Near-Field Scanning Optical Microscopy: a Brief Overview

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Thanks to my former & present collaborators in SPECTRO:


*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:
Confocal: diffraction-limited to 250 nm $10^8$ nm$^3$
NSOM: 30-50 nm $10^5$ nm$^3$

Excitation volume:
Confocal: “large”
NSOM: smaller

Background signal:
Confocal: Small excitation intensity
NSOM: Non-contact microscopy

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

Confocal:
- “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.

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

*Karraï-Grober, 1995*

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**Note:** To beat the diffraction limit, both the aperture size and the tip-surface distance must be \(< \lambda\)
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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

Isolate an interesting feature and map its intensity versus tip position

True spectra of CdTe QDs at 4 K
M. Brun et al.
Near-field optical mapping of exciton wave functions in a GaAs QD (I), Matsuda et al., PRL 91, 177401 (2003)

The NSOM tip

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

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!

Note: a photon is absorbed $\rightarrow$ 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) $\rightarrow$ 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.
Biexcitons XX form at high power when a 2\textsuperscript{nd} exciton is created before the 1\textsuperscript{st} one recombines.


- Exciton and biexciton are elongated along the [-110] direction (anisotropy of the \(\approx 100\) 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)

- **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 $\leq 100$ nm with a large spread of molecules in a domain (up to a factor 60).

Some proteins remain isolated.

Taken from M. Koopman *et al.* FEBS Lett. 573, 6 (2004)
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How to reach a super-resolution?

Decrease $a$?
Not sufficient!
Because:
* transmission $\propto a^4$, therefore one is rapidly missing photons.
* resolution $\rightarrow 2\delta$, $\delta = $ penetration depth ($\delta \approx 10$ nm in Al)

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)
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

  - Optics with a **single molecule** in an organic μ-cristal !!
  - Works only at low temperature, bleaches, limited resolution of 180 nm
  - Extension to a **V-N defect** in a diamond μ-cristal
  - Works at room temperature, no bleaching, but resolution limited to 300 nm
  - 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.

solution of nanoX of concentration \( C_0 \)

\[
\text{dilution} \quad \text{dilution} \quad \text{dilution}
\]

\[
C_0/100 \quad C_0/500 \quad C_0/2500
\]

\[
\text{low-mass PMMA solution}
\]

\[
C_0/100 \quad C_0/500 \quad C_0/2500
\]

Solution of nanoX + PMMA

PMMA + nanoparticles
Very « diluted » active tip
Excitation 458 nm, detection in photon-counting mode at 580 nm-620 nm

Time evolution of the emission of the active probe

- fibre background → nanoX is (are) OFF
A few NCs are active only, perhaps only one ??

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

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

Gold patterns 40 nm thick

SiO$_2$

Topography image (tuning fork)

NSOM transmission image (home-made NSOM)

Reference image of a test sample taken with a regular tip

$\lambda = 647$nm

180 nm aperture tip

transmission = 0.5 %
A second reference image taken with a highly stained active tip

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

$\lambda = 458$ nm

$\lambda = 580 -- 600$ nm

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]

**Topography (rather « thick » tip)**

WHAT ARE THESE BRIGHT PIXELS ?!
On-going step: **optical imaging with a single nanoX (II)** [first images taken by Y. Sonnefraud on 10 November 05]

Signal due to background transmission of the fibre

Useful signal due to a single nanoX “attached” to the tip !!!

Forward scanning

Backward scanning

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..)
Light diffraction has long been considered as a fatal limitation hindering the development of optics over dimensions smaller than $\approx$ 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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