



**The Abdus Salam
International Centre for Theoretical Physics**



2223-23

Winter College on Optics in Imaging Science

31 January - 11 February, 2011

On breaking the Abbe diffraction limit in Optical Nanopatterning & Nanoscopy

R. Menon
*University of Utah & MIT
USA*

On breaking the Abbé diffraction limit in Optical Nanopatterning & Nanoscopy

Rajesh Menon

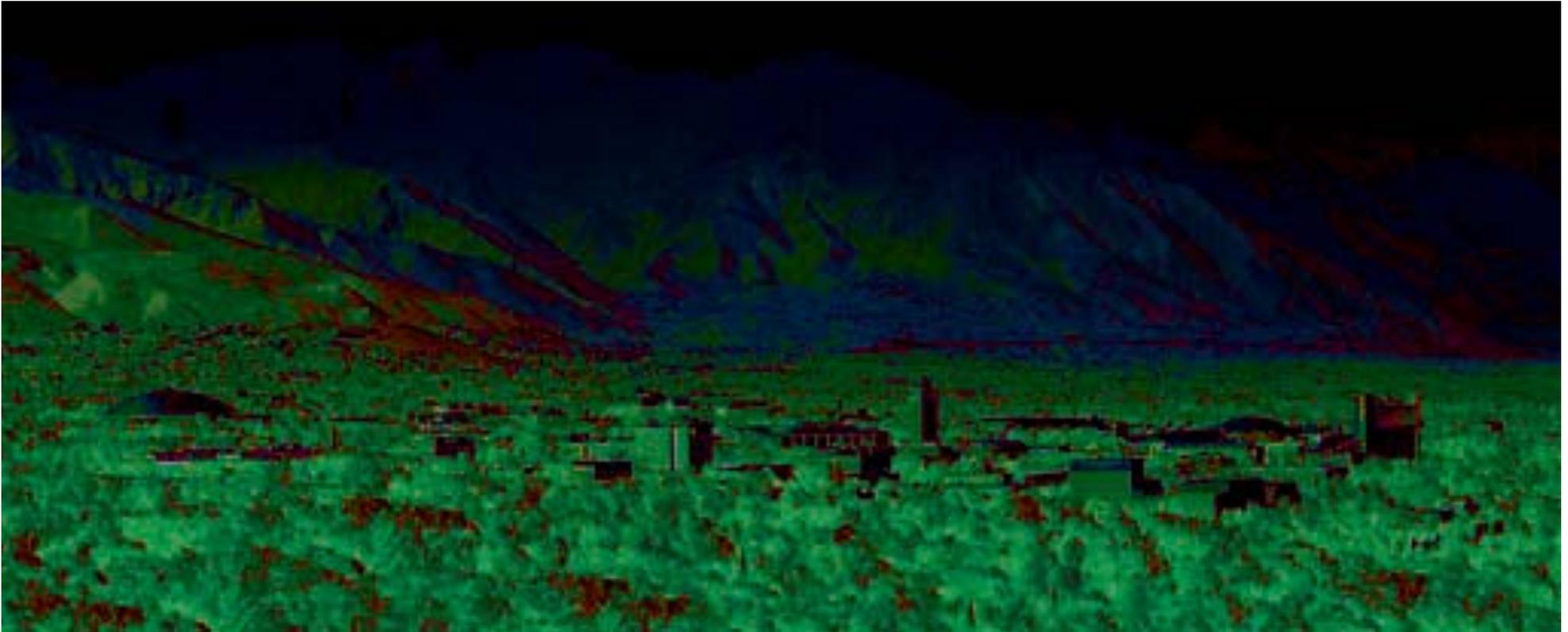
rmenon@eng.utah.edu

<http://lons.utah.edu>

Laboratory for Optical Nanotechnologies
Department of Electrical and Computer Engineering
University of Utah

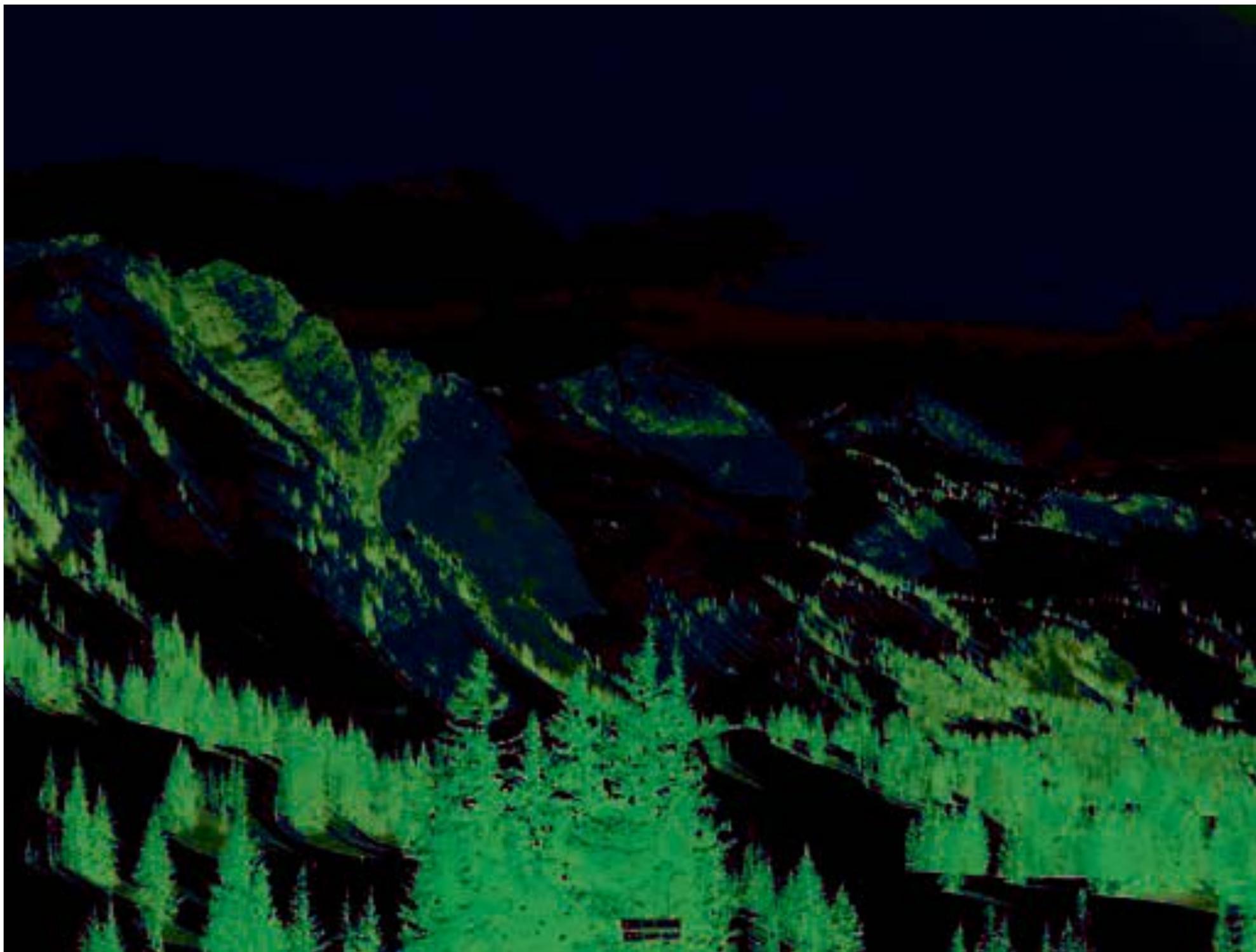
Research Laboratory of Electronics
MIT

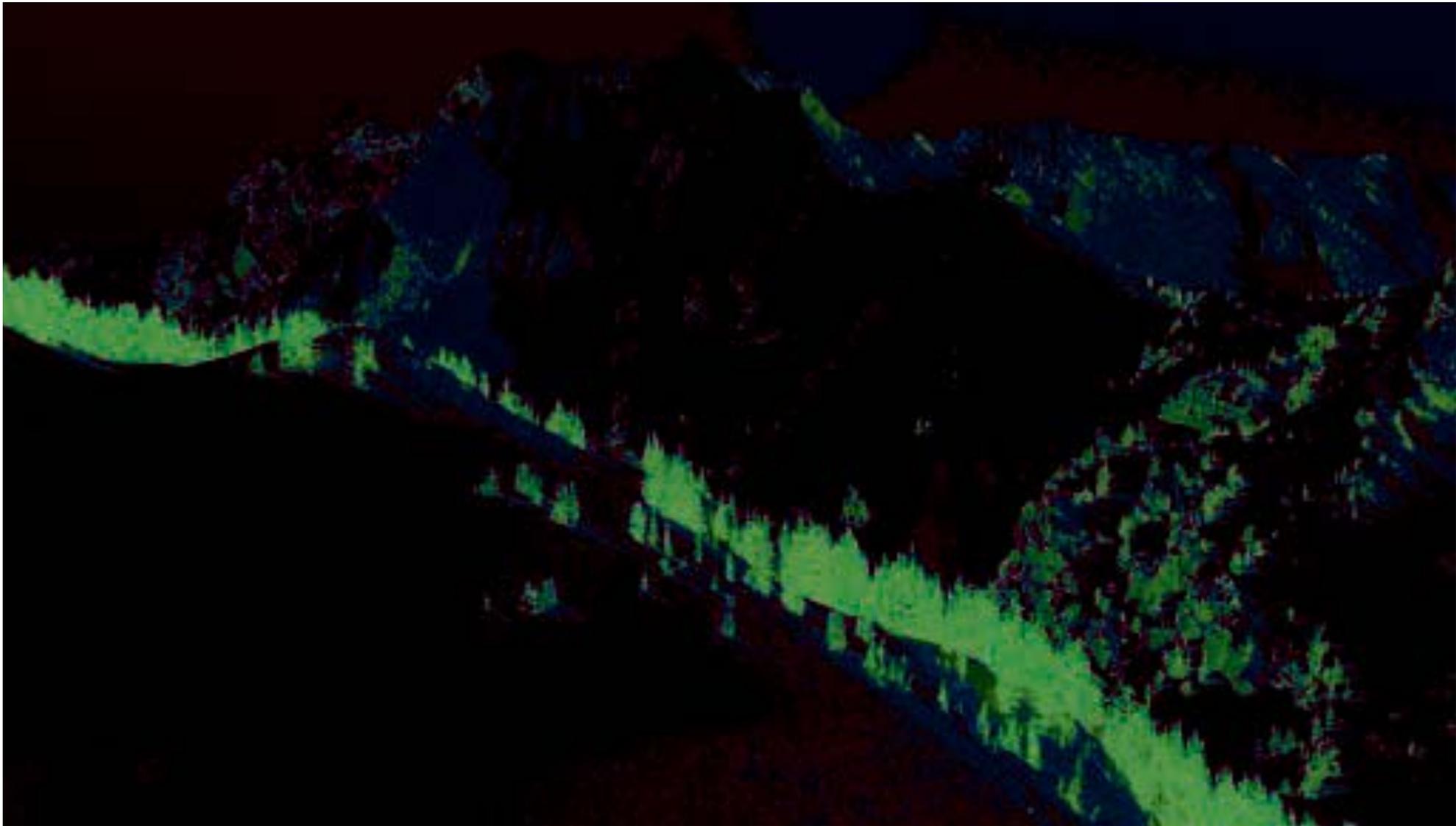
The University of Utah

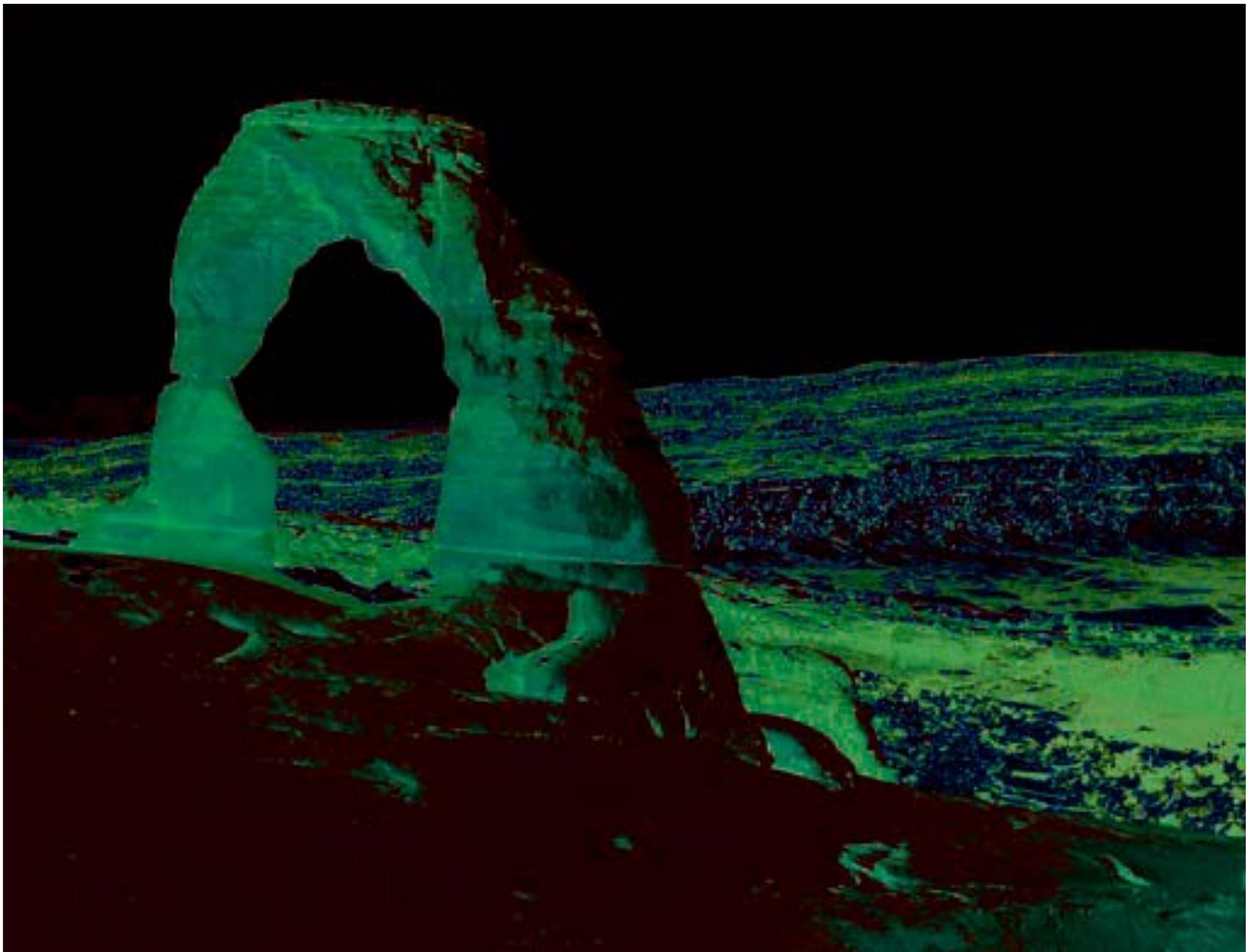


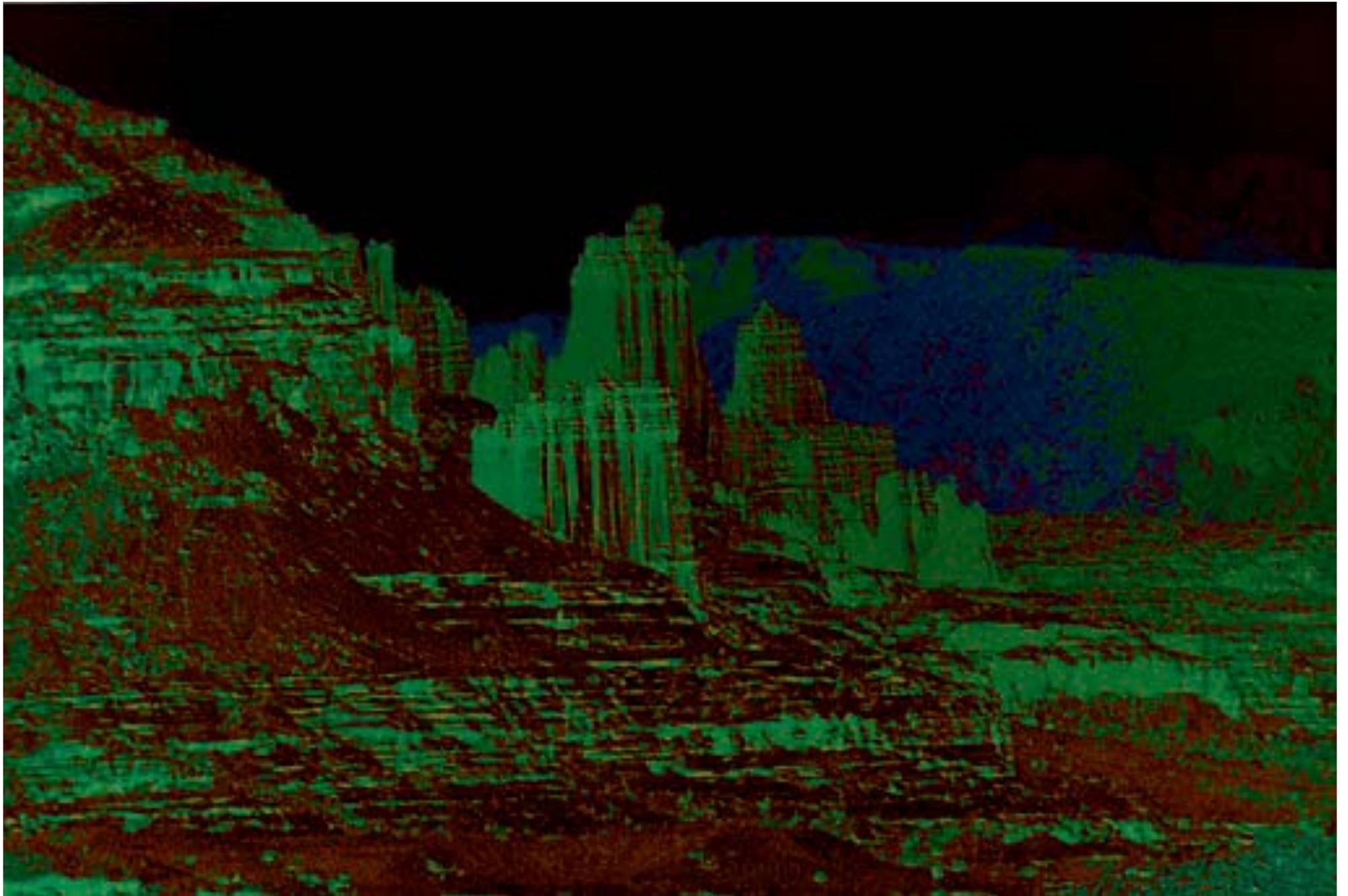
- 1st year fellowship open to the best PhD students (including international students)
- Infra-structures & facilities
- Highly inter-disciplinary research
- Entrepreneurship opportunities





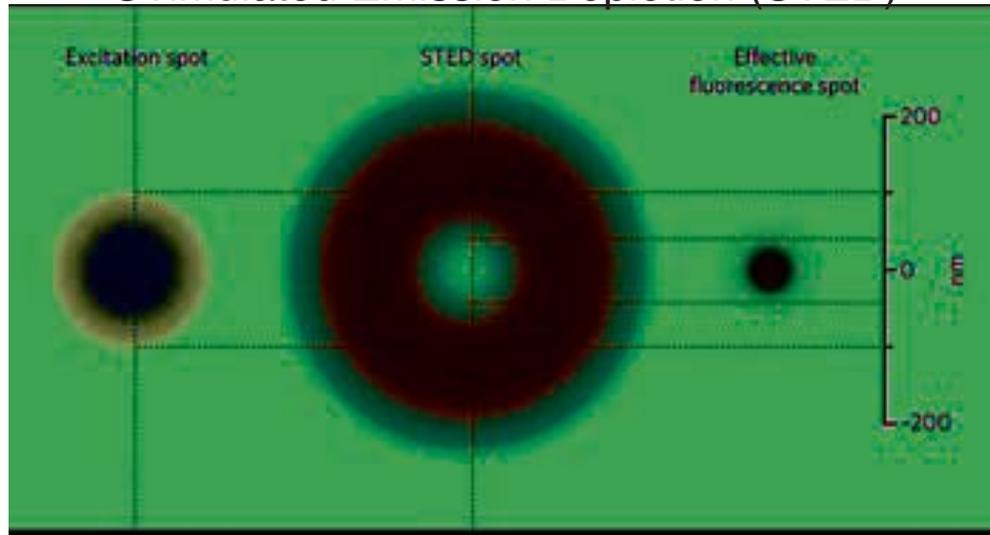




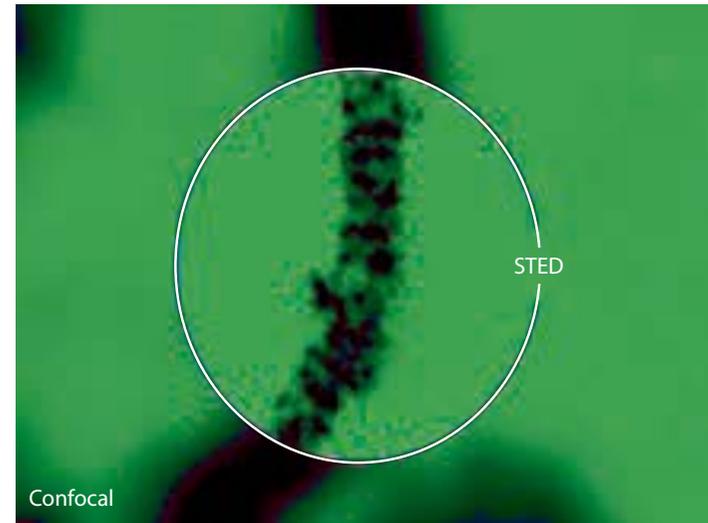


Superresolution using Fluorescence

STimulated Emission Depletion (STED)



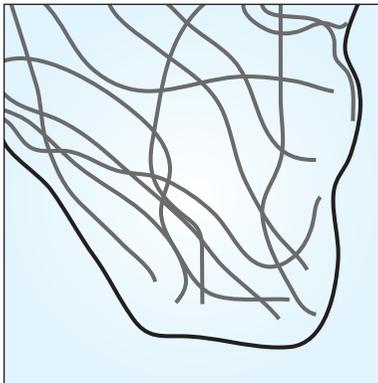
Folds of the mitochondrial inner membrane



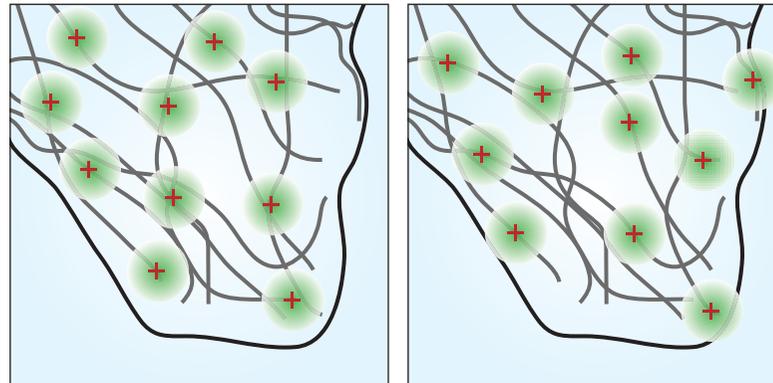
T. A. Klar, *et. al*, *PNAS* 97, 8206 (2000).

STochastic Optical Reconstruction Microscopy (STORM)

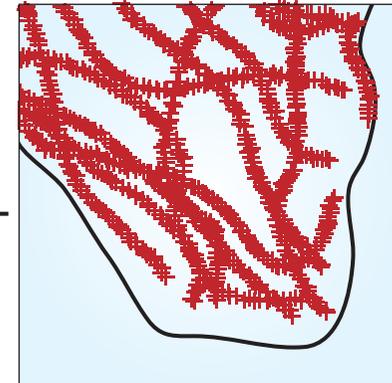
Target structure



Localizing activated subset of probes



STORM image



X. Zhuang, *Nat. Photonics*, 3, 365 (2009).

Acronyms

STED = stimulated emission depletion

GSD = ground state depletion

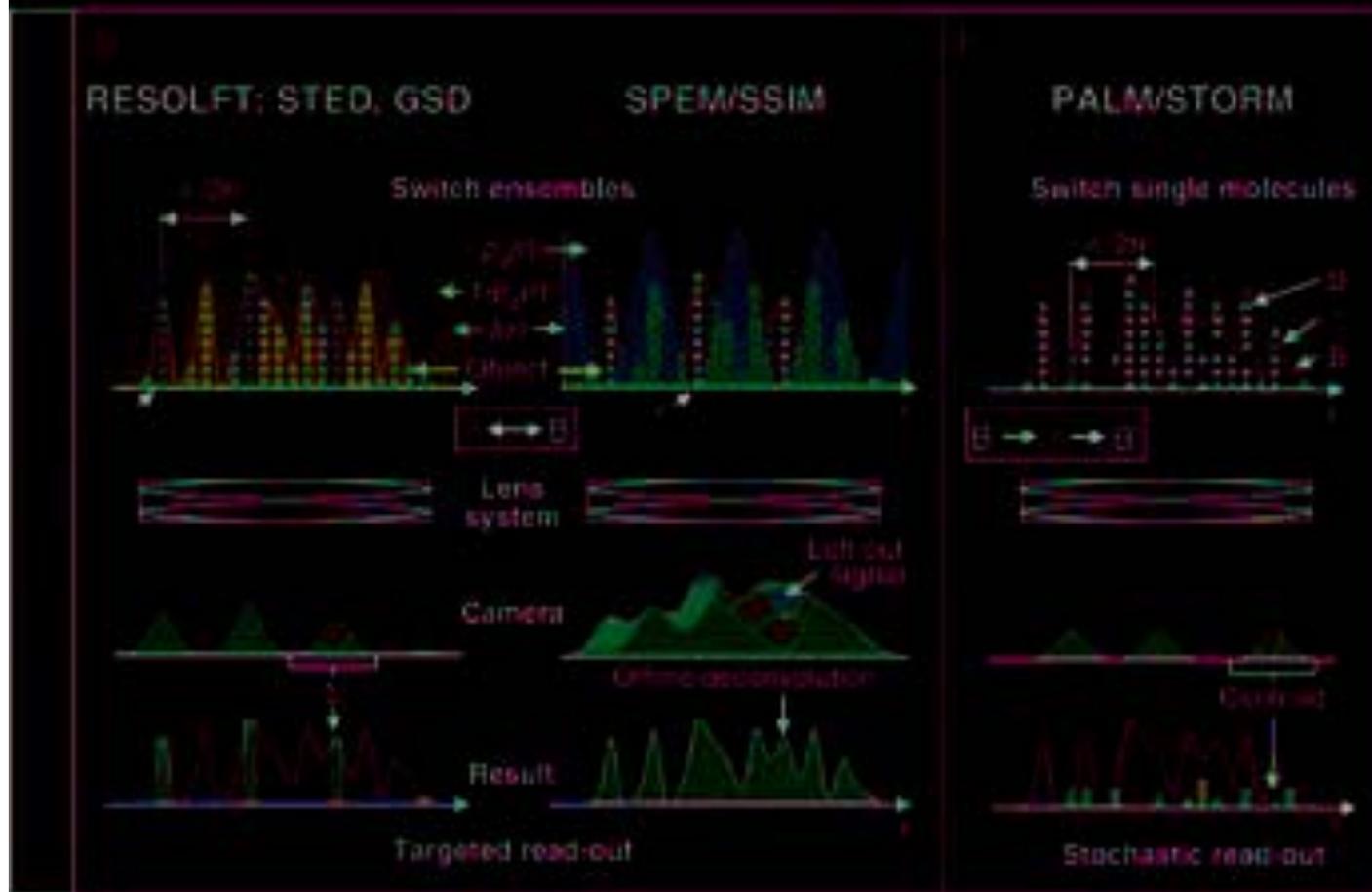
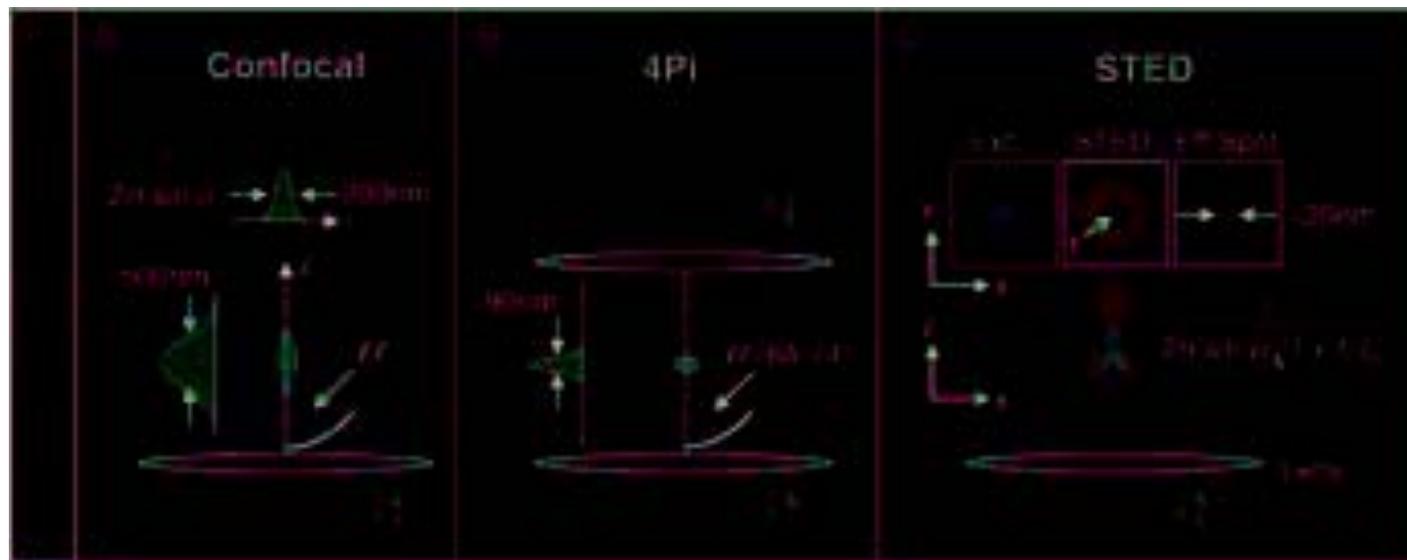
RESOLFT = reversible saturable optically linear fluorescence transitions

