# BL04B2 High-energy X-ray Diffraction

BL04B2 was designed for diffraction and scattering experiments at high-energy more than 37 keV. Four kinds of experimental stations are equipped in tandem: Two-axis diffractometer for disordered materials, Weissenberg camera, and Imaging Plate system for the X-ray diffraction study under high pressure conditions and small angle X-ray scattering of supercritical fluids.

**Area of research**

Structural analysis of glass, liquid, and amorphous materials
X-ray diffraction under ultra high-pressure
Precise single crystal structure analysis

**Keywords**

*Scientific field*

High-energy X-ray diffraction, Disordered materials, Single crystal, High-pressure, Supercritical fluids, Small angle scattering

*Equipment*

Two-axis diffractometer, Weissnberg camera, DAC (diamond anvil cell), IP (Imaging plate)

## Source and optics

Light source is a bending magnet and horizontal divergence of the beam is limited to 0.73 mrad by a fixed mask. To carry out the experiments with high-energy, modest-resolution, and focused X-rays, we adopt a single-bounce, bent-crystal monochromator which deflects the beam horizontally. The available Bragg angle is fixed to 3 deg, so that we can change energy discretely by selecting reflection net-planes. The Si 111 (37.8, 113.3 keV) and Si 220 (61.7 keV) crystals are available. The adjustable bending radius (320 m to 430 m) of Si crystals installed at 46 m from the light source, enables us to accord the focus position at 10 m to 15 m in the experimental hutch from the monochromator.

### X-rays at sample

Energy

Si 111 : 37.8 keV , Si 220 : 61.7 keV , Si 511 : 113.3 keV (*planned*)

Energy resolution

$\Delta E/E = 10^{-3}$ @15 m from the monochromator (at the incident beam size : 0.2 (H) × 4 (W) $mm^2$)

Photon flux

· 37.8 keV Flat : $2.2 \times 10^{10}$ (photons/sec/1 × 1 $mm^2$ @100 mA) , Bent (at focus point) : $7.1 \times 10^{11}$

· 61.7 keV Flat : $3.4 \times 10^{9}$ , Bent (at focus point) : $9.2 \times 10^{10}$

