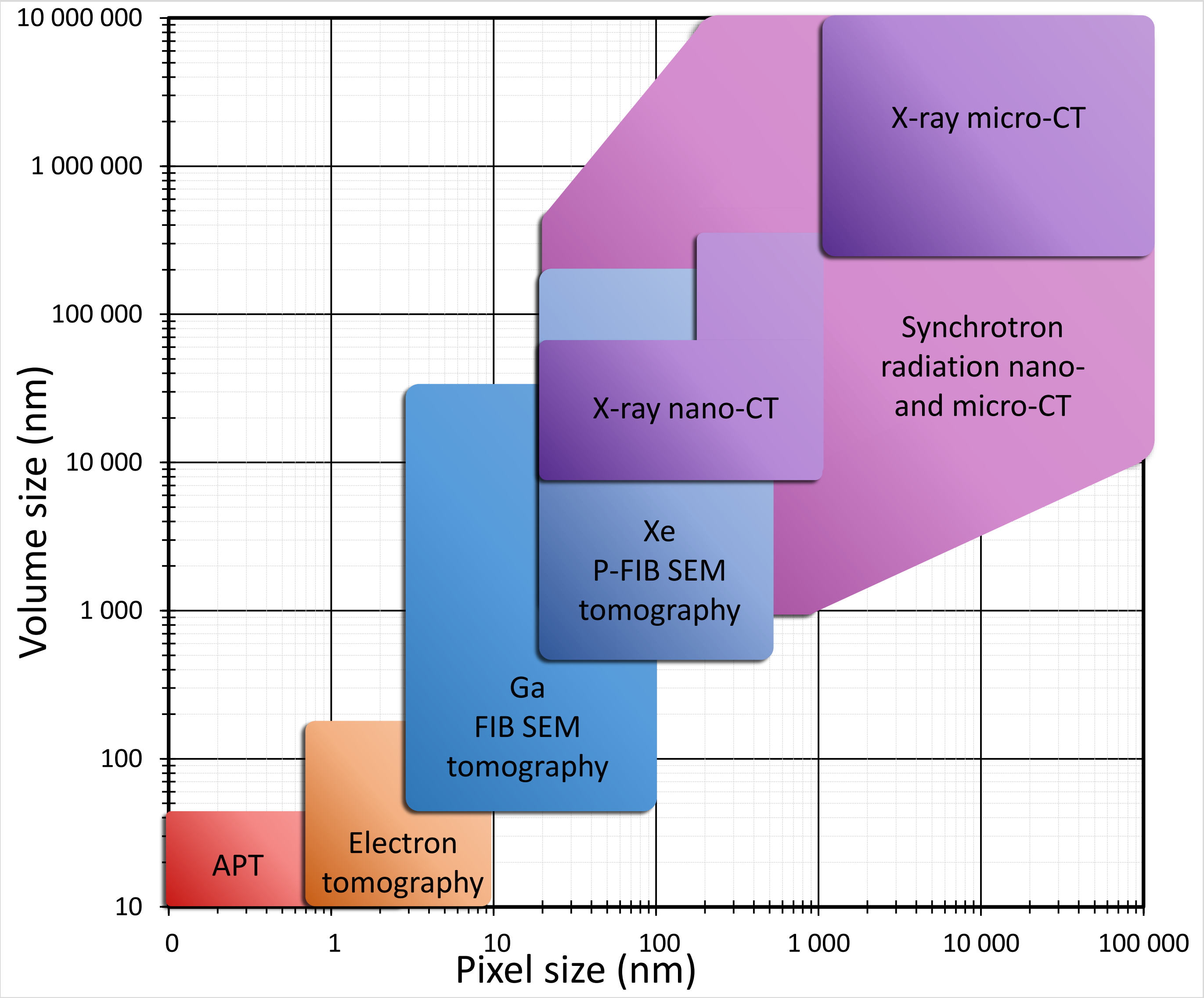
Zineb SAGHI (CEA-Leti)

3D analysis of nanoscale materials using electron and FIB-SEM tomography techniques

3D characterization techniques at the nanoscale

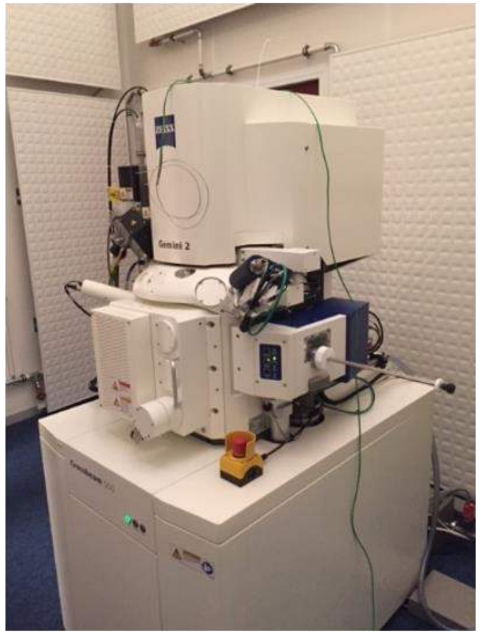


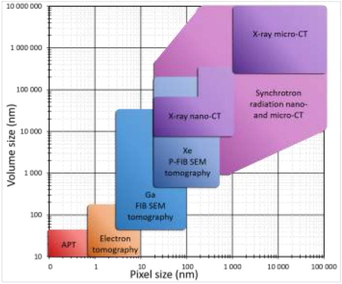
3D characterization techniques at the nanoscale

Atom probe tomography

Electron tomography

Slice-and-view tomography





Field ion microscope combined with a mass spectrometer

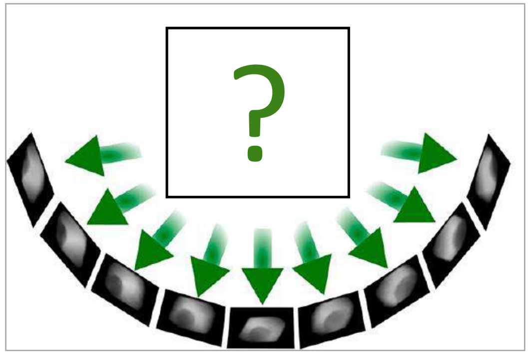
Transmission electron microscope (TEM)

Focussed ion beam combined with a scanning electron microscope (FIB-SEM)

Dimensions / Morphology: serial sectioning by FIB-SEM, BF-TEM tomography, HAADF-STEM tomography
Composition / Doping: APT, STEM-EDX/STEM-EELS tomography, FIB-EDX tomography

Key steps in electon tomography (ET)

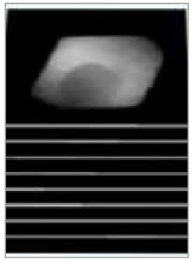
Data acquisition

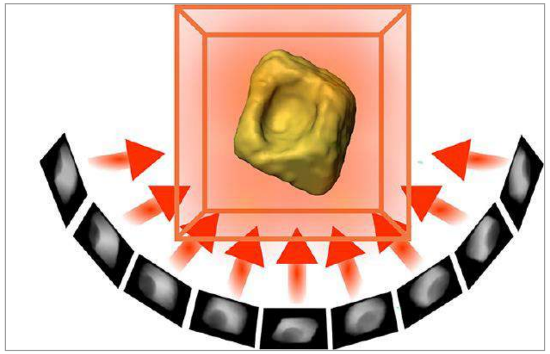


(dedicated) TEMs

Dedicated specimen holders

Pre-processing, alignment, reconstruction





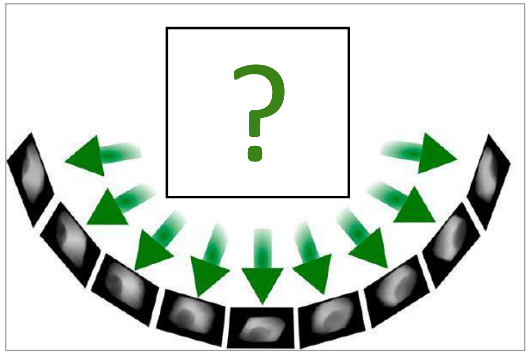
Pre-processing tools (ex: spectral analysis tools)

Alignment and reconstruction algorithms

Tools for segmentation and quantification

Key steps in electon tomography (ET)

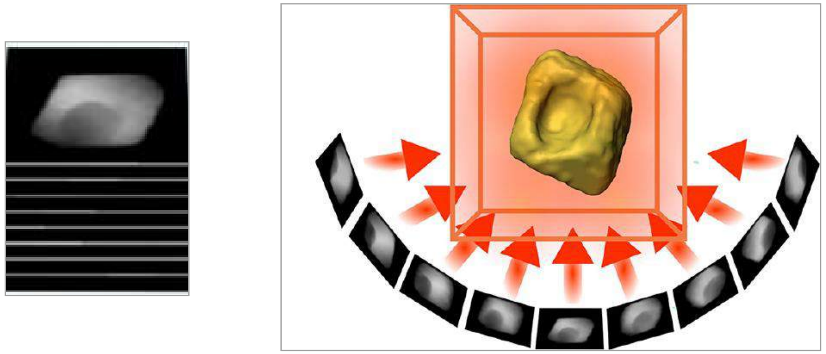
Data acquisition



(dedicated) TEMs

Dedicated specimen holders

Pre-processing, alignment, reconstruction



Pre-processing tools (ex: spectral analysis tools)

Alignment and reconstruction algorithms

Tools for segmentation and quantification

Projection requirement

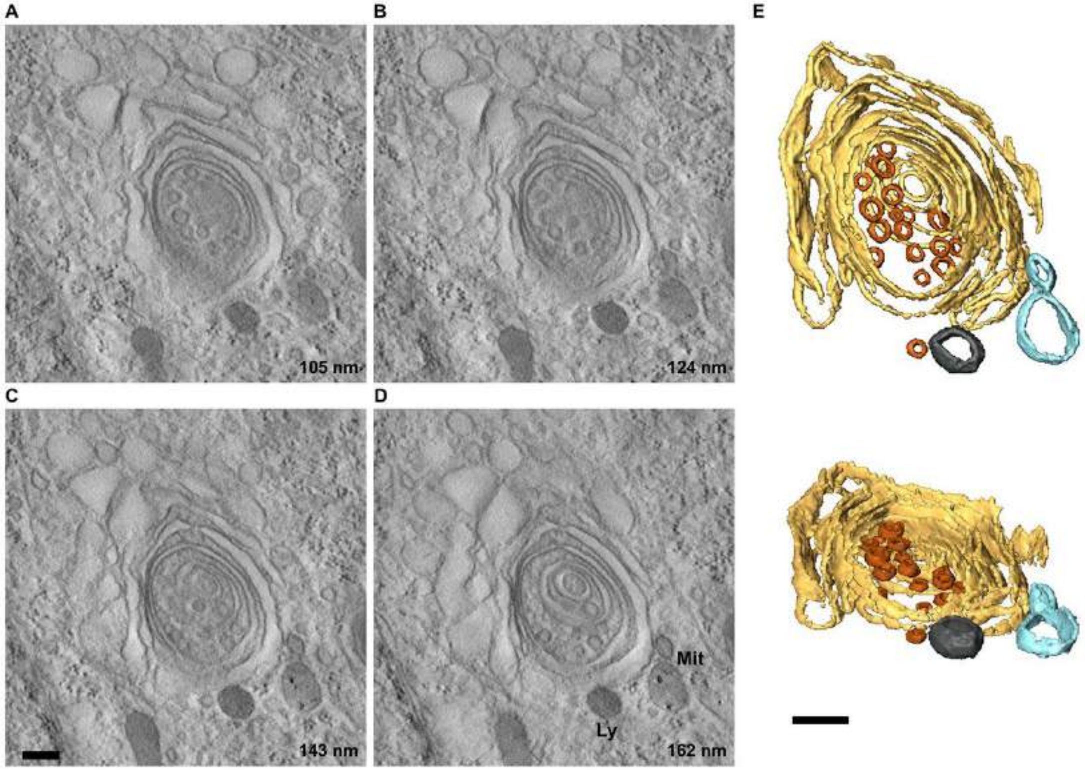
Projection requirement for ET:

"The signal used for tomographic reconstruction should be a monotonic function of a projected physical quantity."

Standard imaging modes in ET:

- Biology: BF-TEM (bright field transmission electron microscopy)

3D electron tomography of brain tissue



M.R Fernandez-Fernandez et al. Journal of Cell Science 2017, 130:83.

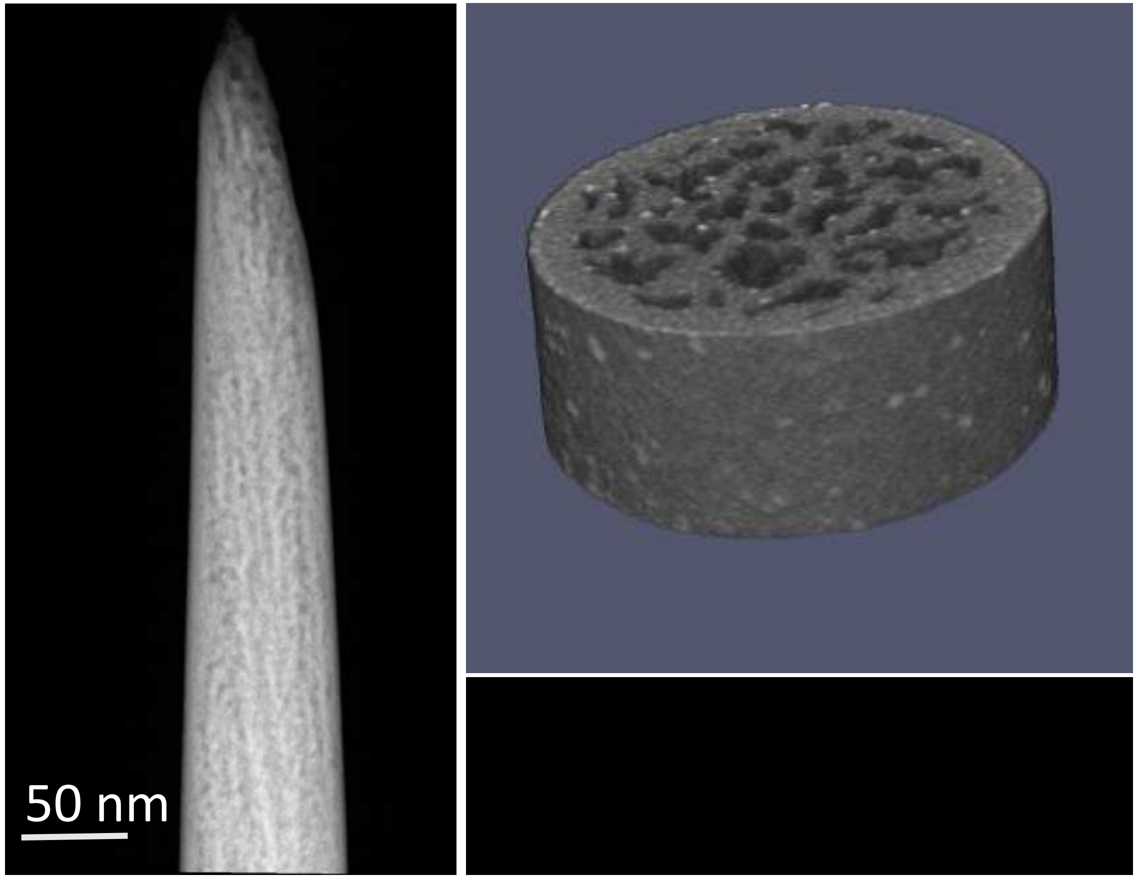
Projection requirement

Projection requirement for ET:
"The signal used for tomographic reconstruction should be a monotonic function of a projected physical quantity."

