

# Imaging Surface Plasmons

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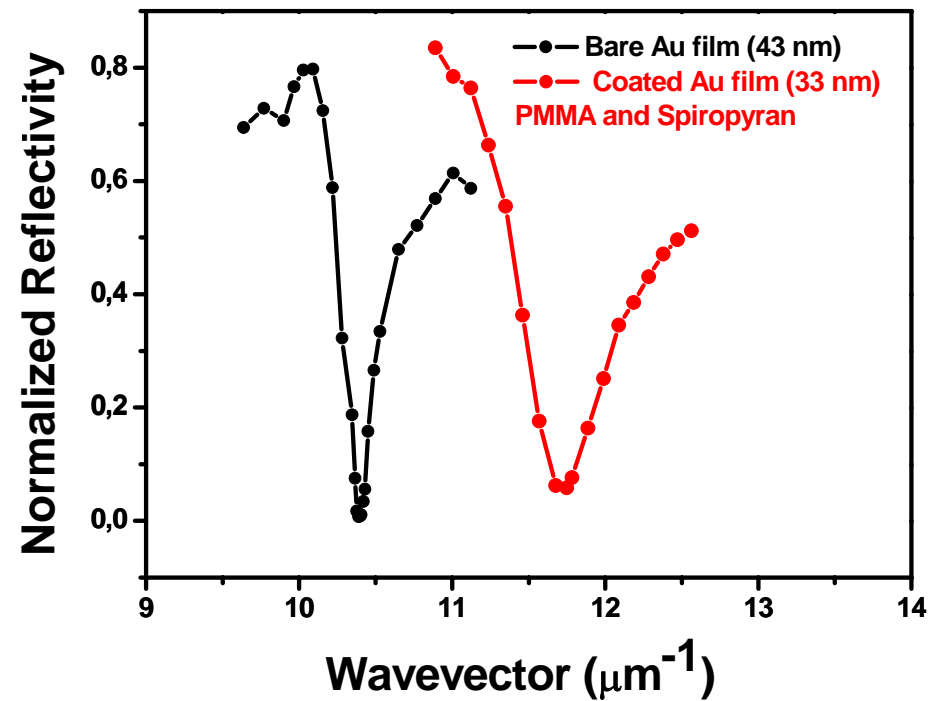
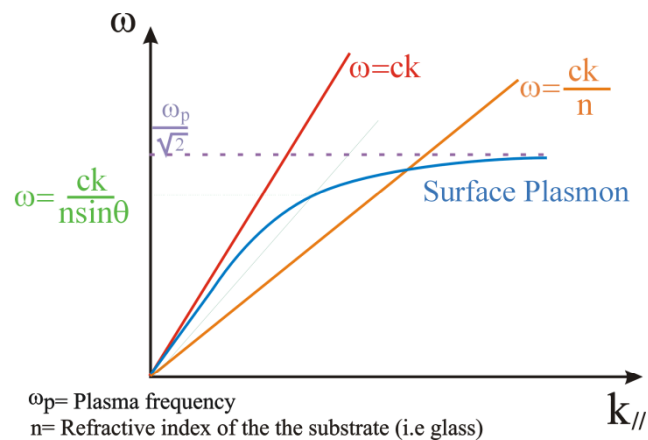
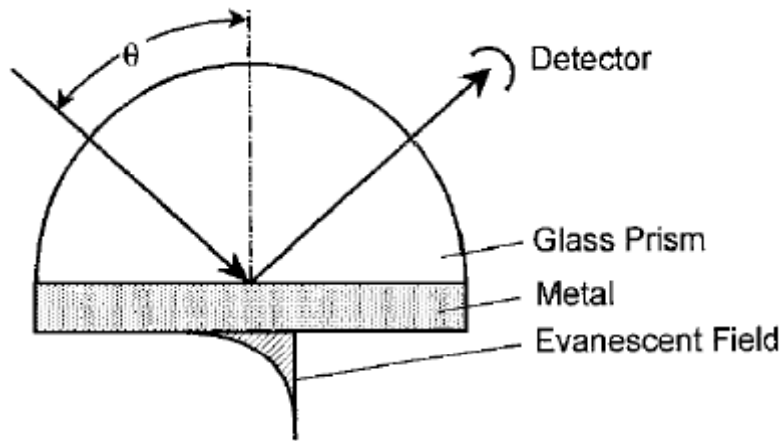
Université de Bourgogne, Dijon

France

# (almost) All there is to know about plasmons in 3 curves

1

## Reflectivity curve (Attenuated Total internal Reflection geometry)



# (Almost) all there is to know about plasmons in 3 curves

The reflectivity curve can be approximated as a Lorentzian

$$R = 1 - \frac{4\Gamma_i\Gamma_{rad}}{[k_x - (k_{spp})]^2 + [\Gamma_i + \Gamma_{rad}]^2}$$



$$k_{spp} = \frac{\omega}{c} \left( \frac{\varepsilon_1\varepsilon_2}{\varepsilon_1 + \varepsilon_2} \right)^{1/2} \pm \Delta k_x = k_x^0 \pm \Delta k_x$$

$$\Delta k_x = \left[ \frac{\omega}{c} \frac{2}{(1 + |\varepsilon_1|)} \left( \frac{|\varepsilon_1|}{|\varepsilon_1| - 1} \right)^{3/2} \exp(-2|k_x^0|d_1) \right] r_{01}^p(k_x^0)$$

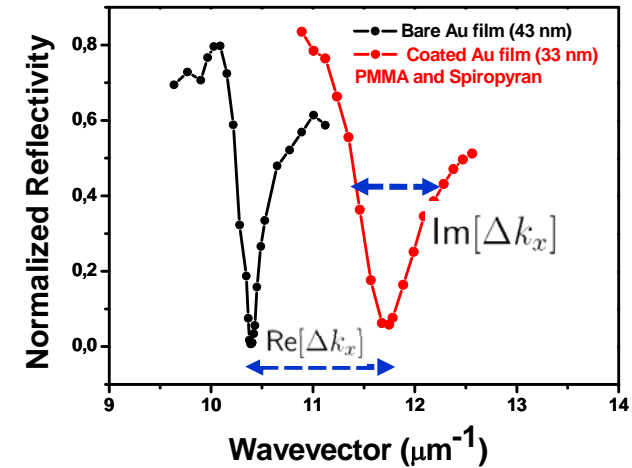
- Shift of the resonance:

$$\text{Re}[\Delta k_x] = \text{constant} \cdot \text{Re}[r_{01}^p(k_x^0)]$$

- Damping of the resonance (roughness scattering can be negligible):

Radiative losses:  $\Gamma_{rad} = \text{Im}[\Delta k_x] = \text{constant} \cdot \text{Im}[r_{01}^p(k_x^0)]$

Intrinsic losses:  $\Gamma_i = \text{Im}[k_x^0]$



Heinz Raether

## Surface Plasmons

on Smooth and Rough Surfaces  
and on Gratings

With 113 Figures

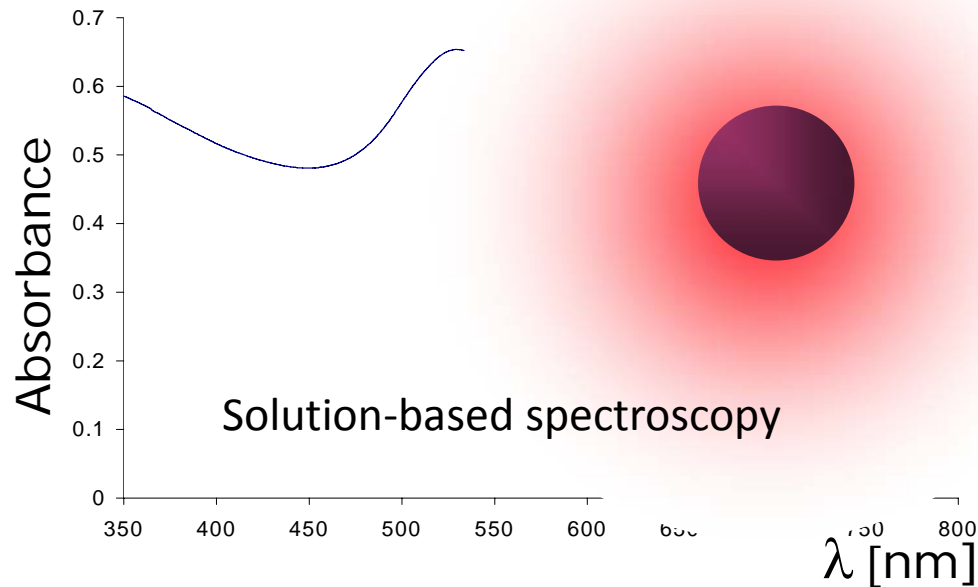


Springer-Verlag  
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London Paris Tokyo

# (Almost) all there is to know about plasmons in 3 curves

2

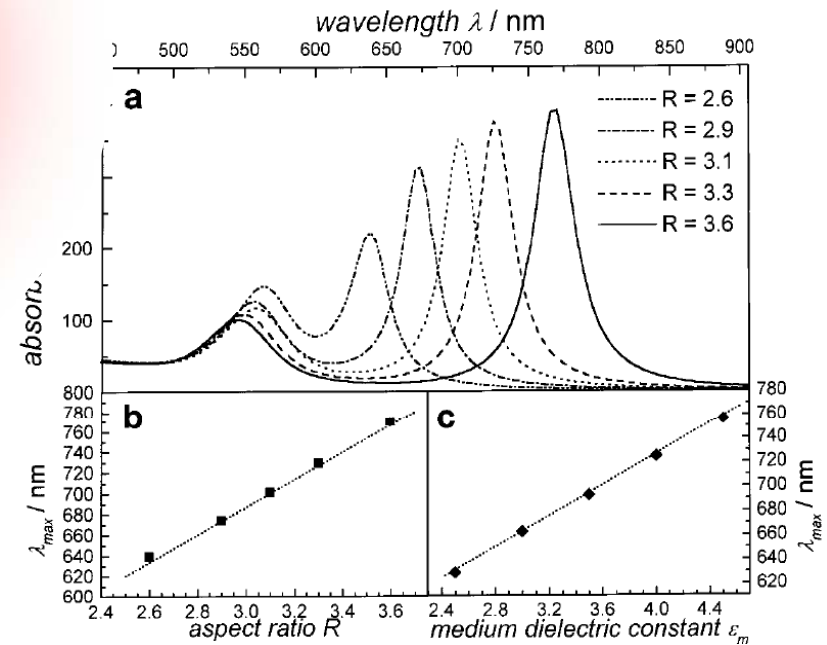
## Extinction spectroscopy



Halas Nanophotonics Group  
Rice University



C. Radloff



S. Link & M. El Sayed, J. Phys. Chem. B, 103, 8410 (1999)



# (Almost) all there is to know about plasmons in 3 curves

Mie theory

$$C_{abs} = \frac{6\pi V}{\lambda} \frac{3\epsilon_m''}{(\epsilon_m' + 2)^2 + \epsilon_m''^2}$$

Bohren & Huffman, *Absorption and Scattering of Light by Small Particles*, Wiley Inter Science

- Fröhlich frequency:  $\omega_f \Rightarrow \epsilon_m' = -2$

- Size correction  $\omega_f \Rightarrow \epsilon_m' = -2 + \beta \left(\frac{2\pi na}{\lambda}\right)^2$

- Surface polarization responsible for the field enhancement effect

$$\mathbf{E} = 4\pi a^3 \frac{\epsilon_m - 1}{\epsilon_m + 2} \mathbf{E}_0 + \mathbf{E}_0$$

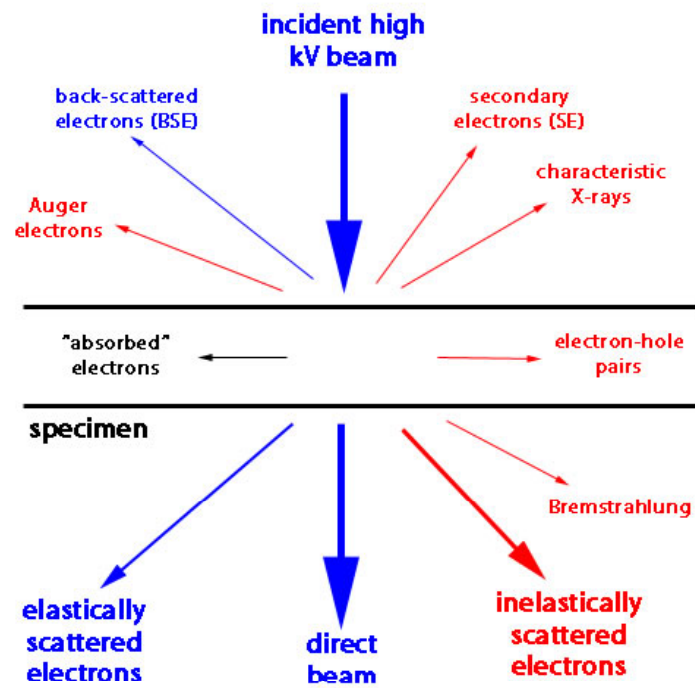
- Life time of the plasmon (dephasing time T2)

$$\tau_{sp} = T2/2 = \hbar/\Gamma$$

# (Almost) all there is to know about plasmons in 3 curves

3

## Electron energy-loss spectroscopy (EELS)



modified from Williams & Carter (1996) Fig. 1.3

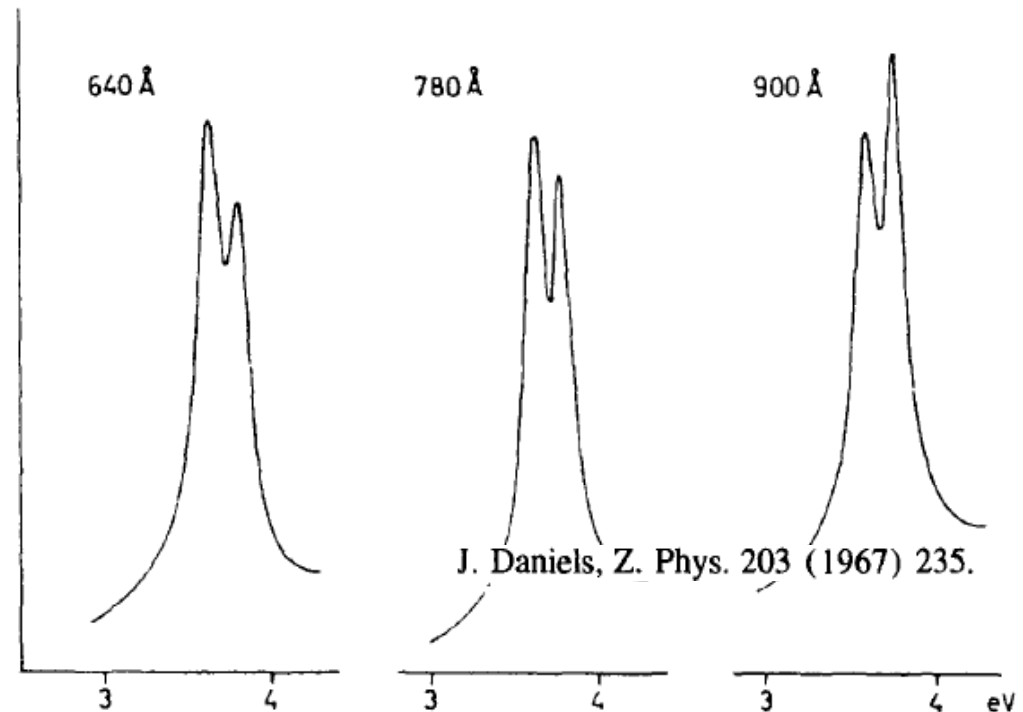
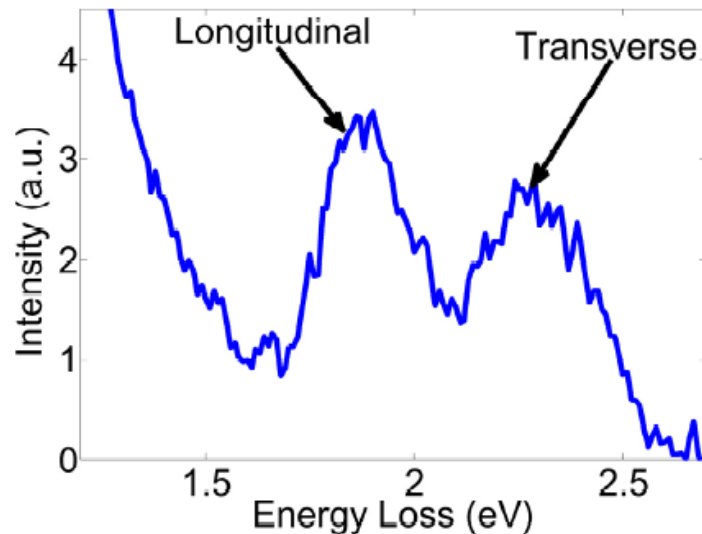


Fig. 5. EEL spectrum recorded with swift electrons ( $E_i = 50$  keV) for thin Ag foils of different thickness at room temperature. Bulk and surface plasmons are clearly resolved, at  $\hbar\omega_p = 3.78$  eV and  $\hbar\omega_{sp} = 3.63$  eV as  $\epsilon_2(\omega)$  nearly vanishes in this frequency range (from Ref. [65], used with permission).

# (Almost) all there is to know about plasmons in 3 curves



M. N'Gom & T. B. Norris, *The emerging frontier at the intersection of optics and electron microscopy*, SPIE Newsroom, 10.1117/2.1200901.1493

- Peaks in the EELS spectrum:

$$[\text{Im}(1/\epsilon)]^{\text{max}} \text{ (bulk)}$$

$$[\text{Im}(1/(\epsilon+1))]^{\text{max}} \text{ (surface)}$$

- Measure of the resonances (bulk, localized, surface)

- Measure of the relation dispersion at large wave-vectors (electron momentum) by momentum-resolved EELS

# Why the heck do we want to image surface plasmons?

With imaging comes control!

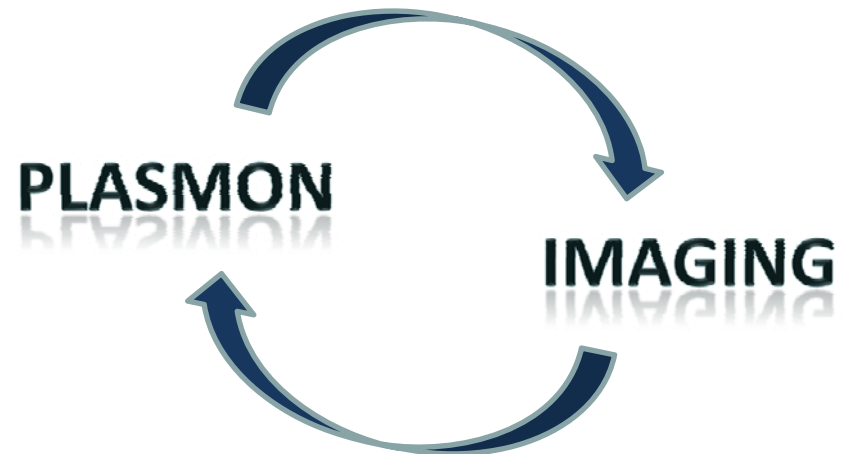
- ↳ Control over the propagation  
(plasmonic circuitry)
- ↳ Control over the localization  
(active substrates)

(the rebirth of plasmonics was partially triggered by our ability to visualize surface plasmons)

# Surface plasmon and imaging: an ambivalence



Early microscope



# Plasmon's principal imaging characteristics

- Surface plasmons are **evanescent** waves confined at a **surface** **Near-field microscopy**
- Surface plasmons can be **leaky** **Leakage microscopy**
- Surface plasmons can **interact** with molecules **Fluorescence microscopy**
- Localized surface plasmons can be scattered **Dark-field microscopy**  
**Cathodoluminescence**
- Surface plasmons can induce **non-linear** phenomena **Confocal microscopy**  
**Photoelectron microscopy**
- Surface plasmons are **loss** channel for  $e^-$  **STEM EELS**
- Surface plasmons can trigger photochemical reactions **Photochemical imaging**

# 1992: the plasmon microscope

VOLUME 68, NUMBER 4

PHYSICAL REVIEW LETTERS

27 JANUARY 1992

## Scanning Plasmon Near-Field Microscope

M. Specht, J. D. Pedarnig, W. M. Heckl, and T. W. Hänsch

*Sektion Physik der Universität München, Schellingstrasse 4/III, D-8000 München 40, Germany*

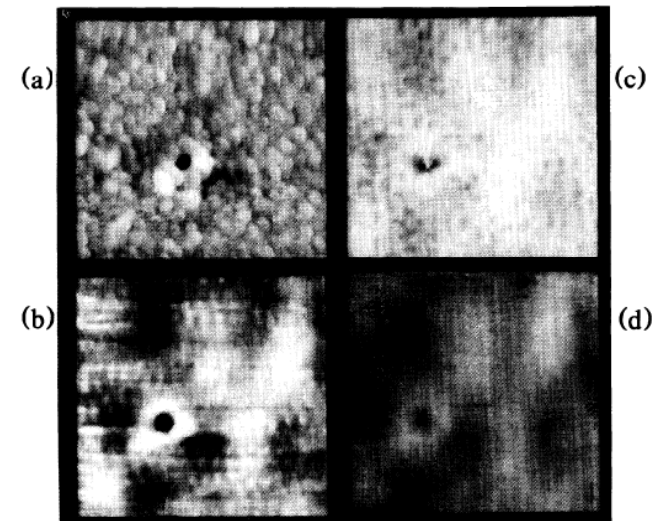
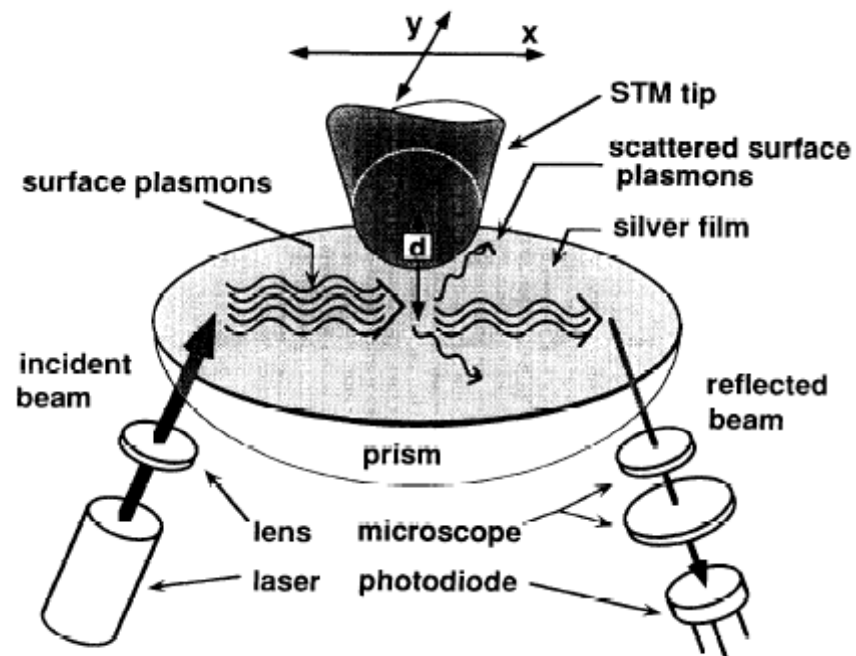


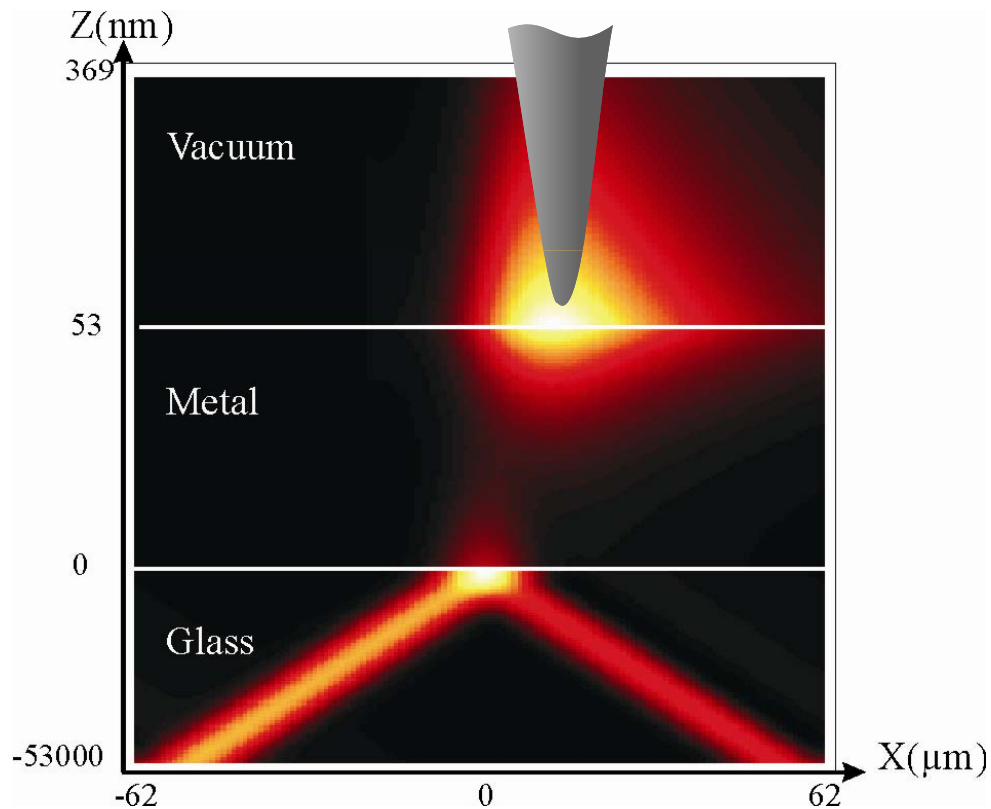
FIG. 2. (a) STM image taken at a  $600 \text{ nm} \times 600 \text{ nm}$  area on a silver surface (tip-sample spacing below  $1 \text{ nm}$ ); (b), (c) SPNM images taken at the same area (tip-sample spacing  $3$  and  $10 \text{ nm}$ , respectively; wavelength  $632.8 \text{ nm}$ ); (d) image recorded in the "STM-SPNM hybrid mode" (tip-sample spacing below  $1 \text{ nm}$ ).