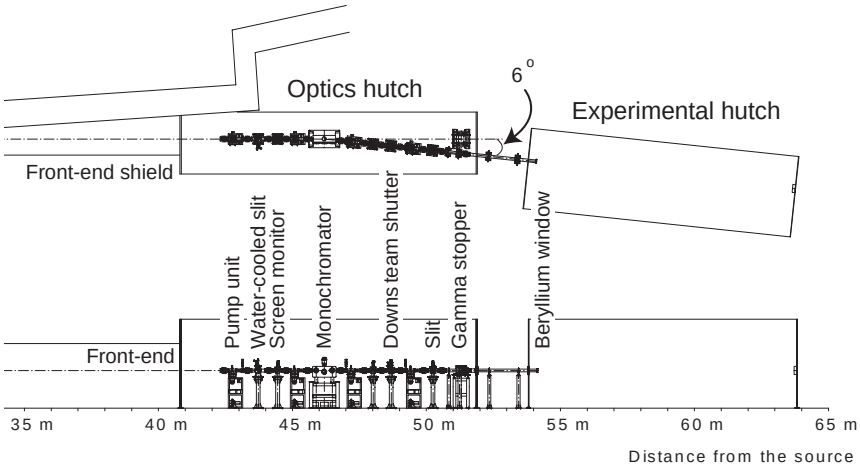
Beam size at focus position

· 37.8 keV : 0.220 mm

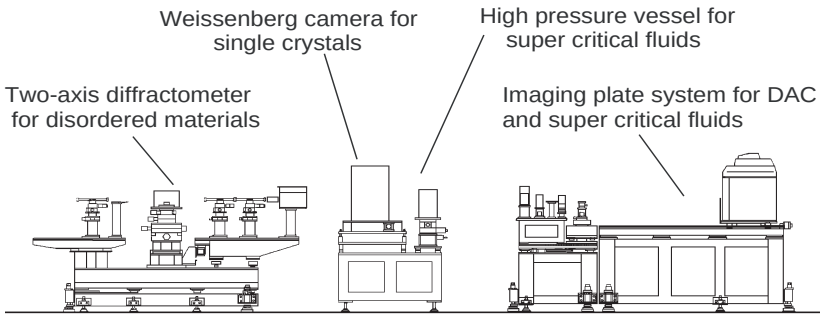
· 61.7 keV: 0.375 mm (at the incident beam size : 0.2 (H) × 4 (W) $mm^2$)

### Reference

[1] M. Isshiki, Y. Ohishi, S. Goto, K. Takeshita and T. Ishikawa Nuclear Inst. and Methods in Physics Research, A:(2001) 467-8, 663-666




Schematic view of beamline




End station in the experimental hutch

## Experimental stations

### Two-axis diffrractometer for high-energy X-ray diffraction of disordered materials

The two-axis diffractometer for glass, liquid, and amorphous materials (Fig. 1) is installed to measure total structure factor S(*Q*) up to high *Q*, providing high real space resolution in Fourier transformed function. The diffractometer was designed with a horizontal scattering plane for an easier use of heavy equipment, although horizontal scattering plane has the disadvantage of the polarization factor for large scattering angle. In high-energy X-ray diffraction ($E$ > 30 keV), however, the experiment can be performed in the rather small angle region where the polarization factor is not so significant.

Table 1. The specification of two-axis diffractometer installed at BL04B2

| | | | |
|---|---|---|---|
| 2θ arm | $2\theta_{main}$ | -10 ~ 150° | Δθ = 0.001 |
| | $2\theta_{sub}$ | -120 ~ 10° | Δθ = 0.001 |
| Sample stage | θ | -180 ~ 180° | Δθ = 0.001 |
| | X | ± 10 mm | |
| | Y | ± 10 mm | |
| | Z | ± 10 mm | |
| | $R_x$ | ± 3° | |
| | $R_y$ | ± 3° | |
| Collimator | $X_c$ | ± 10 mm | |
| | $Z_c$ | ± 10 mm | |
| | $R_{cy}$ | ± 10° | |
| | $R_{cz}$ | ± 180° | |
| | $X_t$ | ± 15 mm | |

Table 2. Available photon energies and accessible *Q* range

| Photon energy (keV) | Accessible *Q* range (Å$^{-1}$) |
|---|---|
| 37.8 keV (Si 111) | 0.1 ~ 22 |
| 61.7 keV (Si 220) | 0.16 ~ 36 |
| 113.3 keV (Si 333*) | 0.3 ~ 40 |

*obtained with 3rd harmonic of Si 111 reflection

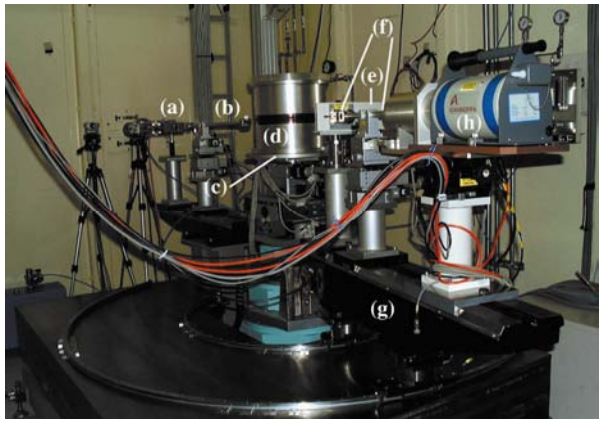



Fig.1. A two-axis diffractometer installed at BL04B2 [1]
(a) Ionization chamber, (b) Incident slit. (c) θ stage, (d) Vacuum chamber, (e) Beam stop, (f) Receiving slits, (g) 2θ arm, (h) Ge detecter

