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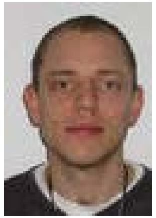
Ana Diaz :: Beamline Scientist :: Paul Scherrer Institut

X-ray Ptychography: a powerful tool for imaging

Workshop on coherence at ESRF-EBS, Grenoble, 9th September 2019

PAUL SCHERRER INSTITUT PSI

The Coherent X-ray Scattering Group


Andreas Menzel


Ana Diaz


Manuel Guizar-Sicairos

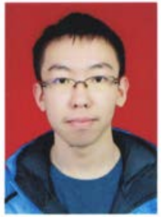

Mirko Holler


Xavier Donath


Johannes Ihli


Klaus Wakonig


Mariana Verezhak


Zirui Gao


Dmitry Karpov

Alumni

- M. Odstrcil
- V. Lütz-Bueno
- E. Tsai
- M. Liebi
- I. Rajkovic
- R. Jacob
- J. da Silva
- J. Han
- O. Bunk
- H. Deyhle
- T. Ikonen
- C. Kewish
- F. Pfeiffer
- P. Thibault
- M. Dierolf

Collaborations within PSI

- J. Raabe (SLS, Pollux)
- C. David (LMN)
- G. Tinti (detectors)
- G. Aeppli (SLS)
- S. Shahmoradian (BIO)
- T. Ishikawa (BIO)
- P. Trtik (SINQ)
- E. Müller (BIO)
- Scientific software

External collaborations (PI's)

- T. Sheppard (KIT, Germany)
- R. Wepf (ETH Zürich, Switzerland)
- J. R. Bowen (Technical Univ. Denmark)
- A. Sepe (Uni. Fribourg, Switzerland)
- H. Help (Uni. Helsinki, Finland)

Postdoc position available at cSAXS

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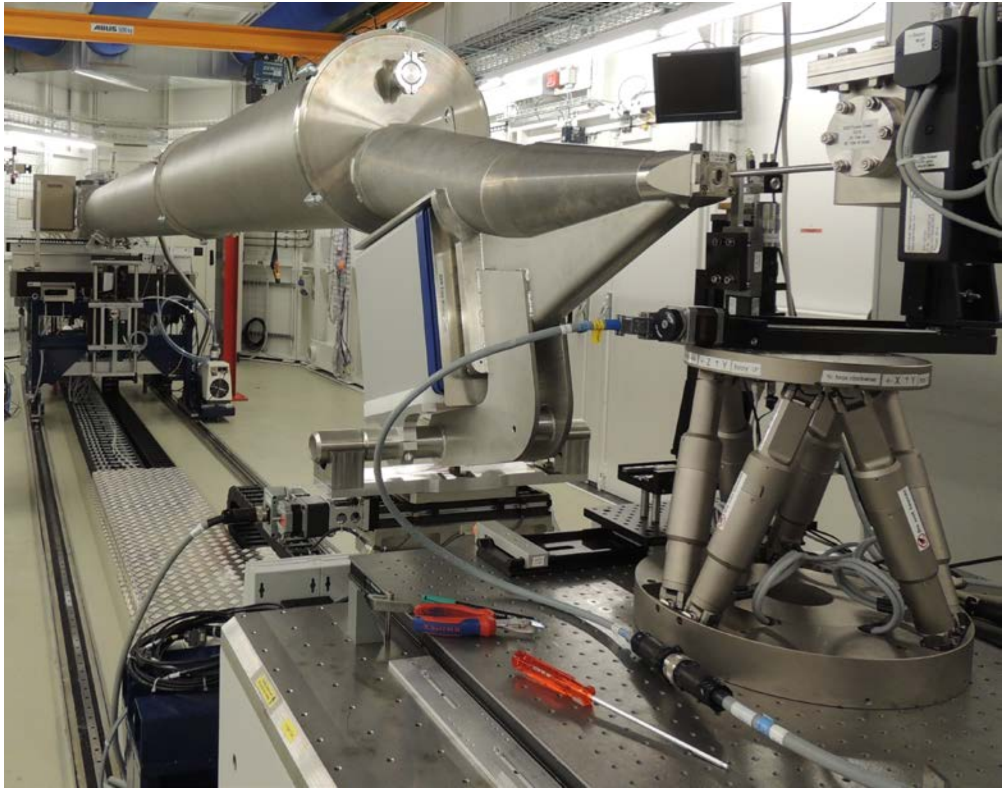
The cSAXS beamline at the Swiss Light Source

cSAXS: coherent small-angle X-ray scattering



Photon energy: 5-18 keV

Main techniques:

- Ptychography
- Spatially resolved SAXS

Exploring Bragg geometry: presentation by Mariana Verezhak tomorrow

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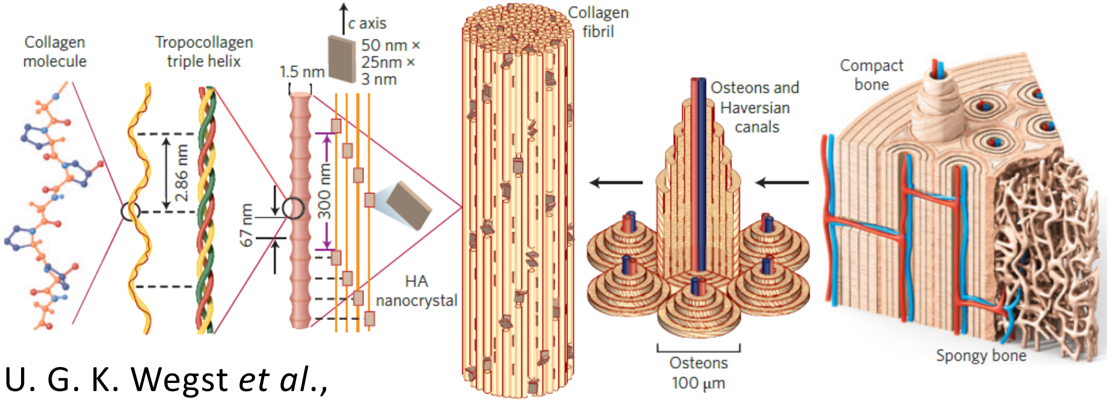
Outline

- Motivation to hard X-ray microscopy
- X-ray ptychography (and tomography)
- Challenges
  - Positioning accuracy
  - Data processing
  - Speed
- Examples:
  - In-situ nanoporous Au coarsening in 2D
  - Ex-situ SOC electrode during full cycle
  - Frozen hydrated biological tissue
- Future improvements

Hard X-ray microscopy



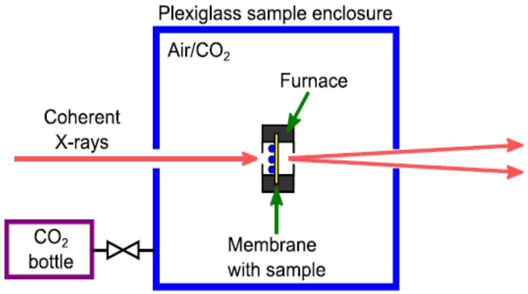
U. G. K. Wegst *et al.*, Nat. Mater. **14** (2014) 23

Hierarchical structures

3D imaging of bulk samples

Thickness from 10 to 100 μm

Resolution from 10 to 100 nm



In-situ reactions in harsh environments

2D imaging of thin samples

Resolution down to 10 nm

CHALLENGES:

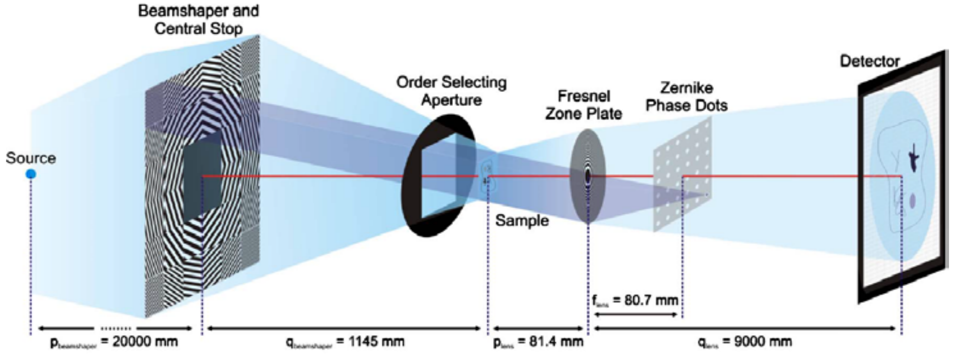
- Fabrication of aberration-free and efficient lenses
- Low absorption contrast

K. Hoydalsvik *et al.*, Appl. Phys. Lett. **104** (2014) 241909

Hard X-ray phase contrast microscopy

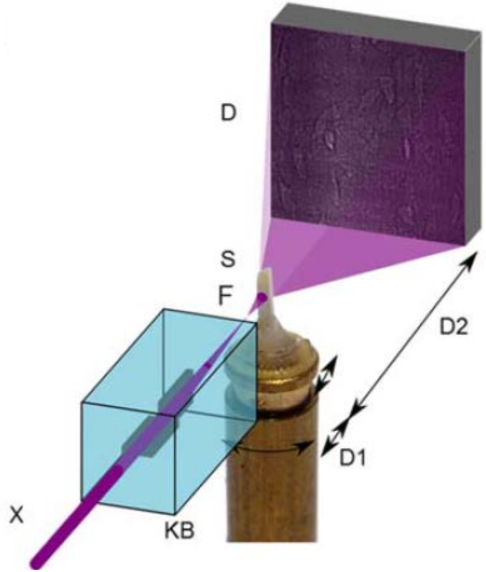
Zernike full-field microscopy @ 10 keV
Objective lens to magnify image
Phase-shifting structure for phase contrast

