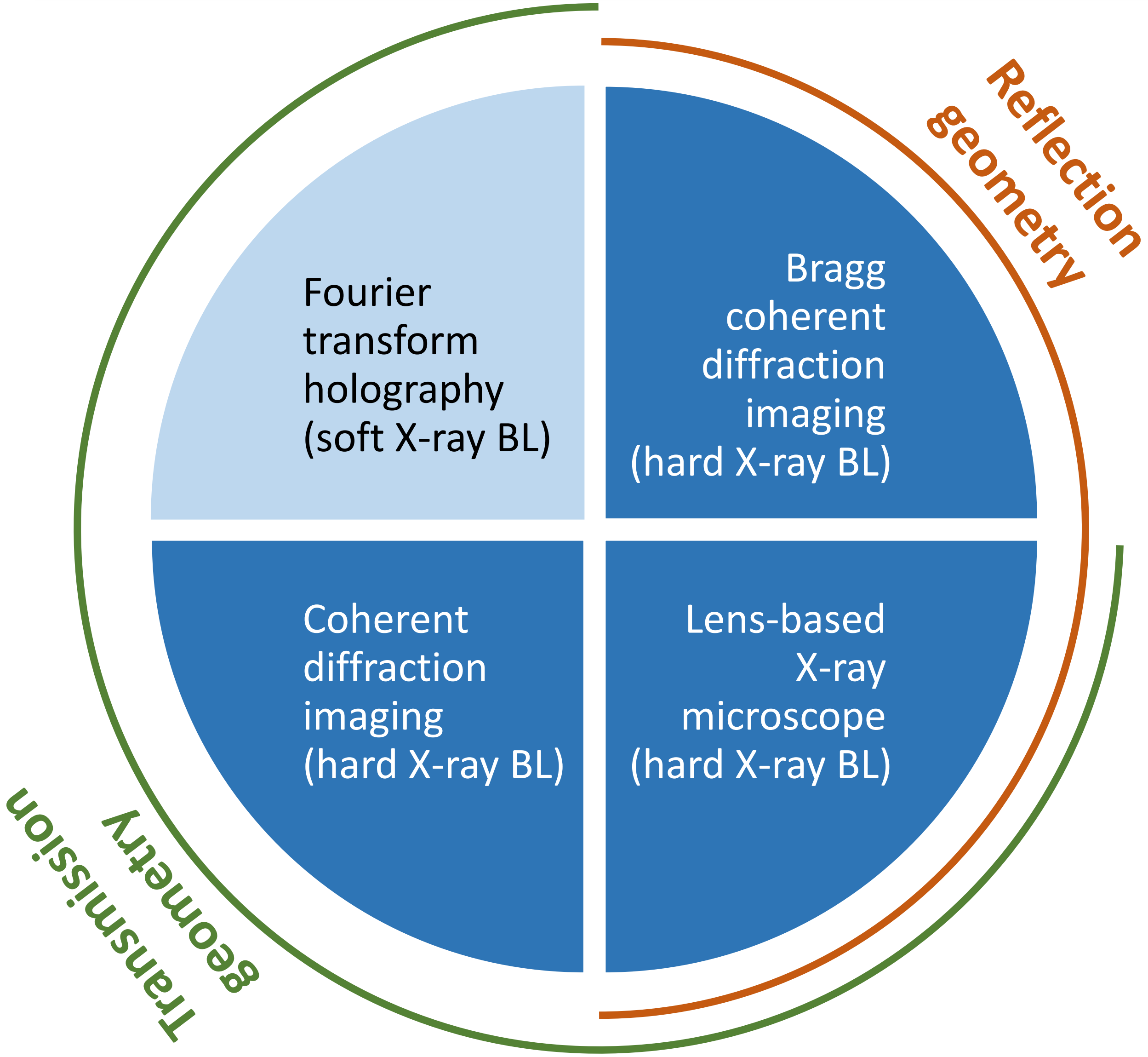
Imaging Science at PAL-XFEL
:Visualization of ultrafast phenomena at nanoscale resolution

