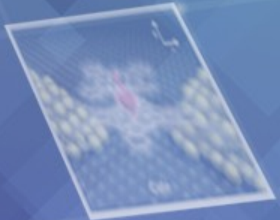

Julio Cesar da Silva

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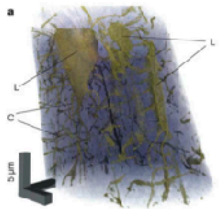
julio-cesar.da-silva@neel.cnrs.fr

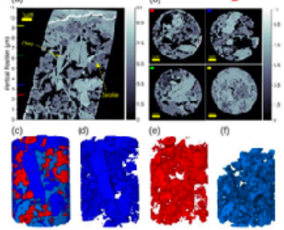
Far-field ptychographic tomography

Bone



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

Technical catalyst

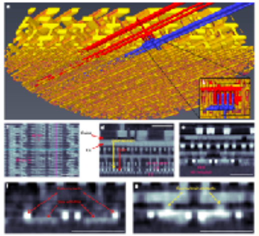


J. C. da Silva et al, ChemCatChem 7 (2015) 413.

Cement

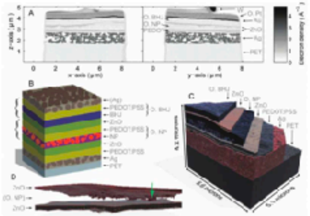
J. C. da Silva et al., Langmuir 31 (2015) 3779.

Integrated circuits



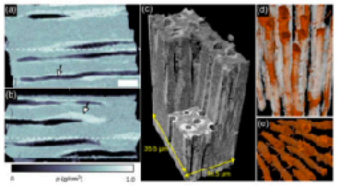
M. Holler *et al.*, Nature 543, 402-204 (2017)

Tandem solar cell



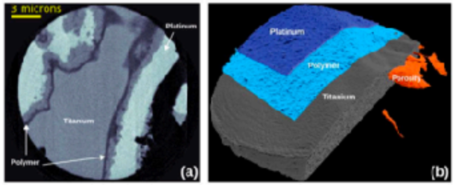
E. B. L. Pedersen et al., Nanoscale 7 (2015) 13765.

Human dentine



I. Zanette et al., Sci. Rep. 5 (2015) 9210.

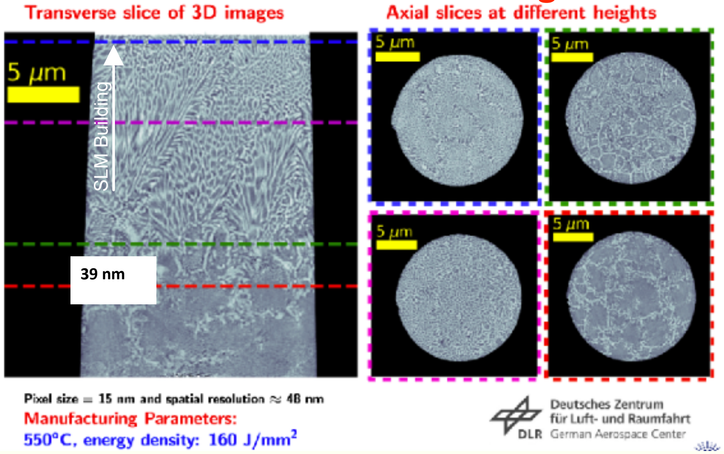
Polymer-metal welding



J. Haubrich et al., Appl. Surf. Sci. 433 (2018) 546.

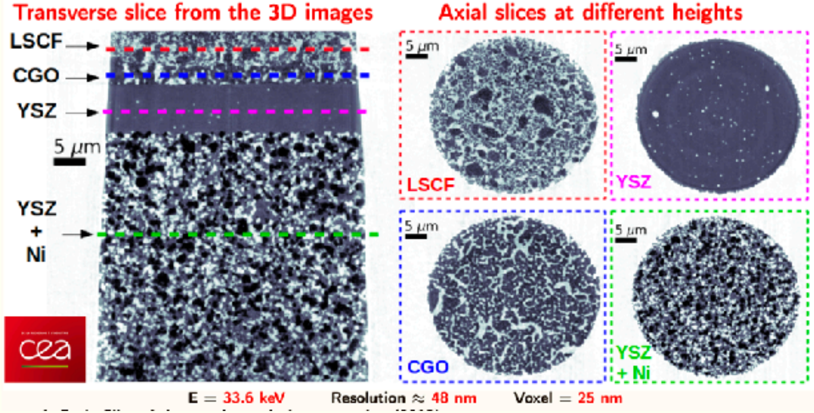
Near-field ptychographic tomography

Additive manufacturing



J. Gussone *et al.*, Appl. Mat. Today 20, 100767 (2020)

Solid Oxide Fuel Cell



F. Monaco et al., Materials Characterization 187, 111834 (2022)

The sampling requirements for ptychography

The spread in the real space has the size of the probe

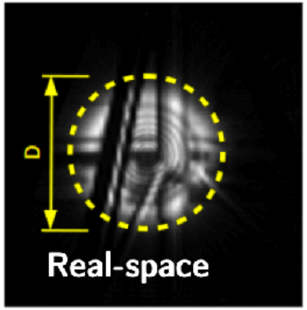
$$\sigma_{r_j} = D_j = probe\ size$$

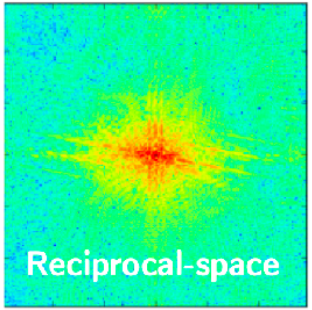
The equivalent spread in the Fourier space would be

$$\sigma_{q_j} = q_j = \frac{2\pi}{D_j}$$

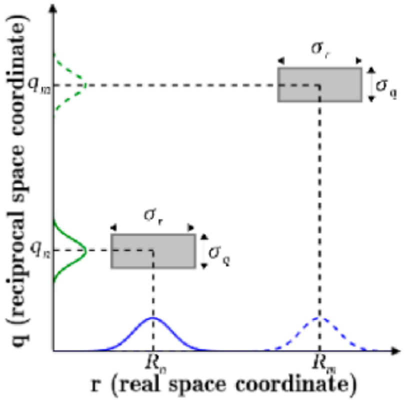
Area of a box:

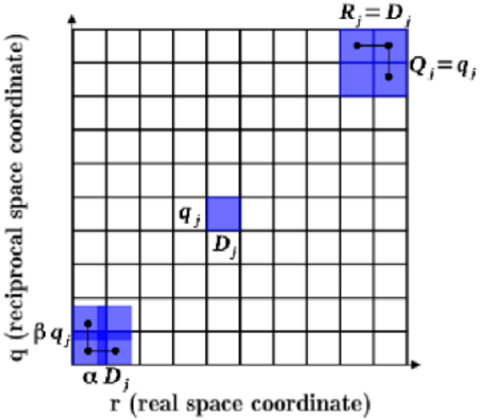
$$D_j\, q_j = 2\pi$$





$$\psi(\vec{r};\vec{R}) = O(\vec{r})P(\vec{r}-\vec{R}) \qquad I(\vec{q}) = |\mathcal{F}\{\psi(\vec{r};\vec{R})\}|^2$$





