X-ray Nano-Analytics and Microscopy

Part II

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DESY & Universität Hamburg

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

Universität Hamburg
DER FORSCHUNG | DER LEHRE | DER BILDUNG

DESY.

Current State of X-ray Microscopy

Conventional X-ray microscopy

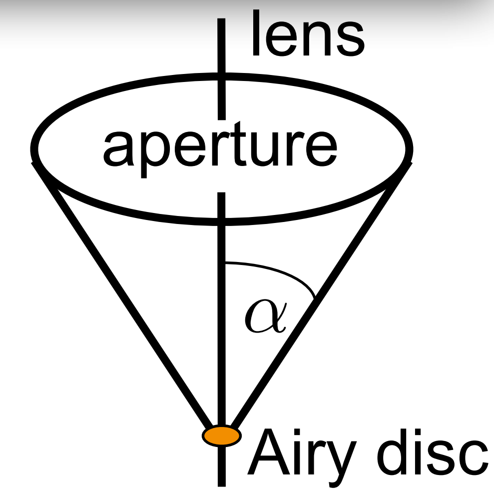
→ optics limit spatial resolution: diffraction limit

$d = \frac{\lambda}{2n\sin\alpha}$ (typically: a few tens of nanometers)

optics are technology limited!

Theoretical extrapolation of X-ray optical performance to the atomic level.

[PRB **74**, 033405 (2006); H. Yan, et al., PRB **76**, 115438 (2007)]



Coherent X-ray imaging techniques (CXDI, ptychography)

→ no imaging optic!

→ limited by statistics of far-field diffraction patterns ...

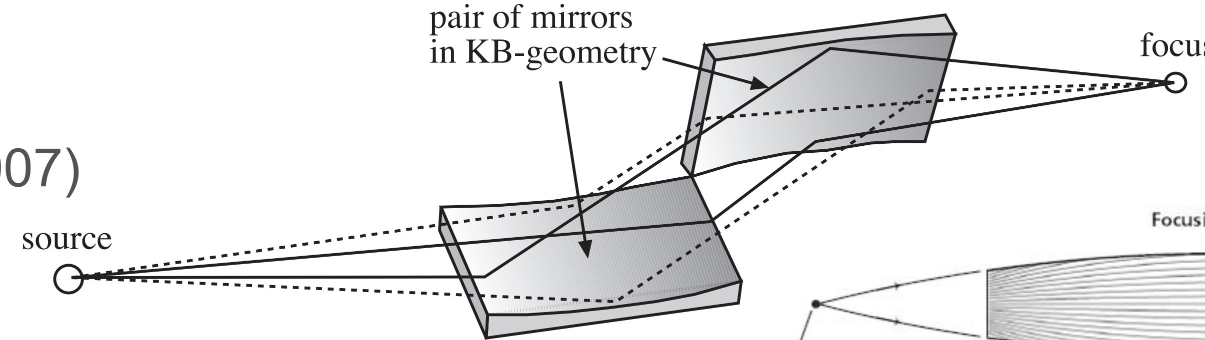
highest resolution: a few nanometers, focusing coherent beam

[PRL **101**, 090801 (2008); Y. Takahashi, et al., PRB **80**, 054103 (2009);
A. Schropp, et al., APL 100, 253112 (2012)]

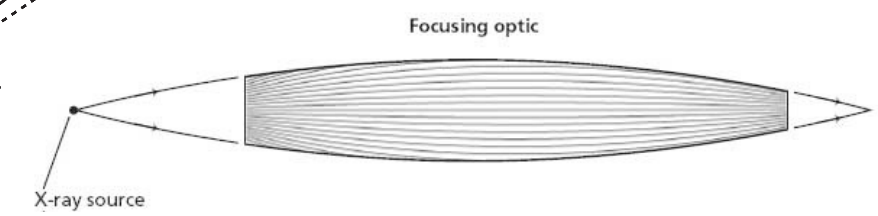
Nanofocusing Optics

reflection:

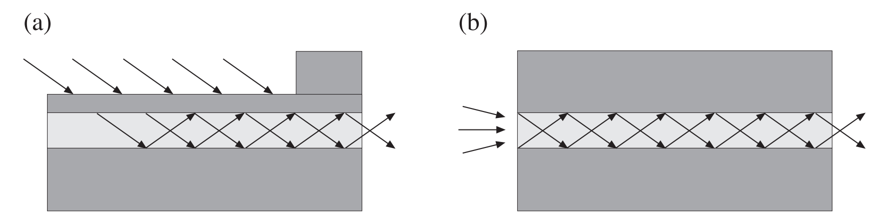
- mirrors (25 nm)
  H. Mimura, et al., APL **90**, 051903 (2007)



- capillaries

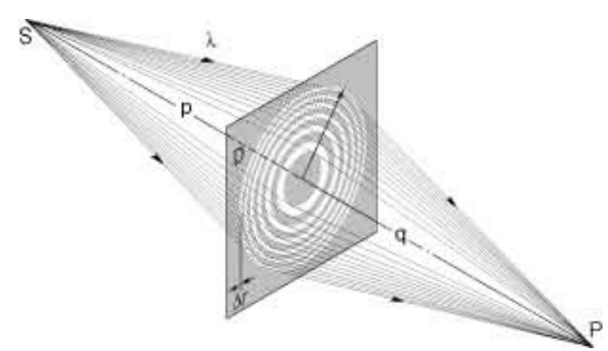


- wave guides (~10 nm)
  S. P. Krüger, et al., J. Synchrotron Rad. **19**, 227 (2012)

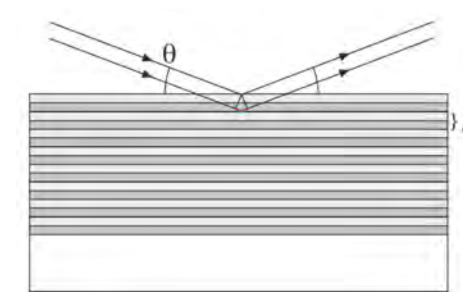


diffraction:

- Fresnel zone plates (< 10 nm)
  J. Vila-Comamala, et al., Ultramic. **109**, 1360 (2009)



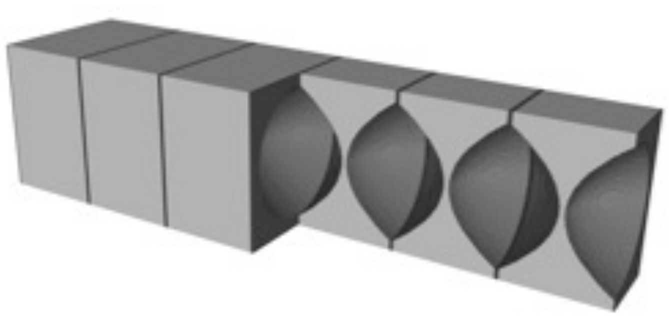
- multilayer mirrors (7 nm)
  H. Mimura, et al., Nat. Phys. **6**, 122 (2010)

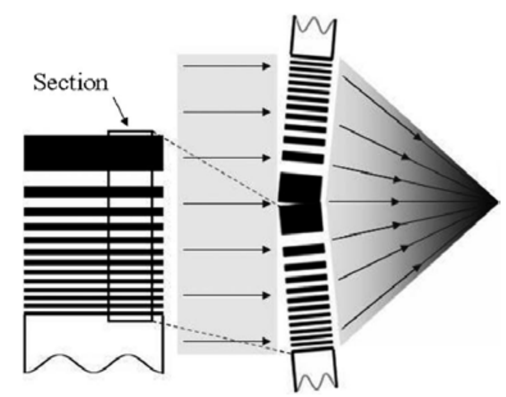


- multilayer Laue lenses (8 x 7 nm)
  S. Bajt, et al., Light: Sci. & App. **7**, 17162 (2018)
- bent crystals

refraction:

- lenses (43 nm, 18 nm)
  C. G. Schroer, et al., AIP Conf. Ser. **1365**, 227 (2011)
  J. Patommel, et al., APL **110**, 101103 (2017)



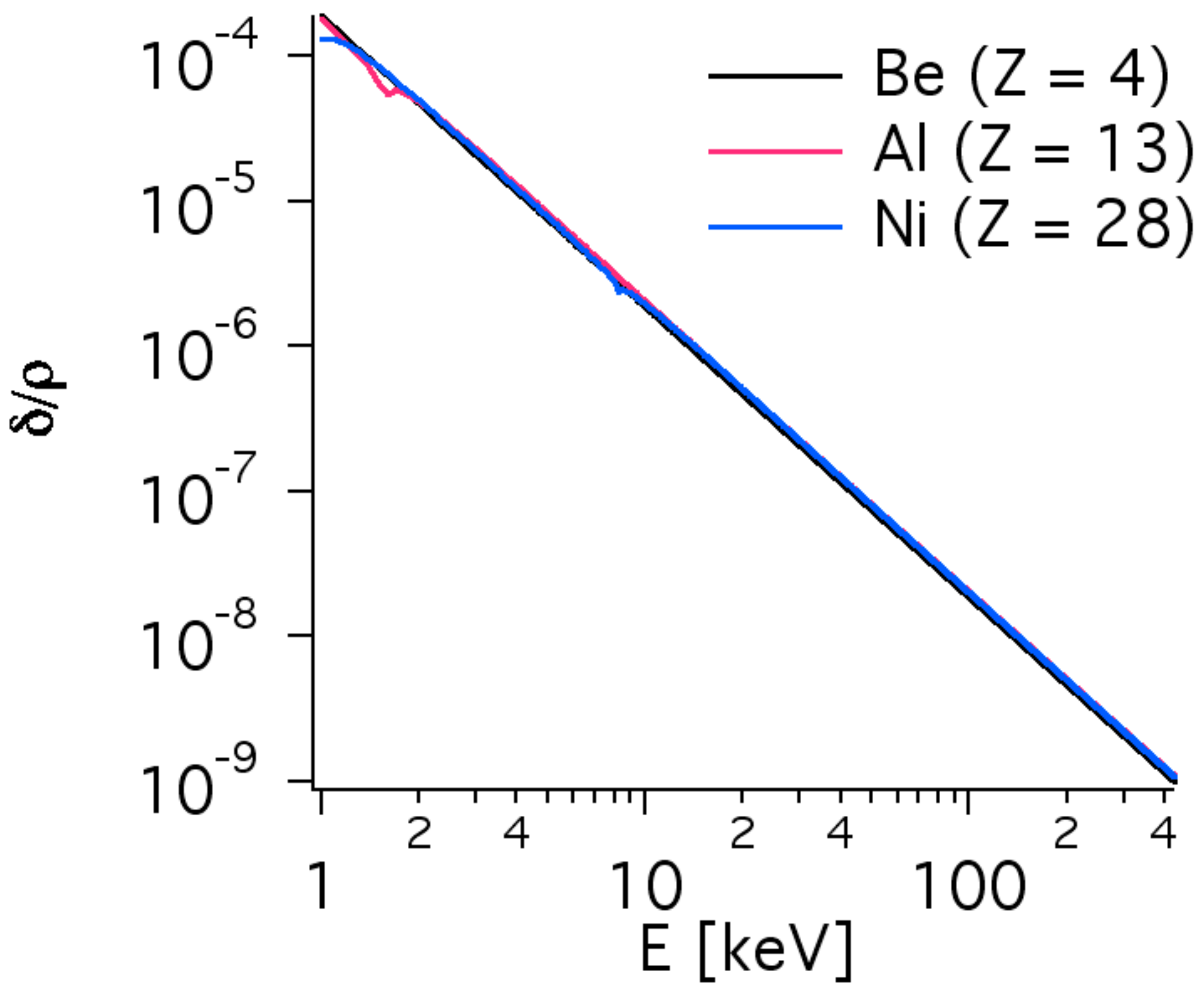


Refraction

$$n = 1 - \delta + i\beta, \quad \delta > 0$$

Vacuum is optically denser than matter!

$$\delta = \frac{N_\mathrm{A}}{2\pi} r_0 \lambda^2 \rho \frac{Z + f'}{A}$$



specific refraction:

- independent of material
- very week

Absorption

$$n = 1 - \delta + i\beta, \quad \delta > 0$$

Lambert-Beer law:

$$I(x) = I_0 e^{-\mu x}$$

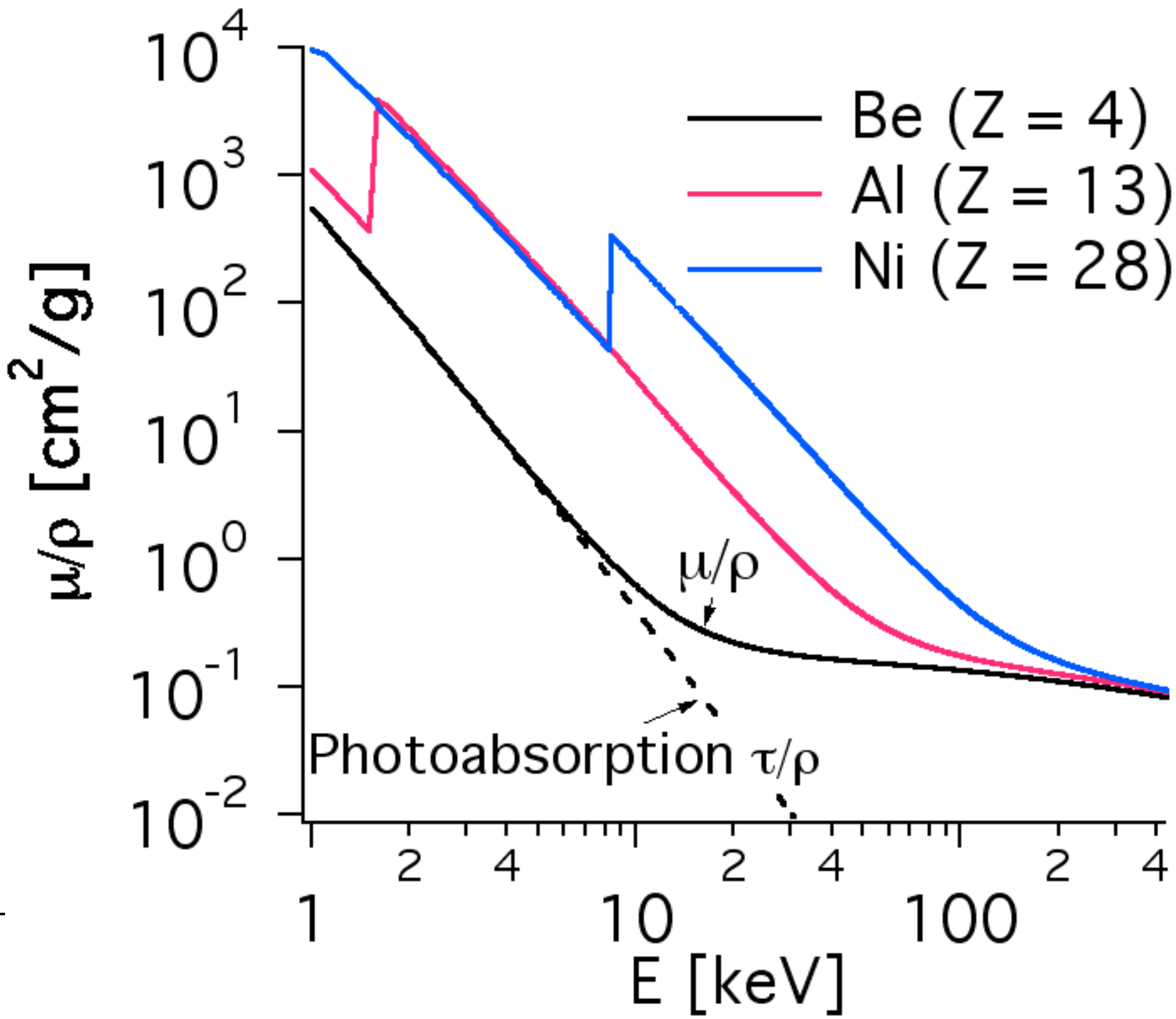
attenuation coefficient μ:

$$\mu = \frac{4\pi\beta}{\lambda}$$

two main contributions:

> photoabsorption $\tau \propto \frac{Z^3}{E^3}$

> Compton scattering μ_C



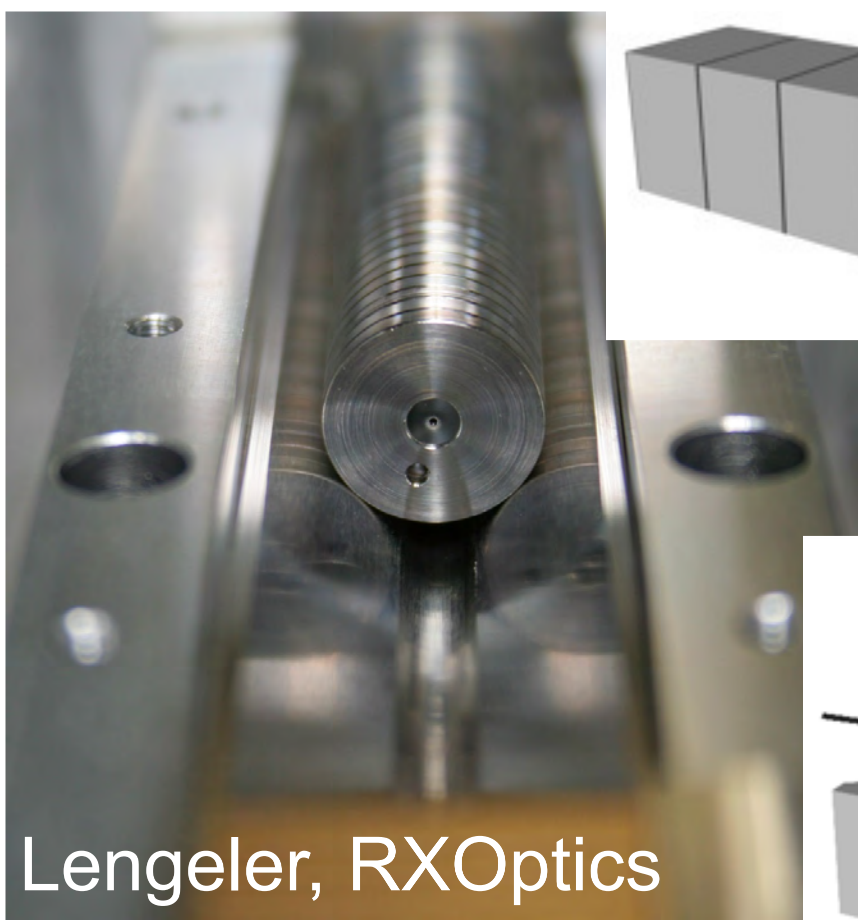
for comparison: $\mu_{glas} = 10^{-7}$ cm^{-1}
for visible light!

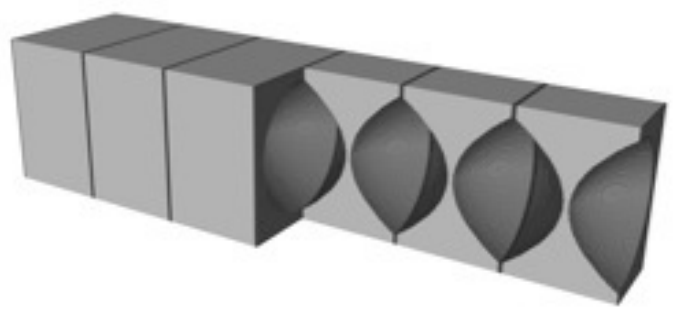
Refractive X-Ray Lenses

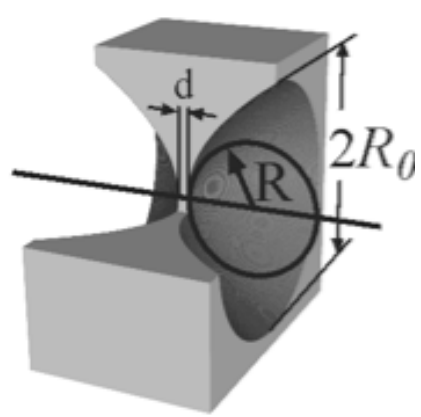
- first realized in 1996 (Snigirev et al.)
- a variety of refractive lenses have been developed since
- applied in full-field imaging and scanning microscopy
- most important to achieve optimal performance:

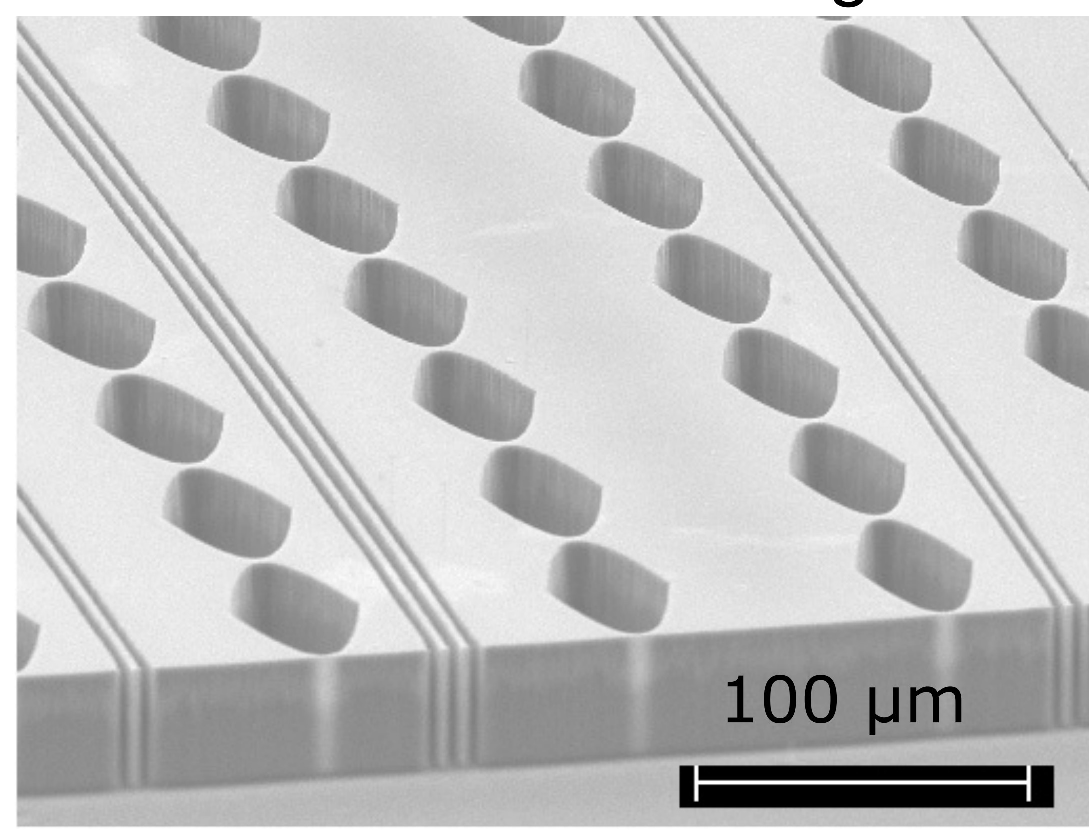
parabolic lens shape

nanofocusing lenses





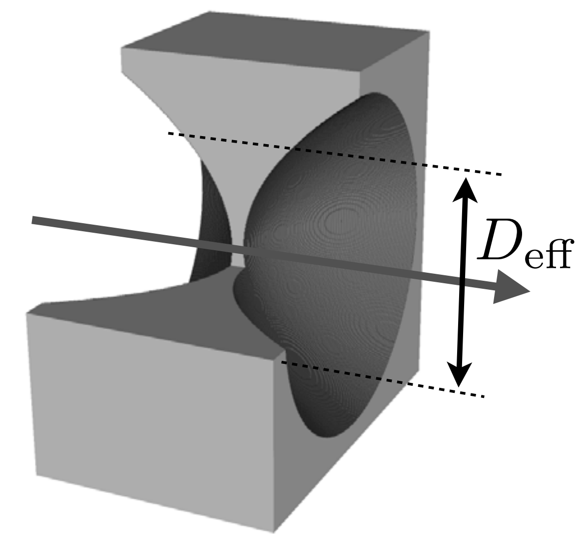




Nanofocus

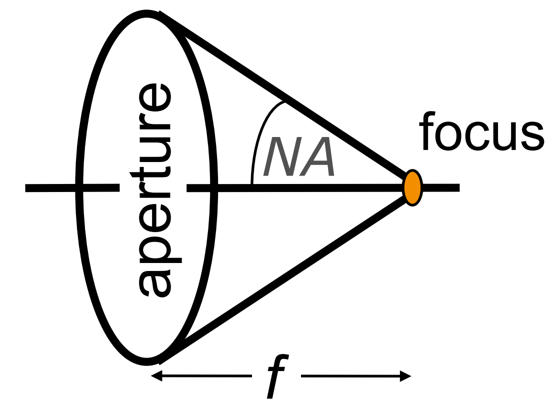
Large focal length *f*: aperture limited by absorption

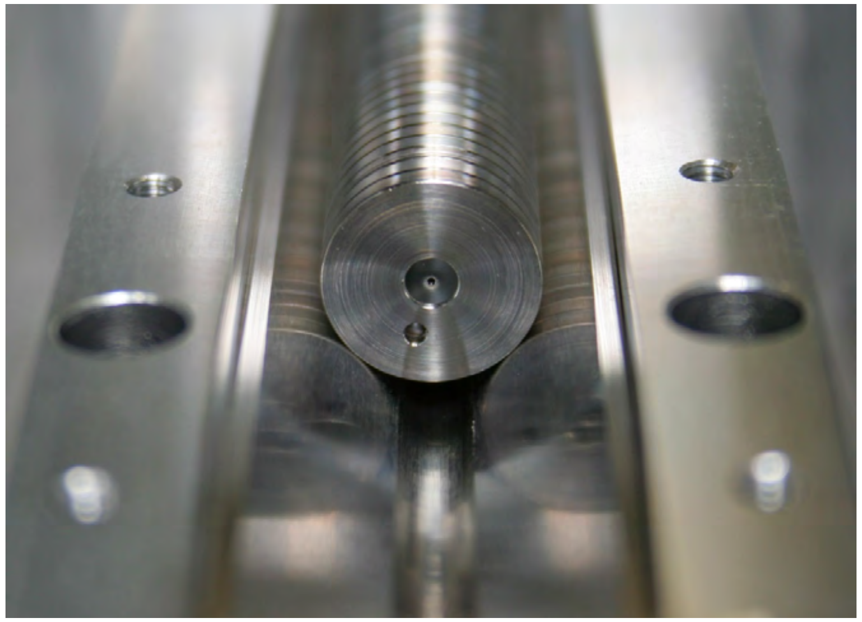
$$D_{\text{eff}} = 4\sqrt{\frac{f\delta}{\mu}} \propto \sqrt{f}$$



→ minimize *μ*/*δ* (⇒ small atomic number *Z*)

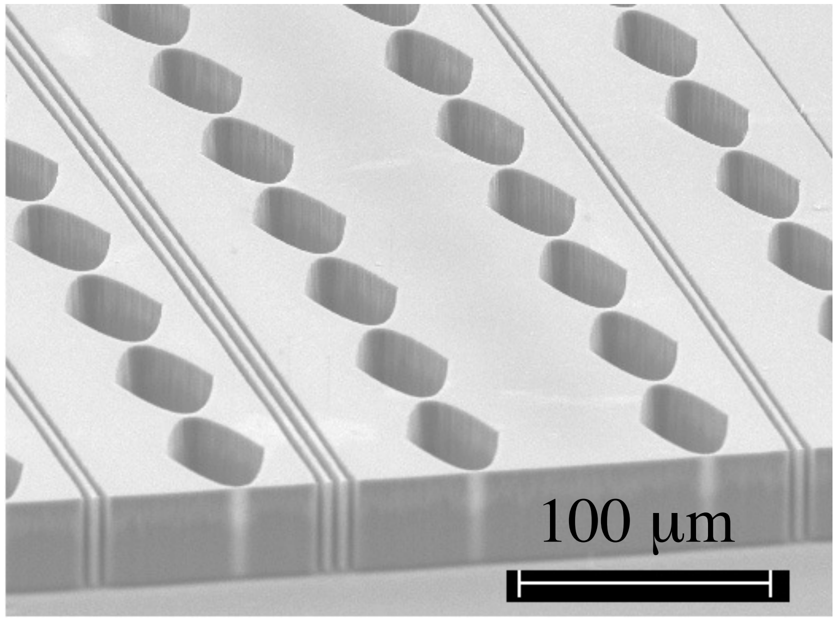
→ $NA = \frac{D_{\text{eff}}}{2f} \propto \frac{1}{\sqrt{f}}$ (⇒ minimize focal length *f*)



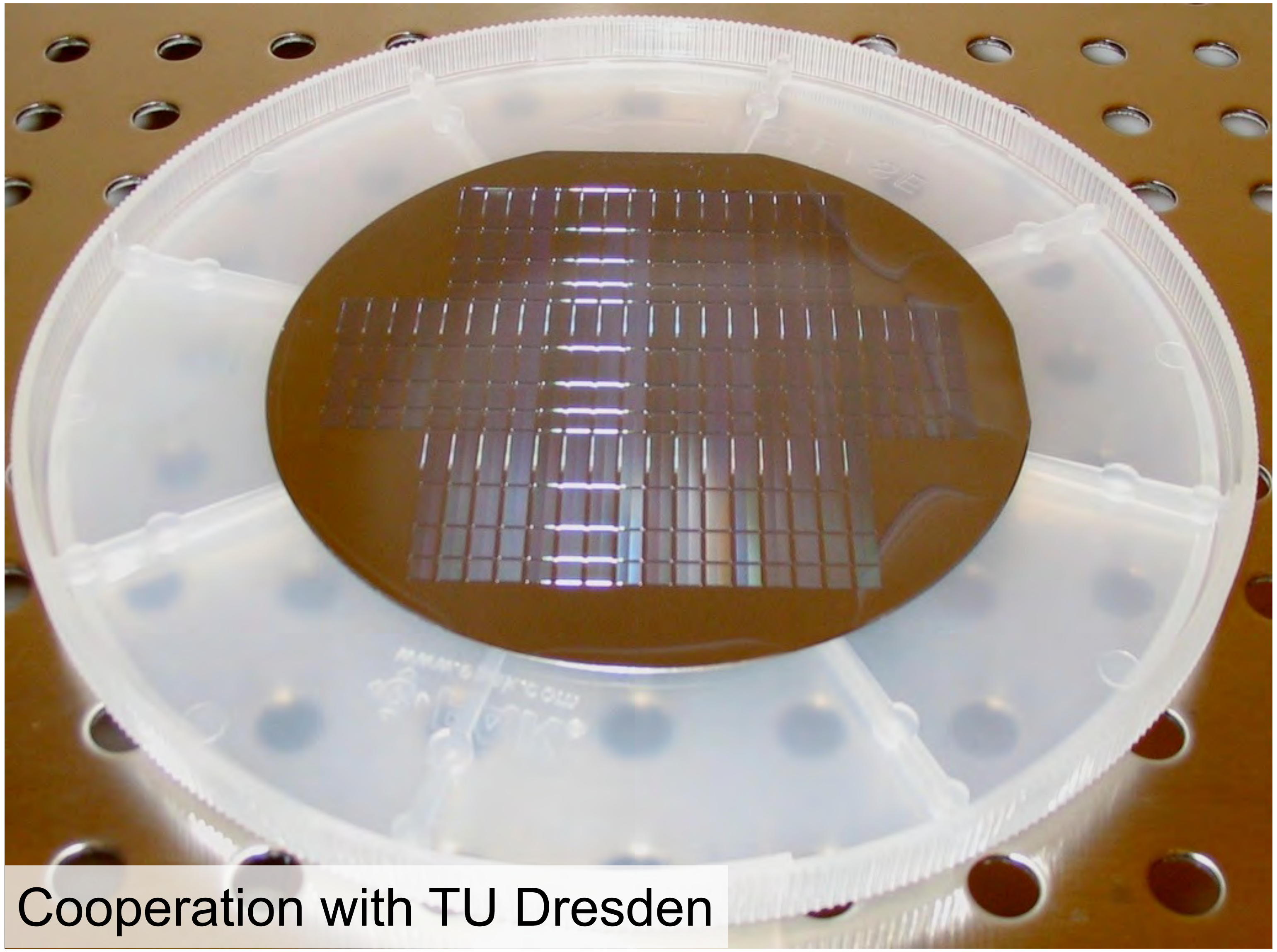


transition to nanofocusing lenses (NFLs)

→

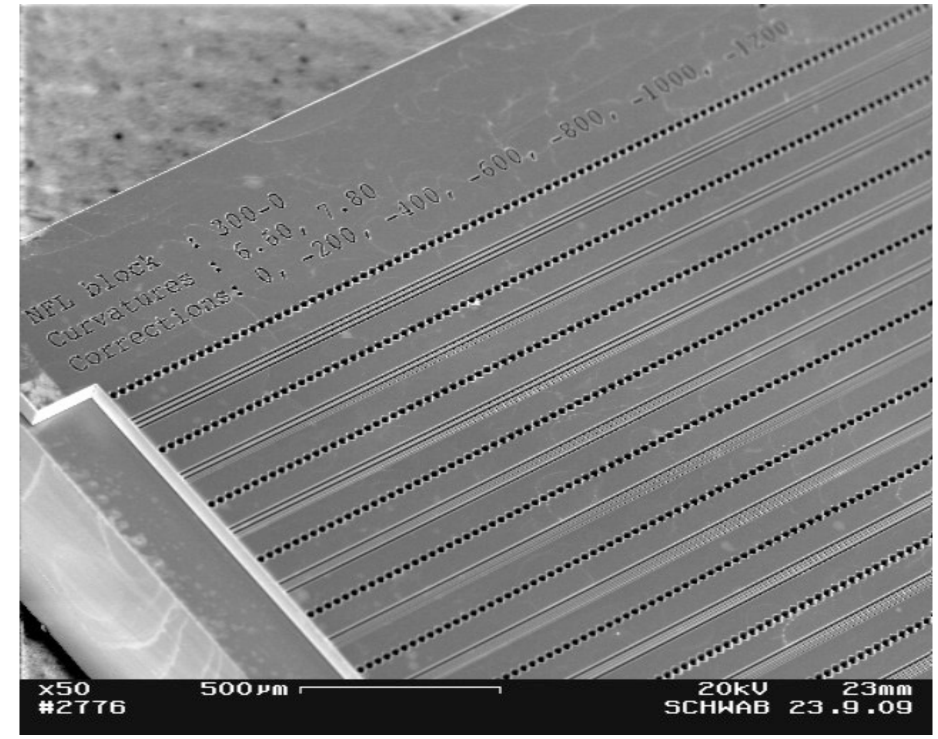


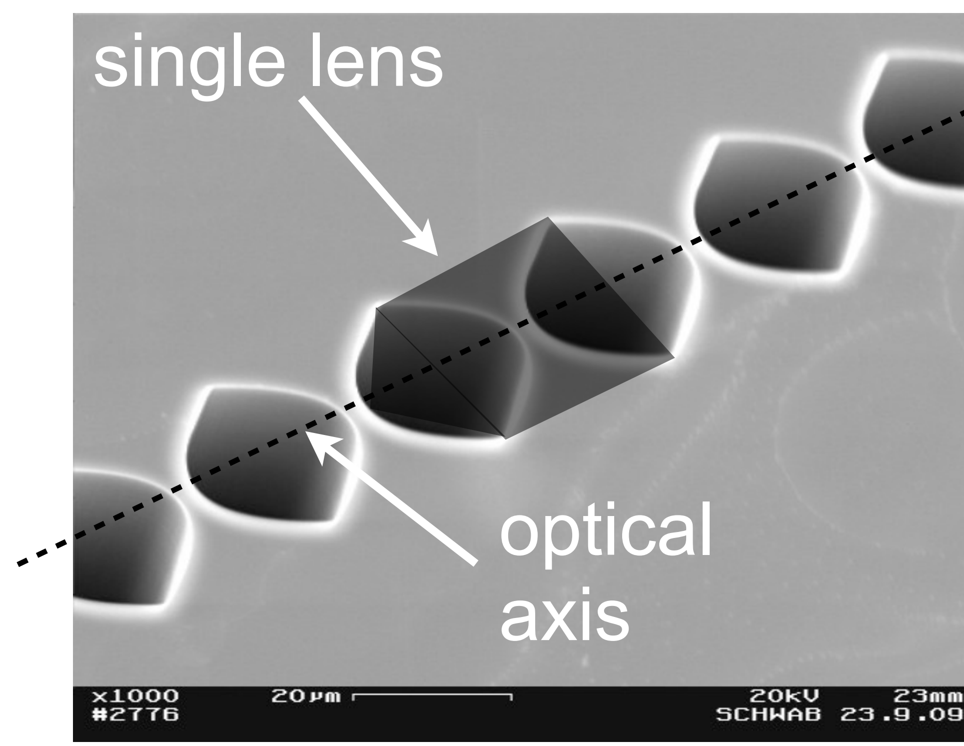
Nanofocusing Lenses (NFLs) Made of Silicon



