

Microscope de champ proche à pointe diffusante

Yannick De Wilde

Institut Langevin, Ondes et Images

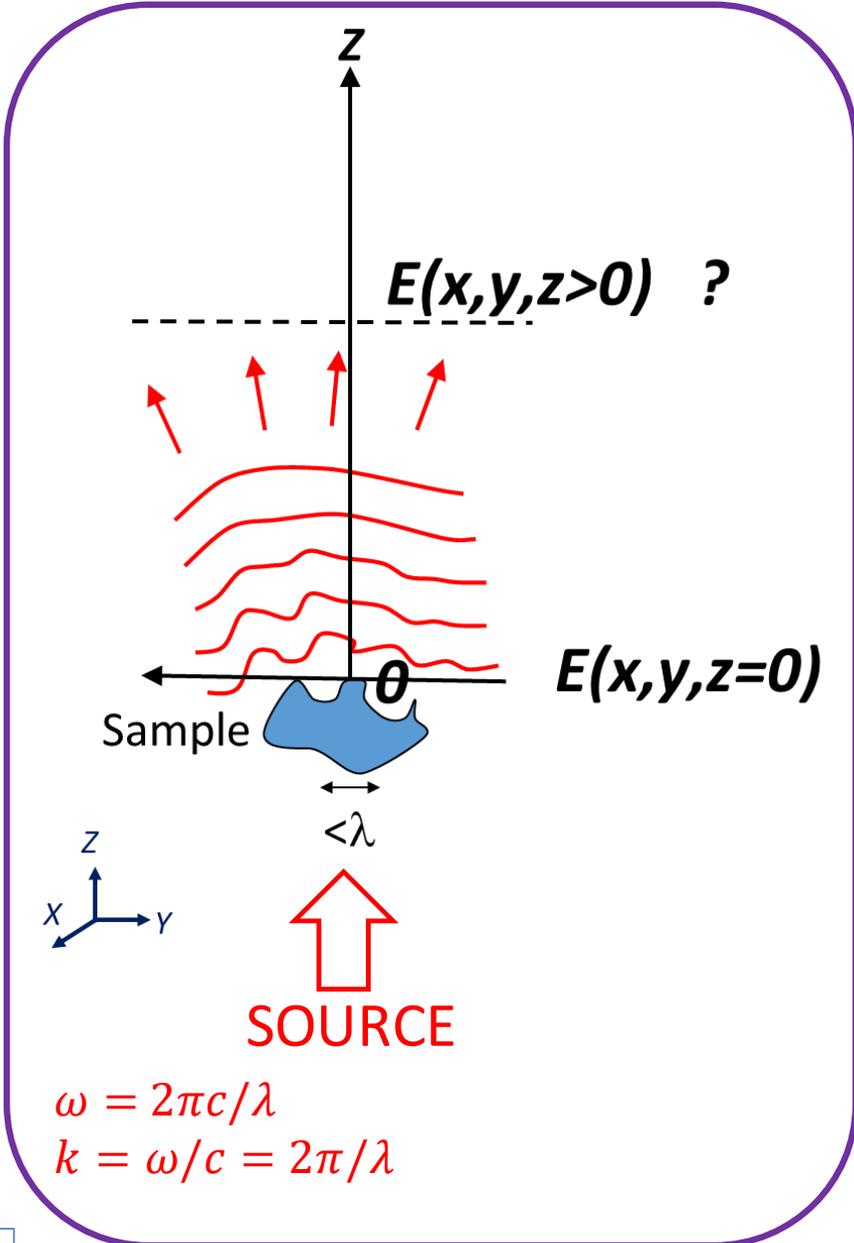
yannick.dewilde@espci.fr



Institut **Langevin**

ONDES ET IMAGES

Basic notions: Angular spectrum representation

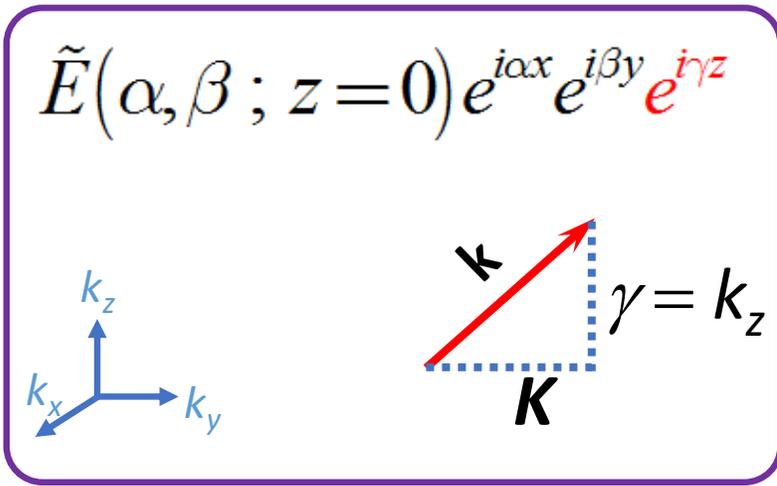


Helmholtz equation :

$$\Delta E + \frac{\omega^2}{c^2} E = 0$$

Solution:

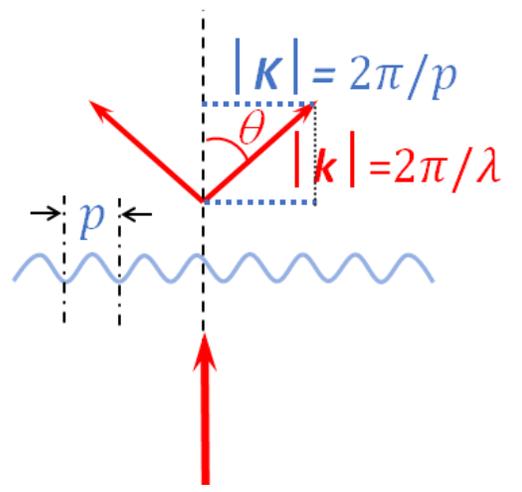
Superposition of plane waves



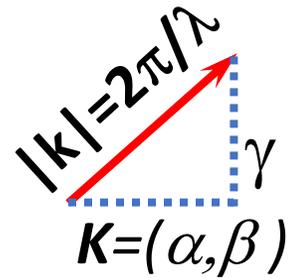
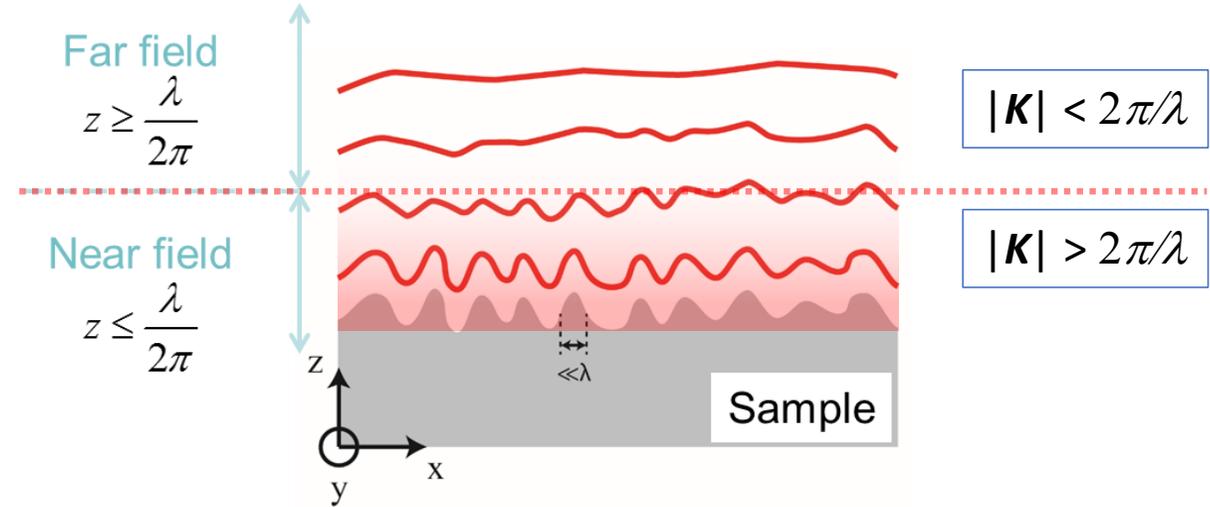
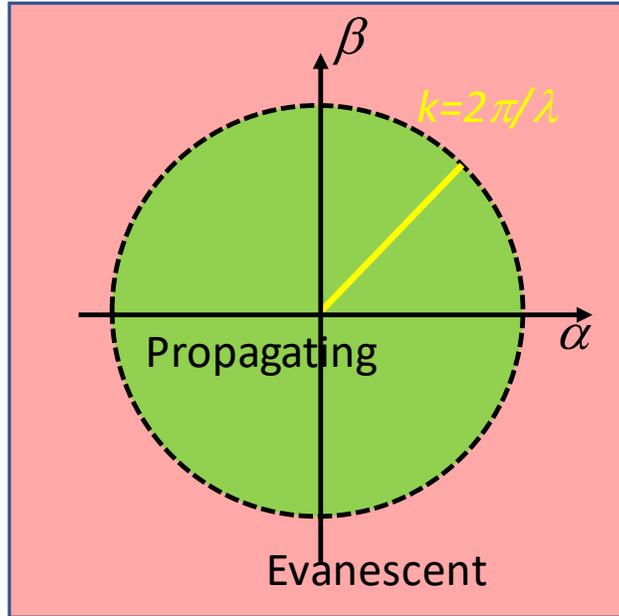
$\mathbf{K}=(\alpha, \beta)$ Transverse (in-plane) wavevector

└─> Spatial frequencies

$$\sin \theta = \frac{\lambda}{p}$$



Evanescent vs. propagating EM fields



$$\left(\frac{2\pi}{\lambda}\right)^2 = |\mathbf{K}|^2 + \gamma^2$$

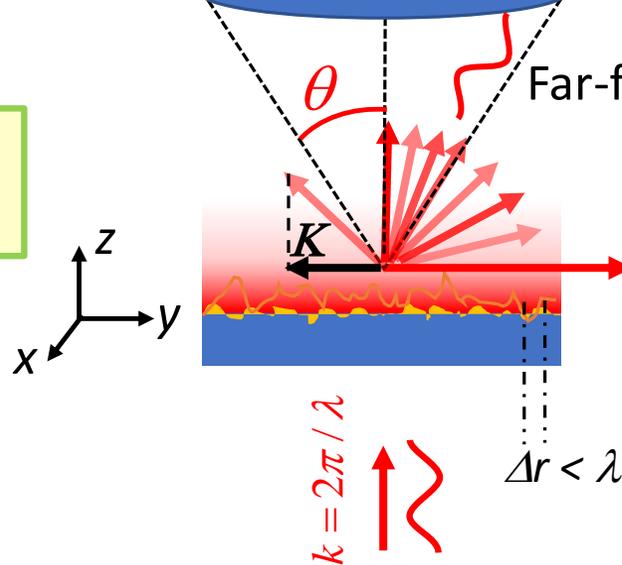
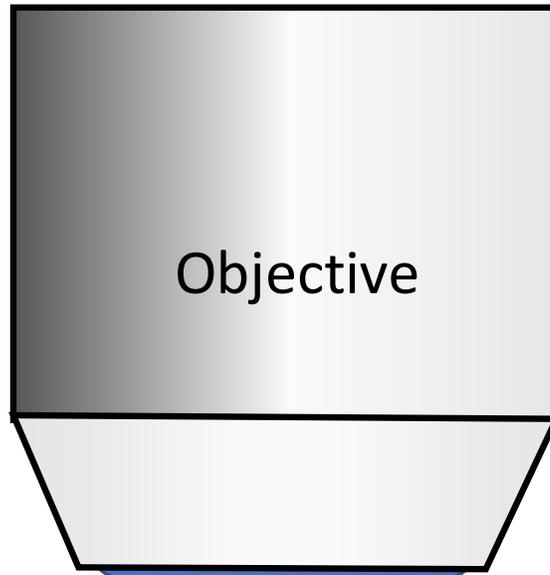
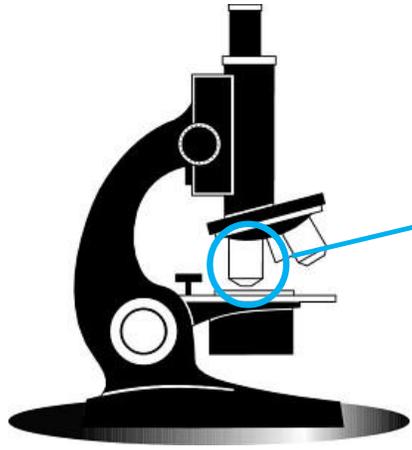
$$\tilde{E}(\alpha, \beta; z=0) e^{i\alpha x} e^{i\beta y} e^{i\gamma z}$$

$|\mathbf{K}| < 2\pi/\lambda$ \longrightarrow Propagating (γ real)

$|\mathbf{K}| > 2\pi/\lambda$ \longrightarrow Evanescent (γ imaginary)

Limits of classical microscopy :

Classical microscope:

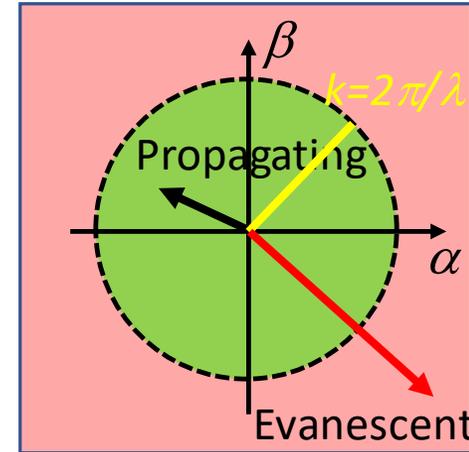


Far-field:
Resolution $\approx 1.22 \lambda / (2 \text{ NA})$

Far-field ($|\mathbf{K}| < |k| = 2\pi/\lambda$)
propagating

Near-field ($|\mathbf{K}| > |k| = 2\pi/\lambda$)
evanescent

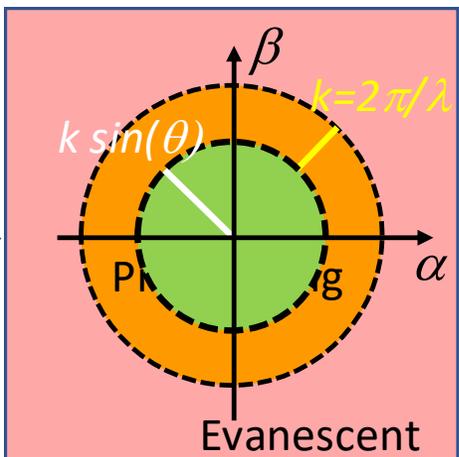
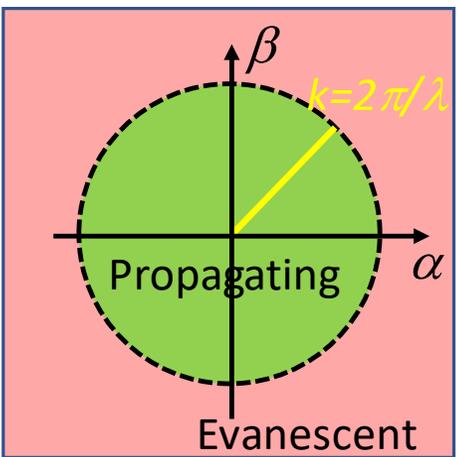
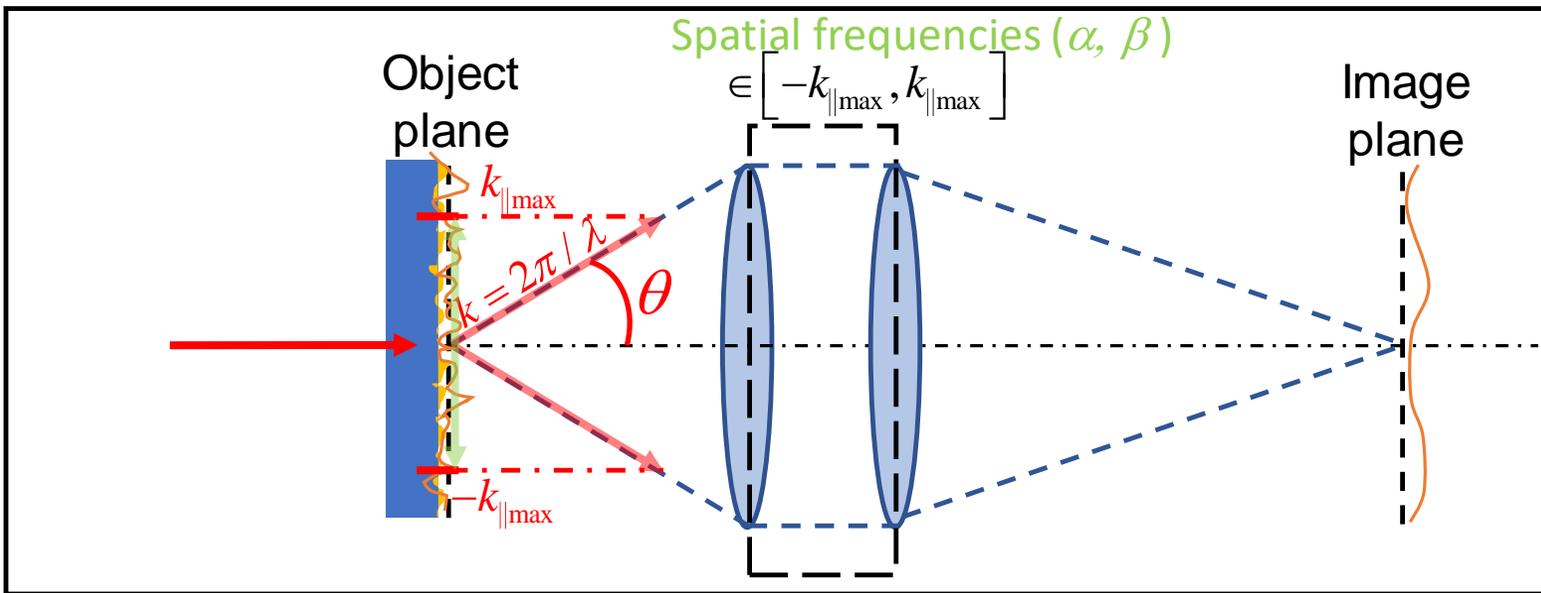
Spatial frequencies in xy plane



$$\gamma^2 = (k_z)^2 = |\mathbf{K}|^2 - (2\pi/\lambda)^2$$

1. Distance= low-pass filter on evanescent fields
- Characteristic decay distance: $\delta \approx \Delta r / 2\pi$

Far-field regime: resolution limit

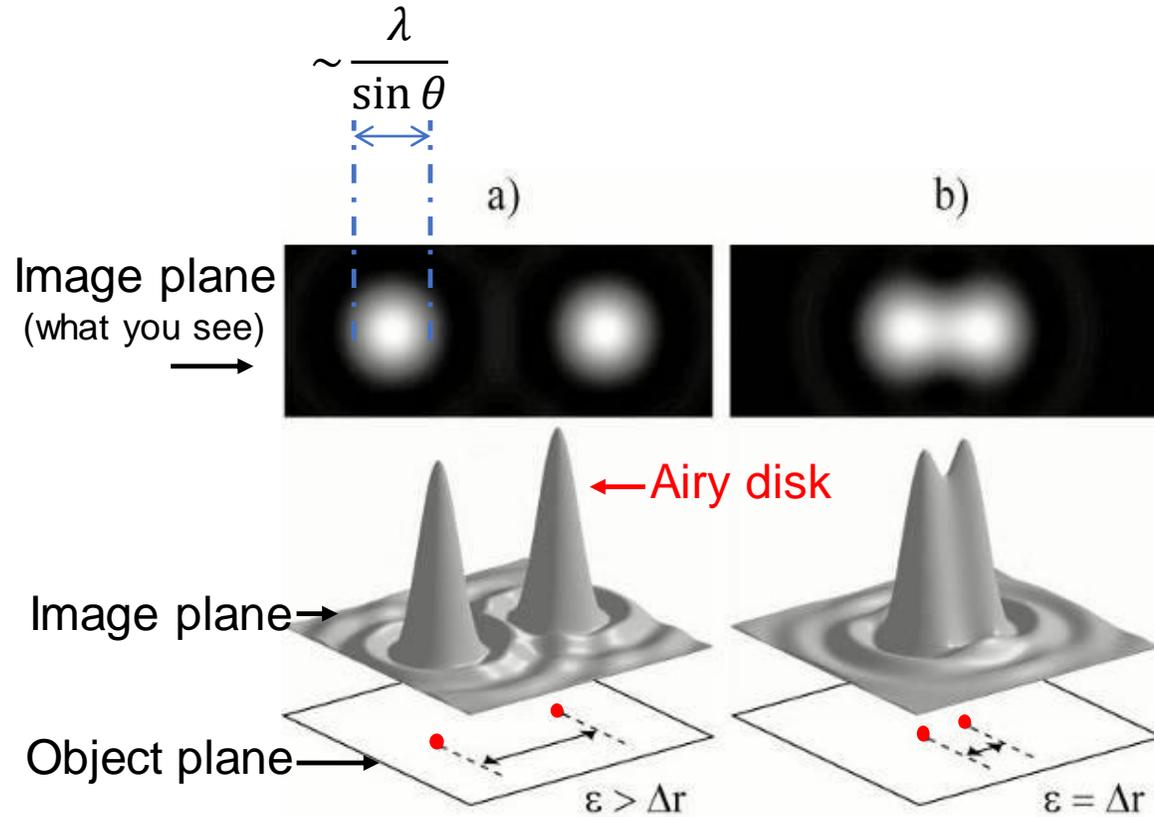


2. The far-field instrument is a low-pass filter of propagating fields. **Diffraction limit.**

Rayleigh criterion :

$$\Delta r = \frac{1.22 \cdot \lambda}{2n \sin \theta}$$

Far-field regime: diffraction limit

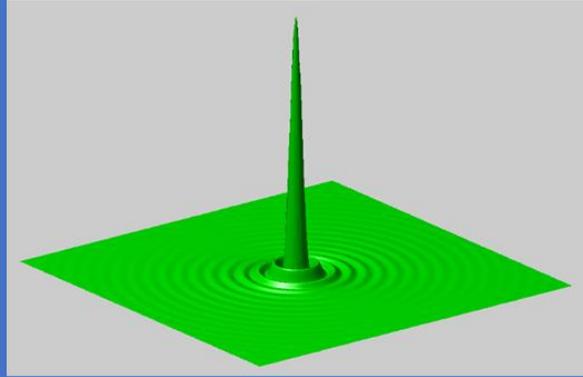


Rayleigh criterion :

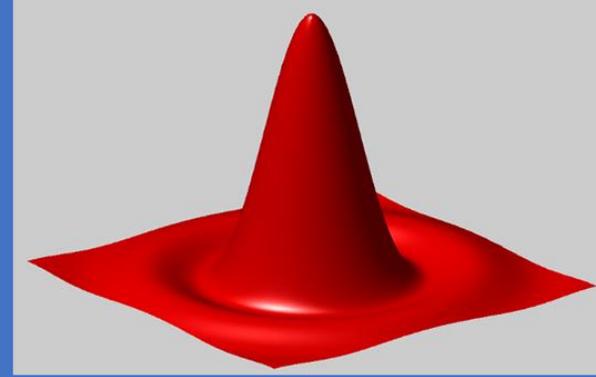
$$\Delta r = \frac{1.22 \cdot \lambda}{2 \sin \theta}$$

Rayleigh criterion (λ dependence)

Image of 1 point : $\lambda = 550$ nm (green)

