscopy

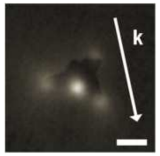


Nanometric objects
3D colloidal NanoStars
Optical antennas, SERS substrate

Objectives
Manipulate light on the nanoscale
Generate electric field hotspots,
Measure of field enhancement factors

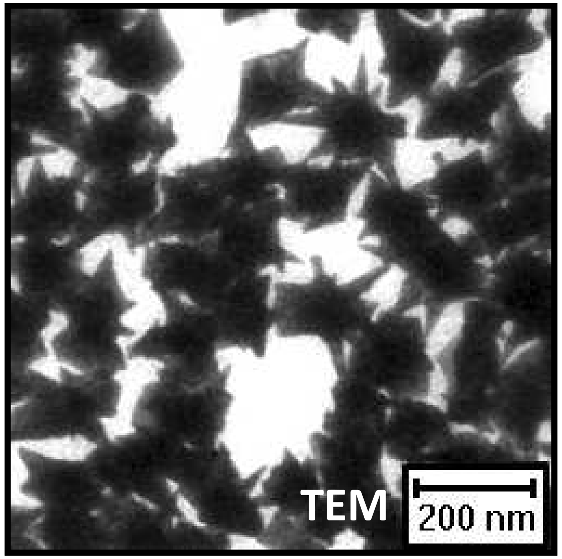
Degree of freedom
Laser light polarization & wavelength (p, λ)
Selective addressing of tips with polarization & wavelength

Fundamental aspects
Spectrometric differences between near- (PEEM) and far- (DFM) field signatures



Near-field mapping of single objects – PEEM

Colloïdal Au nanostars



- **Star shaped colloidal Au nanoparticles**
  - Near-spherical core
  - 5 to 8 sharp tips pointing outwards
  - Mean diameter 120 - 140 nm
  - Tip curvature radius ~ 5 nm
  - Monocrystalline 3D objects (twinned)

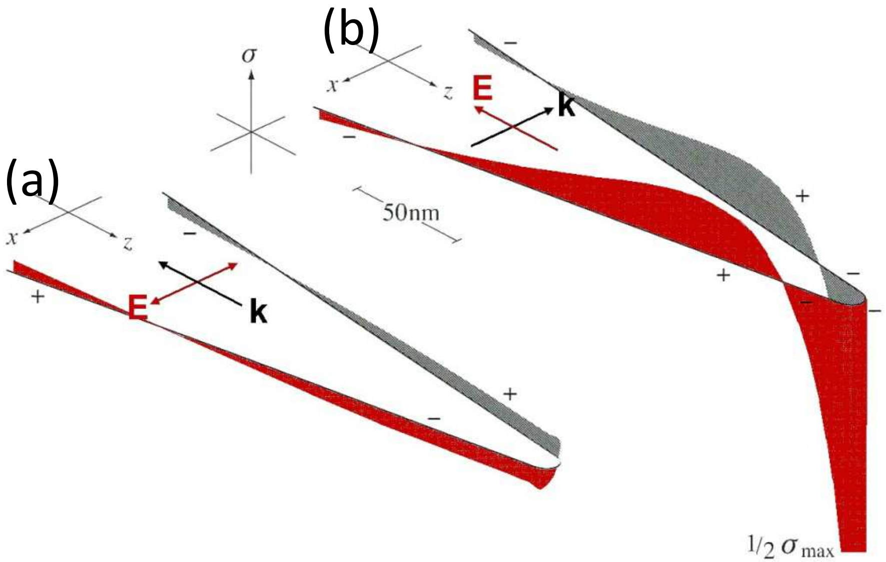
F. Jäckel
Ludwig-Maximilians-Universtität
München, Germany

- **Interest.** Nanostar particles are expected to provide hotspots of highly localized and strongly enhanced electromagnetic field under light excitation,

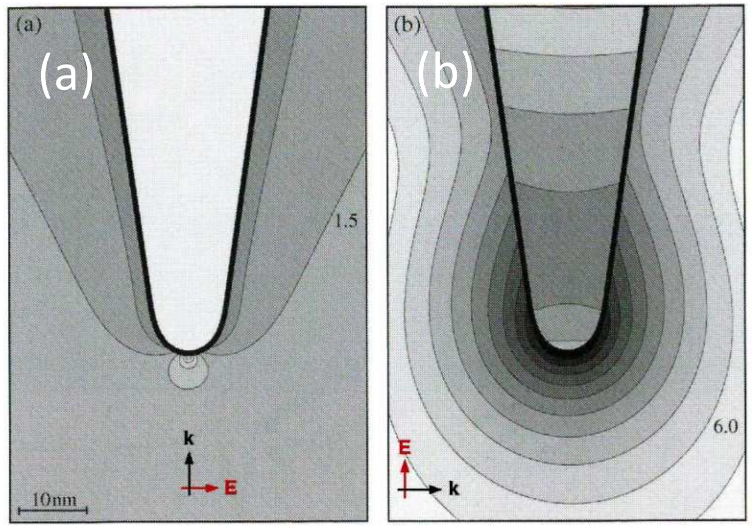
- **Application example.** Nanostar particles = High SERS (*surface enhanced Raman spectrometry* n = 2) signal amplification for single molecule sensitivity. $E/E_0 \sim 50$

C. Hrelescu et al., APL **94** (2009) 153113

Plasmon resonance of a solid Au tip
Theory – *Multiple multipole Method (MMP)* simulation



Induced surface charge density of a Au tip (5 nm tip radius) illuminated by a monochromatic wave at λ = 810 nm. (a) Transverse polarization, (b) Axial polarization.

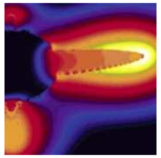


Near-field of a Au tip (5 nm tip radius) illuminated by a monochromatic wave at λ = 810 nm. (a) Transverse polarization, (b) Axial polarization.