Let us define:

$$R_j = \alpha D_j$$

$$Q_j = \beta q_j$$

where: $\beta = \dfrac{1}{\mathcal{O}}$

Density of boxes:

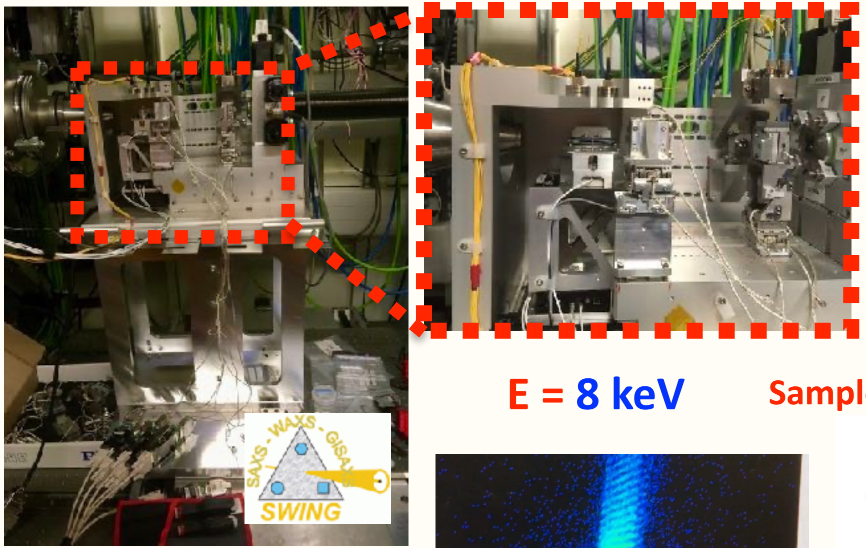
$$\frac{2\pi}{(\alpha D_j)(\beta q_j)} \geq 1$$

$$\boxed{\frac{1}{\alpha\beta} > 1}$$

Oversampling condition

J. C. da Silva, A. Menzel, Opt. Express 23(26): 33812-33821 (2015)
I. Daubechies, A. Grossmann, Y. Meyer, J. Math. Phys. 27,1 271–1283 (1986)

The nanoprobe at SWING beamline SOLEIL



Testing on the Siemens Star

FOV: 20 x 20 μm²
Beam size: 6 μm
Step size: 1 μm
Exp. Time p/ pt: 100 ms
scan points: 322
Total Time: ~42 s

Checking the sampling conditions:

$$\alpha = 0.17$$

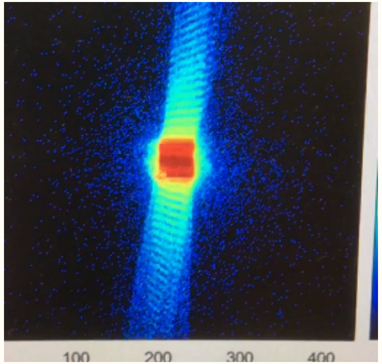
$$\beta = 1/2.27 = 0.44$$

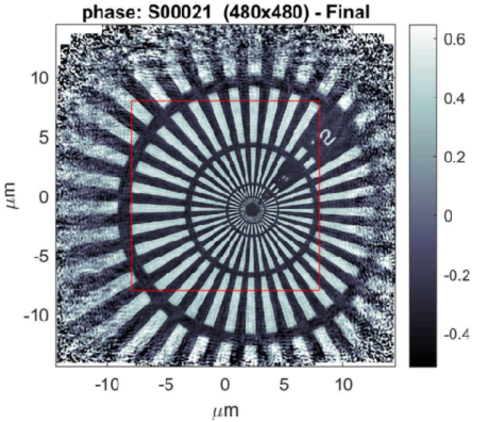
The oversampling ratio:

$$1/\alpha\beta = 13.4$$

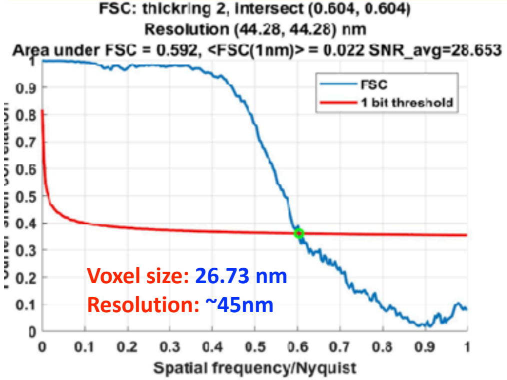
E = 8 keV

Sample-detector distance: 6.6 m





Resolution estimate by FSC

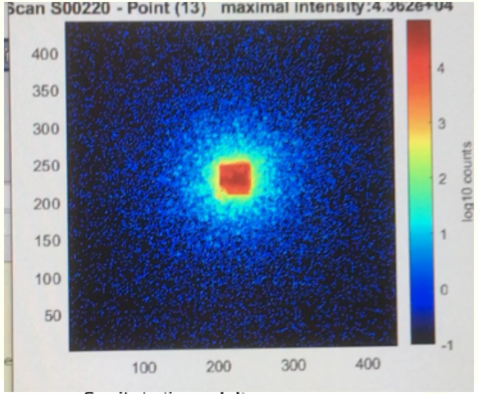


3D imaging of Hydrodesulfurization catalyst

FOV: 26 x 26 µm²
Beam size: 6 µm
Step size: 2 µm
Exp. Time p/ pt: 100 ms
scan points: 195
Scan duration: ~26 s

The oversampling ratio:

