TECHNISCHE UNIVERSITÄT DRESDEN

New Techniques and Methods

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DESY

New Techniques and Methods

Explore the opportunities for different x-ray analytical techniques:

- high brilliance:

  many photons in small phase space volume

- diffraction limit:

  spatial coherence: all photons prepared in same initial state (for given color)

  in practice: coherence length exceeds lateral beam size

Obvious advantages for high-brilliance applications:

- improved photon statistics:

  low background

  large signal in short time

→ high sensitivity, high spatial and temporal resolution

New Techniques and Methods

Explore the opportunities for different x-ray analytical techniques:

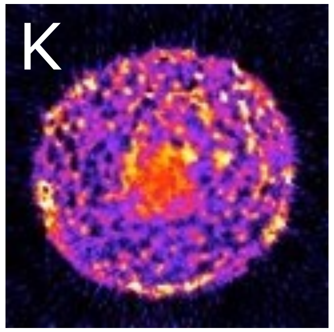
- correlation spectroscopies: XPCS
- scanning probes (diffraction limited nanoprobes)
- coherent x-ray diffraction techniques (CXDI, ptychography, …)
- macromolecular crystallography (MX)
- …

New Techniques and Methods

Challenges:

- beamline: stability
- beamline: background radiation
- sample: radiation damage
- ...

X-Ray Microscopy on the Nanometer Scale



Broad field of applications:

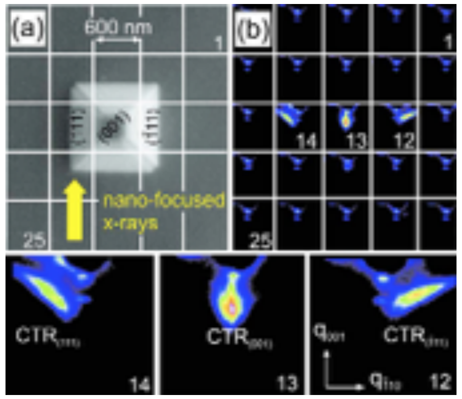
- Physics, chemistry, biology, earth- and environmental science, nano-technology, ...
- Main advantage: large penetration depth

  → in-situ and in-operando studies

- X-ray analytical contrasts: XRD, XAS, XRF, ...

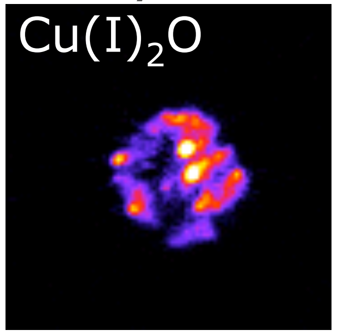
  → atomic scale resolution

quantum dots



M. Hanke, et al.,
APL **92**, 193109 (2008)

catalysts



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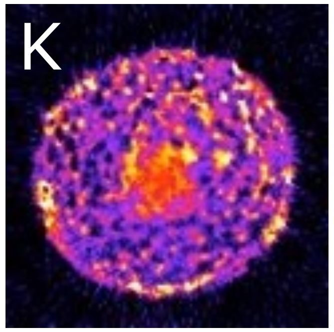

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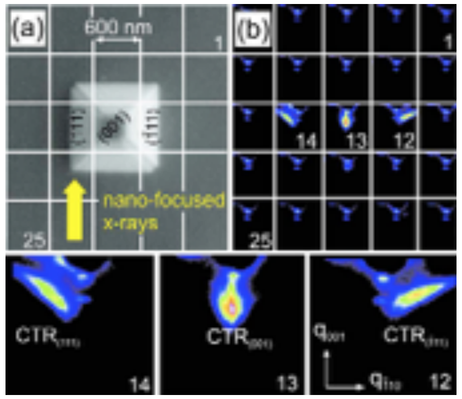

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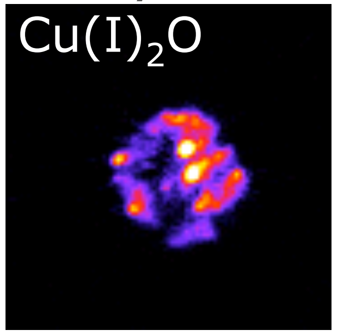
Today: “Mesoscopic gap”

real-space resolution: down to about 10 nm

XRD and XAS: atomic scale

What happens on (mesoscopic) 1 - 10 nm scale?

catalysts



X-Ray Microscopy on the Nanometer Scale

Today: “Mesoscopic gap”

real-space resolution: down to about 10 nm

XRD and XAS: atomic scale

What happens on (mesoscopic) 1 - 10 nm scale?

Many interesting physics and chemistry questions:

investigate local states:

- individual defects (0D): changes in electron density, charge ordering
- (structural) domain boundaries (2D), e. g., in multiferroics
- mesoscopic dynamics at (solid-state) phase transitions
- catalytic nanoparticles (activity in operando)
- ...

Mesoscale also very important for nanotechnology (e. g., defects in devices)!

Current State in X-Ray Microscopy

Conventional x-ray microscopy

→ optics limit spatial resolution: diffraction limited focusing

$$d_t \approx \frac{\lambda}{2NA}$$ (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)]

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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)]

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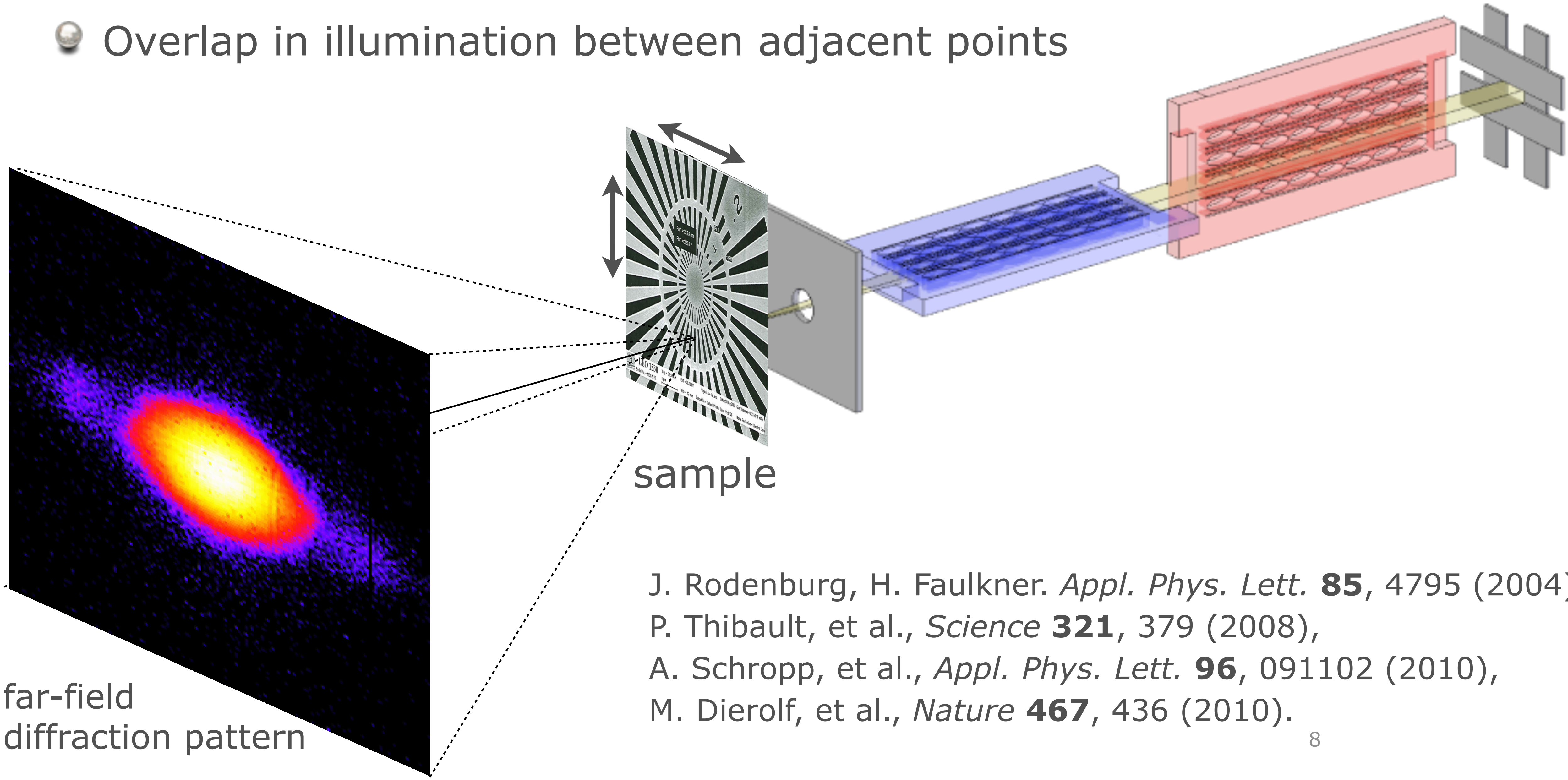
Scanning Coherent X-Ray Diffraction Imaging: Ptychography

- 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),
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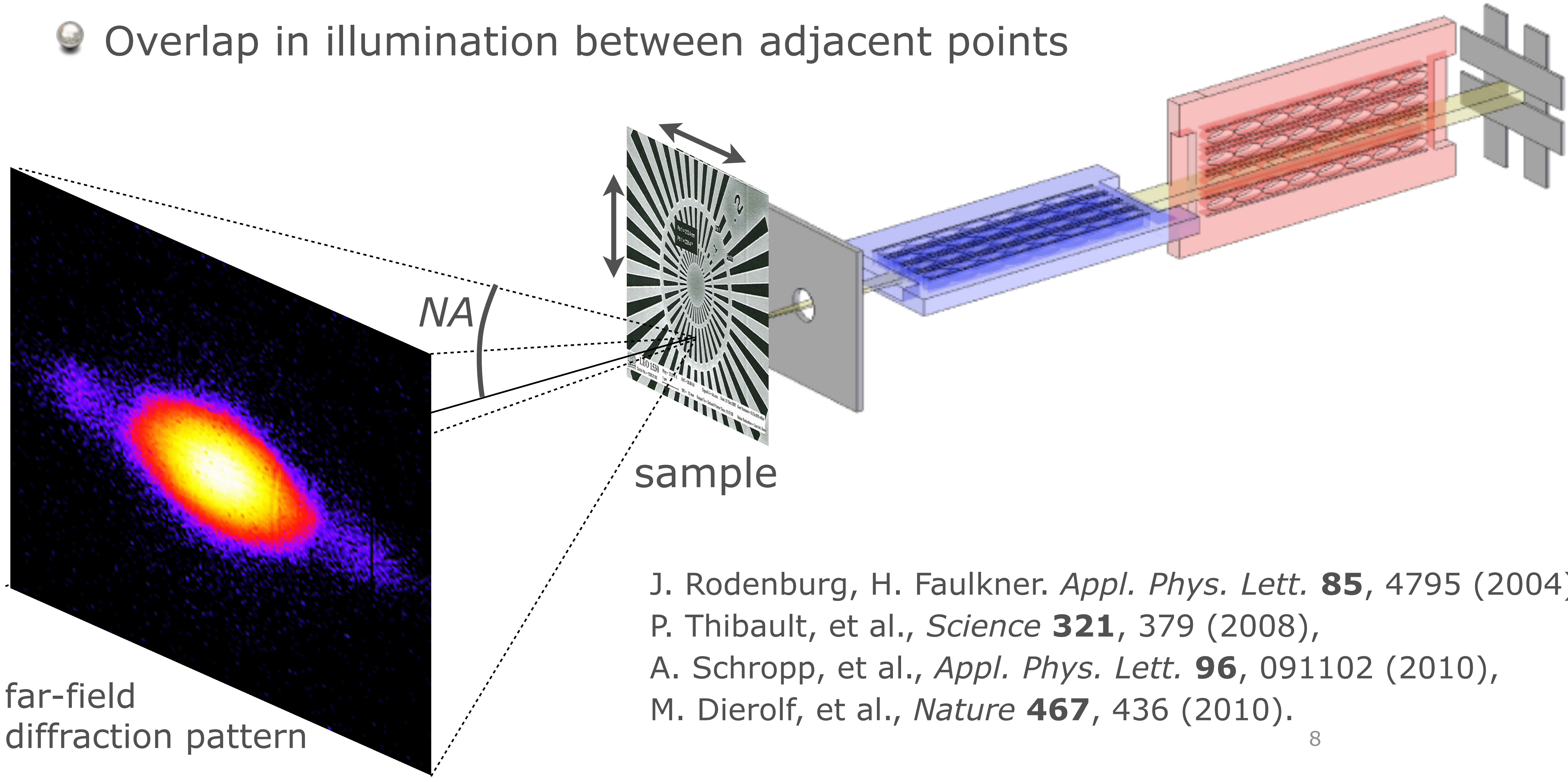

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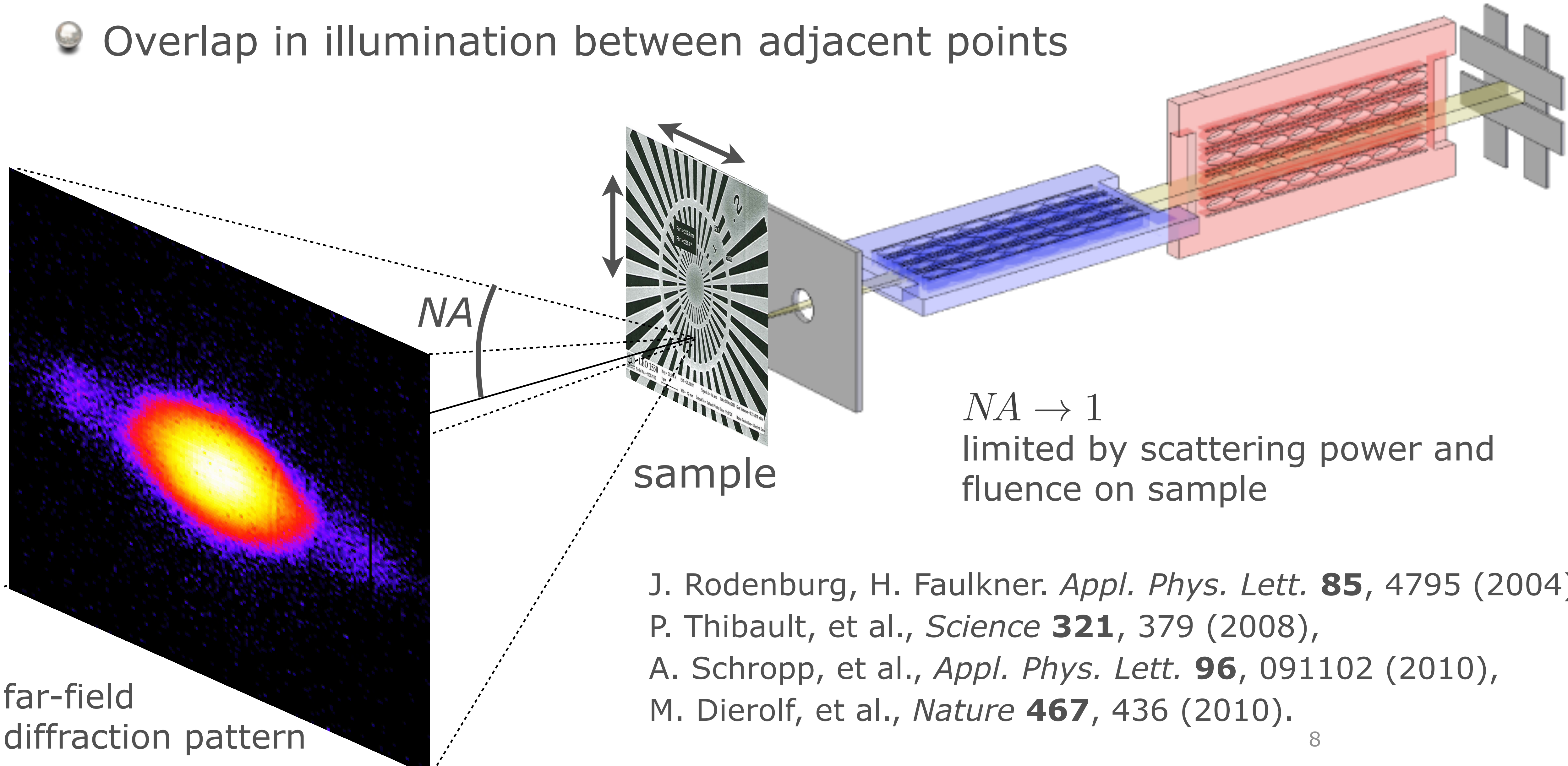

