Near-Field Scanning Optical Microscopy: a Brief Overview

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Thanks to my former & present collaborators in SPECTRO:

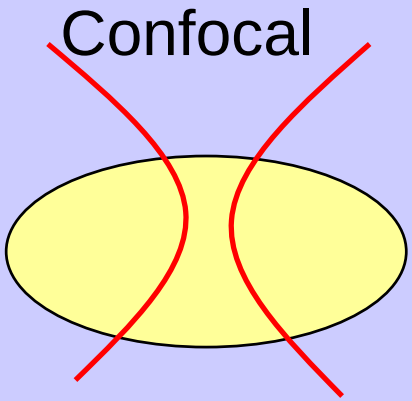
M. Brun, N. Chevalier, A. Drezet, J. F. Motte, *M.J. Nasse, Y. Sonnefraud,* J.C. Woehl

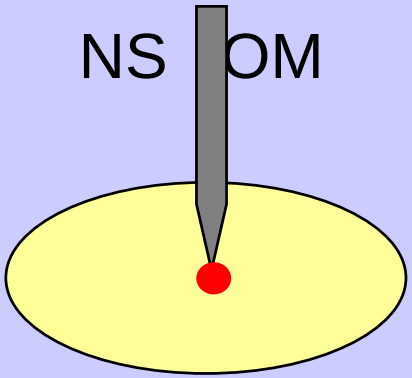
Italics stand for graduate students

Outline

1. Introduction to NSOM (Near-Field Scanning Optical Microscopy)
2. Local spectroscopy of semiconductor nanostructures
3. A short journey through biology
4. Search for the ultimate resolution in optics

Near-Field Scanning Optical Microscopy (NSOM) versus confocal microscopy





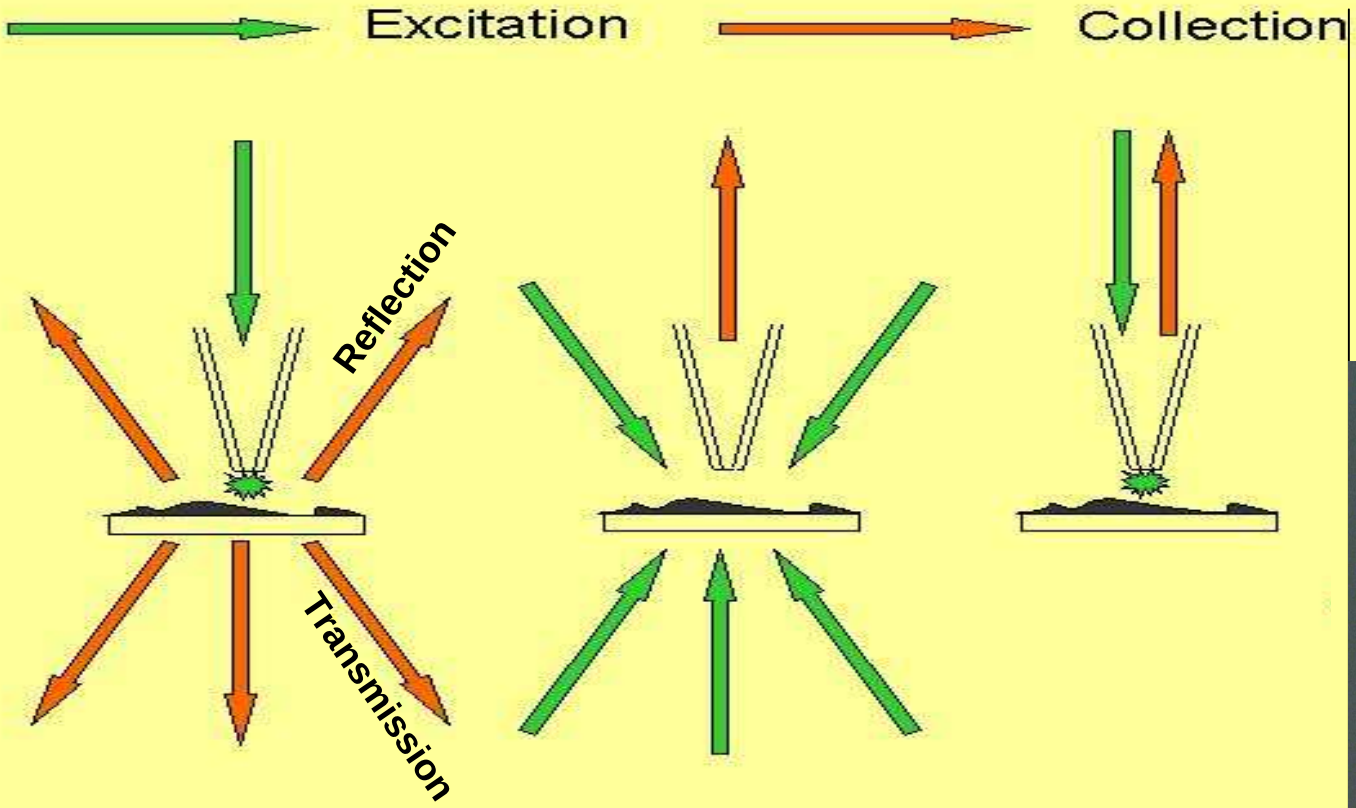
Lateral resolution:	diffraction-limited to 250 nm	30-50 nm
Excitation volume:	10^8 nm^3	10^5 nm^3
Background signal:	“large”	smaller

NSOM

- Optical resolution (10 nm ?)
- Correlation with topography
- Nano-manipulations
- Non-contact microscopy

- “Difficult” to operate (tip !)
- Small excitation intensity
- Slow method

Some configurations in aperture NSOM



Illumination mode: local excitation but far-field collection (used in my lab.)

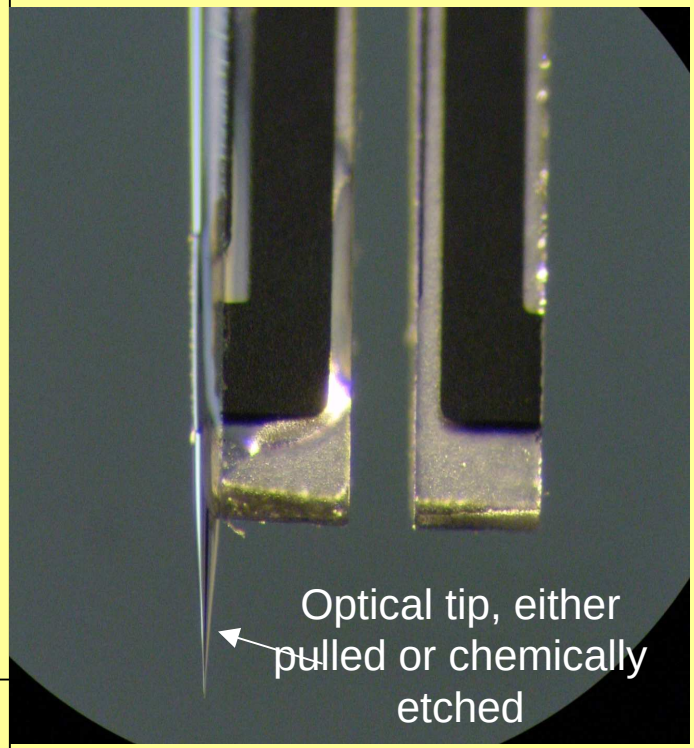
Collection mode

Hydrid I+C mode: the « best » resolution, but difficult to set up.

Note: To beat the diffraction limit, both the aperture size and the tip-surface distance must be << λ

Vertical positionning of the optical tip in the near-field is often achieved by a **TUNING FORK**

Karraï-Grober, 1995



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1. Introduction to NSOM
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Why NSOM on semiconductor nanostructures ? The example of a single quantum dot

1) High spatial resolution. See below the demonstration of the high resolution of NSOM with self-organized CdTe quantum dots → detailed spectroscopic study possible

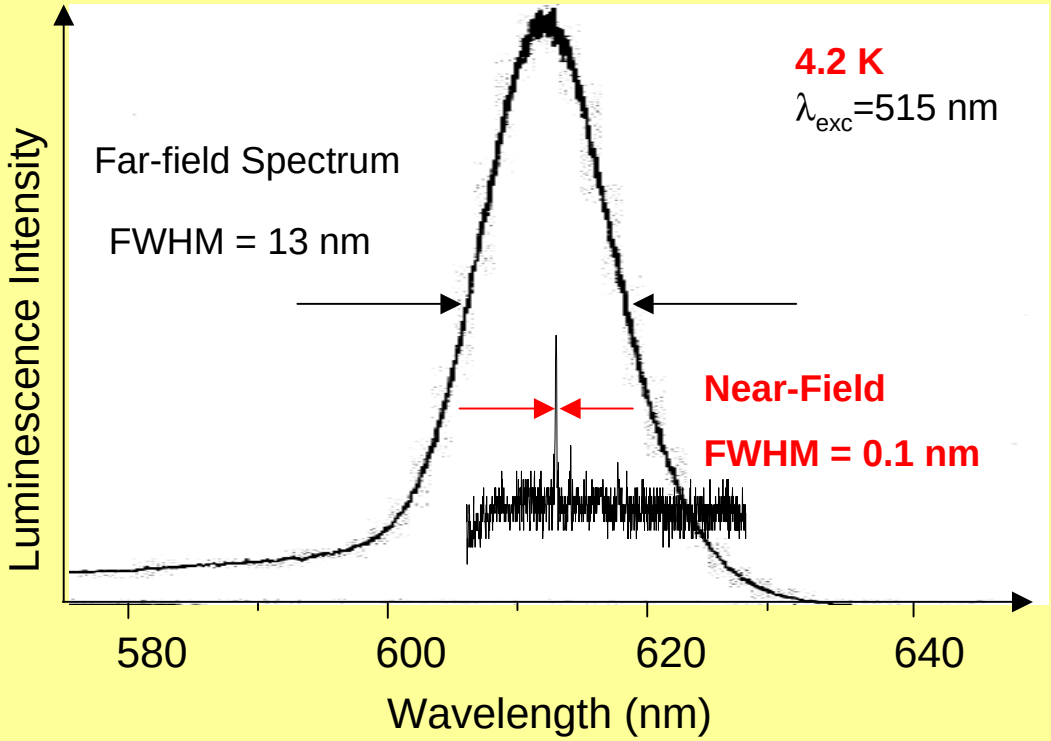
2) Imaging of the spectroscopic properties.