$$1/\alpha\beta = 6.89$$



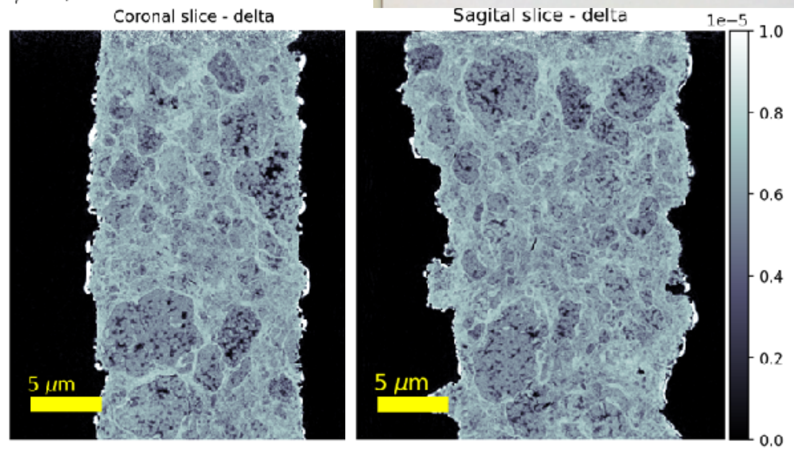
Number of projections: 640

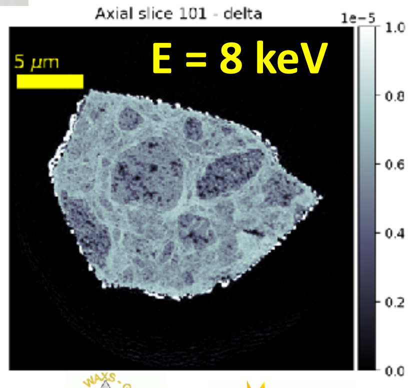
Total tomo time: 5h40min

$$N > \frac{\pi}{2}\frac{D}{\Delta x}$$

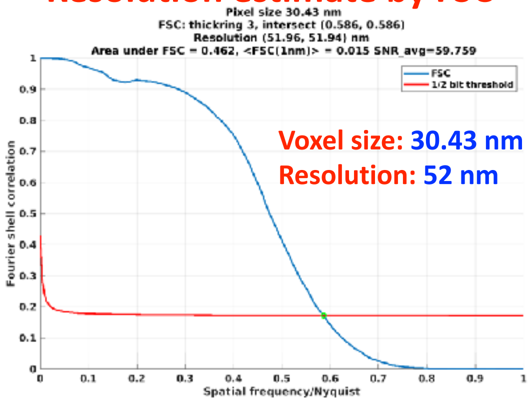
$$D = 16\mu m \quad \Delta x = 40nm$$

$$N > 629$$





Resolution estimate by FSC



Voxel size: 30.43 nm
Resolution: 52 nm

3D imaging of Hydrodesulfurization catalyst

GDCh Zuschriften Angewandte Chemie

Zitierweise: Angew. Chem. Int. Ed. 2020, 59, 17266–17271
Internationale Ausgabe: doi.org/10.1002/anie.202008030
Deutsche Ausgabe: doi.org/10.1002/ange.202008030

Heterogeneous Catalysis

Hierarchical Structure of NiMo Hydrodesulfurization Catalysts Determined by Ptychographic X-Ray Computed Tomography

Johannes Ihli, Leonid Bloch, Frank Krumeich, Klaus Wakonig, Mirko Holler, Manuel Guizar-Sicairos, Thomas Weber, Julio Cesar da Silva, and Jeroen Anton van Bokhoven**

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38000 Grenoble (France)
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Institute for Biomedical Engineering, ETH Zürich, and University of Zürich, 8093 Zürich (Switzerland)
Prof. Th. Weber
Shell Technology Centre Amsterdam
1031HW Amsterdam (The Netherlands)
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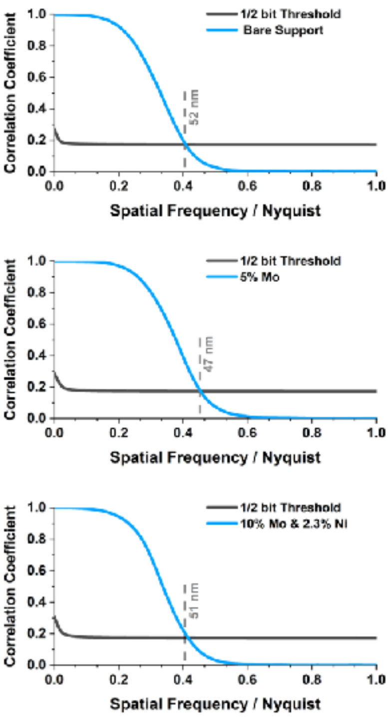
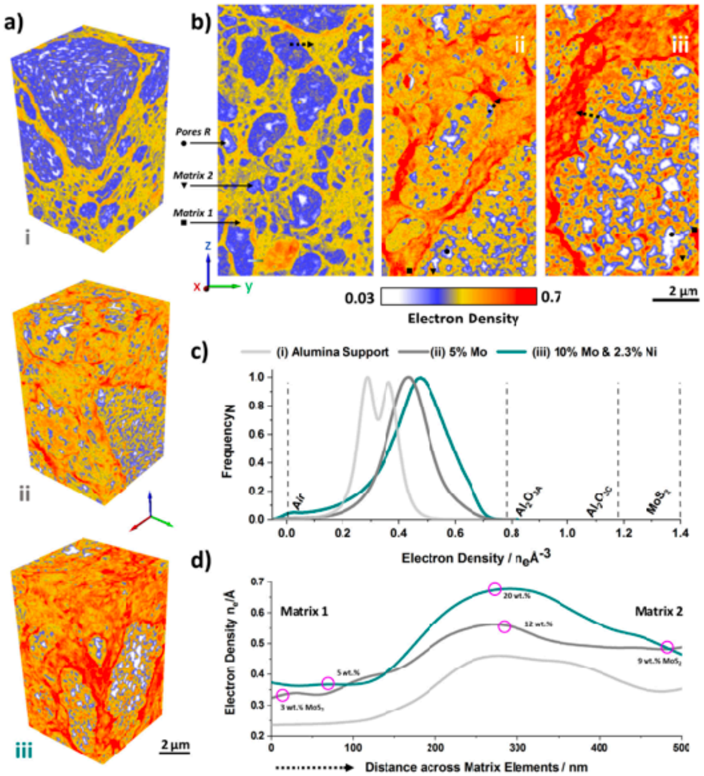
Institut Néel CNRS UPR2940, 38042 Grenoble (France)
Supporting information and the ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/anie.202008030.

Angew. Chem. 2020, 132, 17419–17424 Wiley Online Library 174



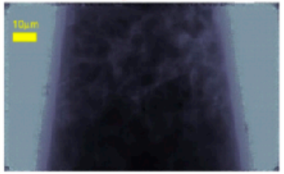


PAUL SCHERRER INSTITUT

Quantitative Ptychography X-ray Computed Tomography

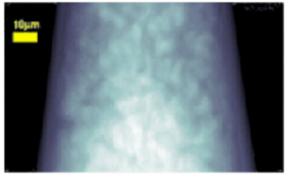
Absorption-contrast

Absorption index β



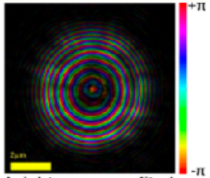
Phase-contrast

Refractive index decrement δ



Probe

Complex-value beam



brightness : amplitude
color : phase

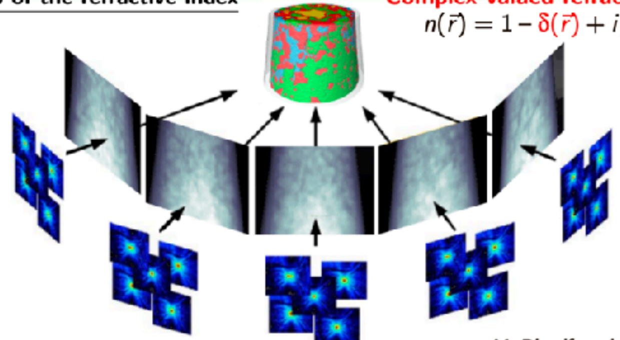
J. C. da Silva et al. Langmuir 31 (2015) 3779

Ptychographic X-ray Computed Tomography (PXCT)

