Beamline Renovation: New HRPD-II Program of BL4B

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Beamline Renovation Project

❖ **Proposal to renovate an existing beamline and convert analysis techniques**

- Switching the existing BM(bending magnet) beamline which has been in operation over 15 years to new analytical techniques for user requirement and research competitiveness

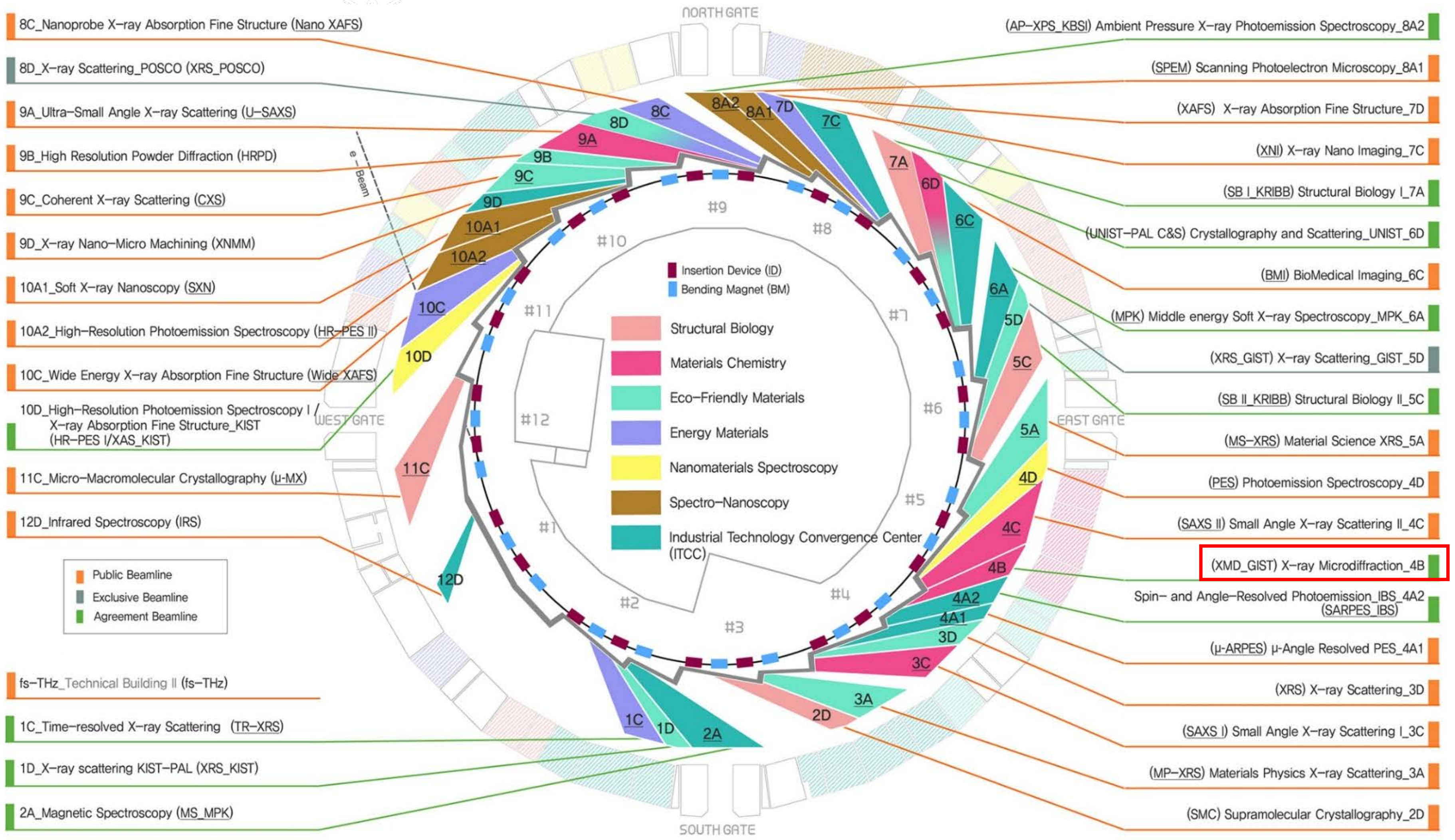

Figure 1. PLS-II beamline map

4B X-ray Microdiffraction beamline ('02~'23)

→ 4B High Resolution Powder Diffraction-II beamline ('27~

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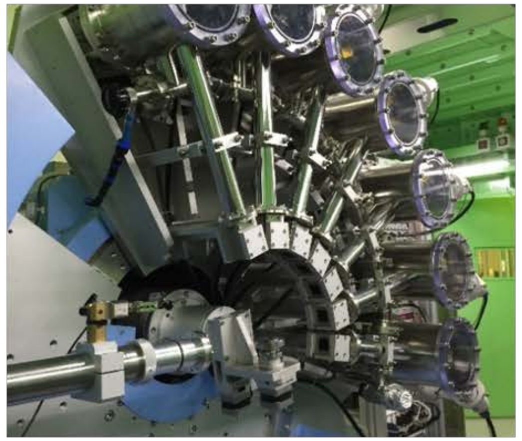
Introduction of HRPD-II beamline

❖ Needs for NEW PD beamline

- One of the techniques with the highest user demand for academic and industrial field
- Only one beamline (9B HRPD) is dedicated to PD experiments among the 36 beamlines of PLS-II.
- A new PD beamline with fast data acquisition capable of real-time experiments and various environmental experiments is required.

HRPD Gen. I
Multi-detector system with analyzer crystals
: Extremely high angular resolution but slow

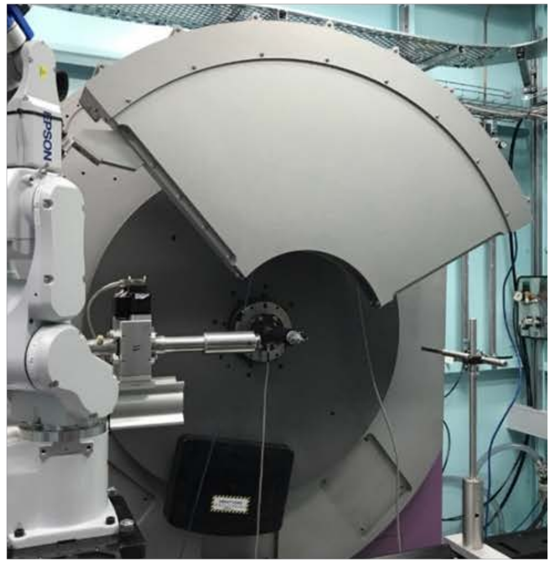
- Angular resolution < 0.015° (9B)
- Long measurement time
  ≈ 2h for Rietveld Refinement Analysis
- Bragg-Brentano geometry
- Large amount of samples required



HRPD Gen. II
1D Micro-strip detectors (MYTHEN)
: High angular resolution and fast

- Angular resolution ≈ 0.02~0.03°
- Fast data acquisition < 5 min.
  → High throughput by using robotic arms
- Transmission geometry
- Tiny samples (in a capillary tube)
  with high S/N ratio