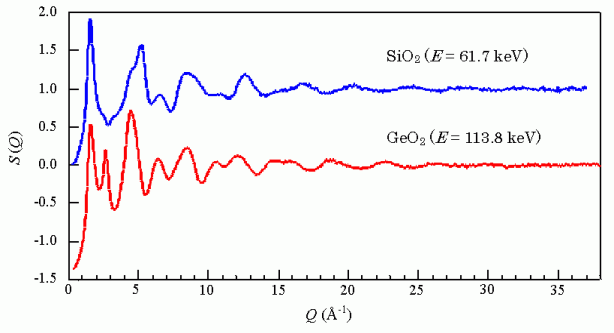



Fig.2. Total structure factors, *S*(*Q*), of typical oxide glasses [1, 2]

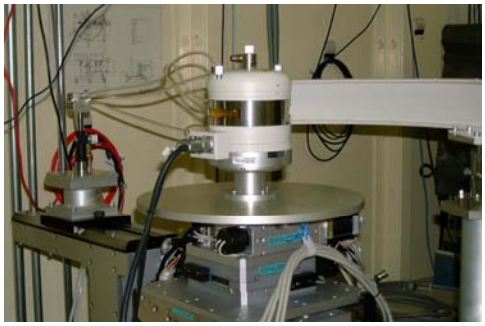


Fig.3. High-temperature furnace (~1200˚C) for a two-axis diffractometer

References

[1] S. Kohara, K. Suzuya, Y. Kashihara, N. Matsumoto, N. Umesaki, and I. Sakai, Nucl. Instr. and Meth. A 467-468, 1030 (2001).

[2] S. Kohara and K. Suzuya: Nucl. Instr. and Meth. B, 199 (2003) 23.


### Weissenberg camera

The Weissenberg camera installed at the BL04B2 beamline provides a facility for single crystal structure analyses of small molecules. It utilizes the highly focused high-energy (> 37.8 keV) X-rays supplied by the beamline. It is equipped with a cylindrical image-plate and an integrated image-plate reader, allowing fully automatic data collection. A low temperature device (cold $N_2$ stream, dry $N_2$ extracted from air) is also available. Obtained diffraction images are automatically processed using the program DENZO. As the image-plate covers the 2θ range from -60 to 140 degrees, diffraction data up to 2.86 in sinθ/λ can be collected, allowing high-angle data refinement for precise structure determinations. The specifications of the camera are summarized in Table 1. For most elements, the absorption coefficients for the 37.78 keV X-rays are approximately the eighth of those for the MoKα radiation. Therefore, the advantage of this camera can be fully exploited when it is used for the structure determinations of crystals that contain heavy elements. The focusing mechanism of the monochromator of this beamline also facilitates the measurements of smaller sized crystals.

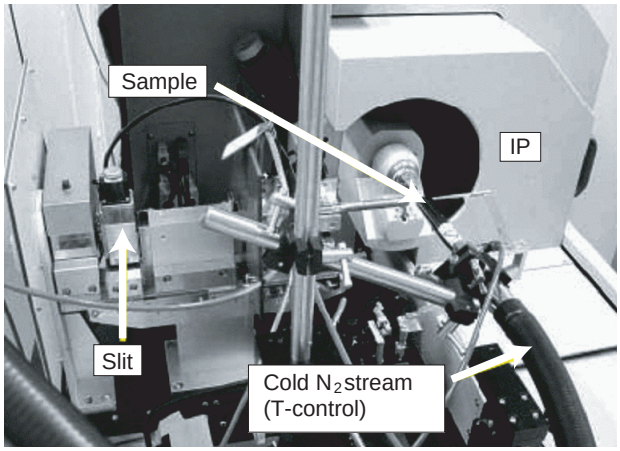



Fig.1. Weissenberg camera installed at BL04B2

Table 1. The specifications of Weissenberg camera

| | |
|---|---|
| Temperature | 90 K to room temperature |
| Crystal size | 10 μm × 10 μm × 40 μm to 500 μm × 500 μm × 500 μm (depends on the content, crystallinity and cell parameters) |
| $(\sin\theta/\lambda)_{max}$ | 2.86 |
| Camera radius | 120 mm |
| Spatial resolution of the detector | 100 μm |