Standard imaging modes in ET:

- Biology: BF-TEM (bright field transmission electron microscopy)
- Materials science: HAADF-STEM (high angle annular dark field scanning TEM)
  - Incoherent (no diffraction contrast)
  - Z contrast ($I \sim Z^2$)

3D electron tomography of Er-doped porous Silicon



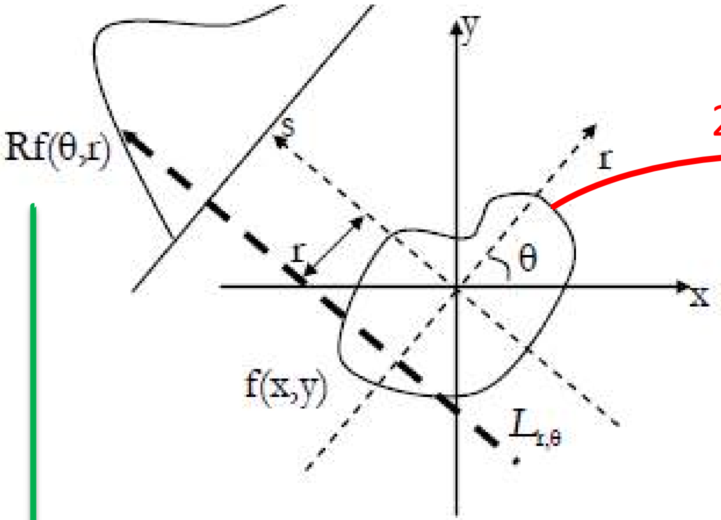
G. Mula *et al. Scientific Reports 2017,* 7:5957

Fourier slice theorem

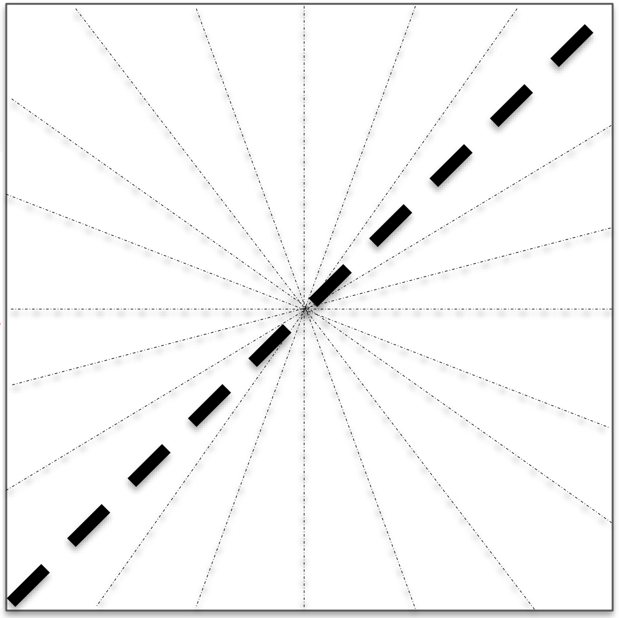
Spatial domain

Frequency domain

$$Rf(\theta,r)=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}f(x,y)\delta(x\cos\theta+y\sin\theta-r)dxdy$$



2D FT



1D FT

$$G(\theta,u)=F(u\cos\theta,u\sin\theta)$$

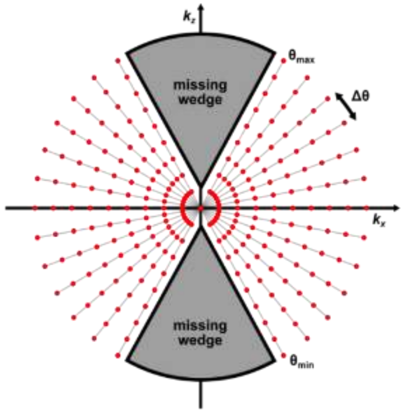
The 2D FT of the projections fill the 3D Fourier space the sample

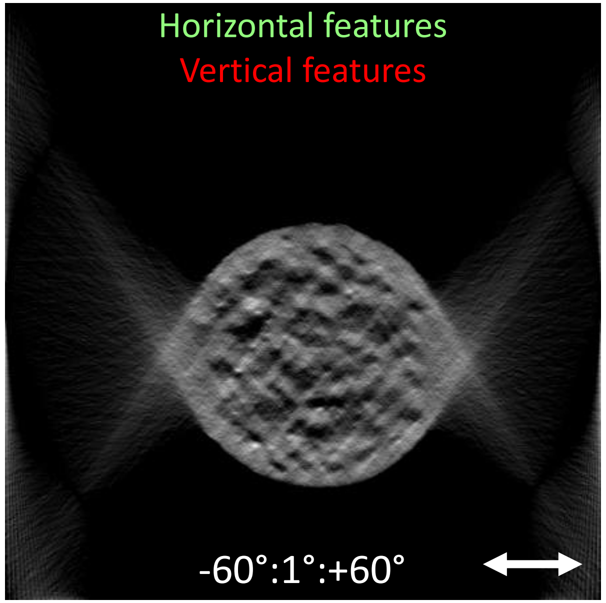
↓

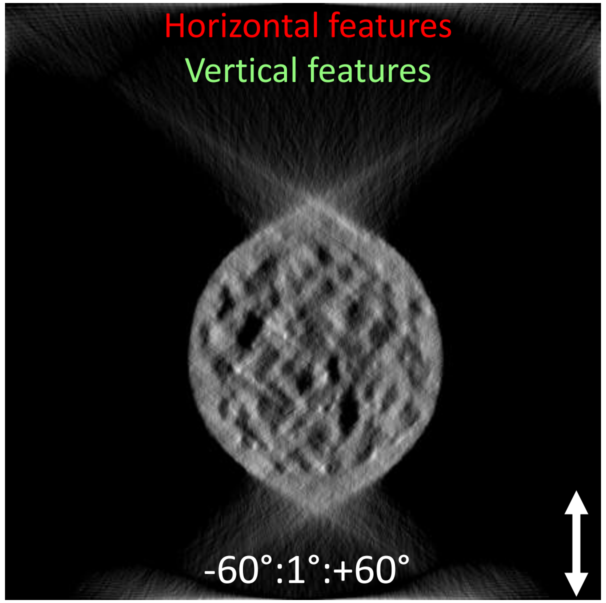
We need to acquire as many projections as possible

Dedicated specimen holders

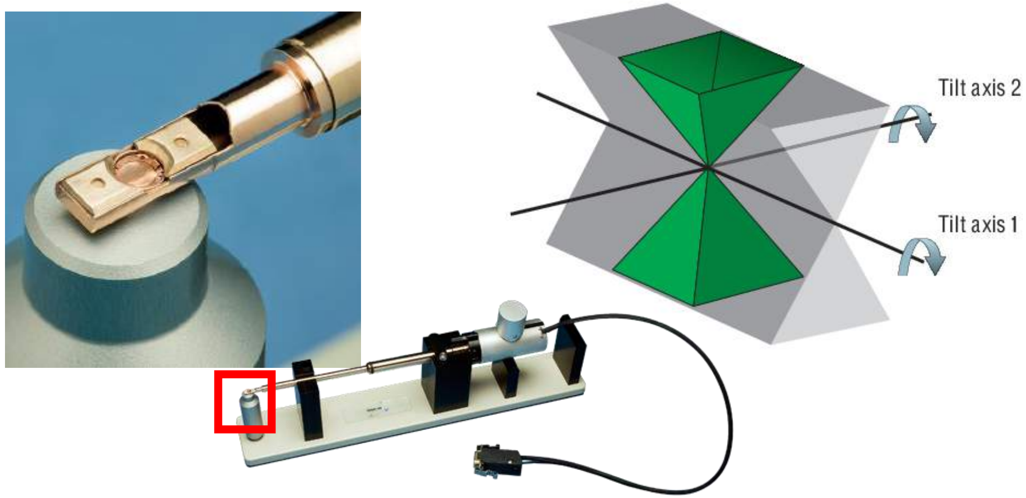
Single-axis tomography holder





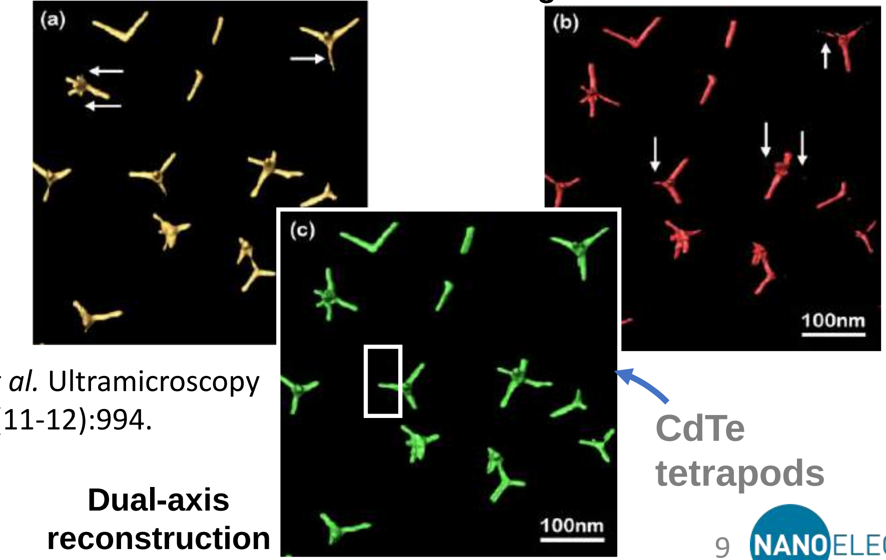


Dual-axis tomography holder



Single-axis reconstruction (1)

Single-axis reconstruction (2)



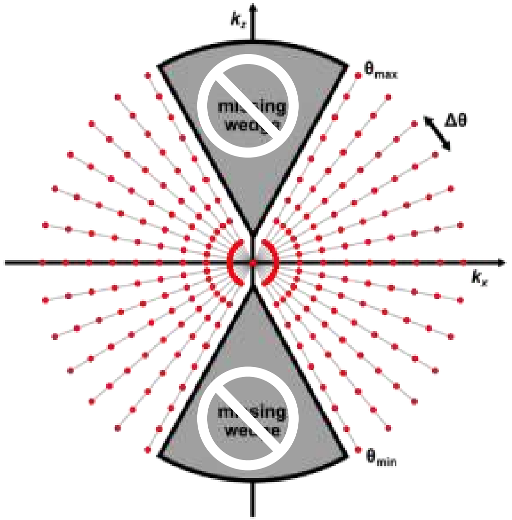
I. Arslan *et al.* Ultramicroscopy *2006,* 106(11-12):994.

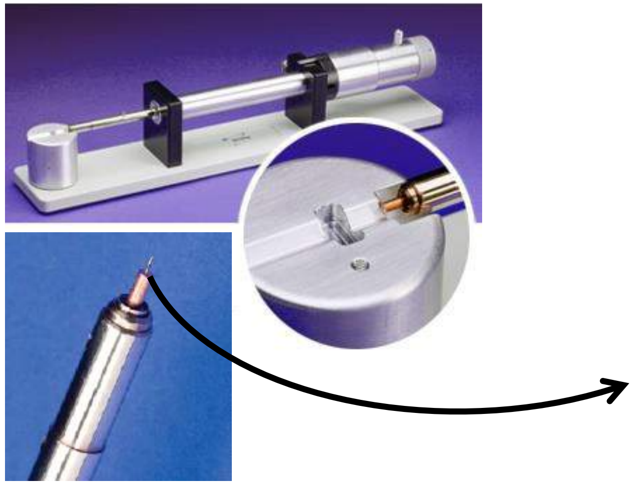
Dual-axis reconstruction

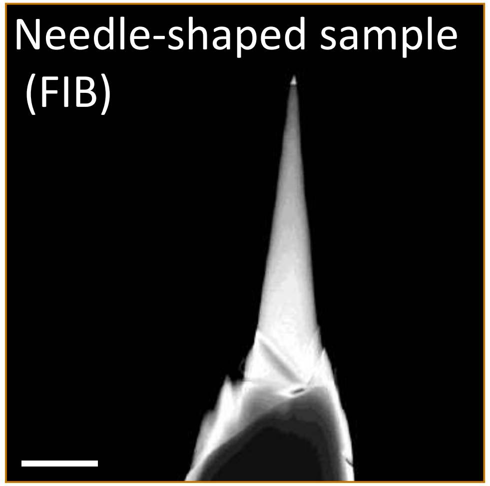
CdTe tetrapods

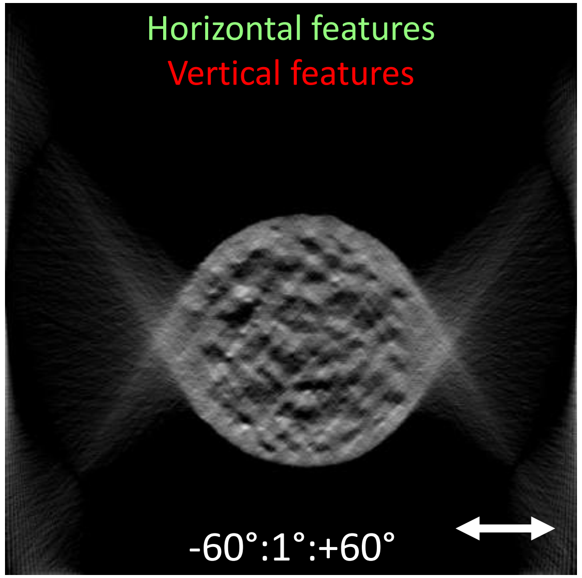
Dedicated specimen holders

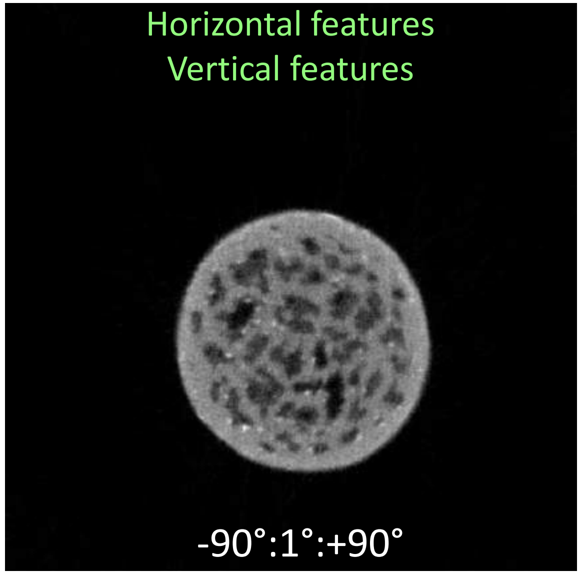
On-axis tomography holder

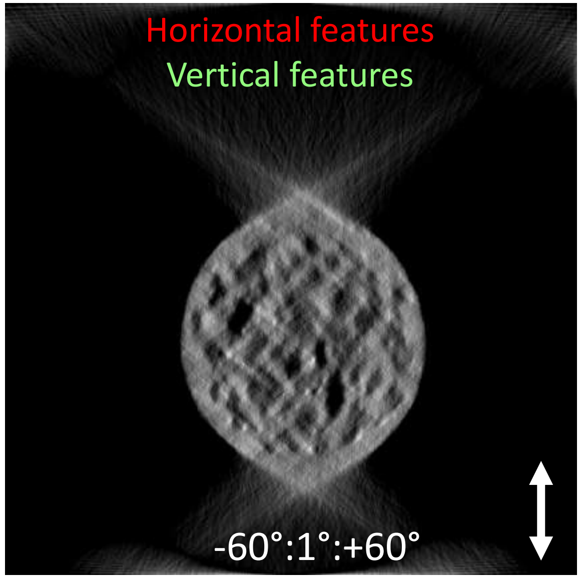




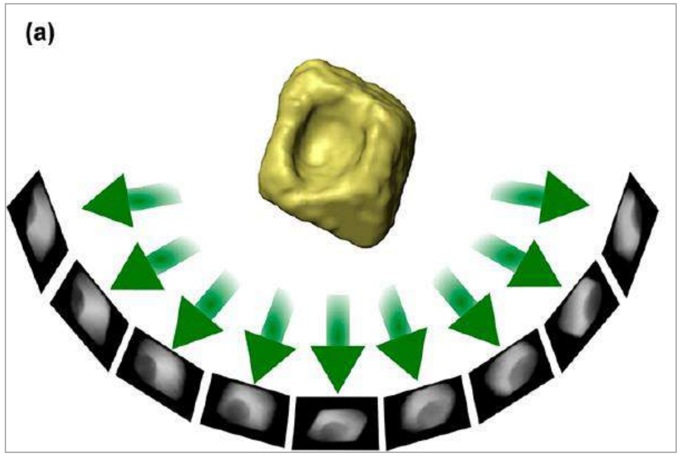




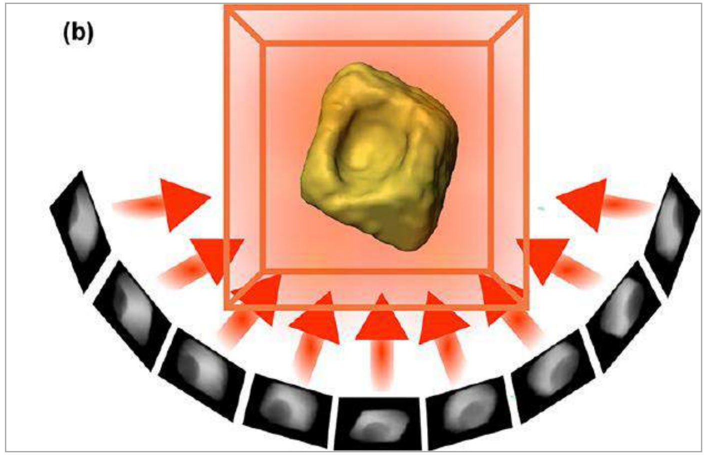




Reconstruction approaches



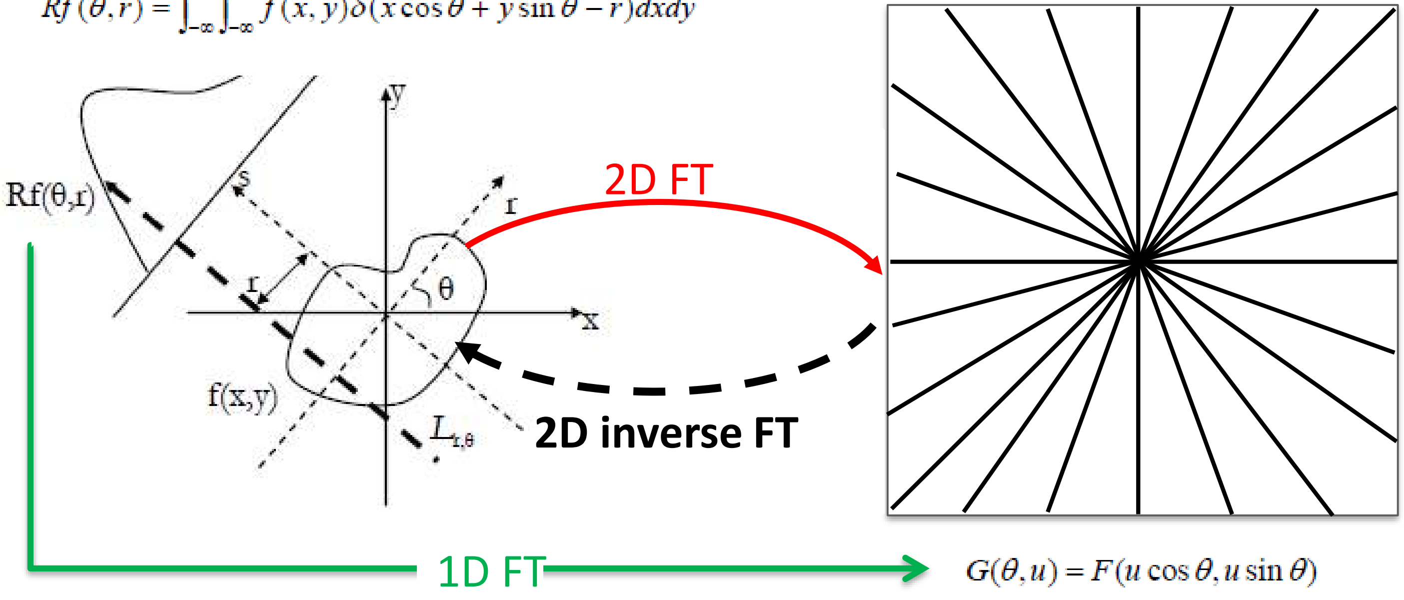
- Fourier approaches
- Analytical approaches
- Algebraic approaches