$NA \to 1$
limited by scattering power and fluence on sample

J. Rodenburg, H. Faulkner. *Appl. Phys. Lett.* **85**, 4795 (2004),
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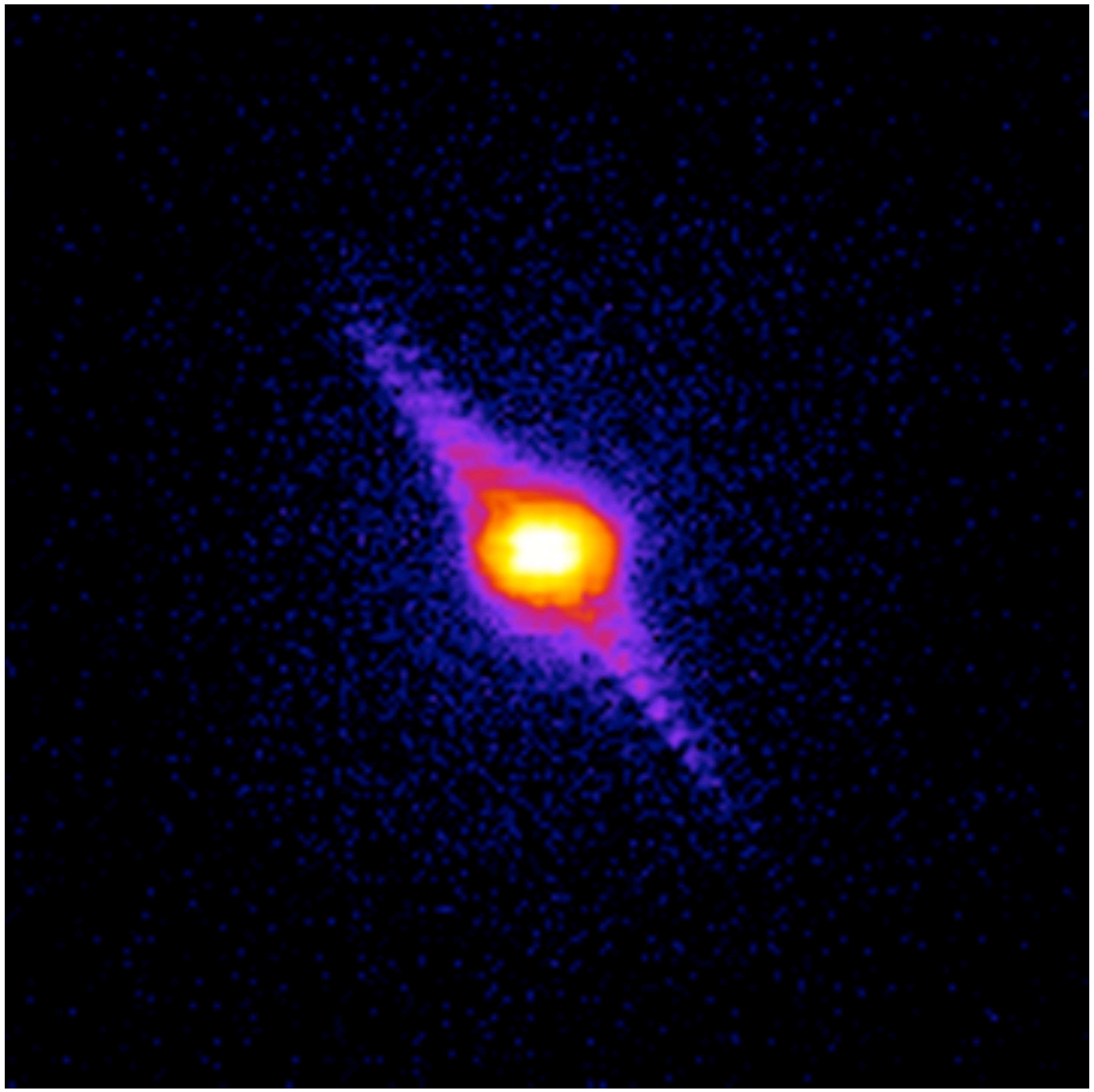
Ptychographic Microscopy



Experiment at P06:

detector:
Pilatus 300k (172µm pixel size)

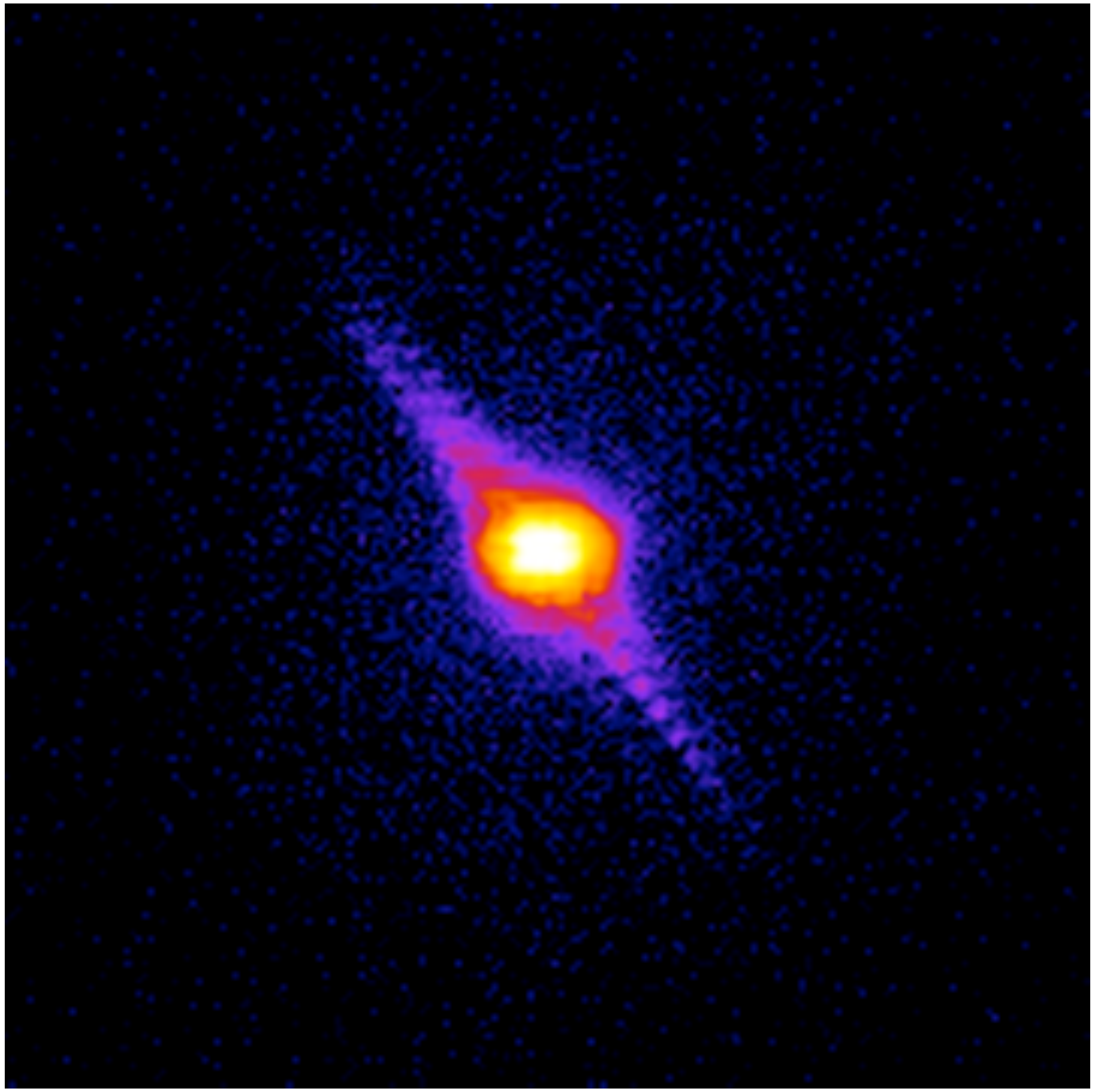
sample-detector distance:
2080 mm

exposure time:
1.5 s per point

Sample: NTT AT test pattern

SEM image (I. Snigireva)

Ptychographic Microscopy



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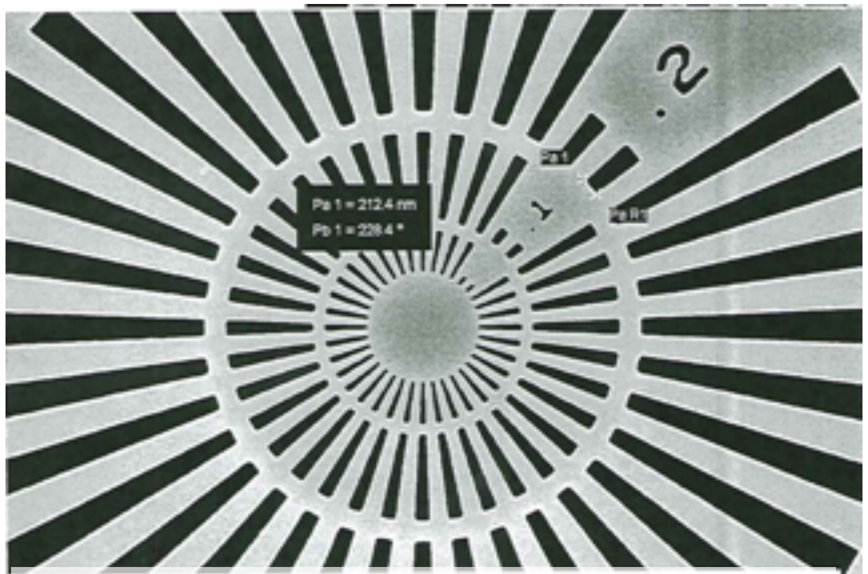
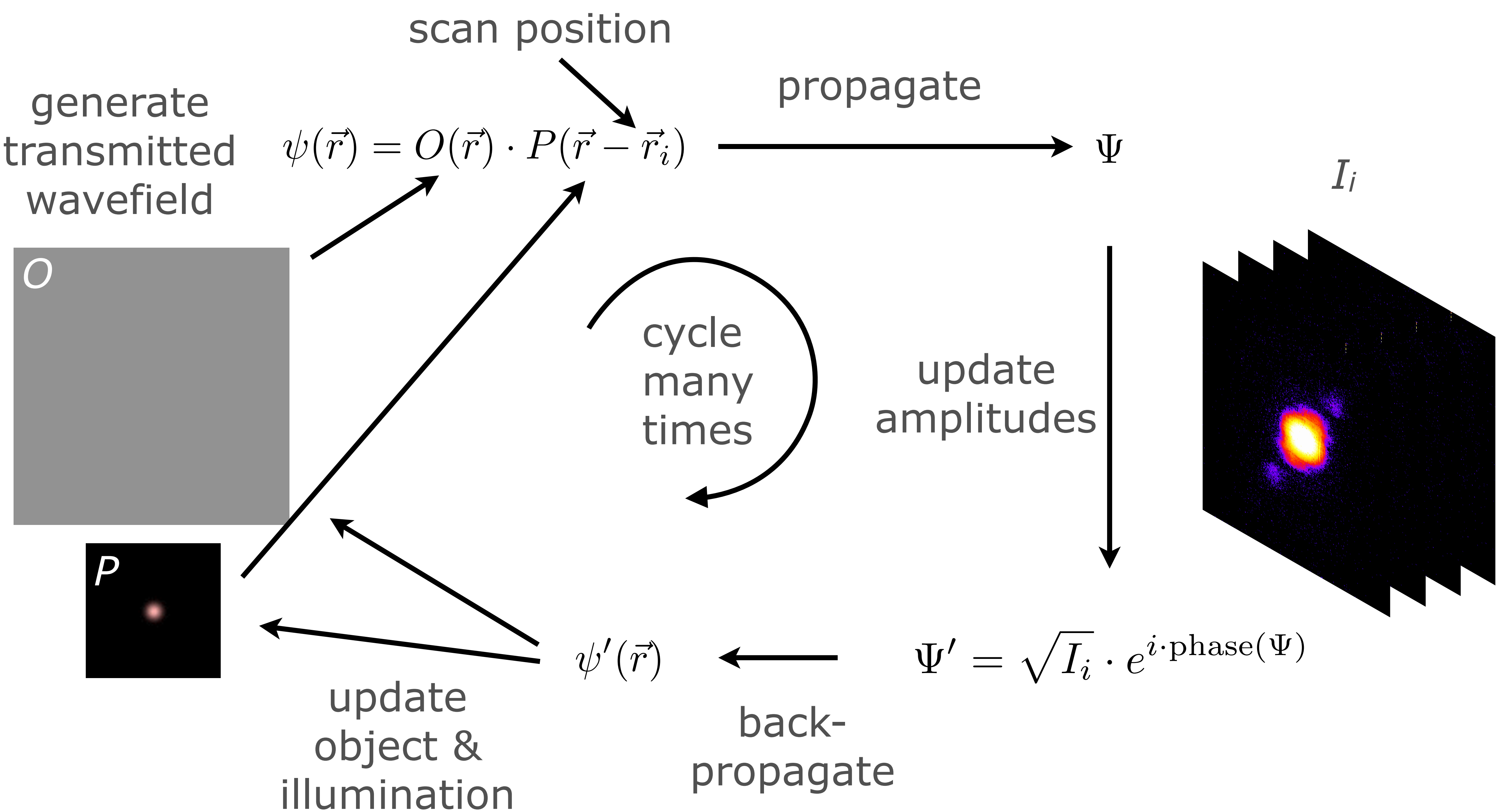


SEM image (I. Snigireva)