M. Stampanoni *et al.*,
Phys. Rev. B **81**,140105(R) (2010)



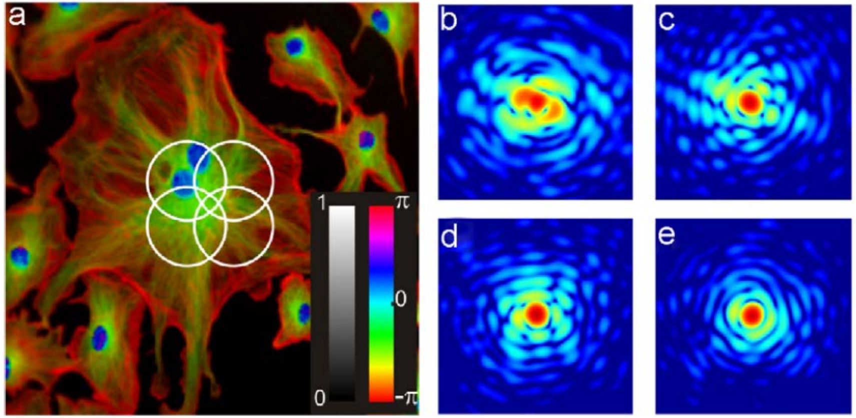
Holo-tomography @ 17 keV
Magnification through divergent beam
Phase contrast by propagation

M. Langer *et al.*, PLOS ONE **7**, e35691 (2012)



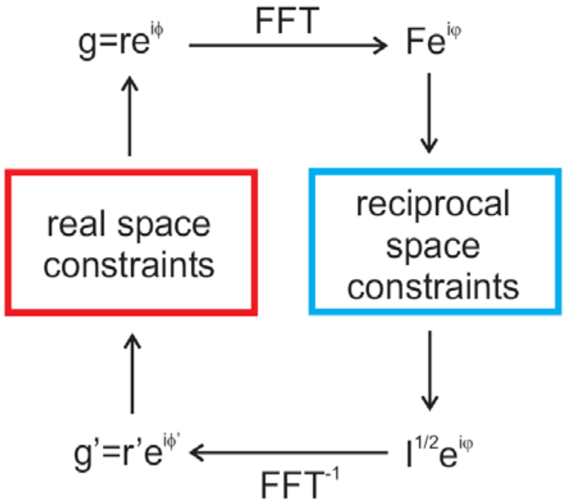
Ptychography

Coherent diffraction patterns from overlapping illuminated areas



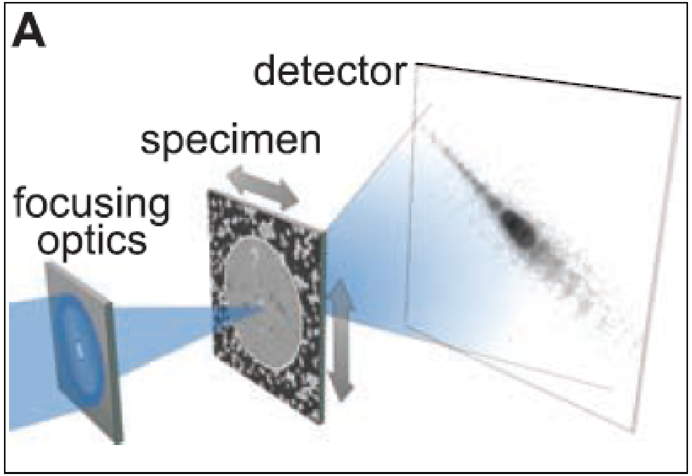
- Absorption and phase contrast
- Resolution not limited by a lens
- In practice limited by mechanical stability and thermal drifts

Iterative phase retrieval algorithms to reconstruct complex-valued transmissivity

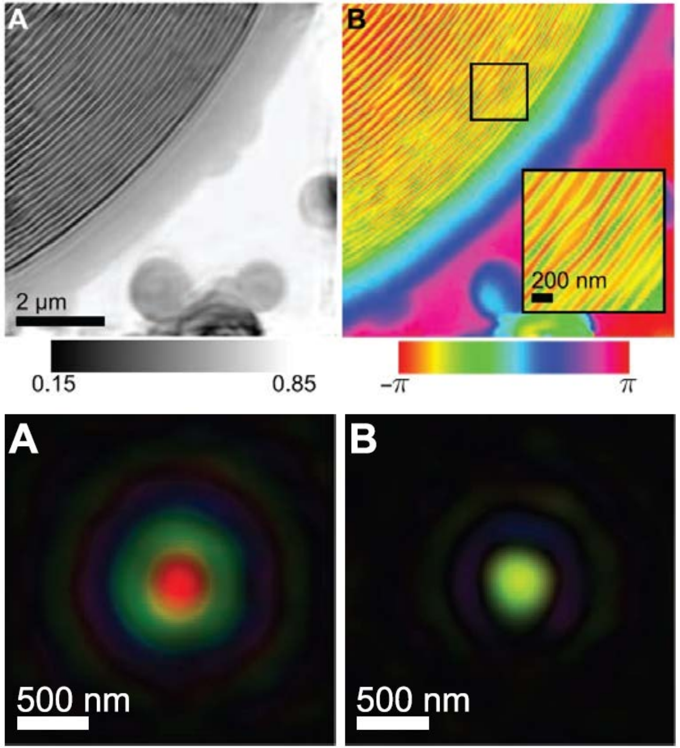


H. M. L. Faulkner & J. M. Rodenburg, Phys. Rev. Lett. **93** (2004) 023903

Ptychography with probe retrieval

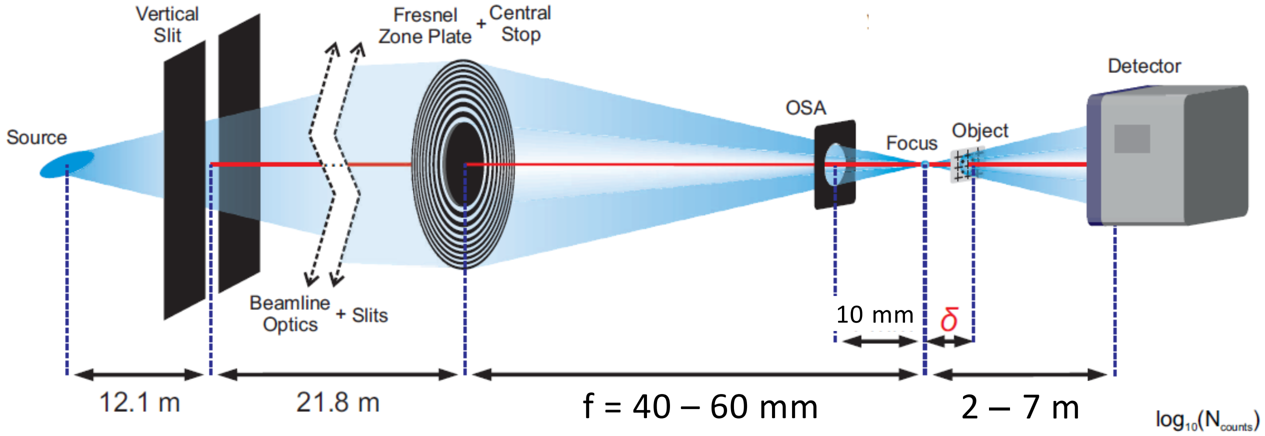


- Enough information to retrieve complex-valued illumination simultaneously
- Effective illumination deconvolution