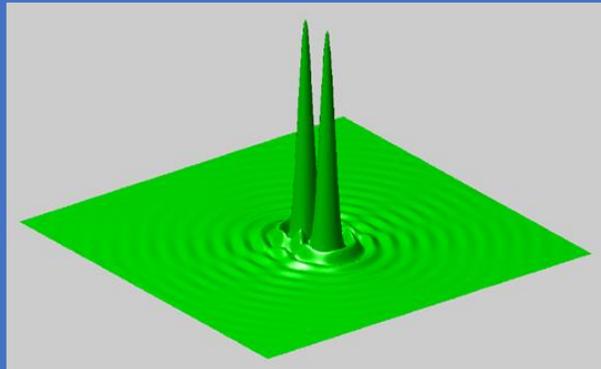


$\lambda = 10$ μm (Infrared)

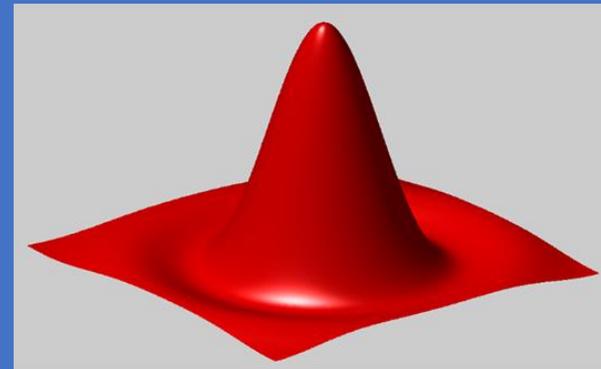


(@ Gilles Tessier)

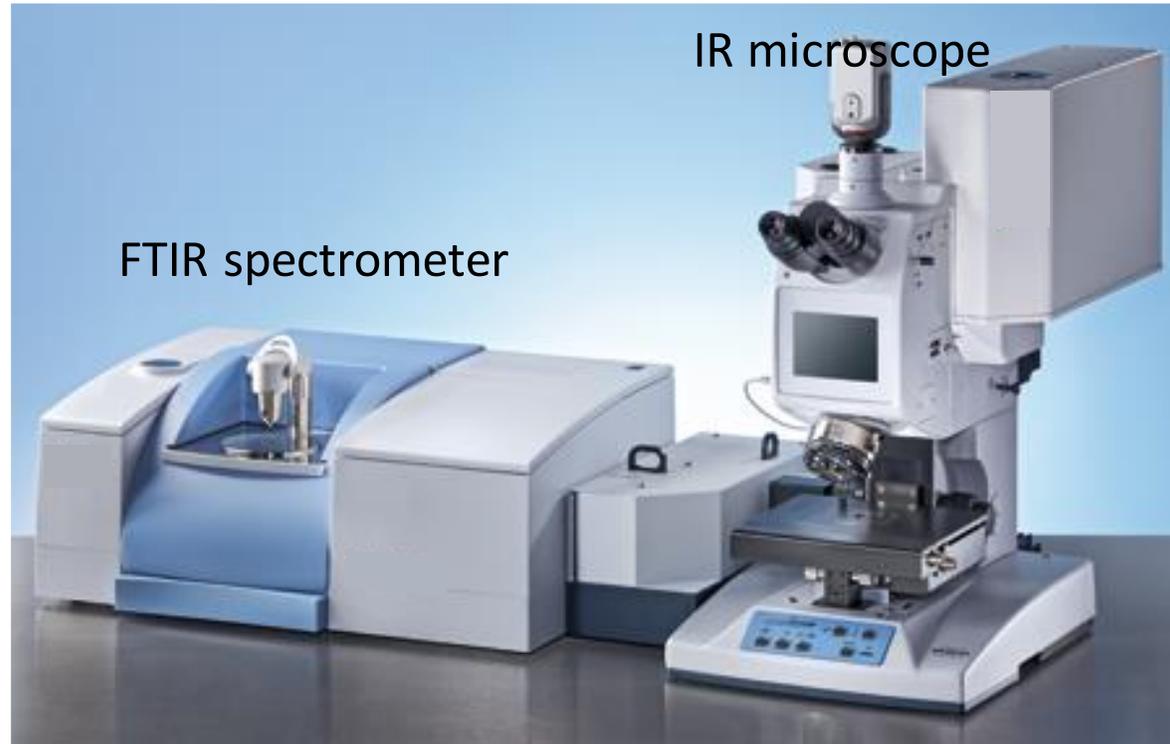
2 points 1000 nm apart :



$\lambda \sim 0.5$ μm (visible)
 $n = 1$; $\sin \theta = 0.95$
 $\Delta r_{vis} \approx 320$ nm

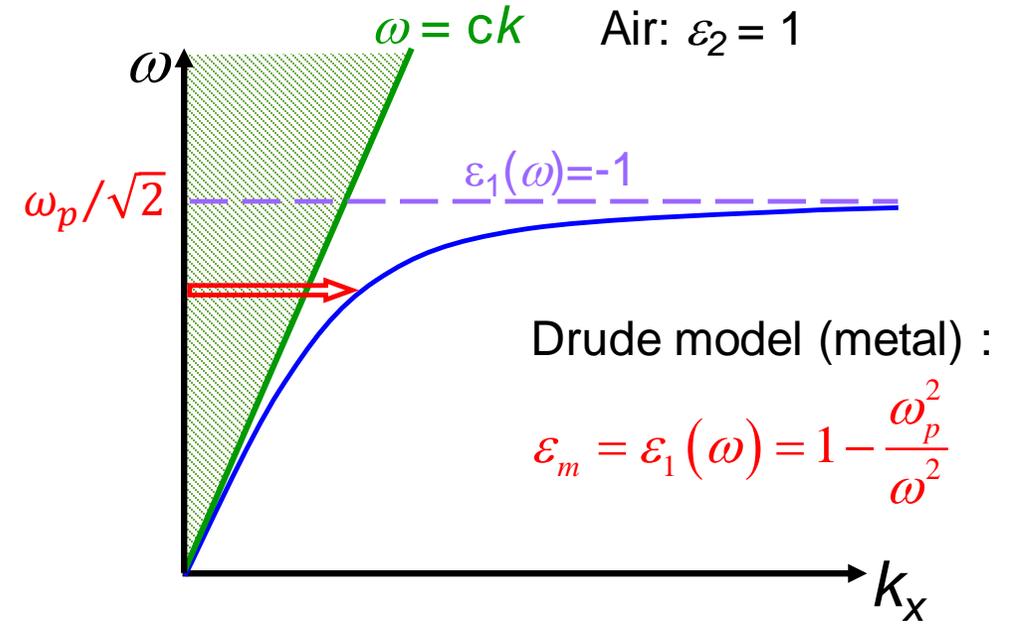
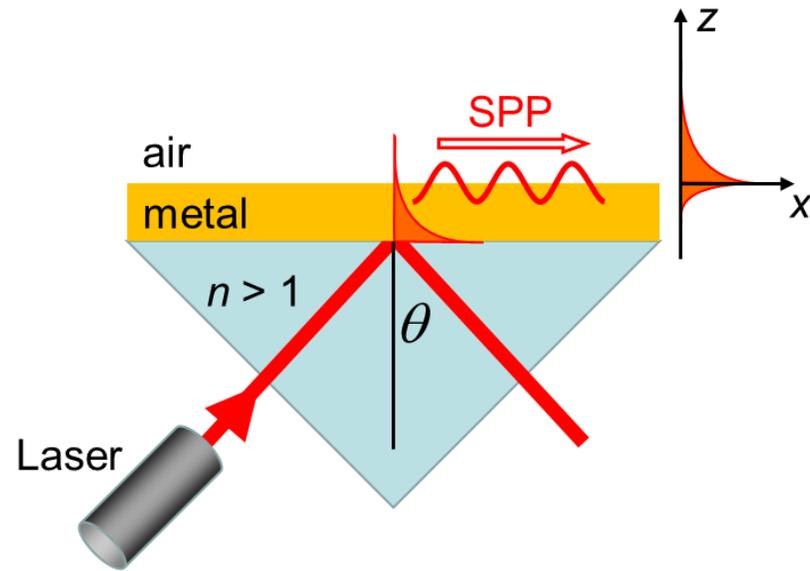


$\lambda \sim 10$ μm (IR)
 $n = 1$; $\sin \theta = 0.75$
 $\Delta r_{IR} \approx 9$ μm

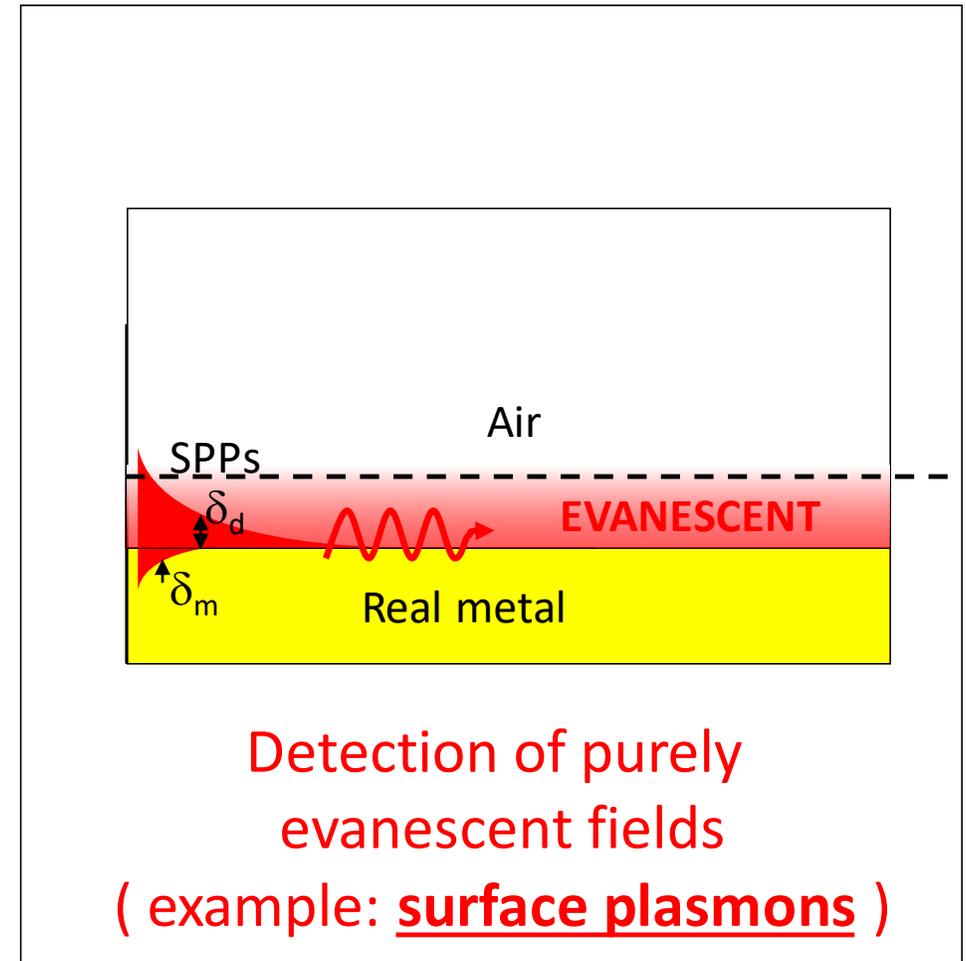
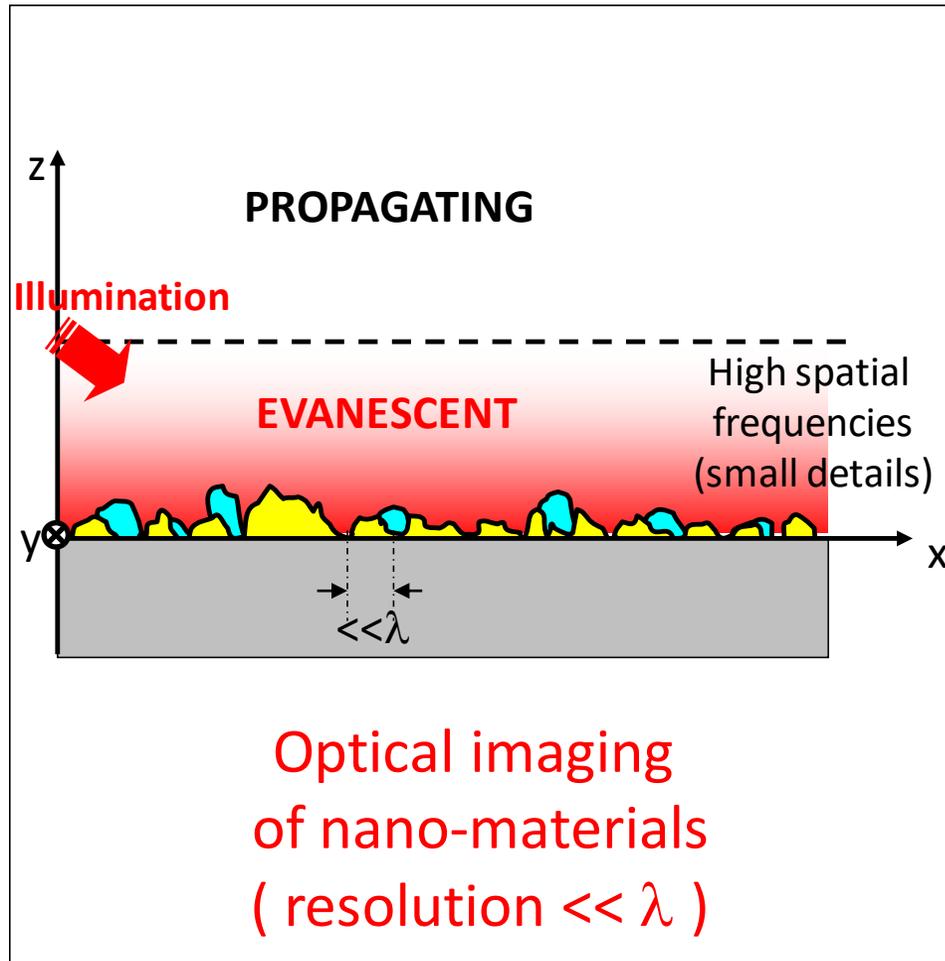


SPATIAL RESOLUTION STRONGLY
LIMITED IN THE INFRARED ($\sim 10 \mu\text{m}$)
+
CANNOT DETECT SURFACE WAVES

Surface plasmons polaritons:

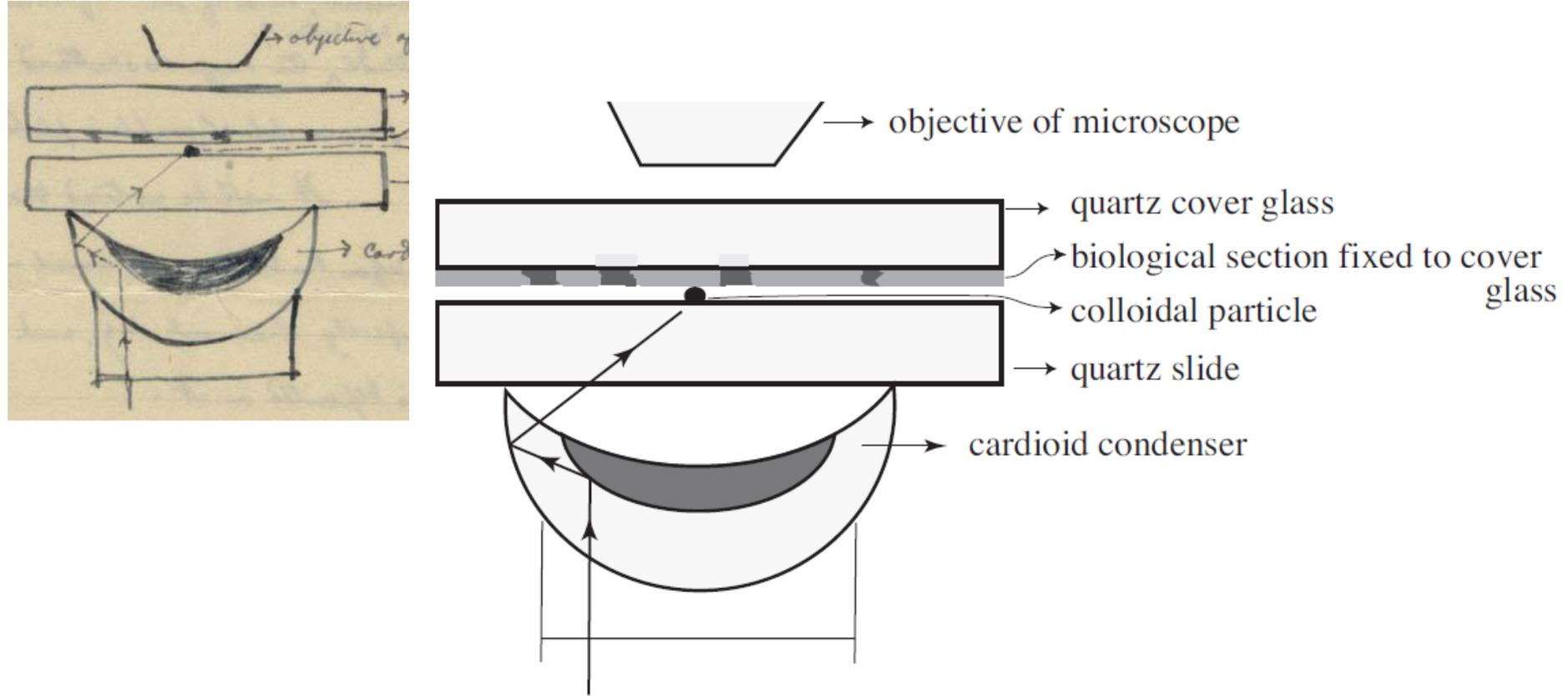


How can we probe the near-field ?



Synge original idea (1928)

Sub- λ LOCAL PROBE = OPTICAL ANTENNA

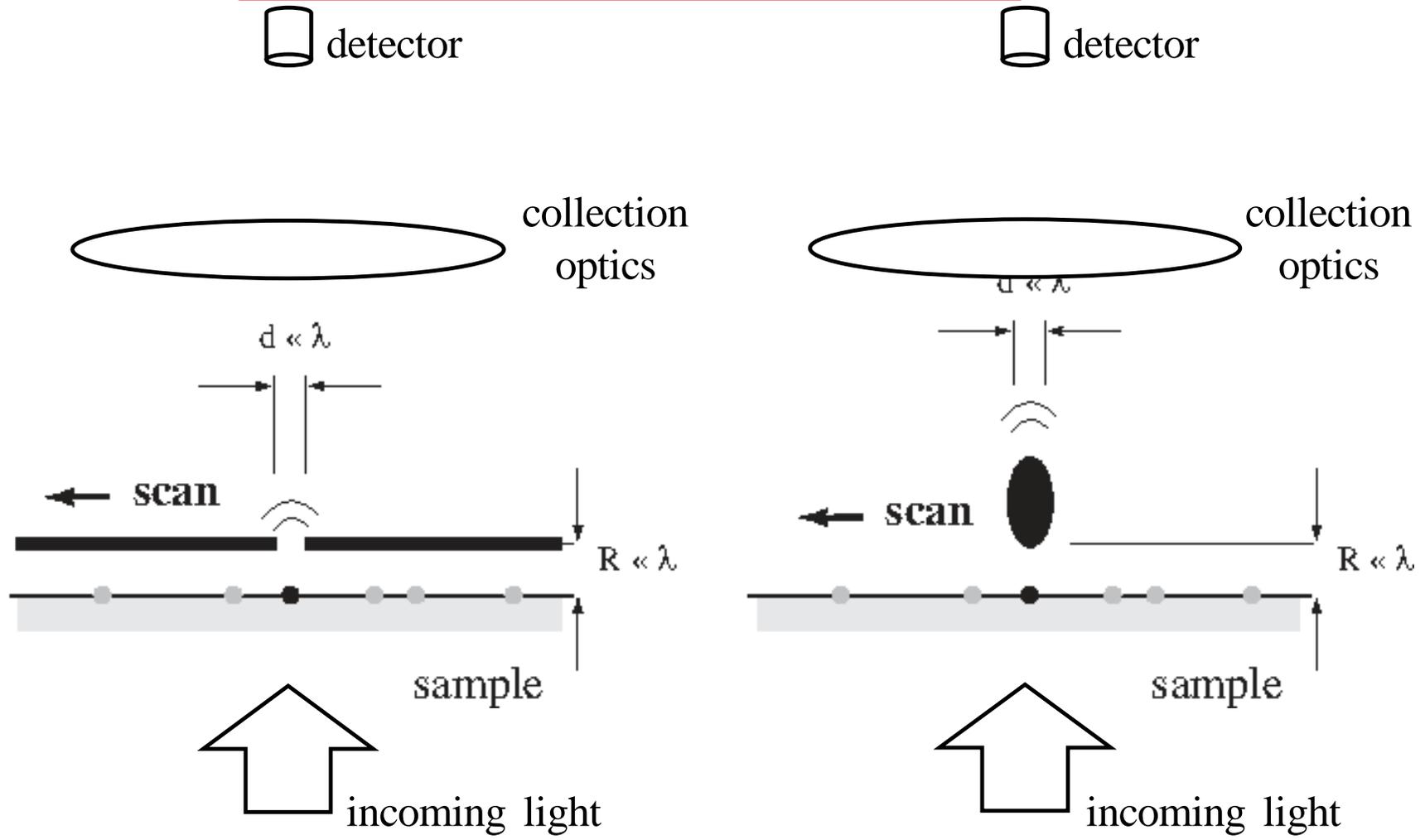


Novotny, "The History of Near-field Optics," *Progress in Optics 50*, E. Wolf (ed.), 2007.

Letter from E.H. Synge to A. Einstein (April 1928)

Syngé original idea (1928)

Sub- λ LOCAL PROBE = OPTICAL ANTENNA

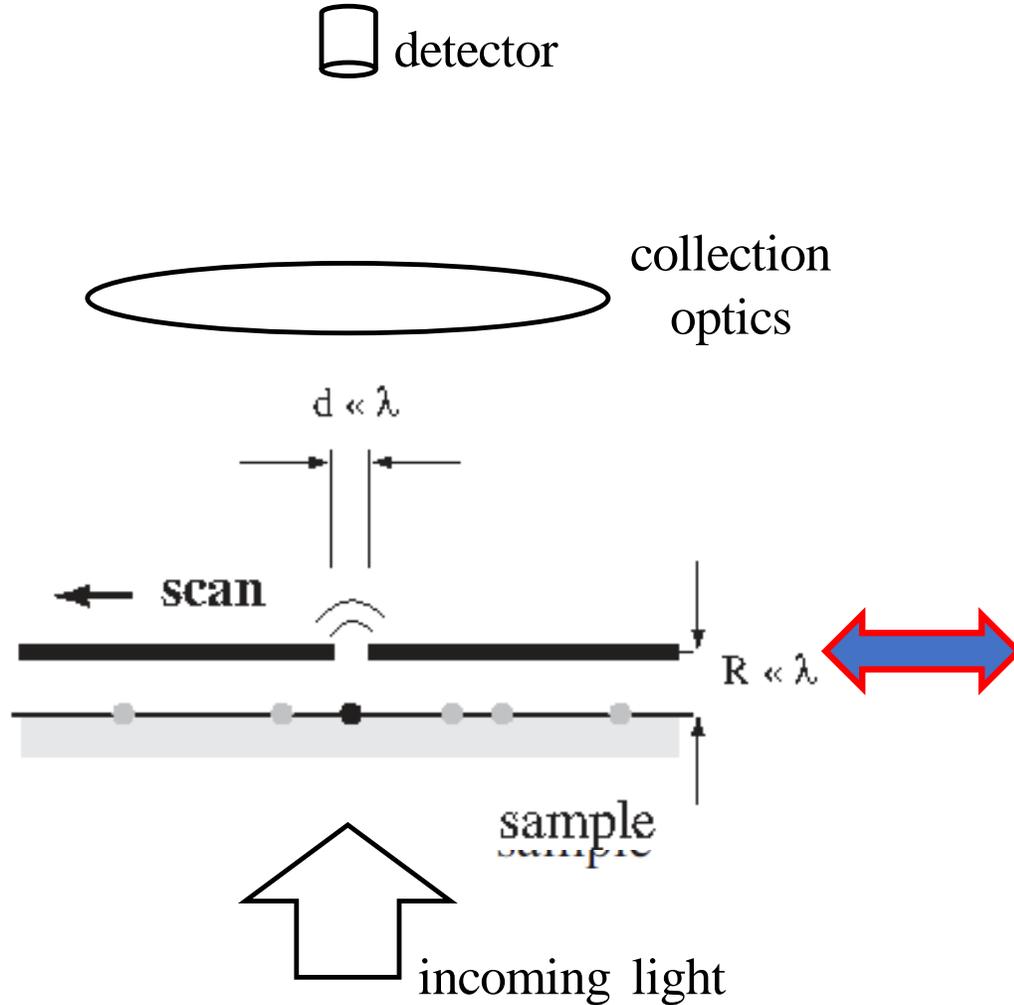


E. H. Syngé, *Phil. Mag.* S.7, **6**, 356 (1928).

Novotny, "The History of Near-field Optics," *Progress in Optics* 50, E. Wolf (ed.), 2007.