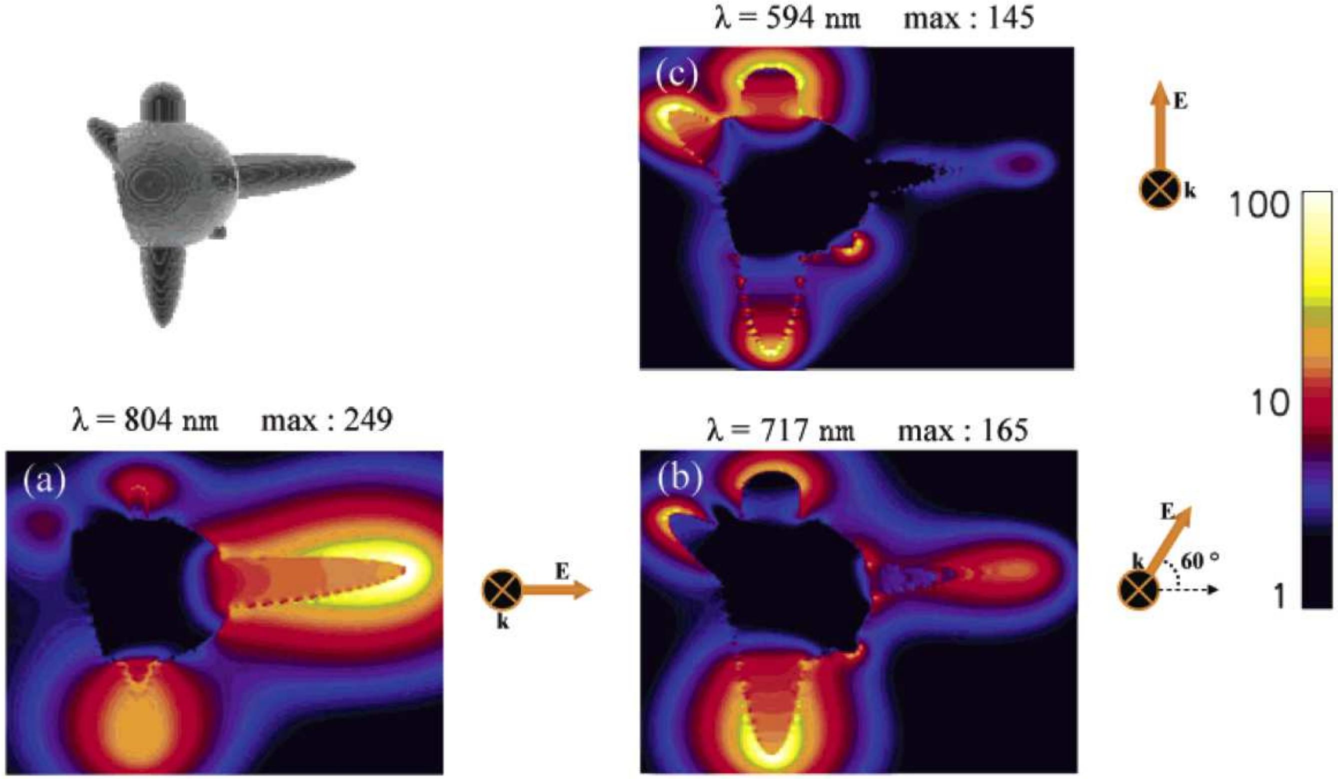
Light incident polarization parallel to the tip axis

- ➢Accumulation of surface charges at the end of the tip,
- ➢ Strong field enhancement at the end of the tip.

L. Novotny, B. Hecht (2006) Cambridge Univ. Press; PRL **79** (1997) 645



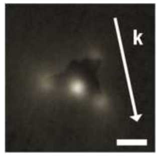
Plasmon resonances of a Au nanostar
Theory – *finite-difference time-domain* simulation



Normalized field enhancement of a Au nanostar (spherical core + tips = prolate spheroids) 100 nm in diameter under light excitation

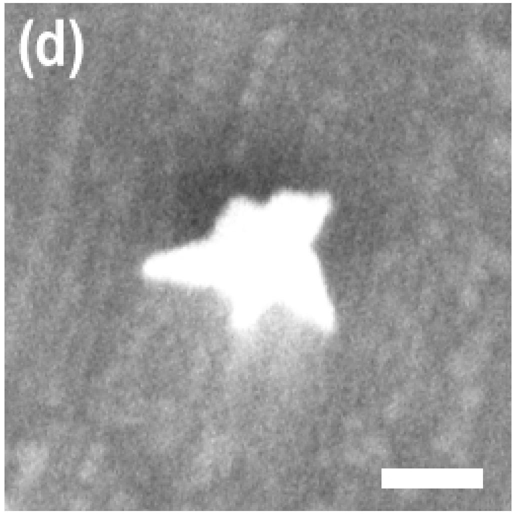
- Strong **polarization-** and **wavelength-** dependences,
- Field enhancement factors $E_{int}/E_0 \sim$ 10 - 20, $E_{ext}/E_0 \sim$ 100 - 200.

Feng Hao *et al. Nano Lett.* **7** (2007) 729

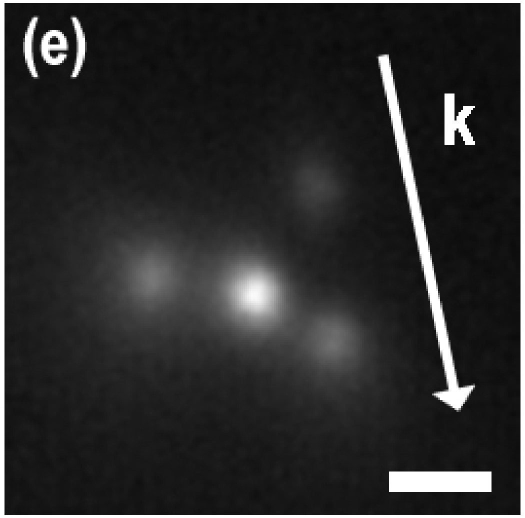


Near-field mapping of single objects - PEEM

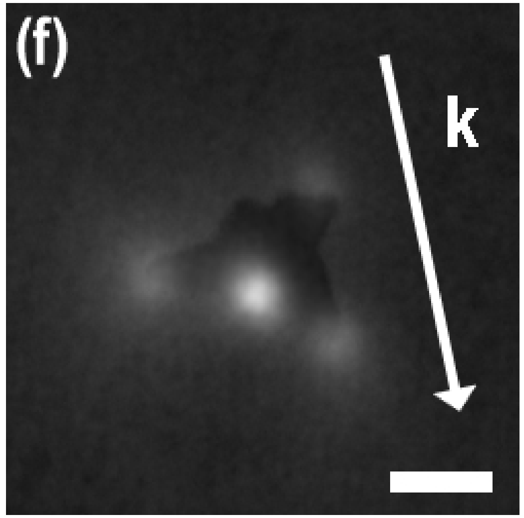
Experiment - NanoStars & Field hotspots at tip ends



SEM – Topographic
scale bar 100 nm



PEEMs – Near-field
wavelength 780 nm inc. 15° p
wavelength 860 nm inc. 15° p - 42°



LEEM, SEM + PEEM superposition

- Field hotspots at the ends of the object's tips,
- Field enhancement factor estimates

(i) PEEM using quality factors Q (minimal value, pole effect)

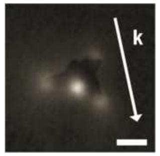
E_{int}/E_0 = 1.3 $\pm$ 0.3 $E_{ext}/E_0 \sim$ 10 (E_{int}/E_0) = 13 $\pm$ 3

(ii) PEEM using reference flat Au surface (true value, pole + shape effects)