Figure 2. HRPD detector systems



Beamline Renovation Project Overview

❖ 4B HRPD-II beamline

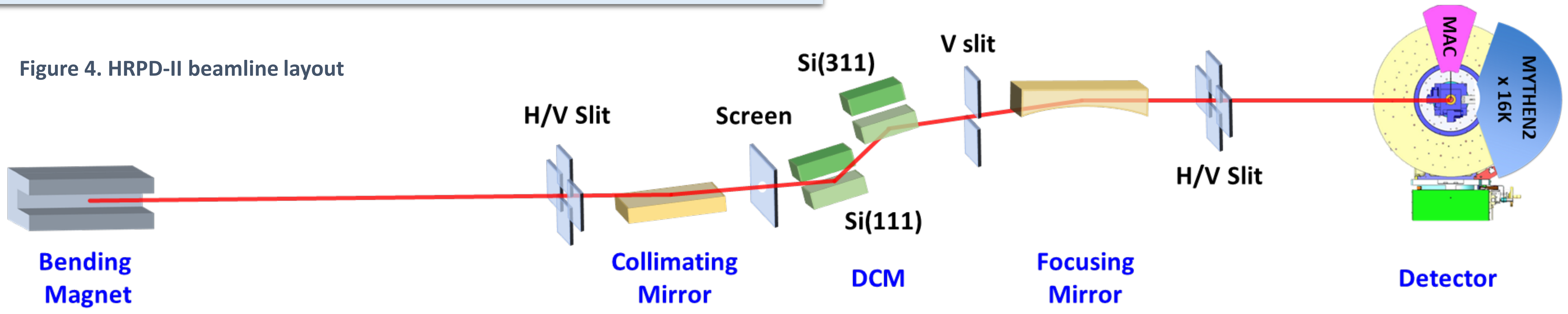
- Meet for energy/environmental materials research and industry samples
- Focusing optics and 1D microstrip detector system for fast data acquisition
- New HRPD-II program can distribute PD user beamtime and expand new research groups
- Total budget : 3 billion won ($ 2.2 million)

Year	Plan
1st year ('23)	Beamline CDR & TDR review, National budget review
2nd year ('24)	Completion of beamline drawing, Radiation safety assessment, FE/PTL rearrangement
3rd year ('25)	Optics (M1, M2, DCM) installation and alignment
4th year ('26)	End-station/Controls/DAQ completion and commissioning
2027~	Starting user service

Figure 3. HRPD-II beamline project plans

Concept of New HRPD-II Beamline

Figure 4. HRPD-II beamline layout



Source	Bending magnet (PLS-II BL4B port)
Available energy range	8-30 keV (1.55-0.4Å)
Photon flux	~10^{10} (minimum)
Optics	Si(111)/Si(311) DCM, Vertical collimating mirror (M1), Toroidal focusing mirror (M2)
Energy resolution (ΔE/E)	2 x 10^{-4}
Beam size (@sample position)	100 ㎛ (H) x 100 ㎛ (V)
Angular resolution (LaB_6 @15keV)	<0.015˚ (Analyzer Crystal), <0.03˚ (MYTHEN-II)
End station	High resolution position sensitive detector, MYTHEN2 (2θ ~80˚, 16K modules) + Ge(111) MAC (Multi-Analyzer Crystals) with 3-circle (ω, 2x2θ) diffractometer

Science and Application 1

❖ **Crystal structure analysis from high resolution powder X-ray diffraction patterns**

- Structure determination of new powder samples
- Structure refinement of polycrystalline materials
- Quantitative phase analysis of multiphase materials
- Tracing phase transition process from crystalline lattice changes

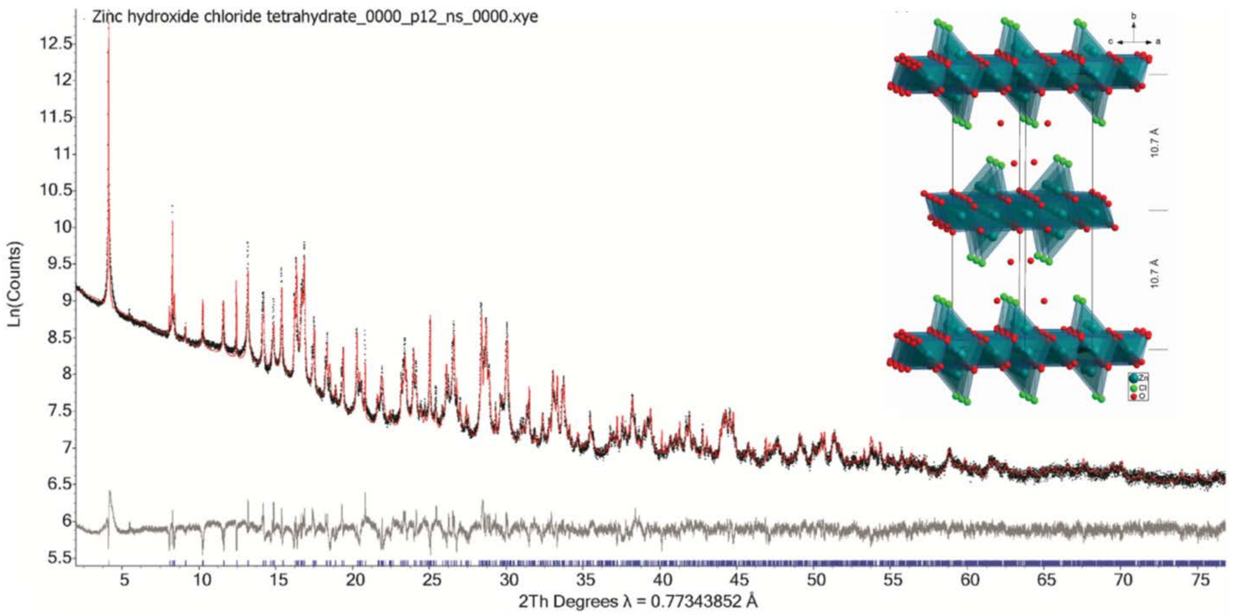


Figure 5. Structure determination of Zn hydroxide chloride samples from SPXRD (0.3mm capillary@16keV, MYTHEN2), J. Solid State Chem. 290, 121483 (2020).