Synge original idea (1928)

Sub- λ LOCAL PROBE = OPTICAL ANTENNA



PERFORM THE LATERAL
SCANS USING A PIEZOELECTRIC
ELEMENT

E. H. Synge, *Phil. Mag.S.7*, **13**, 297 (1932).

E. H. Synge, *Phil. Mag. S.7*, **6**, 356 (1928).

History of NSOM

(NSOM = near-field scanning optical microscopy)

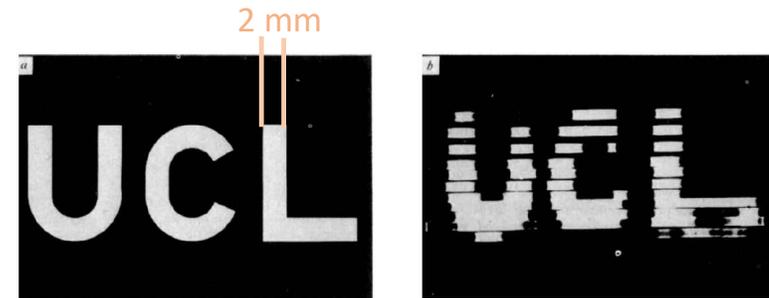
➤ **Concept of « ultramicroscopy » instrument**

E.H. Synge, "A suggested method for extending the microscopic resolution into the ultramicroscopic region," *Phil. Mag.* 6, 356 (1928).

E.H. Synge, "An application of piezoelectricity to microscopy," *Phil. Mag.* 13, 297 (1932).

➤ **Proof of Concept with microwaves ($\lambda=3\text{cm}$)**

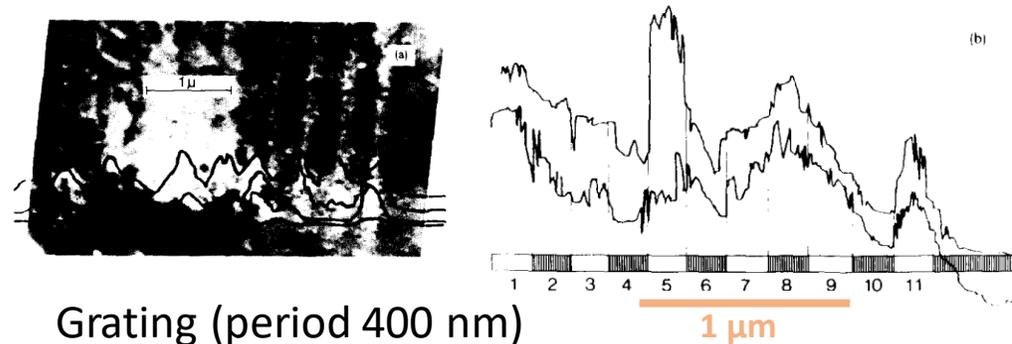
E.A. Ash and G Nichols, *Nature* 237, 510 (1972).



➤ **Sub- λ imaging in the visible ($\lambda=488\text{ nm}$)**

D.W. Pohl, W. Denk, and M. Lanz, « Optical stethoscope », *Appl. Phys. Lett.* 44, 651 (1984).

Resolution = 25 nm
($\lambda/20$)

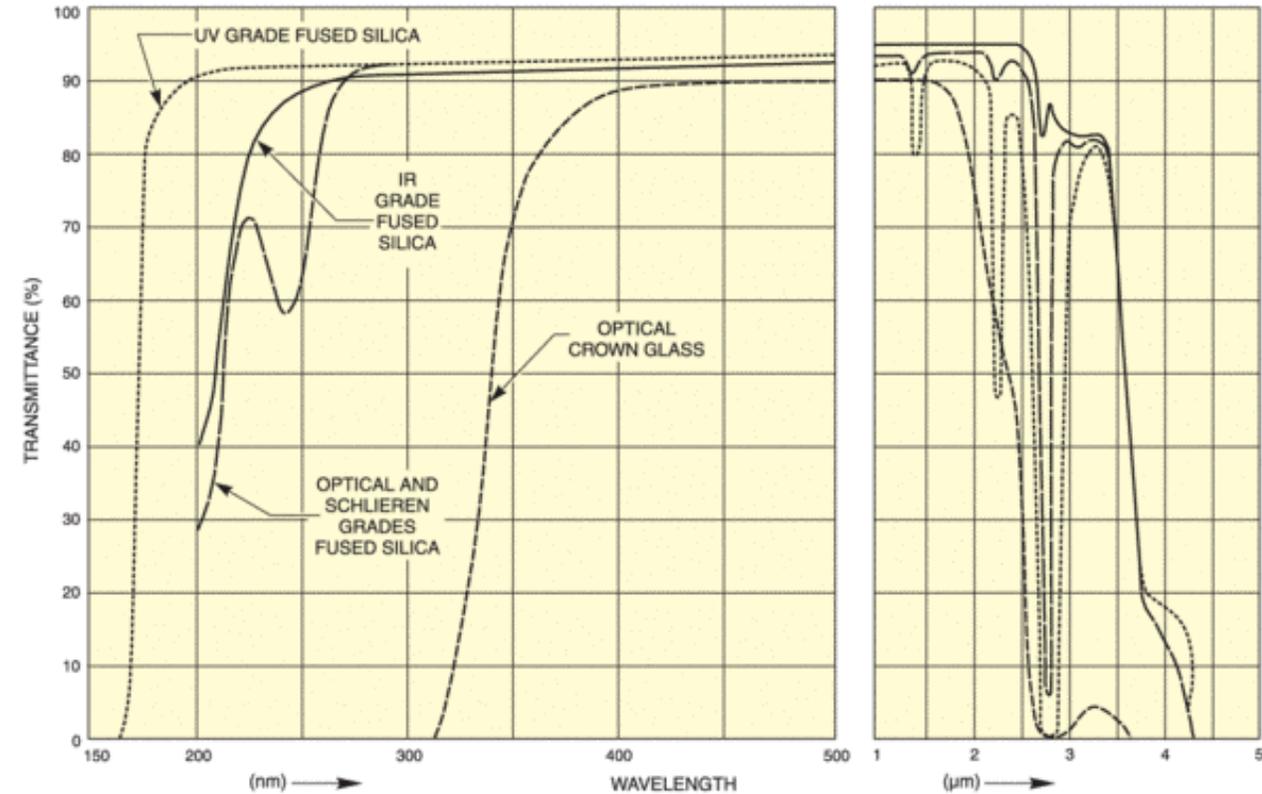
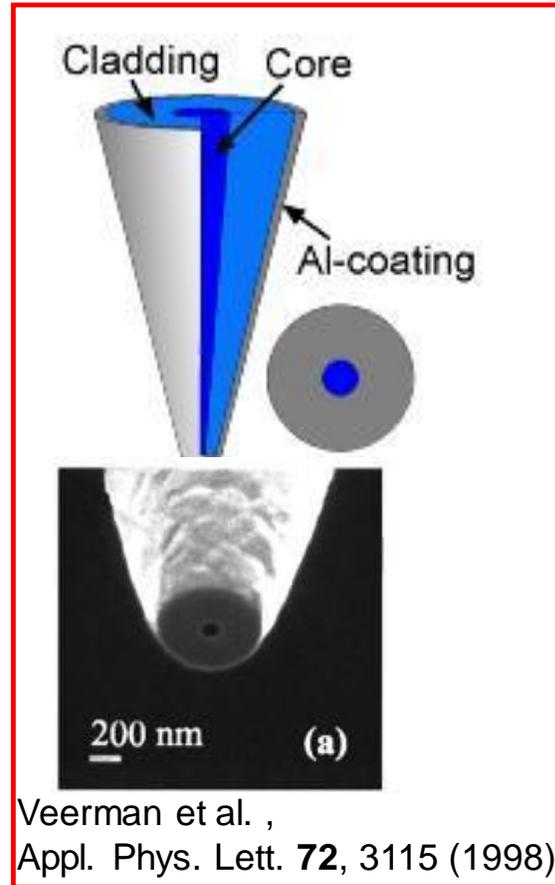
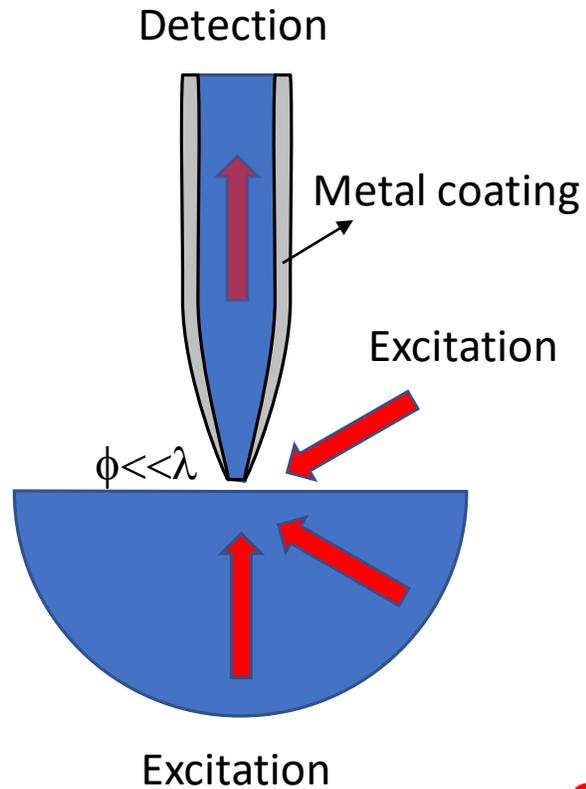


➤ **« Routine » NSOM instrument:**

E. Betzig, J.K. Trautman, *et al.*,
Science 251, 1468 (1991)

Aperture NSOM

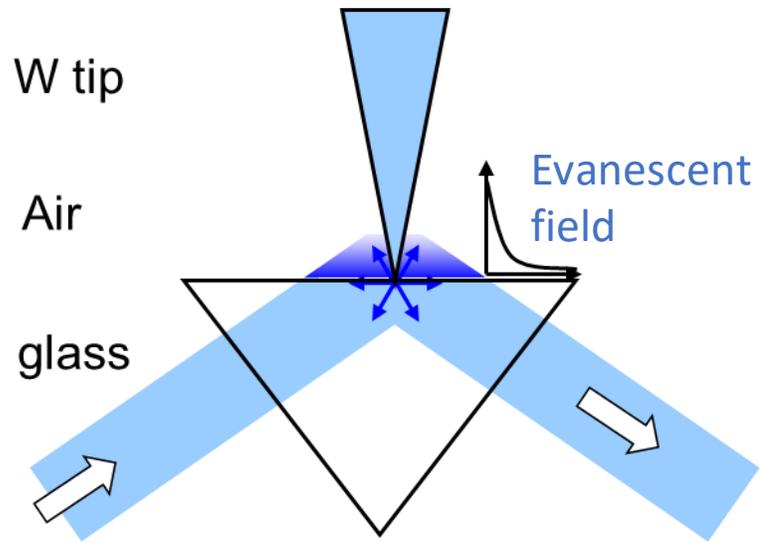
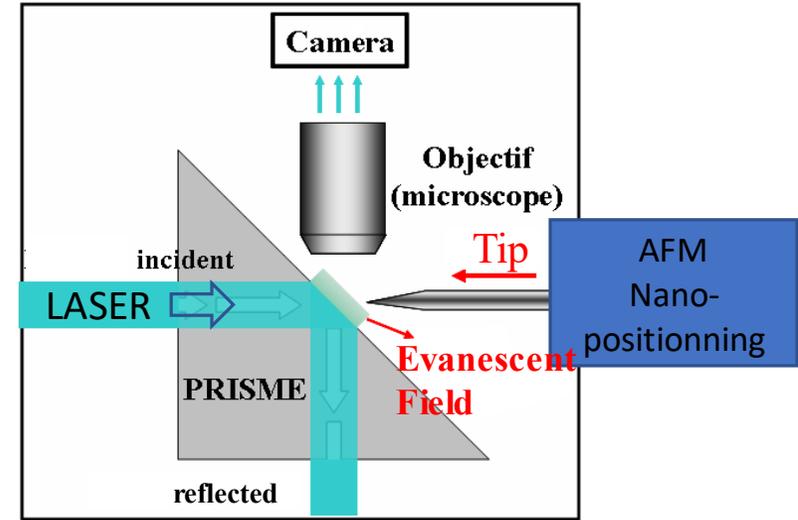
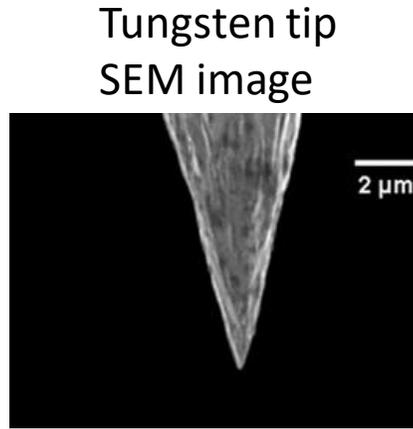
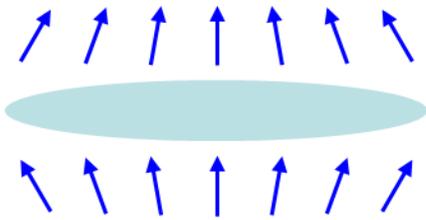
Tapered
optical fiber



Spectral limitation

Silica fiber : Well-suited for visible and near-IR but not for the mid-IR (nor for THz) !!!

Photon tunneling experiment with scattering tip



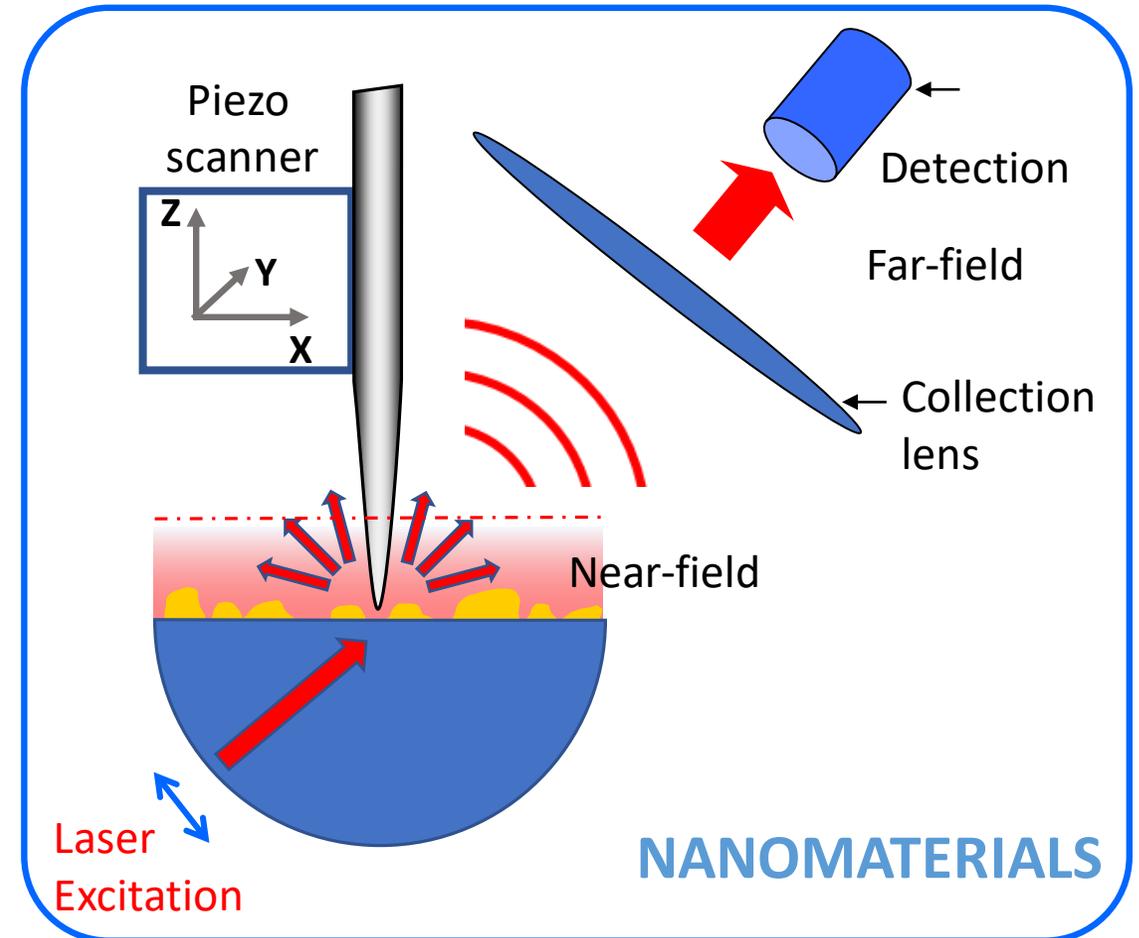
Scattering-type near-field scanning optical microscope (Scattering-type NSOM)

Principle : controlled scanning of a scattering nano-object (tip apex)

$$I_{scat.}(x_t, y_t) = \sigma |E(x_t, y_t)|^2$$

By measuring $I_{scat.}(x_t, y_t)$,
the local field $E(x_t, y_t)$ at the
tip's position is probed.

Optical imaging at sub- λ scale
« **SUPER-RESOLUTION** »



Claude Boccara

Wikramasynghé et al., APL 65,1623 (1994)

Bachelot, Gleize, Boccara, Microanal. Microstruct. 5, 389 (1994)



Scattering-type near-field scanning optical microscope (Scattering-type NSOM)

Principle : controlled scanning of a scattering nano-object (tip apex)

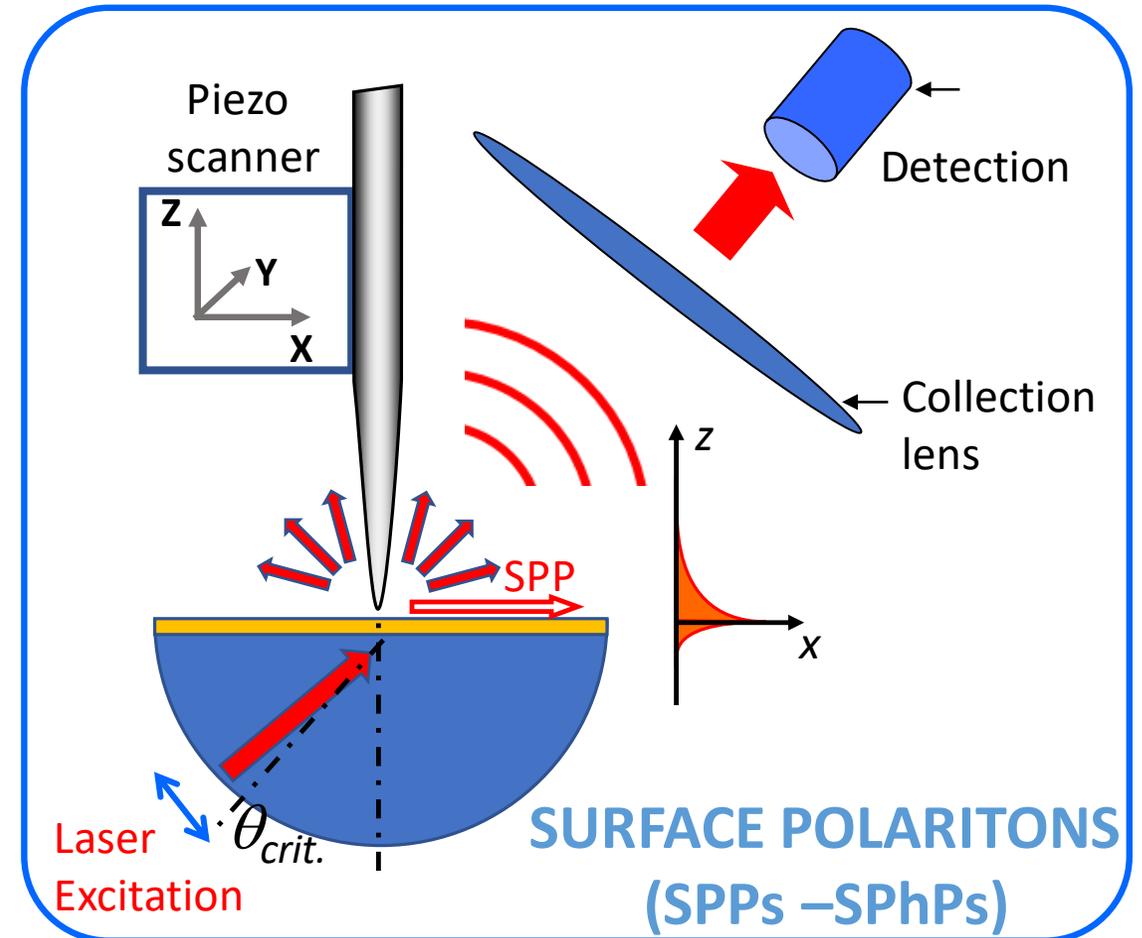
$$I_{scat.}(x_t, y_t) = \sigma |E(x_t, y_t)|^2$$

By measuring $I_{scat.}(x_p, y_t)$,
the local field $E(x_p, y_t)$ at the
tip's position is probed.