E_{int}/E_0 = 8 $\pm$ large error $E_{ext}/E_0 \sim$ 80

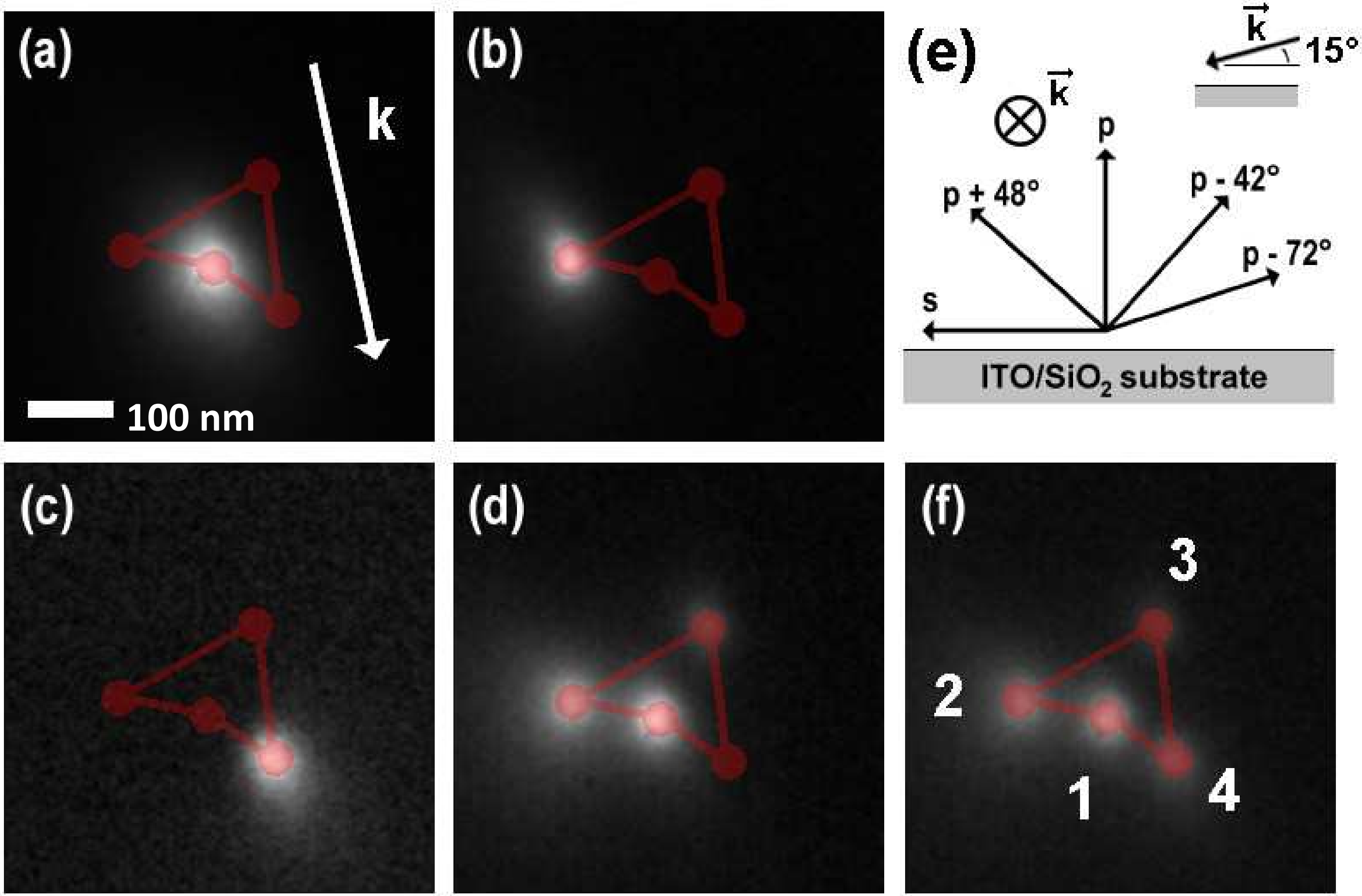
RAMAN $E_{ext}/E_0 \sim$ 50 - 75 (LMU Munchen, APL **94** (2009) 153113)

C. Hrelescu *et al.* *Nano Lett.* **11** (2011) 402



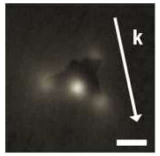
Near-field mapping of single nanostars – PEEM

Selective addressing of object tips (p, λ) - Polarization



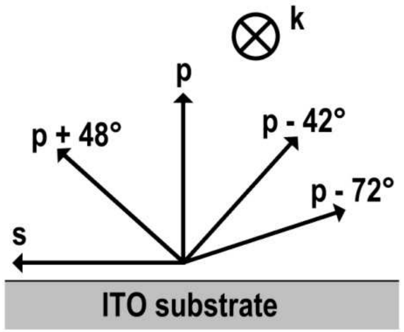
Selective excitation of individual plasmonic hotspots at the tips of single gold nanostars