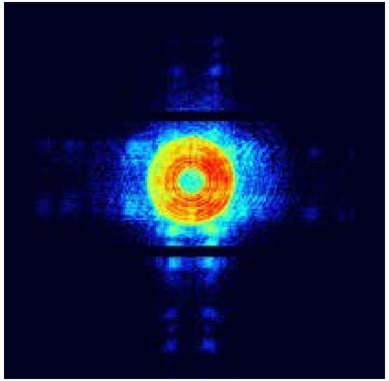


P. Thibault *et al.*, Science **321** (2008) 379

Setting up a 2D ptychographic experiment



J. Vila-Comamala *et al.*, Opt. Express **19** (2011) 21333

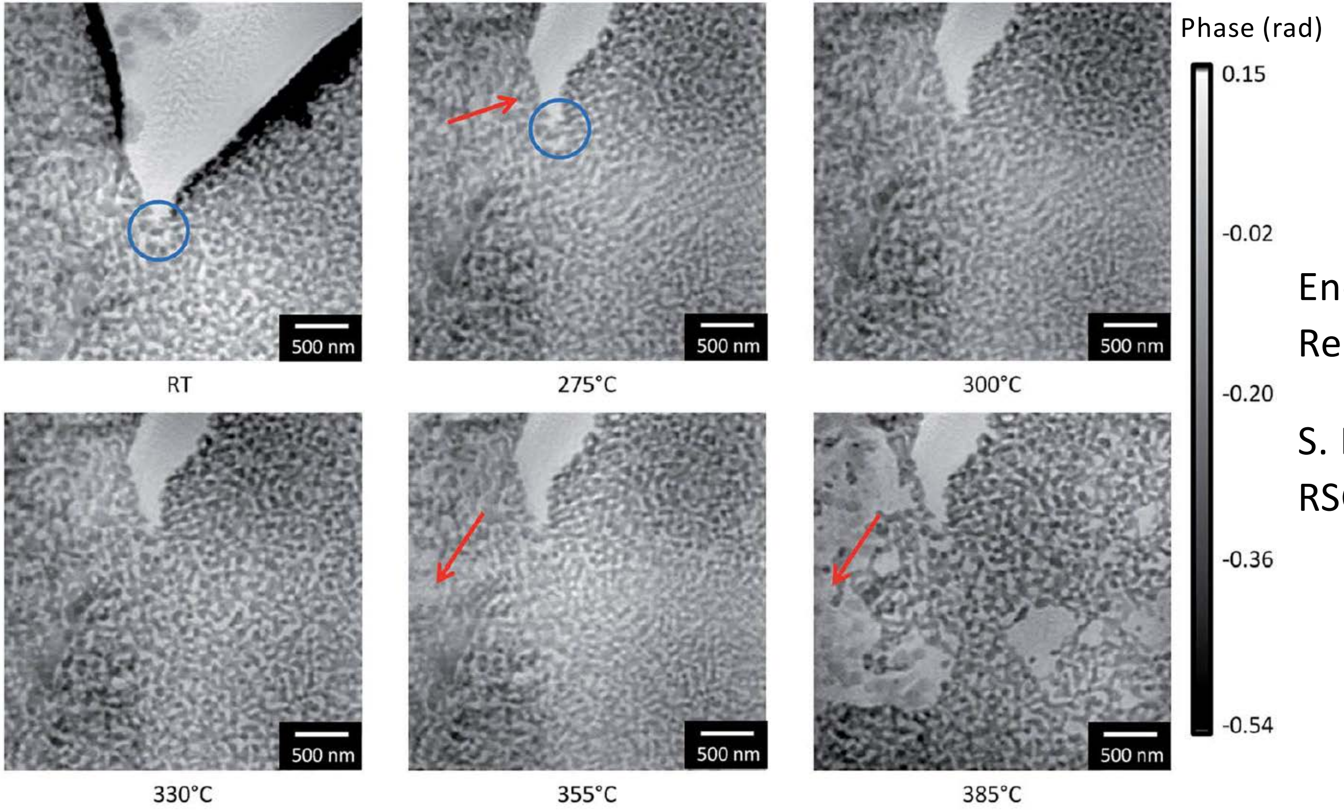


coherent flux:
5×10^8 photons/s
@ 6.2 keV

- Upstream slit defines a small horizontal source for coherent illumination
- Sample scanned by 2D piezo stage
- Sample downstream from focus for efficient scanning
- Large sample-detector distance spreads flux on detector and allows large illumination

Thermally induced coarsening of nanoporous Au

CeO_2/npAu sample, *in situ* heating with a flow of 3 mL/min 20% O_2/He

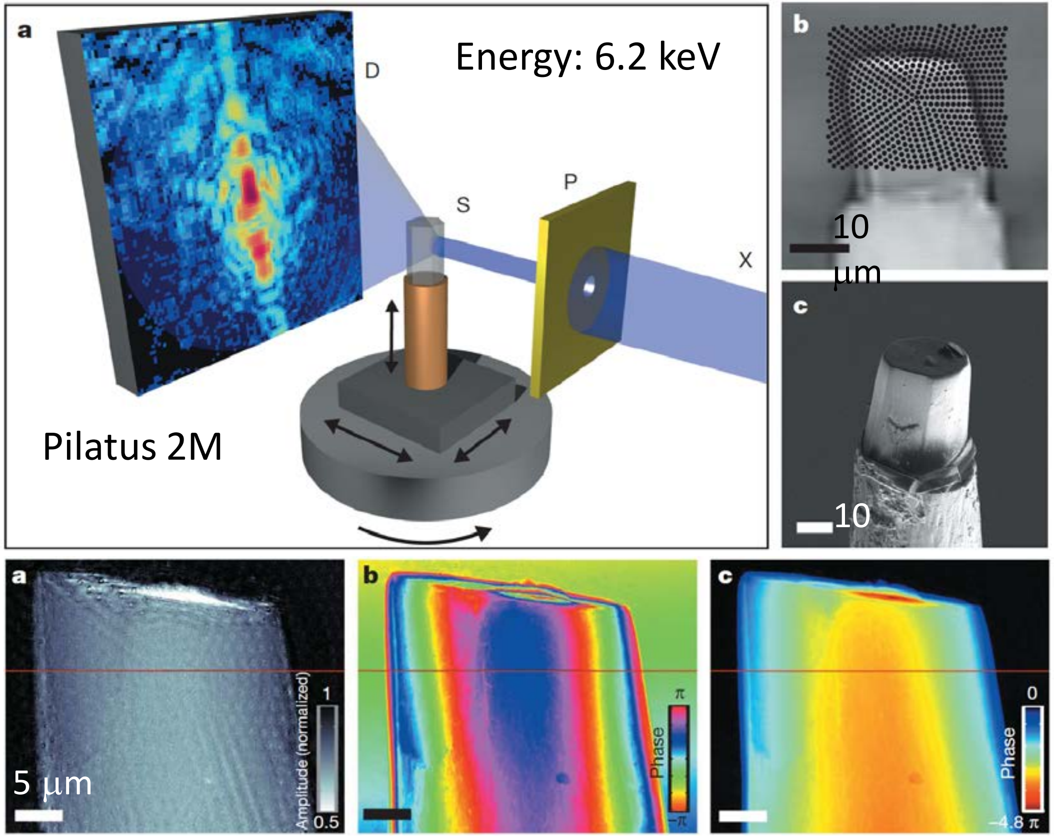


Energy: 5.72 keV

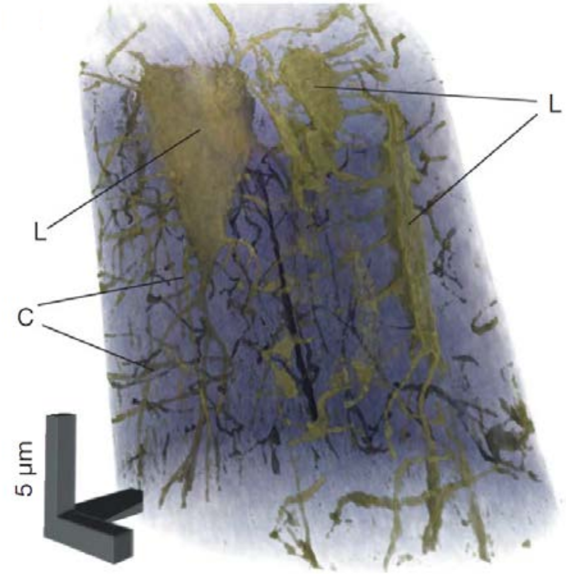
Resolution: ~20 nm

S. Baier *et al.*,
RSC Adv. **6** (2016) 83031

Ptychographic X-ray tomography



M. Dierolf *et al.*, Nature **467**, 436 (2010)



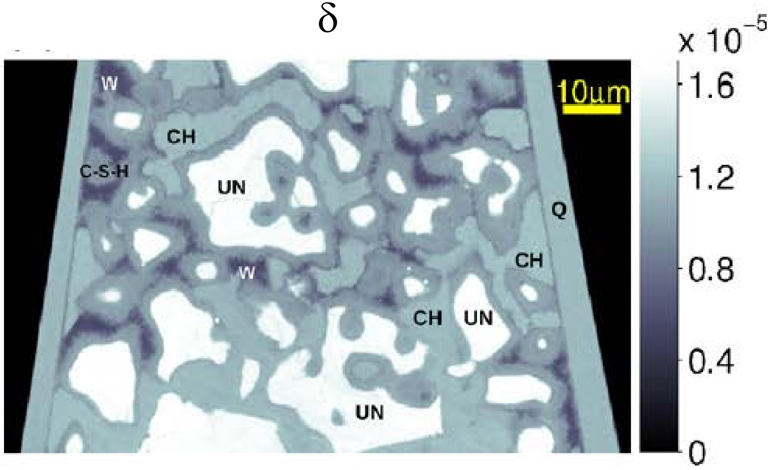
Mouse bone specimen

Voxel size: 65 nm
Resolution: 120 nm
Dose: 2MGy

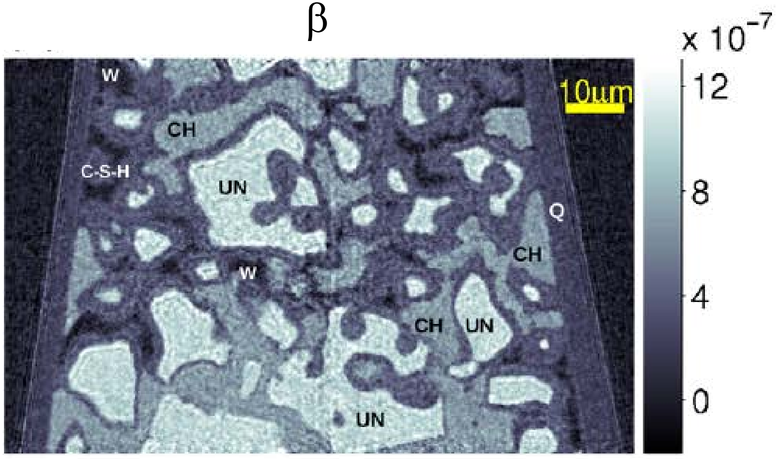
Quantitative contrast

- Hydrated cement phase
- 3D distribution of refractive index: $n(\mathbf{r}) = 1-\delta(\mathbf{r})+i\beta(\mathbf{r})$

δ



β



Identification of material phases:

UN: unhydrated alite particles

W: porosity (mostly water)