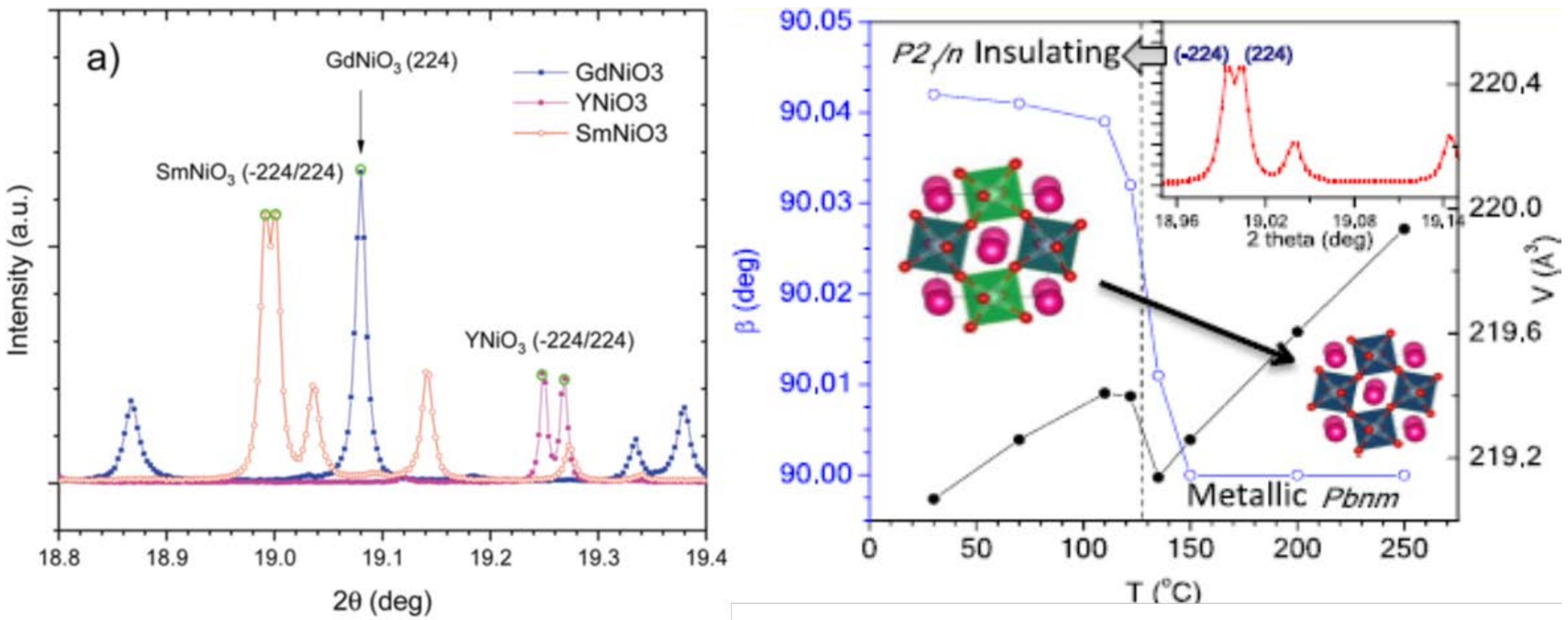


Figure 6. Structure changes during metal-insulator transition of $RNiO_3$ perovskites from SPXRD (0.7mm capillary @28keV), Dalton Trans. 50, 7085 (2021). Inorg. Chem. 58, 11828 (2019).

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Science and Application 2

❖ In situ (Operando) experiments

- Understanding phase transformation mechanism during real-time/environmental experiments

  : temperature, gas adsorption/desorption, photo/electrochemical reaction

- Time-resolved synchrotron powder X-ray diffraction studies (sub-minute scale)

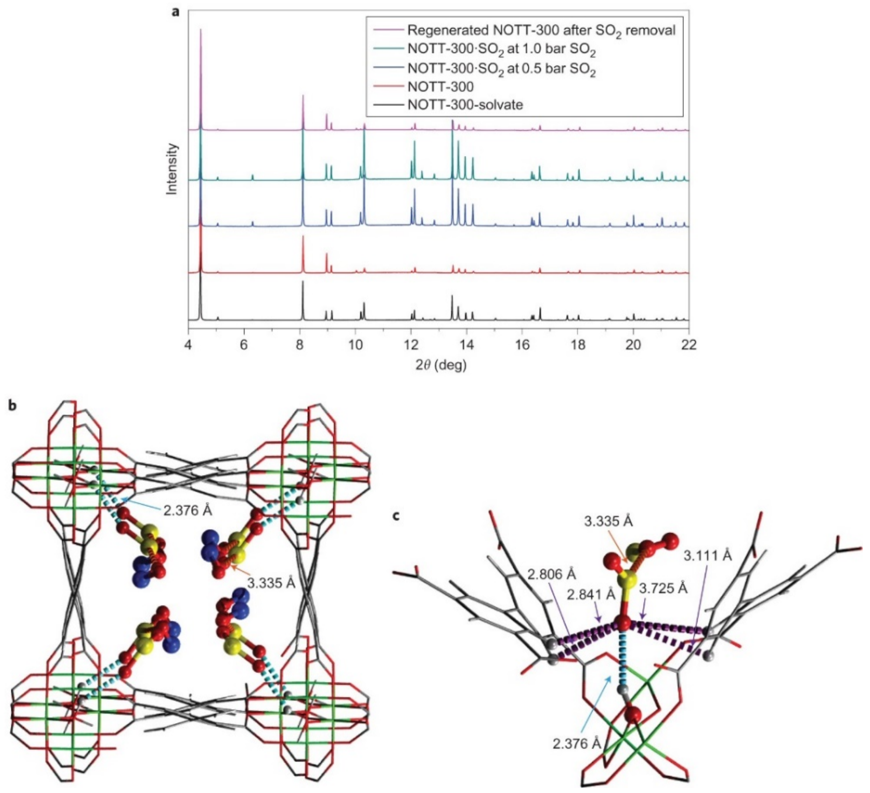


Figure 7. SPXRD patterns and crystal structure of MOF samples depending on SO_2 adsorption, Nature Chem. 4, 887 (2012).

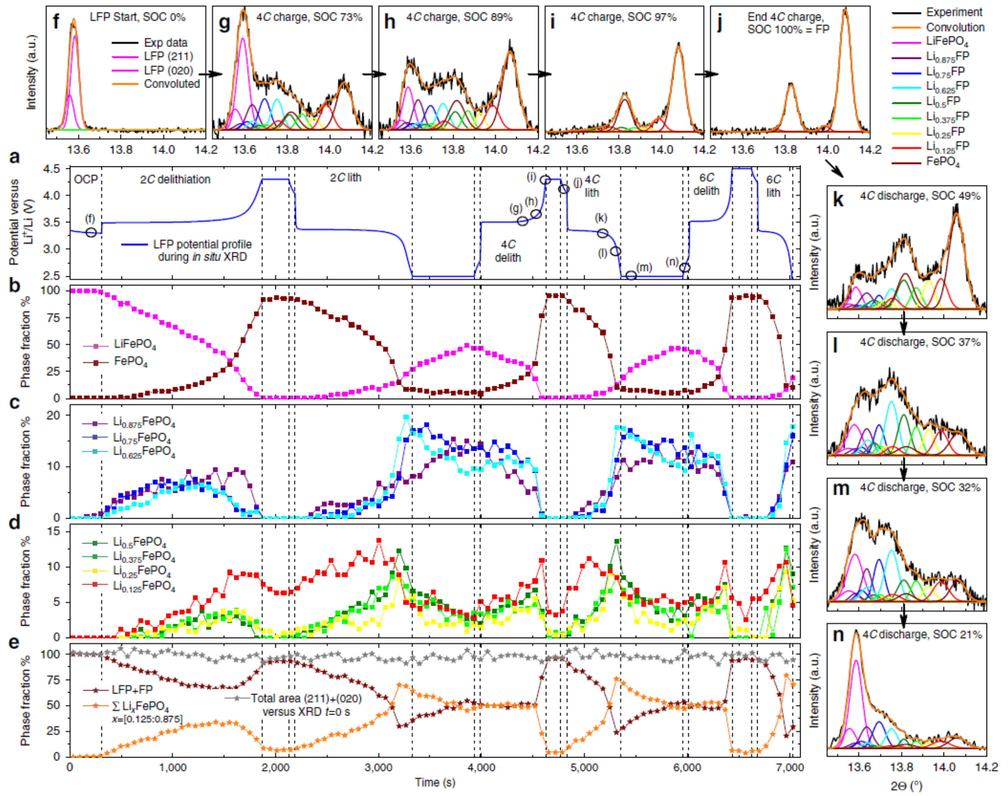


Figure 8. Deconvolution of 9 intermediate-phases of $LiFePO_4$ during dis/charging at a rate of 4C, Nature Commun. 6:8169 (2015).

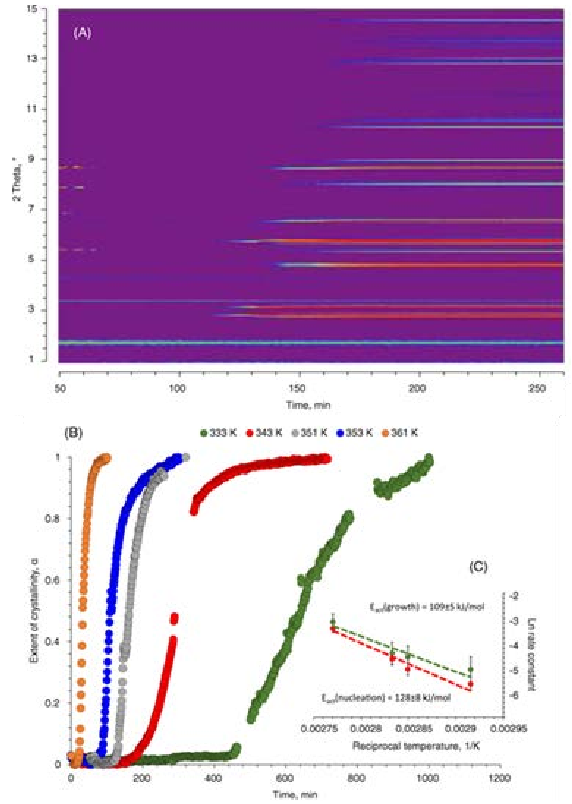


Figure 9. Time-resolved SPXRD of MOFs (MIL-53) crystallization, Nature Commun. 13:3762 (2022).