C. Hrelescu *et al. Nano Lett.* **11** (2011) 402

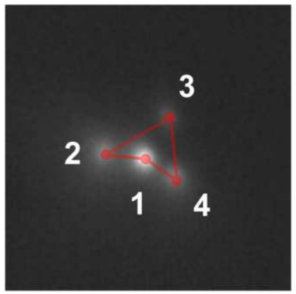


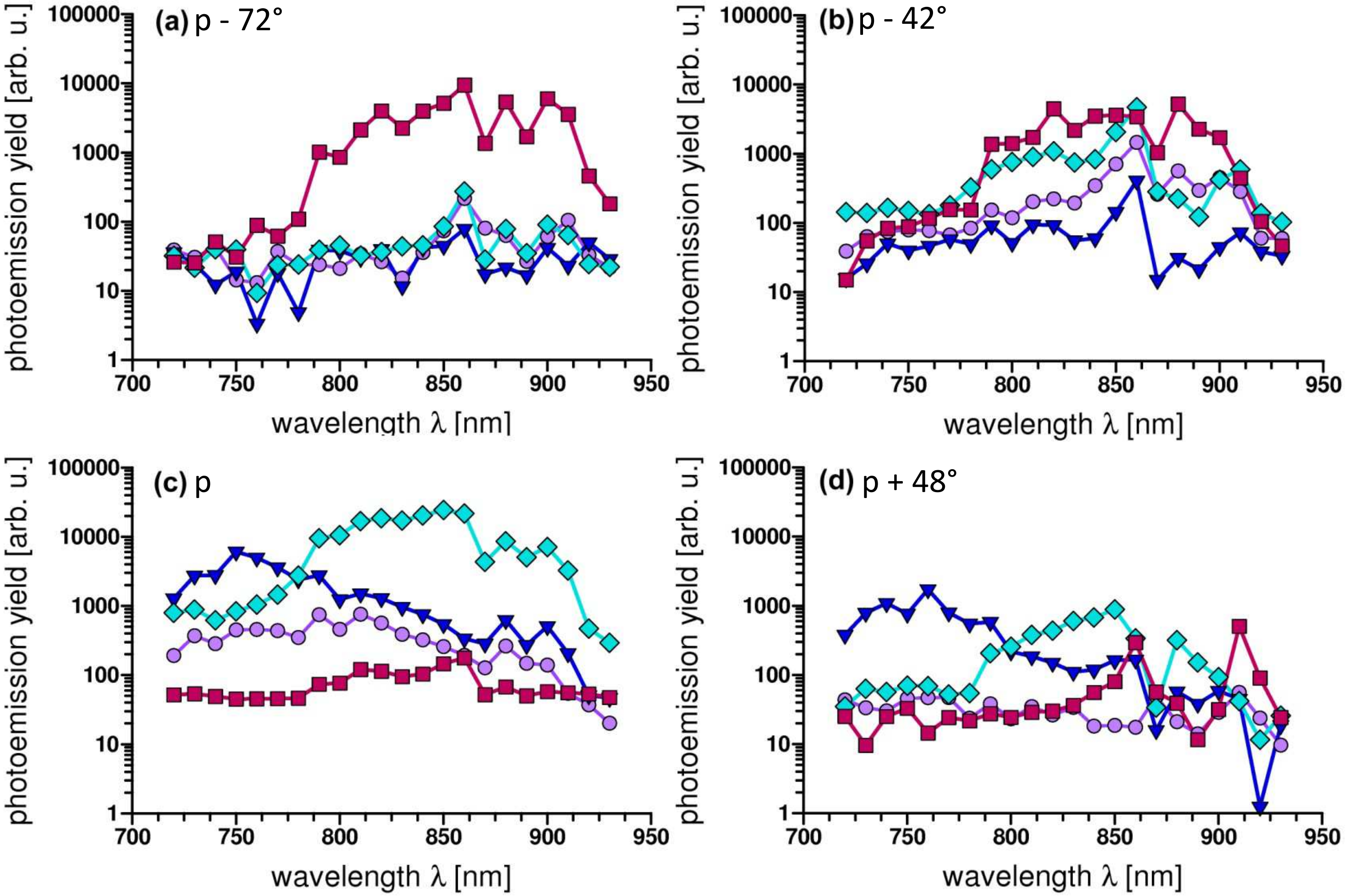
Near-field mapping of single nanostars – PEEM

Selective addressing of object tips (p, λ) - Wavelength

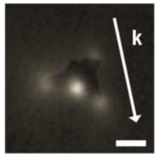


(a) Polarization p - 72°
(b) Polarization p - 42°
(c) Polarization p
(d) Polarization p + 48°



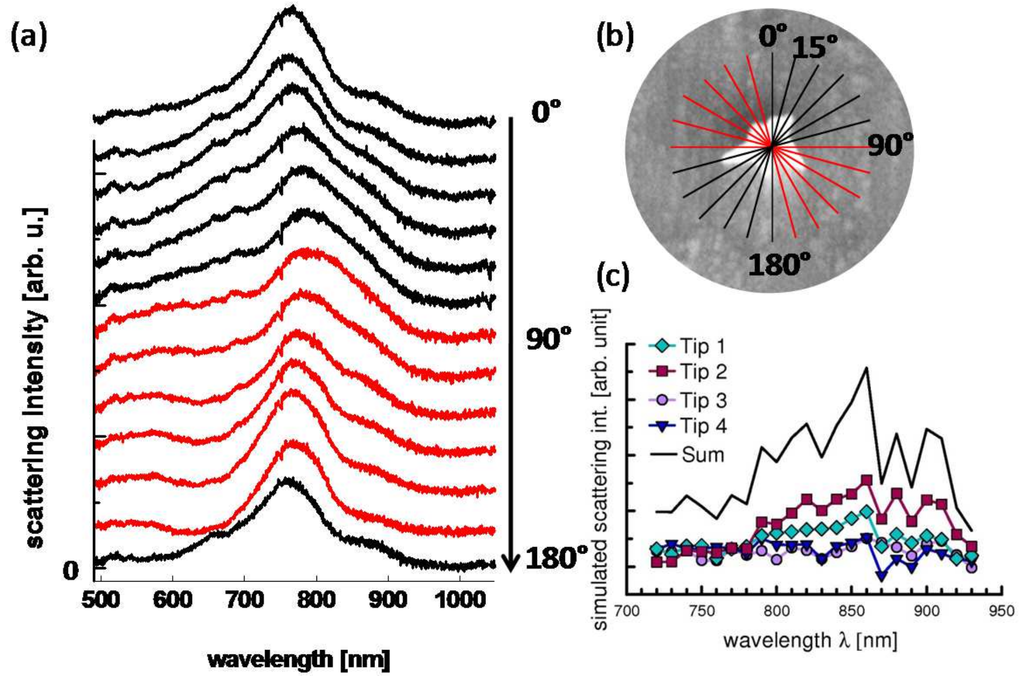


Selective excitation of individual plasmonic hotspots at the tips of single gold nanostars
C. Hrelescu *et al. Nano Lett.* **11** (2011) 402



Near-field mapping of single nanostars – PEEM

PEEM vs DFM (dark field microscopy)



(a, b) Polarization dependent white light Rayleigh scattering spectra of a nanostar particle
(c) Rayleigh scattering spectrum simulated from photoemission experiment data

- Comprehensive understanding of DFM vs PEEM spectra,
- Difference in spectrometric signature positions : near field (PEEM, absorption) signature is blueshifted in respect to the far field (DFM, scattering) signature,
- **Size and shape effects beyond the quasi static approximation?**

B.M. Ross, L.P. Lee *Opt. Lett.* **34** (2009) 896; B.J. Messinger *et al. Phys. Rev. B* **24** (1981) 649

Plasmonics of single nanometric objects
Near field mapping by photoemission electron microscopy

- **Light manipulation at the nanoscale** by selective excitation of different LSPR eigenmodes of simple finite objects

  Degrees of freedom = light polarisation and wavelength,

- **Group theory descriptions** of LSP resonances of regular finite objects

- **PEEM microscopy**. An alternative tool to SNOM in plasmonics for near field investigations at subwavelength scale:

  .non intrusive method (no probing tip),

  .high spatial resolution ~ 20 nm (3 nm on AC instruments)

  .spectrometric resolution ~ 10 meV (fs laser pulse width)