# NEAR-FIELD OPTICAL MICROSCOPY

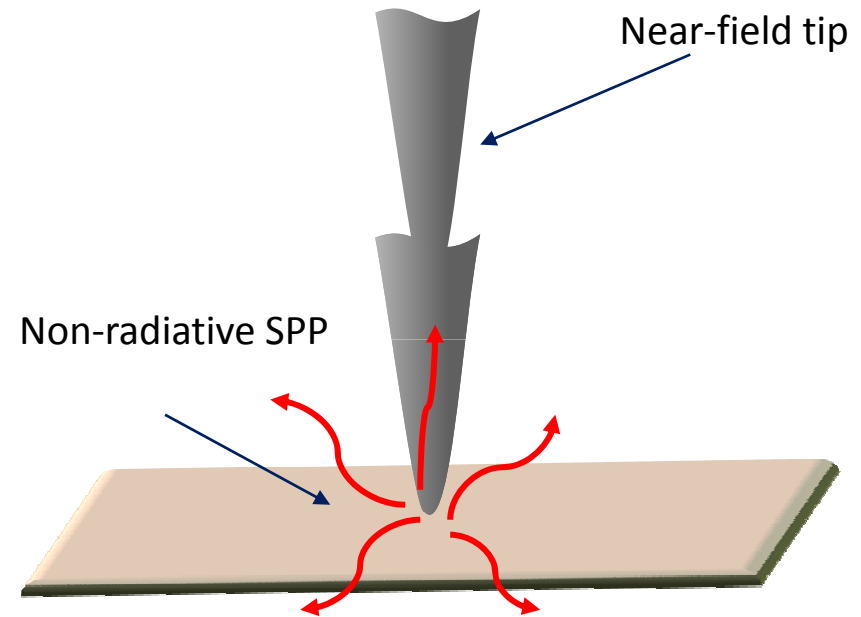
(the rebirth of plasmonics)



# Principle of near-field microscopy in the context



Intensity distribution of the plasmon field excited by Kretschmann configuration, Courtesy of Prof. F. Baida, FEMTO-ST, Besançon-France



**Requirement:** for a imaging a thin-film SPP, the spatial extend of the SPP  $>$  the excitation area (local excitation)

# Near-field microscopy: the first glance at a plasmon

VOLUME 72, NUMBER 18

PHYSICAL REVIEW LETTERS

2 MAY 1994

## Imaging of Surface Plasmon Propagation and Edge Interaction Using a Photon Scanning Tunneling Microscope

P. Dawson,<sup>1</sup> F. de Fornel,<sup>2</sup> and J-P. Goudonnet<sup>2</sup>

<sup>1</sup>*Department of Pure and Applied Physics, Queen's University of Belfast, Belfast BT7 1NN, United Kingdom*

<sup>2</sup>*Faculté Sciences Mirande, Laboratoire Physique du Solide, Equipe Optique Submicronique,  
Université de Bourgogne, BP 138, 21004 Dijon Cedex, France*

Excitation  
spot

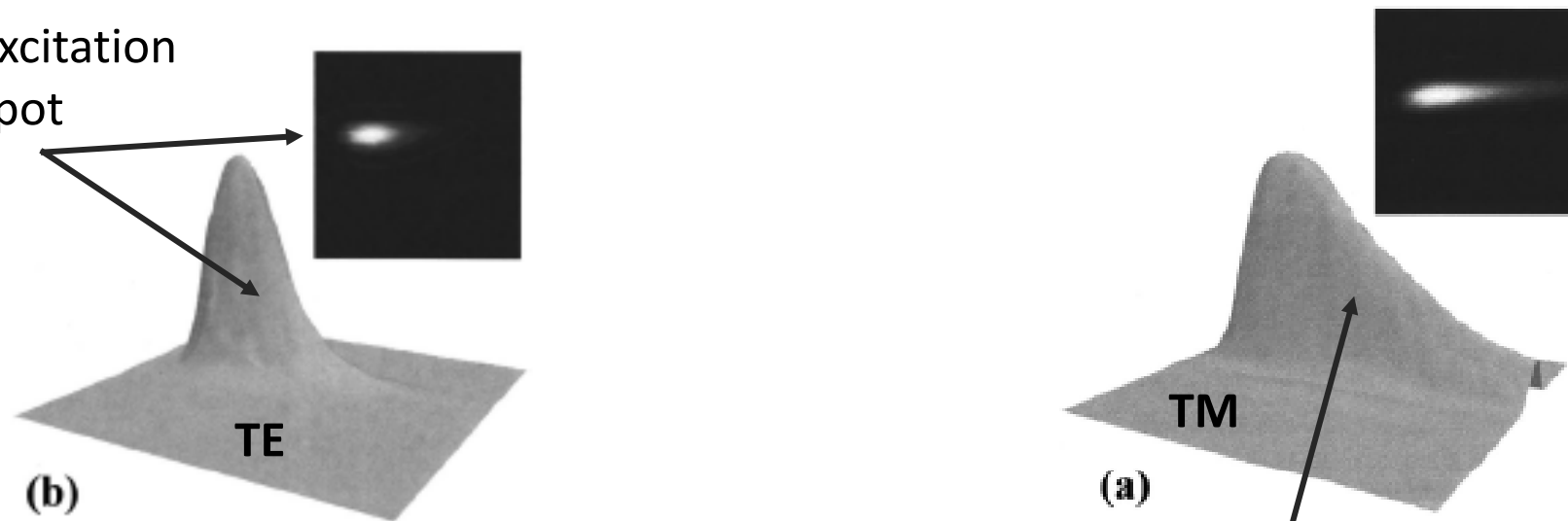
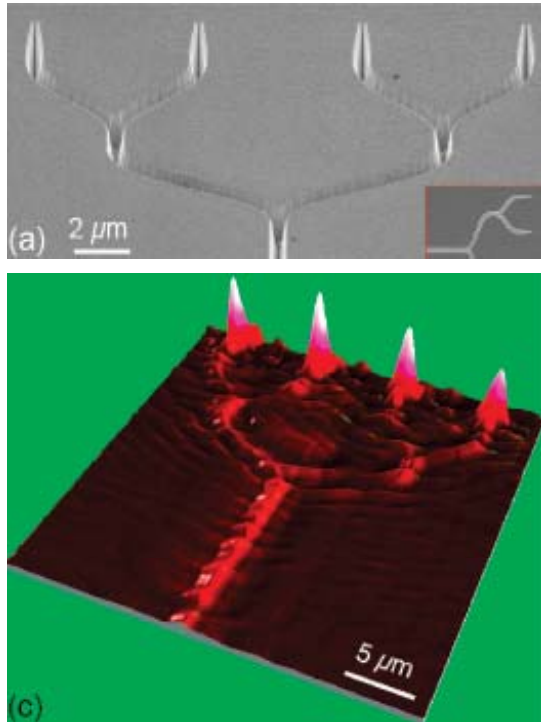


FIG. 5. PSTM images of SPP propagating (left to right) from a localized launch site at  $\lambda_1 = 632.8$  nm, the triangular coupling prism used in the setup is shown in Fig. 1. The scan range is  $36 \times 36 \mu\text{m}$  and insets show two-dimensional views of images. (a) Sample B1; measured  $1/e$  propagation length is  $13.8 \mu\text{m}$ . (b) Sample B3; measured  $1/e$  propagation length is  $4.9 \mu\text{m}$ .

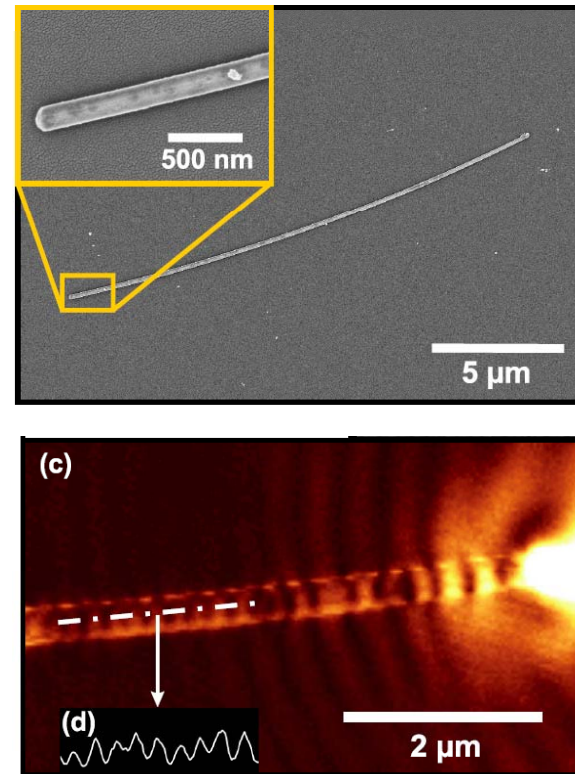
Exponentially decaying  
tail

# Understanding SPP propagation and interactions

SPP waveguides (here V-grooves and nanowires)



V. Volkov et al., *Nano. Lett.*, ASAP (2009)

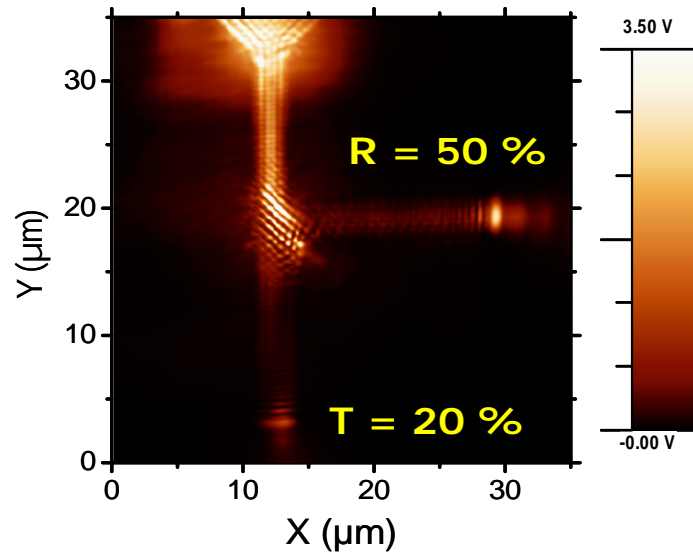
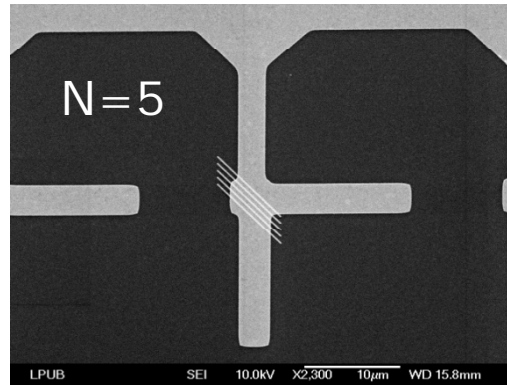


H. Ditlbacher et al., *Phys. Rev. Lett.*, 95, 257403 (2005)

Here, near-field imaging enabled the demonstration of true-subwavelength propagation in nanowaveguides

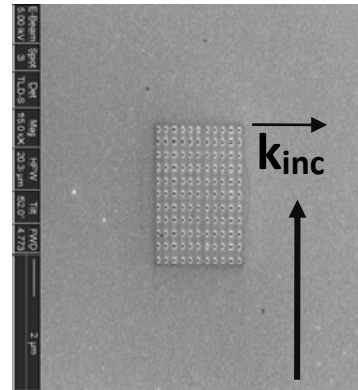
# Understanding SPP propagation and interactions

## SPP splitters



J-C Weeber, et al., *Appl. Phys. Lett.*, 87, 221101 (2005)

## SPP Launchers



J-Y. Laluet, et al., *Appl. Phys. Lett.*, 15, 3488 (2007)

Control over the propagation with passive elements

# Tracking SPP in space and time

## Heretodyned detection

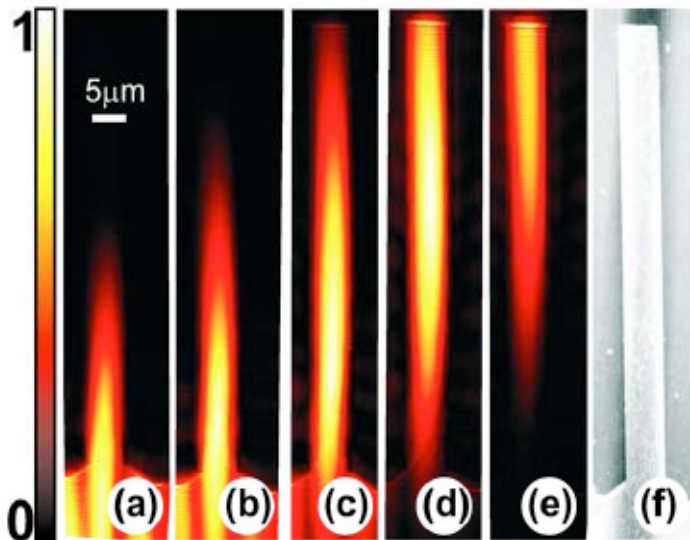
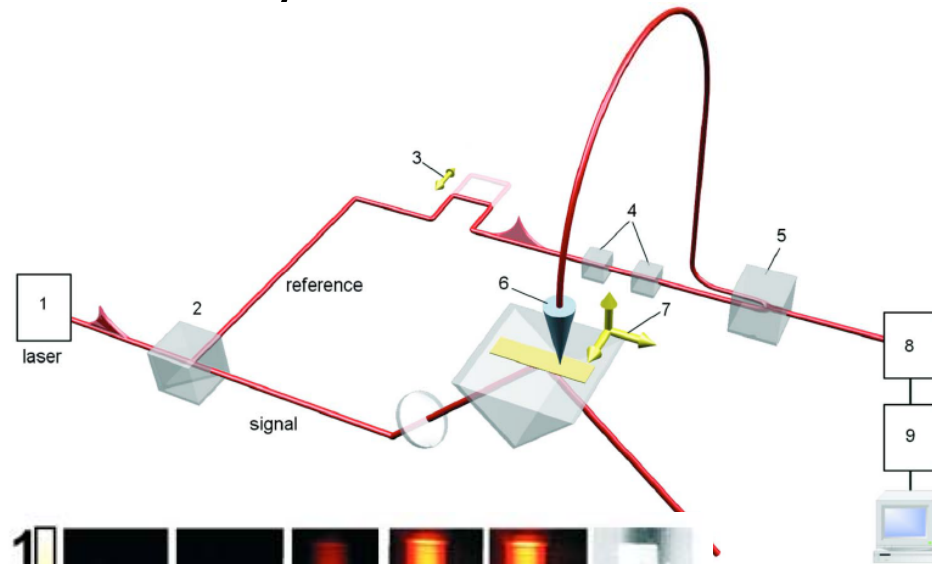


FIG. 9. (Color online) (a)–(e) Normalized amplitude information of the SPP wavepacket E-field. Succeeding frames are new scans of the probe. In between the frames the delay line is lengthened to  $14.4 \mu\text{m}$ . Therefore, the time between two frames is 48 fs. The scan frame is  $15 \times 110 \mu\text{m}^2$ , scan lines run from top to bottom. (f) Topography of the SPP waveguide obtained by shear-force feedback.

M. Sandke et al., *Rev. Sci. Instr.* **79**, 013704 (2008)

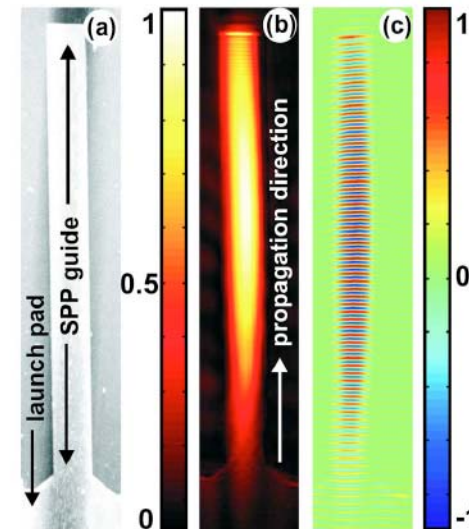


FIG. 8. (Color online) (a) Topography of the SPP waveguide obtained by shear-force feedback. It consists of a 55 nm thick Au guide,  $6 \mu\text{m}$  wide and  $80 \mu\text{m}$  long. (b) Normalized amplitude measurement of the E-field of the SPP wavepacket inside the  $6 \mu\text{m}$  waveguide. (c) Combined phase and amplitude information [ $\sim A \cos(\phi)$ ] of the same measurement as (b). Scan lines run from top to bottom and the scan frame is  $15 \times 110 \mu\text{m}^2$ , wavelength in air used to excite the SPPs: 1500 nm pulses with 20 nm bandwidth.



# Looking a confined plasmons using near-field optics

APPLIED PHYSICS LETTERS

VOLUME 83, NUMBER 2

14 JULY 2003

## Coherent imaging of nanoscale plasmon patterns with a carbon nanotube optical probe

R. Hillenbrand<sup>a)</sup>

*Nano-Photonics Group, Max-Planck-Institut für Biochemie, 82152 Martinsried, Germany*

F. Keilmann

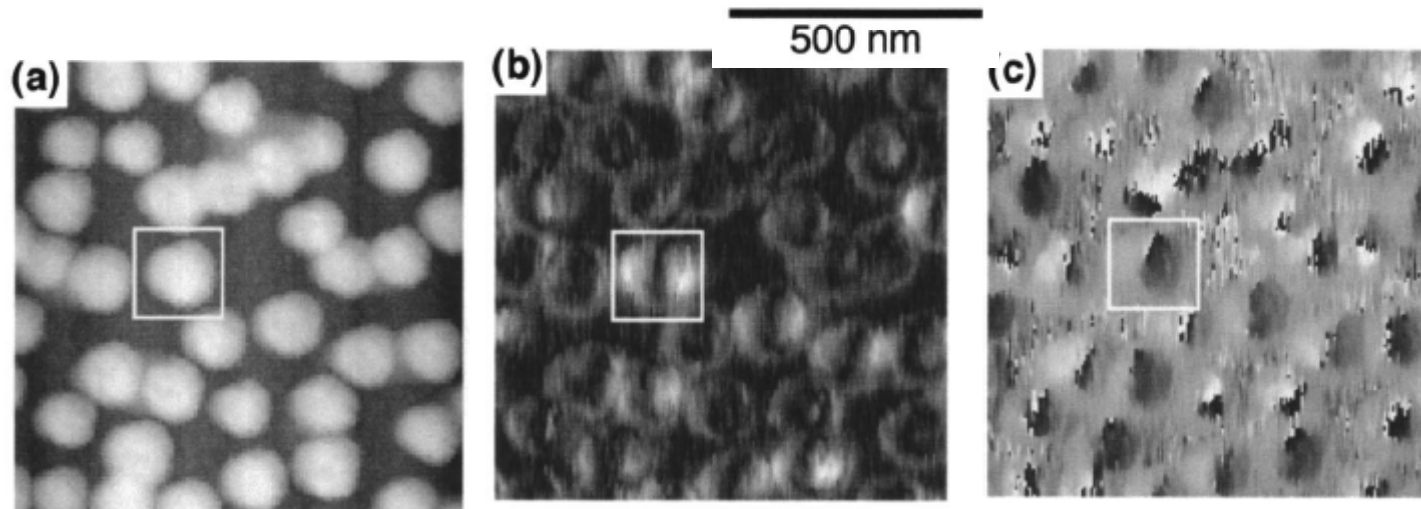
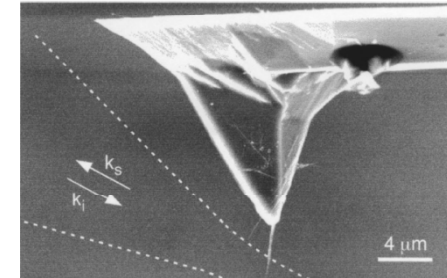
*Abteilung Molekulare Strukturbiologie, Max-Planck-Institut für Biochemie, 82152 Martinsried, Germany*

P. Hanarp and D. S. Sutherland

*Department of Applied Physics, Chalmers University of Technology, 41296 Göteborg, Sweden*

J. Aizpurua

*National Institute of Standards and Technology, Gaithersburg, Maryland 20899-8423*



Localized SPP have dipolar-like distribution (amplitude and phase)