Daewoong Nam

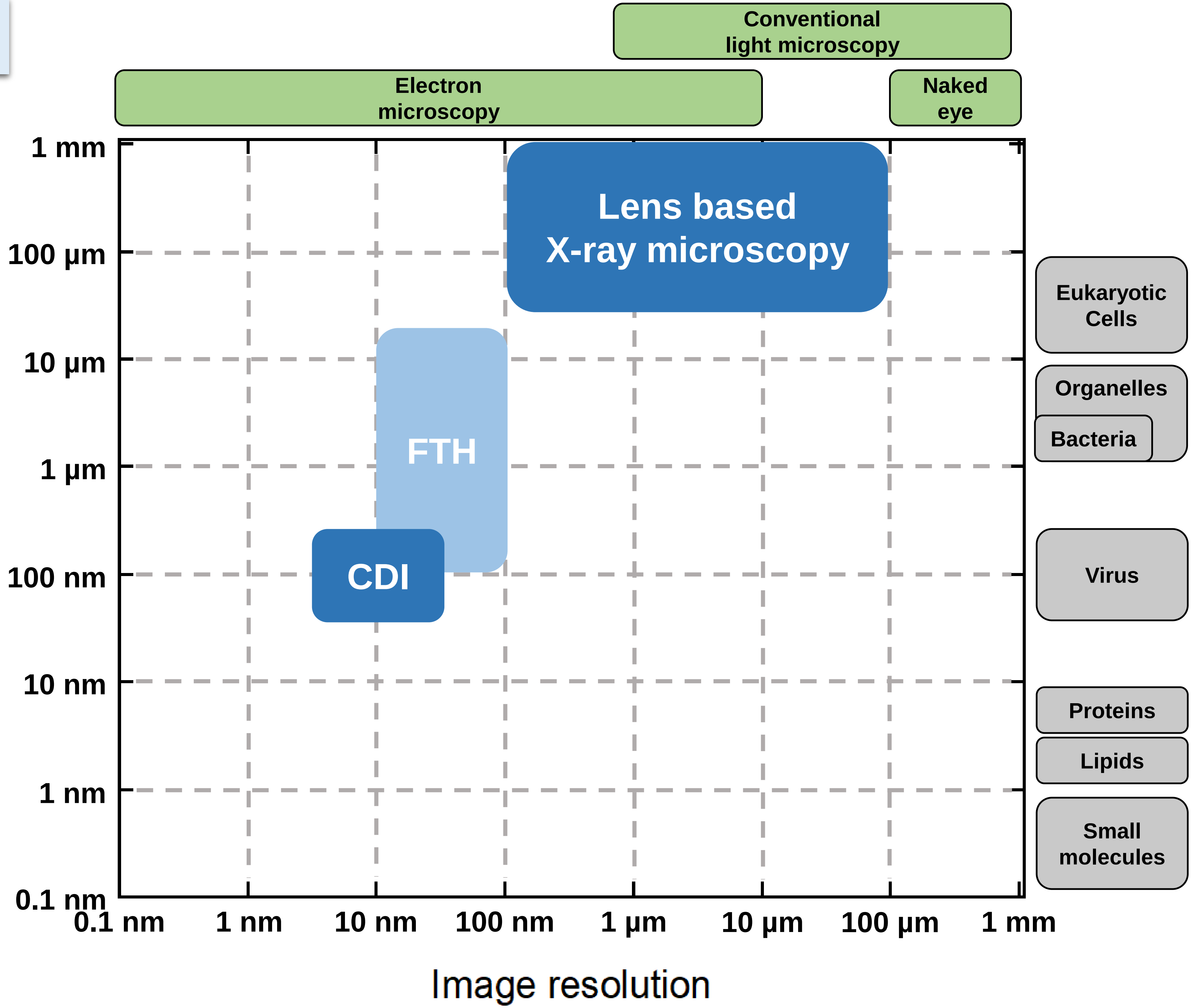
November 14, 2023

Contents

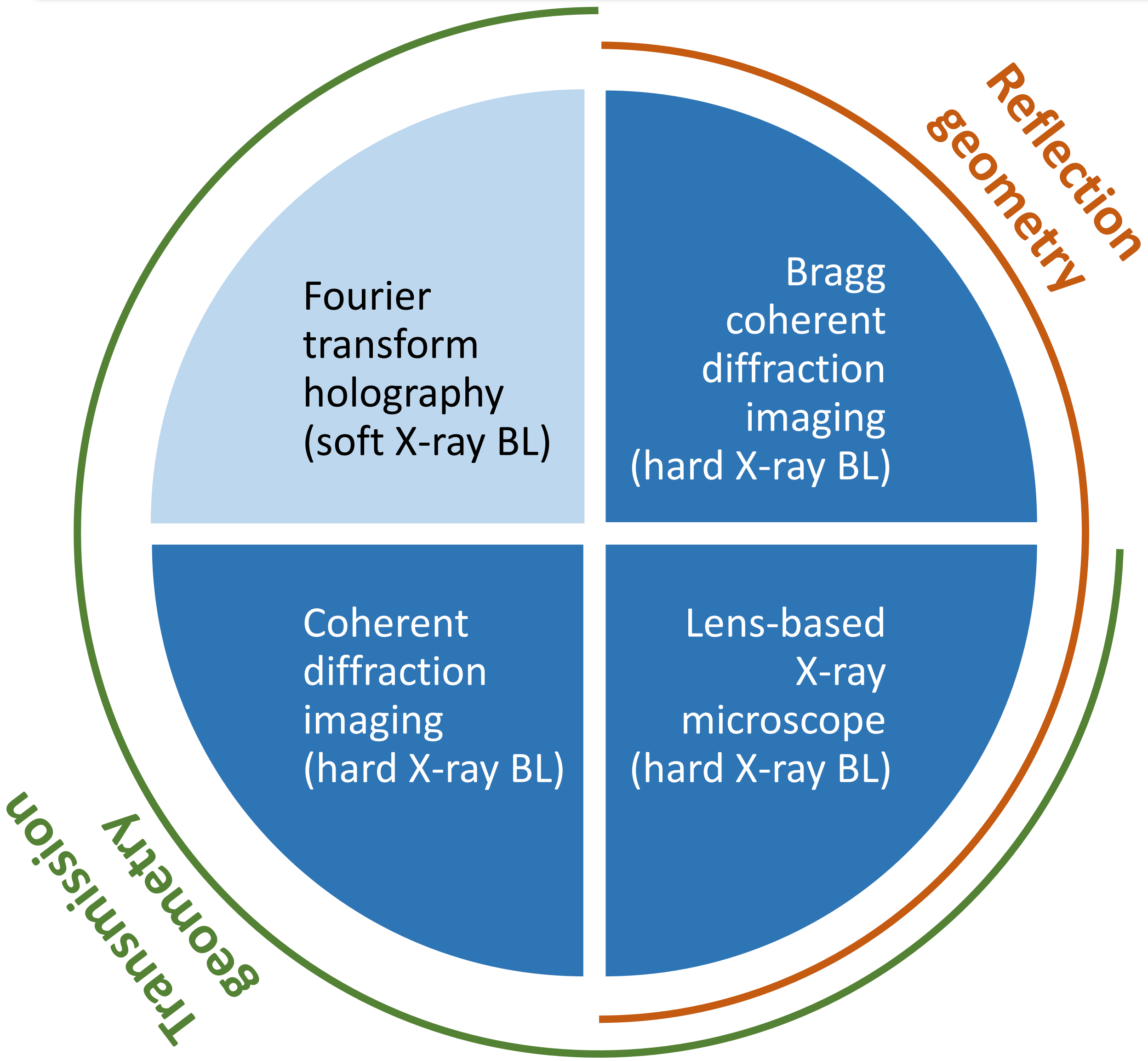
Imaging science programs at PAL-XFEL

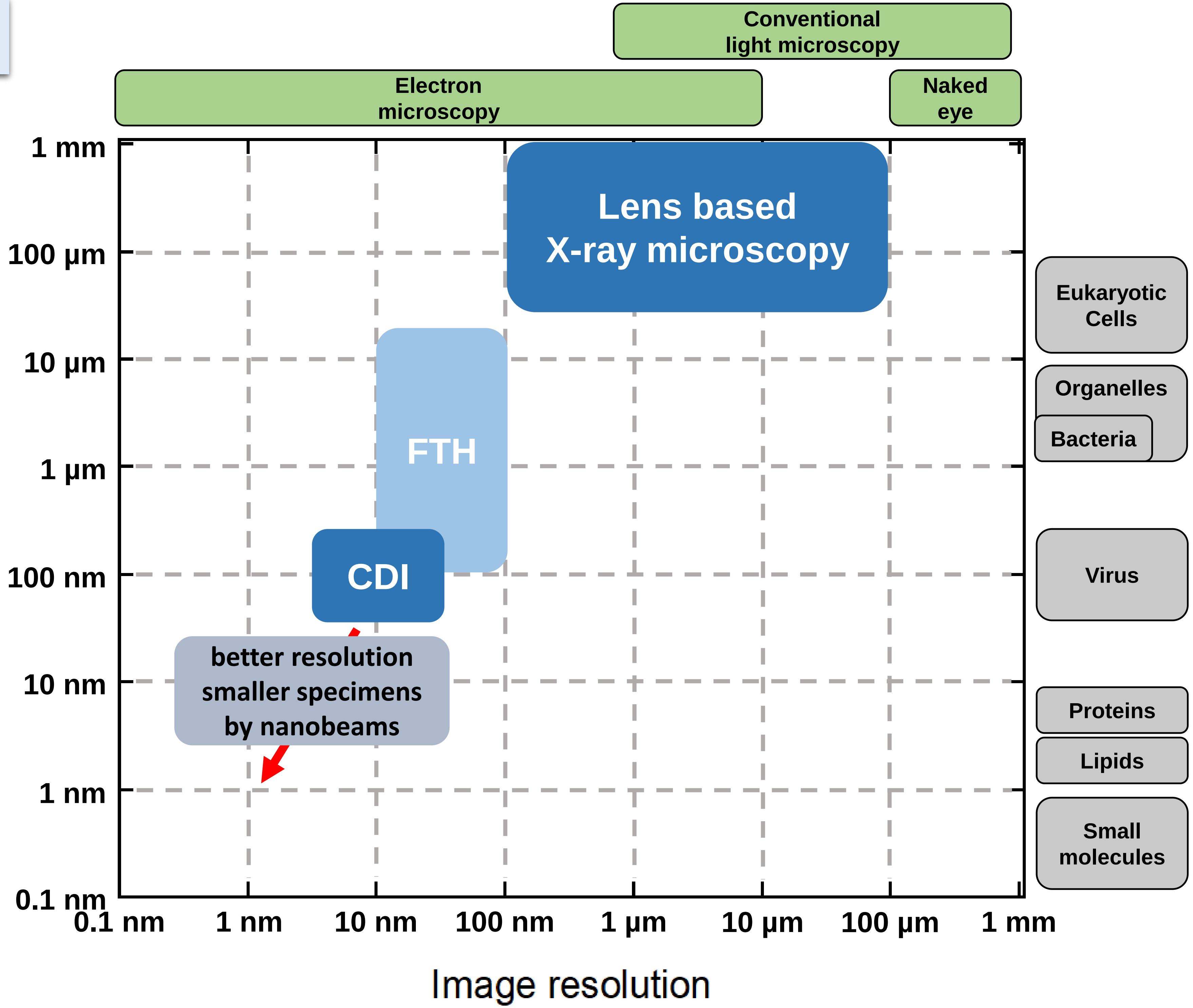


"We recommend the most suitable imaging method to uncover users' specific interests."



Imaging science programs at PAL-XFEL



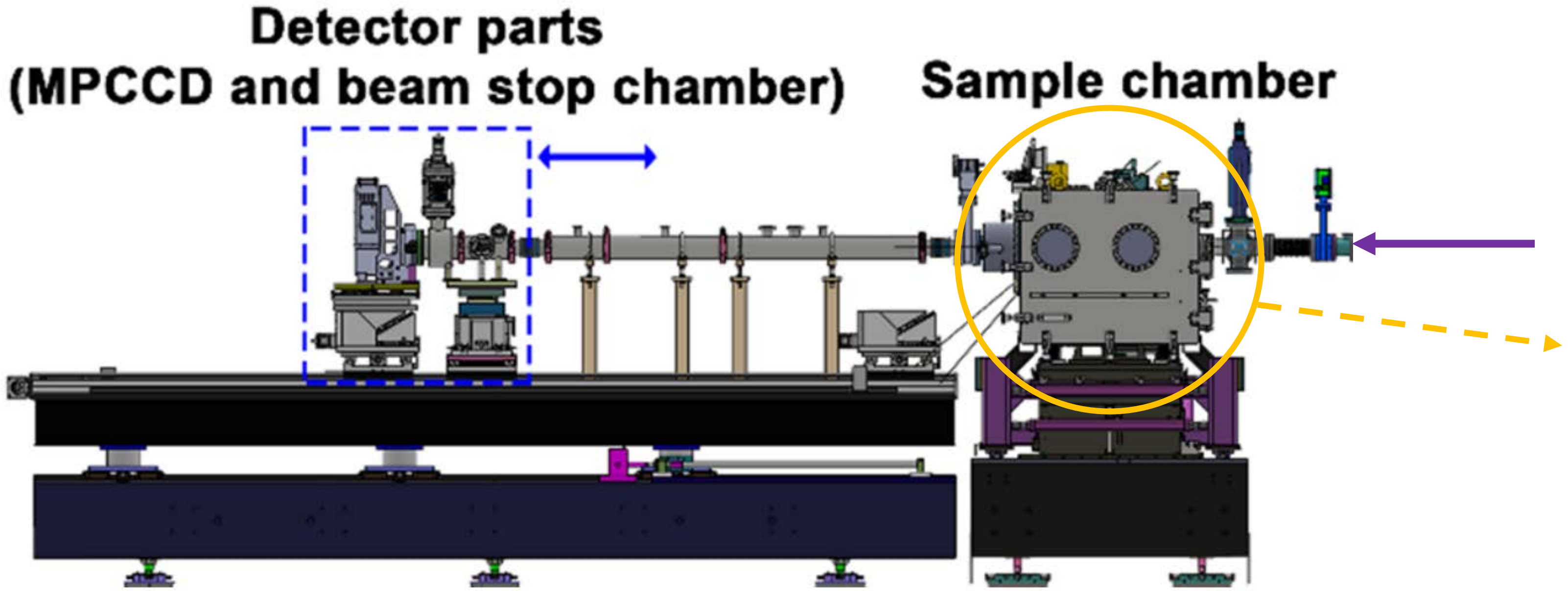


"We are constantly improving our instrument and developing new ones."