Plasmonics of single nanometric objects
List of collaborators

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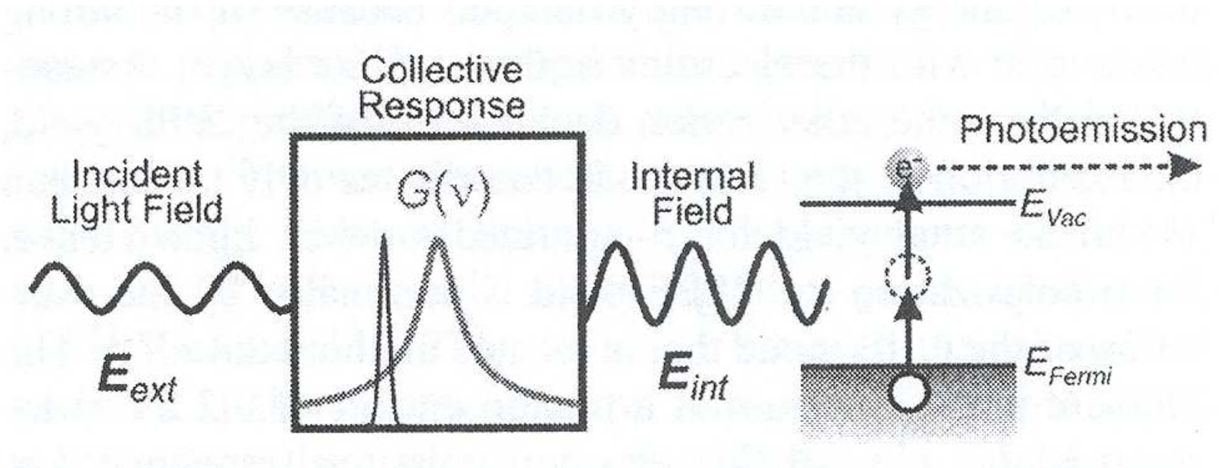
C. Hrelescu, T. Sau, F. Jäckel

Photoemission electron microscopy, a tool for plasmonics

Basic principle



(i) Electron collective response
⇔ plasmon excitation

(ii) Non linear photoemission process

Plasmon res. $E_{plasmon} \approx$ 1,55 eV (IR 800 nm)

Metal work function $\Phi_{\text{noble metal}} \approx$ 4,5 eV

$$E_{\text{int.}}(\nu) = G(\nu).E_{ext.}(\nu), \;\; G_{Sphere} = \frac{3\varepsilon_m}{\varepsilon + 2\varepsilon_m}$$

$E_{\text{int.}}$ Internal electric field [V/m]

G Response function of the many electron system

$E_{ext.}$ Incident electric field [V/m]

$$n.h\nu > \Phi_{Metal}$$

n order of non linearity,

hν photon energy [eV],

Φ_{Metal} metal work function [eV]

M. Merschdorf *et al. Phys. Rev. B* **70** (2004) 193401

Photoemission electron microscopy, a tool for plasmonics

Basic principle

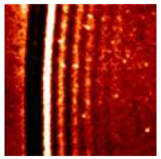
- The non linear photoemission process is proportional to the $2n^{th}$ power of the internal electric field $E_{int.}$ (electron reservoir is imaged)
- Multiphotonic cascade absorption $\neq$ Coherent absorption

$$I_{e^-} \propto (\vec{p}.\vec{E}_{\text{int.}})^{2n} \propto (\vec{p}.G\vec{E}_{ext.})^{2n}$$

p electron momentum [kg.m/s],
E_{int} internal electric field [V/m],

n non linearity order
$E_{ext.}$ incident electric field [V/m]

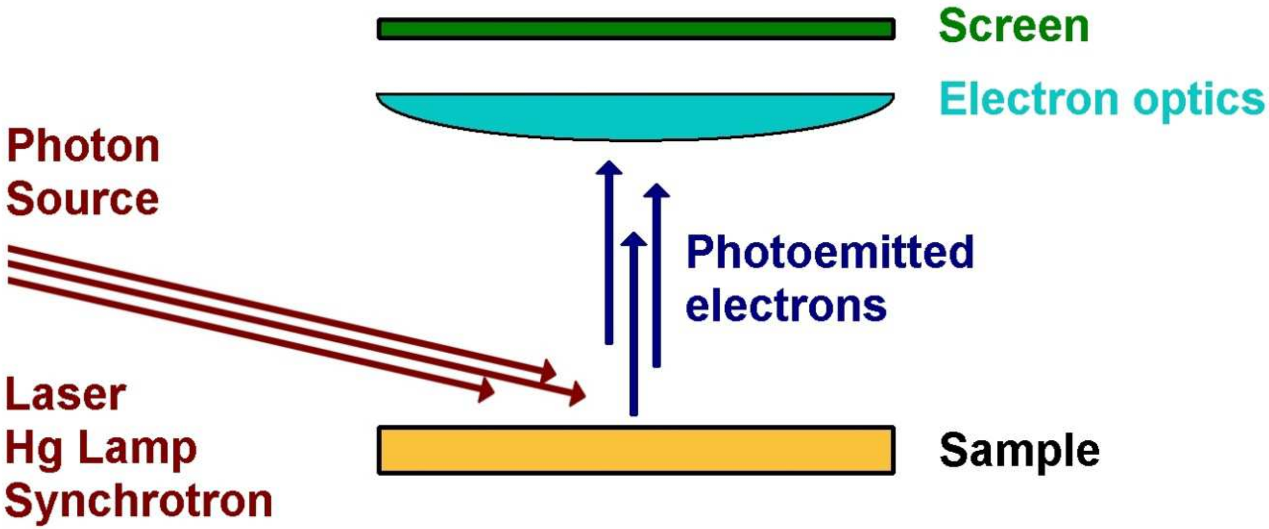
For large objects $\geq \lambda_{h\nu}$, the internal electric field of the plasmon excitation $\vec{E}^{SPP}_{\text{int.}}(r,t)$ (group velocity v_{SPP} < c) interferes with a 2^{nd} component linked to the incident field $\vec{E}^{h\nu}_{\text{int.}}(r,t)$ (group velocity c / refractive index).



- For large objects $\geq \lambda_{h\nu}$, observation of beating interference patterns between $(\vec{E}^{SPP}_{\text{int.}}(r,t),\ \vec{E}^{h\nu}_{\text{int.}}(r,t))$

PEEM – photoemission electron microscopy (1933)

Instrumentation



➢ **Basic principle. Mapping of the photoemitted electron distribution in two dimensions** (photon in, electron out).

