Supplemental Information for

Ptychography at the carbon K-edge

Nicolas Mille,[1] Hao Yuan,[2] Jaianth Vijayakumar,[1,3] Stefan Stanescu,[1] Sufal Swaraj,[1] Kewin Desjardins,[1] Vincent Favre-Nicolin,[4,5] Rachid Belkhou[1] and Adam P. Hitchcock[2*]

[1] Synchrotron SOLEIL, L'Orme des Merisiers, Saint-Aubin, BP 48, 91192 Gif-sur-Yvette Cedex, France
[2] Department of Chemistry & Chemical Biology, McMaster University, Hamilton, ON, Canada, L8S 4M1
[3] UGCT/Radiation Physics, Dept. Physics and Astronomy, Ghent University, Proeftuinstraat 86 - Building N12, 9000, Ghent, Belgium
[4] ESRF, The European Synchrotron, 71 Avenue des Martyrs, 38000 Grenoble, France
[5] University Grenoble Alpes, Grenoble, France

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* Corresponding author: Adam Hitchcock, aph@mcmaster.ca

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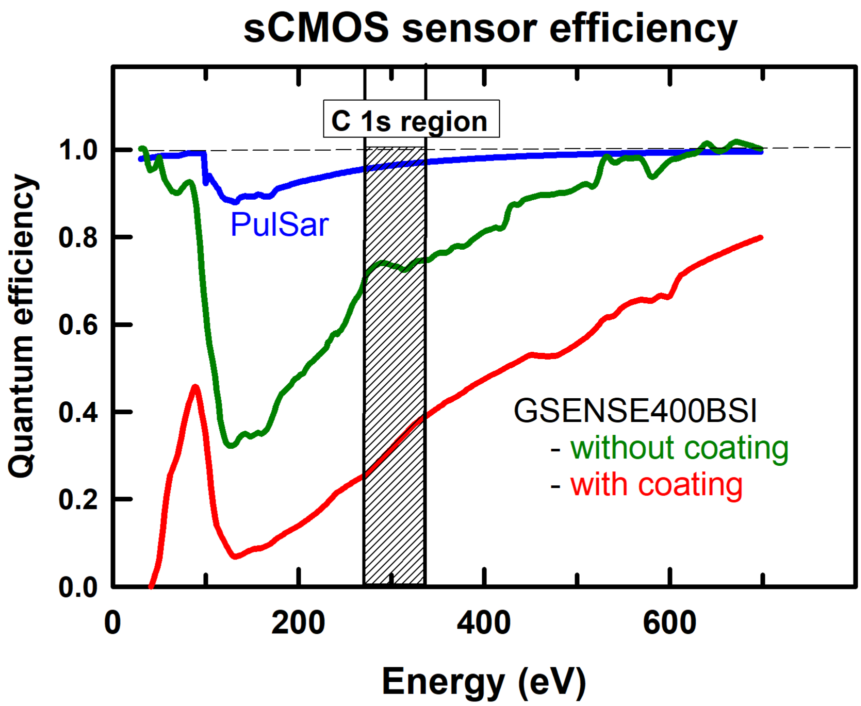
Supplementary Note 1: Photon Energy Dependence of Coated and Uncoated Sensors

Supplementary Figure 1 plots the measured response of the coated (green) and uncoated (red) GSENSE400BSI sensors (manufactured by Gpixel[1]). The third curve in Supplementary Figure 1 is a simulation of the best possible performance.[2] A Tucsen Dhyana-95 camera.[2] equipped with a prototype, uncoated GSENSE400BSI-GP sensor[1] was used for the carbon K-edge ptychography measurements reported in this paper.