Science and Application 3

- Structure analysis of radiation-damaged/non-ambient samples
- A tiny amount of sample (mg scale) measurement
- Materials screening & designed based on high throughput SPXRD data
- SPXRD data for industrial mass production powders

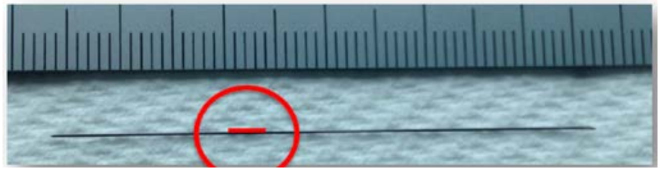


Figure 10. An example of powder samples in a capillary tube for transmission mode SPXRD

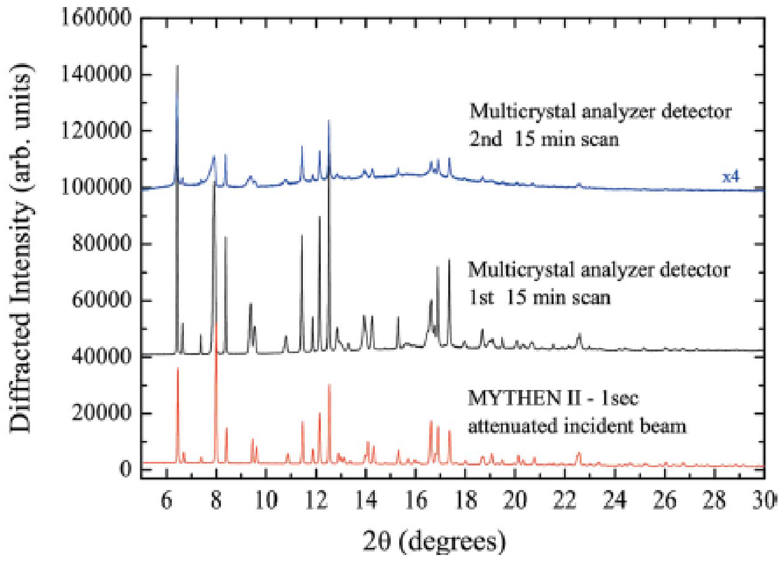


Figure 11. SPXRD patterns of radiation-damaged samples (Bupivacaine hydrochloride form D, 1mm capillary @12 keV), J. Synchrotron Rad. 16, 849, (2009).

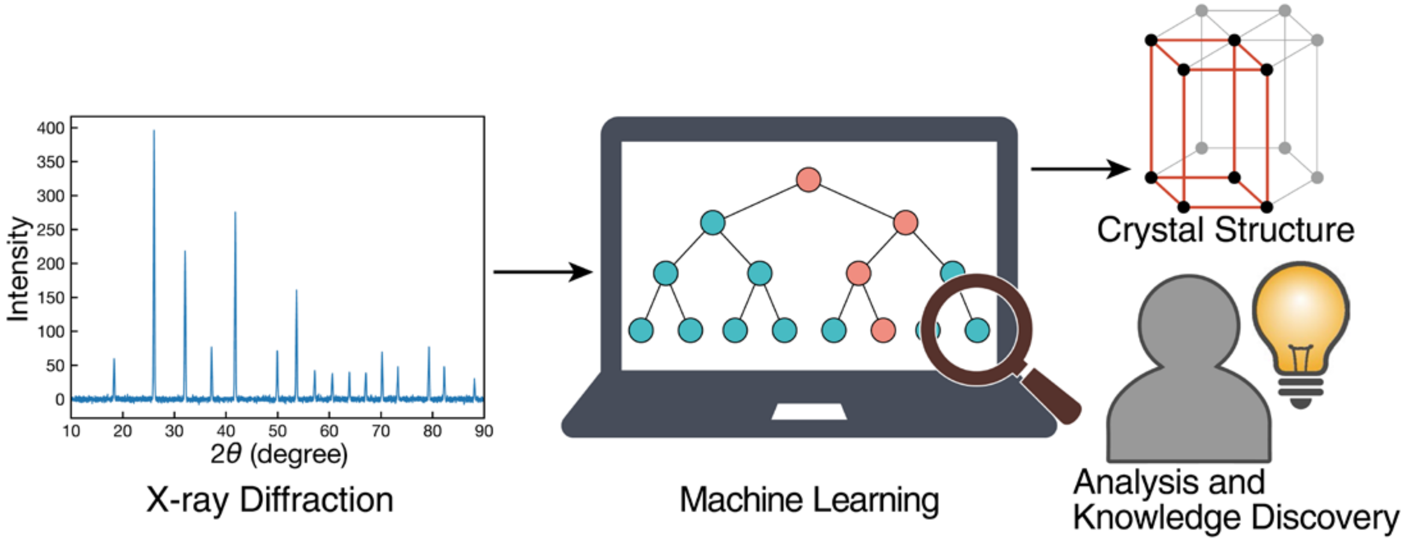


Figure 12. Machine-learning methodology for crystal system and space group classification based on PD patterns, Sci. Rep. 10:21790 (2020).

Optics design 1. Mirrors

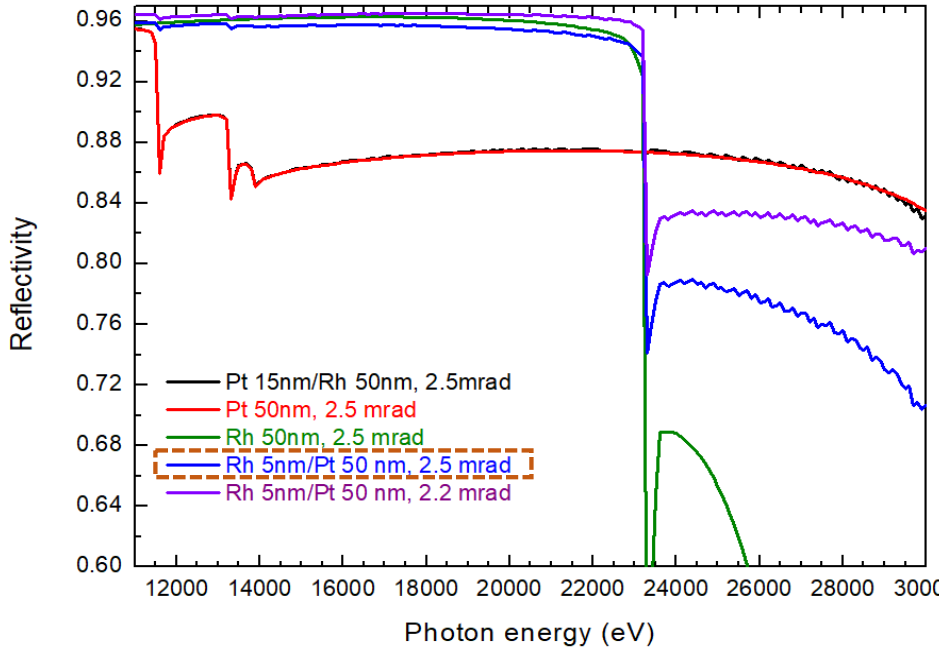
❖ Collimating mirror (M1)

- Cylindrical mirror with water cooling for vertical collimation
- Rh on Pt coating, Si substrate, Upward
- Grazing incidence angle 2.5 mrad