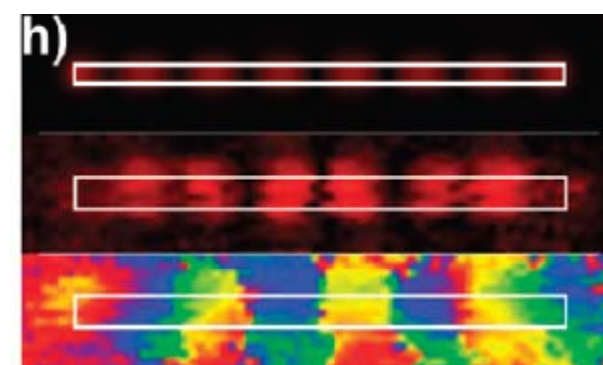
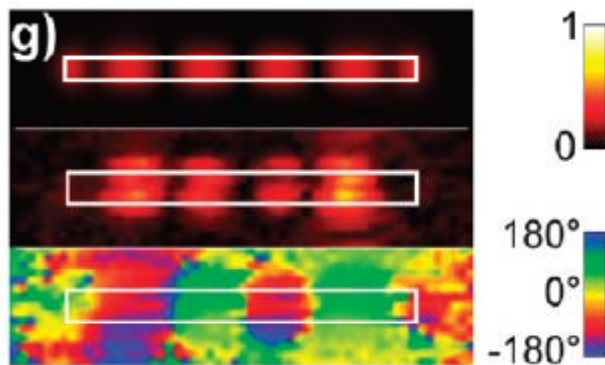
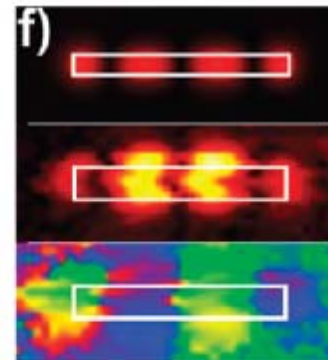
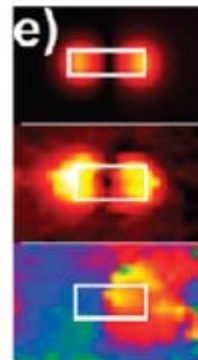
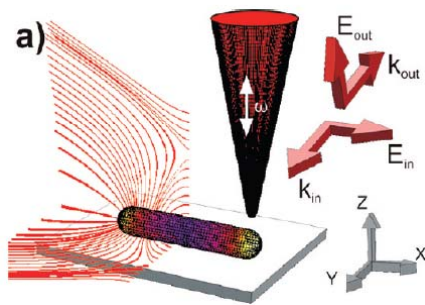
# Looking at confined plasmons using near-field optics

## Fabry-Pérot Resonances in One-Dimensional Plasmonic Nanostructures

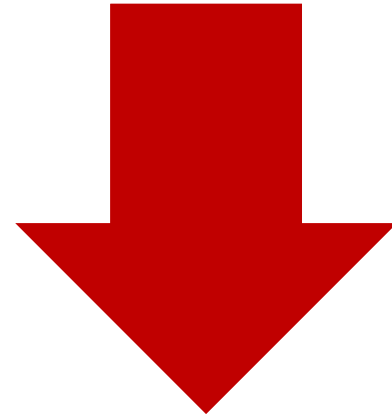
NANO  
LETTERS

2009  
Vol. 9, No. 6  
2372-2377

Jens Dorfmueller,<sup>\*,†</sup> Ralf Vogelgesang,<sup>\*,†</sup> R. Thomas Weitz,<sup>†</sup> Carsten Rockstuhl,<sup>‡</sup>  
Christoph Etrich,<sup>¶</sup> Thomas Pertsch,<sup>¶</sup> Falk Lederer,<sup>‡</sup> and Klaus Kern<sup>†,§</sup>



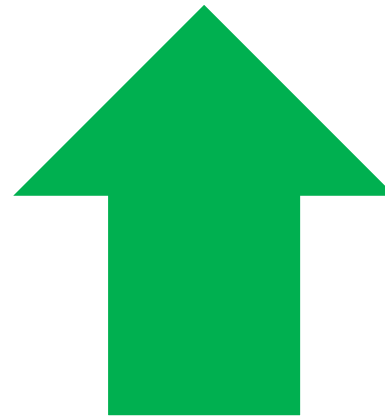
# Pro et contra of near-field imaging



Resolution (still) strongly  
depends on tip quality  
Complex measurements  
Perturbative



Access to the bound E.M. field  
Spectroscopic data  
Space and time investigations  
Flexible with sample  
requirements  
Phase measurement

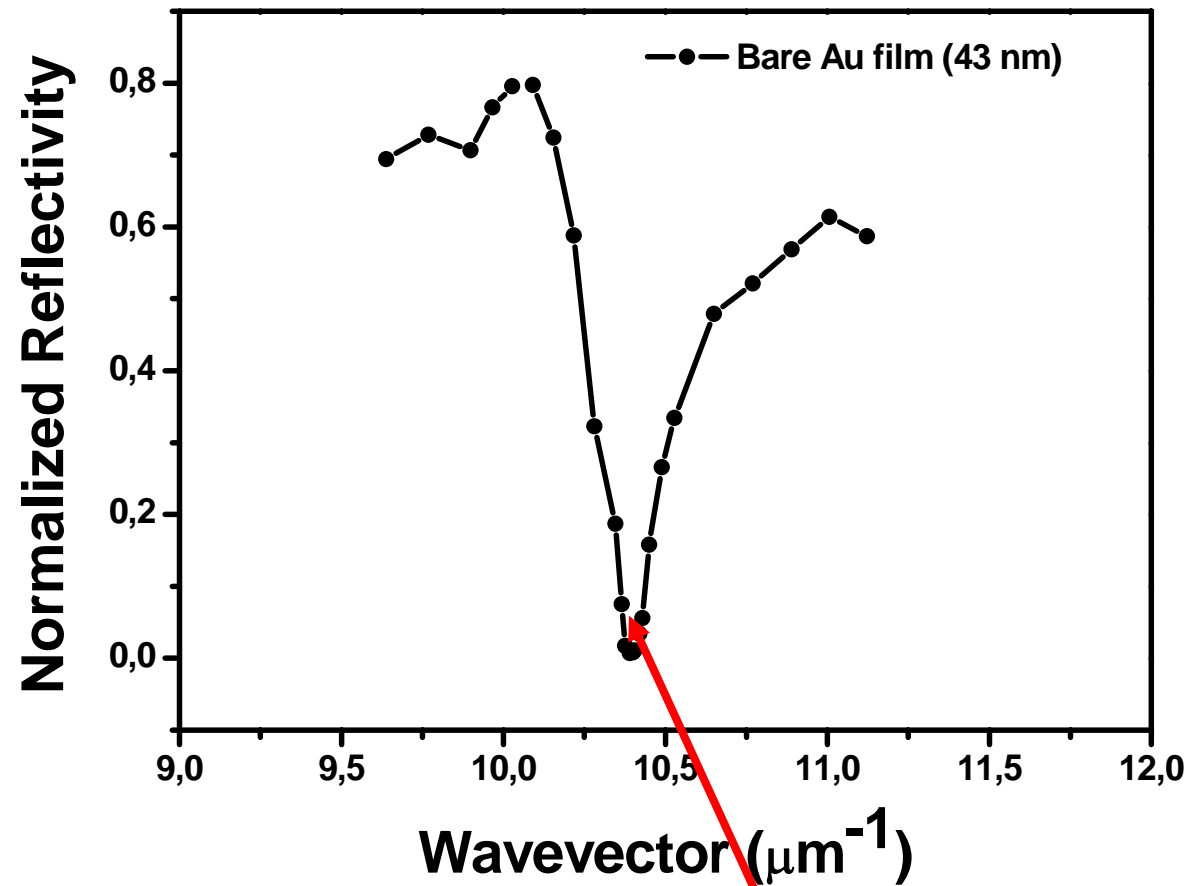




# LEAKAGE RADIATION MICROSCOPY

(naked-eye plasmons)

# Hand-wavy argument

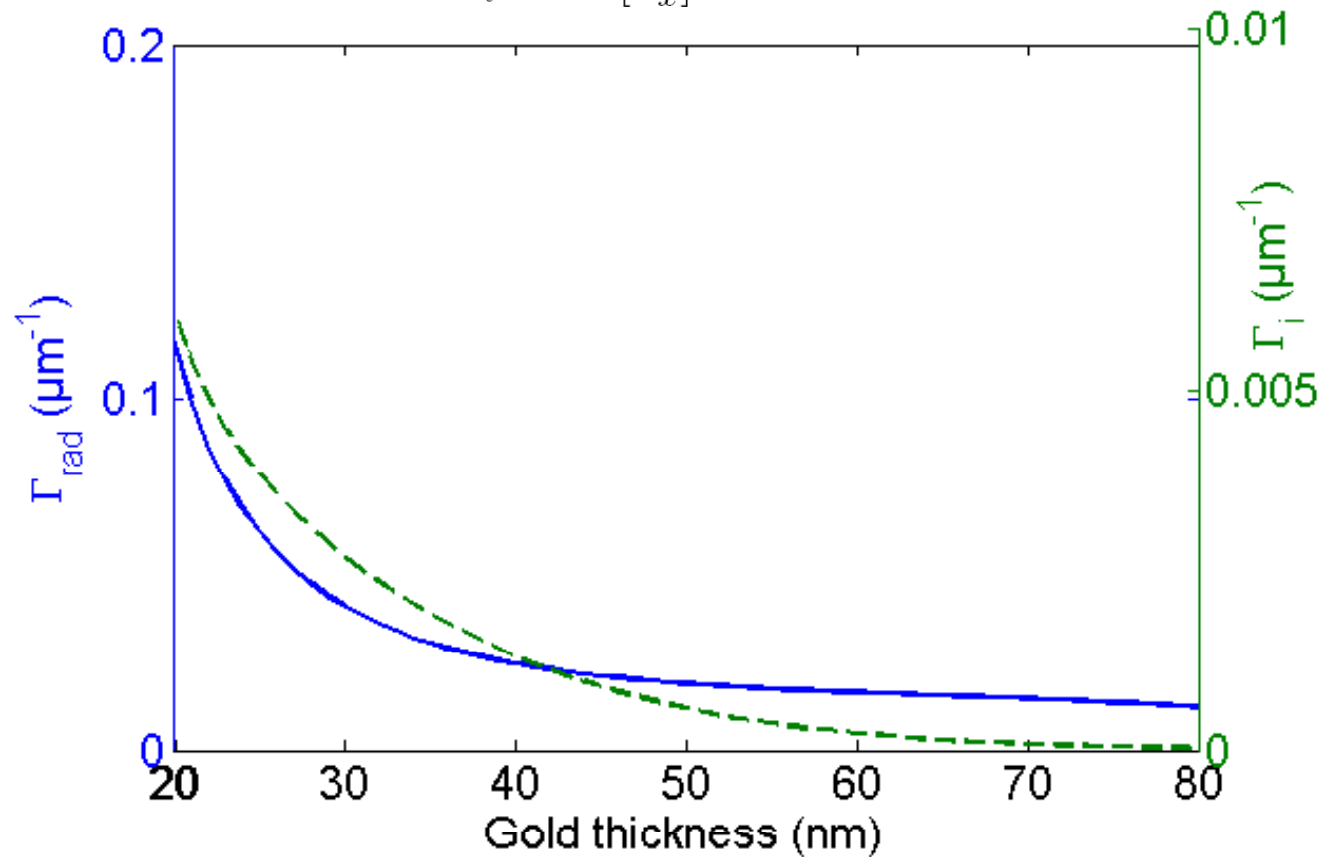


**Signature of the SP; the incident energy is coupled to the plasmon**

# Hand-wavy argument

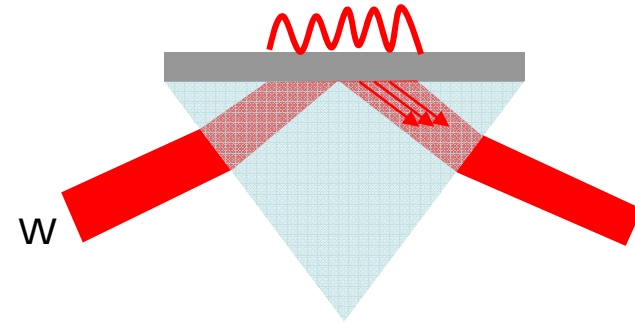
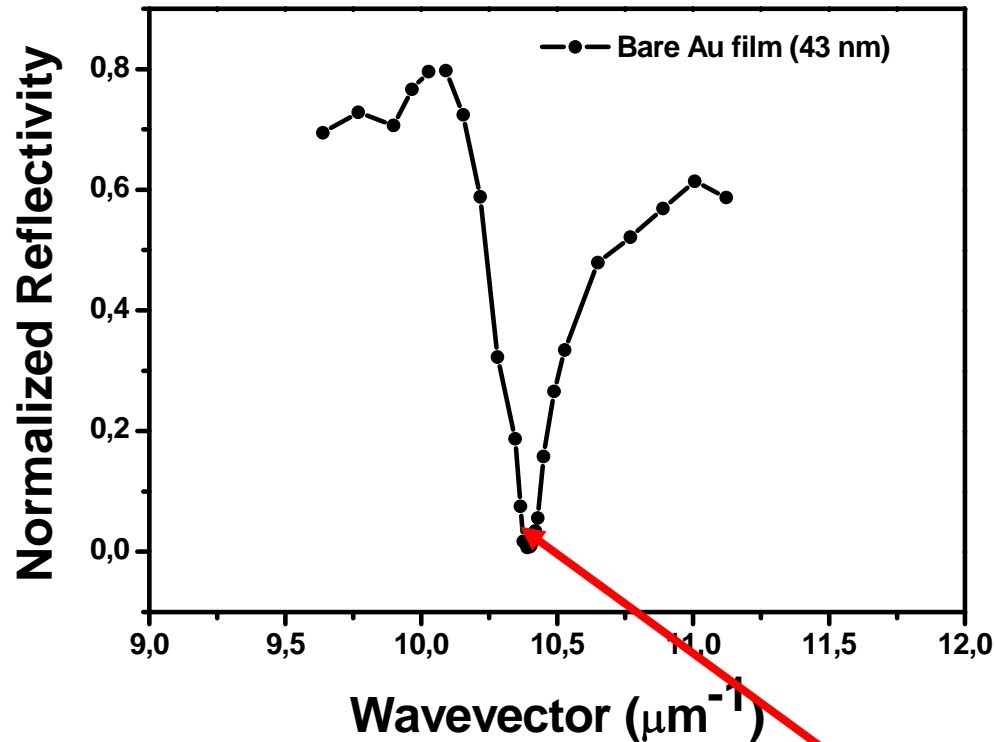
Radiative losses:  $\Gamma_{rad} = \text{Im}[\Delta k_x] = \text{constant} \cdot \text{Im}[r_{01}^p(k_x^0)]$

Intrinsic losses:  $\Gamma_i = \text{Im}[k_x^0]$



J. Grandidier, PhD thesis 2009, Université de Bourgogne

# Hand-wavy argument

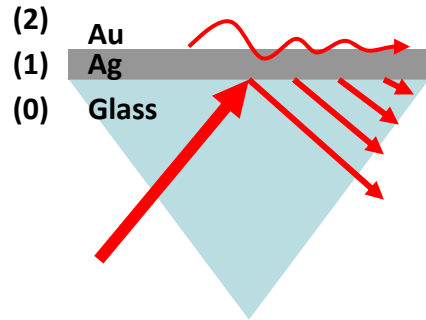


Finite-size beam ( $w=\text{mm}$ )  $\gg$   
SPP attenuation length

Signature of the SP; leakage  
radiation are interfering  
destructively with reflected beam

$$T+R+A=1 \quad T=R=0 \quad \rightarrow \quad A=1$$

# Rigorous basis



$$k_{z0} = \left( \varepsilon_o \left( \frac{\omega}{c} \right)^2 - k_x^2 \right)^{1/2}$$

$$k_{z1} = \left( \varepsilon_1 \left( \frac{\omega}{c} \right)^2 - k_x^2 \right)^{1/2}$$

$$k_{z2} = \left( \varepsilon_2 \left( \frac{\omega}{c} \right)^2 - k_x^2 \right)^{1/2}$$

$$k_x = k_{spp} = \sqrt{\varepsilon_o} \frac{\omega}{c} \sin \theta$$

$$k_{spp} = \frac{\omega}{c} \left( \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \right)^{1/2}$$

$$k_{z2} = \left( \frac{\omega}{c} \right) \sqrt{(\varepsilon_2 - \varepsilon_o \sin^2 \theta)}$$

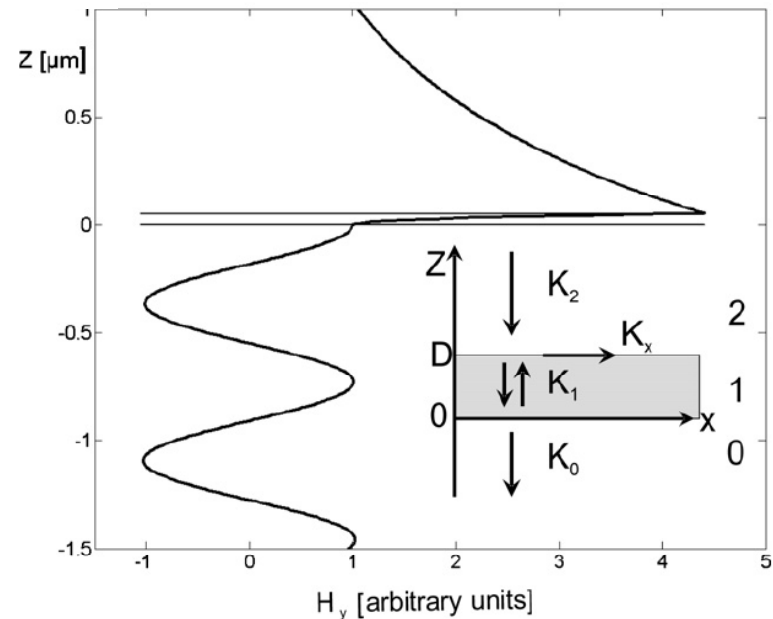
inhomogeneous wave

$$k_{z1} = \left( \frac{\omega}{c} \right) \sqrt{(\varepsilon_1 - \varepsilon_o \sin^2 \theta)}$$

inhomogeneous wave

$$k_{z0} = \sqrt{\varepsilon_o} \left( \frac{\omega}{c} \right) \sqrt{(1 - \sin^2 \theta)}$$

homogeneous wave

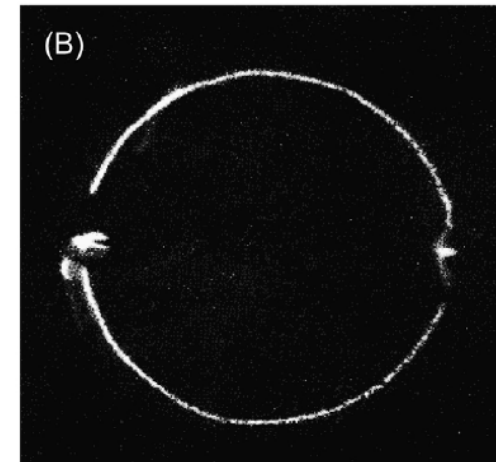
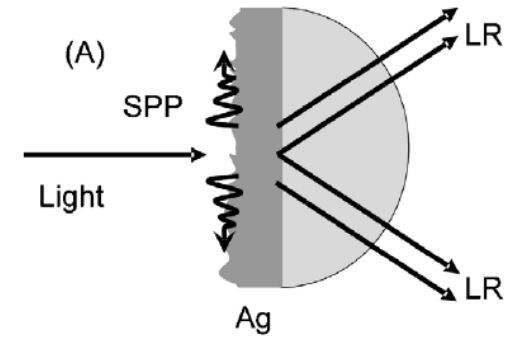
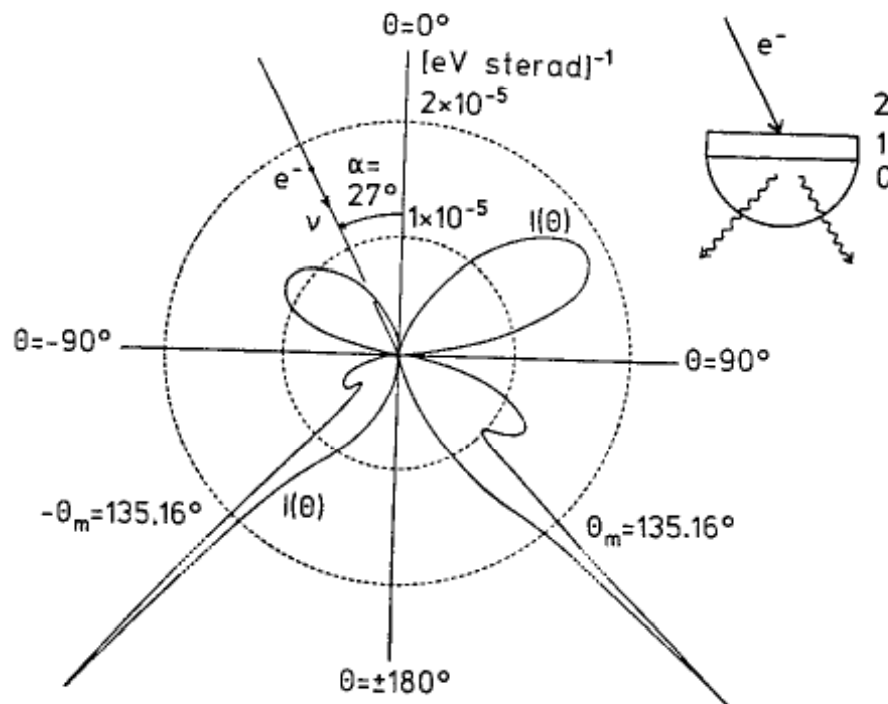


# First measurements

Z. Physik 244, 1-5 (1971)  
 © by Springer-Verlag 1971

## Light Emission from Non Radiative Plasmons Excited by Electrons on Smooth Surfaces

E. KRÖGER and H. RAETHER  
 Institut für Angewandte Physik der Universität Hamburg



Simon & J.K. Guha, *Opt. Comm.*, 18, 391 (1976)  
 A. Drezet et al., *Mat. Sci. Eng. B.* 149, 220 (2008)

# Two-dimensional mapping of SPP by LRM

VOLUME 77, NUMBER 9

PHYSICAL REVIEW LETTERS

26 AUGUST 1996

## Local Excitation, Scattering, and Interference of Surface Plasmons

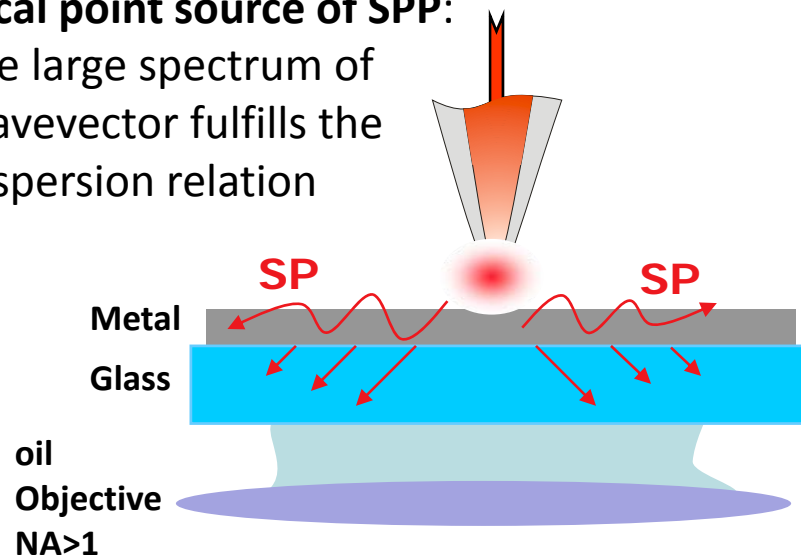
B. Hecht,<sup>1</sup> H. Bielefeldt,<sup>1</sup> L. Novotny,<sup>2</sup> Y. Inouye,<sup>1,\*</sup> and D. W. Pohl<sup>1,†</sup>