3) Free selection of nano-objects with a super-resolution.

4) Nanomanipulation (mechanical, electrical ..) with the optical tip.

5) Possible correlation with topography (if applicable).

…

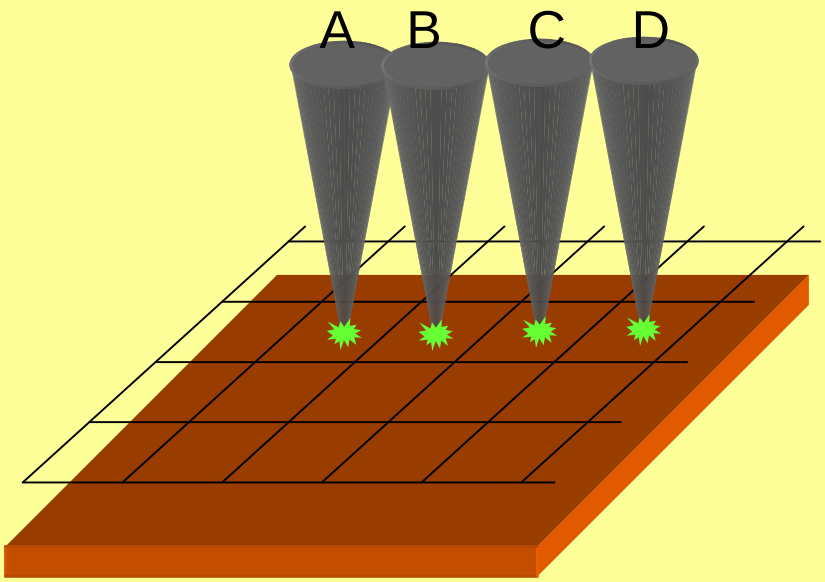


M. Brun *et al.*, *J. Microscopy* **202**, 202 (2001)

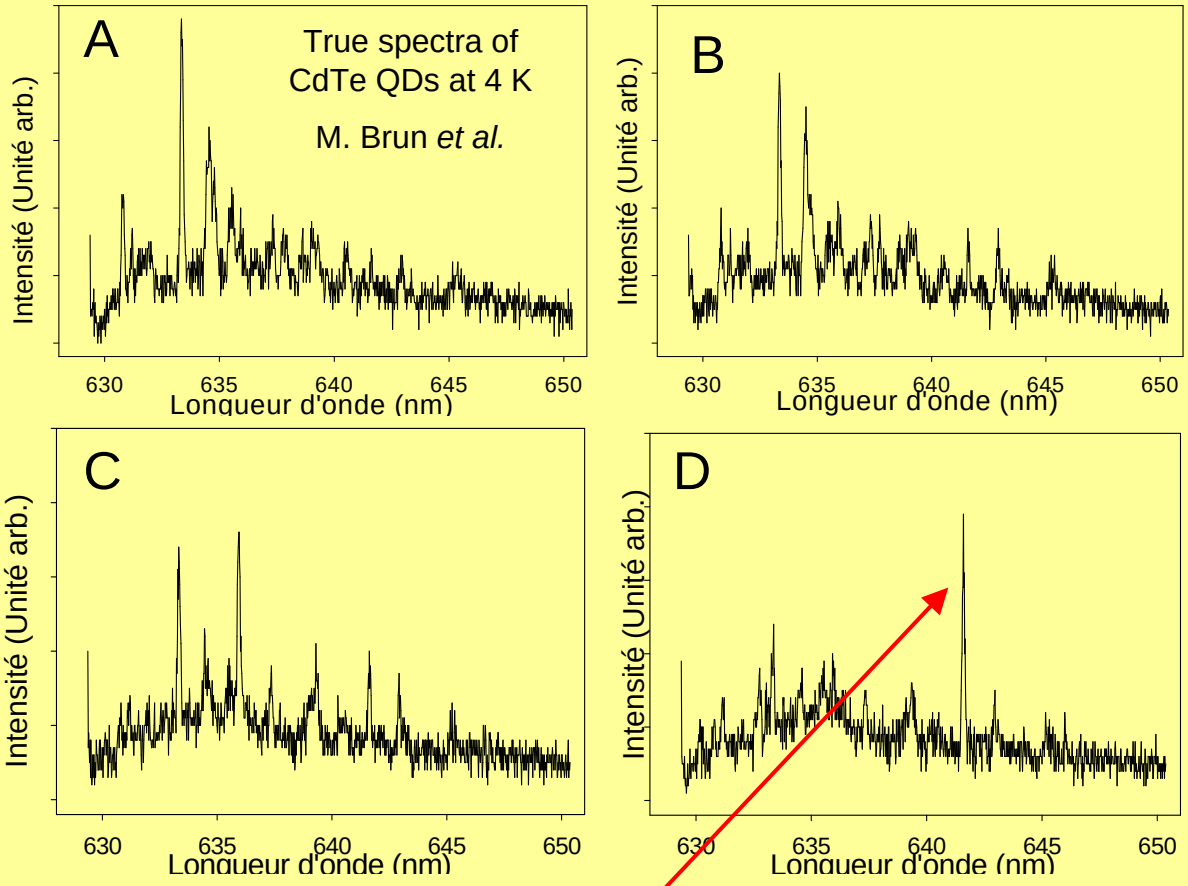
Why NSOM on semiconductor nanostructures ? The example of a single quantum dot

2) Imaging of the spectroscopic properties.

Imaging of the spectroscopic properties: principle



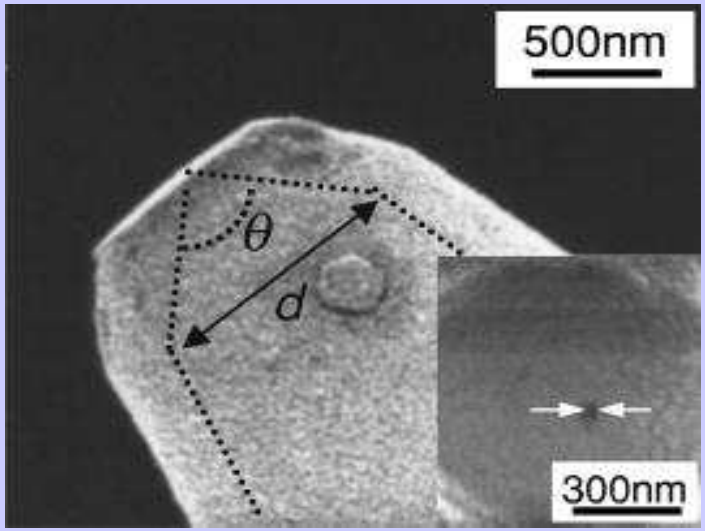
active medium such
as quantum dot sample



Isolate an interesting feature and map its intensity versus tip position

Near-field optical mapping of exciton wave functions in a GaAs QD (I), Matsuda et al., PRL 91, 177401 (2003)

The NSOM tip

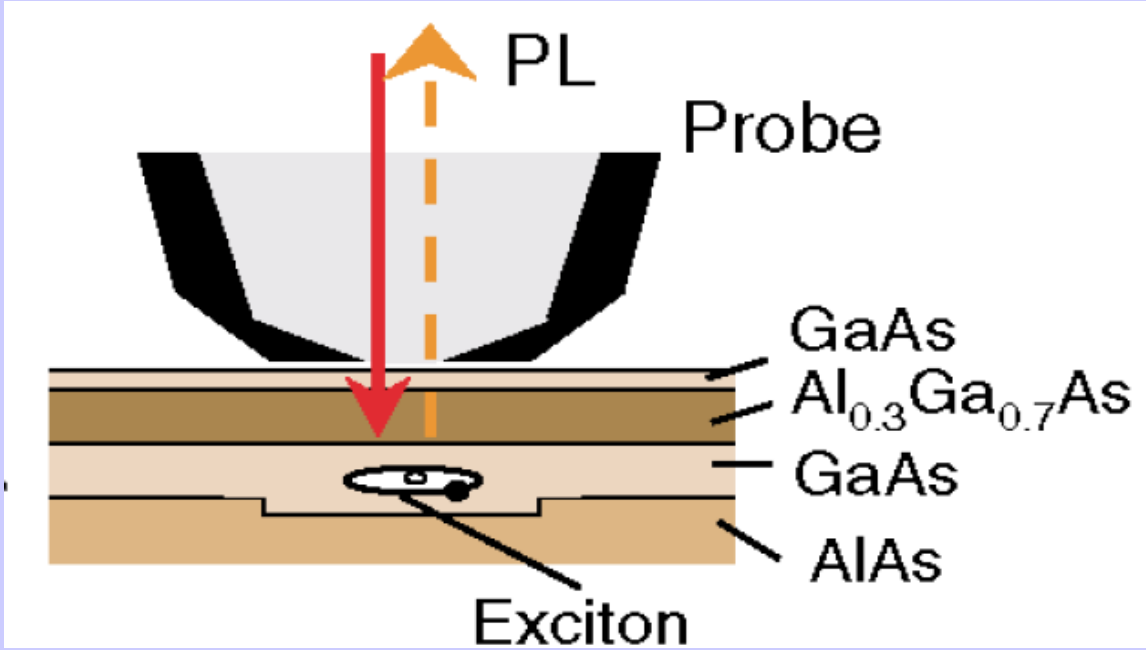


Special optical probe obtained by chemical etching with shape control of the (double) taper, a clear aperture of 30 nm in the Au coating.

