Soft X-ray Ptychography
Development at SSRF

Physics and Environment Division of SSRF

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Introduction to Ptychography
Coherent diffractive imaging — lensless imaging

New X-ray imaging method for non-periodic samples. The highest resolution reaches 2 nm.

Successful applications have been achieved in material, life, chemistry and magnet, etc.
Coherent Diffractive Imaging

A coherent X-ray beam illuminates a non-periodic sample, producing a far-field diffraction pattern. When the pattern is sampled enough finely (oversampling ratio greater than 2), the sample image can be reconstructed by an iterative phase retrieval algorithm.

Merits:

- Resolution is only limited by X-ray wavelength and the maximal diffraction angle: \( d = \frac{\lambda}{\sin \theta_{\text{max}}} \)
- Suited for 3D imaging of Non-periodic, amorphous or biological samples
Six CDI methods

At present there are mainly six kind of CDI methods: plane-wave CDI, Fresnel CDI, scanning CDI (ptyhography), partial coherence CDI, Brag CDI and reflection CDI.
Ptychography

1. Sample size is unlimited laterally, no need to meet with isolated sample requirement.

2. Convergence is better. Unique solution in most cases.

3. Probe and object reconstructed simultaneously.

4. Overlap between neighbor illuminated position is required, usually larger than 60%.

5. Impacted largely by the position errors and thermal drifts of sample motors.

6. High stability of the light source and the beamline is required.
Applications of Ptychography
Alloy nanoparticles analyzed by Ptychography (dual energy)

Chemical contrast of Ptychography

Ptycho imaging at multiple energies near the absorption edge, implementing the spatial resolution of chemical states. The figure shows the ptycho spectromicroscopy at O edge, resolved five PMMA balls and four SiO2 balls.


Fe2+ and Fe3+ distribution in a Li battery electrode material by Ptychography spectromicroscopy, the resolution is about 10 nm. From Tolek (ALS 2013)
ptychography spectromicroscopy of Partially delithiated LiFePO4 nanoplate

Sample is scanned at 60nm ZP focus with 40 nm steps, and the CCD at 8 cm downstream of sample. The movement precision is 1.5nm. Short/long time exposure combination (30/800ms)

a. STXM image, b. ptycho absorption image at 710eV, c. ptycho phase at 709.2 eV, d. LiFePO4 and FePO4 distributions by energy stack analysis of ptycho image series.

D. Shapiro, Y.-S. Yu, T. Tyliszczak, et al., *Nature Photonics*, DOI: 10.1038/NPHOTON.2014.207
Cell spectromicroscopy by ptychography

3D ptychography of bone

Scanning CDI CT imaging of bone sample, resolution ~100nm.

Dierolf et al., Nature 2010

Swiss Light Source
Magnet dichroism by Ptychography

Resonant X ray diffraction image of magnet film using linear polarized X-ray, from which the magnet domain image can be reconstructed.

PNAS 108, 13393, 2011
Building and Development of SSRF Soft X-ray Ptychography
A ptychography platform is set up based on STXM

High resolution imaging with low-dose

Hardware facility setup

Various softwares written

~10 nm resolution achieved, greatly improved from the original 30nm resolution

Reconstructed star pattern from the first experiment dataset

Scanning CDI facility setup

Diagram of the scanning CDI based on STXM. The sample is usually out of focus, making a larger light spot (3-5um).

Photo of ptycho setup

CCD square cone mask—blocking ambient noise

Poster in 12th International Conference on X-ray Microscopy, 26-31, 2014, Melbourne, Australian
First successful ptychography experiment results in SSRF

Two typical diffraction patterns (of total 49 in this dataset)

STXM image of a Siemens star

Reconstructed amplitude image
Reconstructed phase image
Reconstructed probe amp
Recon. probe pha

Results of scanning a star pattern with a 716eV 3um spot and 800nm steps. We can see that the recon amp image (absorption) contrast of the Siemens star is as good as that of STXM image, showing the stained spots clearly. The scanning mode of big-spot big-step could significantly reduce the radiation dose on the sample during imaging. Presented in an XRM 2014 poster.
Ptychography has a much higher tolerance to low-frequency signal missing than the conventional plane-wave CDI.

Left: Reconstruction error of probe-defined single CDI (full-field CDI) changing with the beamstop size and probe size; Right: Reconstruction error of Ptycho changing with the beamstop radius and probe size (overlap 70%), where the results of single CDI also included in the upper-right panel. The lower reconstructed images corresponding to the points denoted by arrows in the upper curves panels, respectively.

Resolution of Ptycho much better than STXM

STXM image
20nm steps

PCDI image
3um light spot, 800nm steps

250*250 exposure positions, each expo is 2 ms, total exposure time is 125 s. The total data acquisition time is 15-20 minutes.

7*7 exposure positions, each expo is 200 ms, total is 9.8 s. The total data acquisition time is 5-6 minutes.

12th International Conference on X-ray Microscopy, 26-31, 2014, Melbourne, Australian
Rastergrid pathology can be reduced by increasing overlap

<table>
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<tr>
<th>Step Size</th>
<th>Efficiency</th>
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<tr>
<td>500nm steps</td>
<td>83%</td>
</tr>
<tr>
<td>400nm steps</td>
<td>87%</td>
</tr>
<tr>
<td>200nm steps</td>
<td>93%</td>
</tr>
<tr>
<td>100nm steps</td>
<td>96.7%</td>
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Diameter of the light spot is 3 μm

The sample is the PtCo alloy nanoparticles on a carbon film of Cu grid.
Optimization of relaxation parameters alpha and beta in EPIE

The used dataset is from an experiment with a 4um light-spot and 75% overlap.