SPEM = saturated pattern excitation microscopy

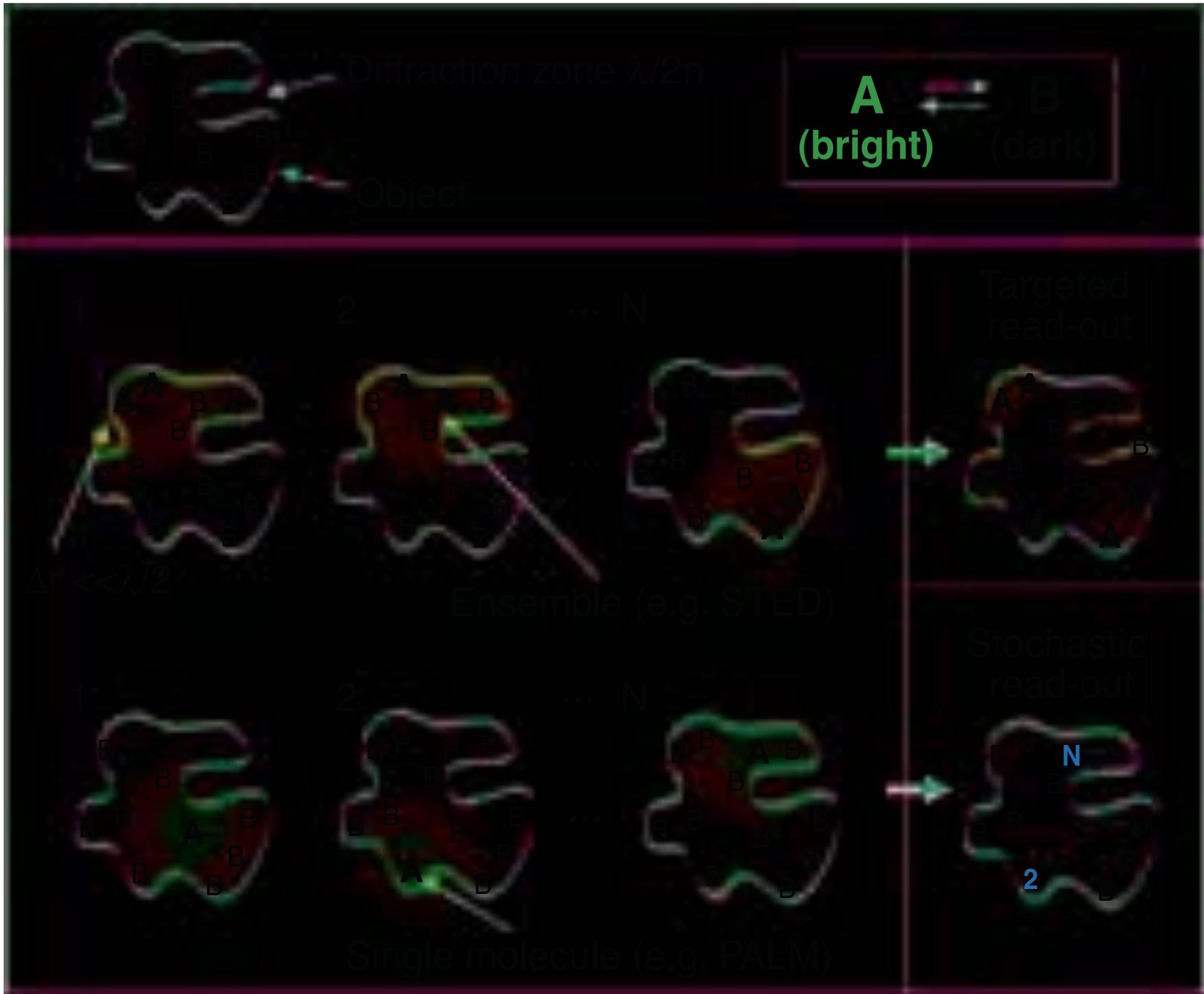
SSIM = saturated structured illumination microscopy

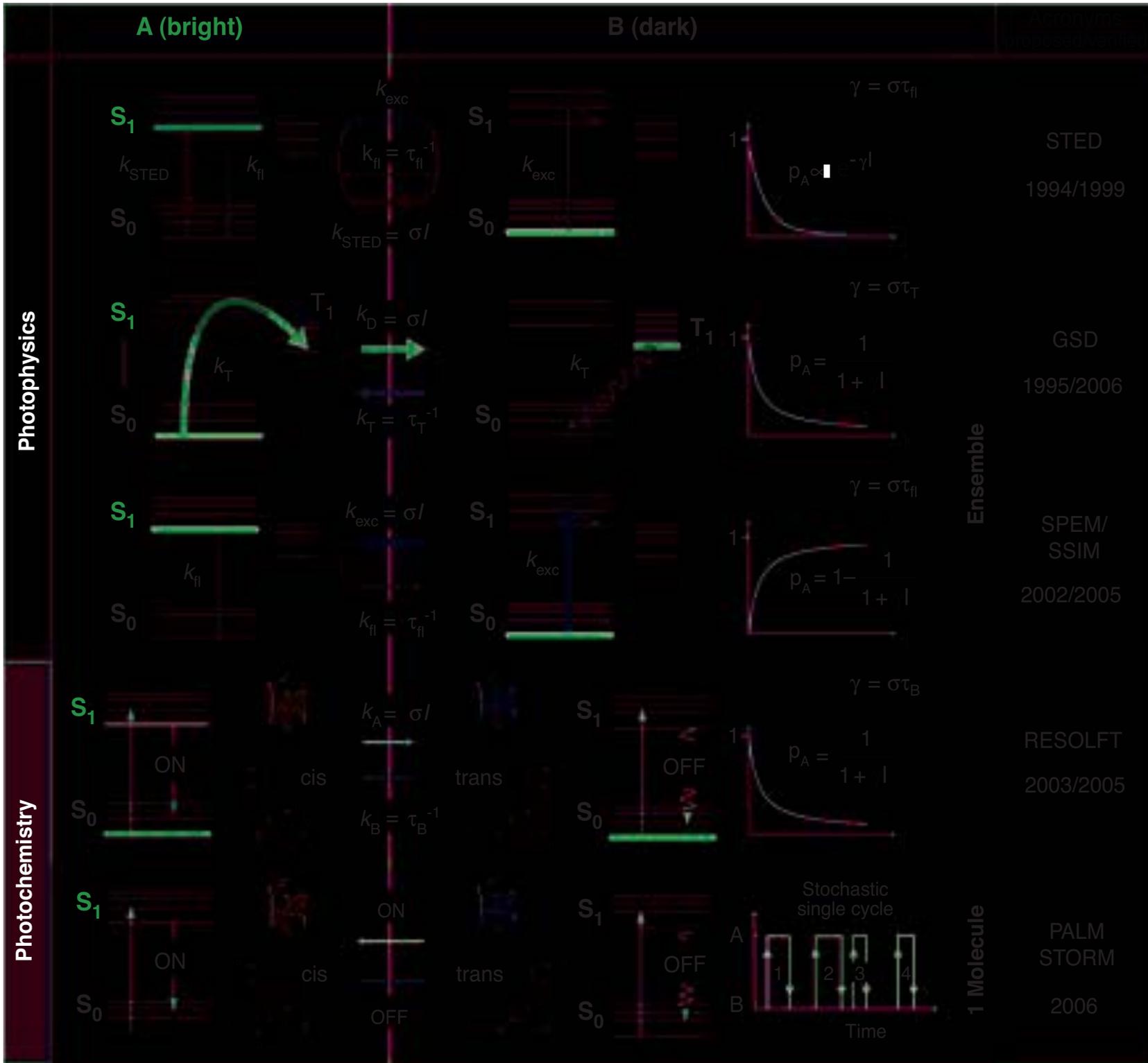
PALM = photoactivatable localization microscopy

STORM = stochastic optical reconstruction microscopy



Scanning vs non-scanning: STED vs PALM





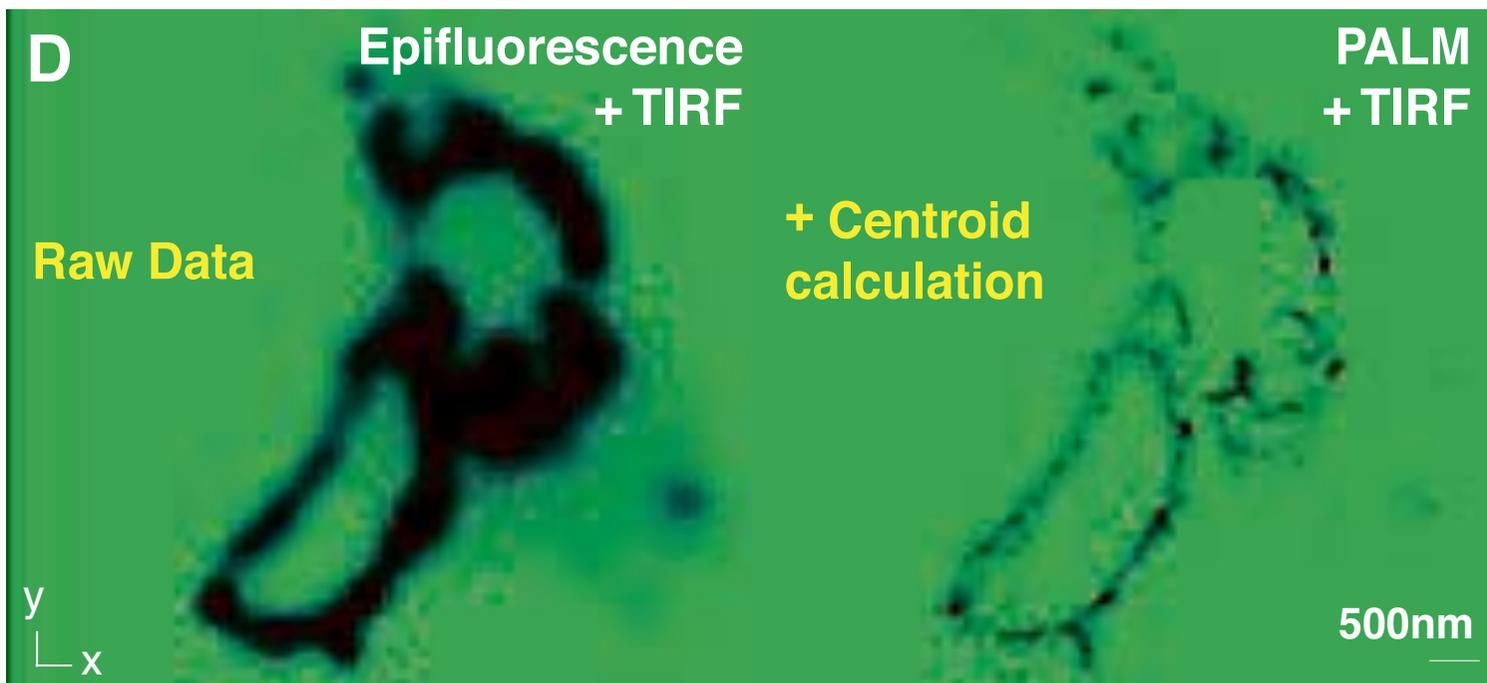
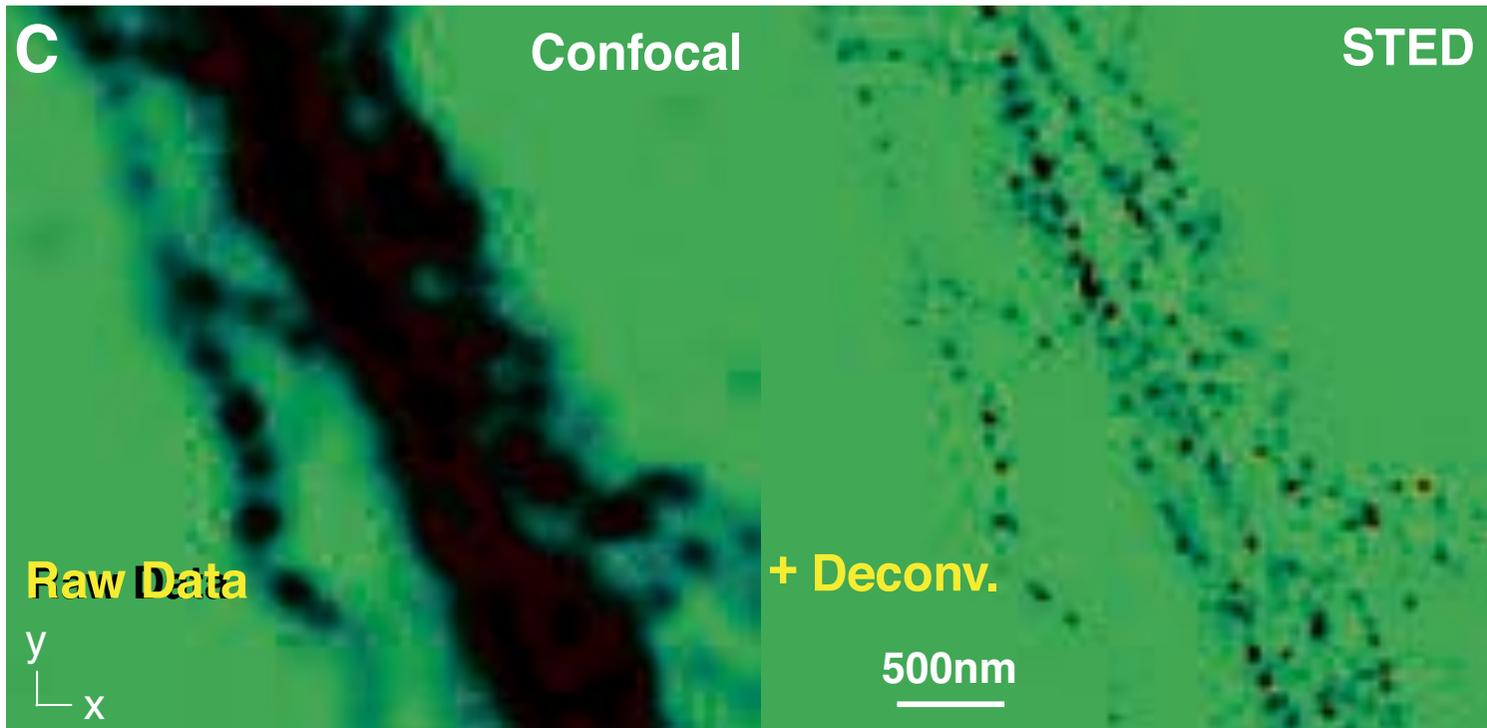
$\tau_{fl} \approx 10^{-9} \text{ s}$
 $\sigma \approx 10^{-16} \text{ cm}^2$
 $I_s = (\sigma \tau_{fl})^{-1}$
 $\sim 10 \text{ MW/cm}^2$
 sub-ns, not femtosecond

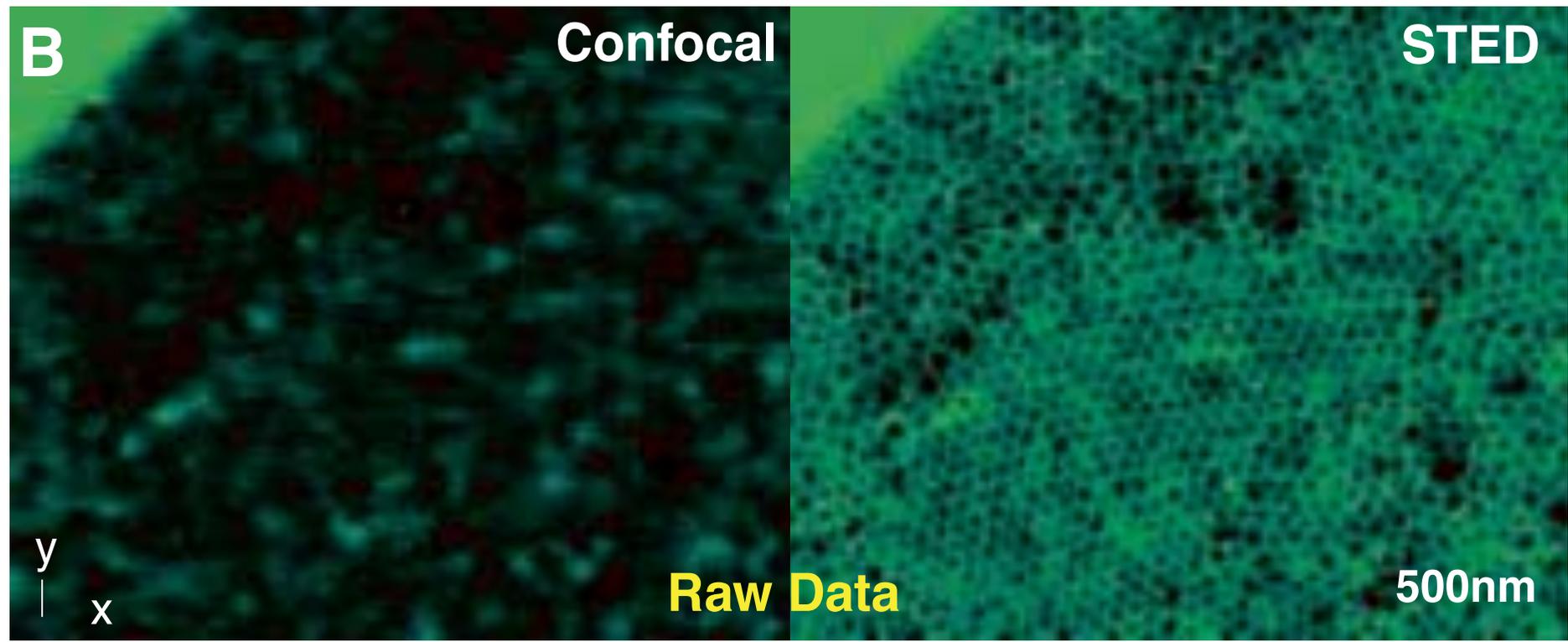
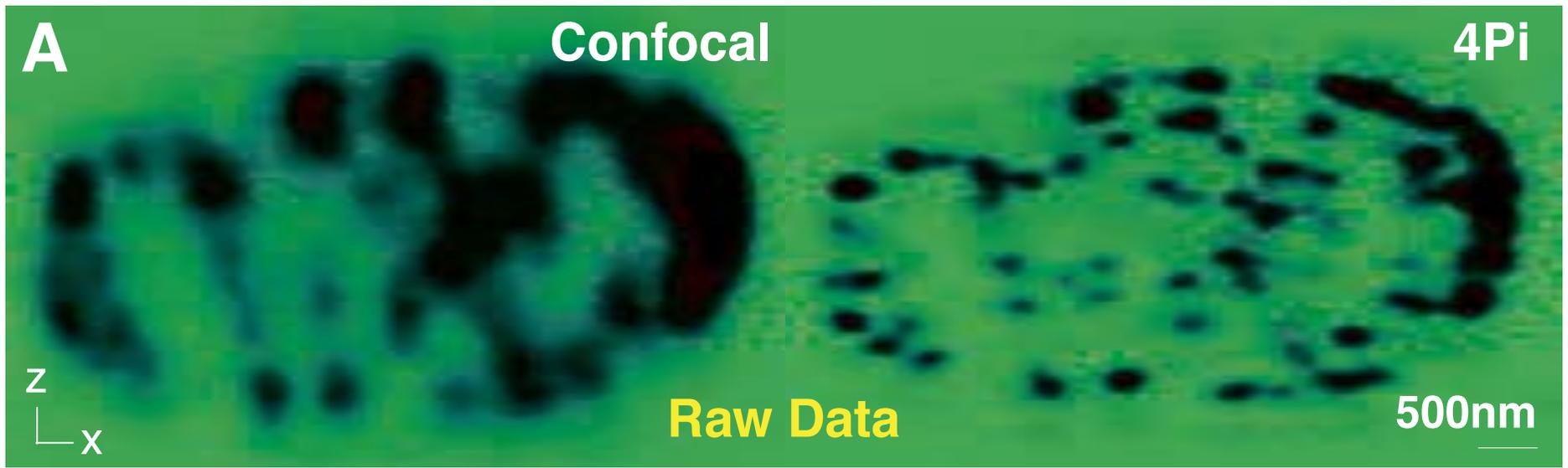
$\tau_T \approx 10^3 \text{ to } 10^6 \text{ ns}$
 $(T_1 \text{ is meta-stable})$
 $\sim 100 \text{ kW/cm}^2 \text{ (CW)}$

Higher SNR requirements

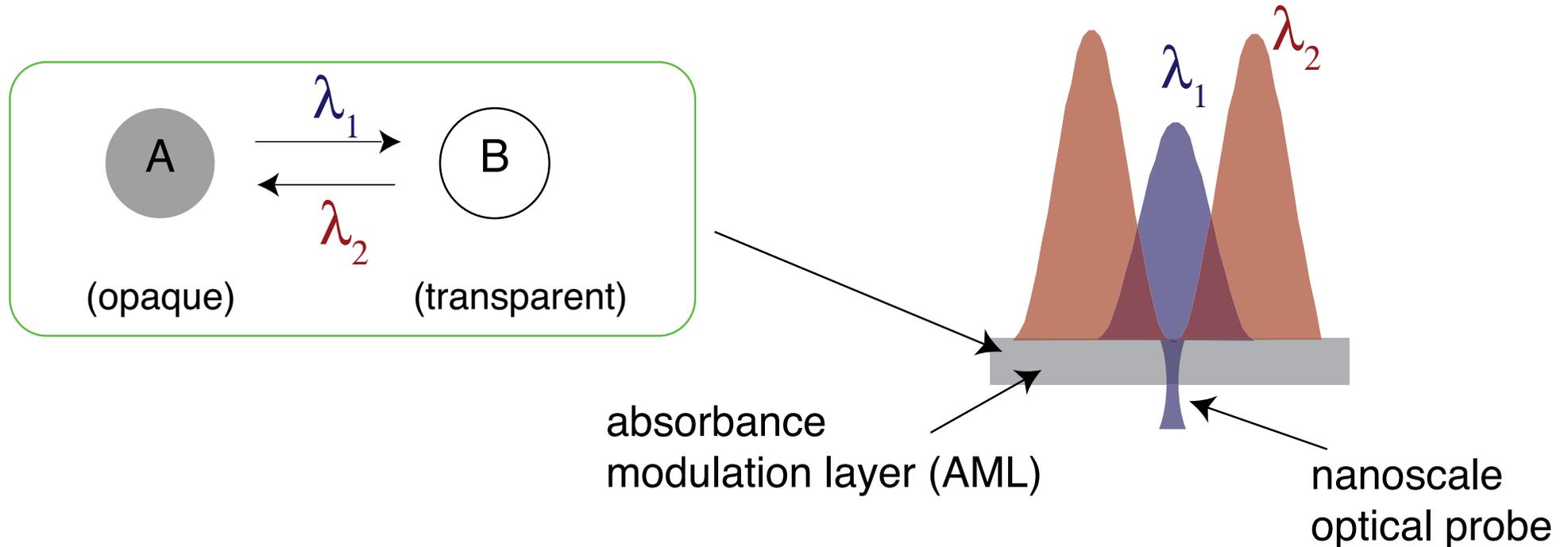
10 W/cm^2
 Photostability?

computation
 long recording times





Absorbance Modulation: Exploiting Wavelength-selective chemistry to overcome the Diffraction limit



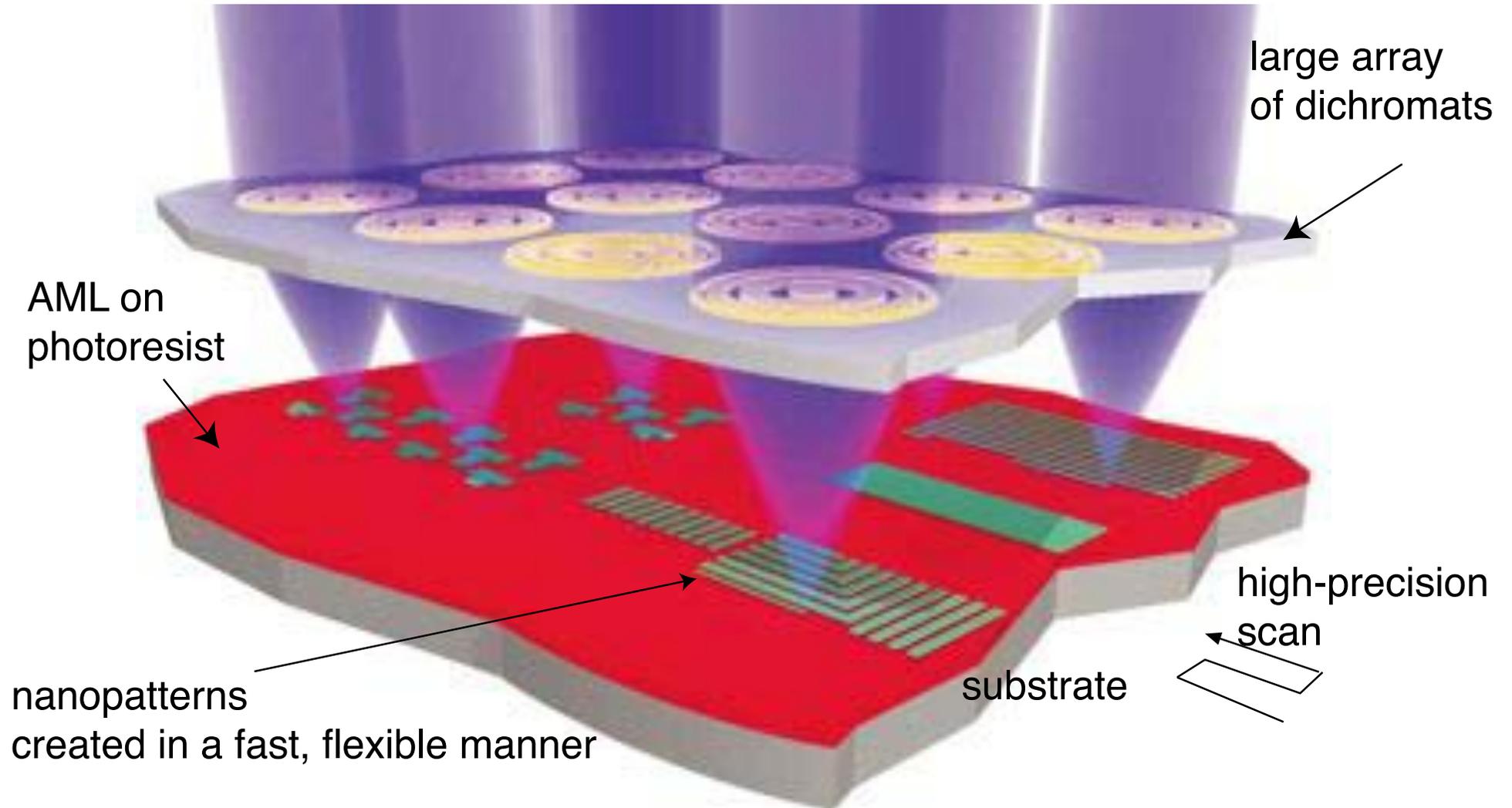
Focal ring at λ_2 in competition with round spot at λ_1 creates a localized sub-wavelength aperture.