Optical imaging at sub- λ scale
« **SUPER-RESOLUTION** »



Claude Boccara

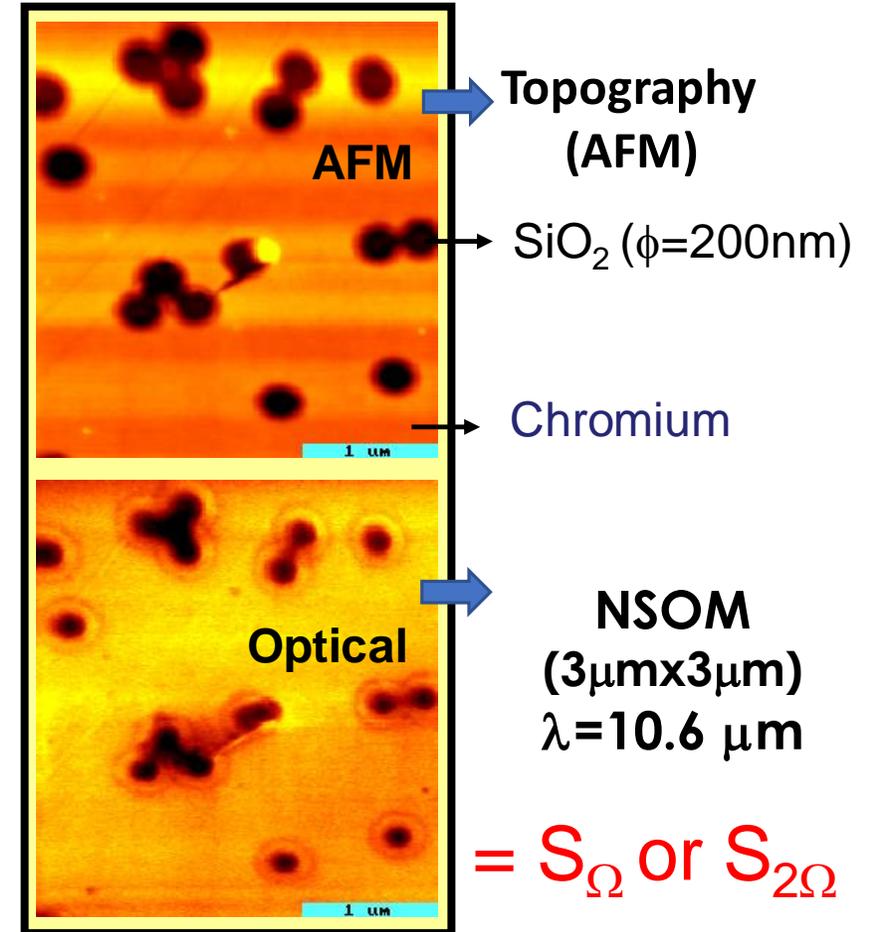
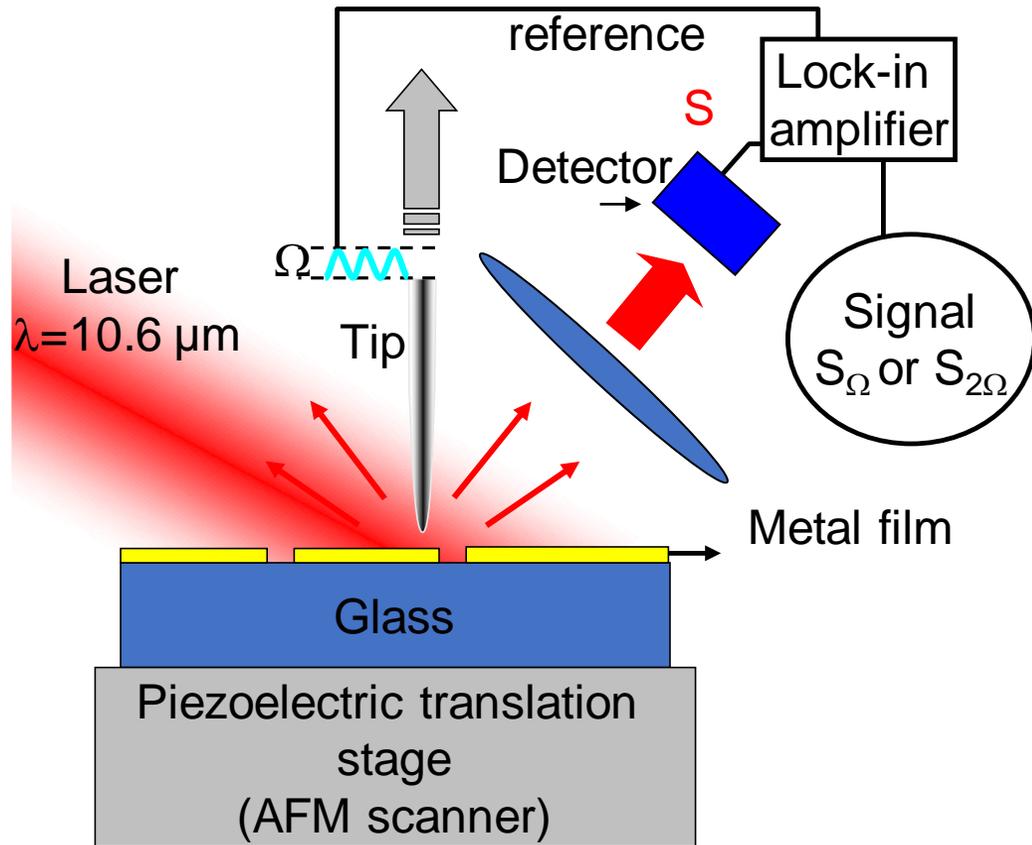


Wikramasynge et al., APL 65,1623 (1994)

Bachelot, Gleize, Boccara, Microanal. Microstruct. 5, 389 (1994)



Sub- λ imaging of nano materials with external IR source



Formanek, et al., JAP **93**, 9548 (2003)

$\lambda = 10.6 \mu\text{m}$



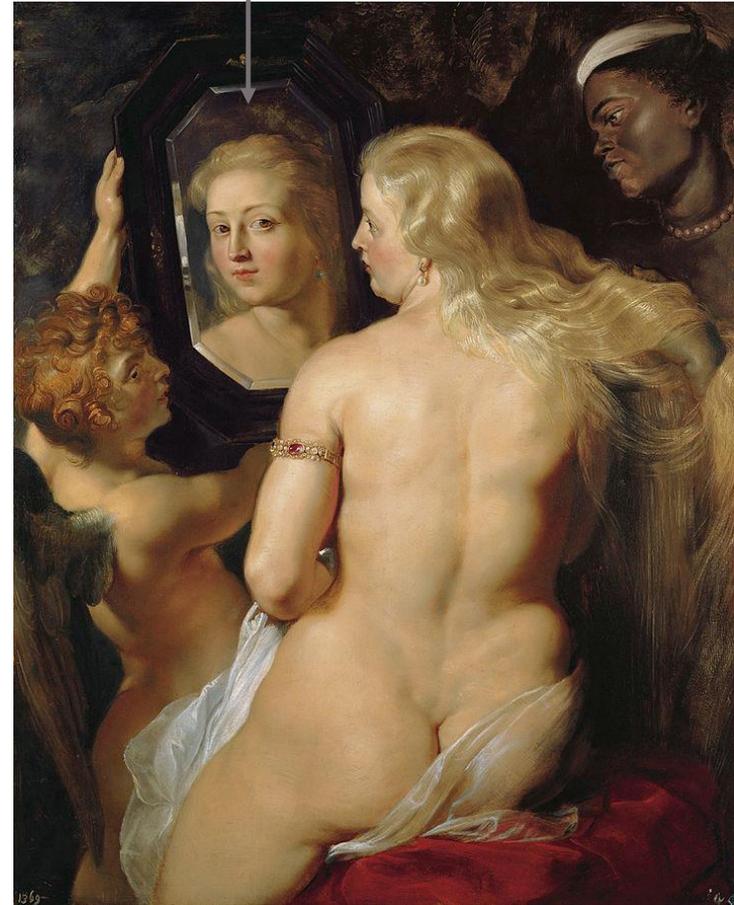
Origin of the contrast

FAR-FIELD



Caravaggio
Narcisse (1598-1599)

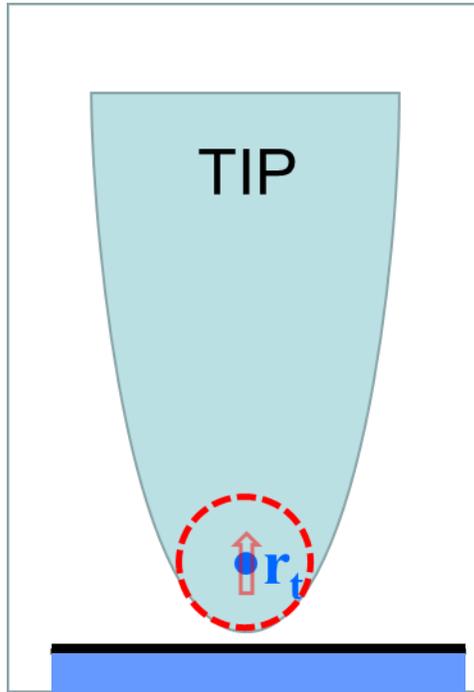
METAL



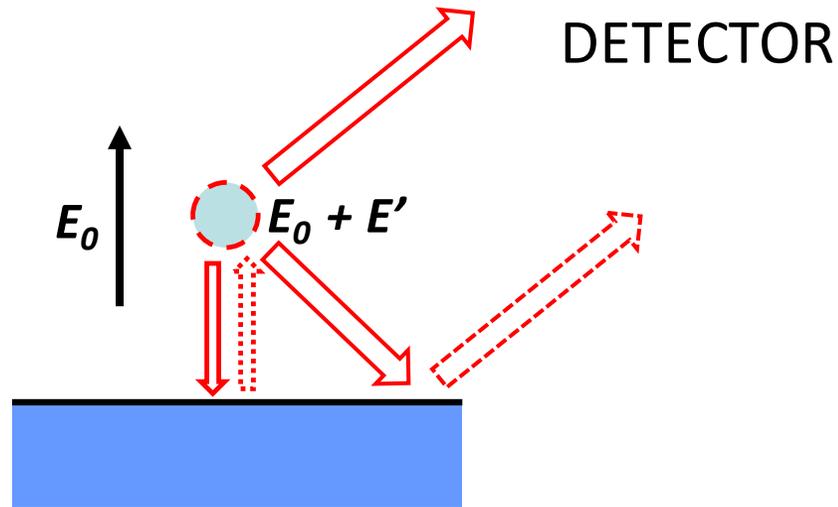
Pierre Paul Rubens
Venus au miroir (1613-1614)

Origin of the contrast

NEAR-FIELD



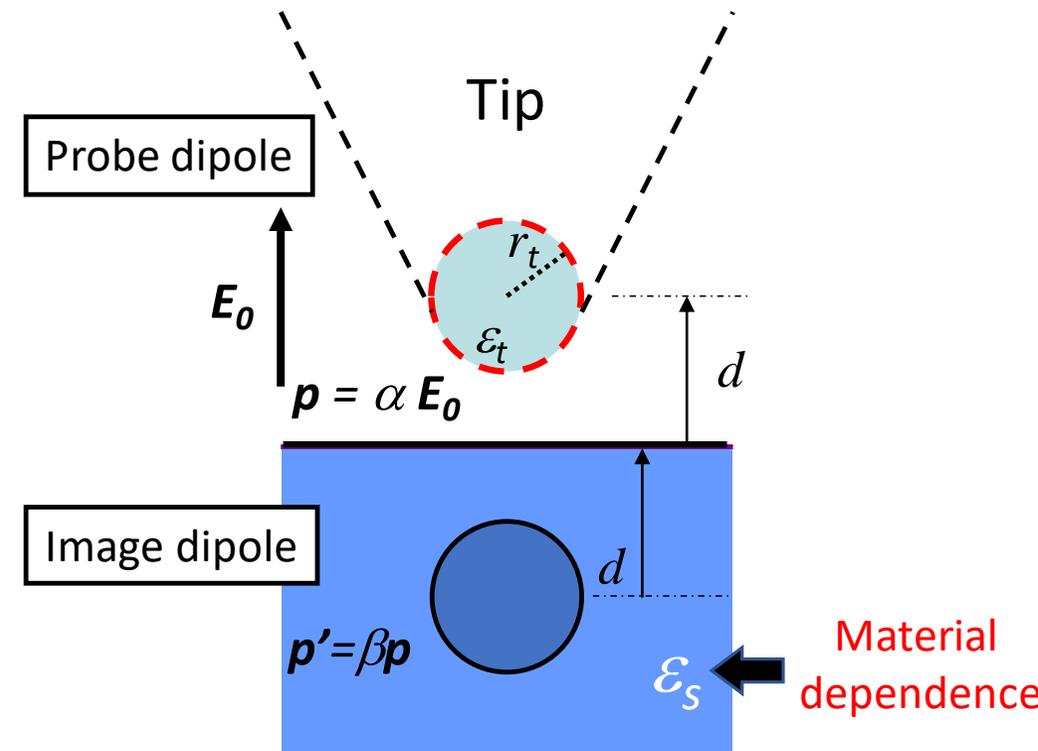
λ



Probe/image dipole model

Effective scattering cross section:

$$\sigma(\varepsilon_t, \varepsilon_s)$$



B. Knoll and F. Keilmann, *Opt. Comm.* **182**, 321 (2000).
 K. Joulain *et al.*, *JQSRT* **136**, 1-15 (2014).

$$E' = E_{dipole} = \frac{p}{2\pi r^3} \quad \text{Avec } r=2d$$

Example: Sample Au + SiC

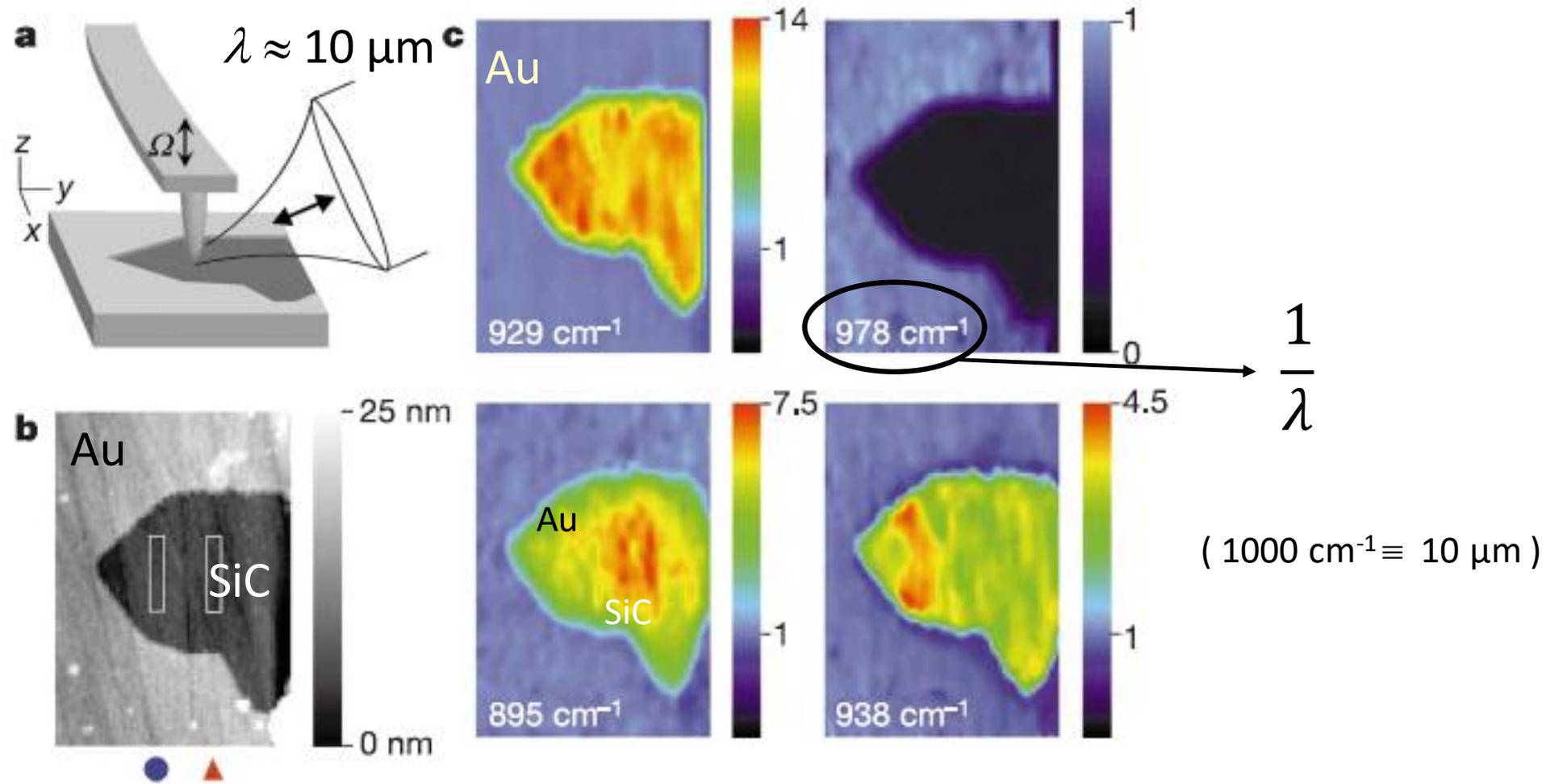
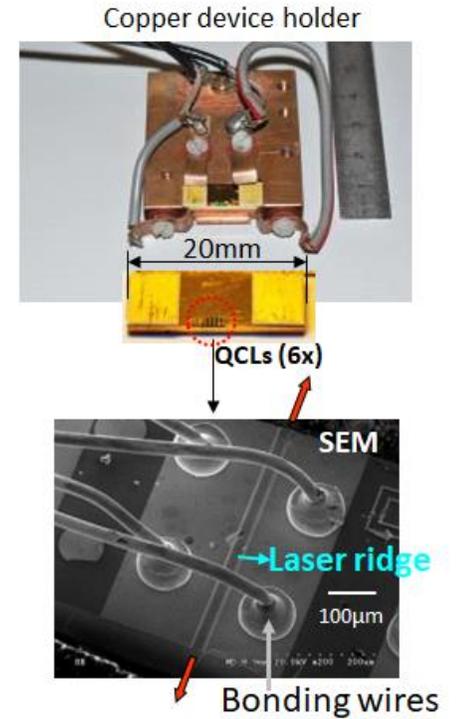
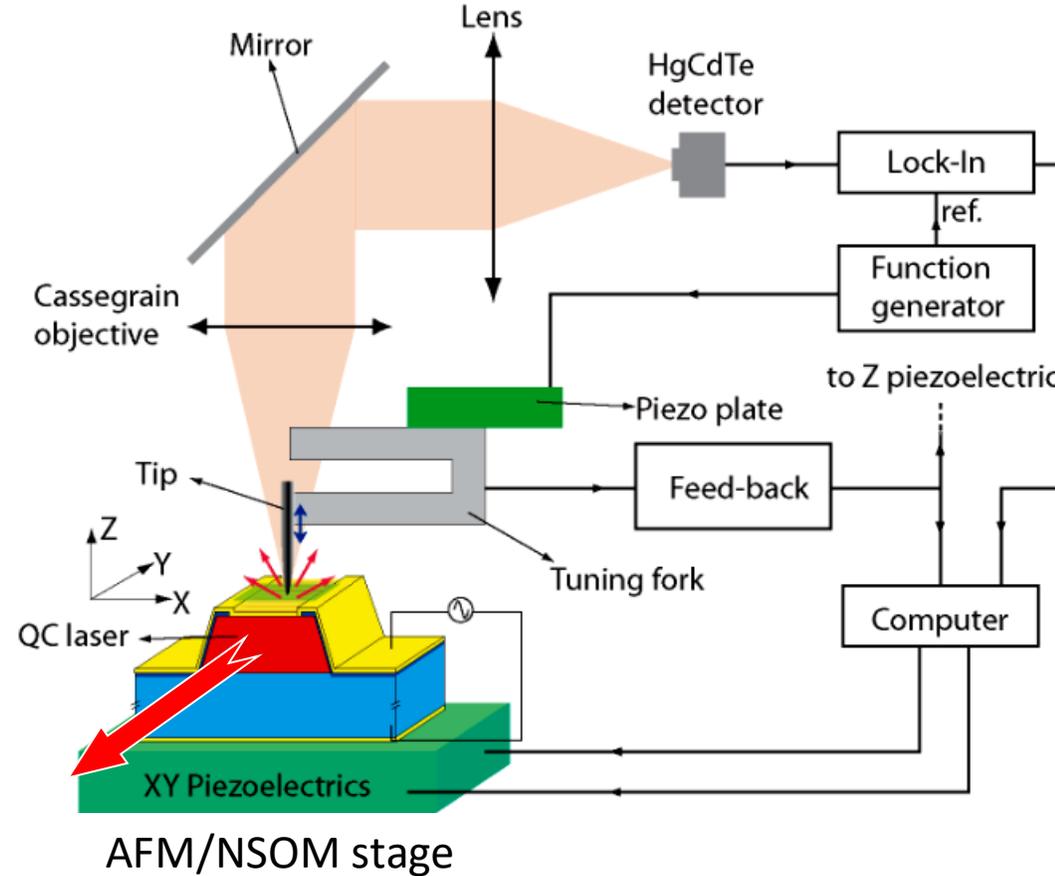
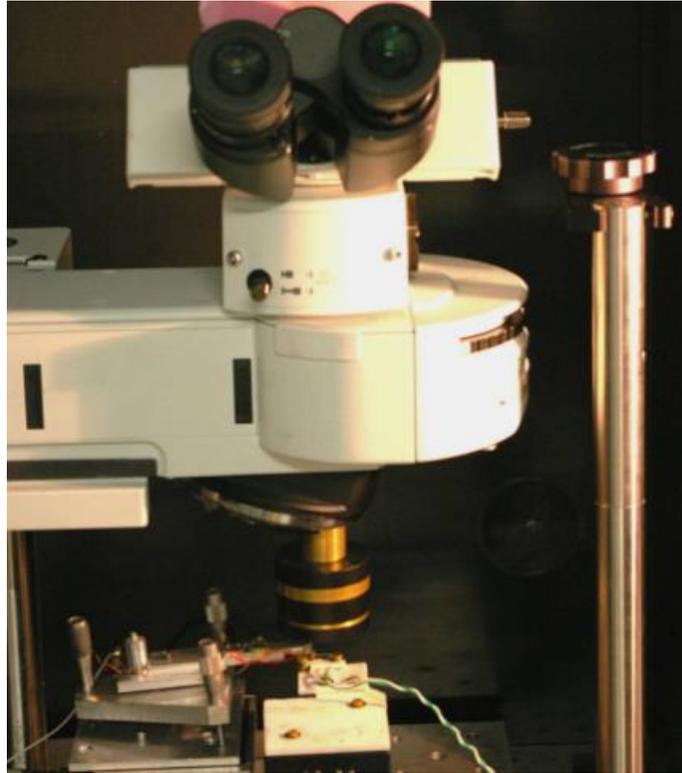


Image size: $1.6 \times 2.3 \mu\text{m}$

Hillenbrand et al. , Nature **418**, 159 (2002)

Scattering-type NSOM (s-NSOM) on active plasmonic devices (QCLs)



Collaboration: R. Colombelli, A. Bousseksou



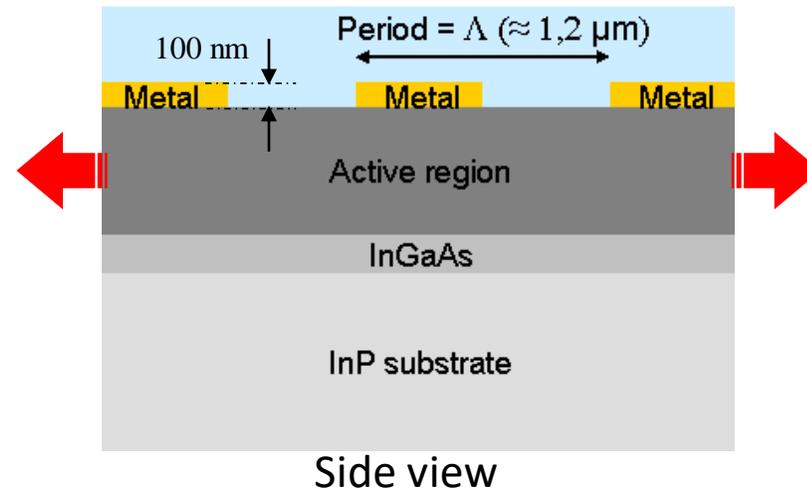
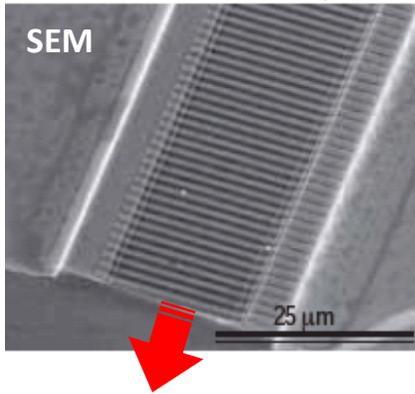
QCLs: Quantum Cascade Lasers