Ptychography: Reconstruction



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

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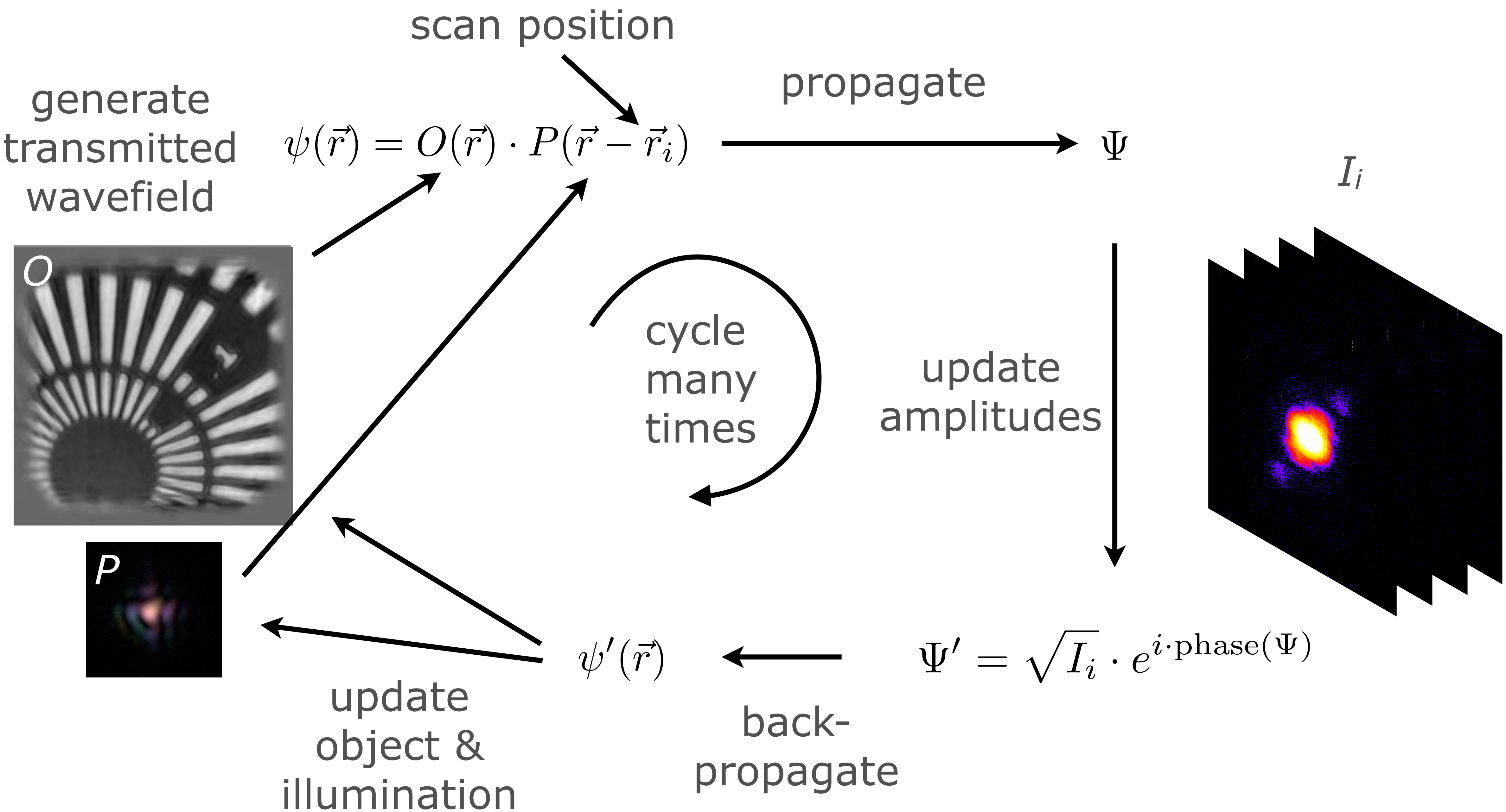

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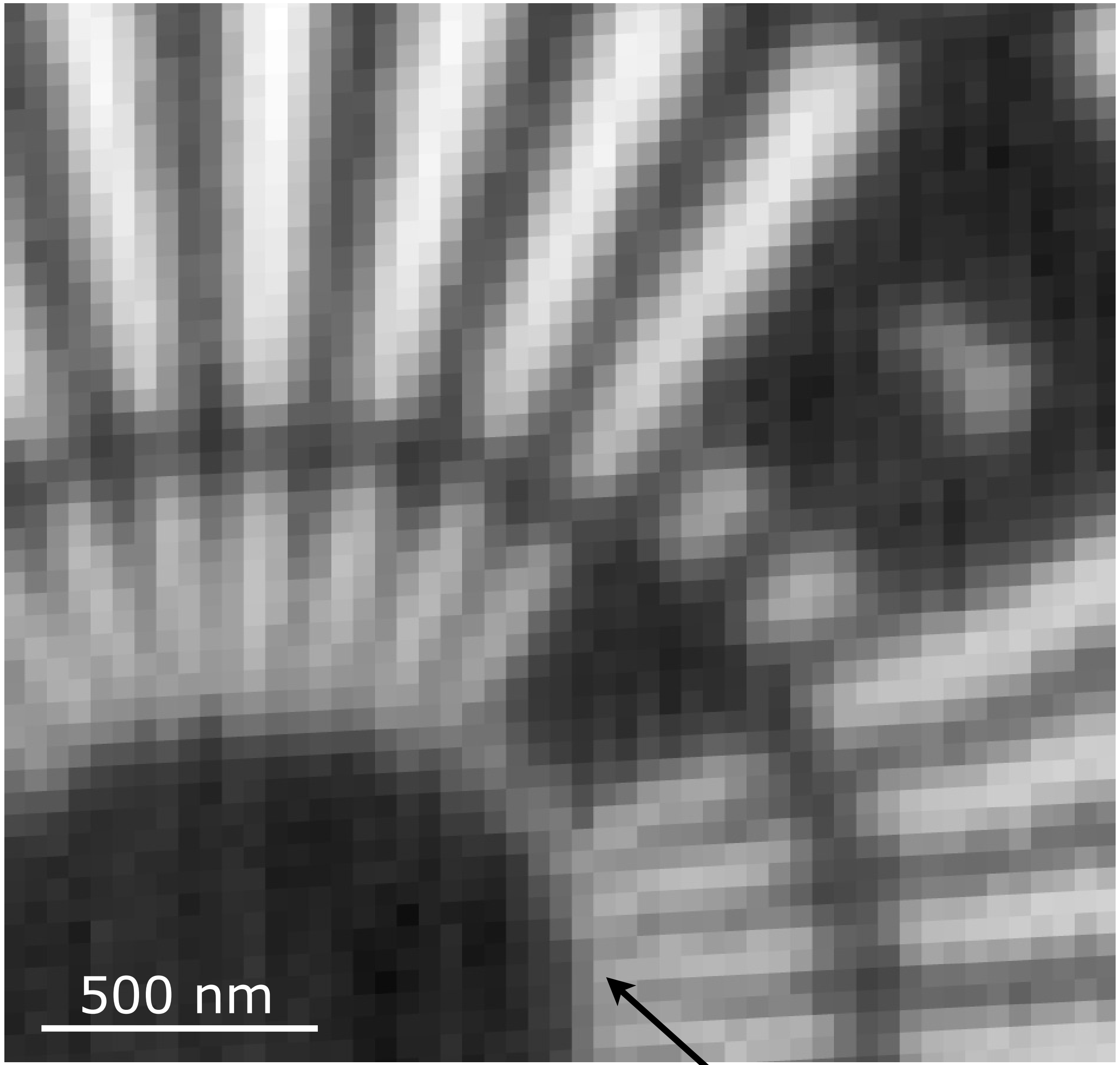
Scanning Microscopy: Fluorescence Imaging



Ta Lα fluorescence

E = 15.25 keV
50 x 50 steps of 40 x 40 nm^2
2 x 2 μm^2 FOV
exposure: 1.5 s per point

50 nm lines and spaces

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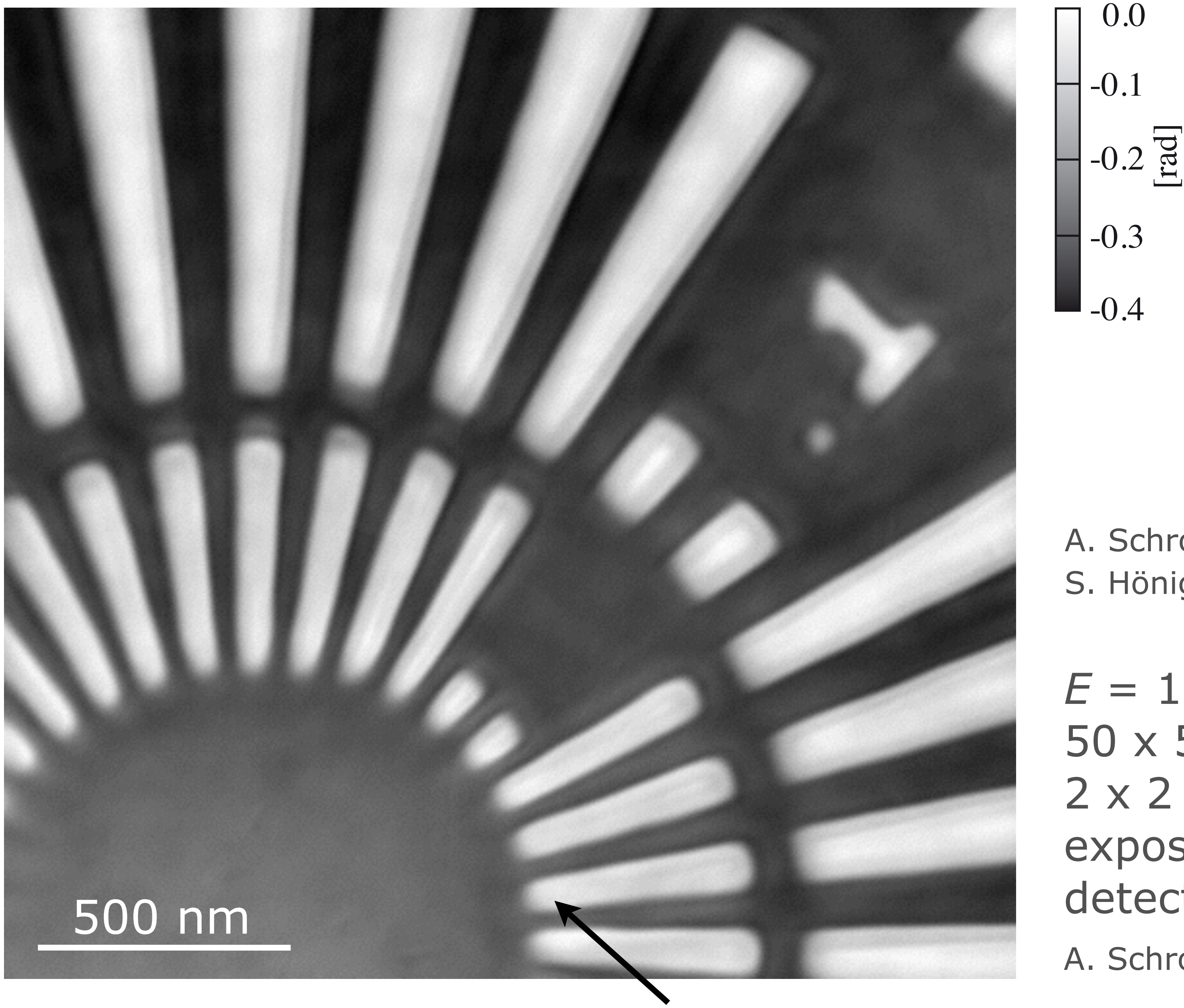
Scanning Microscopy: Ptychography



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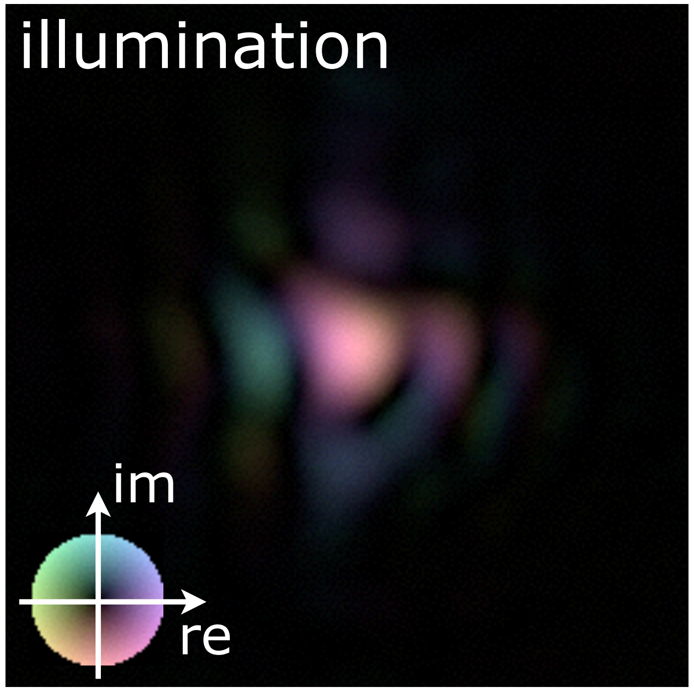

A. Schropp, et al., APL **96**, 091102 (2010),
S. Hönig, et al., Opt. Exp. **19**, 16325 (2011).

E = 15.25 keV
50 x 50 steps of 40 x 40 nm^2
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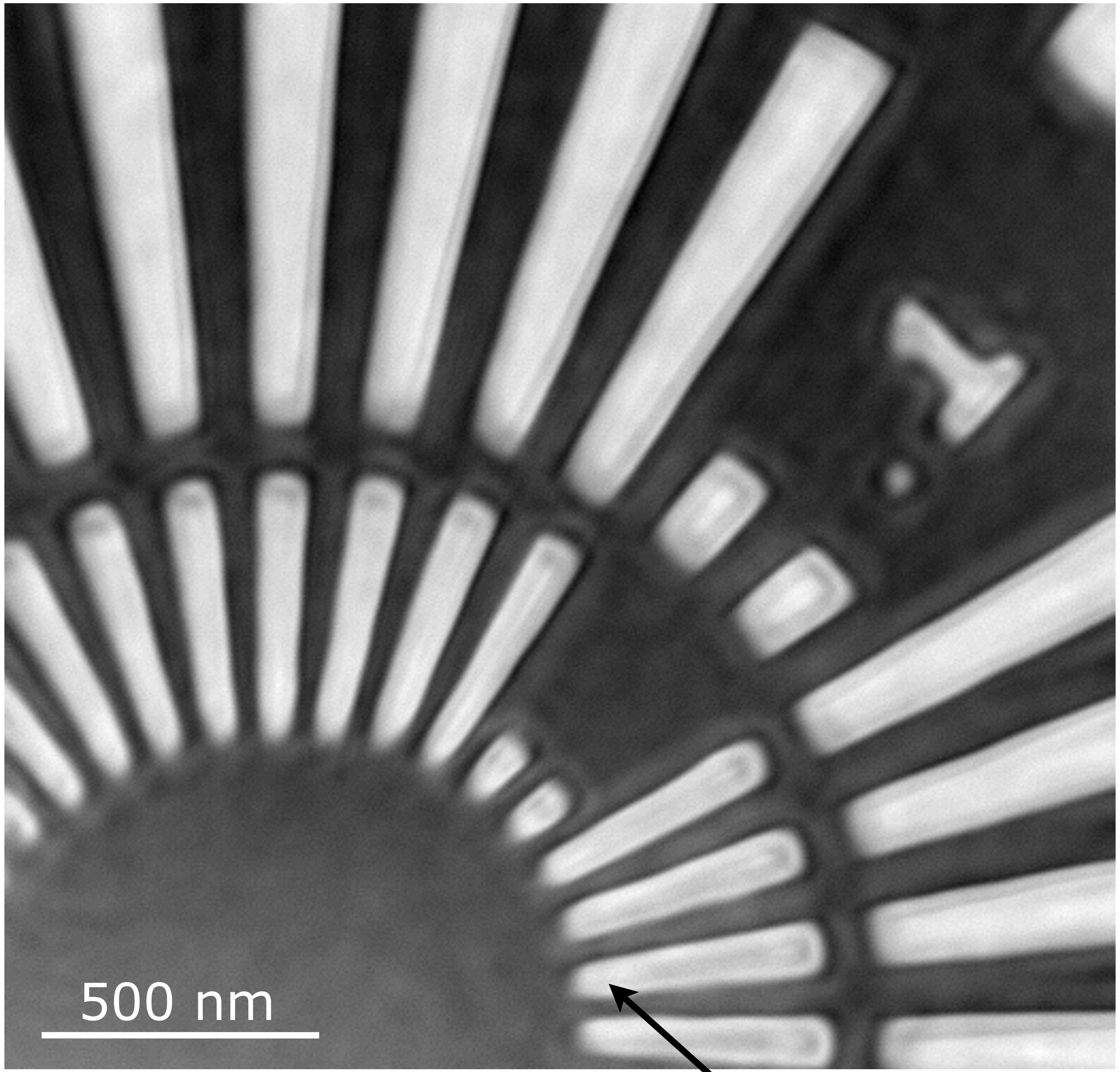
exposure: 1.5 s per point
detected fluence: $2.75 \cdot 10^4$ ph/nm^2