<sup>1</sup>IBM Research Division, Zurich Research Laboratory, CH-8803 Rüschlikon, Switzerland

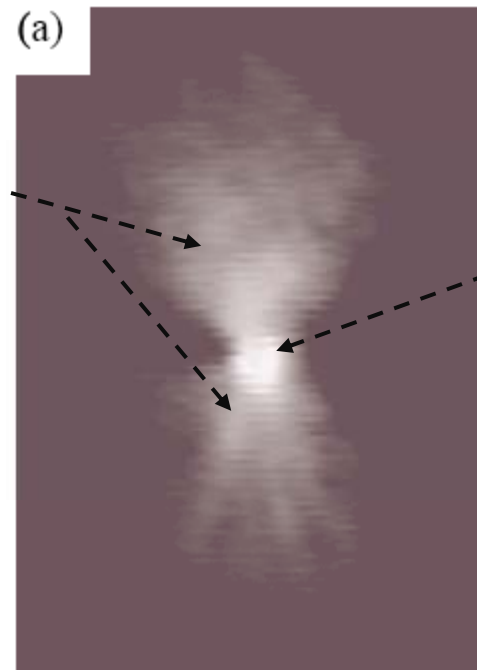
<sup>2</sup>Swiss Federal Institute of Technology, Eidgenössische Technische Hochschule Zürich, CH-8092 Zurich, Switzerland

(Received 14 December 1995; revised manuscript received 29 March 1996)

The near-field tip acts as a **local point source of SPP**: the large spectrum of wavevector fulfills the dispersion relation



SPP "wings"



Launching site  $\ll L_{\text{spp}}$

# Imaging SPP interactions

PHYSICAL REVIEW B, VOLUME 63, 155404

## Plasmon optics of structured silver films

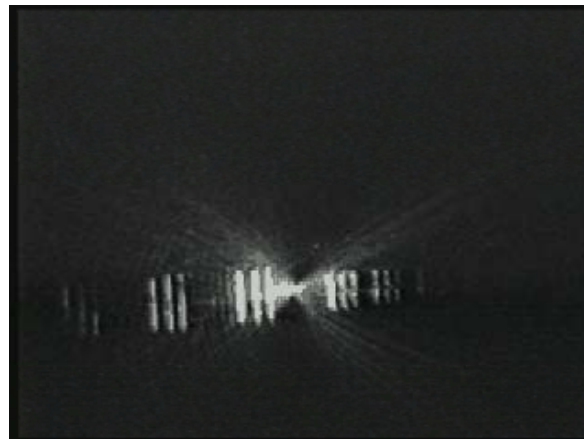
A. Bouhelier,<sup>\*</sup> Th. Huser,<sup>†</sup> H. Tamaru,<sup>‡</sup> H.-J. Güntherodt, and D. W. Pohl  
*Institute of Physics, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland*

Fadi I. Baida and D. Van Labeke  
*Laboratoire d'Optique PM Duffieux, CNRS UMR 6603, Institut des Microtechniques de Franche-Comté, FR0067 CNRS,  
Université de Franche-Comté, F-25030 Besançon Cedex, France*

The launch of a SPP



Polarization control



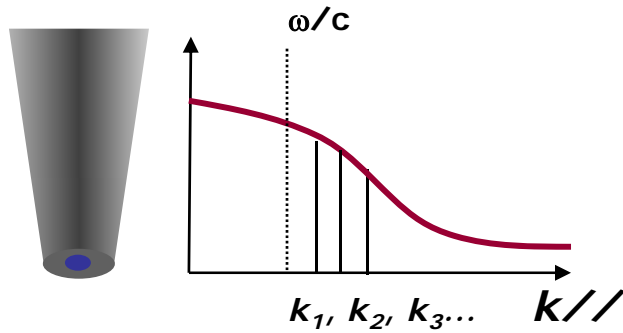
SPP interference



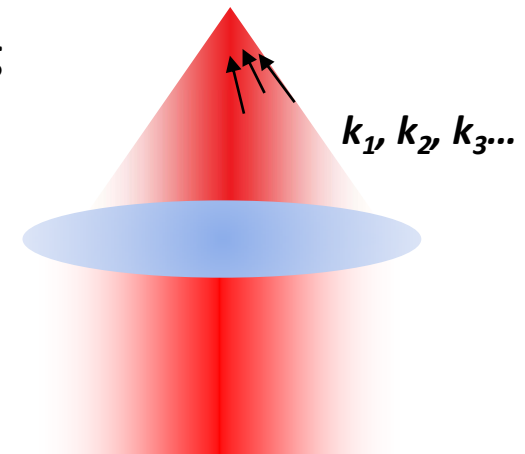


# Going far-field (SPP imaging made easy)

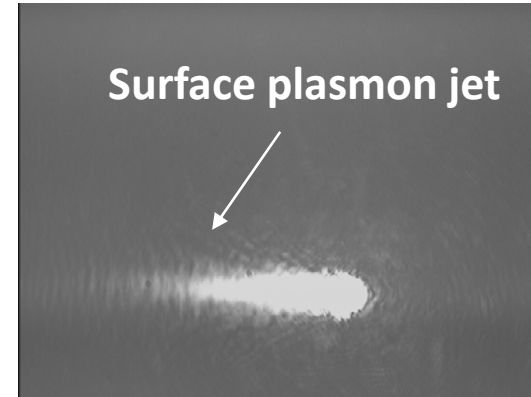
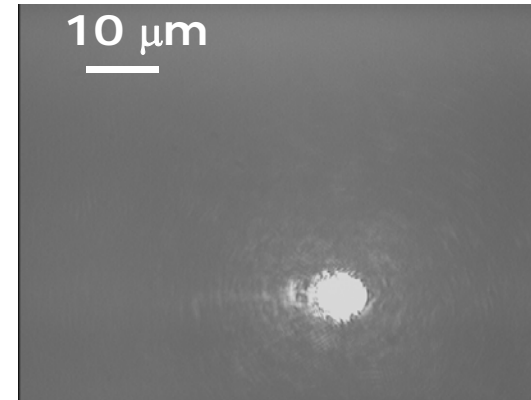
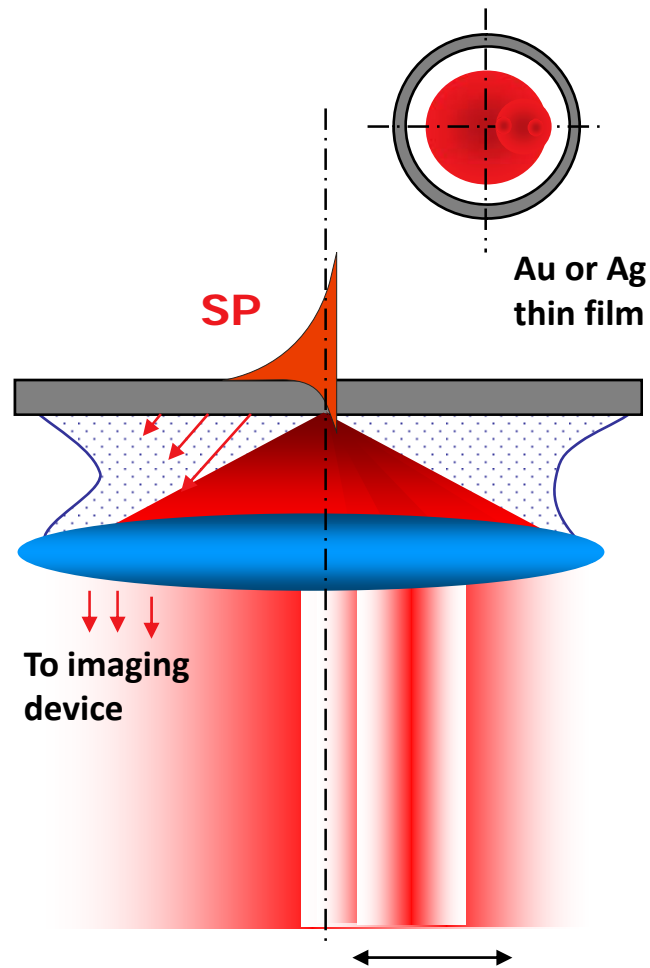
Near-field coupling



Focusing



# Going far-field (SPP imaging made easy)



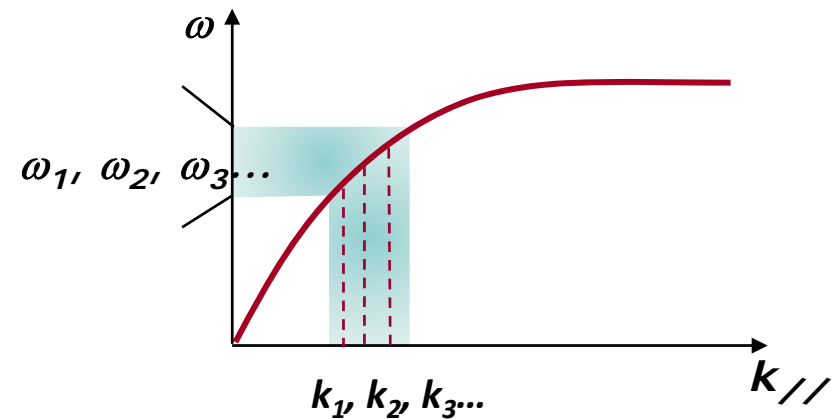
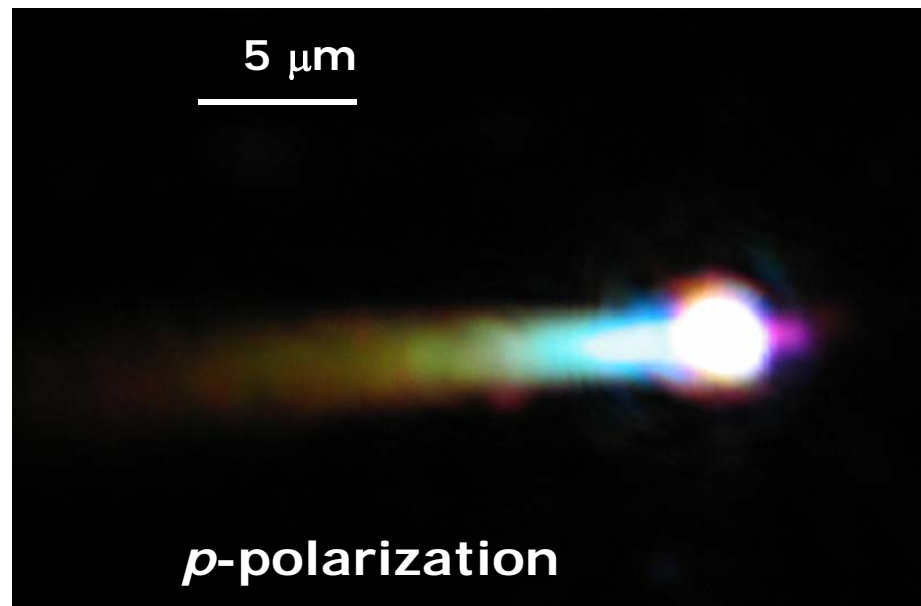
To be adjusted for  
SP excitation ( $k_{spp}$ )

A. Bouhelier and G. P. Wiederrecht, *Opt. Lett.*, **30**, 884 (2005)

A. Hohenau et al, *Opt. Lett.*, **30**, 893 (2005)

# Multicolor plasmon jet

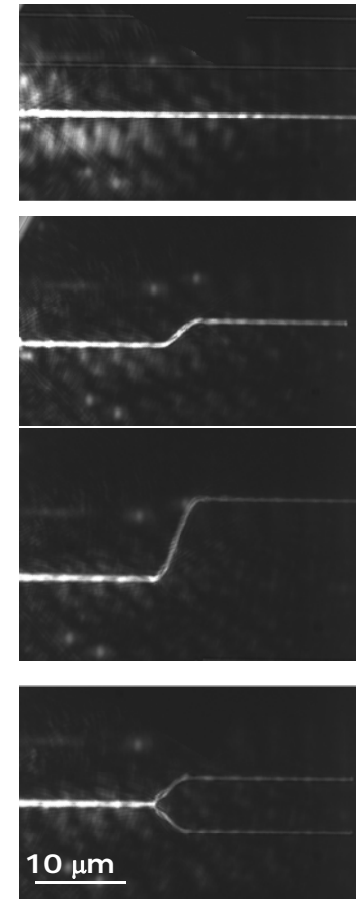
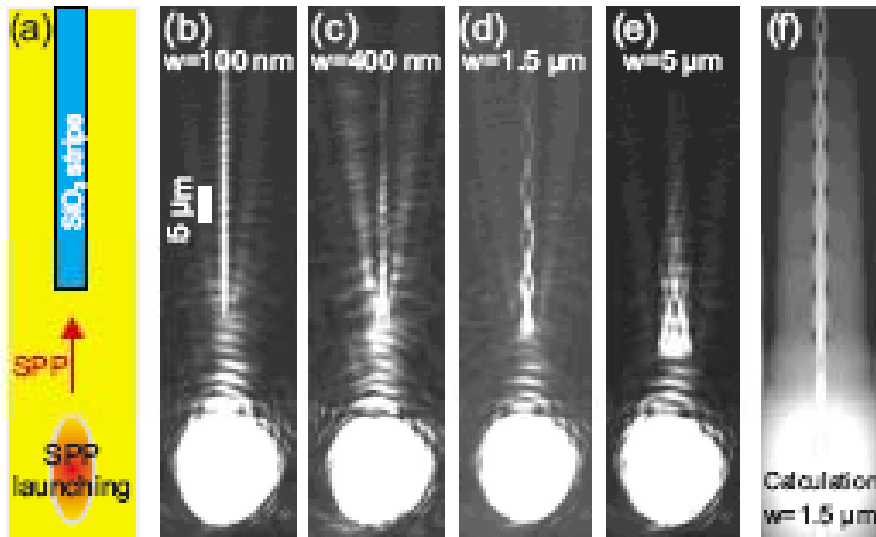
- Broad-band surface plasmon excited from a white-light femtosecond laser source
- Color spread reflects the surface plasmon dispersion: group velocities ( $d\omega/dk$ ) change with color



# SPP waveguiding imaged by LRM

- Details of the propagation of SPP in waveguiding structures

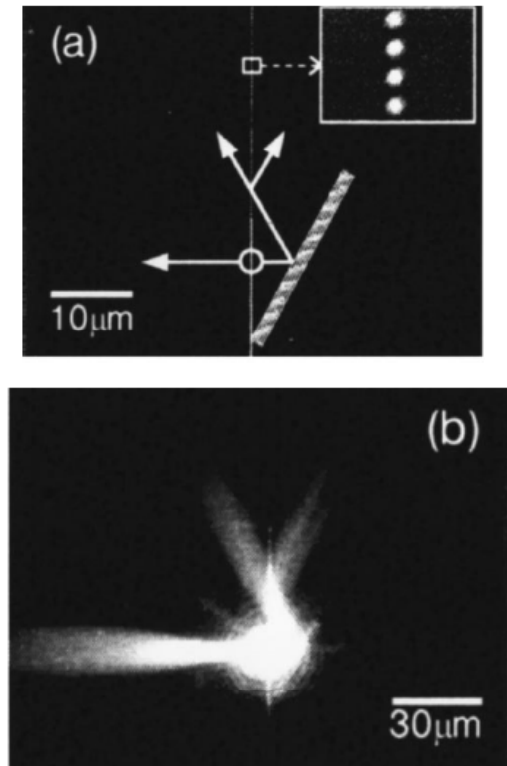
## Dielectric loaded SPP waveguide



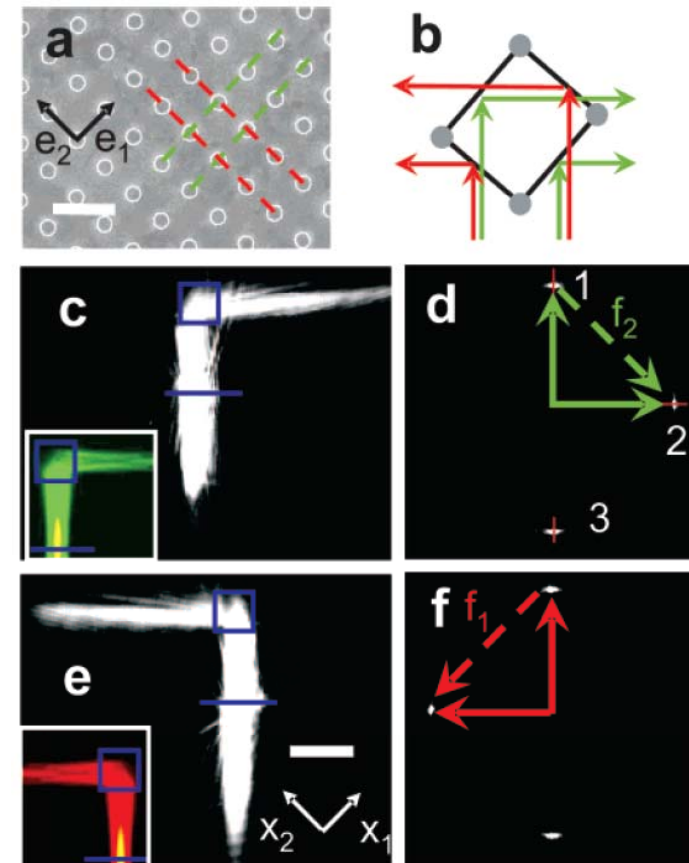
J. Grandier et al, Phys. Rev. B, 78, 245419 (2008)

# SPP control imaged by LRM

- Leakage radiation microscopy allows rapid prototyping of integrated plasmonic components



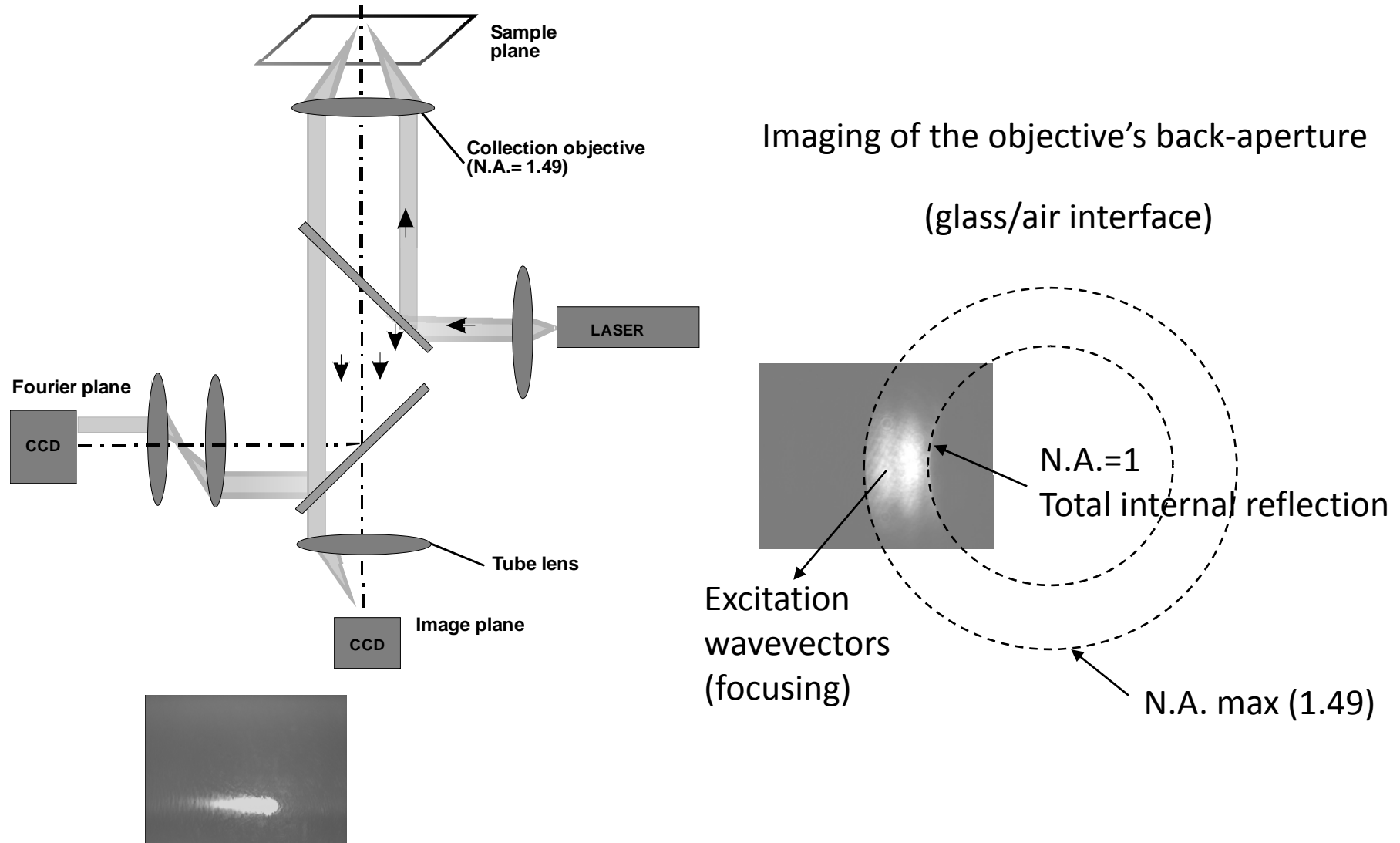
H. Ditlbacher et al, Appl. Phys. Lett. 81, 1762 (2002)



A. Drezet et al, Plasmonics B, 7, 1697 (2007)

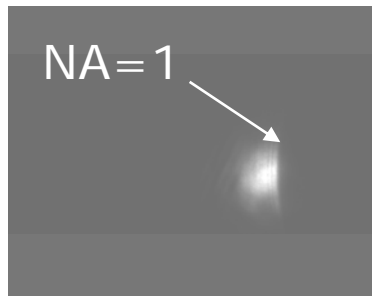
# Beyond propagation: Fourier imaging

- Imaging the Fourier plane gives access to the reciprocal space (wave-vectors)

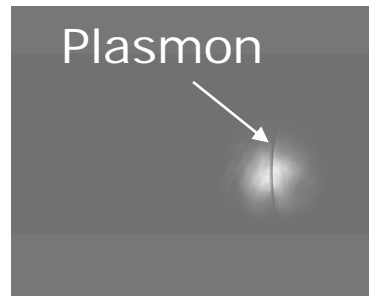


# Beyond propagation: Fourier imaging

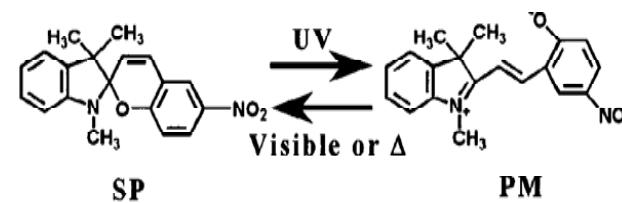
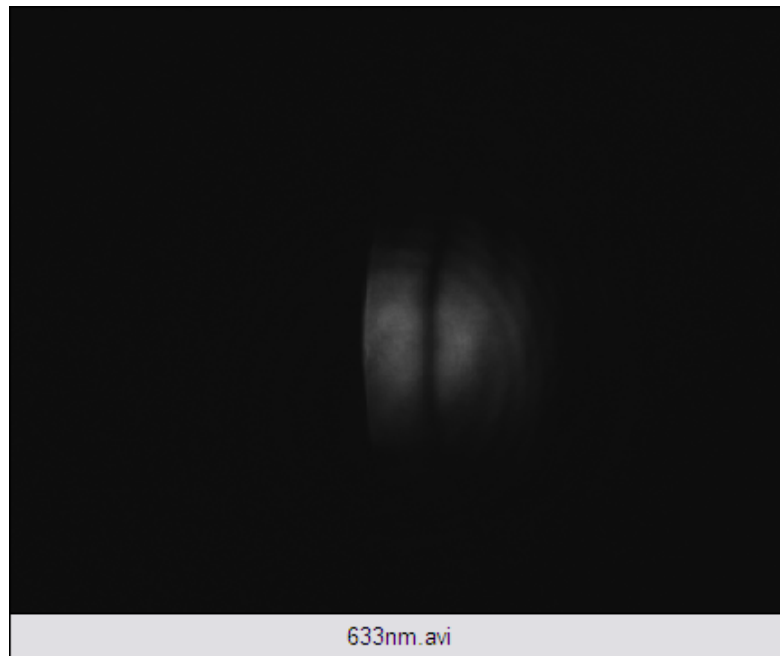
Glass/air interface