Supplementary Figure 1 Quantum efficiency of the Dhyana-95 sCMOS camera as a function of photon energy for different sensors. The curves for GSENSE400BSI with coating (red) and without coating (green) were measured at Soleil.[4] The PulSar efficiency curve (blue) is a simulation based on the results reported by Harada et al.[5]

Supplementary Table 1: Details of the acquisition and reconstruction parameters used for the images presented in the paper

Figure	Sample[α]	Area (µm)	# images	Spot size (nm)[β]	Dwell (ms)	E_{photon} (eV)	Reconst Pixel size)[χ]	$q_{max, reconstruct}$[δ] (nm^{-1})	Overlap factor (%)[ε]	Radiation Dose (MGy)[ϕ]	Reconstruction algorithm[γ]
1. a	Siemens Star[η]	0.1×0.1	100	62	500	280.0	-	0.48	NA	4500	NA
1.b	Siemens Star[η]	2×2	50×50	62	500	280.0	17	0.34	35	110	AP**600
2.a	CNT	0.1×0.1	30	1000	500	285.4	-	0.48	NA	52	NA
2.b	CNT	2×2	20×20	1000	100	285.2	23	0.37	90	3.5	AP**800
3.a	CNT	5×5	50×50	1000	200	285.2	14	0.39	90	7.0	AP**1000
3.b	CNT - STXM	5×5	200×200	31	10	285.2	24	NA	0	36	NA
4.a	CNT	3×3	30×30	1000	100	282 -294[ι]	-	0.37	90	83	AP**1000
4. b,c,d	CNT	5×5	50×50	1000	200	285.2	14	0.39	90	7.0	AP**1000
4. e	CNT - STXM	5×5	200×200	31	20	285.2	24	NA	0	7.2	NA
5.a	CNT	2×2	20×20	1000	100	282 -294[ι]	20	0.37	90	83	AP**1000
S-3a	CNT	3×3	75×75	62	300	285.2	25	0.28	40	65	AP**1000
S-3b	CNT	3×3	12×12	1000	2000	285.2	25	0.28	80	17	AP**1000
S-3c	CNT	10×10	60×60	1000	200	285.2	12	0.28	60	2.4	AP**1000
S-5a	Knife Edge[η]	2×2	50×50	62	200	285.2	15	0.37	35	43	AP**500
S-5b	CNT	3 x 3	75 x 75	62	300	285.2	25	0.39	90	31	AP**800
S-6a	CNT	1.6 x 1.6	40x40	62	100*40[φ]	282 -294[ι]	18	0.39	35	860	AP**800
S-6b	CNT	1.5 x 1.5	40 x 40	1000	100*63[φ]	282 -294[ι]	9	0.28	90	62	AP**800

α. Ptychography is performed using the sCMOS camera to record diffraction images. STXM measurements used a PMT detector coupled with phosphorous scintillator as an indicating detector.

β. The 31 and 62 nm beam spot sizes are the calculated diffraction limited spot size of 25 and 50 nm outmost zone FZPs, respectively. The 1.0 µm (1000 nm) beam spot size is produced by moving the sample 23 µm downstream from the focal plane.

χ. The maximum q-range covered by the camera is 0.6888 nm^{-1}. The camera to sample distance was 56 mm. This column lists the maximum q-range for the ROI used in the ptychographic reconstruction.

ε. The overlap factor is 100*(1-S/P) where S is the step size and P is the beam spot size used for the measurements.

ϕ. The radiation dose is estimated as described in Supplementary Note 2. It includes a multiplicative, extra dose factor of $(P/S)^2$, which accounts for the overlap.

γ. AP stands for Alternate Projection. The number after ** is the number of iterations. NA = not applicable.

η. The radiation dose computed for the Au calibration sample is that which would be absorbed by the CNT sample under the same conditions.

ι. Stacks were recorded with 0.5 eV step, i.e. 24 spectral points were recorded in each stack.

φ. stacks at LH and LV polarization with multiple energies.

Supplementary Note 2: Radiation Dose calculation

Radiation dose is energy deposited per unit mass. Its units are Gray, where 1 Gy = 1 J/kg. Typical doses in soft X-ray microscopy are in the Mega-Gray (MGy) range. The dose delivered to a sample depends on many factors. It can be calculated from[3]:

Dose = (energy absorption rate) *(photon energy)* (time)/(density*volume)

$$= \left(t \cdot (I_o - I) \cdot E / \varepsilon\right) / (\rho \cdot A \cdot d) = \left(t \cdot I_o \left(1 - e^{-OD}\right) \cdot E / \varepsilon\right) / (\rho \cdot A \cdot d)$$

where t is the exposure time (s); I_o is the incident photon flux (ph/s), I is the transmitted photon flux (ph/s). I and I_o are measured by a detector with an efficiency of ε. E is the photon energy (eV), ρ is the sample density, A is the area illuminated (spot size), and d is the sample thickness. An Excel™ spreadsheet (available from the corresponding author upon request), reproduced in Supplementary Figure 2 was used to calculate the deposited dose from the experimental parameters, as summarized in Supplementary Table 1 for all the data presented here. A correction factor is applied to account for the additional dose associated with overlapping illumination. The approach used slightly overestimates the dose.