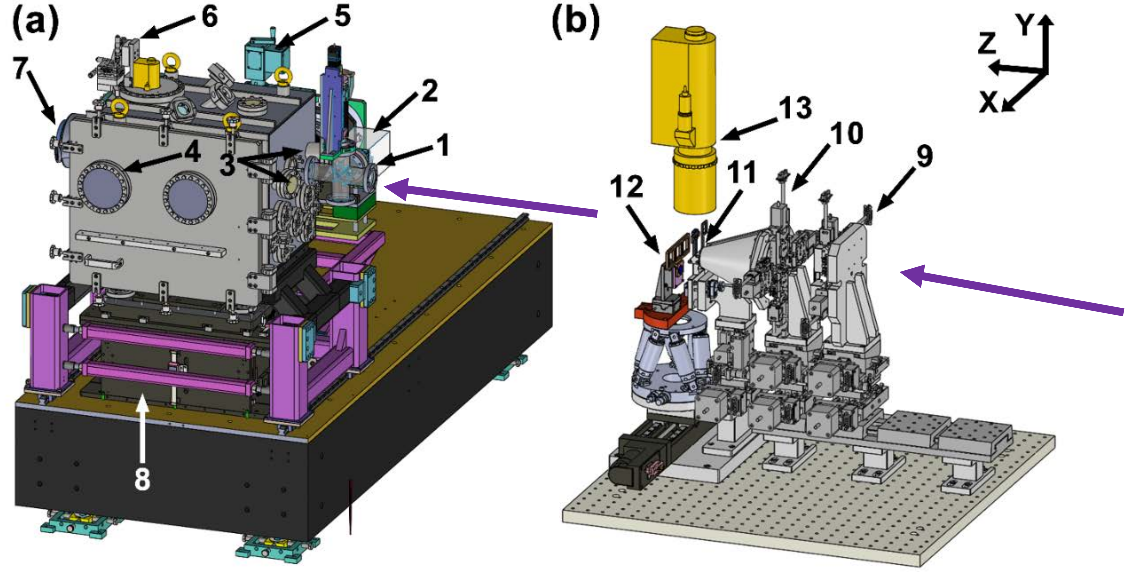
Coherent diffraction imaging at PAL-XFEL

2017

Early user experiment & User beamtime	Instrument	Remarks
CXI chamber commissioning Coherent measurement Single-shot imaging	**CXI chamber** **MPCCD 1M (Kapton film, air gap, Be window)**	Beam characterization (H. Lee *et al.*, **J. Synchrotron Rad. 27** (2020)) 3D single-particle imaging (D. H. Cho *et al.*, **ACS Nano 15** (2021)) Denoising method using machine learning (S. Y. Lee *et al.*, **Phys. Rev. Research 3** (2021))



(figure) Single-shot coherent X-ray imaging instrument at PAL-XFEL. X-ray free electron laser (XFEL) pulses are focused by Kirkpatrick–Baez mirrors (KB mirrors) with a ~5.96 m long focal length and delivered to the sample chamber. Fresh samples are supplied using a thin membrane to each X-ray pulse in the sample chamber. Diffracted signals from samples are collected by the multiport charge-coupled device (MPCCD). To avoid damage to the sensor, the beam stopper is located just upstream of the detector and blocks the direct X-ray beam. The detector parts marked by the blue box are mounted on the rail to easily change the distance between the sample and detector.



(figure) (a) The overall schematics of the sample chamber. (b) Internal components of the sample chamber. (the XFEL beam propagates following the z-axis in the figure)

D. Sung, D. Nam *et al.*, **Appl. Sci. 11**, 5082 (2021)

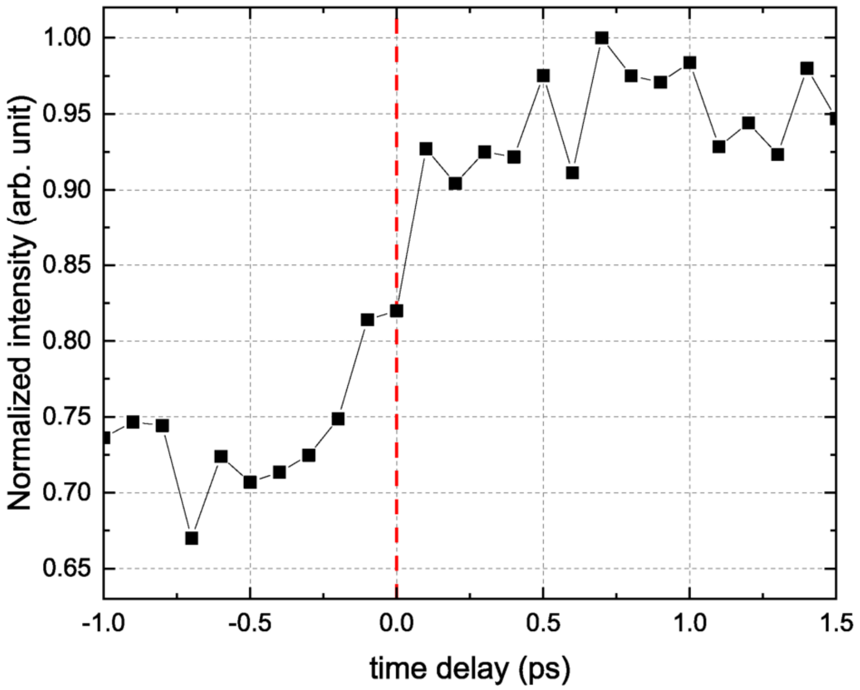
Coherent diffraction imaging at PAL-XFEL

2018

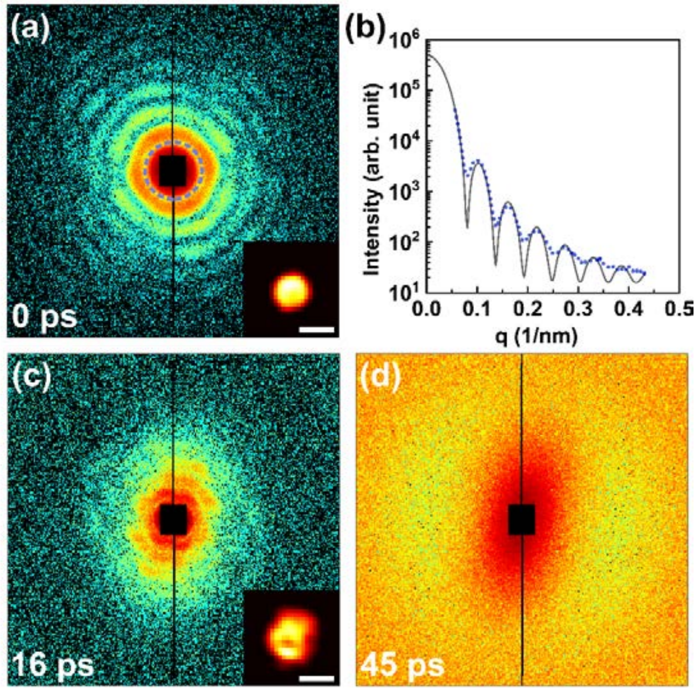
Machine study & User beamtime	Instrument	Remarks
MS: timing using a YAG **Pump-probe imaging**	CXI chamber MPCCD 1M (Kapton film, air gap, Be window)	Established the pump-probe imaging program at PAL-XFEL

2019

User beamtime & R&D beamtime	Instrument	Remarks
Pump-probe imaging R&D: injector commissioning	CXI chamber **JUNGFRAU 4M (Kapton film, air gap)**	Commissioning of an aerosol injector and the JUNGFRAU 4M. First pump-probe imaging result (C. Jung *et al.*, **Sci. Adv. 7** (2021)) CXI instrument (D. Nam *et al.*, **JKPS 76** (2020), D. Sung *et al.*, **Appl. Sci. 11** (2021))



(figure) A Ce:YAG crystal is used to synchronize X-ray and a femtosecond optical laser in the fixed target scheme. It enables to synchronize two light sources with femtosecond precision.



(figure) Demonstration of pump-probe imaging. (a) Diffraction pattern of an intact Ag nanosphere. (b) Radial summation of (a). (c) & (d) The measured diffraction patterns with a time delay of 16 ps and 45 ps.

We demonstrated a single-shot pump-probe imaging, which can **visualize the ultrafast phenomena of the specimens at picosecond temporal and nanometer spatial resolutions**. In particular, it can be applied to investigate **irreversible phenomena** in the case of homogenous specimens.