A. Schropp, et al., APL **100**, 253112 (2012).

Scanning Microscopy: Ptychography



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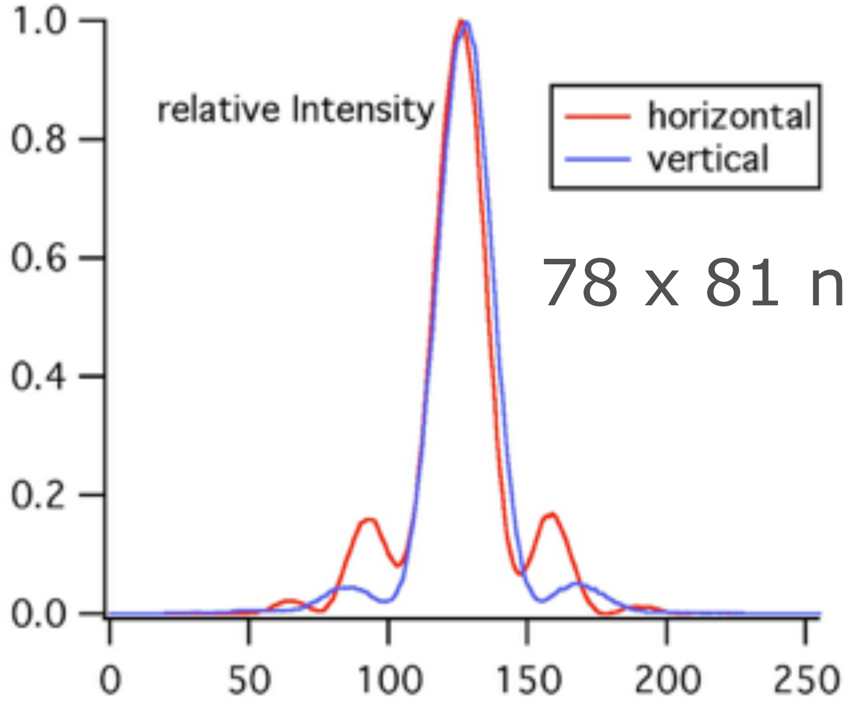
illumination



78 x 81 nm^2

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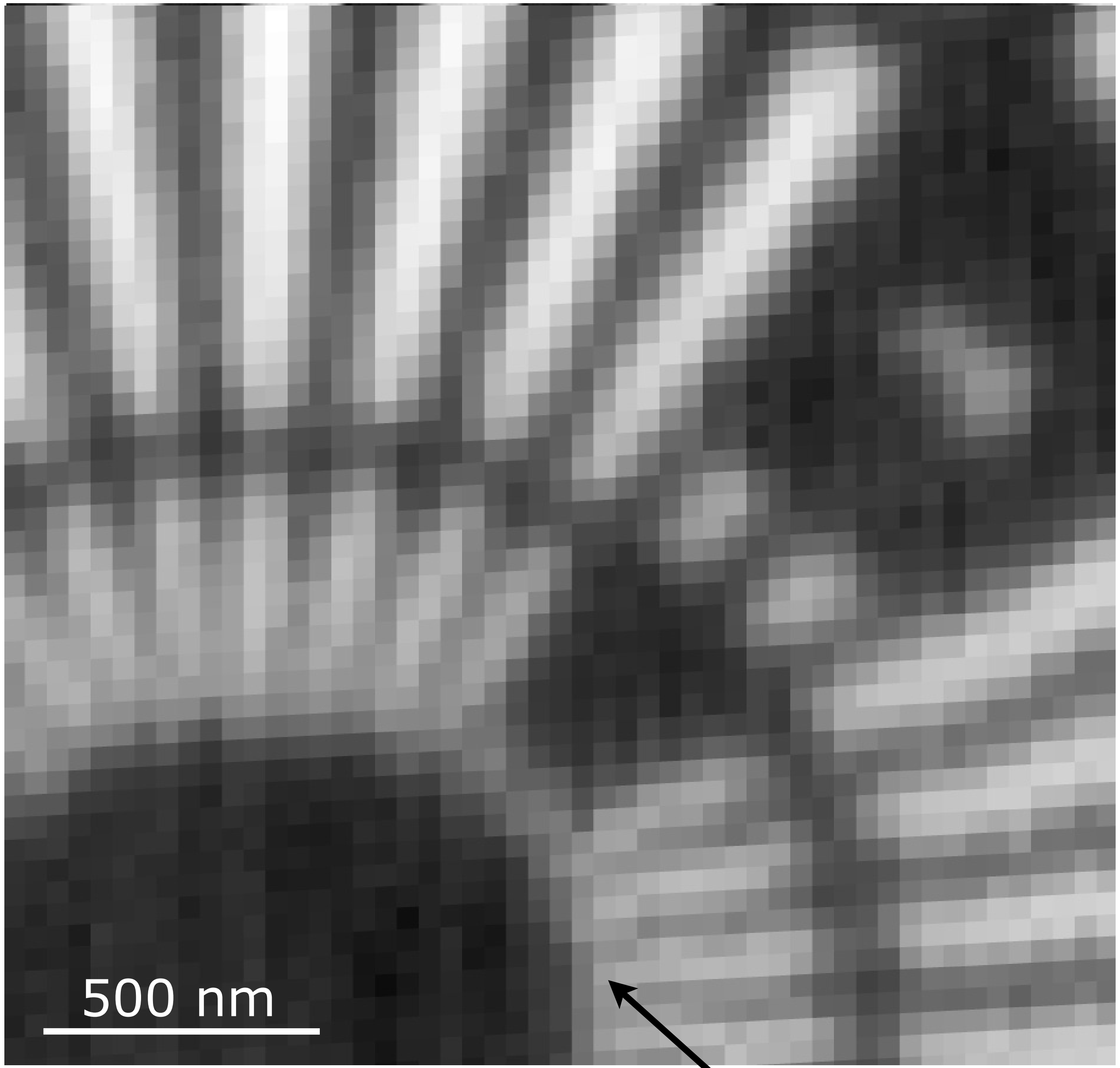

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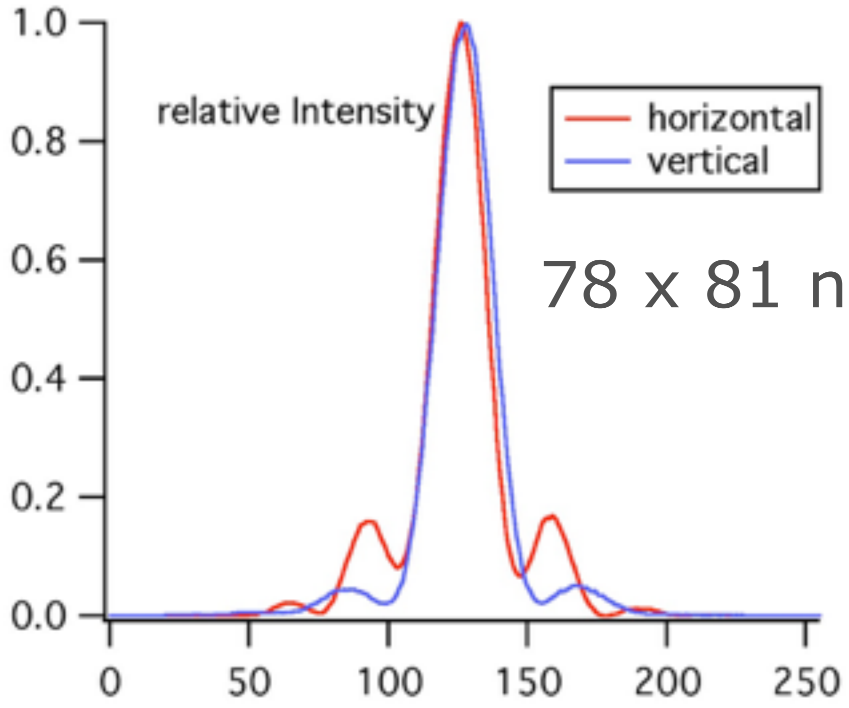

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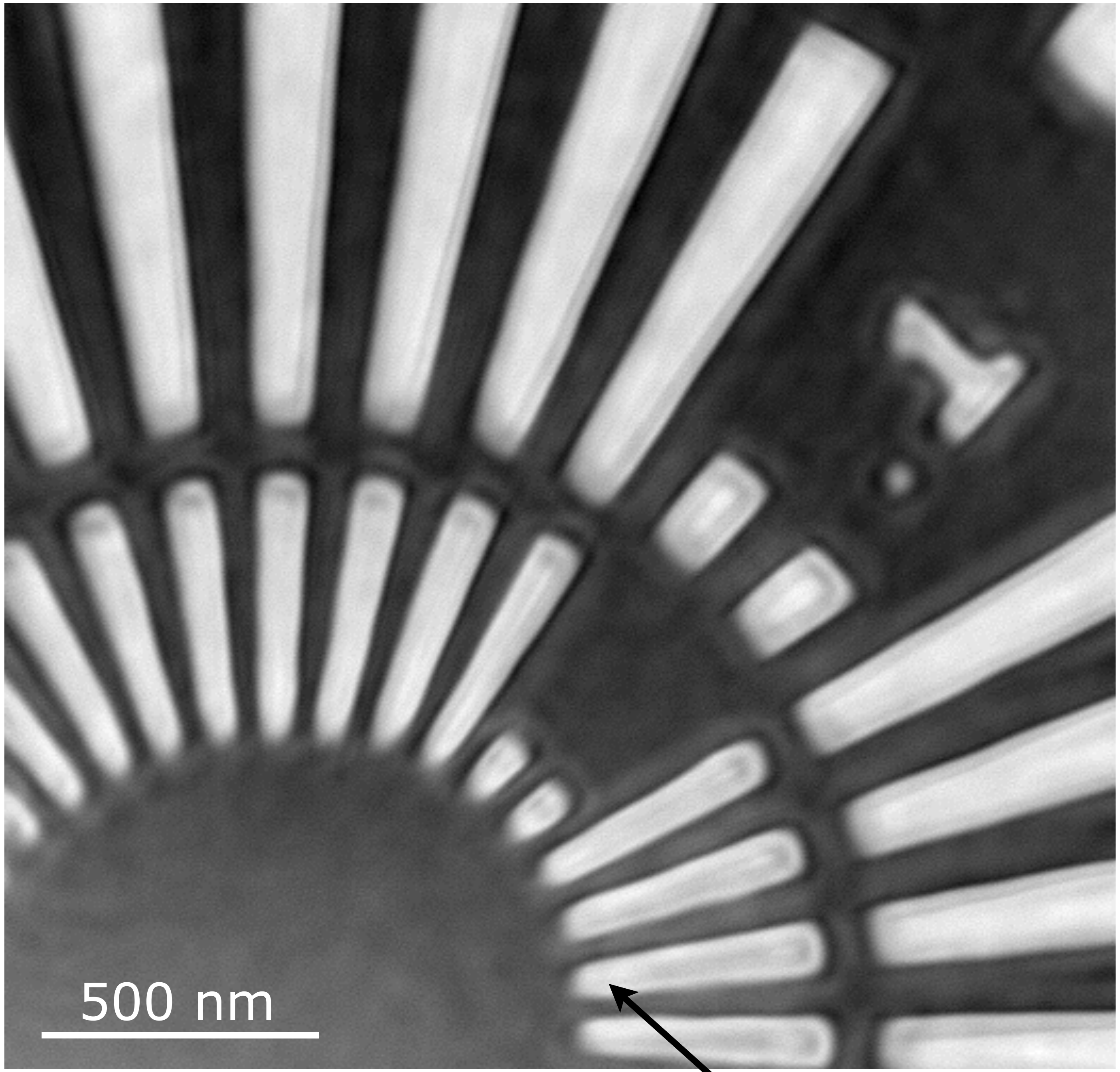

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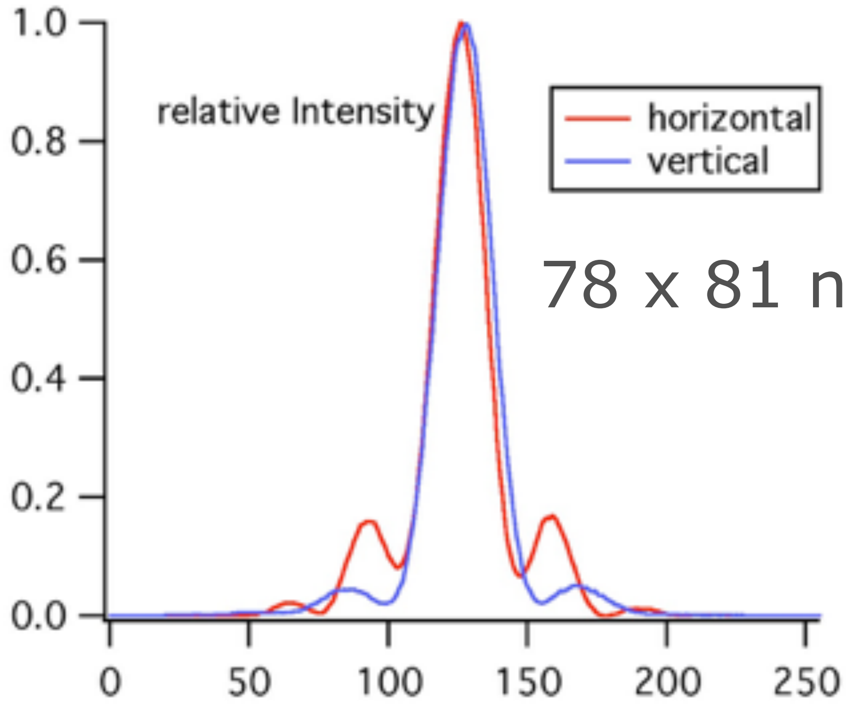