QCL with metal grating

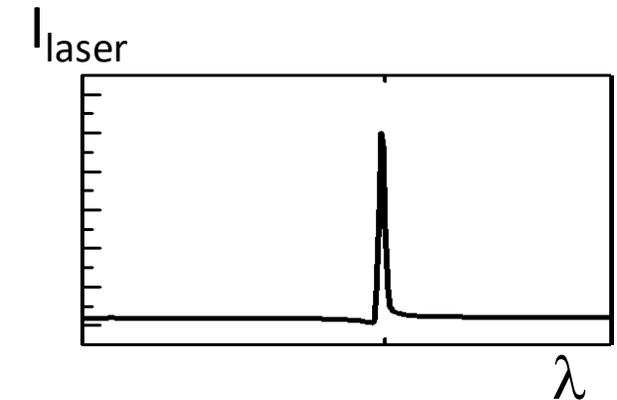
$\lambda \approx 7.5 \mu\text{m}$

$$\Lambda \approx \lambda / 2n_{\text{eff.}}$$

SEM image



Far-field spectrum



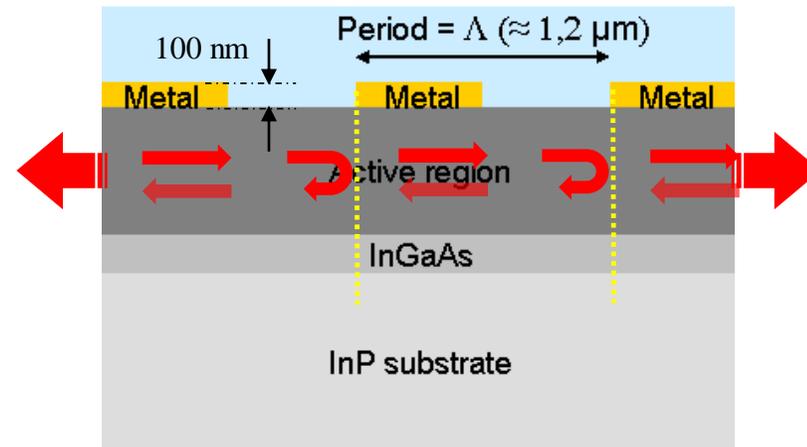
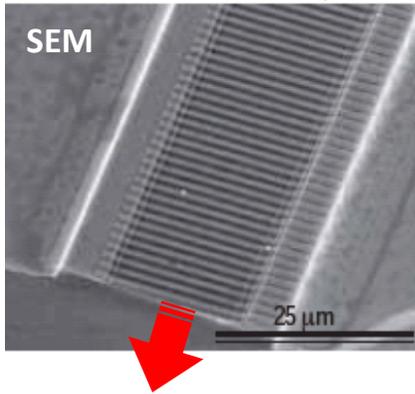
→ Single mode emission

QCL with metal grating

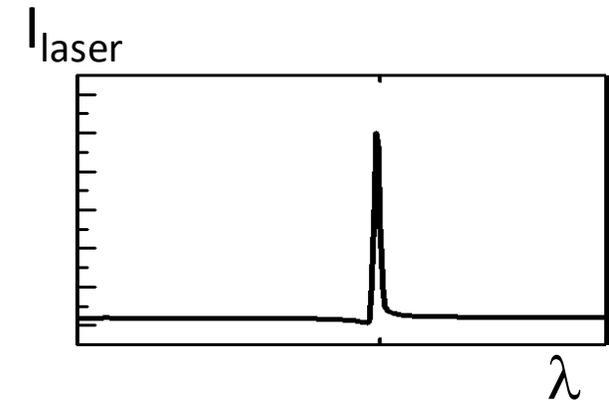
$\lambda \approx 7.5 \mu\text{m}$

$$\Lambda \approx \lambda / 2n_{\text{eff.}}$$

SEM image 

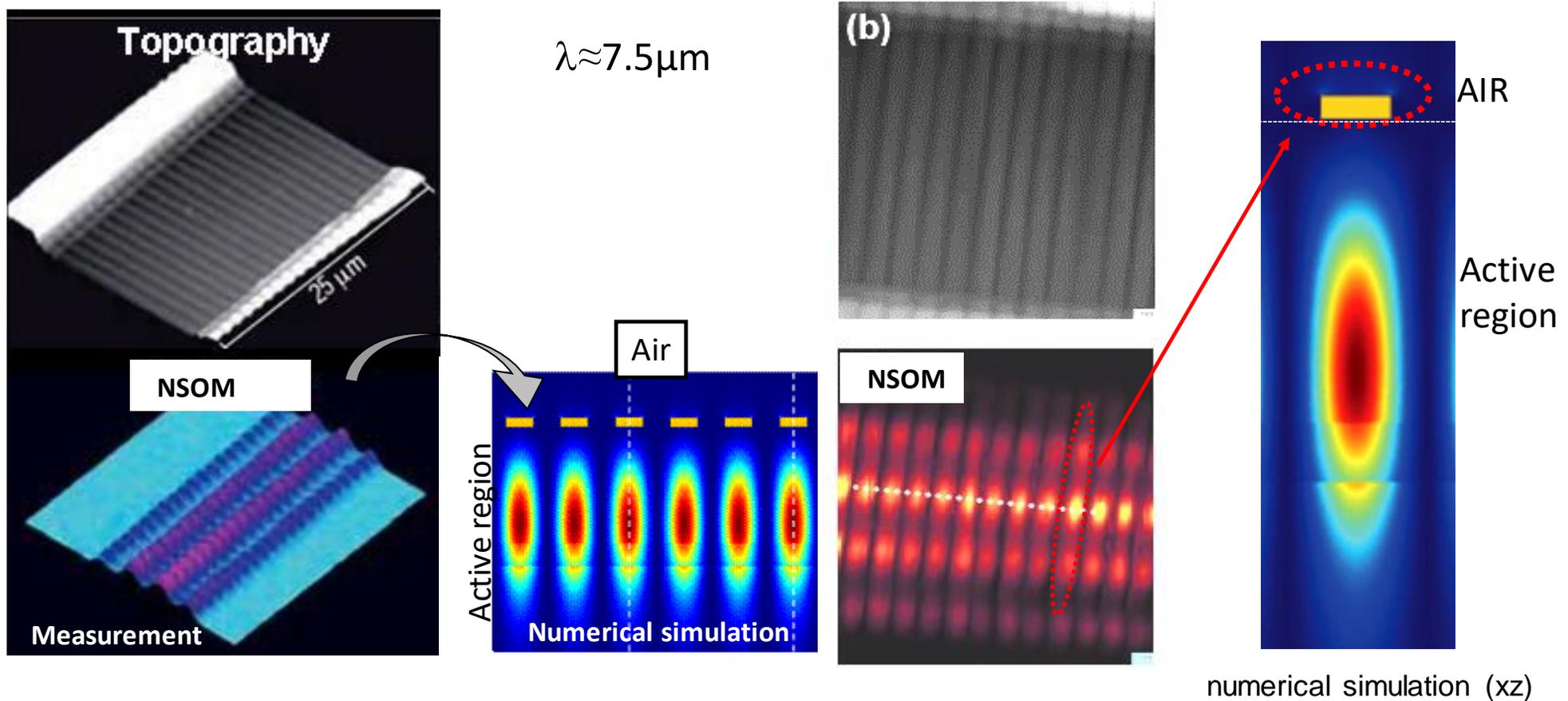


Far-field spectrum



 Single mode emission

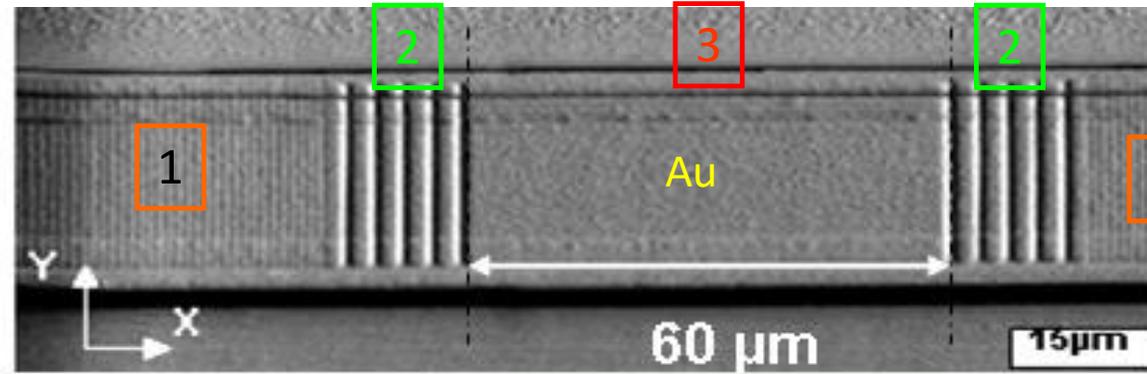
QCL with metal grating : NSOM images



Direct visualisation of the EM modes
Hybrid surface plasmons are generated on the metal grating

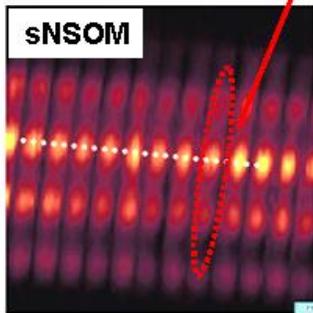
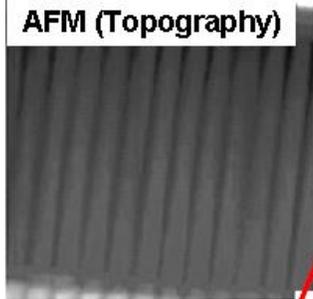
Building block of active plasmonics: generator, coupler, passive SPP waveguide

Measured topography (AFM)



1. DFB grating

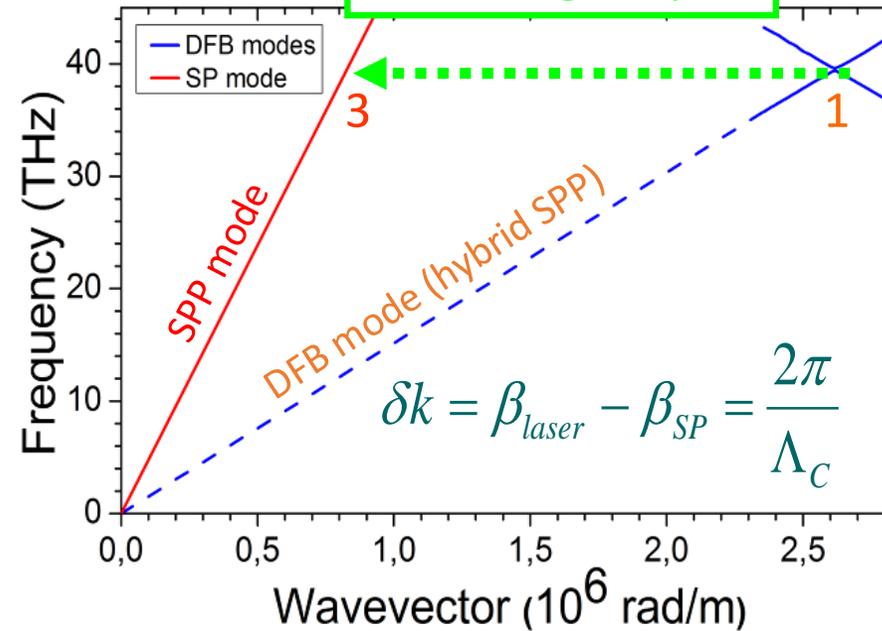
Generator



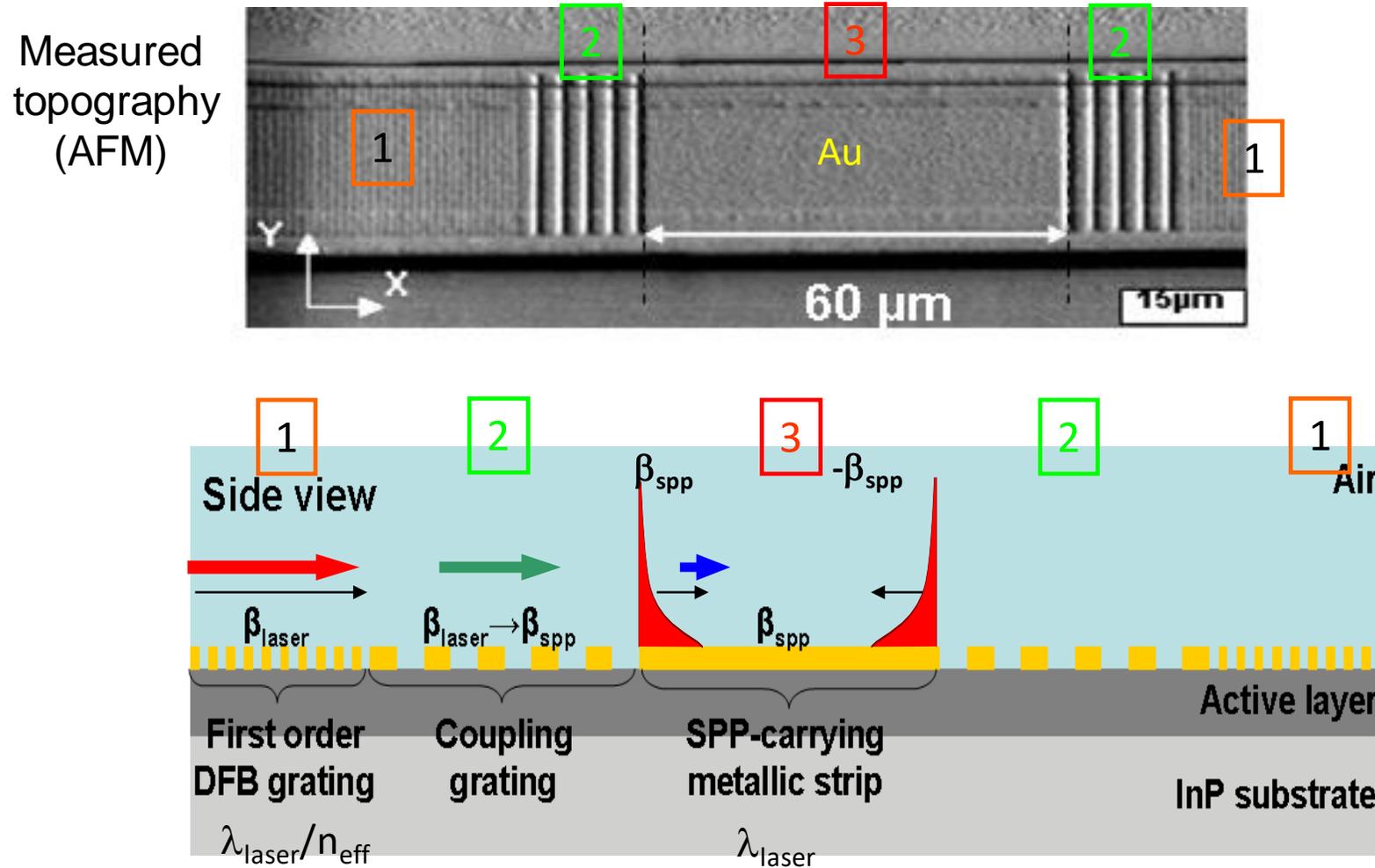
Hybrid SPPs

2. Grating coupler

3. Passive waveguide

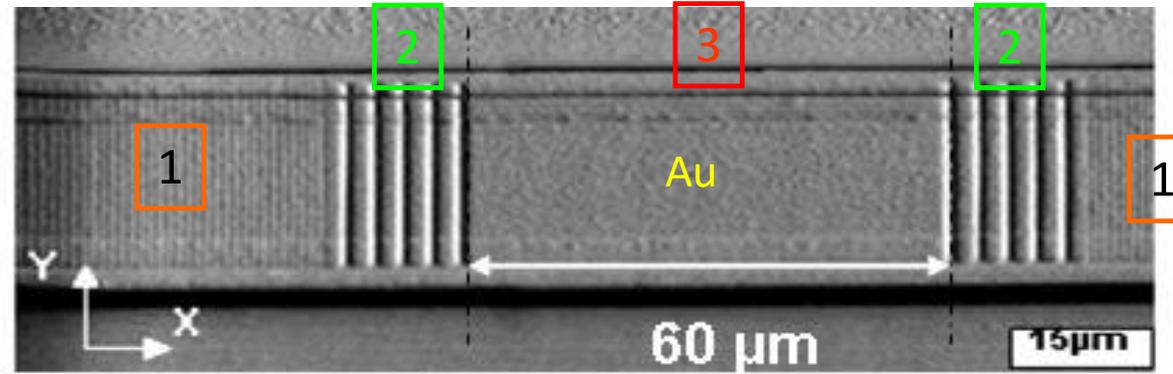


Building block of active plasmonics: Slit doublet experiment

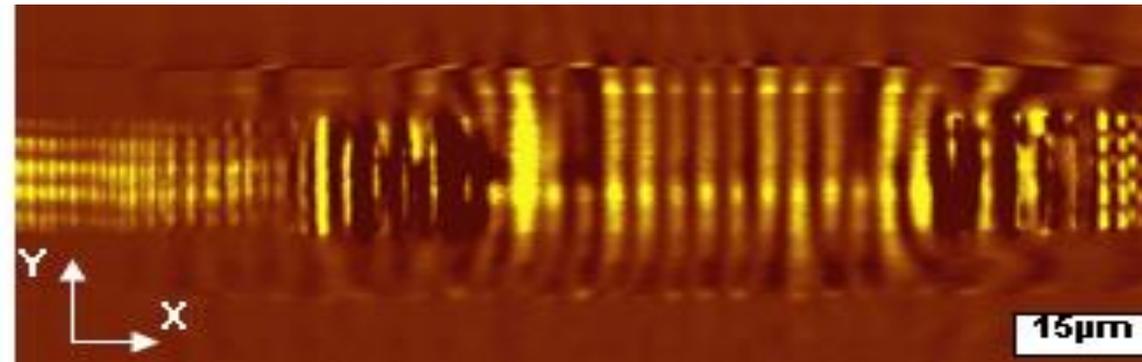


Building block of active plasmonics: Slit doublet experiment

Measured topography (AFM)



Measured near-field
 $\lambda \approx 7.5 \mu\text{m}$



Generation and launching of SPPs

SPP generation at 7.5 μm

Babuty et al., Phys. Rev. Lett. 104, 226806, (2010)

Spoo plasmons at 7.5 μm

Bousseksou et al., Opt. Expr. 20, 13738 (2012)

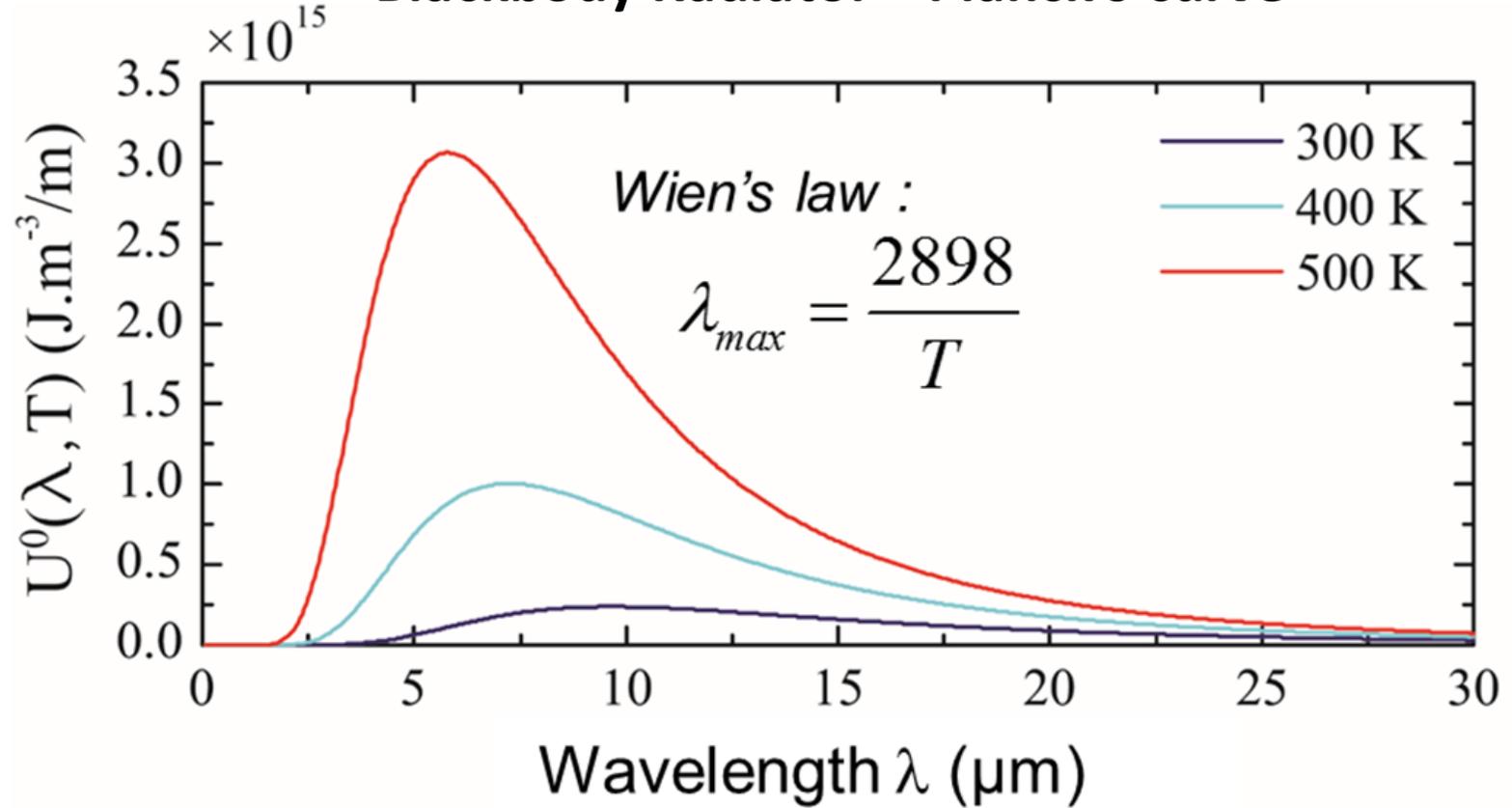
SPP generation at 1.3 μm

Costantino et al., Nano letters 12, 4693 (2012)

Greusard et al., Opti. Expr. 21, 10422 (2013)

