

Active control in Nanophotonics for Super-resolution Imaging

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Acknowledgments

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Electrical Engineering

■ ■ ■ ■ ■ Electronics
■ ■ ■ ■ ■ Computers
■ ■ ■ ■ ■ Communications



Outline

□ Background

- Microscopy over time
- Nanophotonics promises for microscopy
- Control - shaping optical wavefronts

□ Proof of concept for new microscopy

- Controlling nanophotonic/plasmonic wavefronts
- Focusing and scanning microscopy

□ Super-resolution

- Platforms for super-resolution
- Silicon Plasmonics

Microscopy

We started like this



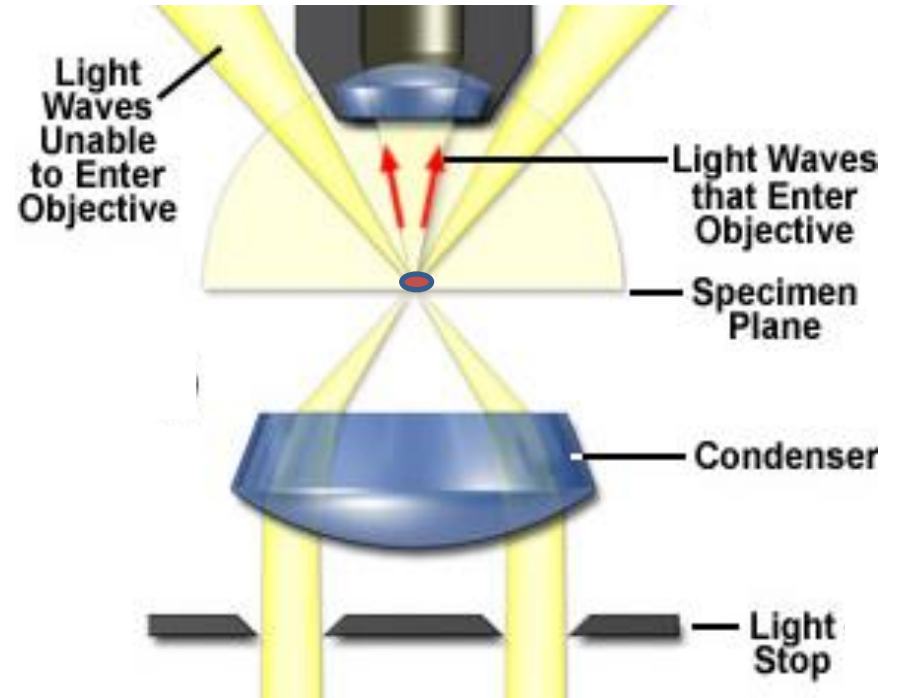
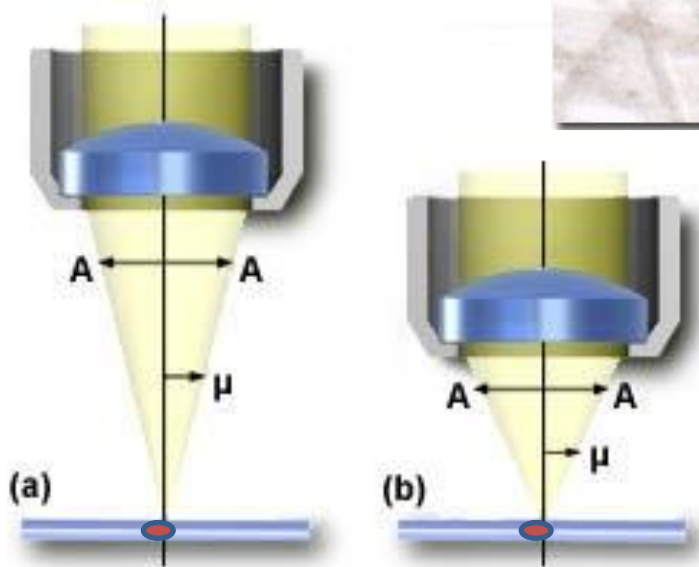
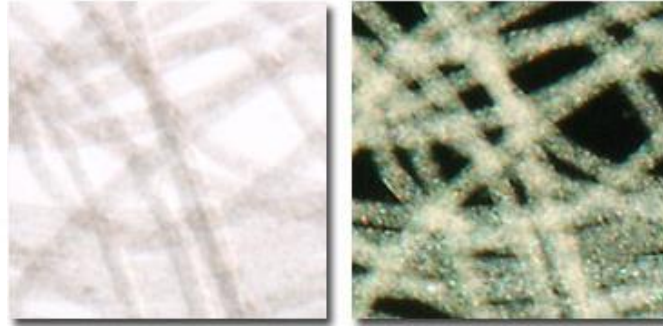
**Few Nobels later
(Nikon Storm)**



Bright field

–

Dark field



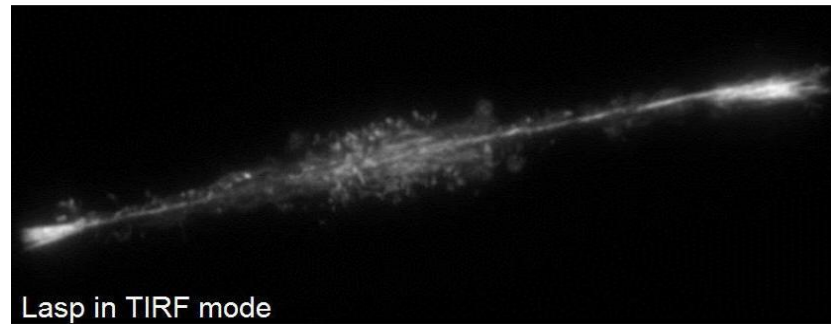
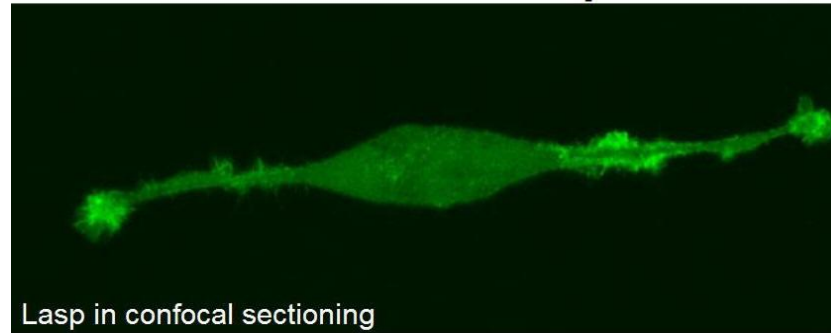
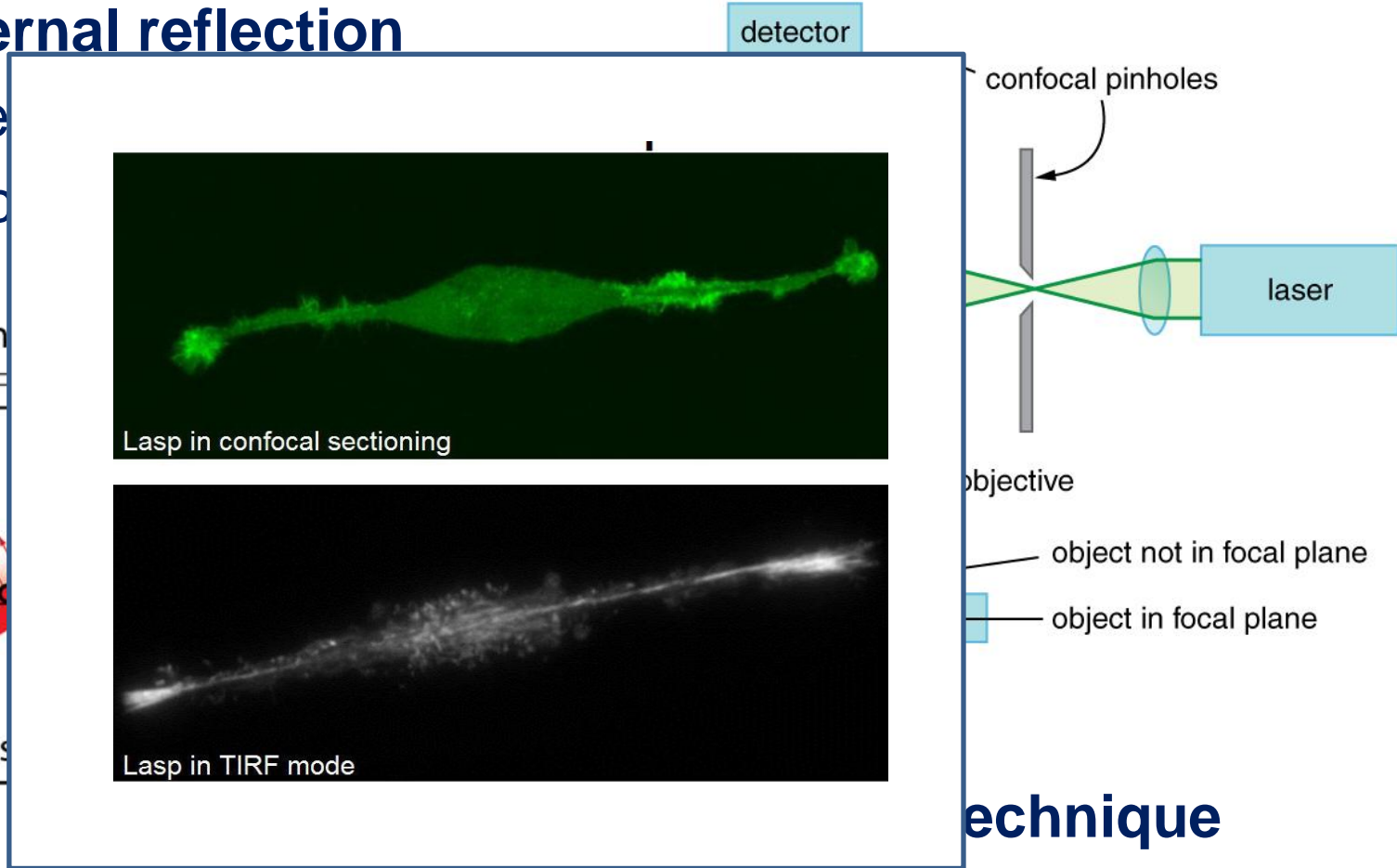
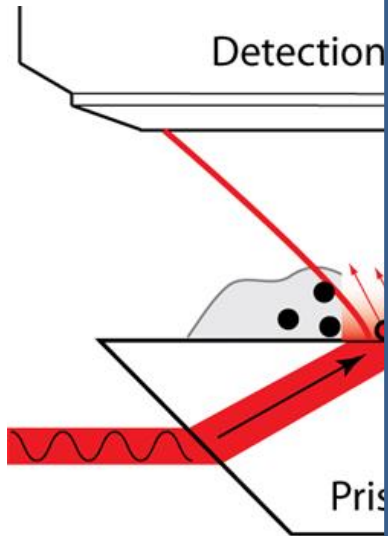
TIRF

&

Confocal

□ Total internal reflection

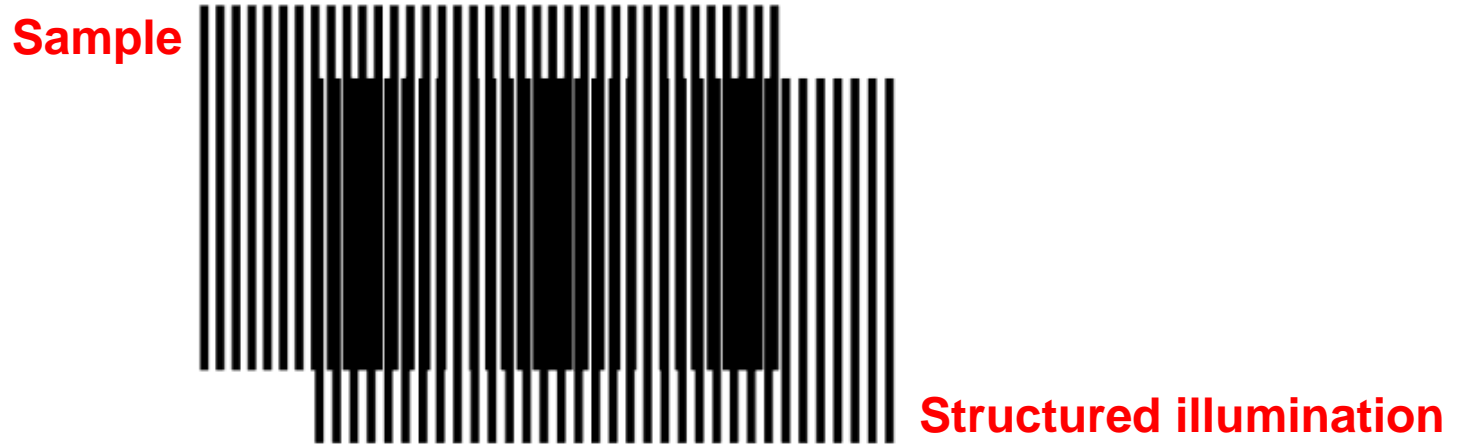
- Evanescent wave
- Surface plasmon resonance



- Focused excitation
- Focused detection

Structured illumination

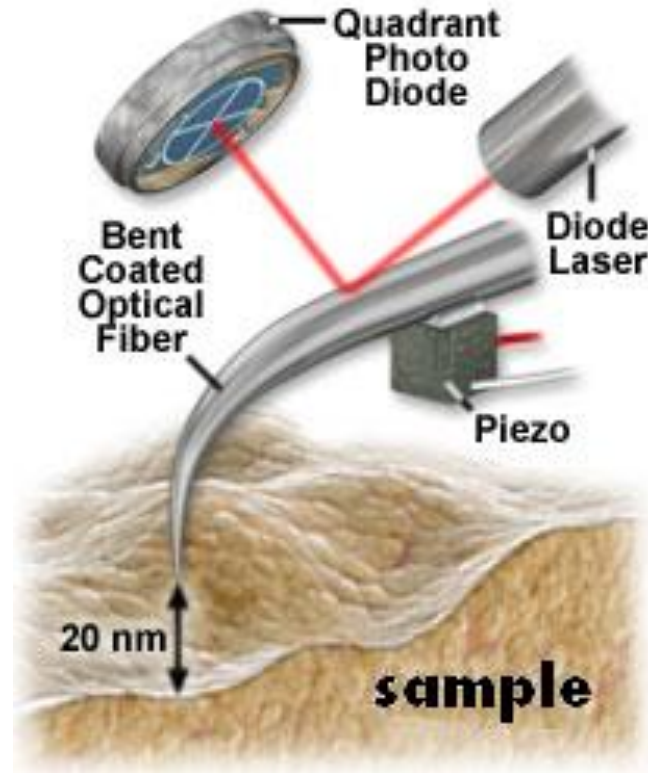
- ❑ Two different gratings generate a third one



If resolution does not allow to resolve one of the gratings,
You can still deduce it from the other two.

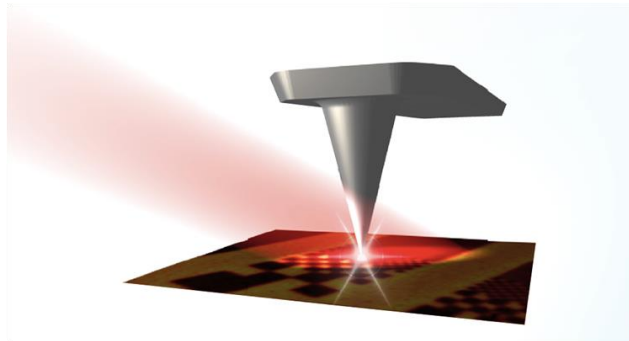
Near field Probes

- ❑ No more lenses
 - Detect with a scanning probe
 - Resolution is given by the probe size



Near – field techniques

Scattering NSOM
Light collected by
lens/parabolic mirror



Advantages:

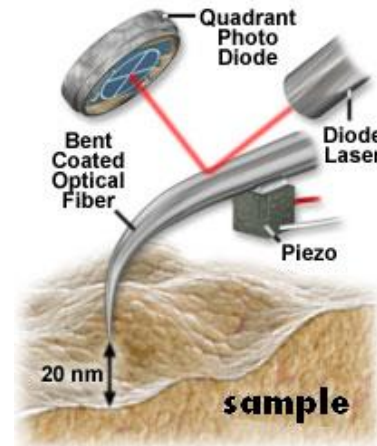
Resolution ($\sim 2-20$ nm)

Disadvantages:

Far field | Background

Scanning Sample

Aperture NSOM
Light coupled into optical fiber



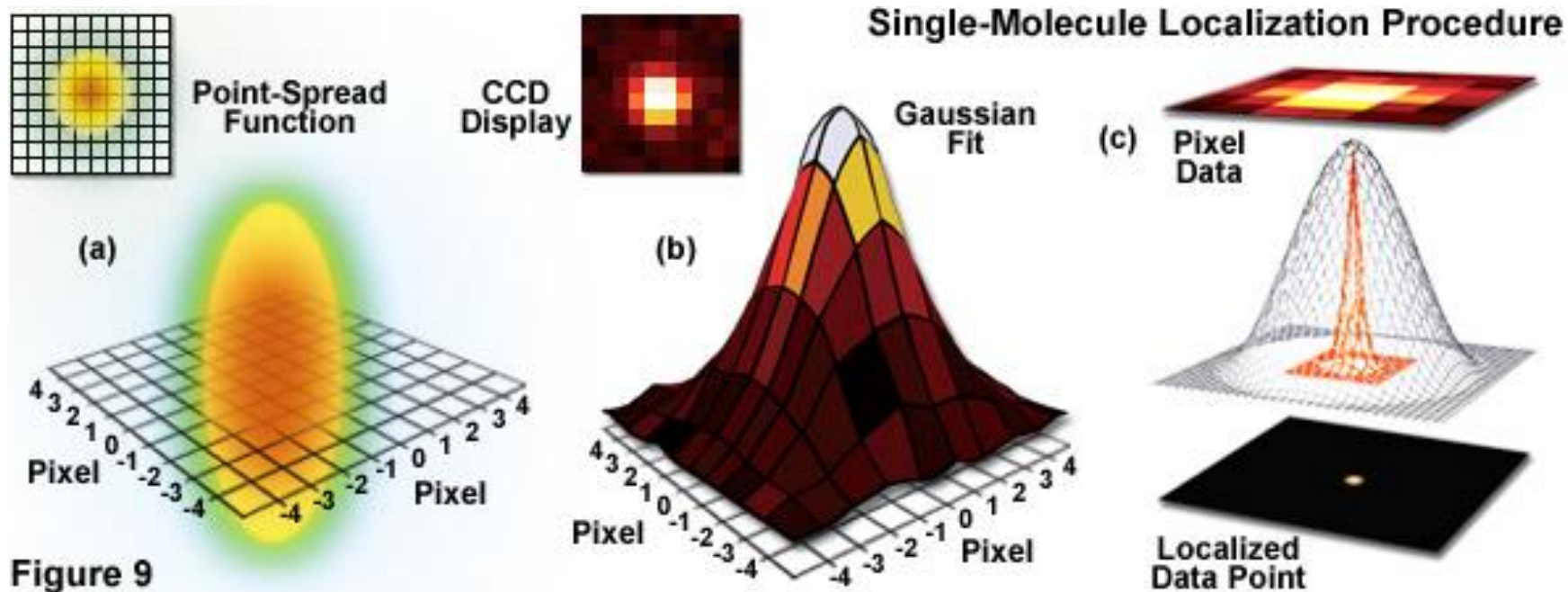
Advantages:

No Background

Disadvantages:

Limited resolution (~ 100 nm)

Single molecule localization



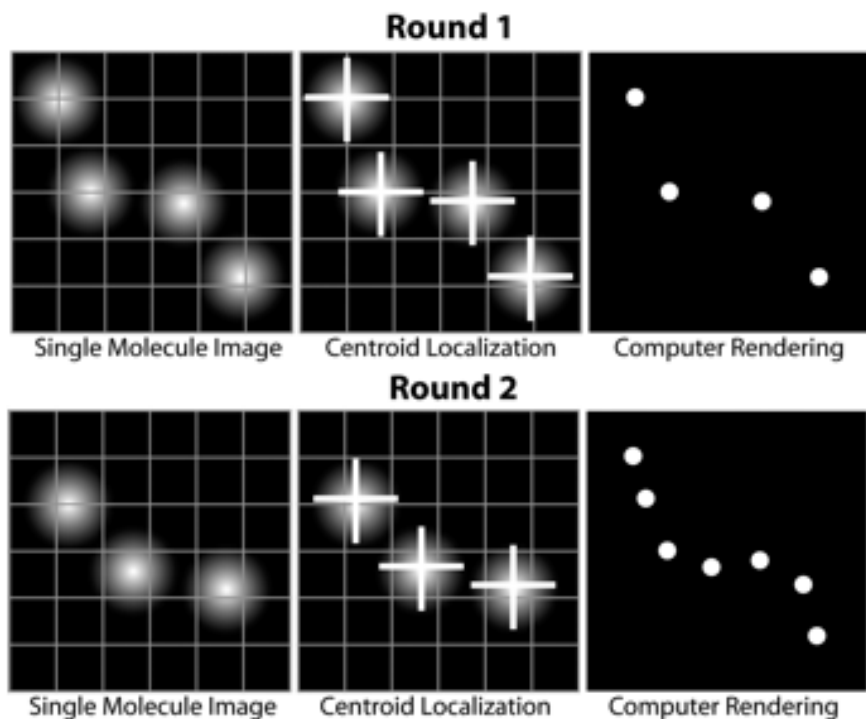
- If two single molecules are well separated in *space*
 - You can localize them by their center of mass

PALM, STORM

STED

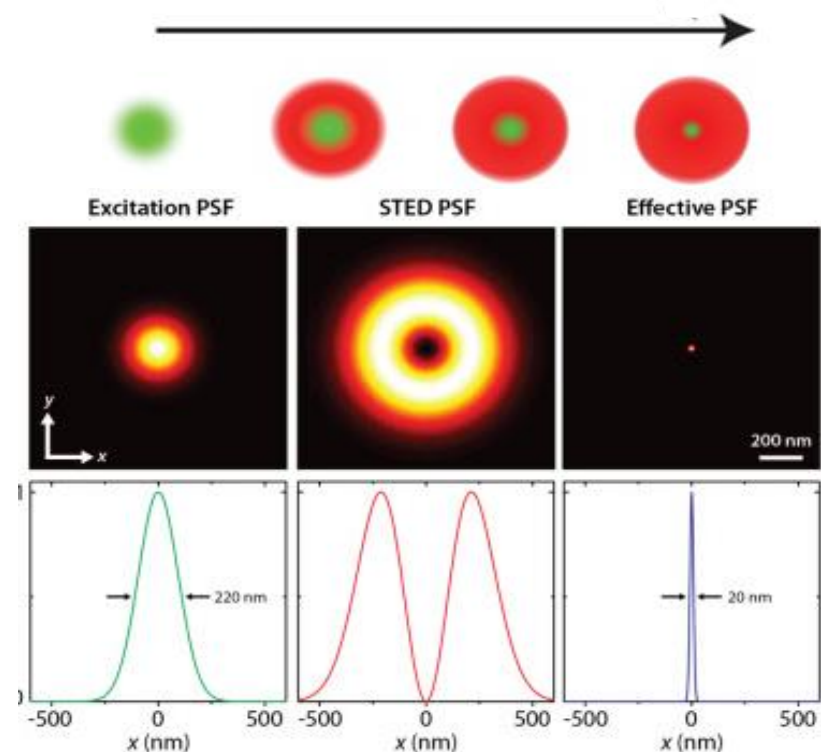
If molecules are separated
in *time or space*

You can sum up an image
(switching, activating)



If molecules are separated
in *energy*

You can scan an image
(saturation, depletion)



Outline

- **Wavefront shaping in Nanophotonics (spatial resolution)**
 - Controlling waves in nanophotonic systems
 - Focusing and scanning microscopy / Super-resolution
 - Goal: towards nanolocalized Raman excitation

- **Wavefront shaping in scattering media (spatio-temporal)**
 - Controlling speckle in time
 - Spatio-temporal focusing at targets
 - Goal: towards burning/imaging targets inside tissue