DOSE CALCULATOR FOR PTYCHOGRAPHY

DOSE (energy/mass)
= {(energy absorption rate)*(time)*(photon energy)}/(density*volume)
= {(Io-I)*E*t/ε}/(ρ*A*d)
{Io(1-exp(-OD))*E*t/ε}/(ρ*A*d)

STXM-ptycho-dose-calculator.xls
LAST CHANGED: 11 May 2021 (aph)

Converting exposure time / conditions to dose

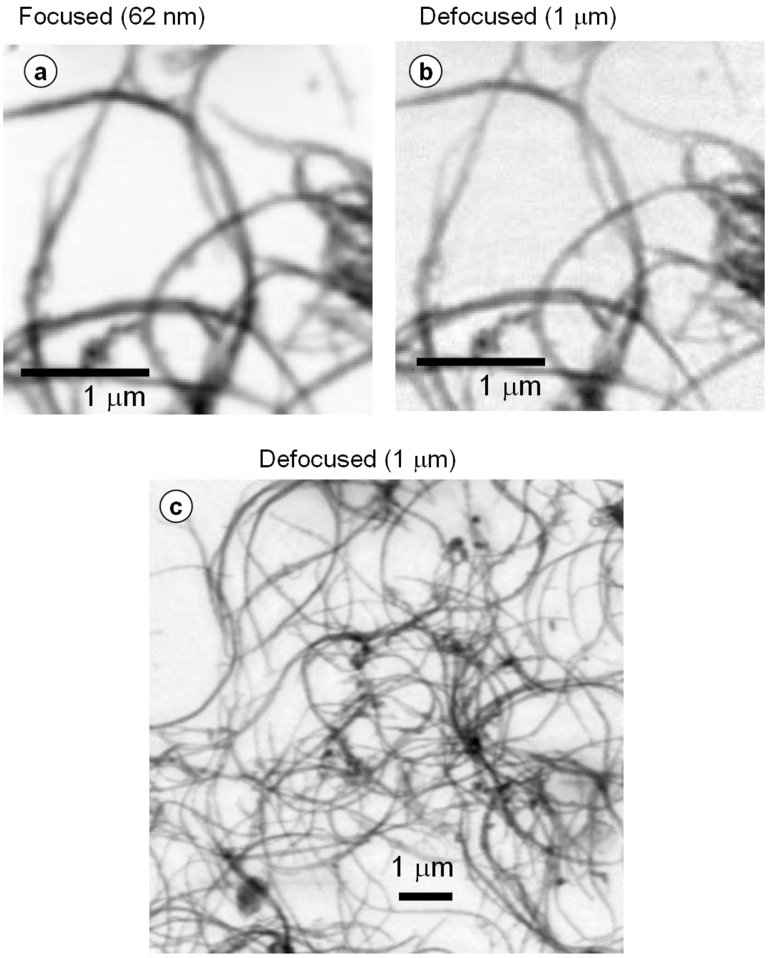
symbol	property	value	source	note
OD at E-dose	optical density	0.5	stacks	
Io	Io (MHz)	1.5	typical (= 50 MHz after det. Eff. Is included)	
Io-I = (Io(1-exp(-OD))	absorbed flux/volume	0.59	calc	
voxel	1 pixel ~ 50 nm	1000		
d	thickness (nm)	200		
V	volume (nm^3)	2.00E+08	calc	A*d
ρ	density (g/cc)	1.3	wikipedia	
m	mass/pixel (kg)	2.60E-16	calc	1.00E+21
E	energy	285		
ε	detector efficiency	0.3	STXM532 paper	
MGy/s	**dose RATE (MGy/s)**	**3.5E-01**		1.60E-19
t	**exposure time (ms)**	**100**		
w	extra dose factor	100	take into account overlapping spots	
D	**Dose (Mgy)**	**3.5E+00**		