Glass/Ag/air interface

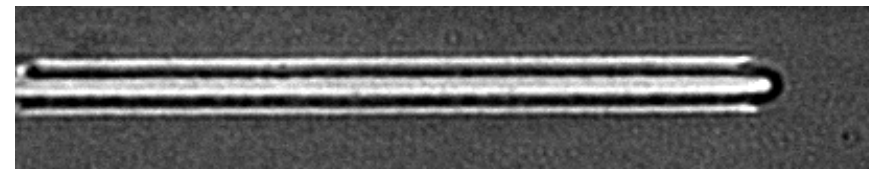
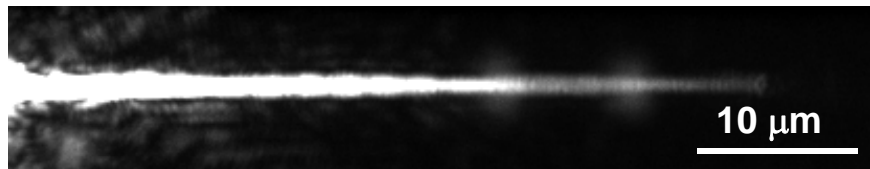


Glass/Ag/**Spiro**/air interface

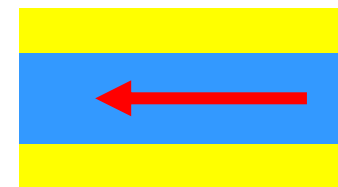
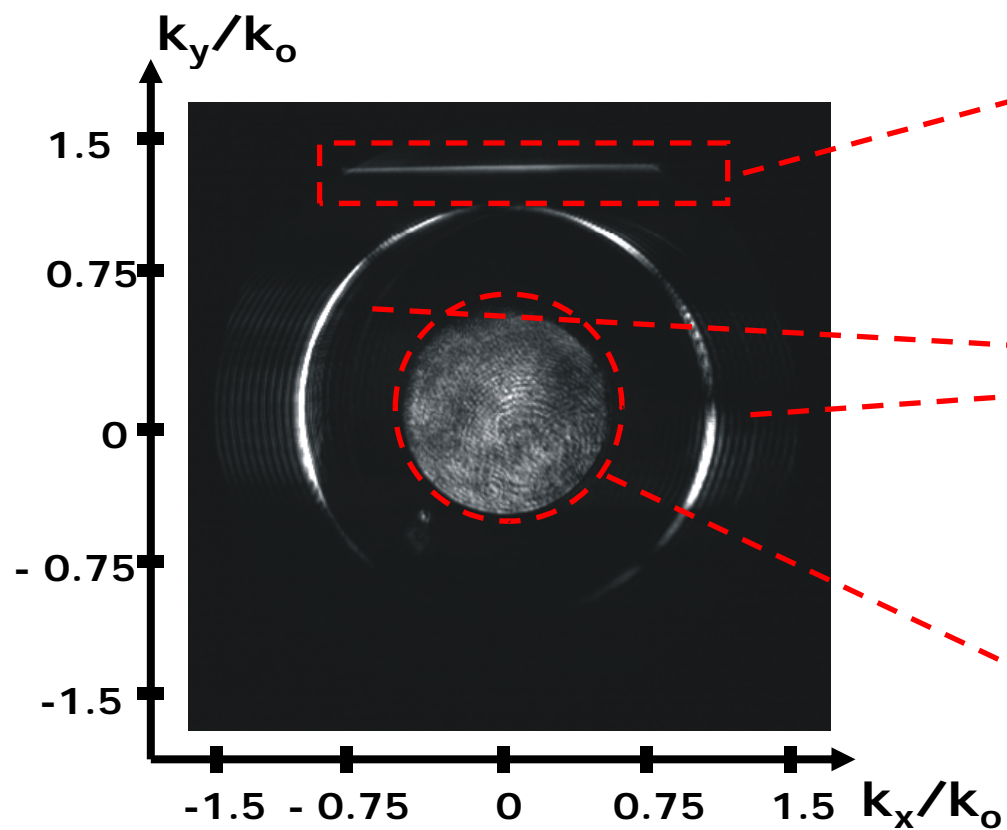


- Optical control of the resonant wavevectors through photochromism

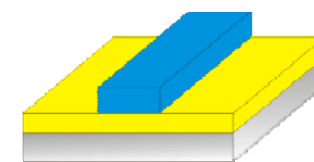
# Beyond propagation: Fourier imaging



Phase condition for  $k_y = \beta$



Au/air SPP

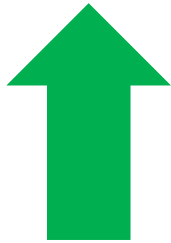


Excitation wavevectors

S. Massenot et al., *Appl. Phys. Lett.* 91, 243102 (2008)



# Pro et contras of Leakage Radiation Microscopy



- Rapid two-dimensional mapping of SPP
- Multi-wavelength, multi-wavevectors
- Only needs an oil immersion objective
- Access to direct and reciprocal space

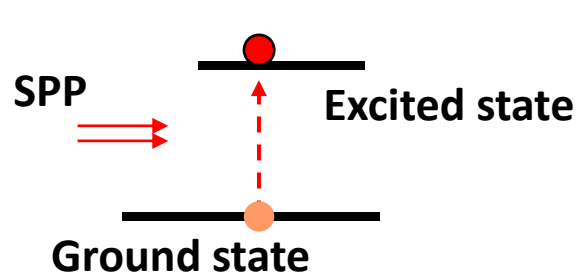


- Limited spatial resolution (diffraction)
- Restricted to leaky plasmons
- Limited to small effective indices (<1.5)

# FLUORESCENCE MICROSCOPY

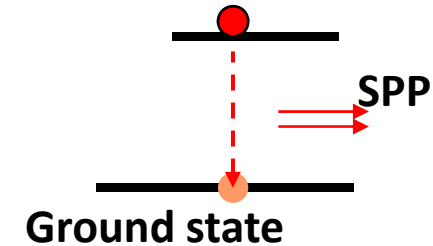
(glowing plasmons)

# Principle: energy transfer to/from fluorescent molecules

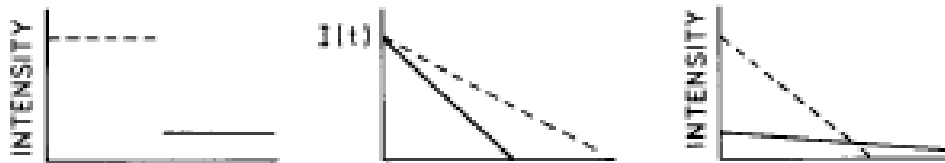


$$\sigma_{abs} = \frac{3\lambda^2}{4\pi} \eta(\gamma, \gamma_r, E_0)$$

$$Q = \frac{\gamma_r}{\gamma_r + \gamma_{nr}}$$



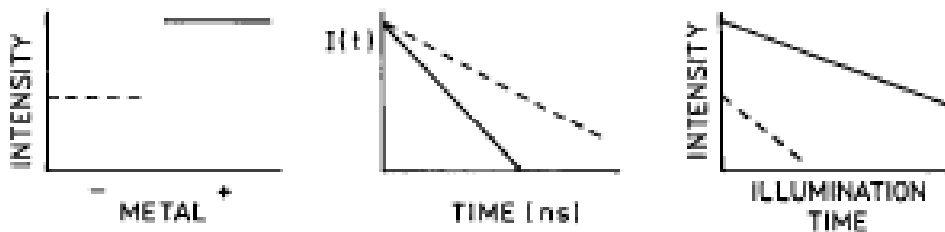
Quenching by metal



Increased excitation field



Increased radiative decay rate



## Quenching:

- Decreased intensity
- Decreased life time
- Increased photostability
- Lower # of photons

## Enhancement:

- Increased intensity from larger E
- Unaffected life time
- Equal # of photons

## Radiative life-time:

- Increased intensity from higher Q
- Decreased life-time
- Increased photostability
- Higher # photons

# Fluorescence intensity distribution: mapping the SPP

APPLIED PHYSICS LETTERS

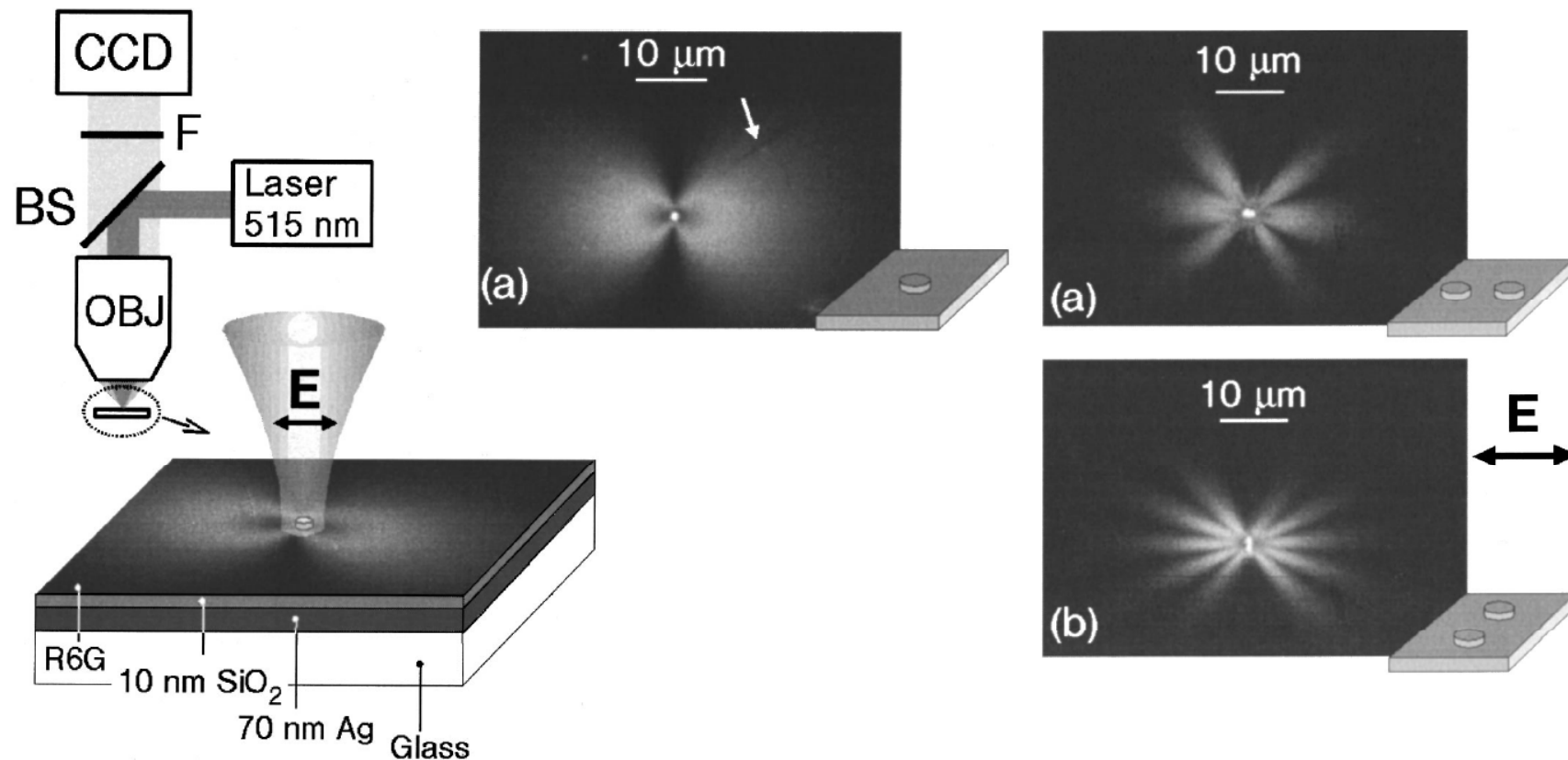
VOLUME 80, NUMBER 3

21 JANUARY 2002

## Fluorescence imaging of surface plasmon fields

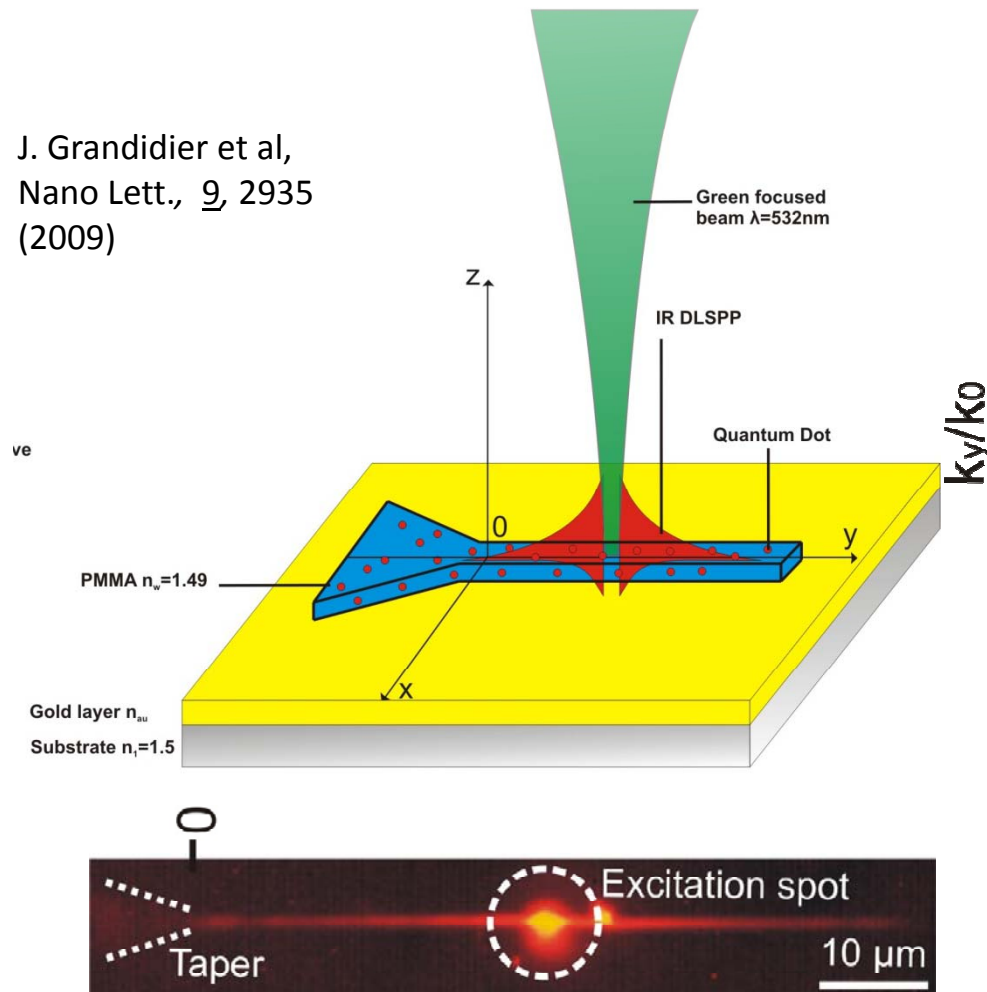
H. Ditlbacher, J. R. Krenn, N. Felidj, B. Lamprecht, G. Schider, M. Salerno, A. Leitner,  
and F. R. Aussenegg

*Institute for Experimental Physics and Erwin Schrödinger Institute for Nanoscale Research, Karl-Franzens-  
University Graz, A-8010 Graz, Austria*

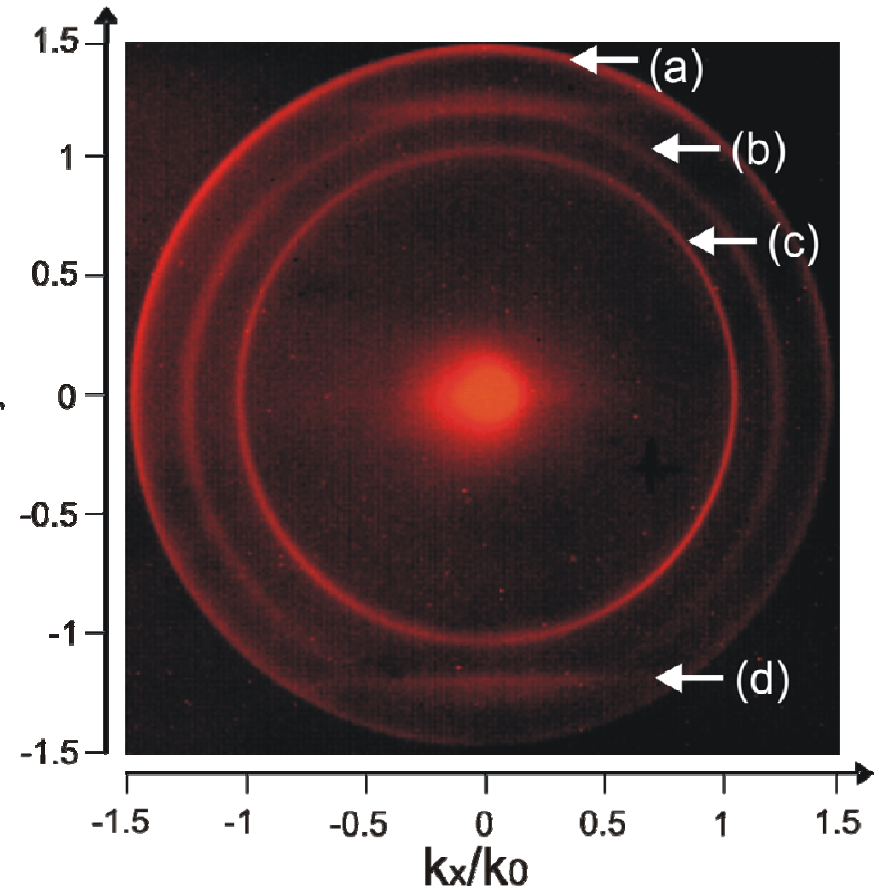


# Revealing mode properties through QDs fluorescence

J. Grandidier et al,  
Nano Lett., 9, 2935  
(2009)

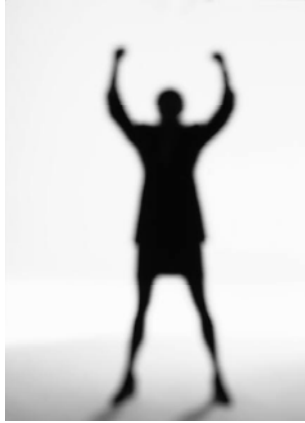


$\text{TM}_{00}$  guided mode excited through excitation of QDs  
Measured  $L_{\text{spp}}=9.21\mu\text{m}\pm 0.82\mu\text{m}$



- (a) Au/Air spp
- (b) Au/PMMA  $\text{TE}_0$  mode
- (c)  $\text{TM}_{00}$  guided mode
- (d) Au/PMMA  $\text{TM}_0$  mode

# Pro et contras of Fluorescence Microscopy



- Local probe
- Large choice of chromophores or QDs (SPP matching wavelength)
- Straightforward excitation and detection
- Access to direct and reciprocal space

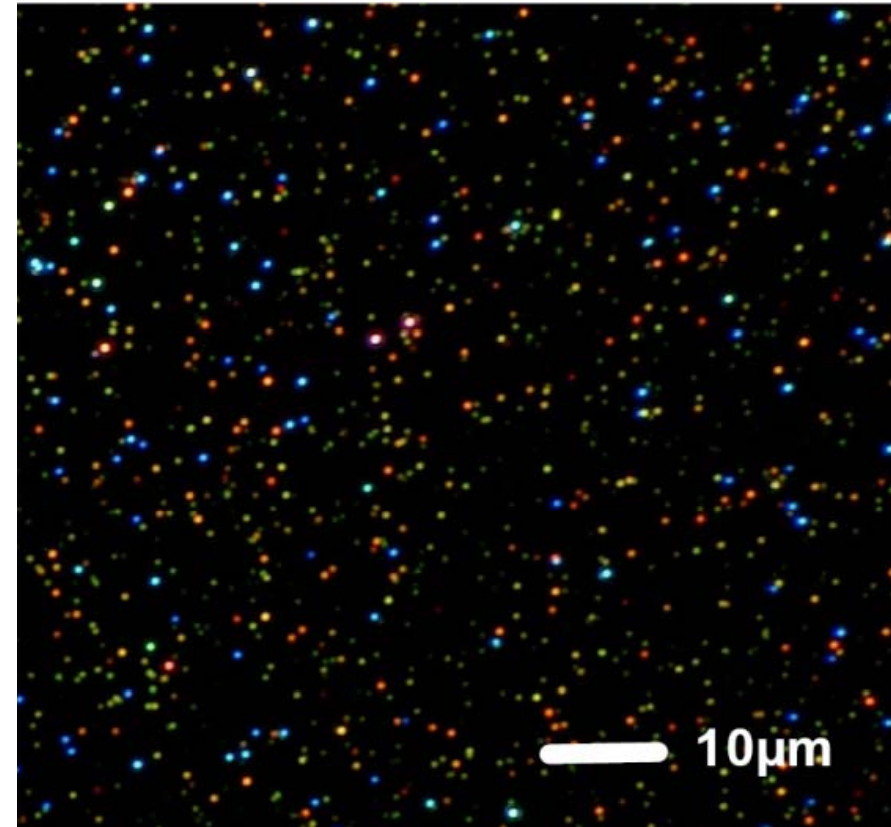
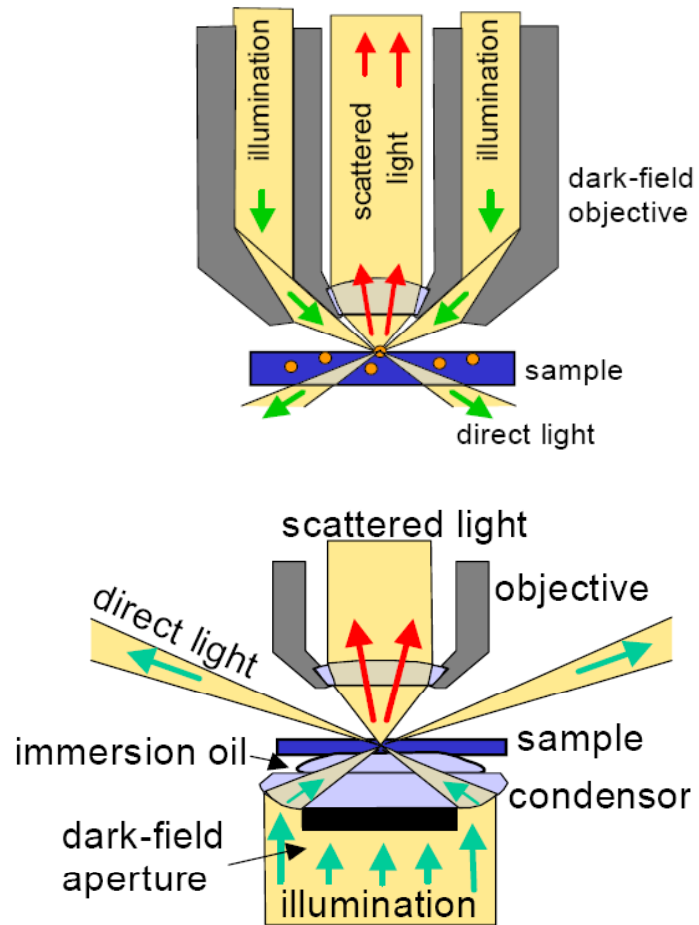