Optical microscopy: optimizations

- **Optics**

 - Oil/water immersion objectives

- **Illumination**

 - Dark Field, Phase Contrast (DIC)

 - Total Internal Reflection (TIRF)

 - Optical Coherence Tomography (OCT)

 - Structured Illumination (SIM)

- **Detection**

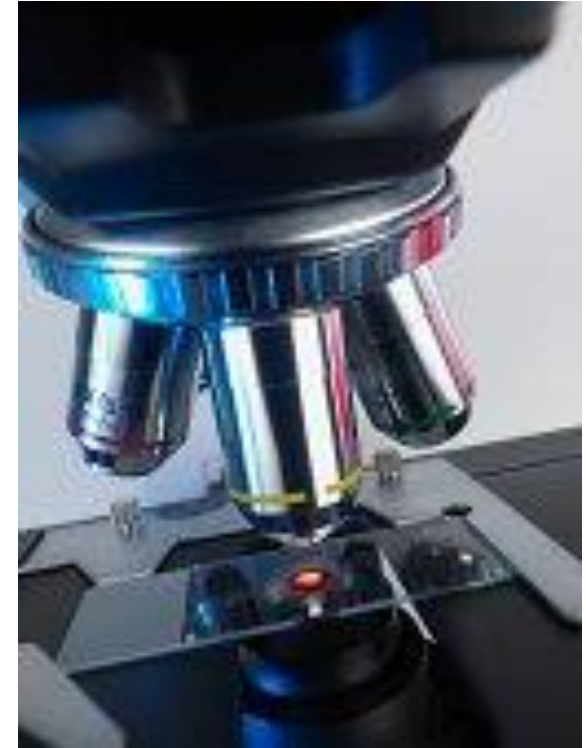
 - Fluorescence,

 - Confocal, Near-Field

- **Light-matter interaction**

 - STED, PALM, STORM

 - Nonlinear, Photo-acoustic

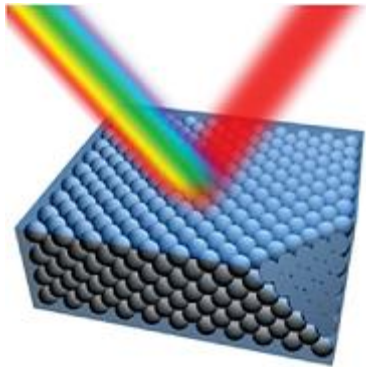


The microscope slide

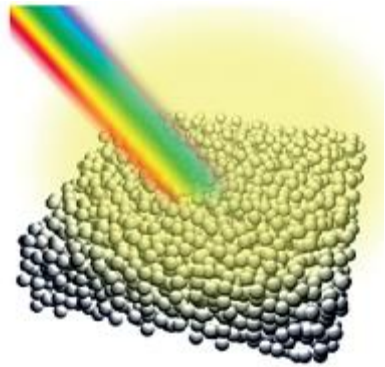
The generalized microscope slide



- ❑ **Conventional**
 - Passive, compensated
 - Cheap and standard

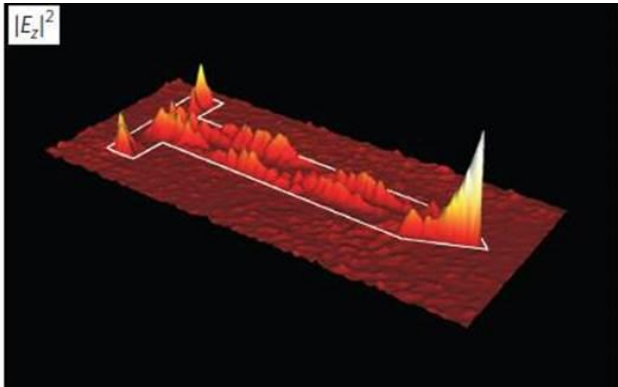


- ❑ **Nanophotonic** *More Control!*
 - Active, focusing, filtering
 - Super, hyper, meta lens



- ❑ **Natural** *More Control!*
 - Omnipresent, random, light scattering
 - Thin slicing *ex-vivo*, other waves

More control in Nanophotonics



Hillenbrand group (Nat Photon, 2012)



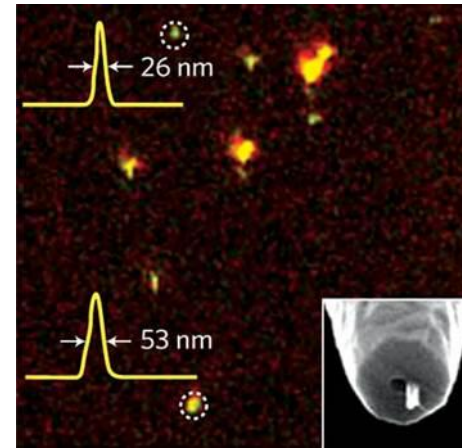
Choo, Yablonovitch (Nat Photon, 2012)

Resolution

- Hot spots 10 – 100 nm
- Fixed, No imaging

Required control

- Image formation



Novotny & Van-Hulst (Nat Photon, 2011)

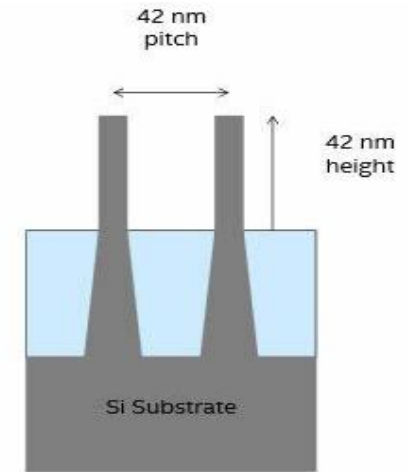
Motivation

□ Why do you need new microcopies?

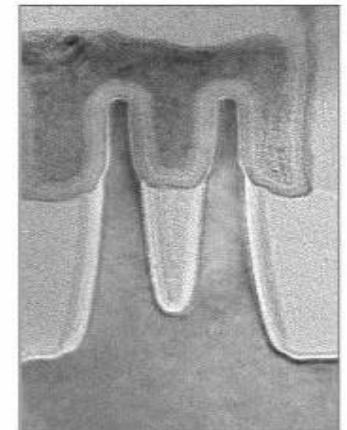
- Applied - latest transistors at 20nm
Cannot be fluorescently labeled

- Fundamental – spectroscopy

The spatial distribution provides the WHAT
You need spectroscopy to provide the HOW

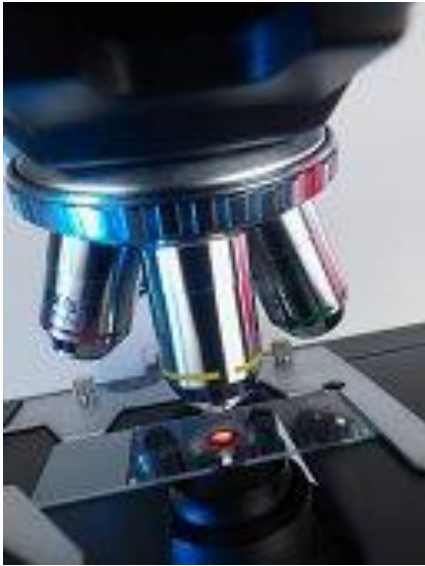


14 nm Process



14 nm 2nd Generation
Tri-gate Transistor

The slide is the lens



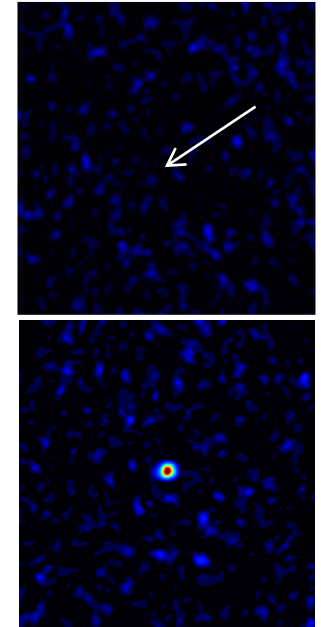
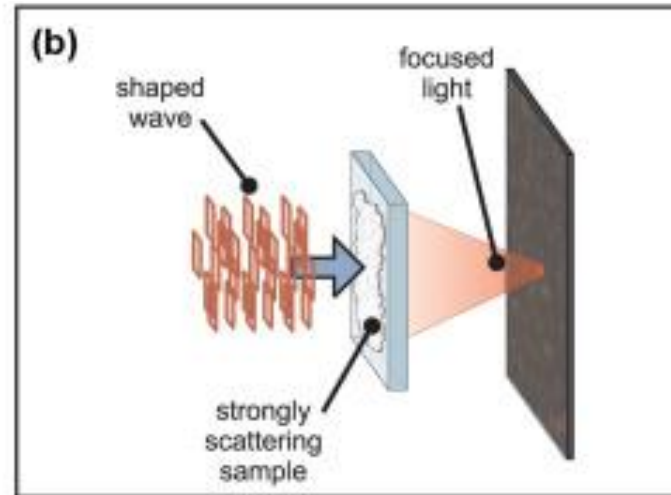
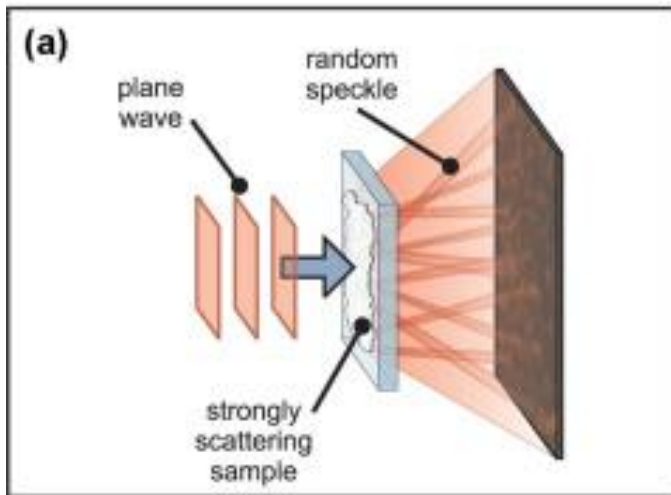
- **Metamaterial slide**
 - Negative refraction materials
 - Perfect lens

 - Hyperbolic material
 - Hyper lens

- **Plasmonics**
 - Short wavelengths
 - Plasmonic lens

Little success in bio imaging – We need more control!

Wavefront Shaping – new control

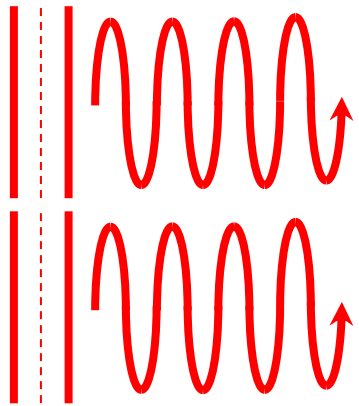


□ Control the incident wavefront

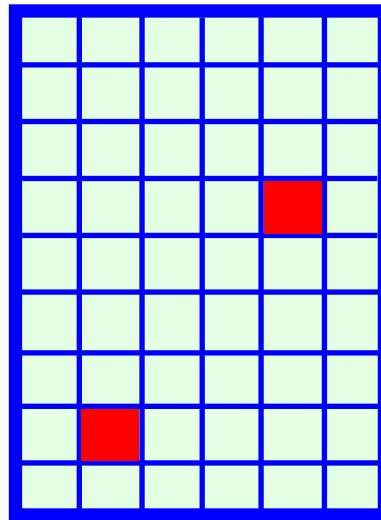
- Use a spatial light modulator to shape the front
- Use feedback to find the right wavefront
- Achieve focusing from scattering media (paper)
- Achieve focusing to a different spot

The wavefront control

Planar front

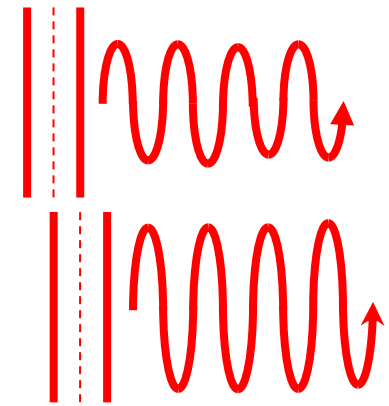


Spatial Light Modulator (SLM)



10 000 channels

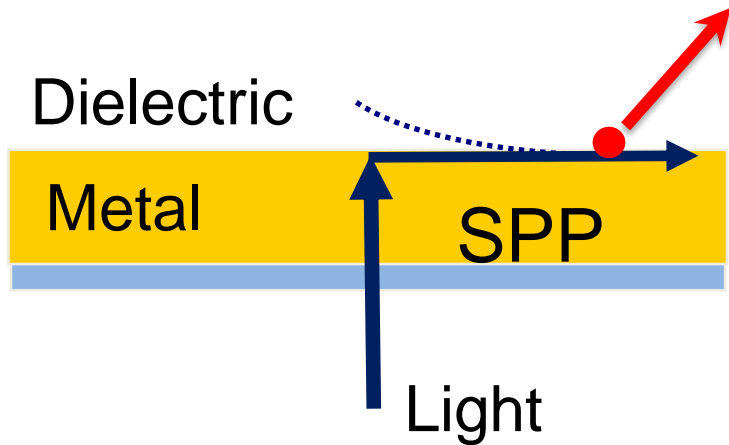
Shaped front
(amp & phase)



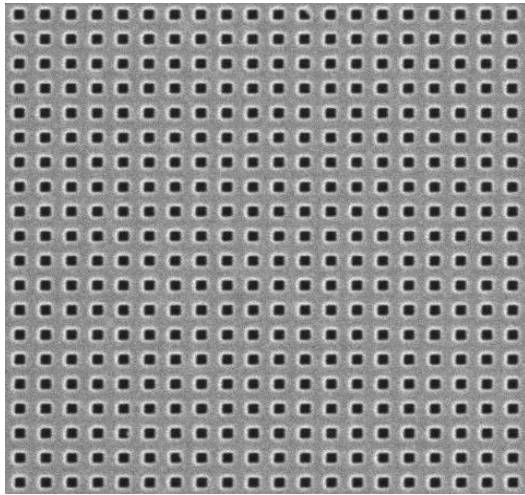
Nanophotonics wavefront control

*Is wavefront control in nanophotonics
applicable for microscopy?*

Surface Plasmon Polaritons (SPP)



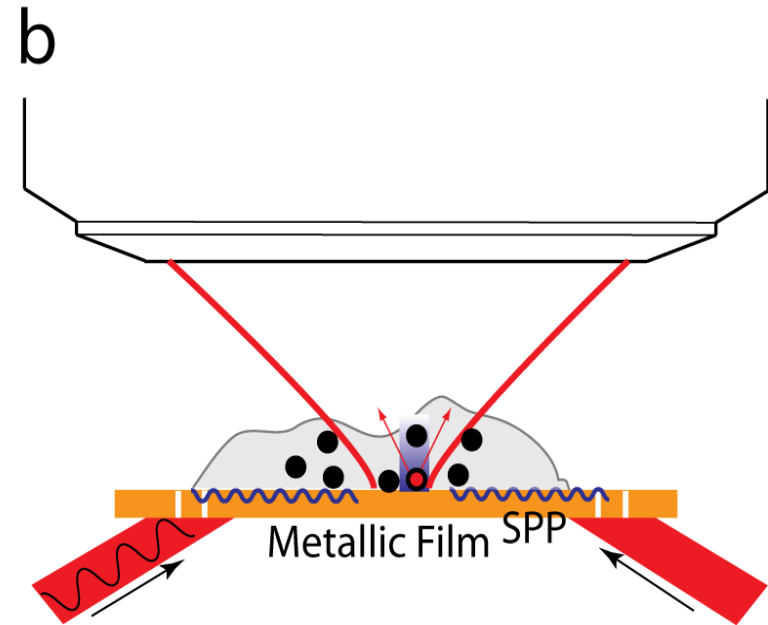
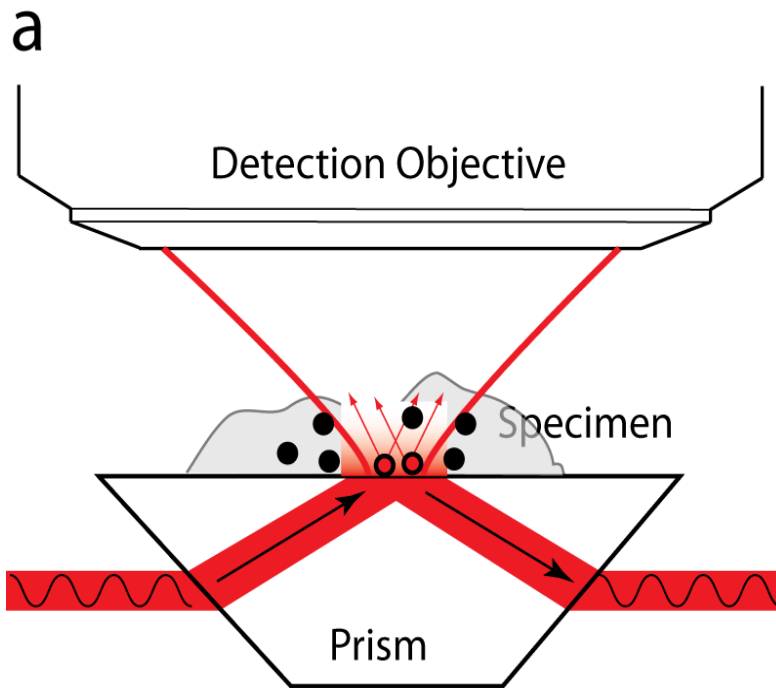
- ❖ Surface waves
- ❖ Evanescent waves
- ❖ Metal-dielectric interface
- ❖ Sensing, SERS, Q. optics



❑ Compared to photons:

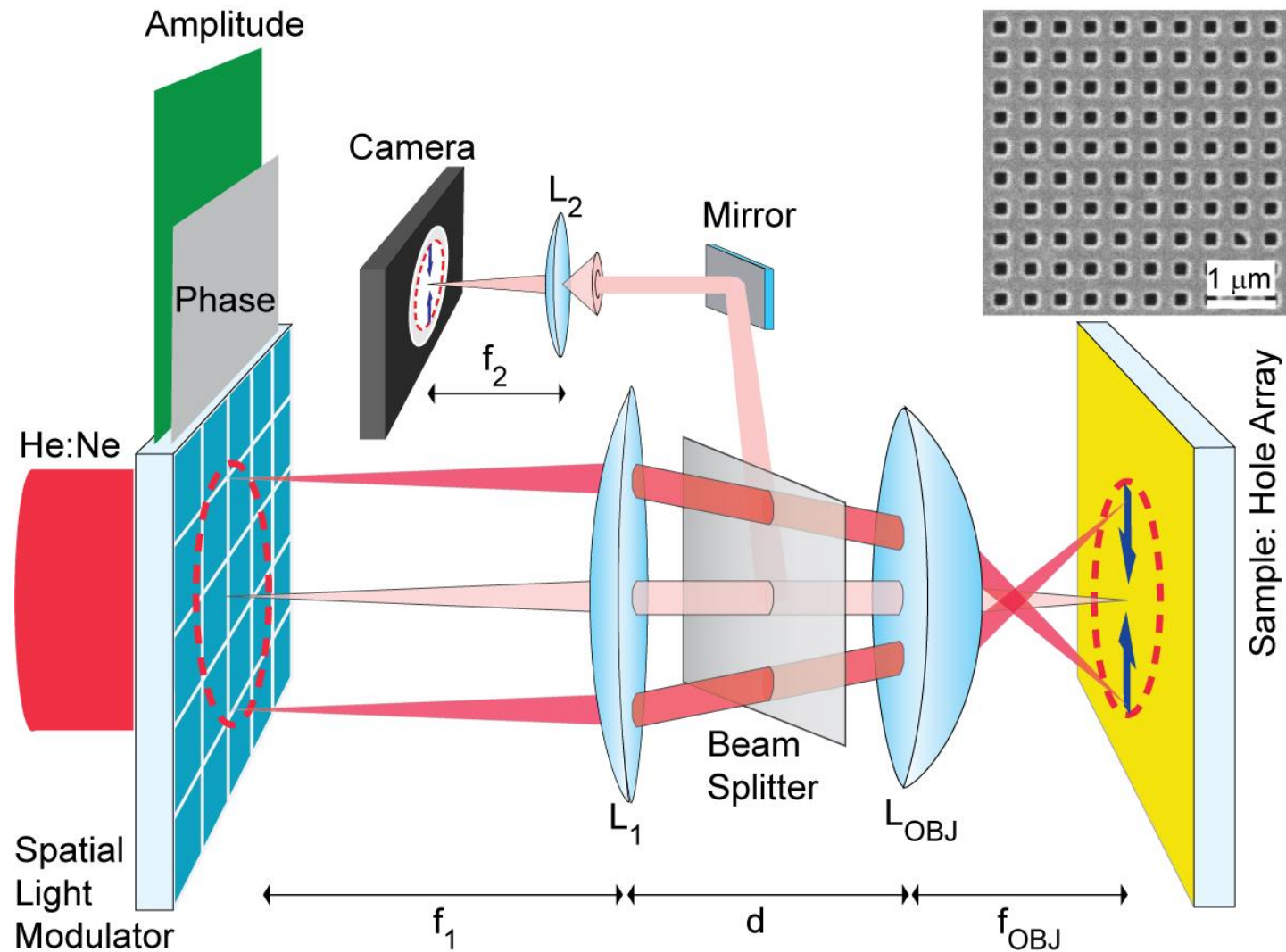
- Same energy (color)
- Shorter wavelength
- Momentum mismatch

Plasmonics for microscopy



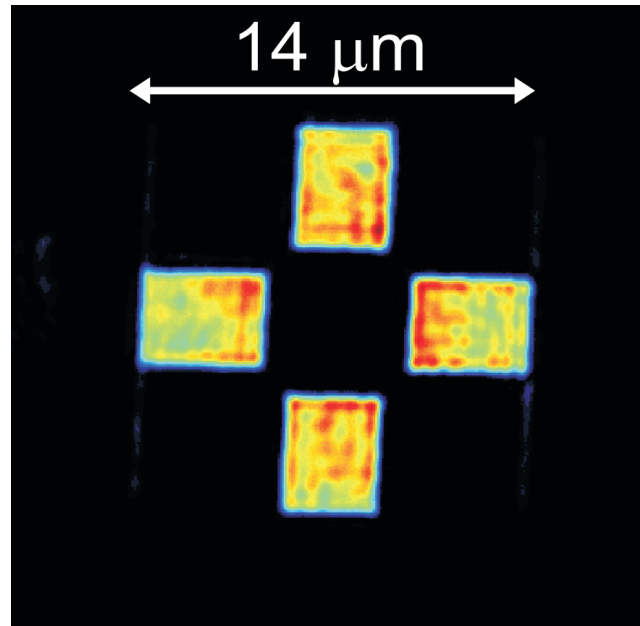
- Shorter wavelength
- Better focusing
- Improved resolution