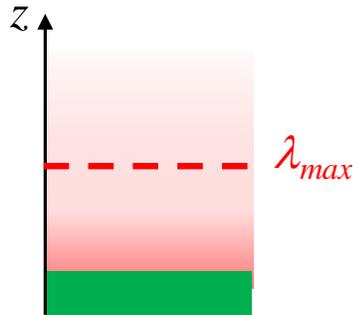
Thermal Radiation

Blackbody Radiator – Planck's curve



Far field:

$$z \gg \lambda_{max}$$

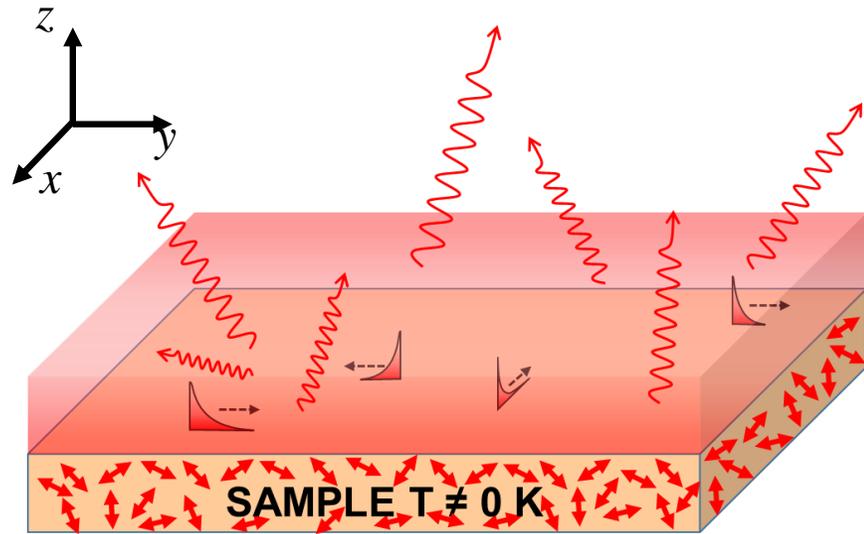


$$U(\omega, T) = \varepsilon^{\text{RM}}(\omega) U^{\text{BB}}(\omega, T)$$

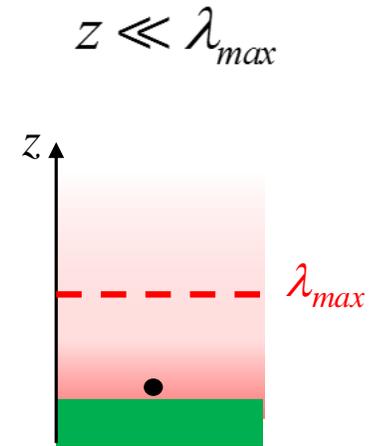
↑
Emissivity

$$0 \leq \varepsilon^{\text{RM}}(\omega) \leq 1$$

Real material radiation:



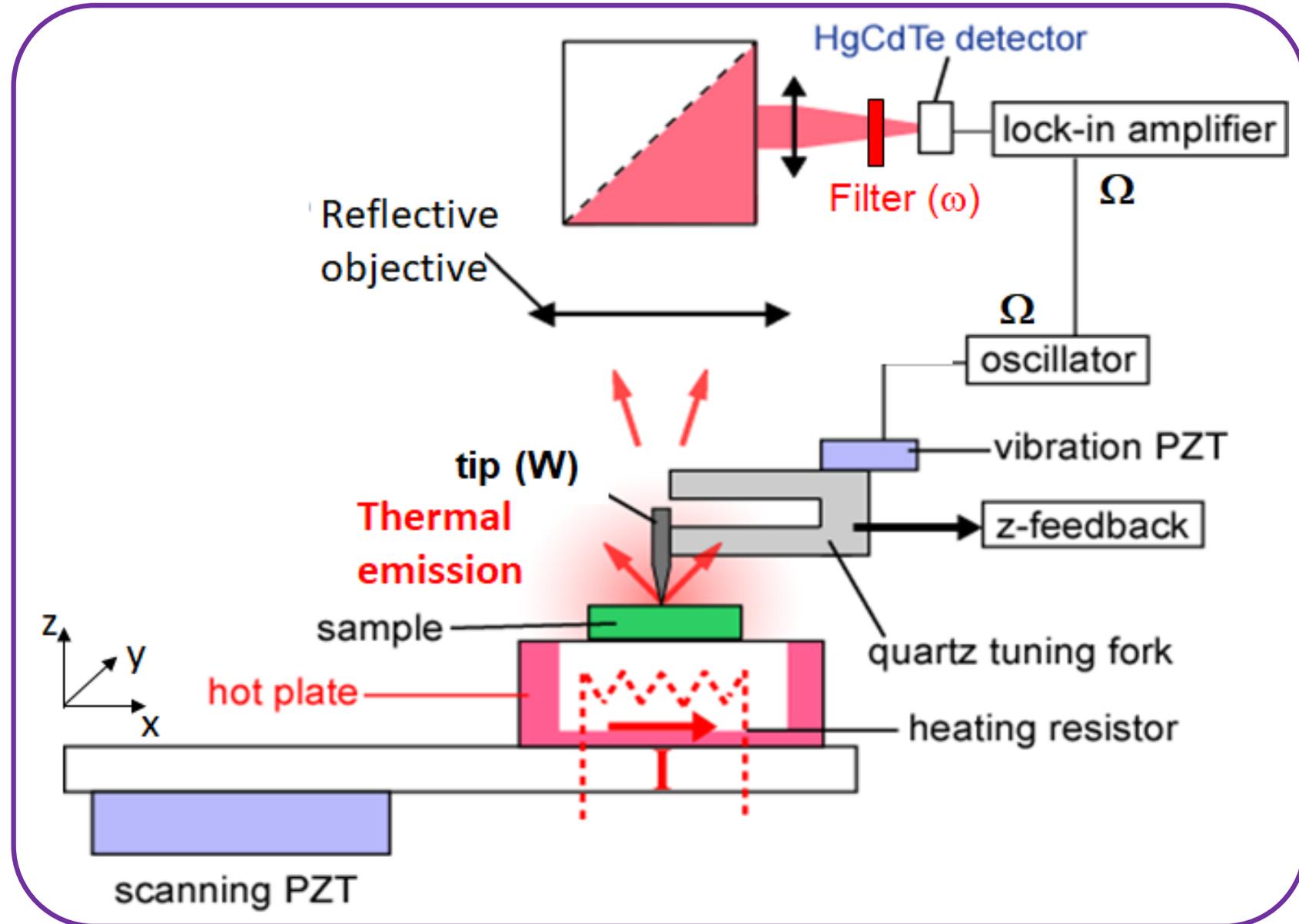
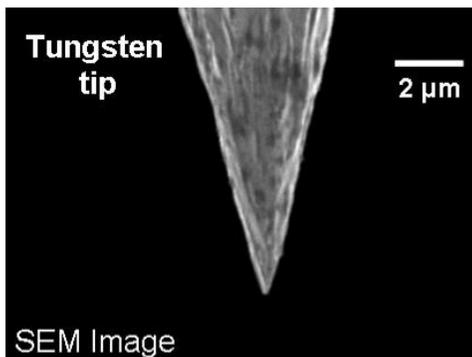
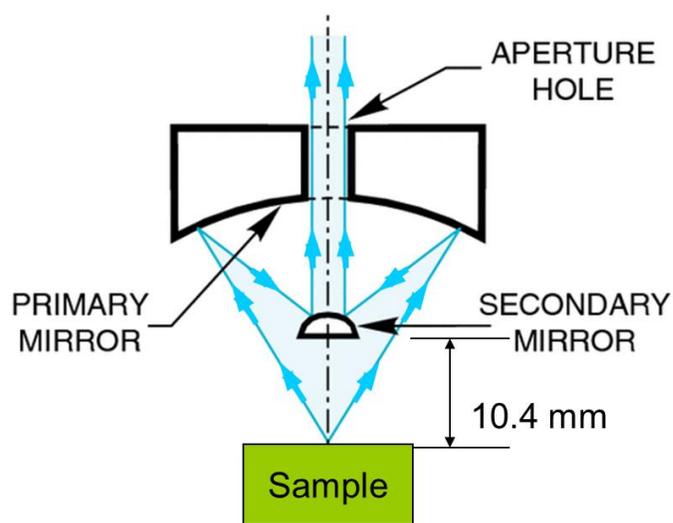
Near field:



Thermal Radiation STM (TRSTM):

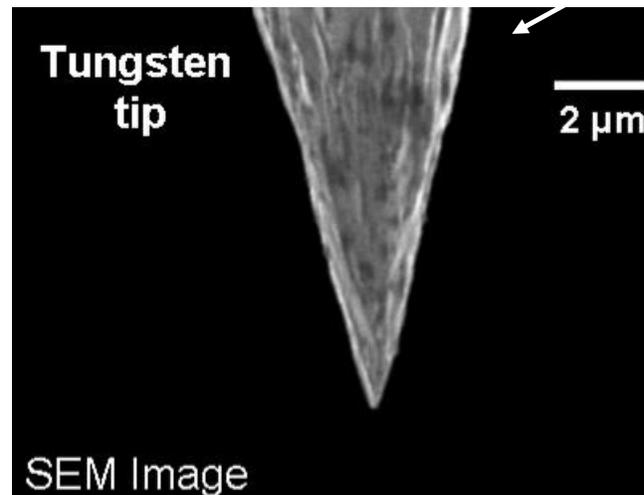
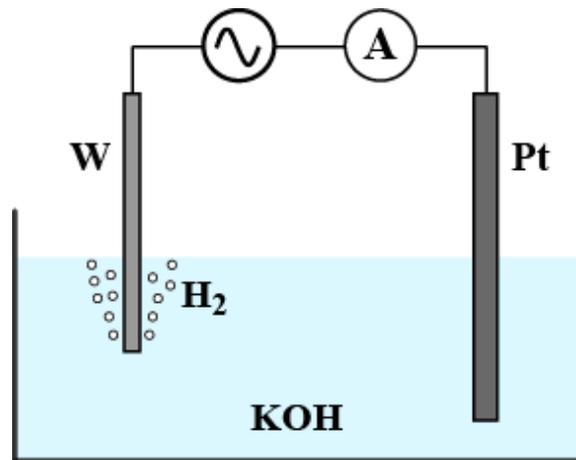
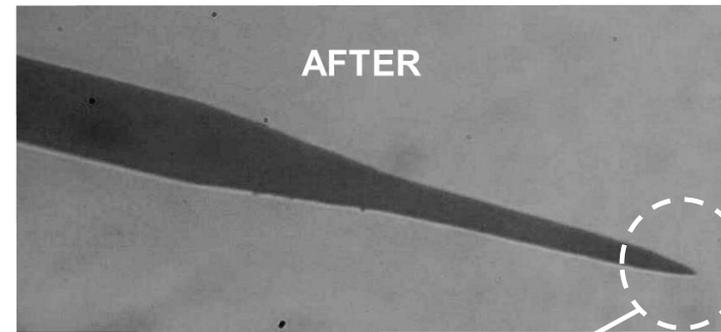
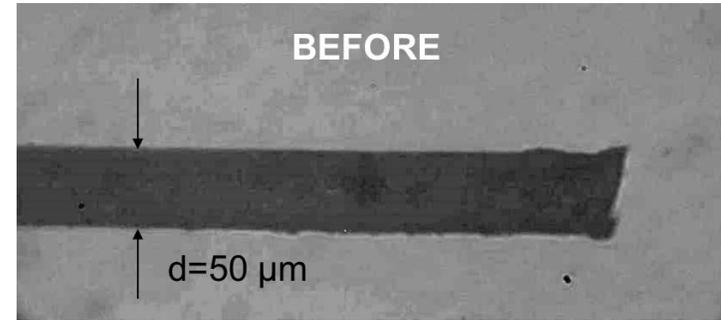
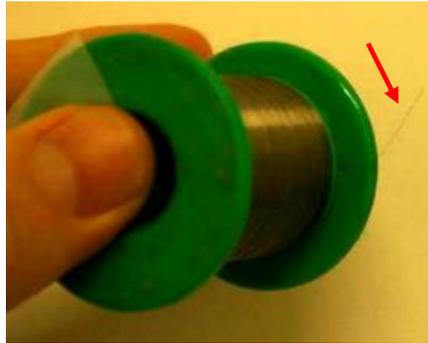
Reflective objective (Cassegrain)

COLLIMATED LIGHT

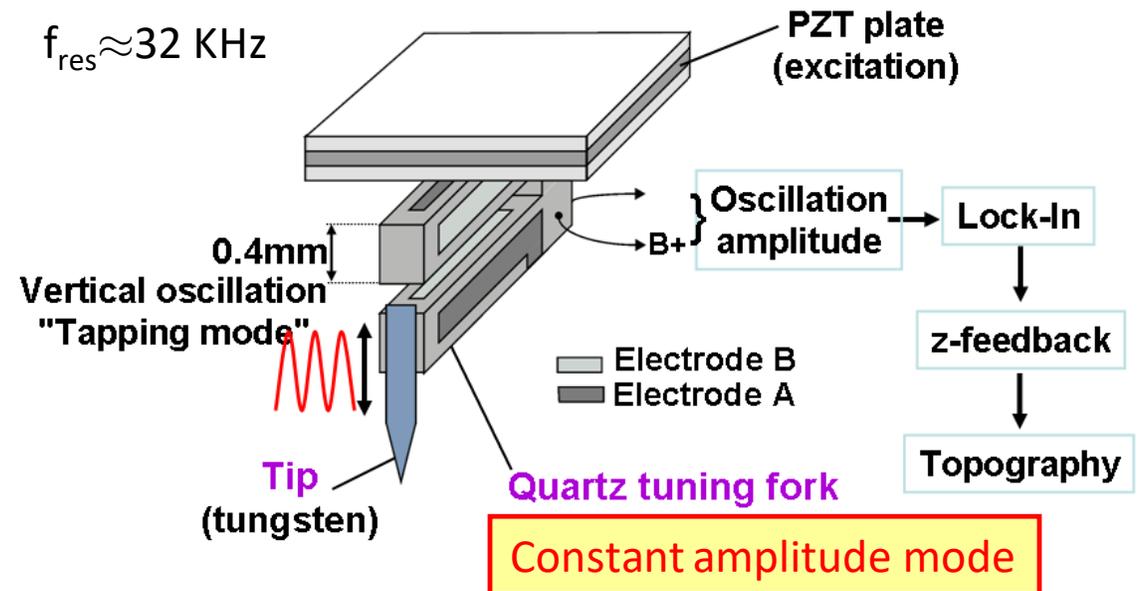
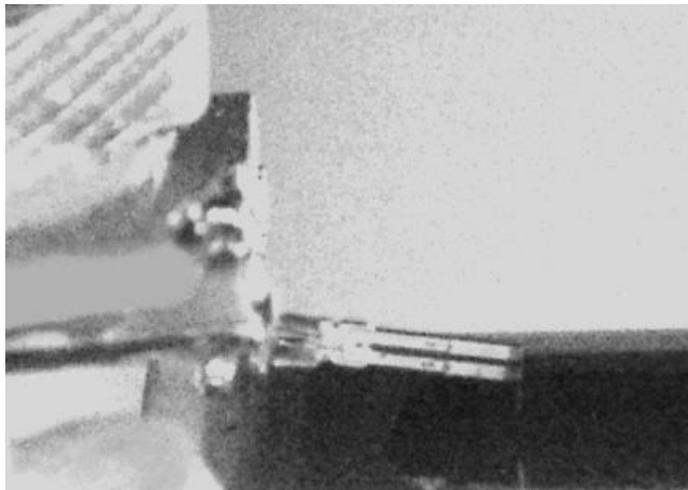
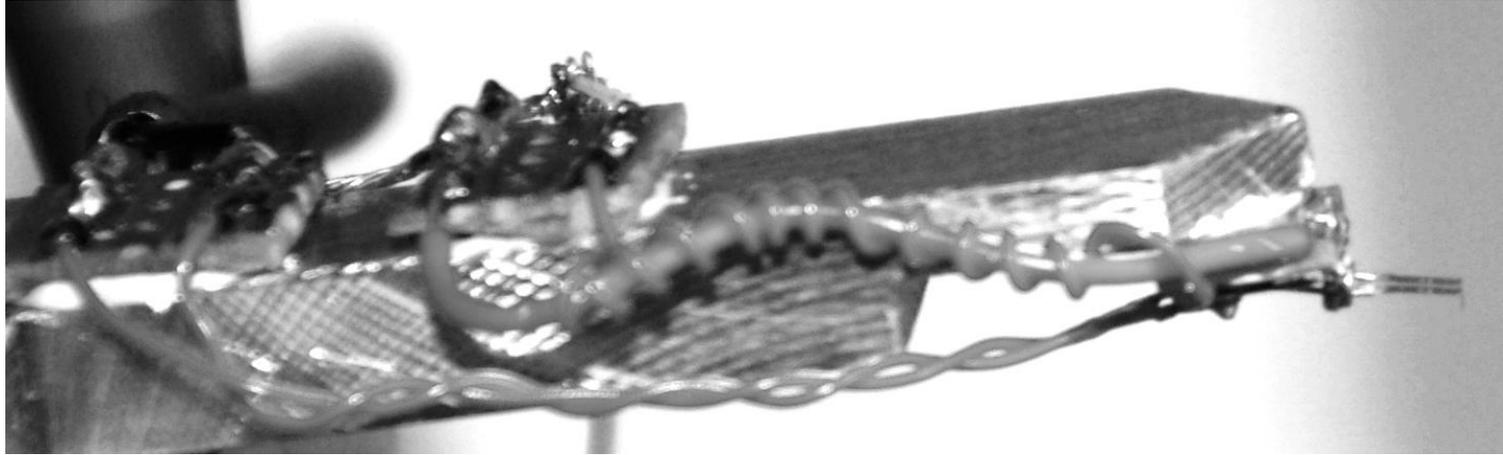


Tip preparation

Tungsten wire



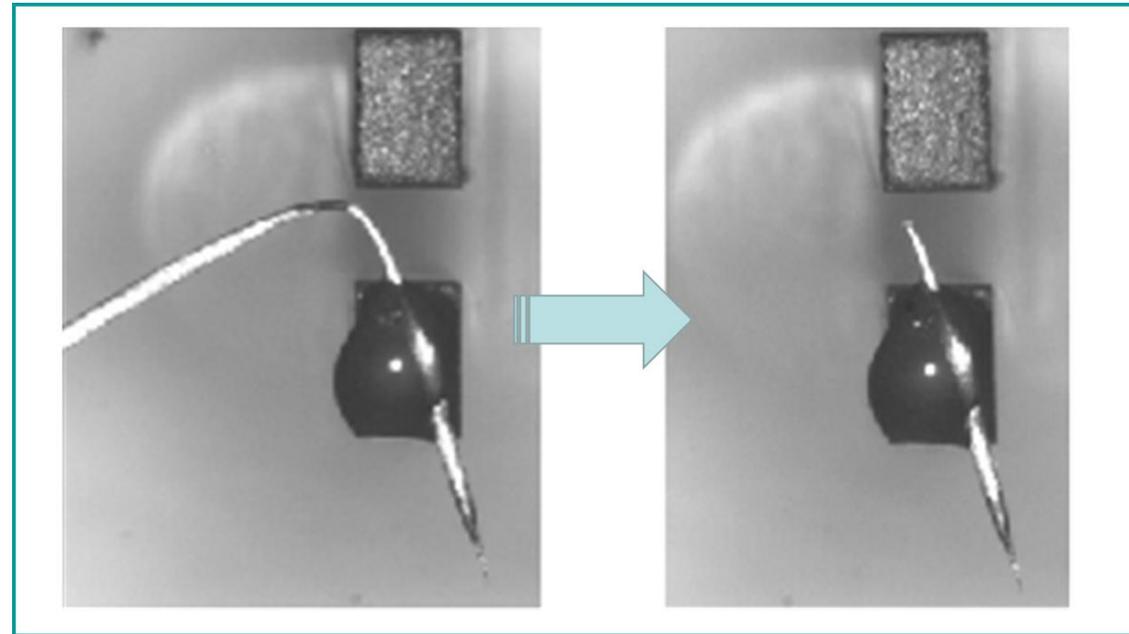
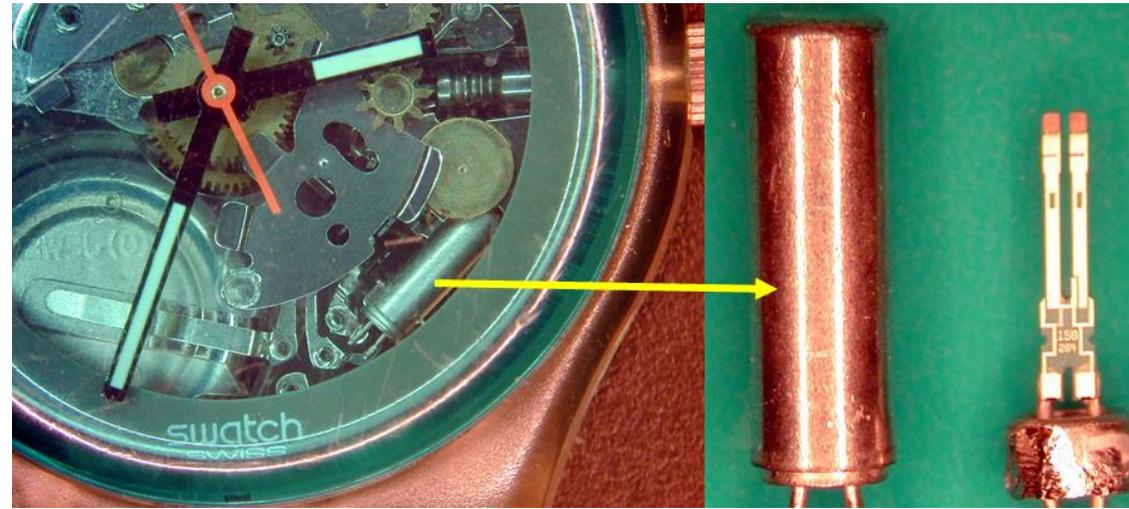
Mid IR s-NSOM – TRSTM: Tip mounting and oscillation detection