3136 NFLs on wafer!
about 600000 single lenses!

→ high accuracy, reproducibility

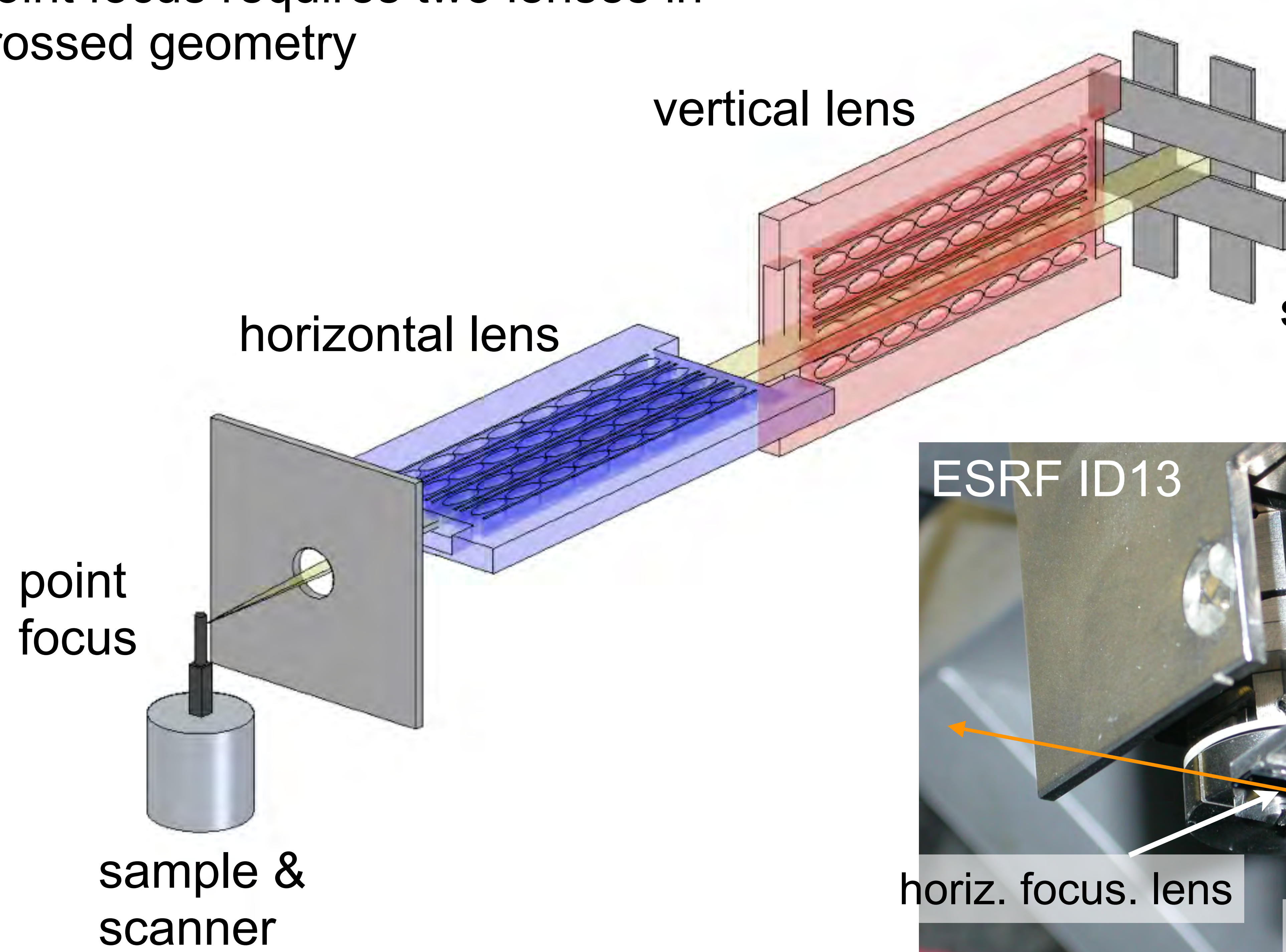


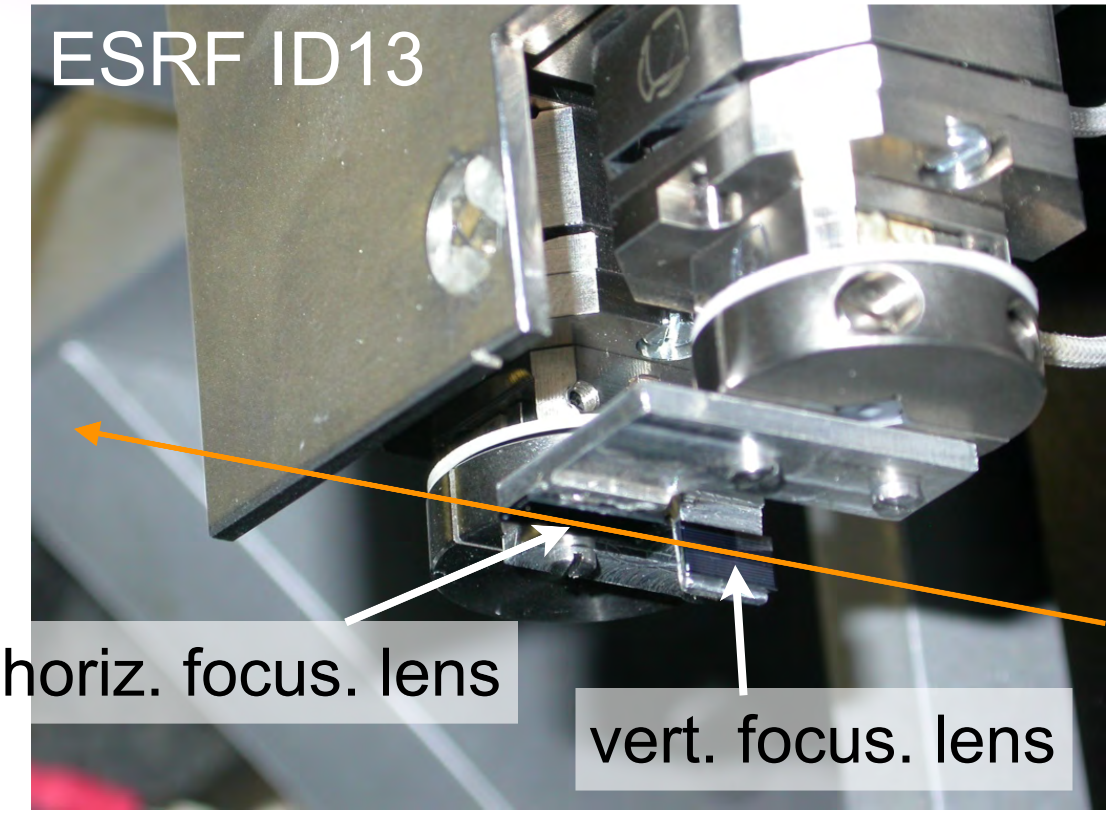


APL **82**, 1485 (2003).

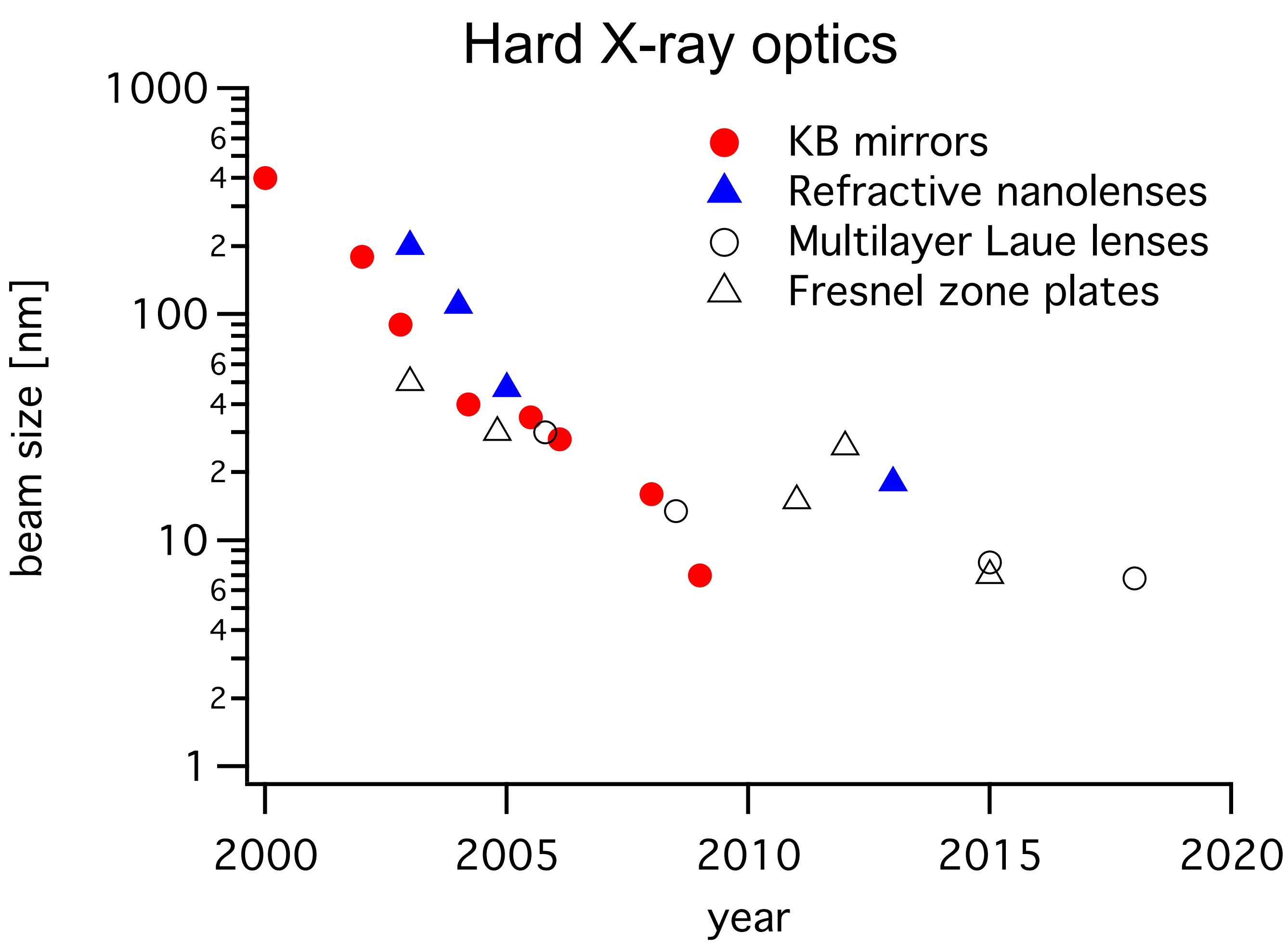
Nanofocusing Lenses (NFLs)

Point focus requires two lenses in crossed geometry



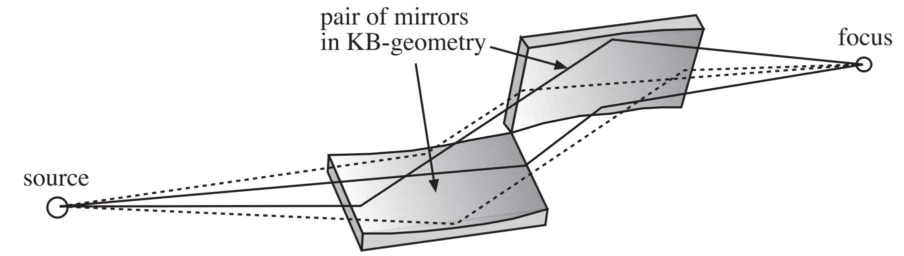


X-Ray Optics: Towards 1 nm Focusing



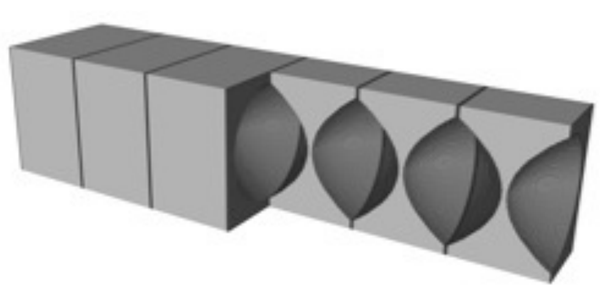
external total reflection

- mirrors (25 nm)
- capillaries
- waveguides (~10 nm)



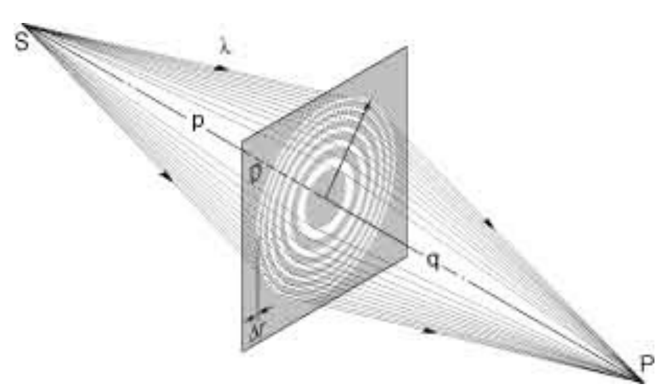
refraction:

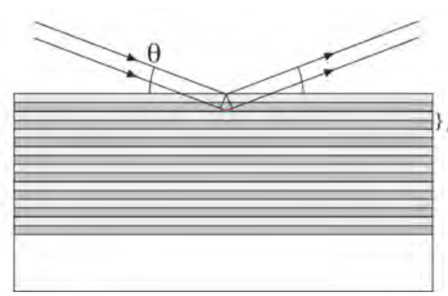
- refractive lenses (43 nm)
- adiabatically focusing lenses (18 nm)



diffraction:

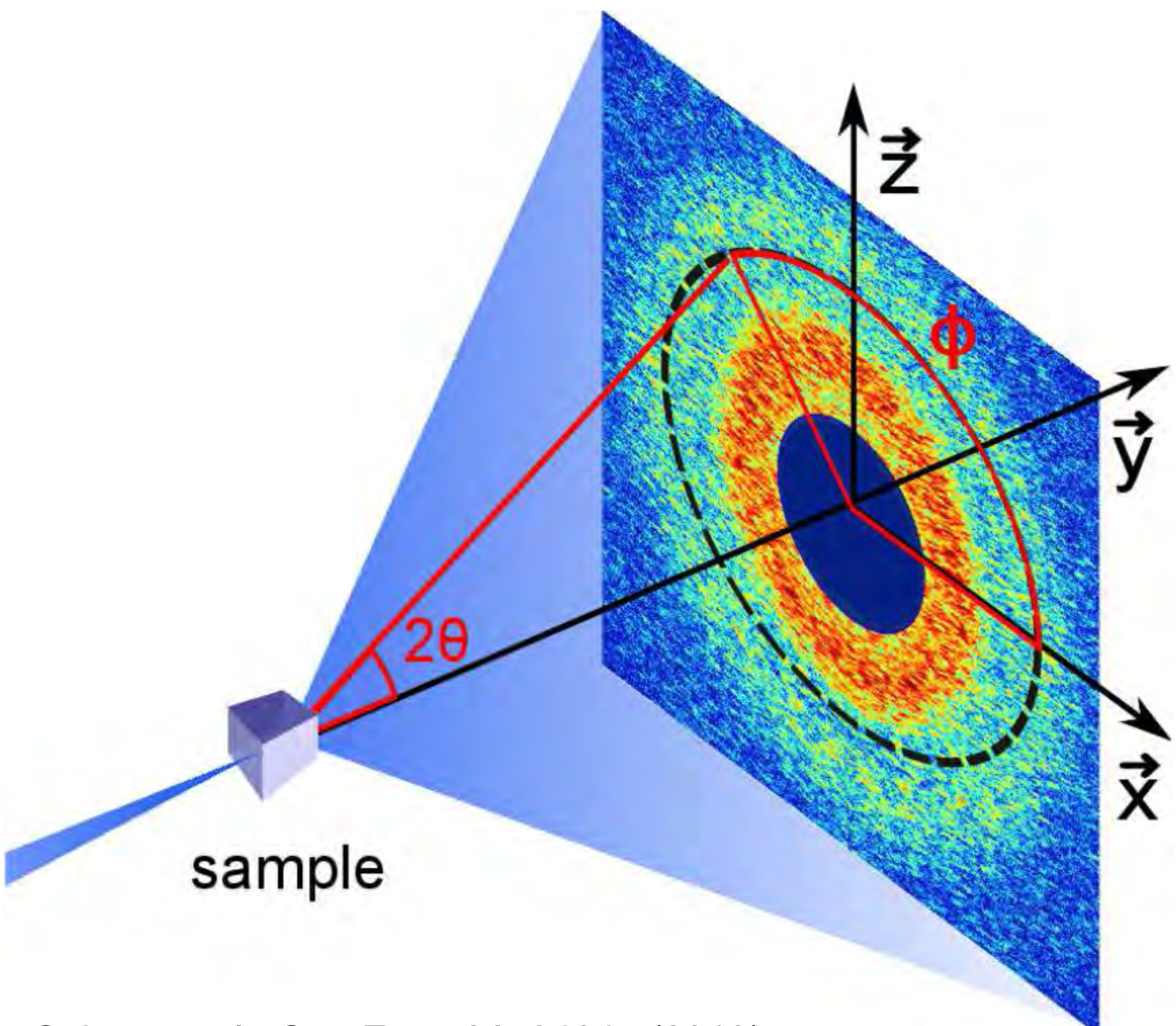
- Fresnel zone plate (~20 nm)
- multilayer mirror (7 nm)
- multilayer Laue lenses (8 nm)
- bent crystals





Workaround: Coherent X-Ray Diffraction Imaging

Sample illuminated by coherent X-rays:



S. Lee, et al., Opt. Expr. **21**, 24647 (2013).

Speckle pattern encodes information about the sample

No optics:

> No limitation of numerical aperture

$$NA = \sin 2\theta$$

➜ limited by intensity in diffraction pattern

> But no direct image of sample

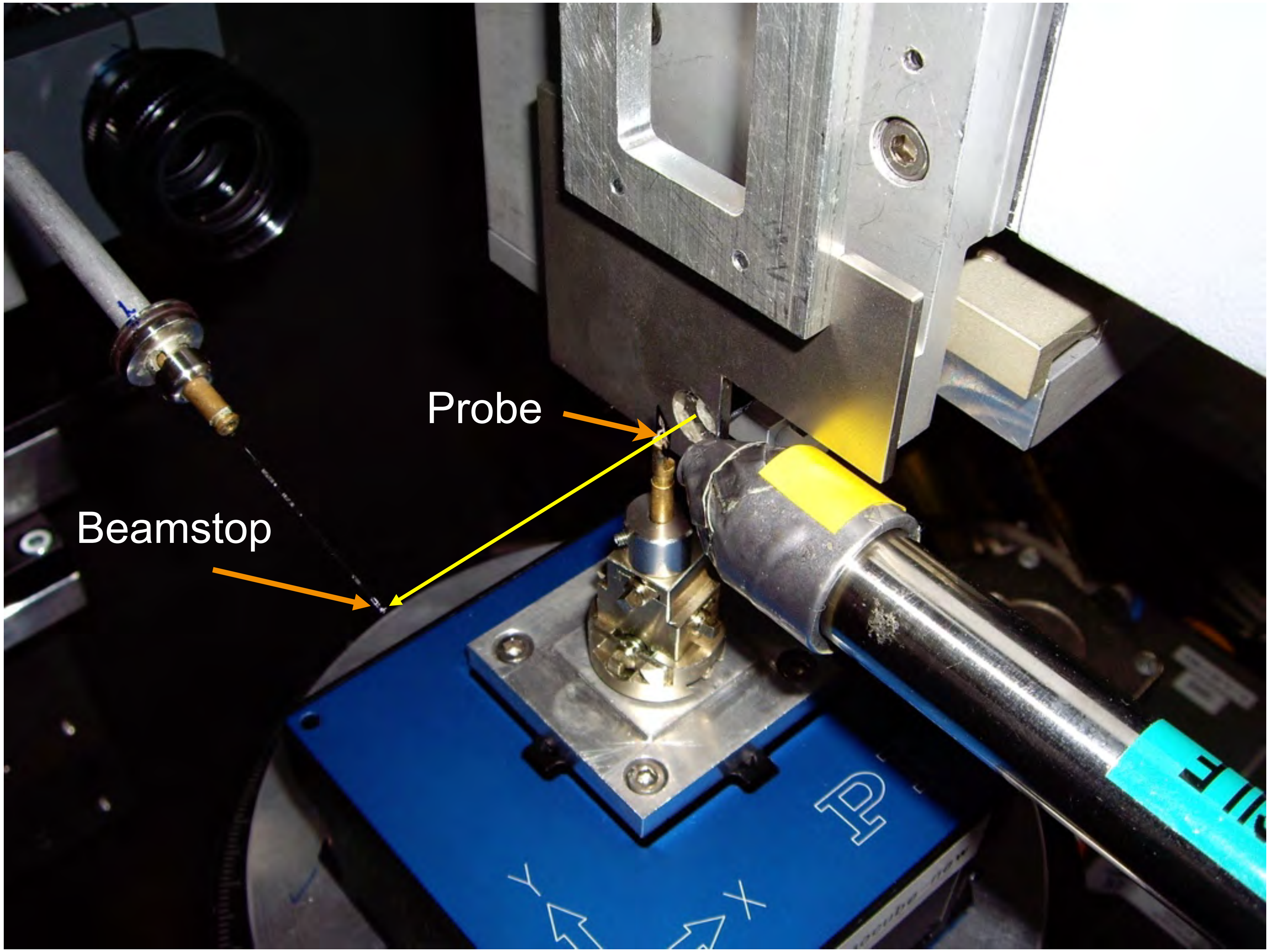
➜ Far-field diffraction pattern

➜ Absolute square of Fourier amplitudes

Important ingredient:

Coherent light

Example: Coherent X-Ray Diffraction Imaging



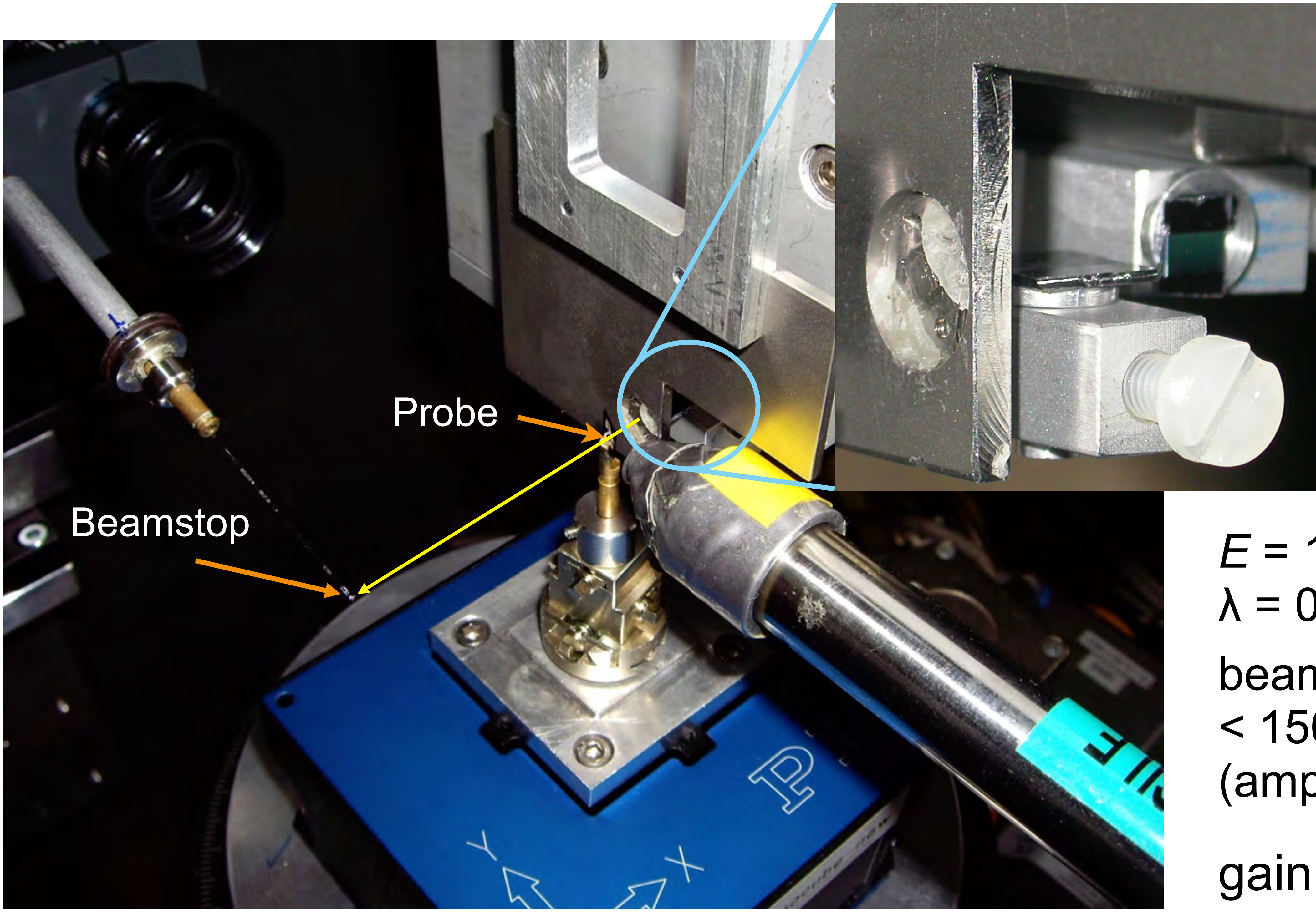
E = 15.25 keV
λ = 0.813 Å

beam size:
< 150 x 150 nm^2
(amplitude)

gain: 10^4

C. Schroer, et al., Phys. Rev. Lett. 101, 090801 (2008).

Example: Coherent X-Ray Diffraction Imaging



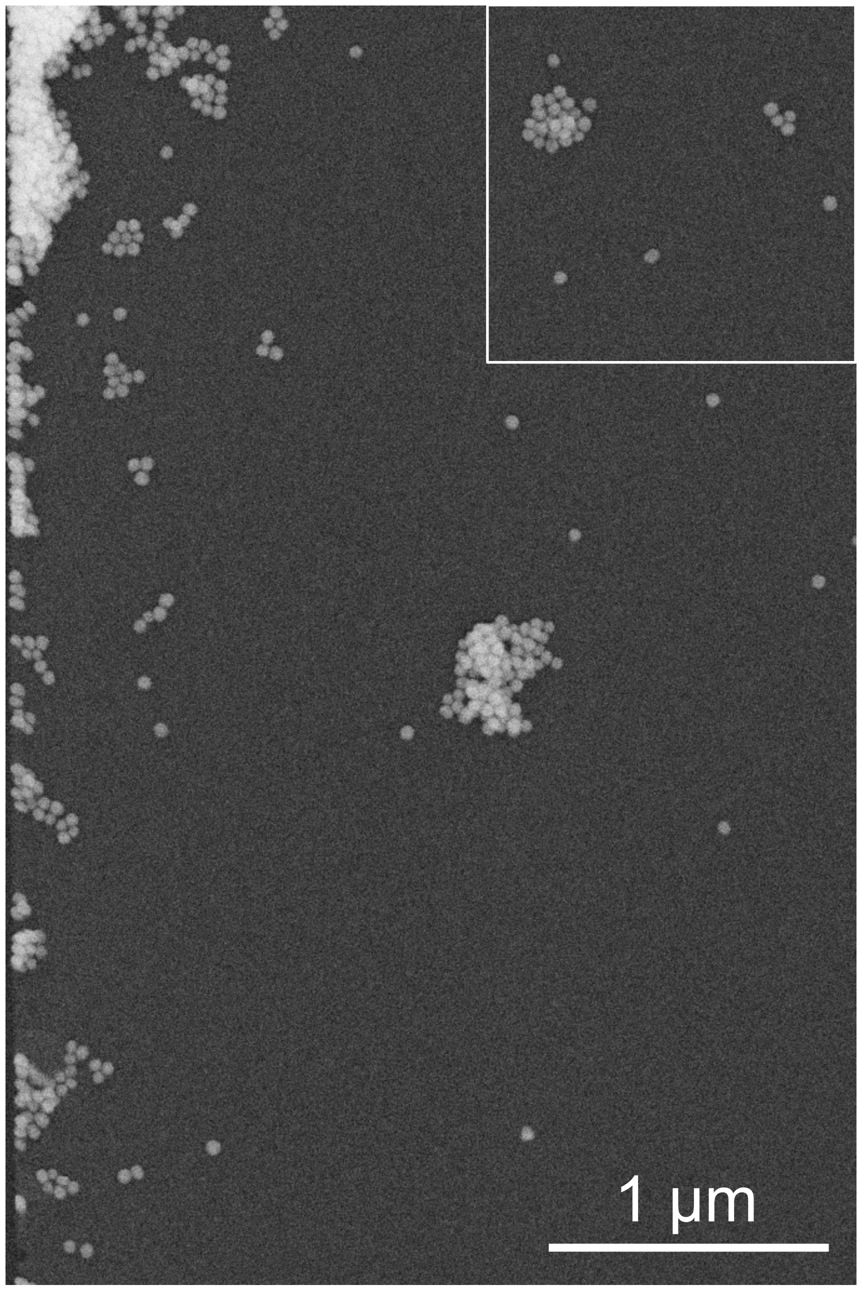
E = 15.25 keV
λ = 0.813 Å

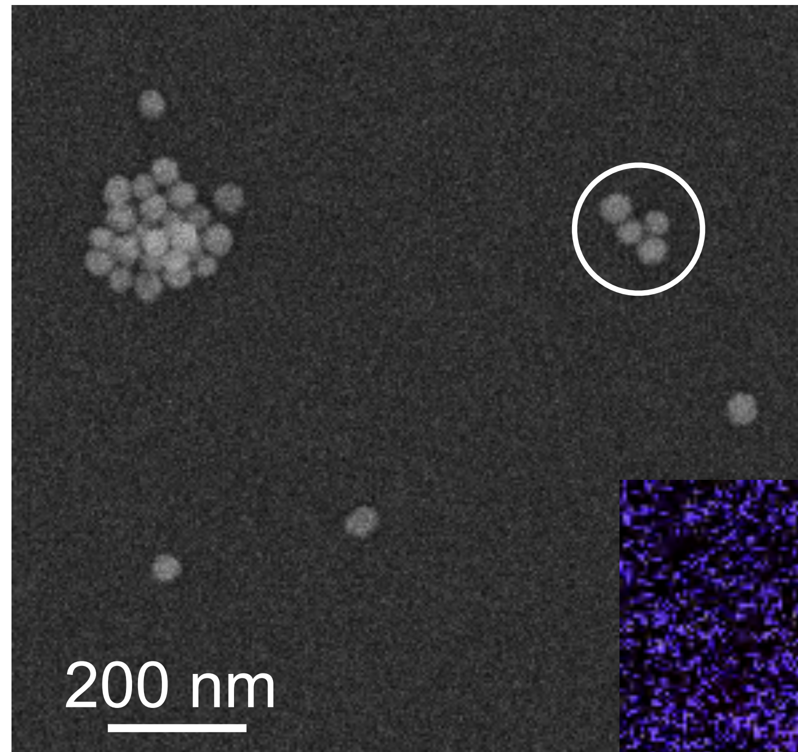
beam size:
< 150 x 150 nm^2
(amplitude)

gain: 10^4

C. Schroer, et al., Phys. Rev. Lett. 101, 090801 (2008).

Imaging of a Small Object





4 gold particles:

diameter ~ 40 nm

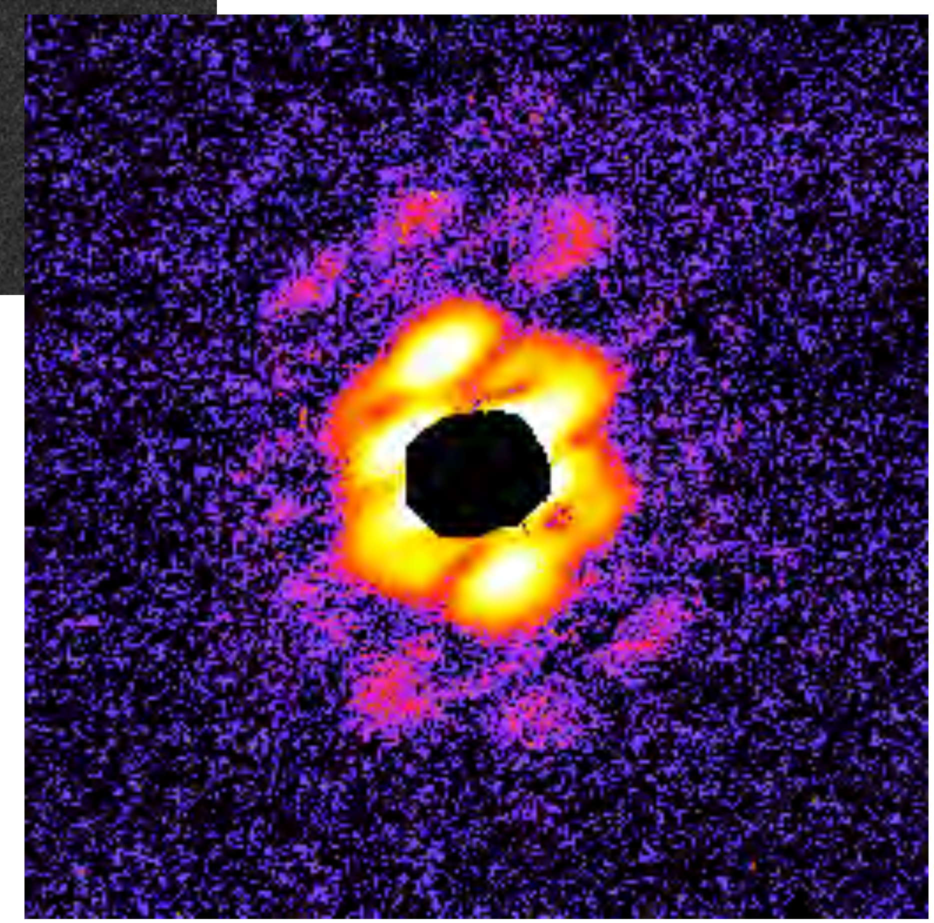
→ $7.5 \cdot 10^6$ atoms

fluence:

$1 \cdot 10^5$ ph/nm^2

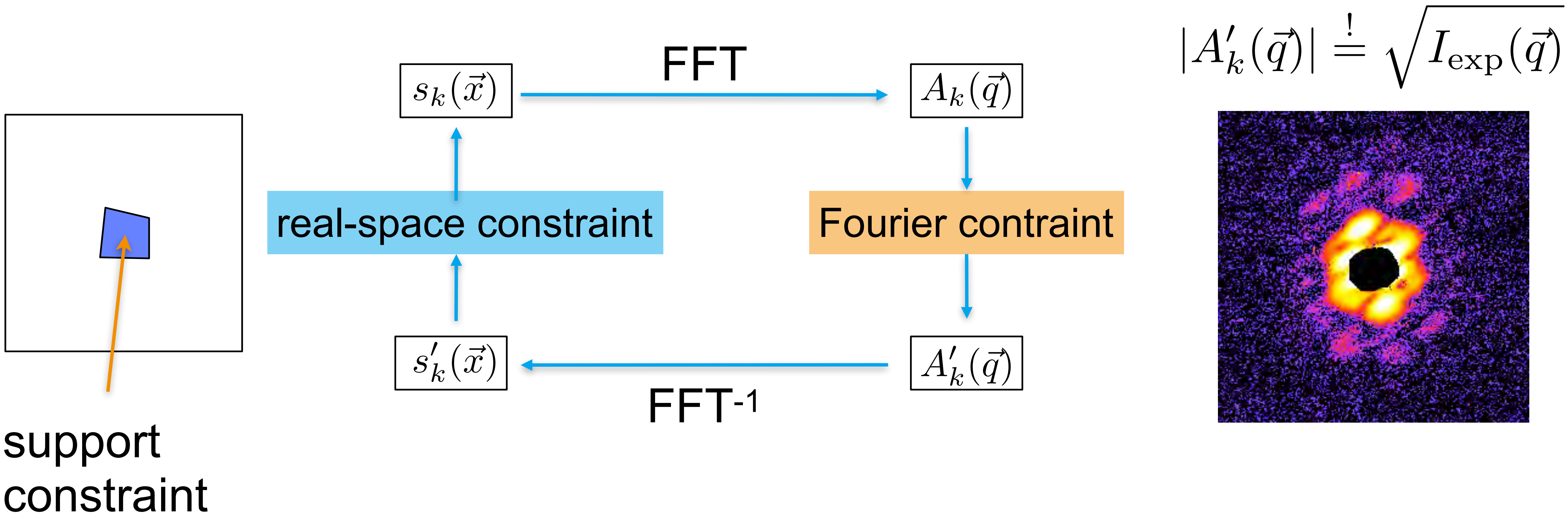
dose:

$1 \cdot 10^{10}$ ph



Coherent X-ray Diffraction Imaging

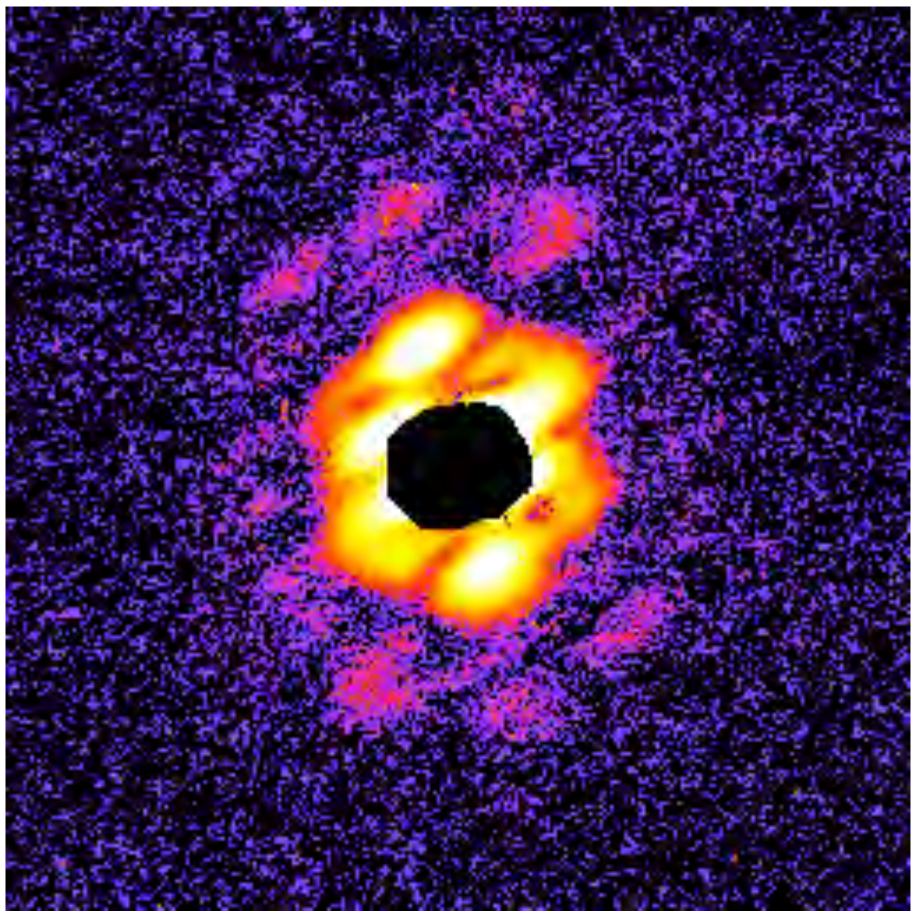
Iterative phase reconstruction:



R. W. Gerchberg & W. O. Saxton, *Optic* (1972) **35**, 237
J. R. Fienup, *Appl Opt.* (1982). **21**, 2758
R. P. Millane & W.J. Stroud, *J. Opt. Soc. Am.* (1997) **A14**, 568

Coherent X-ray Diffraction Imaging

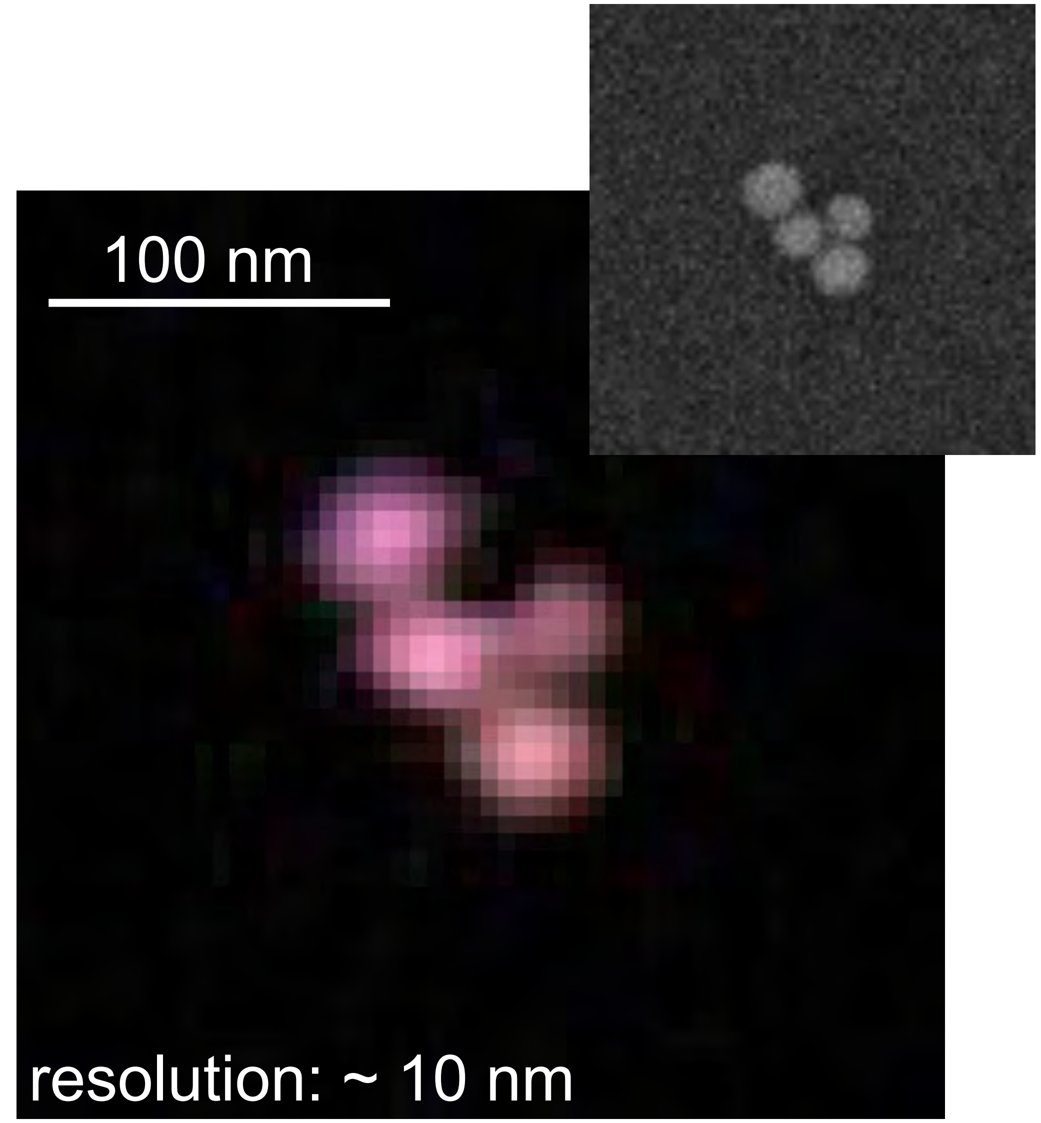
Iterative phase reconstruction:



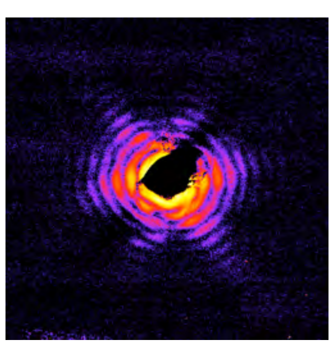
HIO

→

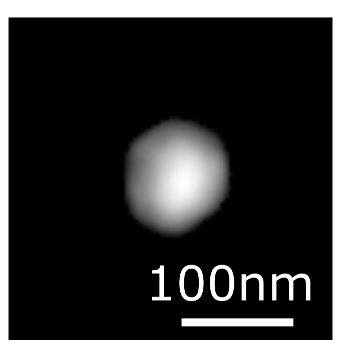
shrink-wrap



record resolution: 5 nm



→



CXDI at an individual gold particle

PRL **101**, 090801 (2008)

Difficulty:

> half of the information is missing

> additional knowledge needed (e. g., support contraint)

Hard X-ray Scanning Microscopy at PETRA III

Microscope:

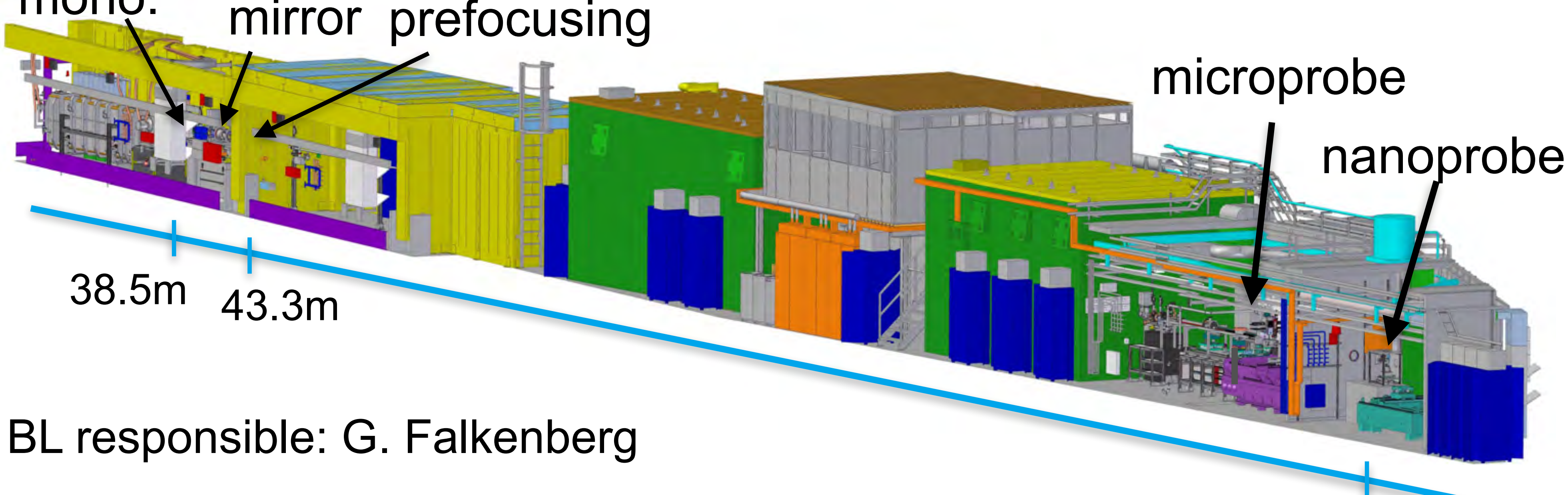
~98 m from source

different contrasts:

- fluorescence
- diffraction (SAXS, WAXS)
- absorption (XAS)
- XBIC/XBIV
- ptychography & CXDI

spatial resolution:
down to < 50 nm
down to < 5 nm (CXDI)

X-ray energy:
10 - 50 keV

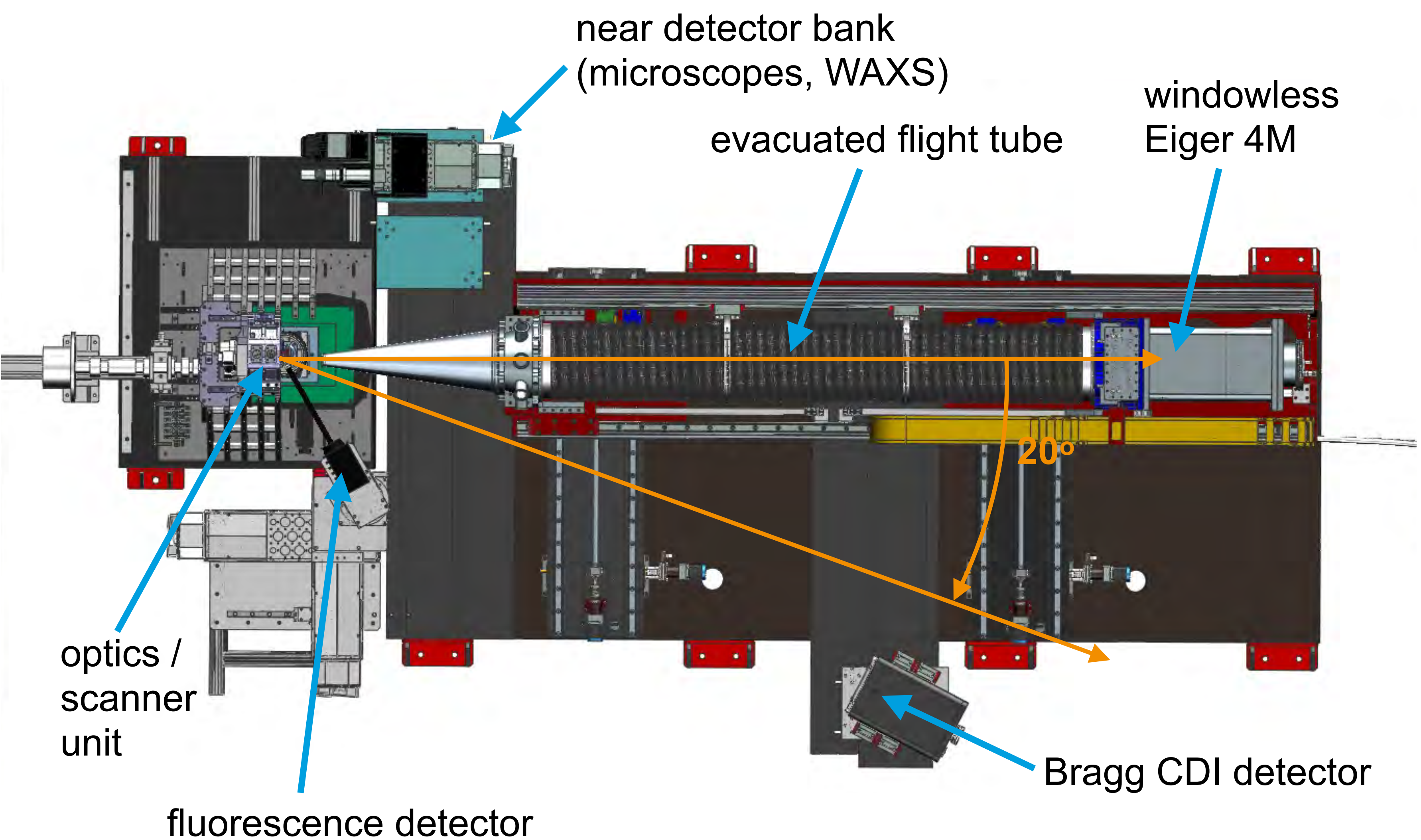


Nucl. Instrum. Meth. A **616** (2-3), 93 (2010).

BL responsible: G. Falkenberg

PtyNAMi: Ptychographic Nano-Analytical Microscope

Ongoing Upgrade Project



Goals:

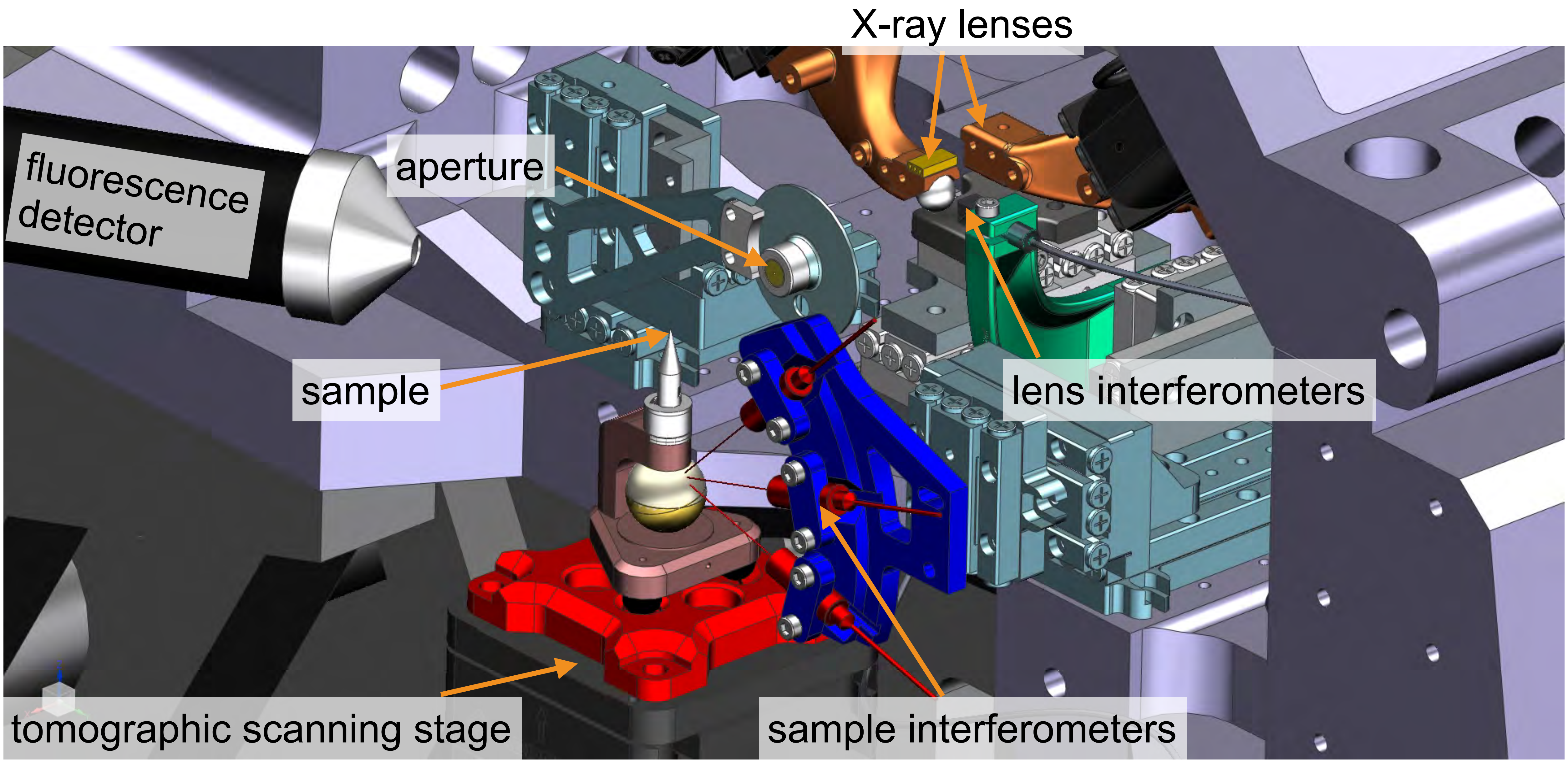
> multimodal: ptychography, XRF, SAXS, WAXS, XAS
> high spatial resolution
> high sensitivity
> 2D and 3D imaging
> *in situ* & *operando*

Experimental requirements:

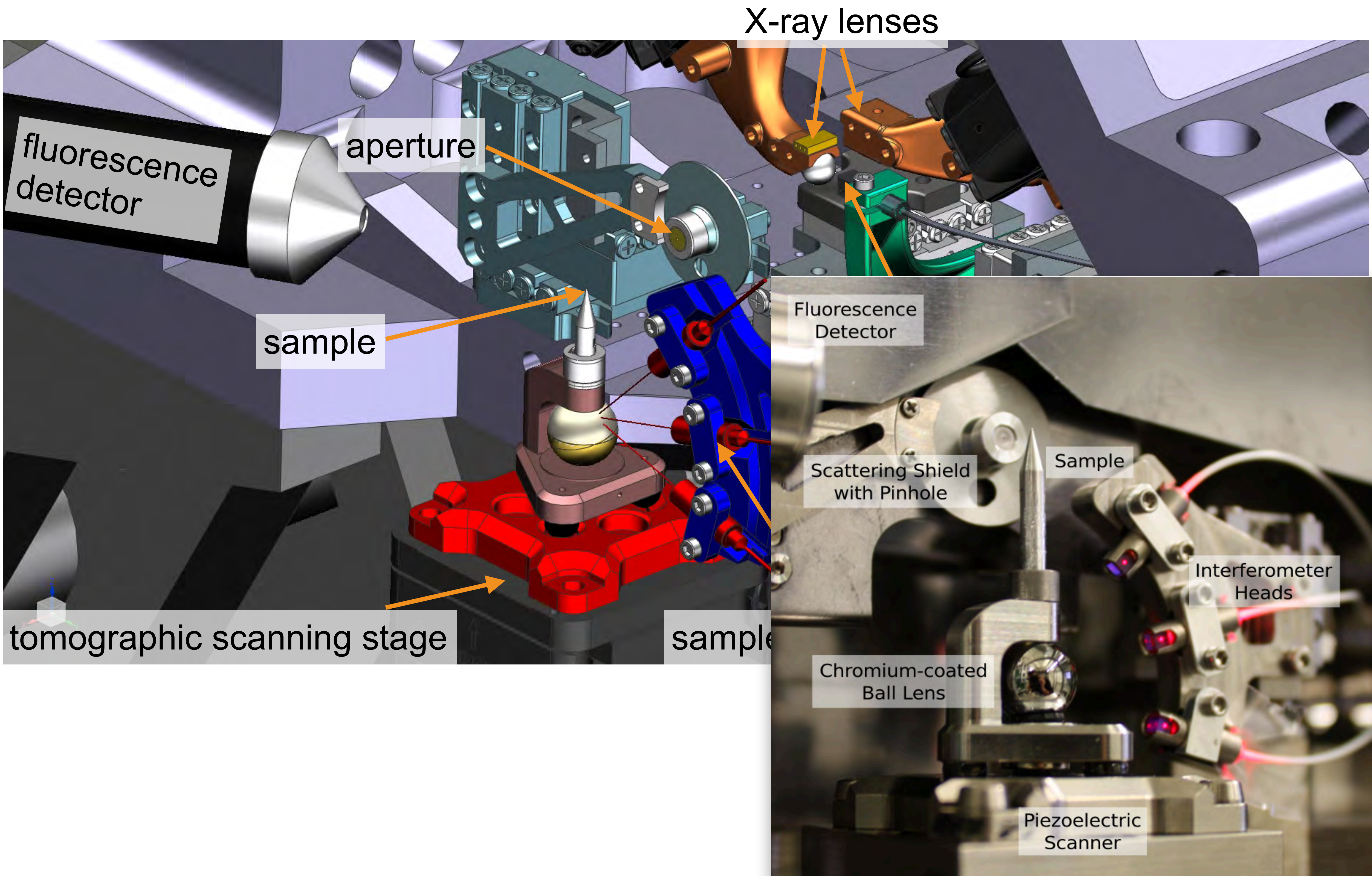
> optimised coherent flux with pre-focusing
> high-performance optics
> high mechanical stability and control
> low background

A. Schropp, et al., *PtyNAMi: Ptychographic Nano-Analytical Microscope*, J. Appl. Crystallogr. **53**, 957 (2020).

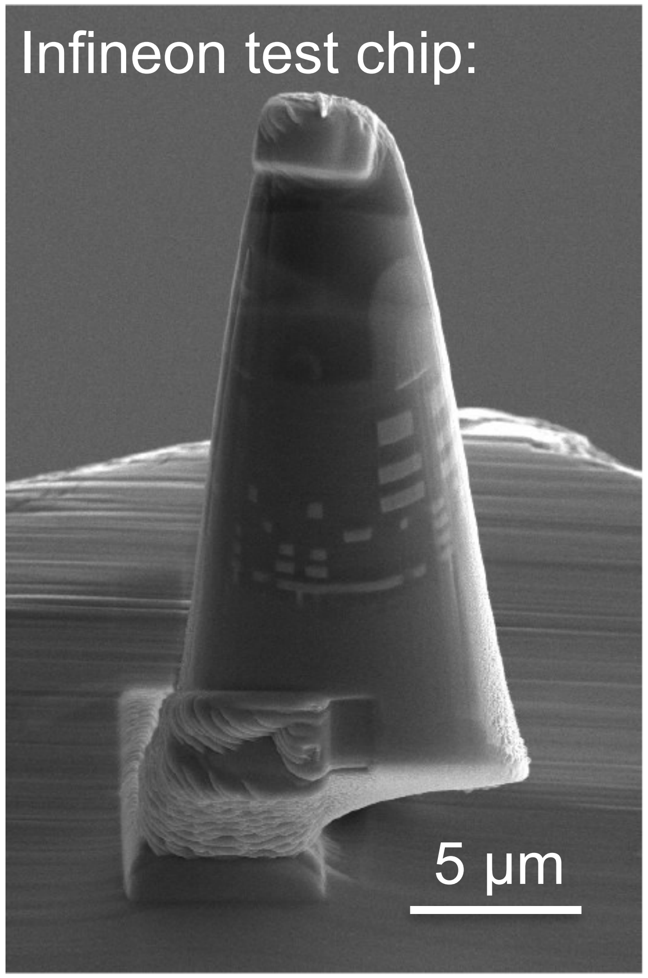
PtyNAMi: Optics and Scanner Unit

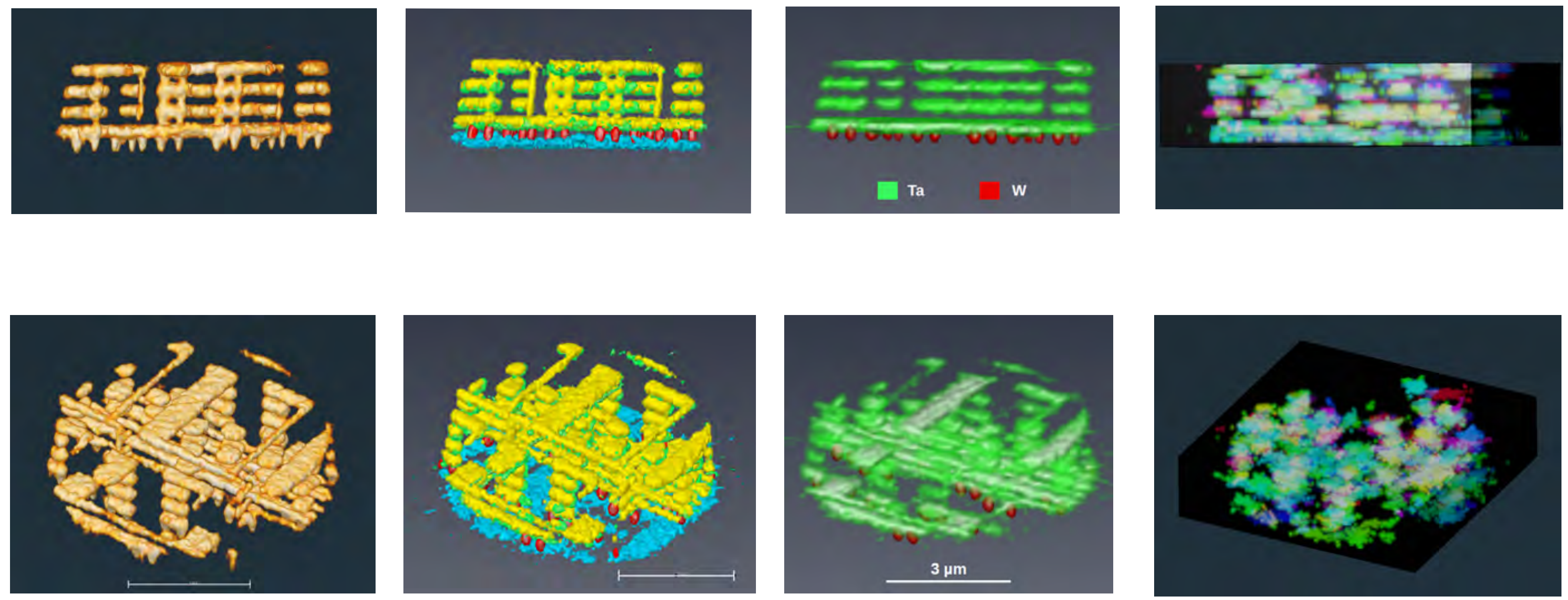


PtyNAMi: Optics and Scanner Unit



Example: Micro-Electronic Devices





E = 18 keV

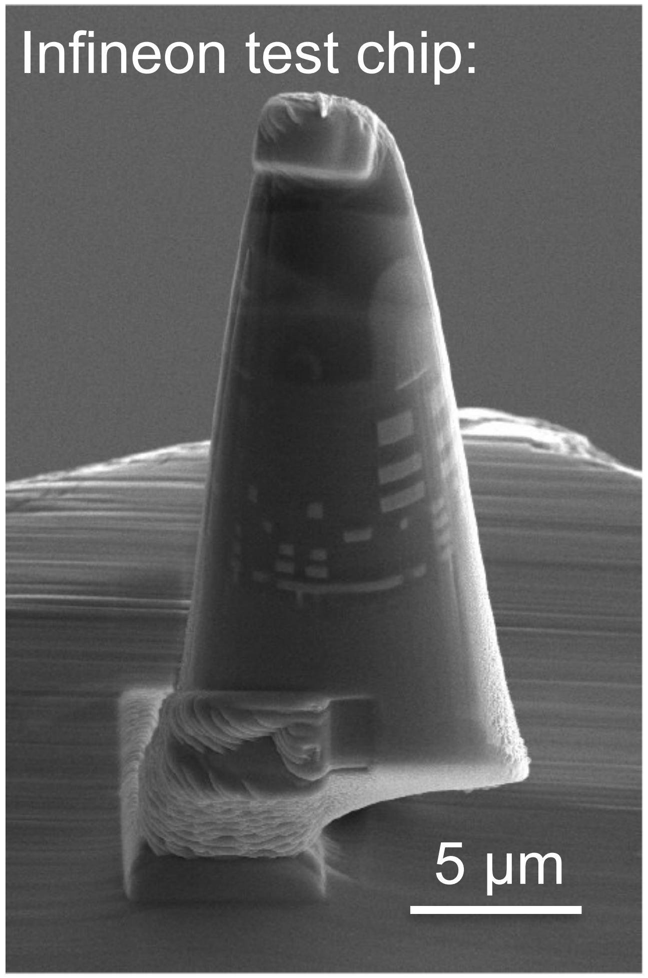
beam size: 61 x 80 nm^2

tomographic scan:

> fluorescence radiation
> diffraction patterns

PhD work: Maria Scholz

Example: Micro-Electronic Devices



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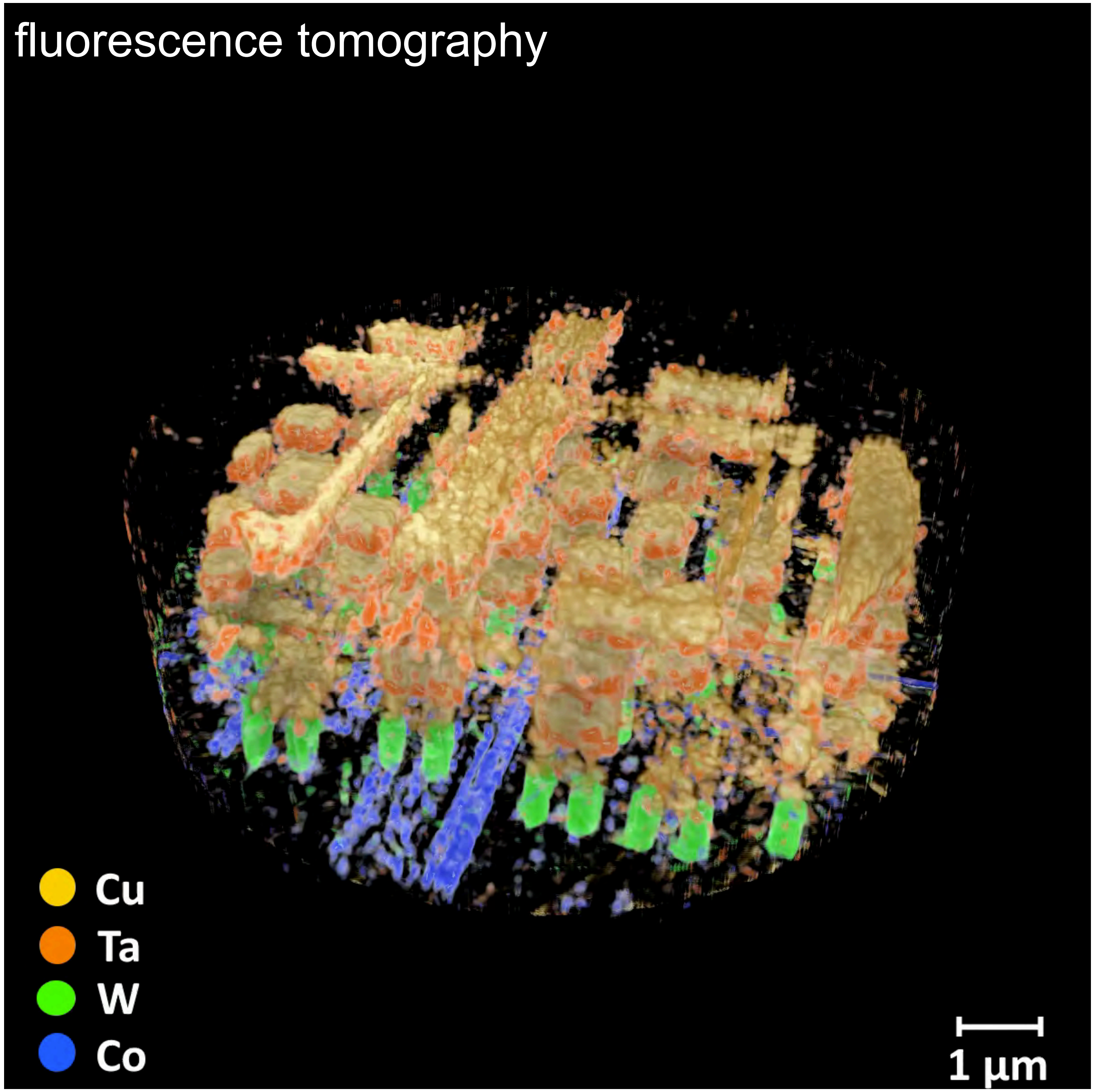
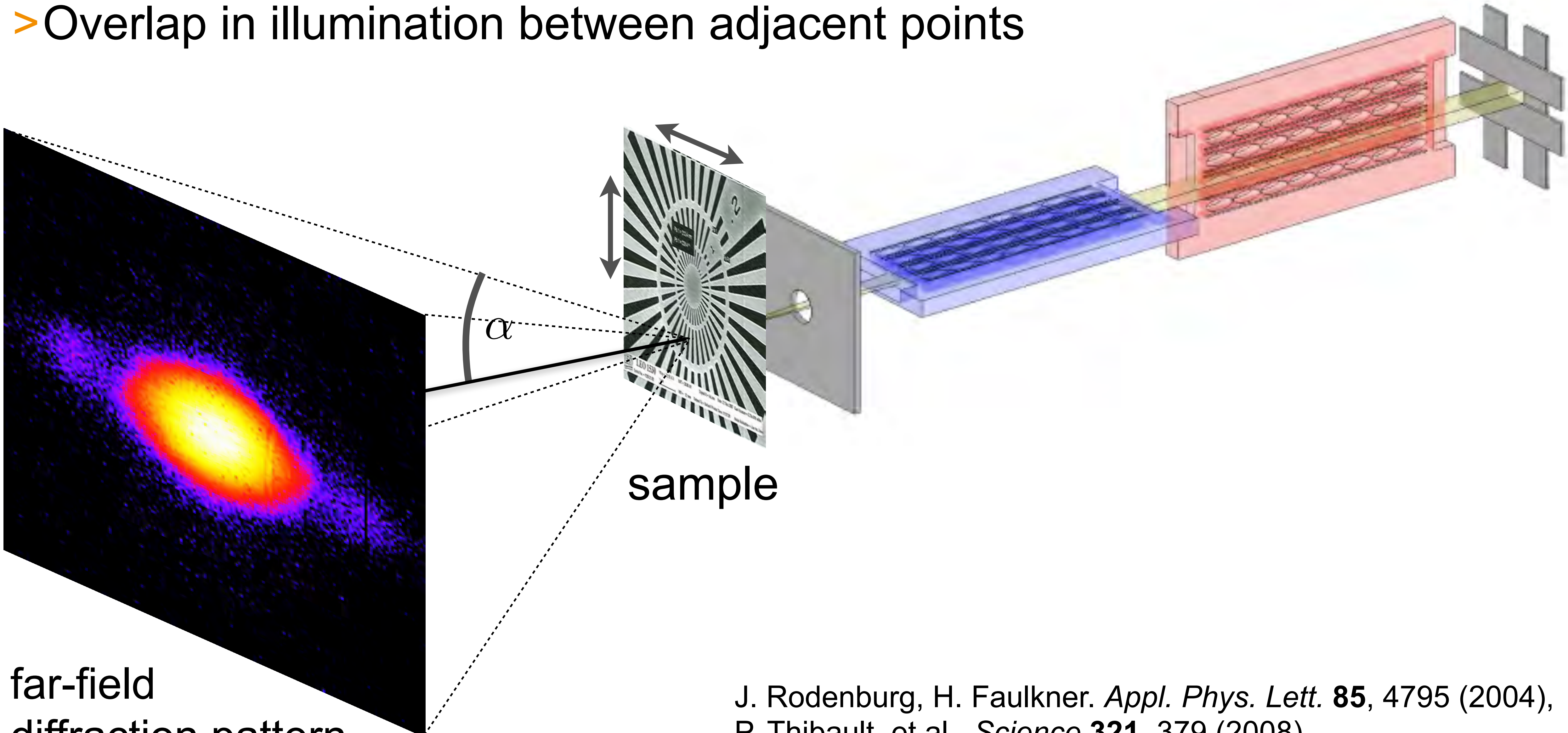
PhD work: Maria Scholz



Master thesis of Lukas Grote

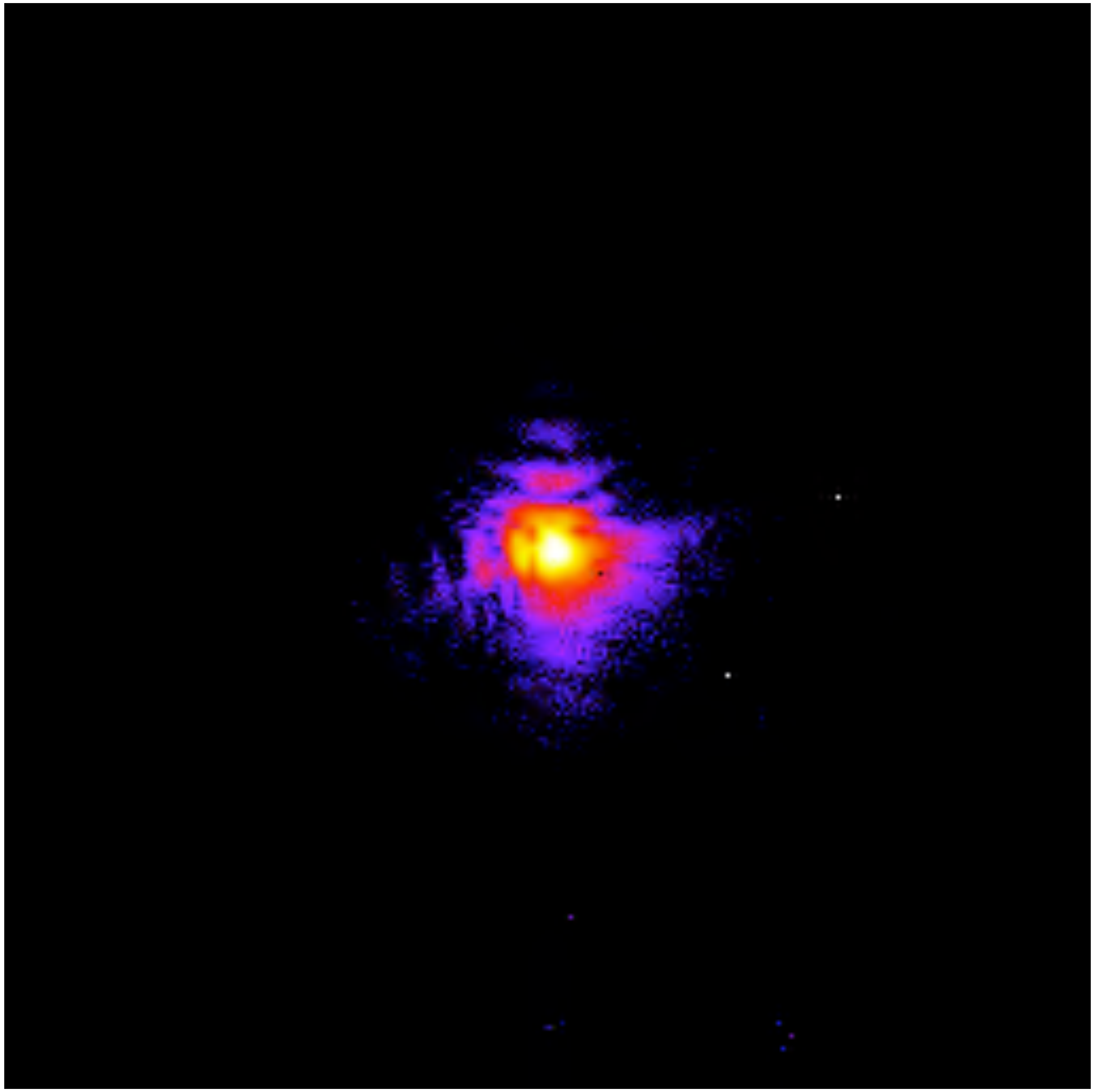
Scanning Coherent Diffraction Imaging: Ptychography

- Sample is raster scanned through confined beam
- At each position of scan: diffraction pattern is recorded
- Overlap in illumination between adjacent points



J. Rodenburg, H. Faulkner. *Appl. Phys. Lett.* **85**, 4795 (2004),
P. Thibault, et al., *Science* **321**, 379 (2008),
A. Schropp, et al., *Appl. Phys. Lett.* **96**, 091102 (2010),
M. Dierolf, et al., *Nature* **467**, 436 (2010).