Plasmonic wavefront shaping

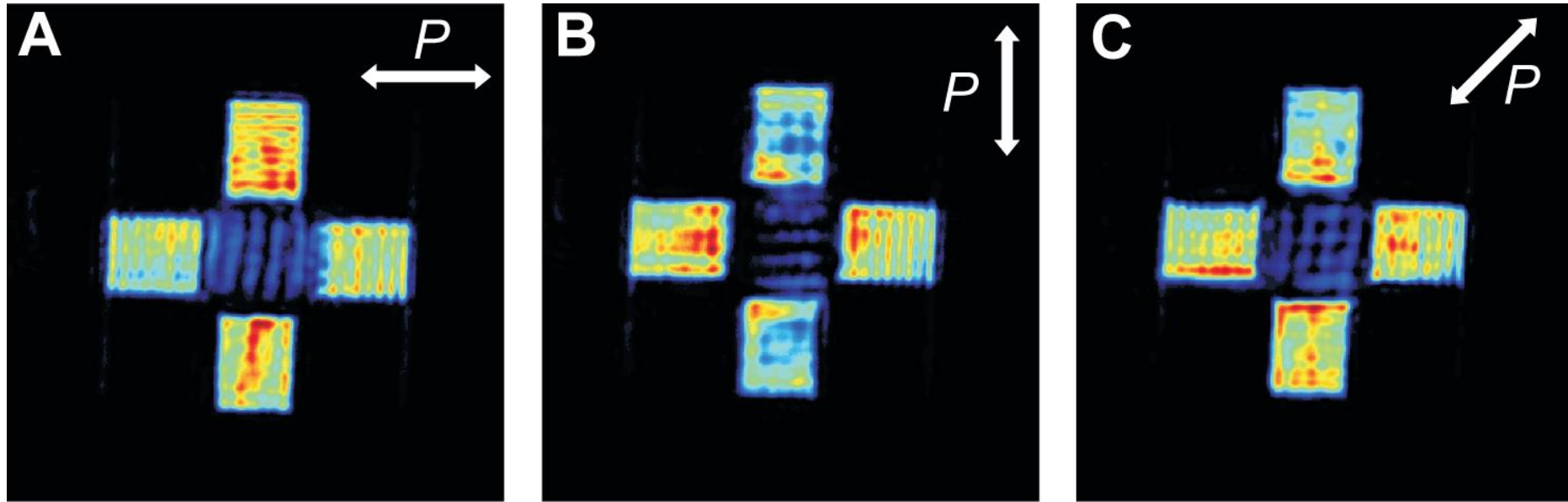


Amplitude design: bare gold

Resolution: 450 nm

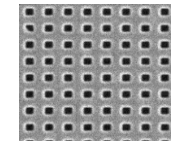
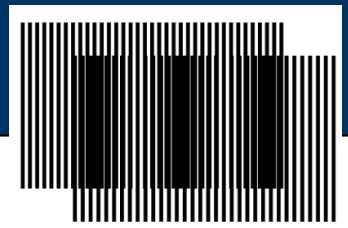


Launching SPPs: hole array

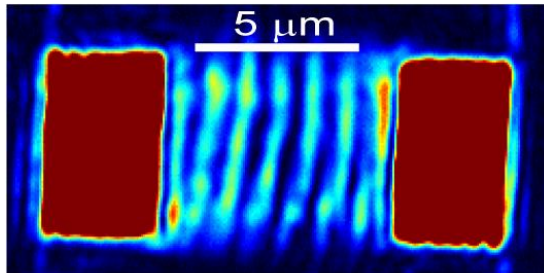


- ❑ SPP plane waves propagate into the SPP arena
- ❑ Polarization dependency
- ❑ Formation of a standing SPP wave

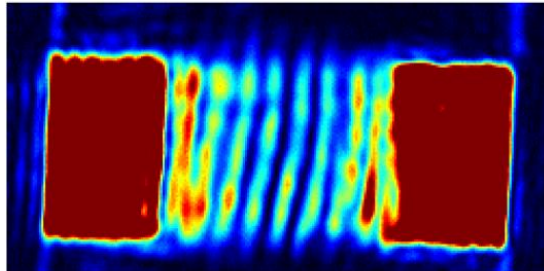
Fringes and gratings



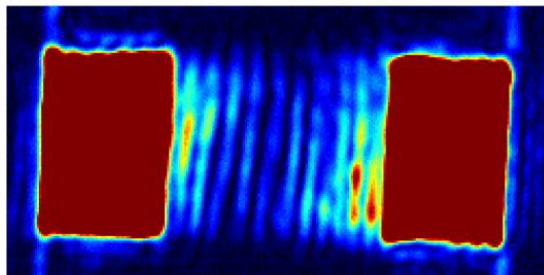
450 nm



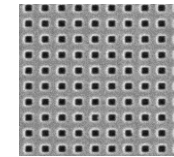
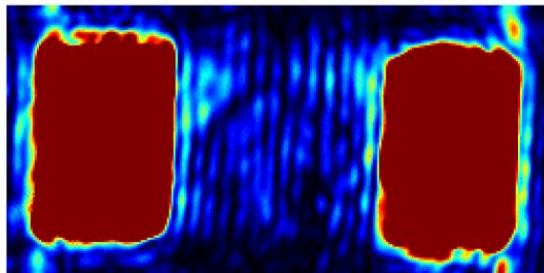
425 nm



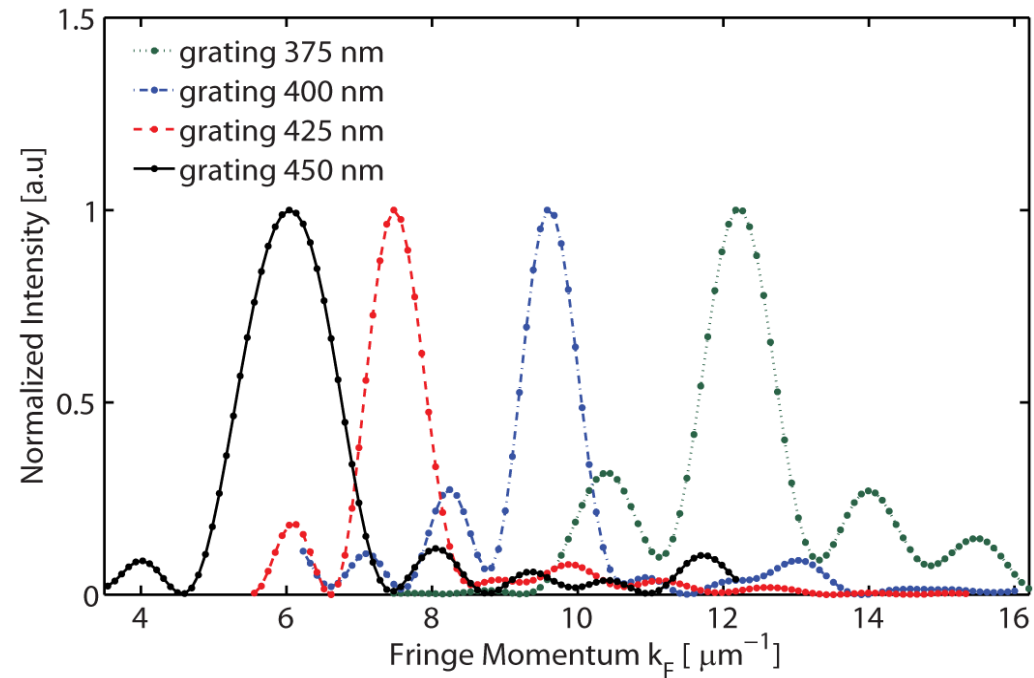
400 nm



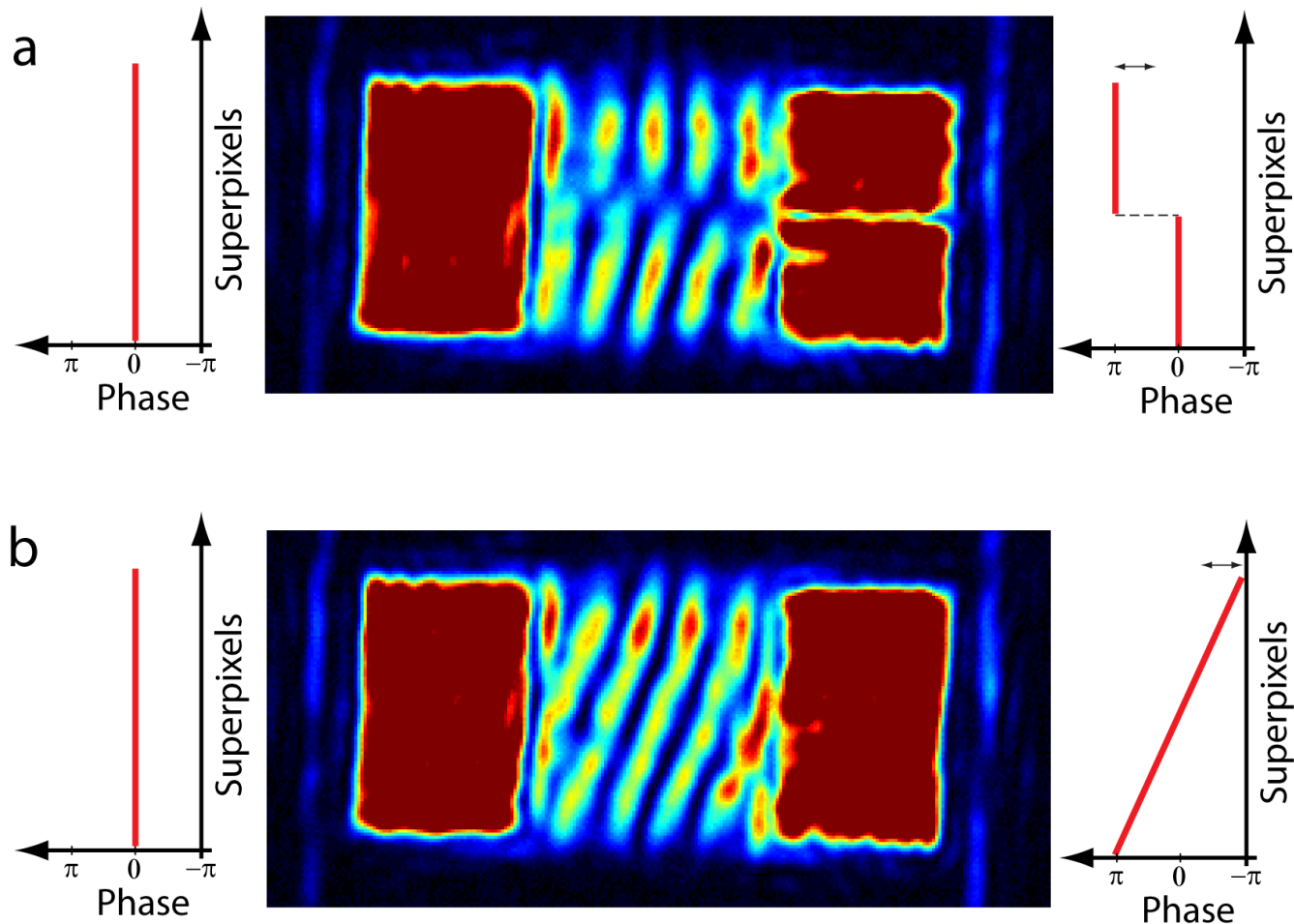
375 nm



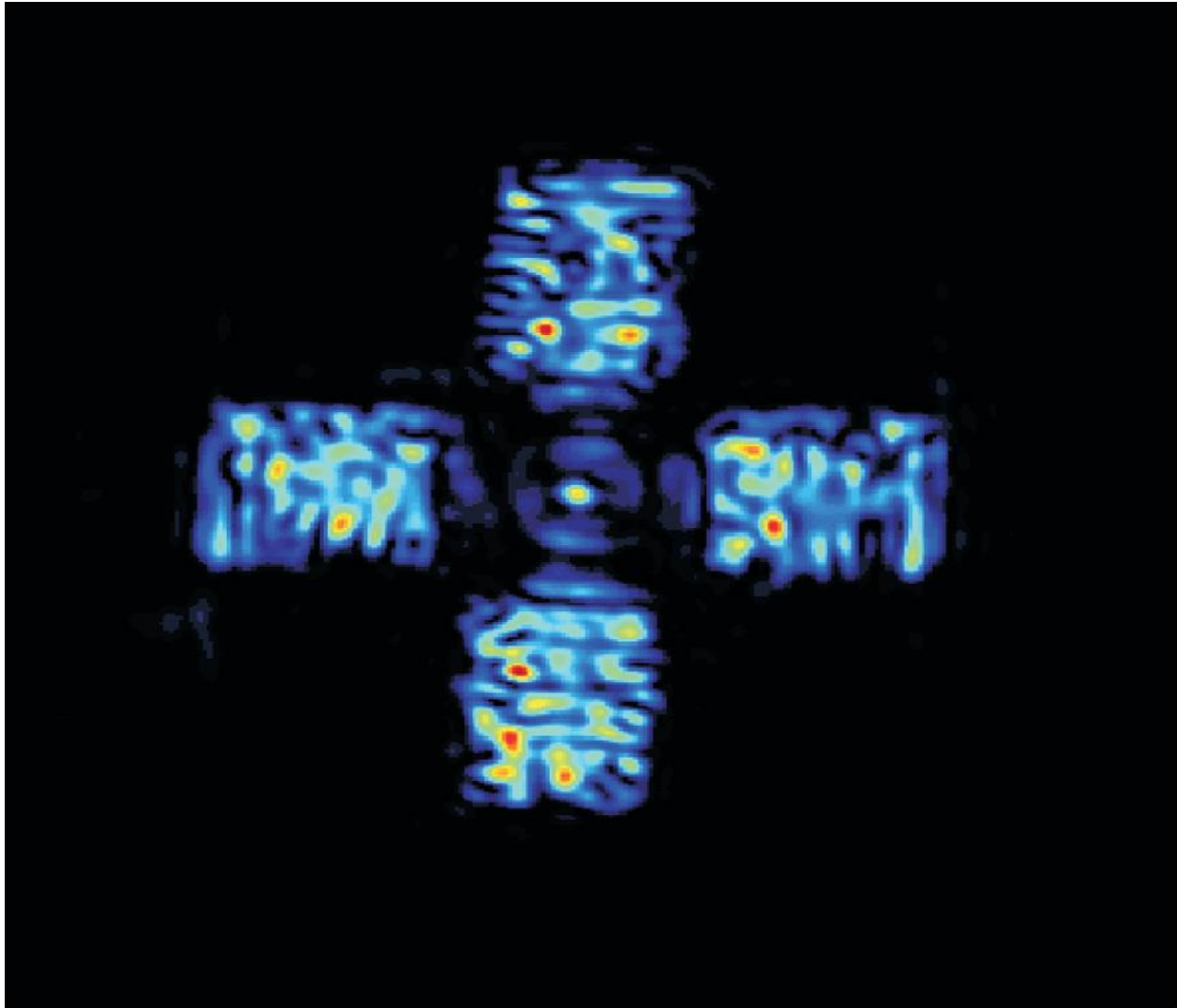
Fringe periodicity = $0.5 * \lambda_{SPP} = 300 \text{ nm}$



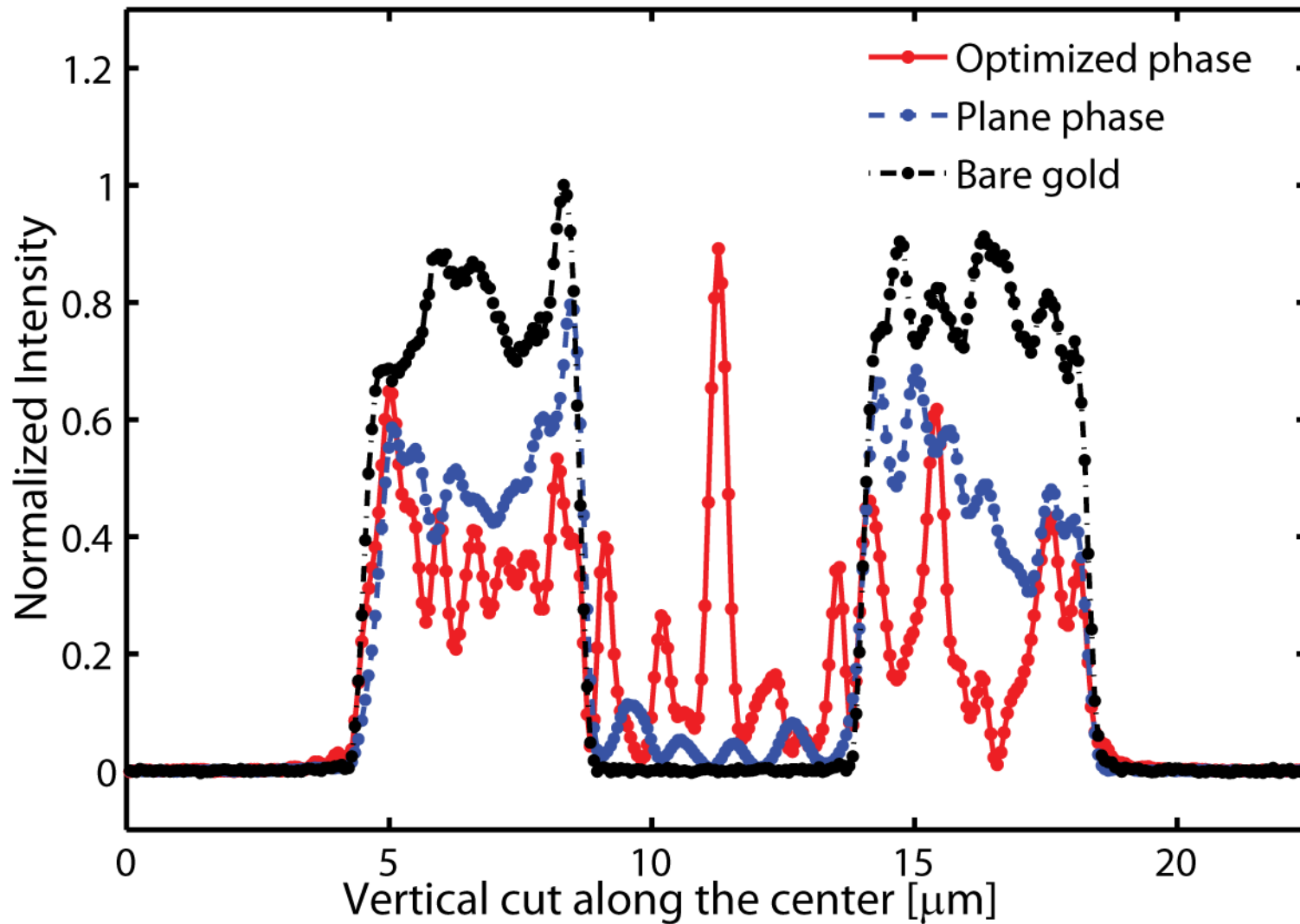
Shift & tilt of the fringes (SSIM)



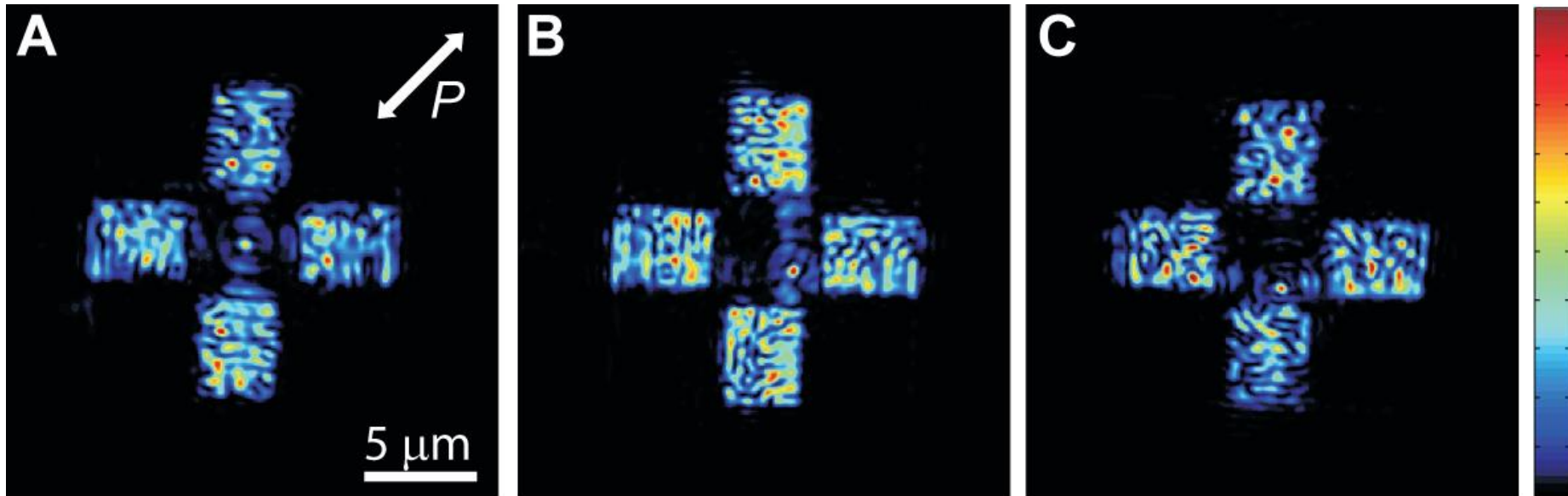
SPP focusing



The SPP focus (with feedback)

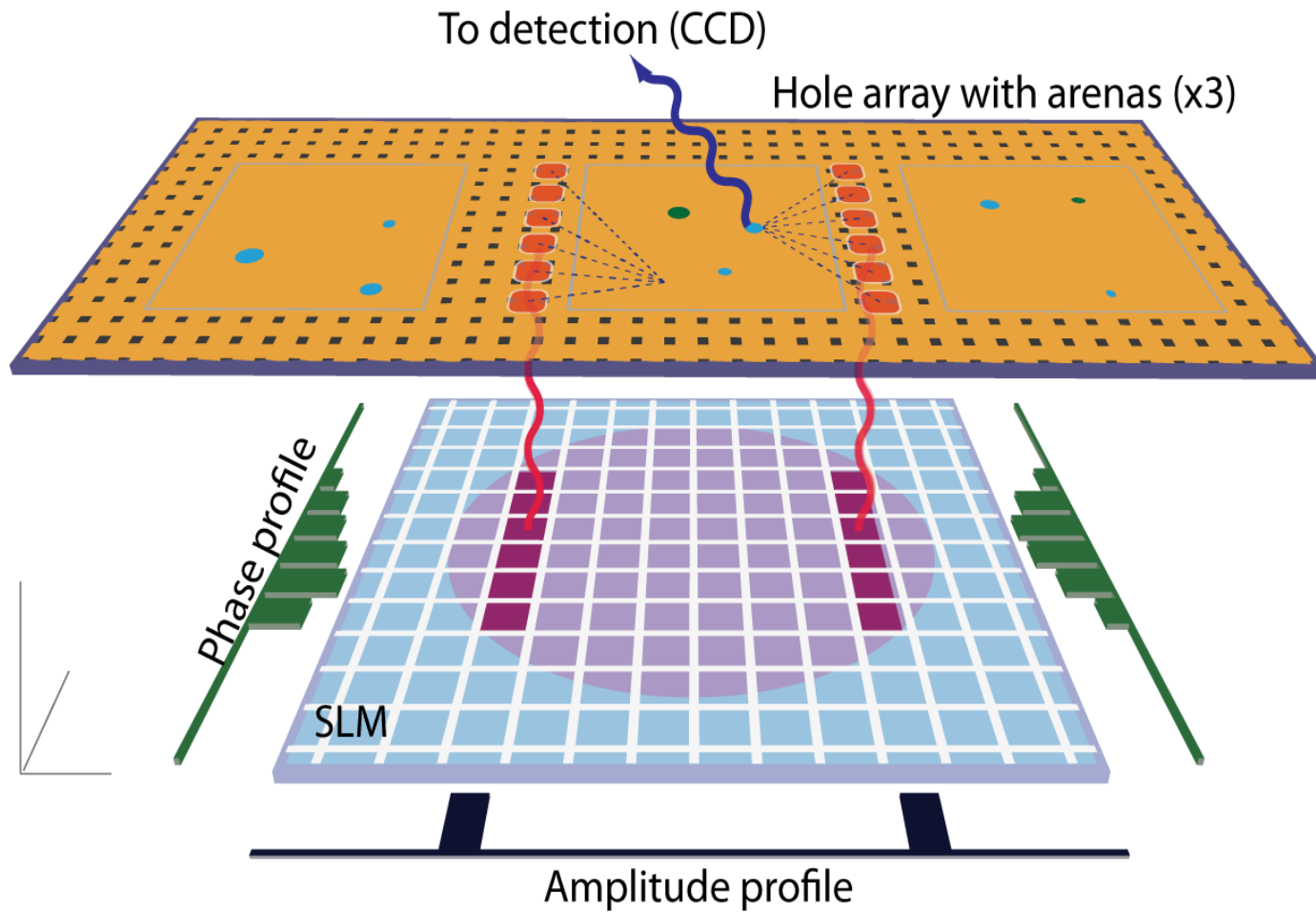


Relocating the focus

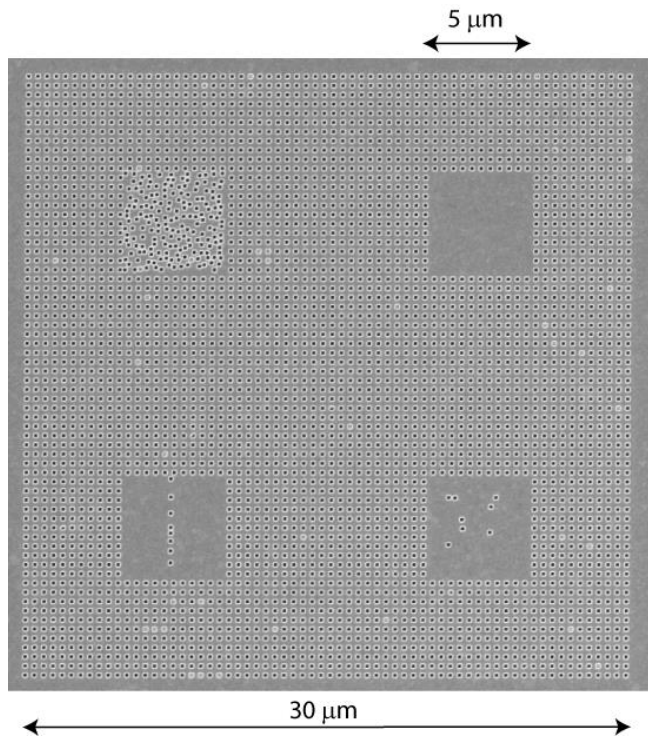


◆ **Everywhere inside the SPP arena**

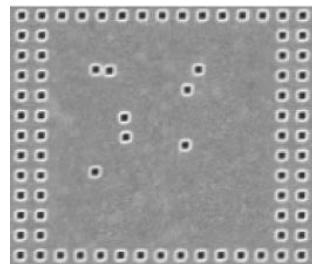
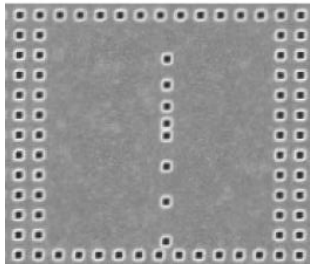
Implementation: microscopy