D. Sung, D. Nam *et al.*, **Appl. Sci. 11**, 5082 (2021)

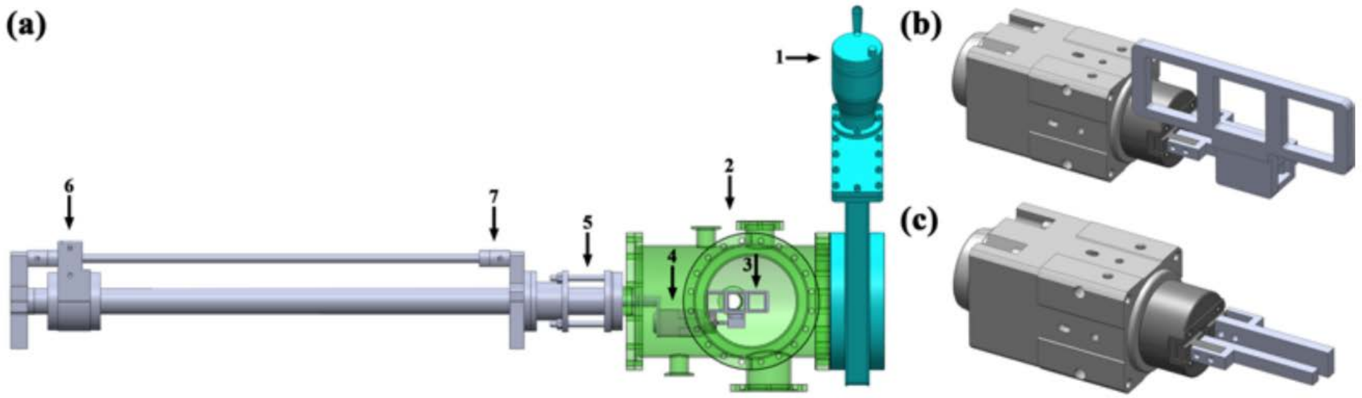
Coherent diffraction imaging at PAL-XFEL

2018

Machine study & User beamtime	**Instrument**	**Remarks**
MS: timing using a YAG **Pump-probe imaging**	CXI chamber MPCCD 1M (Kapton film, air gap, Be window)	Established the pump-probe imaging program at PAL-XFEL

2019

User beamtime & R&D beamtime	**Instrument**	**Remarks**
Pump-probe imaging R&D: injector commissioning	CXI chamber **JUNGFRAU 4M (Kapton film, air gap)**	Commissioning of an aerosol injector and the JUNGFRAU 4M. First pump-probe imaging result (C. Jung *et al.*, **Sci. Adv. 7** (2021)) CXI instrument (D. Nam *et al.*, **JKPS 76** (2020), D. Sung *et al.*, **Appl. Sci. 11** (2021))



(figure) A schematic view of the load lock chamber including a compact gripper and a sample holder for fixed target.

Table 1. Comparison of parameters for Load lock chamber and the existing systems with 30 Hz operation.

	Adopt the fast sample-exchange system	Vent and Pump the sample chamber
Time for data acquisition for three membranes	∼ 3 hours (1 hour/membrane)	∼ 3 hours (1 hour/membrane)
Time to exchange samples	Less than 10 minutes	∼ 30 minutes
Number of diffraction patterns collected in 6 shifts	88,128 diffraction patterns (68 membranes)	79,056 diffraction patterns (61 membranes)

D. Nam *et al.*, **J. Korean Phys. Soc. 76**, 6 (2022)

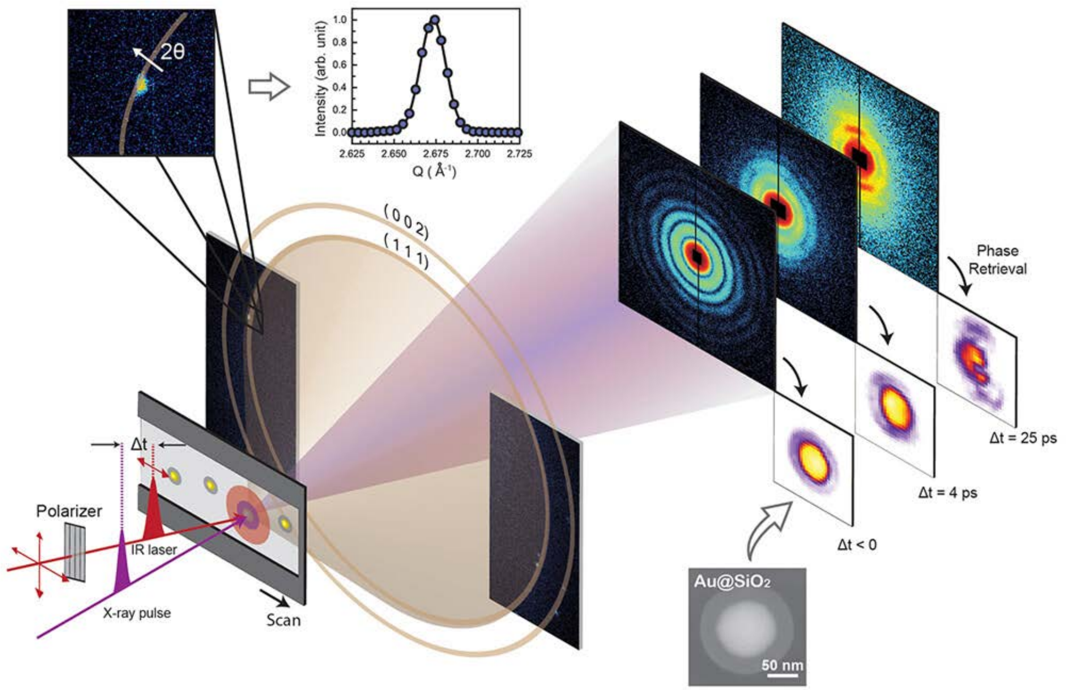
Coherent diffraction imaging at PAL-XFEL

2020

User beamtime	Instrument	Remarks
Pump-probe multiplex experiments	CXI chamber JUNGFRAU 4M for SAXD **JUNGFRAU 0.5M X 2 for WAXD**	Multiplexing experiment; collecting SAXD and WAXD simultaneously First multiplexing experiment result (J. Shin *et al.*, Nano Lett. **23** (2023))

❖ Multiplexing experiment

- Investigation using complementary X-ray techniques for comprehensive understanding of the ultrafast phenomena.
- Imaging: revealing the morphological information at nanoscale resolution
- WAXD: revealing the crystal arrangement at the atomic scale



(figure) Pump-probe multiplex imaging using femtosecond X-ray pulses. A femtosecond IR laser drives the melting process of core-shell nanoparticles and small/wide-angle X-ray diffraction patterns are simultaneously collected while changing the time delay of two light sources.

By modifying the CXI chamber's exit port, we could accommodate **two JUNGFRAU 0.5M**, aimed at collecting Bragg peaks at **a fixed two-theta angle of 30 degrees**.

However, the azimuthal coverage of these two detectors remains partial.

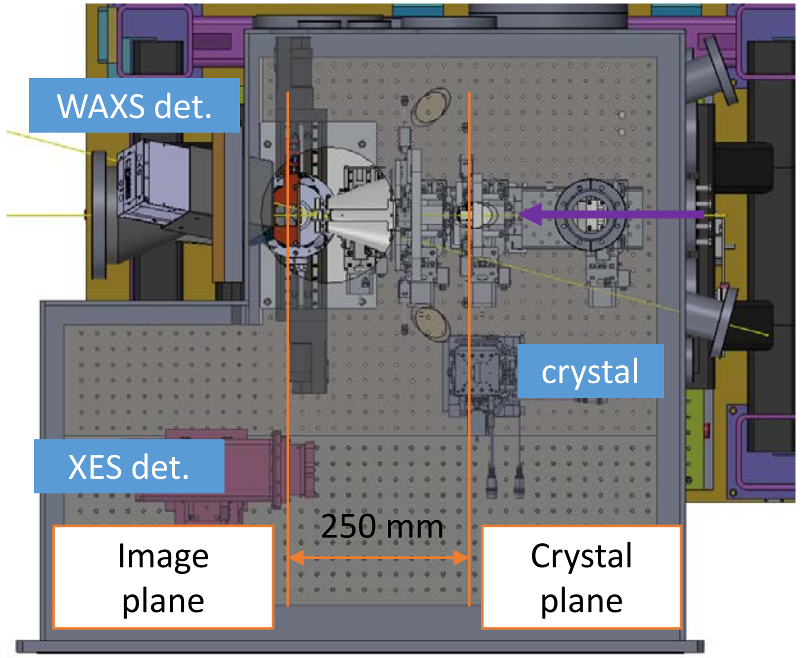
While this setup suffices for comprehending the atomic arrangement of polycrystalline samples, **its effectiveness is limited when studying single crystal samples** due to the random distribution of sample orientations on the thin membrane.

J. Shin *et al.*, **Nano Lett. 23**, 4 (2023)

Coherent diffraction imaging at PAL-XFEL

2021 - 2022

User beamtime & R&D beamtime	Instrument	Remarks
Pump-probe multiplex experiments R&D: multiplex imaging chamber commissioning	CXI chamber **multiplex imaging chamber** JUNGFRAU 4M **JUNGFRAU 0.5M X 3 for WAXD and XES**	Commissioning of a multiplex imaging chamber -SAXD, WAXD, XES



- **Small Angle X-ray Diffraction (SAXD)**
  : for obtaining morphological images
- **Wide Angle X-ray Diffraction (WAXD)**
  : for obtaining the crystal arrangement at the atomic-scale
- **X-ray Emission Spectroscopy (XES)**
  : for identifying corresponding electronic structure