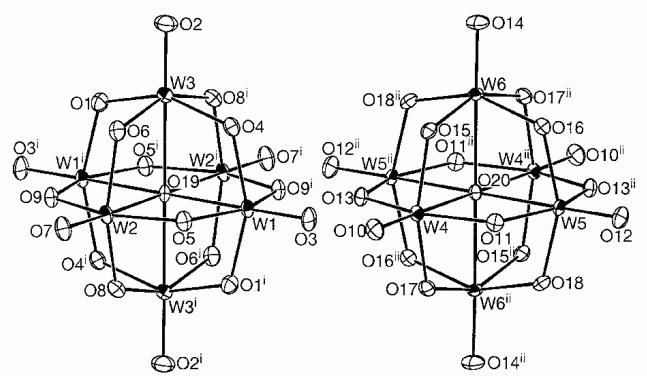



Fig.2. ORTEP drawings of the two independent $[W_6O_{19}]^{2-}$ anions
Displacement ellipsoids are scaled to enclose 50% probability levels. [1]


Reference

[1] T. Ozeki, K. Kusaka, N. Honma, Y. Nakamura, S. Nakamura, S. Oike, N. Yasuda, H. Imura, H. Uekusa, M. Isshiki, C. Katayama and Y. Ohashi, Chem. Lett. (2001) 804.


### Small angle X-ray scattering for supercritical fluids

Small angle X-ray scattering experiments are carried out using 37.8 keV X-rays monochromatized by the flat surface of Si (111) crystal as an incident beam. The scattered X-rays are detected with an imaging plate of 300 mm × 300 mm located about 3 m apart from the sample position. A vacuum path is mounted between the sample and the Imaging Plate. Using a lead beam stop, 5 mm in square, the observable wavenumber k (k = 4απ;sinθ/λ ; λ : wavelength) is estimated to be 0.02 $Å^{-1}$ to 0.9 $Å^{-1}$.

Table 1. The specifications of the high-pressure vessel

| | |
|---|---|
| Temperature | up to 1700°C |
| Pressure | up to 200 MPa (more precisely, 196 MPa) |
| Wavenumber | 0.02 ~ 0.9 $Å^{-1}$ (without the high-pressure vessel) |
| | 0.06 ~ 0.4 $Å^{-1}$ (with the high-pressure vessel) |