S = step size (nm)	P = probe size	Overlap = 1-S/P	Extra Dose Factor = (P/S)^2
9.0	10.0	0.1	1.2
50.0	62.0	0.2	1.5
45.0	62.0	0.3	1.9
40.0	62.0	0.4	2.4
500.0	1000.0	0.5	4.0
400.0	1000.0	0.6	6.3
300.0	1000.0	0.7	11.1
200.0	1000.0	0.8	25.0
100.0	1000.0	0.9	100.0

Supplementary Figure 2 Screen capture from the Excel™ spreadsheet used to calculate the dose rate and dose for the conditions used for STXM and ptychography.

Supplementary Note 3: Focus versus defocus ptychography

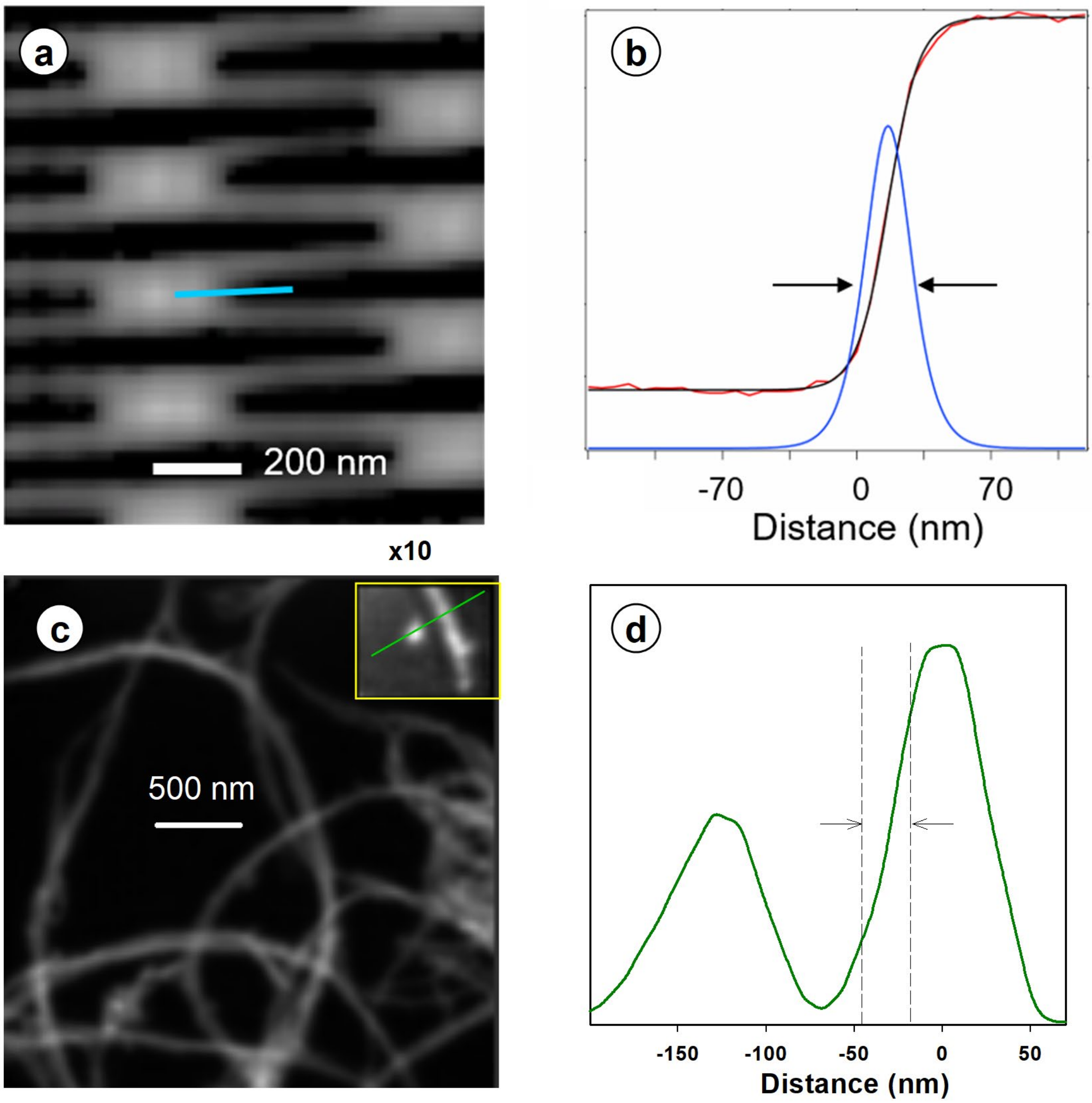
Supplementary Figure 3a presents a ptychographic amplitude image of the CNT sample measured at 285.2 eV with the sample positioned at the focus of the Fresnel zone plate (FZP) with 50 nm outermost zone width, thus giving a 62 nm spot. The parameters employed for the various acquisitions and the ptychographic reconstructions are listed in Supplementary Table 1. Supplementary Figure 3b is the ptychographic amplitude image at 285.2 eV over the same area as in (a), but recorded with the sample moved 23 μm downstream from the focal plane to make the X-ray beam spot 1.0 μm in diameter. Supplementary Figure 3c is the ptychographic amplitude image of a 10 μm × 10 μm area of the CNT sample at 285.2 eV measured using a 1.0 μm defocus spot.