- Limited spatial resolution (diffraction)
  - Molecular photobleaching
- Critical to distance from film
  - Qualitative information

# **DARK-FIELD MICROSCOPY**

(Colored plasmons)

# Principle: rejecting out direct illumination

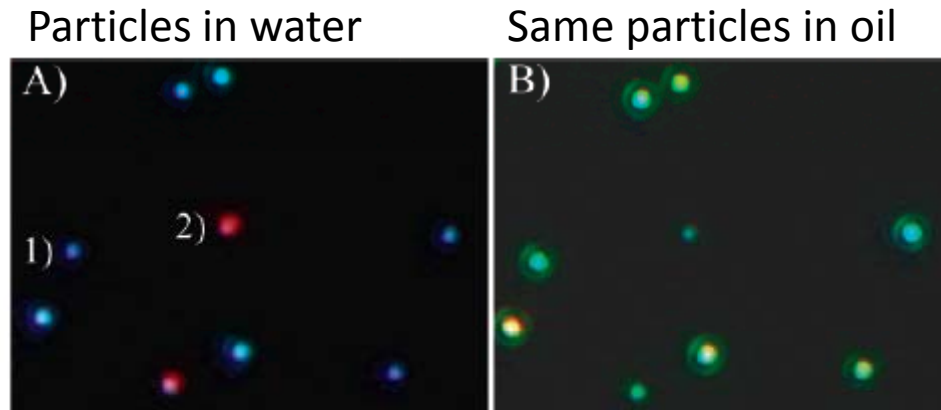


C. Sönnichsen, *Plasmons in metal nanostructures*, PhD thesis, Munich (2001)

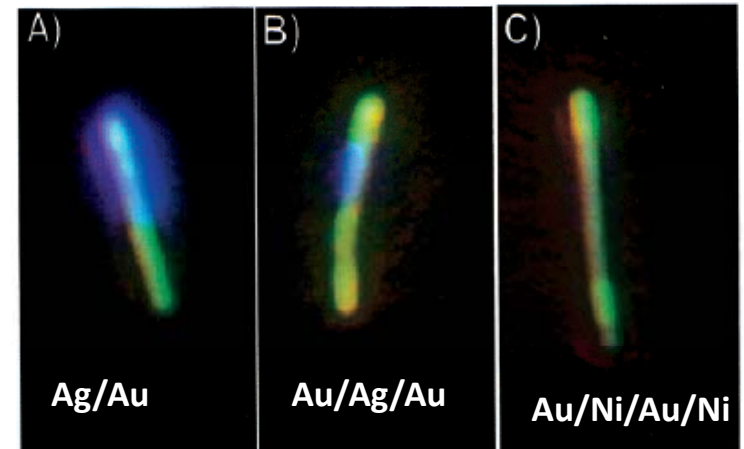
C. Sönnichsen, *Plasmons in metal nanostructures*, PhD thesis, Munich (2001)



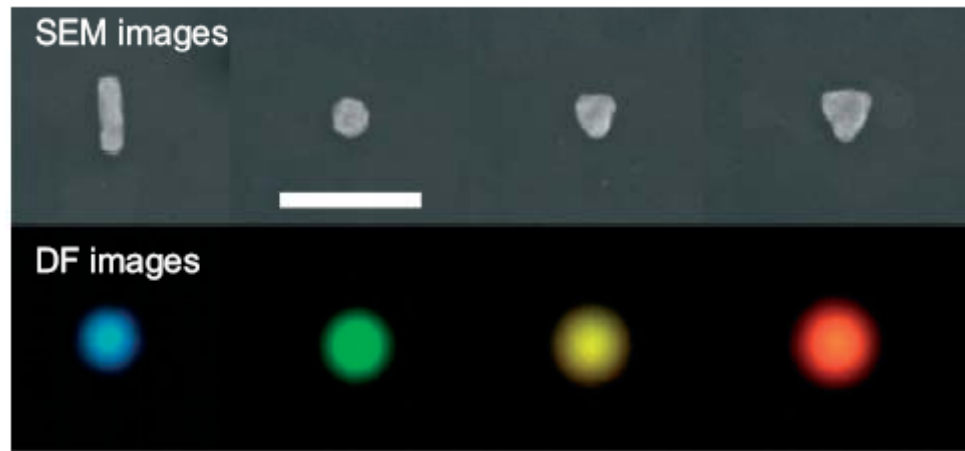
# Dark-field imaging (and spectroscopy)



J. Mock, *Nano Lett.* **3**, 485 (2003)



J. Mock, *Nano Lett.* **2**, 465 (2002)



A. Murray & W. Barnes, *Adv. Mat.* **19**, 3771 (2007)

# Pro et contras of dark-field Microscopy



- Complete spectroscopy
- Array and single particle (multiscale) investigations
- Liquid environment

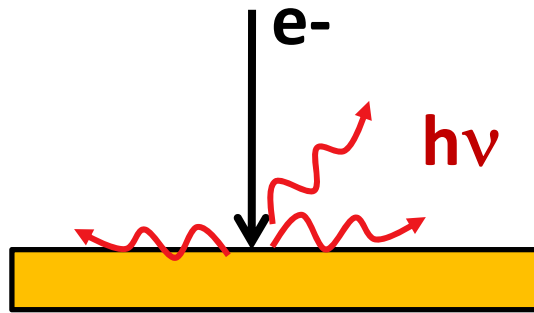


- Limited spatial resolution (diffraction)
- Adapted for localized particle plasmons
- Large particle (>50nm)
- Non-unequivocal

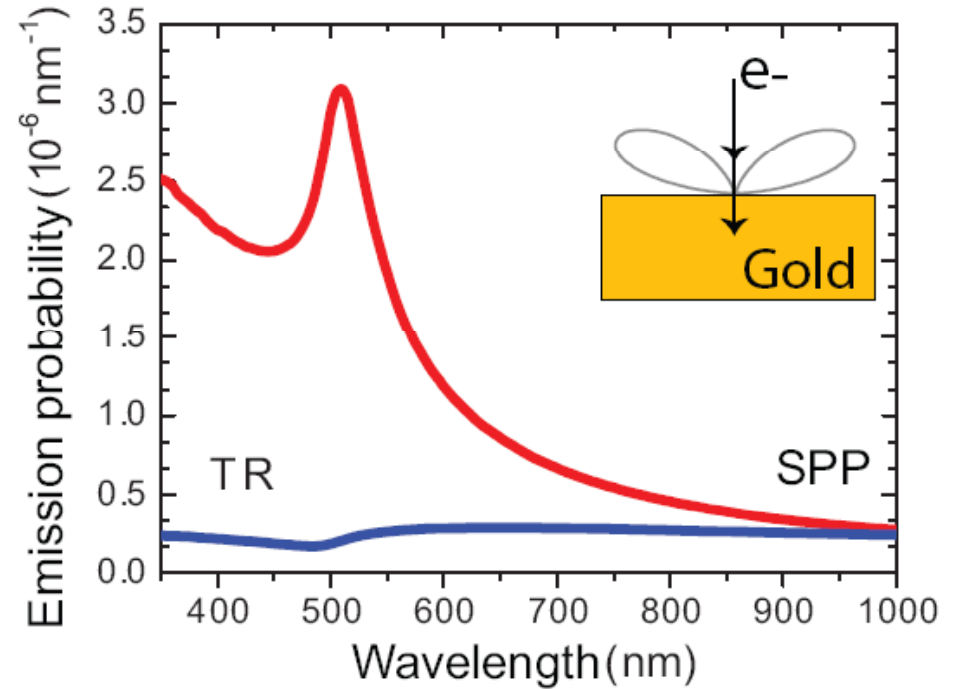
# CATHODOLUMINESCENCE MICROSCOPY

(a valse between an  $e^-$ , a plasmon, and a photon)

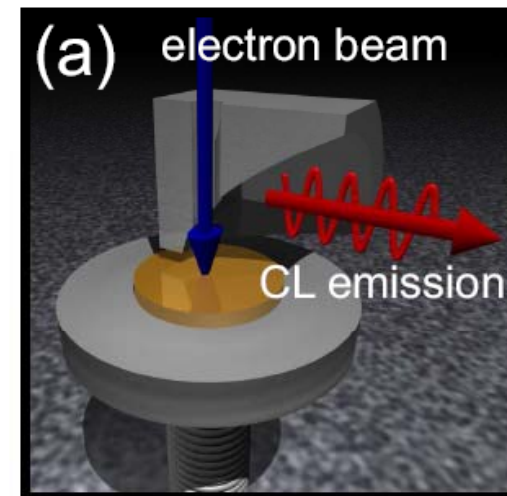
# Principle: transition radiation and surface plasmon



Contribution from transition radiation ( $e^-$  incident on interface) and surface plasmons. Both are coherent with the external field of the electron: interference signal



M. Kuttge, *Cathodoluminescence Plasmon microscopy*, PhD thesis, Utrecht (2009)



# Photon maps and spectroscopy of localized SPP modes

PHYSICAL REVIEW B, VOLUME 64, 205419

## Photon emission from silver particles induced by a high-energy electron beam

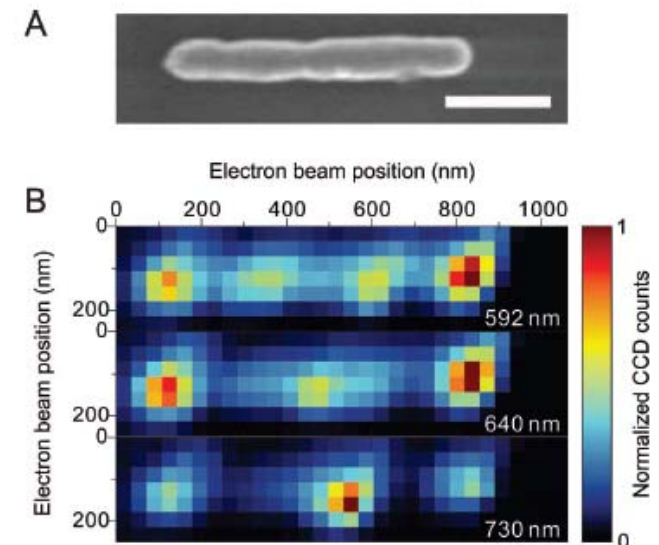
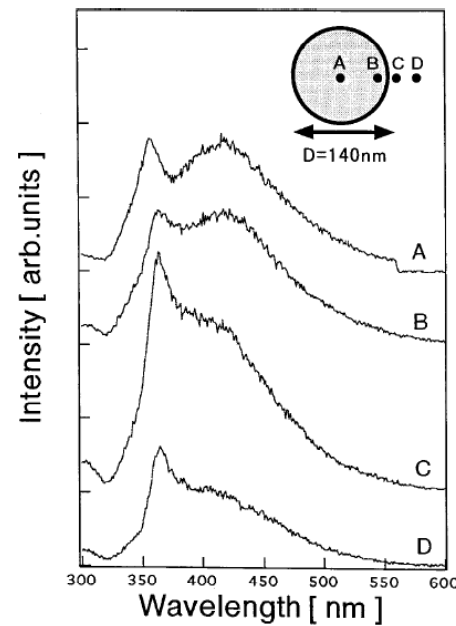
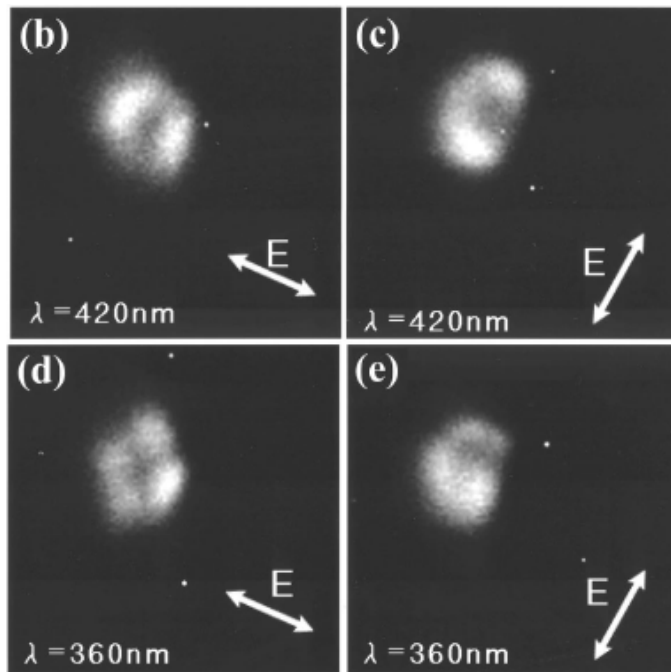
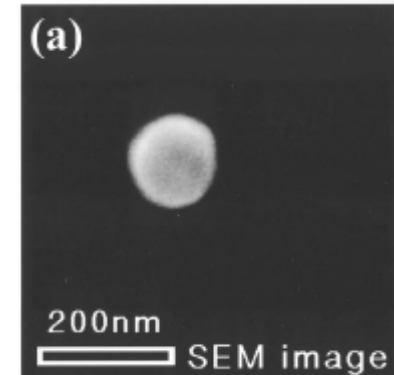
N. Yamamoto and K. Araya

*Department of Physics, Tokyo Institute of Technology, Meguro-ku, Tokyo 152-8551, Japan*

F. J. García de Abajo

*Centro Mixto CSIC-UPV/EHU and Donostia International Physics Center (DIPC), San Sebastián, Spain*

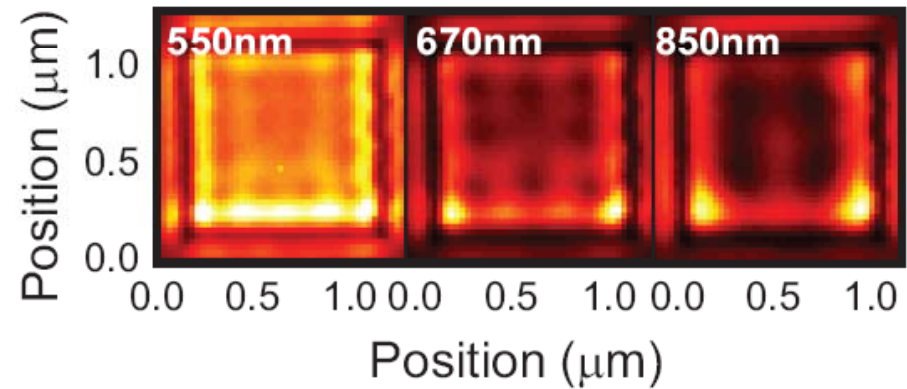
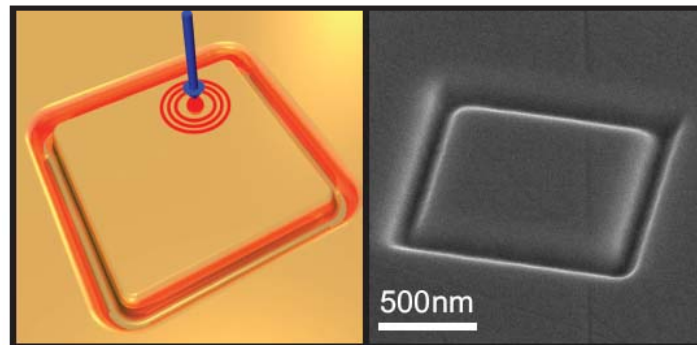
(Received 11 July 2000; published 6 November 2001)



E. Vesseur et al., *Nano. Lett.* **7**, 2843 (2007)

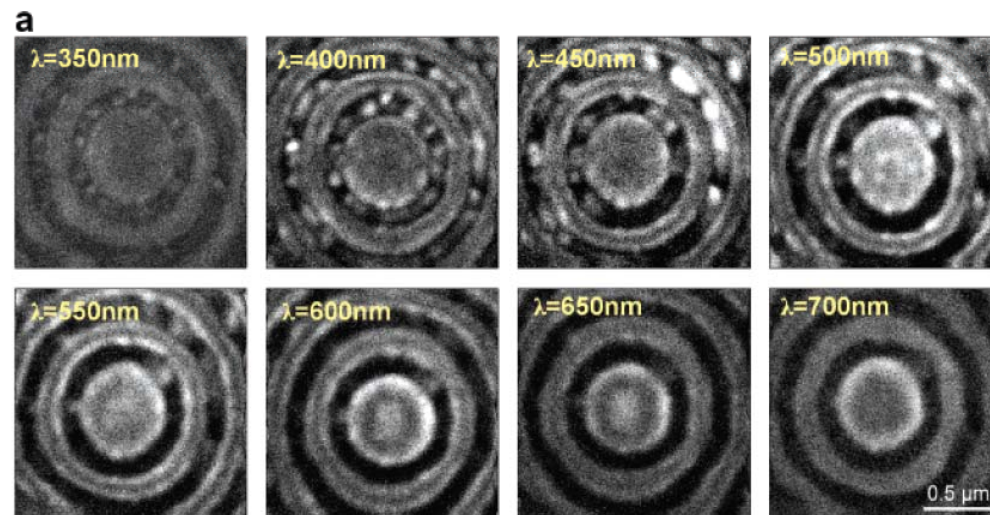
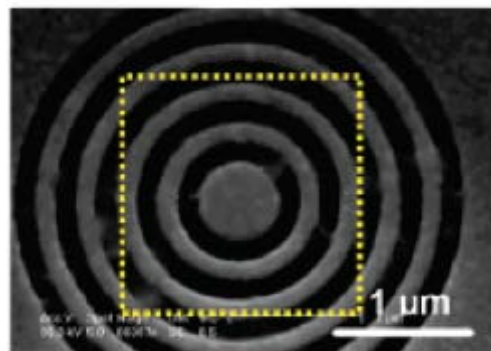
# Photon maps of propagative SPP modes

## Boxed plasmons



M. Kuttge, *Cathodoluminescence Plasmon microscopy*, PhD thesis, Utrecht (2009)

## Resonators



C. Hofmann et al., *Nano. Lett.* 7, 3612 (2007)

# Pro et contras of Cathodoluminescence Microscopy



SPP point source (nm size)  
Spatial and spectral investigations  
Imaging of localized and propagative  
SPP modes



Vacuum environment  
Low efficiency ( $10^{-6}/\text{nm}$ )  
Interference contrast

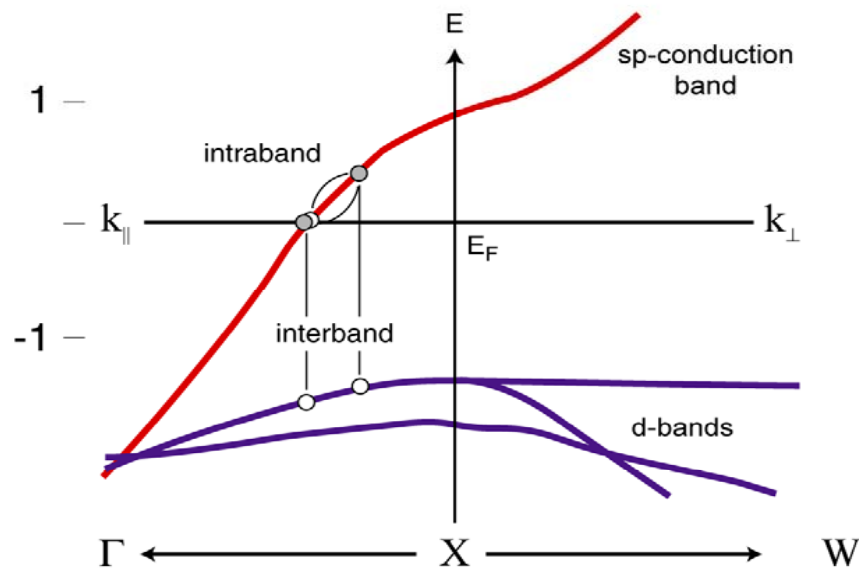
# **(NON LINEAR) CONFOCAL MICROSCOPY**

(Blurred plasmons)

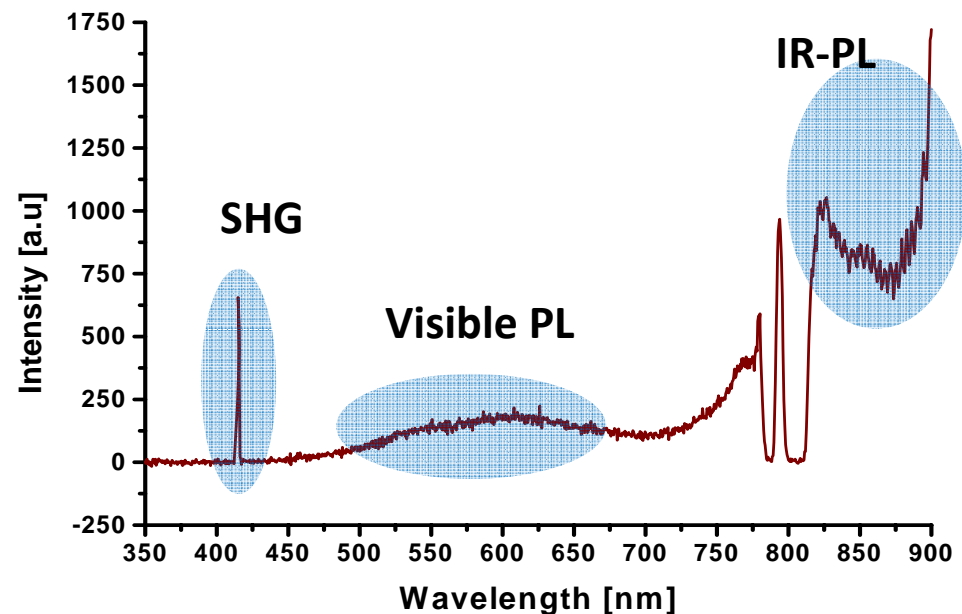


# Gold: a luminescent material!

- Visible photoluminescence originates from interband electronic transitions from  $d$ -band electrons to the conduction band
- IR PL originates from intraband transitions
- Second-harmonic is generated by a non-linear surface susceptibility



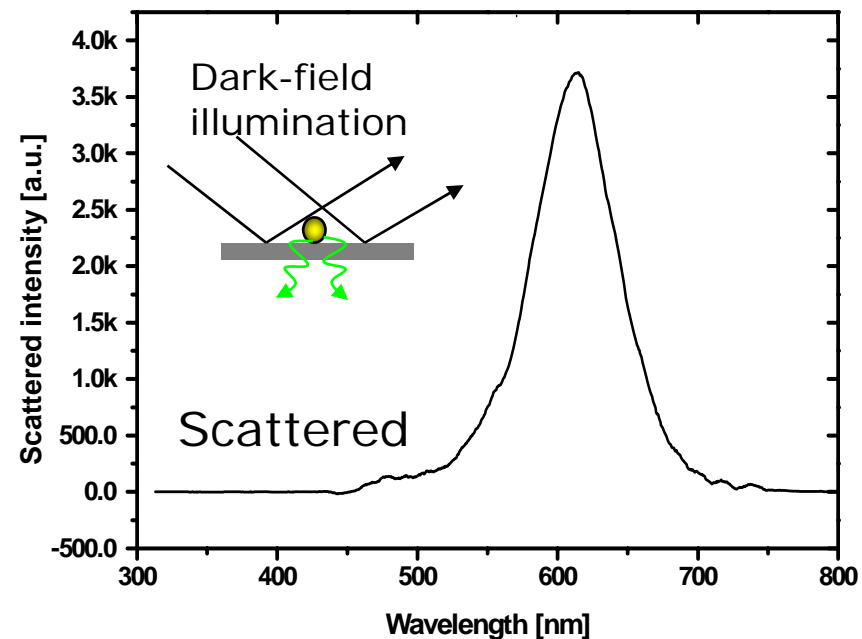
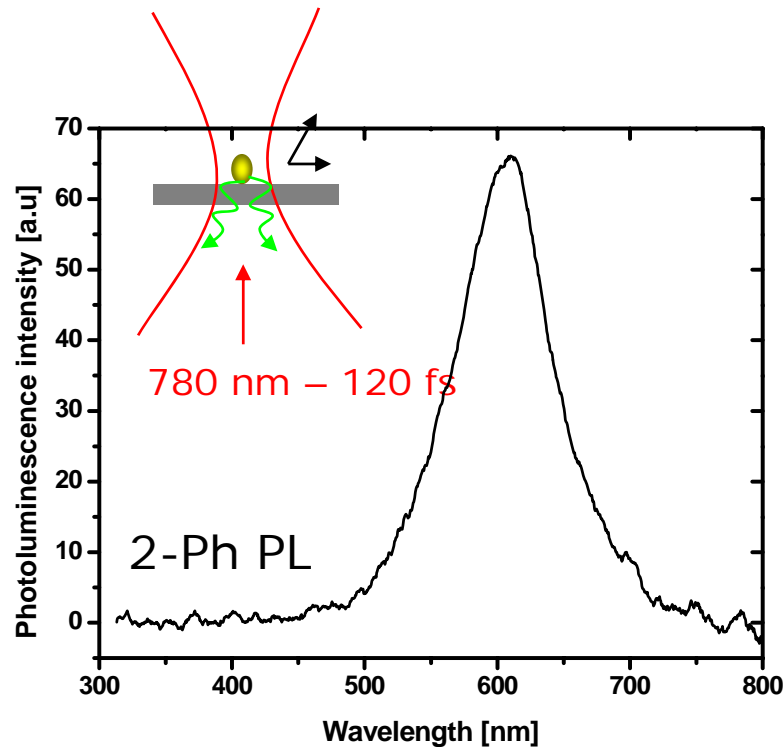
O. Jepsent et al., *Phys. Rev. B*, 23, 2684 (1981).  
M. Guerrisi et al, *Phys. Rev. B*, 12, 557 (1975).