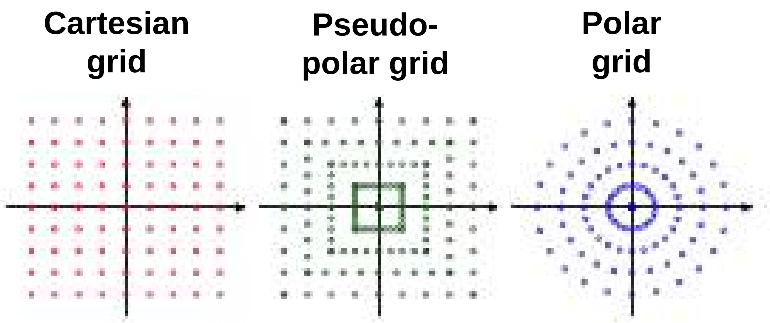
Fourier-based approaches

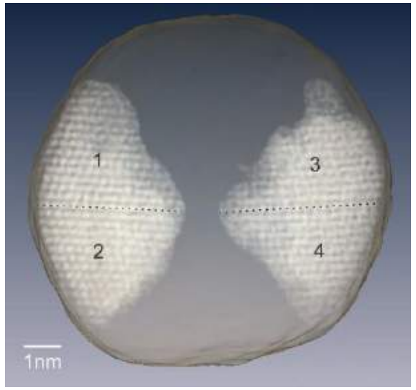
Spatial domain | **Frequency domain**

$$Rf(\theta,r)=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}f(x,y)\delta(x\cos\theta+y\sin\theta-r)dxdy$$



$$G(\theta,u)=F(u\cos\theta,u\sin\theta)$$



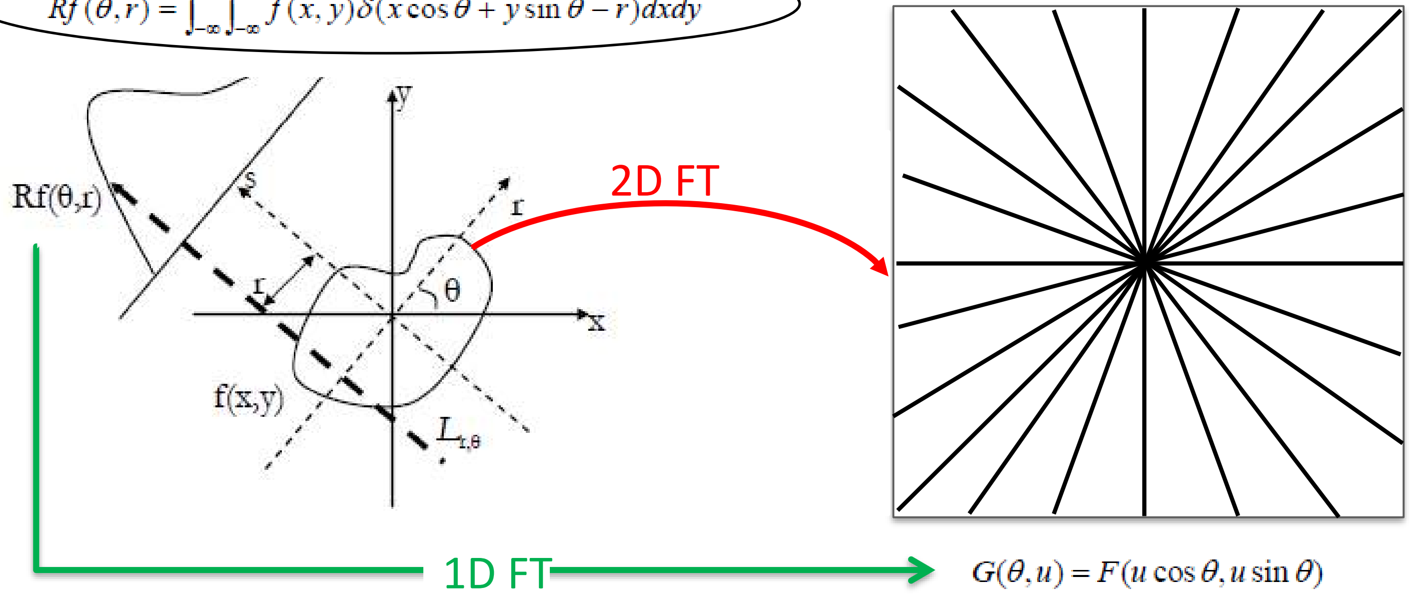


- Direct Fourier methods
  - Polar to cartesian grid interpolation induces many artefacts
- Equally slopped tomography (EST)
  - Equally slopped projections => pseudo-polar FFT (M. C. Scott et al., *Nature*, 2012, 483:444)
- Min-max non-uniform FFT (NUFFT)
  - Convolution of the non-uniformed samples with a kernel + resampling on an oversampled cartesian grid (Fessler, *IEEE T-SP,* 2003, 51(2):560)
  - Widely used in MRI

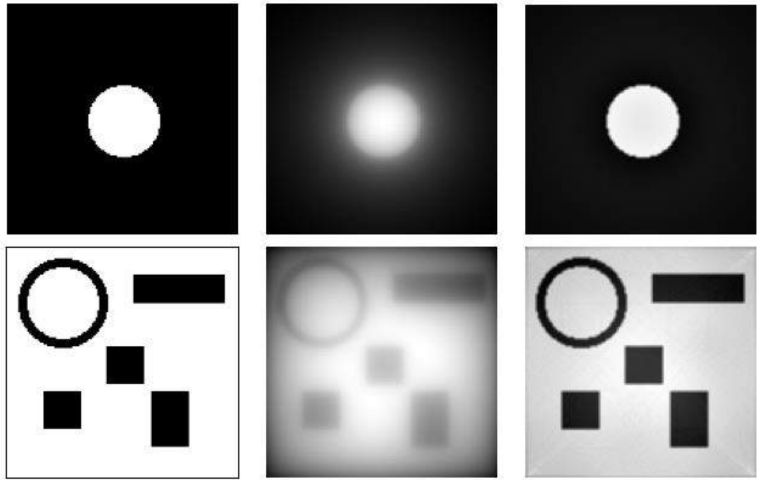
Analytical methods

Spatial domain | **Frequency domain**

$$Rf(\theta,r)=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} f(x,y)\delta(x\cos\theta+y\sin\theta-r)\,dxdy$$

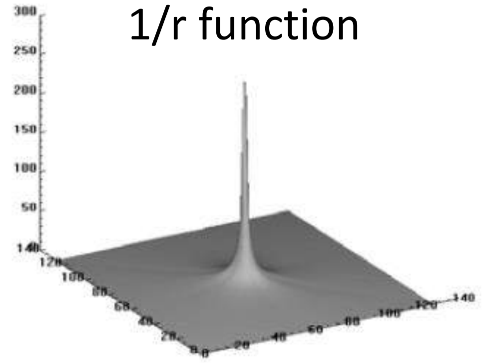


$$G(\theta,u)=F(u\cos\theta,u\sin\theta)$$



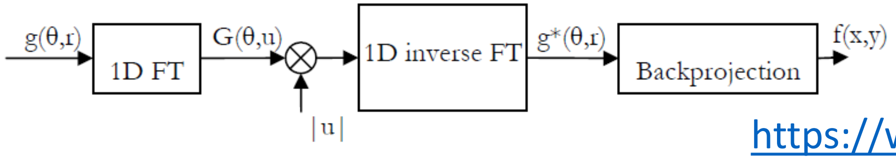
Original | Backprojection | Filtered backprojection

A simple backprojection *b* => a blurred version of the original object *f:* $b(x,y)=f(x,y)\otimes\frac{1}{\sqrt{x^2+y^2}}$



1/r function

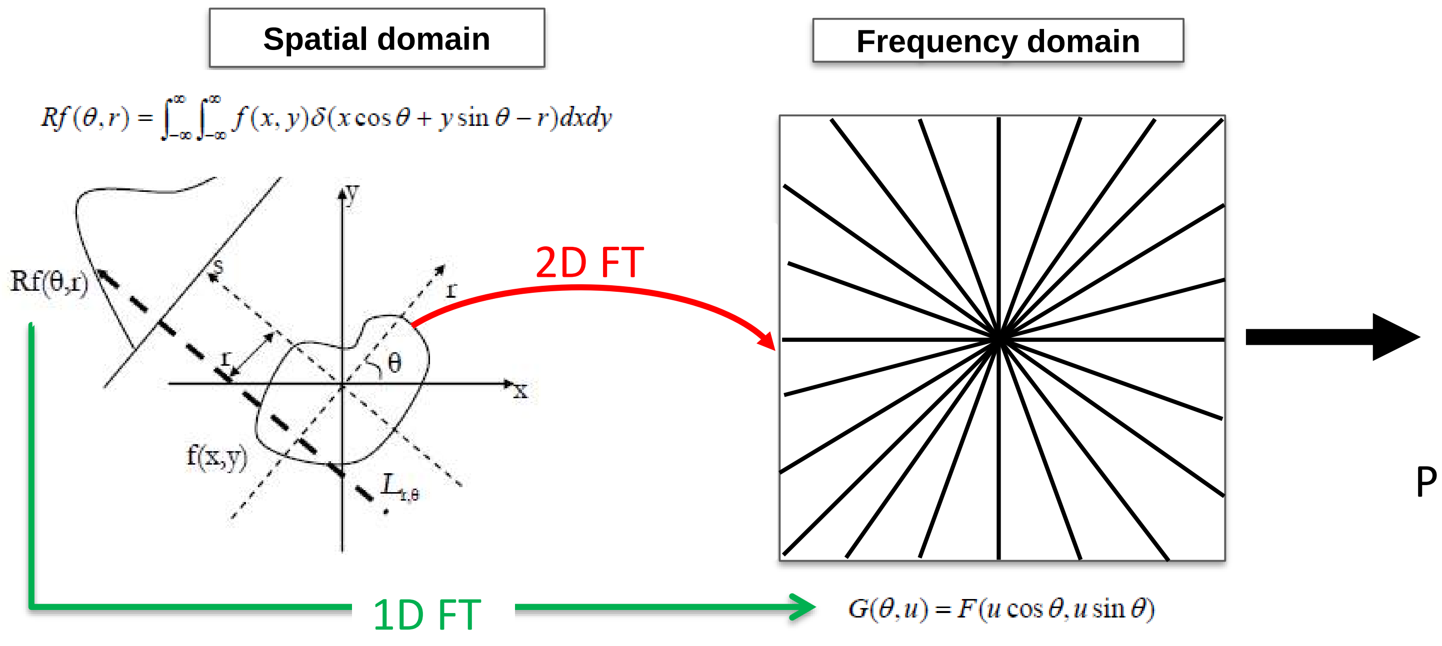
Solution: apply a ramp filter to compensate for this blurring *:* $f(x,y)=FT^{-1}(B(u_1,u_2)\times|u|)$



$$|u|=\sqrt{u_1^{\,2}+u_2^{\,2}}$$

https://web.eecs.umich.edu/~fessler/course/516/l/c-tomo.pdf

Algebraic methods



Solution of a linear equation system:

$$Px = y$$

Projection matrix

The object to reconstruct

Experimental data

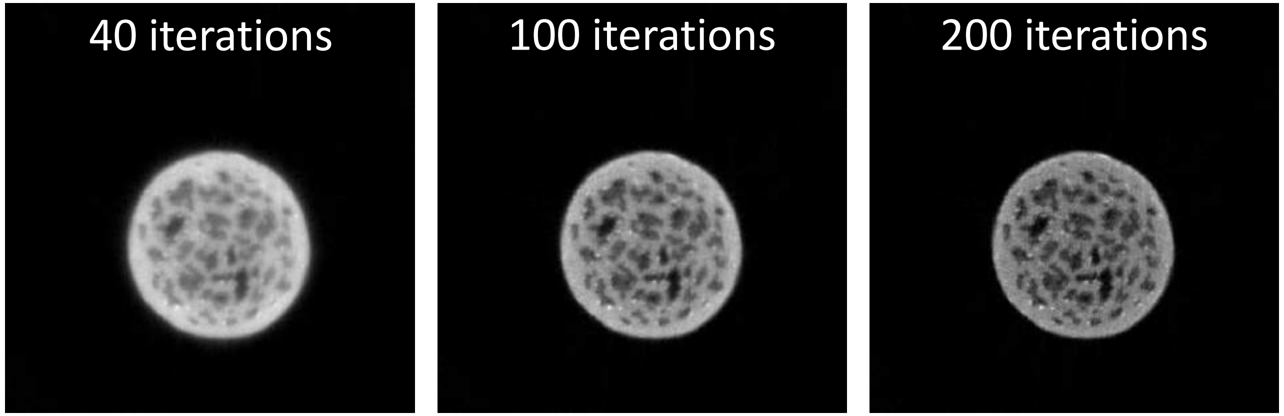
Various algorithms exist for solving this problem: Algebraic reconstruction technique (ART), Simultaneous ART (SART), Simultaneous iterative reconstruction technique (SIRT), etc

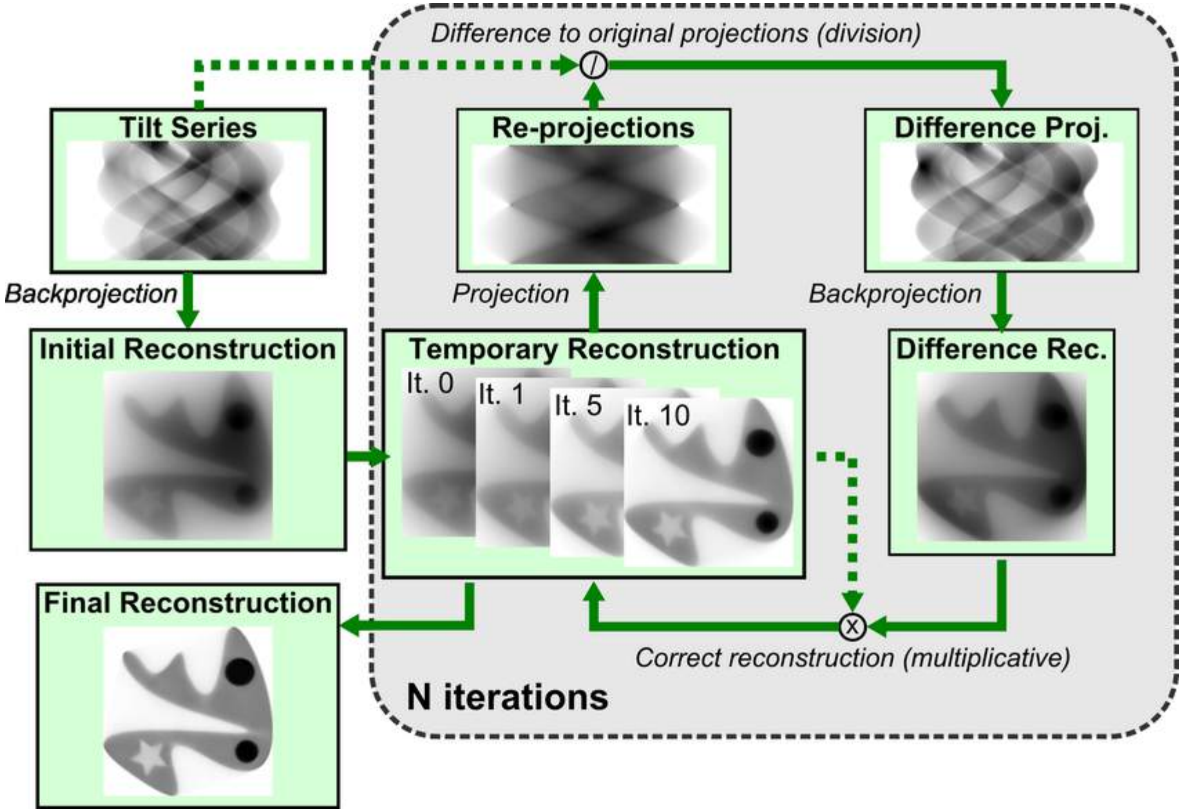
C.OS. Sorzano et al., Biomed Res Int. 2017, 6482567.