Supplementary Figure 3 Comparison of focus and defocus mode ptychographic imaging. (a) Ptychographic amplitude image at 285.2 eV of the CNT sample in focus position (62 nm beam spot size, 300 ms dwell). (b) Ptychographic amplitude image of the CNT sample measured at 285.2 eV with 2000 ms exposure time and a 1 μm defocus beam spot. (c) Ptychographic amplitude image of a large area (10 μm × 10 μm) of the CNT sample measured at 285.2 eV with 200 ms exposure time and a 1 μm defocus beam spot.

Supplementary Note 4: Spatial resolution determined by knife edge and edge profile

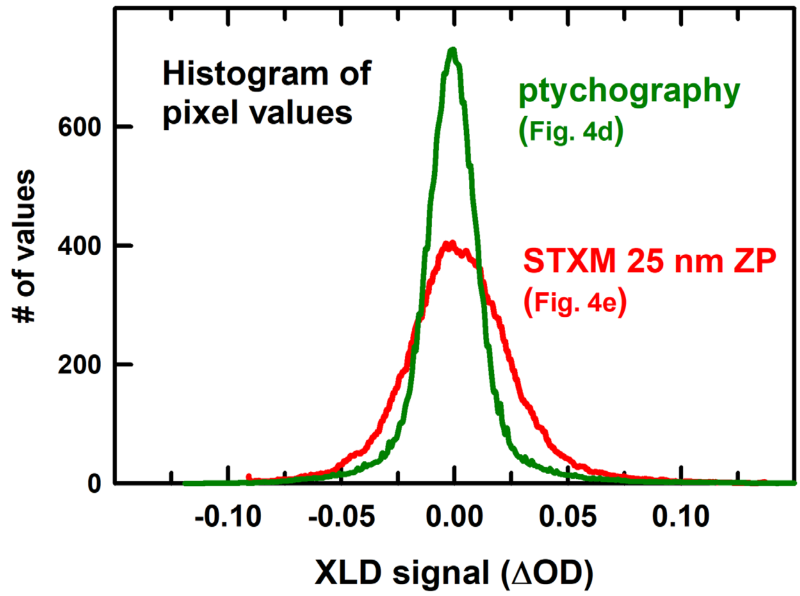
Supplementary Figure 4 reports two analyses of the ptychographic spatial resolution, based on the sharpness of features in the reconstructed images. A knife-edge approach was applied across the sharp edge of a gold structure lithographically generated on a silicon nitride window. Supplementary Figure 4a is the ptychography amplitude image derived by reconstruction of a full-focus ptychography data set measured at 285 eV from the line-space gold pattern. Supplementary Figure 4b plots the intensity of the image across the blue line in Supplementary Figure 4a. The spatial resolution was estimated by fitting the derivative of the smoothed edge profile (blue curve on Supplementary Figure 4b) to a Gaussian function. Its FWHM value is 36±3 nm. The carbon nanotubes are partly transparent to the X-rays and thus a classical knife-edge approach does not apply. Also, most objects in the sample are bundles of AD-SWCNT and thus have a size considerably larger than the spatial resolution. Instead we have estimated spatial resolution from the abruptness of a specific image feature. Supplementary Figure 4c presents an OD image derived from reconstruction of focused ptychography at 285.2 eV (Supplementary Figure 2a is the amplitude image from which Supplementary Figure 4c was derived). The upper right corner insert is that area with the OD scale decreased by a factor of 10 – these are some of the sharpest features in this image. Supplementary Figure 4d is the intensity profile across the green line in the insert to Supplementary Figure 4c. The abruptness of the profile across the CNT is 30±3 nm, as measured from the 20 % to 80 % intensity levels.