Ptychography: Characterization of Nanobeam



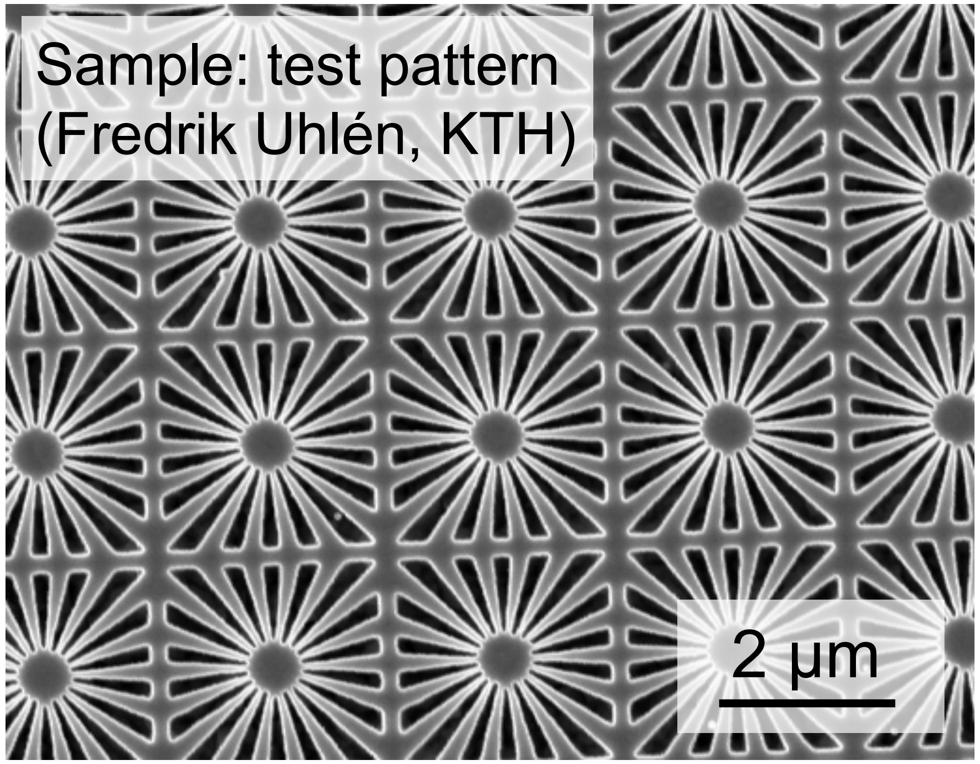
Nanofocusing lenses at PETRA III

E = 8 keV

detector:
Pilatus 300k (172μm pixel size)

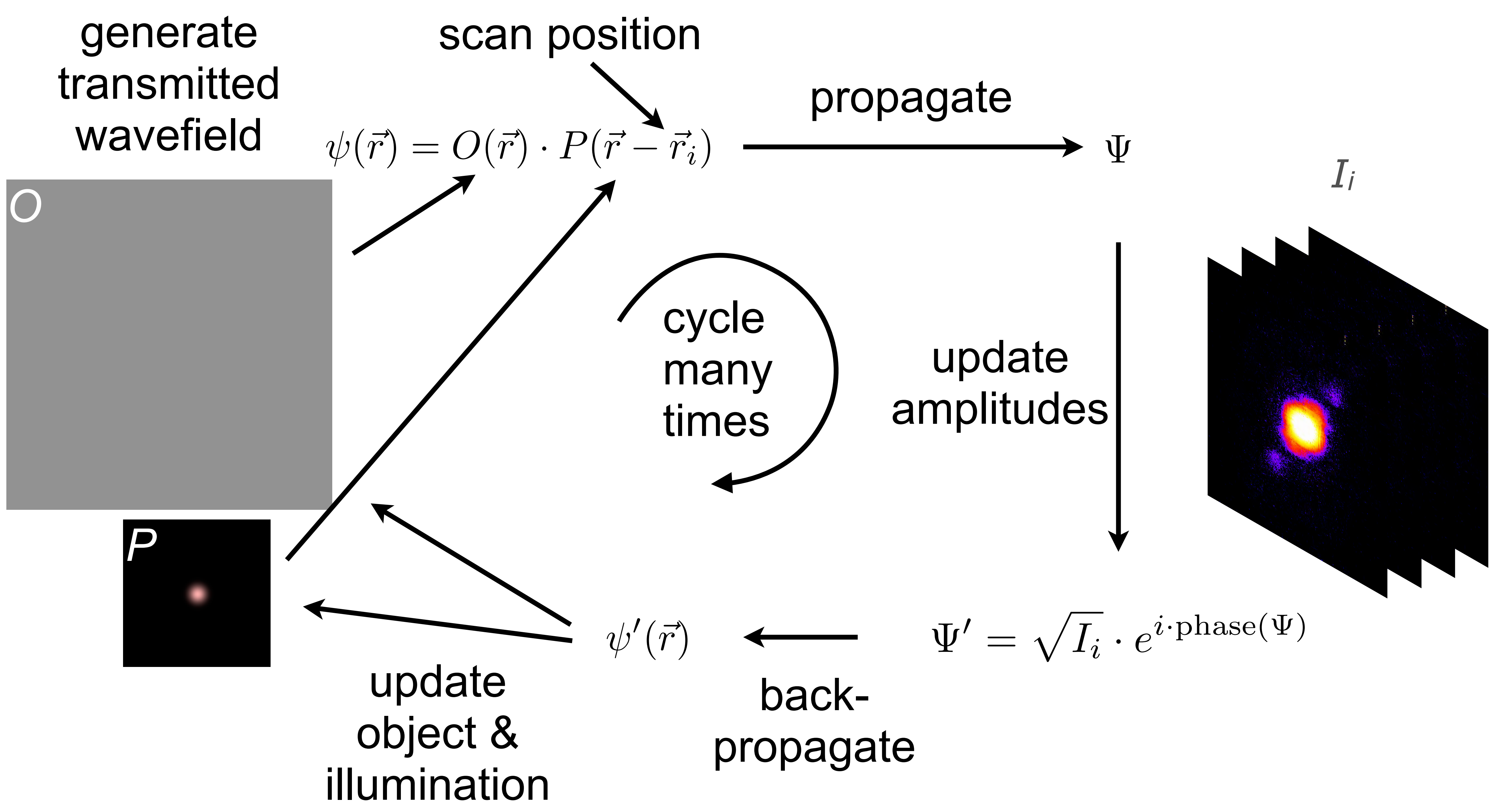
sample-detector distance:
2080 mm

exposure time:
1.0 s per point



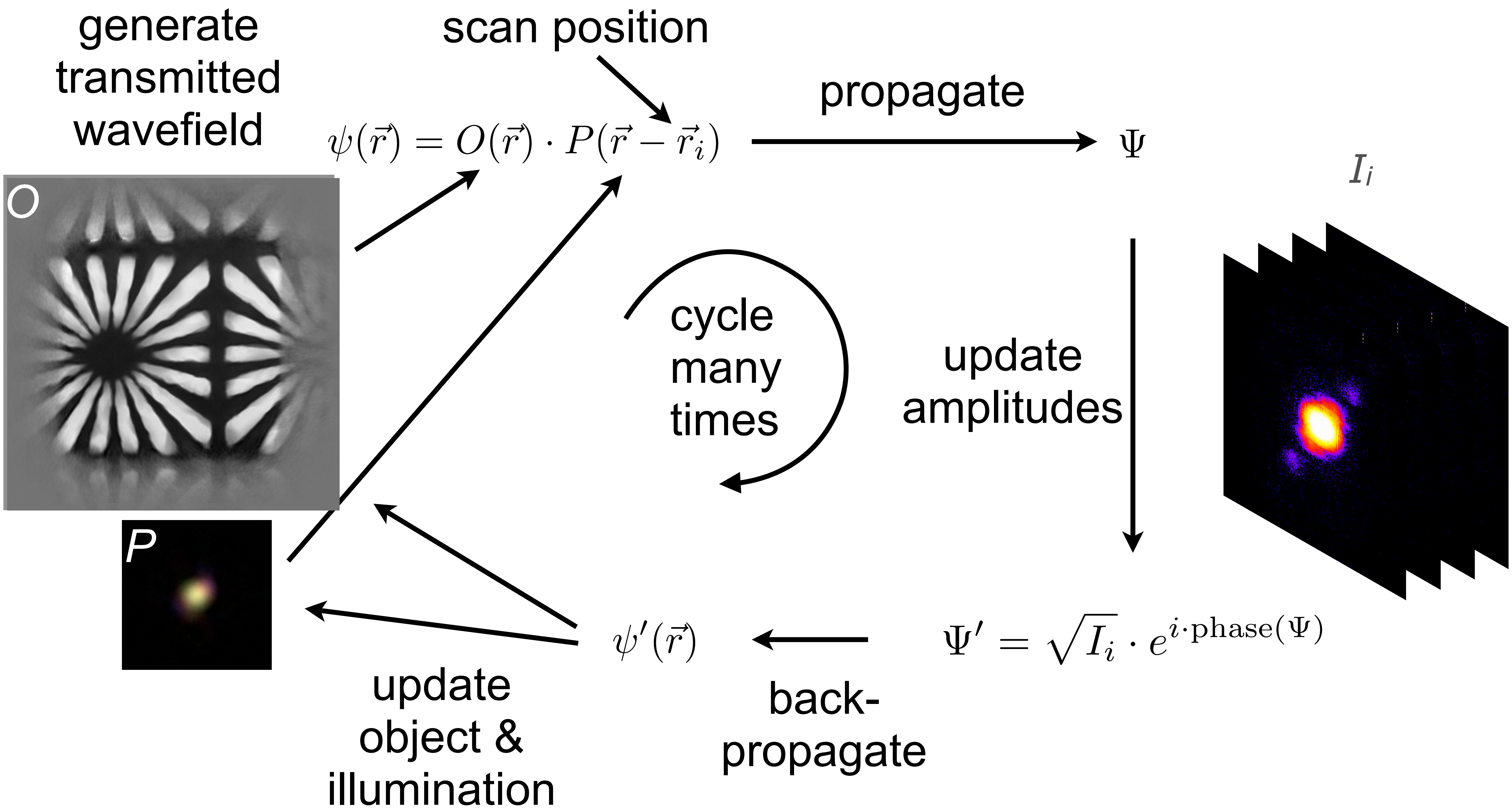
A. Schropp, et al., Appl. Phys. Lett. **96**, 091102 (2010).
C. G. Schroer, et al., Proc. SPIE **8848**, 884807 (2013).

Ptychography: Reconstruction



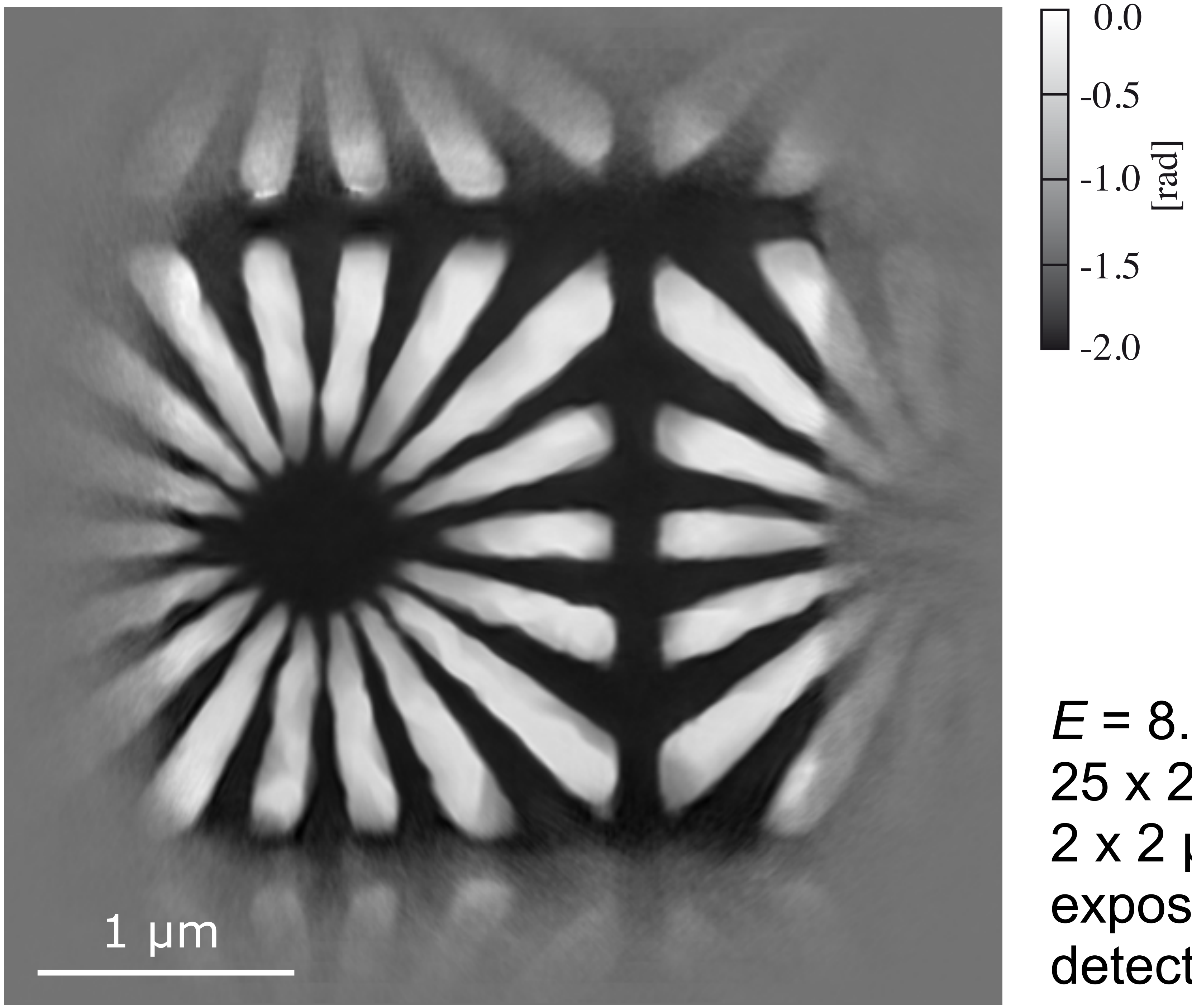
Maiden & Rodenburg, Ultramicroscopy **109**, 1256 (2009).

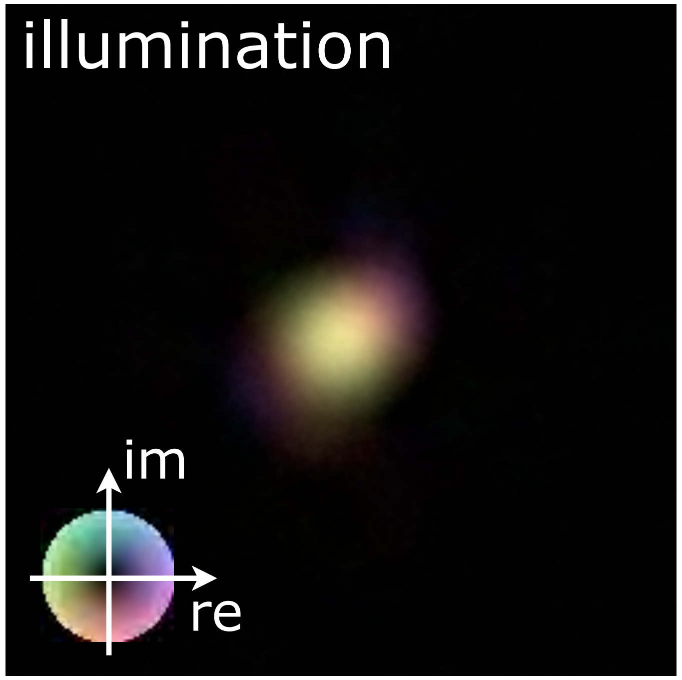
Ptychography: Reconstruction



Maiden & Rodenburg, Ultramicroscopy **109**, 1256 (2009).

Ptychography: Reconstruction of Sample and Probe



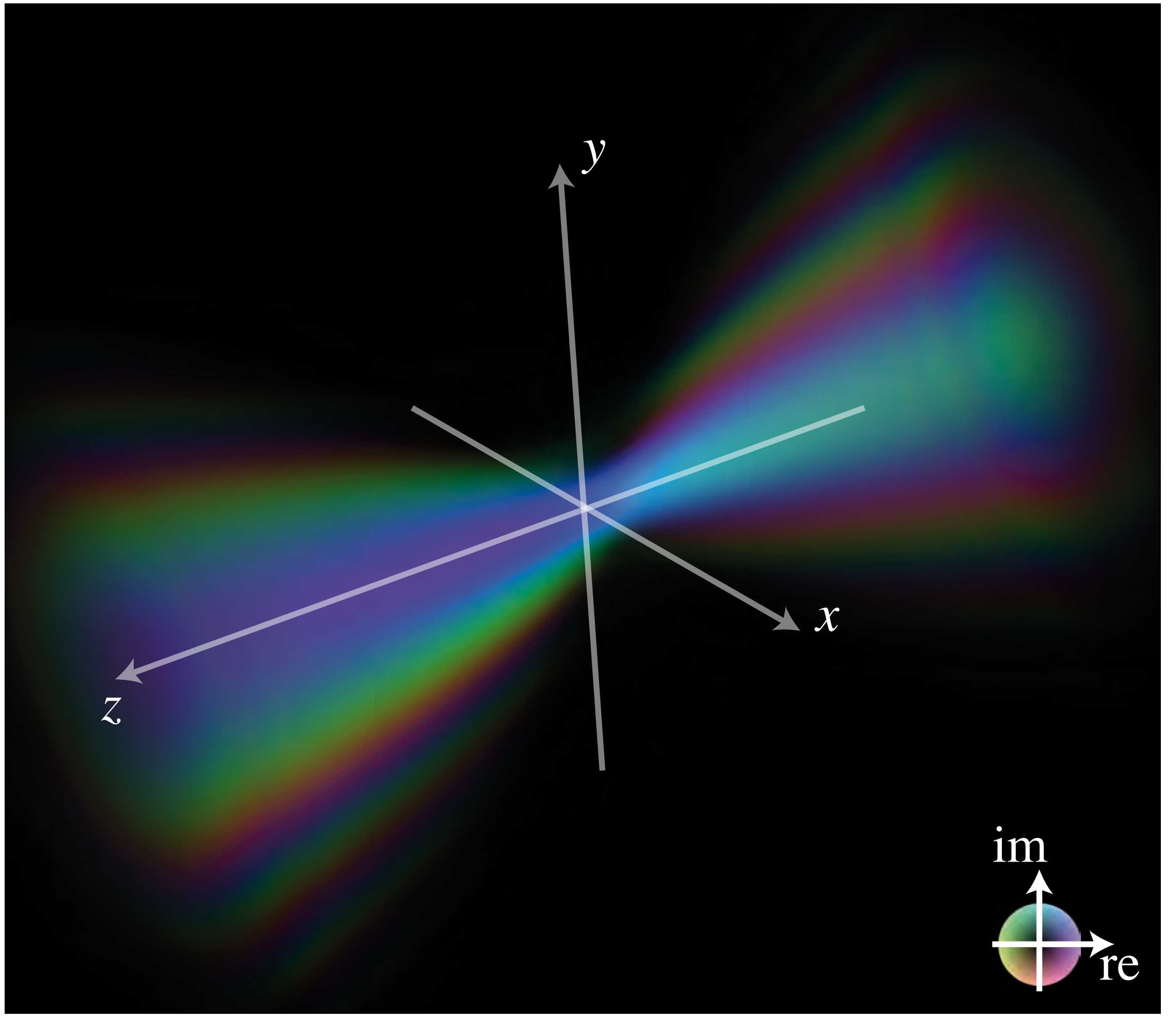


Full state (solution of Helmholtz equation)

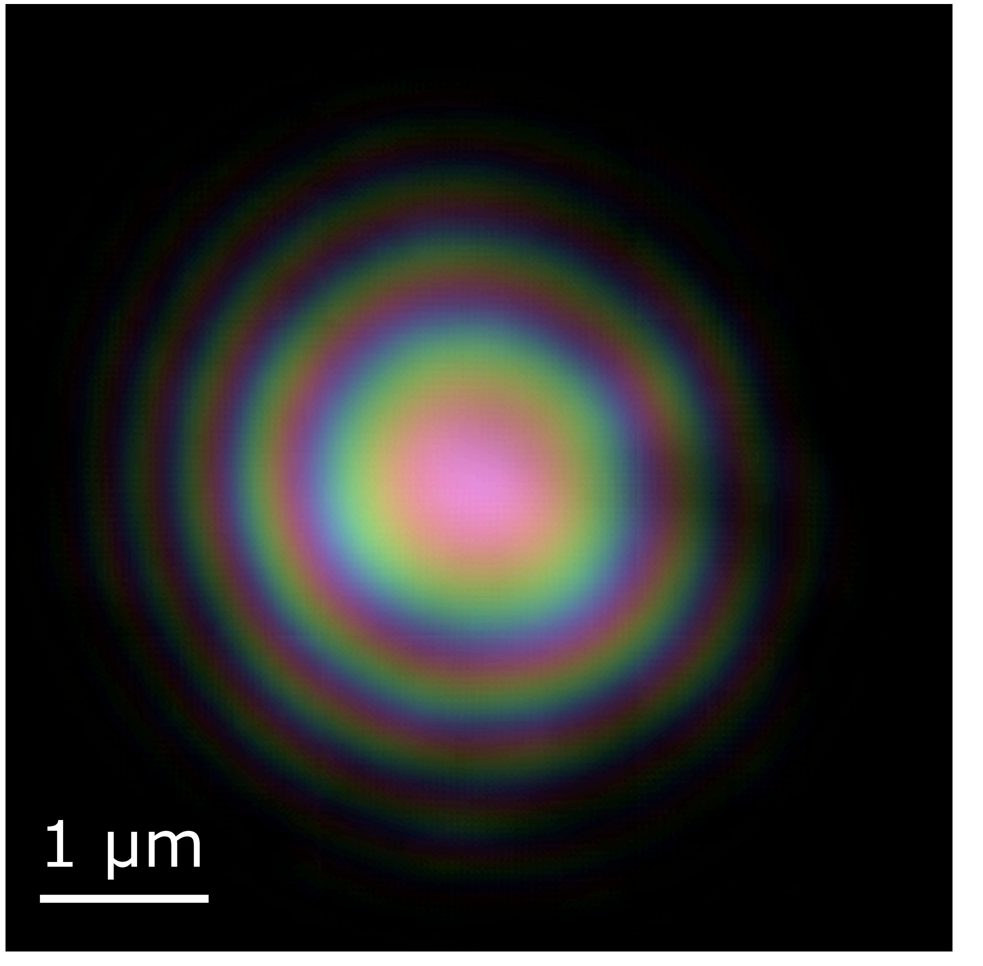
E = 8.0 keV
25 x 25 steps of 80 x 80 nm^2
2 x 2 µm^2 FOV
exposure: 1.0 s per point
detected fluence: 120 ph/nm^2

A. Schropp, et al., Appl. Phys. Lett. **96**, 091102 (2010).
C. G. Schroer, et al., Proc. SPIE **8848**, 884807 (2013).

Reconstructed Wave Field



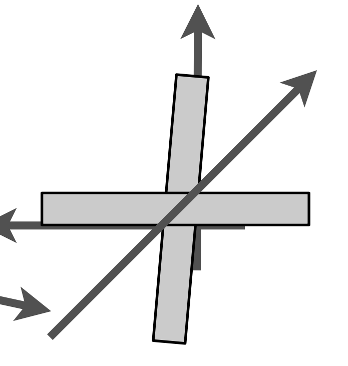
Caustic: -4 mm to 4 mm



Slight astigmatism:

improper lens alignment:

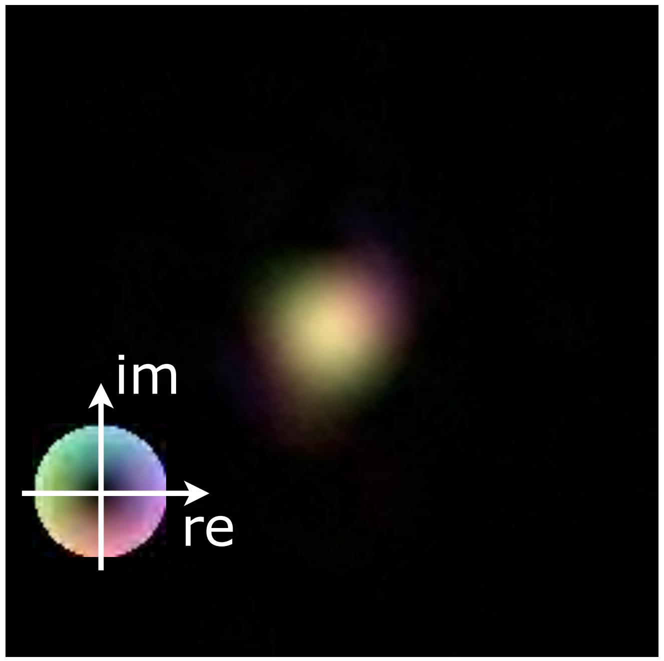
more strongly focusing direction

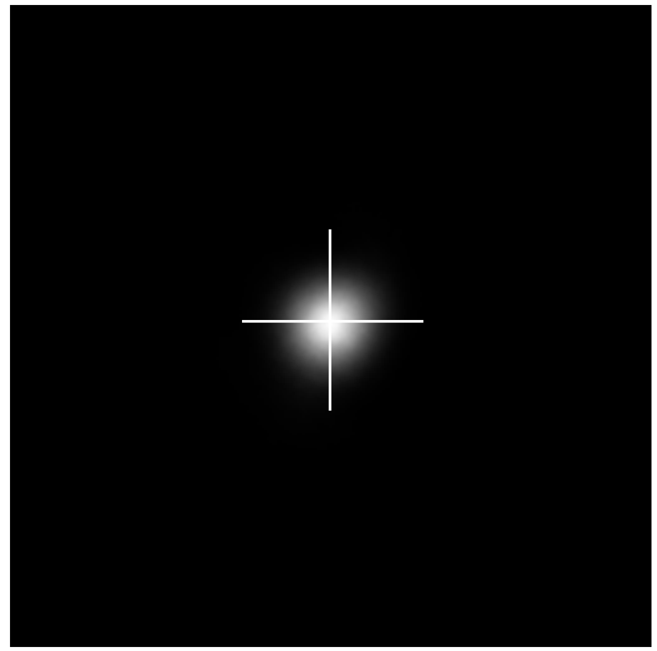


A. Schropp, et al., Appl. Phys. Lett. **96**, 091102 (2010).
C. G. Schroer, et al., Proc. SPIE **8848**, 884807 (2013).

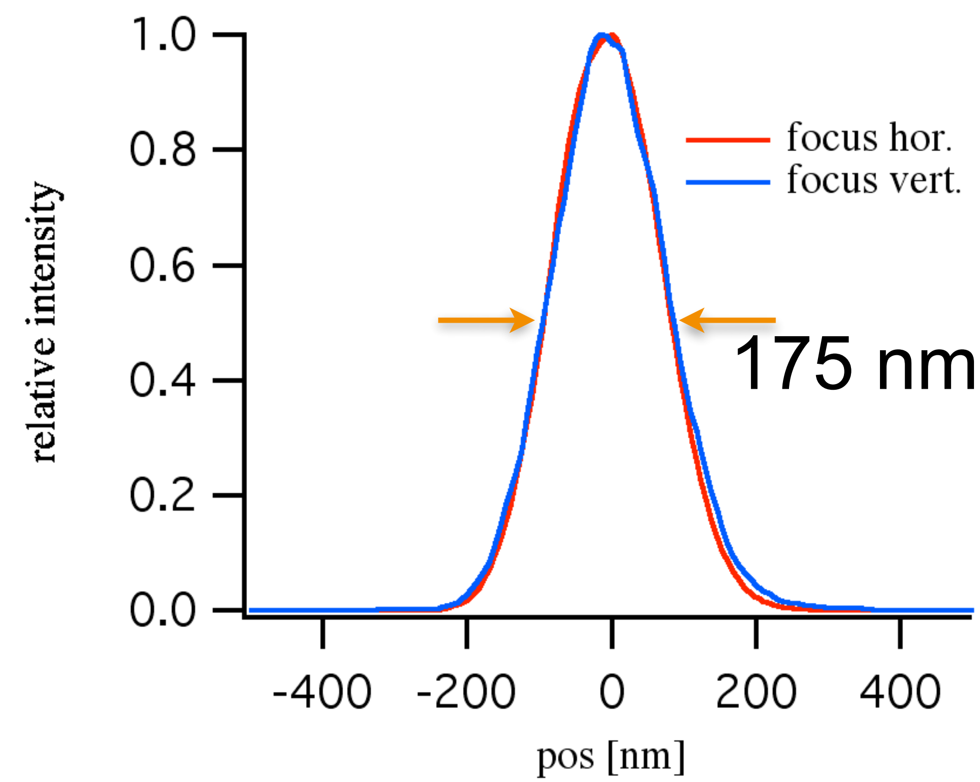
Evaluation of the Complex Wave Field

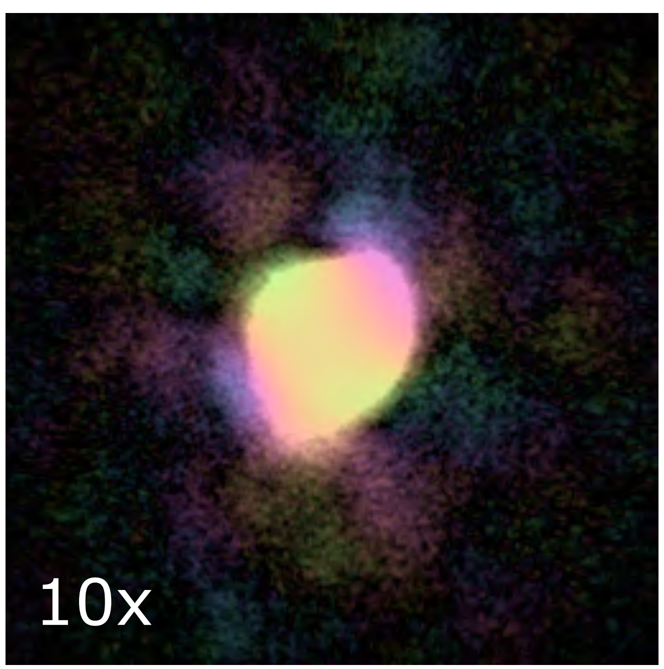
complex amplitude: intensity:





$1/\sqrt{2}\times$ width of amplitude



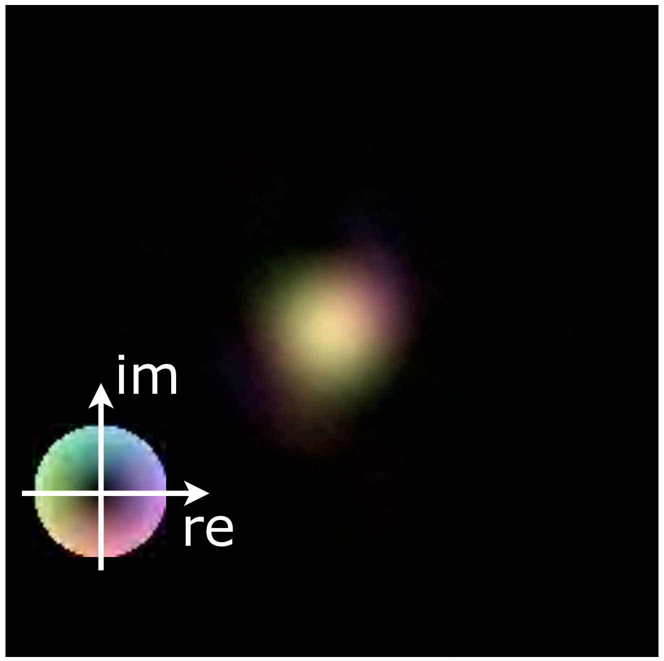


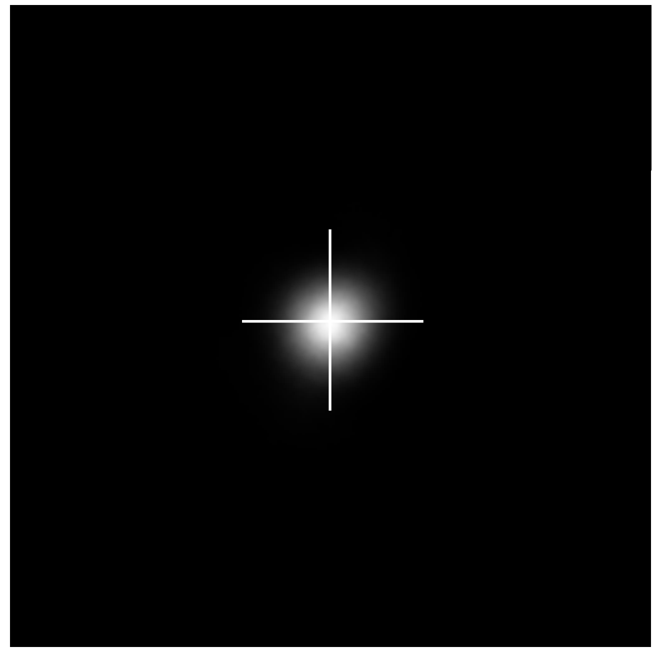
ideal focus: 155 x 175 nm^2

A. Schropp, et al., Appl. Phys. Lett. **96**, 091102 (2010).
C. G. Schroer, et al., Proc. SPIE **8848**, 884807 (2013).

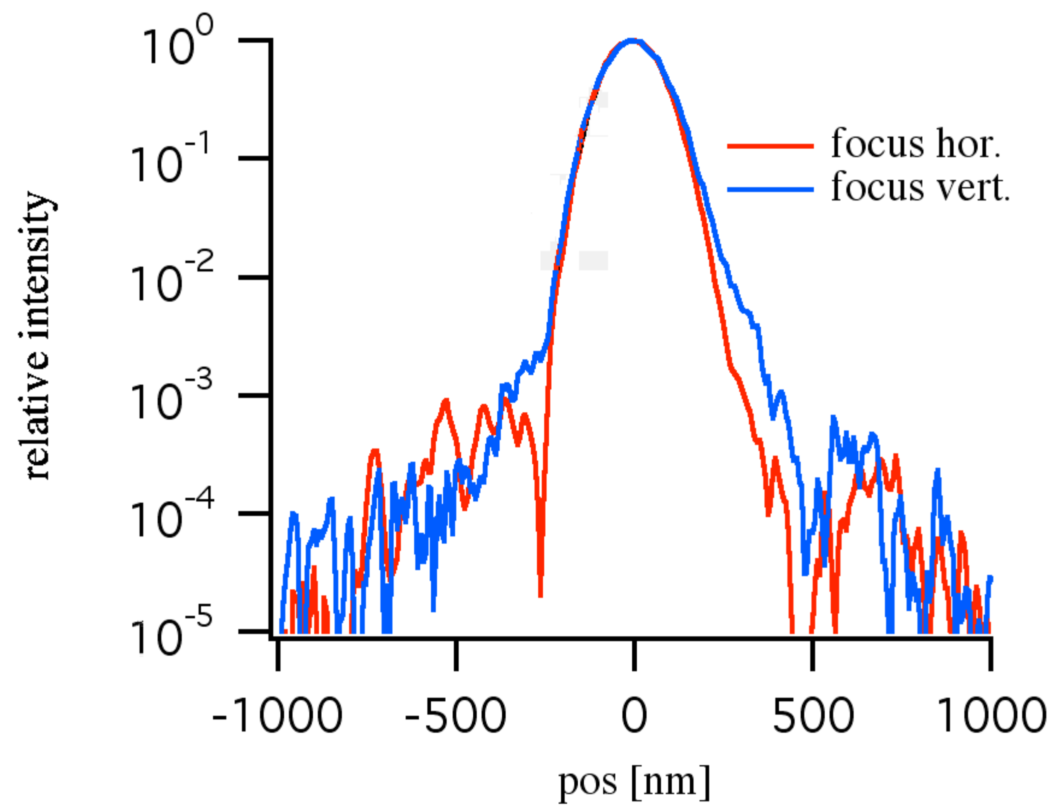
Evaluation of the Complex Wave Field

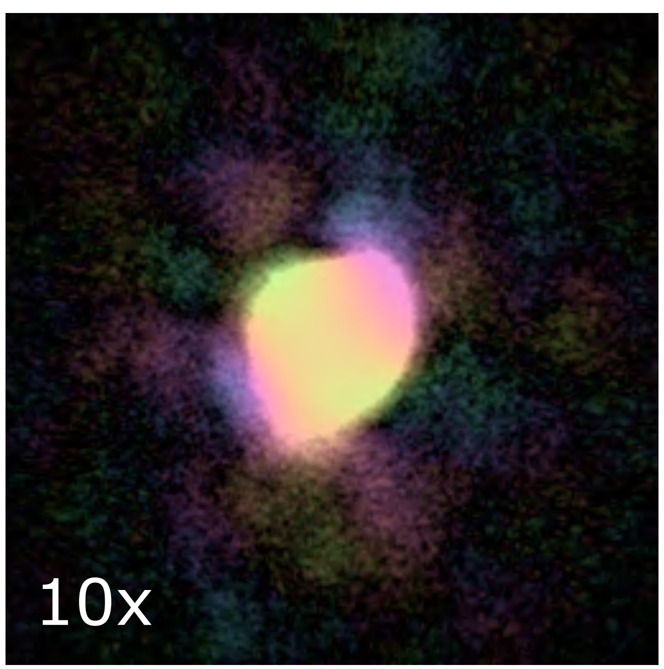
complex amplitude: intensity:





$1/\sqrt{2}\times$ width of amplitude





ideal focus: 155 x 175 nm²

A. Schropp, et al., Appl. Phys. Lett. **96**, 091102 (2010).
C. G. Schroer, et al., Proc. SPIE **8848**, 884807 (2013).

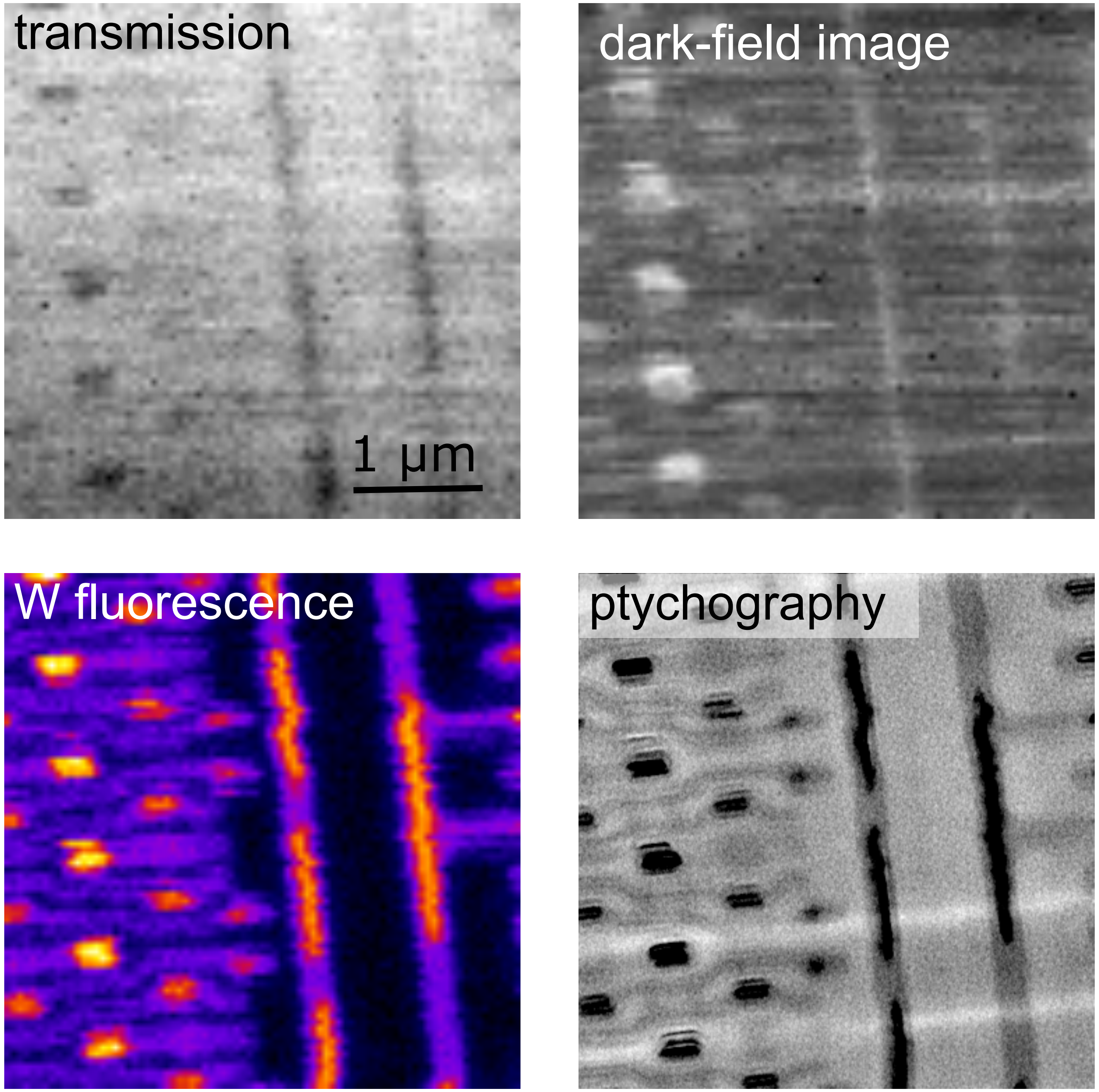
Imaging Electronic Nanostructures in a Microchip

E = 15.25 keV
focus: 81 x 84 nm² (FWHM)
exposure time: 0.1 s
fluence: 800 ph/nm²

scanning parameters:

- 4 x 4 µm² (FOV)
- 80 x 80 steps
- step size: 50 nm

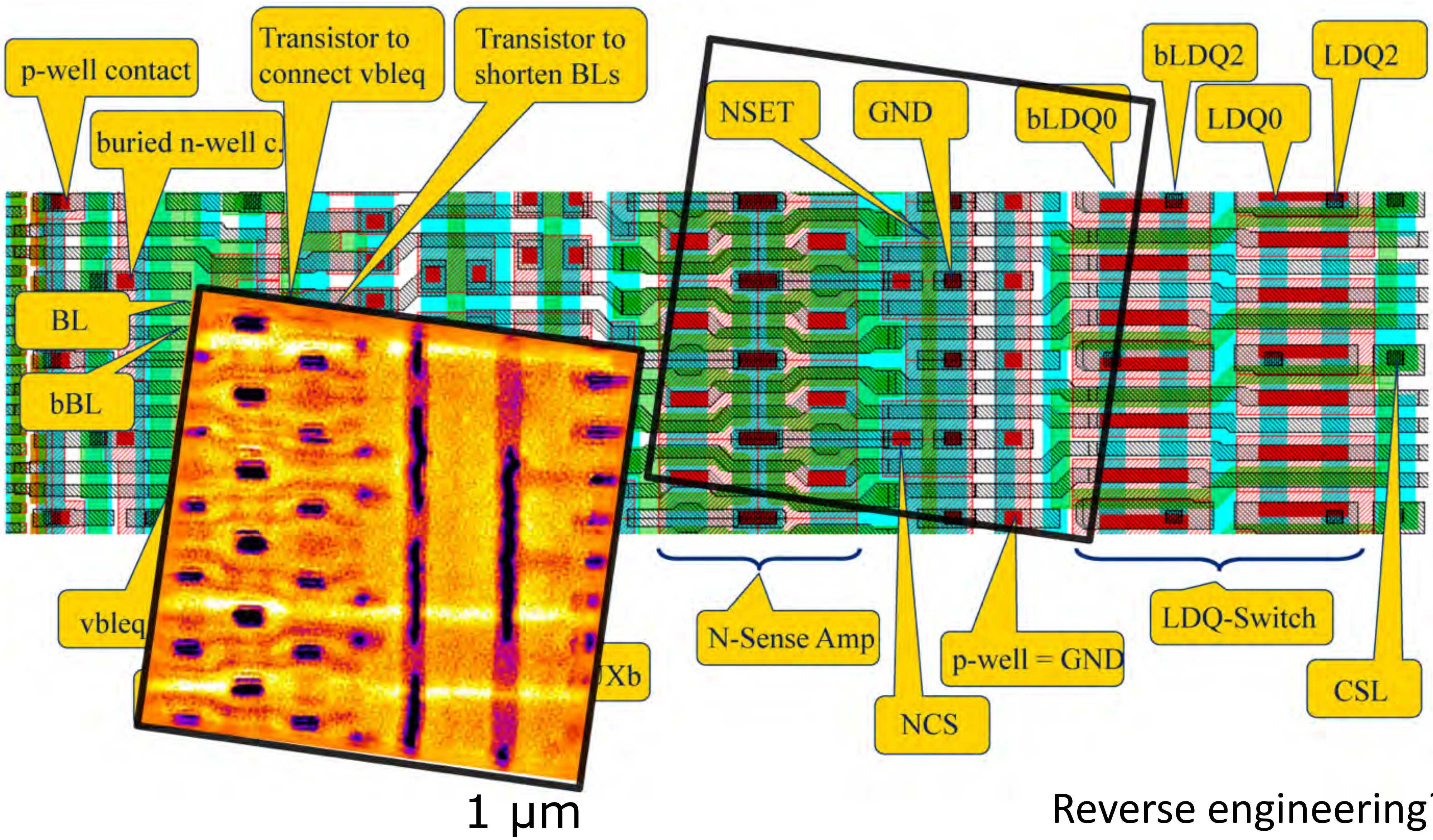
enhanced contrast



A. Schropp, et al., Journal of Microscopy **241**, 9 (2011).

Imaging Electronic Nanostructures in a Microchip

Shared sense amplifier (80 nm)

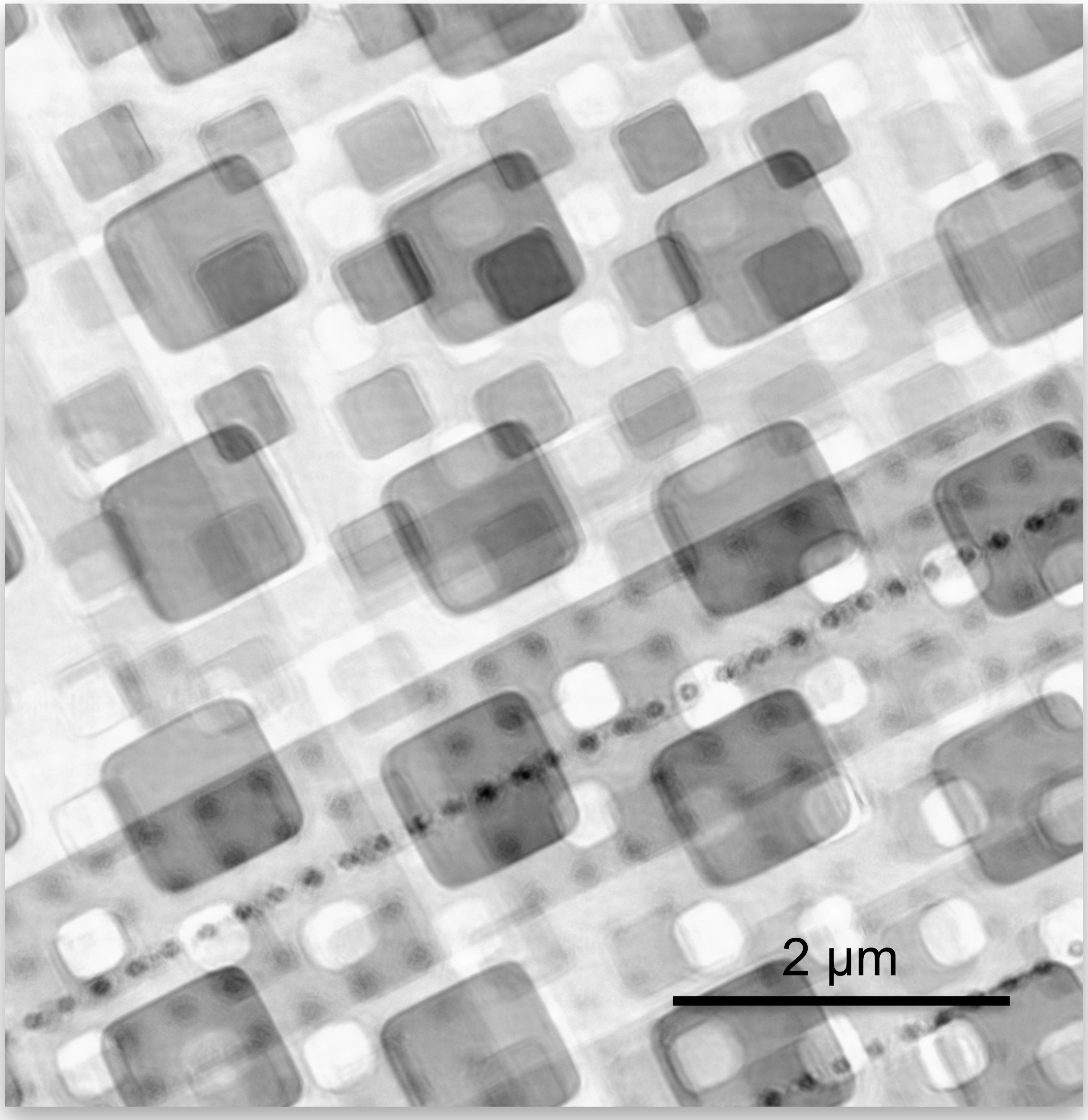


Reverse engineering?!

A. Schropp, et al., Journal of Microscopy **241**, 9 (2011).

Imaging Electronic Nanostructures in a Microchip

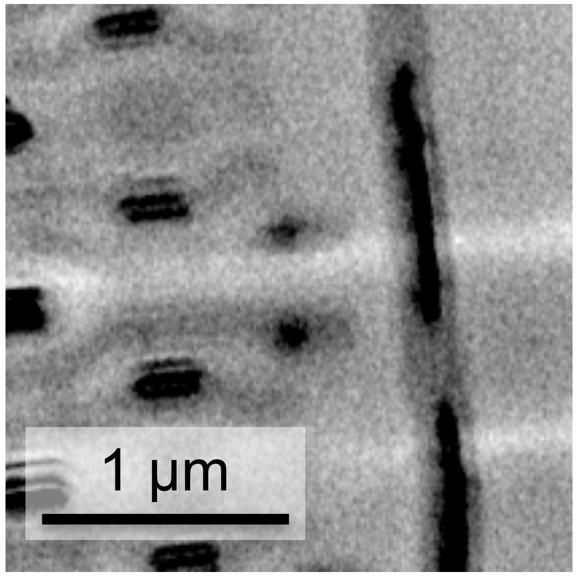
Currently at PETRA III:
imaging nanostructures in test chip



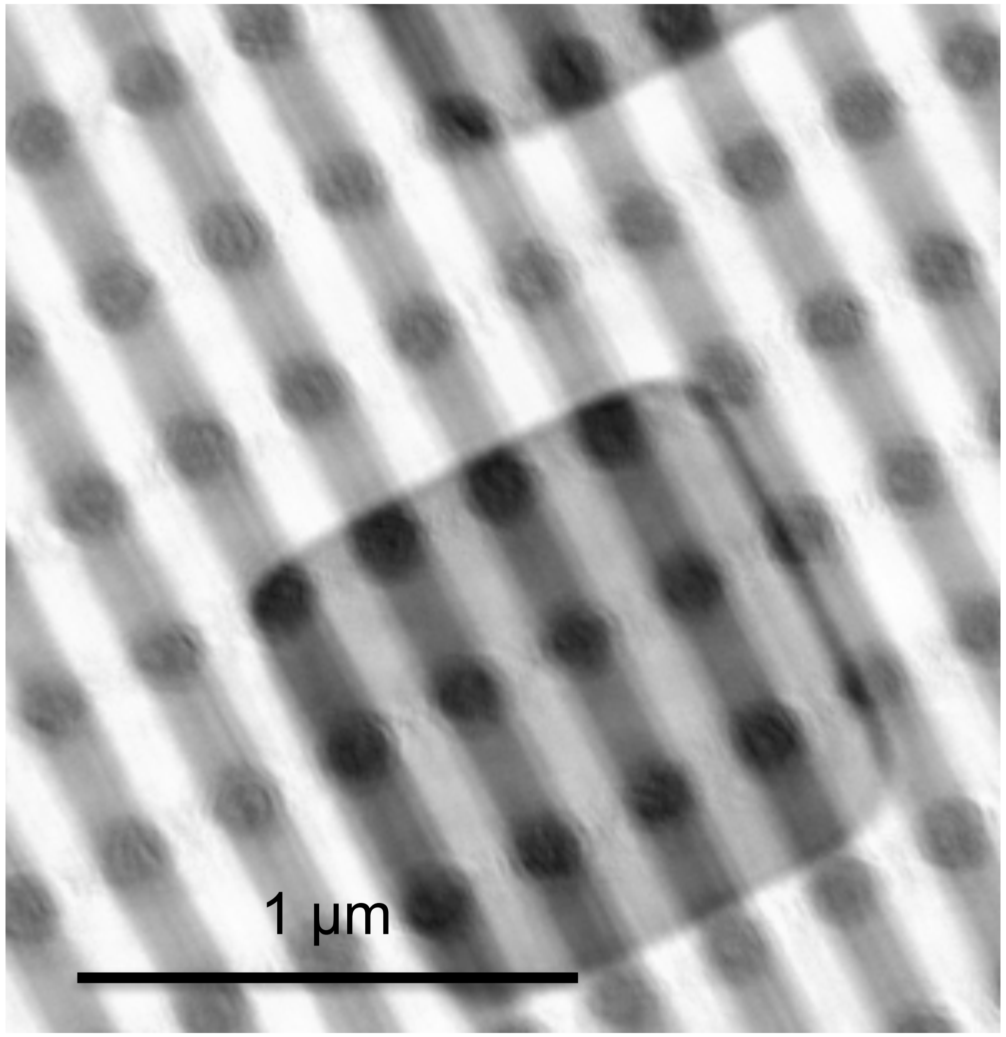
no sample preparation

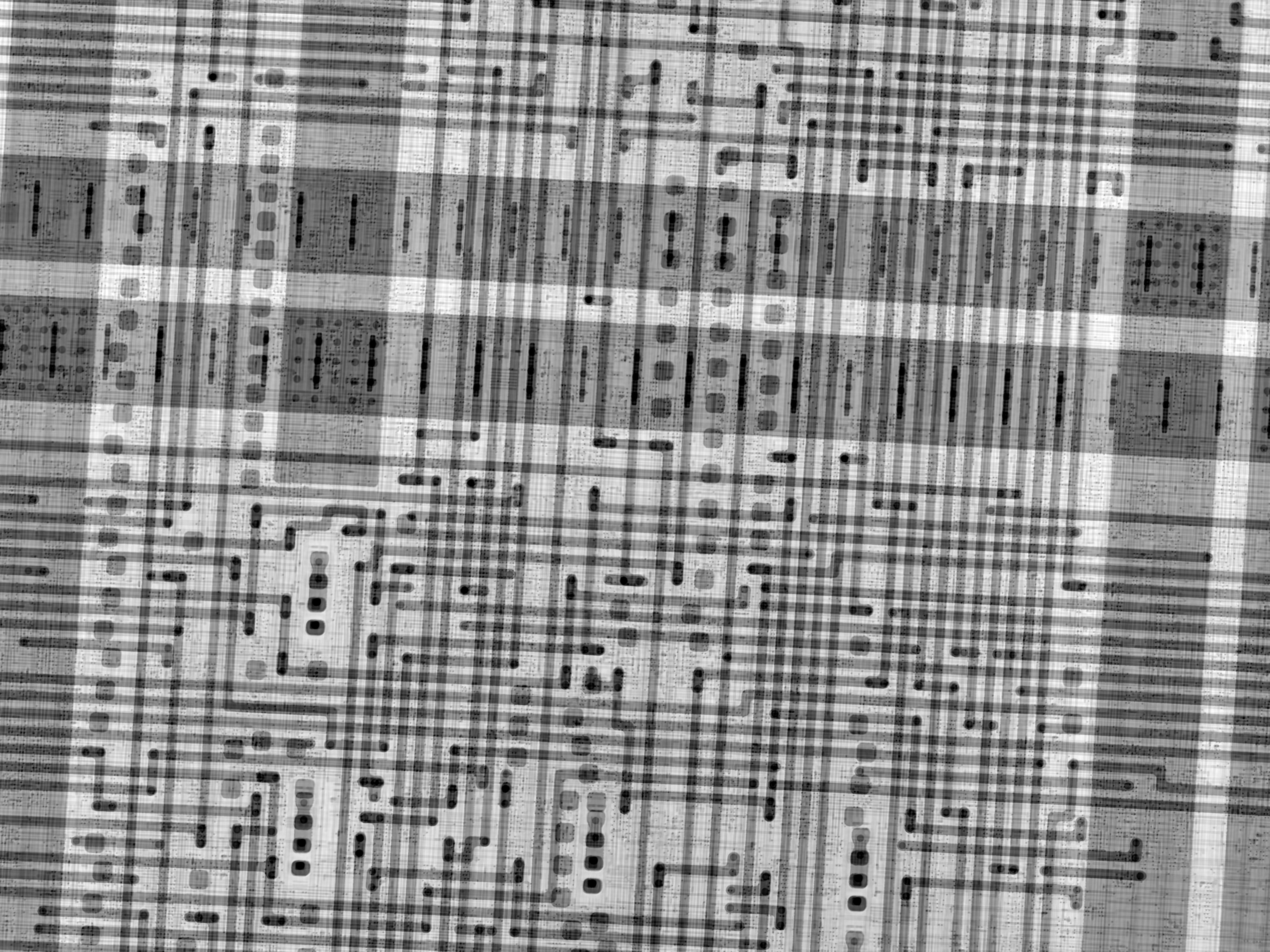
collaboration with Infineon (Dresden)

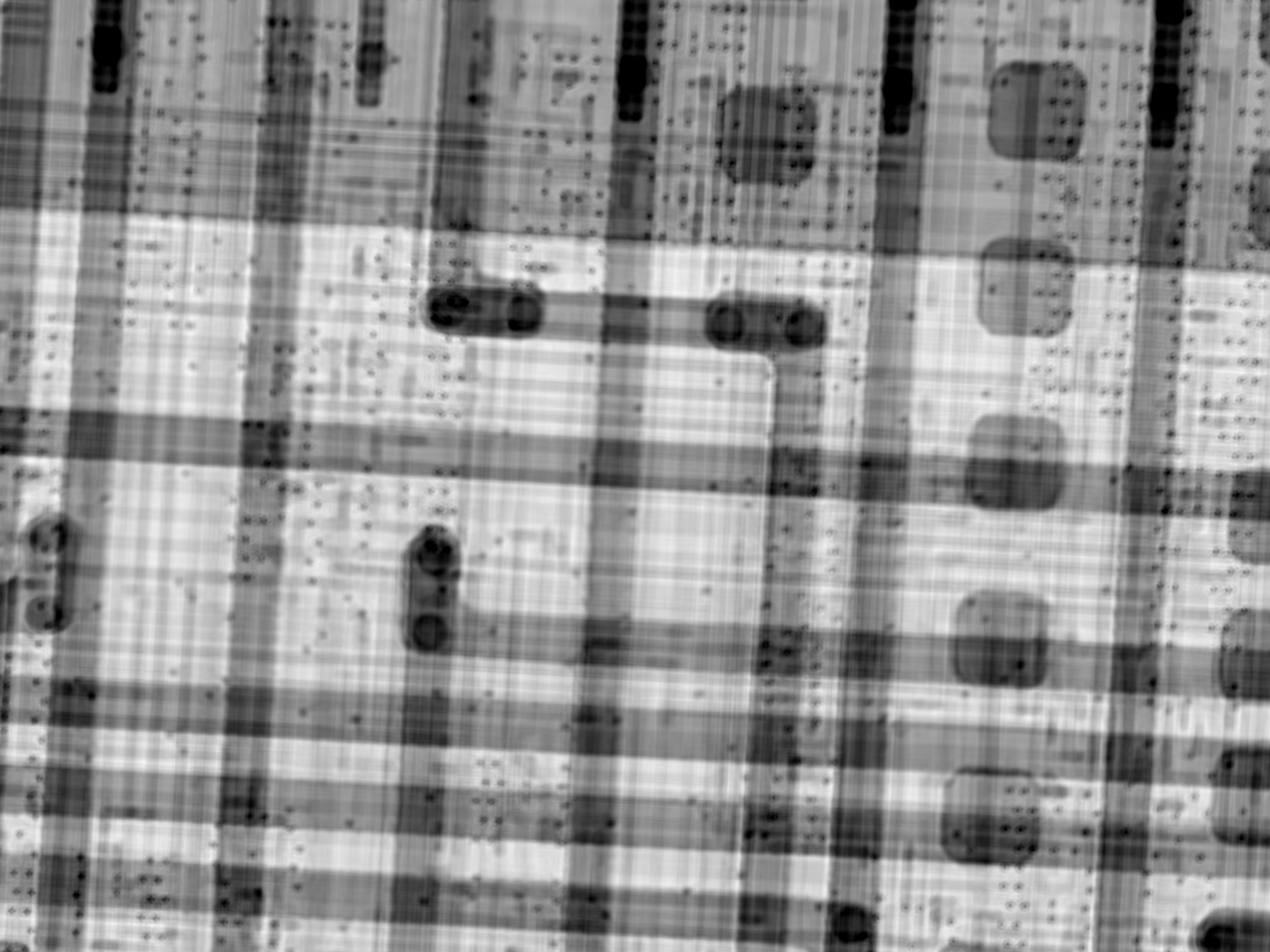
for comparison:



A. Schropp, et al.,
J. Micro. **241**, 9 (2011).

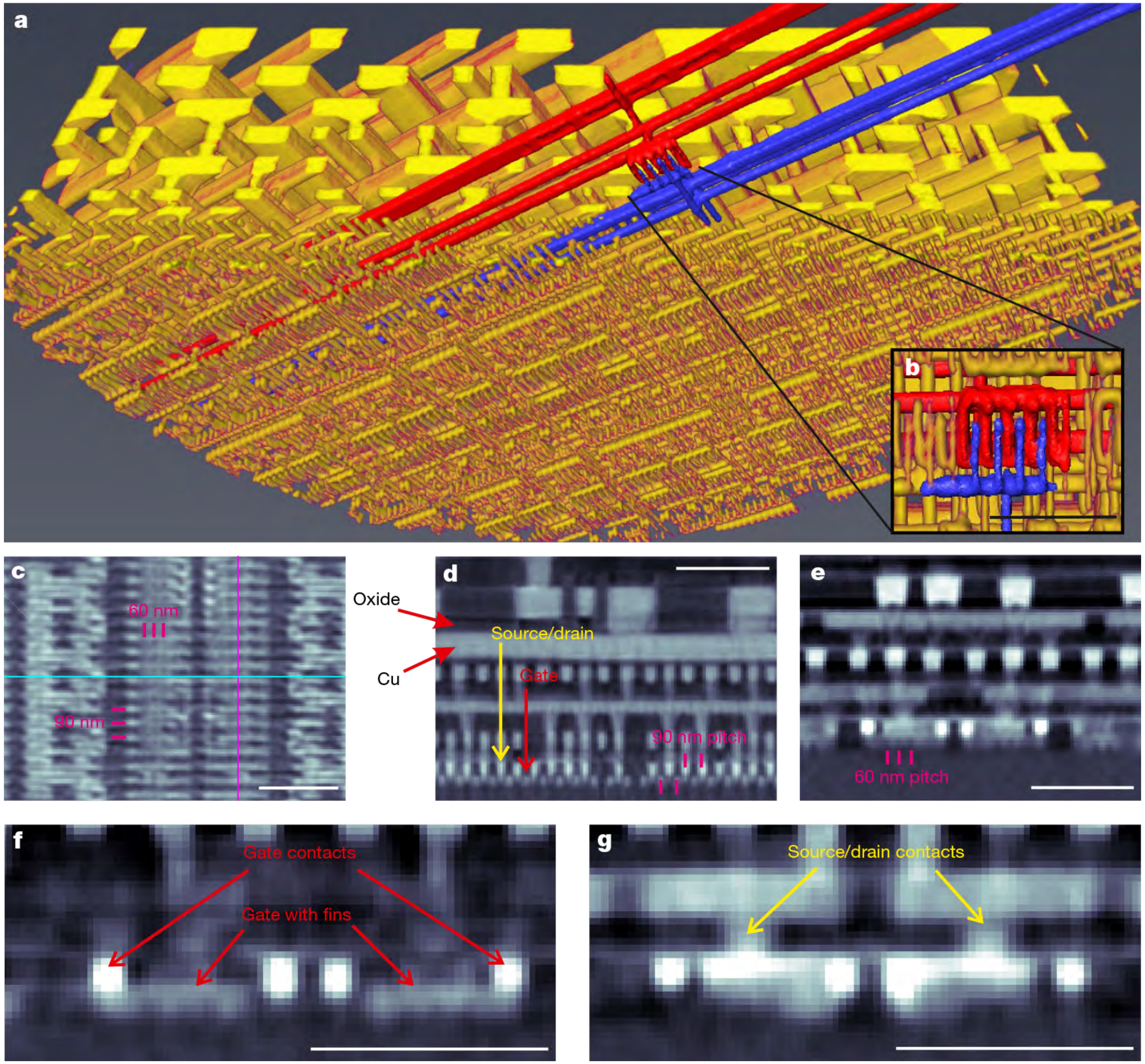






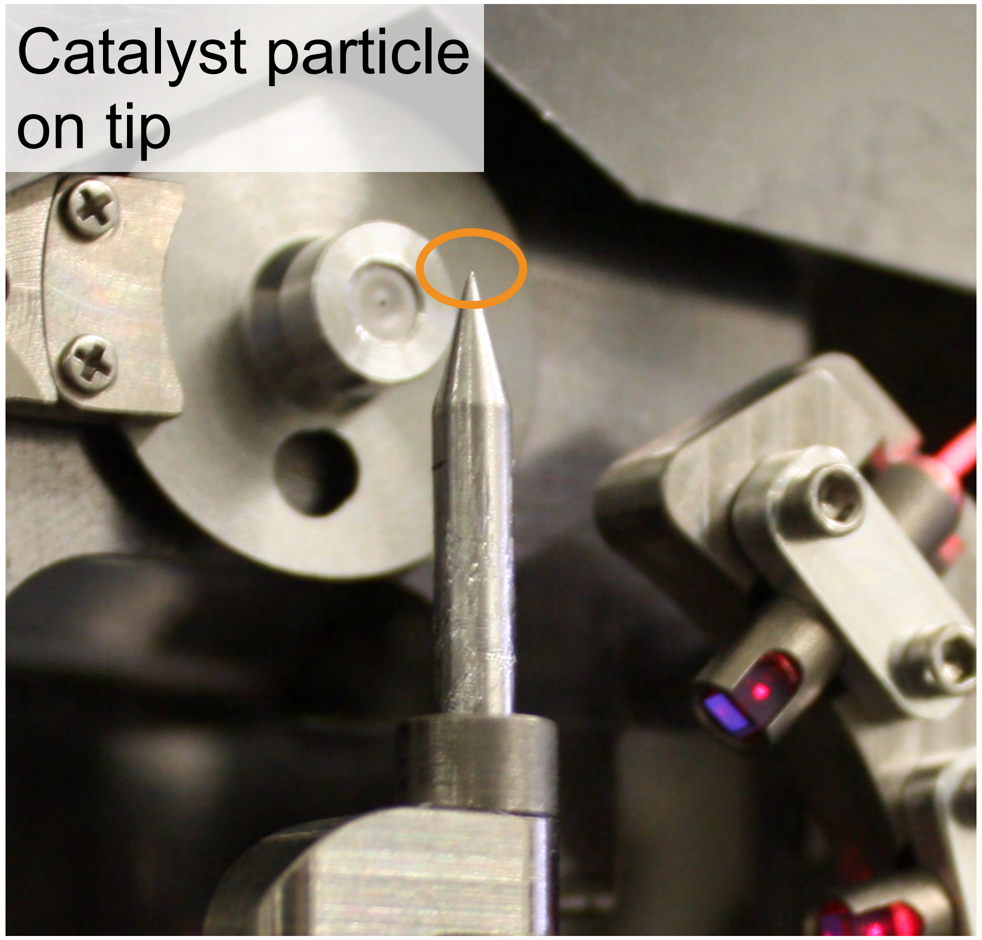
Ptychography Combined with Tomography

- **ptychography:**
  record a series of projections with high spatial resolution
- **tomography:**
  combine the projections to a three



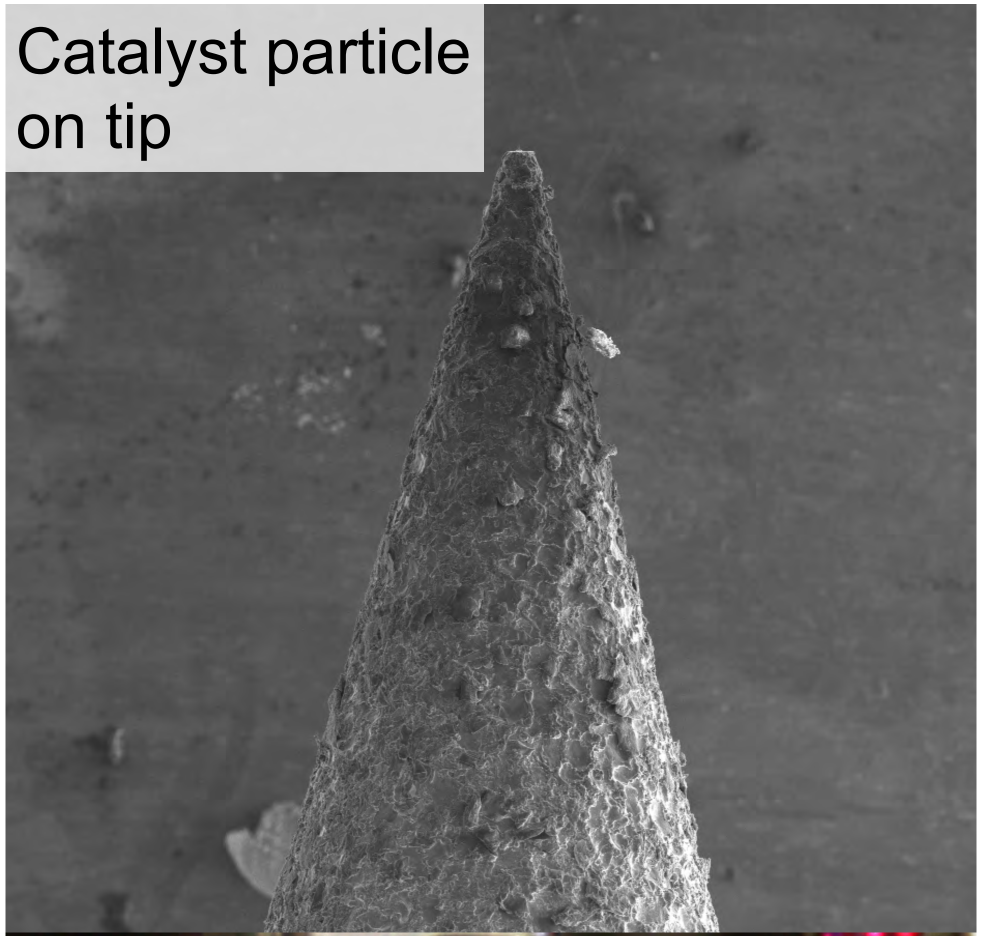
M. Holler, et al., *High-Resolution Non-Destructive Three-Dimensional Imaging of Integrated Circuits*, Nature **543**, 402 (2017).