❖ **SAXD**

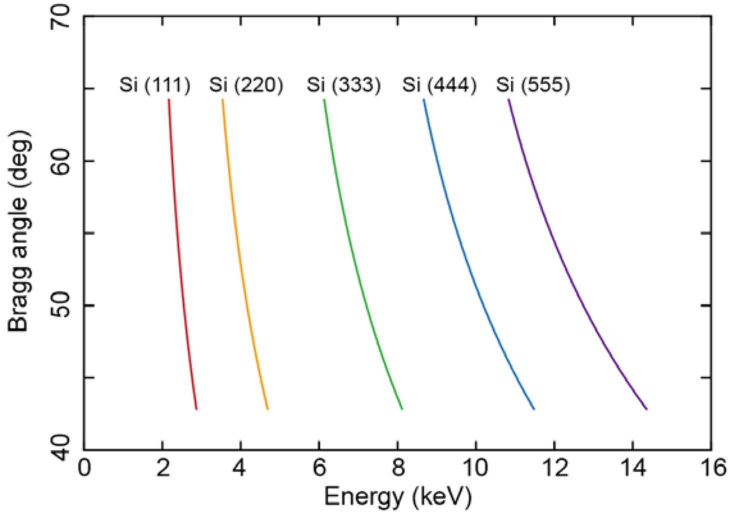
- JUNGFRAU 4M

❖ **WAXD**

- Two JUNGFRAU 0.5M
- 2th is fixed, 30°±7°.
- azimuth angle: ~ 38 % covered by two detectors

❖ **XES (Von Hamos spectrometer)**

- JUNGFRAU 0.5M
- crystal: Si(111) and Si(220)
- Acceptable 2th range: 45° ~ 61° in this setup

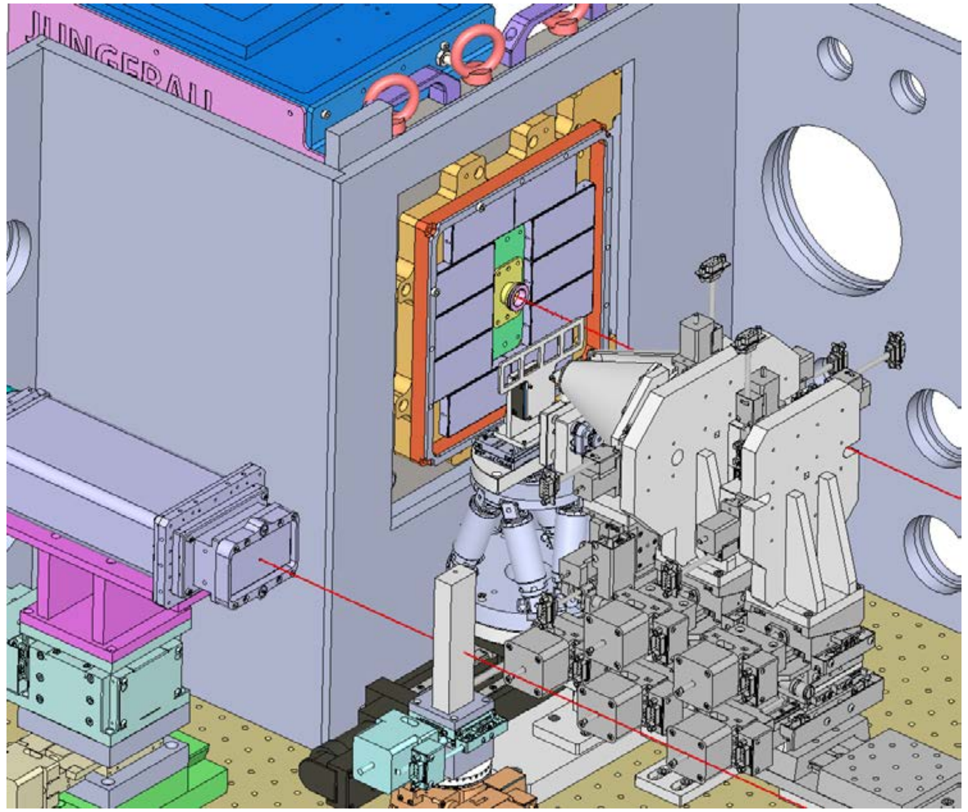


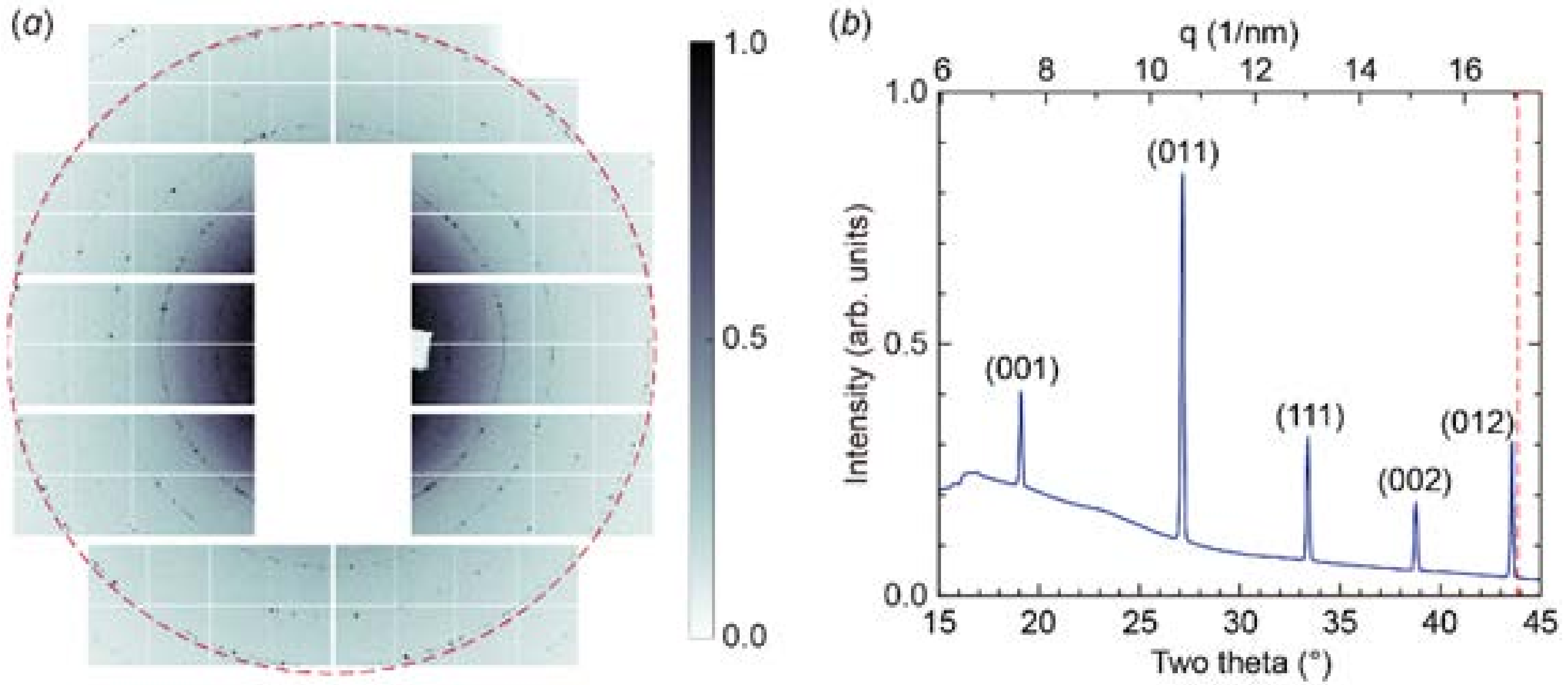
(figure) The spectrometer energy range with the incident angle of X-rays for Si(111) and Si(220) crystal analyzer

Coherent diffraction imaging at PAL-XFEL

2023

User beamtime	Instrument	Remarks
Pump-probe multiplex experiments	multiplex imaging chamber JUNGFRAU 4M for SAXD **JUNGFRAU 5M for WAXD** JUNGFRAU 0.5M for XES	A new large area detector for WAXD





(figure) (a) Diffraction pattern from LaB_6 powder. (b) Radial summation of (a).

❖ WAXD

- Two JUNGFRAU 0.5M
- 2th is fixed, 30°±7°.
- azimuth angle: ~ 38 % covered by two detectors

->

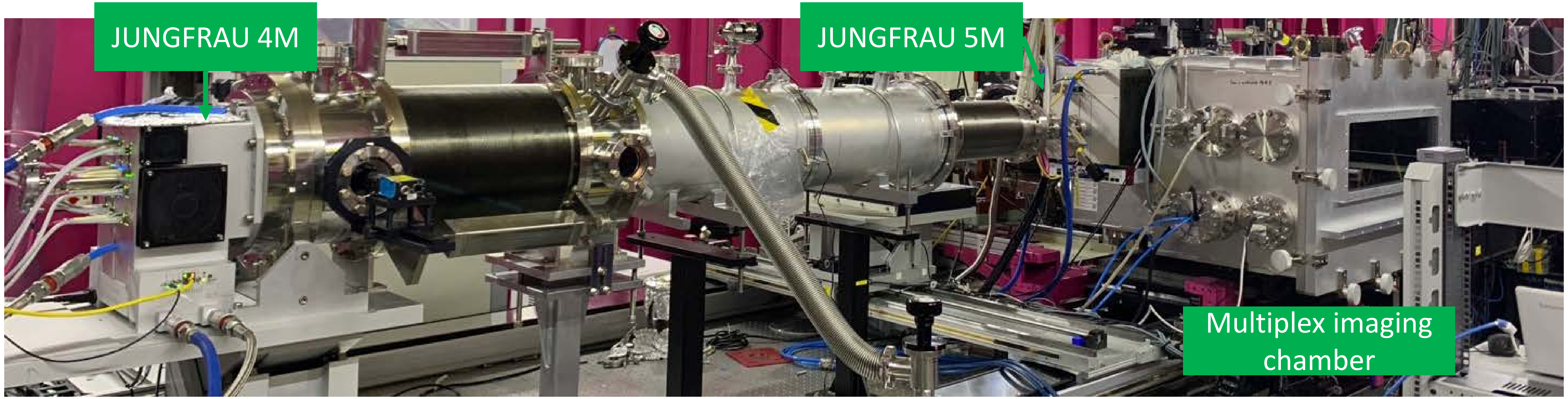
- JUNGFRAU 5M
- 2th range: 18°~ 44°
- Fully covered in azimuthal direction (2th: 33°~ 44°)