3D map of the refractive index

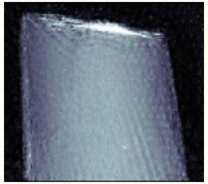
Complex-valued refractive index

$n(\vec{r}) = 1 - \delta(\vec{r}) + i\beta(\vec{r})$

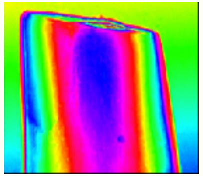


M. Dierolf et al. Nature 467 (2010) 436

Amplitude



Phase



Complex transmittance domain

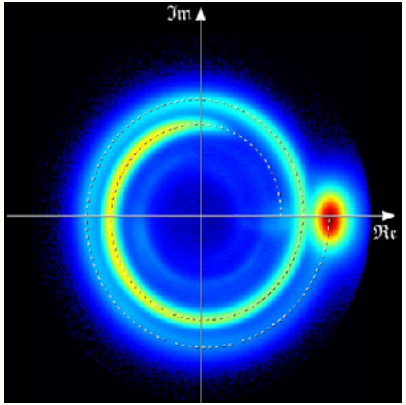


Figure from M. Dierolf et. al., Nature 467 (2010) 436

Complex Transmittance function of object

$$T(x,y) = exp\left\{\frac{2\pi i}{\lambda}\int [n(\vec{r}) - 1]\, dz\right\}$$

$$n(\vec{r}) = 1 - \delta(\vec{r}) + i\beta(\vec{r})$$

Linear attenuation coefficient (cm^{-1})

$$\mu(\vec{r}) = \left(\frac{4\pi}{\lambda}\right)\beta(\vec{r})$$

$\lambda =$ X-ray wavelength

Electron Density (e^{-}/cm^{3})

$$n_e(\vec{r}) = \frac{2\pi\delta(\vec{r})}{\lambda^2 r_0}$$

$r_0 =$ classical electron radius

Mass Density (g/cm^{3})

$$\rho(\vec{r}) = \frac{n_e(\vec{r})}{N_A}\left(\frac{\Sigma_j \alpha_j A_j}{\Sigma_j \alpha_j Z_j}\right)$$

$N_A =$ Avogrado's number $A_j =$ atomic mass

Mass attenuation coefficient (dimensionless)

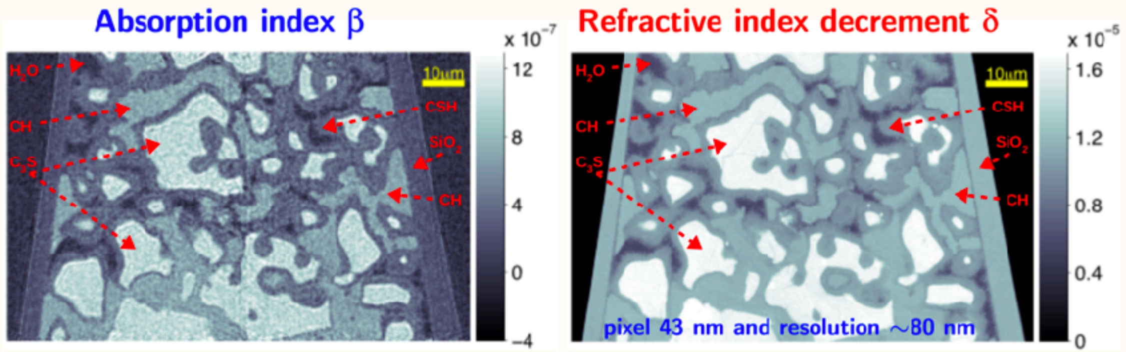
$$\frac{\mu}{\rho} = (const.)\lambda^3 Z^3$$

$Z_j =$ atomic number

B. D. Cullity, Elements of X-ray Diffraction, Addison-Wesley Pub. Company, Inc. (1956)

Chemical Imaging of hydrated ordinary cement

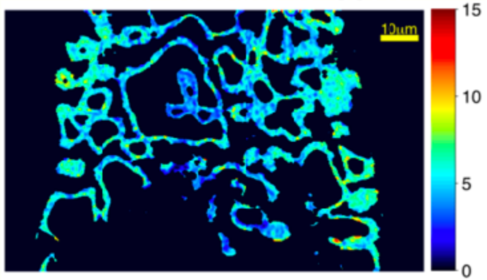
$$\underbrace{Ca_3SiO_5}_{\text{Alite } C_3S} + \underbrace{(1.3 + y)H_2O}_{\text{water}} \rightarrow \underbrace{1.3\,Ca(OH)_2}_{\text{Calcium hydroxide } CH} + \underbrace{(CaO)_{1.7}(SiO_2)(H_2O)_y}_{\text{Calcium-Silicate-Hydrated } CSH}$$



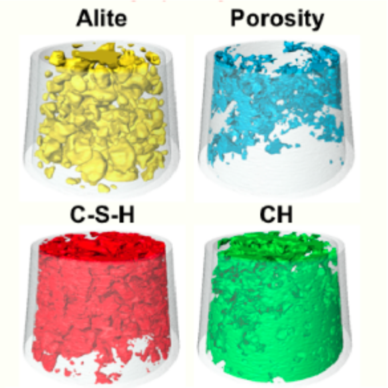
$$\left(\frac{\mu}{\rho}\right)_{CSH} = \omega_{CaO}\left(\frac{\mu}{\rho}\right)_{CaO} + \omega_{SiO_2}\left(\frac{\mu}{\rho}\right)_{SiO_2} + \omega_{H_2O}\left(\frac{\mu}{\rho}\right)_{H_2O}$$

Materials	ρ $(g.cm^{-3})$	$\rho_{ex.}$ $(g.cm^{-3})$
Water	0.99 ± 0.01	1.00
CH	2.18 ± 0.01	2.211
C_3S	3.10 ± 0.01	3.064
Capillary	2.19 ± 0.01	2.203
$CSH_{(y=5.2\pm0.4)}$	1.83 ± 0.01	$1.83\ (y = 5)$