.standard electron optics (electrostatic, magnetic lenses)

.full field microscopy, non intrusive technique, no physical tip in the vicinity of the measuring volume (≠ SNOM)

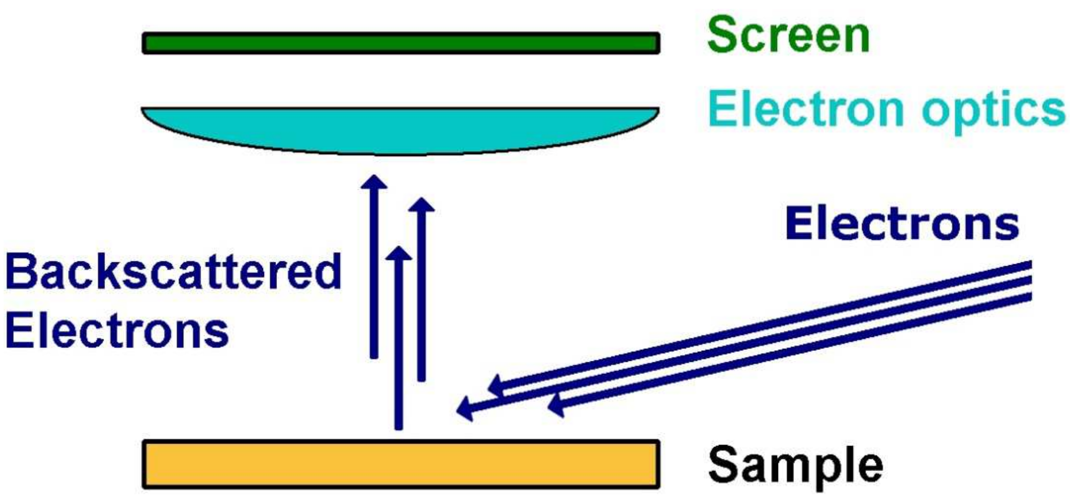
.image contrast: work function Φ (small hν), photoemission lines (large hν)

.lateral resolution (16 / 84 criteria) ≈ 20 nm

E. Brüche, *Z. Phys.* **86** (1933) 448; J. Pohl, *Zeitschr. f. techn. Physik* **12** (1934) 579; E. Bauer *Rep. Prog. Phys.* **57** (1994) 895

LEEM – low energy electron microscopy

Instrumentation



➢ **Basic principle. Mapping of the backscattered electron distribution** (electron in, electron out) - Topographic imaging.

.standard electron optics (electrostatic, magnetic lenses)

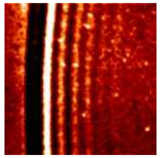
.full field microscopy, no scanning parts, no tips

.contrast = diffraction, interferences (difference in optical paths)…

.lateral resolution (16/84 criteria) ≈ 10 nm (< 2 nm with AC instruments)

.surface sensitivity ~ electron inelastic mean free path

W. Telieps, E. Bauer *Appl. Phys.* A **44** (1987) 55



Surface plasmon-polariton propagation – SNOM

Monocrystalline nanowire – 1D symmetry

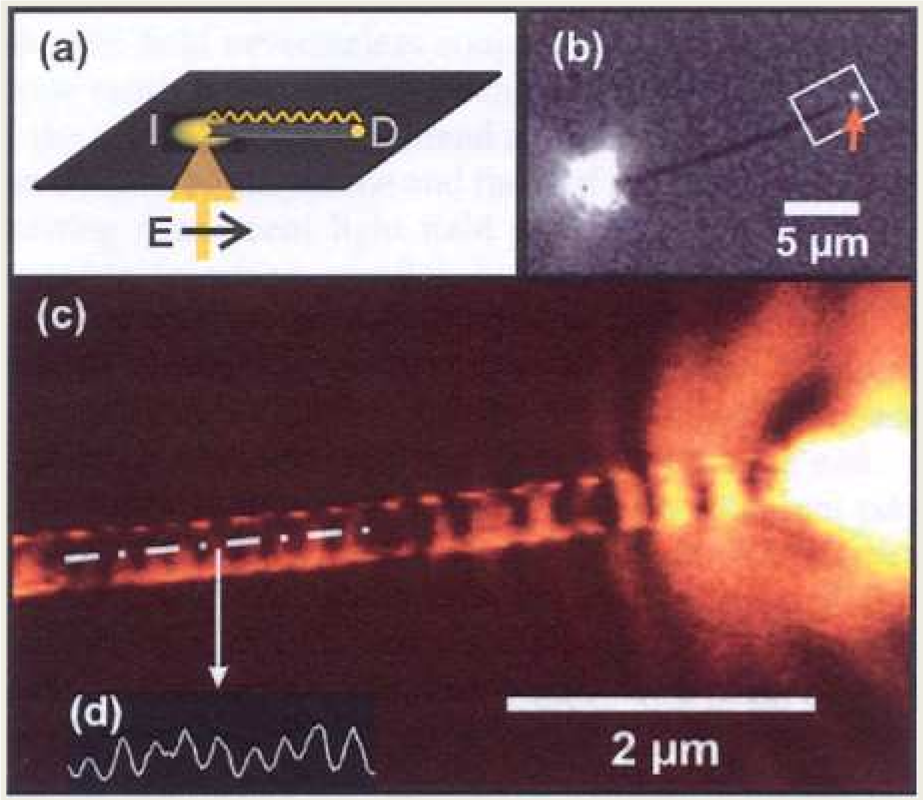


Figure captions

Monocrystalline Ag nanowire (length 18.6 µm, diameter 120 nm) under LASER excitation.

(a) excitation scheme,

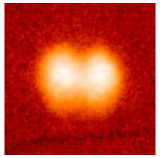
(b) topographic image,

(c) SNOM image of selected area (b),

(d) profile of the SNOM signal along the nanowire axis

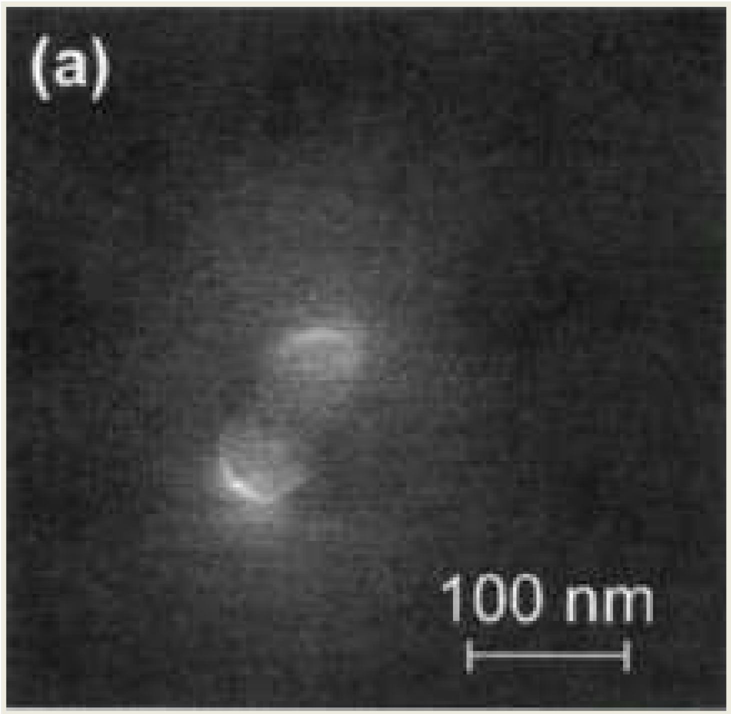
➢ **Surface plasmon polariton propagation along a nanowire**