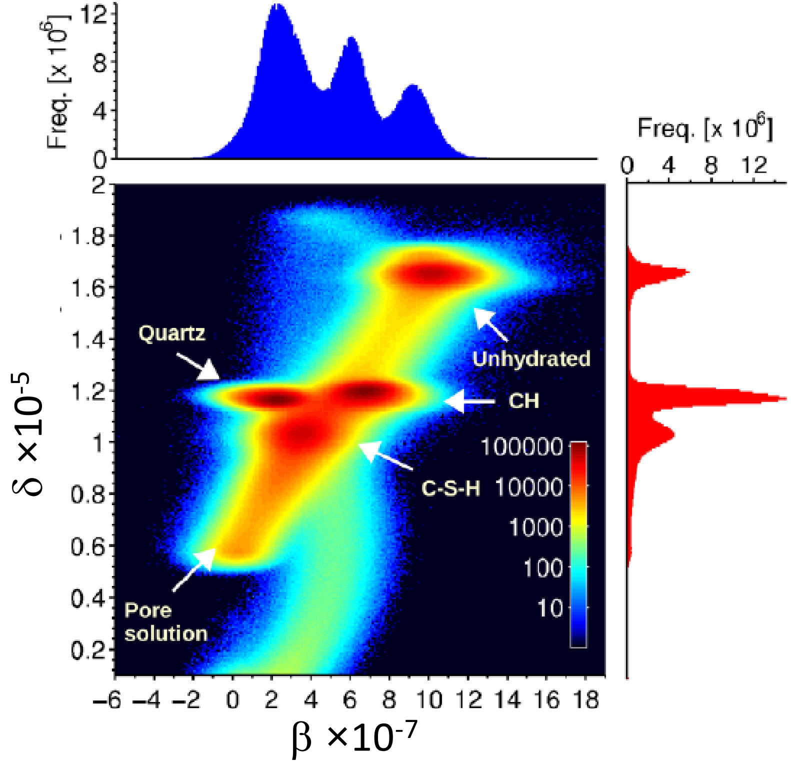
CH: calcium hydroxide

C-S-H: calcium silicate hydrates

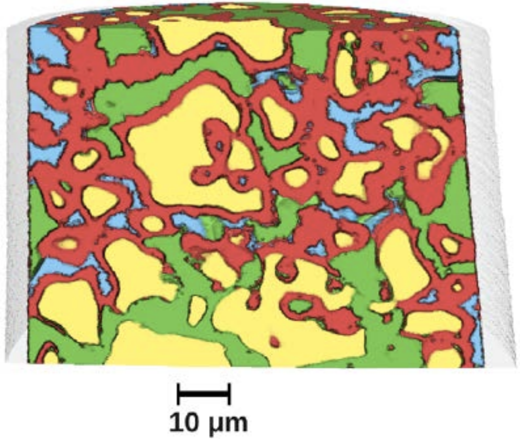
J. C. da Silva *et al.*, Langmuir **31**, 3779 (2015)

Quantitative contrast

Presentation by Miguel Aranda tomorrow



J. C. da Silva *et al.*, Langmuir **31**, 3779 (2015)



UN: unhydrated alite particles

W: porosity (mostly water)

CH: calcium hydroxide

C-S-H: calcium silicate hydrates

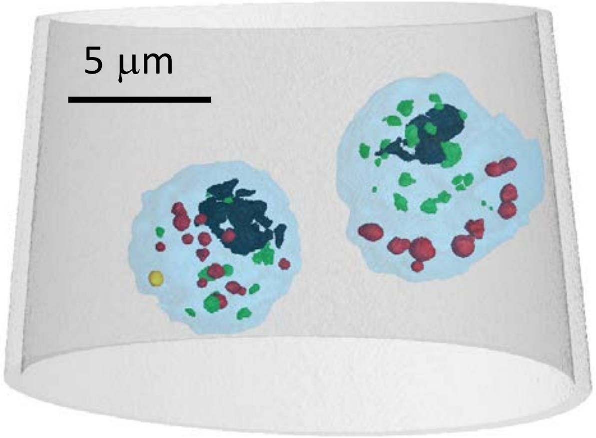
Mass density of C-S-H: 1.828 g/cm^3

Cryo X-ray nanotomography

3D absolute density mapping of intact cells

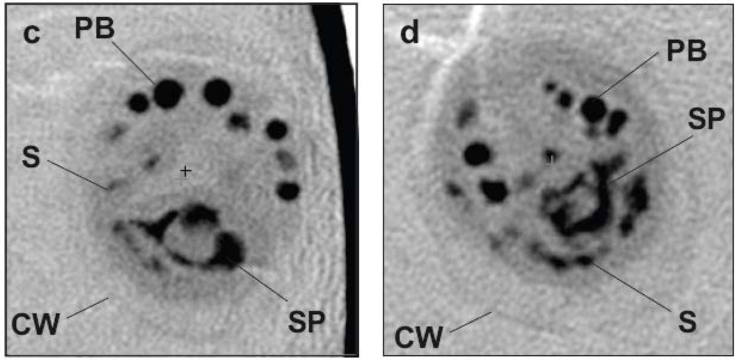
A. Diaz *et al.*, J. Struct. Biol. **192**, 461 (2015)

A. Diaz et al., J. Struct. Biol. **193**, 83 (2016)



Polyphosphate bodies:	**1.56 ± 0.10 g/cm³**
Starch around pyrenoid:	**1.34 ± 0.04 g/cm³**
Other starch granules:	**1.29 ± 0.04 g/cm³**
Cytoplasm:	**1.072 ± 0.012 g/cm³**
Ice matrix:	**0.984 ± 0.010 g/cm³**

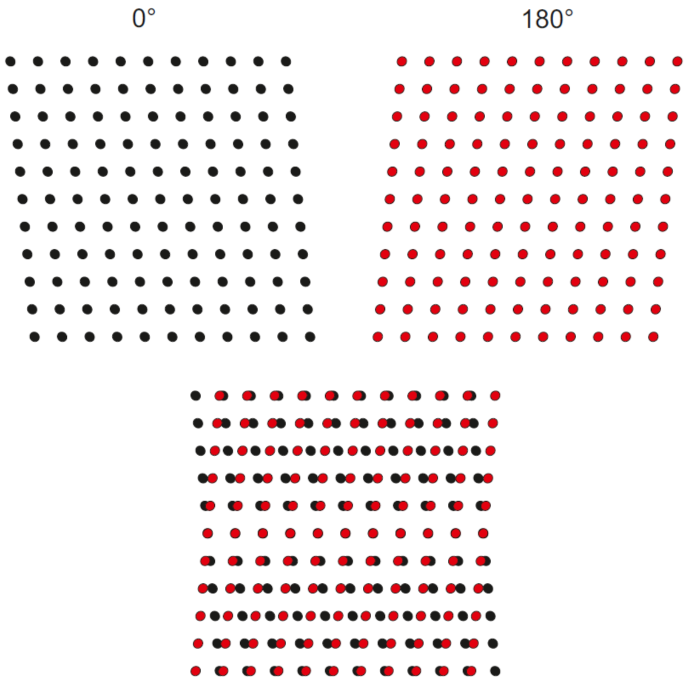
- *Chlamydomonas* unicellular algae
- Solution confined in microcapillary
- Plunge frozen in liquid ethane
- 180 nm resolution limited by thermal drifts in a non-optimized setup



Gray scale:

- quantitative electron density
- conversion to mass density with 6% uncertainty due to high content of H

The challenge of scanning on top of a rotation

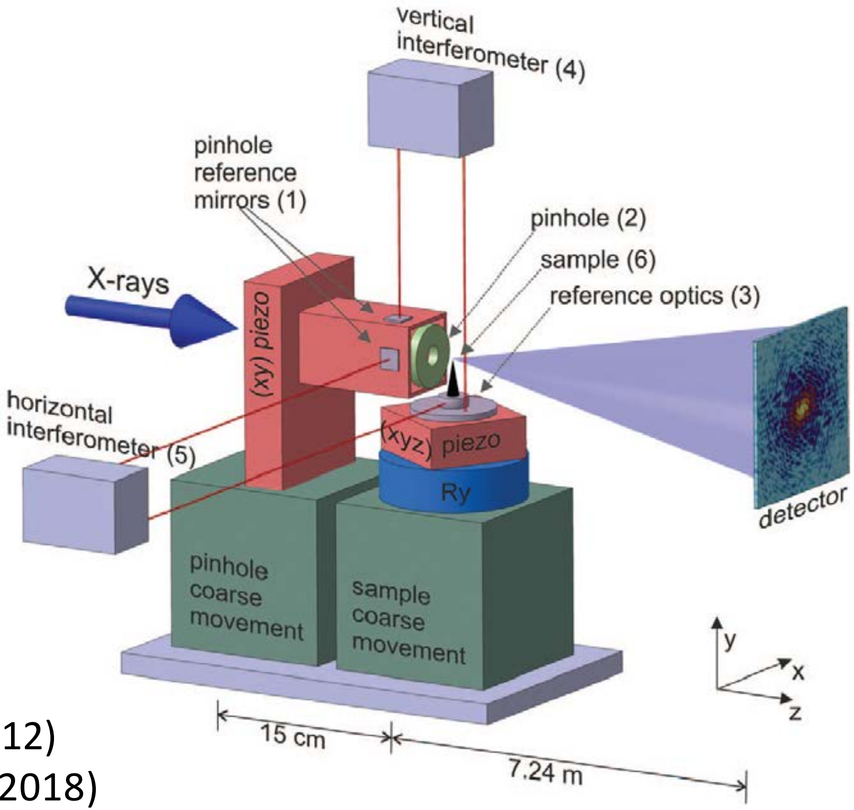


- Piezo scanner error motions and thermal drifts effectively map different positions at different angles
- Distorted positions result in distorted images, also in ptychography
- The 3D resolution is effectively worsen after tomographic reconstruction