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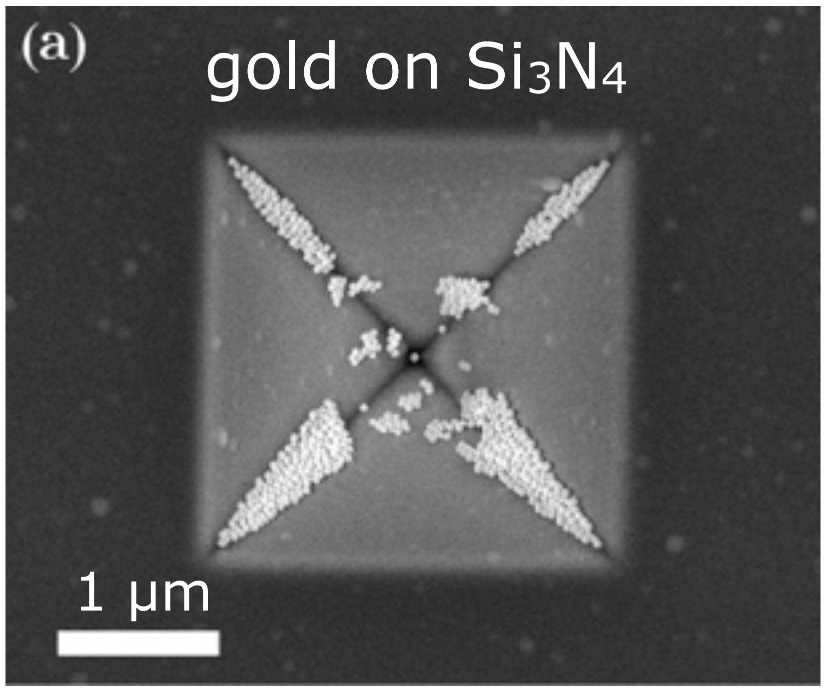
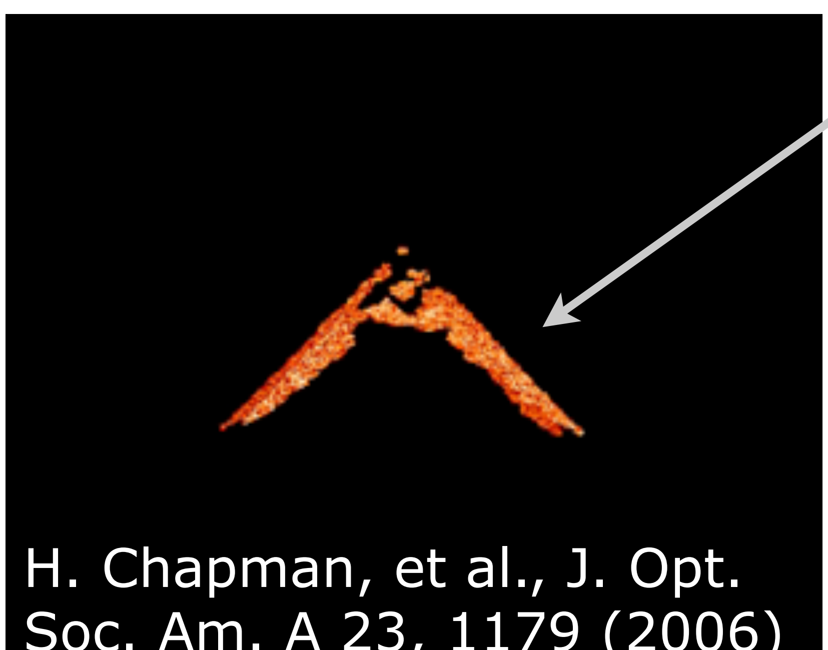
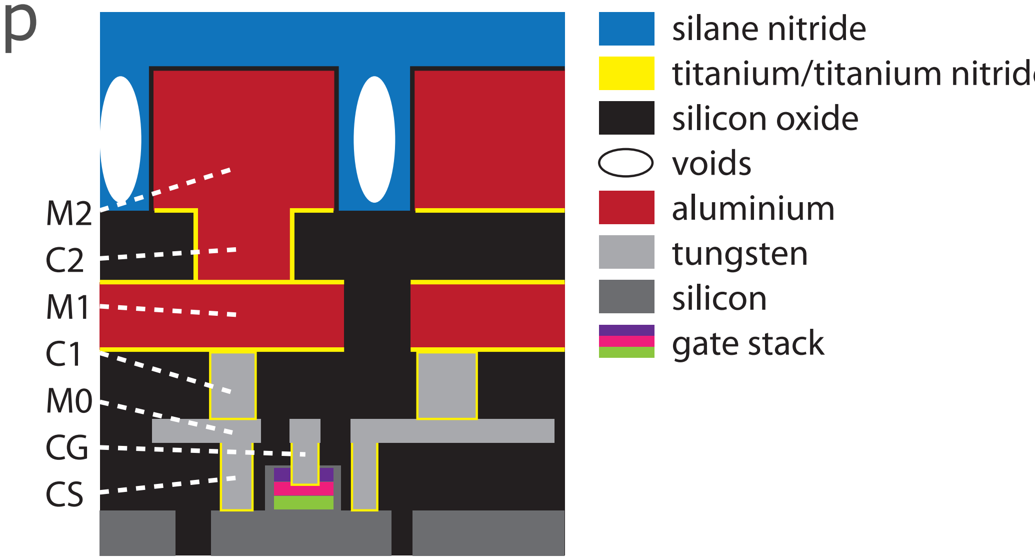
Contrast in Coherent X-Ray Diffraction Imaging



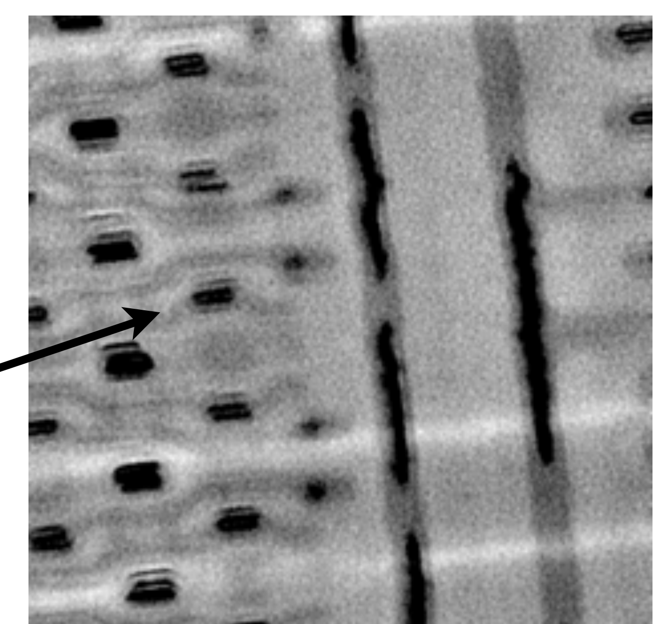


Au particles, only!
No membrane!

Microchip



Only tungsten!
Where is the
Al, Si, ... ?



A. Schropp, et al., J. Microscopy, **241**(1), 9 (2011)

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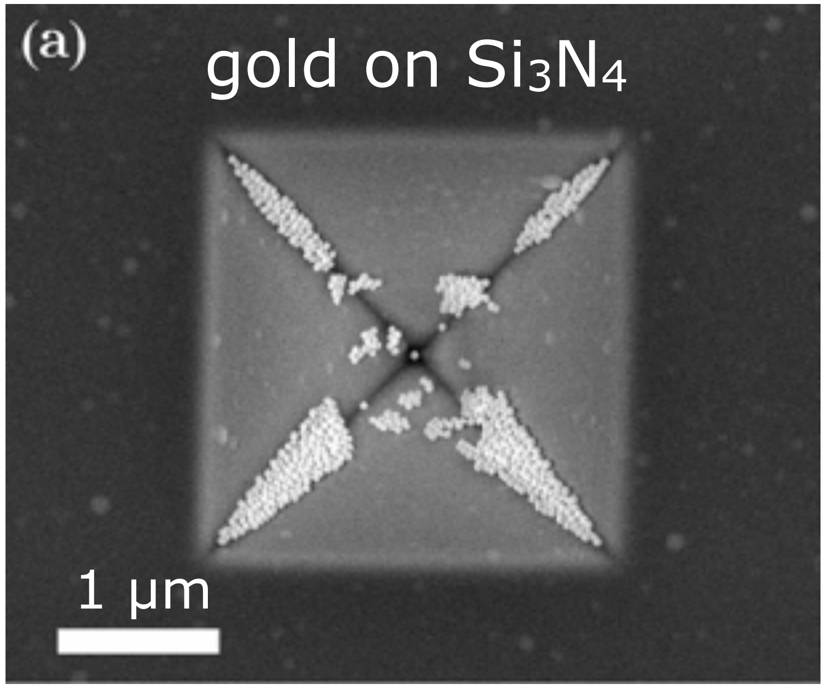

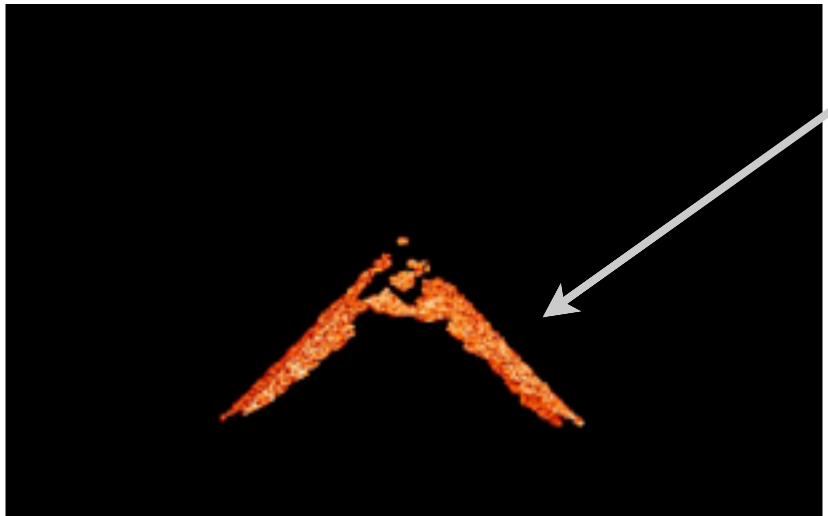

H. Chapman, et al., J. Opt. Soc. Am. A 23, 1179 (2006)

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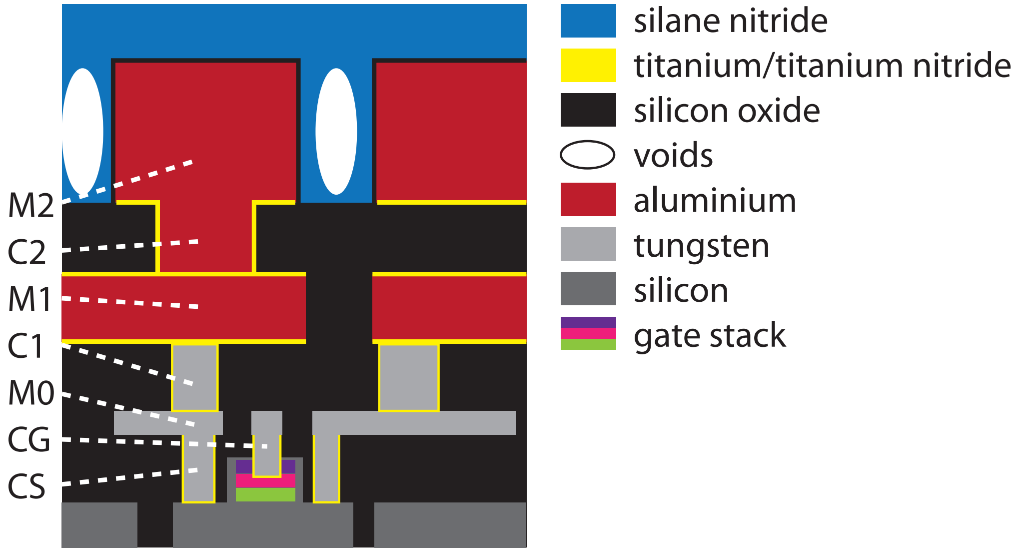

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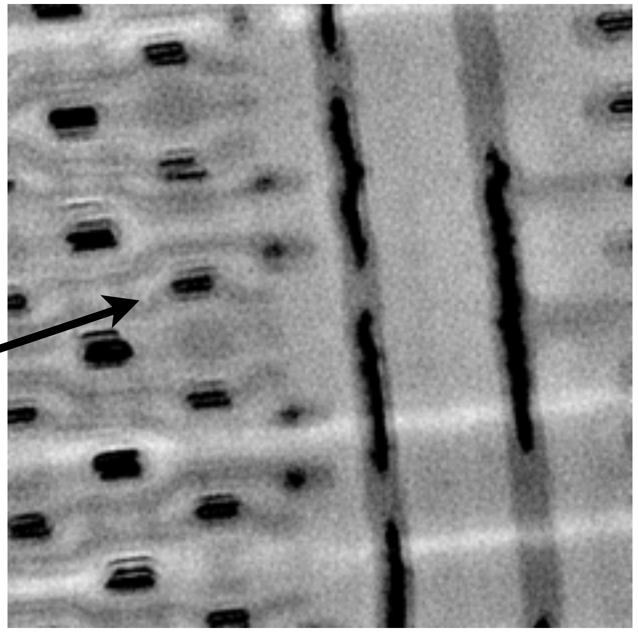

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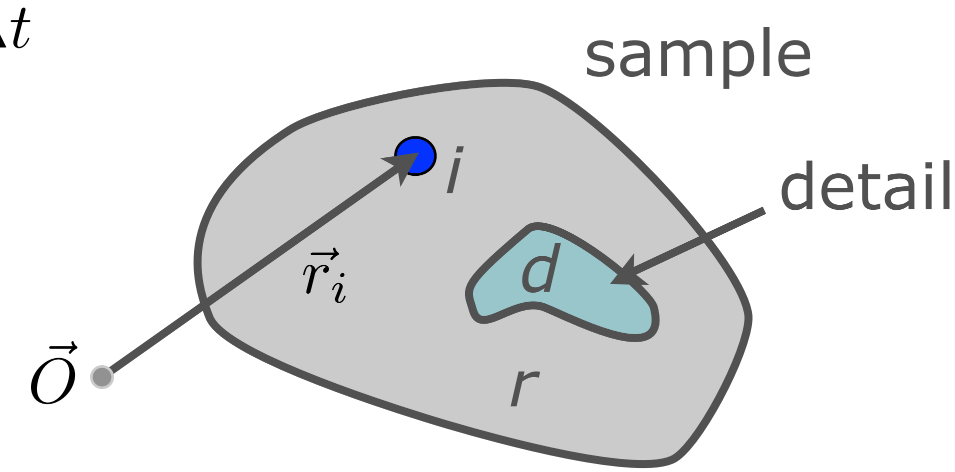
Signal From Sample vs. Signal From Detail

Signal in a given detector pixel:

$$P(\vec{q}) = F(\vec{q}) \cdot \Delta t = I_c \int_{\Delta\Omega} |\psi_d + \psi_r|^2 d\Omega \cdot \Delta t$$

$$= I_c \int_{\Delta\Omega} \left(|\psi_d|^2 + 2\Re(\psi_d \psi_r) + |\psi_r|^2\right) d\Omega \cdot \Delta t$$

$$\psi_d = -r_0 p(\theta) \sum_{i \in d} f_i(\vec{q}) e^{-i\vec{q}\cdot\vec{r}}$$



I_c : coherent flux density
$\Delta\Omega$: solid angle of detector pixel
Δt : exposure time

Detail feature detectable (necessary condition) if

$2\Re(\psi_d \psi_r)$ (heterodyne)
or
$|\psi_d|^2$ (homodyne)

} $\geq \alpha \times$ larger than shot noise from $|\psi_r|^2$

$\alpha = 5$ (Rose criterion)

Schropp & Schroer, NJP **12**, 035016 (2010).

Signal From Sample vs. Signal From Detail

Signal-to-background considerations:

Feature can at best be detected when it can be imaged by itself!

$$I_c|\psi_d|^2 \Delta\Omega_d \cdot \Delta t \geq \frac{\alpha^2}{4}$$

necessary condition!