Used in the excitation-collection mode.

Probably the best aperture tip so far !

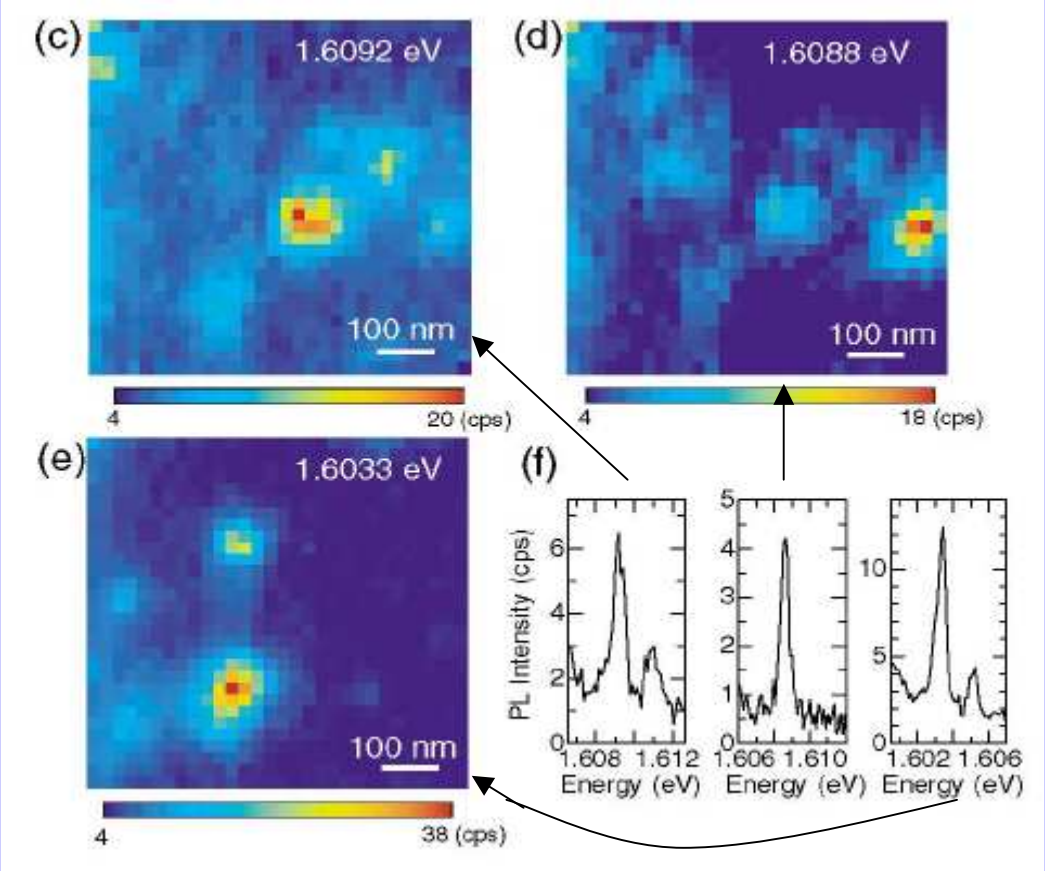
Studied structure= “natural” GaAs quantum dots of size 100 nm



Note: a photon is absorbed → an electron is excited in the conduction band + a hole is left behind

The electron-hole pair form an exciton X that recombines after a few 100 ps (up to 1 ns) → photoluminescence (PL) signature

Near-field optical mapping of exciton wave functions in a GaAs QD (II), Matsuda et al., PRL 91, 177401 (2003)



Scanning area = 1 µm x 1 µm

T= 9 K

Low excitation power (only X excitons form).

Different dots give slightly different spectra and can be located from their PL images.

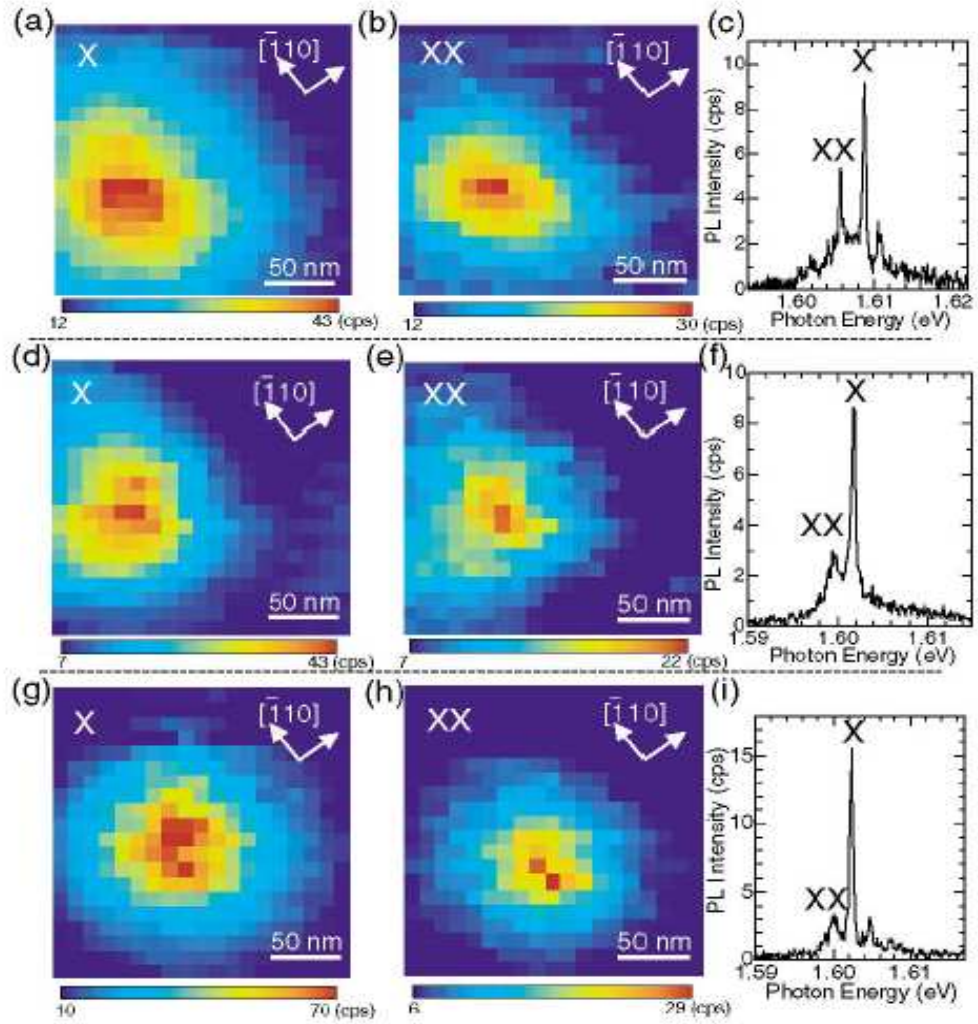


FIG. 3 (color). (a)–(i) Series of high-resolution PL images of exciton state [(a), (d), and (g)], biexciton state [(b), (e), and (h)], and corresponding PL spectra [(c), (f), and (i)] for three different QDs. Scanning area is 210×210 nm^2. Crystal axes along [110] and [$\bar{1}$10] directions are indicated. PL image sizes of biexciton are always smaller than those of exciton.

Biexcitons XX form at high power when a 2nd exciton is created before the 1st one recombines .

Observations of K. Matsuda *et al.*, PRL 91, 177401 (2003):

- Exciton and biexciton are elongated along the [-110] direction (anisotropy of the ≈100 nm natural QD in GaAs).

- XX images are more confined than X images due to exciton correlation (the lighter X particle « roams farther »).

→ The quantum constituents of a luminescence spectrum are spatially identified with no limit due to light diffraction: first reported by H.F. Hess *et al*. Science **264**, 1740 (1994).