Instrumentation for ptychographic tomography

OMNY: tOMography Nano crYo stage

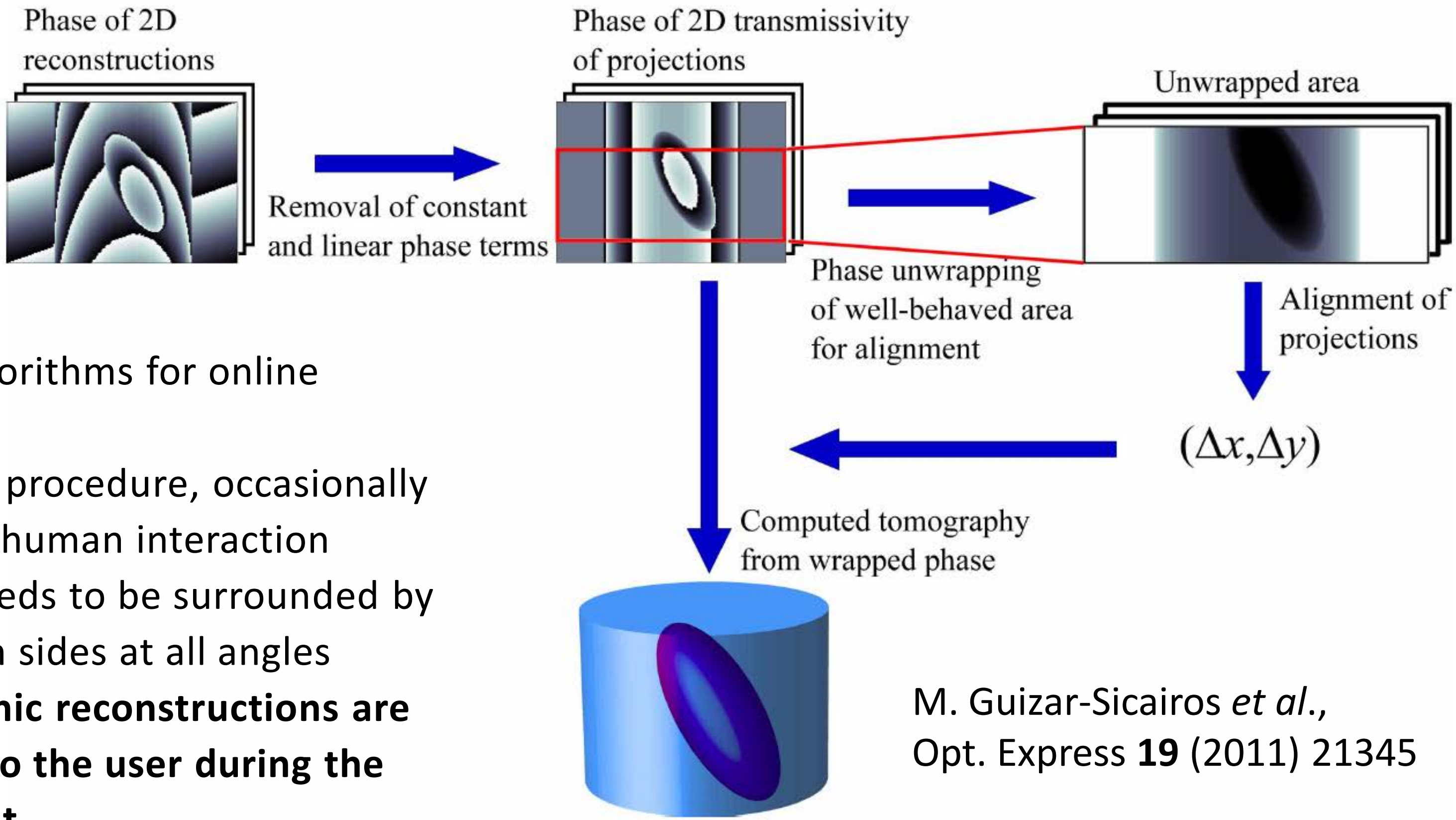
M. Holler and J. Raabe



- Laser interferometry for relative positioning of sample and illumination optics
- Aimed 3D resolution: 10 nm
- Cryo stage in ultra-high vacuum
- First test setup in air at room temperature, still in user operation

M. Holler *et al.*, Rev. Sci. Instrum. **83**, 073703 (2012)
M. Holler *et al.*, Rev. Sci. Instrum. **89**, 043706 (2018)

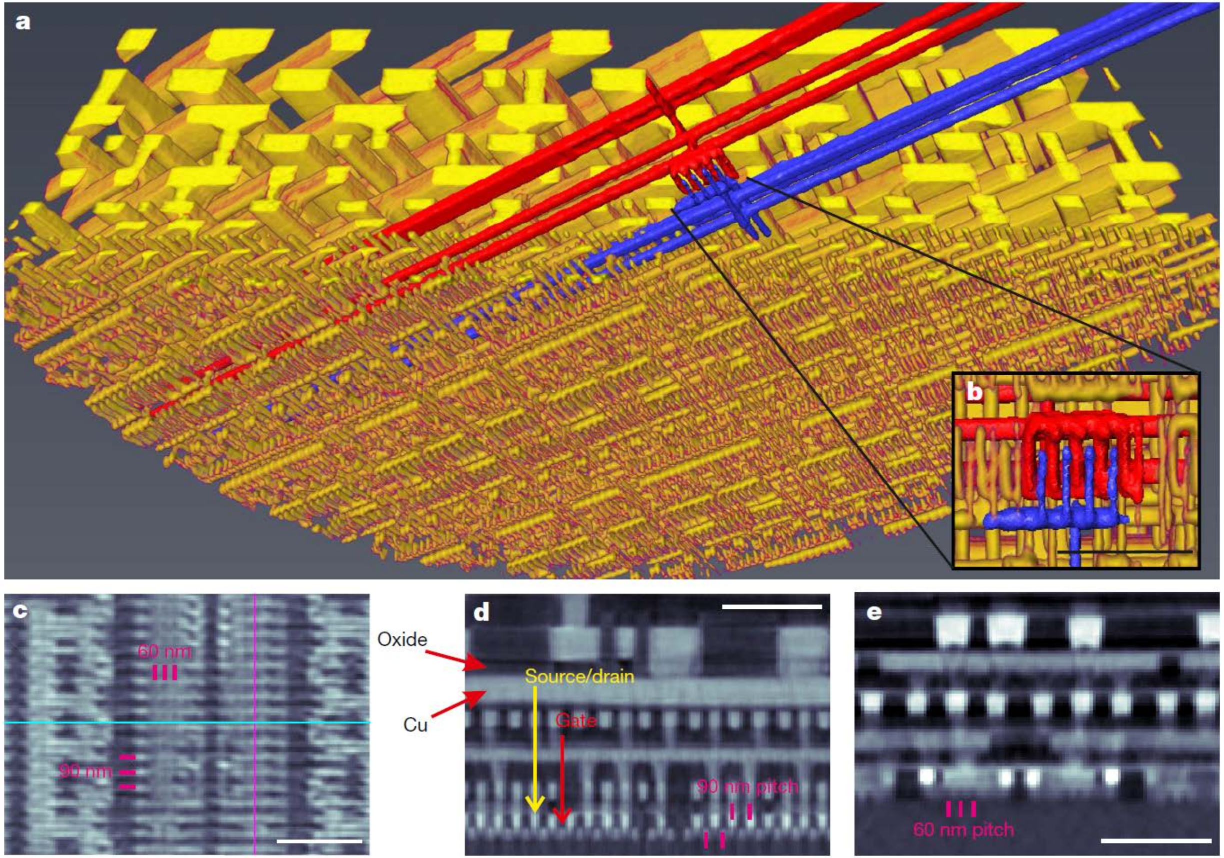
Image processing for tomography



- Robust algorithms for online processing
- Automatic procedure, occasionally still needs human interaction
- Sample needs to be surrounded by air on both sides at all angles
- **Tomographic reconstructions are provided to the user during the experiment**

M. Guizar-Sicairos *et al.*, Opt. Express **19** (2011) 21345

High-resolution nanotomography



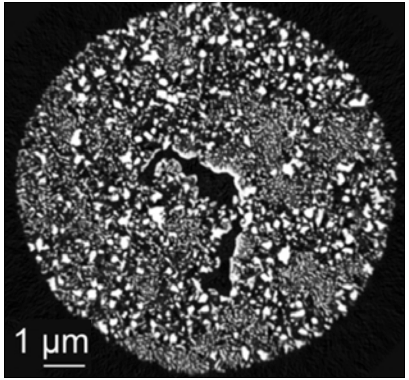
Intel chip,
22 nm technology

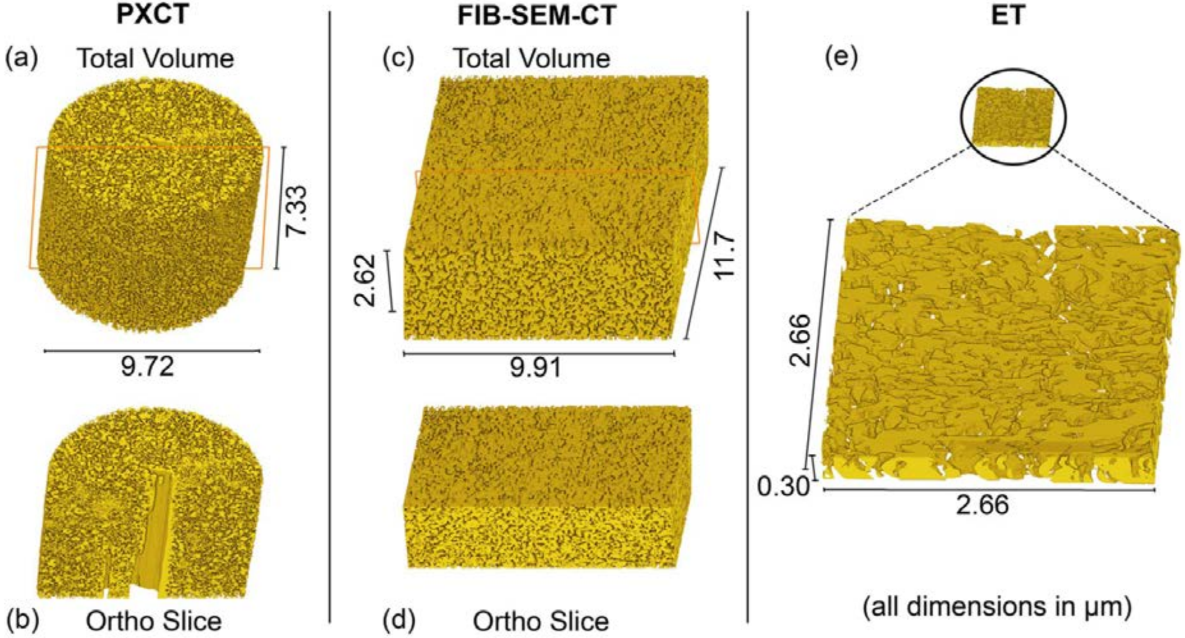
M. Holler *et al.*,
Nature **543**, 402 (2017)

Resolution: 14.6 nm
Scale bars: 500 nm

Nanoporous Au 3D structure

PXCT 2D slice





Estimated 3D resolution: (half-period)