SIRT

SIRT solves the following least squares problem:

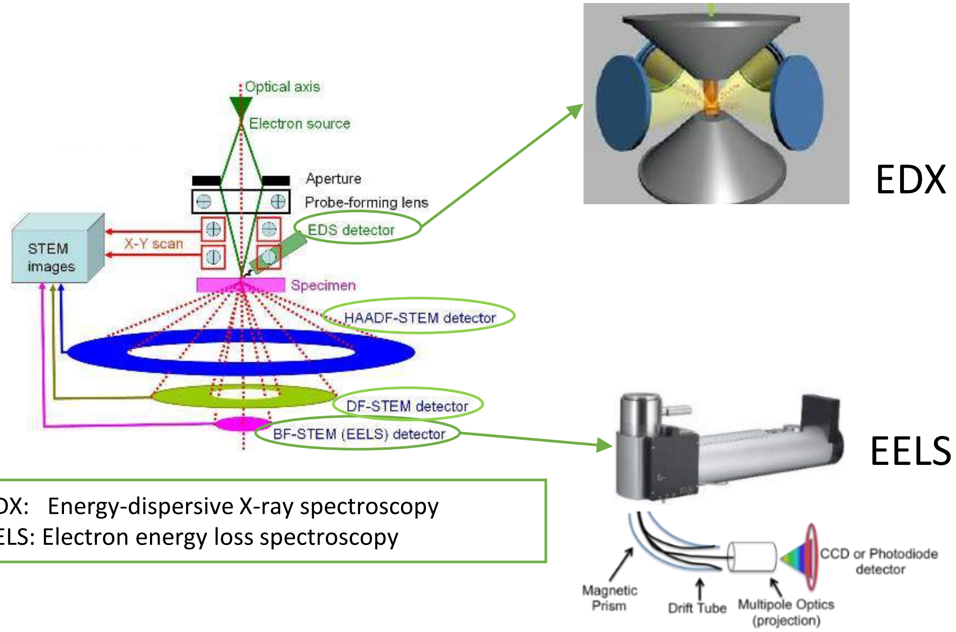
$$\underset{x}{\operatorname{argmin}} \left\{ \frac{1}{2} \, \|Px - y\|_2^2 \right\}$$





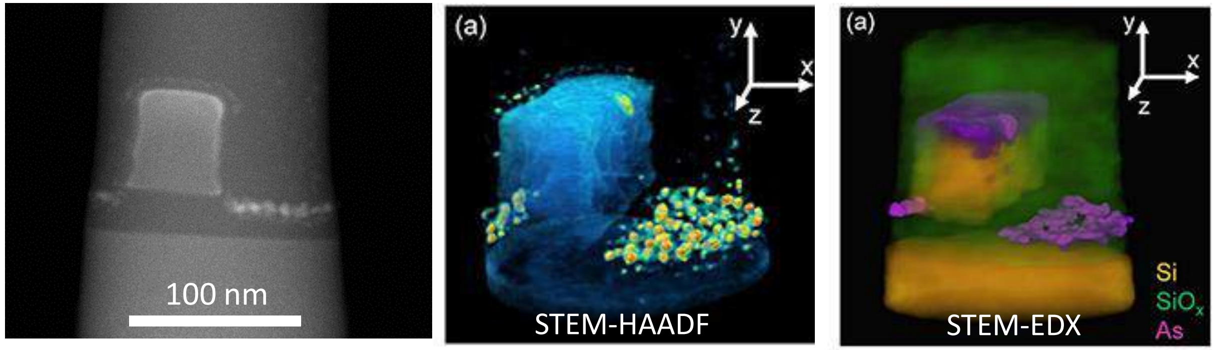
G. Haberfehlner, *3D nanoimaging of semiconductor devices and materials by electron tomography*, 2013.

Spectroscopic ET



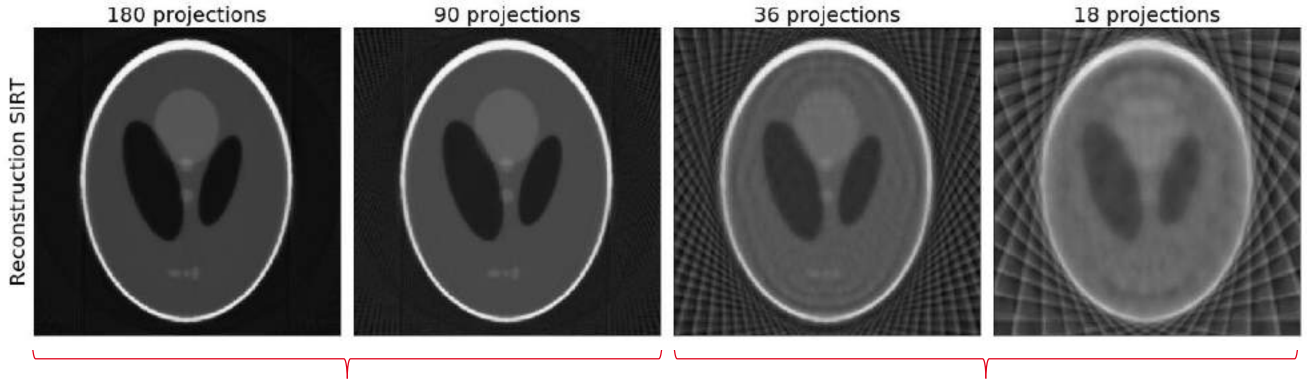
EDX: Energy-dispersive X-ray spectroscopy
EELS: Electron energy loss spectroscopy

2D STEM-HAADF projection	3D morphological information	3D chemical information



M. Jacob et al., Semicond. Sci. Technol. 2021, 36: 035006.

- **Challenges (compared to STEM-HAADF):**
  - Long acquisition times
  - High electron doses
  - Poor-quality reconstructions using standard algorithms



STEM-HAADF tomography | Spectroscopic ET

Advanced algorithms for undersampled datasets

Add prior knowledge

Discrete ART (DART)

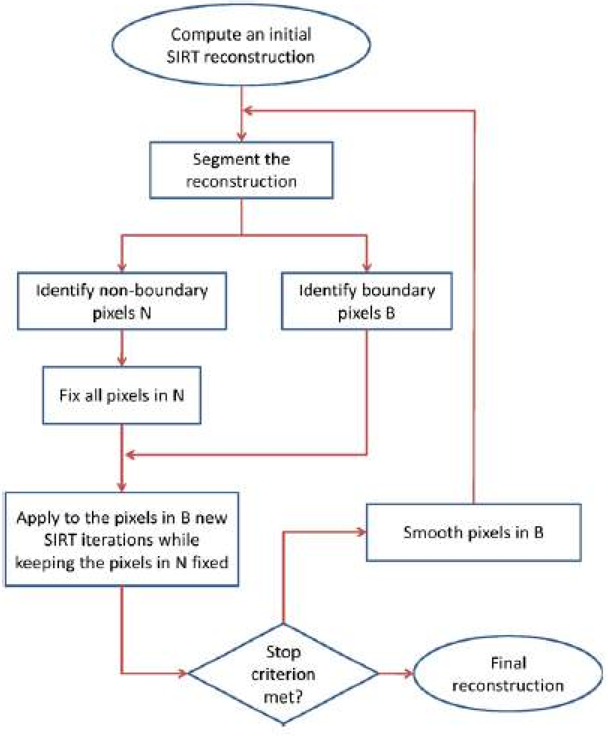
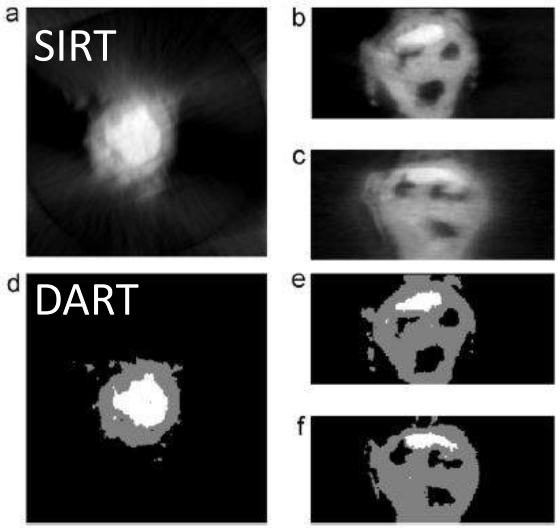


Fig. 2. Flow chart of the DART algorithm.



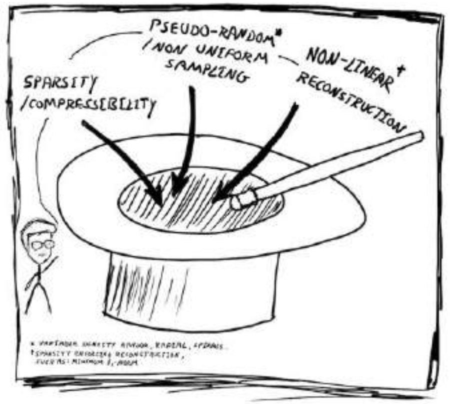
Prior knowledge: number of grey levels, and their intensities.

S. Bals et al. Nano Lett. 2007, 12:3669
K.J. Batenburg et al. Ultramicrosc. 2009, 109: 730

Compressed sensing framework (CS)

$$\underset{x}{\operatorname{argmin}} \left\{ \frac{1}{2} \|Px - y\|_2^2 + \lambda \mathcal{R}(x) \right\}$$

$\mathcal{R}(x) = \|Lx\|_0$ promotes sparsity of x in the transform domain L (few non-zero elements in Lx).



Michael Lustig,
http://www.eecs.berkeley.edu/~mlustig/CS.html

$$\underset{x}{\operatorname{argmin}} \left\{ \frac{1}{2} \|Px - y\|_2^2 + \lambda \|Lx\|_0 \right\}$$

$$\downarrow$$

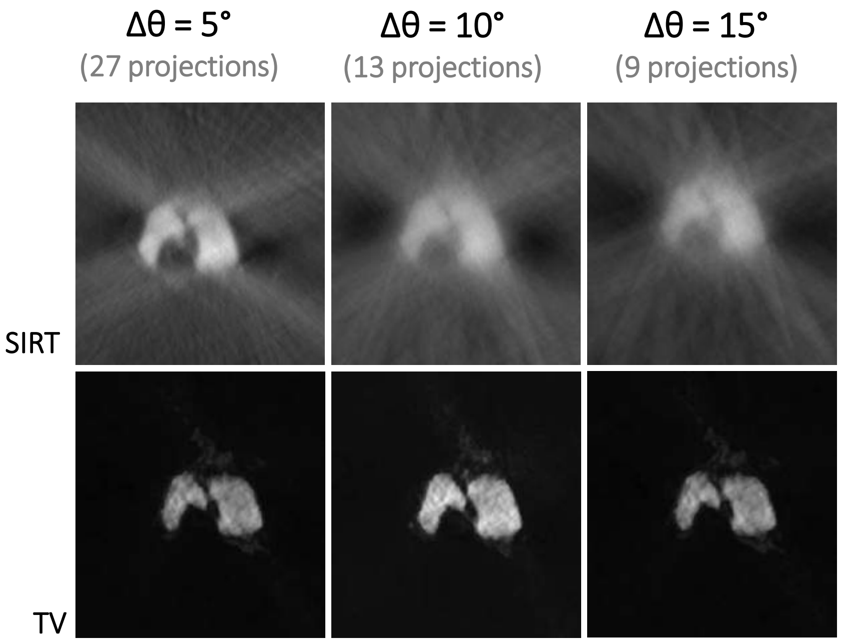
$$\underset{x}{\operatorname{argmin}} \left\{ \frac{1}{2} \|Px - y\|_2^2 + \lambda \|Lx\|_1 \right\}$$

Convex optimization formulation

Compressed sensing for undersampled datasets

Total variation minimization (TV):

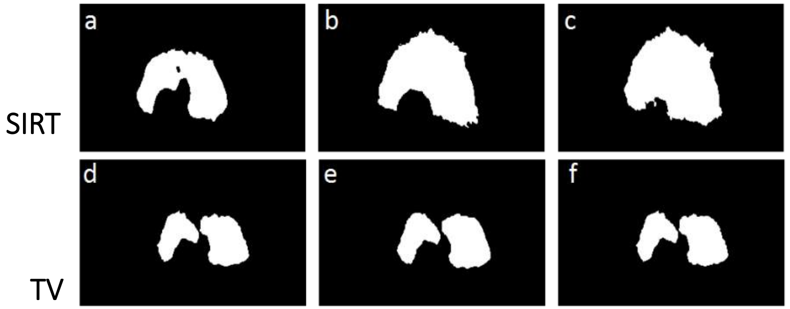
TV promotes sparsity in the gradient domain and is very well suited for objects with *piecewise constant regions and sharp edges*

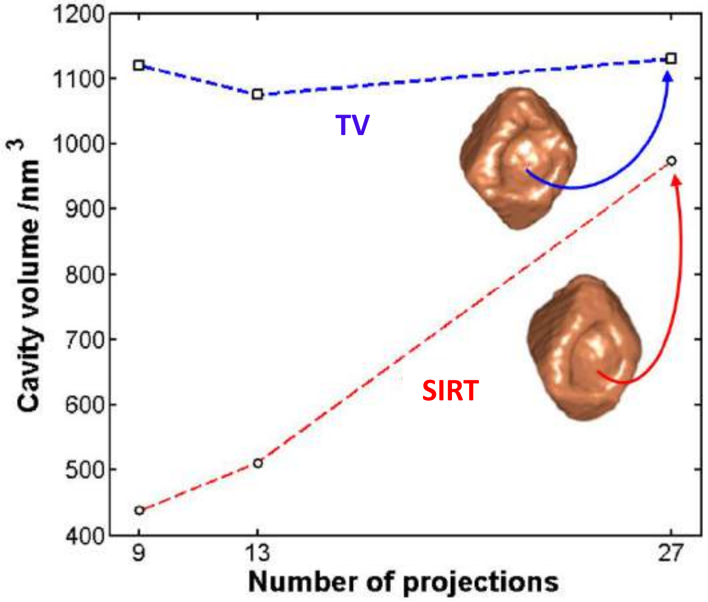


SIRT: $\hat{f} = \underset{f}{argmin} \ \|\mathbb{R}f - p\|_2^2$

Total variation (TV): $\hat{f} = \underset{f}{argmin} \, \lambda\|f\|_{TV} + \|\mathbb{R}f - p\|_2^2$

with: $\|f\|_{TV} = \int |\nabla f| dx$





Z. Saghi et al., Nano Letters 2011, 11(11), 4666.
R. Leary et al., Ultramicroscopy 2013,131, 70.

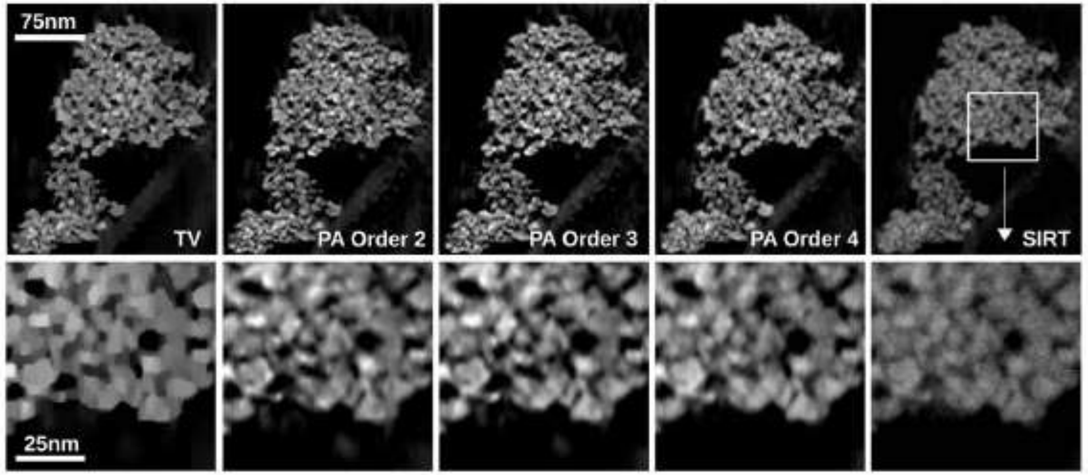
Compressed sensing for undersampled datasets

Limitations of TV:

- Staircase artefacts when the object is not truely piecewise constant.
- Complex structures require more projections than reported for simple objects (Y. Jiang et al. Ultramicrosc. 2017, 186)
- The quality of the reconstructions degrade rapidly in the presence of Poisson noise.