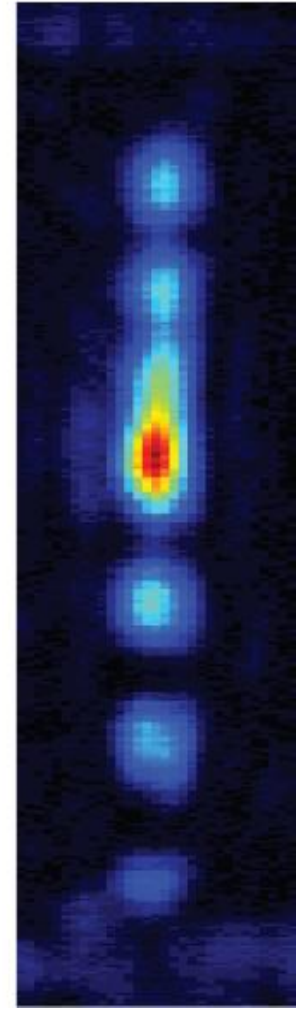
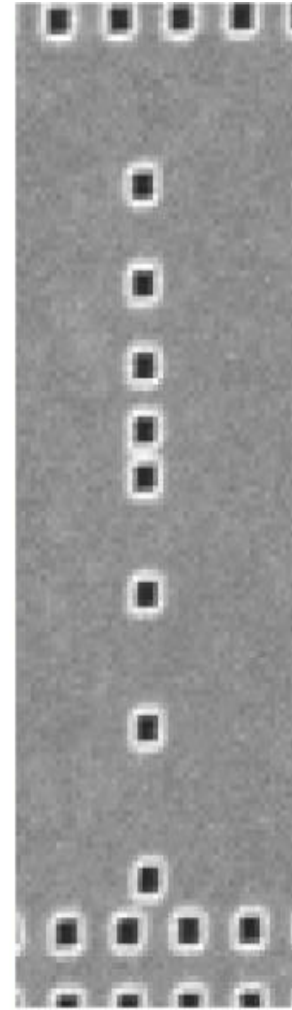
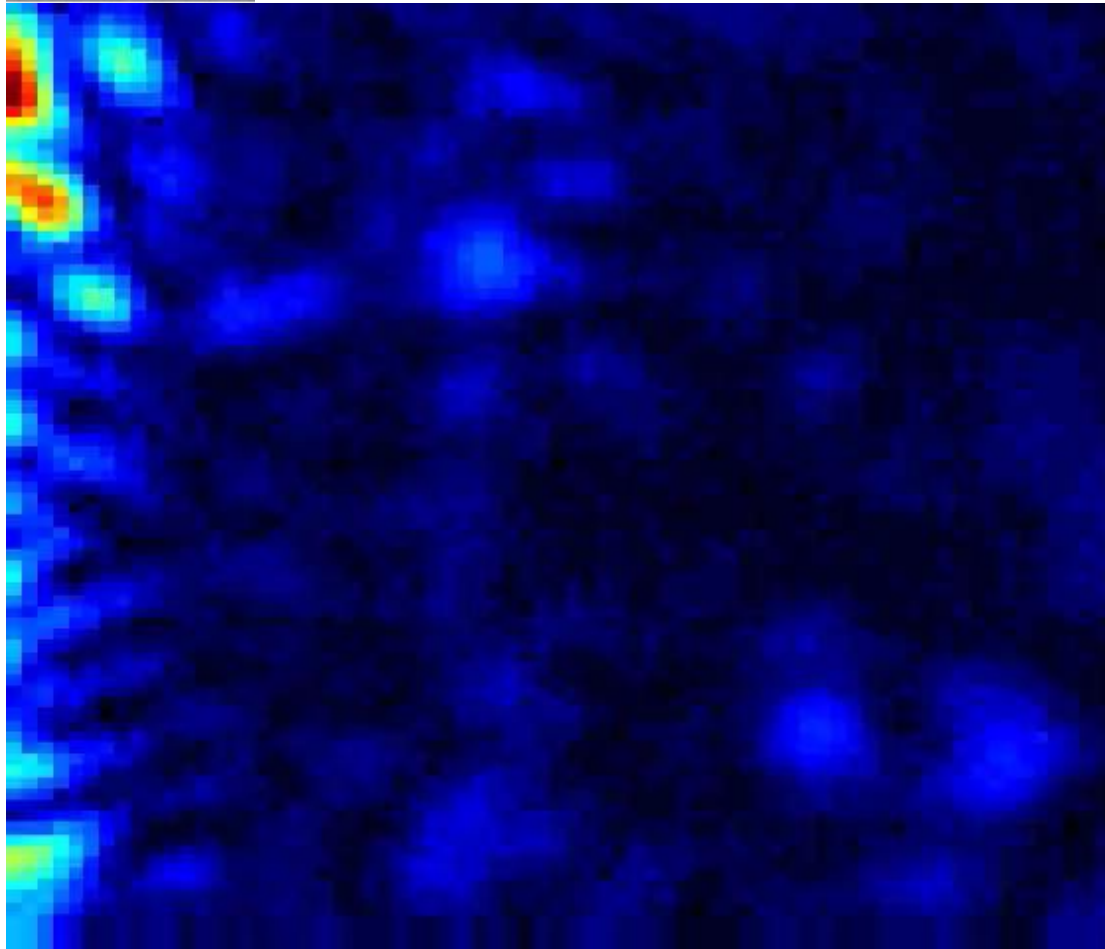
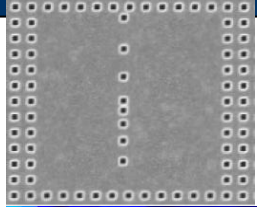
Nanostructured microscope slide



- ◆ Hole grating : SPP launching
- ◆ Bare gold arenas : no feedback
- ◆ Free space GF : calculation
- ◆ Scatterers : to be imaged
- ◆ Metal : functionalize

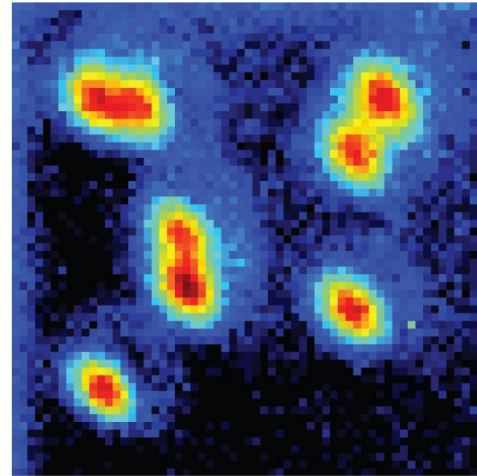
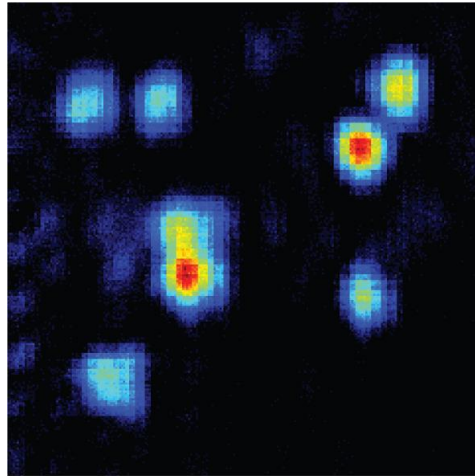


Concept – Scanning the focus

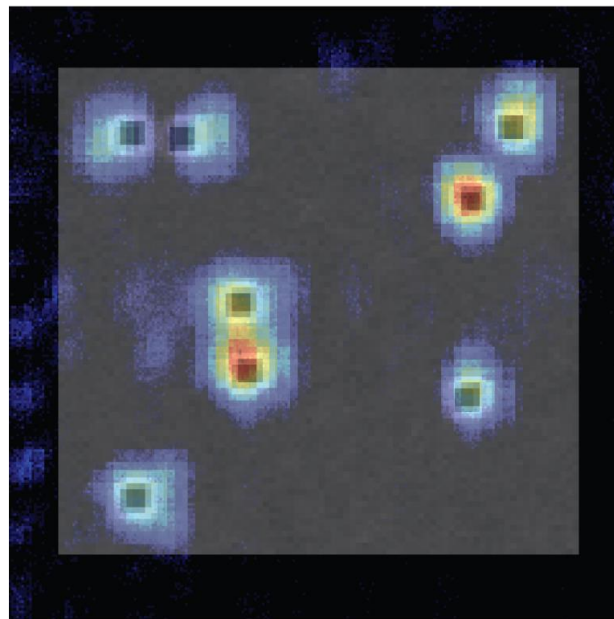
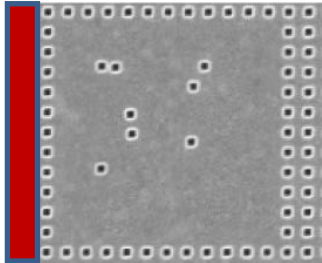


Plasmonic 2D imaging

Focusing
and
scanning

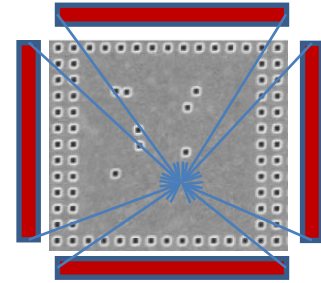
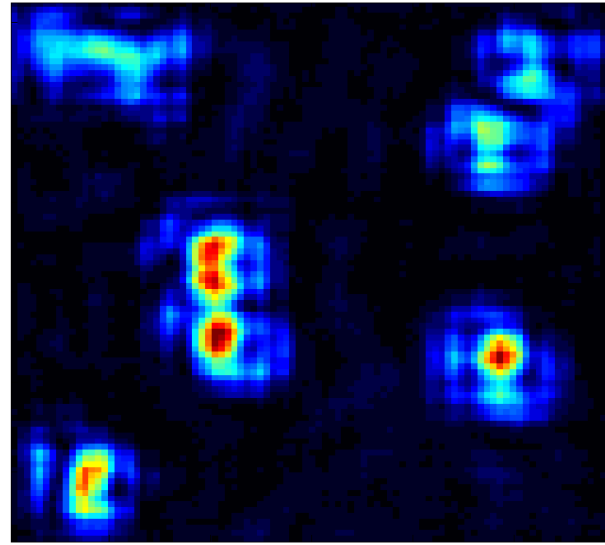
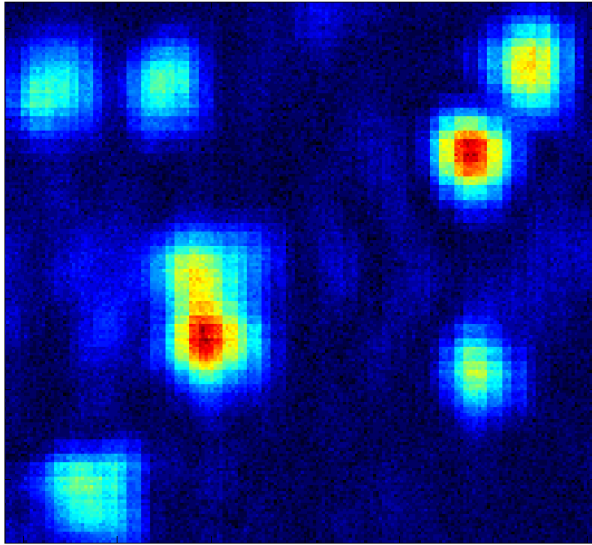
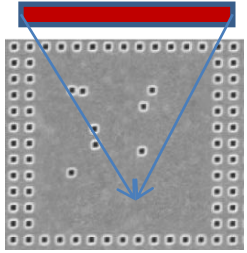


White light
illumination



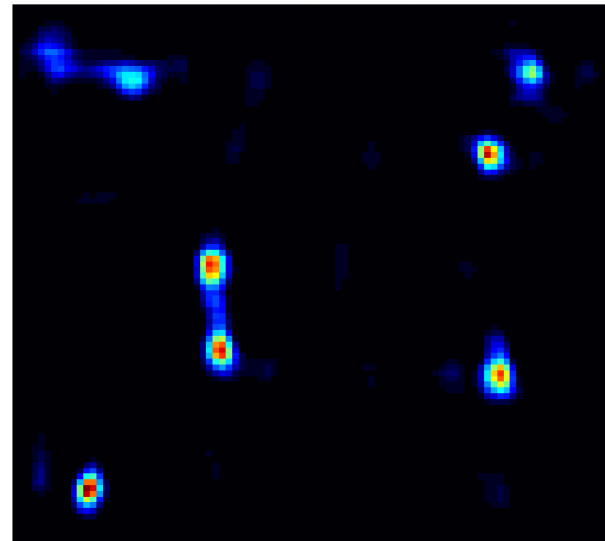
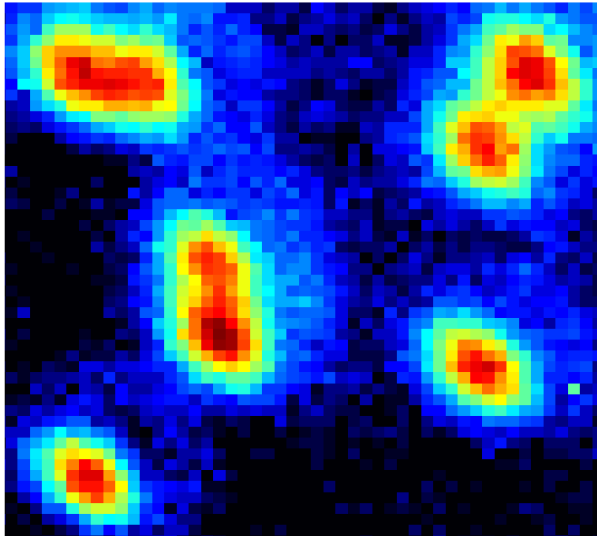
Overlap with
SEM image

Enhancing the focusing/resolution



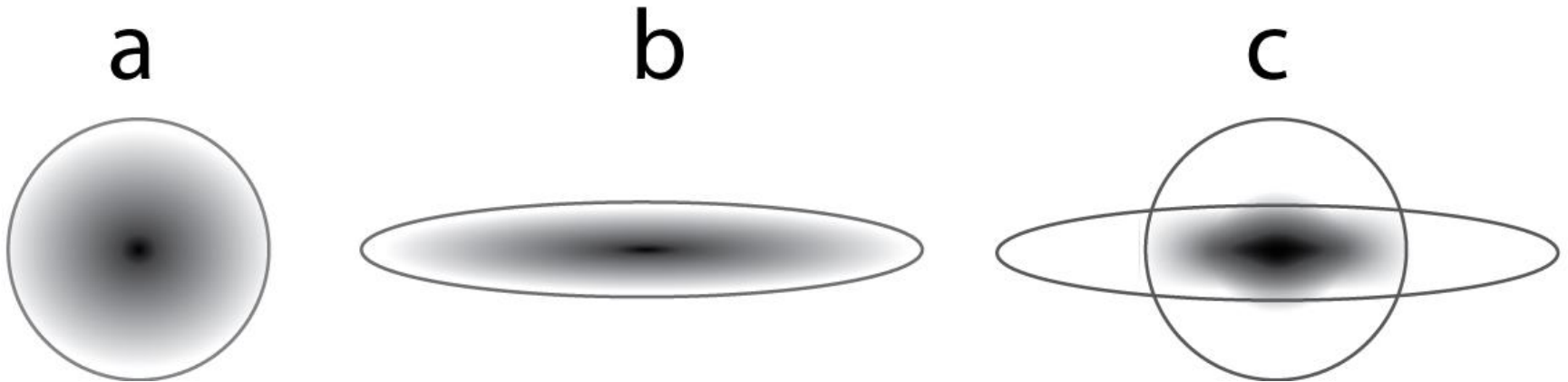
Deconvolution

White light illumination

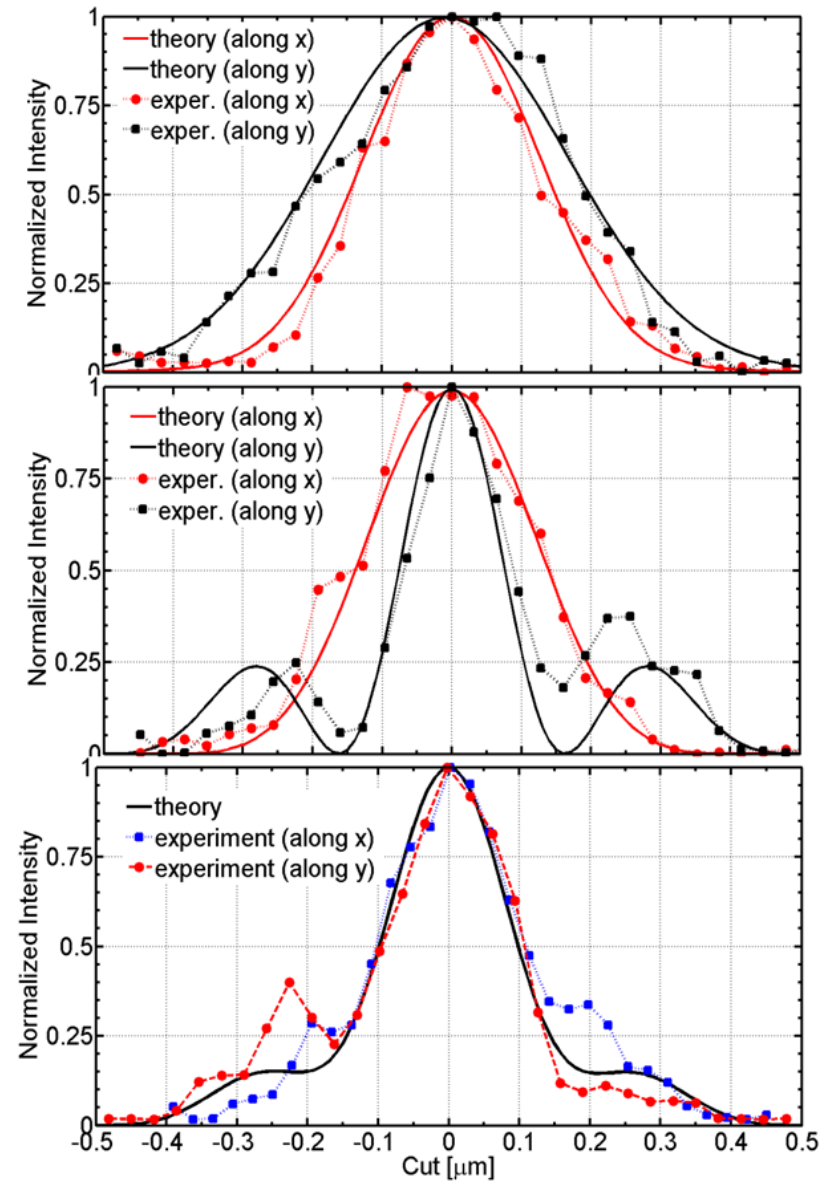
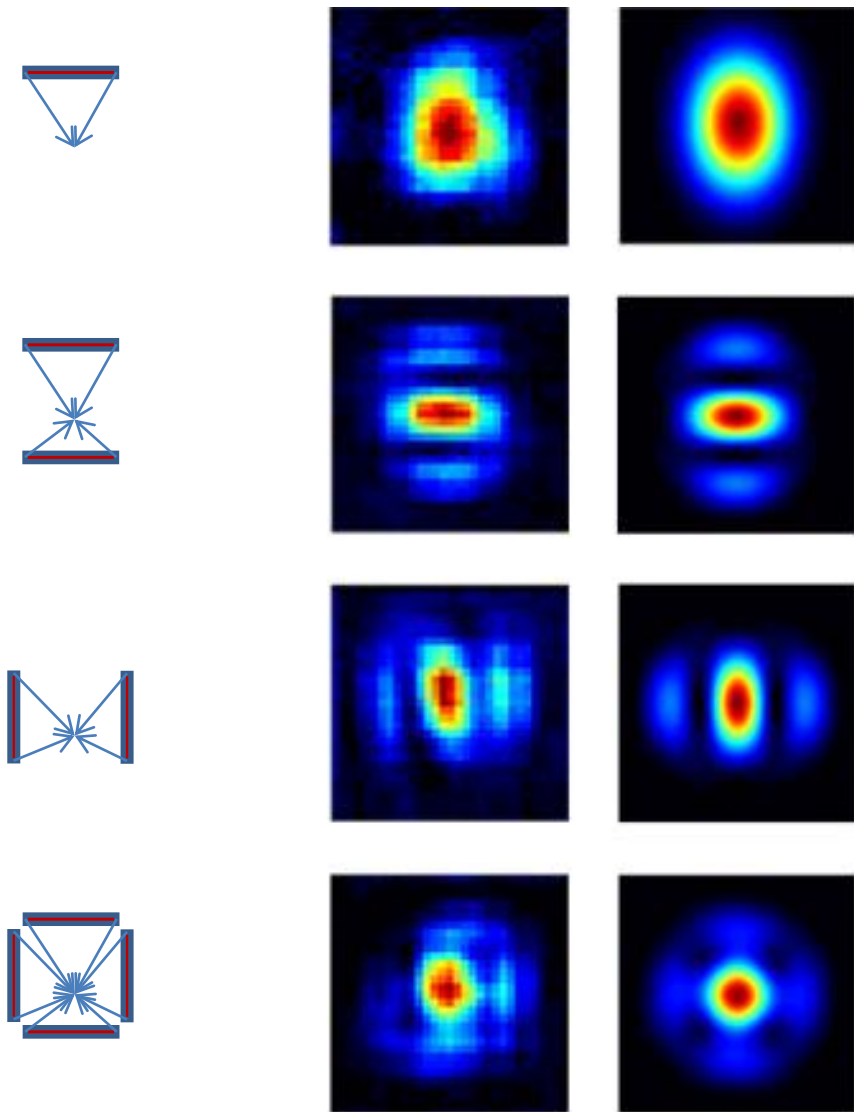


Possible read-outs

- ◆ **Single shot** : **detection psf**
- ◆ **Integrated intensity** : **excitation psf**
- ◆ **Pixel in the focus** : **confocal psf**



Imaging point spread functions



Reorganizing our thoughts

□ Concept

- Simple – scanning a focus to achieve imaging
- Nice – does not require labeling
- Competitive – delivers energy locally (nano-spectroscopy)