Light at λ_1 penetrates through this aperture forming a nanoscale probe

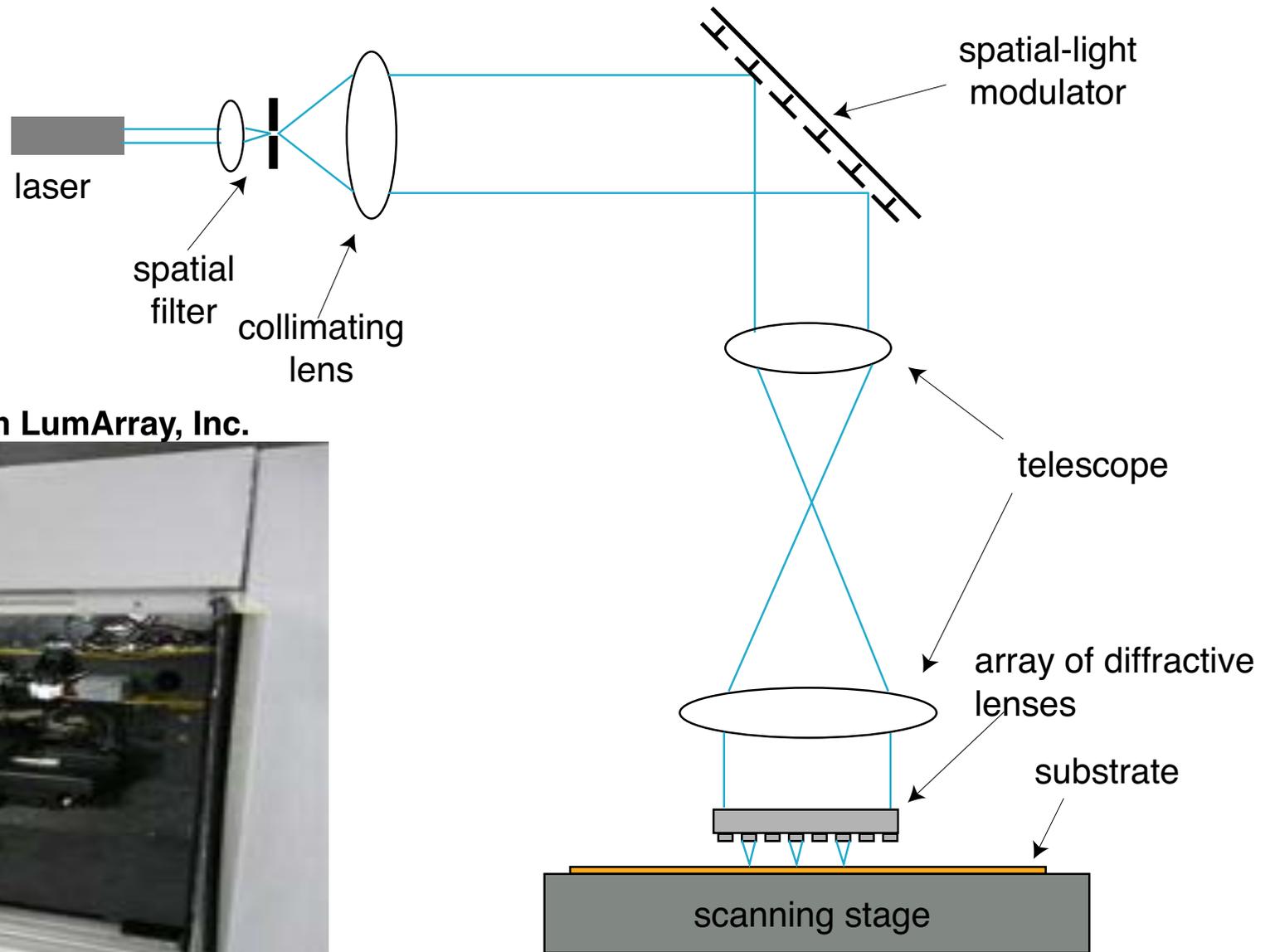
Vision: Massively parallel, independently controllable nanoscale optical probes

λ_1 beamlets individually gray-scaled with spatial-light modulator

λ_2 uniformly illuminates all dichromats



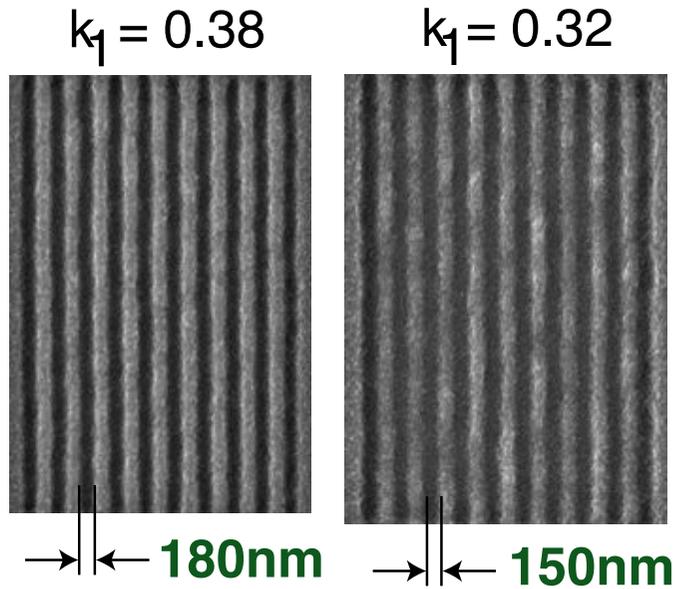
Architecture for massively parallel patterning



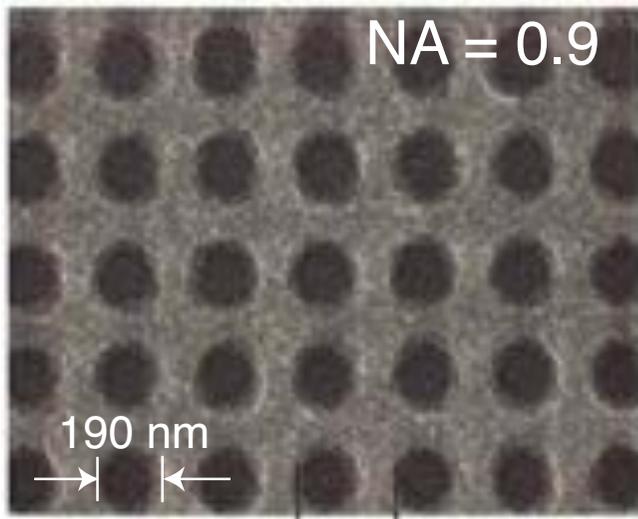
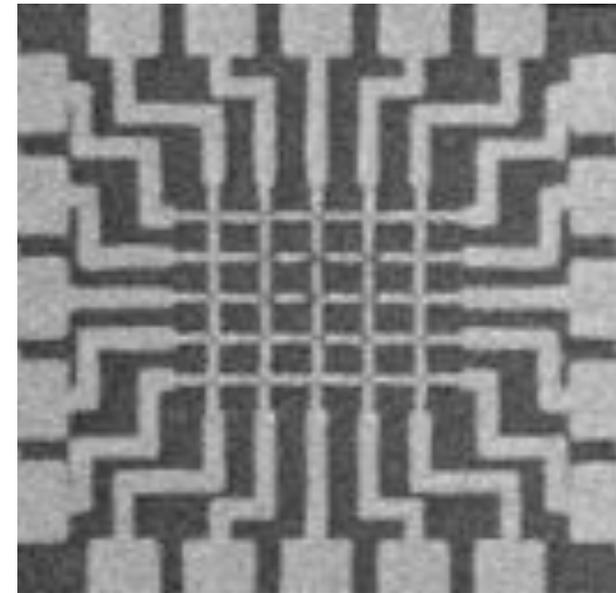
Commercial tool from LumArray, Inc.



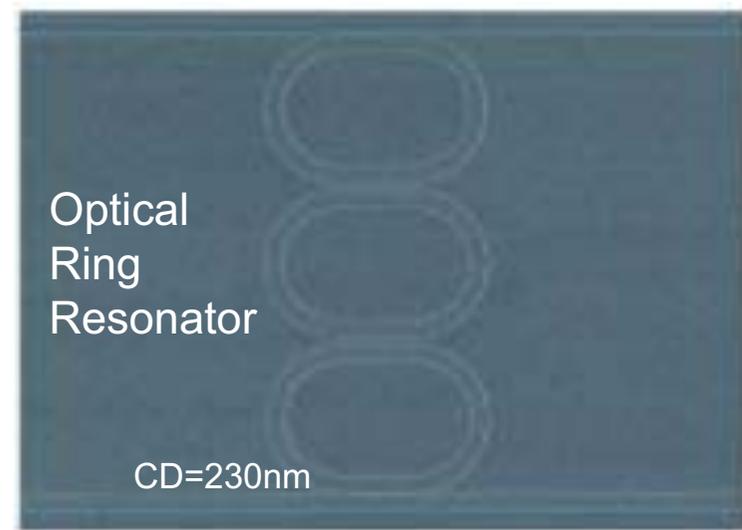
Examples of ZPAL Patterns



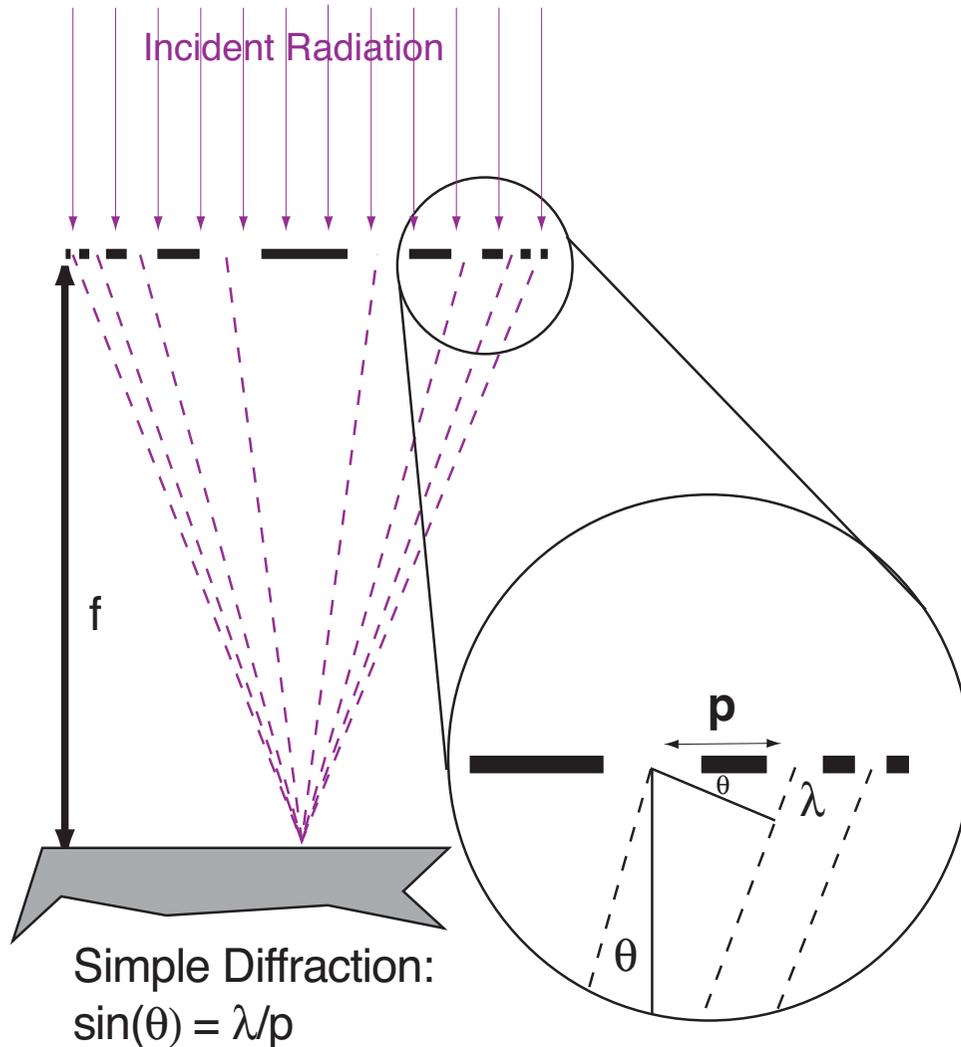
Prototype
MRAM
memory



Array of contact holes

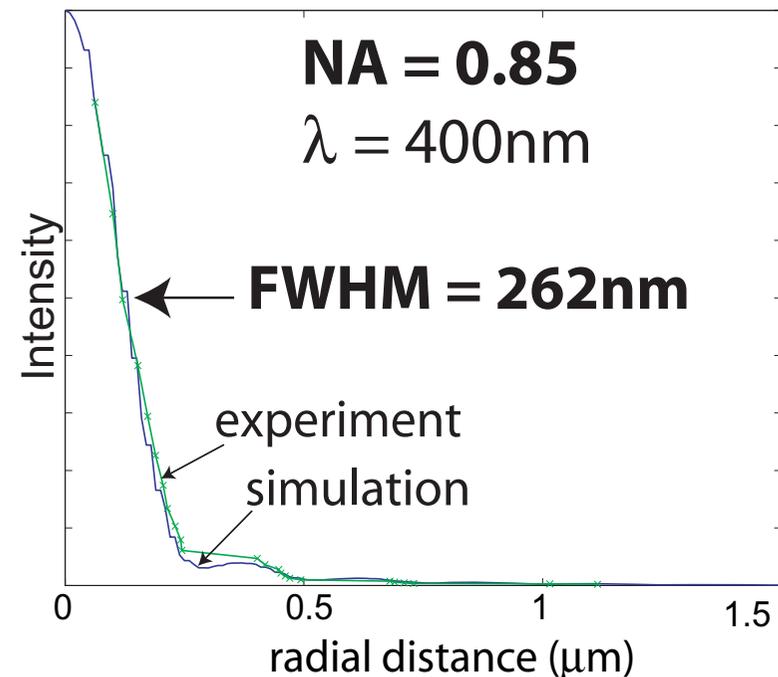


Zone Plate: A Simple Diffractive Lens



Why diffractive optics?

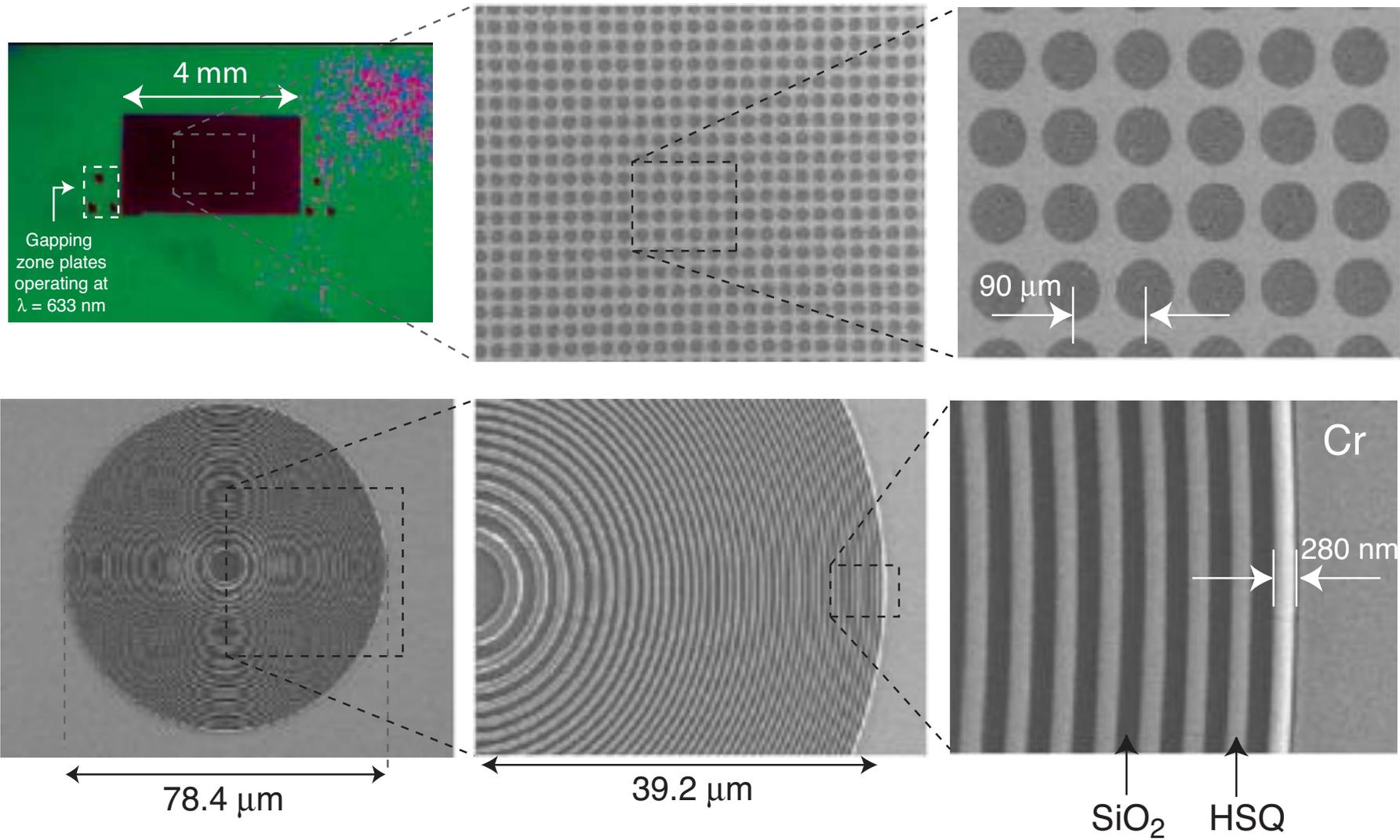
- Abberation-free on-axis.
- High-NA at low cost.
- Fabricated with planar process.
- Focus uniformity across array.
- Wavefront engineering.



Large Zone-Plate Arrays Possible

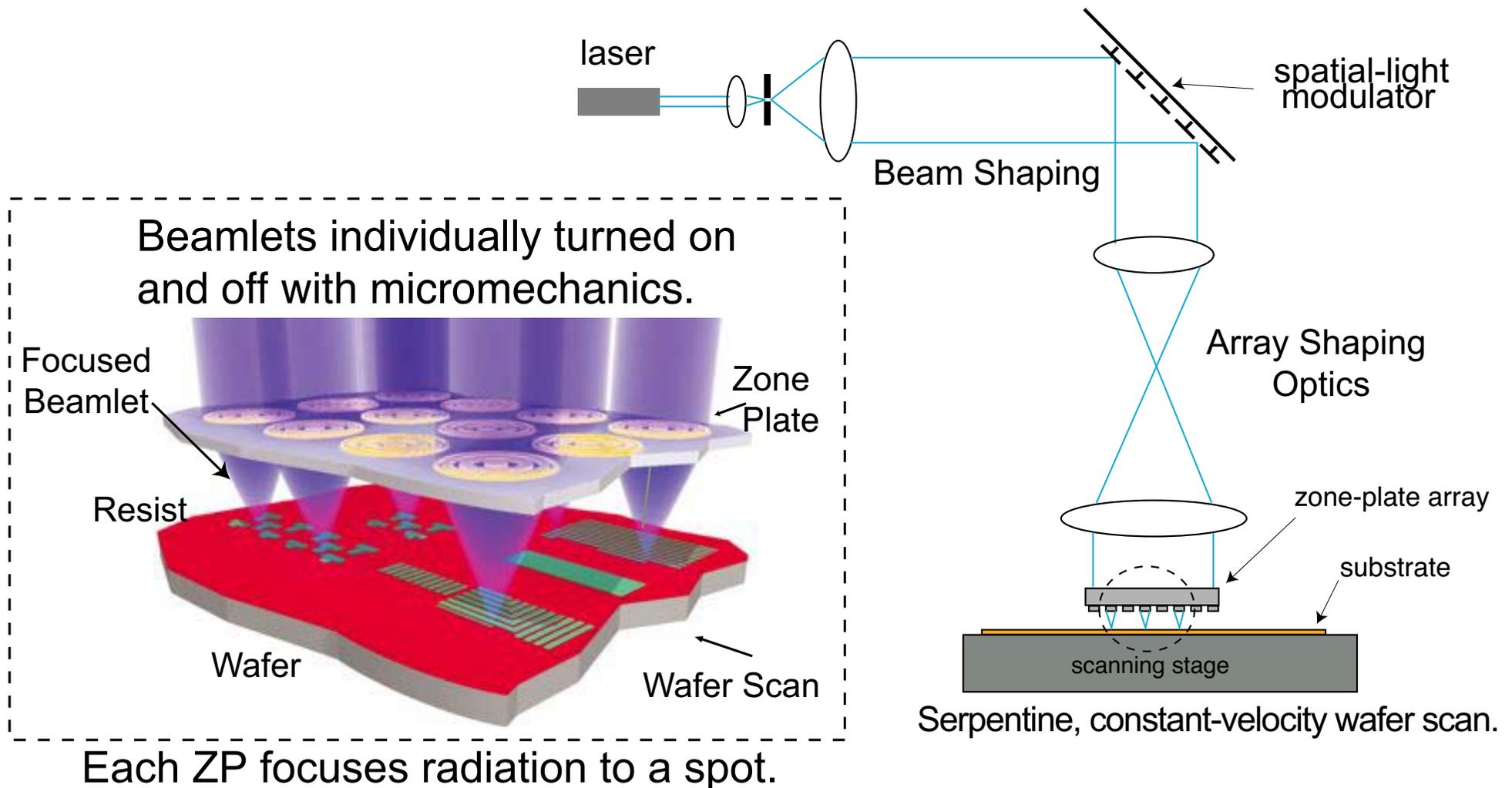
1000 Zone Plates for $\lambda = 400 \text{ nm}$, with $\text{NA} = 0.7$ and Focal Length = $40 \mu\text{m}$

Areal Coverage $\sim 9 \text{ mm}^2$



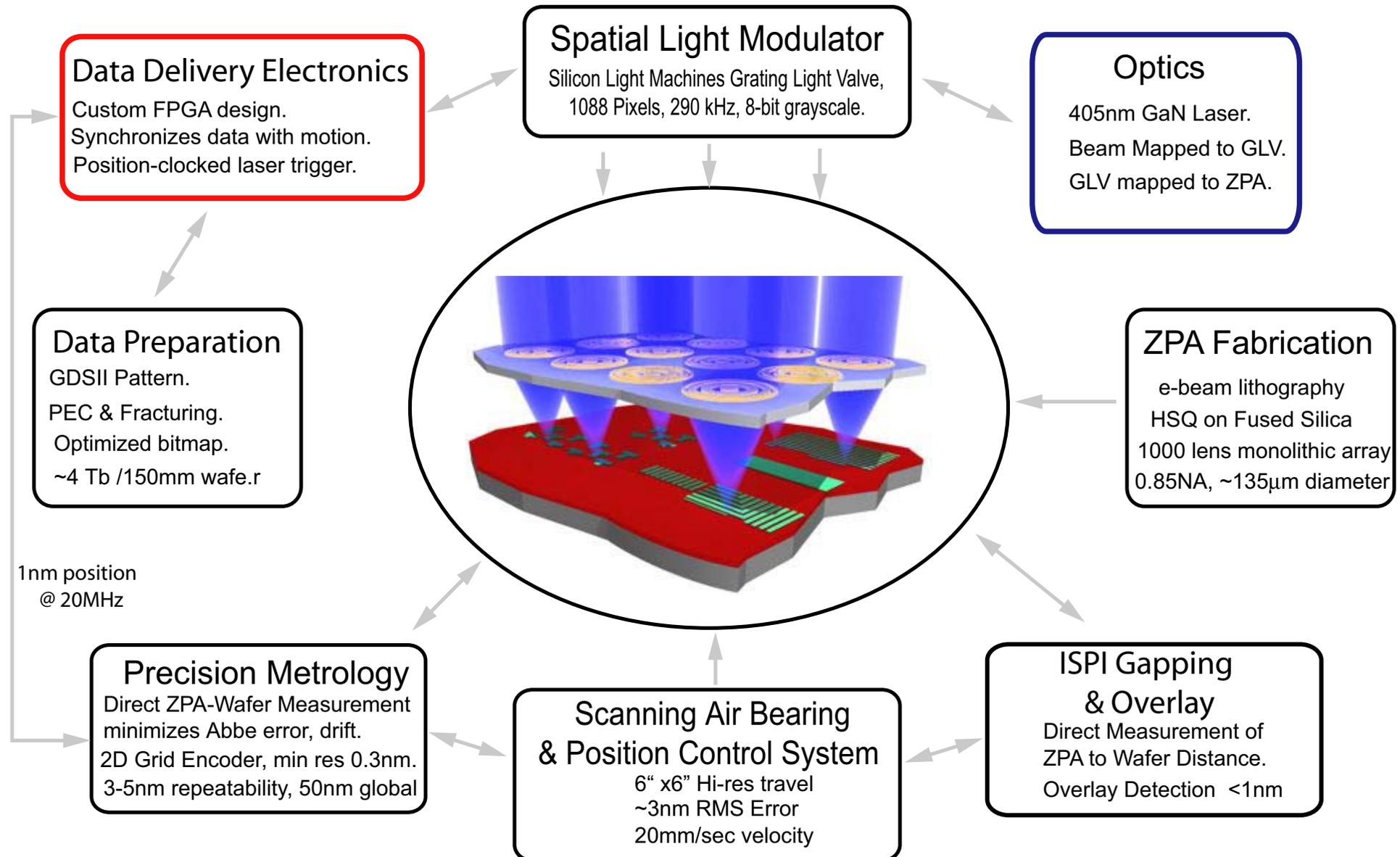
Zone-Plate-Array Lithography

Arbitrary patterns in a dot-matrix fashion as substrates are scanned beneath a fixed array of diffractive microlenses known as zone-plates.



Serpentine, constant-velocity wafer scan.

ZP-150A System Overview



Design for Accuracy

Design

Benefit

Static Lens Array

Monolithic zone-plate array fixes relative positions of all beams on wafer.

- ★ Accurate Stitching
- ★ Loose Tolerances for beams to ZPA.
- ★ Location of beams on wafer determined only by stage position relative to ZPA.

Position-clocked data

Timing of exposure determines location of exposed pixels on wafer

- ★ Only errors normal to scan are printed.
- ★ Position and velocity errors along scan compensated by exposure timing.

Direct Metrology

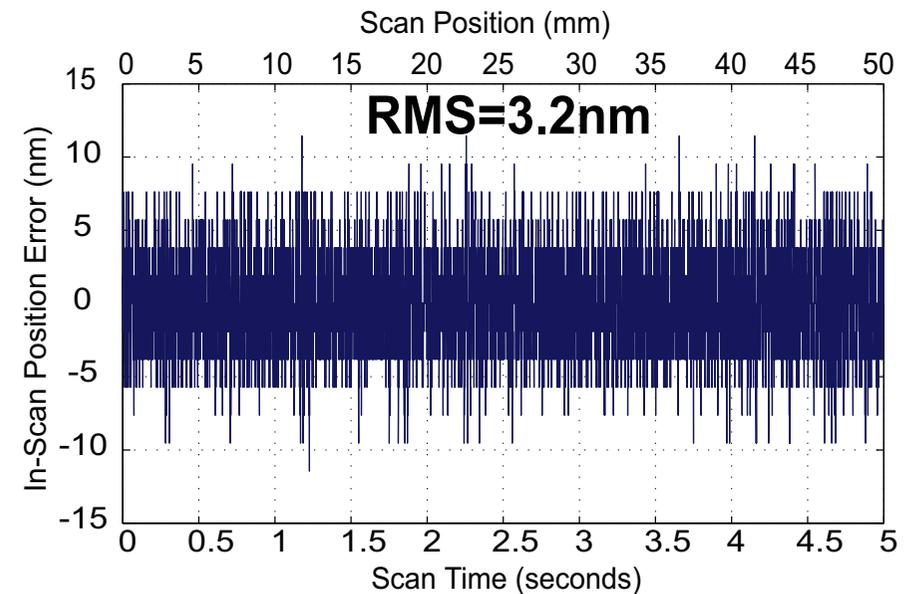
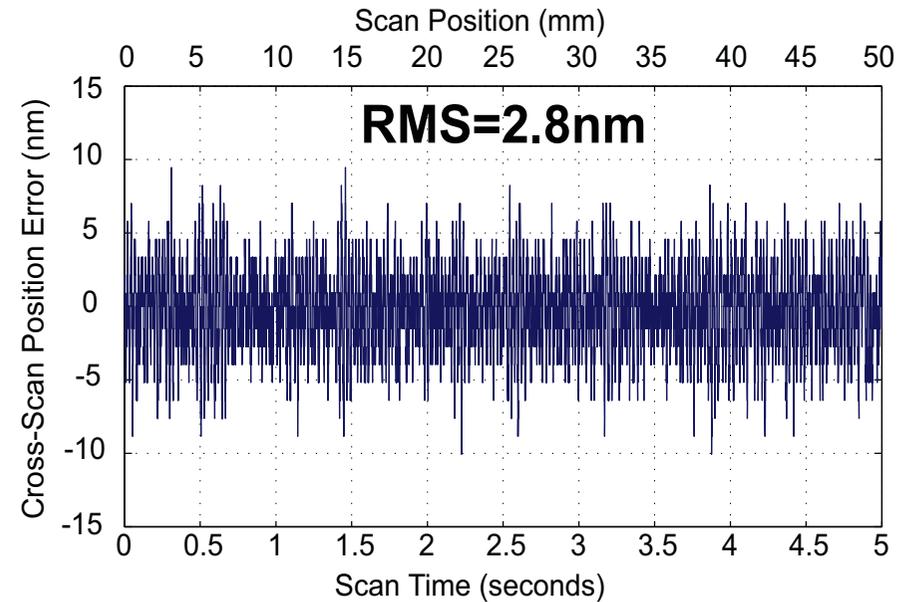
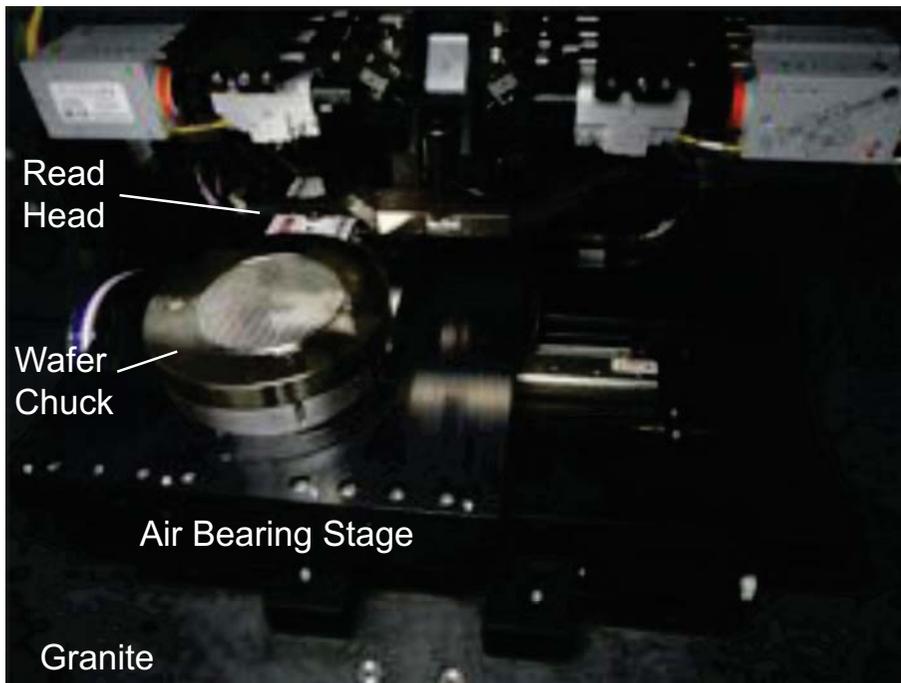
ZPA, wafer chuck integrated in metrology frame with 2D encoder.