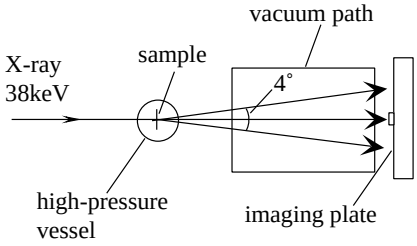



Fig.1. The schematic diagram of the spectrometer

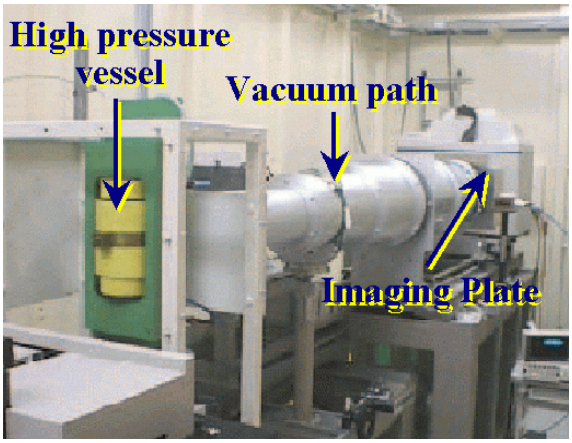



Fig.2. The photograph of the spectrometer

High pressure gas apparatus that permit experiments up to 1700°C and 200 MPa are installed in the experimental hutch. A synthetic diamond 3 mm in diameter of the highest quality is used as a window for the incident beam, and a Be window 10 mm in diameter is used for scattered X-rays to cover larger k. A cell made of sapphire is used for fluid sample such as selenium and mercury because it is transparent to X-rays and resistant to chemical corrosion by the sample at high temperature.

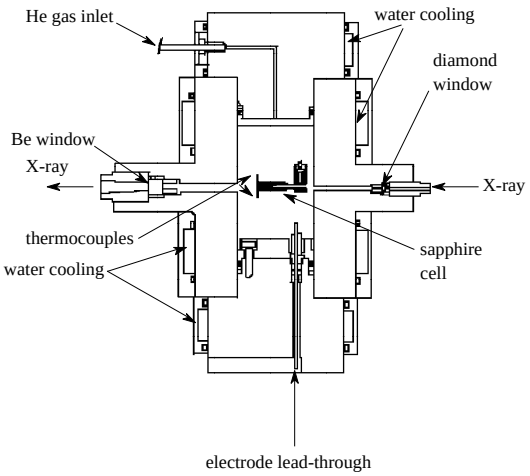



Fig. 3. A side view of the high-pressure vessel for the small angle X-ray scattering measurements

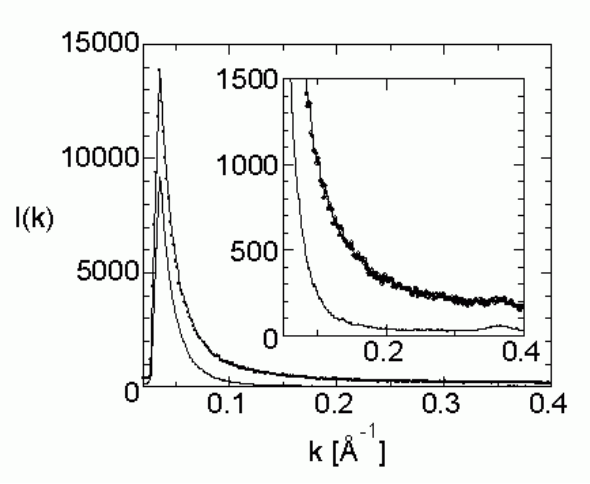



Fig. 4. Small angle X-ray scattering intensity spectra of an empty cell (solid line) at 25°C, and the cell and fluid Se at 1680°C and 457 bar near the critical density (dots and line) after absorption correction [1]

The inset shows the smaller-intensity region on an enlarged scale. The large background intensity from 0.02 Å$^{-1}$ to 0.08 Å$^{-1}$ arises from the Be window of the high-pressure vessel. After deducing the small-angle scattering spectrum by subtracting the intensity of the empty cell, a reliable k-minimum is found: it is 0.06 Å$^{-1}$. A small peak which comes from a Kapton window of the vacuum path is observed at around 0.37 Å$^{-1}$. The maximum k is limited to be 0.4 Å$^{-1}$ due to a view from the sample position through the Be window.

## Reference

[1] K. Tamura and A. Inui, J. Phys. :Condensed Matter, 13 (2001) R337.

## Imaging Plate system for the X-ray diffraction study under high pressure conditions

The Imaging Plate system was designed to study the structure and physical properties of materials under high pressure conditions.