❖ Focusing mirror (M2)

- Toroidal mirror for vertical/horizontal focusing
- Rh on Pt coating, Si substrate, Downward
- Grazing incidence angle 2.5 mrad
- Vertical focal length 10 m
- Horizontal focusing 2.1:1

Figure 13. Reflectivity of coated mirror



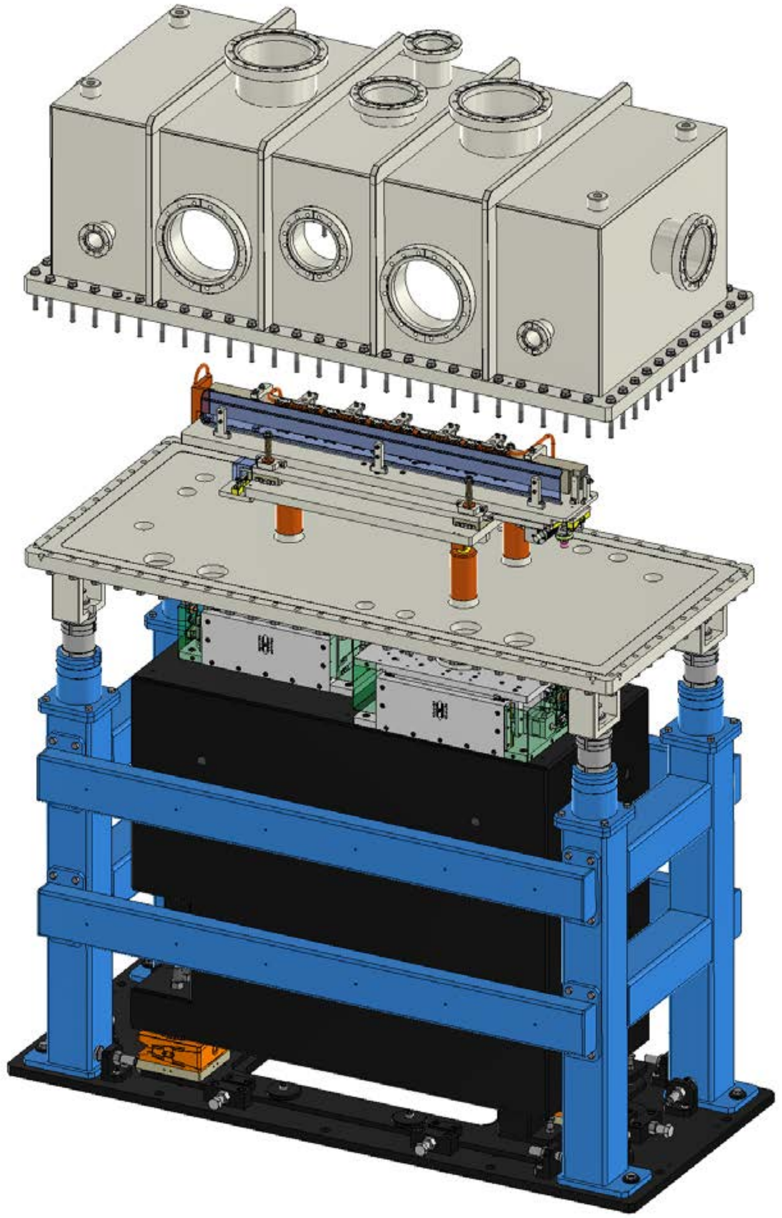
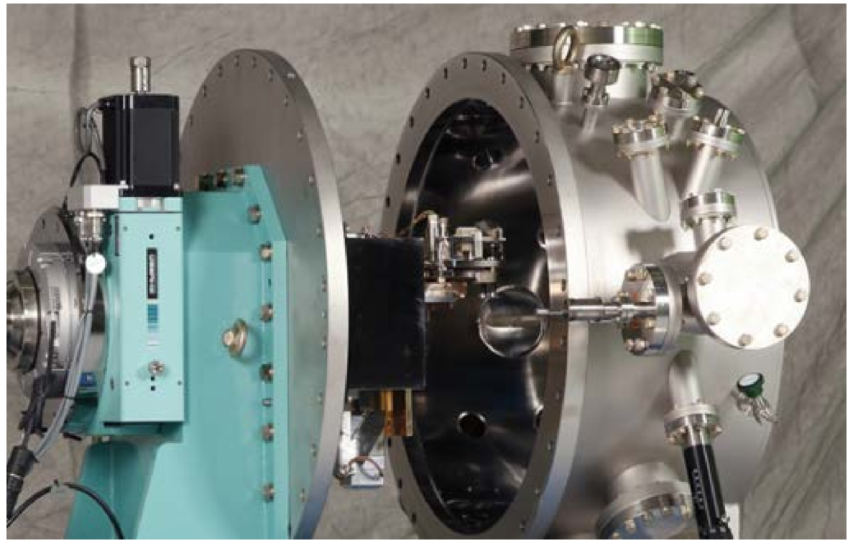
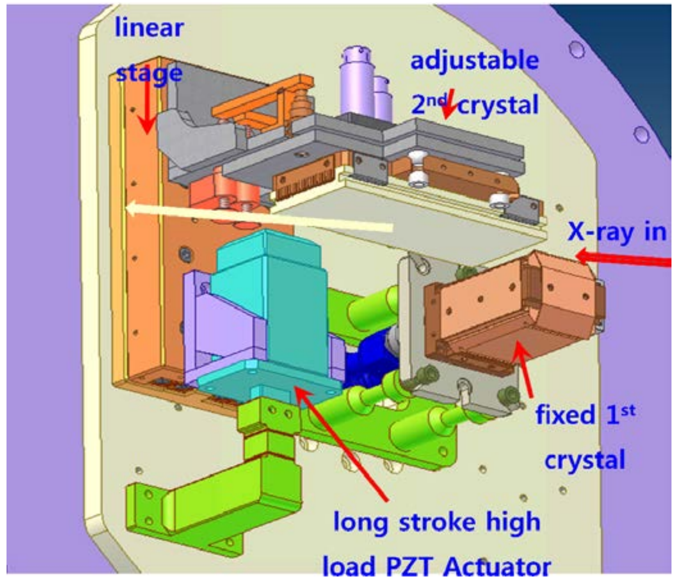


Figure 14. Mirror design for BL4B (in progress)

Optics design 2. DCM

Optical Parameters	
First crystal	Si(111) / Si(311) Footprint: 20 mm (H) x 35 mm (V), Water-cooling
Second crystal	Si(111) / Si(311)
Beam size	35 mm (H) [2 mrad in horizontal] x 2.5 mm (V) [0.2 mrad in vertical]
Beam offset	25 mm upward, fixed-exit
X-ray energy	8 - 25 keV, Si(111) Bragg angle 4-15° 20 - 30 keV, Si(311) Bragg angle 7-11°