Some related works:

- GRENOBLE
  M. Brun *et al.*, *J. Microscopy* **202**, 202 (2001)
  M. Brun *et al., Solid State Commun.* **121**, 407 (2002)

- BERLIN
  F. Intonti *et al.*, *PRB* **63**, 075313 (2001)
  V. Emiliani *et al.*, *PRB* **64**, 155316 (2001)

- LAUSANNE
  A. Feltrin *et al., PRL* **95**, 177401 (2005)

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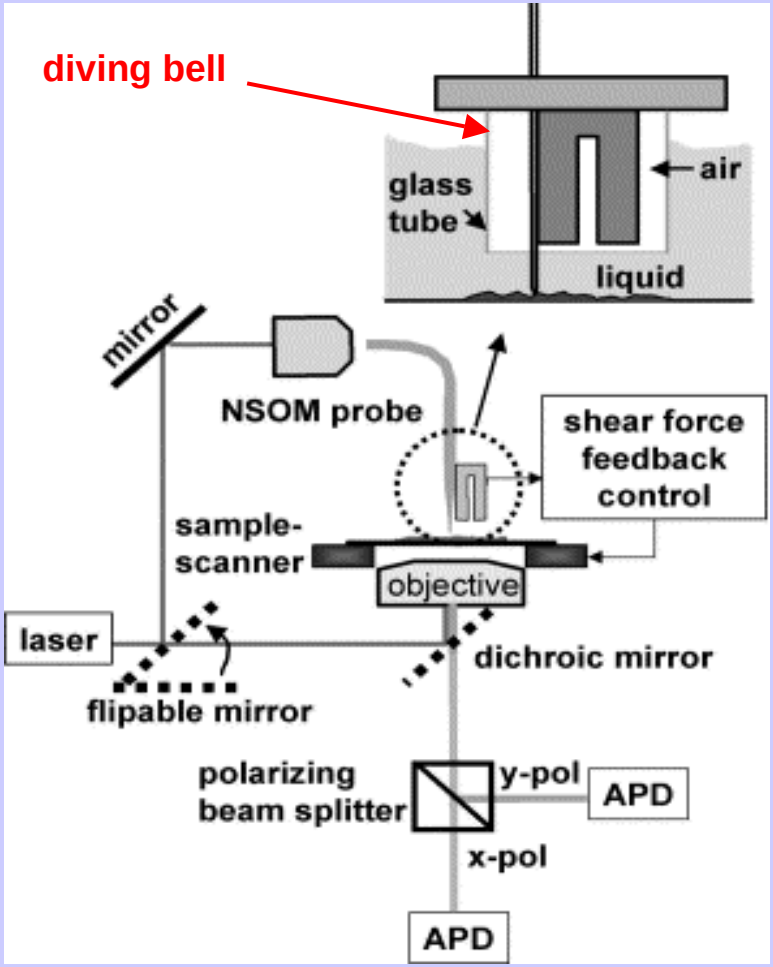
A special design to work in a liquid for biological studies, by M. Koopman *et al.*, *FEBS Lett.* 573, 6 (2004)

NB: group of N.F. van Hulst, Univ. of Twente, NL



Hybrid confocal NSOM microscope with a tuning fork in air and sample in liquid.

→ Weak interaction force of < 300 pN.

Three microscopes in one:

- Confocal
- NSOM
- « AFM » (topography)

The sub-diffraction sized organization of transmembrane proteins on dendritic cells

SPECIMEN:

Dendritic cells from human blood monocytes in buffered solution.

GOAL:

Determine how transmembrane proteins (DC-SIGN) are organized on the cell ?

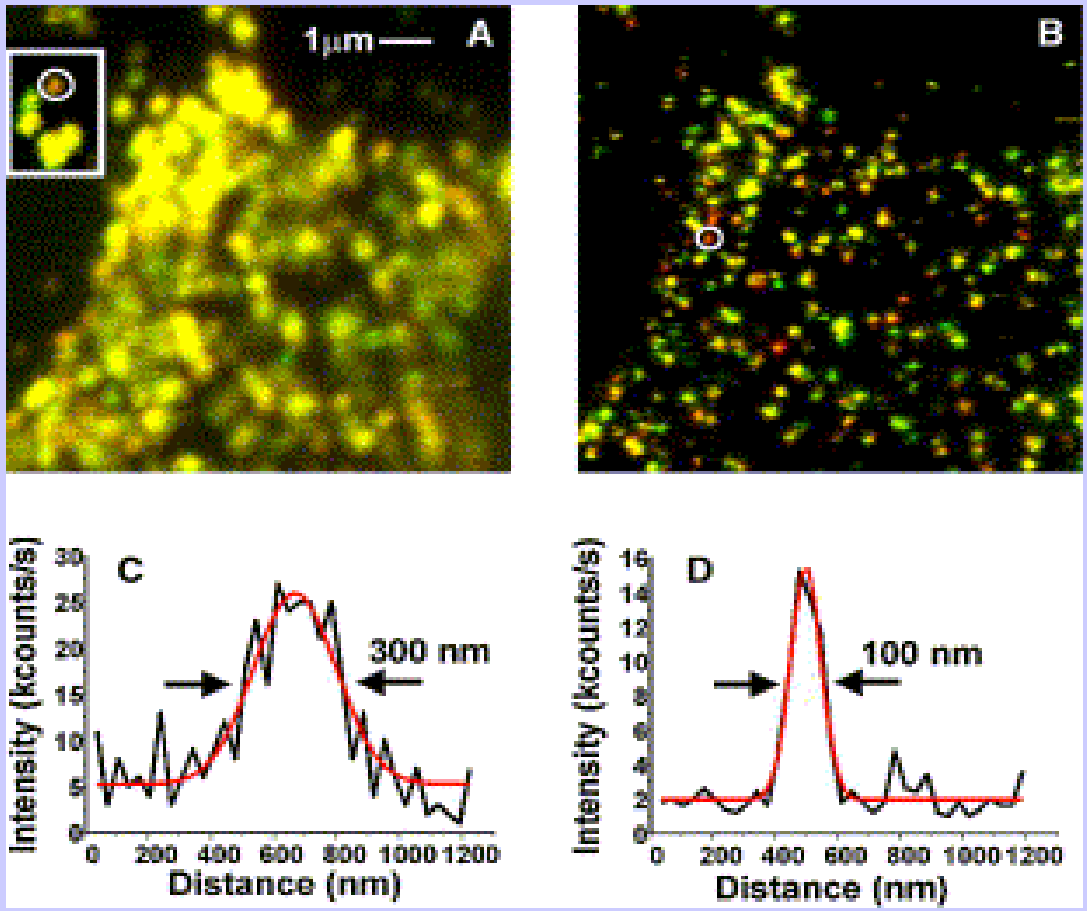
Proteins are labelled with a dye whose fluorescence is imaged.

RESULTS obtained from polarization-conserving **NSOM**:

DC-SIGN are organized in clusters of size ≤ 100 nm with a large spread of molecules in a domain (up to a factor 60).

Some proteins remain isolated.

Confocal NSOM



Taken from M. Koopman *et al. FEBS Lett.* **573**, 6 (2004)

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How to reach a super-resolution?

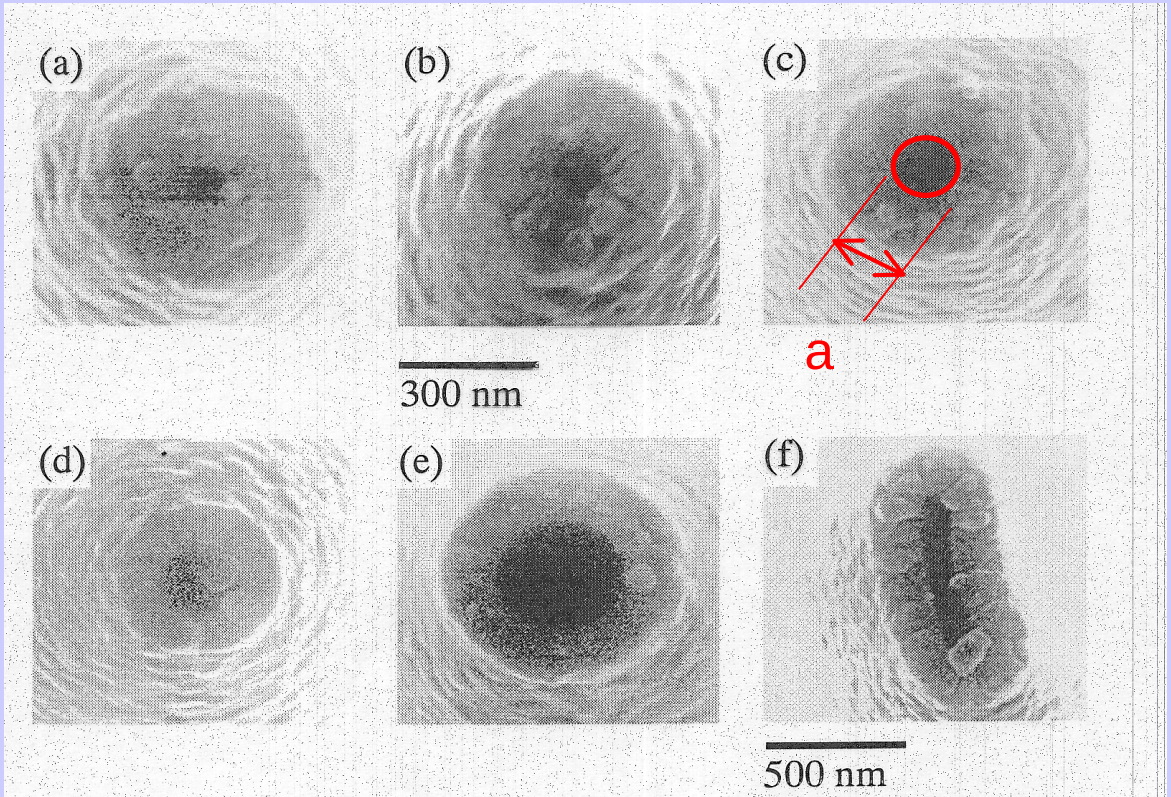


Figure 5.5: SEM images of apertures: (a)–(e) apertures with sizes of 80–300 nm that can be obtained by changing the respective pulling parameters. (f) has been obtained by focusing of the CO_2 laser spot too tightly.

SEM front views of metalized tapered fiber tips taken from Bert Hecht, thesis, University of Zurich (1996)

Decrease a ?