J. Hwang *et al.*, in preparation

Multiplex imaging chamber

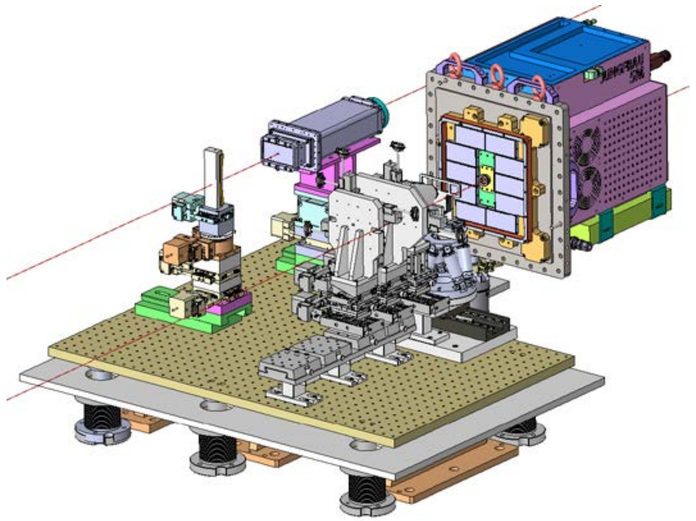
❖ Multiplexing experiment

- Investigation using complementary X-ray techniques for comprehensive understanding of the ultrafast phenomena.



(figure) A photograph of the multiplex imaging chamber and two detectors to collect small/wide-angle X-ray diffraction patterns. The whole instrument is installed on the NCI hutch at hard X-ray beamline, PAL-XFEL.

- **Small Angle X-ray Diffraction (SAXD)**
  : for obtaining morphological images
- **Wide Angle X-ray Diffraction (WAXD)**
  : for obtaining the crystal arrangement at the atomic-scale
- **X-ray Emission Spectroscopy (XES)**
  : for identifying corresponding electronic structure



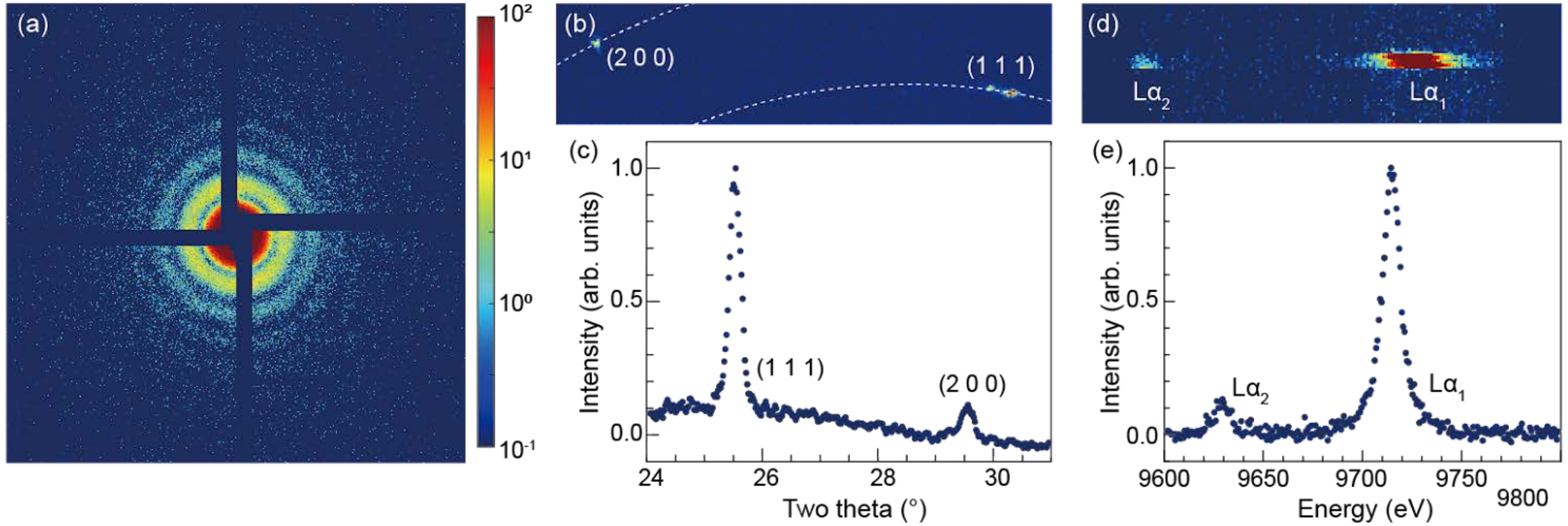
(figure) 3D schematic showing the interior parts of multiplex imaging chamber. JUNGFRAU 5M for WAXD and JUNGFRAU 0.5 M for XES are mounted. Von-Hamos spectrometer is used to reveal electronic structural information of the specimens.

Multiplex imaging chamber

❖ Commissioning of multiplex imaging experiment

- Photon energy is determined by considering following three factors.
: the oversampling ratio for the imaging experiment
: the position of the Bragg peak under the fixed detector geometry
: the X-ray absorption edge of the target atom.

❖ Simultaneous measurement of three different signals of gold nanoparticles

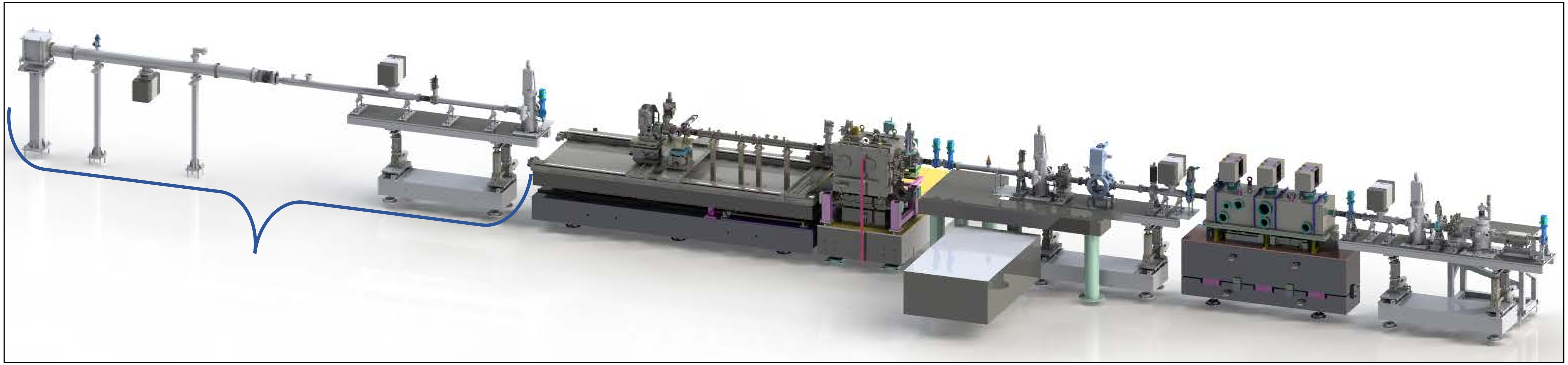


(figure) (a) The small angle diffraction pattern of a single Au NP. (b) The periodic arrangement of the sample formed strong diffraction peaks on the detector. The observed diffraction peaks were (111) and (200) peaks corresponding to atomic spacings of 2.35 Å and 2.04 Å, respectively. (c) The diffraction intensity as a function of the two theta angles is shown by averaged over the azimuth angle. (b & c) Simultaneous measurements were taken with the same particle measured in (a). (d) Using the von Hamos spectrometer with a Si(444) crystal analyzer, Au Lα_1 (9.713 keV) and Lα_2 (9.628 keV) spectrum were collected. More than 1000 X-ray pulses were exposed to improve the signal-to-noise ratio (SNR) of emissions. In addition, the bent crystal allowed the X-rays to be focused vertically for better SNR. (e) The Lα_1 emission intensity is ~ 10 times higher than the Lα_2 emission. The energy resolution from the detector pixel size was 0.68 eV in this von Hamos geometry.

J. Hwang *et al.*, in preparation

New endstation equipped K-B mirrors for nanobeam

❖ **New project with photon science center, POSTECH**



(figure) A schematic of the NCI hutch in 2020.