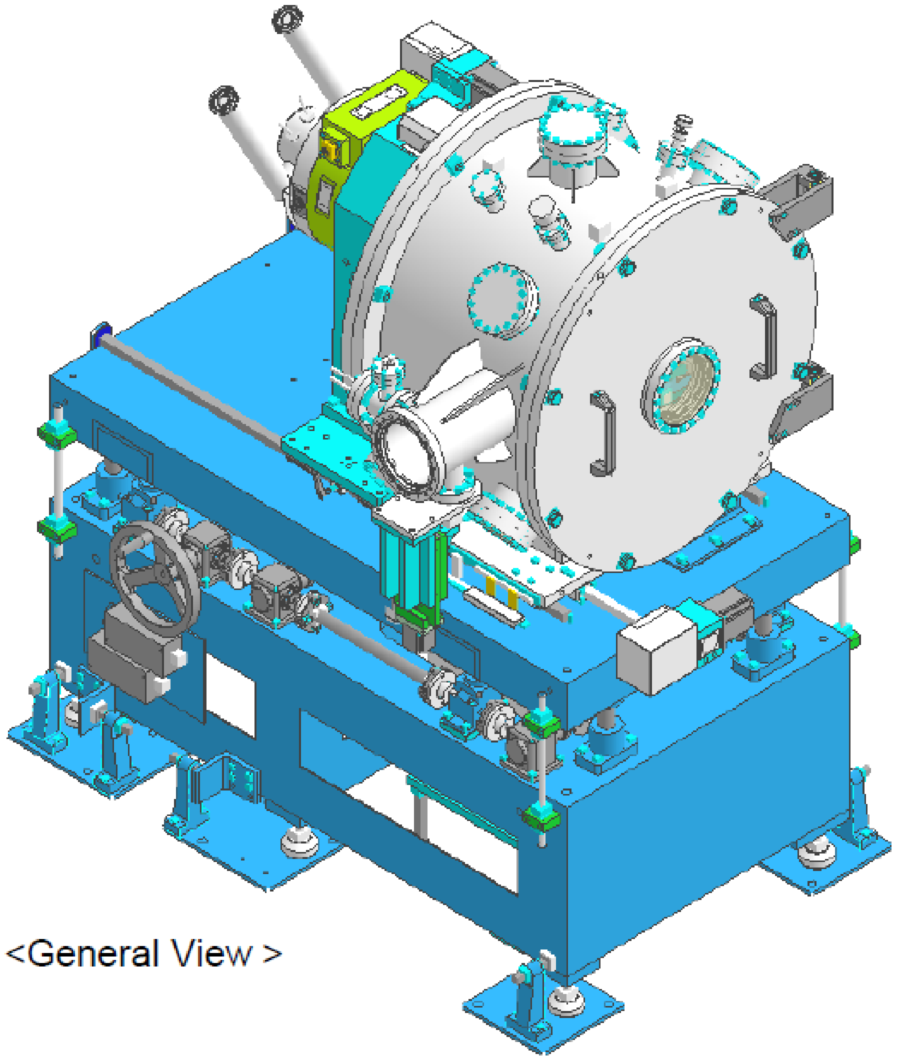


Figure 14. PAL DCM design for BL4B (in progress)

Optics design 3. Angular resolution

❖ Δ2θ (FWHM) with focusing optics in PD beamline

$$\Delta^2(2\theta) = (\Delta\tau_p^{\,2} + \Delta_m^{\,2}/2)(\tan\theta_a / \tan\theta_m - 2\tan\theta / \tan\theta_m)^2 + \Delta_a^{\,2} + \Delta\tau_f^{\,2}$$

$\Delta\tau_p$: beam divergence after reflection on the collimating mirror
$\Delta\tau_f$: beam divergence after reflection on the refocusing mirror
Δ_m, Δ_a : Darwin widths of monochromator and analyzer crystal
θ_m, θ_a : Bragg angles of monochromator and analyzer crystal

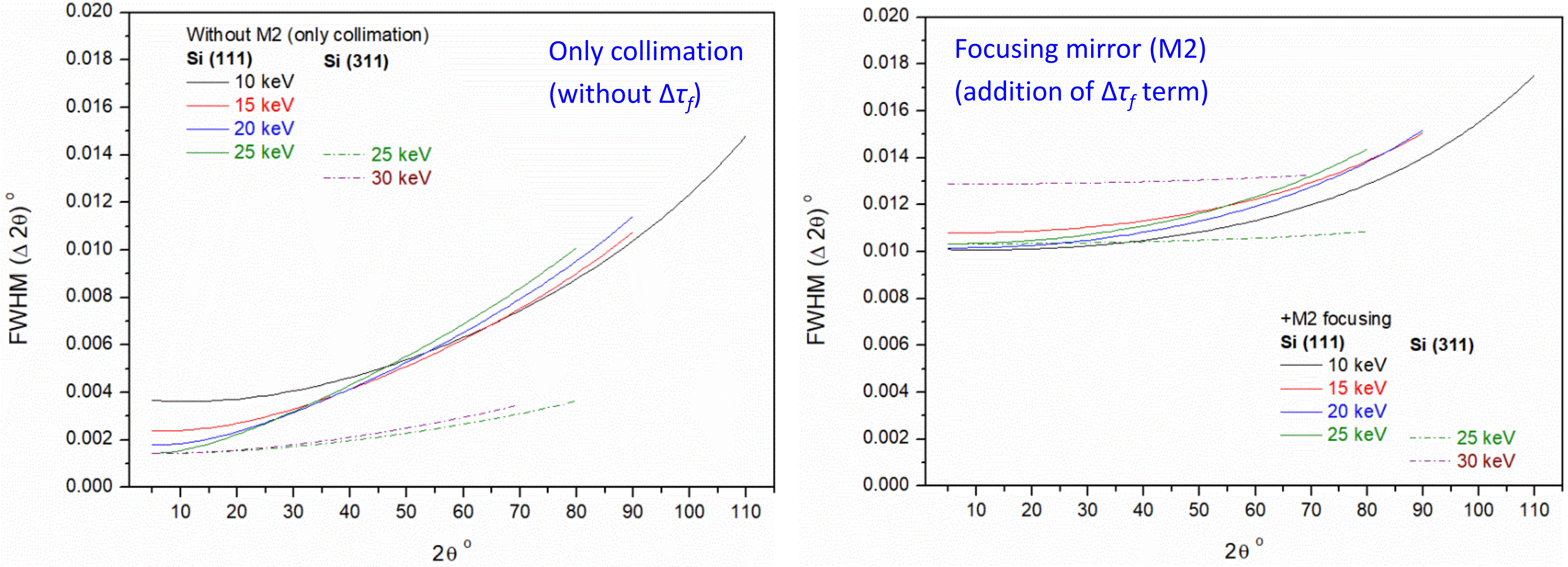
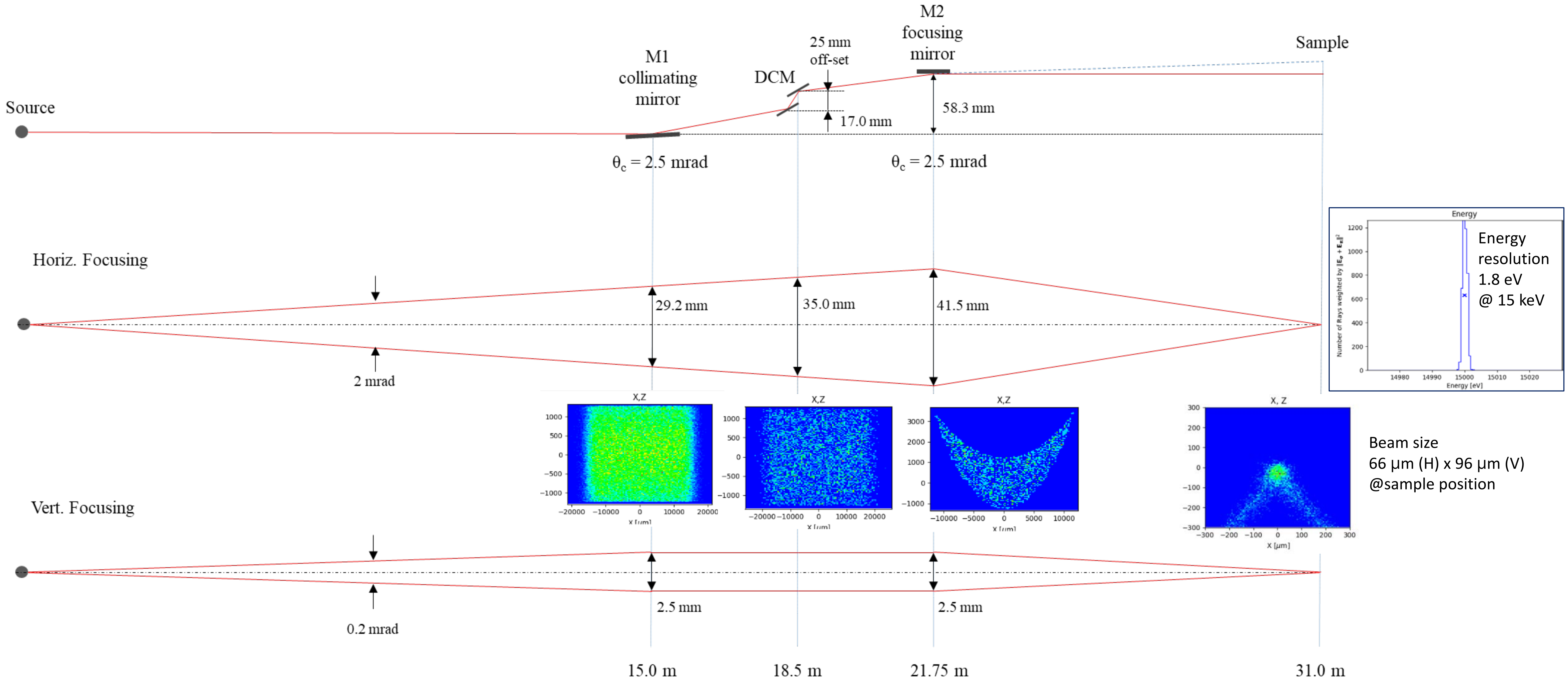