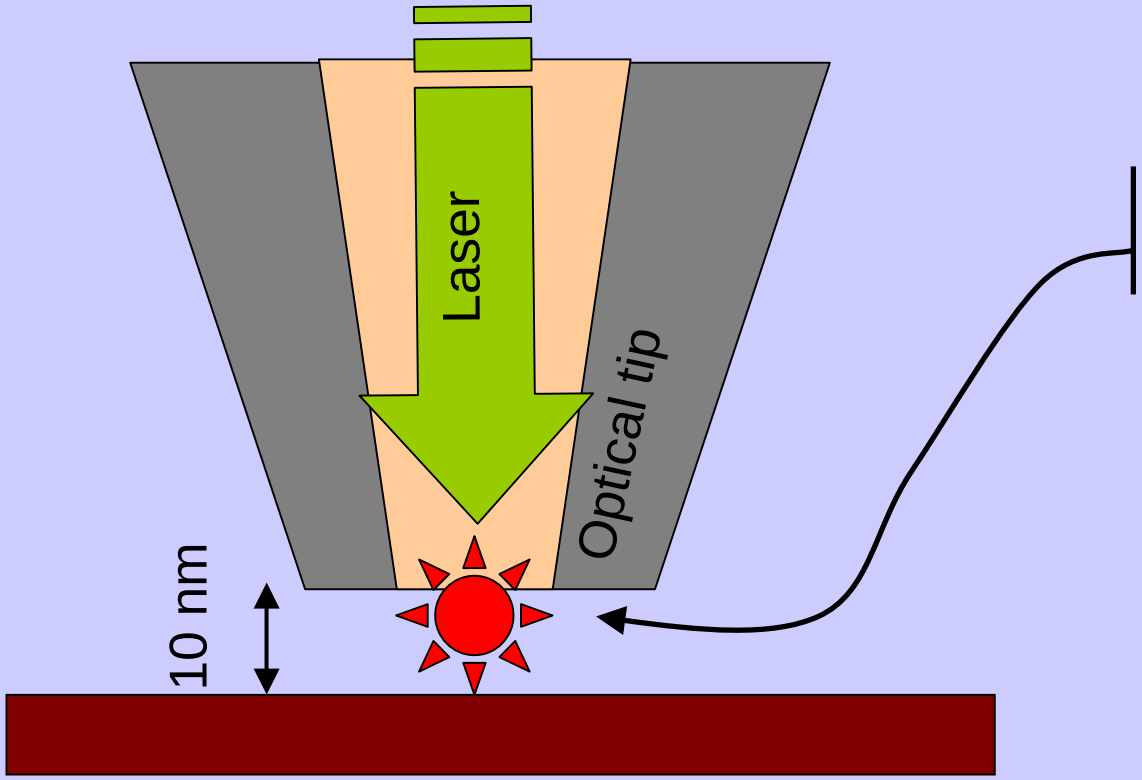
Not sufficient !

Because:

* transmission α a^4, therefore one is rapidly missing photons.

* resolution → 2δ , δ = penetration depth ($\delta \approx$ 10 nm in Al)

An interesting concept: the use of a single nano-object as source of light?



RECIPE:

- Take a NSOM tip
- Take a single fluorescent (nano-)object.
- Attach it at the tip apex.
- Excite it through the fibre tip.
- Use its emission light as nanometre-sized source of light….

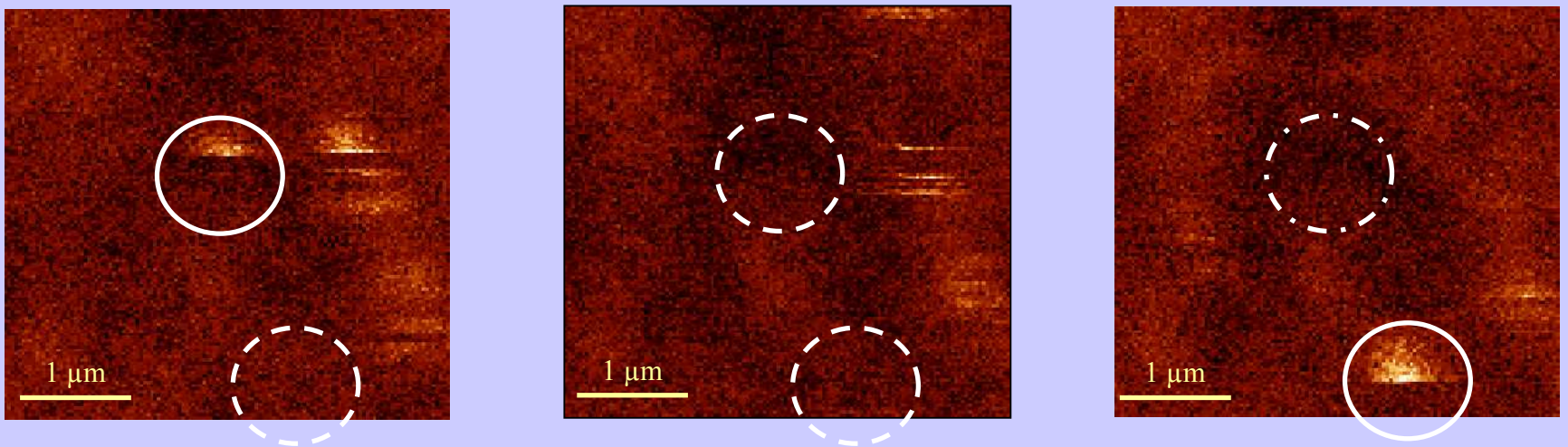
A few previous works

- J. Michaelis *et al*., *Nature* **405**, 325 (2000) [V. Sandoghdar group]
  - *Optics with a **single molecule** in an organic µ-cristal !!*
  - *Works only at low temperature, bleaches, limited resolution of 180 nm*
- S. Kühn *et al.*, *J. Microscopy* **202**, 2 (2001) [V. Sandoghdar group]
  - *Extension to a **V-N defect** in a diamond µ-cristal*
  - *Works at room temperature, no bleaching, but resolution limited to 300 nm*
- L. Aigouy, Y. De Wilde, M. Mortier, *APL* **83,** 147 (2003) [ESPCI Paris]
  - *Micro-particles of **erbium**-doped glass*
  - *Works at 300K, very convincing images, resolution achieved so far 300 nm, but should be improved by using smaller particles*
- Our on-going contribution (collaboration: *CEA Grenoble + Bath Univ. + Troyes Univ.)*
  - *Use of a **single CdSe nanocrystal***
  - *Works at 300 K, nanometre-sized object, very stable, etc..*

    but ***blinks !***

Fluorescence microscopy of single CdSe nanocrystals → blinking

Scanning confocal microscopy
CdSe/ZnSe nanoX dispersed in a thin PMMA layer
Excitation @ 458 nm ; Collection @ [540-620 nm]
3 subsequent images



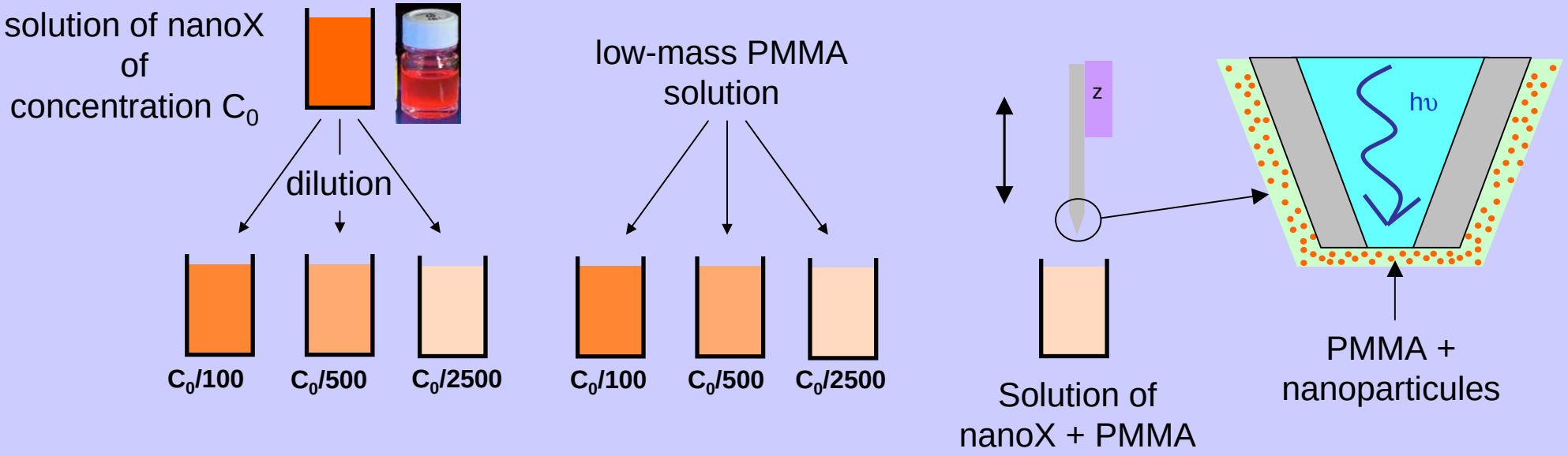
”Blinking” behaviour typical for a single object

See *e.g.* Shimizu *et al.*, *PRB* **63**, 205316 (2001)

Warning: this behaviour should manifest itself in the active tip.