$\Delta\Omega_d$: size of Shannon pixel for given feature

scattering amplitude of feature:

$$\psi_d = -r_0 p(\theta) \sum_{i \in d} f_i(\vec{q}) e^{-i\vec{q}\cdot\vec{r}}$$

Scattering cross section of feature:

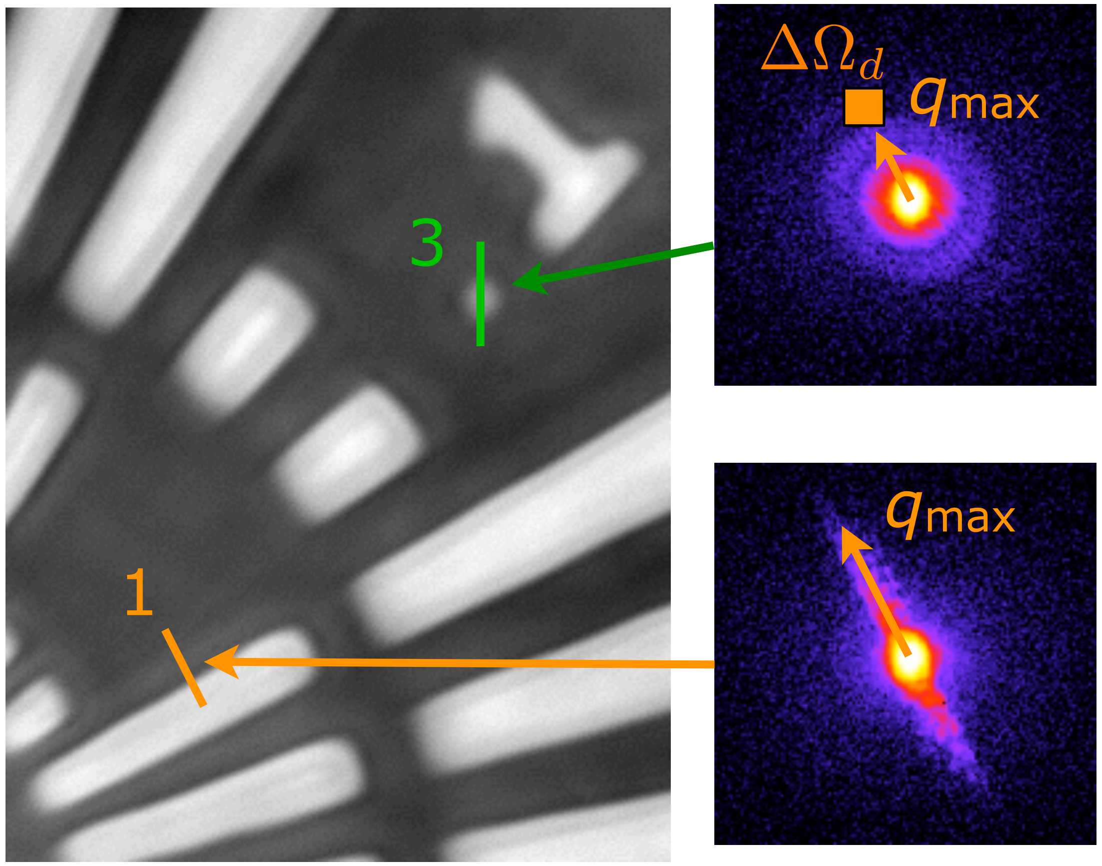
$$\left(\frac{d\sigma}{d\Omega}\right)_d = |\psi_d|^2$$

Schropp & Schroer, NJP **12**, 035016 (2010).

Detecting a feature inside an object

$$\underbrace{I_c \Delta t}\cdot \underbrace{|\psi_d|^2 \Delta\Omega_d} \geq \frac{\alpha^2}{4}$$

~ structure factor of feature:



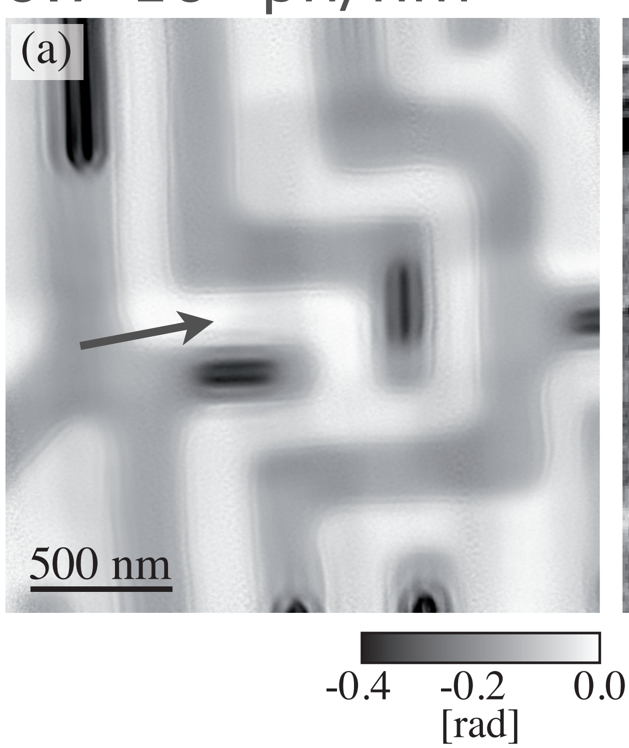
A. Schropp, et al., APL **100**, 253112 (2012).

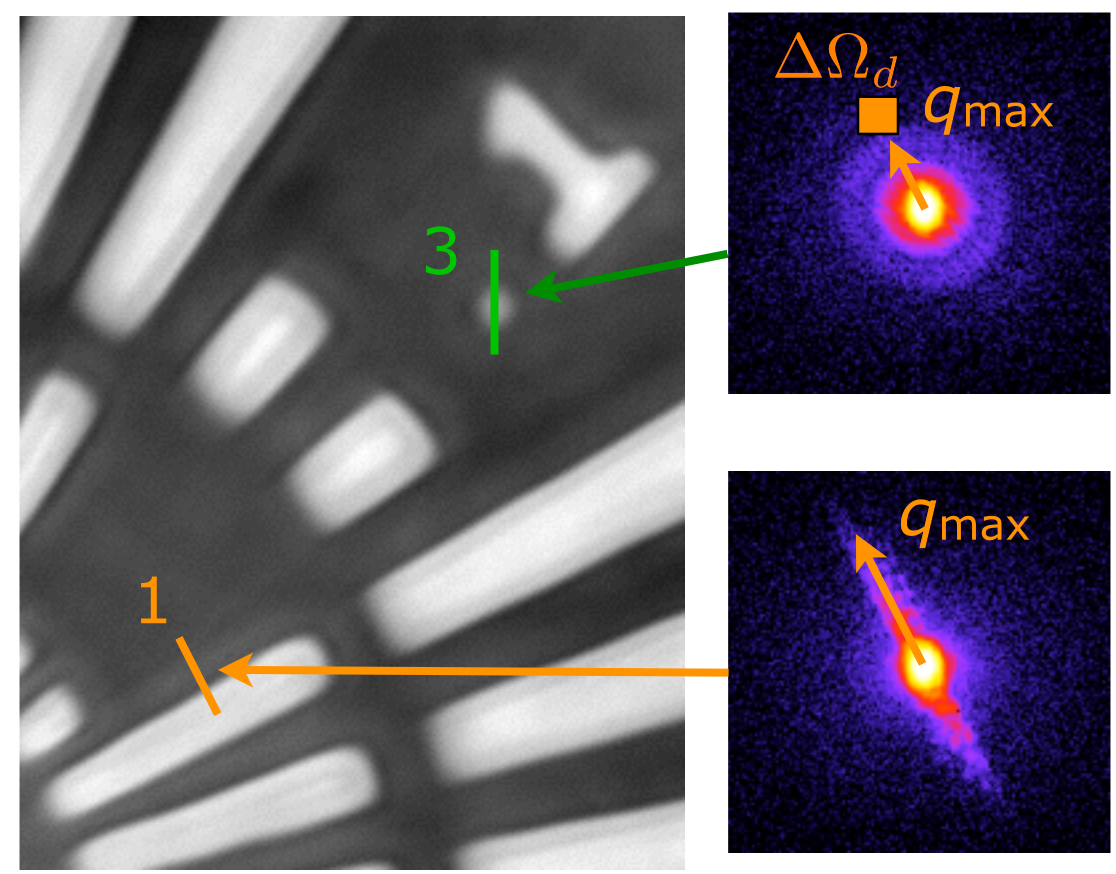
Detecting a feature inside an object

$$\underbrace{I_c \Delta t}\cdot \underbrace{|\psi_d|^2 \Delta\Omega_d} \geq \frac{\alpha^2}{4}$$

coherent fluence on feature: ~ structure factor of feature:

$6.7 \cdot 10^3$ ph/nm^2 $3.9 \cdot 10^2$ ph/nm^2





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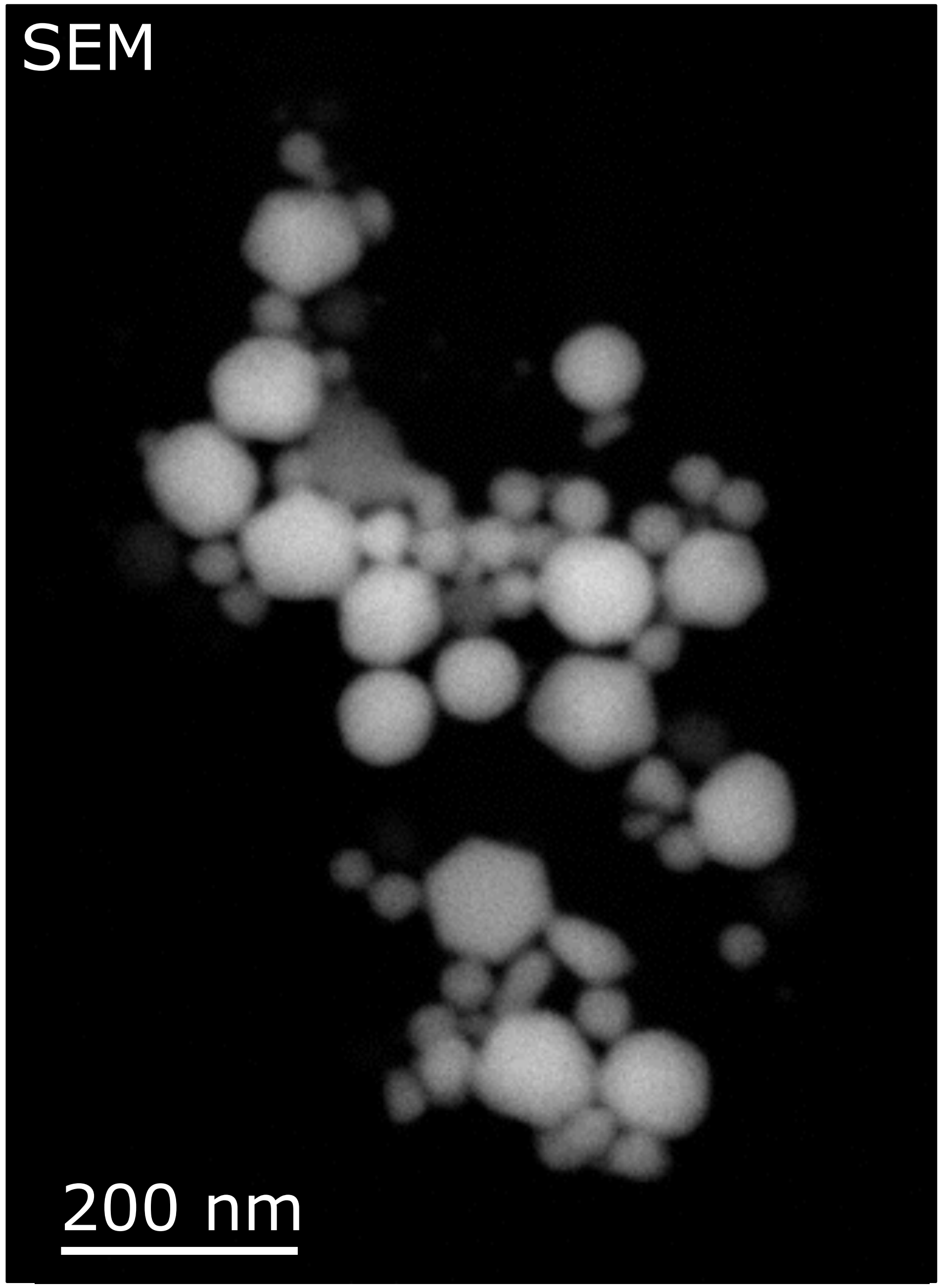
Ptychography: Sensitivity & Resolution

Sample: Pd, Pt, and Au particles



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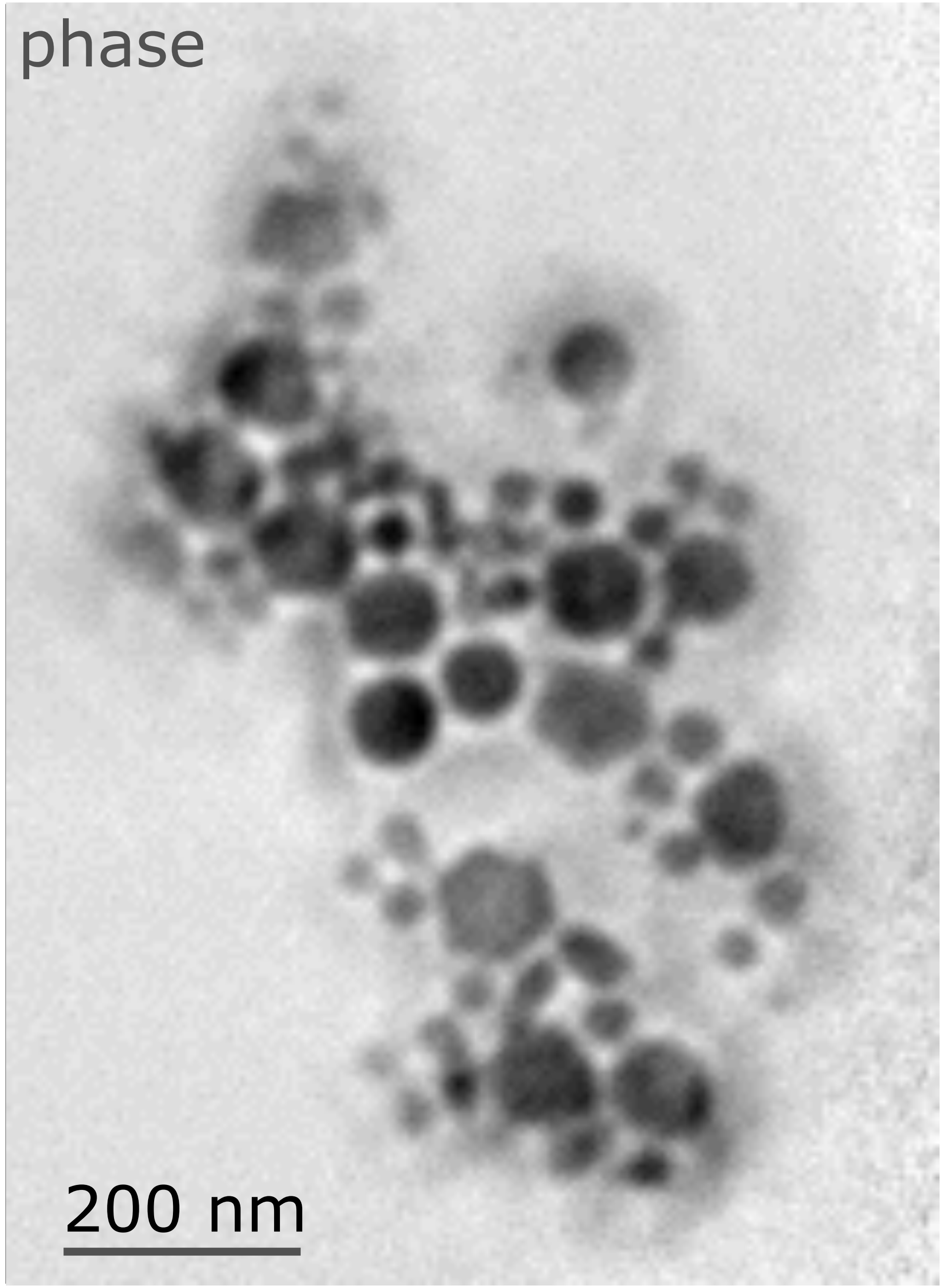