- ★ Directly measures ZPA relative to wafer, not to machine frame
- ★ Reduces Abbe error, simplifies overall system.
- ★ More robust than laser interferometer

Scanning System

Only Cross-Scan error, not along scan, contributes to pattern error

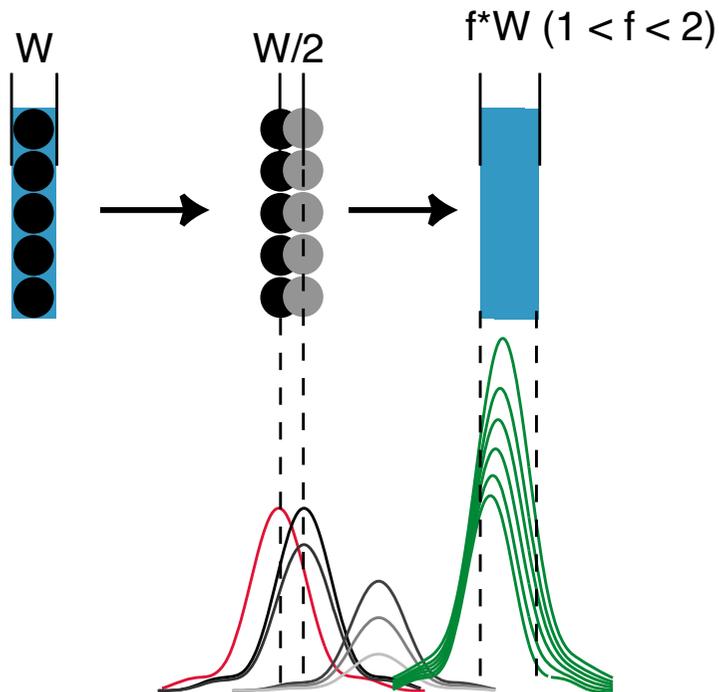
- ◎ Custom XY Air Bearing on Granite Base
- ◎ 1nm resolution at 20MHz
- ◎ 2kHz Control Loop



Pattern Optimization

Proprietary software ensures pattern fidelity, CD linearity by optimizing dose level to every pixel. Also corrects illumination inhomogeneity.

Line-Edge Control

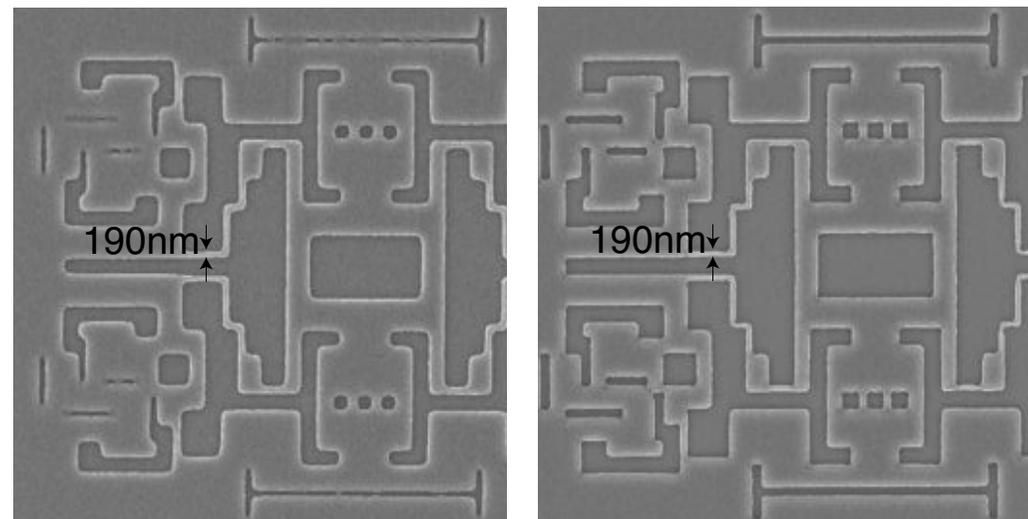


~200 gray-levels for every exposure pixel allows sub-pixel line control

Proximity-Effect Correction

Uncorrected

Corrected

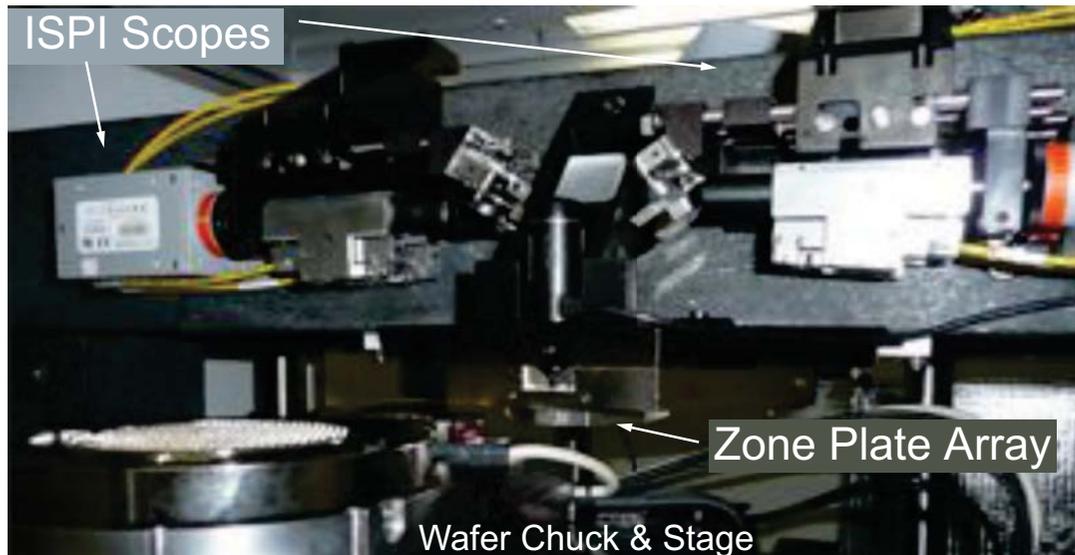
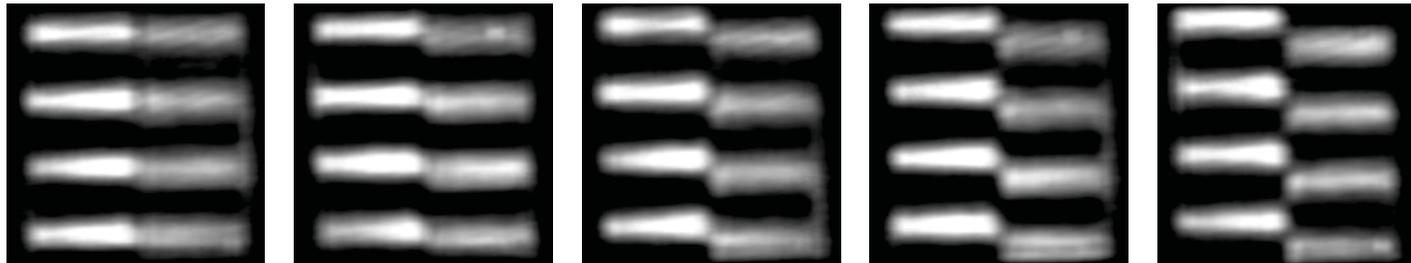


PEC is computationally easier for ZPAL (incoherent) than coherent imaging (e.g. projection litho).

Interferometric Spatial-Phase Imaging

ISPI encodes position in the spatial-phase disparity between a matched pair of interferometric moiré patterns that magnify displacement.

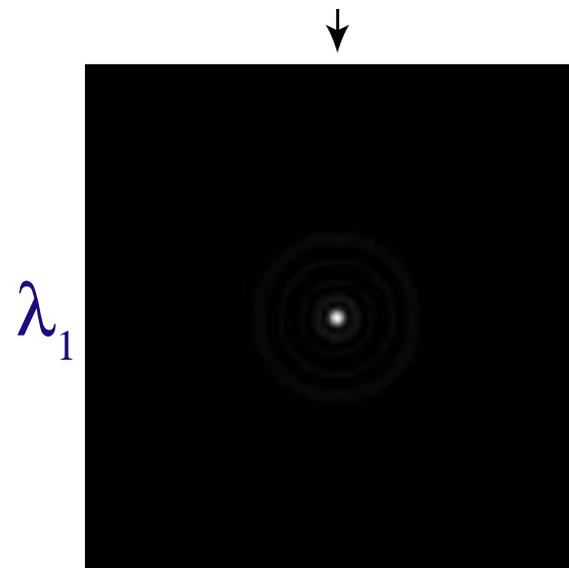
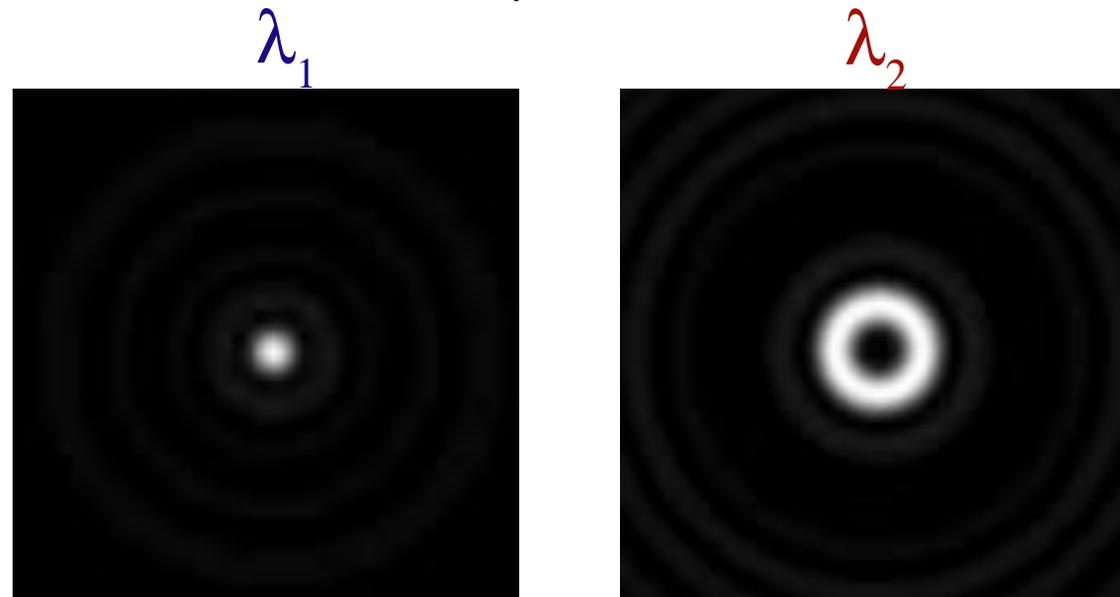
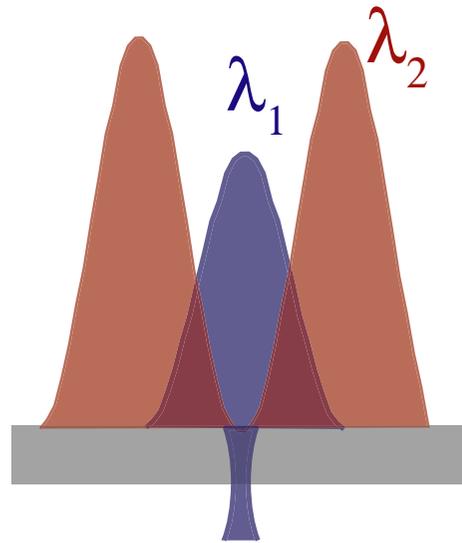
sub-1 nm via
phase-analysis



Benefits of ISPI

- Directly measure working distance.
- Direct ZPA-wafer overlay.
- Dark Field Imaging for High-SNR.
- Low-NA (0.06) optics.
- Robust through multiple layers.

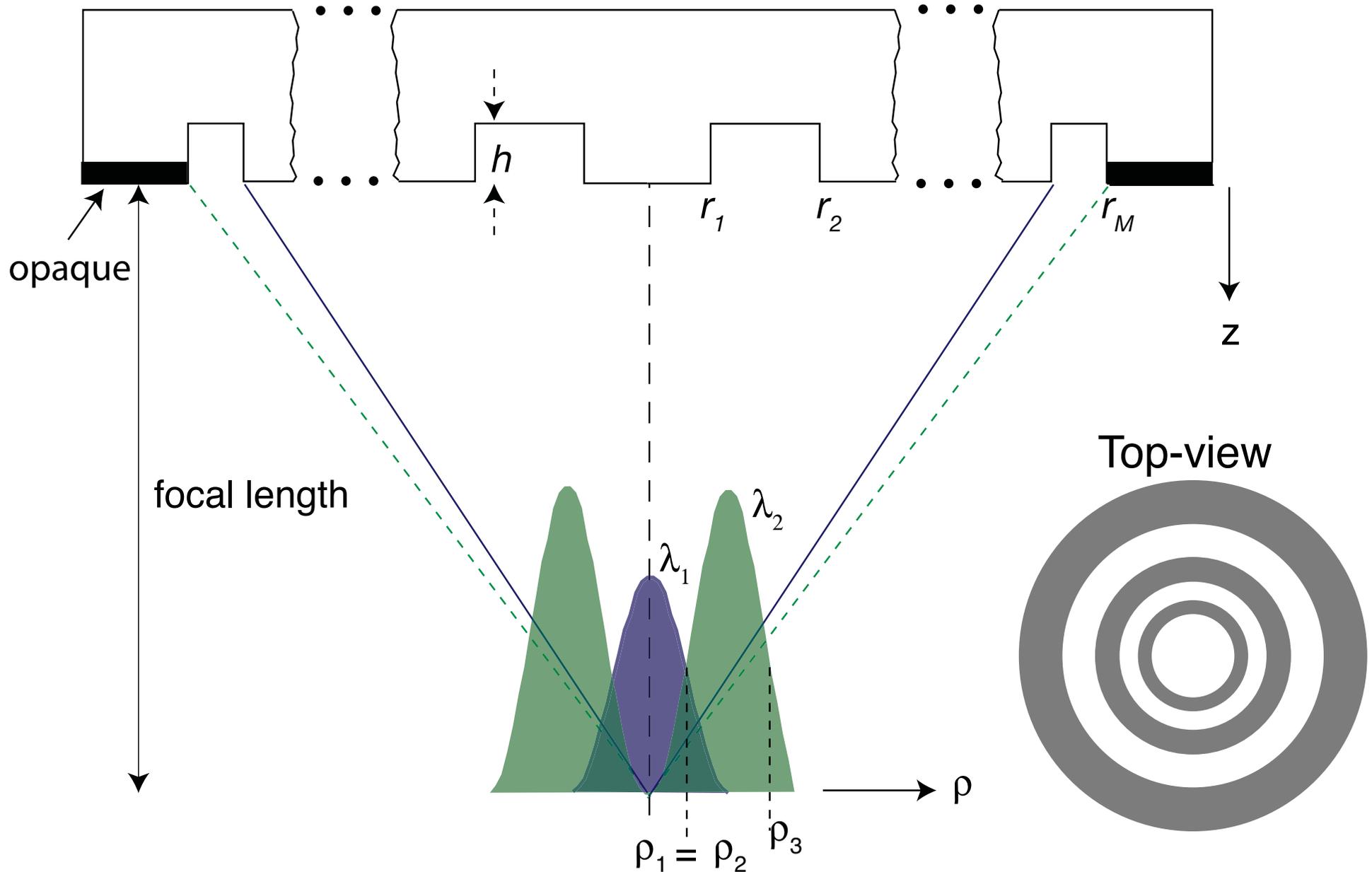
Extension to 2-D requires Ring-shaped spot @ λ_2 &
Round spot @ λ_1



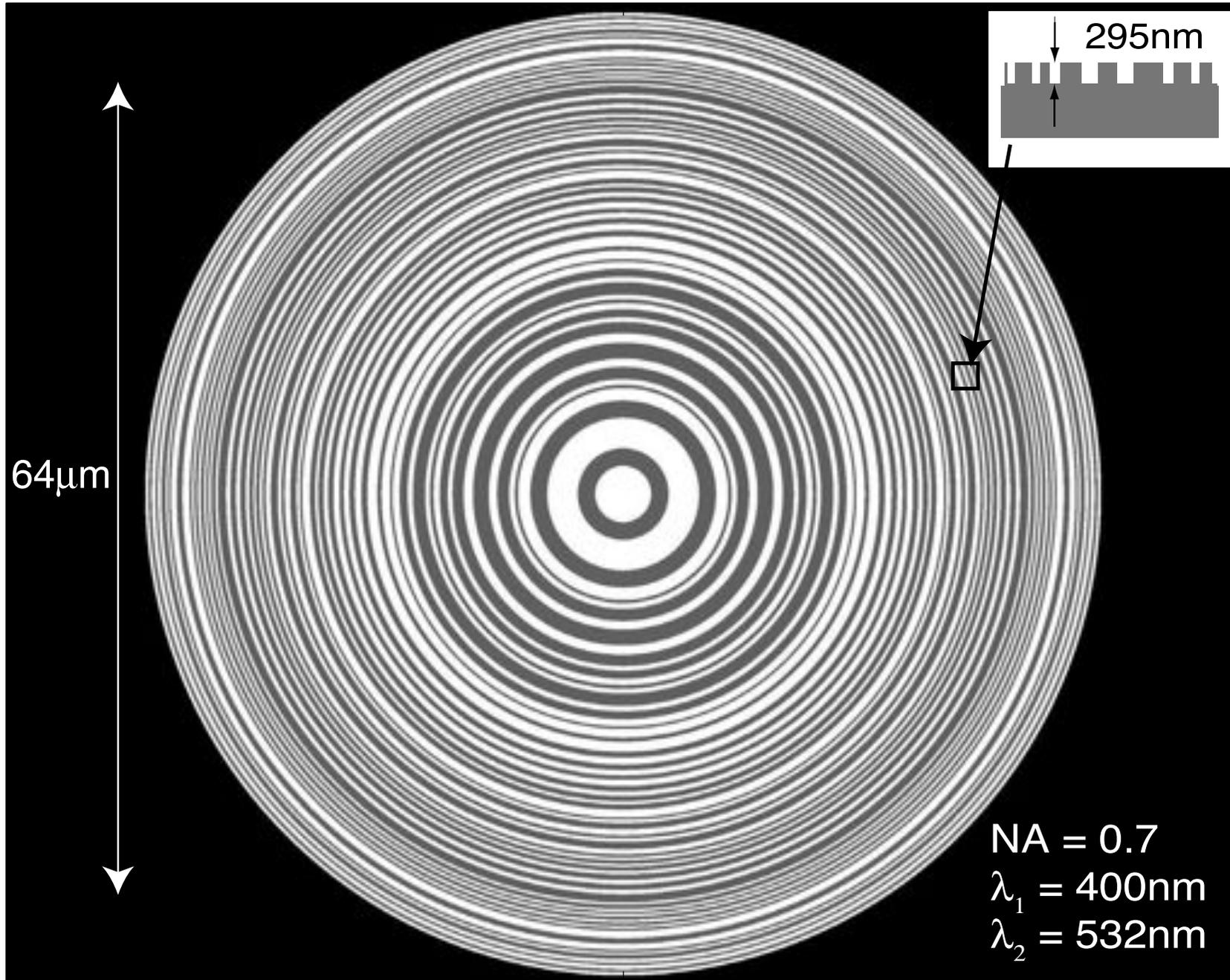
Can we design a single lens for both wavelengths?

Ease of fabrication \longrightarrow binary phase, circular symmetric, diffraction-limited

Cross-section

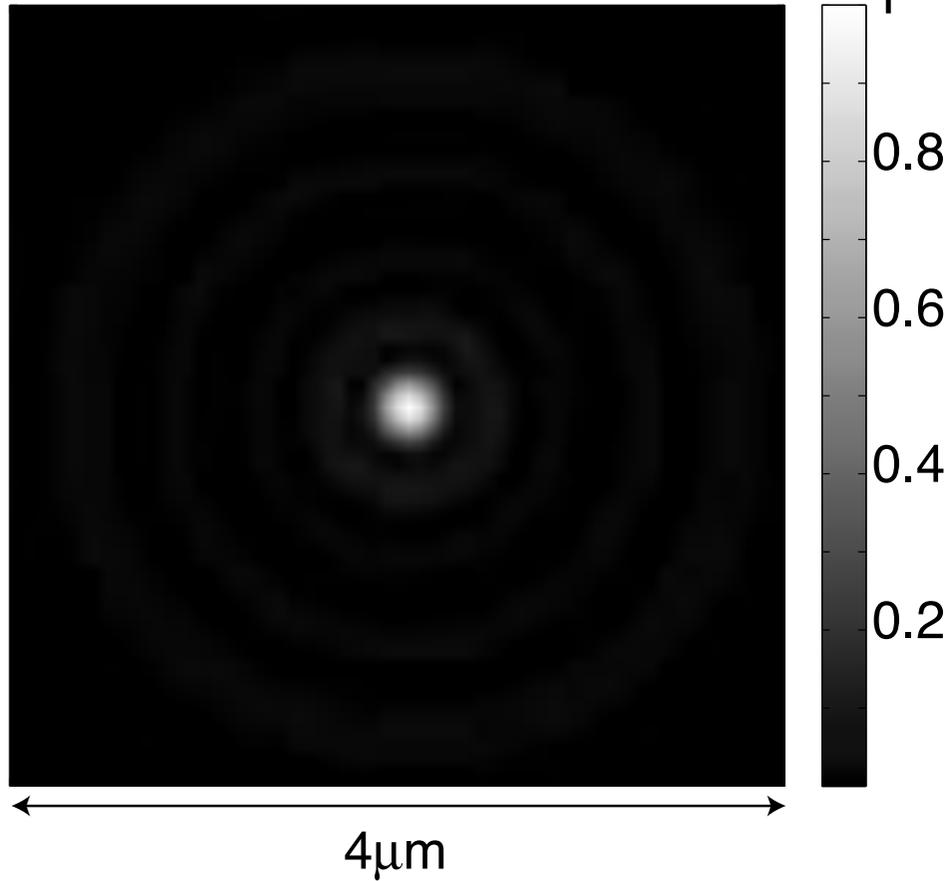


Dichromat: A dual-wavelength phase diffractive lens

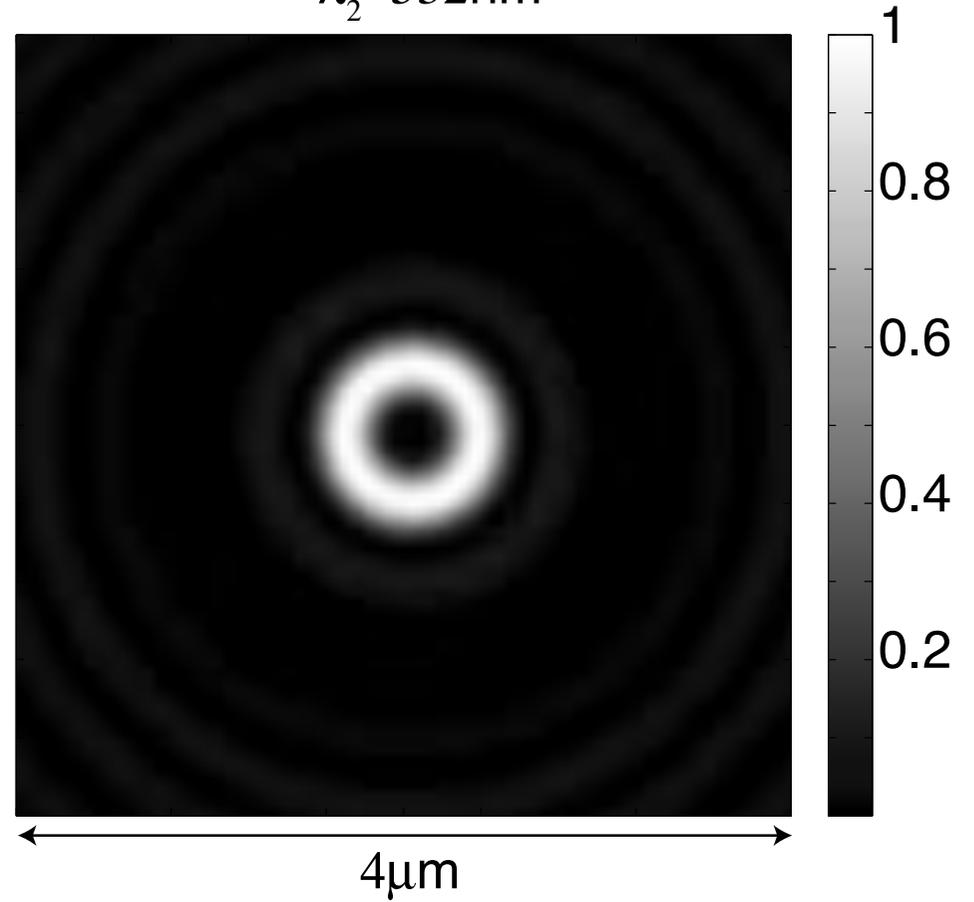


Simulated Focal Spots of the Dichromat

$\lambda_1=400\text{nm}$

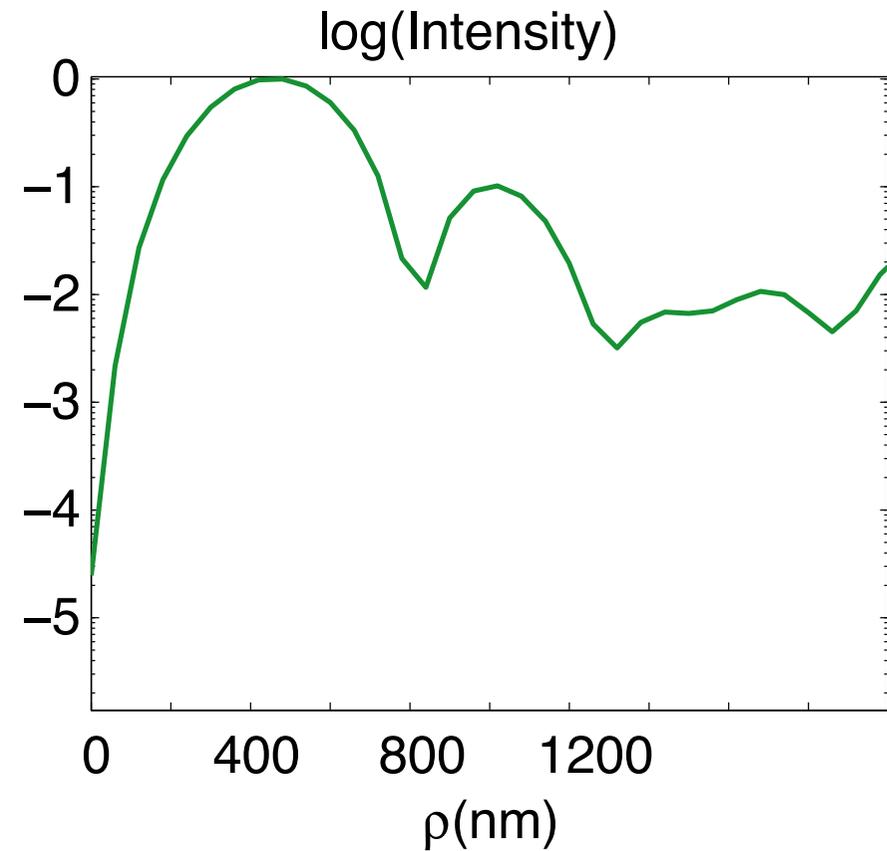
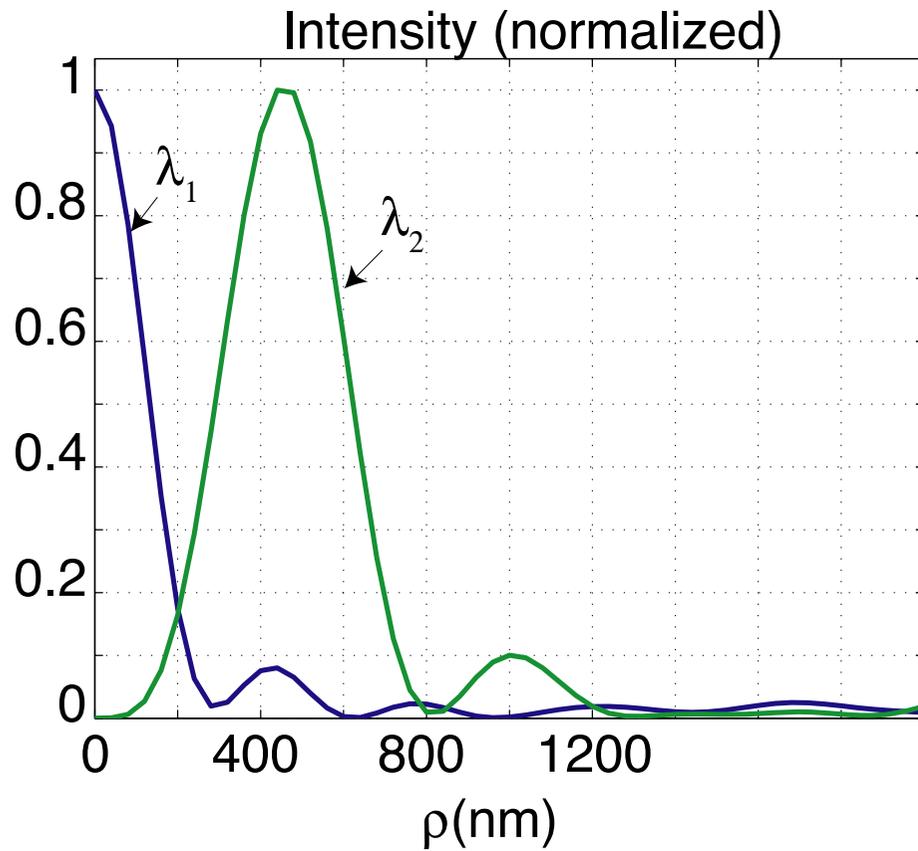