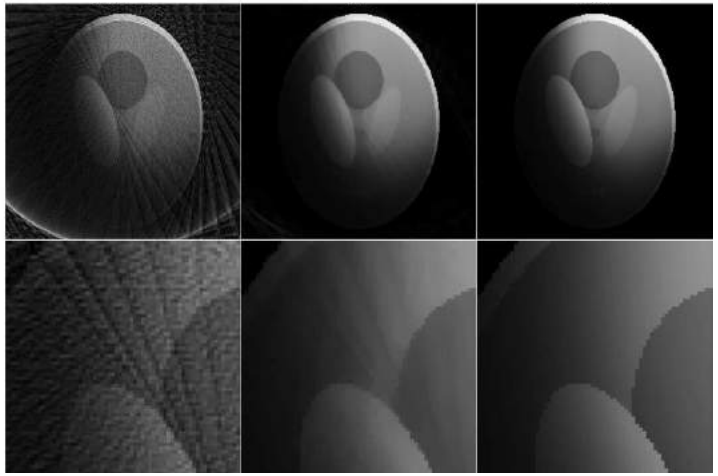
Higher order TV (Incorporation of higher order derivatives):

- ***HOTV*** *(ref: R. Archibald et al. J. Sci. Comput. 2015, 67) and* ***TGV*** *(total generalized variation, see e.g.: M. Benning et al. J. Sci. Comput. 2013, 54:269)*
- *Promote piecewise smooth regions while preserving sharp edges.*

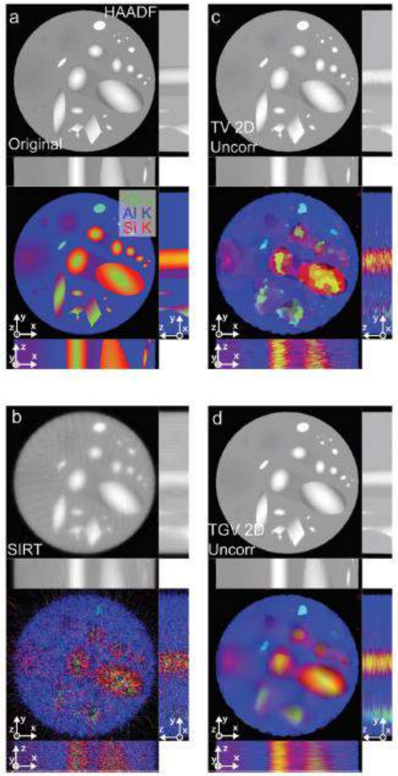


T. Sanders et al., Ultramicrosc. 2017, 174: 97.

NUFFT TV TGV



F. Knoll et al., Magn. Reson. Med 2011, 65(2):480.



TV

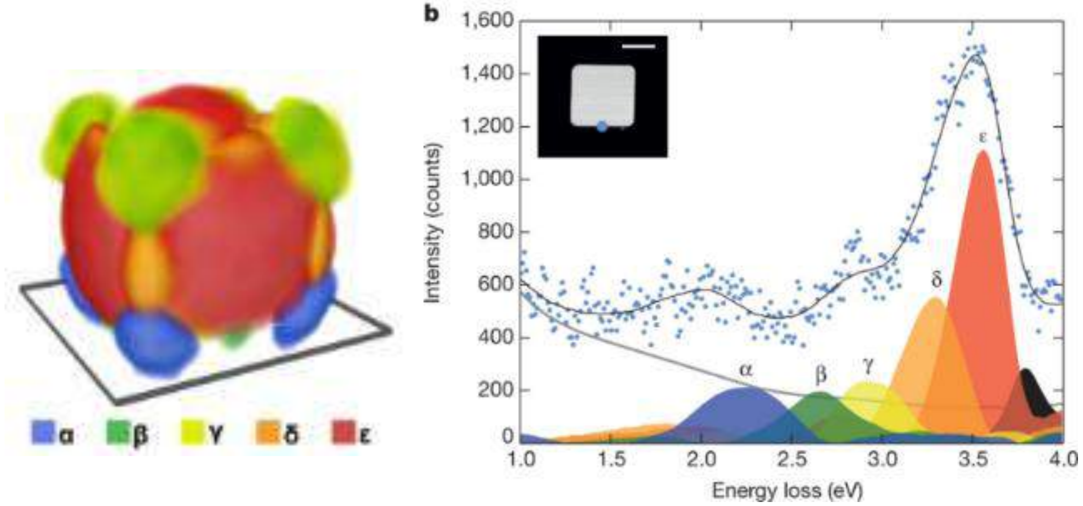
TGV

R. Huber et al., Nanoscale 2019, 11:5617.

Sparsity in the wavelet domain:

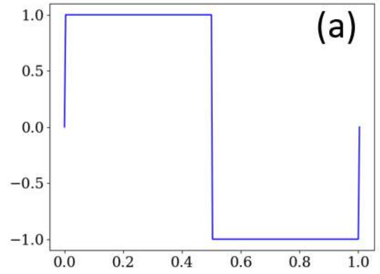
- Multiscale approach used in microscopy mainly for denoising purposes.
- Widely used in MRI and recently applied to CT and ET.
- Applied to a wider range of objects, compared to gradient sparsity.
- Knowledge about the sample => choice of the appropriate wavelet function.

First application in spectroscopic ET using Coiflets



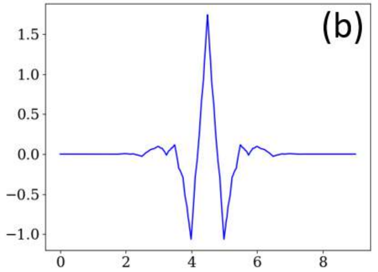
Three-dimensional imaging of localized surface plasmon resonances of metal nanoparticles.
Nicoletti *et al.* Nature 2013, 502:80

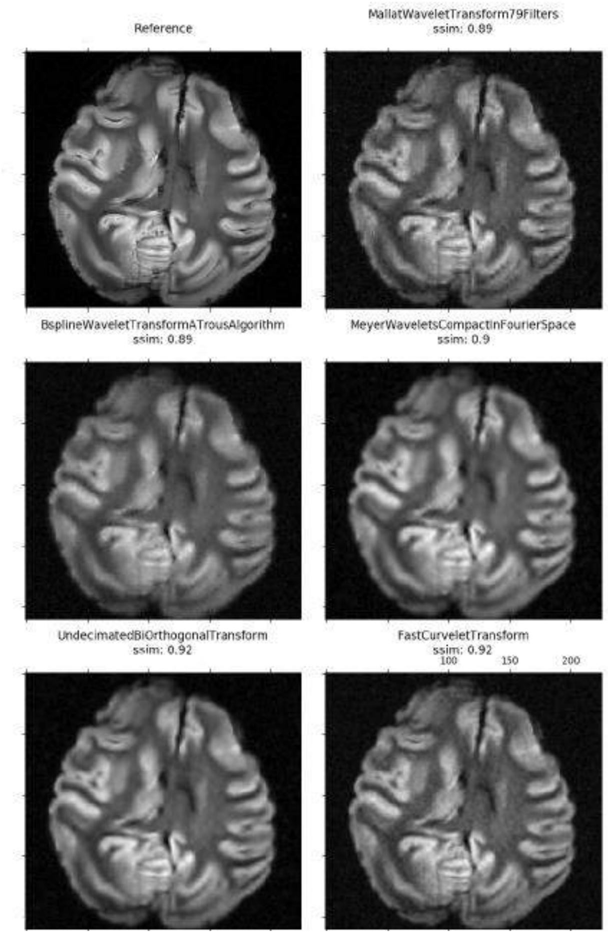
Haar



(multi-scale gradient)

Bior 4.4





H. Cherkaoui et al. EUSIPCO 2018, 36

Open-source packages for 3D reconstruction

Commercial software: Inspect3D (FEI), etc.
Commercial software with advanced algorithms: NONE.

Name	General features	Reconstruction algorithms
Astra toolbox	Initially developed for X-ray tomography GPU-based implementations. Incorporated in Inspect3D (FEI)	Standard reconstruction algorithms + TV plugin
Tomopy	Initially developed for X-ray tomography CPU-based implementations. Recently integrated with Astra toolbox.	Standard reconstruction algorithms
Tomotools	Developed for electron tomography Tools for alignment + reconstruction.	Standard reconstruction algorithms (Astra + Tomopy)
Tomviz	Open source plateform for alignment, reconstruction and visualization.	Standard reconstruction algorithms + TV
Matlab package (T. Sanders et al., Ultramicroscopy 2017, 174: 97)	Alignment, inpainting and denoising.	TV, HOTV, and multiscale HOTV.
Graptor (R. Huber et al., Nanoscale 2019, 11:5617)	Developed for EELS/EDS tomography GPU-based implementations.	TGV algorithm and multi-modal reconstruction approach
PySAP-etomo (M. Jacob et al., Ultramicroscopy 2021, 255: 113289)	*Adaptation of PySAP + Modop libraries* *Includes Astra and Pywavelet.*	*Implementation of gradient-based (TV, TGV, HOTV) and wavelet-based methods.*

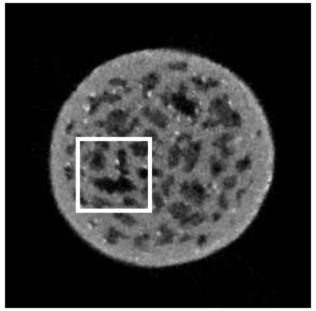
Application 1: HAADF-STEM tomography of Er-doped porous Silicon

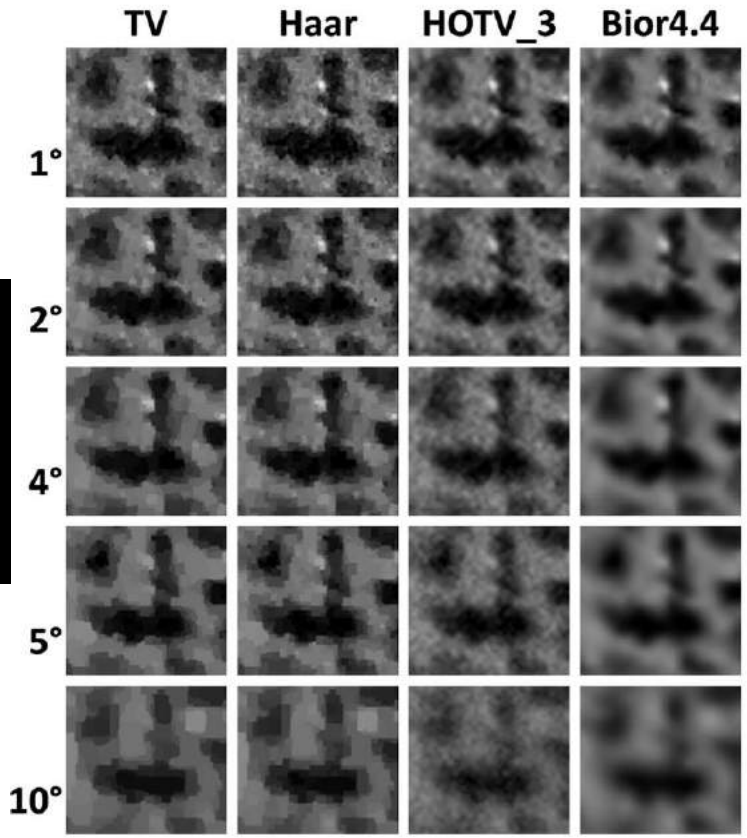
HAADF-STEM mode
Tilt angles: -90°:1°:+90°

TV/Haar: staircasing artifacts due to the piecewise constancy assumption.

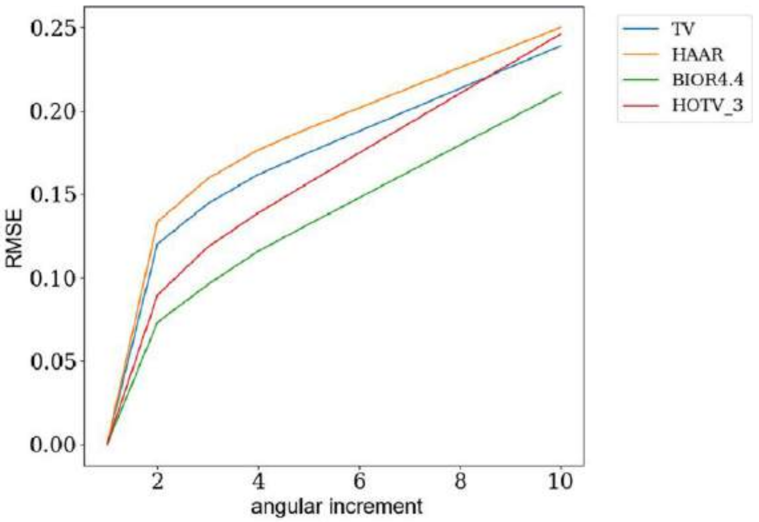
HOTV_3 : no staircasing artifacts. Noise-like oscillations appear with large tilt increments.

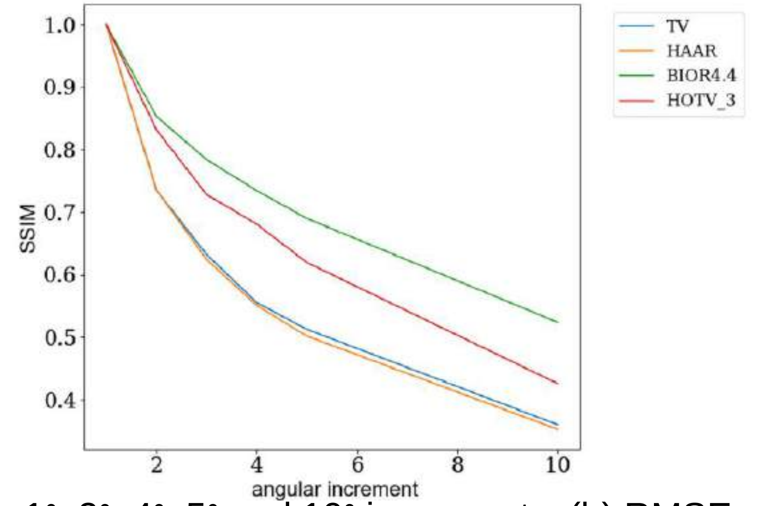
Bior4.4: best results. Induces smoothness with large tilt increments





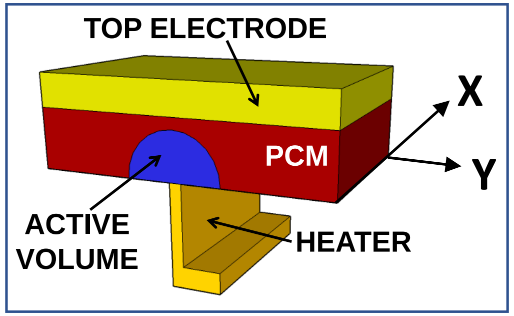
M. Jacob et al., Ultramicroscopy 2021, 255: 113289



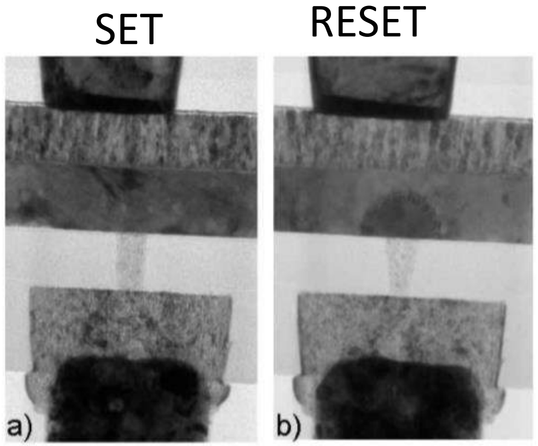


TV, Haar, HOTV_3 and Bior4.4 reconstructions with 1°, 2°, 4°, 5° and 10° increments. (b) RMSE and (c) SSIM scores showing the evolution of the image quality as function of the angular increment (in degrees).

Application 2: STEM-EELS tomography of a Ge-rich GeSbTe (GST) thin film for phase-change memory (PCM) applications

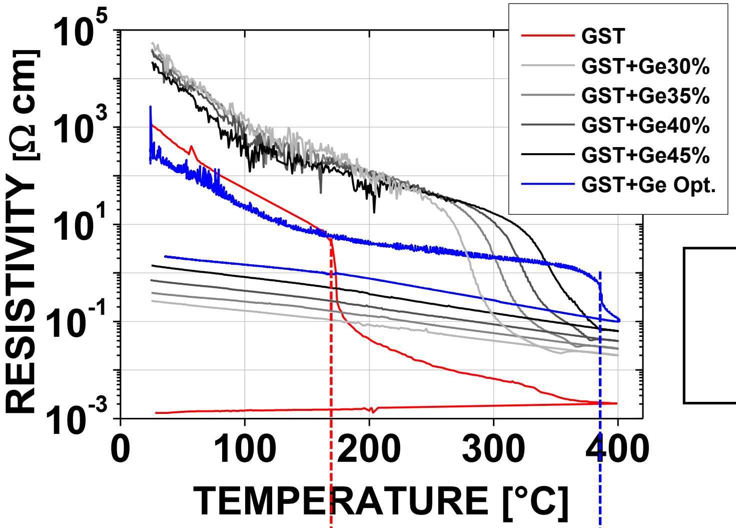


Example of an industrial PCM device



From: Phase change materials: science and applications, S. Raoux and M. Wuttig, Springer Verlag, New York, 2009.

Current challenge: PCM memory devices stability is challenged at high temperature (automotive applications).