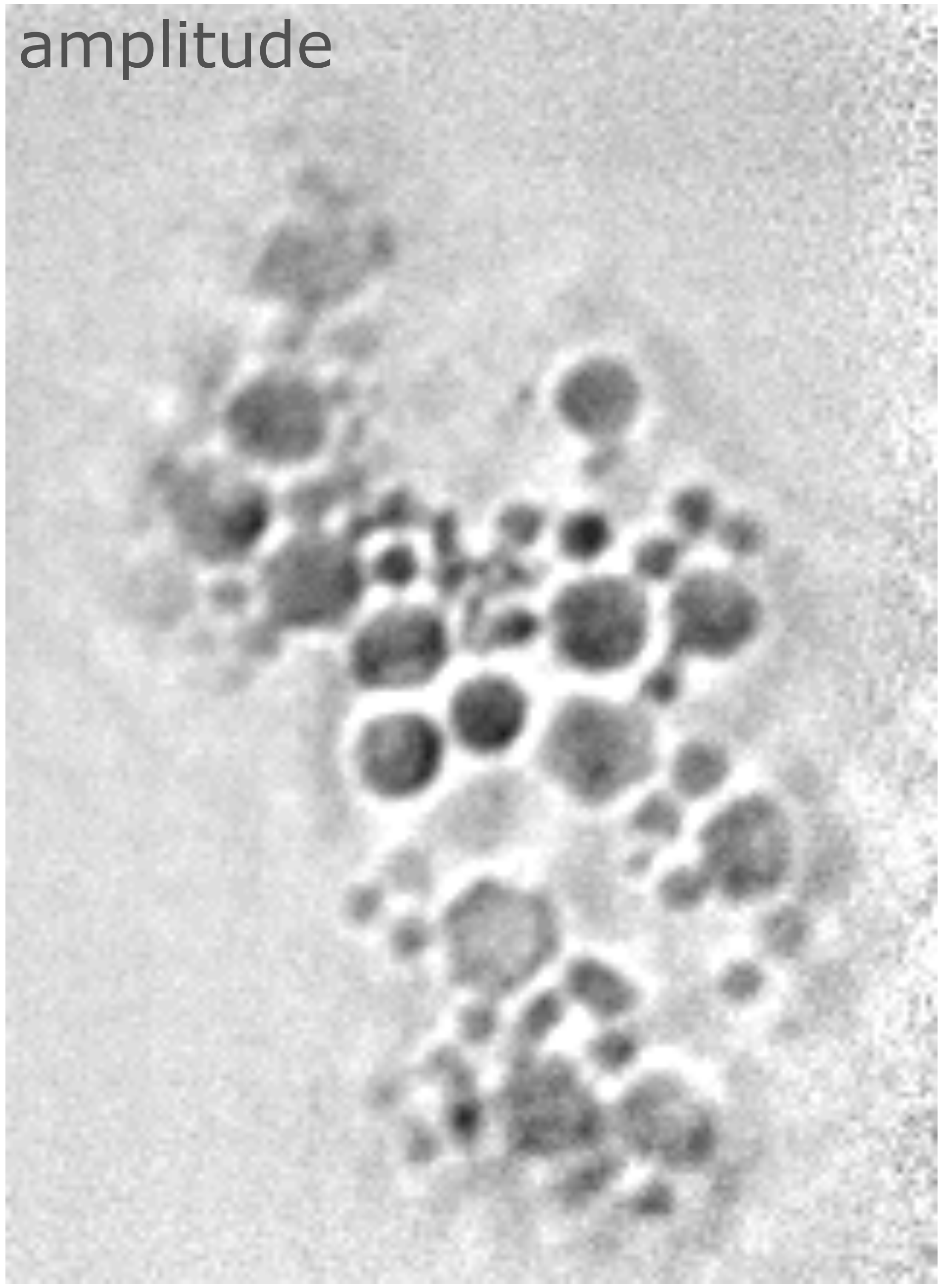

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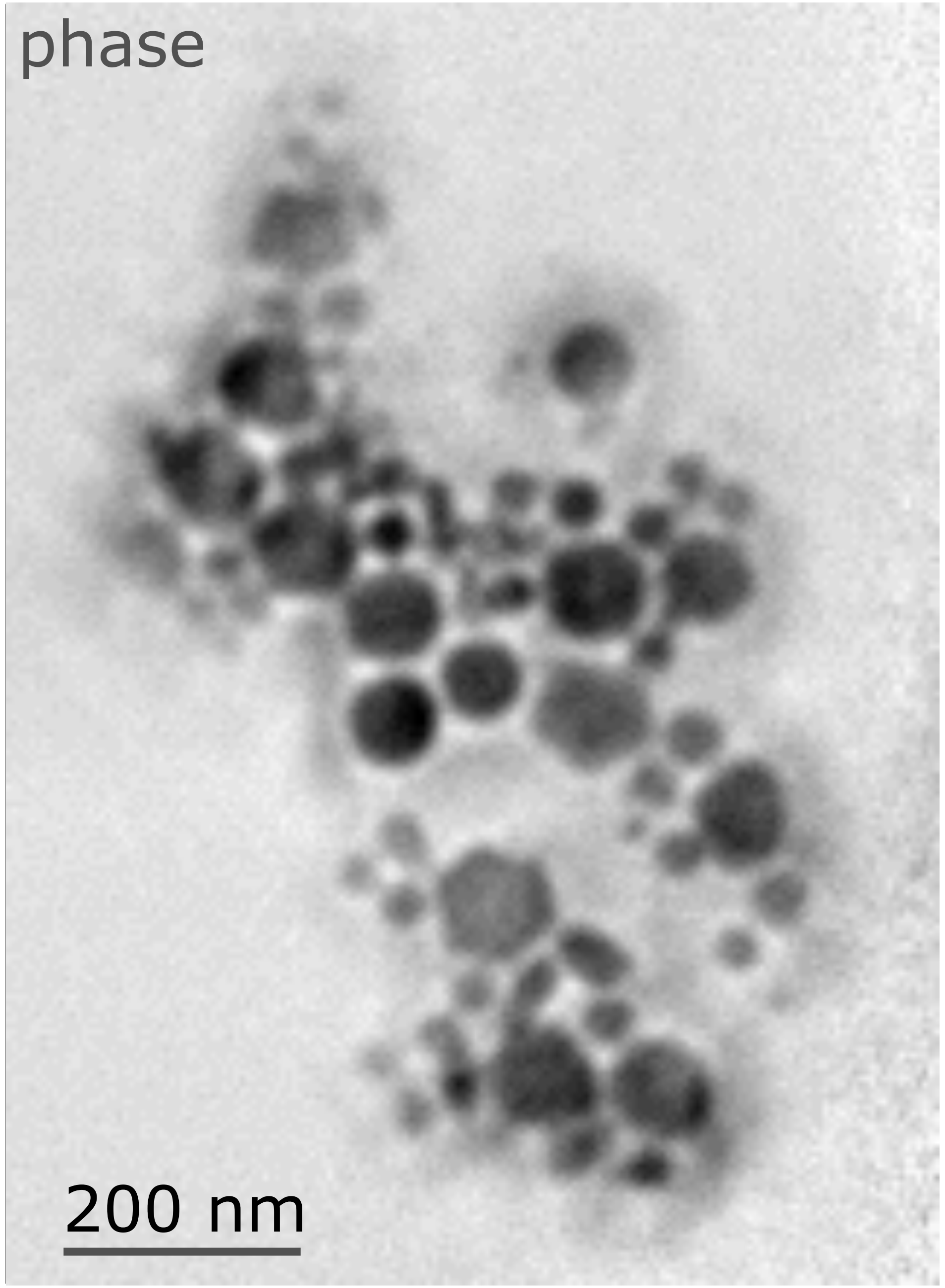

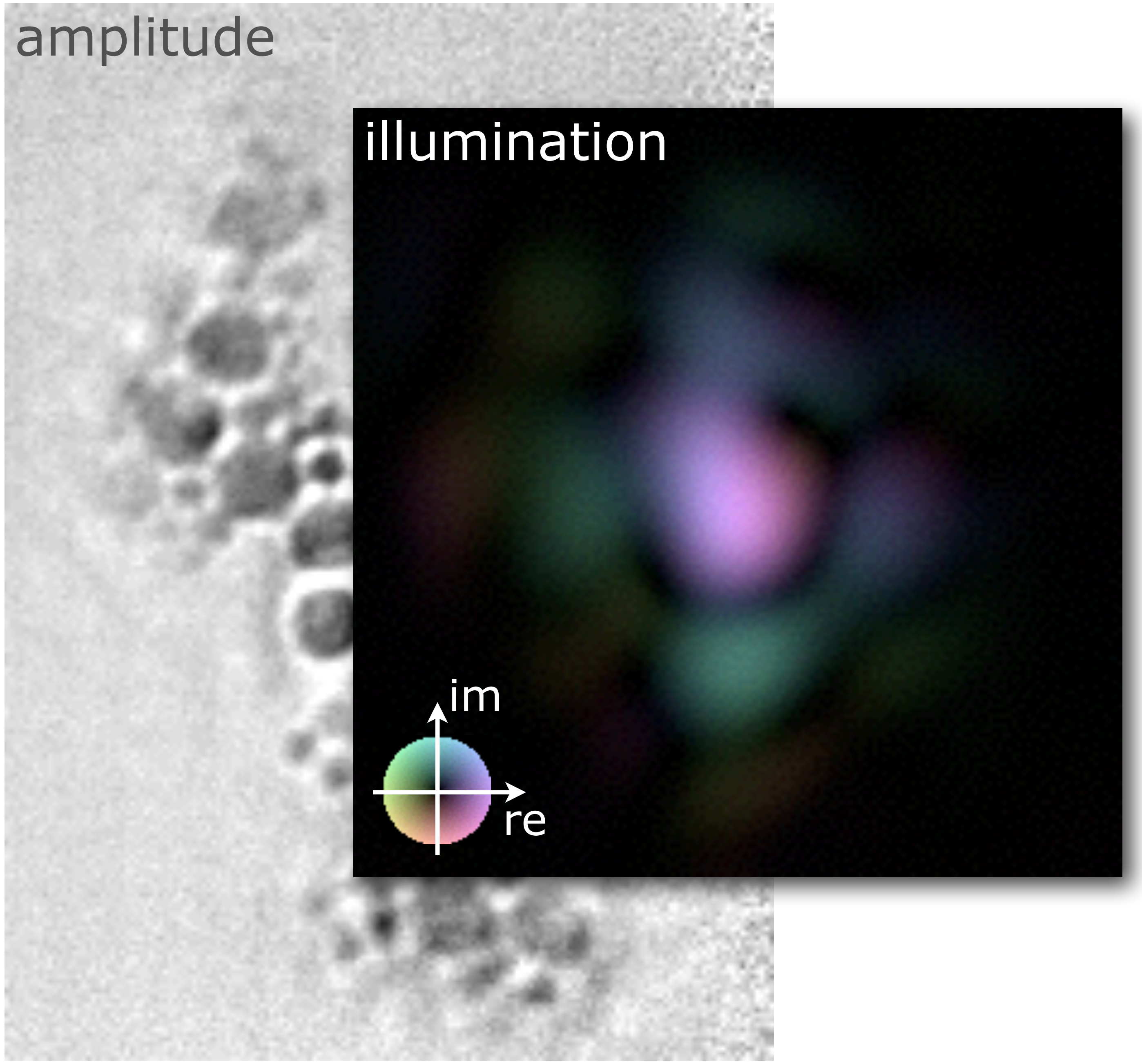

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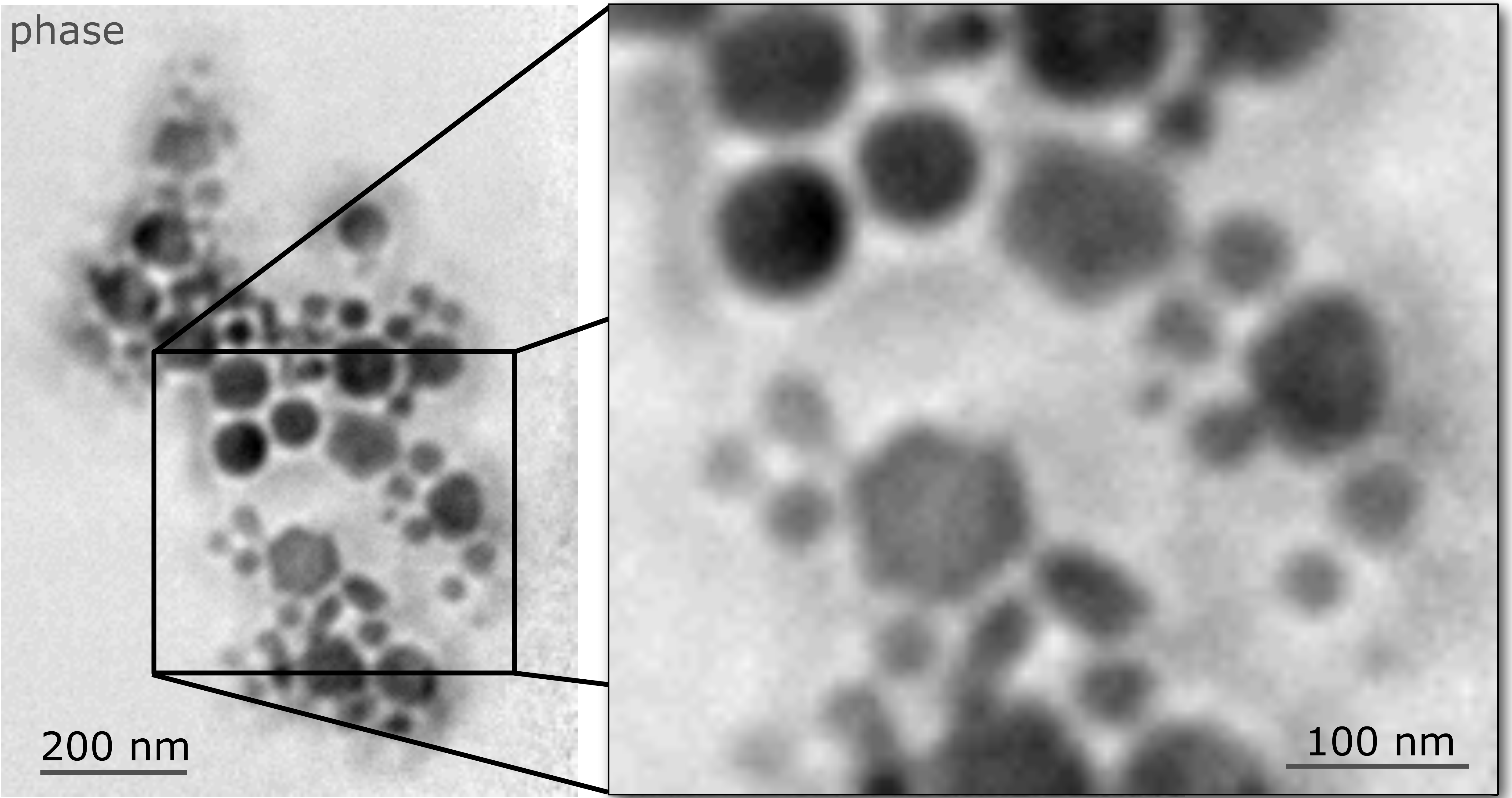