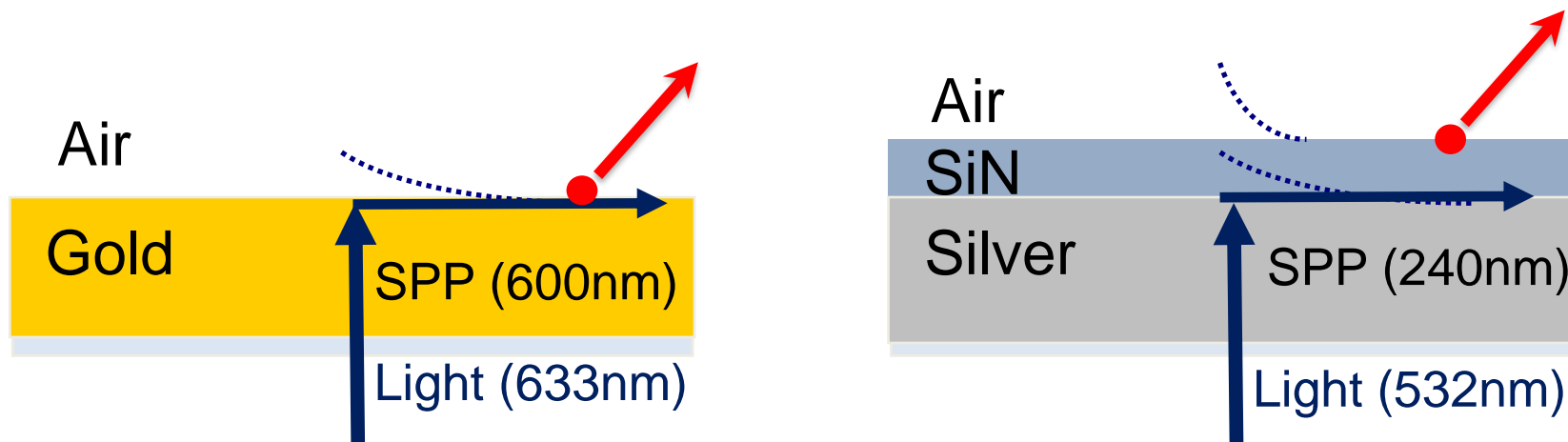
□ Resolution

- Lensing – From 450nm objective to ~ 200nm. Great
- Wavelength – From 633nm to ~ 600nm plasmonic. ($n=1.1$)
- Maturity – STED, PALM, STORM ~ 20nm. Unfit

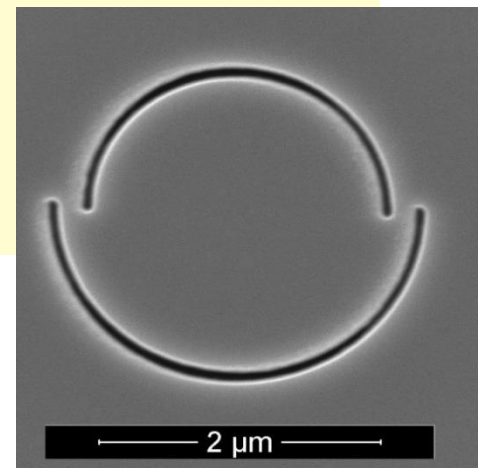
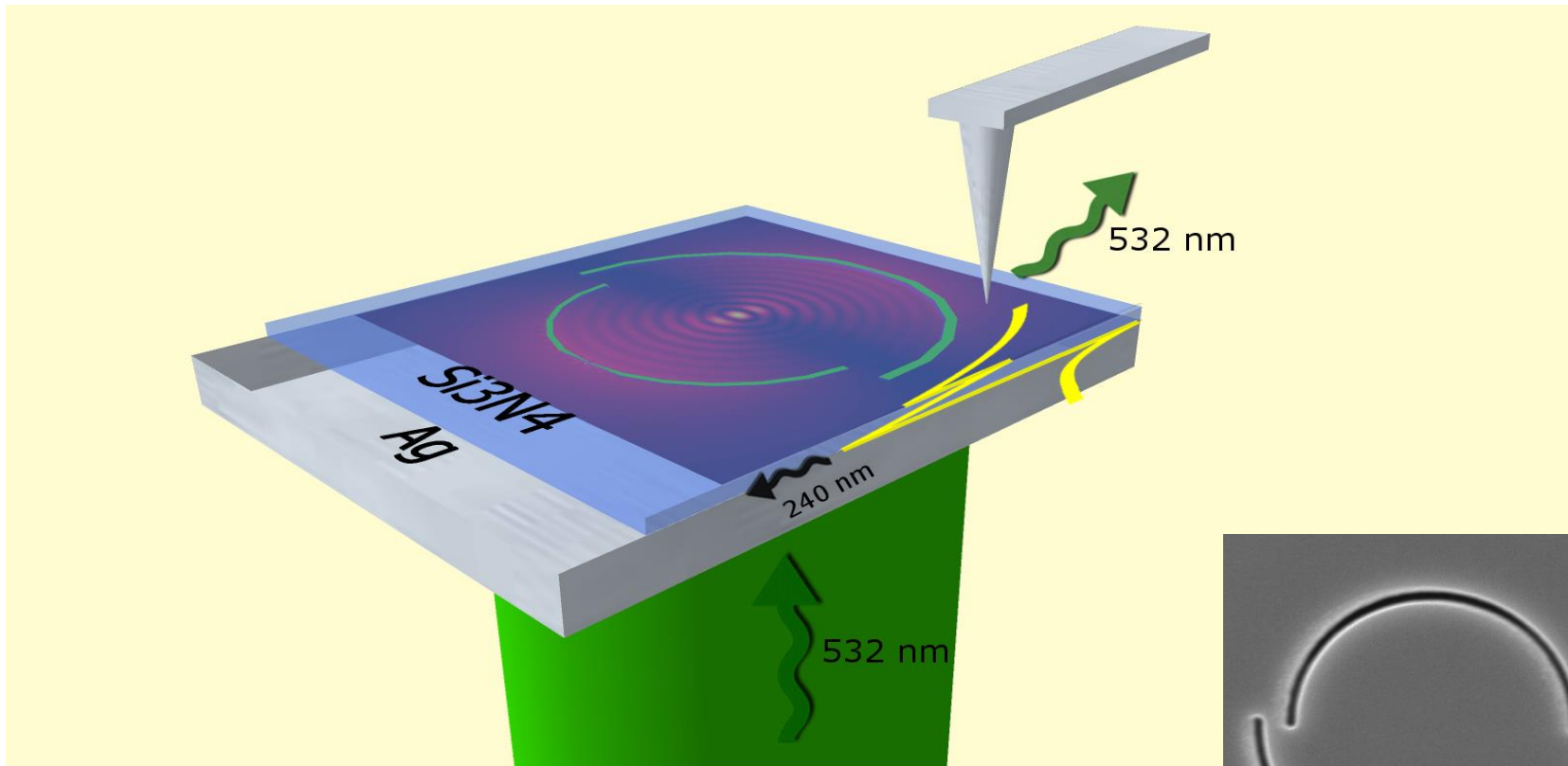
Improved waveguides – Si Nitrite

□ Next: New nanophotonics platforms

1. Wavelength – From 633nm to ~ 180nm. (**$n=3.5$**)
2. Losses – Propagation length > 1 micrometer.
3. Interaction – Should excite a probe or a bio molecule

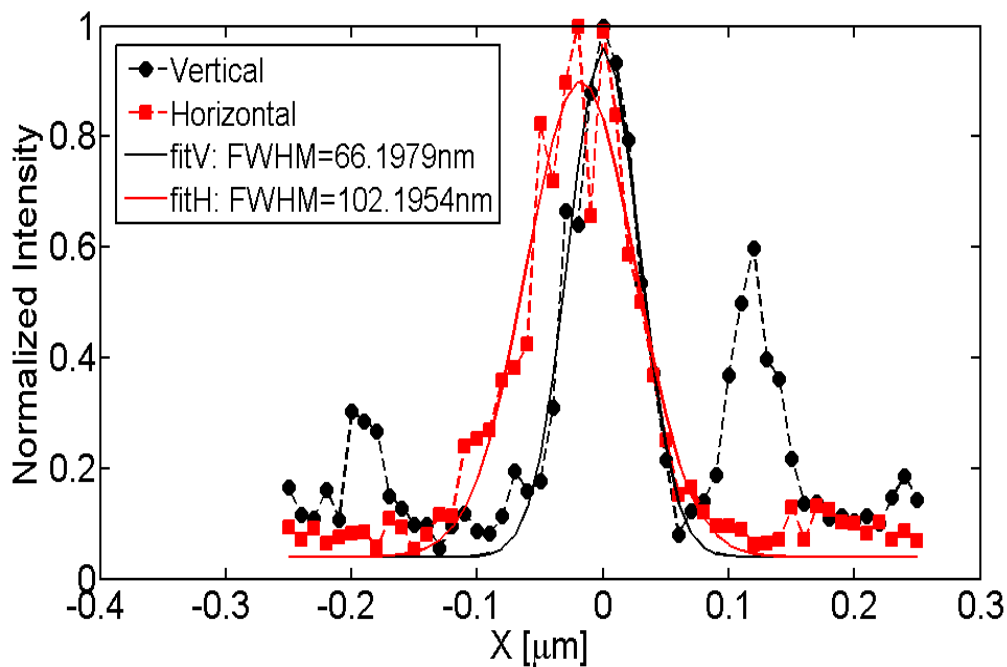
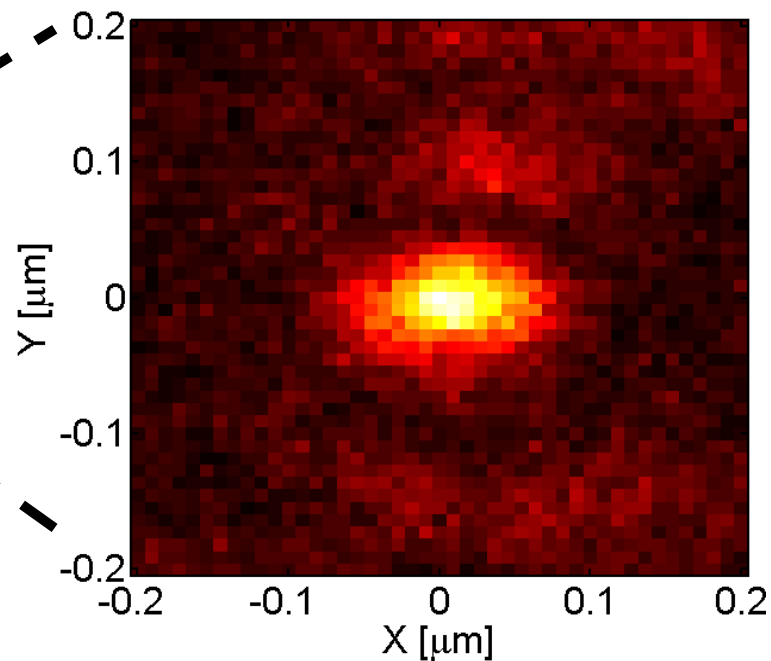
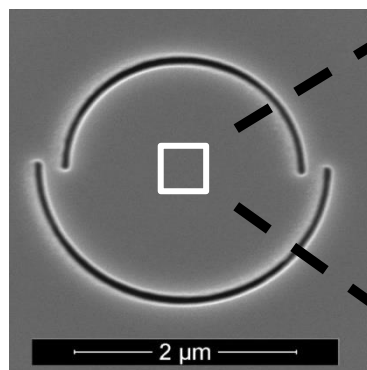
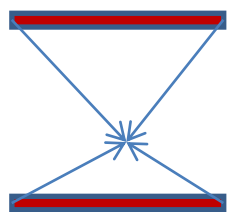


Near field measurements

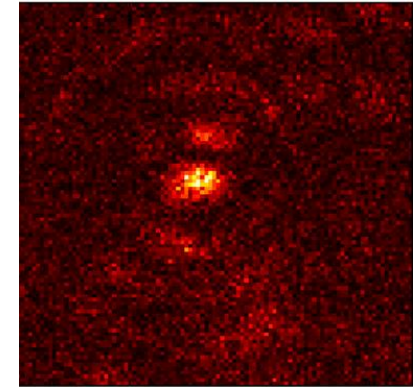
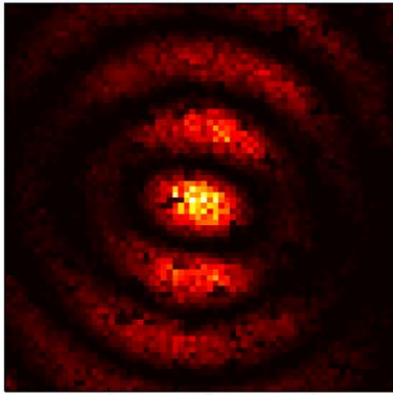


Static focus due to the shape of the slits

Wavelength 240nm; focus 66 nm



Improved waveguides – Si Nitrite



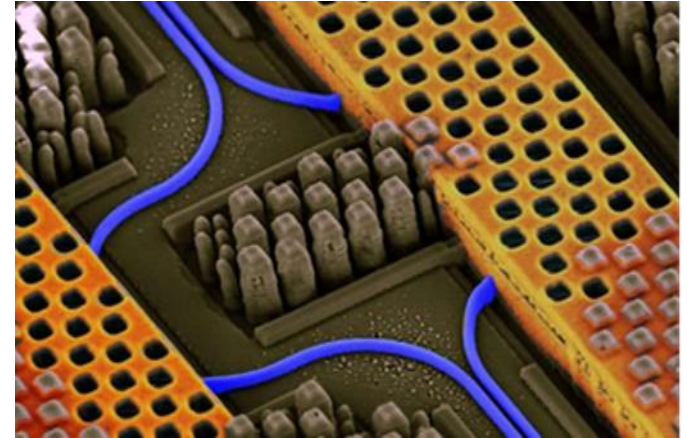
1. Wavelength – From 600nm to ~ 240nm. Almost there
2. Losses – Propagation length < 1 micrometer.
3. Interaction – Should excite a probe or a bio molecule

Super-resolution in Silicon domain

❑ Bulk Silicon

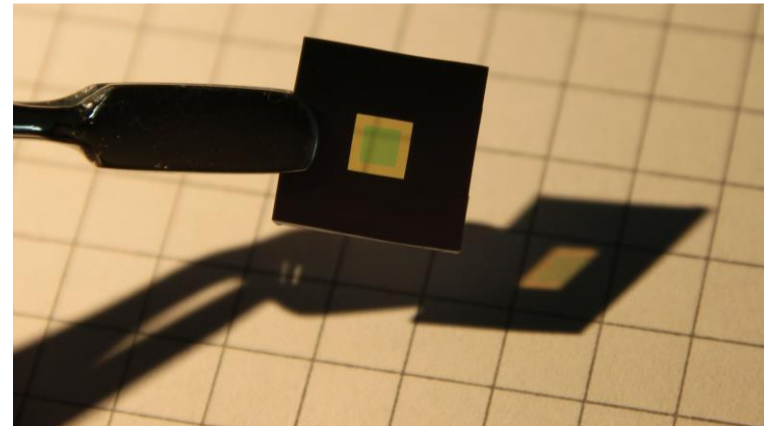
- Refractive index ~ 3.5
- Connection to Si Photonics
- Connection to lithography
- Absorption in the visible
- Hard to harvest light

IBM Research

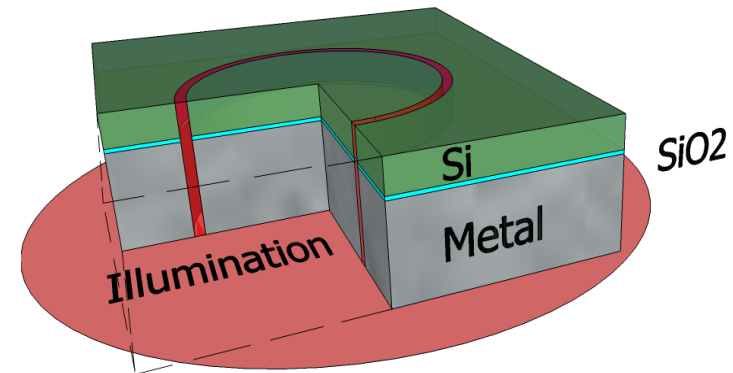
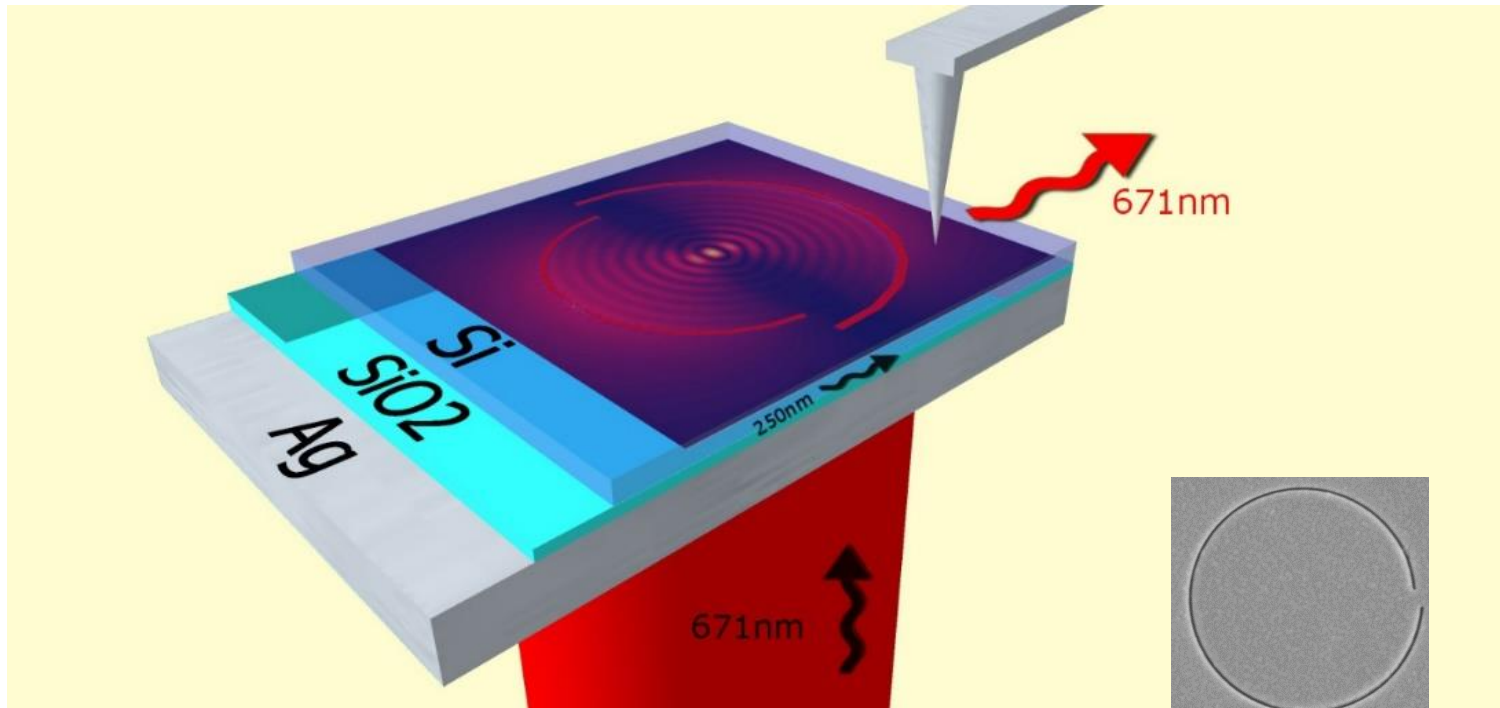


❑ Thin Si waveguide

- Transparent
- Guided modes in 2D
- Propagation up to 10 μm

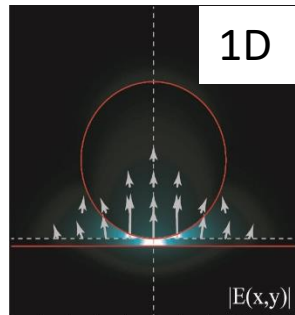


Focusing with Silicon

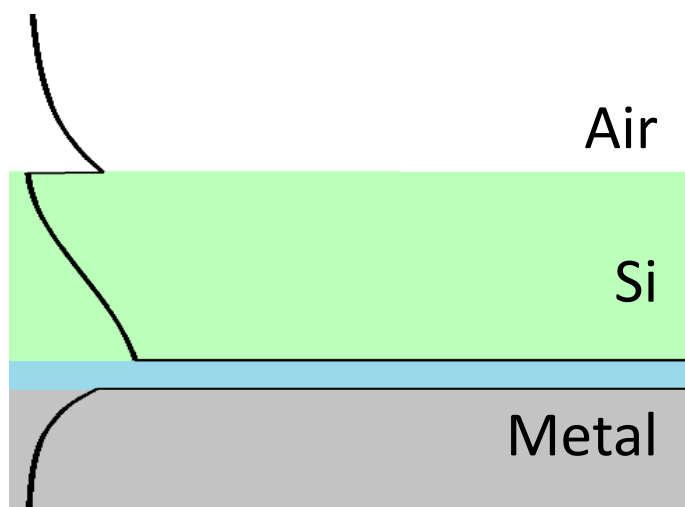


Hybrid photonic plasmonic modes

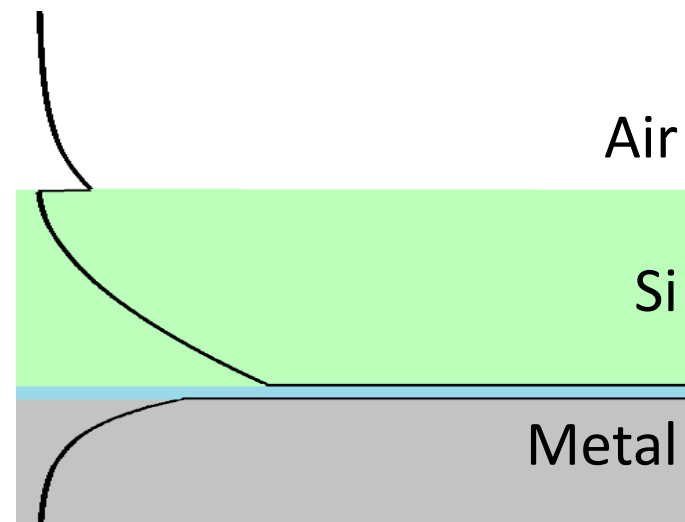
Si - SiO₂ - Ag
60 - 8 - 300 nm



Si - SiO₂ - Ag
60 - 4 - 300 nm



- SiO₂ -



□ Single hybrid mode

- More photonic
- Wavelength 220 nm
- Propagation 5.3 μ m

□ Single hybrid mode

- More plasmonic
- Wavelength 184 nm
- Propagation 3.3 μ m

Resolution: contrast or size?