$Ge_2Sb_2Te_5$
T_c~170°C

Ge-rich GST
T_c~380°C

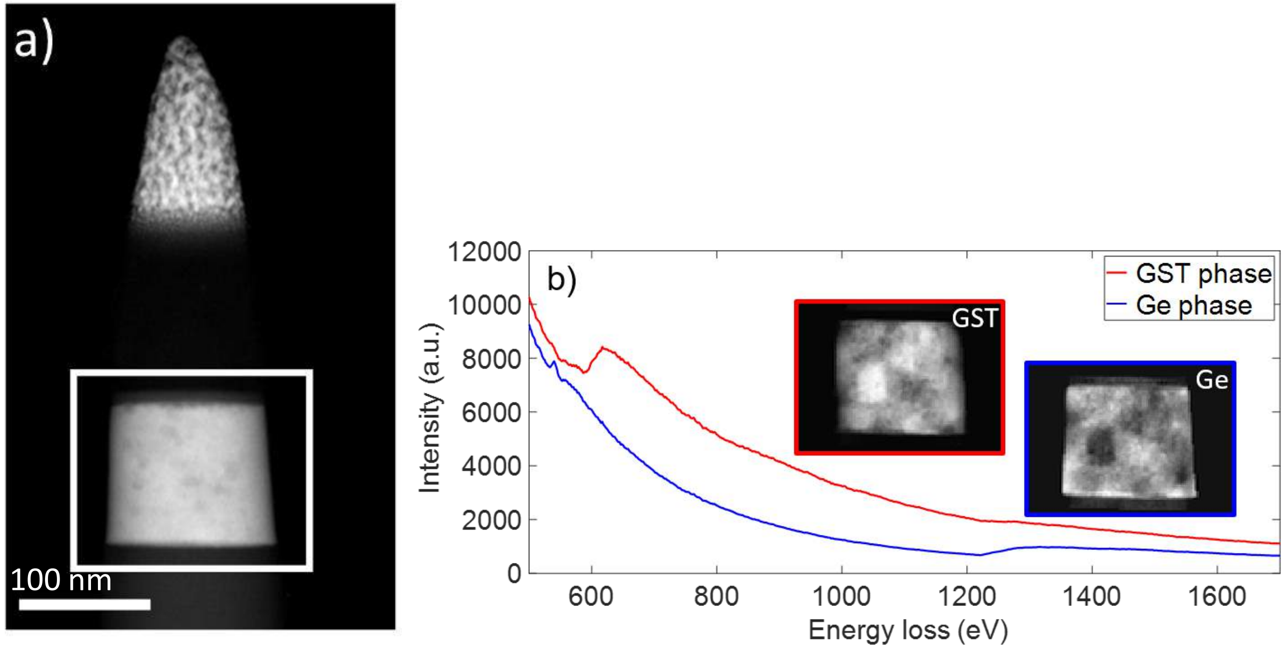
Ge-rich GST crystallisation
⇩
Phase separation GST & Ge

Increase of Ge at. %
⇩
Increase of crystallization temperature (T_c)

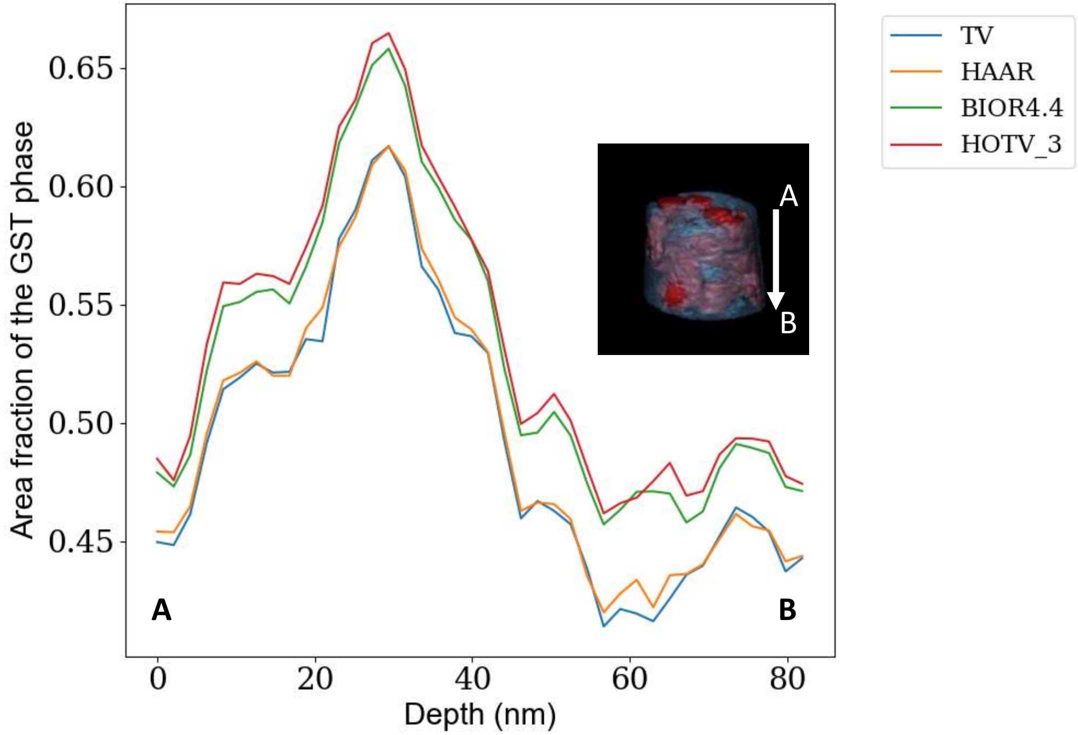
Zineb SAGHI

Application 2: STEM-EELS tomography of a Ge-rich GeSbTe (GST) thin film for phase-change memory (PCM) applications

STEM-EELS mode
Tilt angles: -90°:10°:+90°



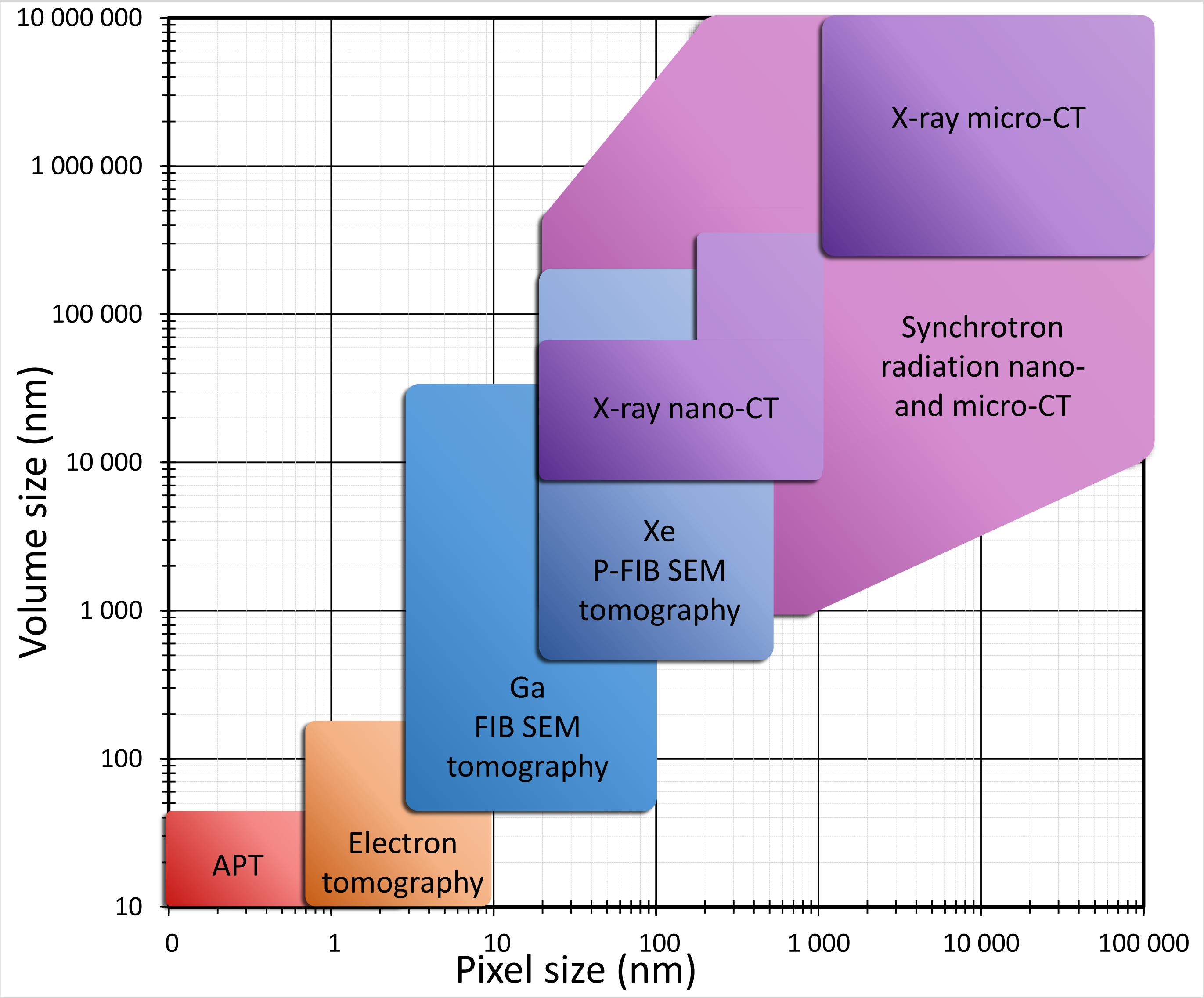
M. Jacob et al., Ultramicroscopy 2021, 255: 113289



Volume fraction of the GST phase:

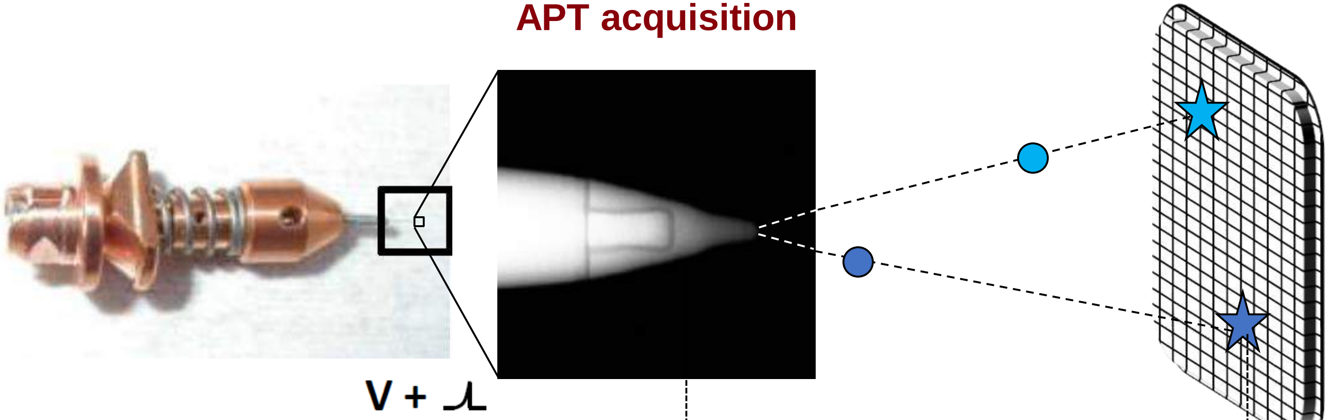
Bior4.4: 0.527 Haar: 0.494
HOTV_3: 0.533 TV: 0.491

3D characterization techniques at the nanoscale



Atom probe tomography (APT)

APT acquisition

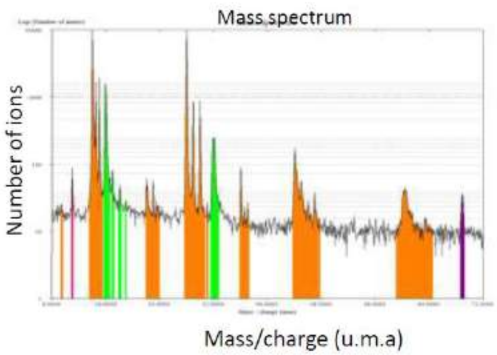


Continuous voltage +
electric or laser impulsion
(V ≈ 3-15 kV, T ≈ 20-80 K, E> 10 V/nm)

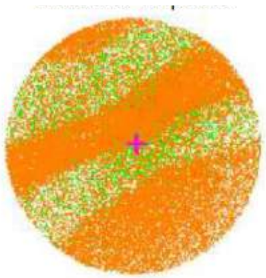
Time Of Flight measure

TOF mass spectrometer

Detector impacts

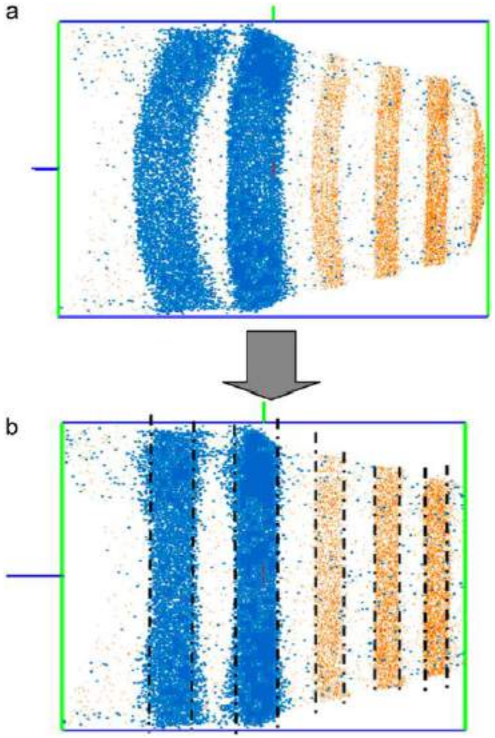


+



APT on heterogenous structures with different fields of evaporation:

- Variations in the local magnification
- Distortions in the reconstructed volume
- Abnormal atom density variations



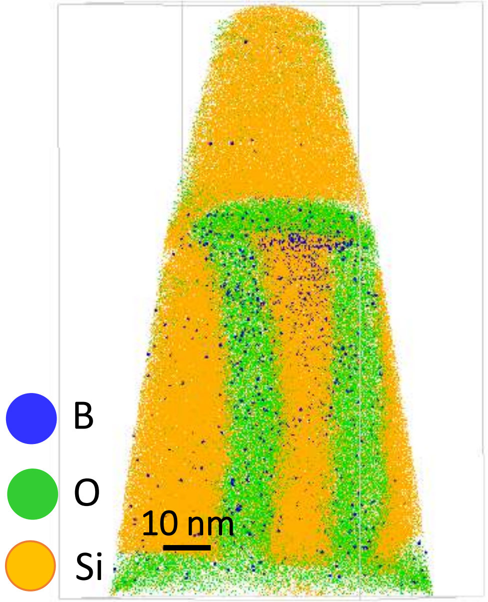
APT of GaN/InGaN/AlGaN multilayers obtained with :
(a) the standard method
(b) the two-step, interface-flatness driven, process.

F. Vurpillot et al., Ultramicroscopy 132 (2013), 19.

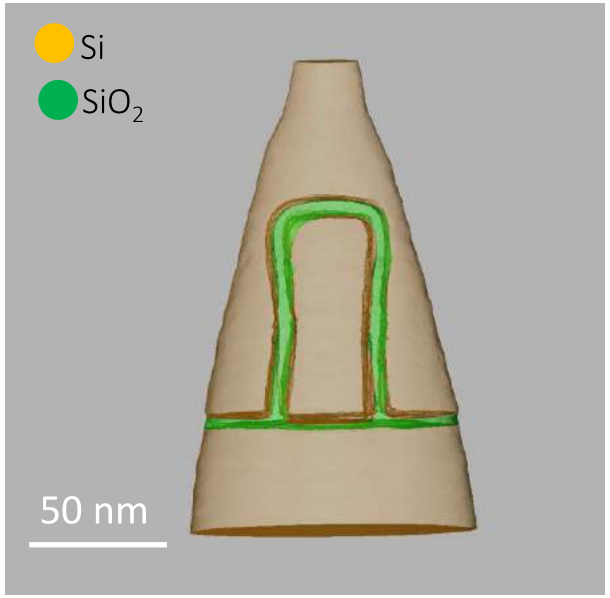
NANOELEC.

Correlative ET/APT

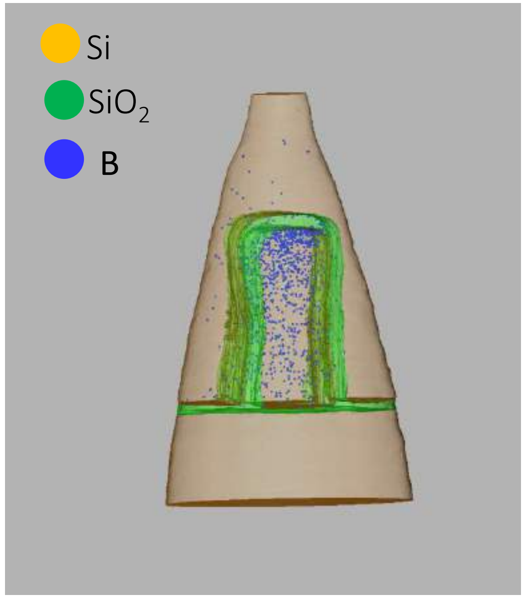
- Improvement of APT reconstruction using the combination of electron tomography reconstruction with density corrected APT algorithm.



3D APT reconstruction of the 45 nm device (Density corrected algorithm)



3D electron tomography reconstruction of the 45 nm device after segmentation.