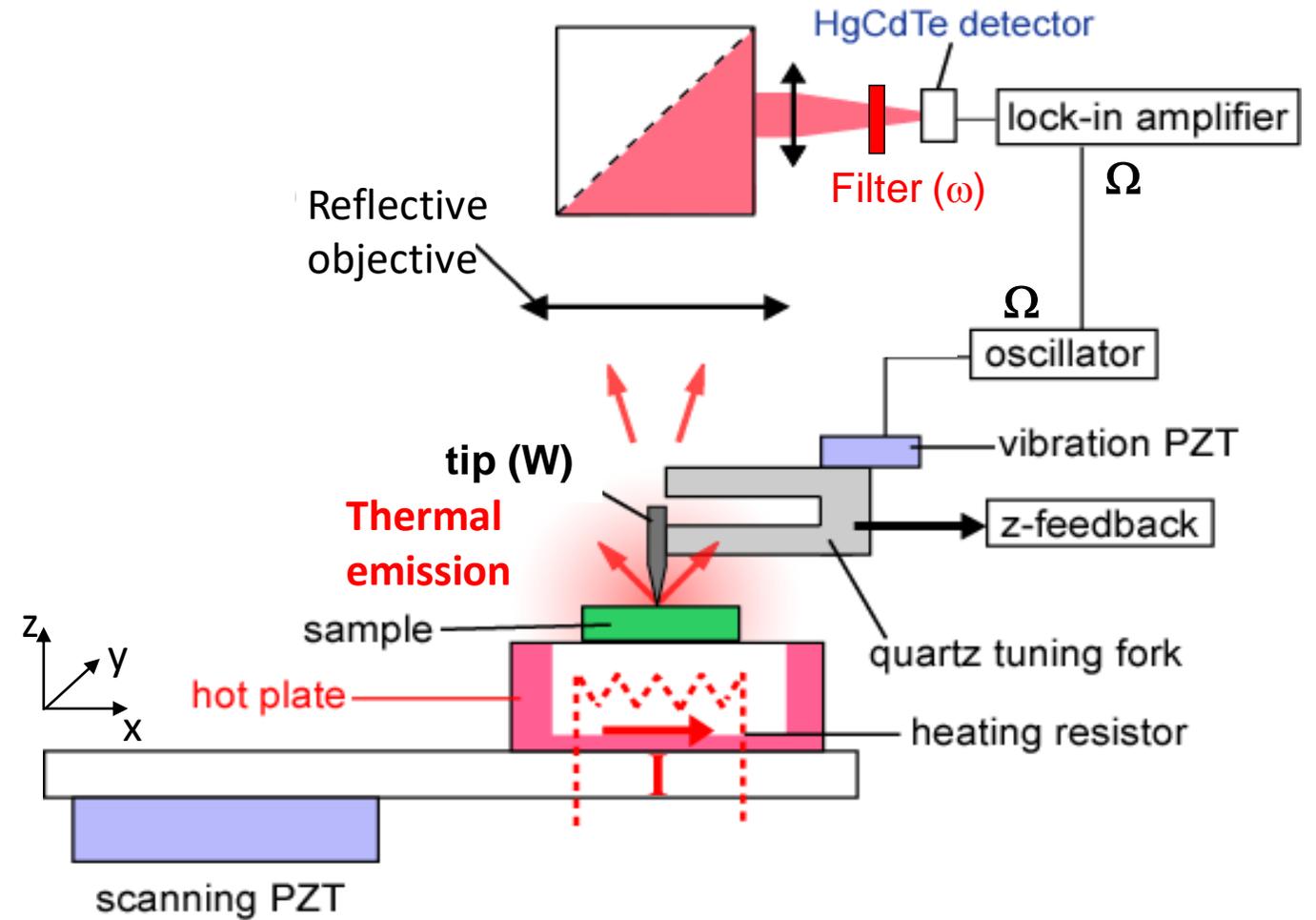
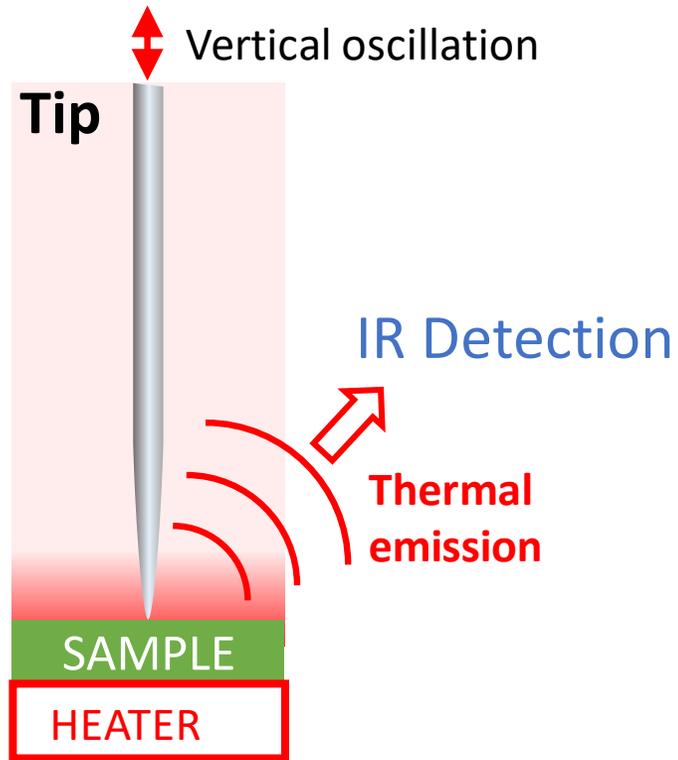
Tip gluing

Quartz tuning fork

$$\Omega_{\text{res}} = 32768 \text{ Hz}$$



Tip modulation



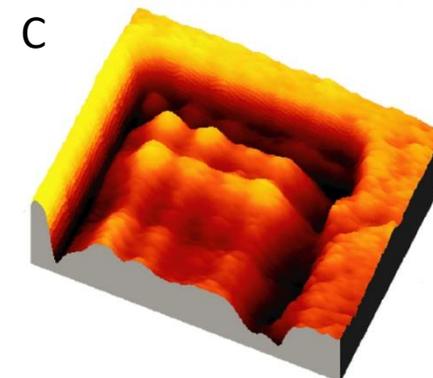
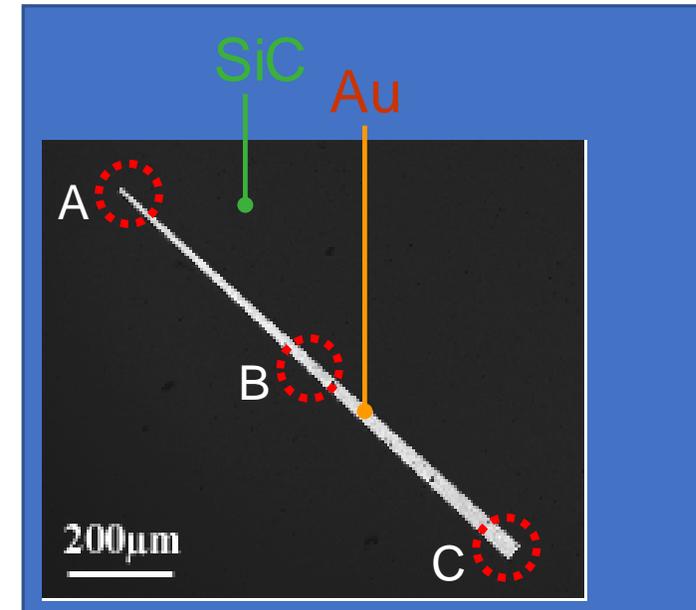
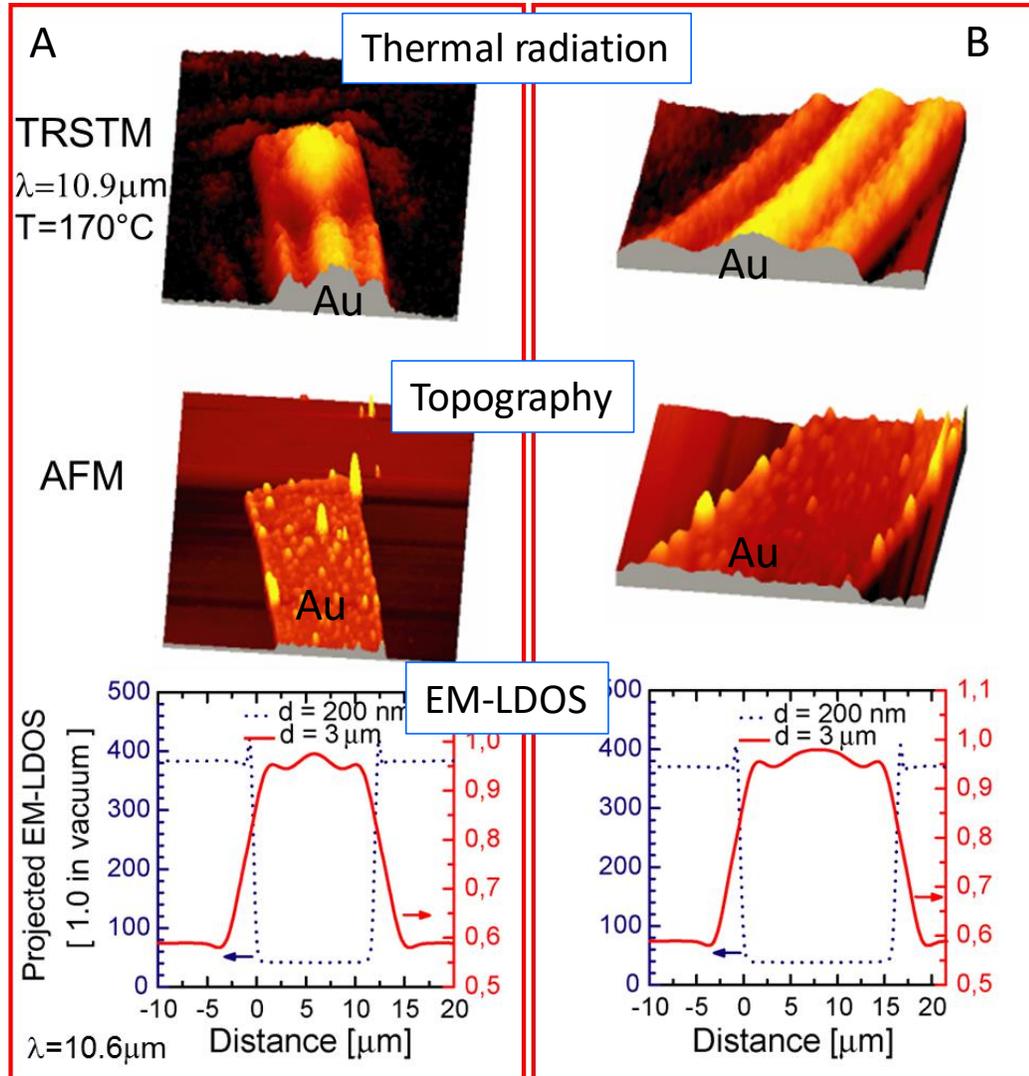
$$S(z_0 + a \cos \Omega t) \propto S_0 + A \frac{dS}{dz} \cos \Omega t + B \frac{d^2S}{dz^2} \cos 2\Omega t + \dots$$

Lock-In signal: $S_\Omega \propto \frac{dS}{dz}$ at z_0 \Rightarrow **Suppression of far-field background.**

Near-field imaging of EM-LDOS

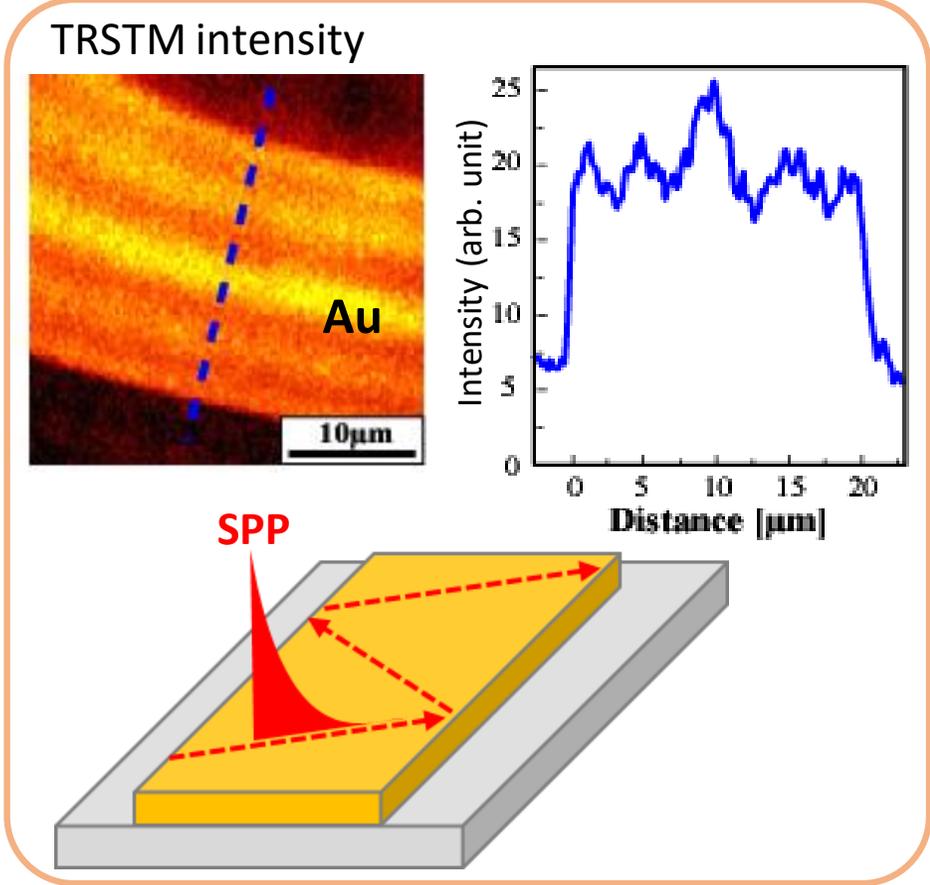
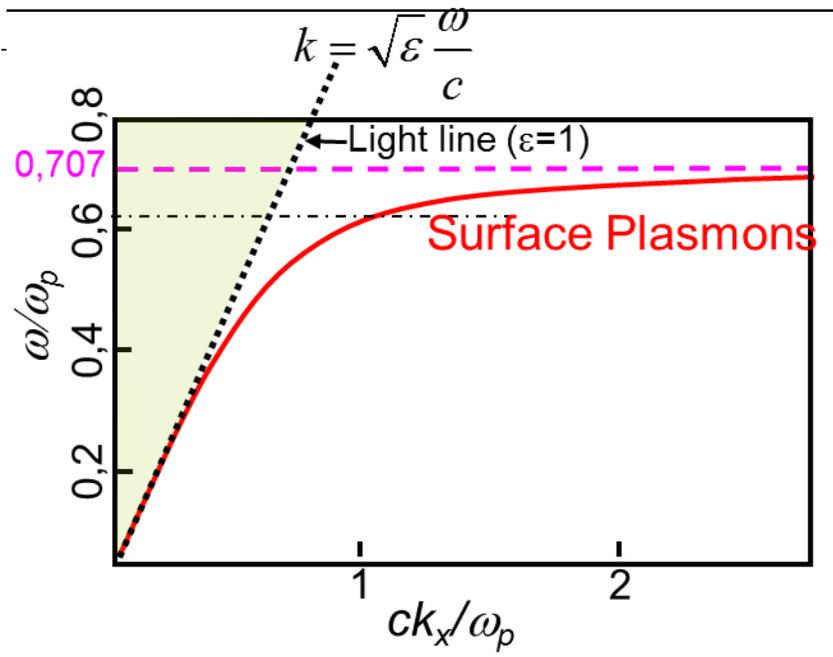
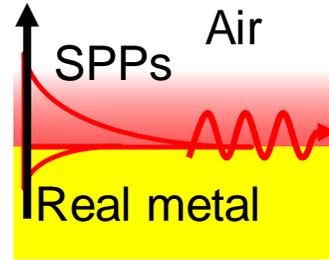


IMAGING ($\lambda \approx 11 \mu\text{m}$, $T \approx 440 \text{ K}$)

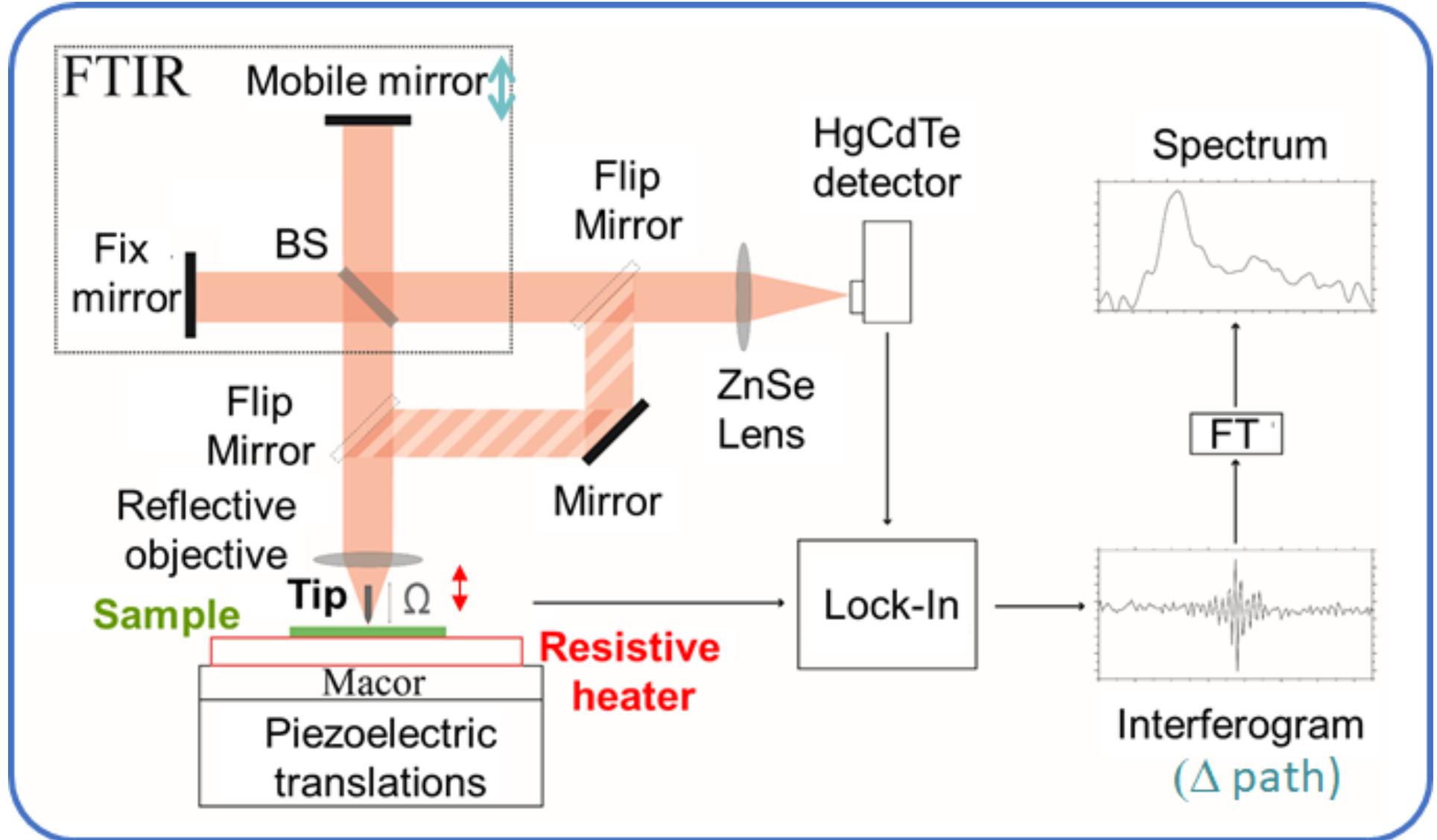
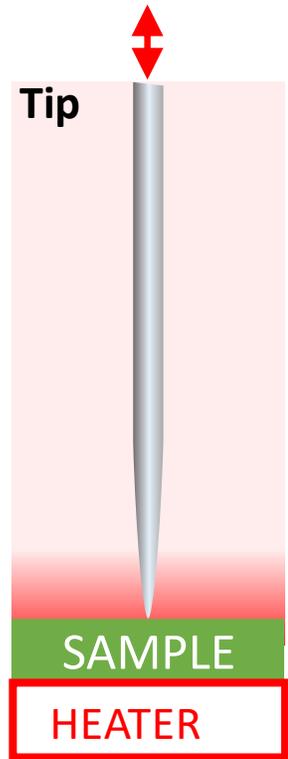


Cavity modes of SPPs on Au

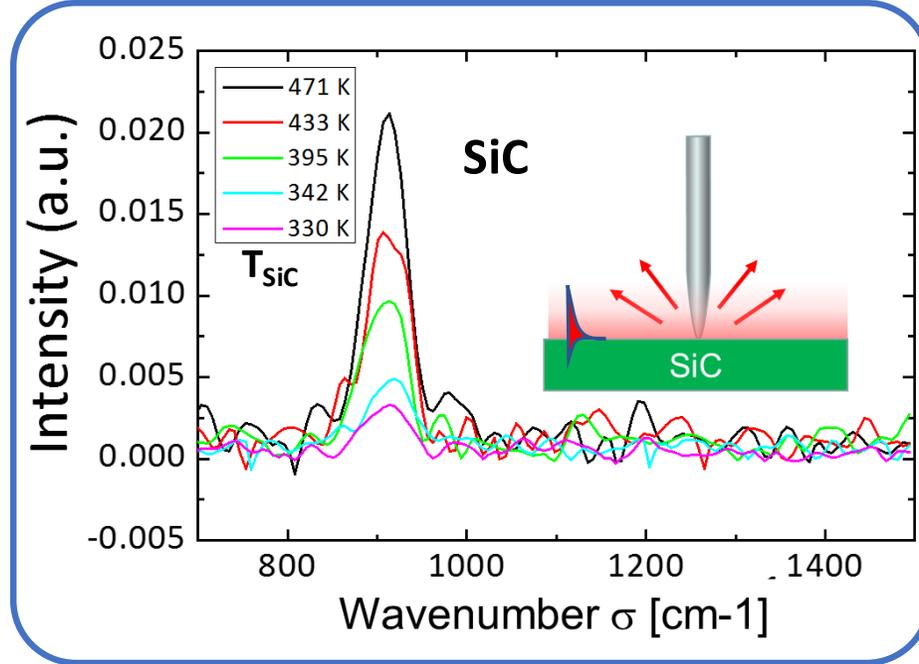
Surface waves



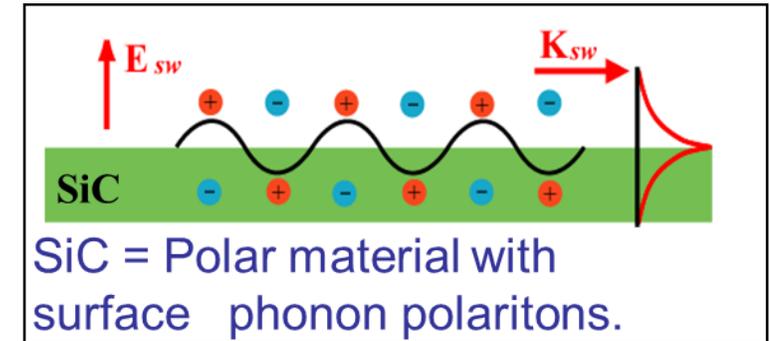
TRSTM coupled with FTIR spectrometer



TRSTM spectra on SiC:



Non-planckian behavior

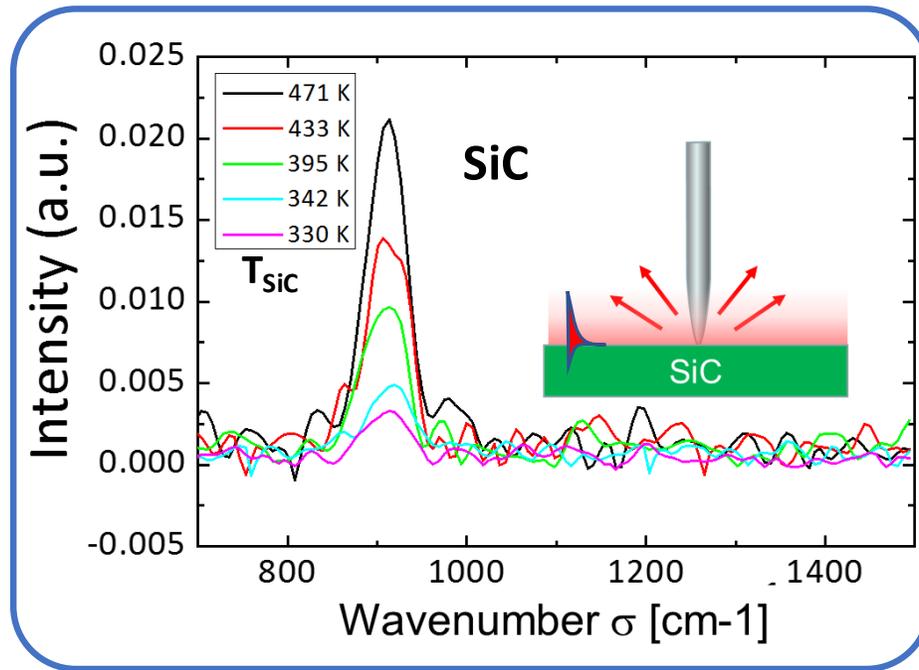


Prediction: Shchegrov *et al.*, PRL 85, 1548 (2000).