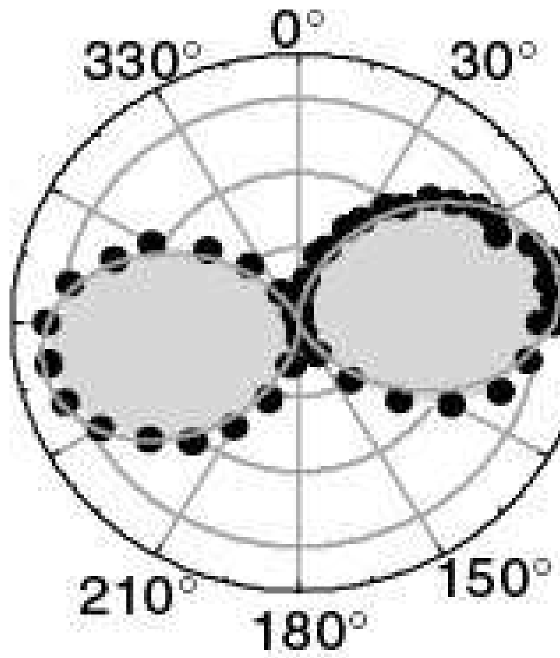
H. Ditlbacher *et al. Phys. Rev. Lett.* **95** (2005) 257403



Near-field mapping of single objects – PEEM

NanoRod antenna – Dipolar mode, m = 1



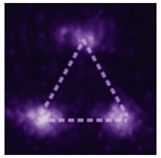


➢ Near-field photoluminescence imaging of an elliptic rod
rod = dimer of Au spheres, ∅ 40 nm.

➢ Polar diagram of the light intensity scattered by one single monocrystalline Au rod (100 x 20 x 20 nm^3).
➢ **dipolar emission** - dark field microscopy.

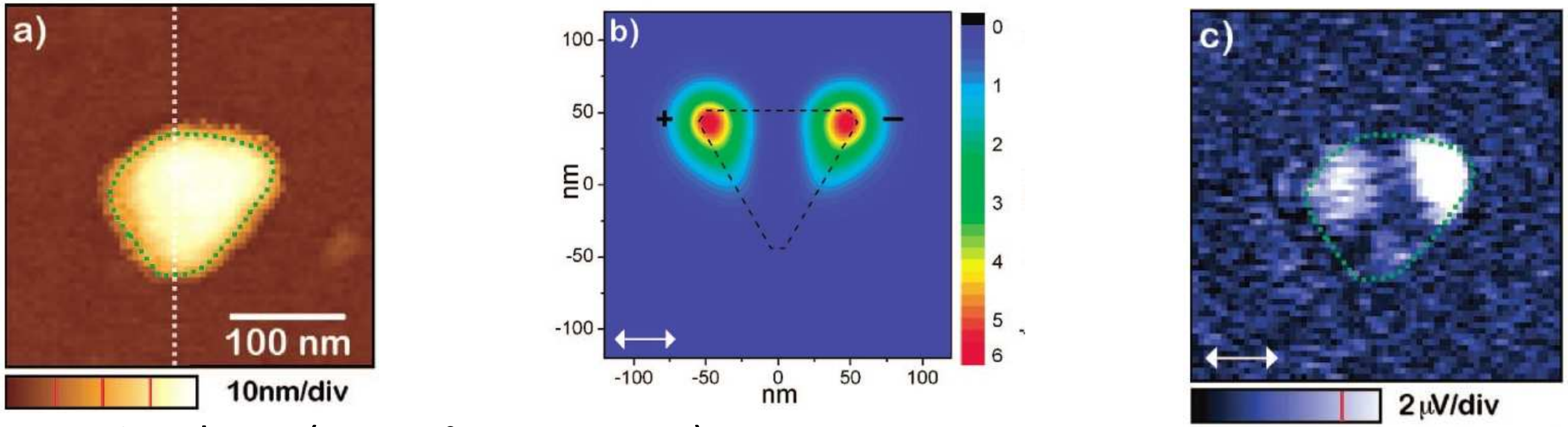
A. Bouhelier *et al. Appl. Phys. Lett.* **83** (2003) 5041

C. Sönnichen *et al. PRL* **88** (2002) 77402



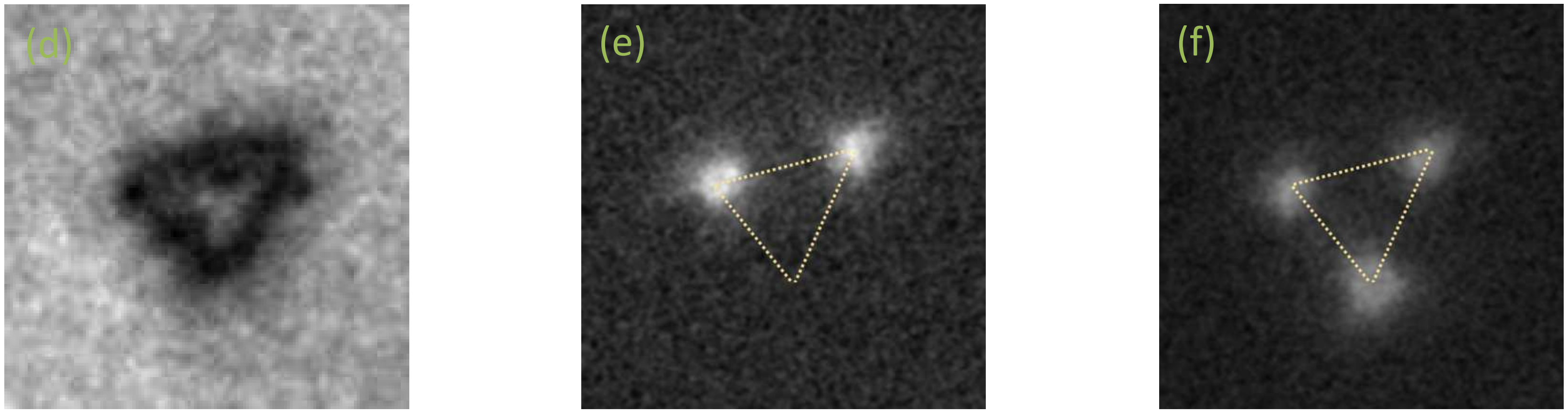
Plasmonics of Regular D_{3h} Nanotriangles - PEEM

Selective excitation of dipolar E' modes



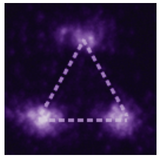
(a) AFM, triangle Ag (α = 60°, L = 104 nm)
(b) Simulation DDA $|E_z^2|$, polarisation S, grazing incidence
(c) **SNOM,** excitation 633 nm, polarisation S, grazing incidence