$\lambda_2=532\text{nm}$

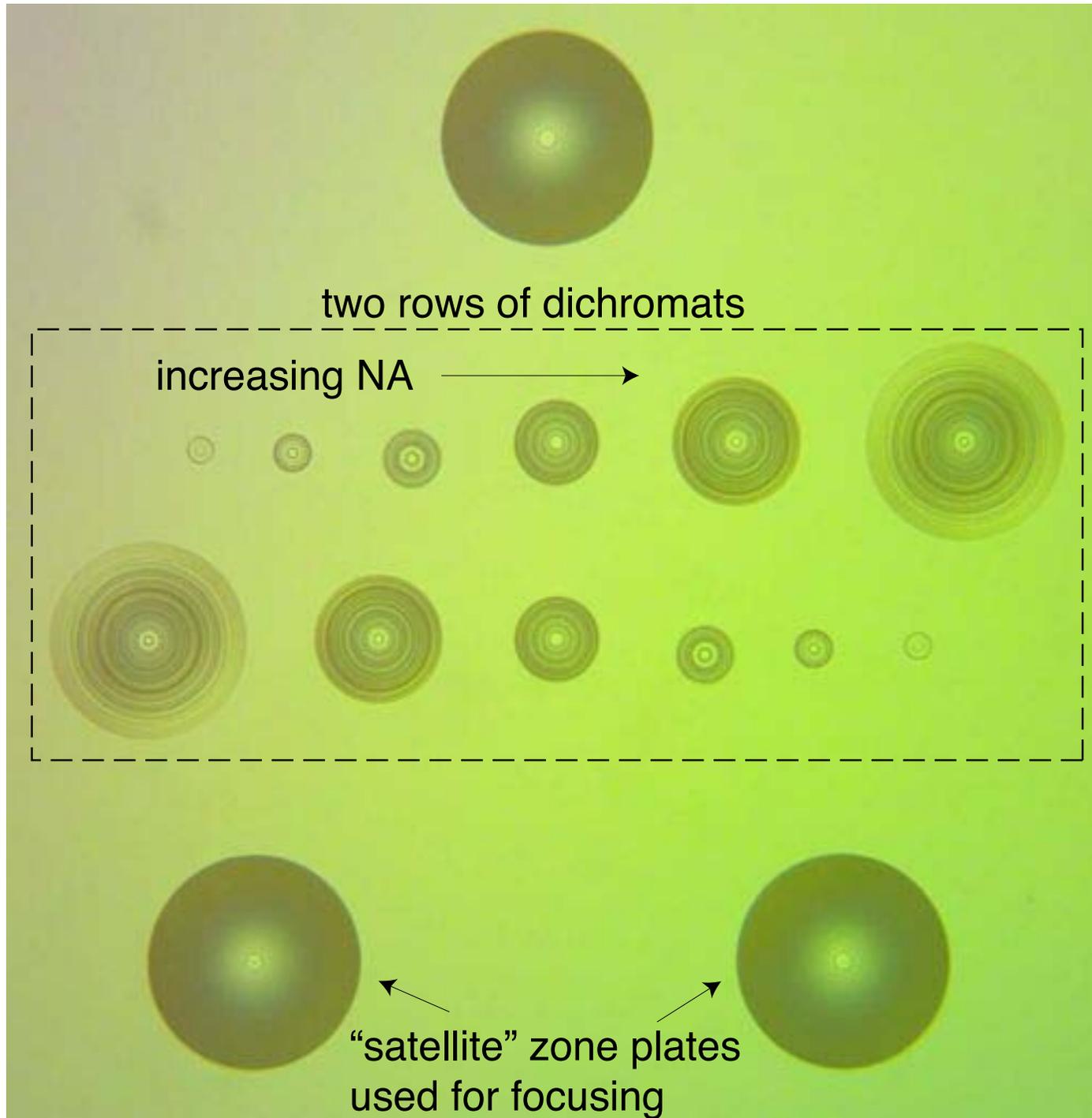


Simulated Focal Spots of the Dichromat

Radial cross-sections



Fabricated Dichromats



Fabricated Dichromats

$\lambda_1 = 400\text{nm}$
 $\lambda_2 = 532\text{nm}$
 $f = 40\mu\text{m}$



NA = 0.7



NA = 0.83

NA = 0.42



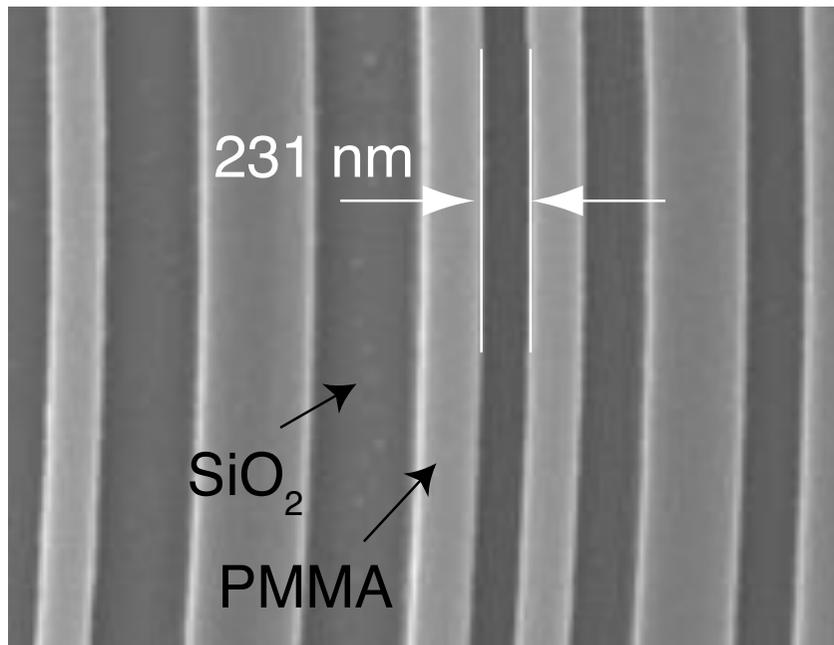
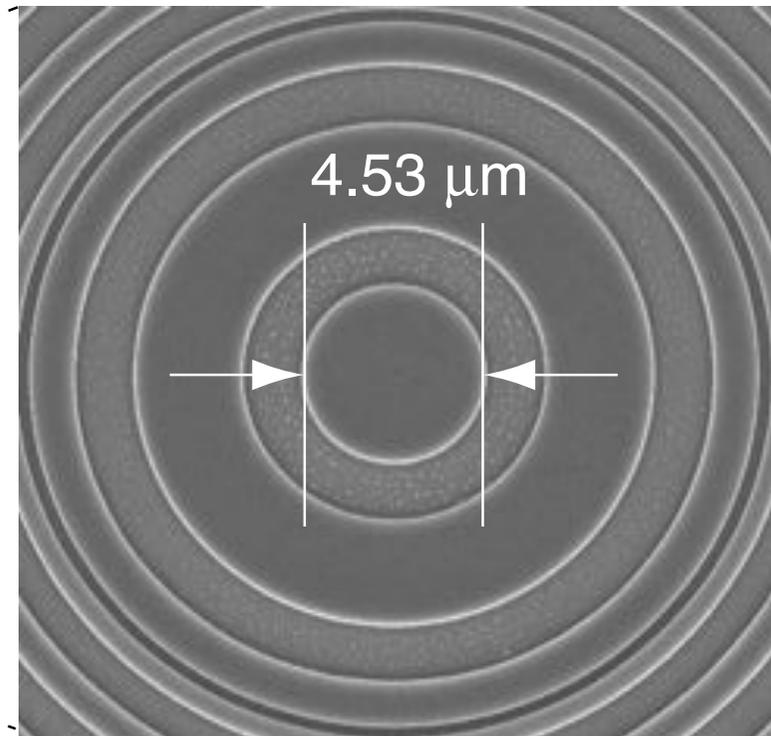
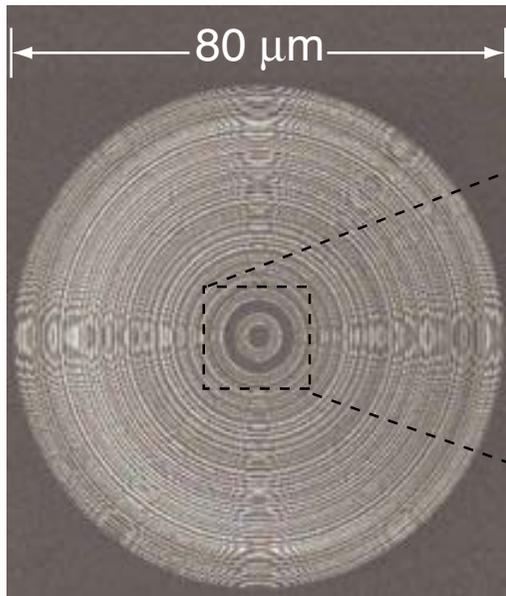
NA = 0.30



NA = 0.22



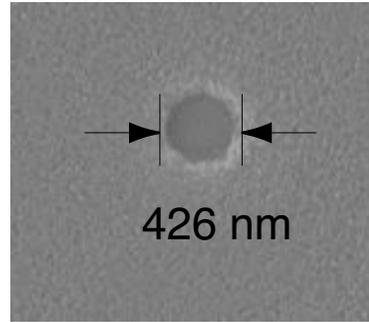
Fabricated Dichromat



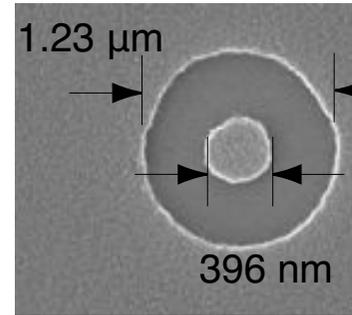
- Patterned with scanning-electron-beam lithography (SEBL)
- PMMA on glass substrate
- $\lambda_1 = 400\text{nm}$, $\lambda_2 = 532\text{nm}$, NA = 0.7

Characterizing the Dichromats

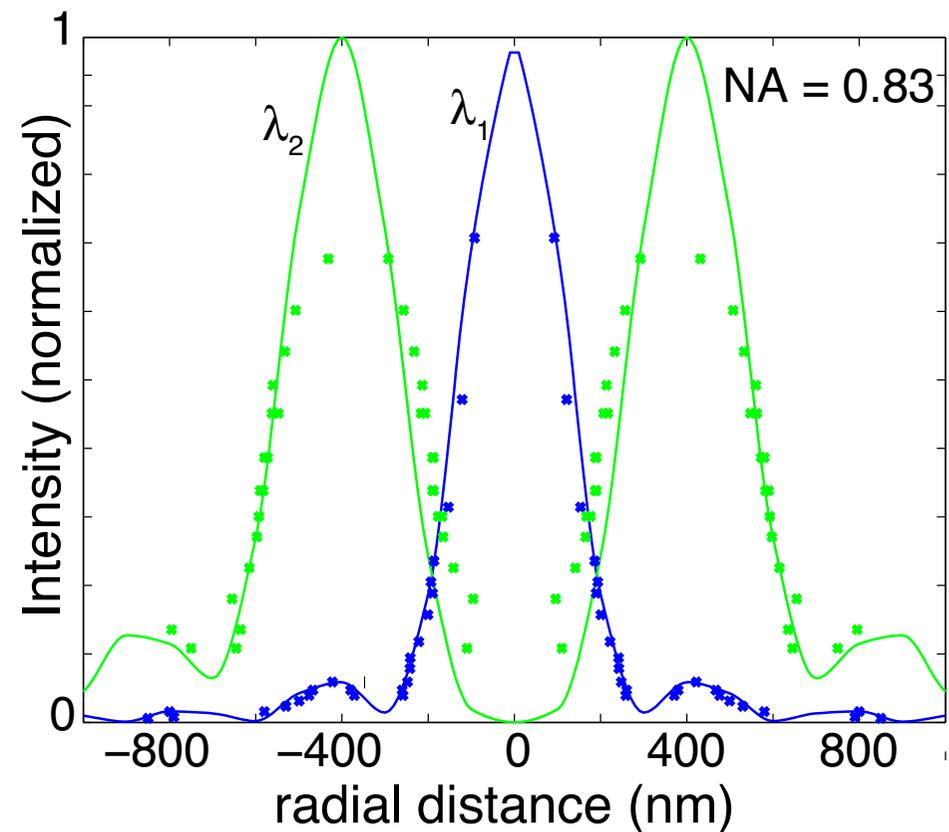
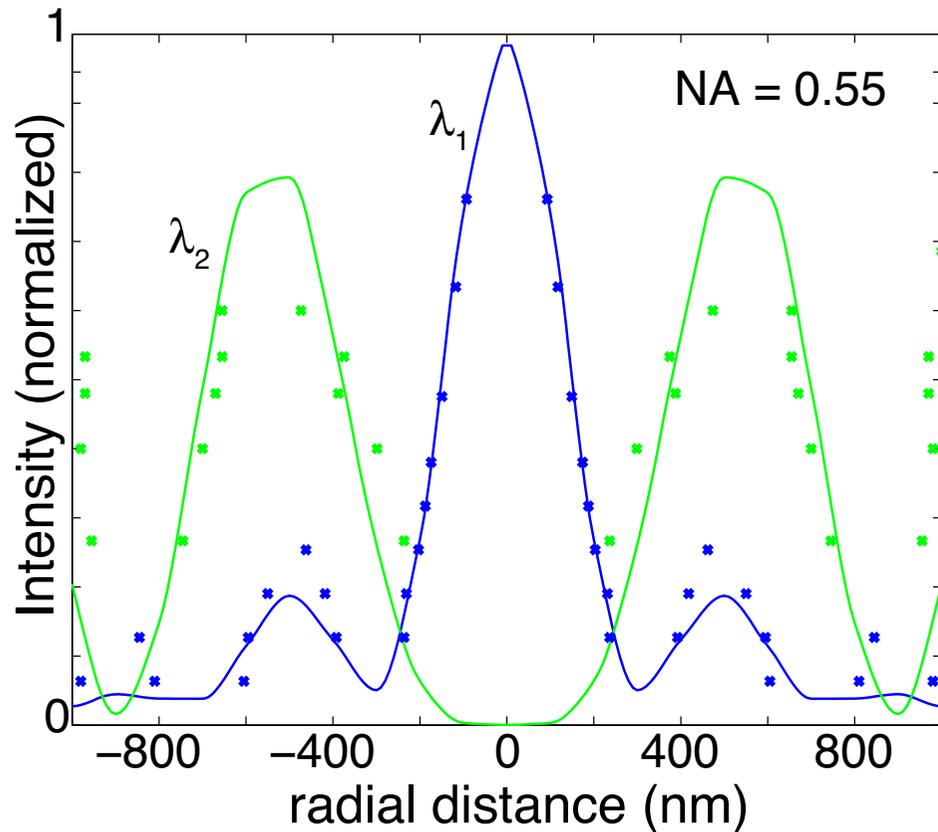
Focal spot at λ_1



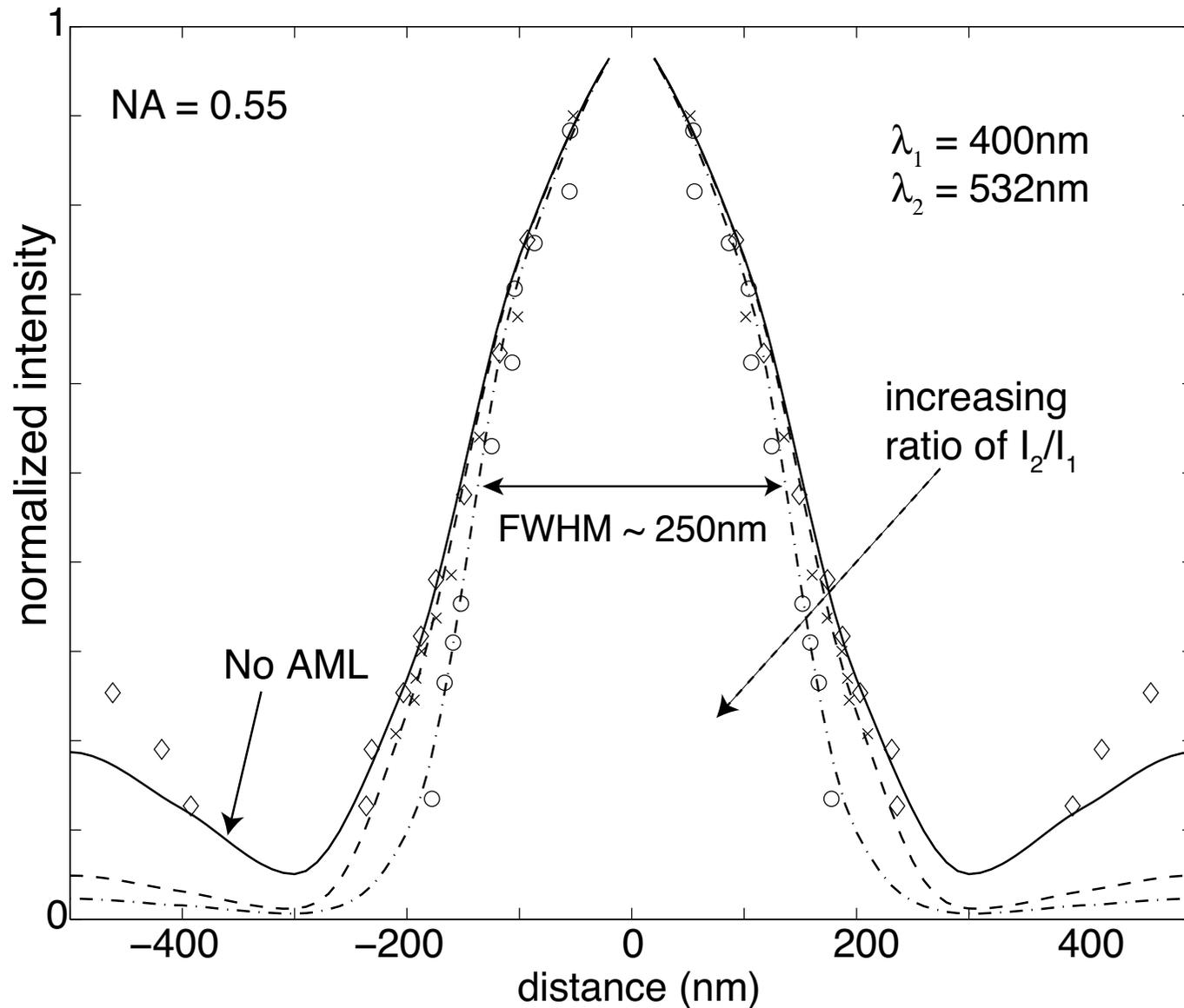
Focal ring at λ_2



Measured Point-Spread Functions



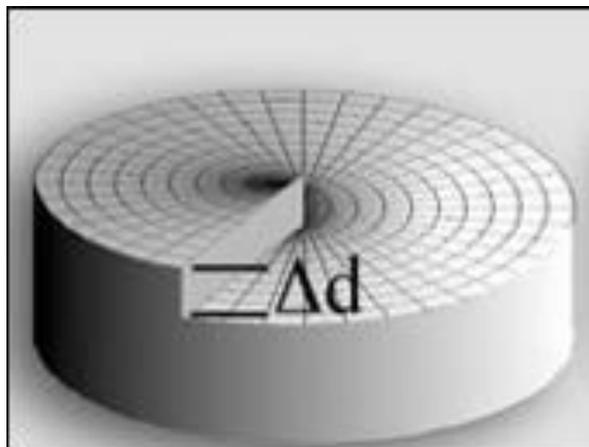
“Squeezed” spot with Dichromat



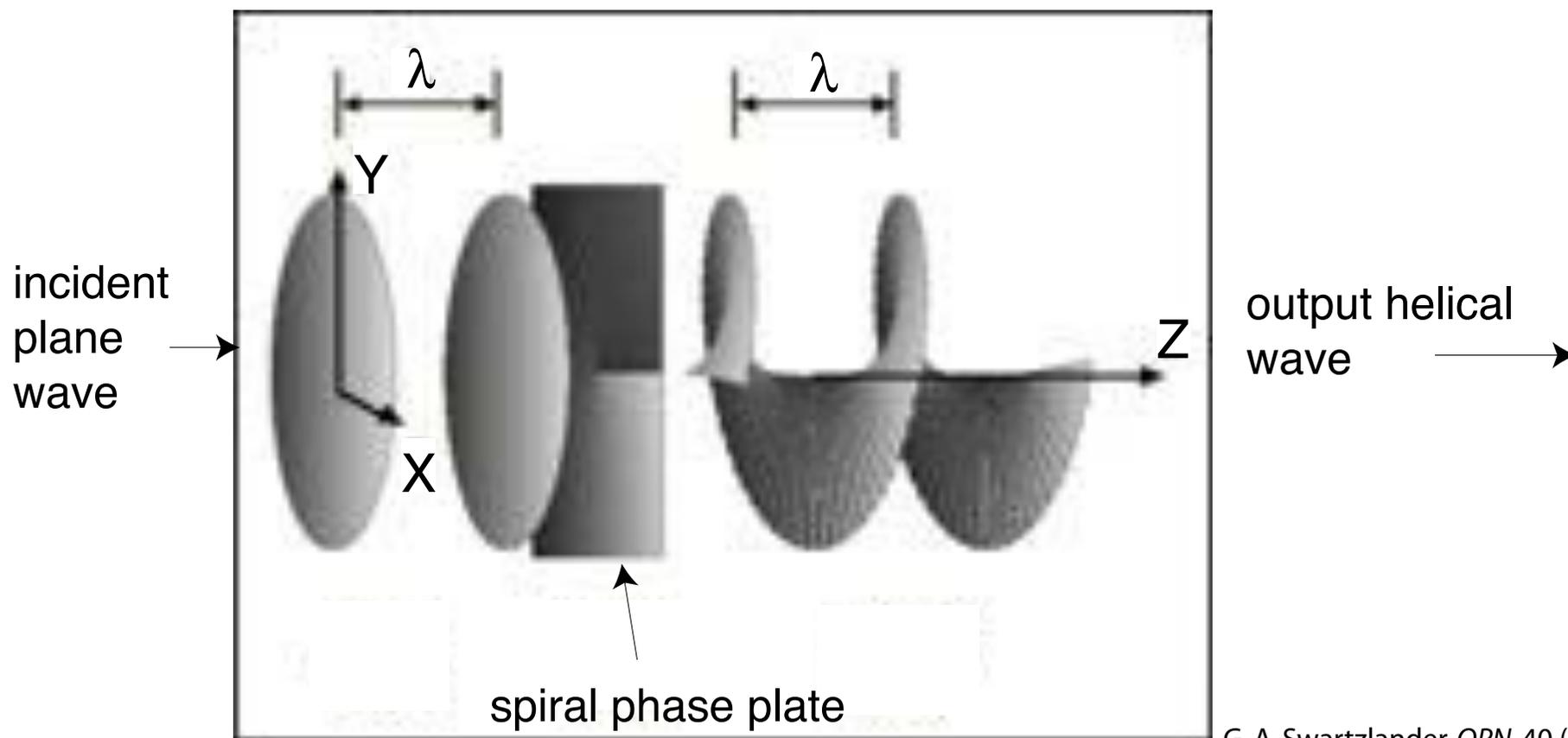
AML = 200nm of azobenzene polymer

Thermal instability of the azobenzene & the sensitivity of the photoresist to λ_2 limit “squeezing.”