Realization of a CdSe-nanoX-based active probe

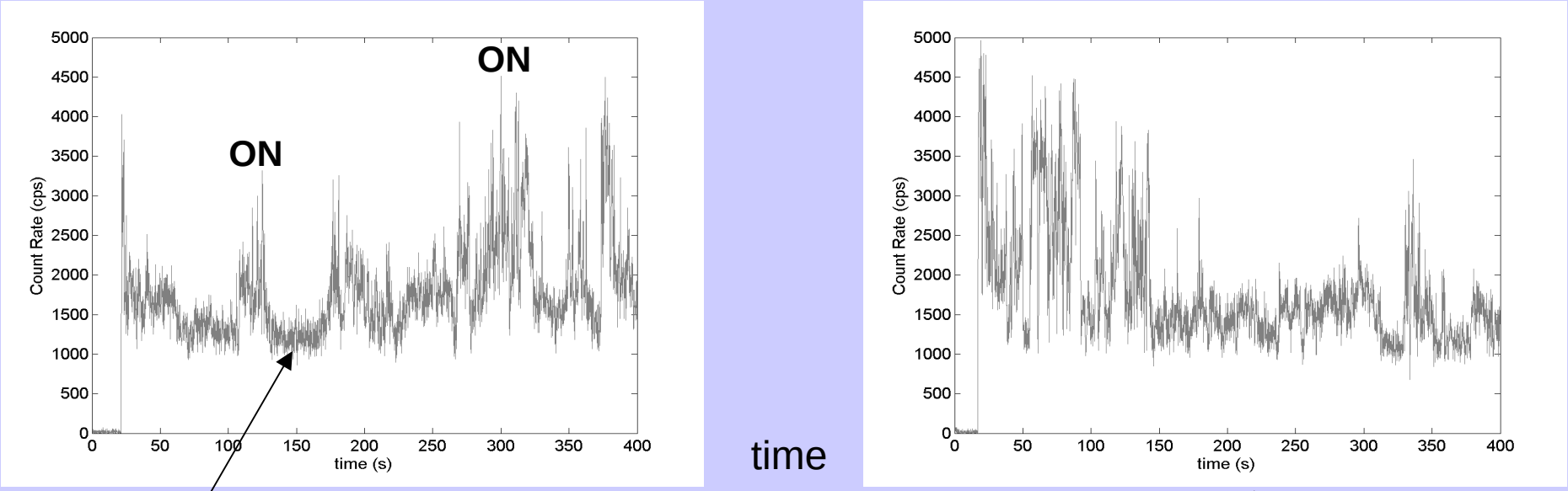
Coating of the initial NSOM tip with a thin PMMA layer stained with CdSe NCs. Progressive decrease of the NC concentration to obtain a few NCs at the tip apex.



Time evolution of the emission of the active probe (I)

Very « diluted » active tip

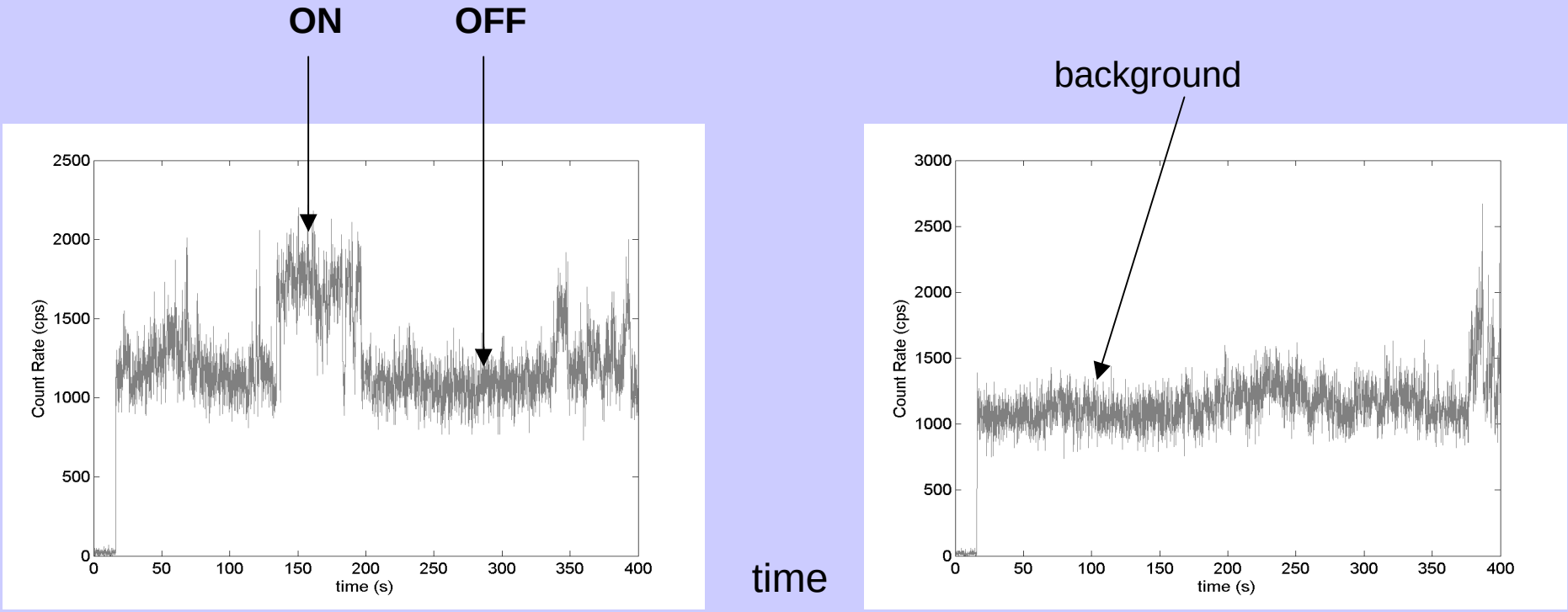
Excitation 458 nm, detection in photon-counting mode at 580 nm-620 nm



fibre background →

nanoX is (are) **OFF**

Time evolution of the emission of the active probe (II)

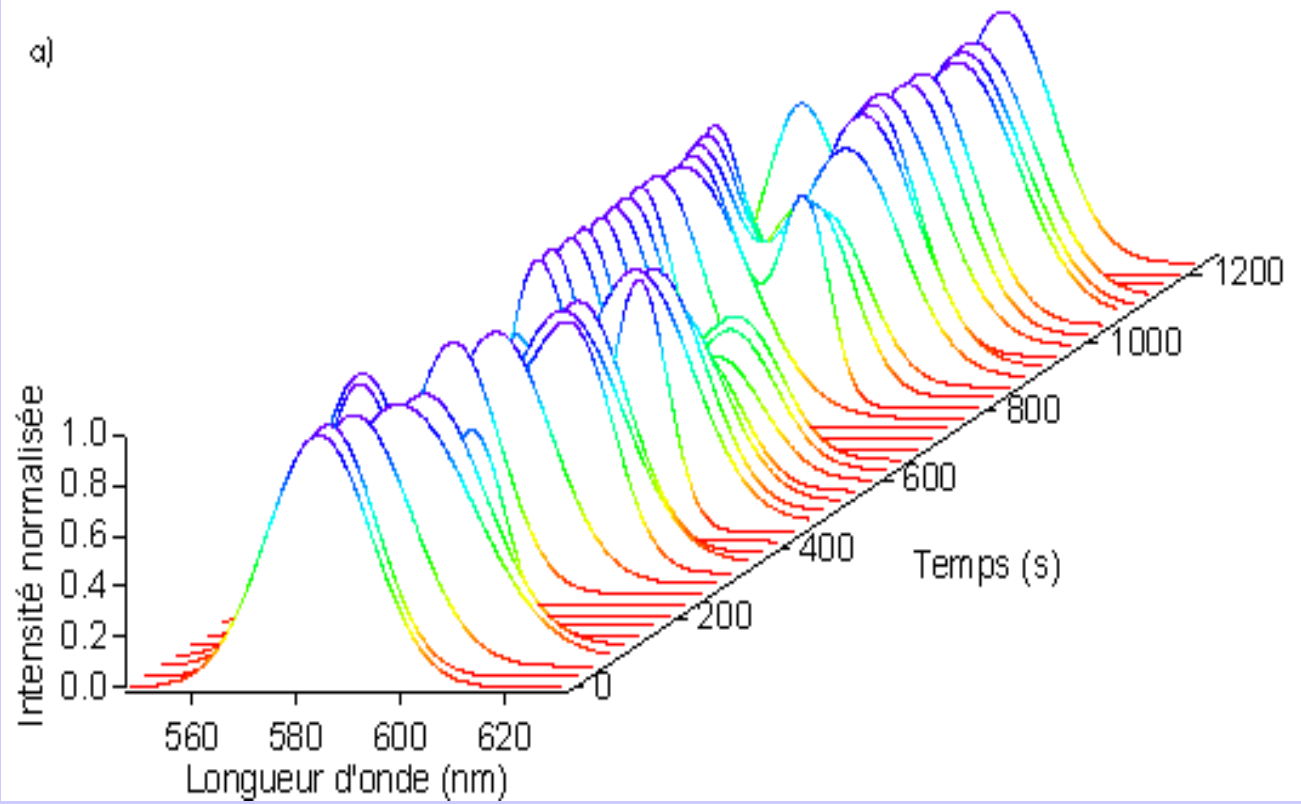


A few NCs are active only, perhaps only one ??