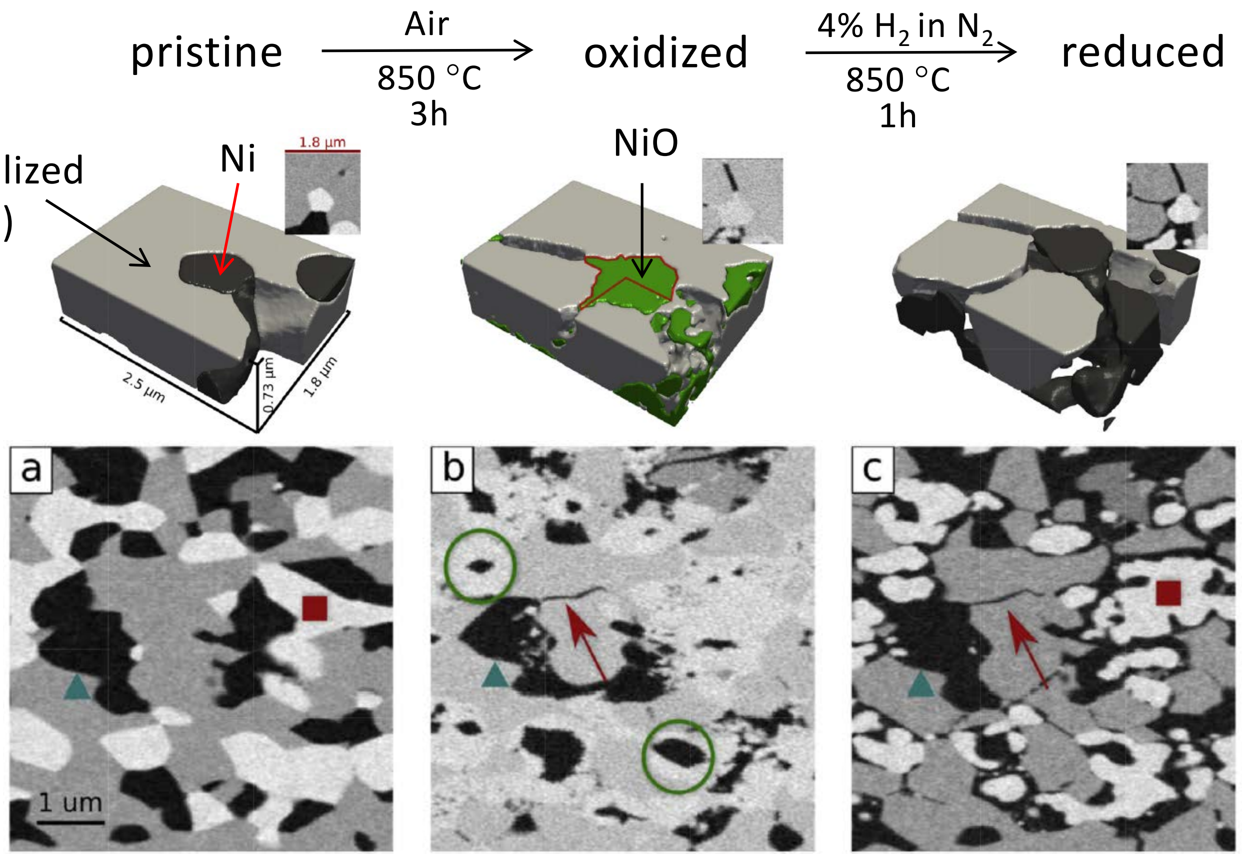
23 nm

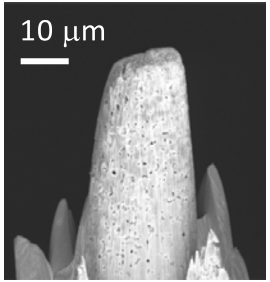
7-22 nm

0.5-1.5 nm

Y. Fam *et al.*, ChemCatChem **10**, 2858 (2018)

Ex-situ SOC electrode microstructure evolution



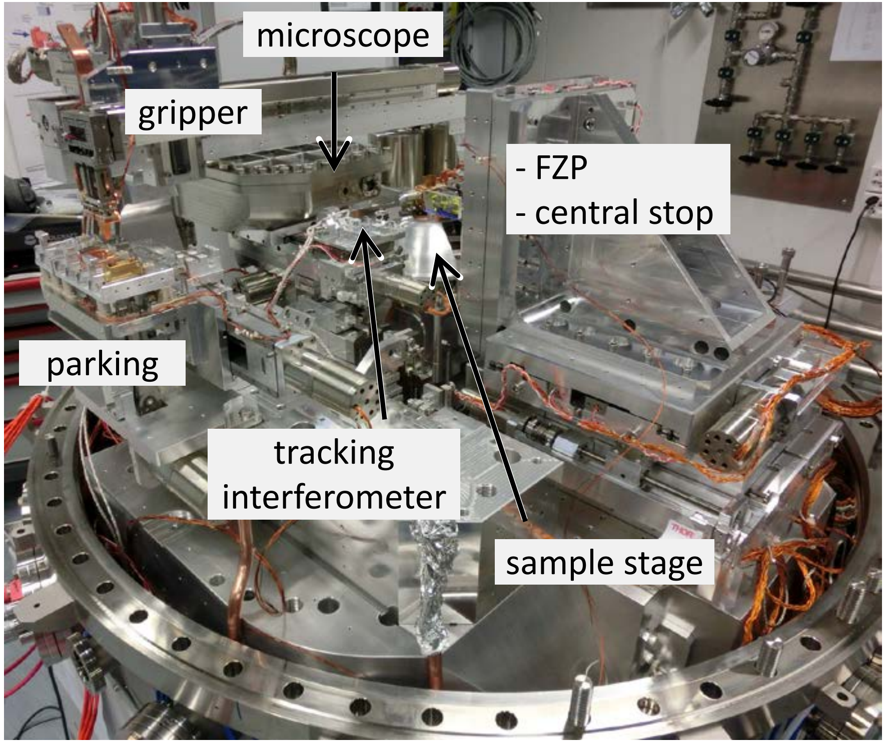


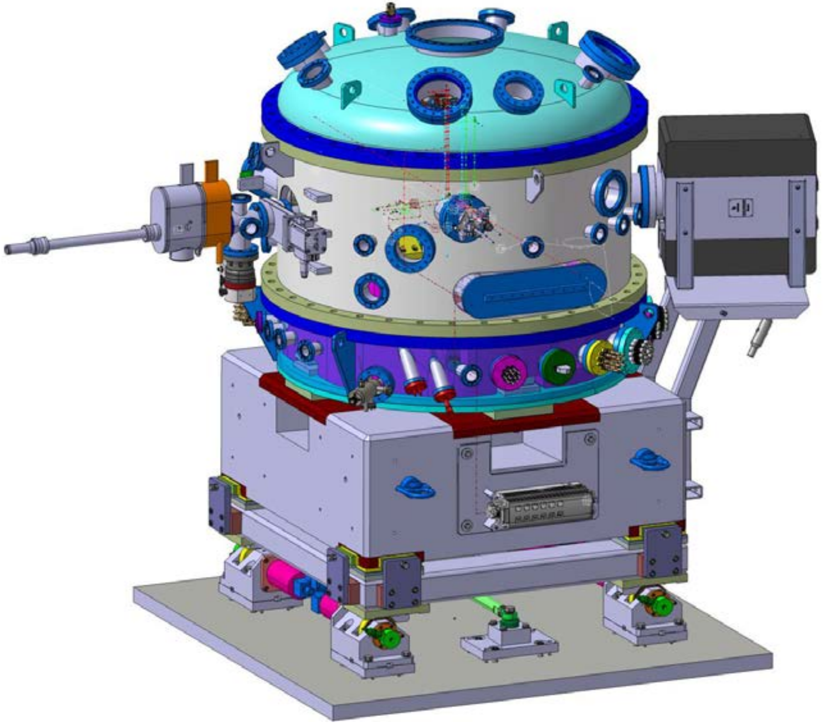
18.4 nm thick slices through 3D dataset

S. De Angelis *et al.*, J. Power Sources **360**, 520 (2017)

OMNY: The cryo-stage instrument

M. Holler, J. Raabe, and engineer team at PSI





M. Holler *et al.*, Rev. Sci. Instrum. **89**, 043706 (2018)

Beetle scale structure: optimized by evolution

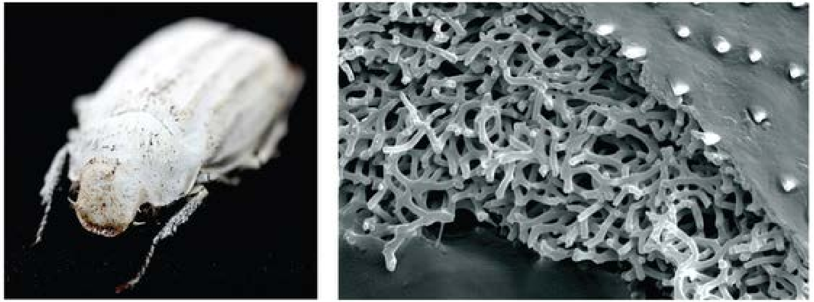
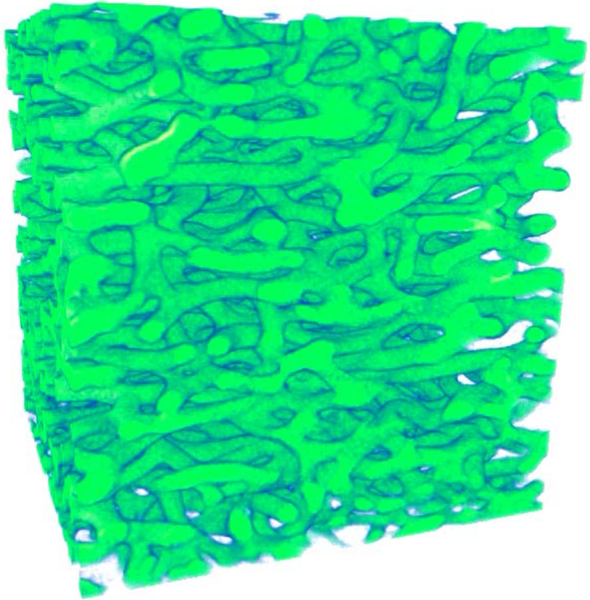