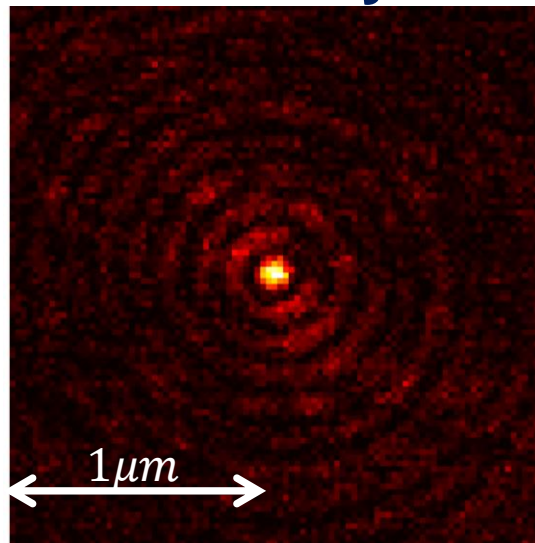
Si - SiO₂ - Ag
60 - 8 - 300 nm

Wavelength 220 nm

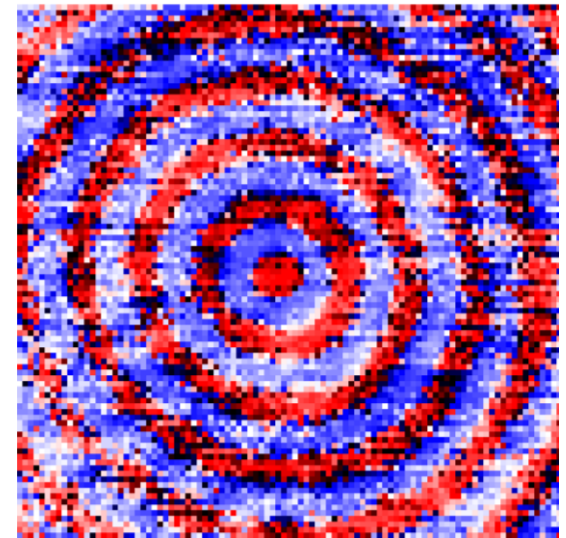
Propagation 5.3 μm

Size (FWHM) 78 nm

Intensity



Phase

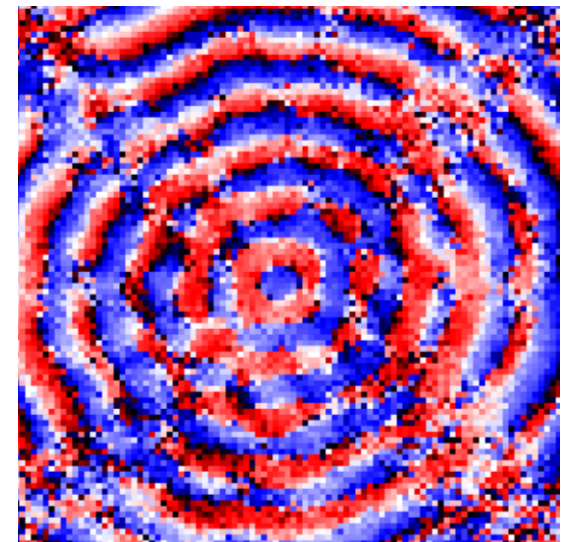
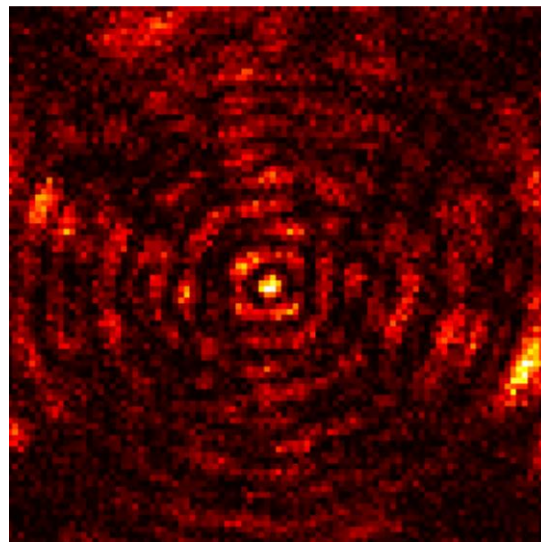


Si - SiO₂ - Ag
60 - 4 - 300 nm

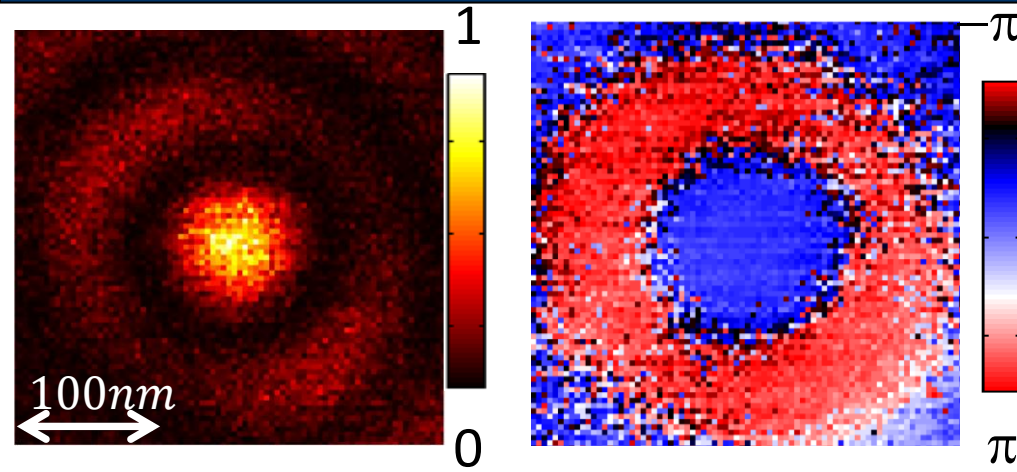
Wavelength 184 nm

Propagation 3.3 μm

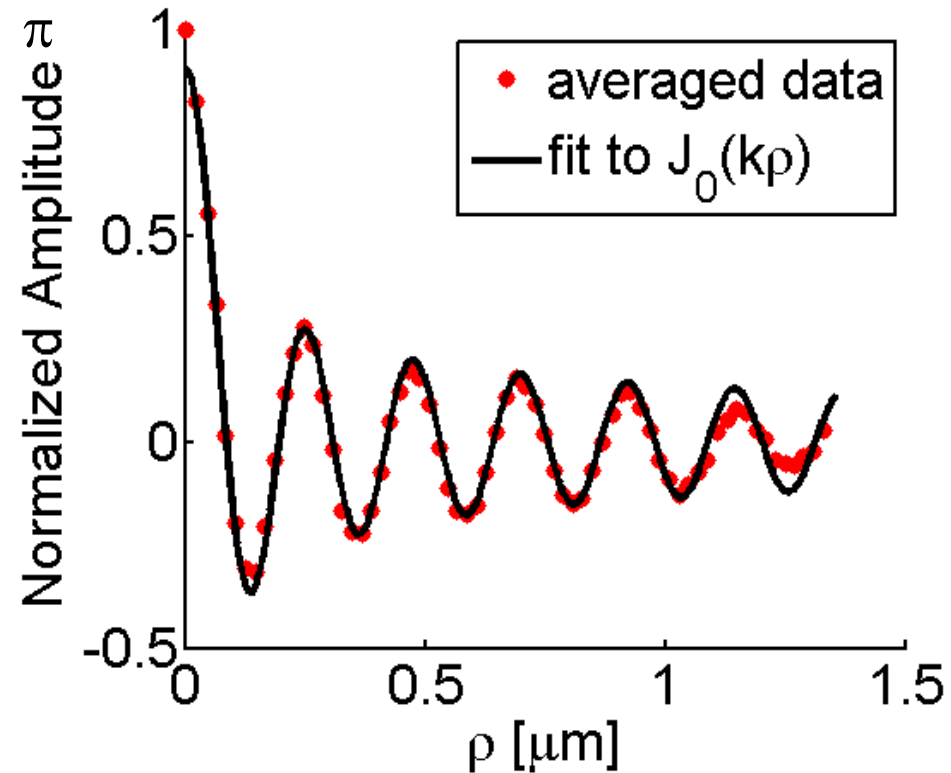
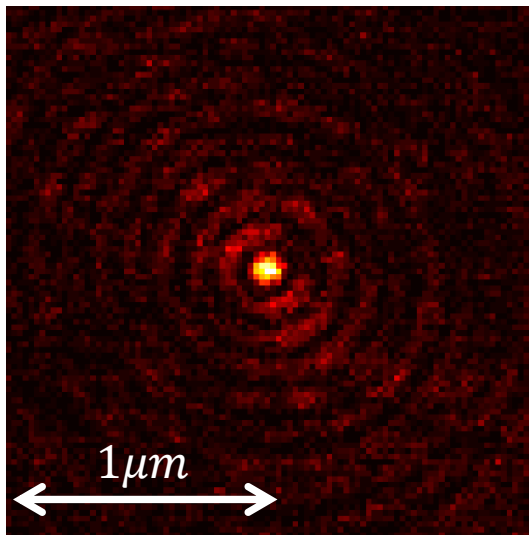
Size (FWHM) 66 nm



Tunability: better contrast focusing

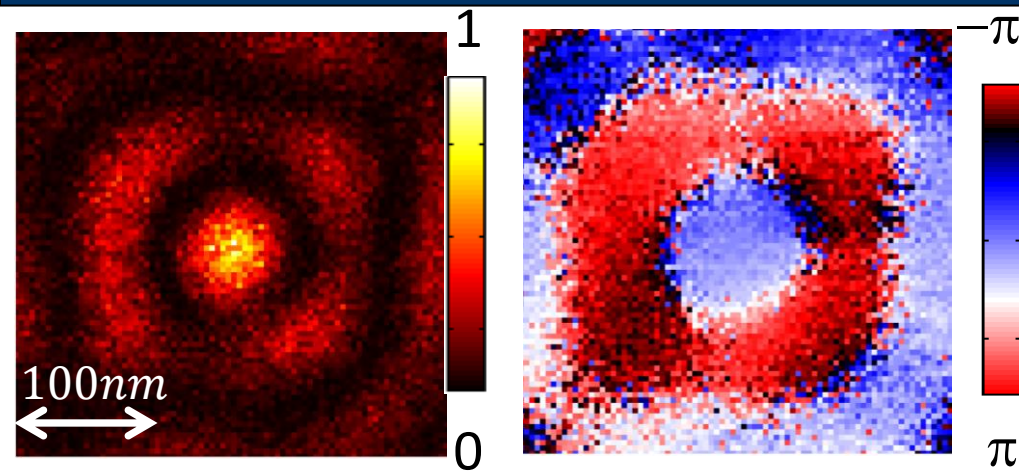


Wavelength **220 nm**
Propagation **5.3 μm**
Size (FWHM) **78 nm**

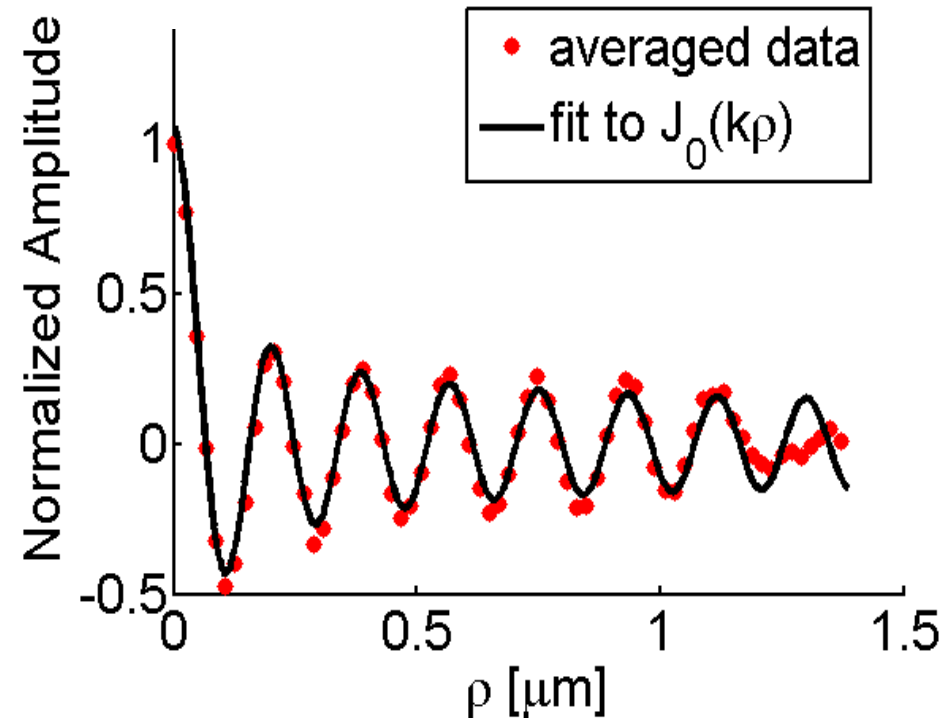
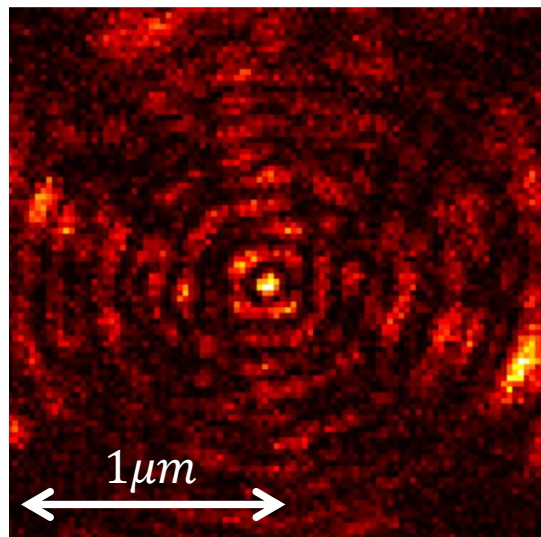


Si	-	SiO ₂	-	Ag
60	-	8	-	300 nm

Tunability: better size focusing



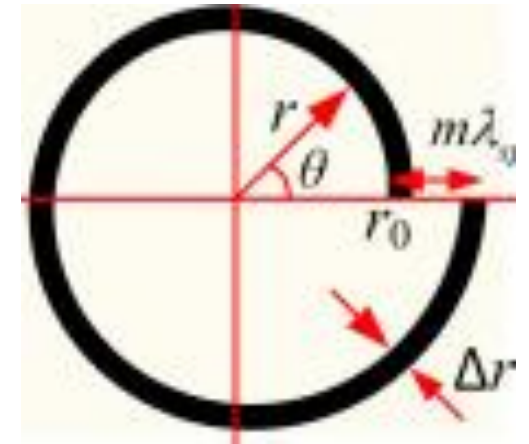
Wavelength 184 nm
Propagation 3.3 μm
Size (FWHM) 66 nm



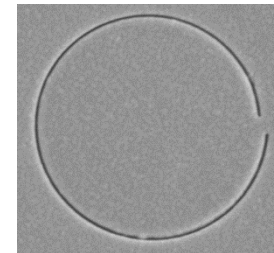
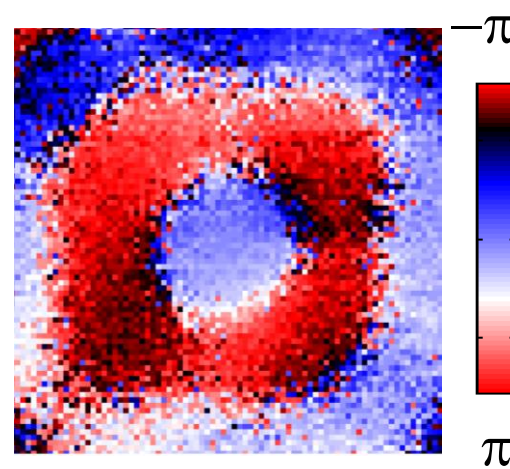
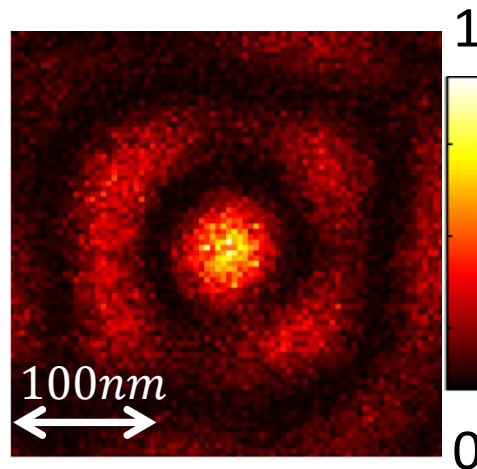
Si	-	SiO ₂	-	Ag
60	-	4	-	300 nm

Orbital angular momentum (spiral)

- **Spiral lens & circular polarization**
 - Angular momentum spiral = -1
 - Angular momentum polarization = 1
 - Net angular momentum $L = 0$



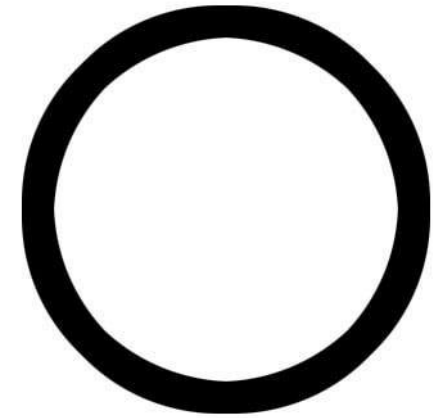
$$E(\rho, \theta) = J_0(k\rho)$$



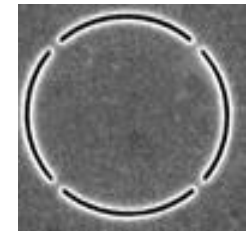
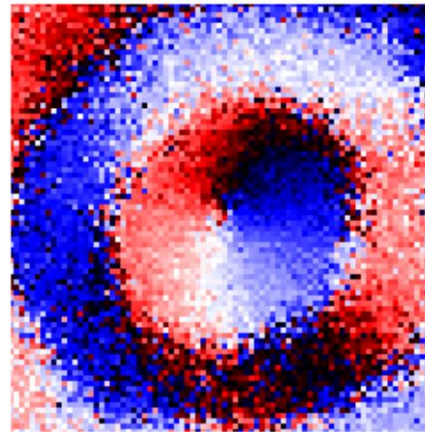
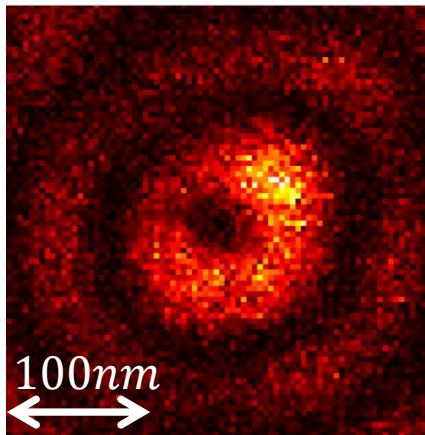
Orbital angular momentum (circle)

□ Circular lens & circular polarization

- Angular momentum circle = 0
- Angular momentum polarization = 1
- Net angular momentum L = 1

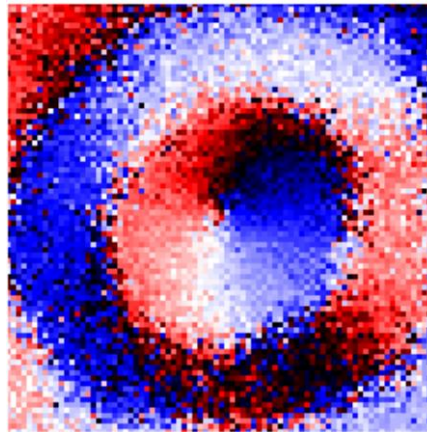
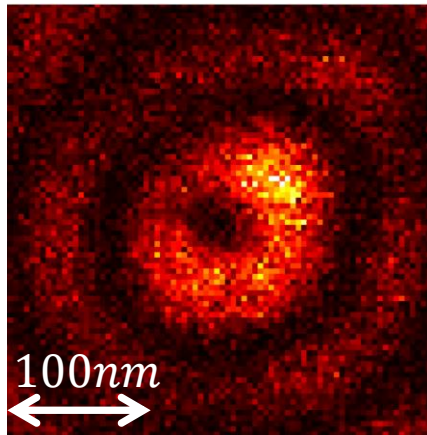


$$E(\rho, \theta) = J_1(k\rho) * e^{i\theta}$$



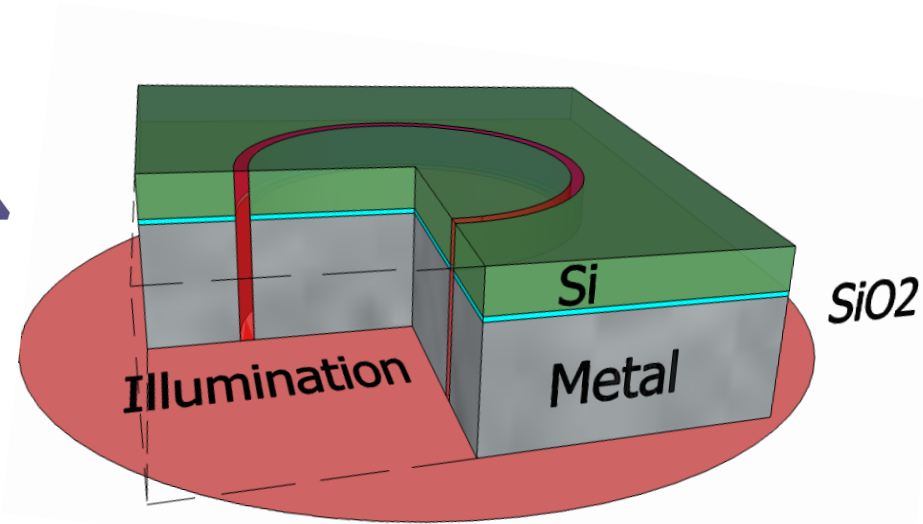
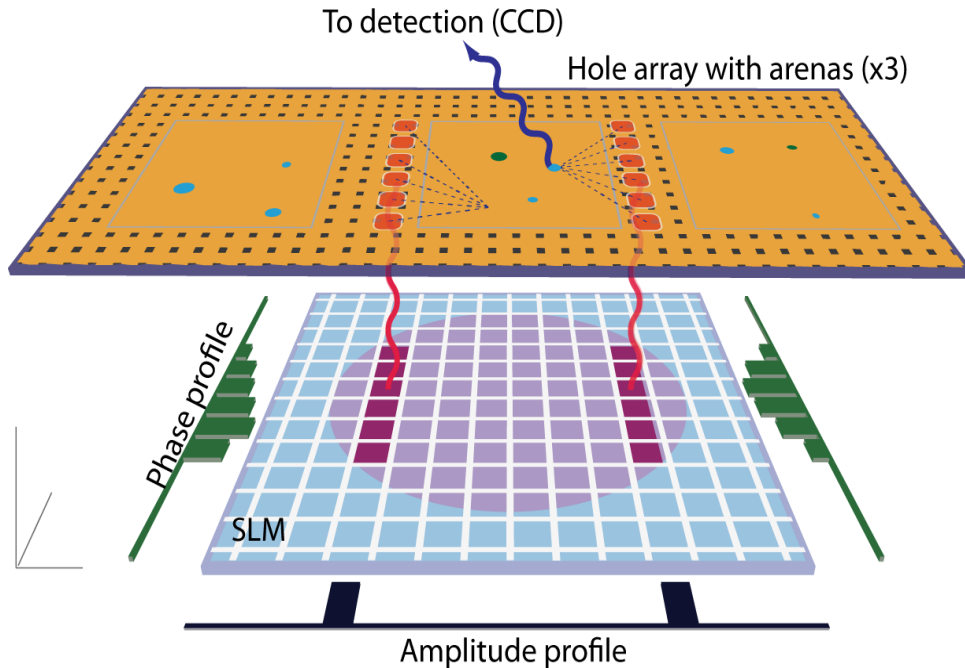
Nano vortex relevance

- ❑ **Optical vortices are well known and used**
 - Hasman, et al Nano Letters (2011)
- ❑ **Nano vortices are new**



- The size (60 nm) is comparable with quantum systems
- For example Quantum Dots
- Beyond the dipole transition
- Accessing new transitions (dipole prohibited)

Joining efforts



□ Active wavefront control

- Raster scanning (focus or vortex)
- Far field microscopy
- Flexibility

□ Silicon waveguides

- Wavelength 184 nm
- Propagation 3.3 μm
- Resolution 66 nm
- CMOS compatibility Al