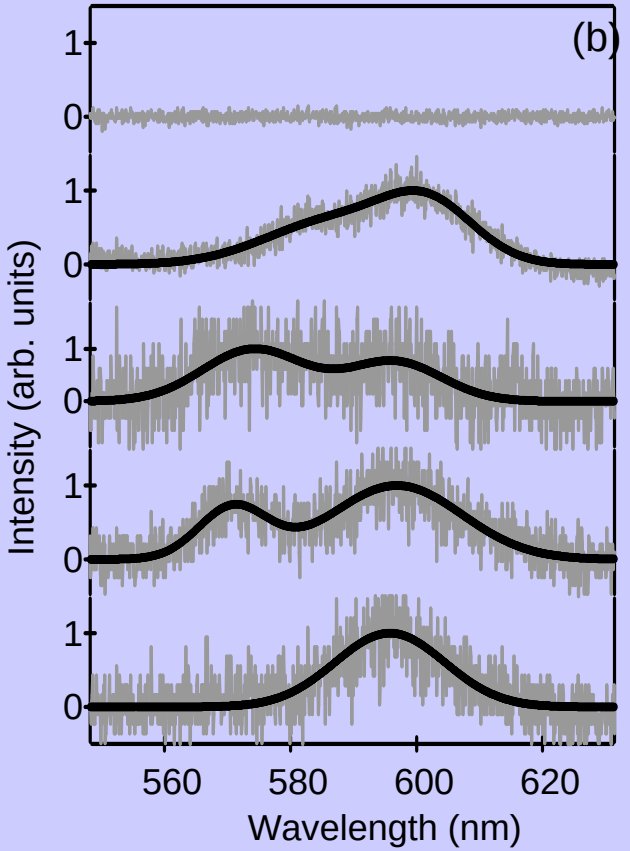
See N. Chevalier *et al.*, *Nanotechnology* **16**, 613 (2005)
See also: http://nanotechweb.org/articles/news/4/3/6/1

PL spectra of a « dilute » active probe: evidence for blinking

40 successive spectra, integration time= 30 s



A selection of 5 spectra

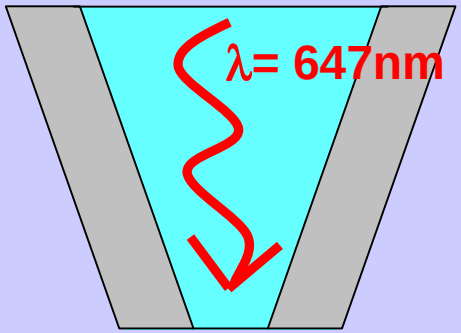


Only 2 or 3 NCs are active !?

NEXT STEP: ATTEMPT TO DO OPTICS WITH THIS ACTIVE TIP !

Reference image of a test sample taken with a regular tip

Test structures prepared in Bath (UK) in collaboration with S. Maier's group

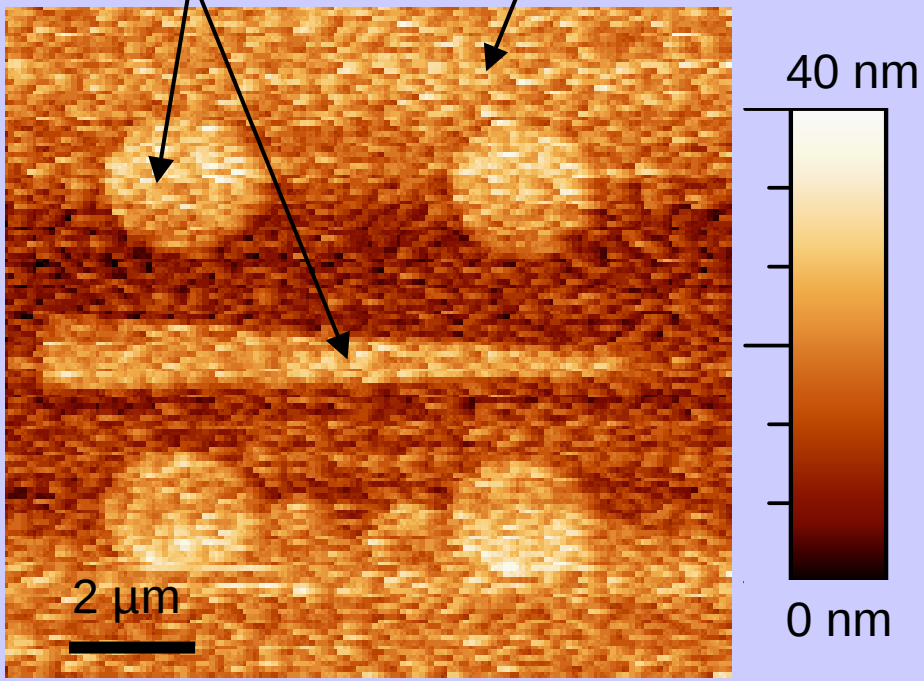


180 nm aperture tip

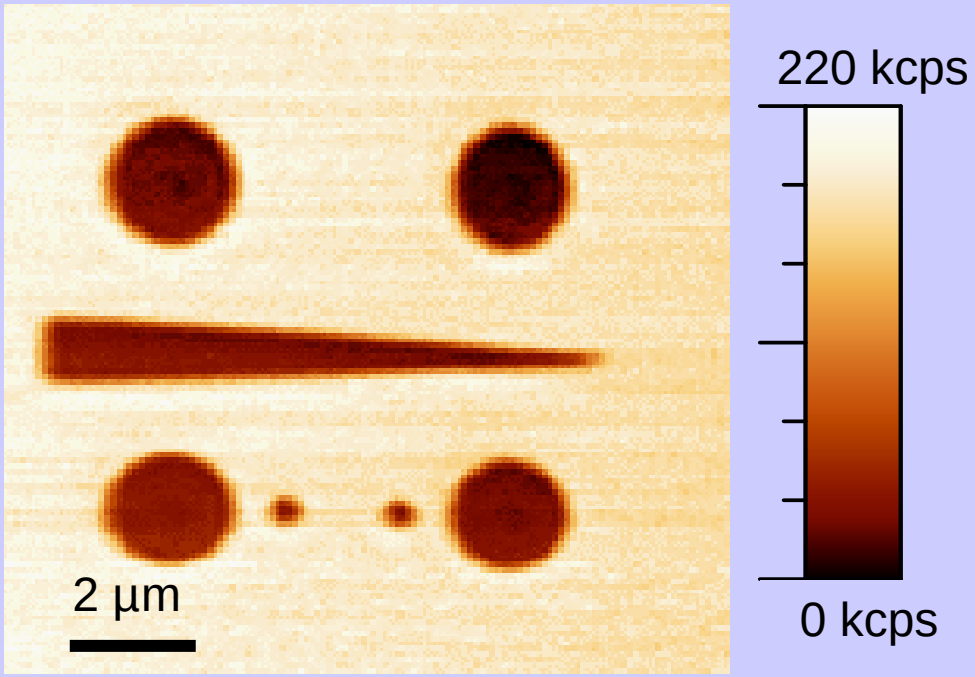
transmission= 0.5 %

Gold patterns 40 nm thick

SiO_2

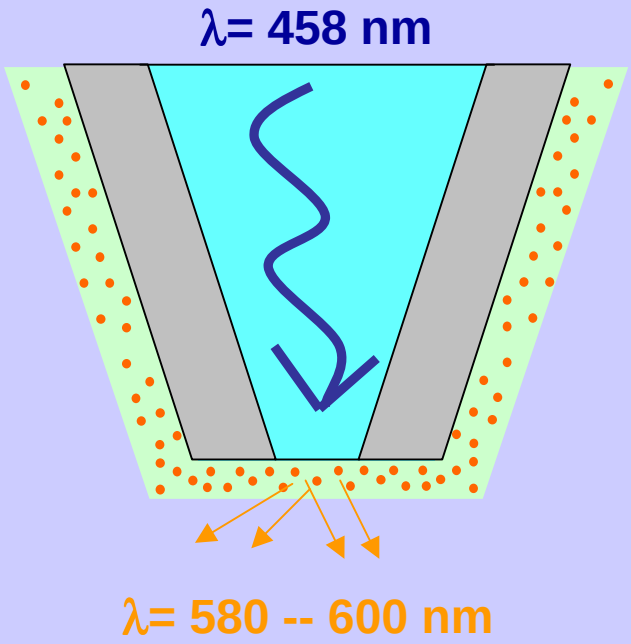


Topography image (tuning fork)

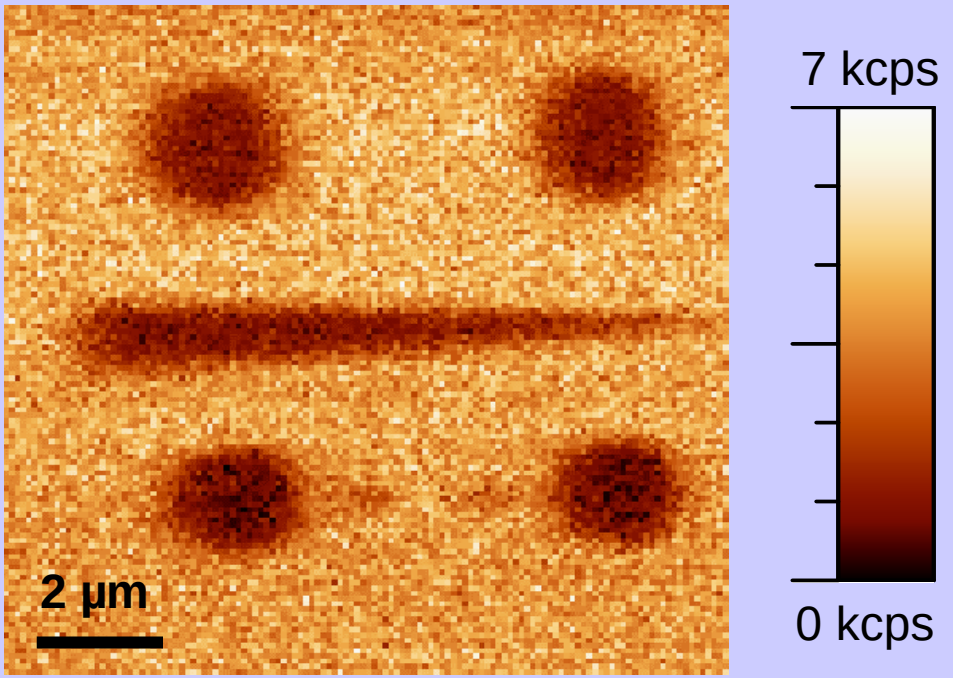


NSOM transmission image (home-made NSOM)