M. Beversluis, A. Bouhelier, and L. Novotny, *Phys. Rev. B*, 68, 115433 (2003)

# Gold PL and plasmons

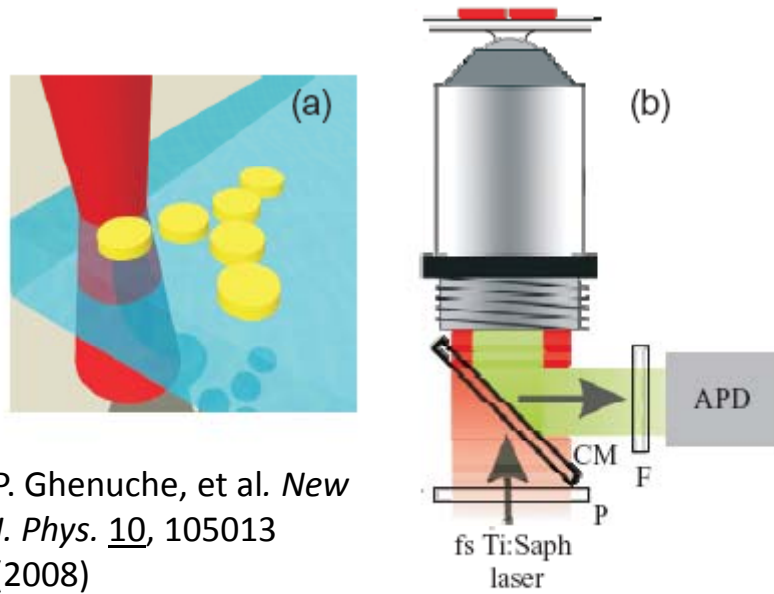
- The PL emission is modulated by the surface plasmon resonance



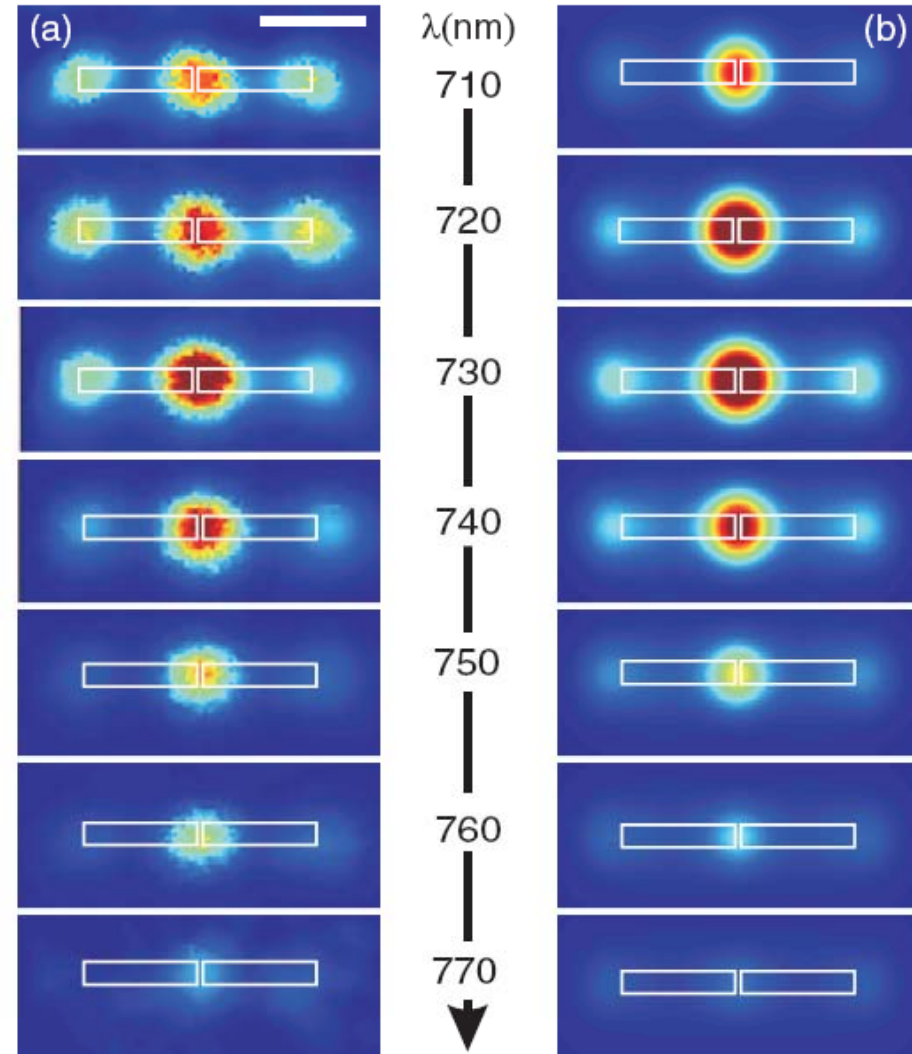
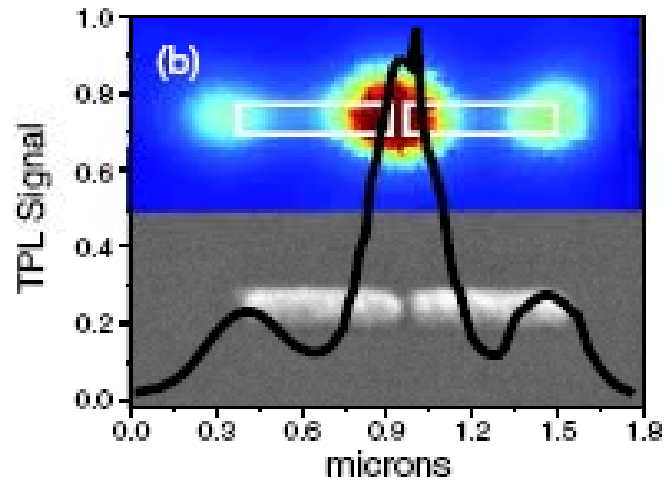
A. Bouhelier, et al. *Phys. Rev. Lett.* 95, 267405 (2005)

The surface plasmon plays a double role here: create a local field enhancement to increase the number of PL photons and also provide a fast radiative  $e^-$  relaxation process through SP emission

# Imaging PL spatial distribution

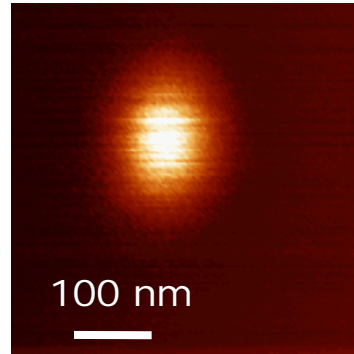
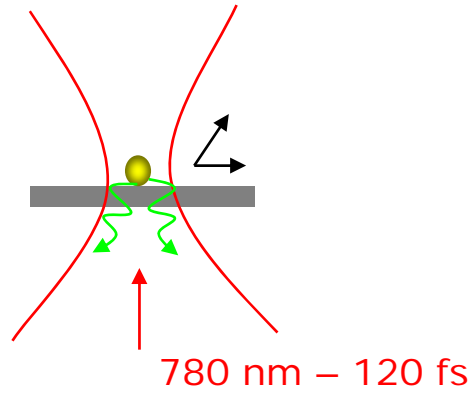


P. Ghenuche, et al. *New J. Phys.* 10, 105013 (2008)

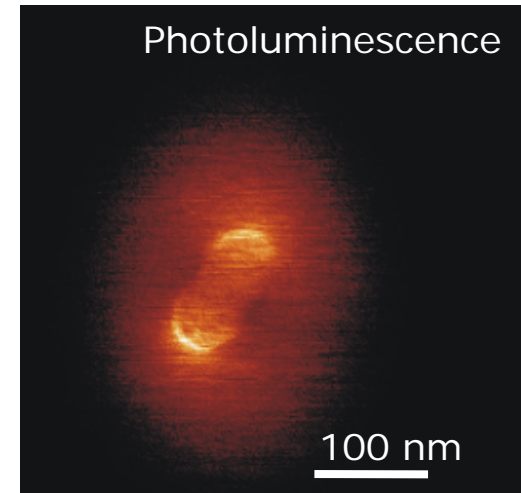
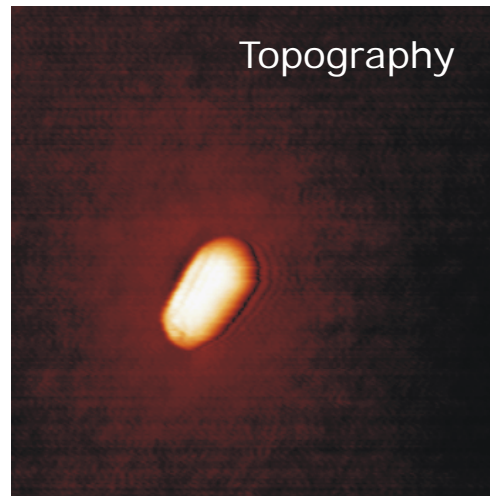
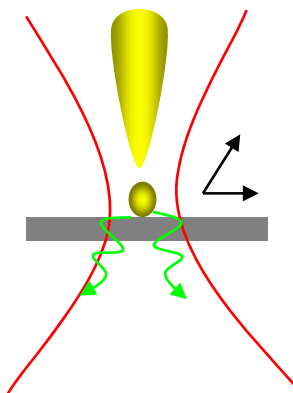


P. Ghenuche, et al. *Phys. Rev. Lett.* 101, 116805 (2008)

# Parenthesis: near-field non-linear imaging

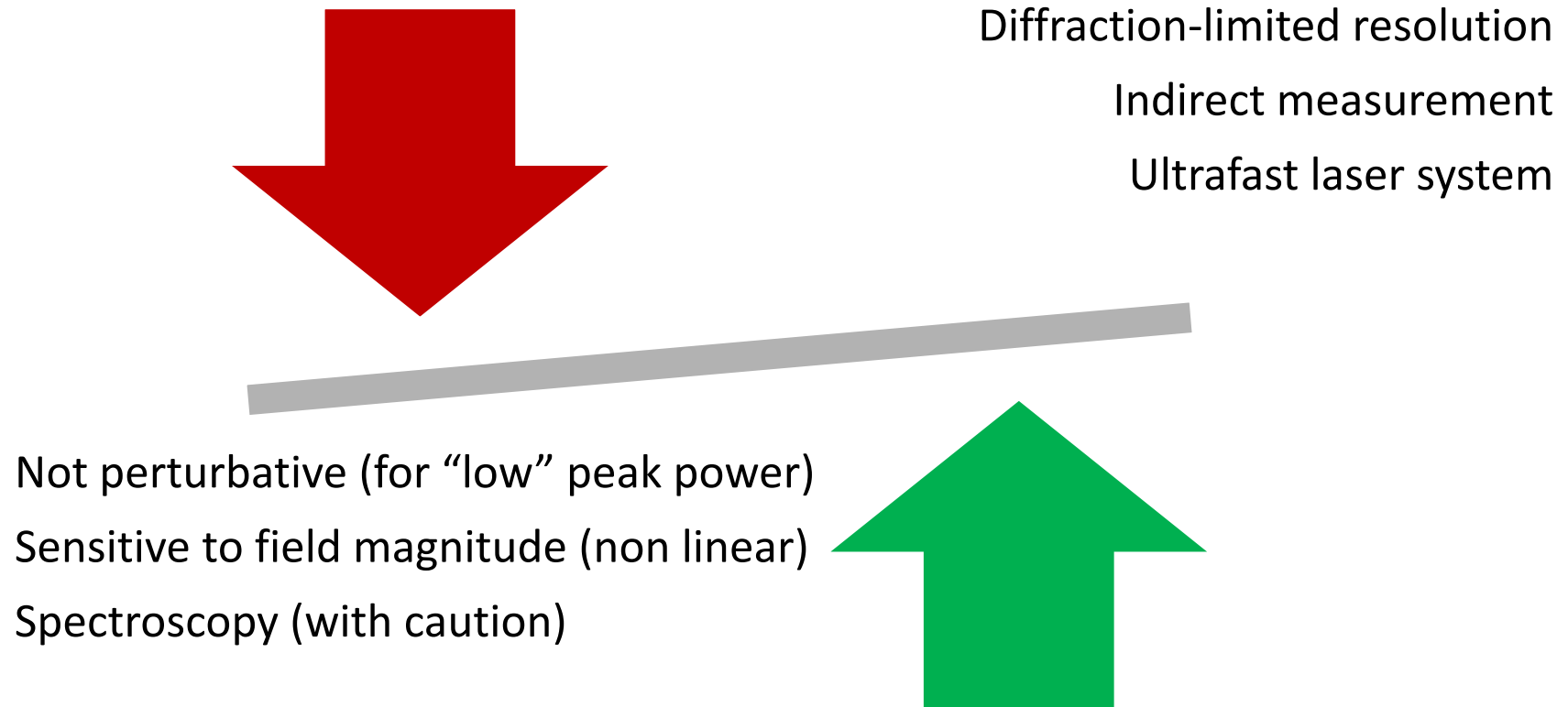


Confocal far field



A. Bouhelier, M. Beversluis, and L. Novotny, *App. Phys. Lett.* 83, 5041 (2003)  
K. Imura, T. Nagahara, and H. Okamoto, *J. Am. Chem. Soc.* **126**, 12730 (2004)

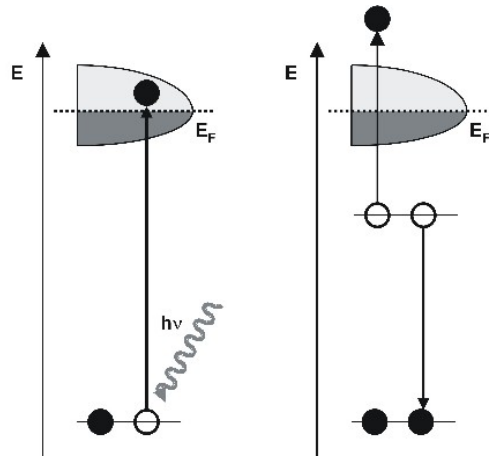
# Pro et contras of confocal Microscopy



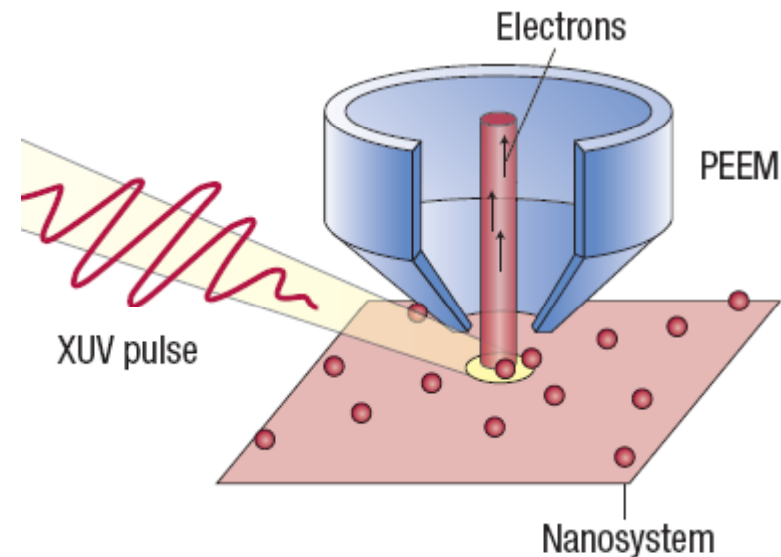
# PHOTOELECTRON EMISSION MICROSCOPY

(a valse between photon(s), plasmons, and e<sup>-</sup>)

# Photo-electron emission



- 1) Upon photo-excitation (Xray, UV), electrons are excited above Fermi level, creating holes on core levels
- 2) Auger scattering leads to electron emission above the work function
- 3) Broad distribution of energy between the excitation energy and the work function



Role of the plasmon: electron yield proportional to  $E^4$

M. Nisoli, Nat. Photo. 1, 499 (2007)

# Ultrafast dynamics in space and time

## Femtosecond Imaging of Surface Plasmon Dynamics in a Nanostructured Silver Film

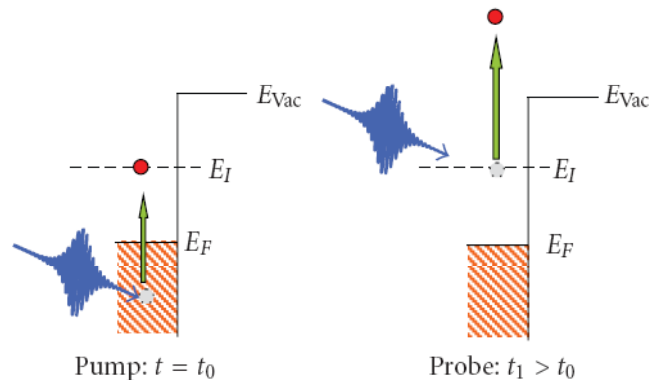
NANO  
LETTERS

2005  
Vol. 5, No. 6  
1123–1127

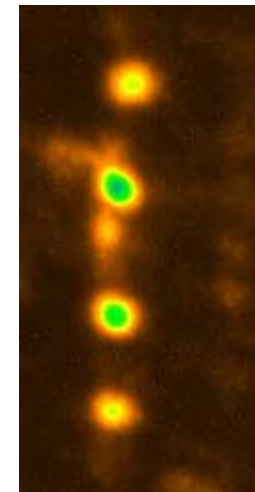
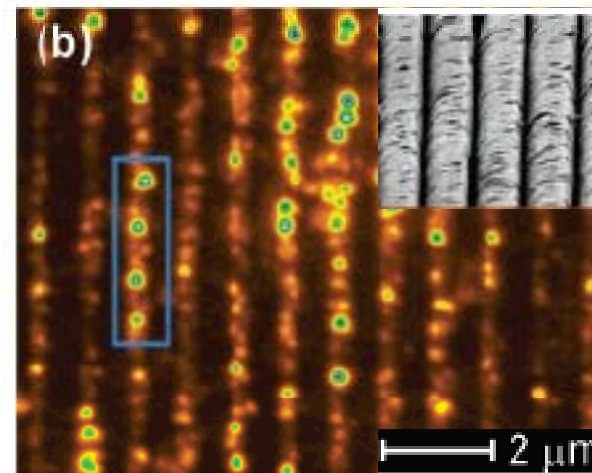
Atsushi Kubo,<sup>†,§</sup> Ken Onda,<sup>†,§</sup> Hrvoje Petek,<sup>\*,†,§</sup> Zhijun Sun,<sup>†,§</sup> Yun S. Jung,<sup>†,§</sup> and Hong Koo Kim<sup>†,§</sup>

*Department of Physics and Astronomy, Department of Electrical Engineering, and Institute of NanoScience and Engineering, University of Pittsburgh, Pittsburgh, Pennsylvania 15260*

Interferometric time-resolved two-photon photoemission (ITR-2PP) and photoelectron emission microscopy (PEEM).