Ptycho-Tomography of Catalysts



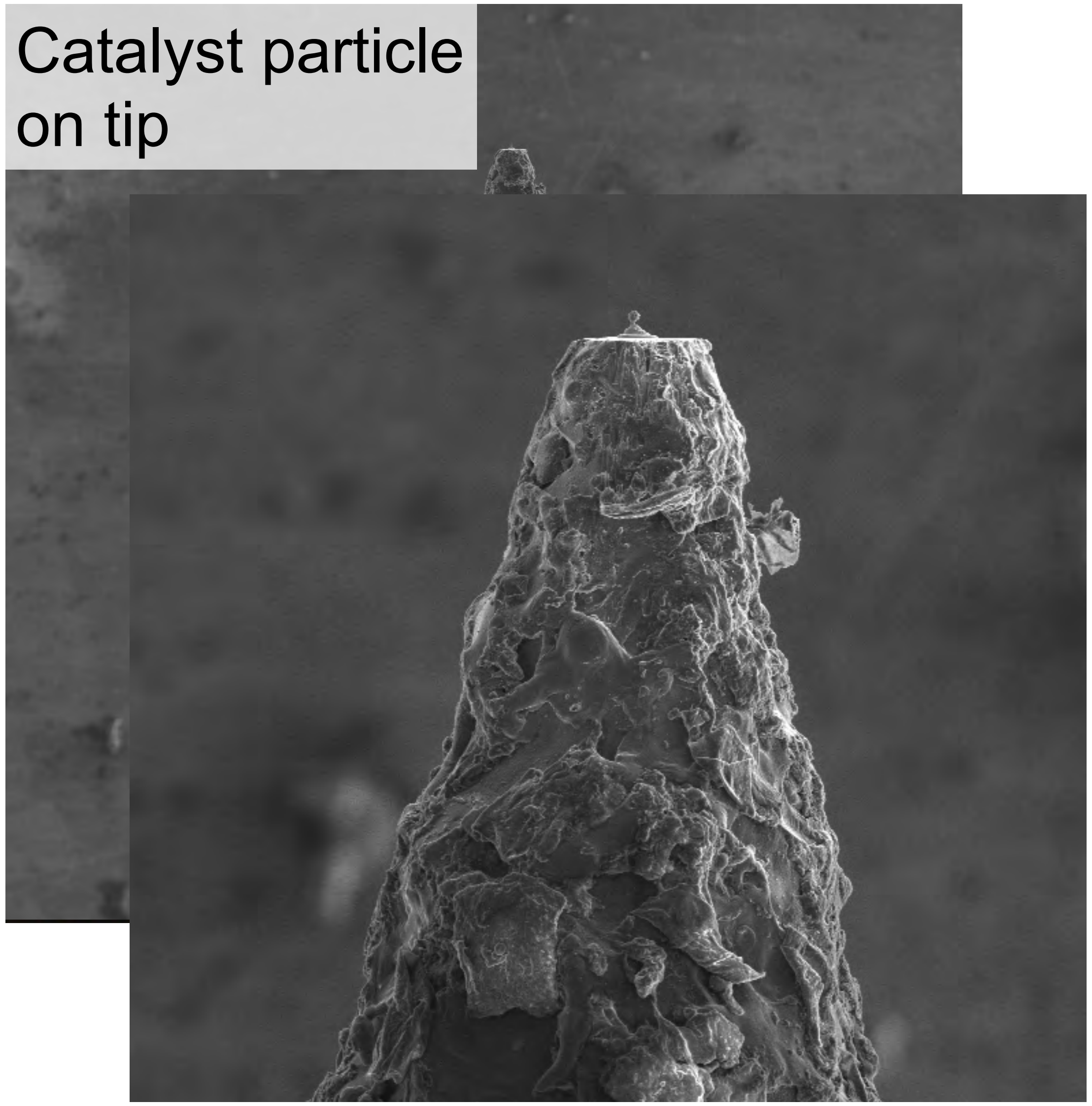
M. Kahnt, et al., *Coupled Ptychography and Tomography Algorithm Improves Reconstruction of Experimental Data*, Optica **6**, 1282 (2019).

Ptycho-Tomography of Catalysts



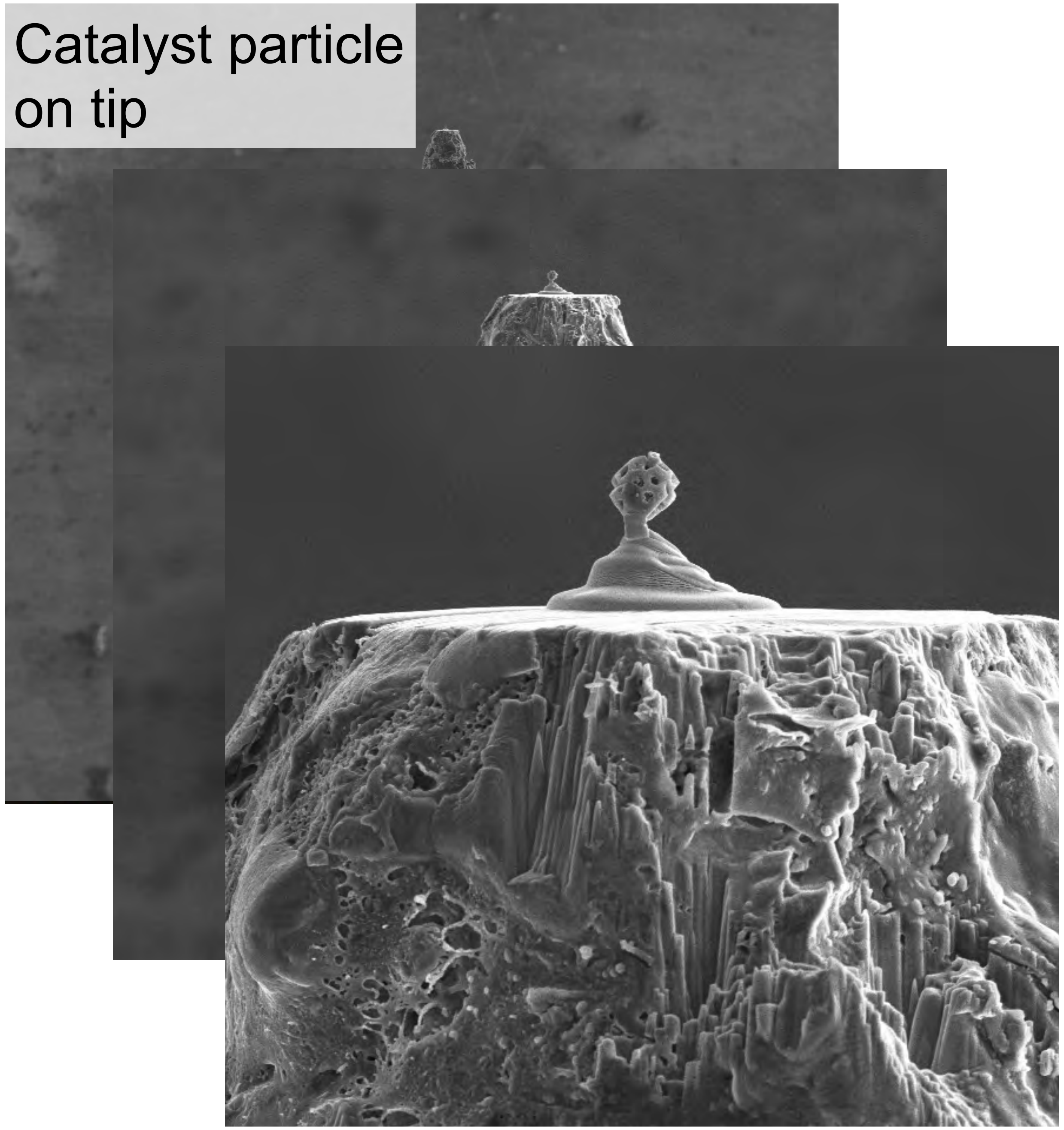
M. Kahnt, et al., *Coupled Ptychography and Tomography Algorithm Improves Reconstruction of Experimental Data*, Optica **6**, 1282 (2019).

Ptycho-Tomography of Catalysts



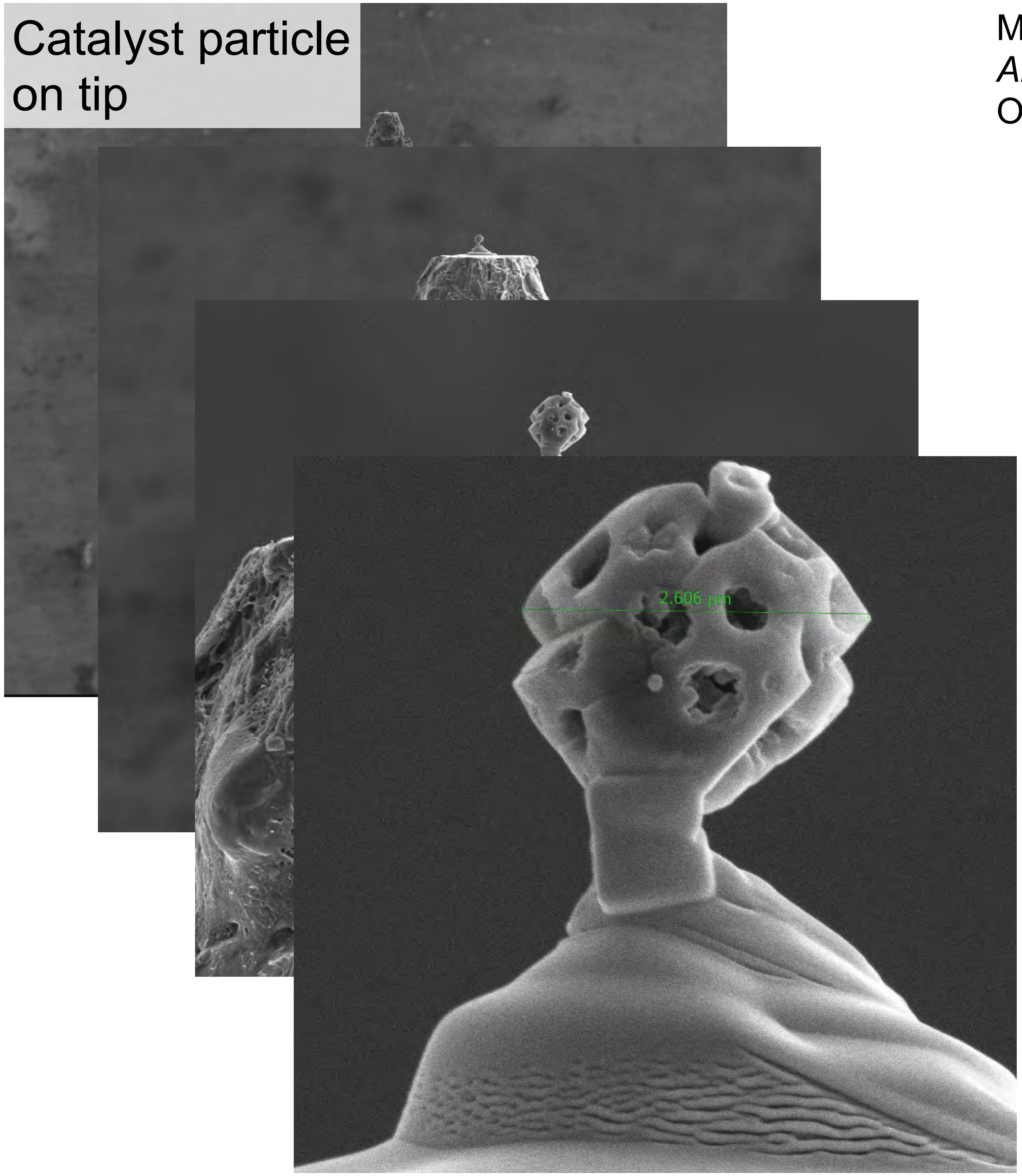
M. Kahnt, et al., *Coupled Ptychography and Tomography Algorithm Improves Reconstruction of Experimental Data*, Optica **6**, 1282 (2019).

Ptycho-Tomography of Catalysts



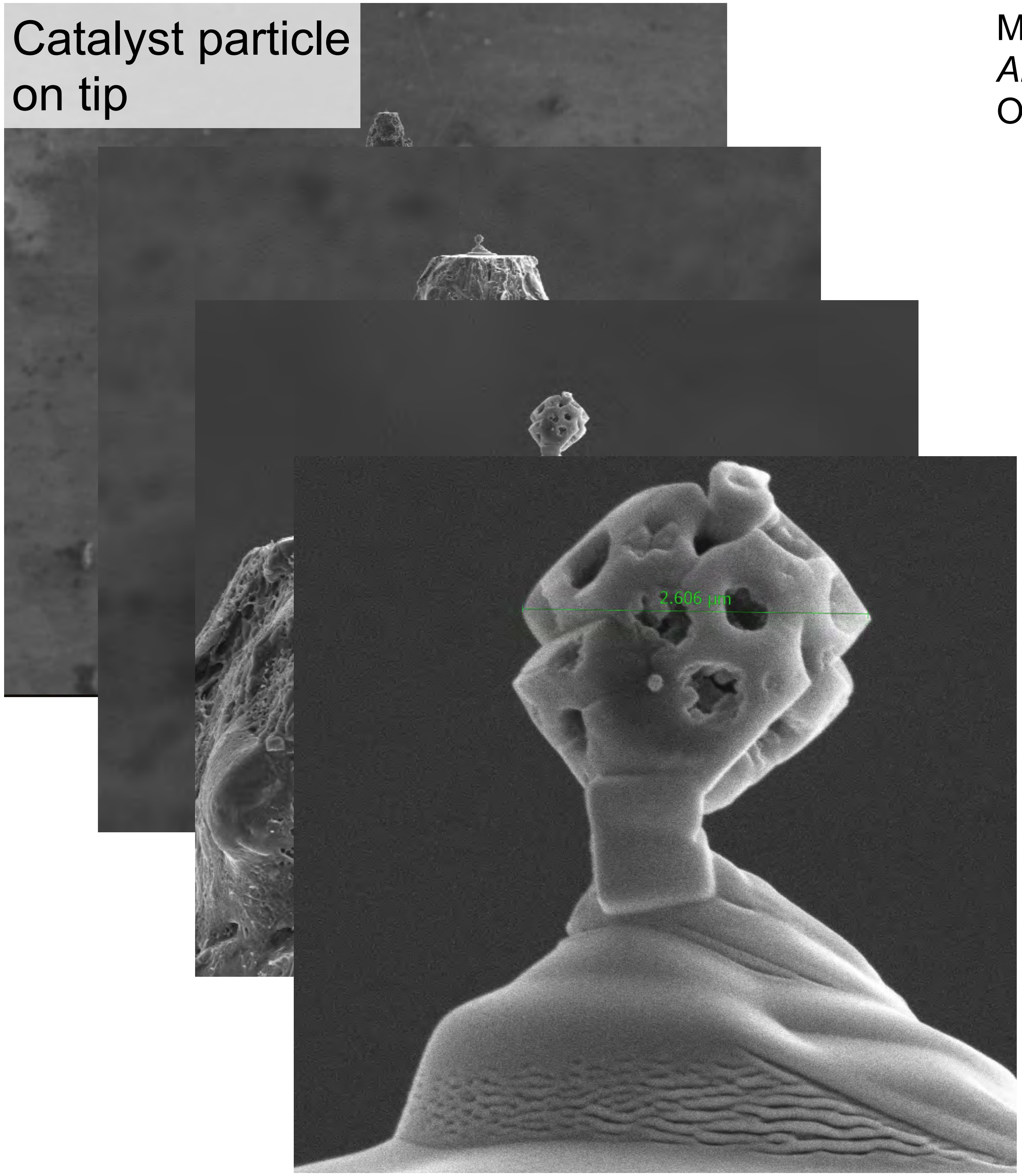
M. Kahnt, et al., *Coupled Ptychography and Tomography Algorithm Improves Reconstruction of Experimental Data*, Optica **6**, 1282 (2019).

Ptycho-Tomography of Catalysts

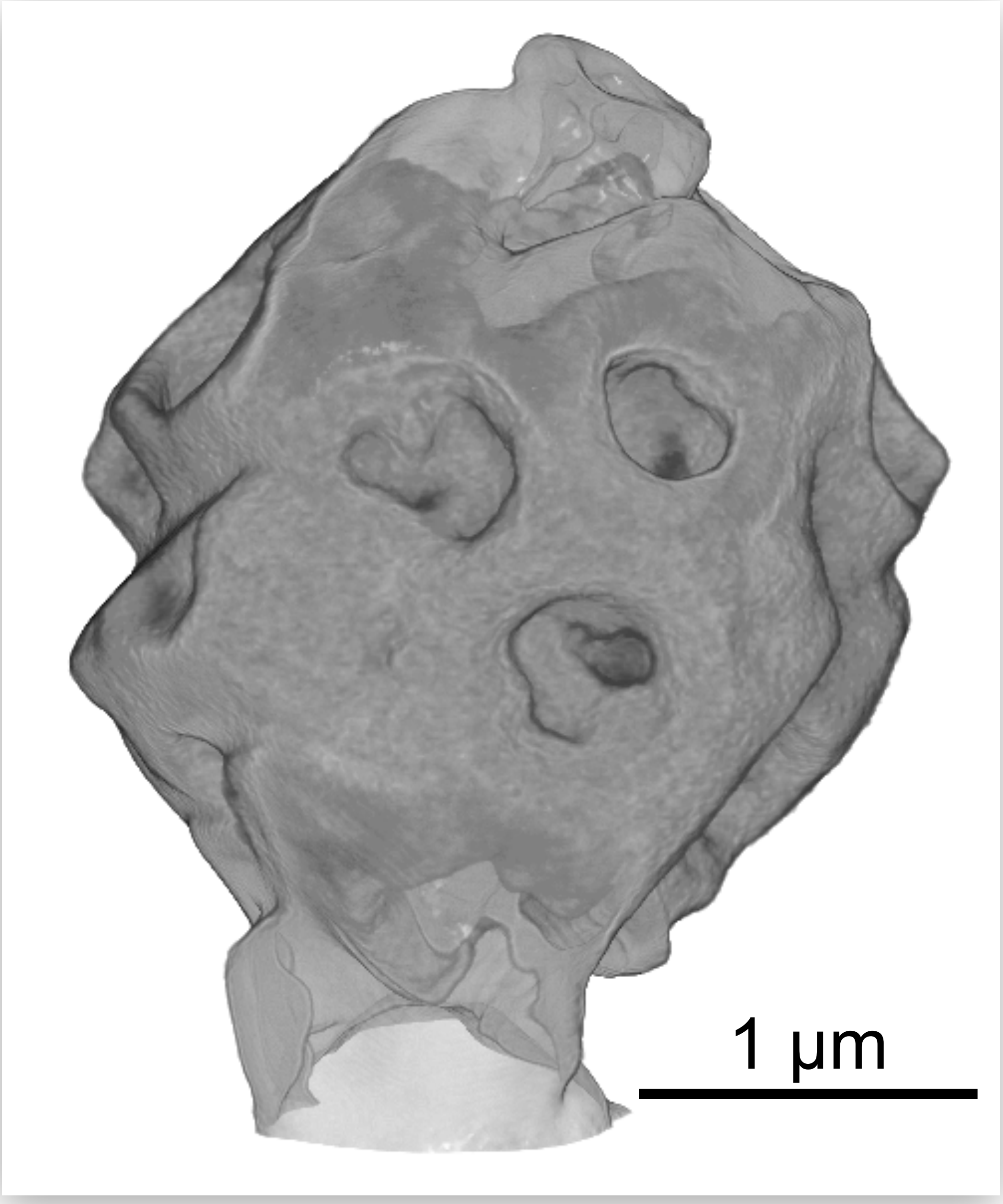


M. Kahnt, et al., *Coupled Ptychography and Tomography Algorithm Improves Reconstruction of Experimental Data*, Optica **6**, 1282 (2019).

Ptycho-Tomography of Catalysts



M. Kahnt, et al., *Coupled Ptychography and Tomography Algorithm Improves Reconstruction of Experimental Data*, Optica **6**, 1282 (2019).



Time-Resolved Imaging at X-Ray Free-Electron Lasers

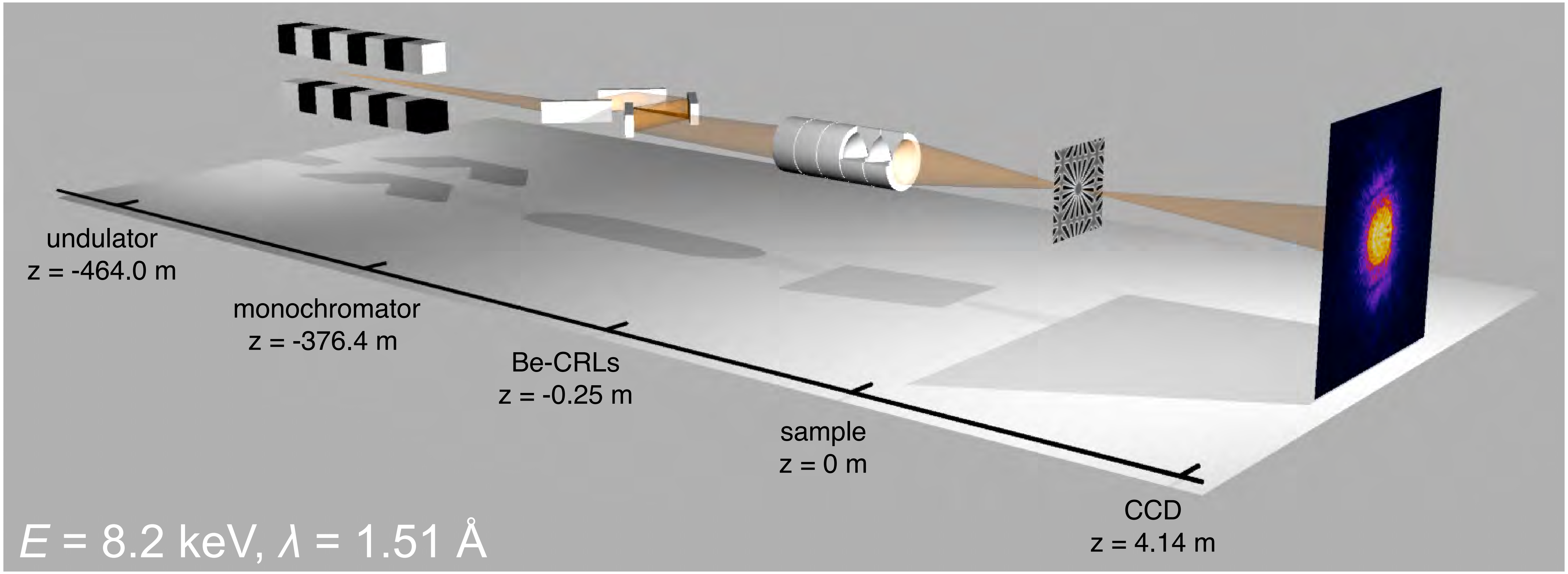
Microscopy at the XFEL

single-pulse imaging of
fast processes in matter

pump-probe experiments:

- excite process in matter
- probe by single-pulse microscopy

Nanofocusing and Nanoimaging at LCLS



> Nanofocusing by Be CRLs

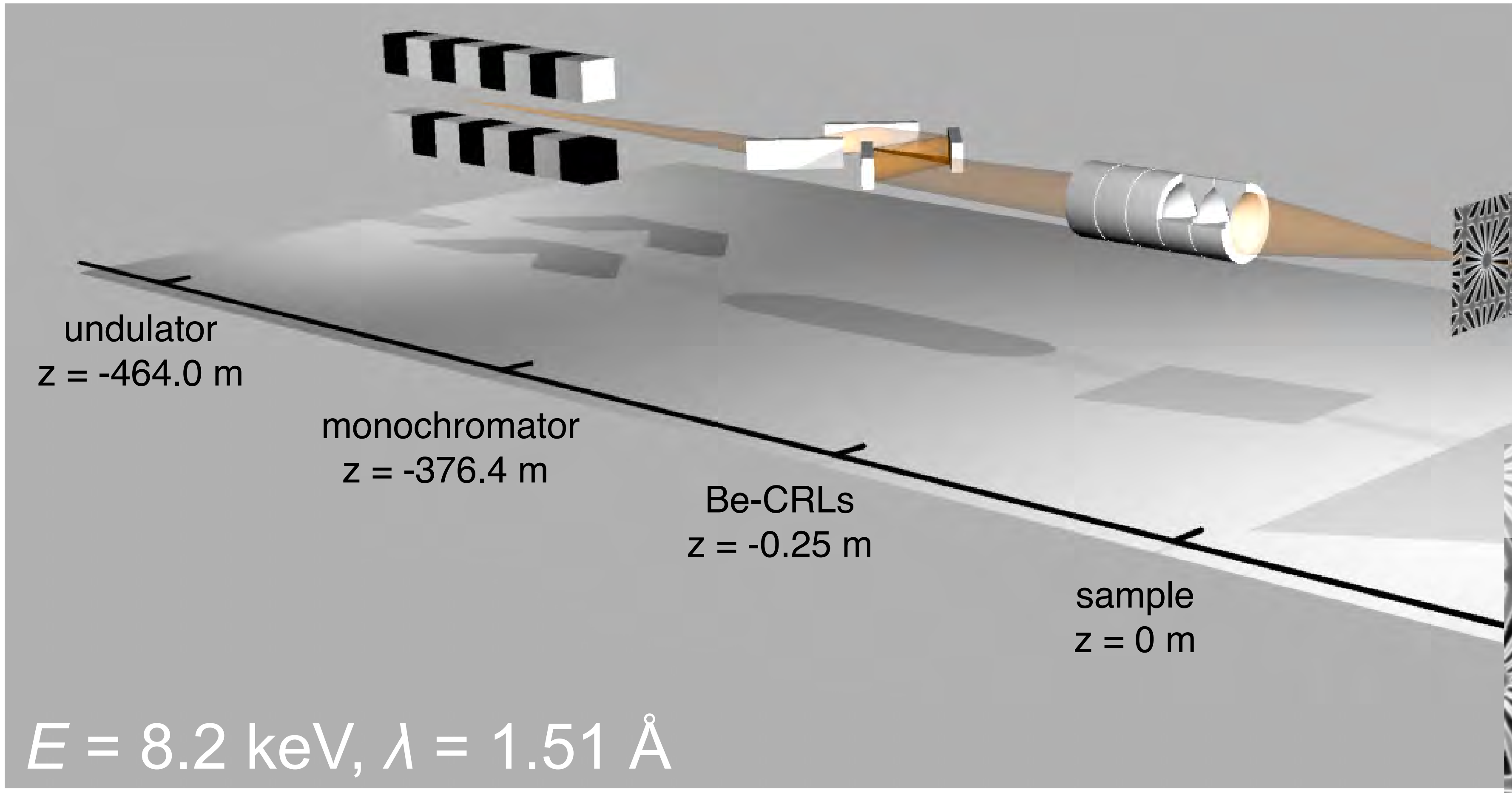
⟶ generate nanobeam for near-field microscopy

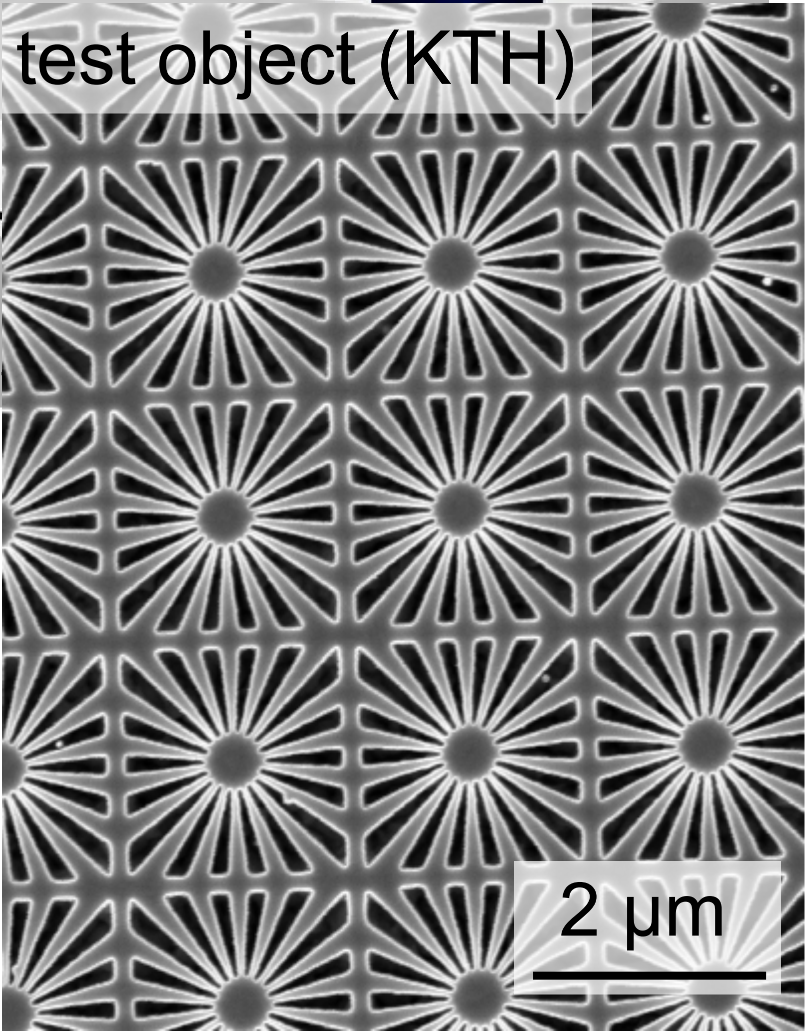
> Characterization by ptychography

⟶ determine full caustic and aberrations of optic

A. Schropp, et al., Sci. Rep. **3**, 1633 (2013).

Nanofocusing and Nanoimaging at LCLS

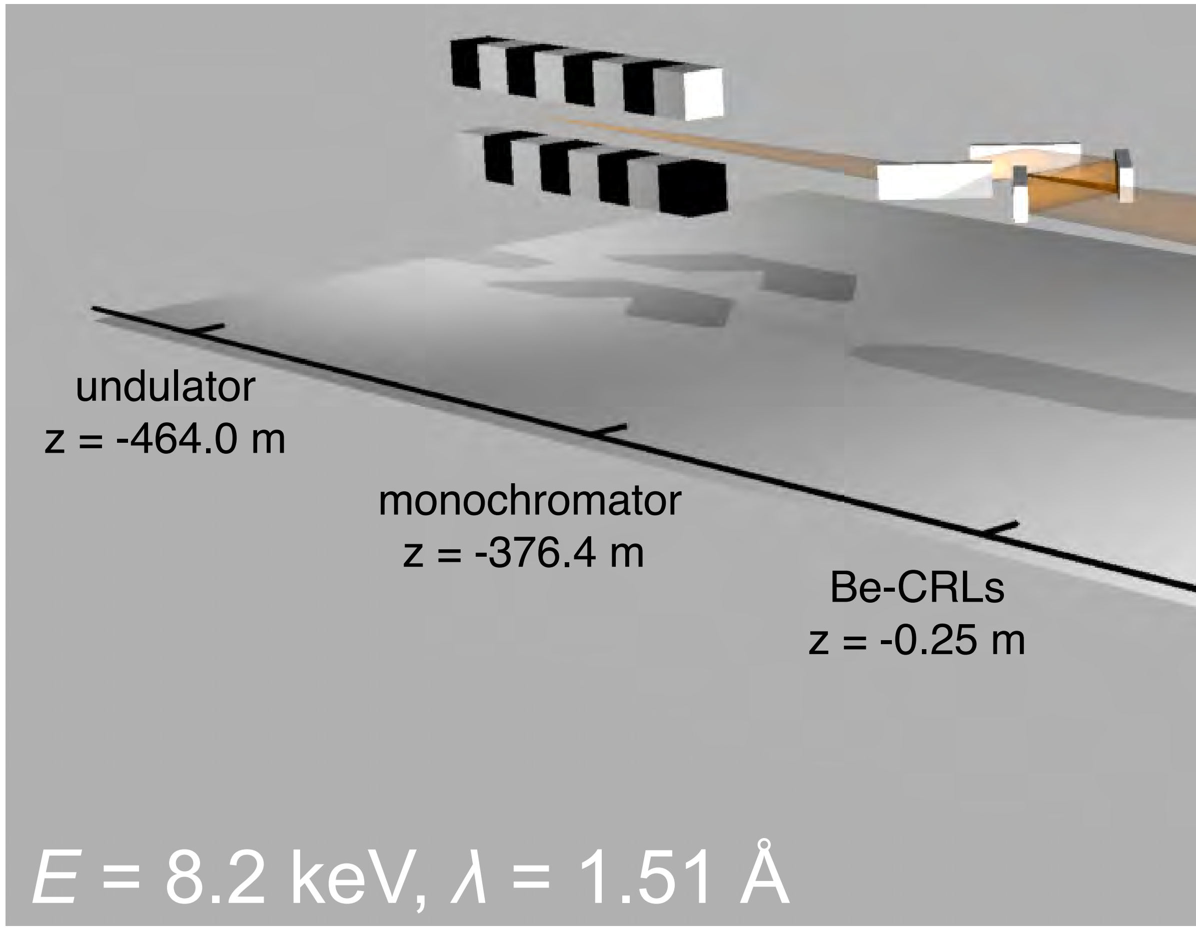


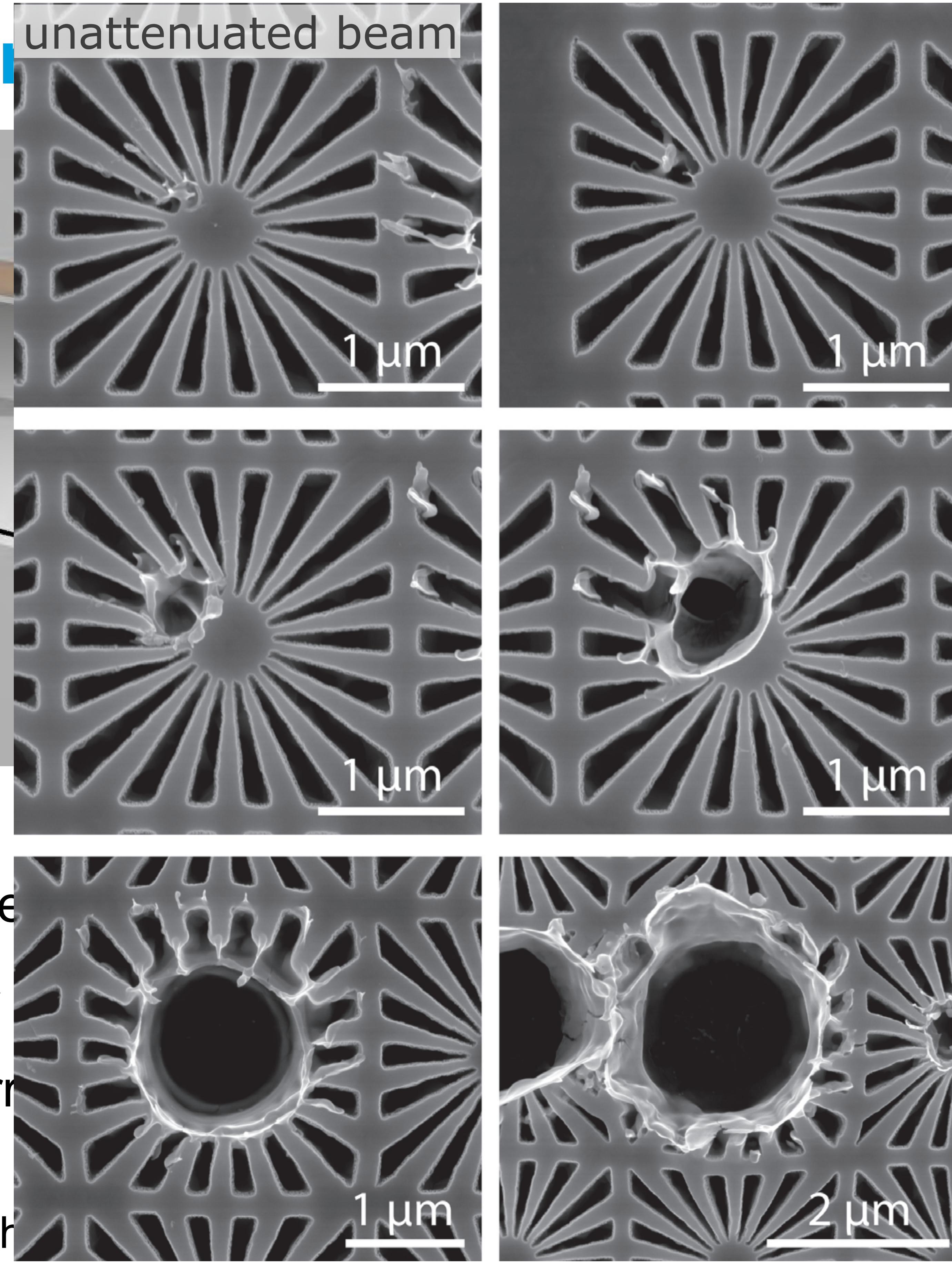


- > Nanofocusing by Be CRLs
  - ⟶ generate nanobeam for near-field microscopy
- > Characterization by ptychography
  - ⟶ determine full caustic and aberrations of optic