A second reference image taken with a highly stained active tip

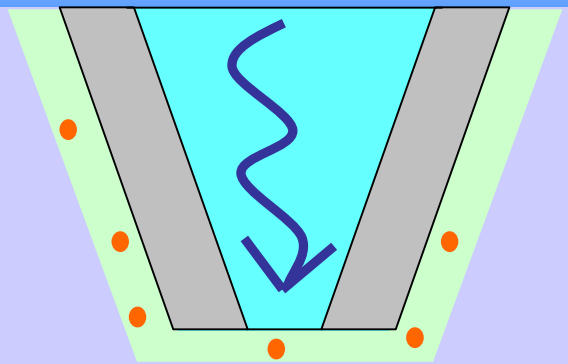


Tip is « doped » with a large (>> 10) number of nanoX at the apex

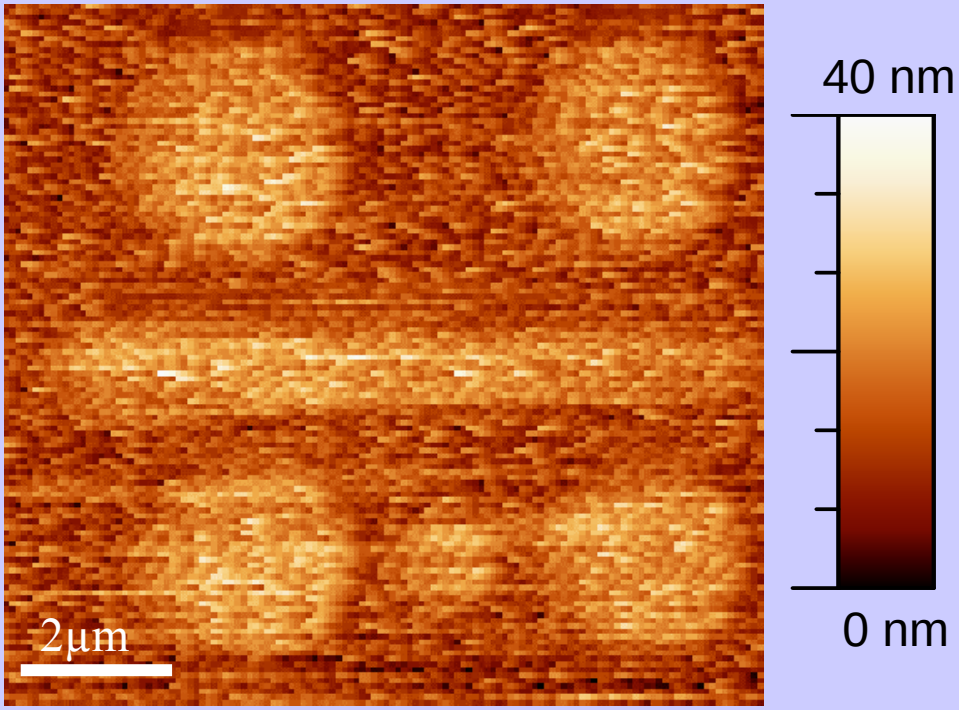


NSOM transmission image @ [540-620 nm], i.e., the fluorescence emission of CdSe nanoX

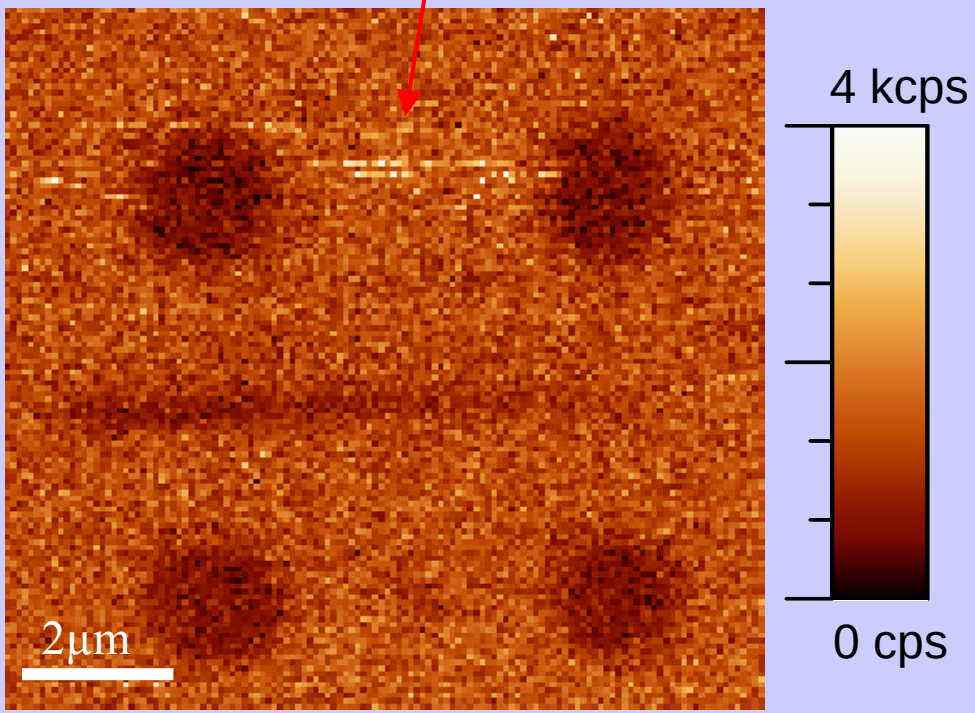
On-going step: optical imaging with a single nanoX (I) *[first images taken by Y. Sonnefraud on 10 november 05]*



WHAT ARE THESE BRIGHT PIXELS ?!

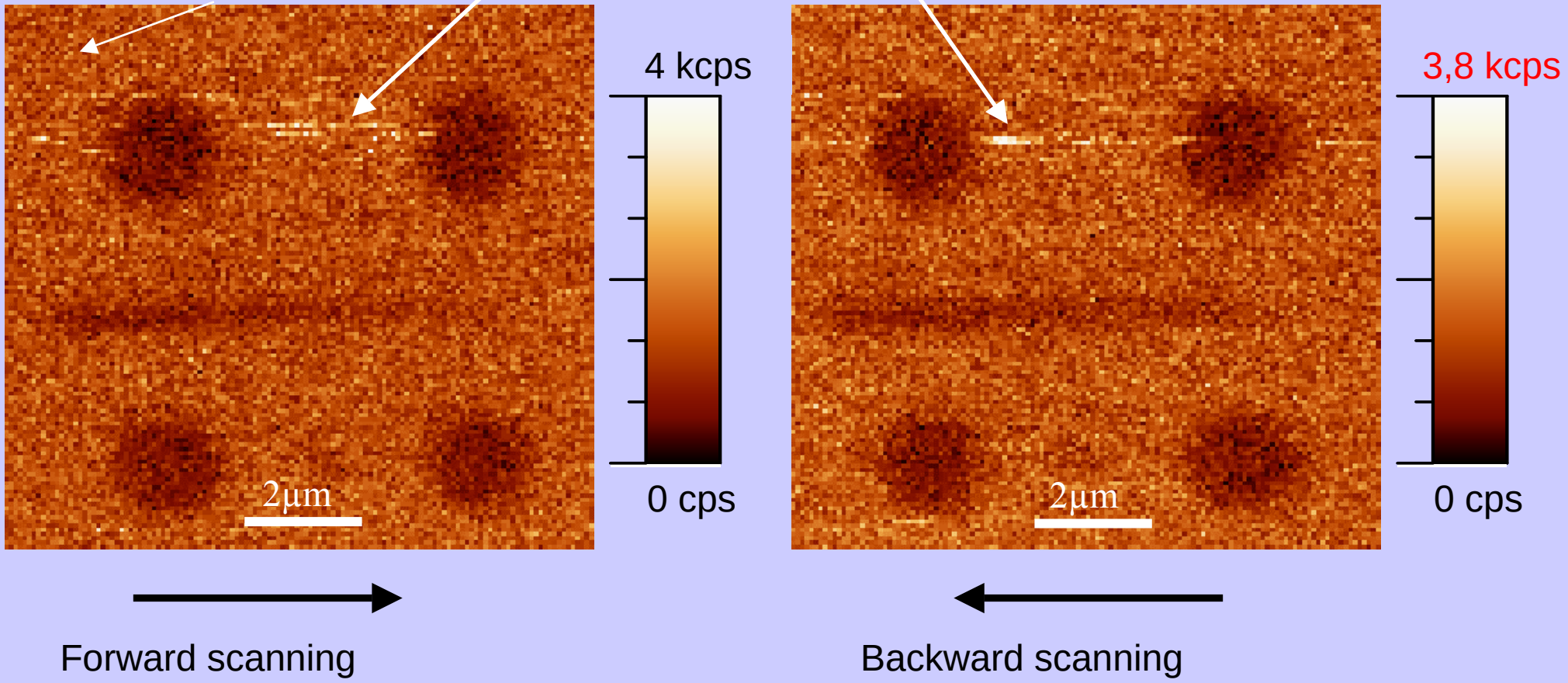


Topography (rather « thick » tip)



NSOM @ [540-620 nm]

On-going step: optical imaging with a single nanoX (II) *[first images taken by Y. Sonnefraud on 10 november 05]*



There is still plenty of room for improvement !!

(control of the initial tip with a FIB with LETI, control of the attached nanoX with CEA &Troyes, etc..)

“La morale de l’histoire”

Light diffraction has long been considered as a fatal limitation hindering the development of optics over dimensions smaller than ≈ half the wavelength of light. This is an old story now !

Thanks to the development of NSOM in the last 2 decades, optics has definitively entered the nanoworld.

“Soon” it will be possible to do optics at a true nanometre scale ….

See more at: http://nsom.online.fr

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