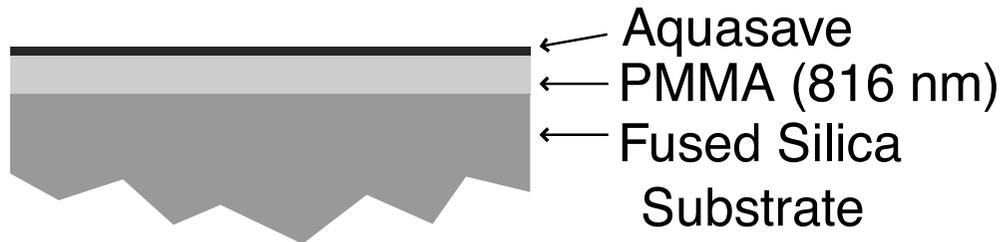
Phase singularities (optical vortices) generate dark beams



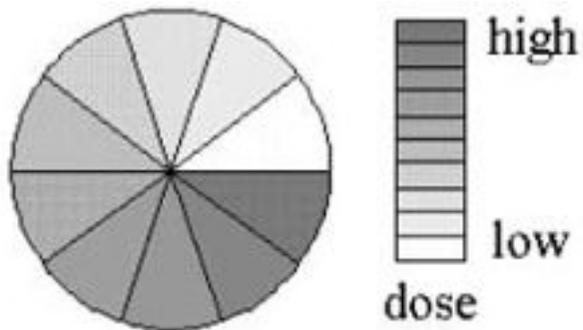
Spiral Phase Plate of topological charge 1



Spiral-Phase Plates (SPPs) are hard to fabricate



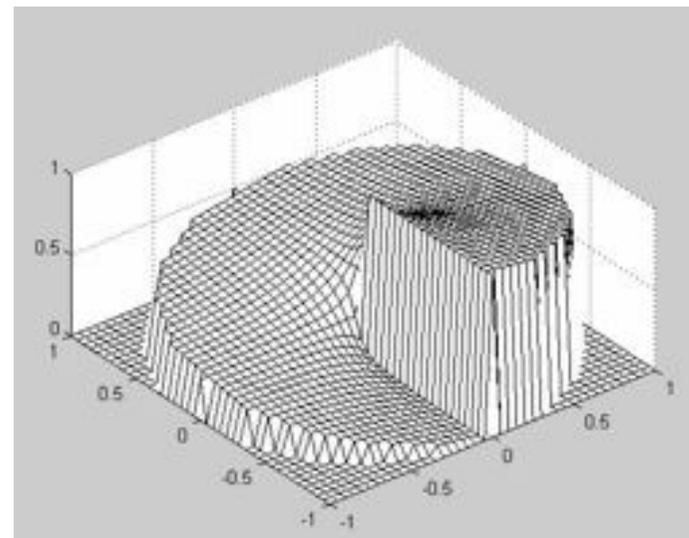
Grayscale E-beam exposure



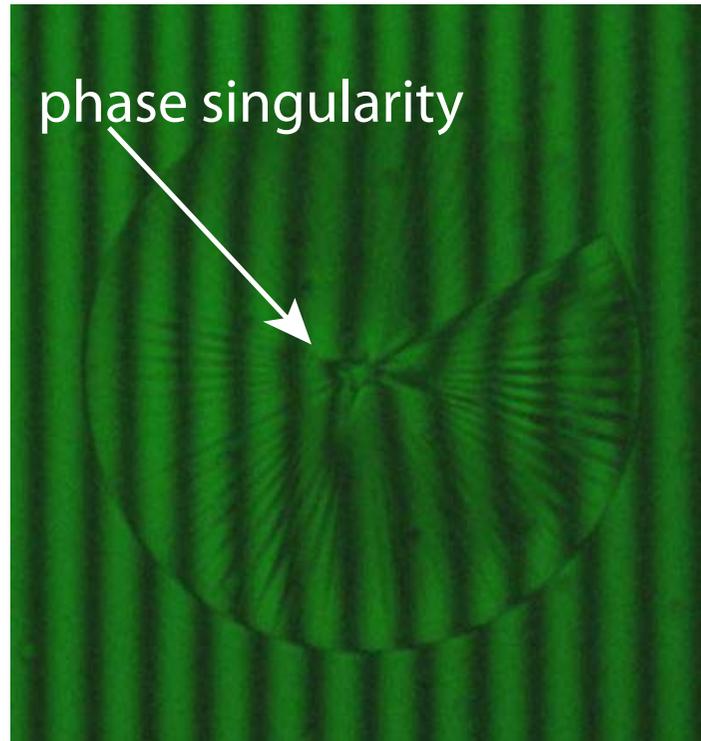
Low Contrast Development
with 1:1 MIBK/IPA



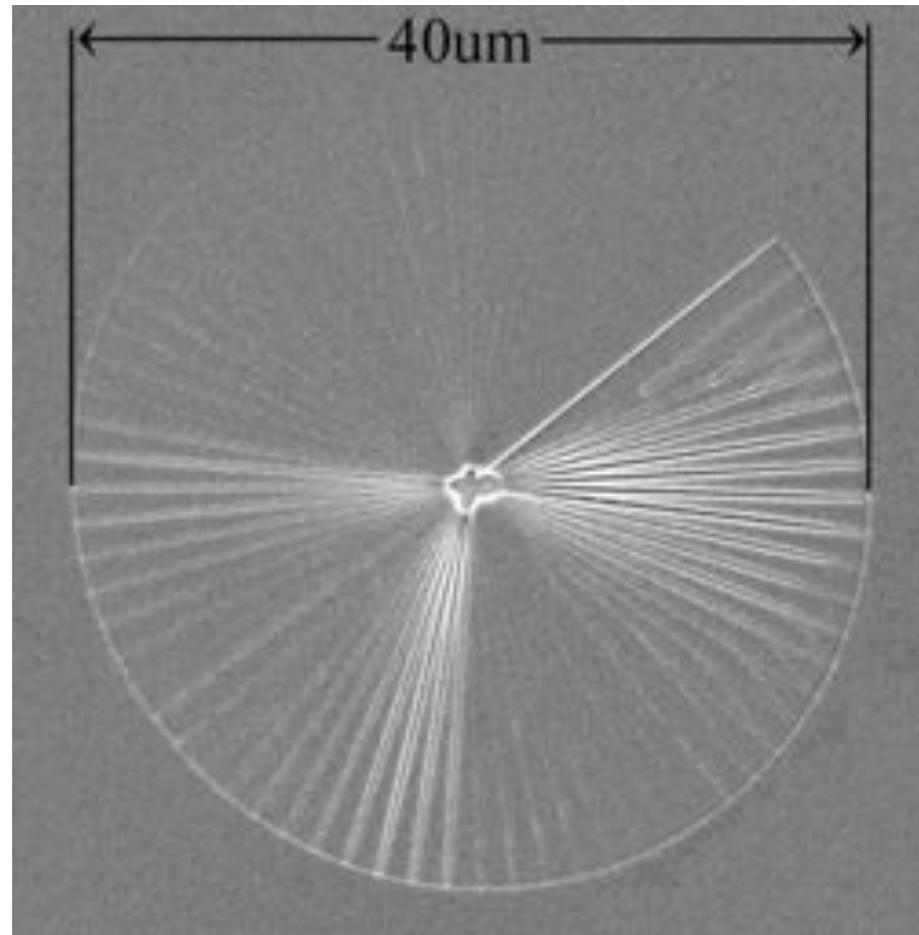
- Spiral Phase Plate (SPP) with grayscale scanning e-beam lithography (SEBL)



Spiral-Phase Plates (SPPs) are hard to fabricate



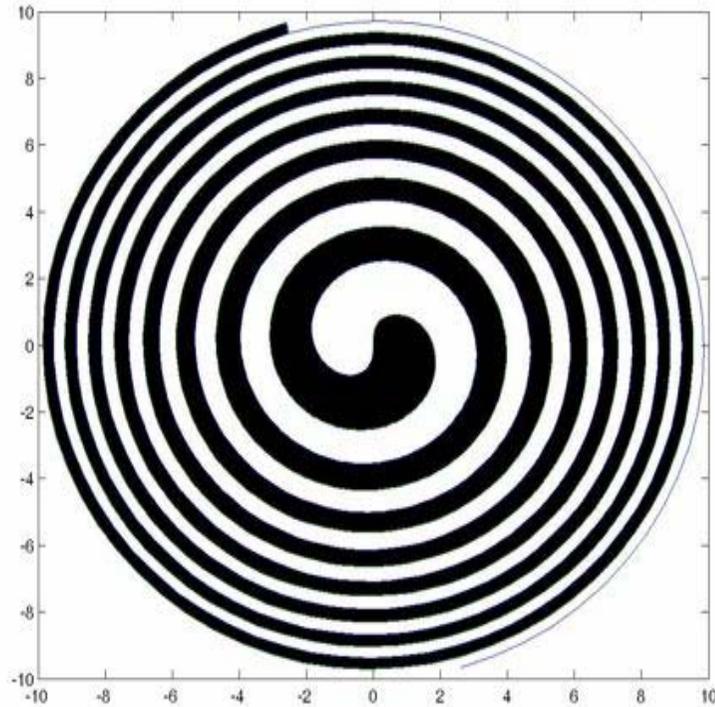
- Linnik interferogram of fabricated SPP



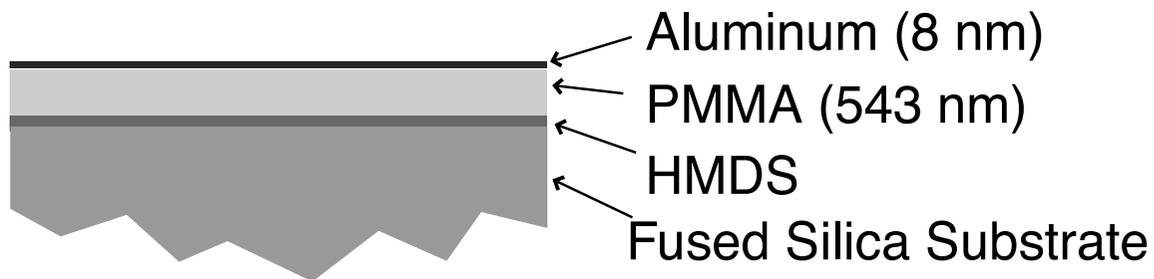
- SEM of a fabricated SPP

Proximity Effects due to backscattered electrons wreak havoc!

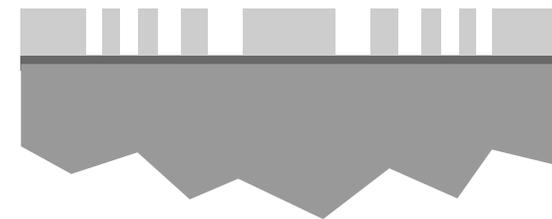
Spiral-Zone-Plate (SZP) is easier to fabricate



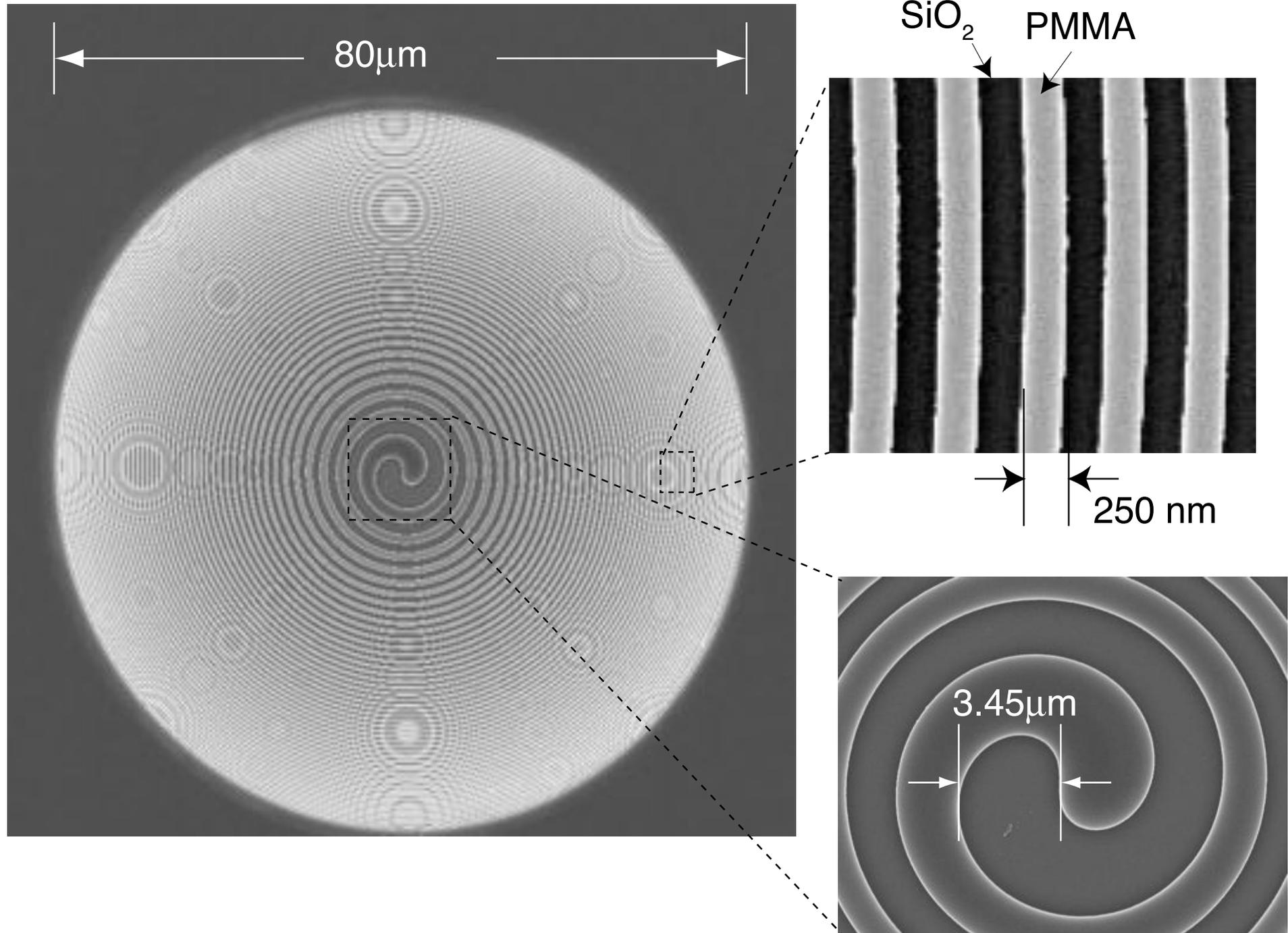
Binary phase spiral-zone plate (SZP) generates a ring-focus.



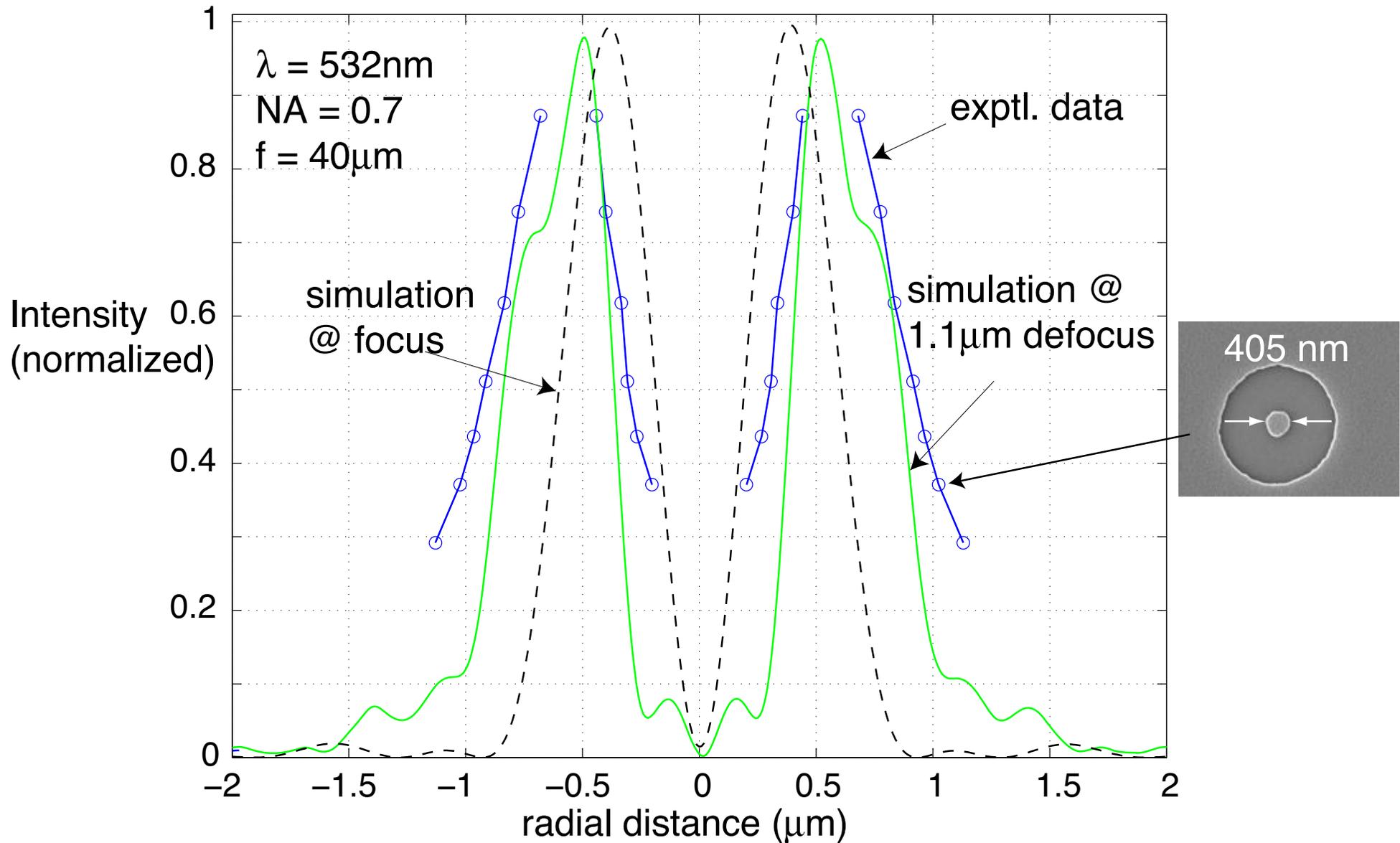
E-beam Pattern PMMA
Develop with 1:3 MIBK/IPA



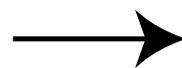
A fabricated Spiral-Zone-Plate (SZP) of NA = 0.7



Characterizing the Spiral-Zone-Plate of NA = 0.7

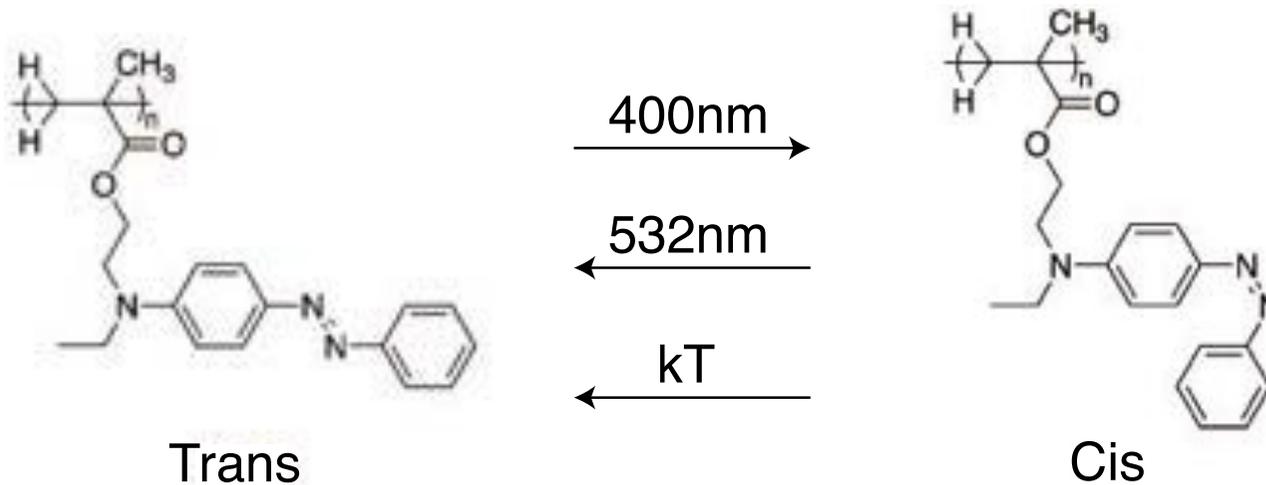


- Focus errors during exposure
- Fabrication errors in phase shift & zone duty cycle



larger ring-diameter & higher noise in null

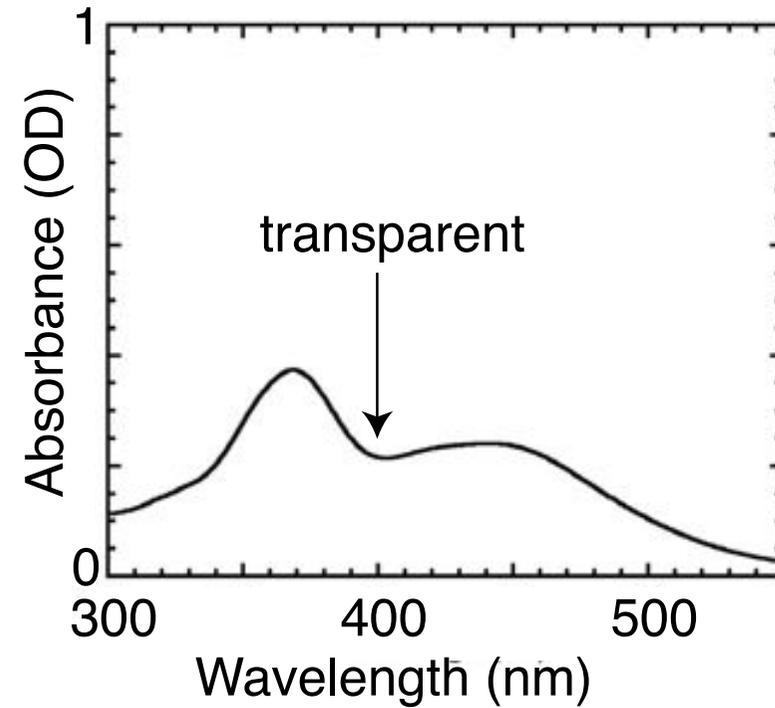
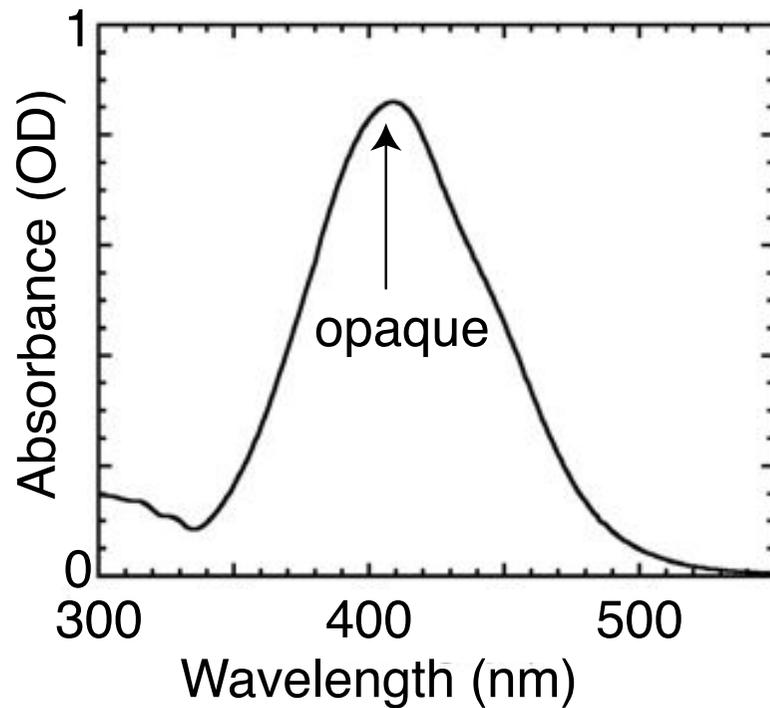
Photochromic molecule: Azobenzene polymer



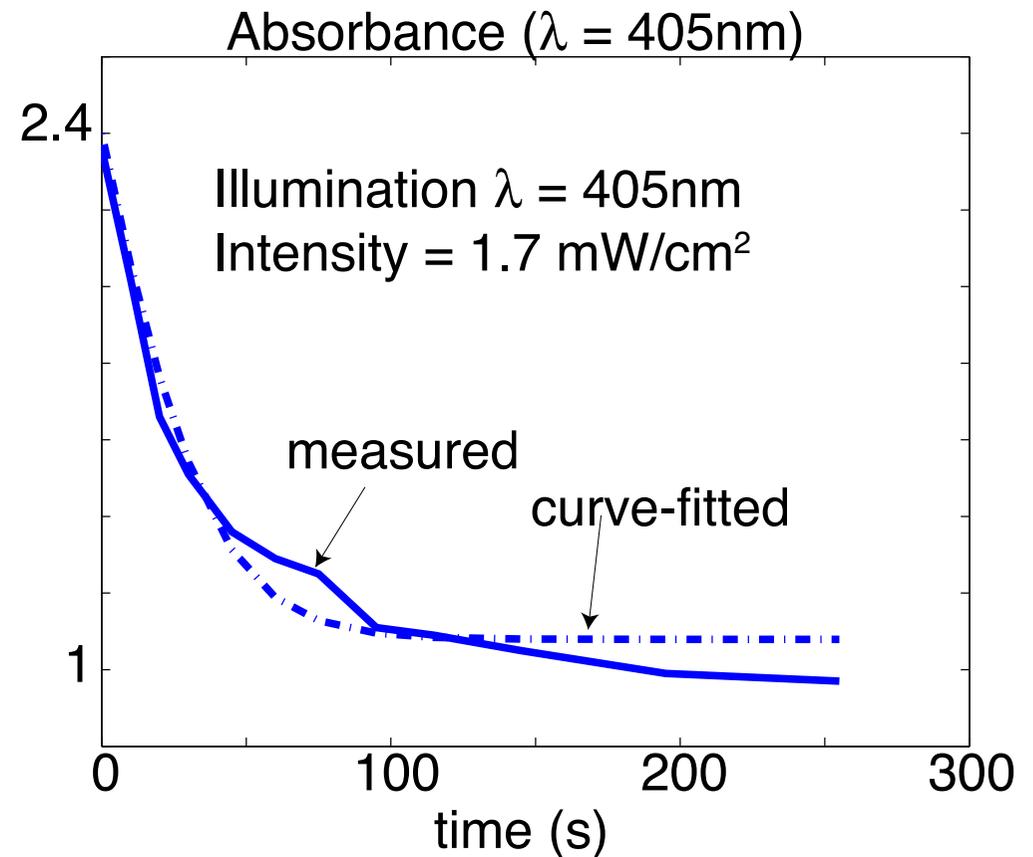
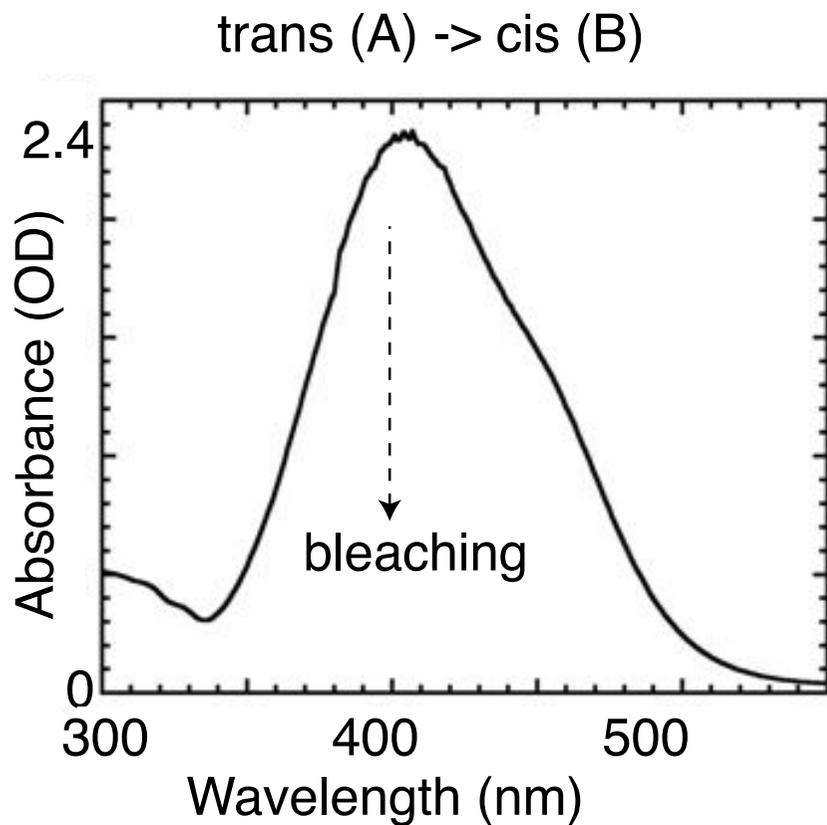
Trans (A)

UV-Vis Spectra

Cis (B)