M. Rang *et al.*
Nano. Lett. **8** (2008) 3357



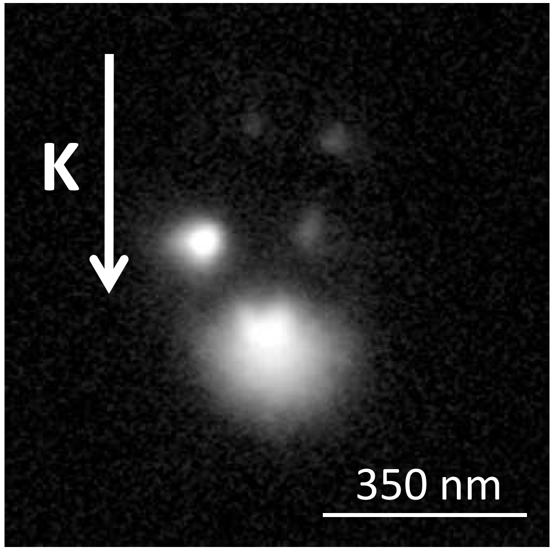
(d) **LEEM**, triangle Au EBL (α = 60°, L = 200 nm)
(e) **PEEM**, excitation 800 nm, S polarisation, grazing incidence
(f) **PEEM**, excitation 800 nm, P & S polarization superposition

C. Awada *et al.*
J. of Phys. Chem. C
16 (2012) 14591

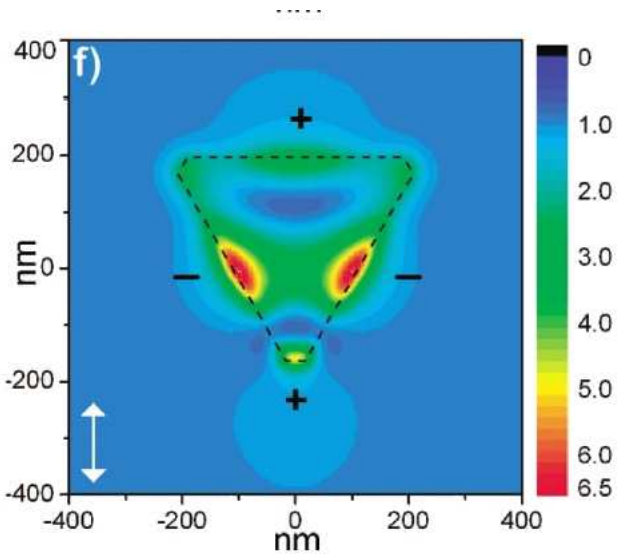


Plasmonics of Regular Nanotriangles

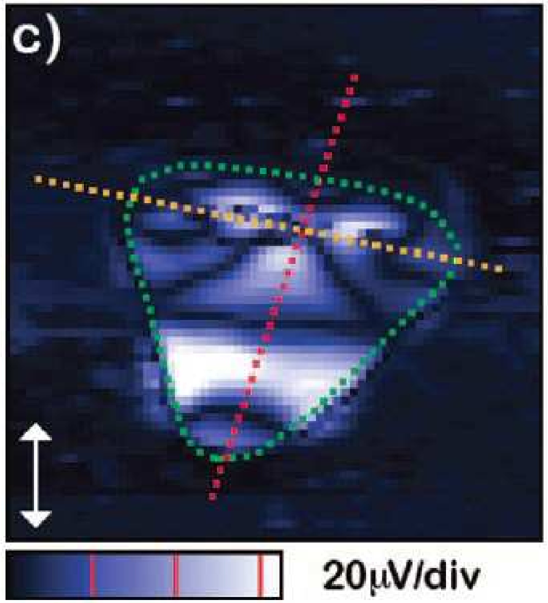
In-plane E' modes – Quadrupolar resonances



Triangle Au 300 nm x 50 nm
3-PPE PEEM
Excitation 730 nm
P polarisation
Incidence angle 14°
Quadrupolar mode



Triangle Ag 450 nm x 35 nm
Simulation DDA $|E_z^2|$
Excitation 633 nm HeNe
P polarisation
Incidence angle 20°
Quadrupolar mode



Triangle Ag 450 nm x 35 nm
SNOM
Excitation 633 nm HeNe
P polarisation
Incidence angle 20°
Quadrupolar mode

C. Awada *et al. J. of Phys. Chem. C* **16** (2012) 14591

M. Rang *et al. Nano. Lett.* **8** (2008) 3357

Plasmonics of Regular Nanotriangles

Group theory & Vectorial selection rules for dipolar resonances

D_{3h}	E	$2C_3(z)$	$3C'_2$	$\sigma_h(xy)$	$2S_3$	$3\sigma_v$	Linear fcts	Quad. fcts
A'_1	+1	+1	+1	+1	+1	+1		$x^2 + y^2$, z^2
A'_2	+1	+1	-1	+1	+1	-1	R_z	
E'	+2	-1	0	2	-1	0	(x, y)	$x^2 - y^2$, xy
A''_1	+1	+1	+1	-1	-1	-1		
A''_2	+1	+1	-1	-1	-1	+1	z	
E''	+2	-1	0	-2	+1	0	(R_x, R_y)	(xz, yz)

Top. Character table for the group D_{3h} **Bottom.** Product table for D_{3h} point group

	A'_1	**A'_2**	**E'**	**A''_1**	**A''_2**	**E''**
A'_1	A'_1	A'_2	E'	A''_1	A''_2	E''
A'_2	A'_2	A'_1	E'	A''_2	A''_1	E''
E'	E'	E'	$A'_1+A'_2+E'$	E''	E''	$A''_1+A''_2+E''$
A''_1	A''_1	A''_2	E''	A'_1	A'_2	E'
A''_2	A''_2	A''_1	E''	A'_2	A'_1	E'
E''	E''	E''	$A''_1+A''_2+E''$	E'	E'	$A'_1+A'_2+E'$

Selection rules: if the i^{th} component of the resonance mode $\mathbf{E_n}$, resp. illuminating light $\mathbf{E_{ext}}$ transforms as the irreducible representation $\Gamma_{n,i}$, resp. $\Gamma_{ext,i}$ (i = x, y, z), then the excitation strength of mode n $\langle E_n | E_{ext} \rangle$ vanishes unless there is a product $\Gamma_{n,i} \otimes \Gamma_{ext,i}$ transforming as the totally symmetric irrep A.

Illuminating wave $\mathbf{E_{ext}} = (E_{ext\ x}, E_{ext\ y}, E_{ext\ z})$ transforms as (x, y, z); the corresponding irreps are $(\Gamma_{ext\ x}, \Gamma_{ext\ y}, \Gamma_{ext\ z}) = (E', E', A''_2)$