Soft X-ray Coherent Diffraction Imaging

Soft X-ray microscopy is a powerful technique that can probe large volumes of materials at high spatial resolution with chemical, electronic and orientation contrasts.[1] Developed recently, coherent diffraction imaging (CDI) in a form based on a soft X-ray scanning transmission X-ray microscope (STXM), the so-called ptychography, is making substantial improvements in spatial resolution and spectral information.[2] It can, in principle, provide a wavelength-limited resolution without a limitation of X-ray lenses. Different from CDI, ptychography relies on collecting a set of diffraction patterns at varied regions of a sample, which become physically overlapped with a known separation.[3] Spatial resolutions 10 nm and below 3 nm were achieved for hard[4] and soft[2] X-rays.

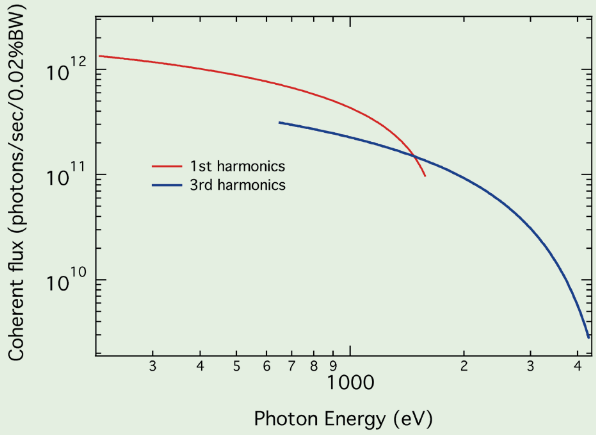


Fig. 1: Estimated coherent flux of an ideal beamline, assuming that the total transmission of the grating and the beamline optics is 1.3%.

Similar to the standard STXM, soft X-ray ptychography can provide 2D chemical mapping and determine the chemical states at each pixel by X-ray absorption spectra (XAS) with high spatial resolution. David Shapiro[2] first demonstrated the greatest resolution of a soft X-ray microscope using ptychography and applied it to study the $LiFePO_4$ phase separation in lithium-ion batteries. The presence of multiple particles with varied chemical states and the cracks along the C-axis were visualized, which indicated that the coupling of kinetics of a phase transformation with mechanical consequences is critical during the lithiation and delithiation processes.

The second example was performed by Xiaohui Zhu[5,6] who investigated XAS associated with X-ray magnetic circular dichroism (XMCD) of individual magnetosomes in a cell of magnetotactic bacteria (MTB) from ptychography image sequences recorded with left and right circular polarization. The results indicated that the magnetic dichroic information could be probed with ptychographic XMCD in both absorption and phase signals for extracellular magnetosomes. Compared with the FeL_3 spectrum measured with a ptychography and a conventional STXM using varied sizes of the outer zone plates, the results clarified that a soft X-ray ptychography can not only improve the spatial resolution but also benefit the spectral and chemical sensitivity.

To perform various diffraction experiments, a sufficient coherent flux is necessary. X-rays delivered from an undulator beamline at a third-generation synchrotron radiation facility are coherent on a micrometer length scale. With appropriate optics and filtering, the EPU of TPS will yield soft X-rays of great intensity with a high degree of coherence. As shown in **Fig. 1**, the estimated coherent flux from an EPU48 source in energy range 400 to 1200 eV is as much as 10^{11} photons/s. The coherent flux is estimated with a formula $I_{coh} = B(\lambda/2)^2(\Delta\lambda/\lambda)$ and assuming the resolving power of the beamline to be 5000. The objective of this project is to take advantage of Taiwan Photon Source to establish advanced coherent diffraction image techniques to study electronic and magnetic properties with great spatial resolution.

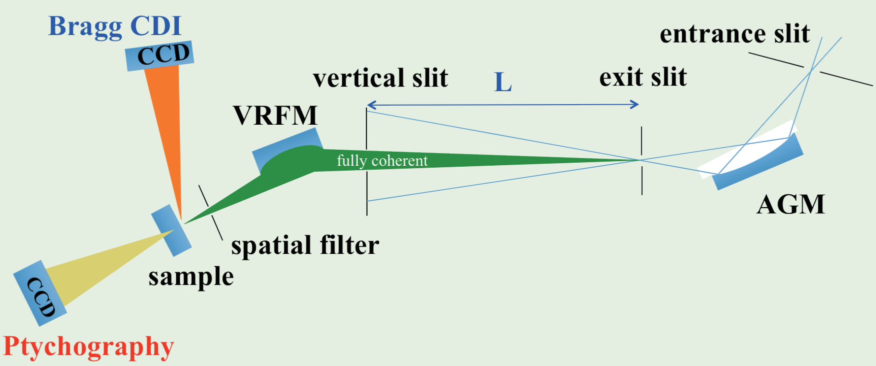


Fig. 2: Principle of beamline design with a variety of operating modes.