It can be seen that the optimized parameter values gathered in the alpha>beta area. The optimal ones are: Alpha=1.7, Beta=0.35 (just for this dataset, maybe different for other dataset).
New background noise removal techniques

Choosing a reference region R, a coefficient is calculated by minimization of the difference between the ref. regions of the pattern image and dark image (background image):

$$\alpha_i = \frac{\sum_{x,y \in R} I_{M,i}(x,y) I_D(x,y)}{\sum_{x,y \in R} I_D^2(x,y)}$$

Subtracting the background from the diffraction pattern:

$$I_{\text{eff},i} = I_{M,i}(x,y) - \alpha_i I_D(x,y)$$

Then, a threshold value of the residual background noise is determined within this ref. region:

$$I_{th} = I_{\text{max}} - 2 I_{\text{std}} = \langle I^{R}_{\text{max},i} \rangle_i - 2 \langle I^{R}_{\text{std},i} \rangle_i$$

After this thresholding, a clear diffraction pattern almost without background noise can be obtained.

*Applied Optics, 56 (2017) 2099*
New background noise removal techniques

Three new background noise removal methods were developed: difference minimization, thresholding, local erasing. By combination of the three methods, we can obtain clean patterns without background noise almost.

Upper: recon. Images of the siemens star; Lower: 2D power spectral density (PSD) images. It can be seen that there is least noise and pseudo information in (c1) image, and the intensity of the third order frequency spectral peak in (c2) is much higher than that in the other two images. These practical applications indicated that our new noise removal methods significantly improve the reconstruction image quality of ptychography.
Resolution improving of ptychography by mixed state algorithm

(a, b) Ptychography amplitude and phase images of the siemens star (mixed state algorithm), (c-e) the central area (red squared region in a) images by CDI (c), SEM (d) and STXM (e), respectively. It can be seen that the innermost 30nm stripes can be distinguished clearly in ptycho image, which is not worse than SEM image, much better than STXM image. The black shadows in amp image center and white speckles in pha image center are due to contaminations by impurity nanoparticles. *Nucl. Sci. Tech.*, 28, 74 (2017).
Resolution improving of MS ptychography relative to STXM

Resolution analysis

(a) PSD analysis of the ptycho amp image, giving 8.1nm resolution.
(b) PSD analysis of the STXM image, giving 32.1nm resolution.
(c) FRC analysis of the ptycho image, giving **11.7nm** resolution based on the halfbit threshold.
Several Proof Experiments
Imaging of Pt-Co nanoparticles (14nm)

Ptycho amplitude image

STXM image

Ptycho image of probe amp

PSD analysis of the ptycho amp, giving 12.4 nm resolution

PSD analysis of the STXM image, giving 38.5 nm resolution

FRC analysis of CDI image, giving 14.6 nm resolution
Au nanowires imaging (8.5nm)

STXM image with 20nm steps

Ptycho image with 3um light spot and 400nm steps.

PSD analysis of CDI image, giving 8.5nm resolution

STXM with 15 nm steps

Local amplified ptycho image
A breast cancer cell stained by ZnSe quantum dots (18.7nm)

(a) STXM image of the whole cell with 100nm steps.
(b) Ptychography phase image (red squared area in (a)).
(c) STXM image of the same area as in (b) with 30nm steps.

(a) PSD analysis of ptycho image, giving 12.7nm resolution; (b) PSD of STXM image, giving 75.8nm resolution; (c) FRC analysis of Ptycho image, 18.7nm resolution.
Magnetotactic Bacteria

It can be seen that ptychography resolved the separated magnetic nano-particles in the MTB, while the STXM image could not. The sample is provided by Prof. Fink of Germany.
Some Users experimental data for biological samples

- A sort of bacteria (H.D. Jiang, Shanghai Tech)
- Functional protein locating in a cervical cancer cell (Y. Zhu, SINAP)
- Bamboo slice (S.M. Yang, ICBR), 28 nm resolution
- Human hair slice (R. Fink, FAU Germany)
Summary

The performance comparison between STXM and PCDI at STXM endstation of SSRF （based on a typical dataset）

<table>
<thead>
<tr>
<th>Performance</th>
<th>STXM</th>
<th>PCDI</th>
<th>STXM: PCDI</th>
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<tr>
<td>Exposure time (dose)</td>
<td>226 s</td>
<td>18.8 s</td>
<td>12:1</td>
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<tr>
<td>Data acquisition time</td>
<td>30 min</td>
<td>11 min</td>
<td>~ 3:1</td>
</tr>
<tr>
<td>Spatial resolution</td>
<td>&gt; 30 nm</td>
<td>~ 8.5 nm</td>
<td>~ 3.5 :1</td>
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- We have successfully set up the soft X-ray ptychography platform, both in hardware and software aspects。
- Ptyhcogaphy shows significant advantages in resolution, radiation dose and experiment efficiency over the traditional scanning imaging method.
- At present we have implemented about 10 nm resolution for material samples and 20 nm resolution for biological samples.
- By optimizing the experimental parameters (the scanning position, the illumination spot size and step size, etc) and the reconstruction parameters, the imaging quality of ptychography could be further improved.
- Spectro-ptychography and 3D-ptychography are being developed at STXM of SSRF to promote its applications in users’ research.
Thank you for your attention