- new endstation for studying of functional nanomaterial and biological specimens

- key instrument: nanofocusing optics

: improve X-ray flux density effectively

: promote high resolution imaging by increasing in X-ray flux density

: develop new science programs utilizing nanobeams

New endstation equipped K-B mirrors for nanobeam

❖ **Focusing optics: diagnostic part**

- Quadrant Beam Position Monitor (QBPM)

: Thin foils (Si_3N_4 membrane and CVD) and four PDs (backscattering geometry)

: Provide estimated the beam position and the incident flux without disturbing X-rays

- Slit

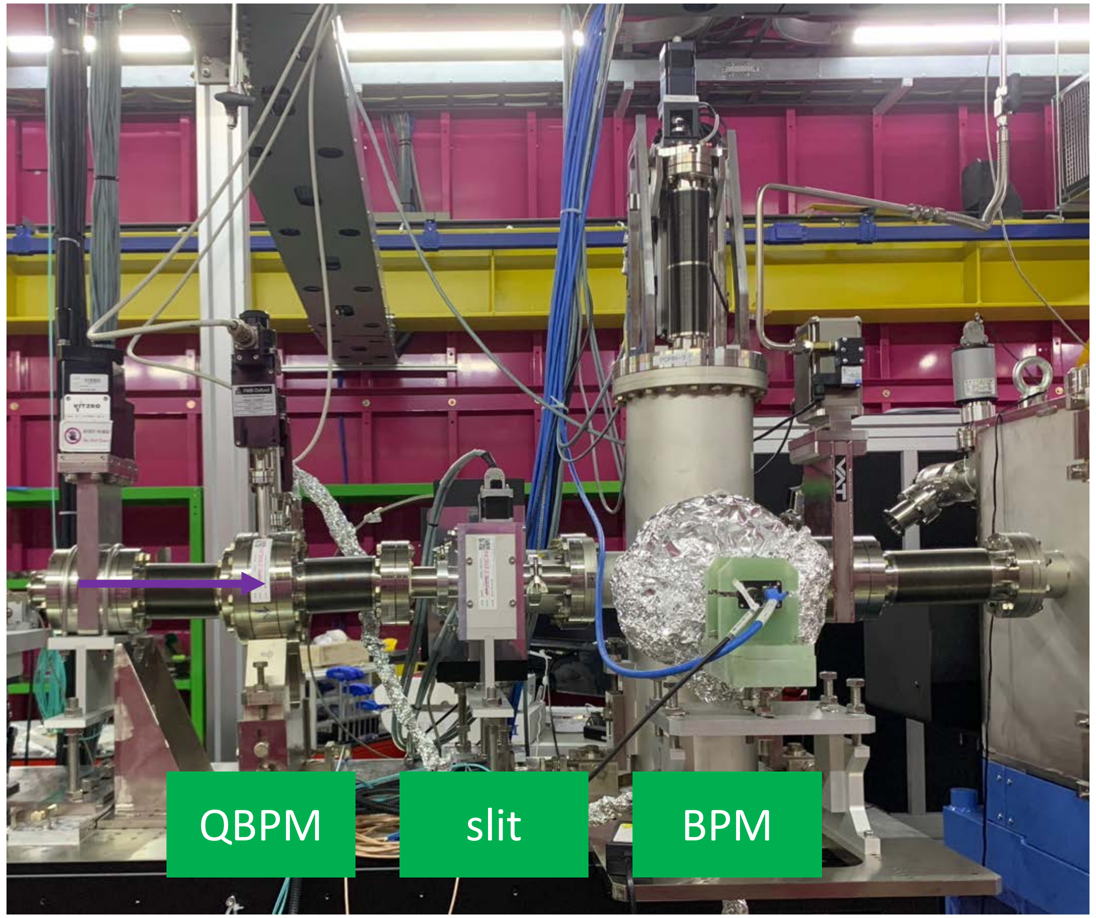
: 2 mm tungsten carbide blades X 4

: Block the parasitic scattering from upstream optics

- Beam Position Monitor (BPM)

: Ce:YAG crystal and vision camera

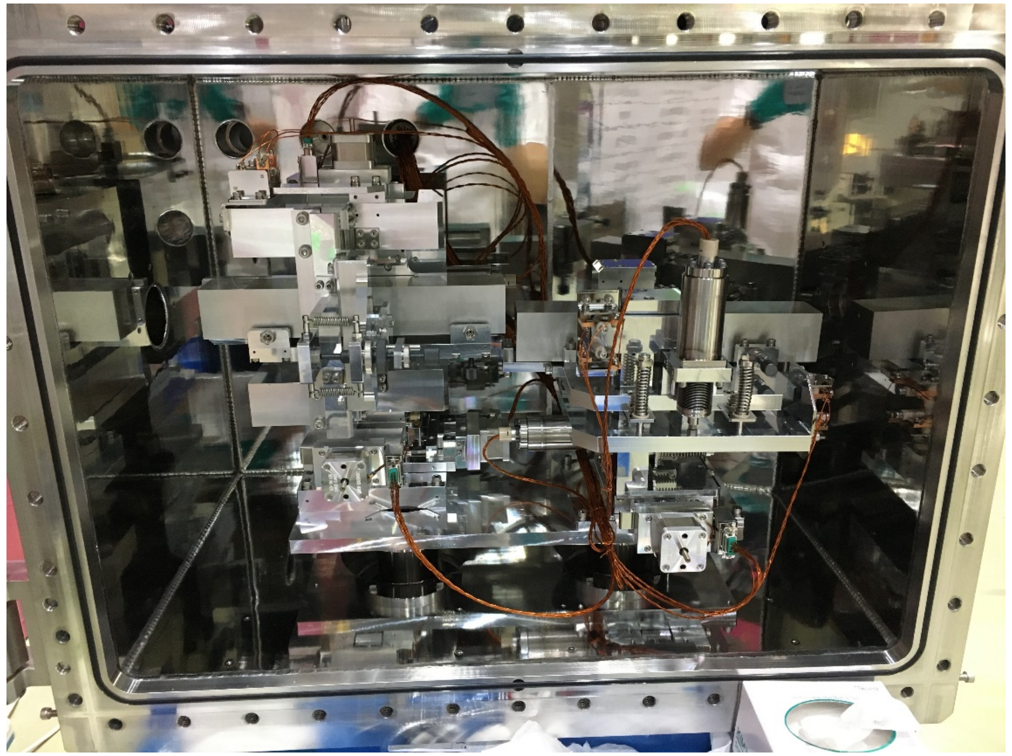
: Intuitively provide the X-ray position on the crystal but disturb X-rays



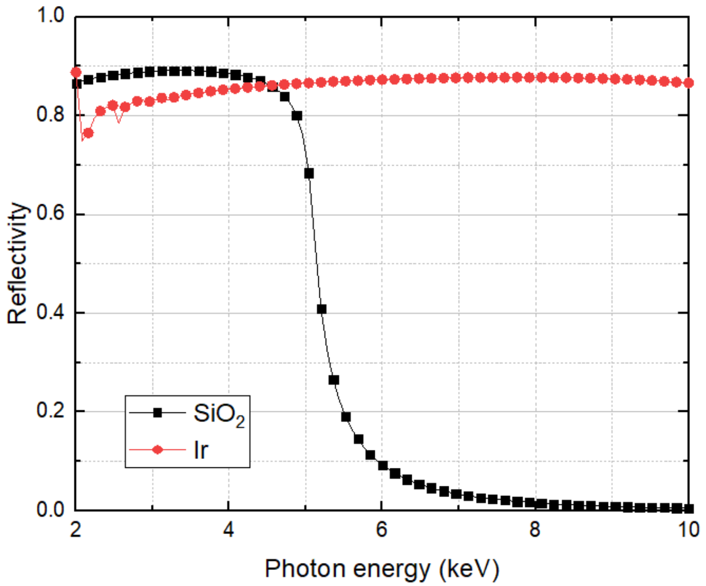
(figure) A photograph of the diagnostic part at the new endstation.

New endstation equipped K-B mirrors for nanobeam

❖ **Focusing optics: K-B mirrors**



(figure) A photograph of a new K-B mirrors chamber.



	Horizontal focusing mirror	Vertical focusing mirror
Surface profile	Elliptical cylinder	
Substrate material	SiO_2	
Surface coating	Ir 50 nm (6 mm width) and non-coated (7 mm width)	
Mirror substrate size	300 mm x 50 mm x 50 mm (L x W x H)	
Effective mirror area	290 mm x 15 mm	
Grazing incidence angle	6 mrad	
Focal length	910 mm	600 mm
Spatial acceptance	1.74 mm	1.74 mm

(table) Specification of new K-B mirrors.