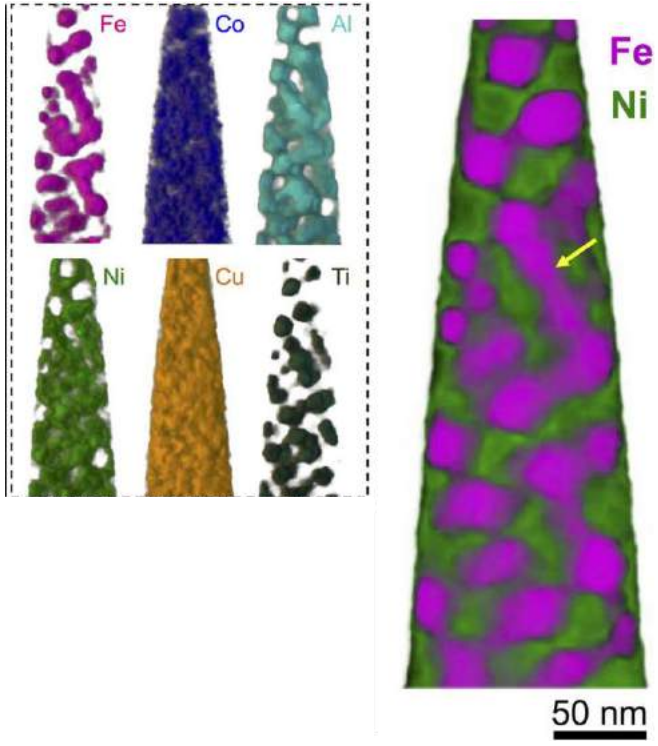


3D boron distribution inside the gate with minimal distortions.

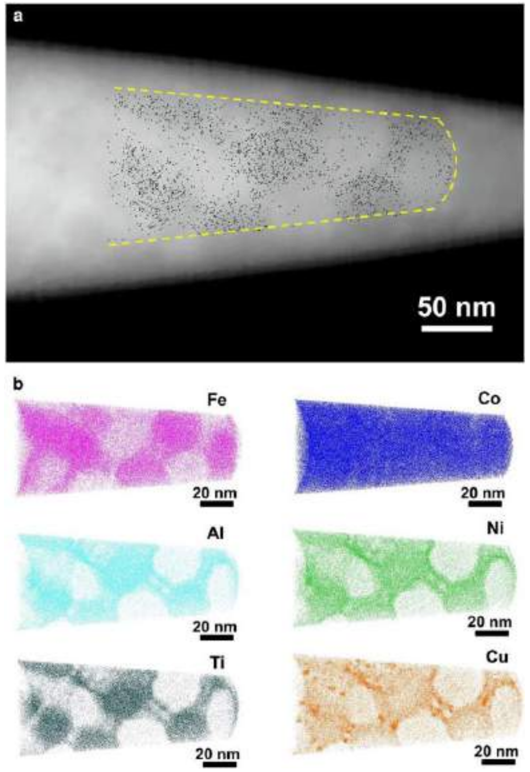
A.Grenier et al., APL, 106, 213102 (2015)

Correlative spectroscopic ET/APT

EDX-STEM tomography



APT

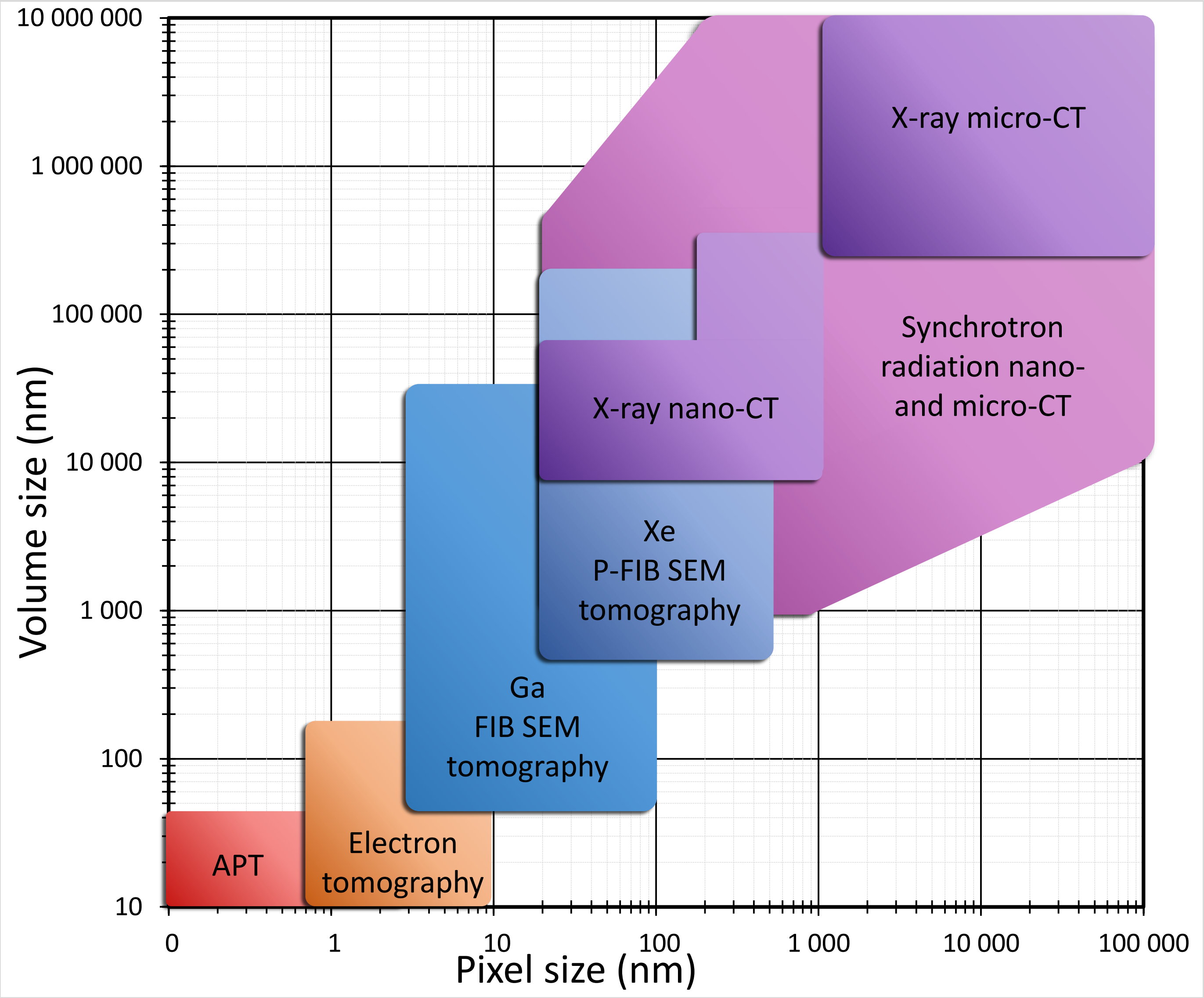


Phase Separation in an Alnico 8 Alloy.
Guo et al., Microsc. Microanal. 22, 1251 (2016)

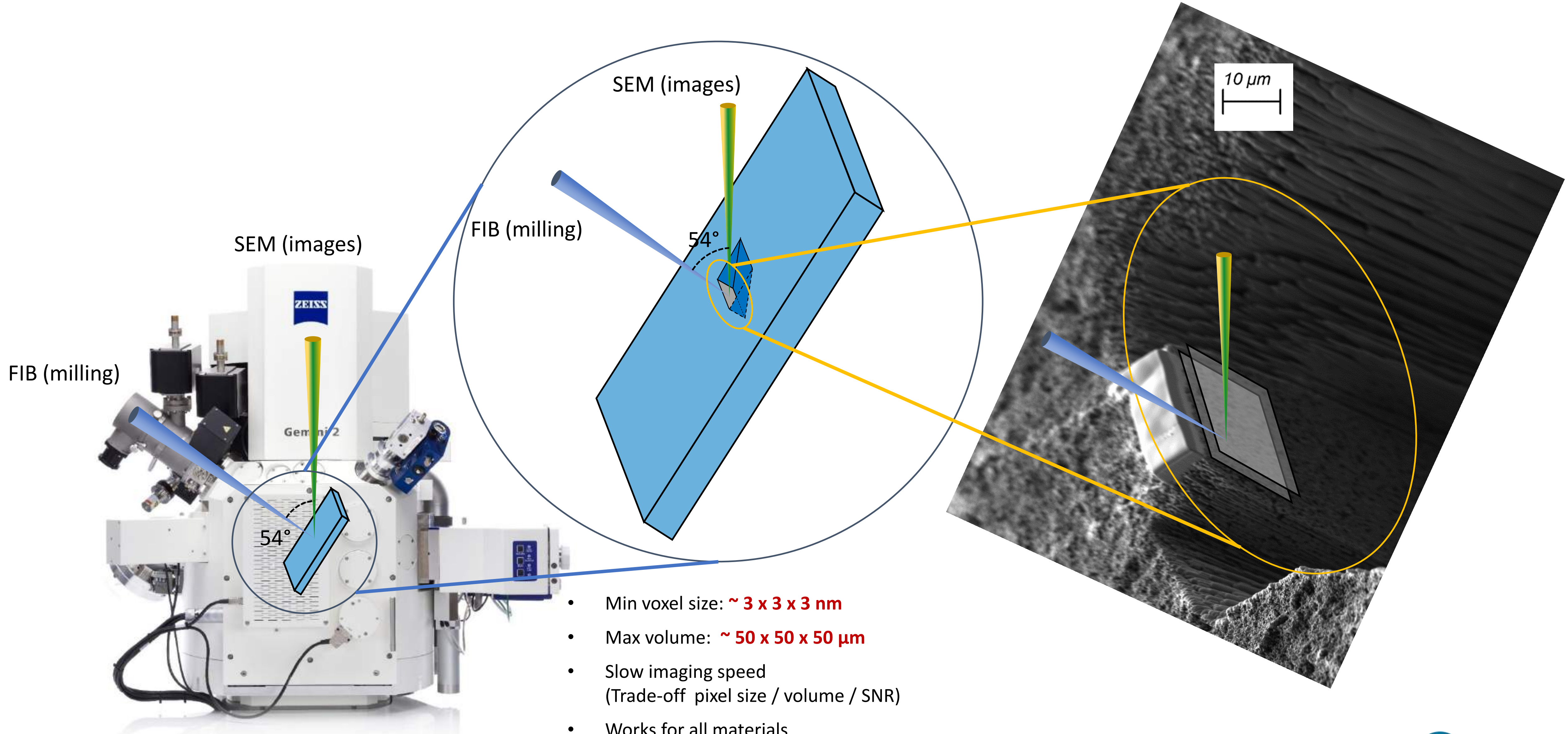
EELS/EDX-STEM tomography:
Non-destructive, large fields of view, high spatial fidelity, limited spatial resolution (few nms), LLD ~1×10^{20} at.cm^{-3}, quantification methods not fully developped.

APT:
Destructive, small fields of view, limited spatial fidelity, high spatial resolution, LLD ~5×10^{18} at.cm^{-3}, quantitative method.

3D characterization techniques at the nanoscale



FIB-SEM tomography

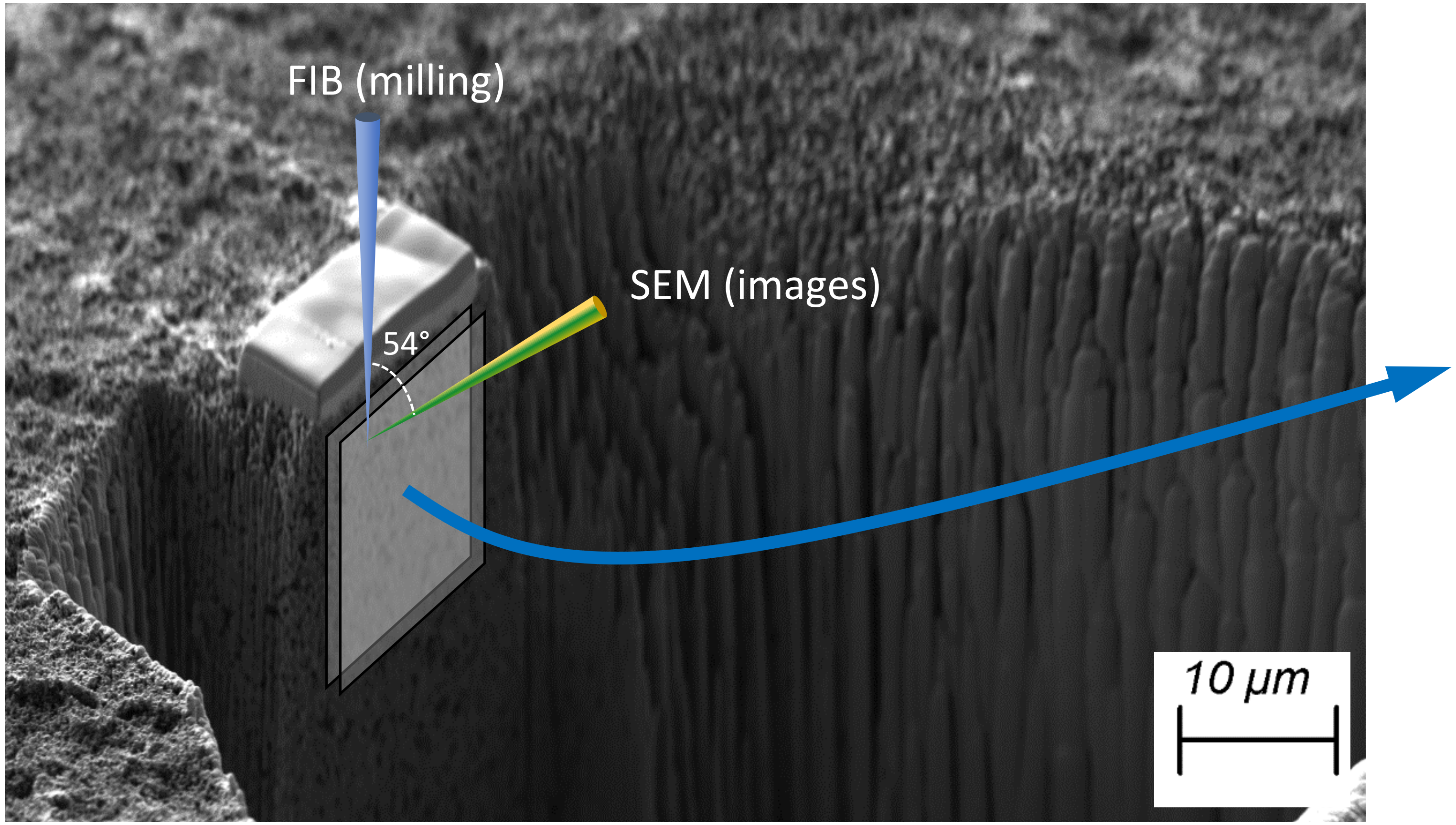


2010 – Zeiss NVision 40
2018 – Zeiss XB 550

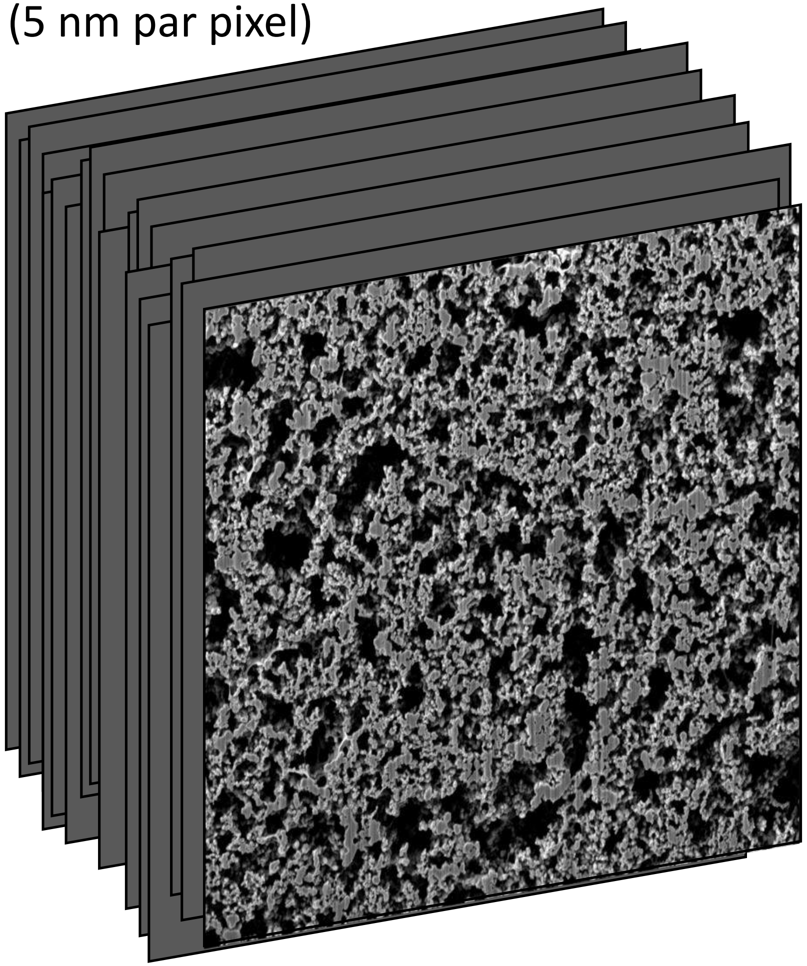
- Min voxel size: **~ 3 x 3 x 3 nm**
- Max volume: **~ 50 x 50 x 50 µm**
- Slow imaging speed
  (Trade-off pixel size / volume / SNR)
- Works for all materials

FIB-SEM tomography

Proton-exchange membrane fuel cells (PEMFC)



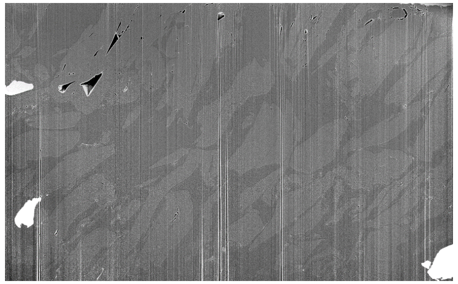
1500 images of 1960x1924 pixels
(5 nm par pixel)



NANOELEC.