Figure from D. S. Wiersma, Nat. Photonics **7** (2013) 188

B. D. Wilts *et al.*, Adv. Mater. **30**, 1702057 (2018)

- *Cyphochilus* beetle scale specimen prepared by focus ion beam milling
- OMNY cryo stage at 92 K in vacuum
- 3D resolution: 28 nm
- Nanophotonic simulations confirm that the structure is optimized by evolution



About 7 x 7 x 7 μm^3

Compare *Chlamydomonas* measurements

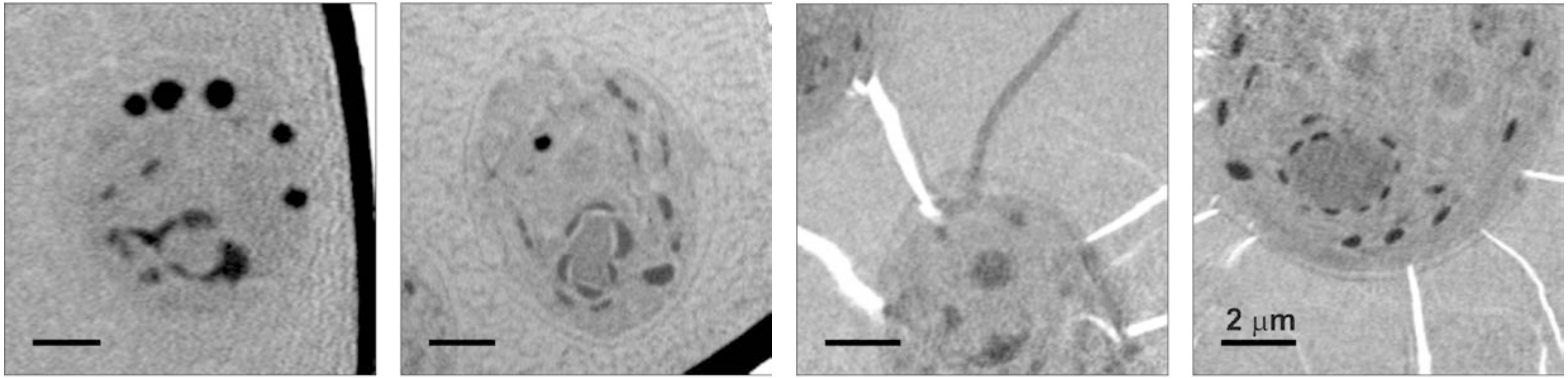
Plunge frozen

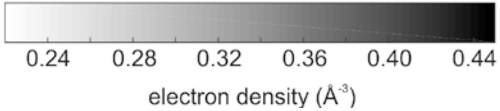
10% glycerol
cryo-jet (1)

10% DMSO
OMNY (2)

High pressure frozen

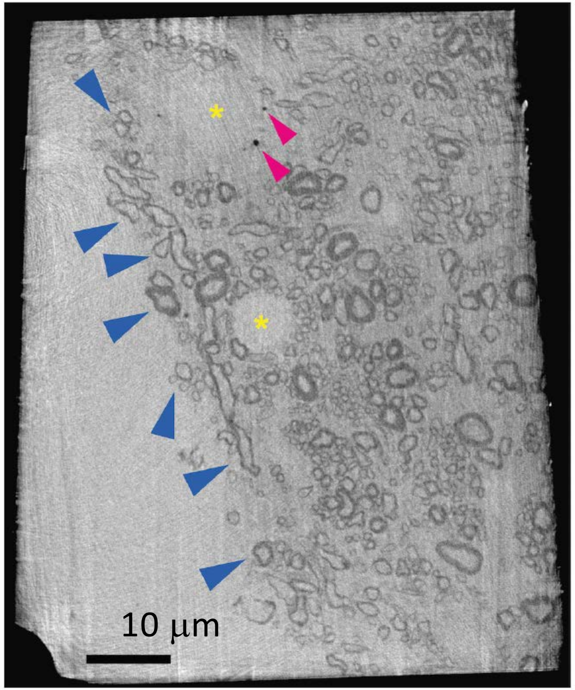
no cryoprotectant
OMNY (3)





(1) A. Diaz et al., J. Struct. Biol. **192**, 461 (2015)
(2) M. Holler *et al.*, Rev. Sci. Instrum. **89**, 043706 (2018)
(3) M. Holler *et al.*, Rev. Sci. Instrum. **88**, 113701 (2017)

Mouse brain tissue



Chemically fixed
Frozen hydrated

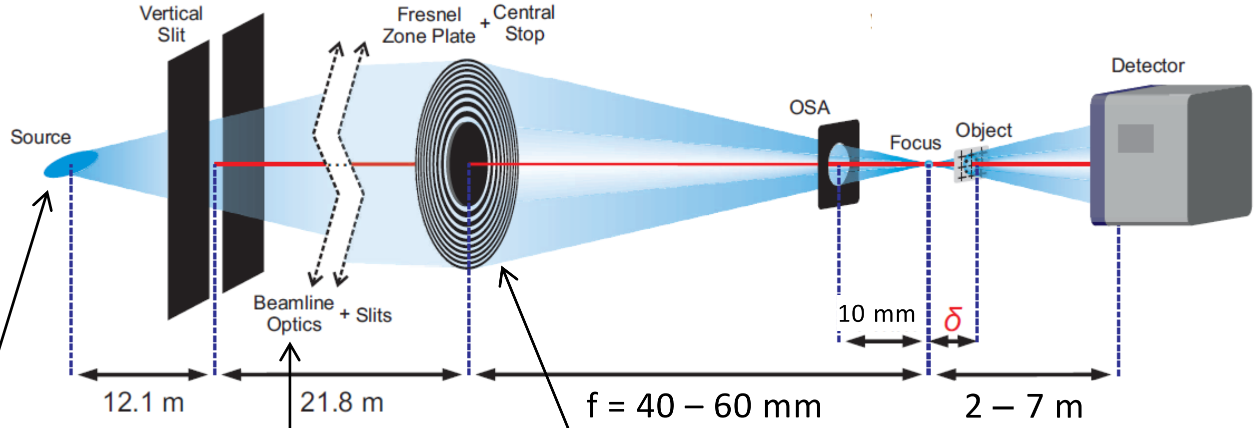
myelinated axons
cell nuclei
lysosomal lipofuscin or pigmented autophagic vacuoles

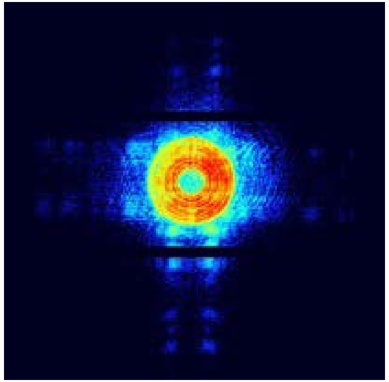
Volume: 80×70×20 μm^3
3D resolution: 120 nm

S. Shahmoradian *et al.*, Sci. Rep. **7** (2017) 6291

Experimental improvements

J. Vila-Comamala *et al.*, Opt. Express **19** (2011) 21333





coherent flux:
5×10^8 photons/s @ 6.2 keV

Storage ring upgrade, new undulator
×100

Broader bandwidth
×10

Efficient optics
×10

gain in coherent flux

A bright future for ptychography

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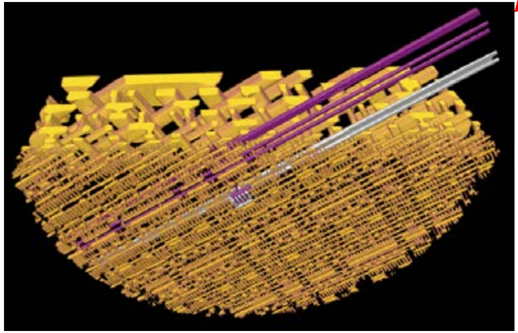
5800 resolution elements/s



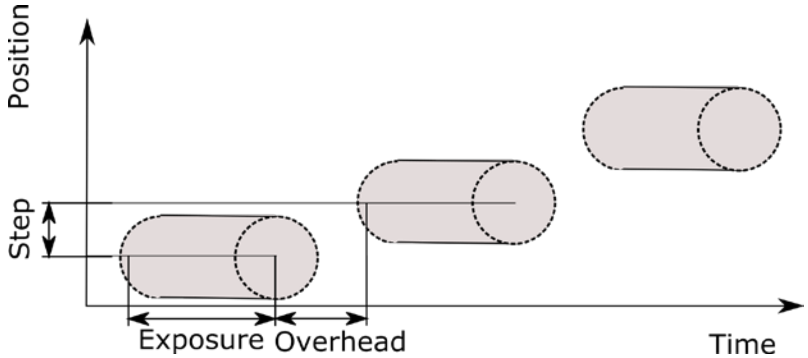
M. Holler *et al.*,
Nature **543**, 402 (2017)

DEVELOPMENT	RESOLUTION (nm)	VOLUME (μm^3)	TIME
State of the art	**14.6**	**15x15x8**	**22 h**
SLS-2	6.2	85x85x8	41 min
+ new undulator	4.6	150x150x8	13 min
+ broadband	2.6	475x475x8	1.3 min
+ efficient optics	1.5	1500x1500x8	8 s

Numbers indicate the gain in one parameter with respect to the **state of the art** when keeping the other two parameters constant

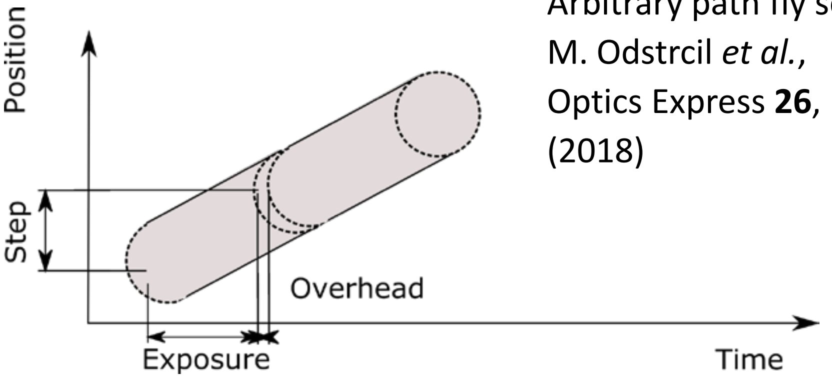
How can we scan faster?