Supplementary Figure 4 Evaluation of spatial resolution from the sharpness of reconstructed ptychographic images. (a) Ptychography amplitude image at 285.2 eV of a lithographed gold line-space pattern with a 50 nm pitch measured using the 50 nm outermost zone FZP, with the sample in the focal plane (see Supplementary Table S-1 for details of the acquisition and reconstruction parameters). The OD changes from 0 to a very large value (>10) at the edge and thus the boundary is valid for knife edge spatial resolution evaluation. (b) Intensity profile across the blue line in (a). The red curve is the raw profile, the black curve is a smoothed version and the blue curve is a Gaussian fit to the derivative of the smoothed line profile. The FWHM from the Gaussian fit is 36 ± 3 nm. (c) OD image of the CNT sample (OD version of the amplitude data presented in Supplementary Figure 2a). The insert is a x10 intensity amplification. (d) Line profile across the green line. The abruptness of the rise in optical density (20% – 80 %) is 30 ± 3 nm.

Supplementary Note 5: Evaluation of contrast in ptychography versus STXM

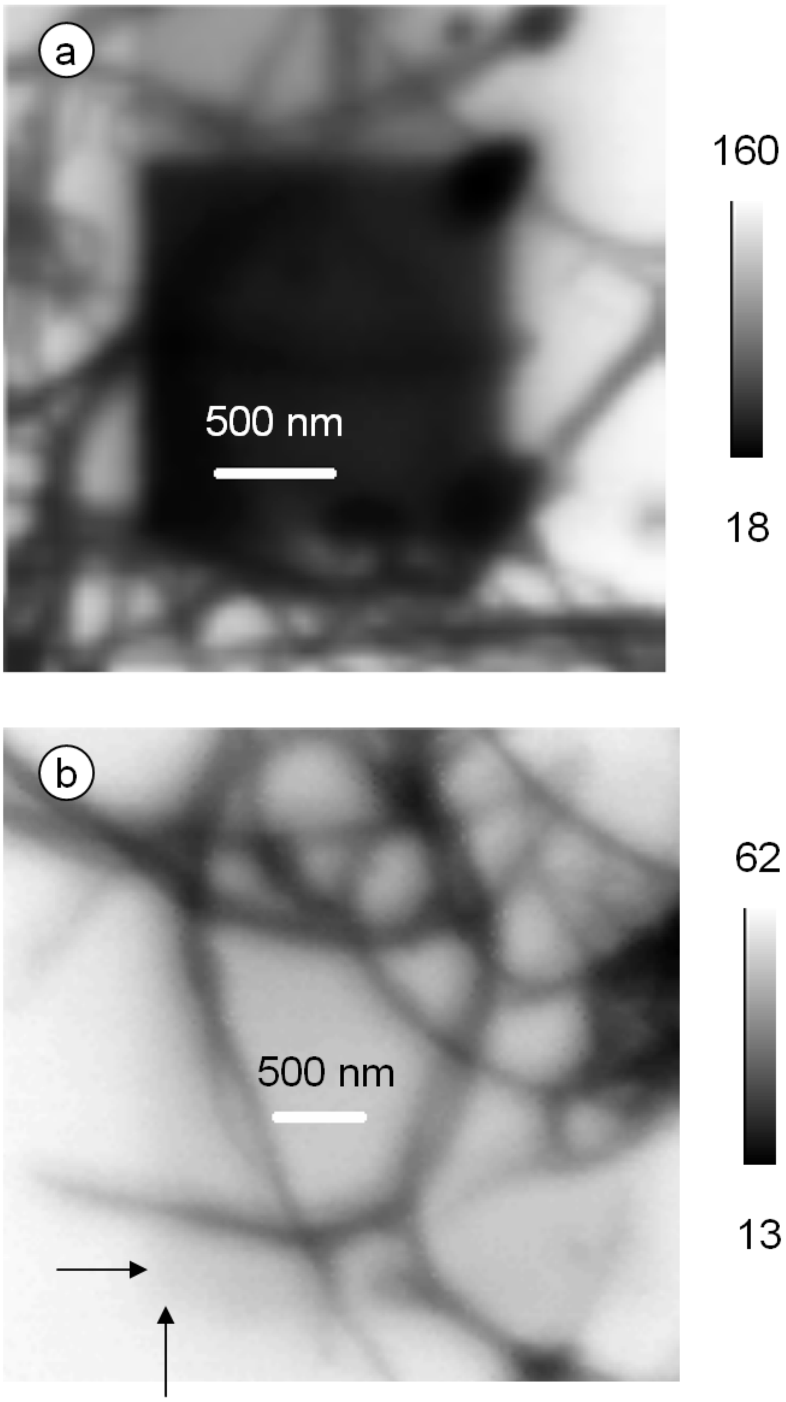
In order to quantify the difference in quality (signal to noise, contrast and sharpness), the histogram of pixel values in the XLD images recorded by ptychography (Supplementary Figure 4d) and STXM (Supplementary Figure 4e) have been compared. Supplementary Figure 5 presents plots of these histograms, normalize to same total number of pixels. The full width at half maximum of the histogram of the ptychography XLD image is 60 % narrower than that of the STXM XLD image.