The principle of the beamline design is shown in **Fig. 2**. The monochromatic beam is generated with an active-grating monochromator (AGM) and focused on the exit slit to define a secondary source. The

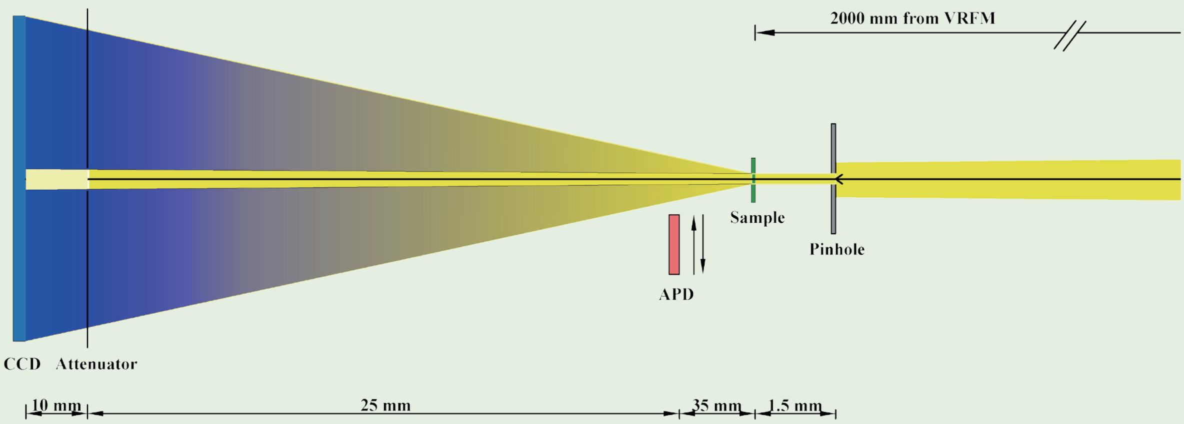


Fig. 3: Schematic of the basic concept of soft X-ray ptychography.

incoherent portion of the beam outside the coherent angle of λ/d, in which d is the slit opening, is rejected with a set of coherent slits in both directions. A spatial filter (pinhole aperture) is placed between the Kirkpatrick-Baes (KB) refocused optics and the sample to clean the coherent beam and to minimize its size. The system operates in three modes – resonance soft X-ray scattering (RXS), Bragg CDI and ptychography. The operating system incorporates a diffractometer in vacuum with two principal rotation axes θ and δ for RXS measurement. On replacing the existing point detector with a 2D CCD (charge coupled device) detector for RXS measurement, the diffraction superstructure can be collected to obtain spatial information about spin, charge and orbital ordering.

The layout of the soft X-ray ptychography is shown in **Fig. 3**. All stages and components for ptychography are located inside the Taiwan-Anglo Coherent Diffraction Endstation (TACoDE). In this design, the KB optics focus a coherent beam onto a sample; a pinhole is placed between to clean the coherence source. With precise 2D sample-scanning stages, Overlap of the probed areas can be ensured. Diffraction data are collected on a retractable point detector for a regular scanning transmission X-ray microscopic (STXM) measurement or on a 2D X-ray CCD for a ptychography measurements. An attenuator is placed between the CCD and the sample to avoid damage and saturation of the CCD and to expand the dynamic range. **Figure 4** shows a photograph of the instrument as implemented at TACoDE. The following major system components are incorporated: pinhole x, y, z stages; sample x, y stages; point detector x stage; attenuator x, y stages and CCD detector.