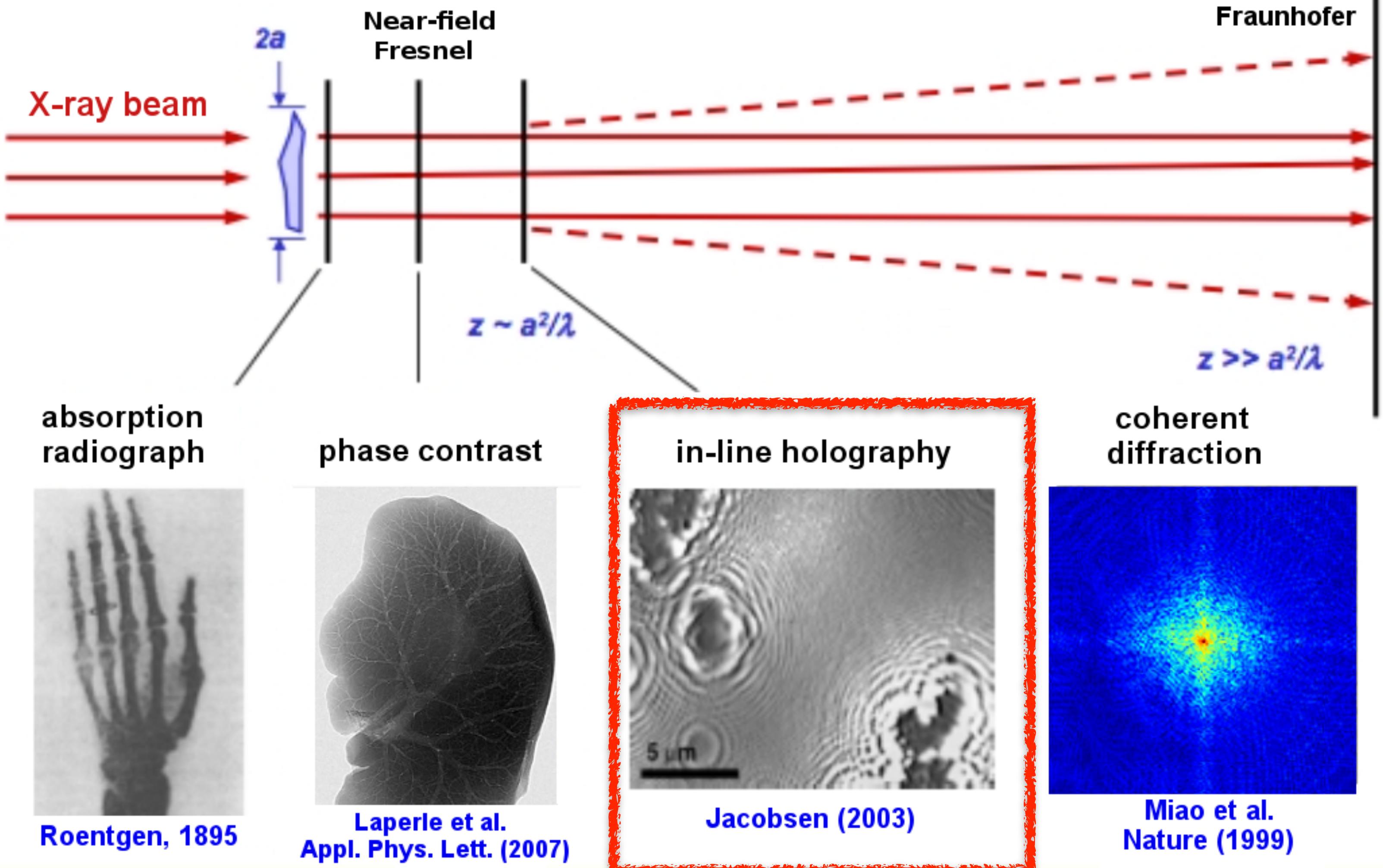
Water content of CSH:



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

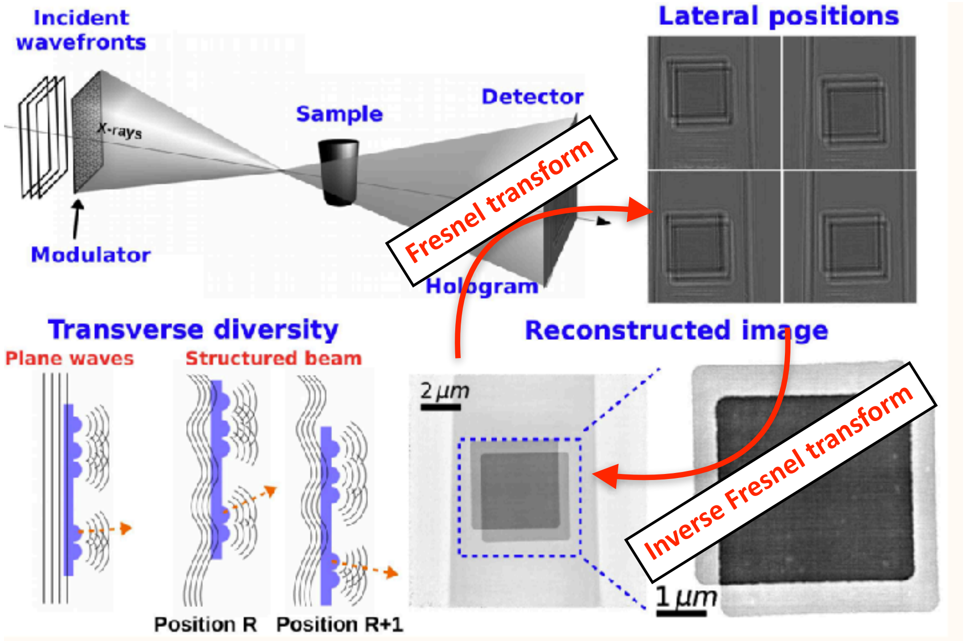


Transmission geometry



Another way to exploit the transverse diversity

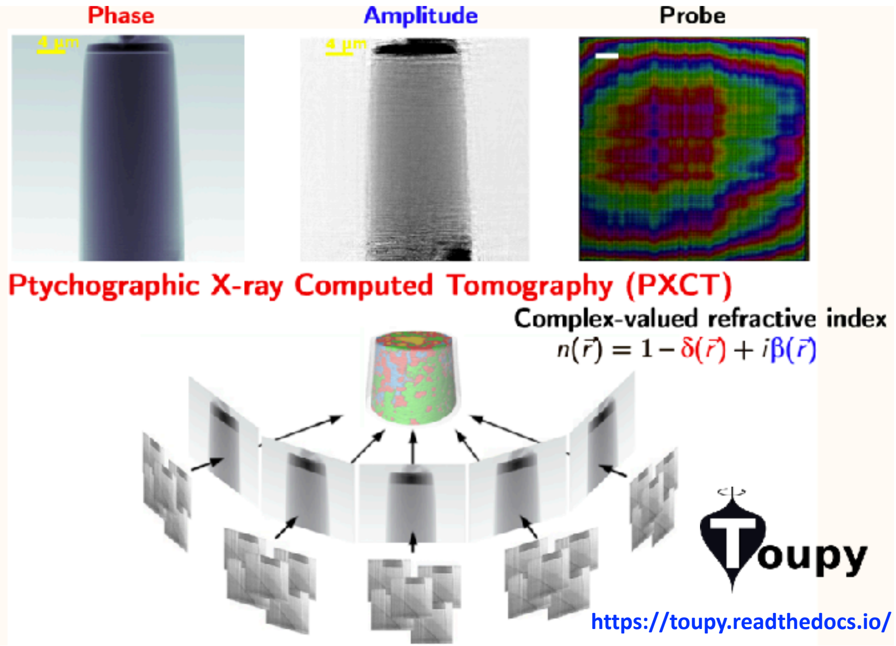
Near-Field Ptychography (NFP)