Simulation

Source to mirror distance = 153,125 mm

Source size = 0.05 mm

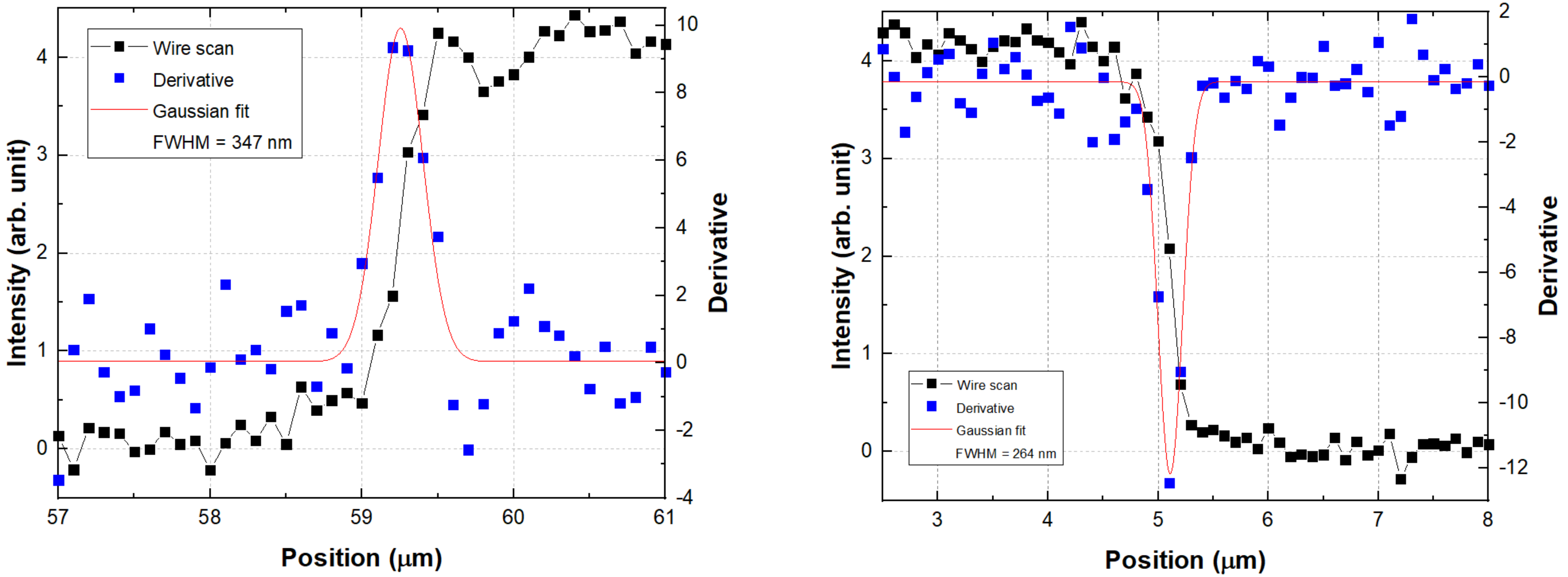
-> expected focused beam size (H X V) ~ 300 nm X 200 nm

New endstation equipped K-B mirrors for nanobeam

❖ **Focusing optics: K-B mirrors**

- photon energy: 9.5 keV
- reflection area: Ir
- focused beam size (FWHM): ~ 350 nm X 270 nm (H X V)

→ **The focused beam size closely matches the simulation value, 300 nm by 200 nm.**

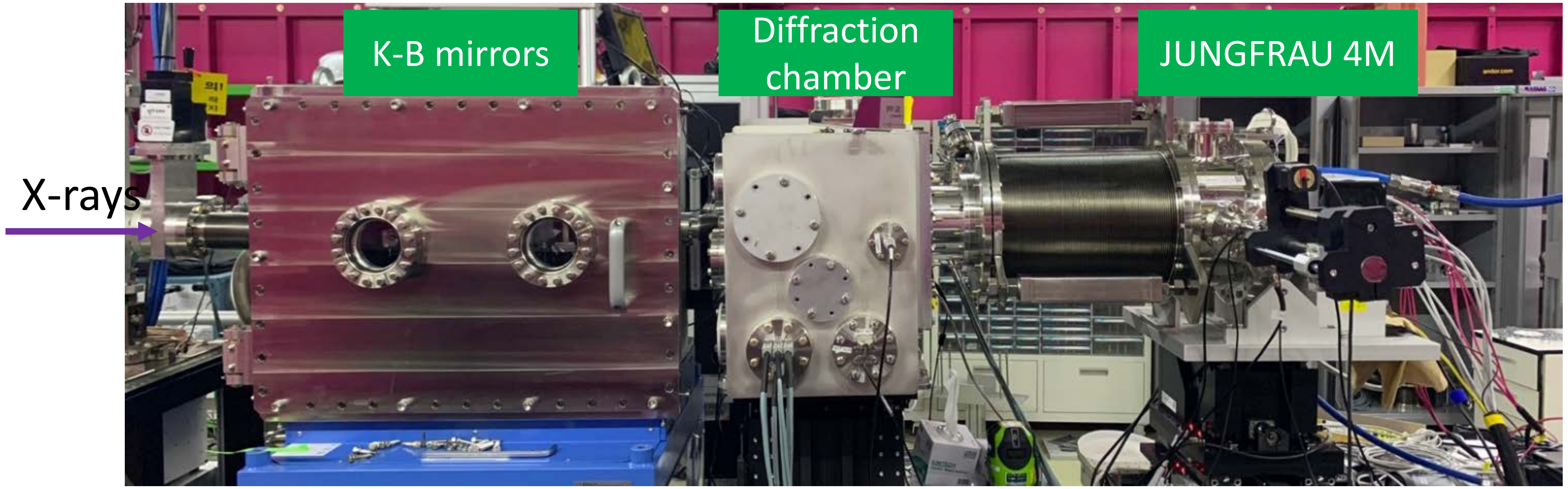


(figure) Focused beam size measurement by wire scan. The beam size is about 350 nm by 270 nm.

New endstation equipped K-B mirrors for nanobeam

❖ Instrument for coherent diffraction imaging

- Diffraction chamber including a slit stage, a mirror stage for microscope and a sample stage is ready.
- Fixed target is available with 60 Hz repetition rate.
- A large area detector, JUNGFRAU 4M, is available.
- Multiplex experiment, SAXD and WAXD, will be available from 2024.

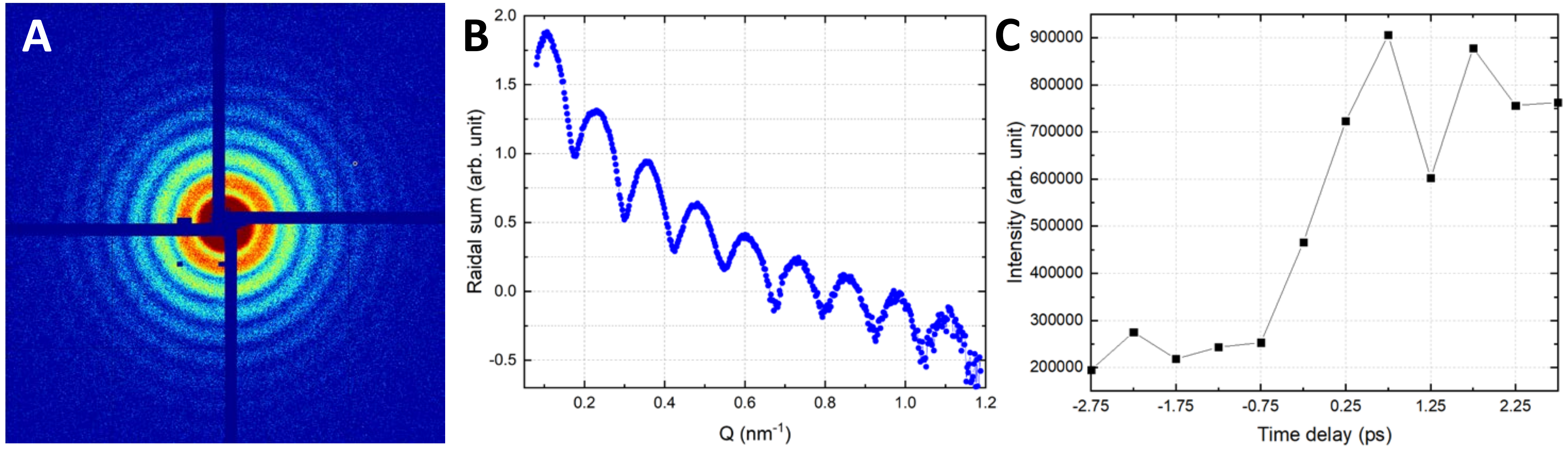


(figure) Imaging instrument installed at the new endstation.

New endstation equipped K-B mirrors for nanobeam

❖ Instrument for coherent diffraction imaging

- Single-shot imaging experiment has been successfully demonstrated at the new endstation.
- Gold nanospheres with 50 nm diameter were dispersed on the Si_3N_4 membrane.
- X-ray energy was set to 5 keV and focused beam size was ~ 500 nm by 650 nm.
- Fs. IR laser pump – X-ray probe experiment with sub ps precision is available now.



A Diffraction pattern of gold nanoparticle with 50 nm diameter.

B Radial summation of the diffraction pattern. The signal extends to ~ 5 nm in spatial resolution.

C Timing synchronization of two light sources, IR laser and X-rays, with femtosecond precision using a Ce:YAG crystal.

Summary

❖ **Multiplex imaging experiment**

- As the result of "Accelerator core technology development project" funded by NRF, the new instrument for multiplexing experiment has been developed.
- This instrument drives to comprehensive understanding of the ultrafast phenomena through three different information.

❖ **XFEL experiments using nanobeam**

- We've successfully constructed the new endstation equipped with K-B mirrors and demonstrated single-shot imaging with nanobeam.
- As the first nanofocusing system using K-B mirrors in Korea, we earned valuable know-how through this project.
- Investment for infra strengthens the capability of PAL staffs.
- 16 beamline scientists form a tie up with user groups to utilize nanobeams and we show the possibility to solve scientific problems using this endstation through internal beamtime.

❖ **CDI at the soft X-ray beamline**

- As another project with Photon Science Center, we are constructing a new endstation at the soft X-ray beamline.
- Tight focusing system by K-B mirrors is employed for single-shot experiment including coherent diffraction imaging.
- Commissioning of focusing system will be conducted in 2024.
- Because of the budget issues, manufacture of a sample chamber is stopped now.

Thanks to:

PAL-XFEL, PAL
Sangsoo Kim, Sunam Kim, Sang Han Park, Minseok Kim, Intae Eom, Tae-Yeong Koo and all staffs of XFEL beamline department

Photon Science Center, POSTECH
Changyong Song and Jae Hoon Park

Sogang Univ.
Hyunjung Kim

RIKEN SPring-8 Center
Makina Yabashi and Tetsuya Ishikawa