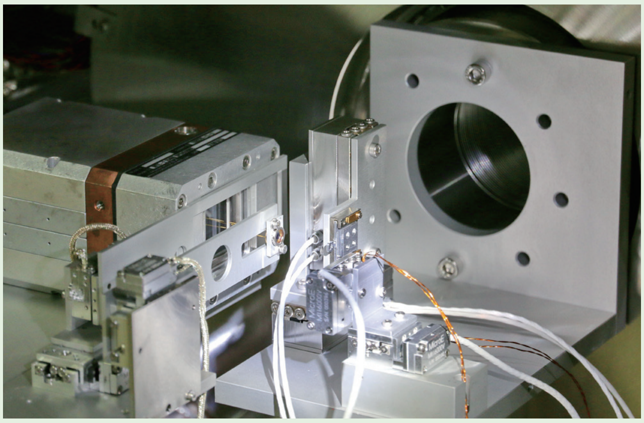


Fig. 4: Photograph of a soft X-ray ptychography.

With this geometry and wavelength, some of the following properties can be estimated.

Resolution limit

The maximum diffraction angle is found to be $\theta_{max} = \tan\theta_{max} = f_{1/2}/z$, in which $f_{1/2}$ is the half frame size of the CCD and z is the distance from sample to detector. In this geometry, $f_{1/2}$ is 13.8 mm, z is 70 mm; thus θ_{max} is 0.197 radian. As mentioned, CDI and ptychography can in principle overcome the limitation of the X-ray optics, but the numerical aperture of the detector limits the spatial resolution, *i.e.* $d = \lambda/2\,N\,A_{CCD} = \lambda/2\theta_{max}$. For $\lambda = 1.4$ nm, the resolution limit is 4 nm.

Coherent flux

In **Fig. 1**, the coherent flux before the pinhole is 1.5 x 10^{11} photons/s with resolving power 5000. For a KB focused beam size 3 x 3 μm^2, a pinhole of diameter 1 µm further decreases the flux by a factor $\pi/36$ leading to a coherent flux 1.3 x 10^{10} photons/s.

Illuminated area

The temporal coherent length in this design is found to be $l_t = \lambda^2/2\,\Delta\lambda = 7$ µm, in which $\lambda = 1.4$ nm and $\lambda/\Delta\lambda = 5000$. The temporal coherent length must be greater than the maximum path difference between incident rays. In this case, l_t should be greater than $w\,\theta_{max}/2$, *i.e.* $l_t > w\,\theta_{max}/2$ in which w is the illuminated area and θ_{max} is the maximum diffraction angle. In this geometry, the maximum illuminated area is 71 x 71 μm^2.

The spatial coherent length is defined as $l_s = \lambda R/2a$, in which λ is the wavelength, R is the distance between pinhole and sample and a is the width of the pinhole. For λ = 1.4 nm, R = 1.5 mm, a = 1 μm, the spatial coherent length l_s is equal to 1 μm; the maximum illumination area is thus 2 x 2 μm^2. In conclusion, the maximum illuminated area is dominated by the spatial coherent length because of the large pinhole.