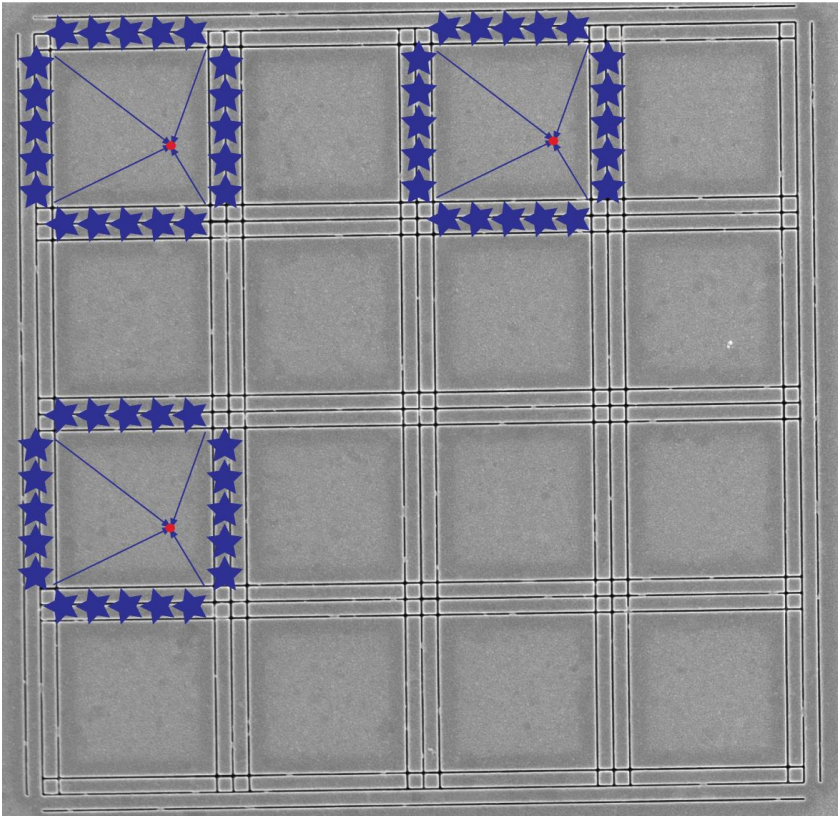
Take home messages

- ❑ **Microscopy is continuously growing**
- ❑ **Nanoscopy is an actuality**
 - Limitations: Fluorecent labelling, invasiveness
 - Open questions : Chips, nanospectroscopy (i.e. Raman)
- ❑ **Wavefront shaping is flexible control**
 - Microscopy by focusing and scanning (label free)
 - Deliver energy suitably for spectroscopy
- ❑ **Resolution is enhanced in Photonic 2D waveguides**
 - Static focusing to ~60 nm with reasonable contrast
 - Tunable by material properties (resolution, losses, CMOS)

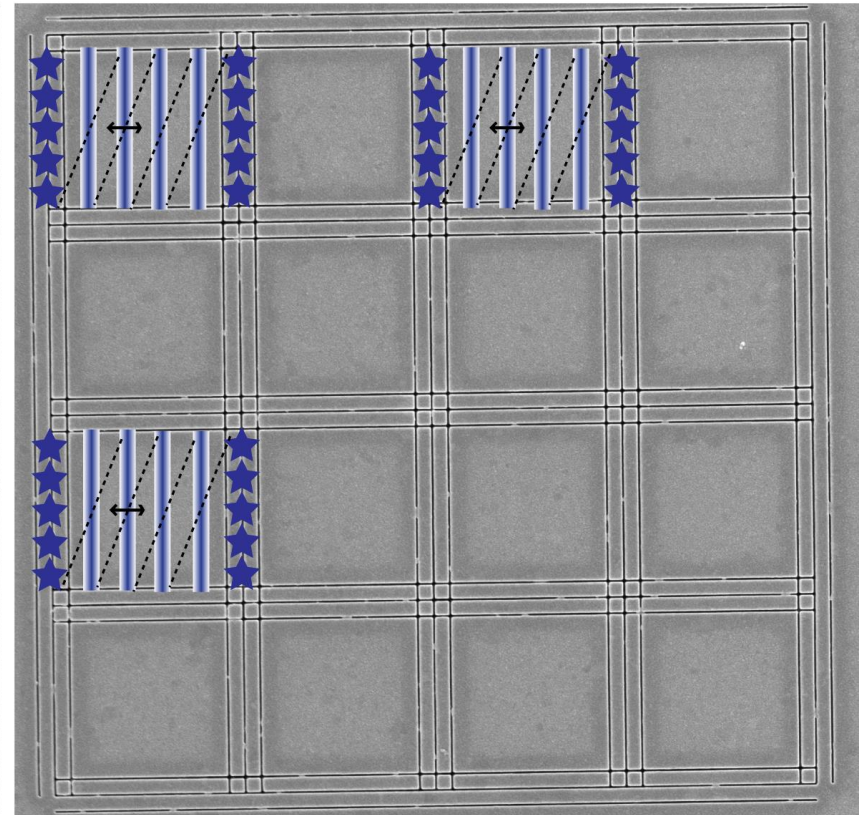
Upgrading an existing microscope

Multiplexed imaging

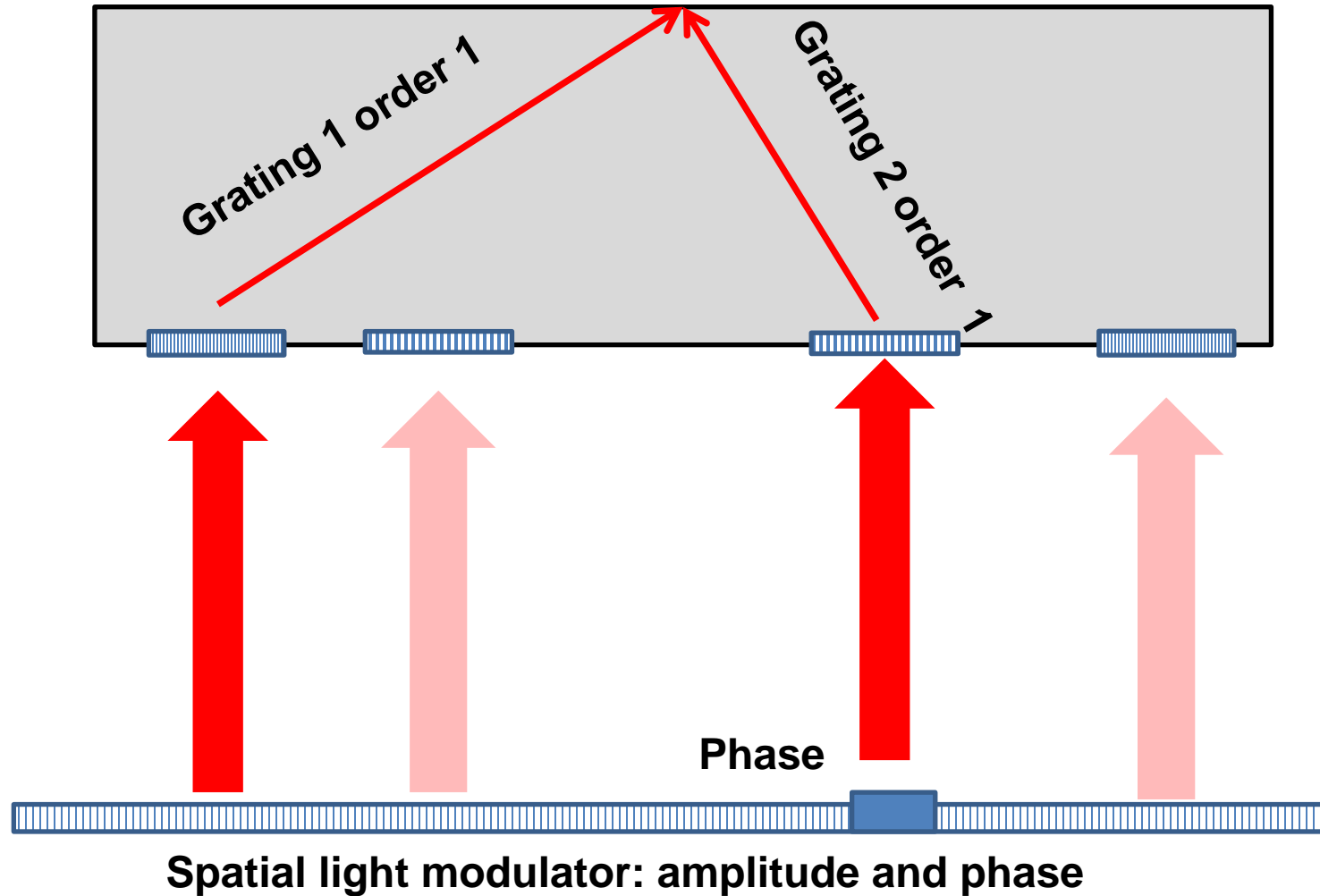
a



b



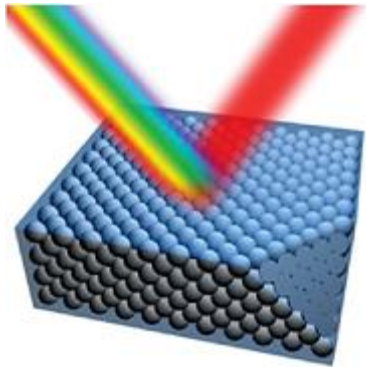
How to achieve it



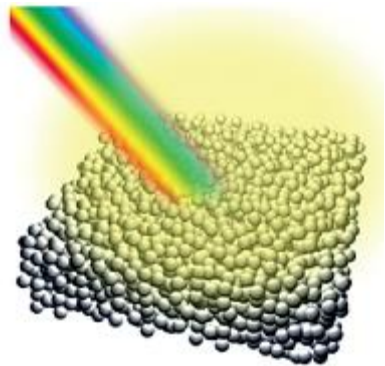
The generalized microscope slide



- ❑ **Conventional**
 - Passive, compensated
 - Cheap and standard



- ❑ **Nanophotonic** *More Control!*
 - Active, focusing, filtering
 - Super, hyper, meta lens



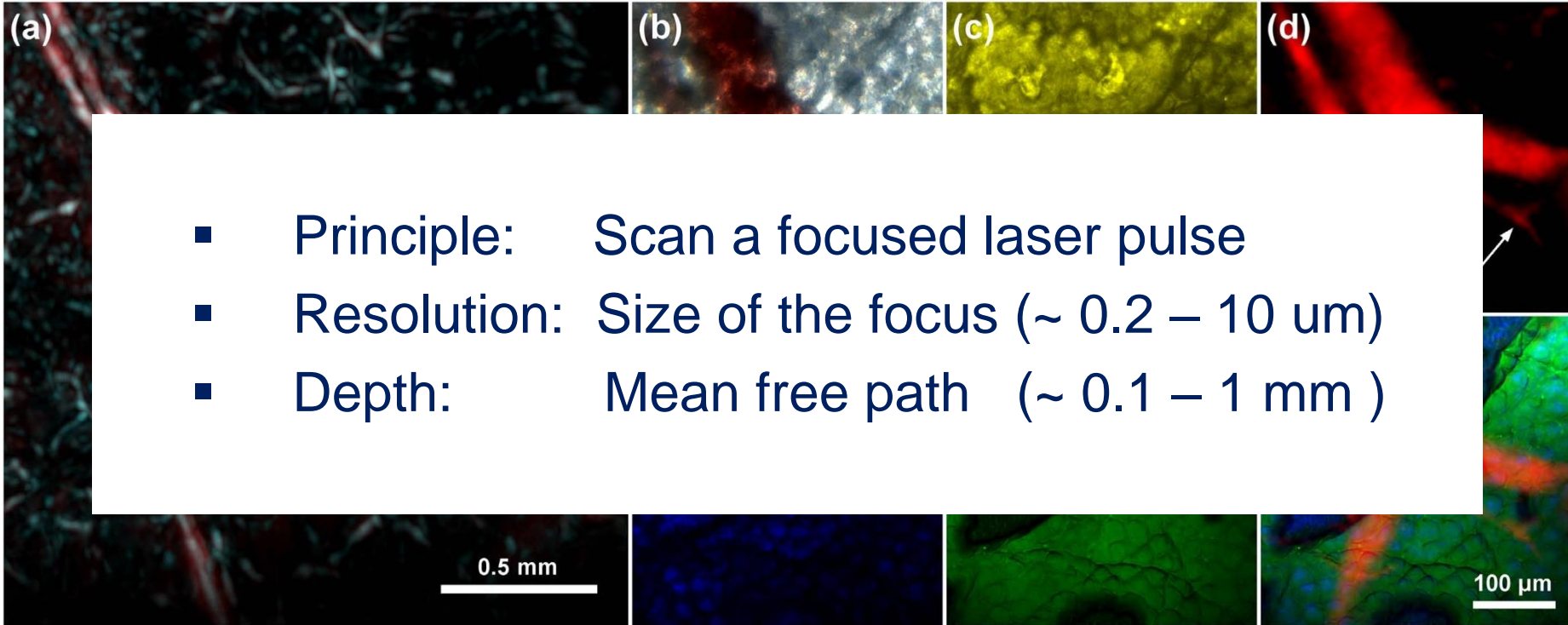
- ❑ **Natural** *More Control!*
 - Omnipresent, random, light scattering
 - Thin slicing *ex-vivo*, other waves

Optical bioimaging – label free

Bright-field

2 Photon
fluorescence

Optoacoustic
microscopy



- Principle: Scan a focused laser pulse
- Resolution: Size of the focus ($\sim 0.2 - 10 \mu\text{m}$)
- Depth: Mean free path ($\sim 0.1 - 1 \text{ mm}$)

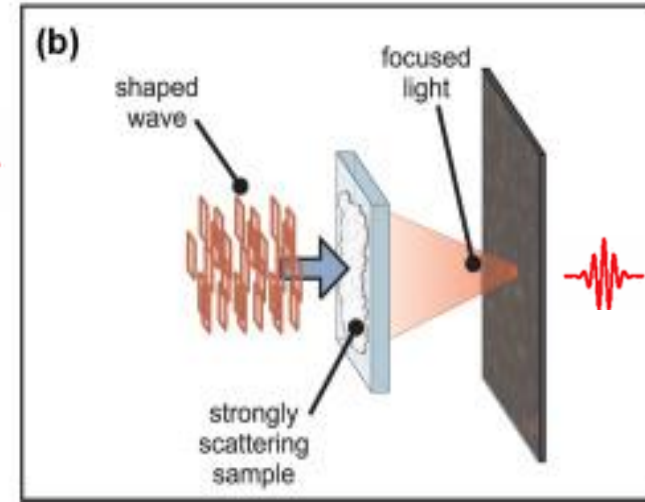
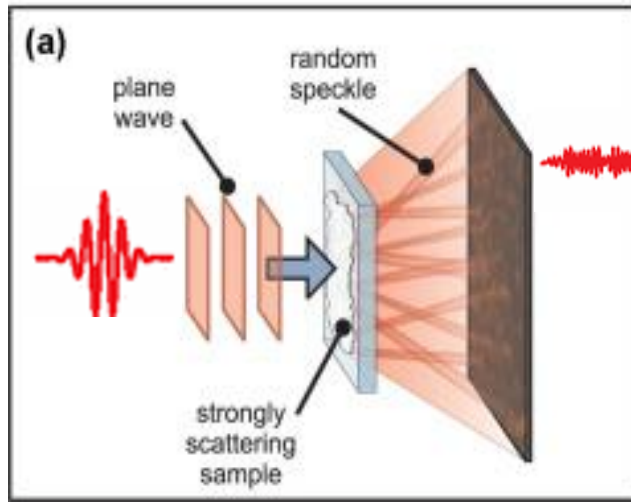
SHG

THG

Overlaid
contrast

Nonlinear harmonic generation

Bypassing diffusion (space – time)



- **Acoustics**

- M. Fink, IEEE (1992)

- A. Derode, et al, PRL (1995)

- **Microwave**

- G. Lerosey, et al, PRL (2004)

- G. Lerosey, et al, Science (2007)

- **Optics (monochromatic)**

- S. M. Popoff, et al, PRL 2010

- **Optics (pulsed)**

- D. J. McCabe, et al, Nat Com (2011)

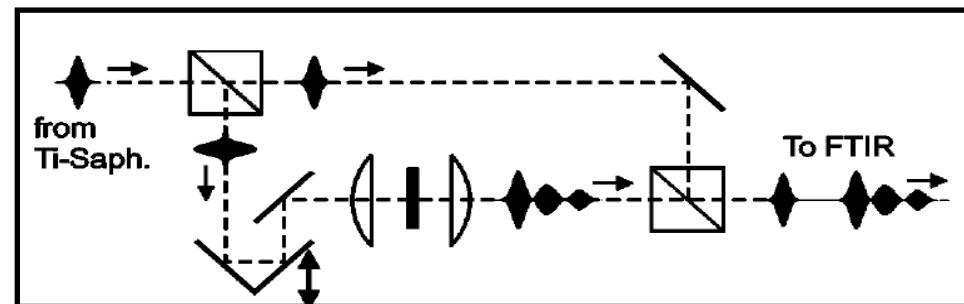
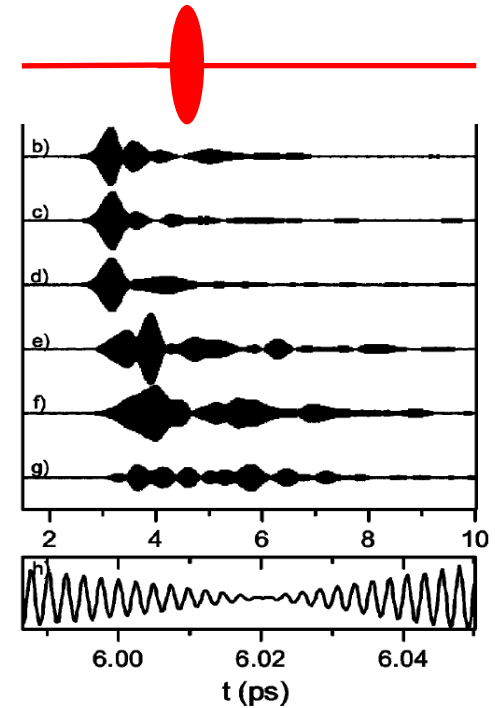
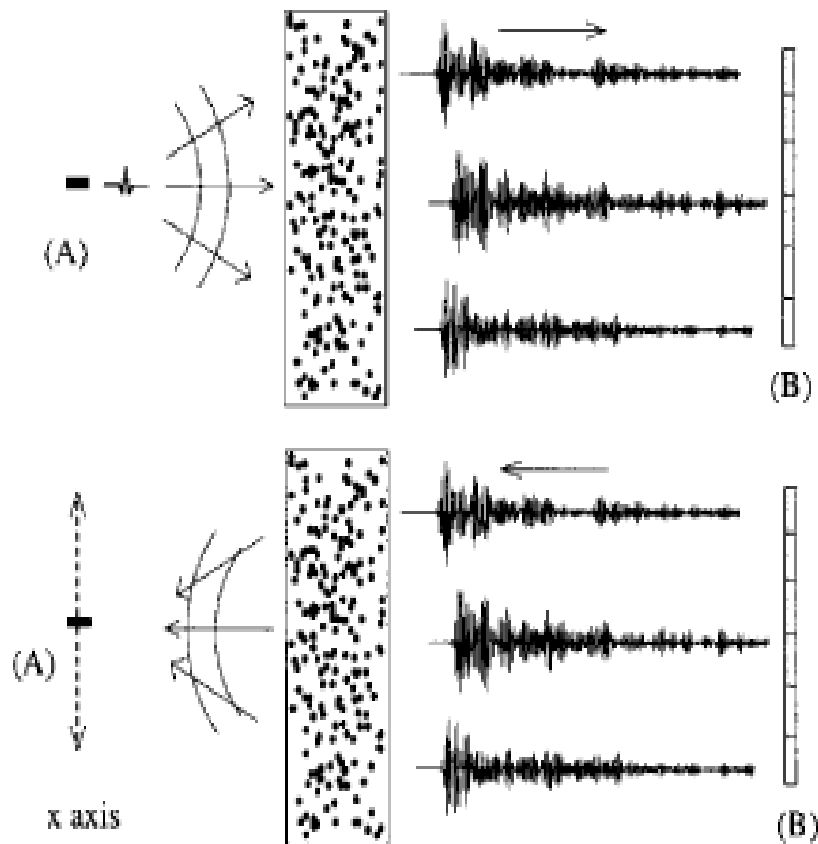
- J. Aulbach, et al, PRL (2011)

- O. Katz, et al, Nat Photon (2011)

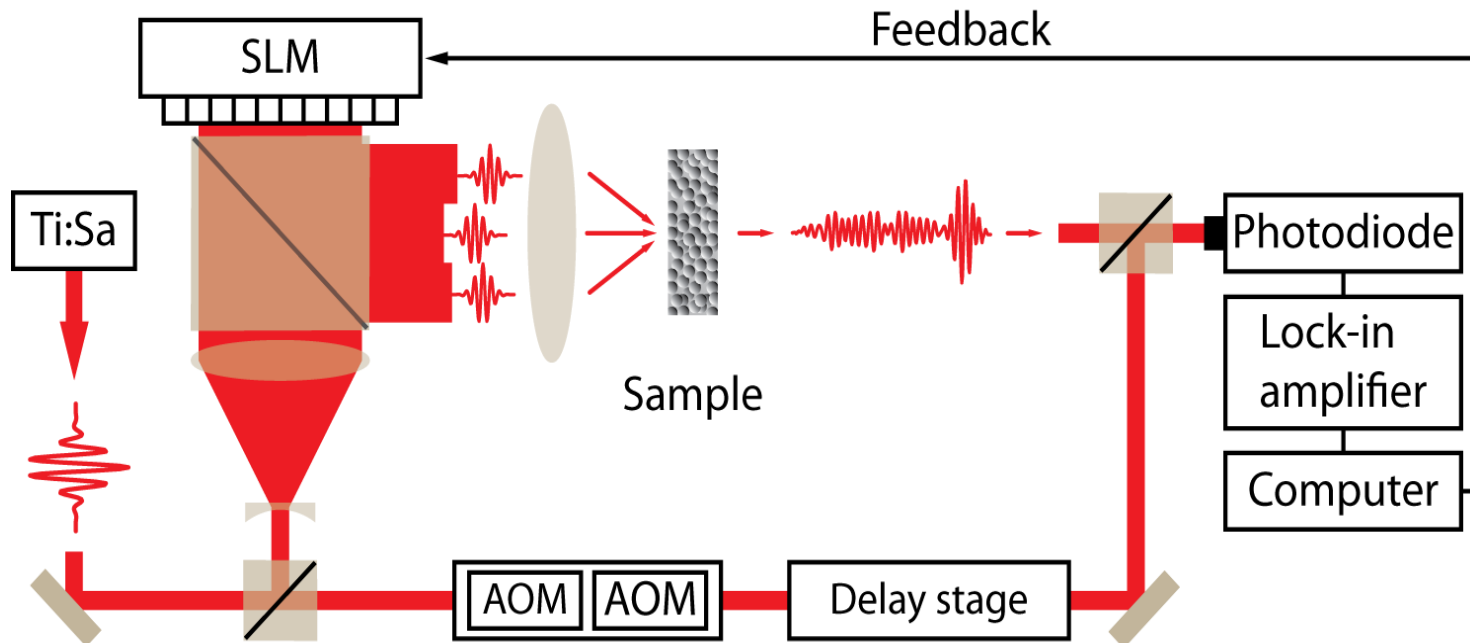
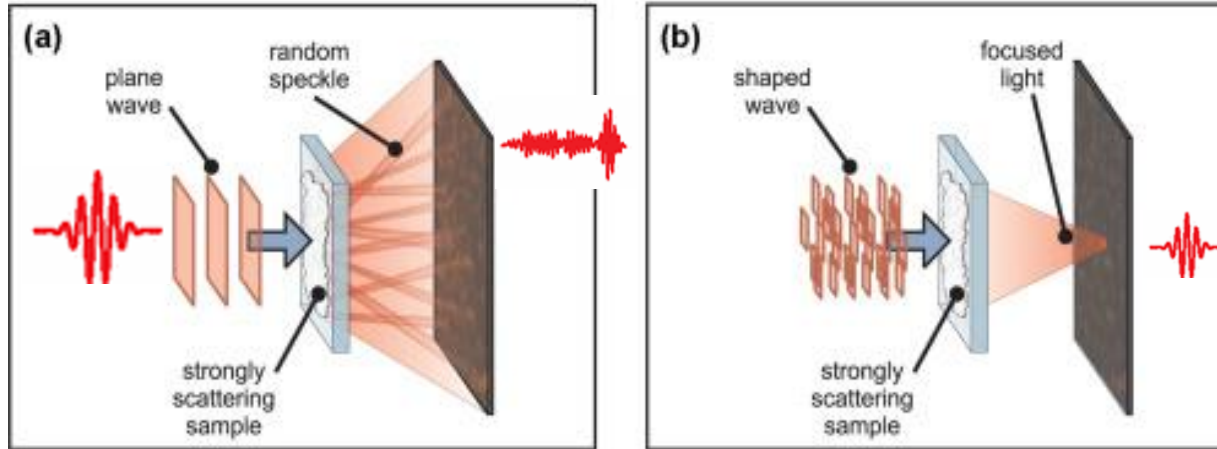
Acoustics