A. Schropp, et al., Sci. Rep. **3**, 1633 (2013).

Nanofocusing and Na





> Nanofocusing by Be CRLs

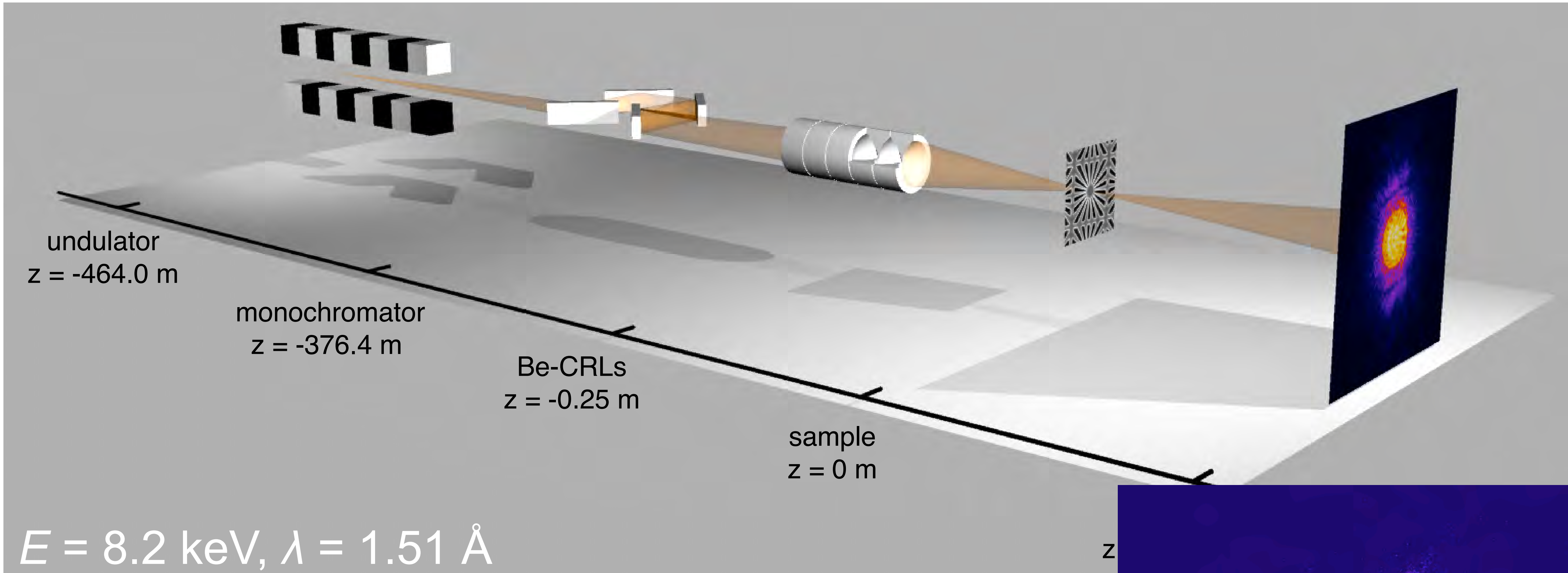
→ generate nanobeam for near-fie

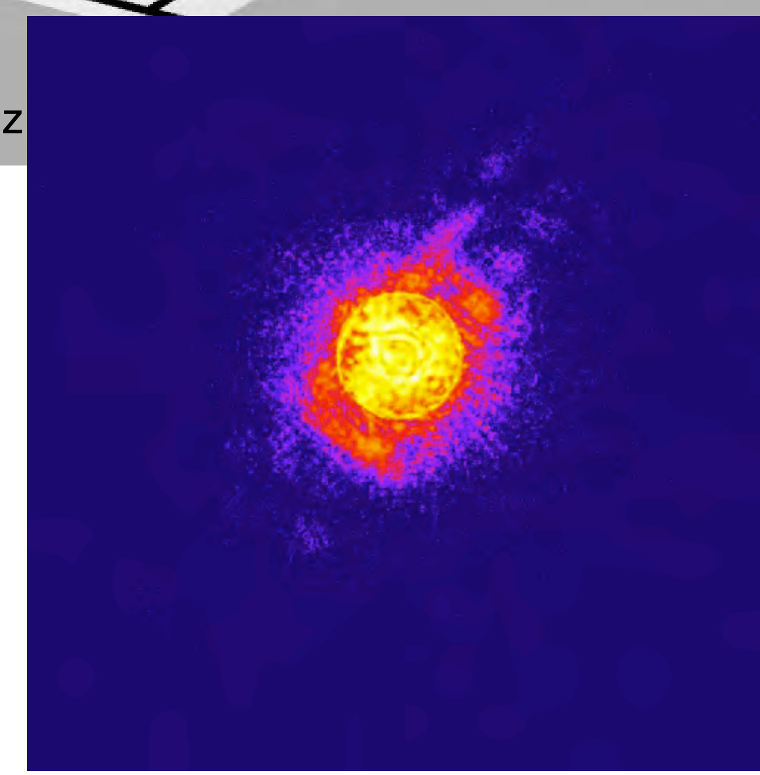
> Characterization by ptychography

→ determine full caustic and aberr

A. Sch

Nanofocusing and Nanoimaging at LCLS

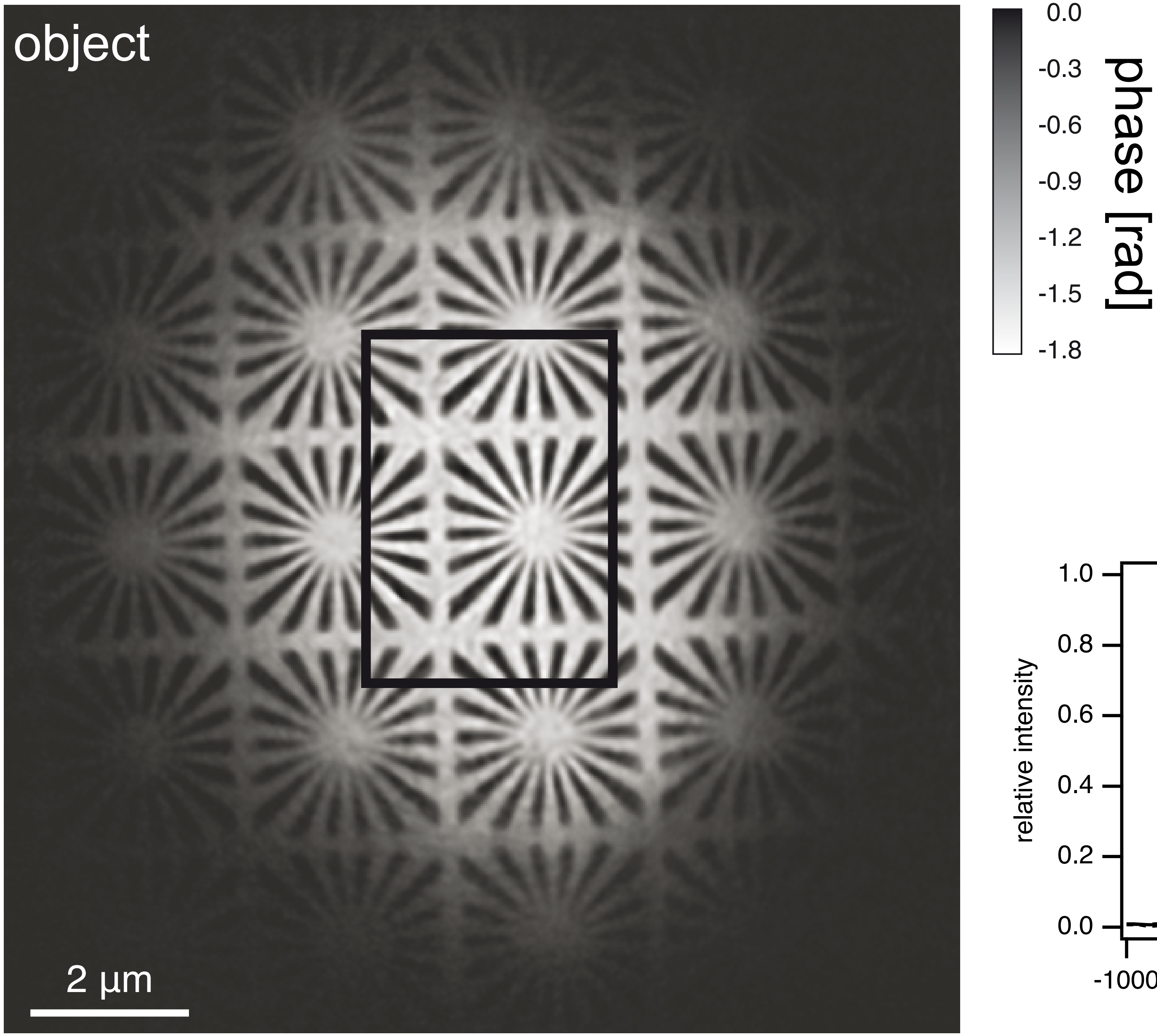


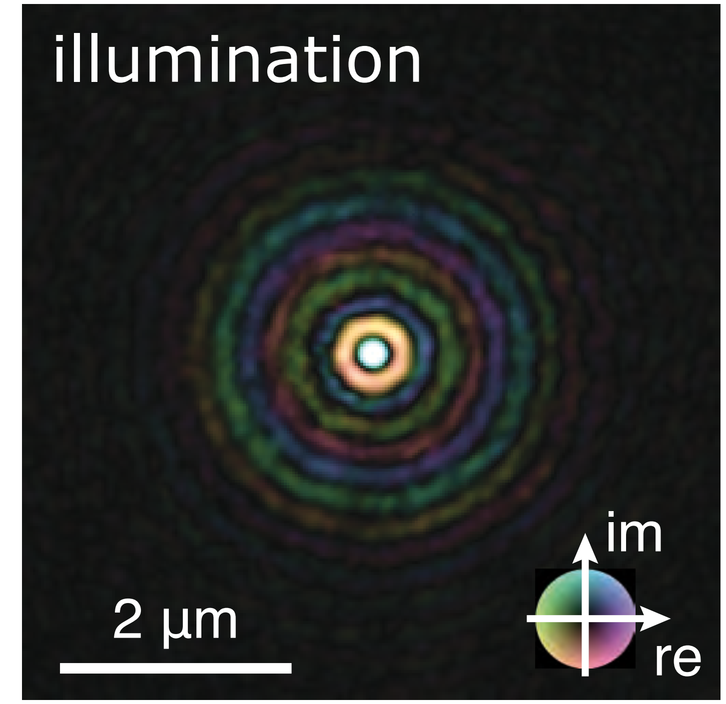


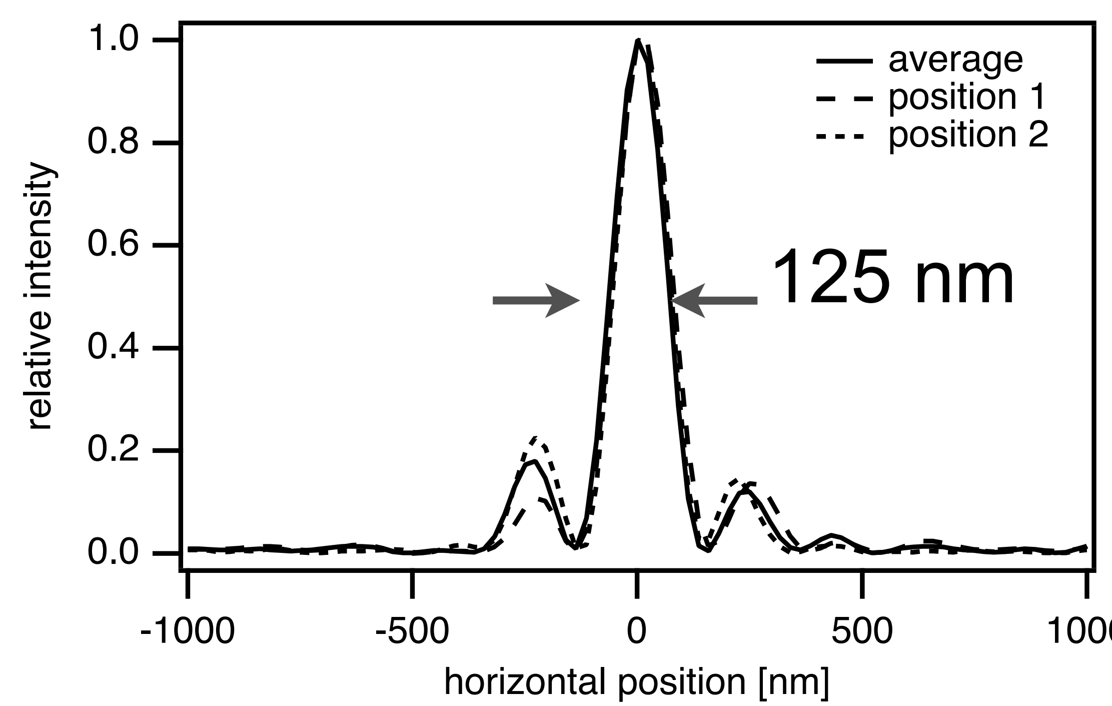
- > Nanofocusing by Be CRLs
  - → generate nanobeam for near-field microscopy
- > Characterization by ptychography
  - → determine full caustic and aberrations of optic

A. Schropp, et al., Sci. Rep. **3**, 1633 (2013).

Ptychographic Reconstruction

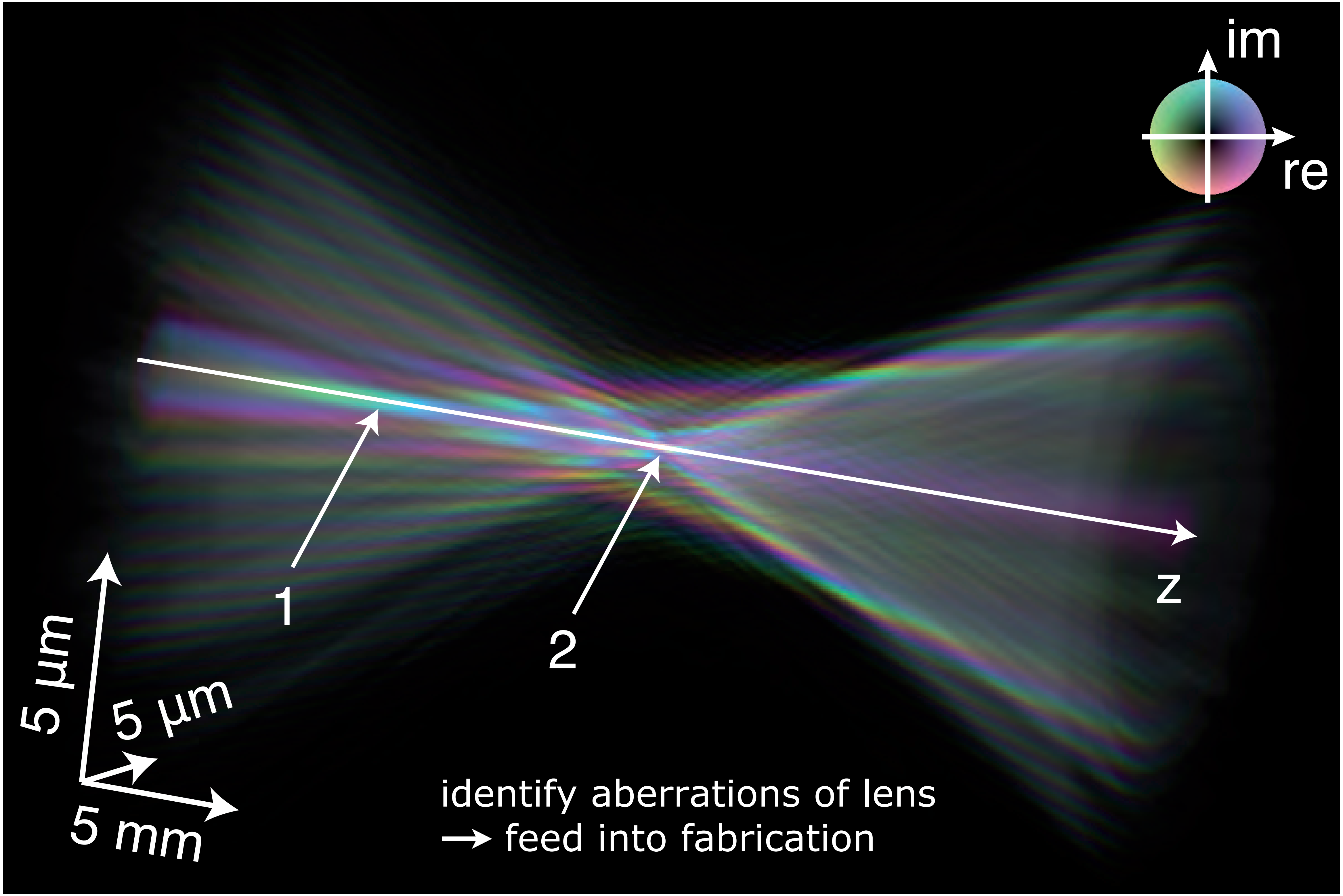






A. Schropp, et al., Sci. Rep. **3**, 1633 (2013).

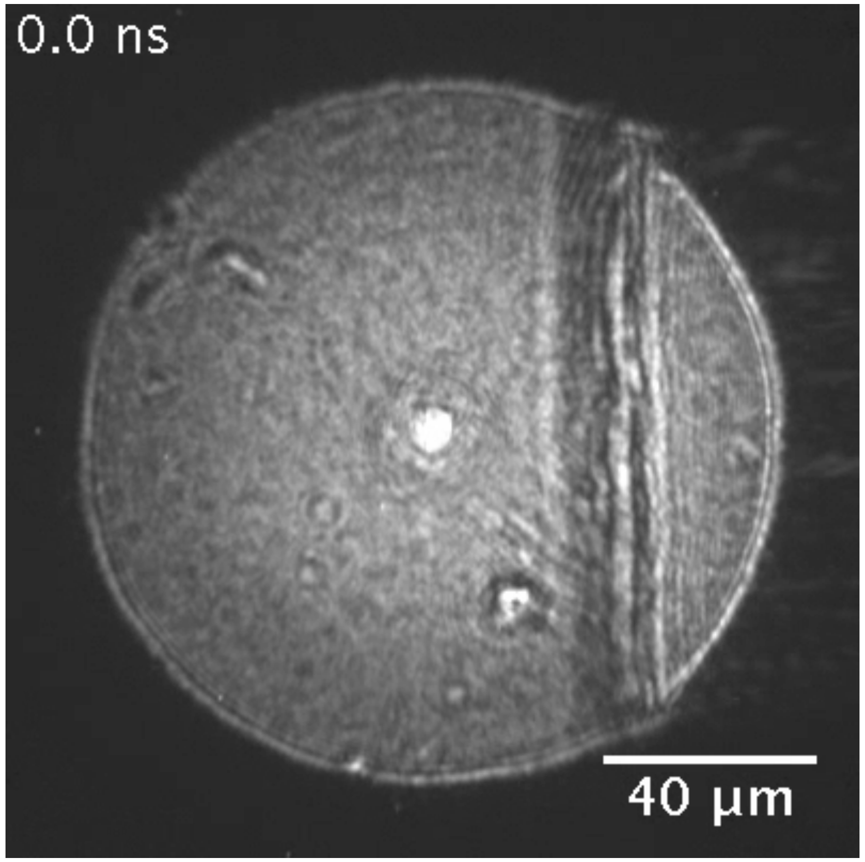
Nanofocused LCLS Beam Profile

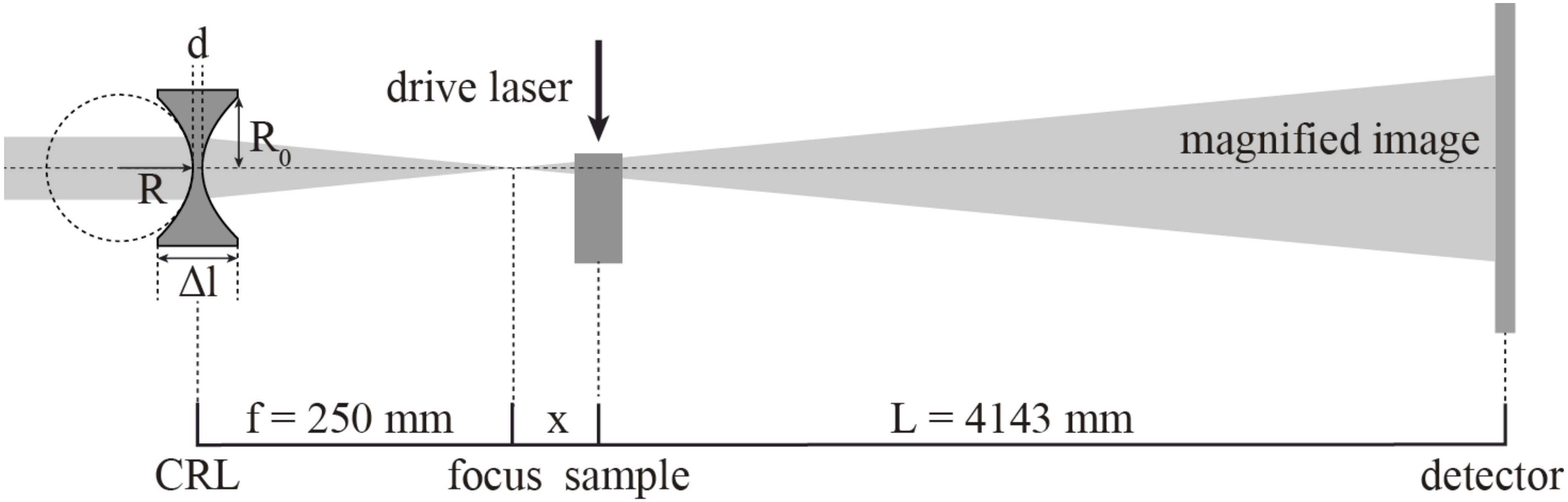


A. Schropp, et al., Sci. Rep. **3**, 1633 (2013).

Elastic Wave in Diamond

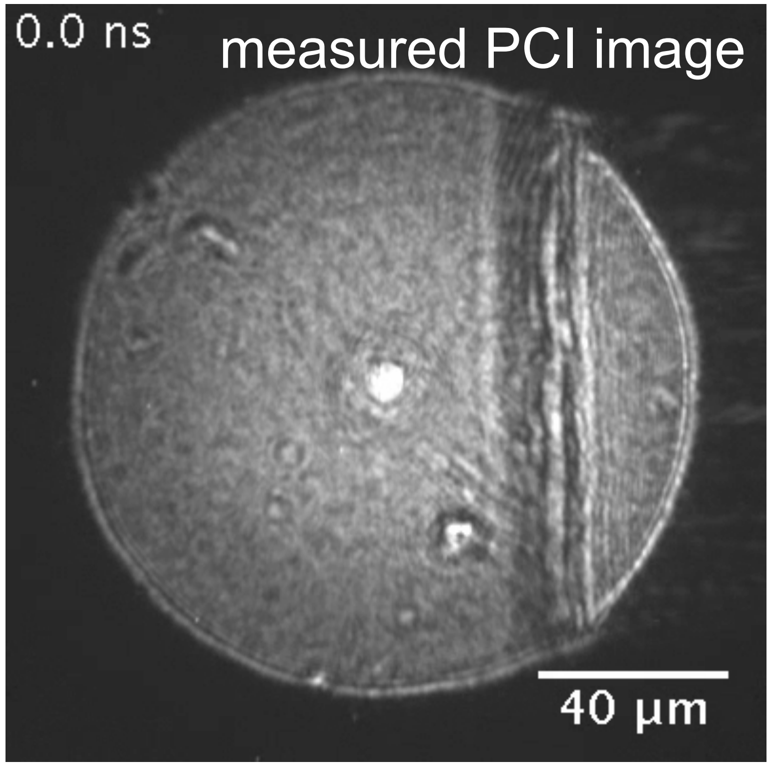
> pump: 150 ps drive laser, 800 nm, 130 mJ
> probe: XFEL beam: E = 8.2 keV, 50 fs
> single-pulse imaging (stop-trick movie)
> pump-probe time delay between 0 ns and 3 ns in steps of 0.2 ns
> high spatial resolution in the phase contrast image of about 300 - 600 nm
> phase retrieval required

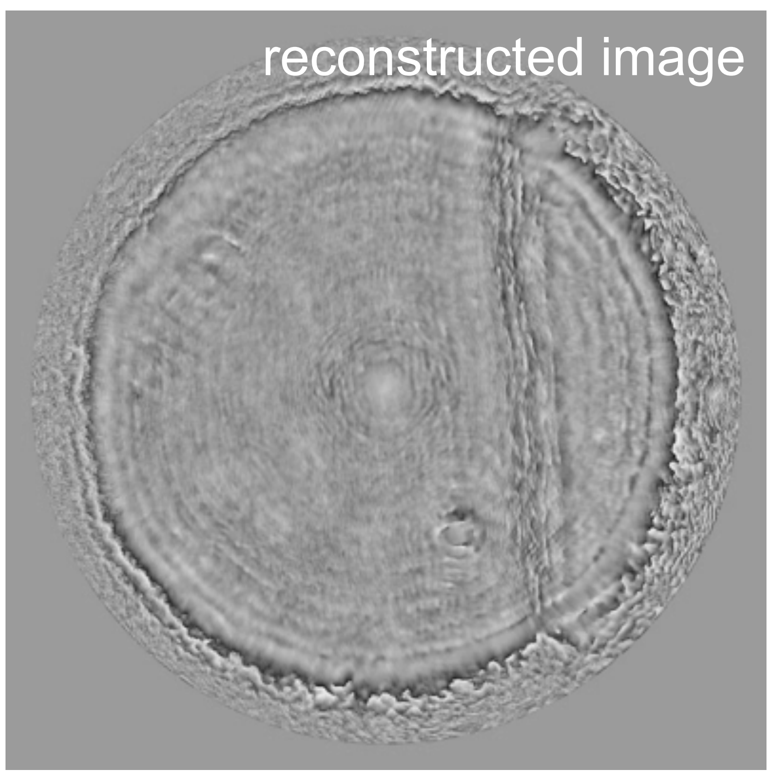


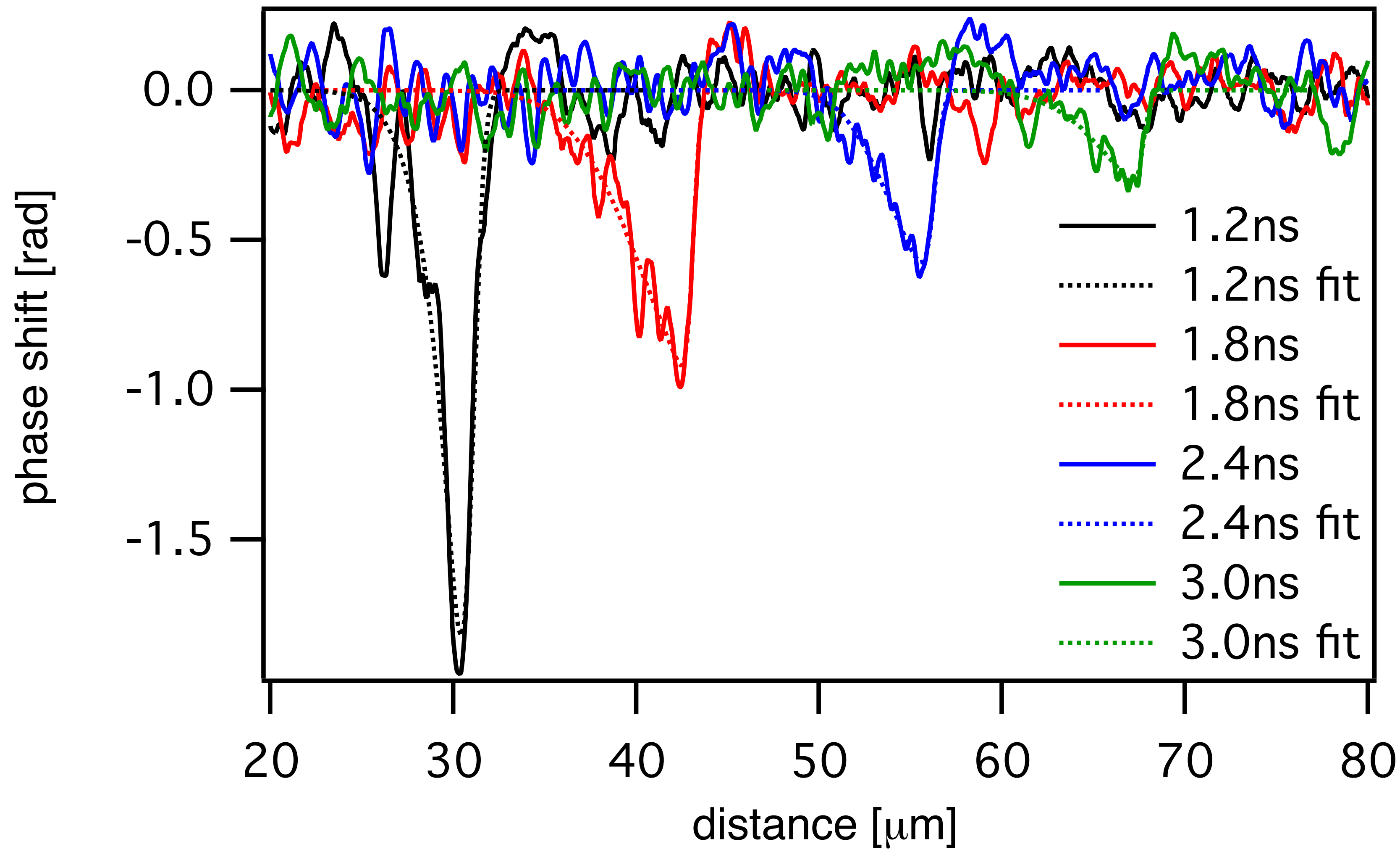


A. Schropp, et al., Sci. Rep. **5**, 11089 (2015).

Elastic Wave in Diamond







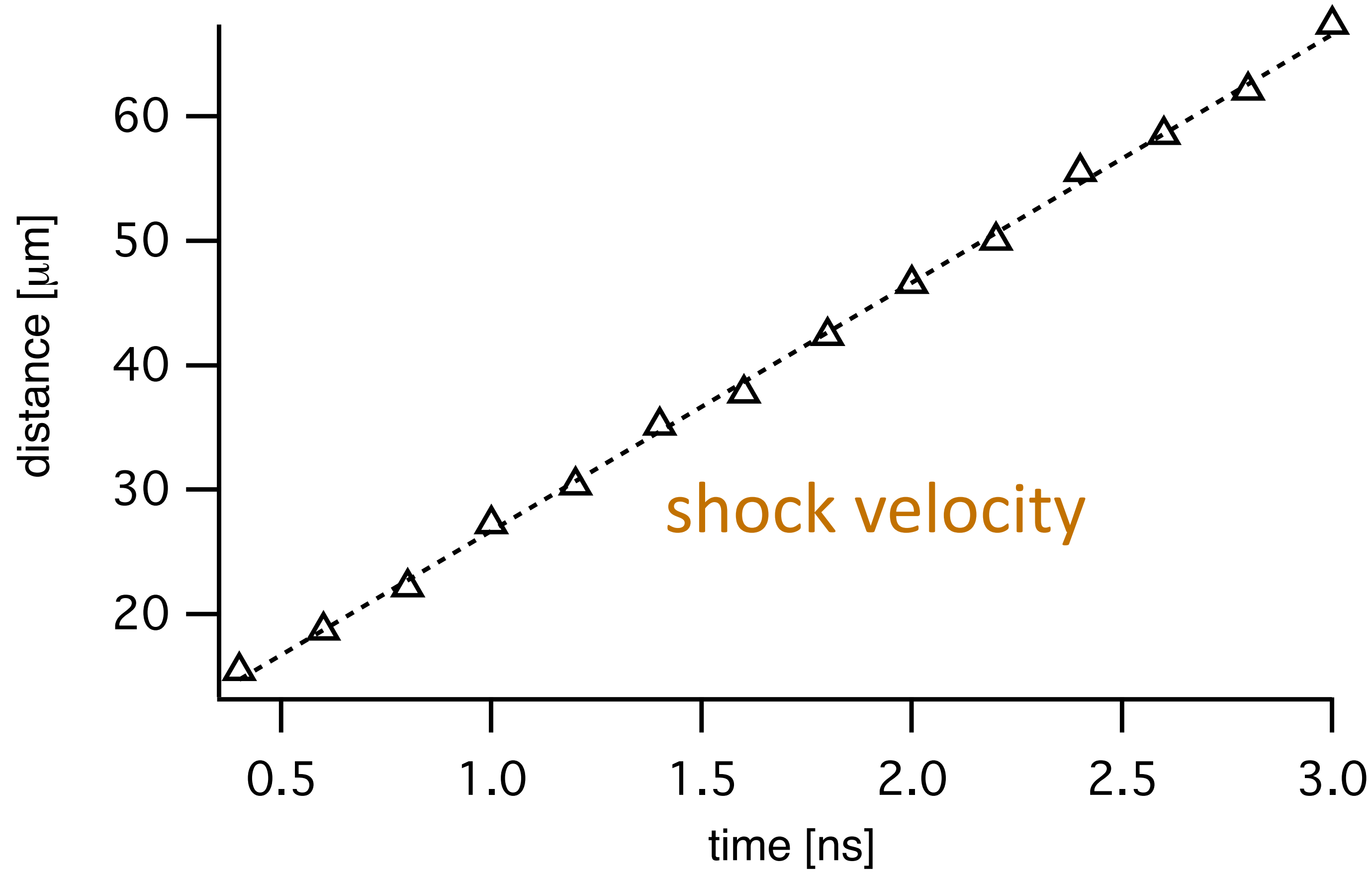
- shock velocity
- density distribution with both high spatial and temporal resolution

A. Schropp, et al., Sci. Rep. **5**, 11089 (2015).

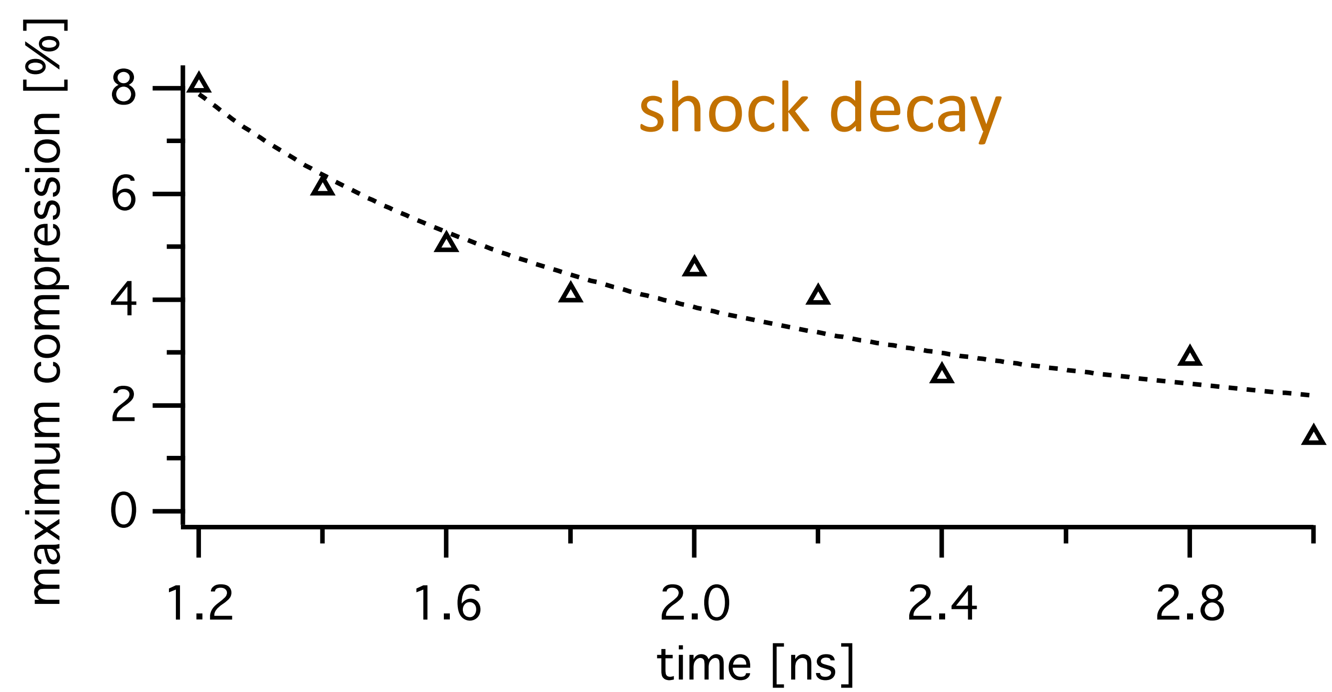
Elastic Wave in Diamond

Quantitative information on

- shock velocity
- compression values
- characteristic time scale of shock decay



- spatial resolution of about 300 nm (SASE)
- PCI: high sensitivity of about 1% lattice compression (not visible in absorption!)