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

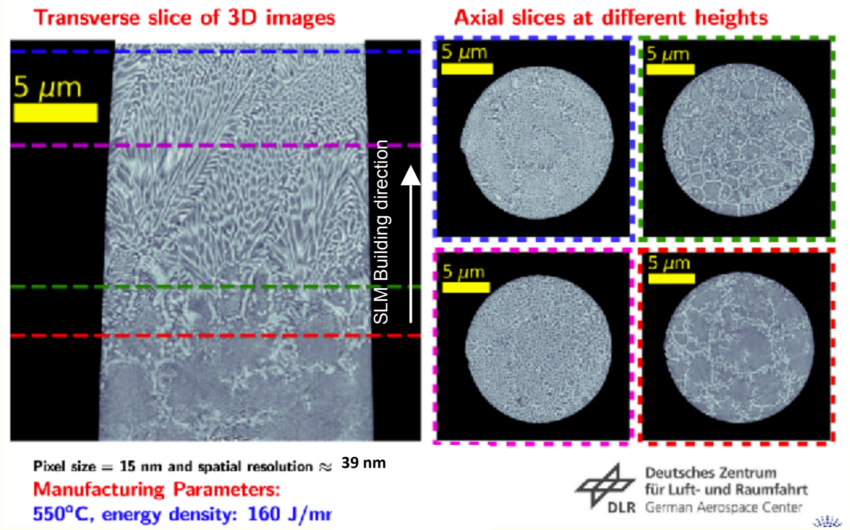
ptypy
https://ptycho.github.io/ptypy/

Near-Field PXCT (nf-PXCT)

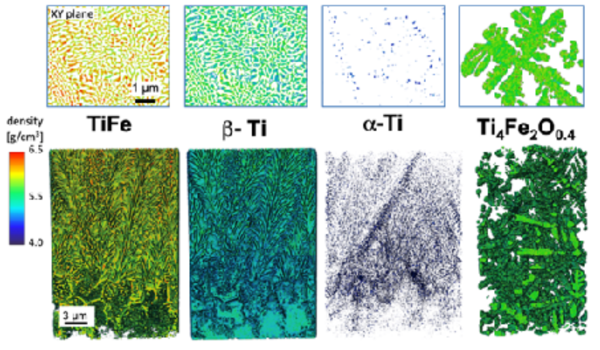


M. Stockmar *et al.*, Opt. Express 23, 12720 (2015)
J. Gussone *et al.*, Appl. Mat. Today 20, 100767 (2020)

Additive manufacturing for metallurgy - FeTi



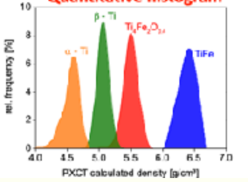
Quantitative analysis



Expected mass density values

Phases	ρ_{HEXRD} [g/cm³]	$\rho_{NF-PXCT}$ [g/cm³]
α - Ti	4.5	4.6 ± 0.08
β - Ti	5.0	5.1 ± 0.09
$Ti_4Fe_2O_{0.4}$	5.7	5.5 ± 0.12
TiFe	6.5	6.4 ± 0.13
LPBF-Ti-32.5Fe	5.63	5.62 ± 0.09