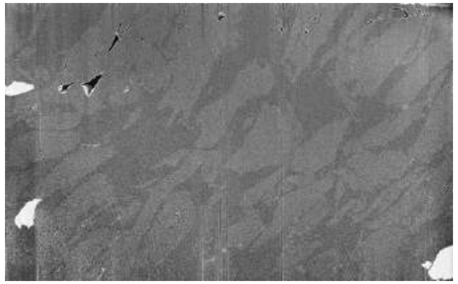
FIB-SEM tomography

Adjustment of brightness/contrast

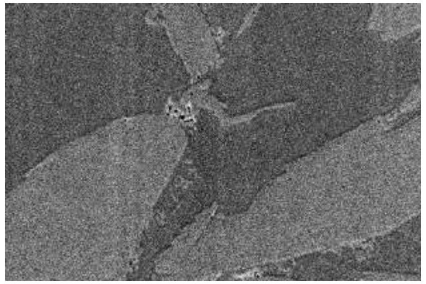


Possible causes: porous materials, rough surfaces, heterogeneous materials (hard+soft), etc

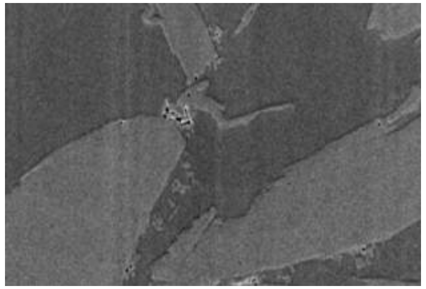
Curtaining effect removal



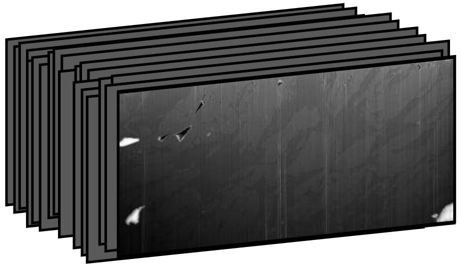
- Fourier-based approaches
- Wavelets
- VSNR (Variational Stationary Noise Remover) - J. Fehrenbach et al., *IEEE Trans. Image Process.* 21(10), 4420 (2012).



denoising



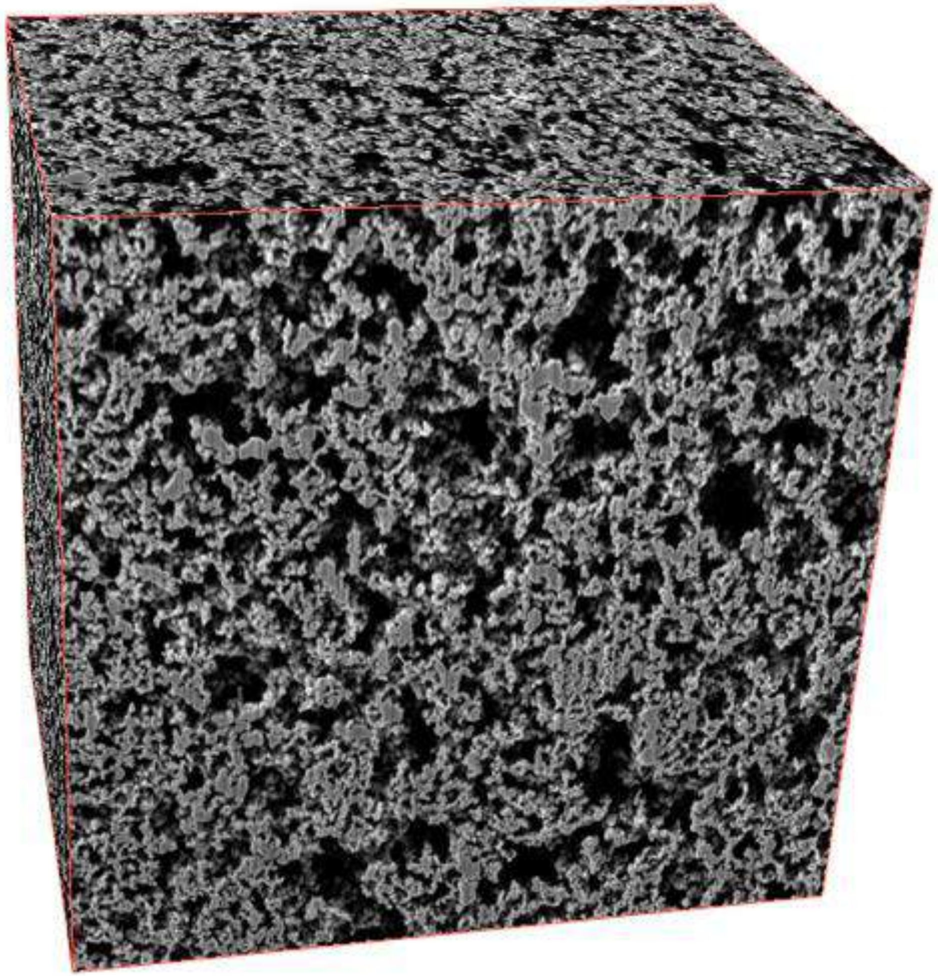
Edge-preserving methods :
- Non-local means
- Anisotropic diffusion
- (deep-learning approaches)



Stack alignment

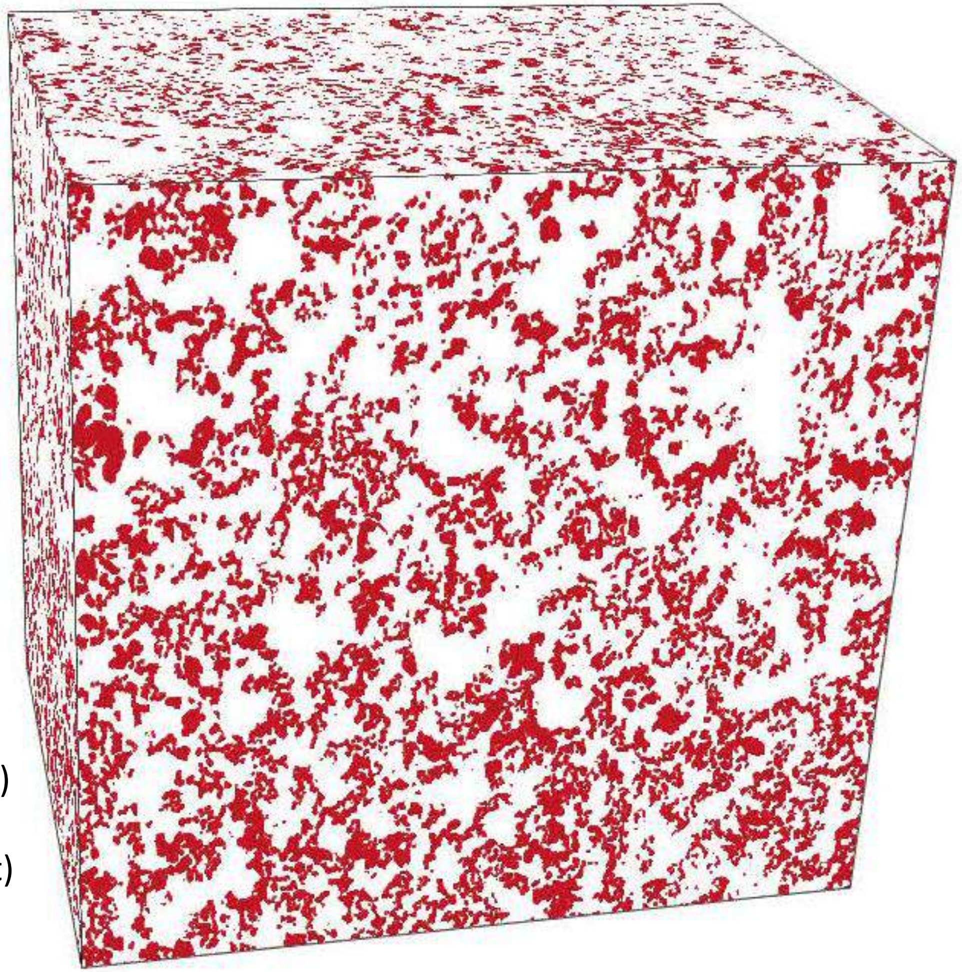
- Cross-correlation
- SIFT (Scale-invariant Feature Transform) - D.G. Lowe, *Int. J. Comput. Vis.* **60**, 91 (2004)
- AMST (Alignment to Median Smoothed Template) - J. Hennies et al., *Sci. Rep.* **10,** 2004 (2020)

FIB-SEM tomography



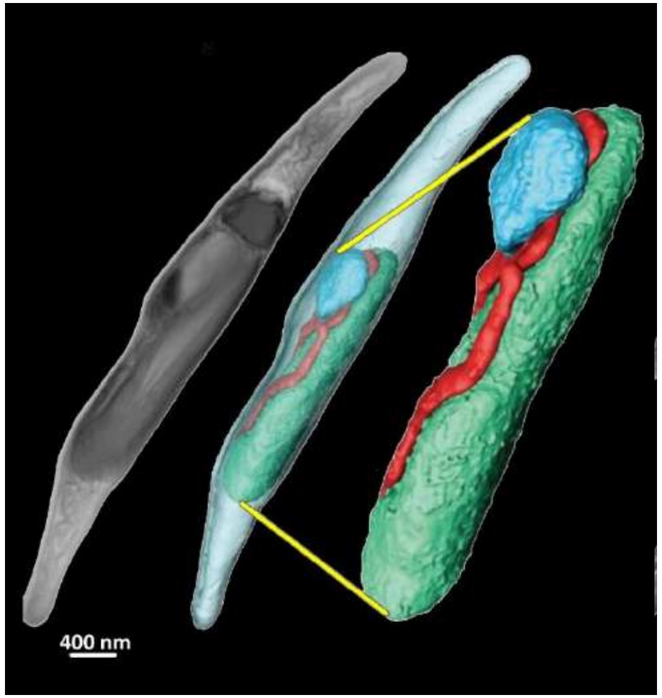
Segmentation

- Basic thresholding
- Watershed algorithm
- Machine learning approaches (e.g.: random forest classification)
- Deep learning approaches (e.g.: U-net, mask R-CNN, Stardist)

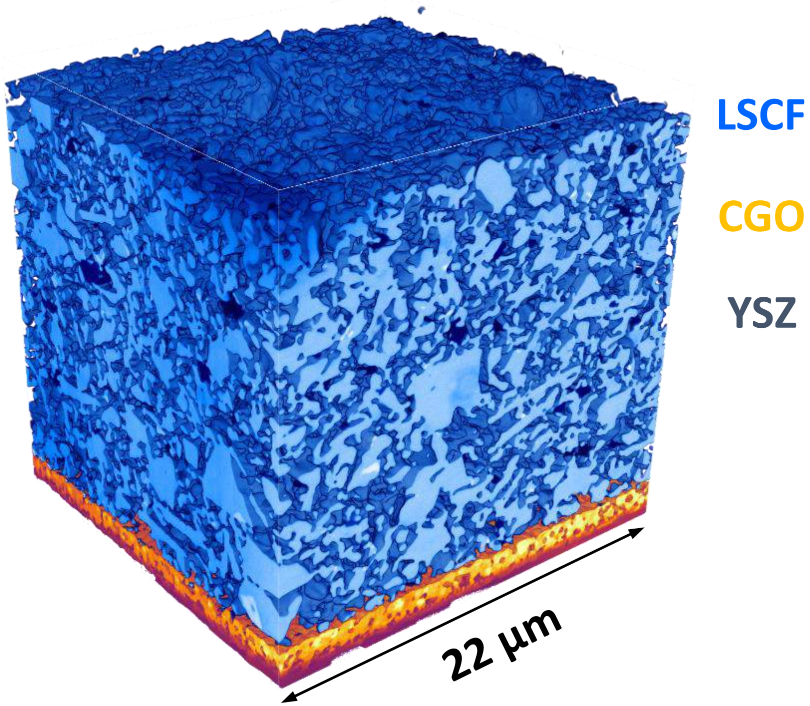


FIB-SEM tomography

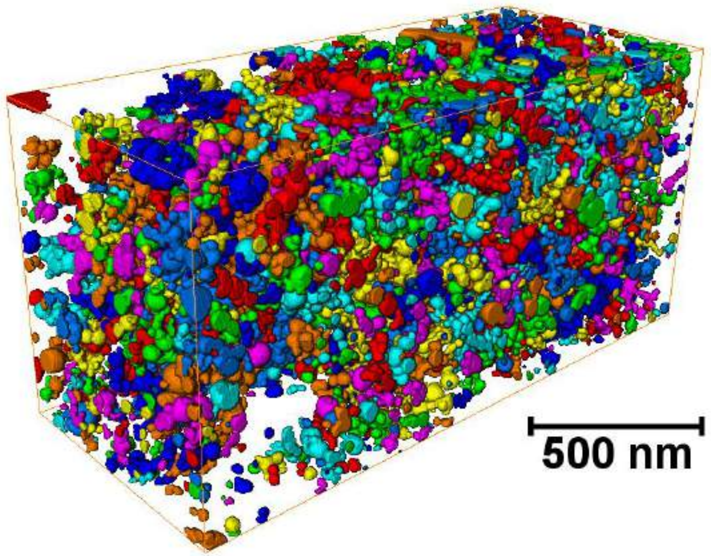
Phytoplankton cells



Solid oxide fuel cell (SOFC) electrodes



Magnetic metal-polymer nanocomposites

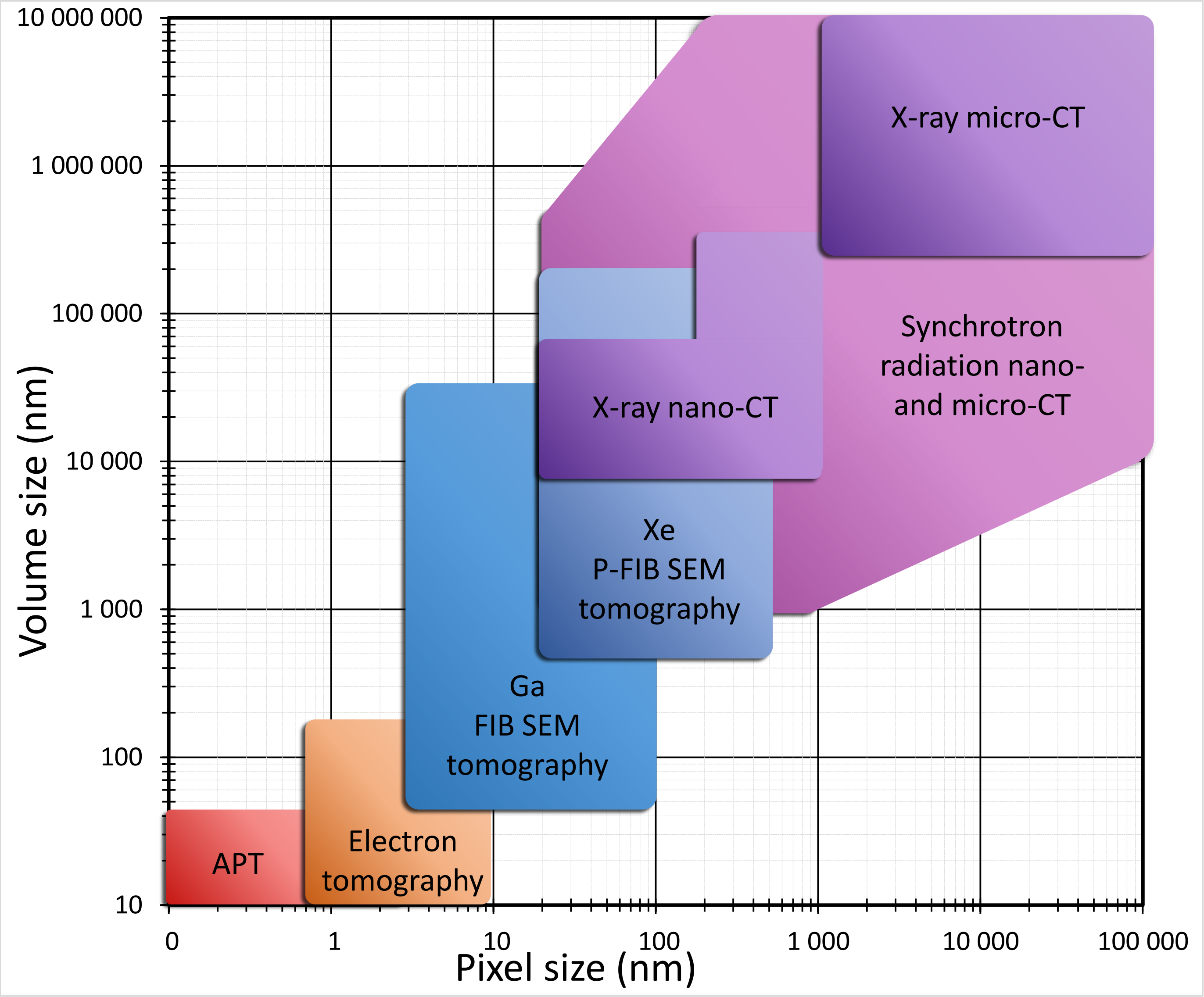


3D characterization techniques at the nanoscale

Multi-scale correlative analysis:

- CT/FIB-3D
- ET/APT
- CT/ET

…



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S. Farrens

Thank you for your attention!

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