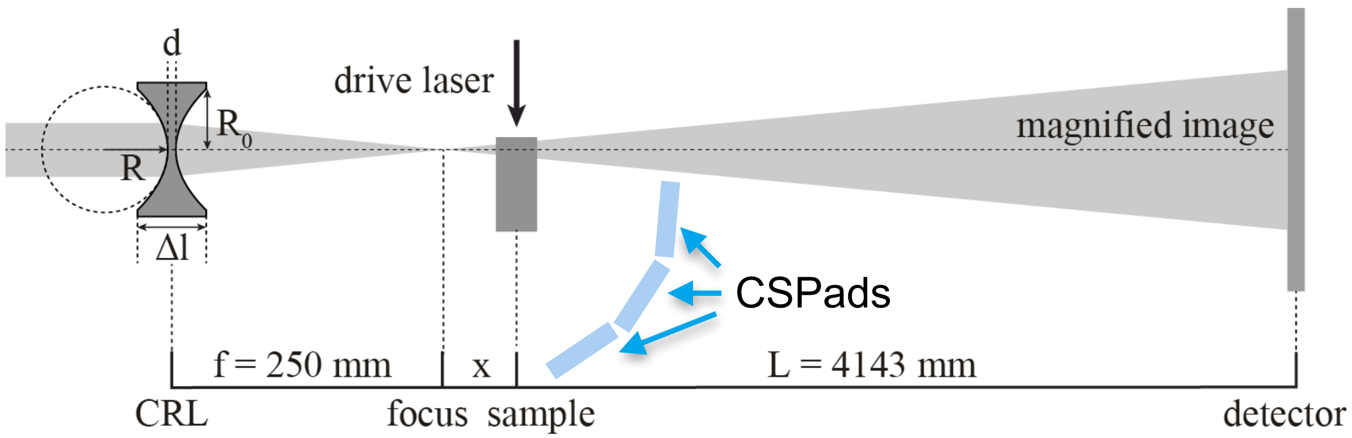


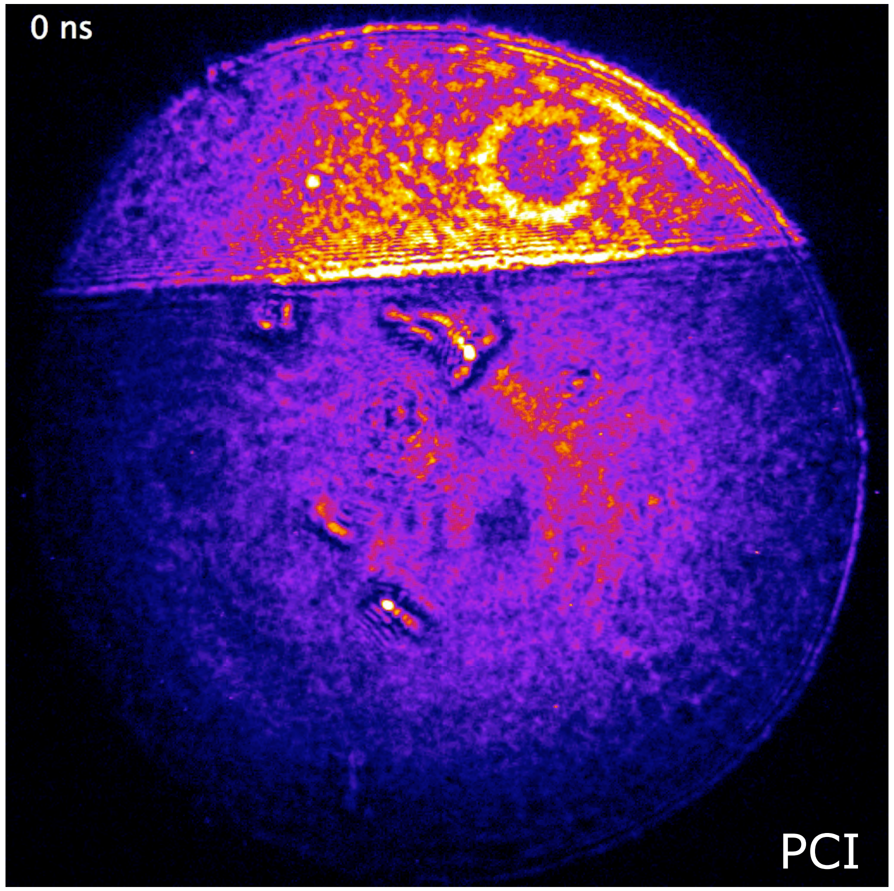
Combining PCI and WAXS - Shock Wave in Silicon

Combining high-resolution phase-contrast imaging with wide-angle X-ray scattering

Drive laser parameters:

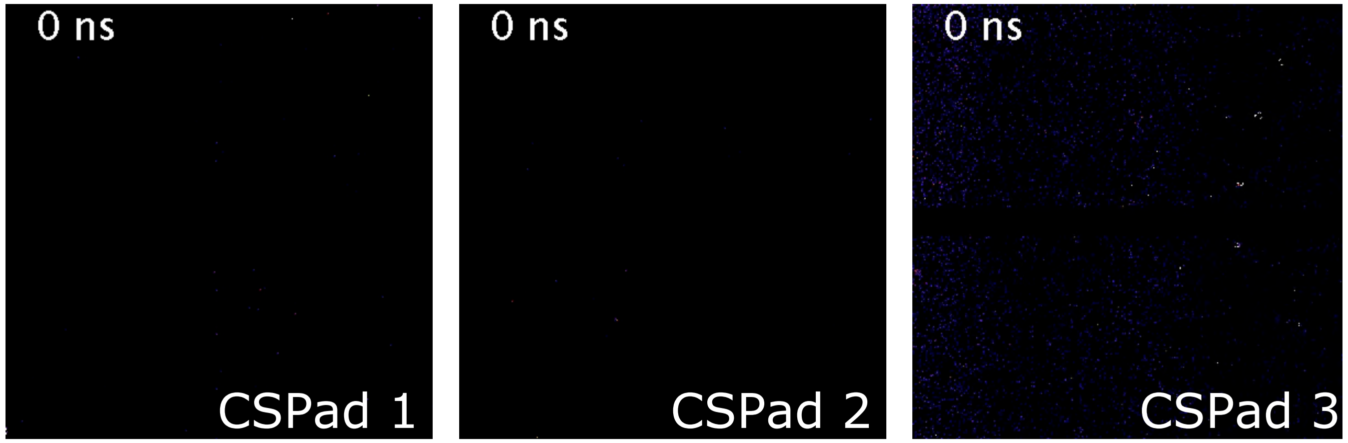
- > long pulse laser (527 nm)
- > 10 ns, 8 J, ramped pulse shape
- > spot size 30 µm (flat top)





Simultaneous measurement of PCI and wide angle X-ray scattering

- > watch phase transformations in real time
- > material recrystallizes (polycrystalline)



nature COMMUNICATIONS

ARTICLE

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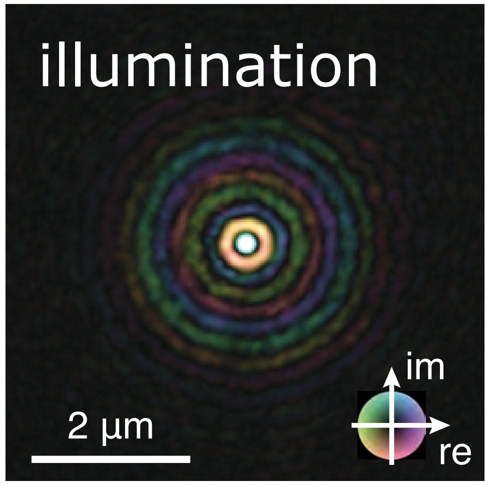
OPEN

Perfect X-ray focusing via fitting corrective glasses to aberrated optics

Frank Seiboth[1,†], Andreas Schropp[2], Maria Scholz[2], Felix Wittwer[1,2], Christian Rödel[3,4], Martin Wünsche[3], Tobias Ullsperger[5], Stefan Nolte[5], Jussi Rahomäki[6], Karolis Parfeniukas[6], Stylianos Giakoumidis[6], Ulrich Vogt[6], Ulrich Wagner[7], Christoph Rau[7], Ulrike Boesenberg[2], Jan Garrevoet[2], Gerald Falkenberg[2], Eric C. Galtier[4], Hae Ja Lee[4], Bob Nagler[4] & Christian G. Schroer[2,8]

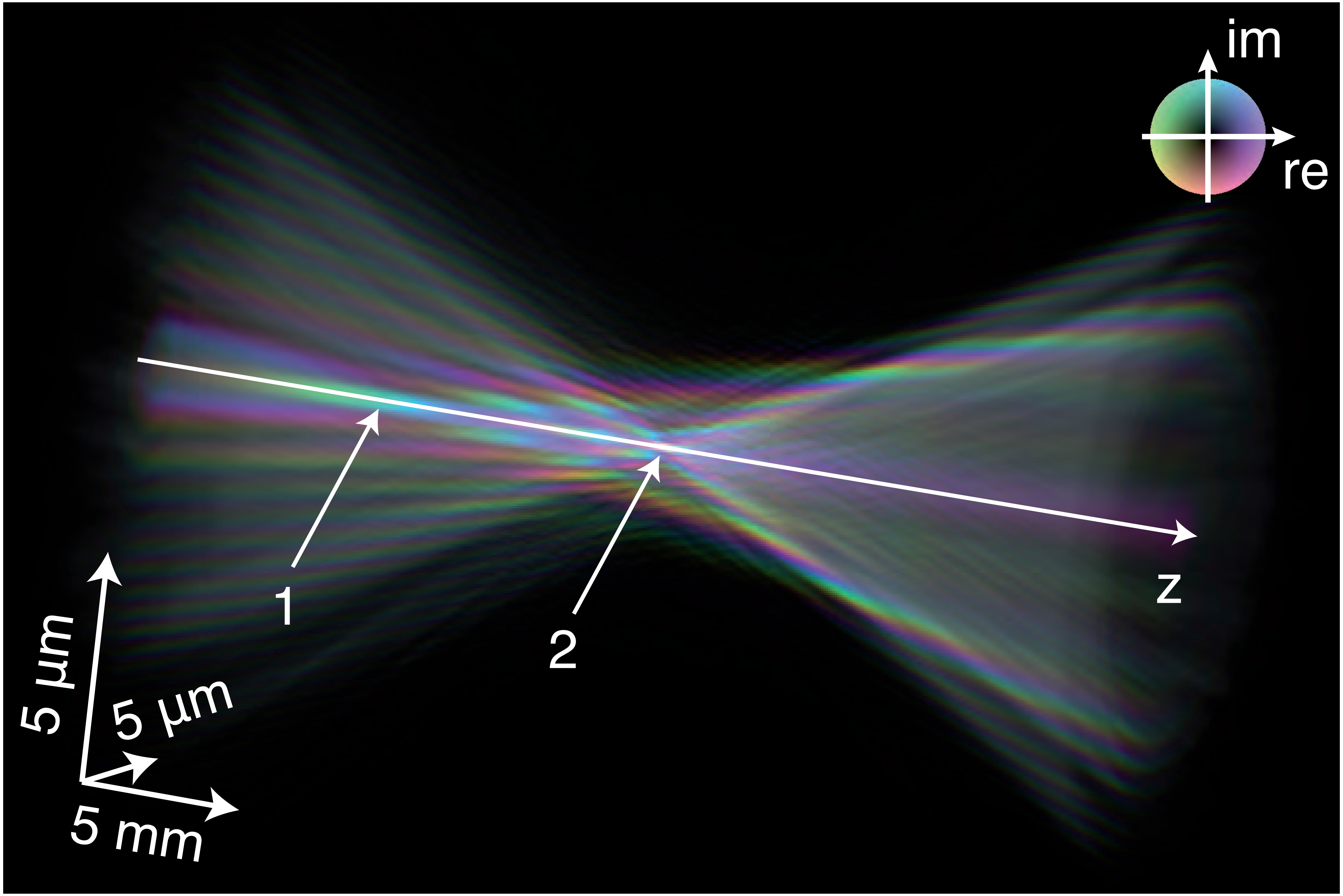
- wave-field near focus known from ptychography
- propagate wavefield to exit of optic → determine phase error
- make corrective phase plate to measure

  → eliminate spherical aberration: perfect focusing



F. Seiboth, *et al.*, Nat. Commun. **8**, 14623 (2017).

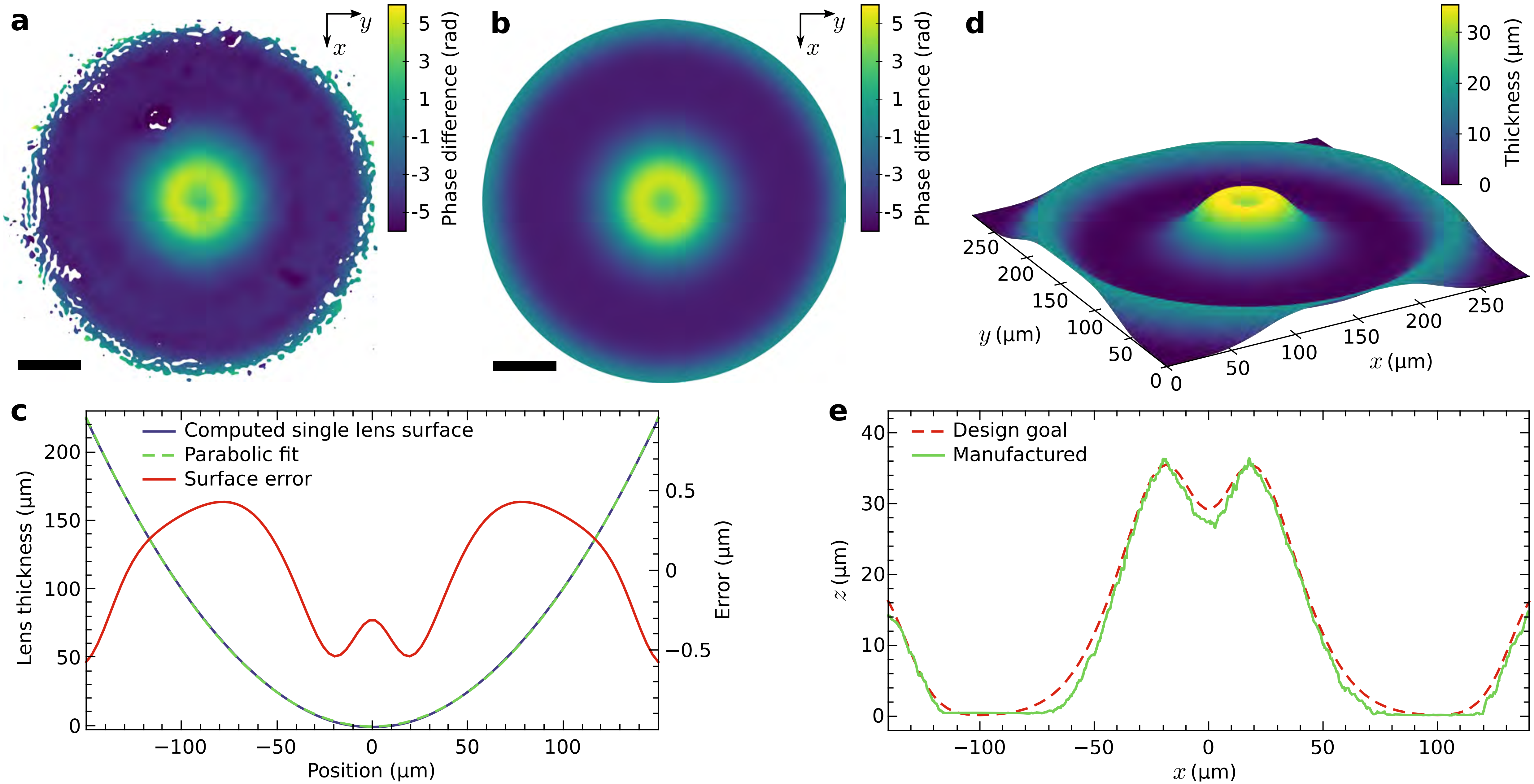
Nanofocused Free-Electron Laser Beam Profile



A. Schropp, et al., Sci. Rep. **3**, 1633 (2013).

Aberrations: Determination of Lens Shape and Error

measured phase error | modelled phase error | modelled phase plate

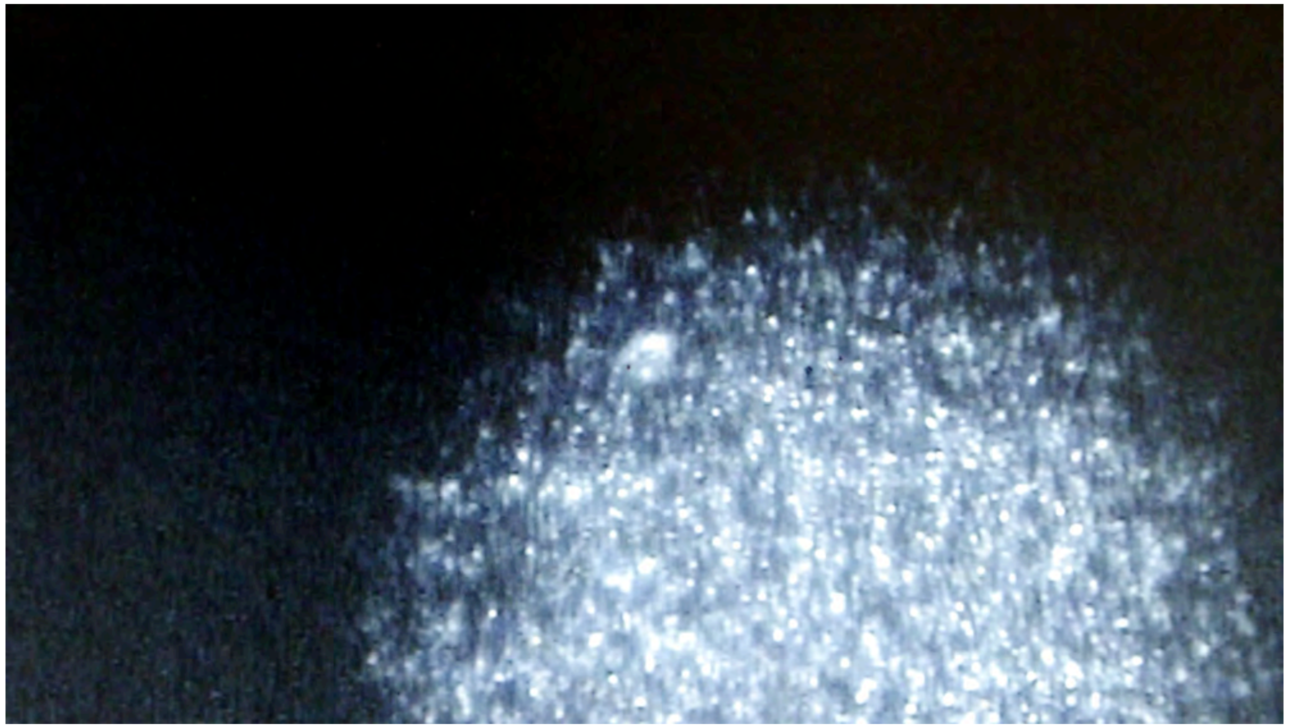


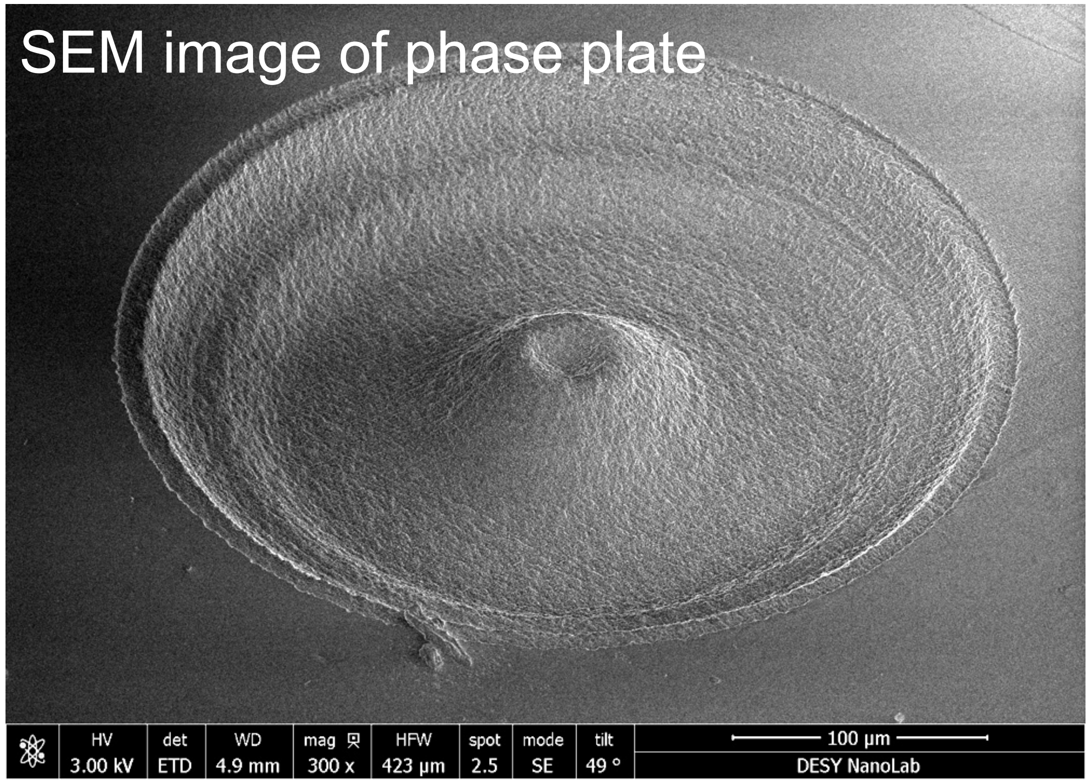
> Shape errors of single Be-CRLs are smaller than 500 nm! Very challenging to improve!
> phase plate for whole stack of lenses easier to fabricate.

F. Seiboth, *et al.*, Nat. Commun. **8**, 14623 (2017).

Aberration Correction: Fabrication of Phase Plate

- For given lens set, make phase plate to measure
- accumulated phase error is more easily corrected
- transmission optic not sensitive to roughness





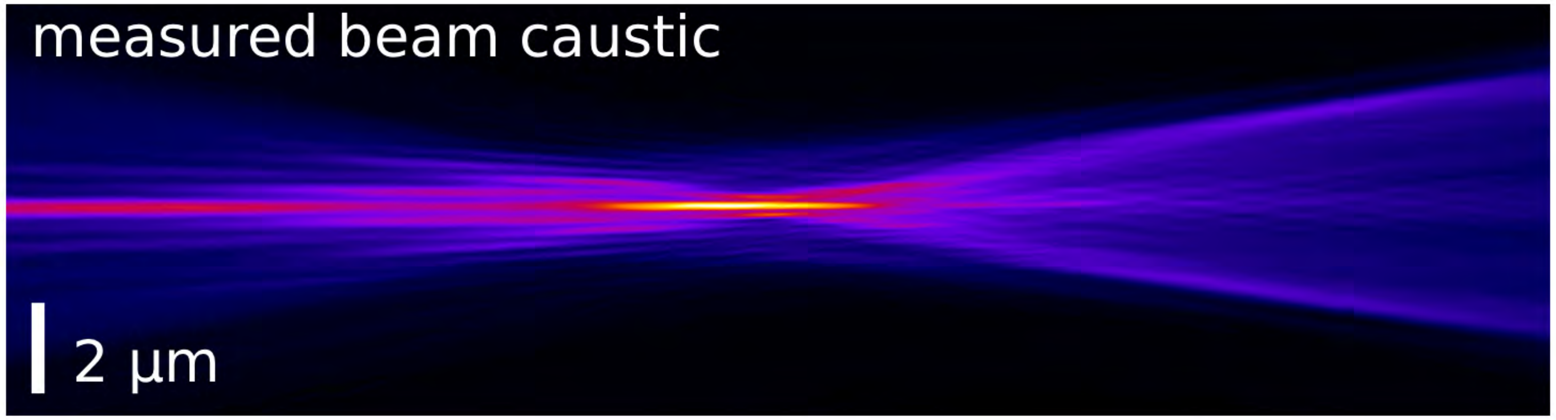
University of Jena:

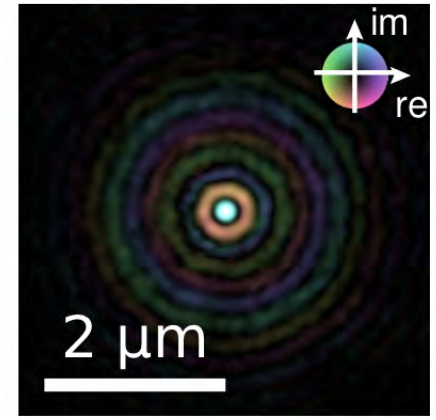
Fabrication of corrective phase plate by laser ablation:

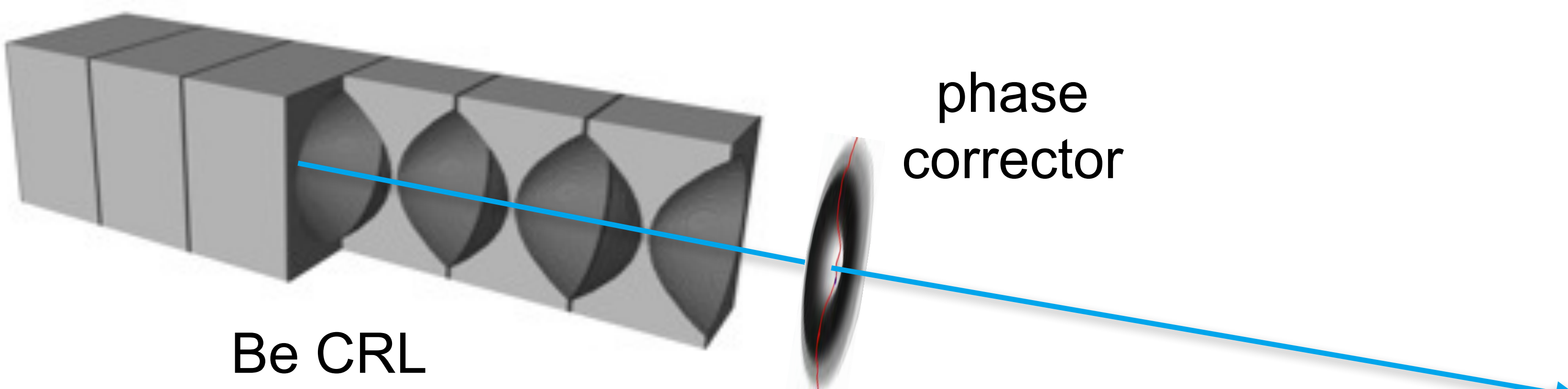
- 8 ps pulses
- 1030 nm wavelength
- 0.2 mJ pulse energy
- focused to substrate with NA = 0.4
- removed layer thickness ~ 1 µm

F. Seiboth, *et al.*, Nat. Commun. **8**, 14623 (2017).

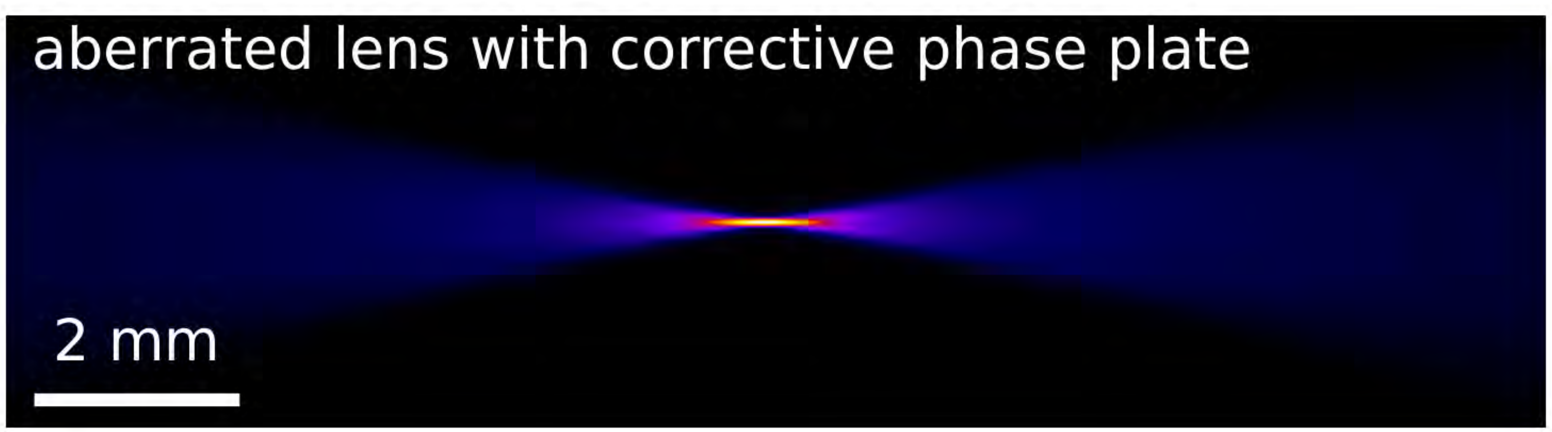
Aberration Correction: Experimental Verification

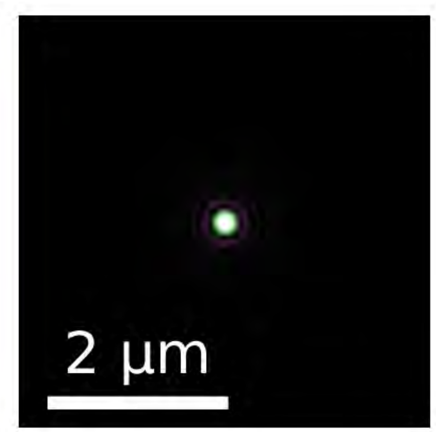






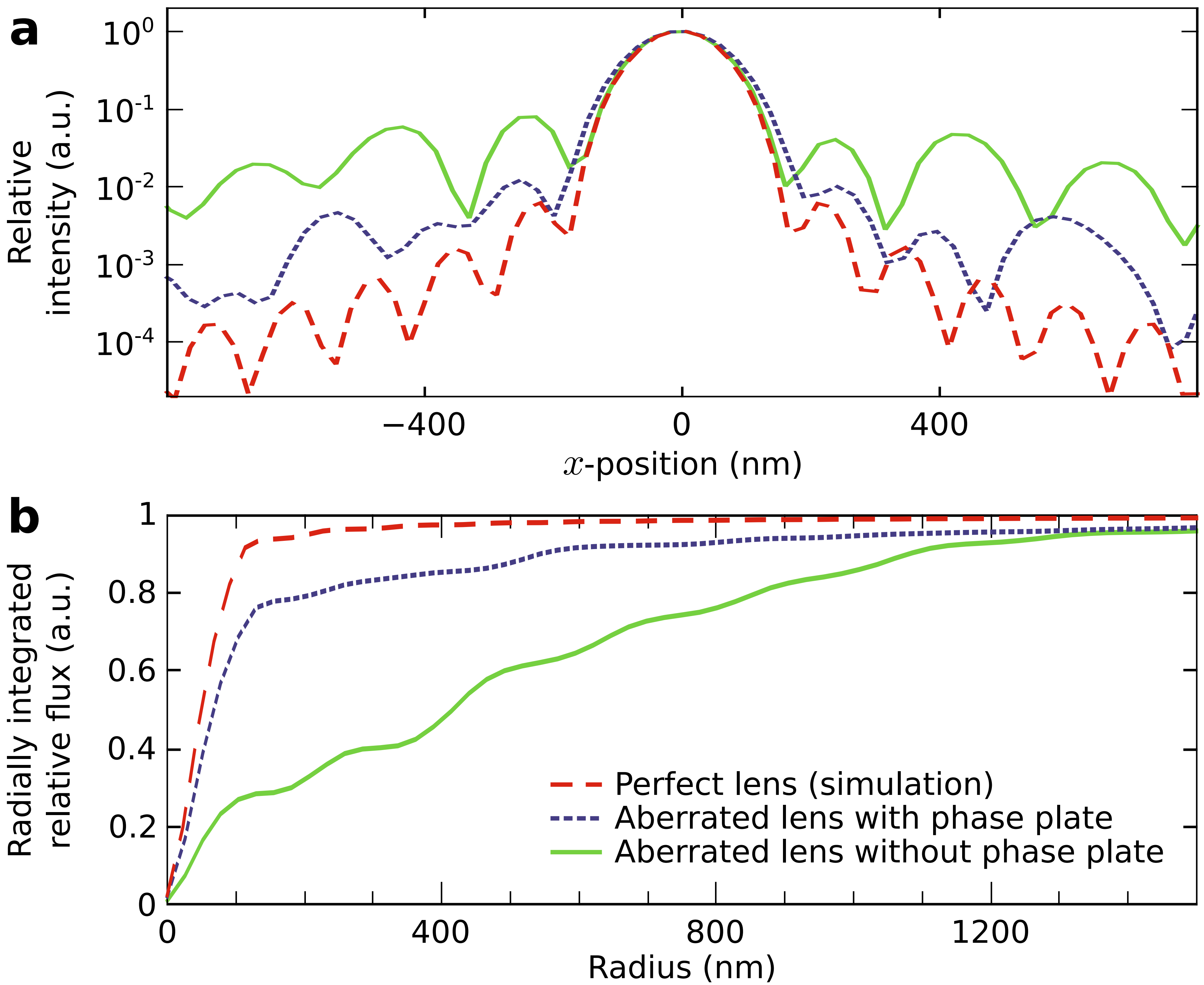
Ptychographic verification of focusing properties





F. Seiboth, *et al.*, Nat. Commun. **8**, 14623 (2017).

Aberration Correction: Experimental Verification



Lens with
Strehl ratio > 0.8!

75 % of the radiation are concentrated in the central speckle!

↓

Focus full beam!

F. Seiboth, *et al.*, Nat. Commun. **8**, 14623 (2017).

Conclusion & Outlook: An X-ray Microscopists Dream

Quantitive in-situ measurement of physical properties of matter

- on all relevant length scales
- on all relevant time scales

→ (in principle) from Å to millimeters

Key technology: brilliant, coherent X-rays with time structure

Requirements:

- high coherent flux
  - → X-ray free-electron lasers
  - → diffraction-limited storage rings (PETRA IV, …)
- efficient nanofocusing
  - → aberration-free optics with high numerical aperture
- stability on nanometer scale

Fusion of real and reciprocal space!