Extraction of parameters from UV-Vis spectra



- 200nm dye spun on glass substrate

- spectrum measured in transmission

Molar Absorptivities

$$\epsilon_{1A} = 3 \times 10^4 \text{ m}^2 / \text{mol}$$

$$\epsilon_{1B} = 1.25 \times 10^4 \text{ m}^2 / \text{mol}$$

Thermal Rate Constant

$$k_{BA} = 2 \times 10^{-3} \text{ s}^{-1}$$

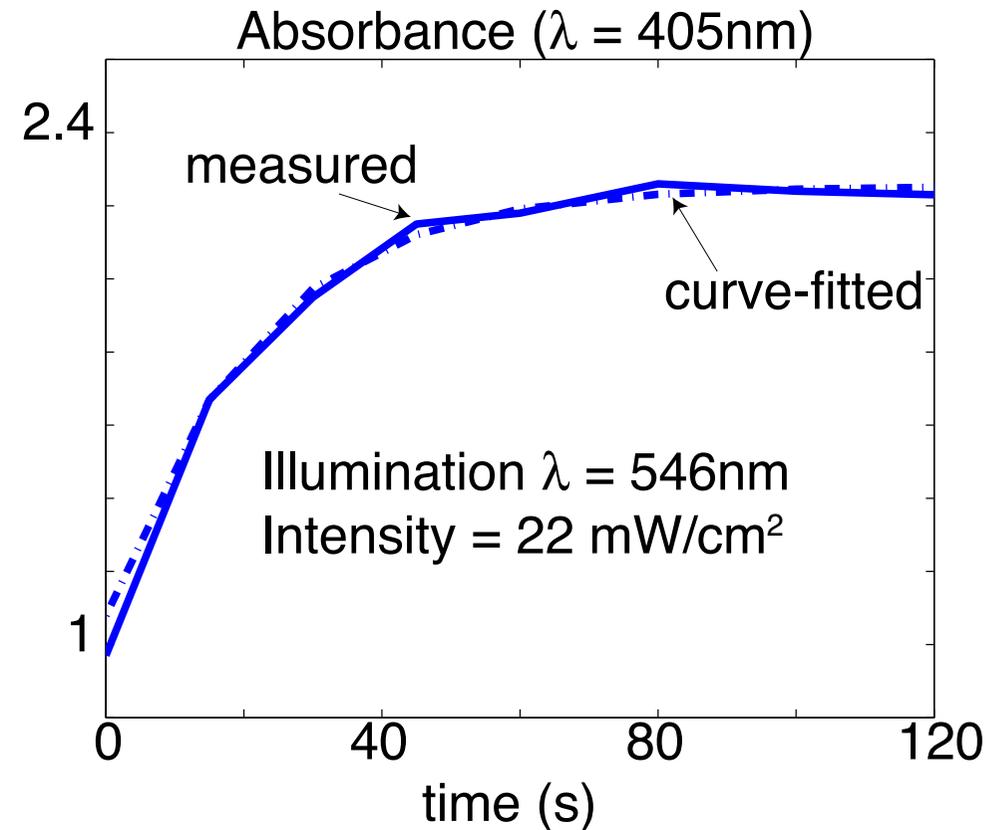
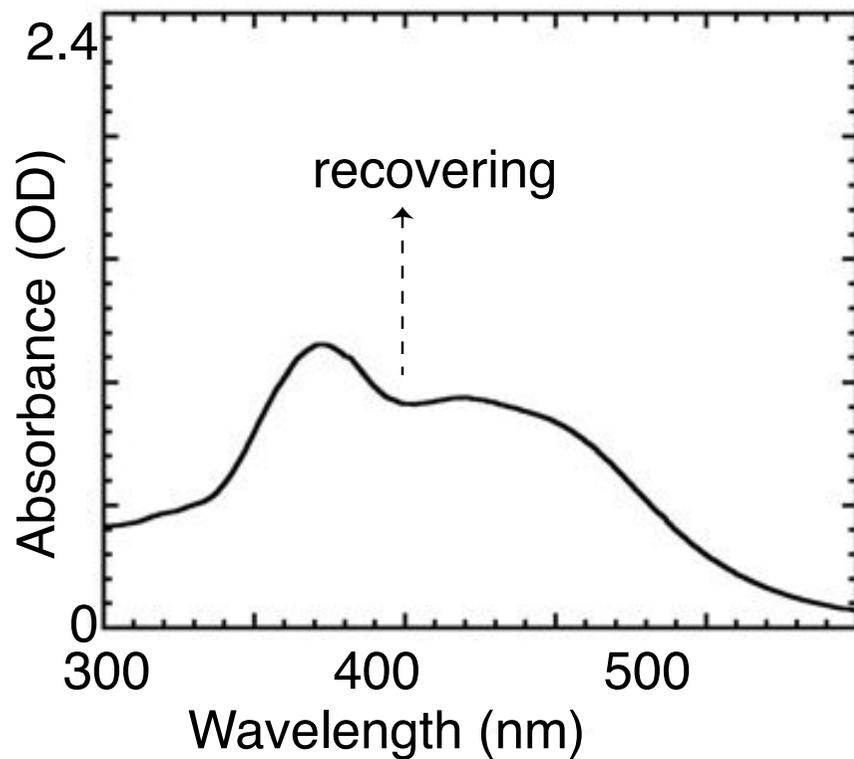
Quantum Efficiencies

$$\phi_{1AB} = 3.7 \times 10^{-3}$$

$$\phi_{1BA} = 2.1 \times 10^{-3}$$

Extraction of parameters from UV-Vis spectra

cis (B) -> trans (A)



- 200nm dye spun on glass substrate

- spectrum measured in transmission

Molar Absorptivities

$$\epsilon_{2A} = 1.25 \times 10^3 \text{ m}^2 / \text{mol}$$

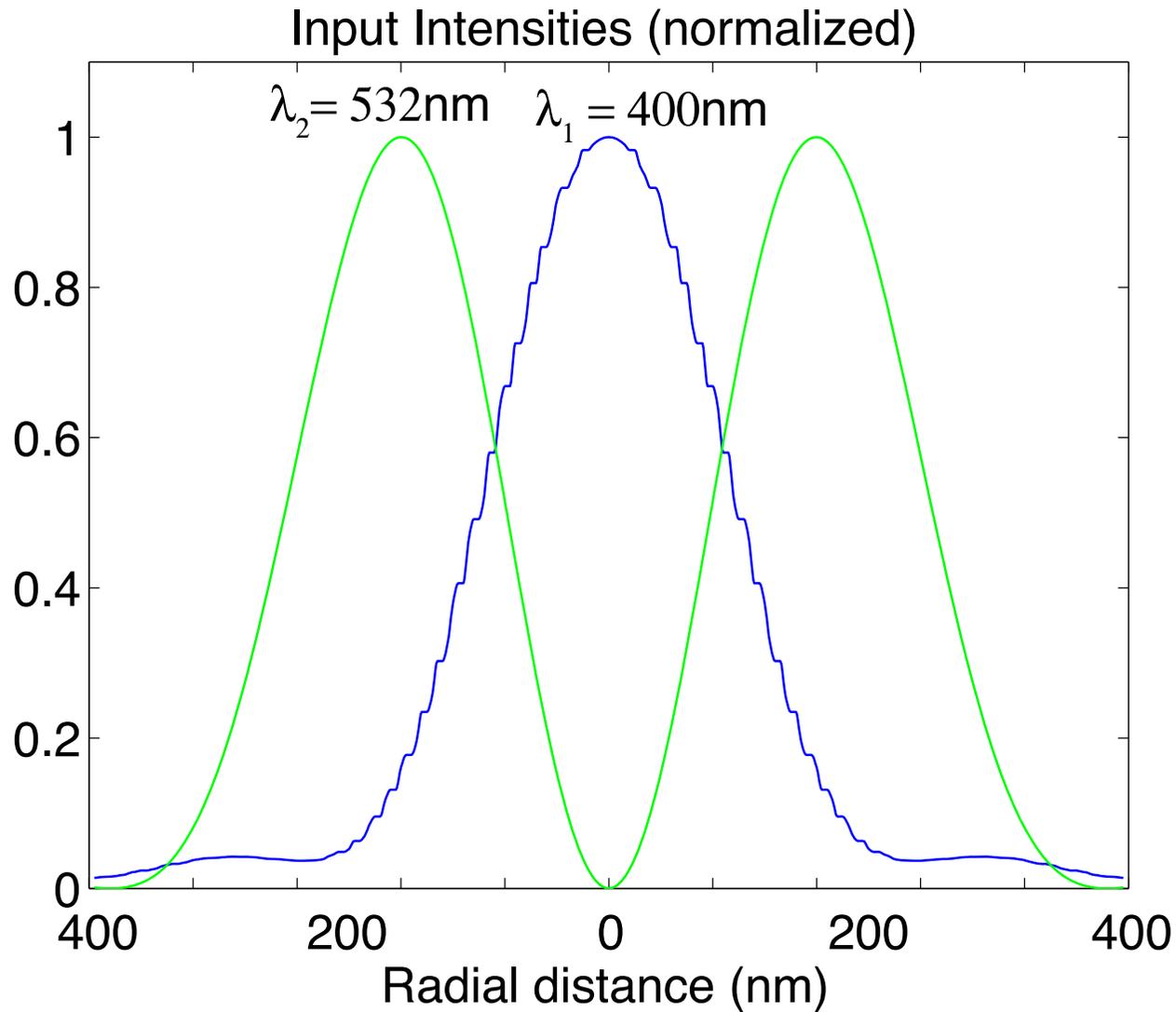
$$\epsilon_{2B} = 3 \times 10^3 \text{ m}^2 / \text{mol}$$

Quantum Efficiencies

$$\phi_{2BA} = 7.2 \times 10^{-3}$$

$$\phi_{2AB} = 2.1 \times 10^{-3}$$

Absorbance Modulation Simulation

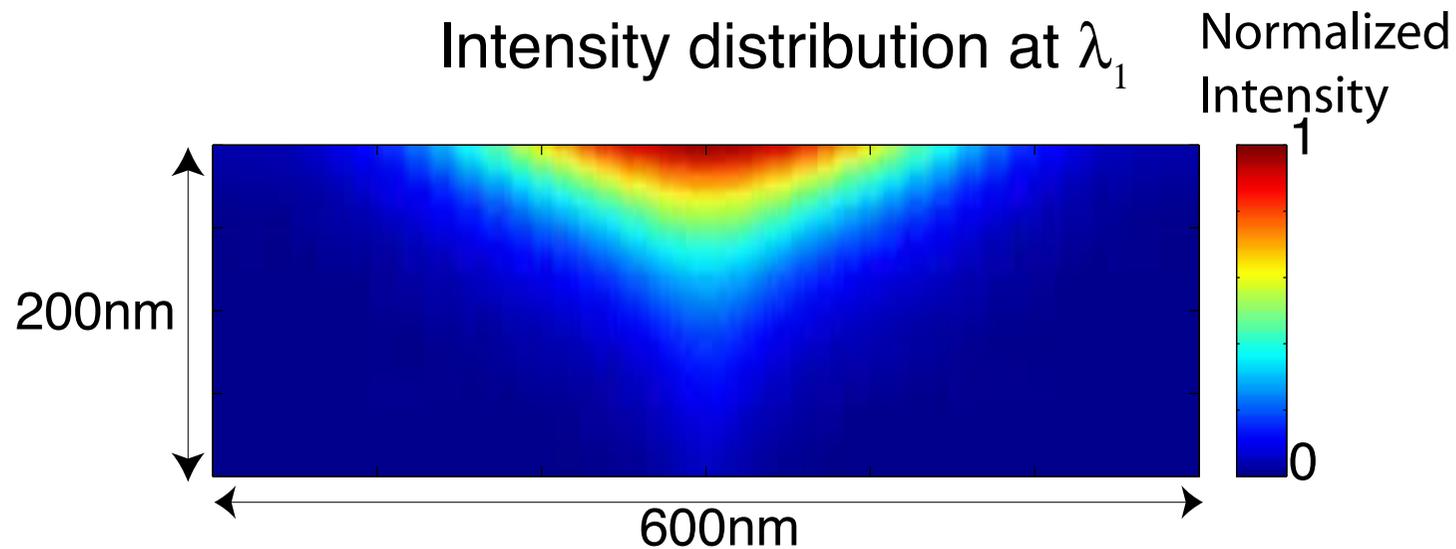
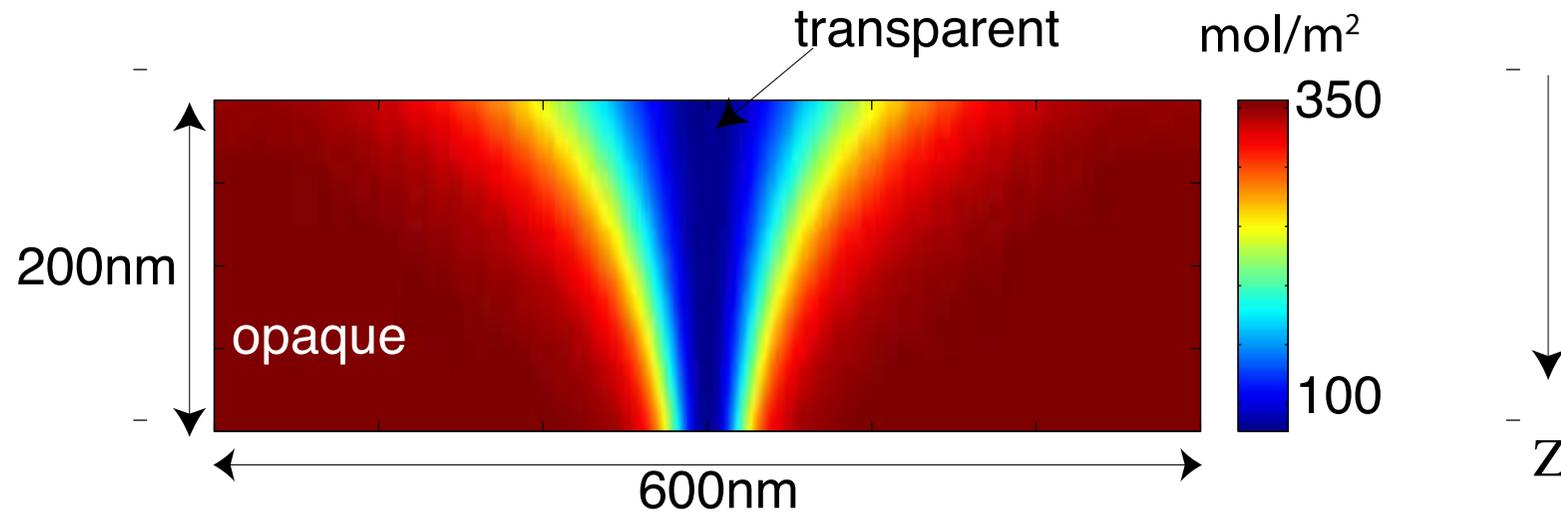


- λ_1 Binary Phase zone plate of numerical aperture (NA) 0.85
- λ_2 Spiral Phase plate of NA 0.72

Absorbance Modulation Simulation

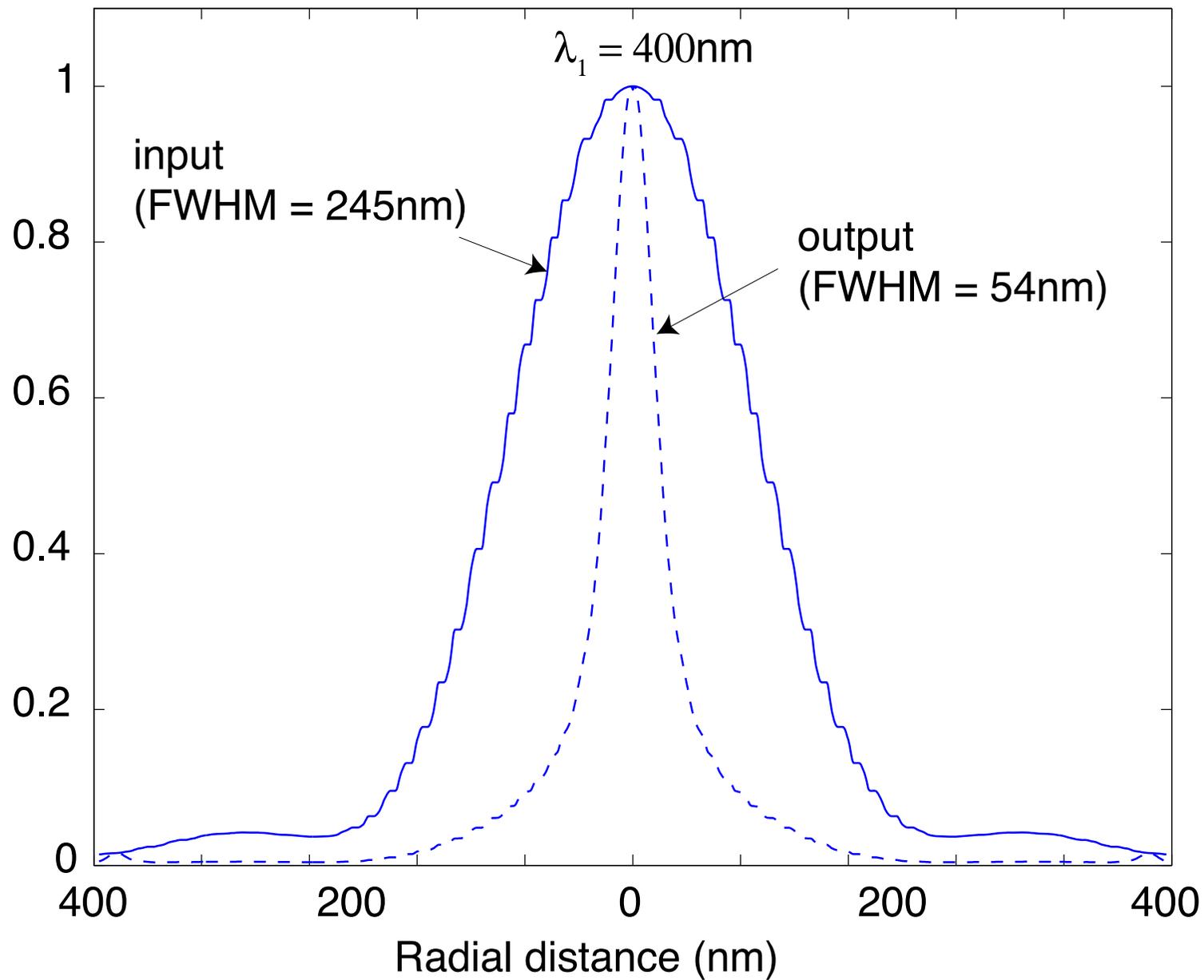
Beam propagation (λ_1) through 200nm of azobenzene polymer (AML)

Concentration of species A = [A]

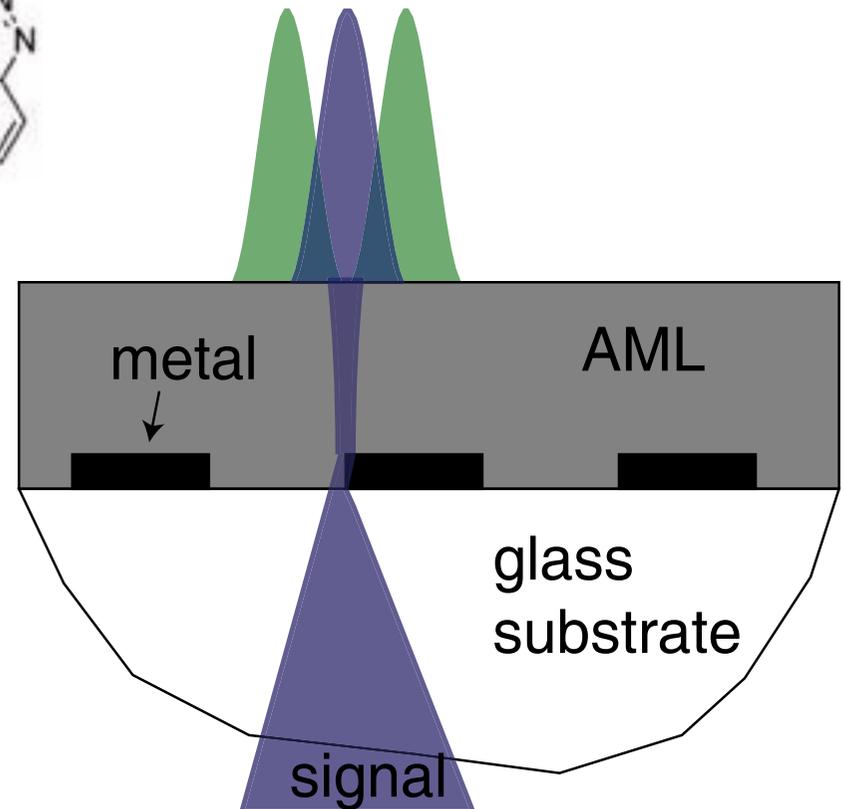
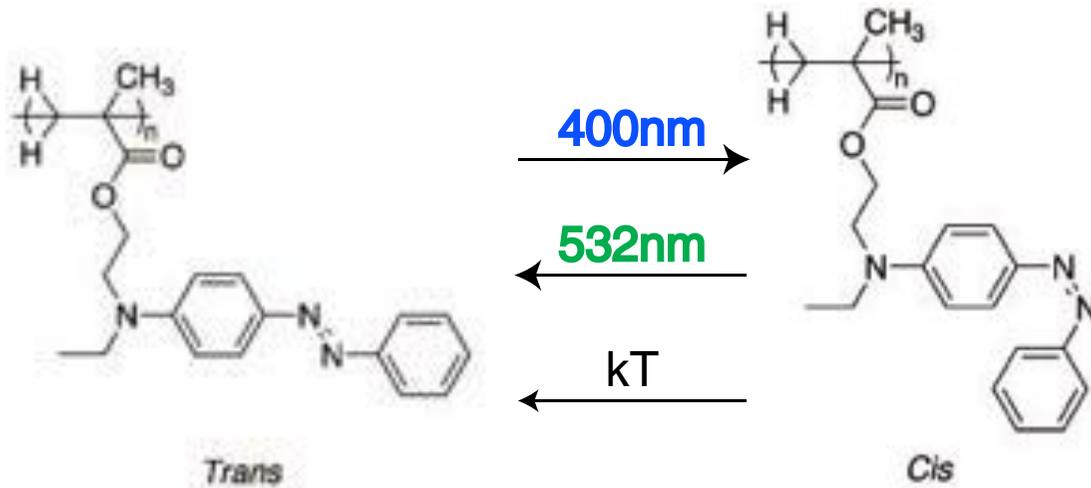
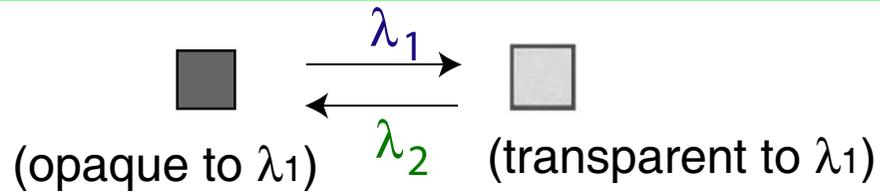


Absorbance Modulation Simulation

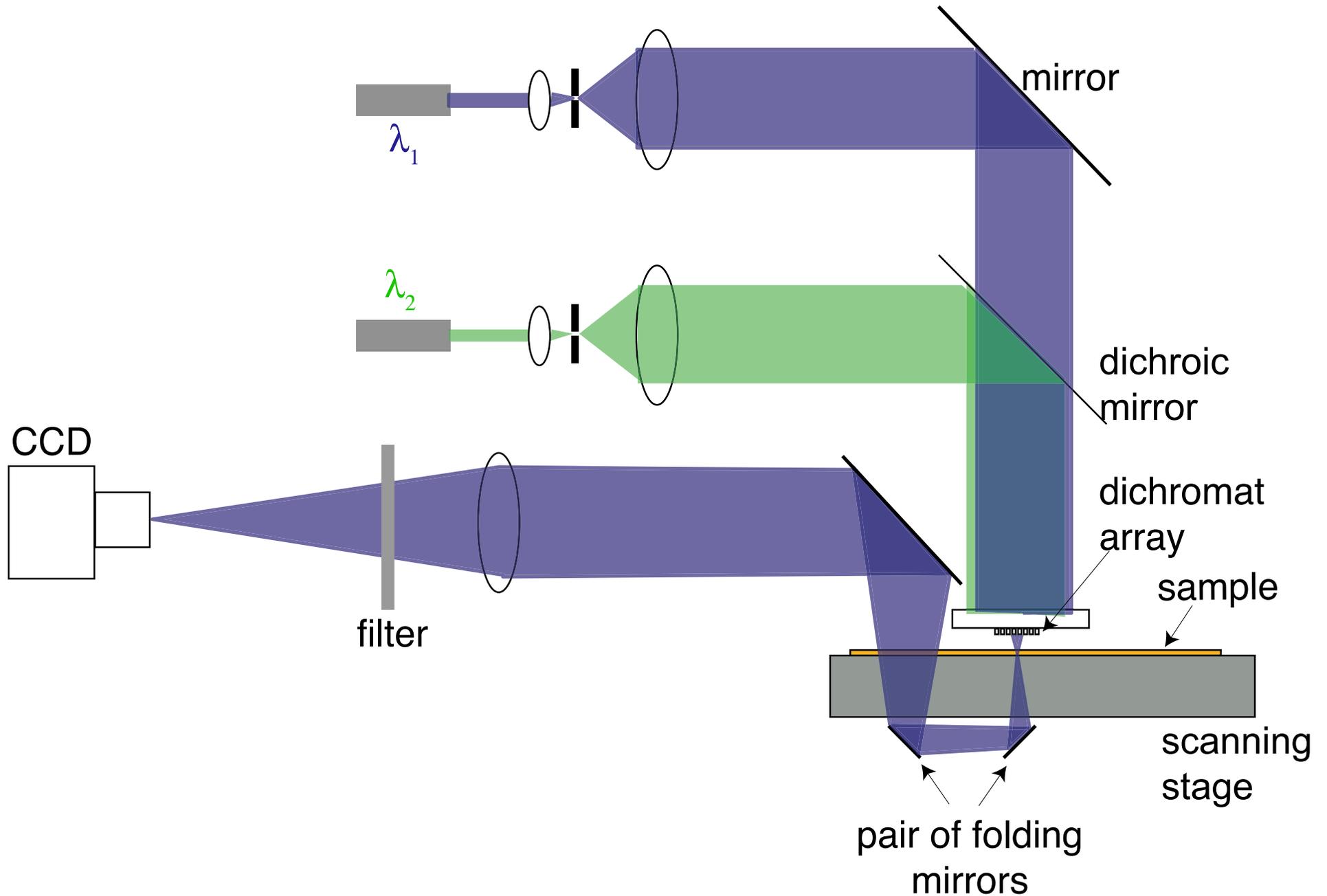
Output Intensity is “squeezed”



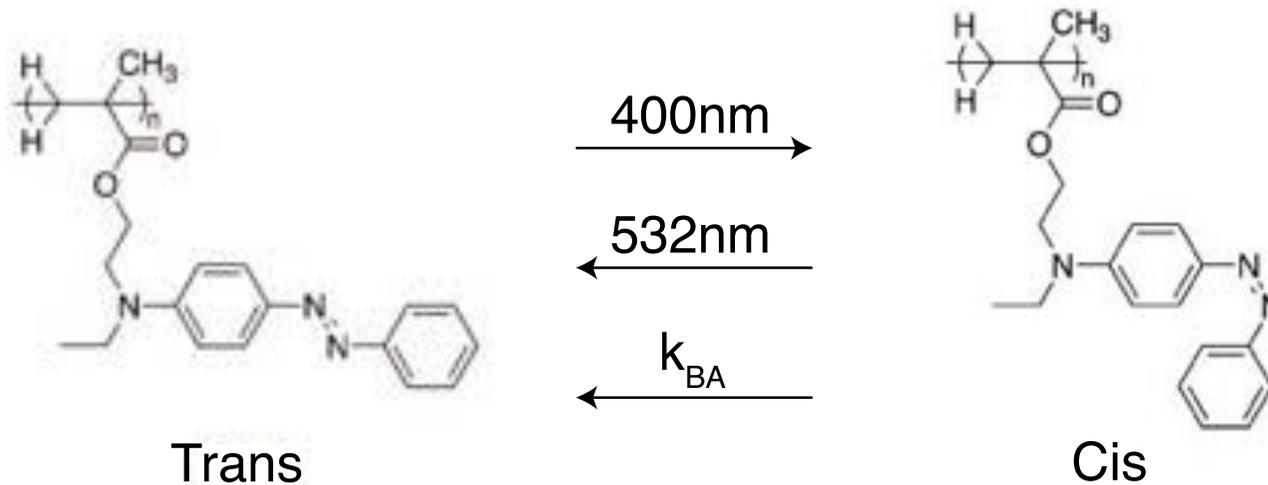
Absorbance Modulation for Imaging



Schematic of current Microscope



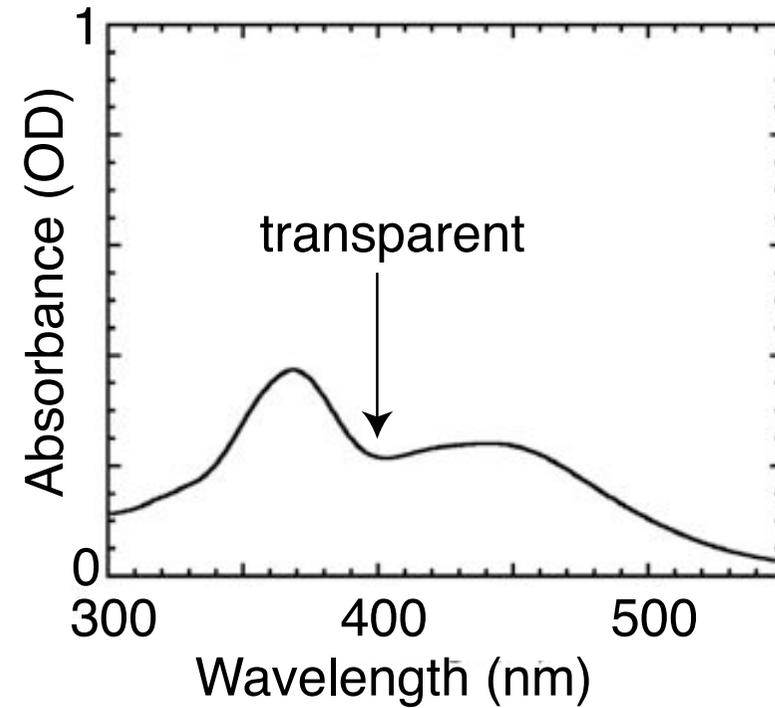
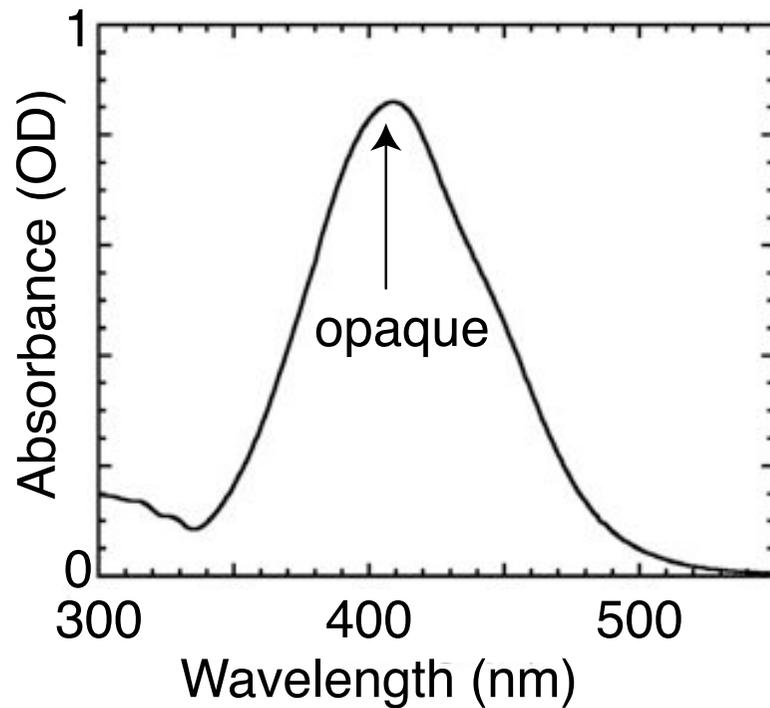
Photochromic molecule: Azobenzene polymer