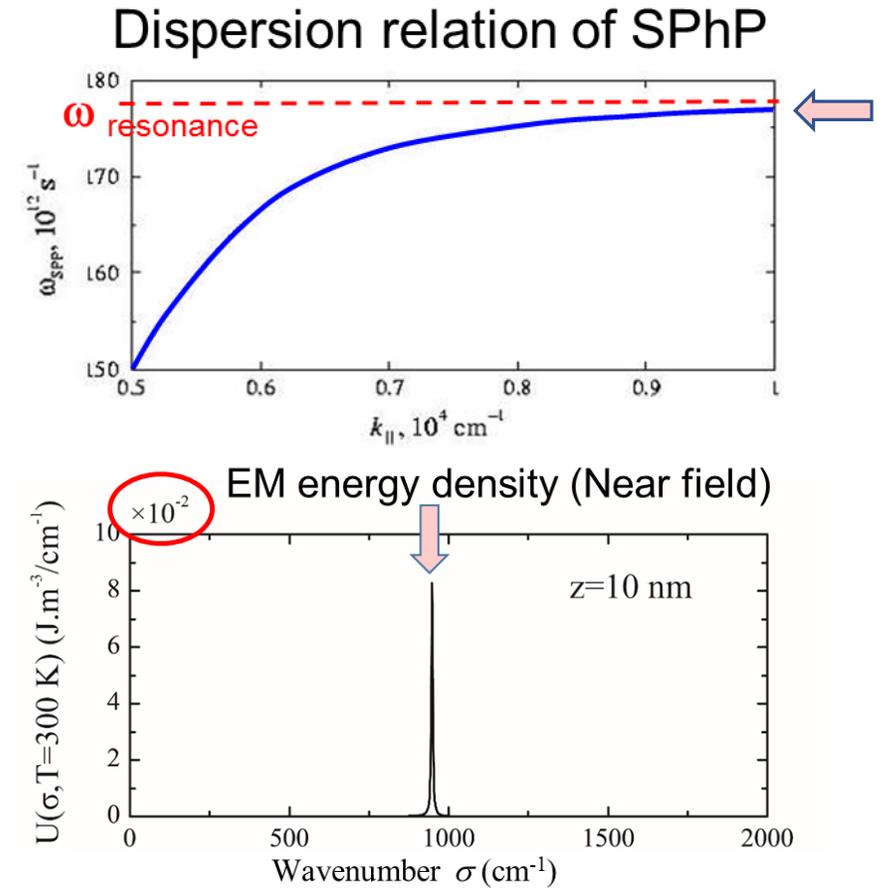
Near-field thermal radiation from surface **phonon** polaritons (SPhP).

Babuty *et al.*, *Phys. Rev. Lett.* **110**, 146103 (2013).
Joulain *et al.*, *JQSRT* **136**, 1-15 (2014).
Peragut *et al.*, *Appl. Phys. Lett.* **104**, 251118 (2014).

TRSTM spectra on SiC:



Non-planckian behavior

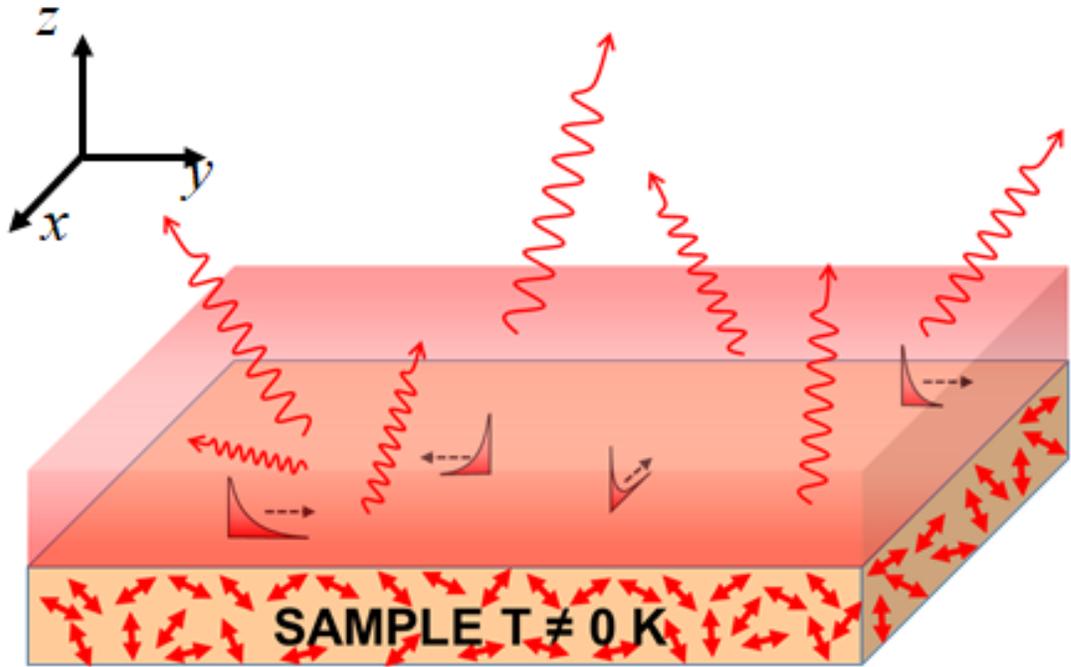


Prediction: Shchegrov *et al.*, PRL 85, 1548 (2000).

Near-field thermal radiation from surface **phonon** polaritons (SPhP).

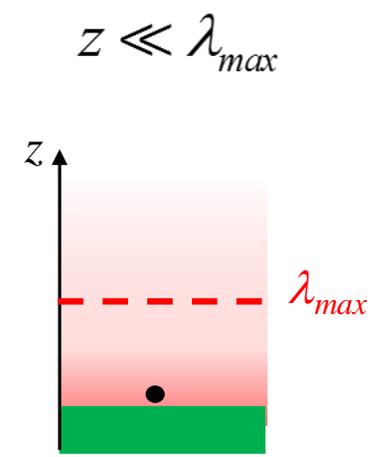
Babuty *et al.*, *Phys. Rev. Lett.* **110**, 146103 (2013).
 Joulain *et al.*, *JQSRT* **136**, 1-15 (2014).
 Peragut *et al.*, *Appl. Phys. Lett.* **104**, 251118 (2014).

Origin of the signal



The TRSTM probes the electromagnetic local density of states (EM-LDOS) in the near-field.

Near field:



- Predictions:
- EM-LDOS
 - Contribution of surface waves
 - **Coherence properties**

$$U(\mathbf{r}, \omega, T) = \underline{\underline{\rho(\mathbf{r}, \omega)}} \theta(\omega, T)$$

Local density of state (EM-LDOS)

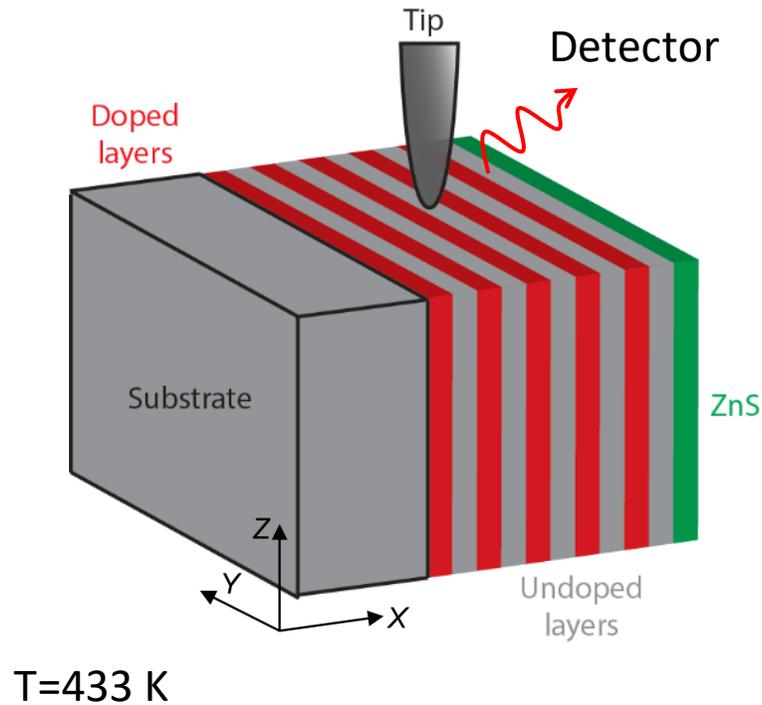
$$\theta(\omega, T) = \hbar\omega \frac{1}{\exp(\hbar\omega/kT) - 1}$$

Shchegrov, Joulain, Carminati, Greffet, *Phys. Rev. Lett.* **85**, 1548 (2000)

Joulain et al., *Phys.Rev.B* **68**, 245405 (2003).

Carminati, Cazé, Cao, Peragut, Krachmalnicoff, Pierrat, De Wilde, *Surf. Sci. Rep.* **70**, 1 (2015)

TRSTM study of doped/undoped semiconductor multilayer:



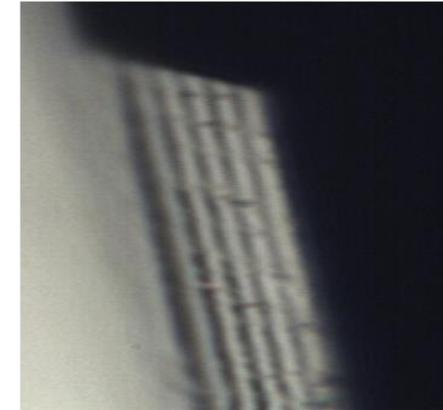
5 pairs InAs/InAs(Si)

Undoped InAs layers: $\sim 10^{16} \text{ cm}^{-3}$, thickness: 290 nm

Doped InAs layers: $5 \cdot 10^{19} \text{ cm}^{-3}$, thickness: 370 nm

ZnS layer, thickness: 2 μm

Cleaved edge



Collaboration :

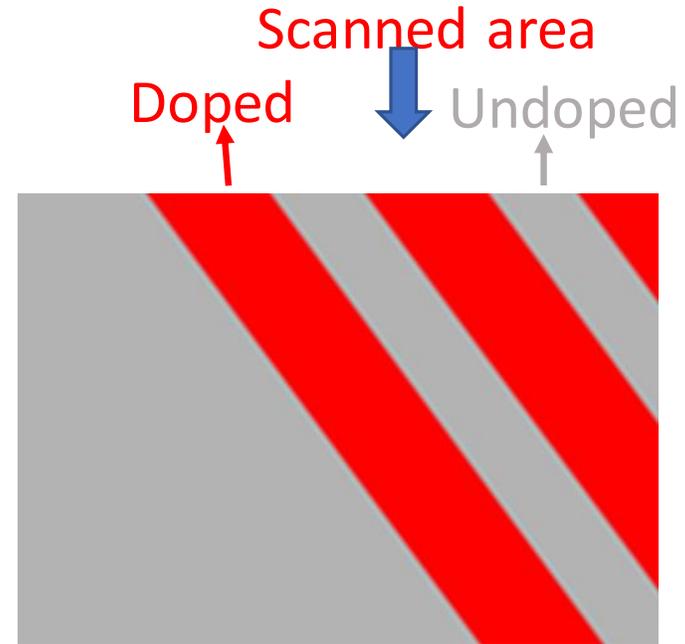
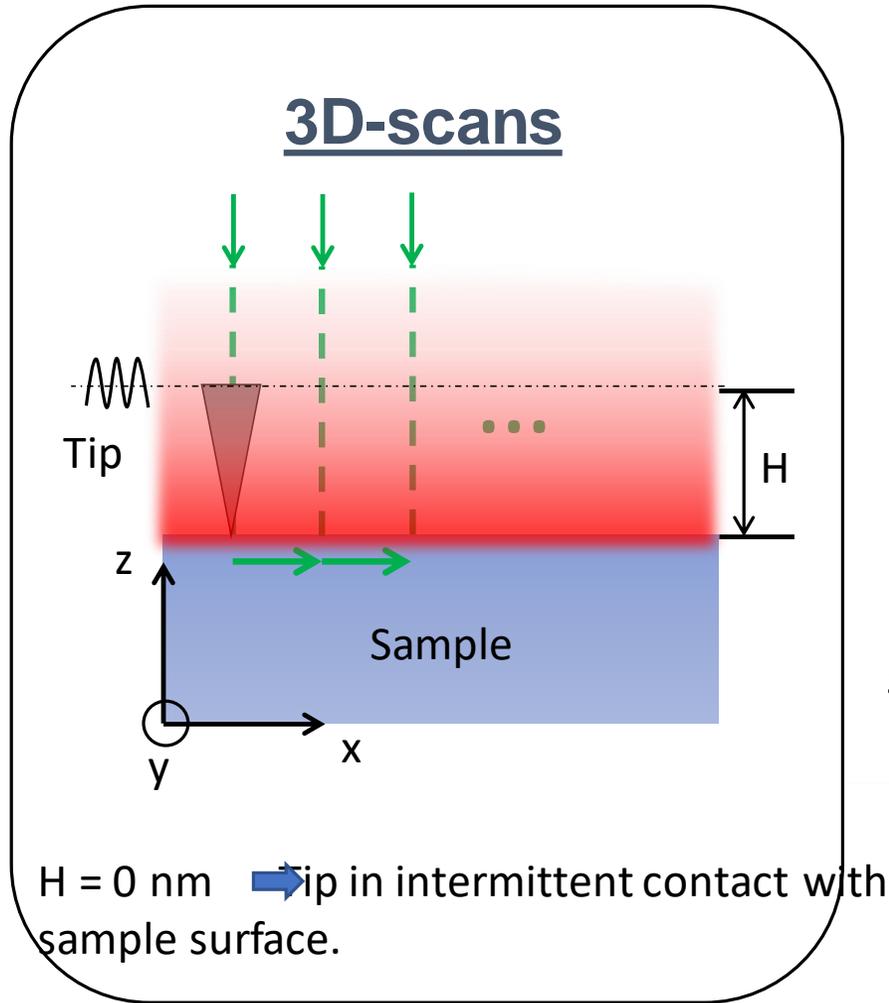


T. Taliercio
L. Cerutti
A. Baranov



J.-J. Greffet
J.-P. Hugonin

TRSTM imaging from far-field to near-field

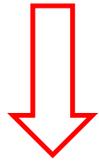


Scan size = $2 \mu\text{m} \times 0.7 \mu\text{m} \times 200 \text{ nm}$

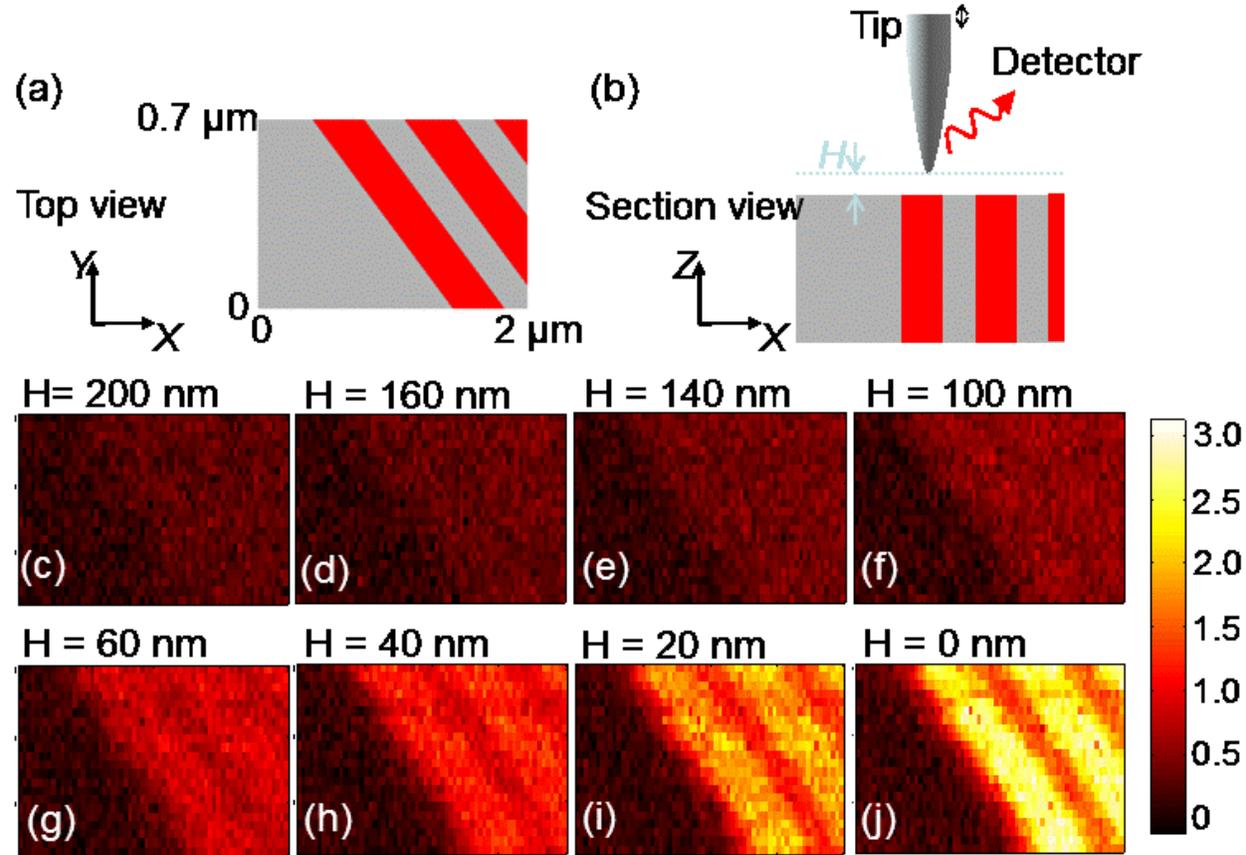
From $H=200 \text{ nm}$ to 0

TRSTM imaging from far-field to near-field

$$\left(\frac{2\pi}{\lambda}\right)^2 = |\mathbf{K}|^2 + \gamma^2$$



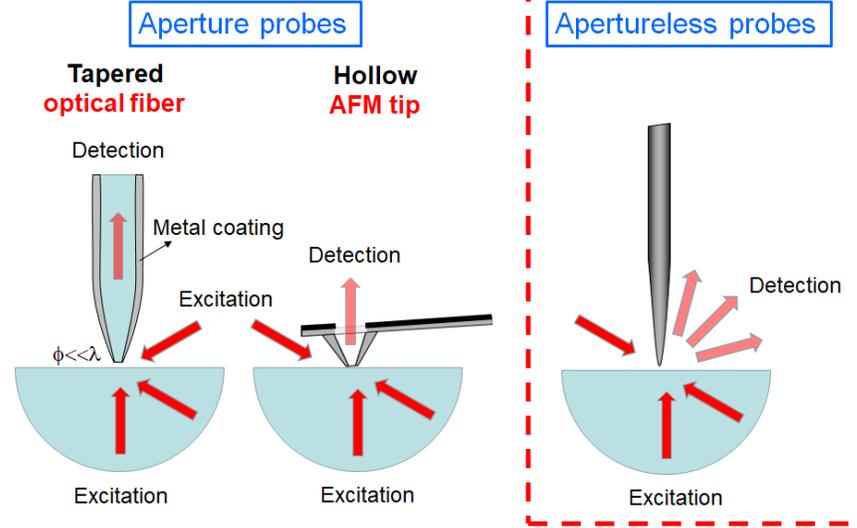
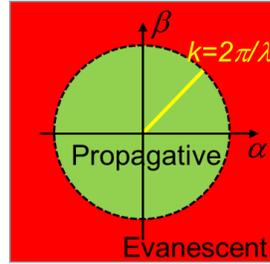
$$z^* = \frac{\Delta r_{\parallel}}{2\pi}$$



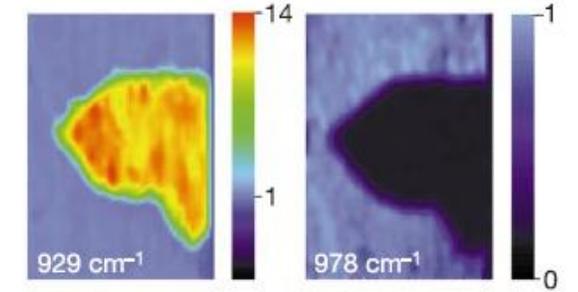
- Distance = Low-pass filtering.
- Homogeneous to heterogeneous EM-LDOS transition.

Conclusion

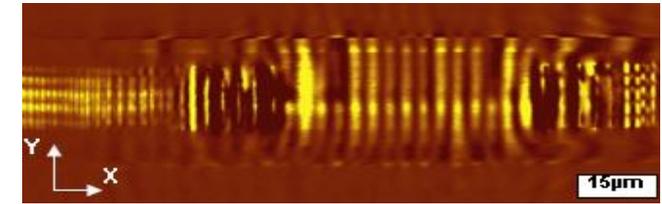
- NSOM information on evanescent fields



Materials at sub- λ scale



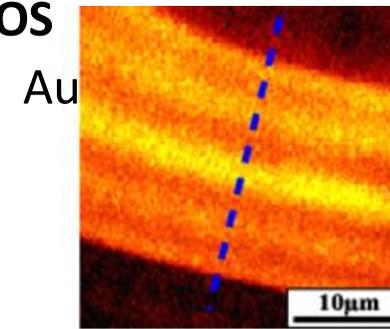
Surface wave studies



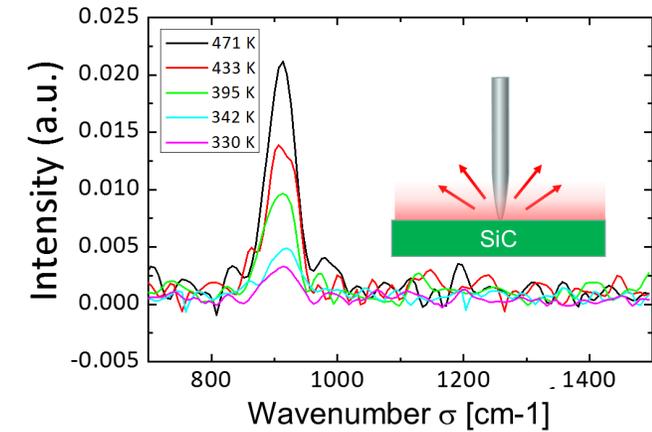
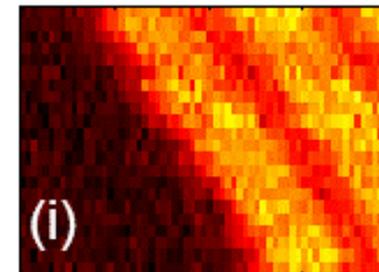
- TRSTM: Thermal radiation in the near-field probes the EM-LDOS

+ Coherence effects

- Super-resolution is achieved both for imaging and spectroscopy



H = 20 nm



NSOM in FRANCE

Liste non-exhaustive:

Lille: IEMN

Paris: ESPCI (Institut Langevin, LPEM),
Sorbonne Université (INSP)

Paris-Saclay: C2N, ONERA, CEA,
SOLEIL, Inst. Chimie Physique (UPS)

Versailles: GEMaC (UVSQ)

Troyes: L2n laboratory (UTT)

Dijon: Laboratoire ICB, Université de Bourgogne

Besançon: FEMTO St

Nantes: IMN

Lyon: STMS/INL (Ecole Centrale Lyon), ILM

Grenoble: Inst. Néel

Marseille: Inst. Fresnel

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