Figure 15. Computation of the instrumental resolution function (IRF) without and with focusing optics

Beamline ray tracing (@ 15keV)



Experimental station 1. MYTHEN2 module

❖ **MYTHEN2 (Microstrip sYstem for Time rEsolved experimNts) detector**

- Operating in single-photon-counting type
- Consisting of 1280 silicon microstrips with strip size of 50 μm

BL4B HRPD-II requirements	
Energy range	**8-30 keV**
Efficiency at 15 keV	> 70 %
Angular resolution (Δ2θ)	≤ 0.01°
Angular coverage (2θ)	**80°**
Number of MYTHEN2 modules	**16**
Channels per module	1280
Sample to Detector distance	**760 mm** (~0.004° intrinsic resolution)
Frame rate (MYTHEN2 single)	100-900 Hz
Multi-analyzer-crystal (MAC)	3 Ge(111) with 3 scintillation detectors + slit- and anti-scatter system

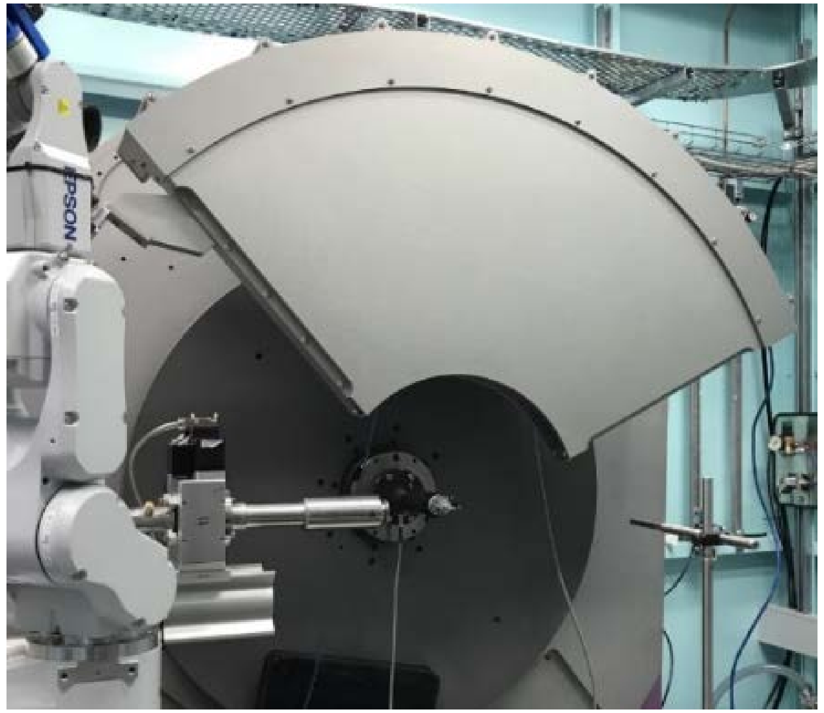


Figure 16. MYTHEN2 in PD beamline at Australian Synchrotron

Experimental station 2. 3-circle diffractometer

❖ **Specially designed 3-circle diffractometer for 4B HRPD-II beamline**

- Double axis goniometer system (inner rotation platform & middle rotation platform, ω x 2θ) + One-circle goniometer (2θ2)
- Detector housing for radial arrangement of MYTHEN2x16K, offset between modules for gapless data including mounting, support on detector circle and counterweight
- MAC (Multi-Analyzer-Crystal) System
  : required to design for HRPD-II beamline, compact system with Ge(111) analyzer crystals with detector array, simultaneous AC/detector rotation, slit- and antiscatter system, sealed housing for MAC system