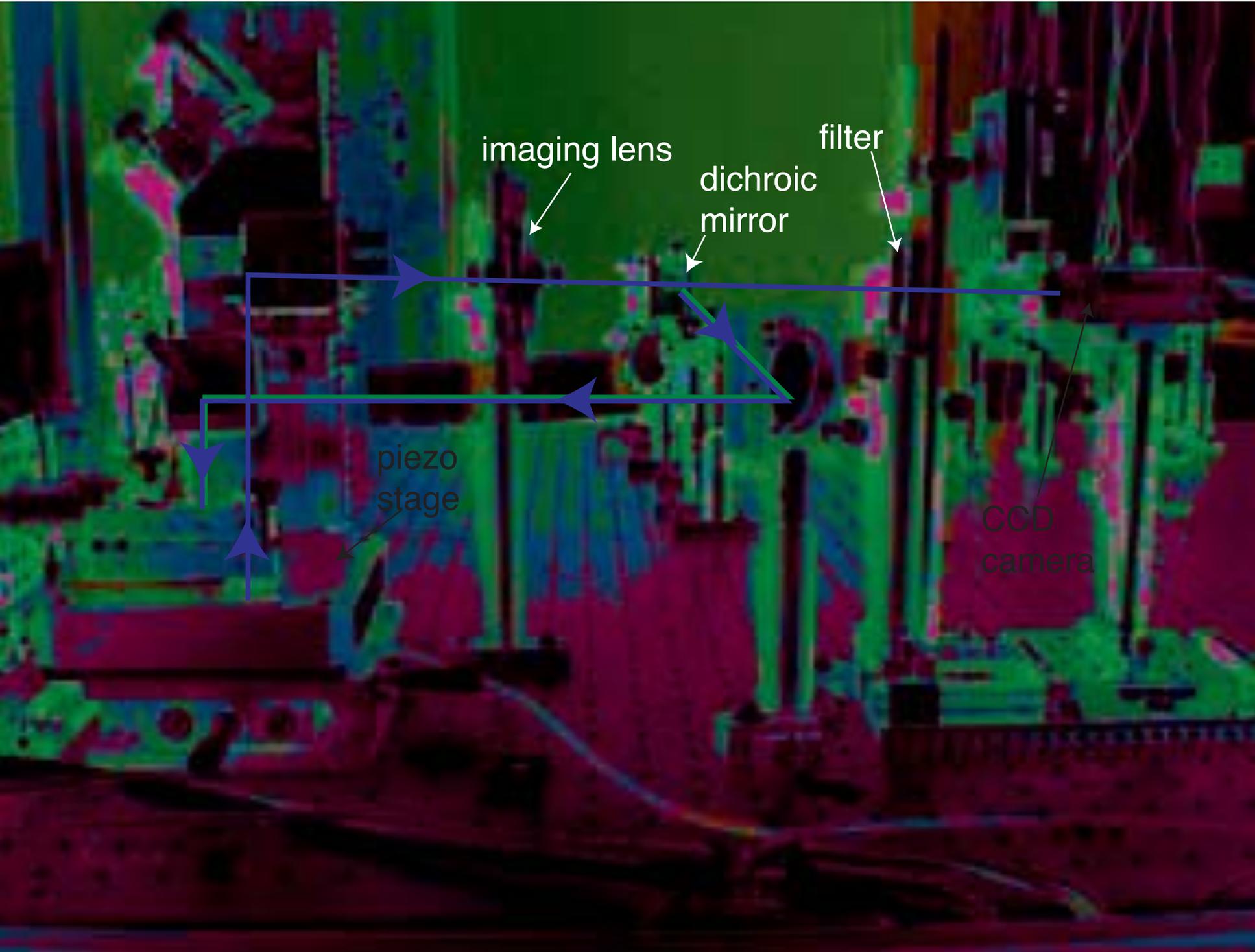


Trans (A)

UV-Vis Spectra

Cis (B)





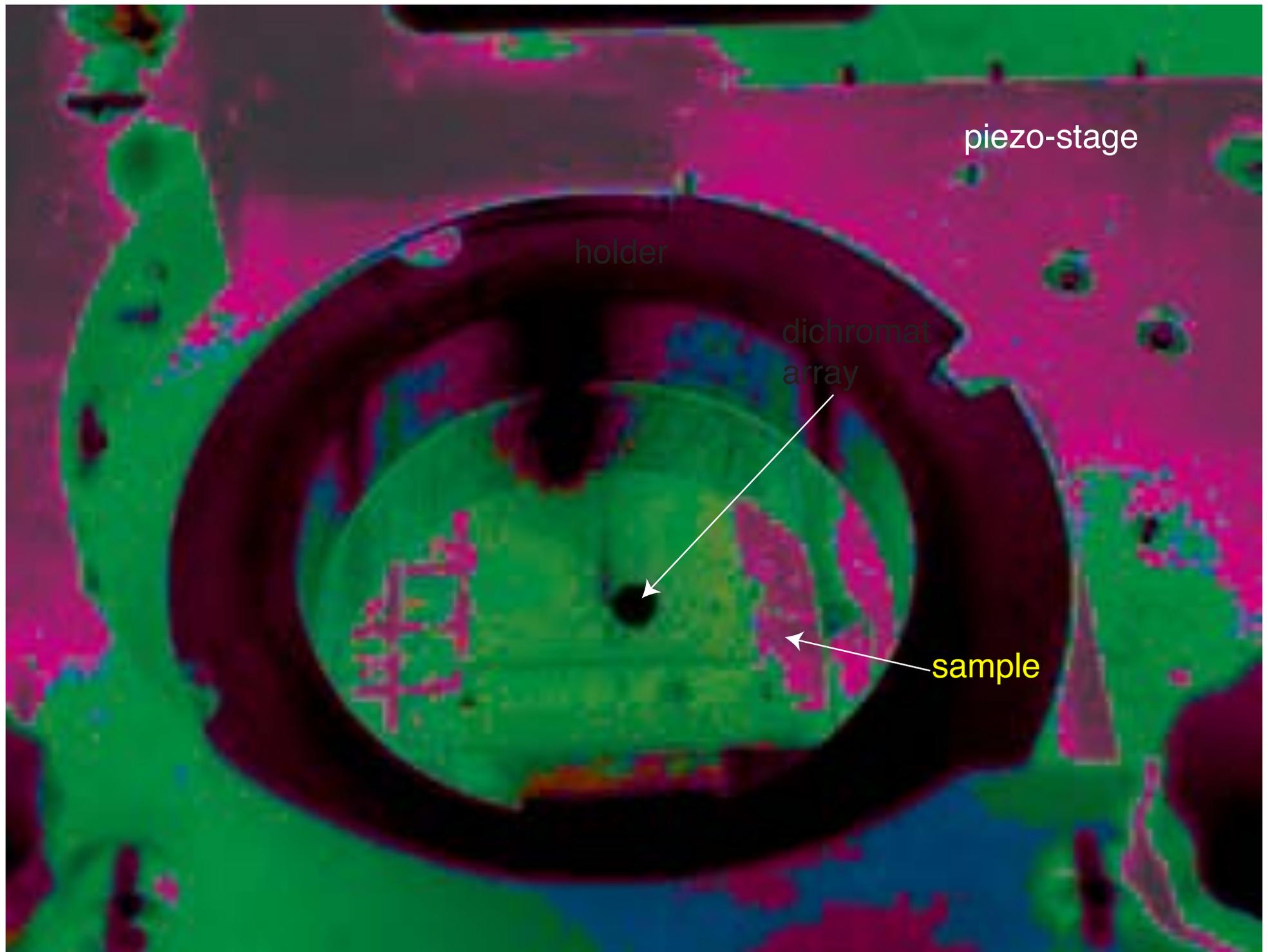
imaging lens

filter

dichroic mirror

piezo stage

CCD camera



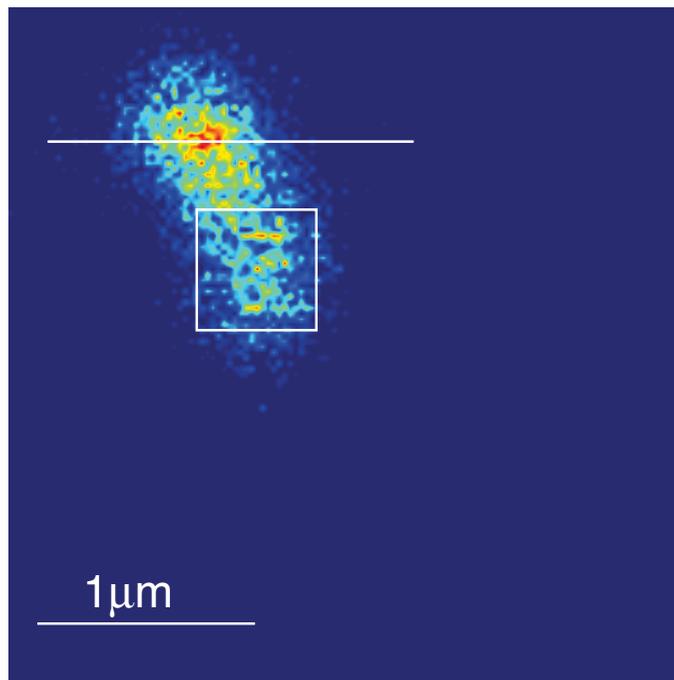
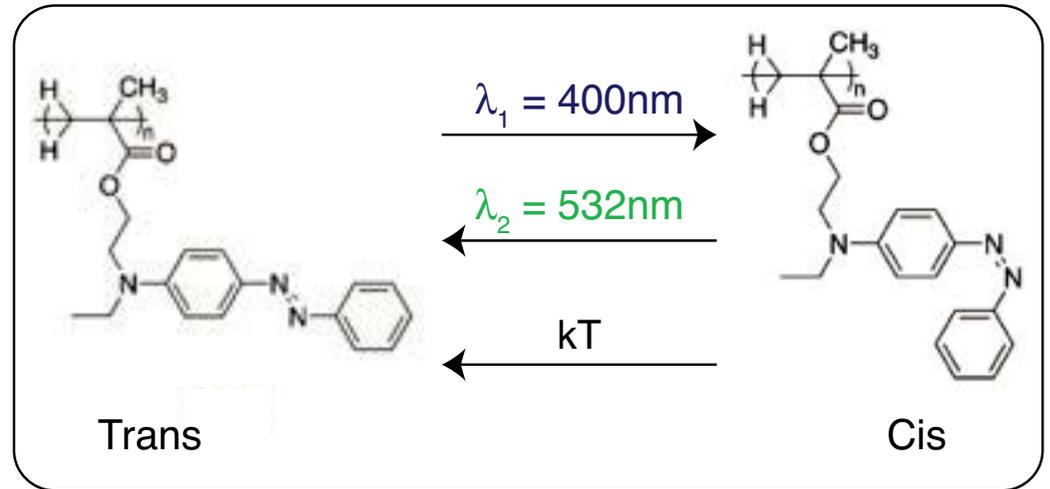
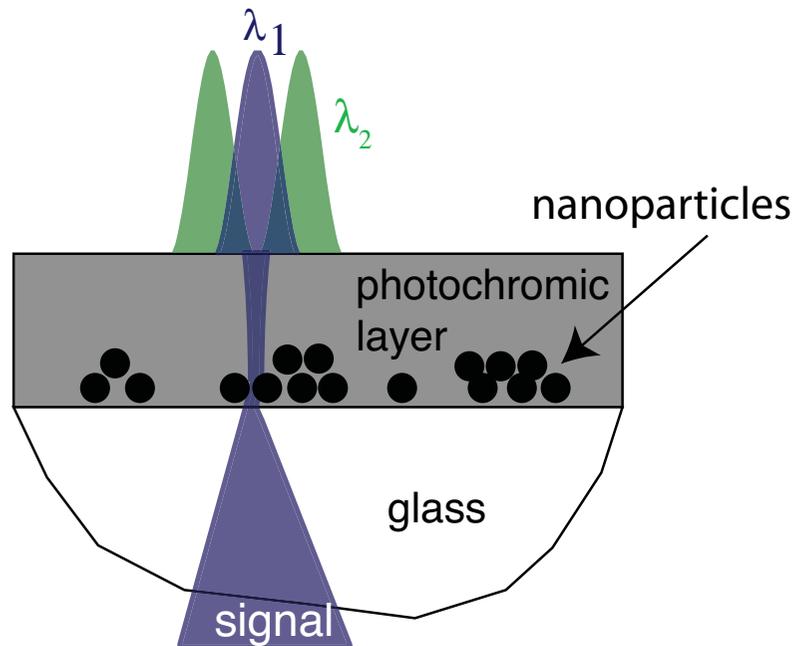
piezo-stage

holder

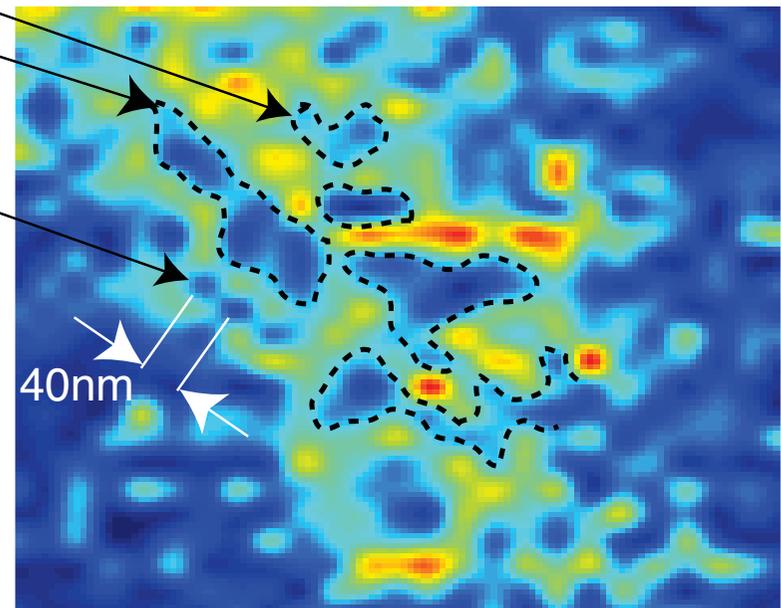
dichromat
array

sample

Absorbance-Modulation Nanoscopy

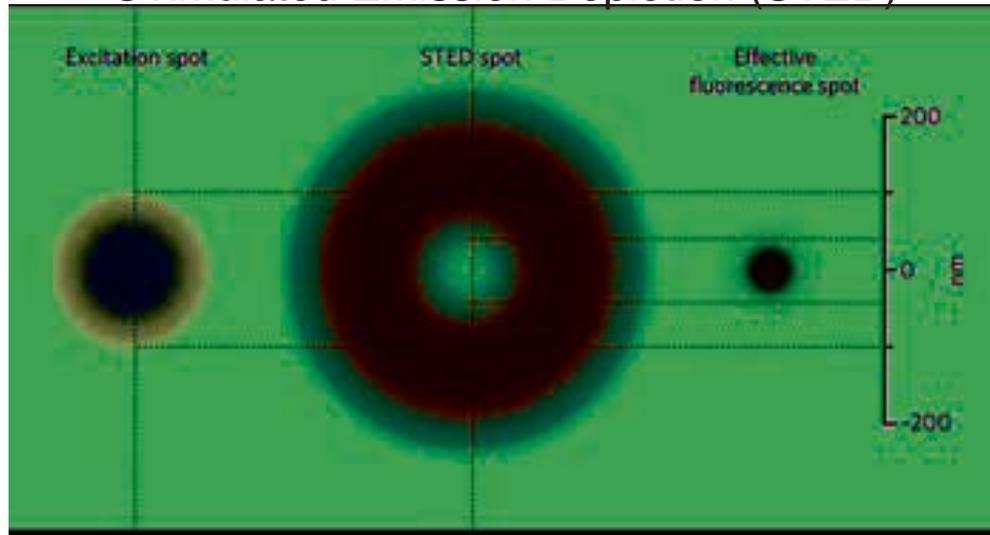


nanoparticle clusters
possible single nanoparticle

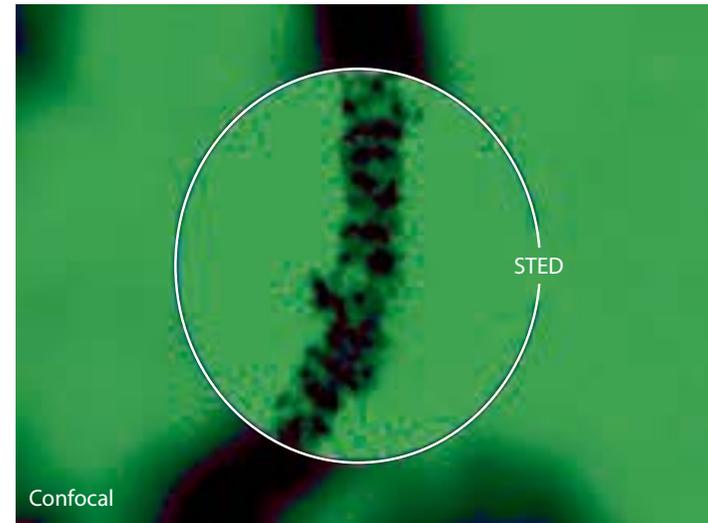


Superresolution using Fluorescence

STimulated Emission Depletion (STED)



Folds of the mitochondrial inner membrane



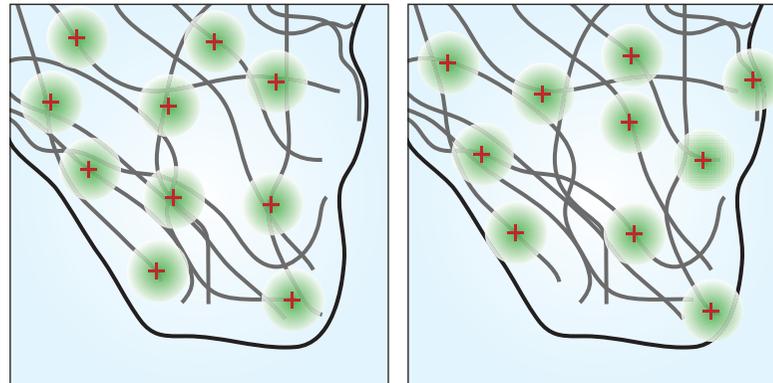
T. A. Klar, *et. al*, *PNAS* 97, 8206 (2000).

STochastic Optical Reconstruction Microscopy (STORM)

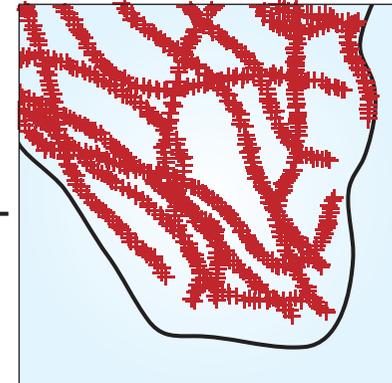
Target structure



Localizing activated subset of probes

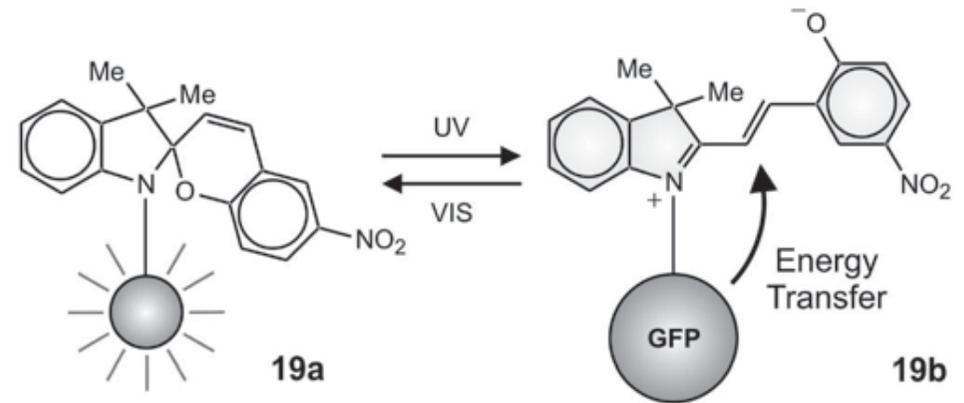
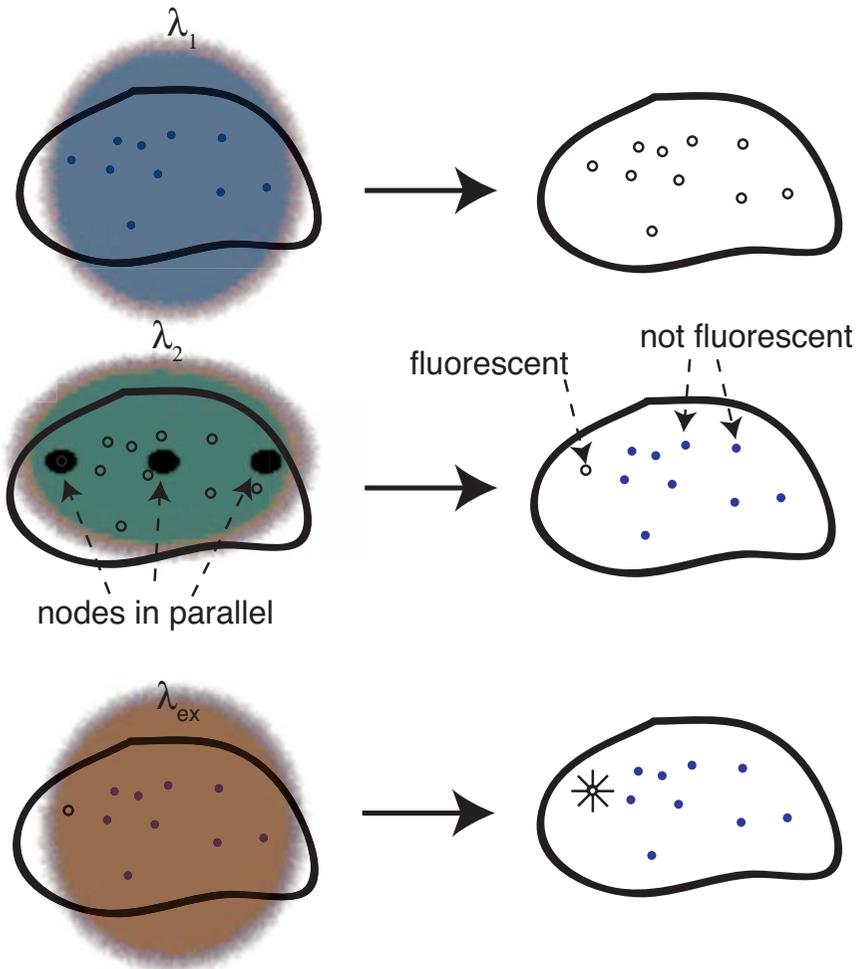
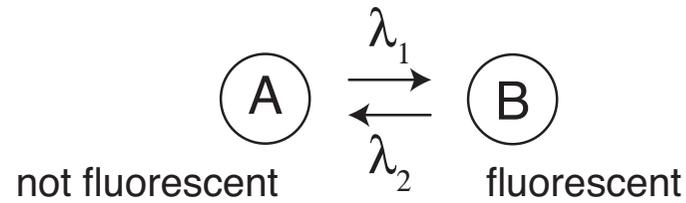


STORM image



X. Zhuang, *Nat. Photonics*, 3, 365 (2009).

Fast 3-D Optical nanoscopy with optically switchable fluorophores



- Single molecule resolution in 3-D
- 3-D node at λ_2 created by fast spatial-light modulator (SLM). Node may be scanned by SLM.
- Massively parallel - 1000s of nodes at 100Hz-25kHz frame rates -> live-cell imaging
- Low power levels (CW laser or lamp sources) compared to STED (& related approaches)
- Deterministic raster scan (smaller data sets, much lower post-processing) compared to STORM/PALM. Real-time imaging feasible.

Ganghun Kim



Jason Kleinschmidt



Precious Cantu



Raja Manthena



Sidney Tsai



Trisha Andrews

Milli Grimhall



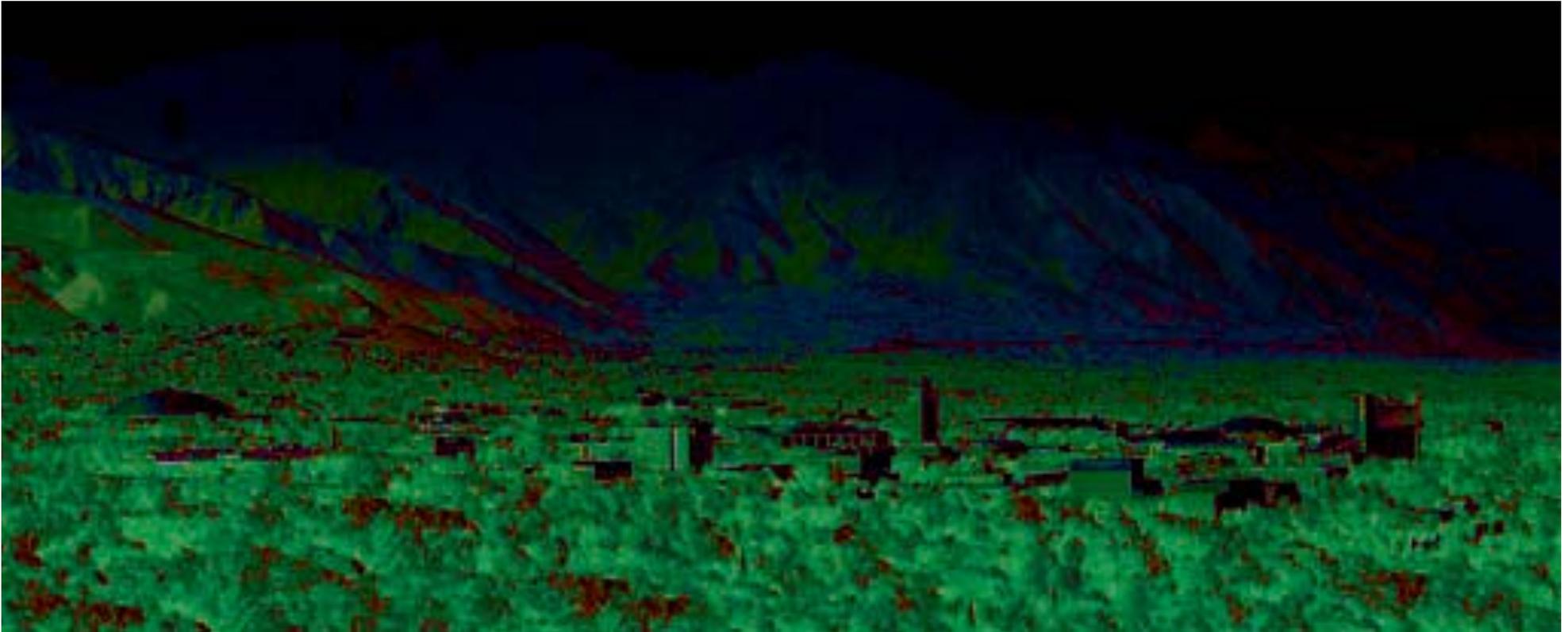
Mohit Diwekar



Farhad Mahdavi



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- Infra-structures & facilities
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