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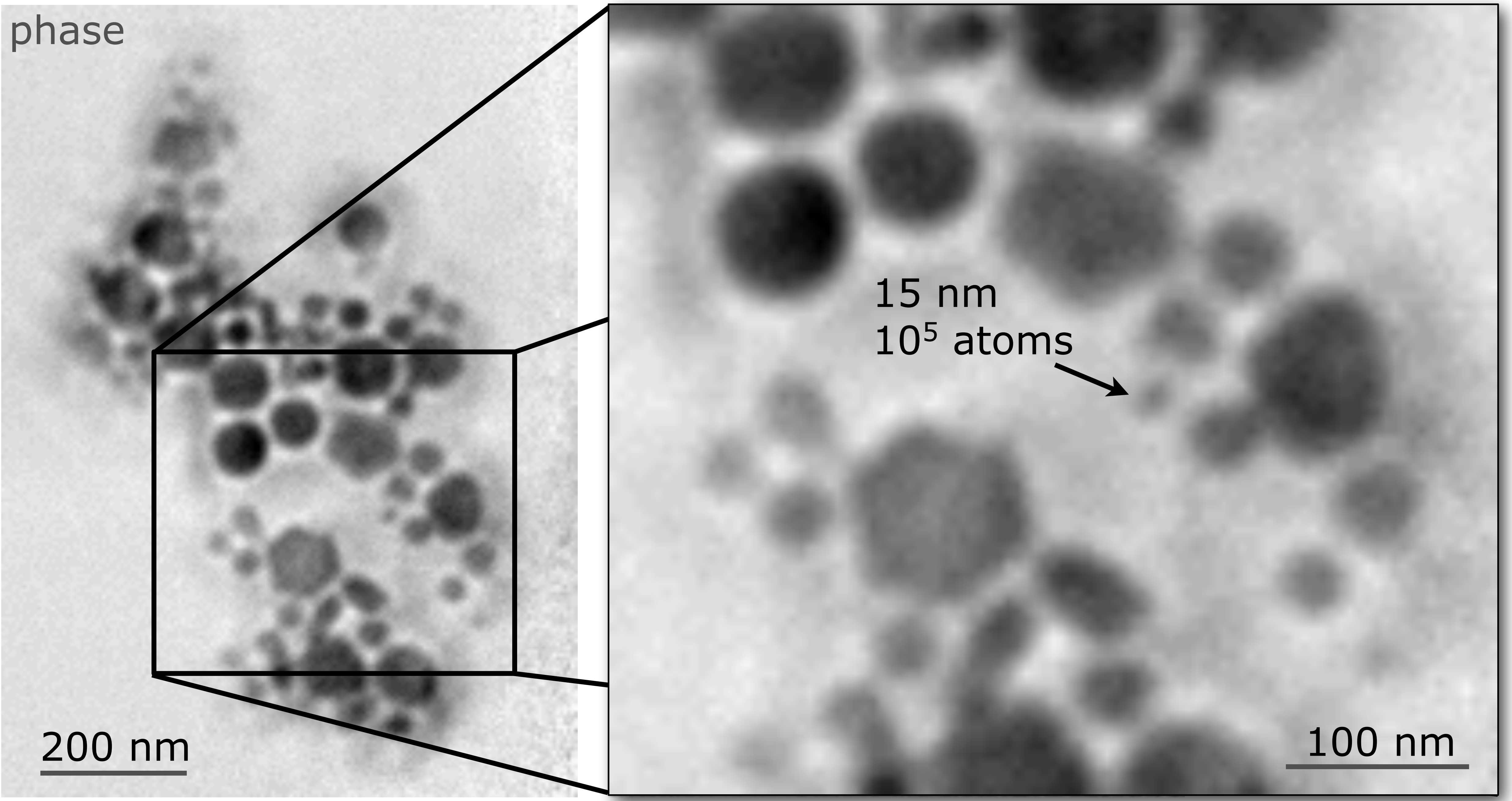
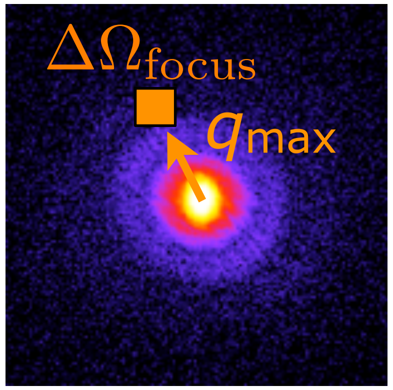


Locating a Feature at High Spatial Resolution

Signal from small feature in given speckle at high $q_{\max}$:



$$I_{\mathrm{c}} \cdot t \cdot \frac{d\sigma}{d\Omega}(\vec{q}_{\mathrm{max}}) \cdot \Delta\Omega_{\mathrm{focus}} \geq \frac{\alpha^2}{4}$$

- $I_{\mathrm{c}} \cdot t$: coherent fluence on feature
- $\frac{d\sigma}{d\Omega}(\vec{q}_{\mathrm{max}})$: scattering cross section of feature at highest q
- $\Delta\Omega_{\mathrm{focus}}$: Speckle size defining a piece of information in reciprocal space

Figure of merit of x-ray microscope:

$I_{\mathrm{c}} \cdot t \cdot \Delta\Omega_{\mathrm{focus}}$ coherent fluence per entropy in diff. pattern

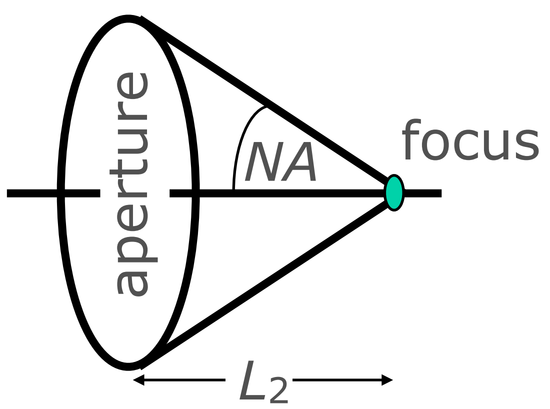
Locating a Feature at High Spatial Resolution

Coherent fluence:

$$I_c \cdot t = \frac{F_c}{A} \cdot T \cdot t \qquad F_c \propto Br \cdot \lambda^2 \cdot \frac{\Delta E}{E} \quad \text{coherent flux}$$

optic's transmission

illuminated area



$$A \propto d_t^2 \propto \frac{\lambda^2}{NA^2}$$

Diffraction limited focus:

$$I_c \cdot t \propto Br \cdot \frac{\Delta E}{E} \cdot \underbrace{NA^2 \cdot T}_{} \cdot t$$

source

optic

Size of speckle in diffraction pattern:

$$\Delta\Omega_{\text{focus}} = \pi \frac{\lambda^2}{4d_t^2} = \pi NA^2$$

optic

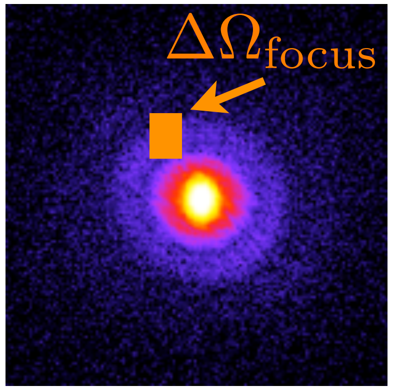


Figure of Merit of X-Ray Microscope

Signal from small feature in given speckle at high $q_{\max}$:

$$I_{\mathrm{c}} \cdot t \cdot \frac{d\sigma}{d\Omega}(\vec{q}_{\max}) \cdot \Delta\Omega_{\mathrm{focus}} \propto \underbrace{Br}_{\text{source}} \cdot \frac{\Delta E}{E} \cdot \underbrace{NA^4 \cdot T}_{\text{optic}} \cdot t$$

Improve scanning coherent diffraction microscope:

- upgrade the source:

  ⟶ diffraction limited storage ring

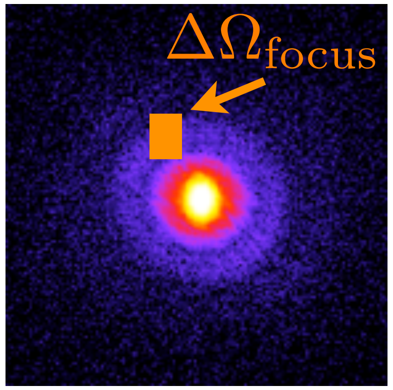
- improve optic:

  ⟶ $\propto NA^4$ strong dependence on numerical aperture!
  ⟶ transmission

Atomic Resolution?

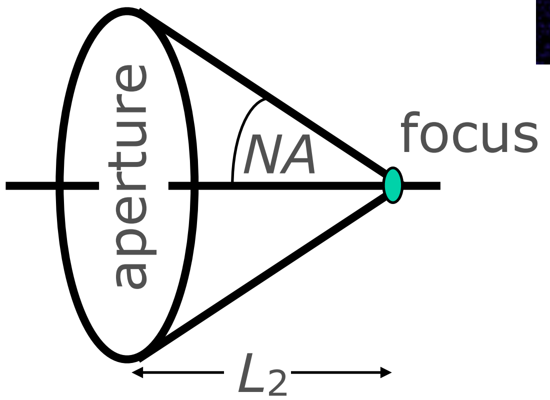
Have sufficiently many scattered photons in speckle at large q!

Size of speckle in diffraction pattern:



$$\Delta\Omega_{\text{focus}} = \pi \frac{\lambda^2}{4 d_t^2} = \pi NA^2$$

NA: numerical aperture of optic



given example:

$$\Delta\Omega_{\text{focus}} \approx 10^{-6} \text{ srad}$$

elastic scattering of gold atom to 1 Å:

$$I_c \frac{d\sigma}{d\Omega} t = 0.4 \cdot 10^{-3} \text{ srad}^{-1}$$

Current experiment:

$$I_c \cdot t \cdot \frac{d\sigma}{d\Omega} \Delta\Omega_{\text{focus}} \approx 4 \cdot 10^{-10}$$

is about 10 orders of magnitude off!!!

Route to Atomic Resolution...

Coherent fluence:

$$I_c \cdot t = \frac{F_c}{A} \cdot T \cdot t \qquad F_c \propto Br \cdot \lambda^2 \cdot \frac{\Delta E}{E}$$ coherent flux

A: illuminated area; T: optic's transmission

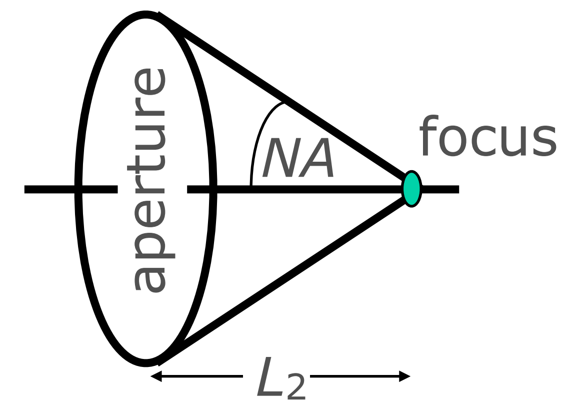
Ways to increase fluence:

- increase brilliance *Br*
- decrease illuminated area *A*
- improve transmission *T* of optic
- longer exposure *t*

Diffraction limited focus:

$$I_c \cdot t \propto Br \cdot \frac{\Delta E}{E} \cdot \underbrace{NA^2 \cdot T}_{\text{optic}} \cdot t$$

Br: source



$$A \propto d_t^2 \propto \frac{\lambda^2}{NA^2}$$

T: transmission of optic

Route to Atomic Resolution...

Signal in diffraction pattern:

$$I_c \cdot t \cdot \frac{d\sigma}{d\Omega} \cdot \Delta\Omega_{\text{focus}} \propto \underbrace{Br}_{\text{source}} \cdot \frac{\Delta E}{E} \cdot \underbrace{NA^4 \cdot T}_{\text{optic}} \cdot t$$

speckle size:

$$\Delta\Omega_{\text{focus}} = \pi \frac{\lambda^2}{4d_t^2} = \pi NA^2$$

fluence:

$$I_c \cdot t \propto Br \cdot \frac{\Delta E}{E} \cdot NA^2 \cdot T \cdot t$$

Improvements (compared to current experiment):

- upgraded sources (DLSR):

  $Br \to \times 40 - \times 1000$ — $10 - 10^3$

- improve optic:

  $T \to \times(7 \cdot 100)$ — $10 - 10^3$

  (7: efficiency; 100: relaxing spatial filtering of source)

  $NA \to \times 10$ (10 nm focus) — 10^4

overall improvement: 10^{10}

Route to Atomic Resolution...

atomic resolution not completely out of reach!

- requires diffraction limited storage ring
  increase in brilliance & matching of coherence length
  (Thibault, Menzel, Nature **494**, 68 (2013))
- significant gain by extreme focusing

$$\propto NA^4$$

Here: optical aspects covered!

Other issues:

- **modification/destruction** of the sample by imaging
- stability of mechanics & beam
- background (e. g., Compton scattering)

Holy Grail in X-Ray Microscopy

In-situ quantitative measurement of physical properties of matter on

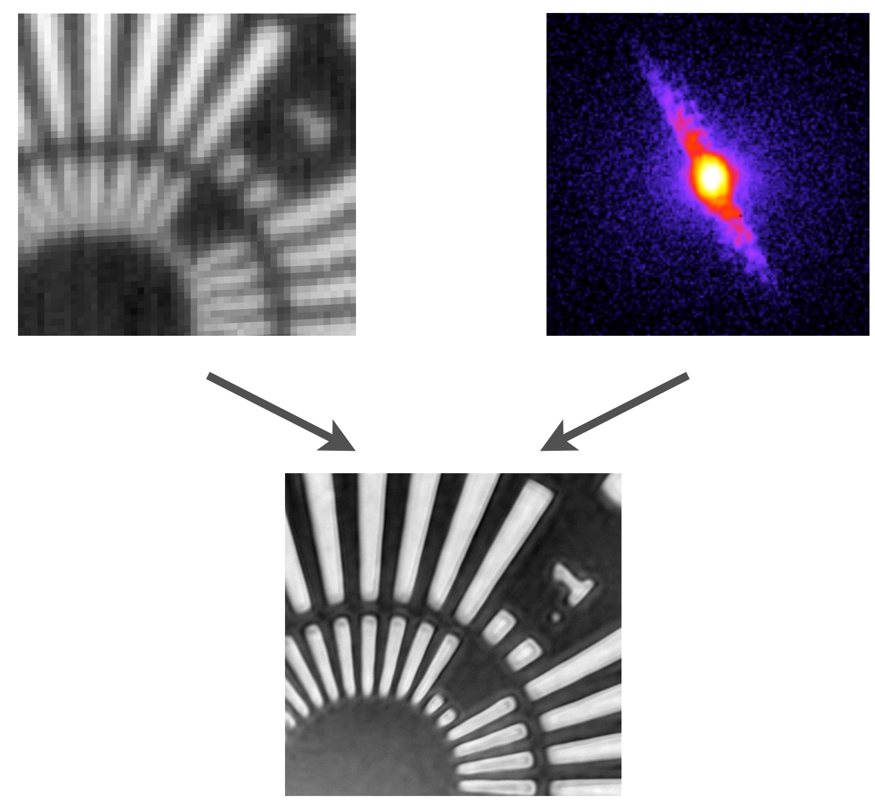
→ all relevant length scales and

→ all relevant time scales!

Key technology: brilliant coherent x-rays with time structure

Fusion of real and reciprocal space:

speckle size matches
real space resolution



→ access to all length scales:

(in principle) from Å to millimeters