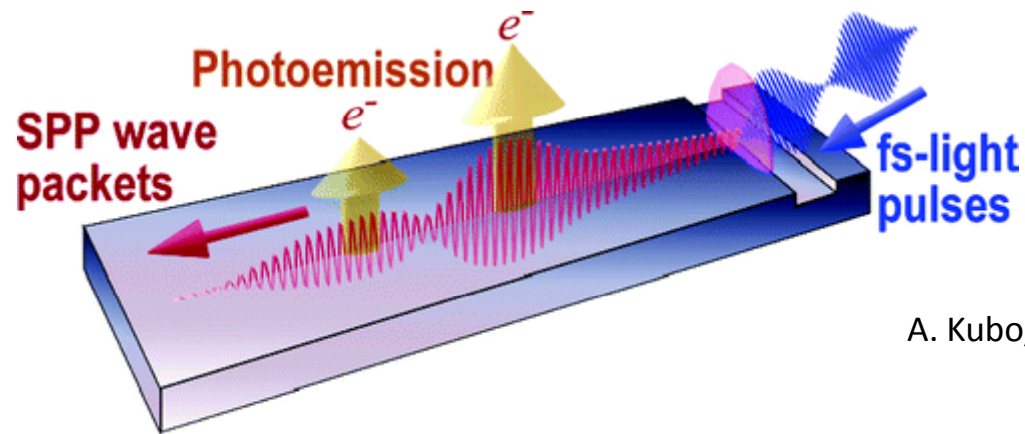


D. Bayer, et al. *J. Nano. Mat.* 249514 (2008)

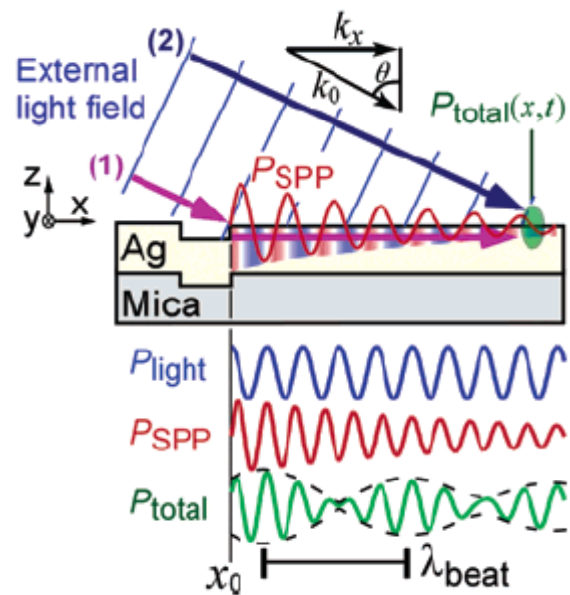




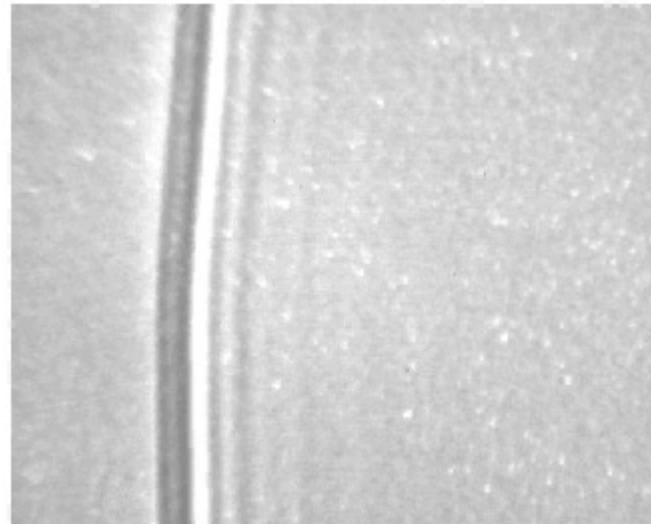
# Ultrafast dynamics in space and time



A. Kubo, et al. *Nano Lett.* 7, 470 (2007)



$$\tau_d = 0.00 \times 2\pi$$



# Resolving mode structures

## Short Range Plasmon Resonators Probed by Photoemission Electron Microscopy

Ludovic Douillard,\* Fabrice Charra, and Zbigniew Korczak†

*Commissariat à l'Energie Atomique Saclay Direction des Sciences de la Matière - Institut Rayonnement Matière de Saclay - Service de Physique et Chimie des Surfaces et Interfaces, F-91191 Gif sur Yvette, France*

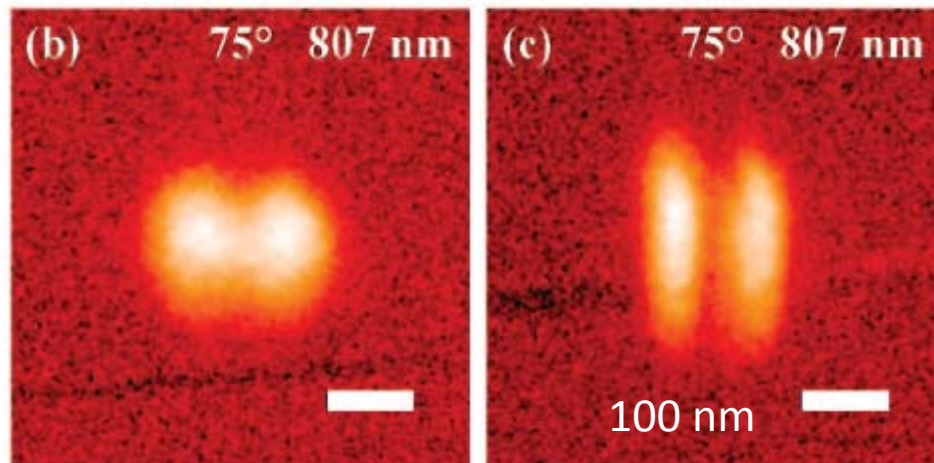
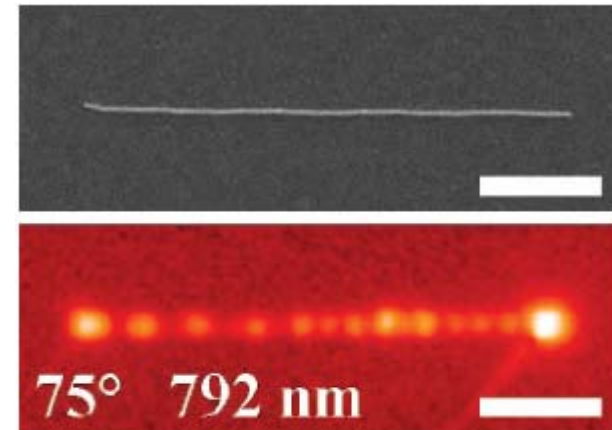
Renaud Bachelot, Sergei Kostcheev, Gilles Lerondel, Pierre-Michel Adam, and Pascal Royer

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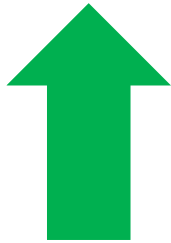
NANO  
LETTERS

2008  
Vol. 8, No. 3  
935-940



Dipolar mode in short nanorods

# Pro et contras of PEEM



- Nanometer scale resolution (~25 nm)
- Sub-fs time resolution
- Energy resolution (TOF-PEEM)
- Coherent control of SPP field

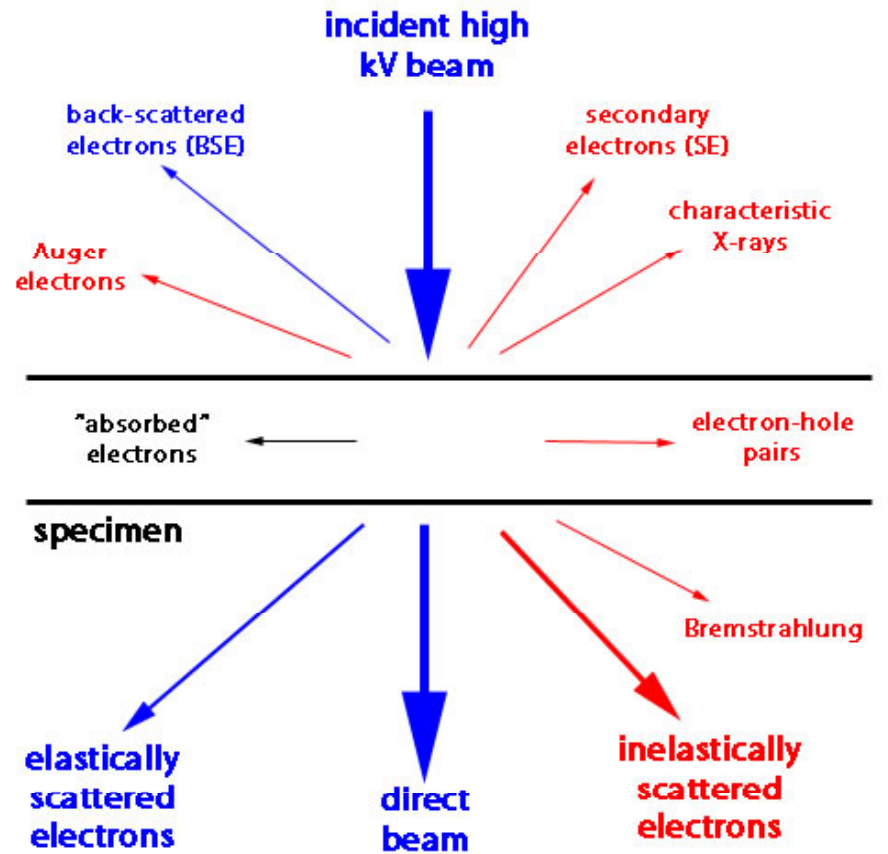


- UHV environment
- Chromatic aberrations
- Limited to short-wavelength resonance (multi-photon process, electron yield to  $E^4$ )

# **ELECTRON-ENERGY LOSS IMAGING**

(a nanoscale detective)

# Principle of operation



modified from Williams & Carter (1996) Fig. 1.3

- Raster scanning the electron beam across the sample
- EELS spectrum @ each pixel
- fix energy, plot the amplitude of the spectra for each pixel to reconstruct 2D-image

The sampling volume (beam size) is smaller than the spatial variation of the field

# EELS Mapping

Mapping surface plasmons on a single metallic nanoparticle

nature [physics](http://www.nature.com/naturephysics) | VOL 3 | MAY 2007 | [www.nature.com/naturephysics](http://www.nature.com/naturephysics)

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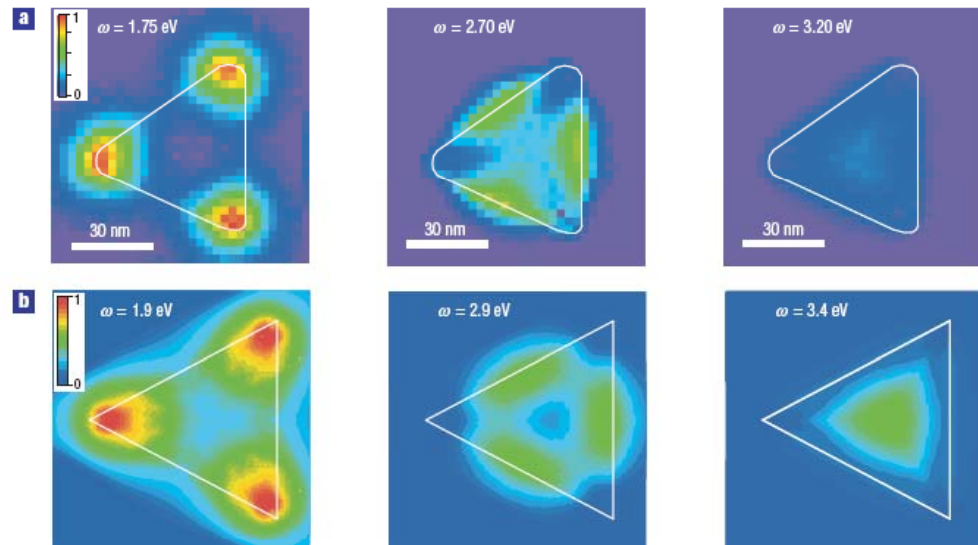
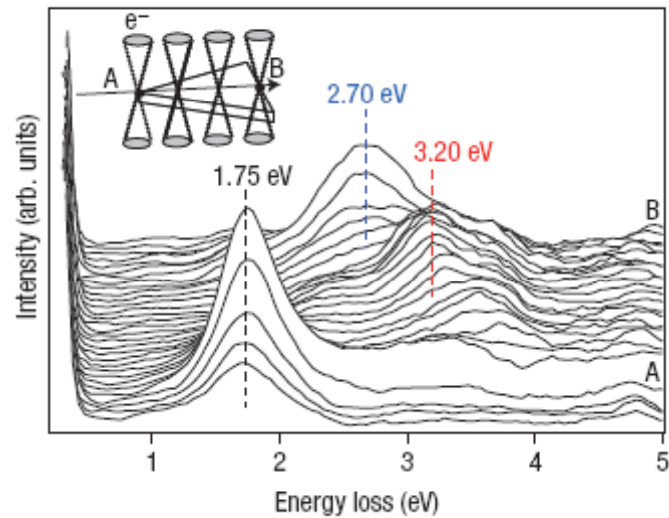
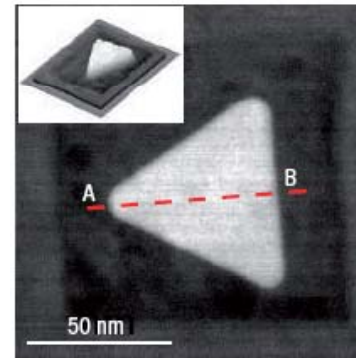
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# EELS Mapping

IOP PUBLISHING

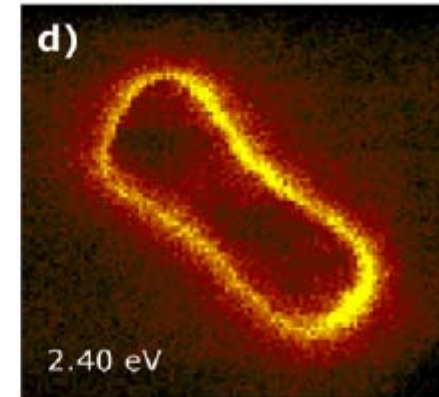
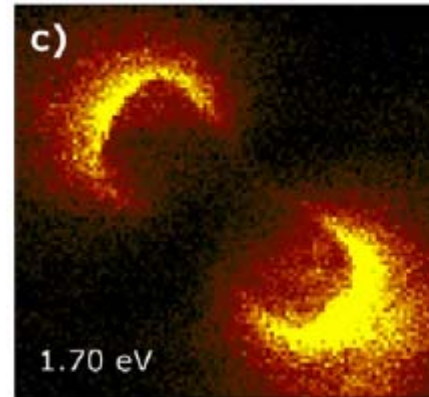
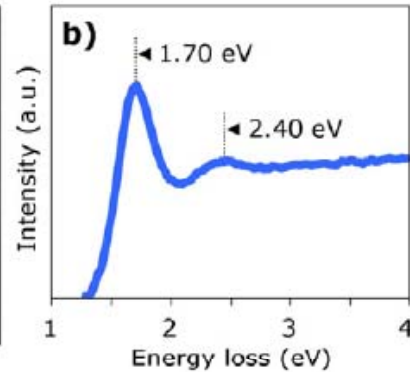
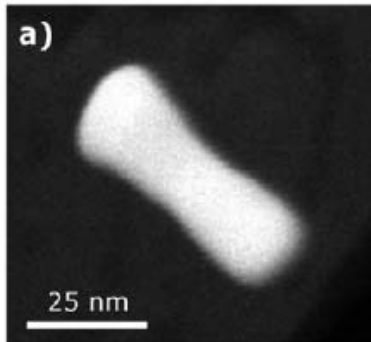
NANOTECHNOLOGY

Nanotechnology 18 (2007) 165505 (5pp)

doi:10.1088/0957-4484/18/16/165505

## Mapping surface plasmons at the nanometre scale with an electron beam

Michel Bosman<sup>1</sup>, Vicki J Keast<sup>2</sup>, Masashi Watanabe<sup>3</sup>,  
Abbas I Maaroo<sup>4</sup> and Michael B Cortie<sup>4</sup>



# Pro et contras of EELS mapping



- Unprecedented resolution ( $< 5\text{nm}$ )
- Relationship between shape, localization & energy
- Investigation to high energies (bulk plasmon)
- Direct measurement of energy exchange (access to the dielectric function of the material)



- UHV environment
- Deconvolution for plasmon bands close to the zero-loss peak

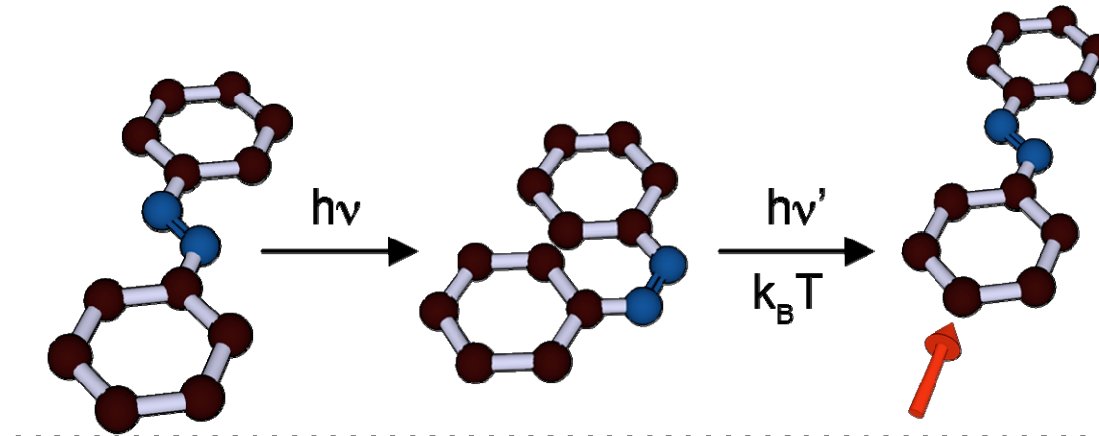


# PHOTOCHEMICAL IMAGING

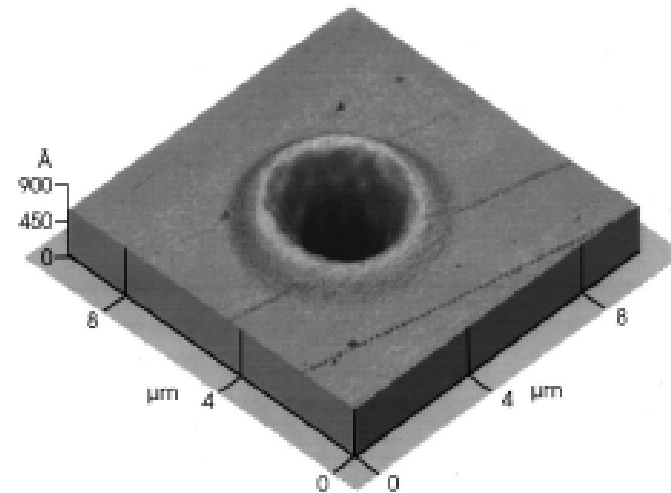
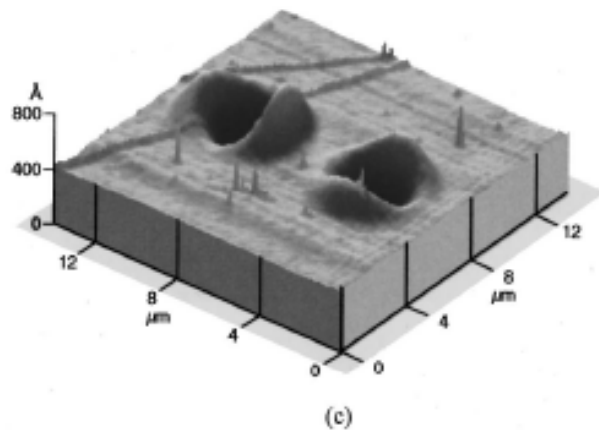
(through topography)

# Principle of operation: isomerization and diffusion

- Repetitive *Trans-Cys-Trans* cycles introduce translational molecular diffusion

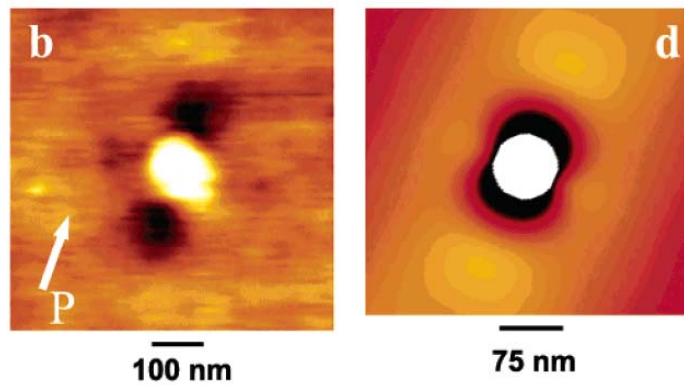


Courtesy of Prof. Plain, UTT Troyes

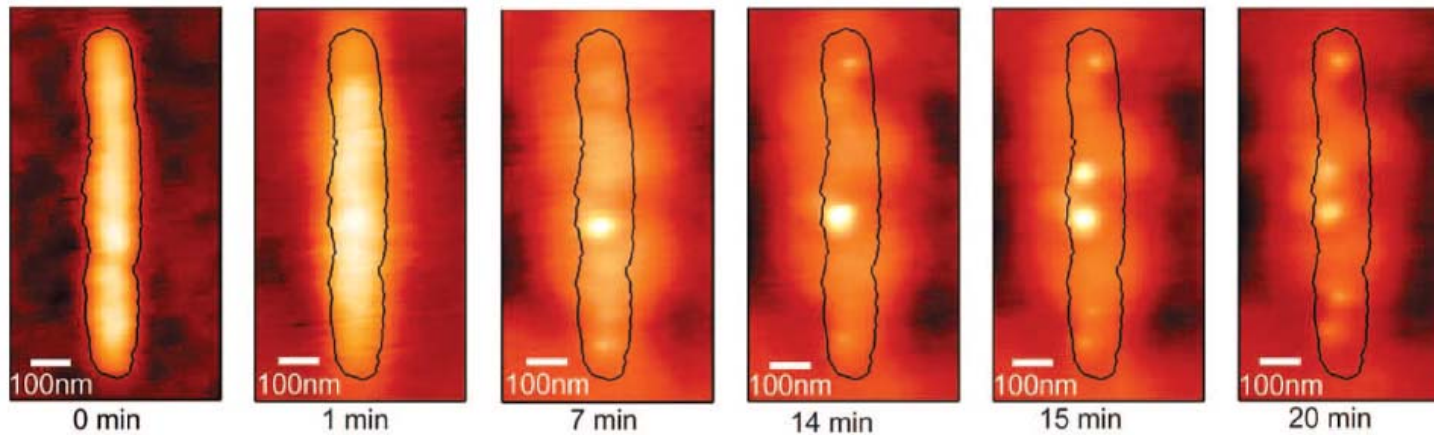
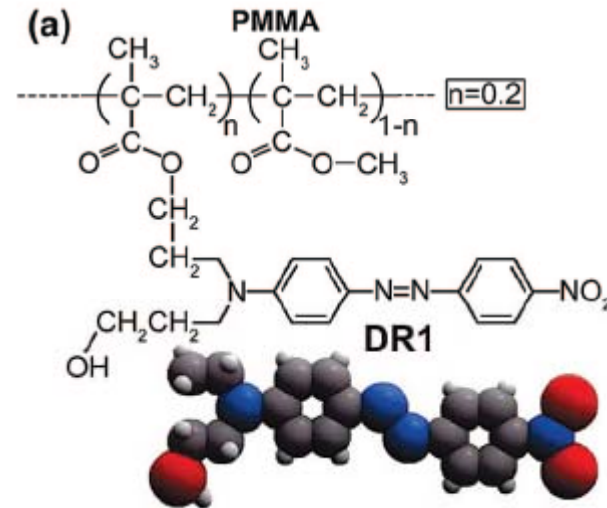


S. Bian, et al. *J. Appl. Phys.* 86, 4498 (1999)

# Photochemical imaging of localized SPs



C. Hubert, et al. *Nano Lett.* 5, 615 (2005)



M. Juan, et al. *J. Phys. Chem. A* 113, 4647 (2009)

# Pro et contras of photochemical imaging



- Vectorial approach (field sensitivity)
- Control over molecular mass transport
- In principle, molecular resolution



- Limited wavelength range
- Dependence to temperature
- Second-order measurement
- Relatively complex modeling required for interpretation

# Conclusion

## Reflectivity

- Interface sensitivity
- Dispersion
- Indirect access to  $L_{sp}$
- Application to « sensing »

## Spectroscopy

- Influence of shape, materia, polarization, environment...
- Enhancement
- Life-time

## EELS

- Electronic surface excitations(transitions)
- Dispersion
- Crystallinity

## Imaging

- Spatial distribution
- Controlled enhancement
- Guiding and processing
- Plasmonic circuitry

**POST-DOC(S)  
AND  
PHD(S) WANTED!!**



Bio-plasmonics, integrated plasmonics  
& molecular plasmonics

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