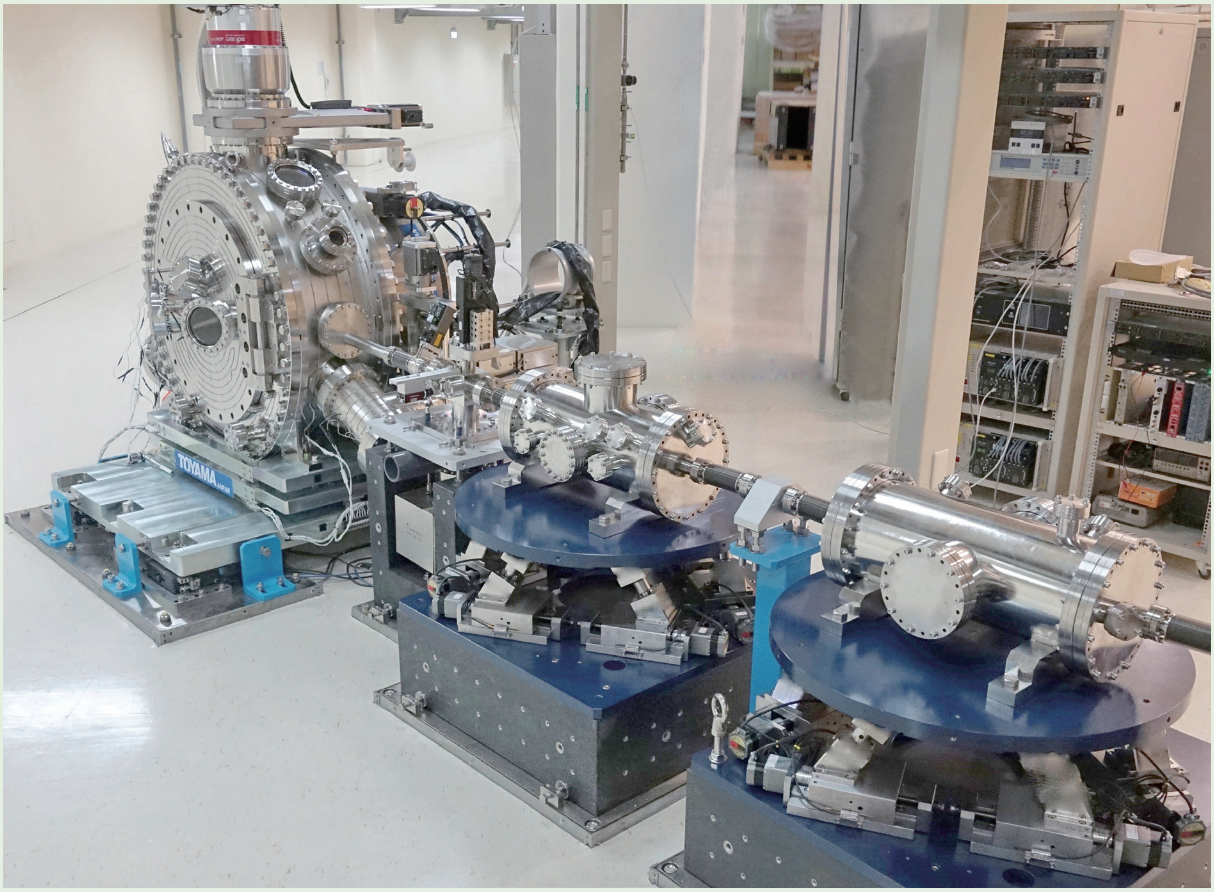
Speckle pattern

The Shannon interval[7] for frequency-space sampling of the intensity is $1/2\,w = \Delta/\lambda z$, in which w is the illuminated area, Δ is the pixel size of the CCD, z is the distance from sample to detector. For λ = 1.4 nm, z = 70 mm, w = 1 μm, the pixel size of the CCD is equal to 49 μm; *i.e.* to provide correct Shannon sampling of the diffraction plane intensity, the pixel size of the CCD must be less than 49 μm. The pixel size of the CCD that we intend to use is 13.5 μm.

In summary, the soft X-ray ptychography at TPS will enable studies that can be defined as true nano-science with spatial resolution less than 10 nm. The combination of great spatial resolution, great chemical sensitivity and a large field of view of spectro-ptychography will be a promising probe for detailed analysis of nature in various fields. At present, the design and construction is almost finished; commissioning and opening to users will begin next year. (Reported by Hung-Wei Shiu)

| References |

1. A. P. Hitchcock, and J. Electron, Spectrosc. Relat. Phenom. **200**, 49 (2015).
2. D. A. Shapiro, Y.-S. Yu, T. Tyliszczak, J. Cabana, R. Celestre, W. Chao, K. Kaznatcheev, A. L. D. Kilcoyne, F. Maia, S. Marchesini, Y. S. Meng, T. Warwick, L. L. Yang, and H. A. Padmore, Nat. Photon **8**, 765 (2014).
3. J. M. Rodenburg, A. C. Hurst, A. G. Cullis, B. R. Dobson, F. Pfeiffer, O. Bunk, C. David, K. Jefimovs, and I. Johnson, Phys. Rev. Lett. **98**, 034801 (2007).
4. Y. Takahashi, A. Suzuki, N. Zettsu, Y. Kohmura, Y. Senba, H. Ohashi, K. Yamauchi, and T. Ishikawa, Phys. Rev. B **83**, 214109 (2011).
5. X. Zhu, A. P. Hitchcock, D. A. Bazylinski, P. Denes, J. Joseph, U. Lins, S. Marchesini, H.-W. Shiu, T. Tyliszczak, and D. A. Shapiro, Proc. Natl. Acad. Sci. **113**, E8219 (2016).
6.. X. H. Zhu, T. Tyliszczak, H.-W. Shiu, D. Shapiro, D. A. Bazylinski, U. Lins, and A. P. Hitchcock, AIP Conf. Proc. **1696**, 020002 (2016).
7. V. Friso van der, and P. Franz, J. Phys. Condens. Matter **16**, 5003 (2004).



TPS 41A Soft X-ray Scattering