Fig. 1 shows a schematic view of the Imaging Plate system installed at BL04B2. This system consists of 4D slit, X-ray shutter, ionization chamber, collimator, goniometer and imaging plate detector (detector area is 300 mm × 300 mm, Rigaku R-AXIS IV++). A camera length (distance from the sample position to the detector) can be changed from 270 mm to 1470 mm. A diamond anvil cell (DAC) is mounted on this diffractometer. A sample is enclosed in a DAC, which can generate high pressure, and the diffraction from the sample is detected by the imaging plate detector.

The imaging plate detector is also used as a detector for small angle X-ray scattering for supercritical fluids in the high pressure vessel.

Table 1. The specifications of DAC

| | |
|---|---|
| Temperature | Room temperature |
| | High temperature (up to 1000 K using the electric heating) |
| Pressure | up to 200 GPa |
| Beam size | more than 0.04 mm × 0.04 mm |

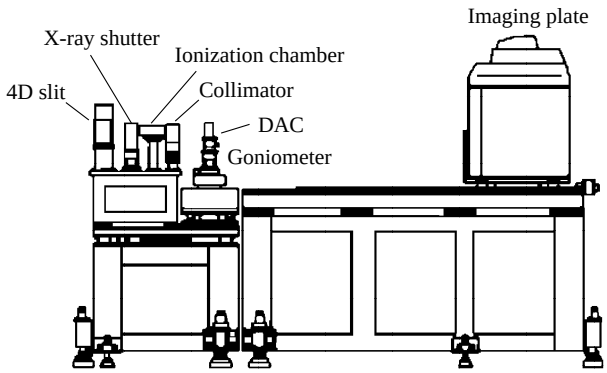



Fig.1. A schematic view of the imaging plate system installed at BL04B2

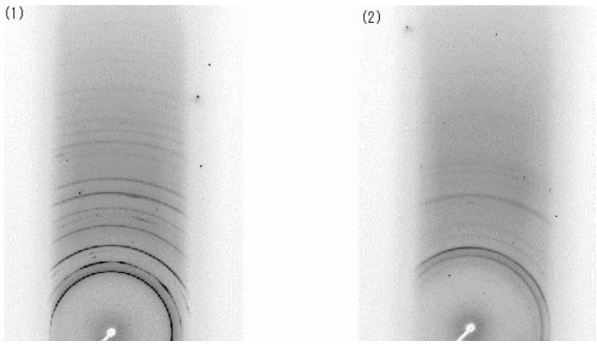



Fig.2. X-ray diffraction patterns of CeSb at 4.2 GPa (1) and at 24 GPa (2)

(1) $a$ = 6.26 Å, Fm3m, NaCl-type structure

(2) $a$=3.73 Å, $c$=3.11 Å, P4/mmm, Tetragonal-type structure

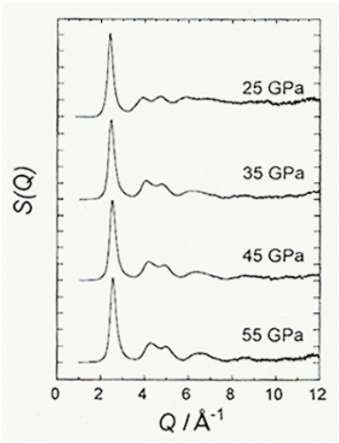



Fig.3. Faber-Ziman total structure factors, $S(Q)$, for $SnI_4$ at 35, 45 and 55 GPa on compression and at 25 GPa on decompression

The high energy x-ray of 61.7 keV makes possible to cover a wide range of the wave number, $Q$ (= $4\pi\sin\theta/\lambda$), up to ~ 16 $Å^{-1}$ even on high pressure experiments using diamond anvil cell (DAC) which restricts the maximum value of Bragg angle.

## References


[1] Y. Ohishi, M. Isshiki, N. Ishimatsu and N. Hamaya: SPring-8 user report #1999B0148-ND-np.

[2] A. Ohmura, N. Hamaya, K. Sato, C. Ogawa, M. Isshiki and Y. Ohishi: J. Phys.: Condens. Matter, 14 (2002) 10553.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