Supplementary Figure 5 Evaluation of the relative quality of X-ray linear dichroism (XLD) measured by ptychography and STXM. Peak area normalised histograms of the XLD images derived from ptychography (Supplementary Figure 4d) and STXM (Supplementary Figure 4e).

Supplementary Note 6: Demonstration of reduced radiation damage with defocus ptychography

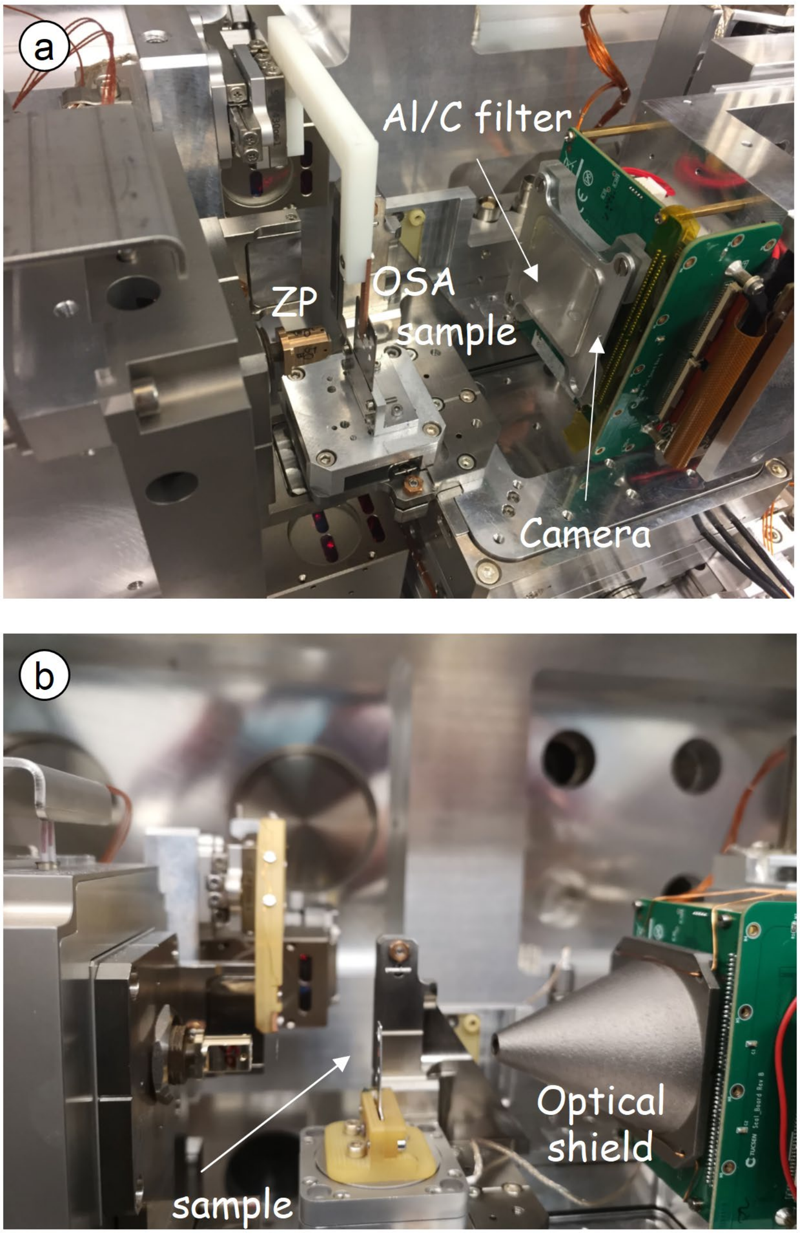
When we used full focus ptychography with an estimated dose of 860 MGy, the carbon build up made the region imaged completely opaque, as shown in the post-stack image at 288.5 eV, Supplementary Figure 6a. When a similar area was measured by defocus ptychography with an estimated dose of 62 MGy, the post-stack image (at 285.2 eV) showed very little carbon build-up (Supplementary Figure 6b). From a practical point of view, the defocusing is a big help because it greatly reduces the radiation dose and dose rate.