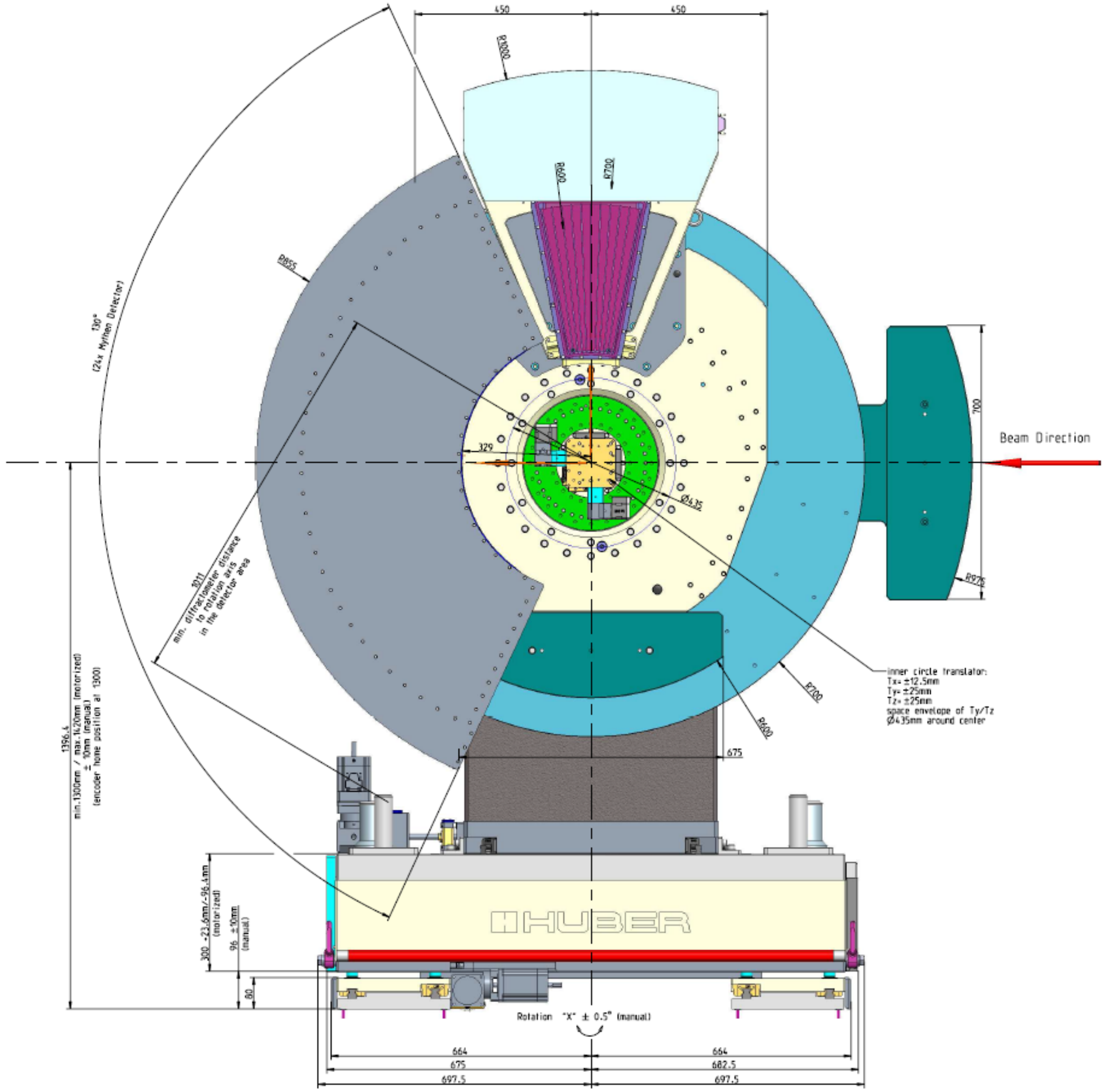
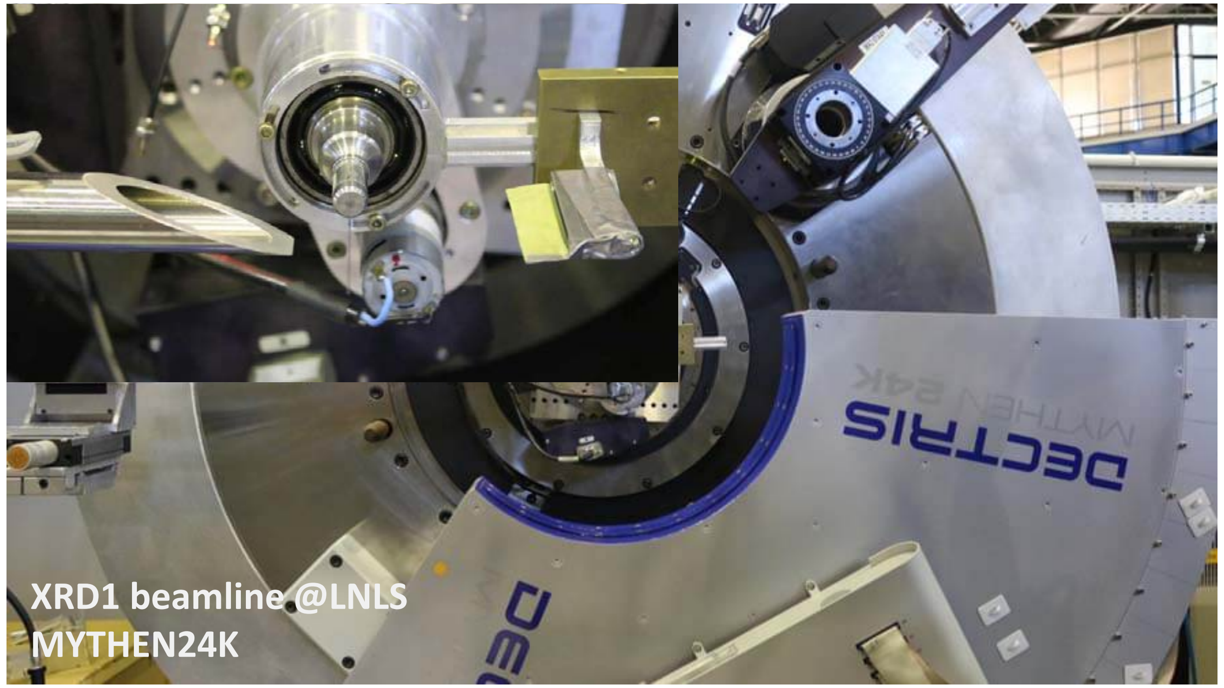


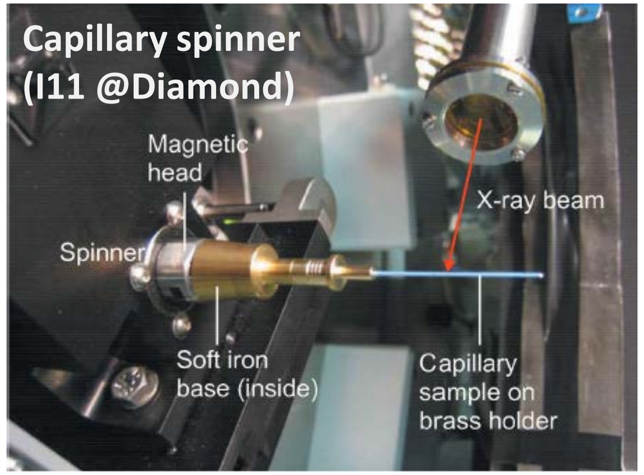
Figure 17. An example of 3-circle diffractometer with MYTHEN2 and MAC detector systems design for BL4B (in progress)

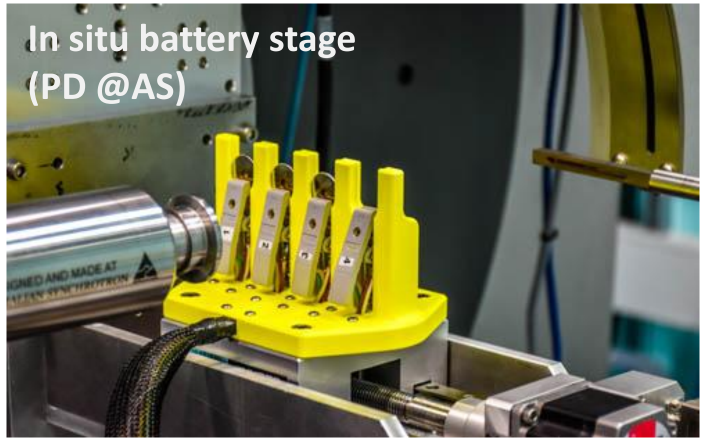
Experimental station 3. Sample environments

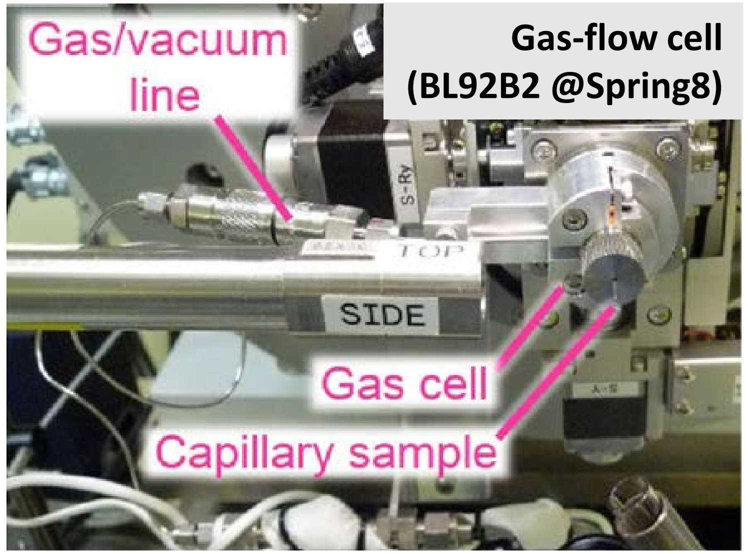
❖ Transmission geometry for PD experiments

- Sample spinners for capillary & flat plates
- Gas-flow cell
- Hot air blower (for capillary)
- Cryostream
- Heating furnace
- In situ battery stage
- Auto sample changer with a robotic arm (phase2)
  → automation set-up









Cryostream

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