to

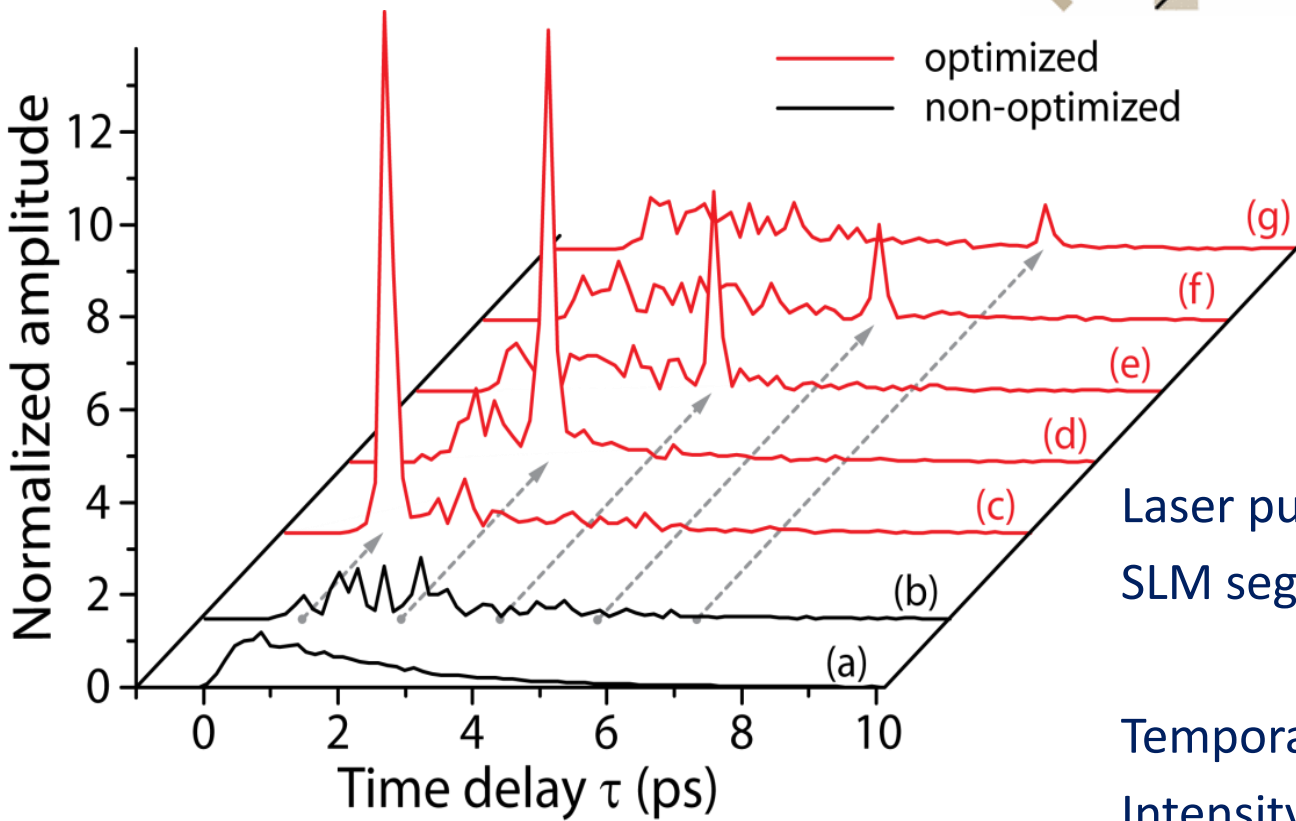
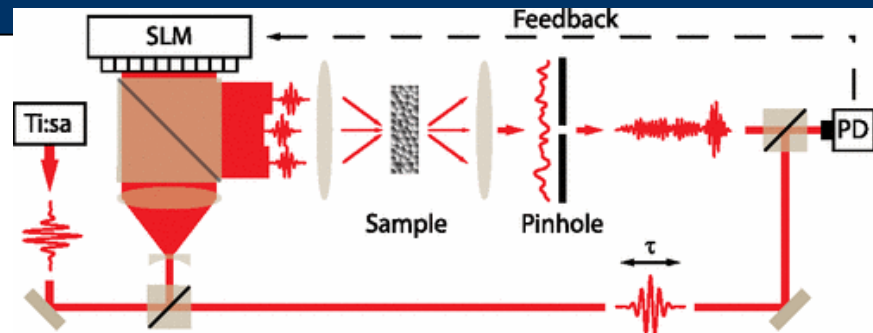
optics



Bypassing diffusion (space – time)



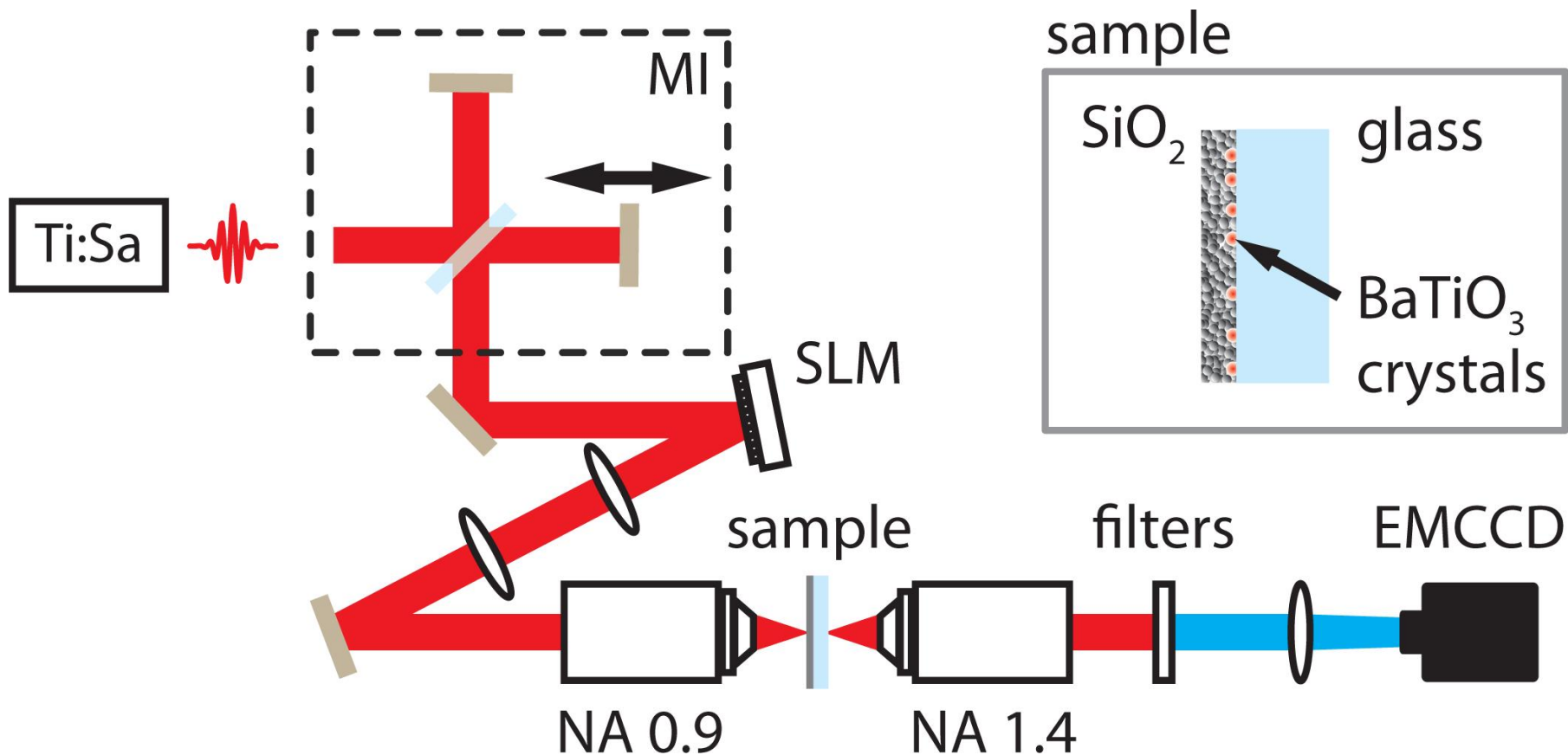
Time focusing



Laser pulse ~ 64 fs
SLM segmentation $N \sim 300$
Temporal focusing ~ 115 fs
Intensity enhancement $\sim 10^2 - 10^3$

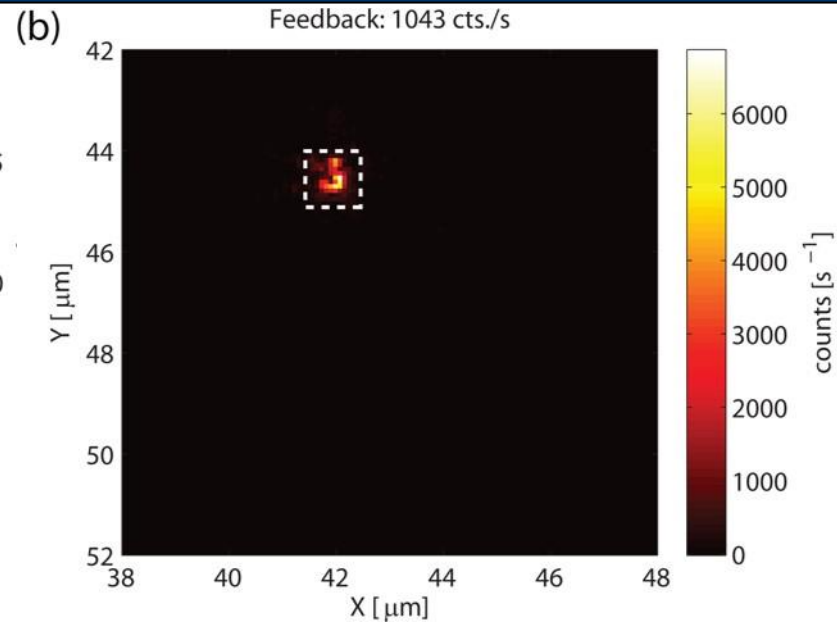
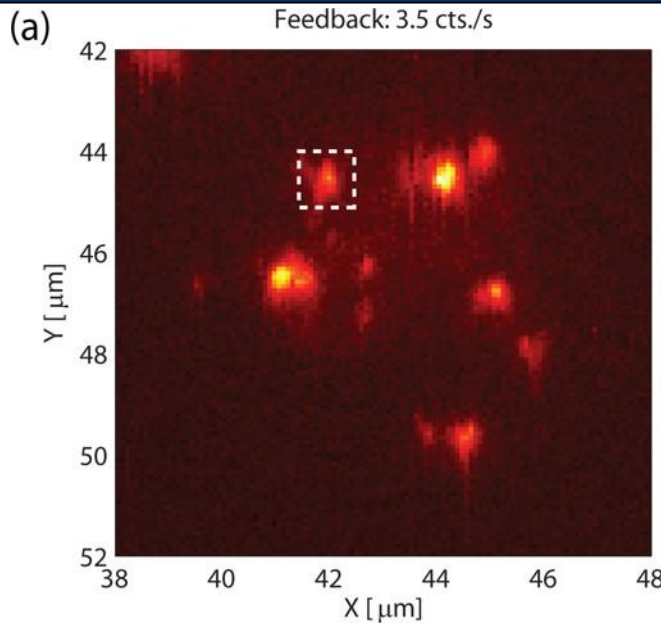
Space-time addressing of targets

Second Harmonic particles (size 200 nm)

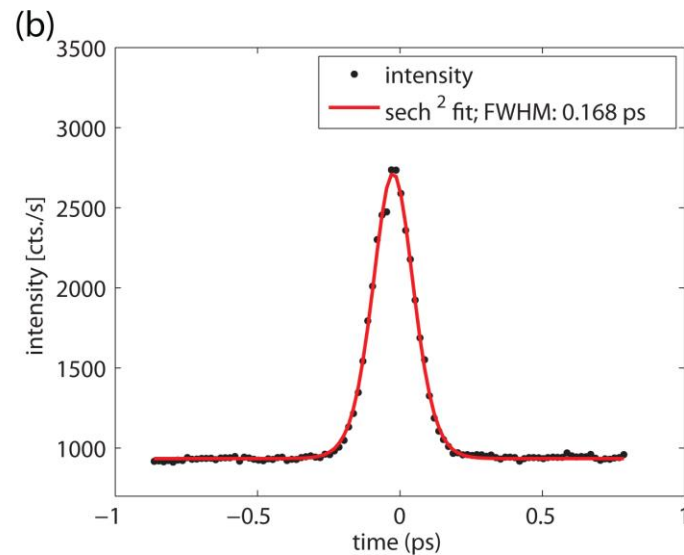
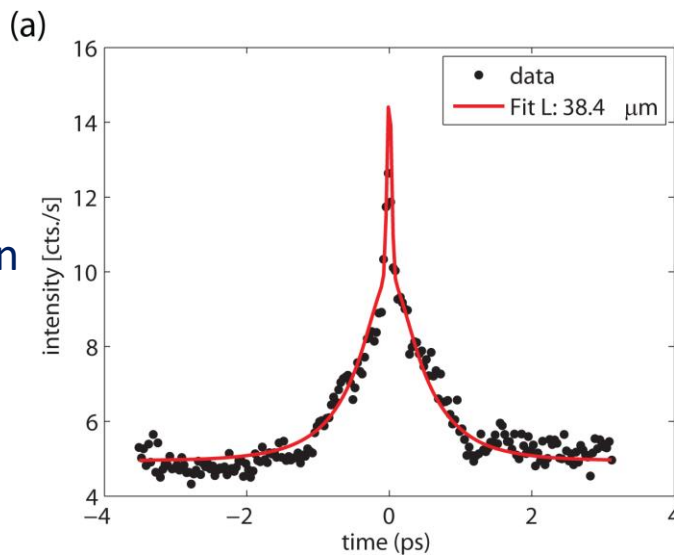


Space-time focusing

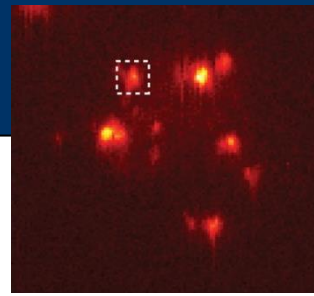
SH images
(@ 2ω)



Autocorrelation
(@ 2ω)



Reproducibility



Part. #	τ_{pulse} (fs)	η_{exp}	η_{model}	η_{cw}
1	110	$2.5 \cdot 10^2$	$3.1 \cdot 10^2$	$1.6 \cdot 10^4$
2	102	$0.7 \cdot 10^2$	$3.2 \cdot 10^2$	$1.7 \cdot 10^4$
3	111	$2.7 \cdot 10^2$	$2.7 \cdot 10^2$	$1.4 \cdot 10^4$
4	104	$0.7 \cdot 10^2$	$2.9 \cdot 10^2$	$1.5 \cdot 10^4$
5	109	$3.0 \cdot 10^2$	$3.8 \cdot 10^2$	$2.0 \cdot 10^4$
6	109	$5.5 \cdot 10^2$	$6.5 \cdot 10^2$	$2.4 \cdot 10^4$

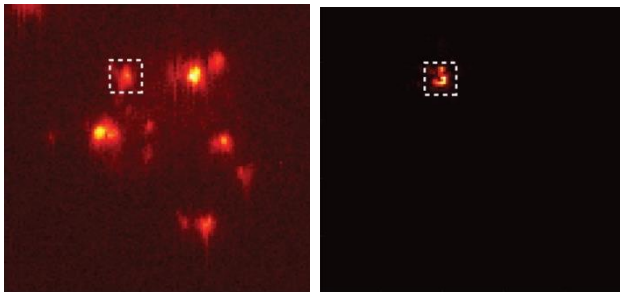
The enhancement, η , depends on the SLM segmentation.

We contained the SLM segmentation to $N = 800$,
because above it ($N > 1000$) **the particles are burned and destroyed**

Biomedical opportunities

Depth ~ **10 - 100** mean free paths

Tissue equivalent **0.1 - 5 cm**



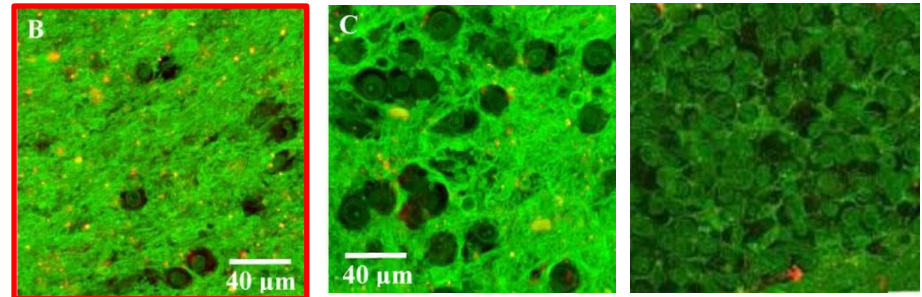
Can we burn inside?

Nanoparticle labelling ?

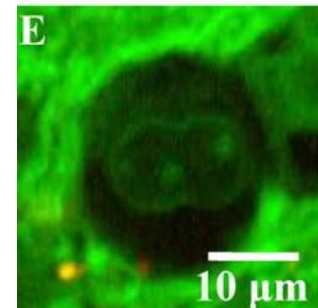
Can we image inside (THG)?

Focusing and memory effect?

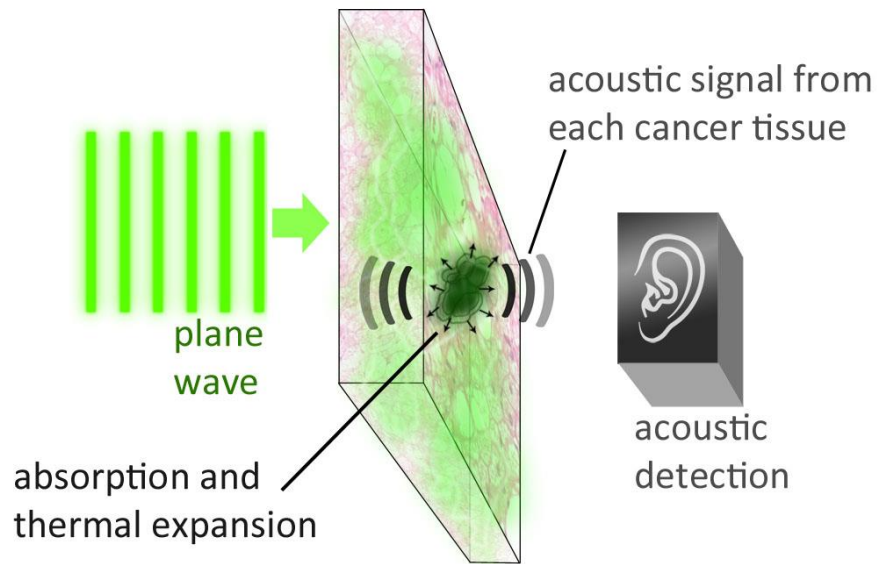
Human brain tumour (dark)
Intra-operational THG image (40 mW)



- Intra-operational (in depth)
 - High tumour density: CUT MORE
 - Low tumour density: CLOSE UP
- Early stage (in depth)
 - Degeneration: YES/NO

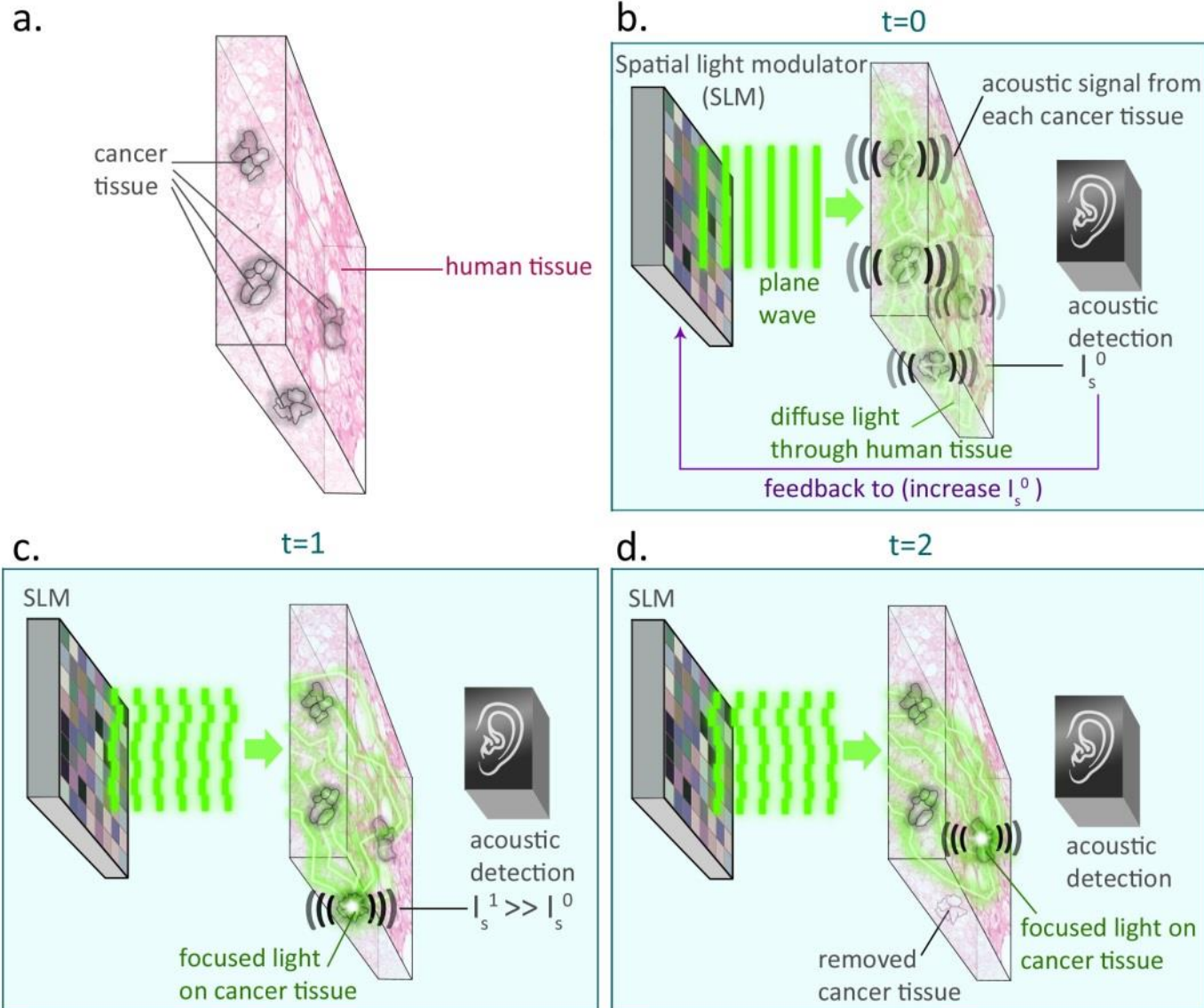


The right feedback



- **Challenges/requisitions:**
 - Diffusion of light by tissue
 - Local power delivery
 - **Specificity – tumours only**
 - Photoacoustic contrast
 - Multi-sensorial (light, sound)
 - Single detector (image-free)
 - Only optical diffusion

Ant-burning of tumours



Therapy, imaging & sensing

□ Photoacoustics space-time focusing:

- Contrast agents: Hemoglobin – brain, eye, breast
Melanin – skin, lymph nodes, circulating tumor cells
External – dyes, plasmonic particles
- Imaging : SHG, **THG**, (memory effect - high res, limited range)
- Sensing: Oxygen saturation of Hemoglobin (sO_2), temperature

One case of success is sufficient!

Conclusions



Generic

- Microscopy is important and is challenging
- 'SIM is the future' – *Betzig said.*



Specific to this work

- High index materials ($n > 3$) are great for SIM
- A double Moire illumination is the way to go
- 5x better resolution (down to 45nm)
- Relatively simple and fast
- The required control is feasible