Supplementary Figure 6. Influence of spot size on carbon deposition. (a) presents a STXM transmission image at 288.5 eV recorded after a full focus ptychography stack measured with an estimated dose of 860 MGy. (b) presents a transmission image at 285.2 eV recorded after a ptychography stack was measured with a 1.0 µm spot size, delivering an estimated dose of 62 MGy. The arrows in Supplementary Figure 6b mark the lower left corner of the area where the defocused ptycho stack was measured.

Supplementary Note 7: Implementation of the Dhyana-95 camera in the Hermes STXM

Supplementary Figure 7 presents two photographs of the ptychography setup employed at HERMES' STXM. The standard STXM setup elements - the FZP, the OSA (50 µm) and the sample plate are labeled. As detailed elsewhere,[4] the standard housing of the Tucsen Dhyana95 camera is removed and the main electronic components are cooled down using a



Supplementary Figure S-7 Photographs of the set-up used for ptychography. (a) Photograph of the Tucsen Dhyana model 95 sCMOS camera, mounted without casing in the Hermes STXM. The Al-supported parylene in front of the camera is a visible light filter which is only used for measurements above 500 eV. (b) View of the setup used for carbon K-edge ptychography. The visible light filter is replaced with a Ti printed cone whose small opening (< 10 mm) prevents stray light reaching the camera sensor.

specially designed metallic heat exchanger which is the cold point for the Peltier stage used to keep the sensor at -20°C during the measurements in order to reduce thermal noise. To avoid mechanical vibrations, the cooling system, a low-pressure closed-circuit chiller from Julabo (www.julabo.com), is placed as far as possible from the STXM vacuum tank (6 meters), and the inlet and outlet pipes pass through a passive damper made of sand. For the same reason, the camera is attached rigidly to the detector stage of the STXM which allows optimization of the XY position of the sCMOS camera and the detector to sample distance. The sensor socket is fully accessible which allows easy exchange of the sensor. The backside interface of the sensor with the Peltier stage was made using special thermal paste. As shown in Supplementary Figure 7a, for measurements above 500 eV, the sensor was protected by a parylene supported Al filter against visible light, which arises from the He-Ne laser used for the interferometric position control system. This kind of setup was used during early ptychography tests at higher energies (> 500 eV) but was unsuitable for the C K-edge measurements. As shown in Supplementary Figure 7b, this filter was replaced by a 3D printed conical shield made of Ti. Its 55 mm length and < 10 mm opening was adapted for the geometrical constraints around 300 eV. With this shield, dark images recorded with long exposure times (> 1 s) did not exhibit signs of visible light background.

Supplementary References

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