Quantitative histogram



J. Gussone *et al.*, Appl. Mat. Today 20, 100767 (2020)

J. Gussone *et al.*, Appl. Mat. Today 20, 100767 (2020)

Technical catalysts for oil refinery

KAUST
King Abdullah University of Science and Technology

Removed unpublished results

A. Kulow, R. Boudjehem, J.L. Hazemann, J. C. Da Silva, et al. in preparation

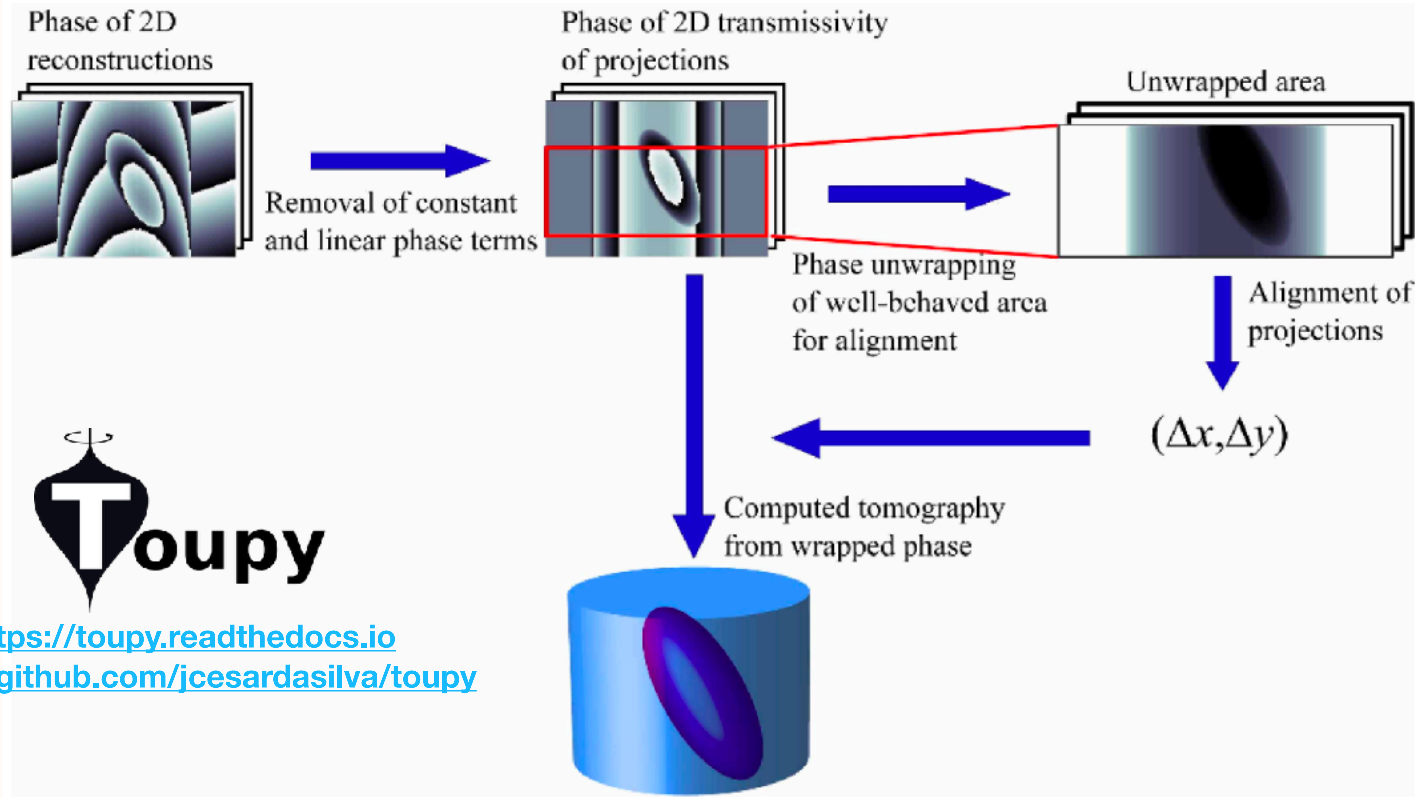
Eco-friendly cementitious materials

Removed unpublished results

UNIVERSIDAD DE MÁLAGA

S. Shirani, J. C. Da Silva, M. Aranda, et al., in preparation

Tomographic pre-processing steps

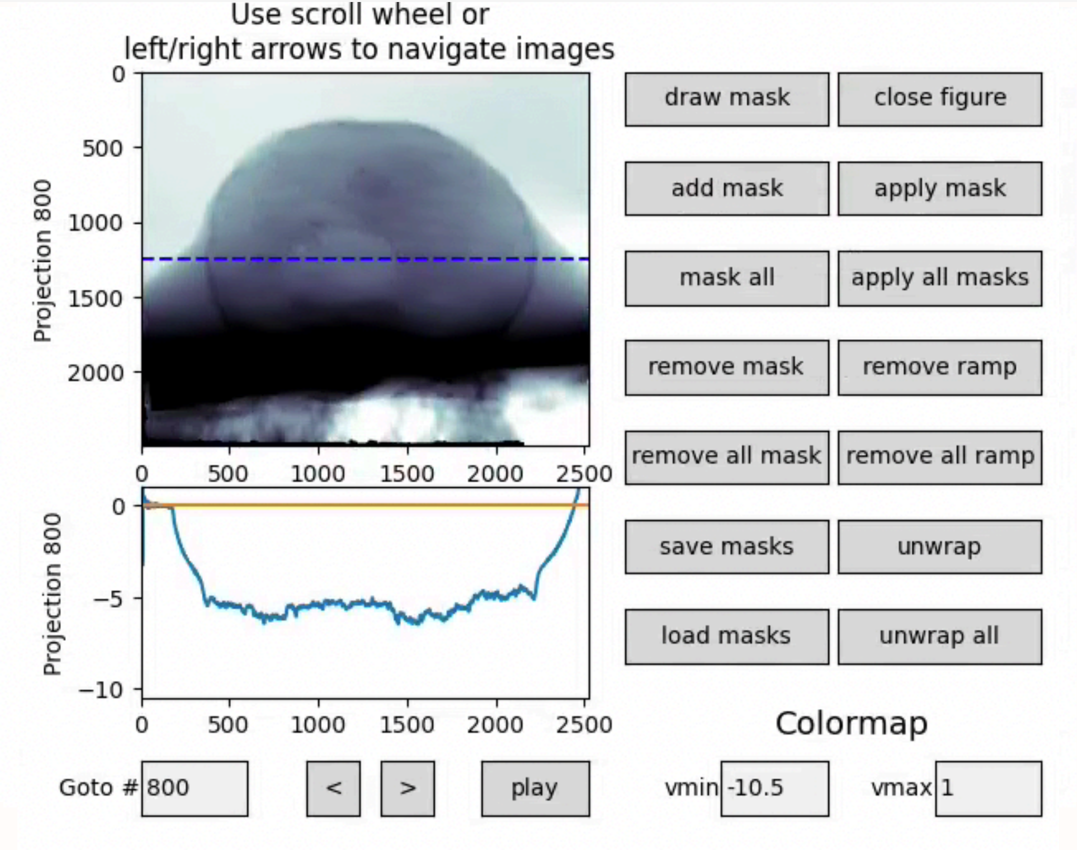


Toupy

https://toupy.readthedocs.io
https://github.com/jcesardasilva/toupy

Correction of the phase projections

```python
from toupy.restoration import gui_plotphase
stack_phasecorr = gui_plotphase(projections,vmin=-1.6,vmax=1.6, crop_reg = None, colormap='bone')
```



1) Linear phase ramp removal

(Phase ambiguities)

Object: $O'(x,y) = O(x,y)e^{[2\pi i(a+bx+cy)]}$

Probe: $P'(x,y) = P(x,y)e^{[-2\pi i(a+bx+cy)]}$

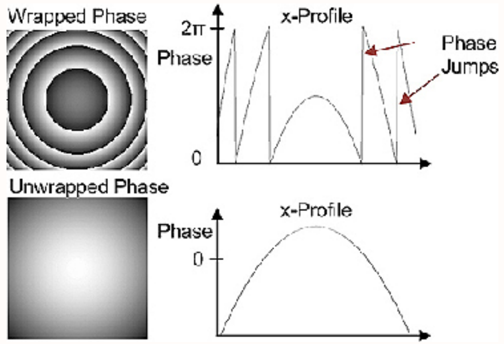
Exit wave:

$$\psi(x,y) = P'(x-x_n, y-y_n)O'(x,y)$$

$$= P(x-x_n, y-y_n)O(x,y)e^{[-2\pi i(bx_n+cy_n)]}$$

2) Phase unwrapping

(Module 2π phase recovery)



M. A. Herraez et al., J. Appli. Optics 41, 7437 (2002)

Also in https://scikit-image.org

Subpixel precision alignment of projections

Vertical alignment

Helgason-Ludwig consistency condition:

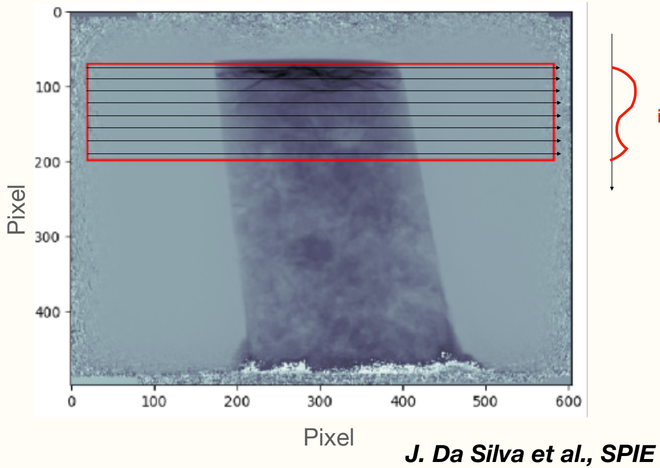
$$\int \mathcal{R}(f)(\theta,p)dp = \iint f(x,z)dxdz,$$

D. Ludwig, Comm. Pure Appli. Math 19, 49-81 (1966)
S. Helgason, Acta Math. 113, 153-180 (1965)

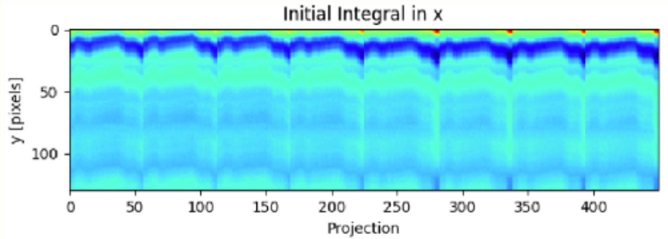
```
from toupy.registration import alignprojections_vertical
shiftproj, valignproj = alignprojections_vertical(projections,shiftproj,**params)
```

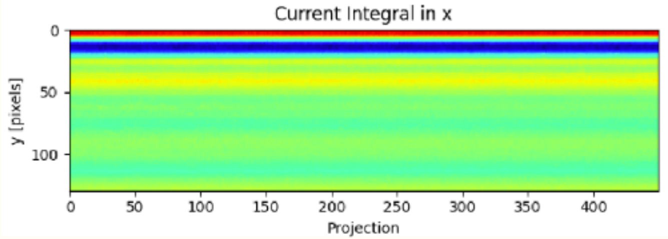
Iterative vertical alignment using the Vertical "mass" distribution:

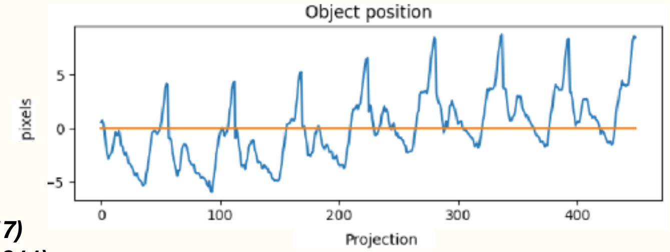
$$vmd_\theta(y) = \int \varphi_\theta(x,y)dx = -\frac{2\pi}{\lambda}\iint \delta(x\cos\theta - z\sin\theta,\ y,\ z\cos\theta + x\sin\theta)dxdz,$$



1D function which is independent of the angle







J. Da Silva et al., SPIE proc. 10391, 1039106 (2017)
M. Guizar-Sicairos et al., Opt. Express 19, 21345 (2011)

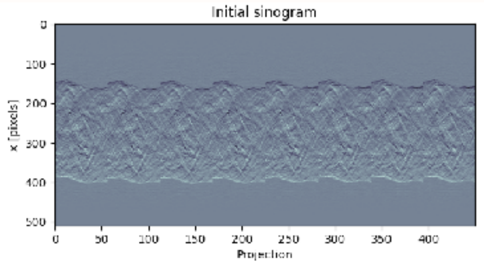
Subpixel precision alignment of projections

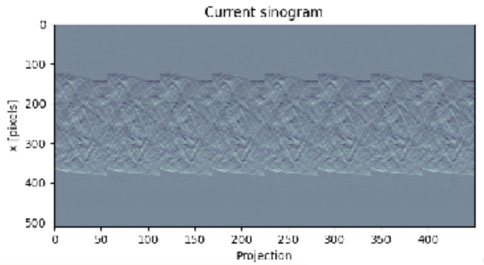
Horizontal alignment

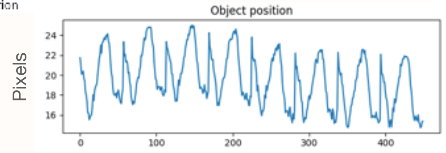
Tomographic consistency:

"The inverse Radon transform by FBP followed by a Radon transform only retrieves the original sinograms if they are consistent with a 3D representation, which is not the case for misaligned projections."

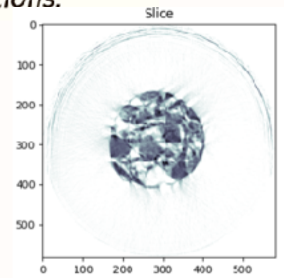
```python
from toupy.registration import alignprojections_horizontal, compute_aligned_horizontal
shiftproj = alignprojections_horizontal(sinogram, theta, shiftproj, **params)
alignedproj = compute_aligned_horizontal(valigndiff, shiftproj, shift_method='fourier')
```





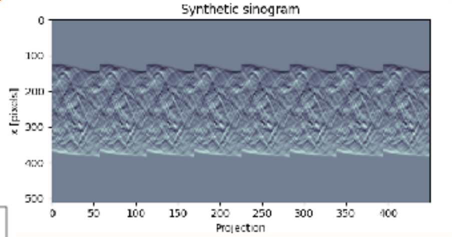


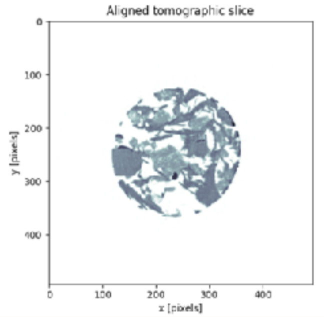
Inverse Radon Transform

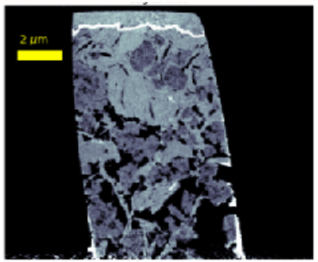


Iterative refinement of positions

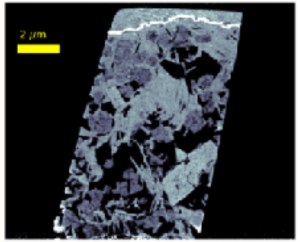
Forward Radon Transform

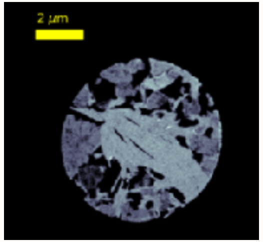







```python
from toupy.tomo import full_tomo_recons
tomogram = full_tomo_recons(projections, theta, **params)
```





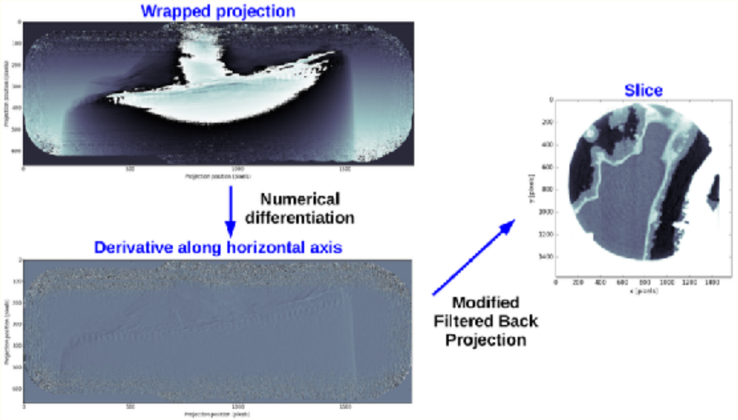
J. Da Silva et al., SPIE proc. 10391, 1039106 (2017)
M. Guizar-Sicairos et al., Opt. Express 19, 21345 (2011)
J. Dengler, Ultramicroscopy 30, 337 (1989)

Modified Filtered Back projection algorithm

"Traditional" Filtered back projection (FBP):
(R. N. Bracewell, A. C. Riddle, The Astrophysical J. 150 (1967) 427)

$$-\frac{2\pi\Delta}{\lambda}\delta(\vec{r}) = \int_0^\pi \mathcal{F}_u^{-1}\{|u|\mathcal{F}_{x'}\{\phi(x', y, \theta)\}\}d\theta$$

where Δ is the length of the side of the voxel.



Using the derivative property of the Fourier transform: $\mathcal{F}_x\{\partial\phi/\partial x\} = i2\pi u \mathcal{F}_x\{\phi\}$

Modified Filtered back projection (FBP)

$$-\frac{2\pi\Delta}{\lambda}\delta(\vec{r}) = \int_0^\pi \mathcal{F}_u^{-1}\left\{\frac{1}{i2\pi\ \mathrm{sgn}(u)}\mathcal{F}_x\left\{\frac{\partial\phi(x, y, \theta)}{\partial x}\right\}\right\}d\theta$$

Conclusion and prospects

➜ **Ptychographic X-ray Computed Tomography is an already mature technique.**

➜ **Appropriately sampling the data allows us to extract the most information from it.**

➜ **The optimization of the instrument also significantly impacts the speed of data acquisition and analysis.**

➜ **Next challenges: Faster data acquisition, nanoimaging of large volumes, *in situ/operando* in 3D (gases, temperature, charge/discharge).**

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https://github.com/jcesardasilva

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QR code