Step scan



Fly scan

- step size = resolution element to preserve resolution
- ×100 faster acquisitions to reach current performance
- Hardware would limit an increase of speed by another ×100



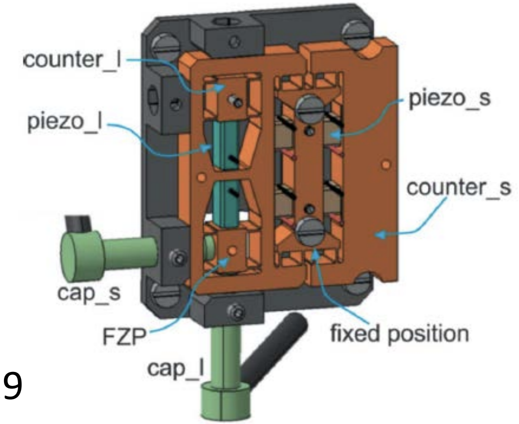
Arbitrary path fly scan:
M. Odstrcil *et al.*,
Optics Express **26**, 12585
(2018)

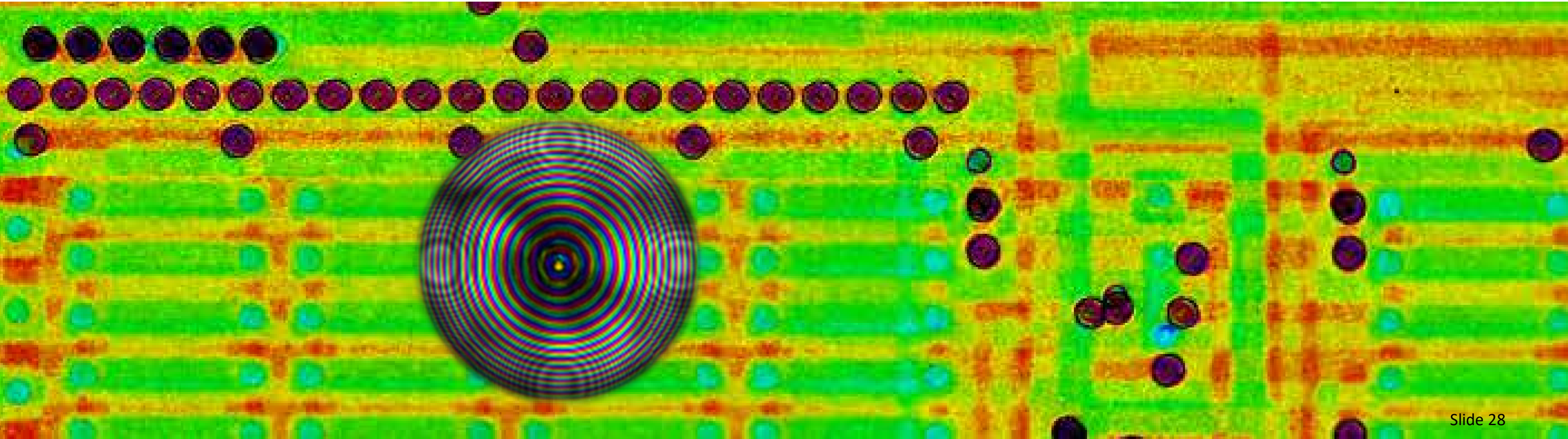
How can we scan faster?

A hardware approach:

- Hybrid sample and optics motion system for 500Hz scanning
- **Up to 50x reduced scan overhead without quality reduction**

M. Odstrcil *et al.*, J. Synchrotron Rad **26**, 504 2019

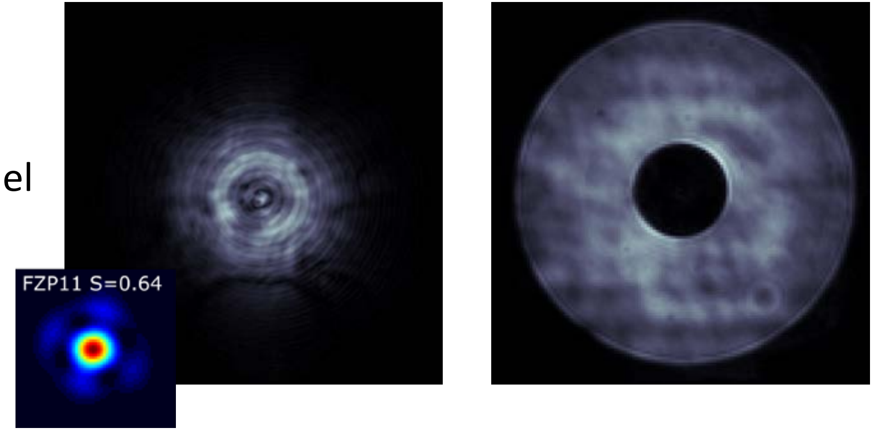




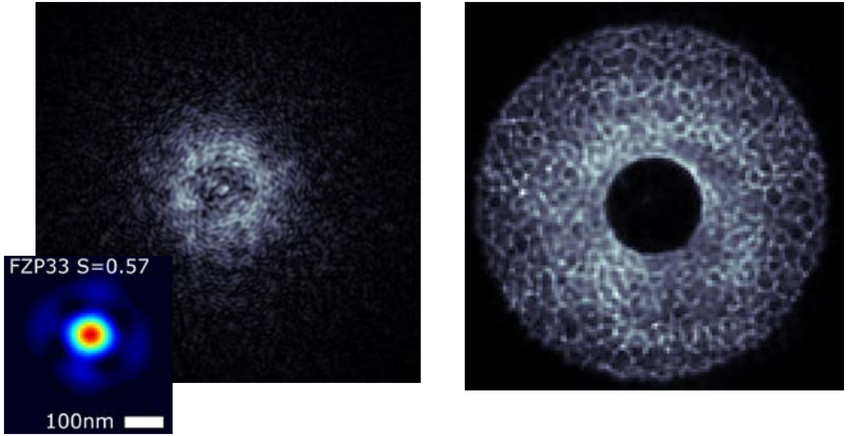
Optimization of the illumination

M. Odstrčil *et al.*,
Optics Express **27** 14981 (2019)

Conventional Fresnel
zone plate



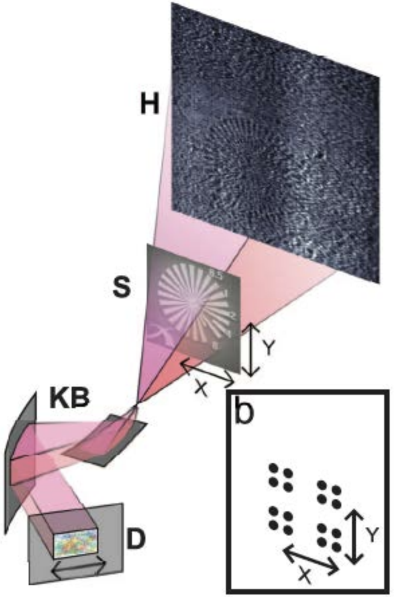
Modified
Fresnel zone plate



- More dose efficient
- Accelerates convergence
- Mitigates effect of beam instabilities

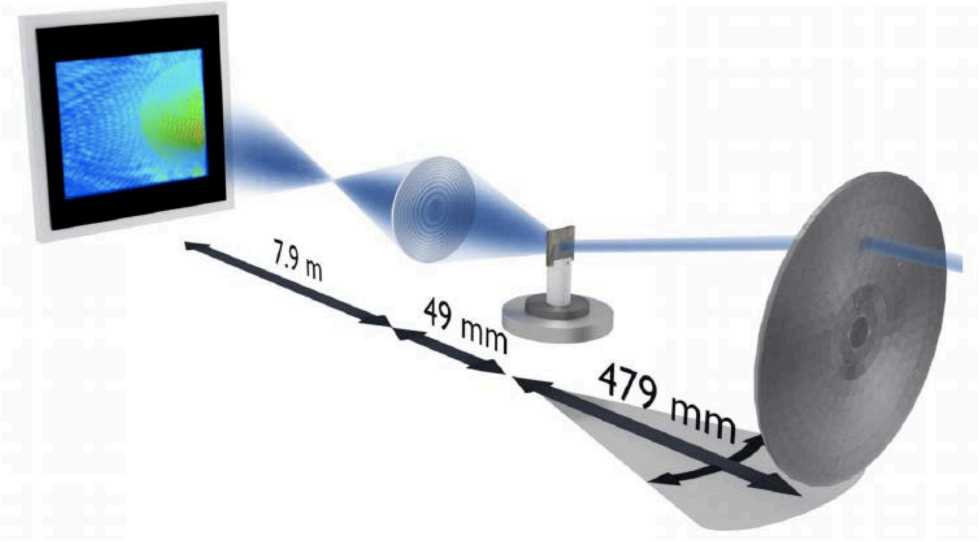
Explore different types of X-ray ptychography

Near-field ptychography



M. Stockmar *et al.*, Sci. Rep. **3** 1927 (2013)

Fourier ptychography



K. Wakonig *et al.*, Sci. Adv. **5** eaav0282 (2019)

Conclusions

- Ptychographic tomography (PT) is a powerful nanotomography technique:
  - High resolution
  - High phase sensitivity
  - Quantitative contrast
- Requirements:
  - Positioning accuracy beyond what is commercially available
  - High computing power to do online image reconstructions
  - Sample preparation on custom mounts
- At the cSAXS beamline we have successfully implemented PT for non-expert users
- Upgraded sources with further experimental improvements can push the performance of PT by orders of magnitude
- Method development in X-ray ptychography is mandatory to fully benefit from these upgrades

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