BL13XU
X-ray Diffraction and Scattering I

1. Introduction

BL13XU is a beamline designed for the structural analyses of various materials in atomic scale using X-ray diffraction techniques. Originally, this beamline targeted mainly the study of surface and interface structure [1]. Recently, the demands for advanced techniques such as *in situ*, operando, and automatic measurements have been increasing by many users in various fields. To meet these requests, we integrated and rearranged the various diffraction systems in this beamline in FY2022. At the same time, we upgraded the optical components such as monochromator and mirrors for use with high-energy X-rays up to 72 keV.

After the upgrade, BL13XU has four experimental hutches (EHs): the multi-axis diffractometer at EH1, diffraction measurement multipurpose frame at EH2, high-resolution powder diffractometer at EH3, and nanodiffraction system at EH4.

Below, we report the upgrades in FY2023 on the diffraction measurement multi-purpose frame (EH2) and high-resolution powder diffractometer (EH3).

2. Diffraction measurement multipurpose frame

In recent applications of X-ray diffraction, the demand regarding *in situ*/operando measurements with the control of the sample environments has been increasing, such as those of heating, cooling, and voltage applications. These improvements make sample environment devices large and heavy, thus necessitating more space around the sample than in the traditional multi-axis diffractometer. To meet these requirements, we developed a new diffraction system named the diffraction measurement multipurpose frame in FY2022.

The multipurpose frame is composed of a sample stage and a robot arm (Fig. 1). The sample stage is a combination of XZ stages, a rotational stage, and a hexapod. The maximum load of the sample stage is 250 kg. The robot arm for an area detector is combined with the sample stage. The robot arm has a maximum load of 25 kg and a reach of 2 m. The camera distance from the sample to the detector can be controlled from 150 to 1000 mm. When the camera distance is aligned at 1000 mm, the robot arm can cover the scattering angle by ±60° and 60° in the horizontal and vertical directions, respectively. PILATUS X 300k is mounted at the tip of the robot arm.

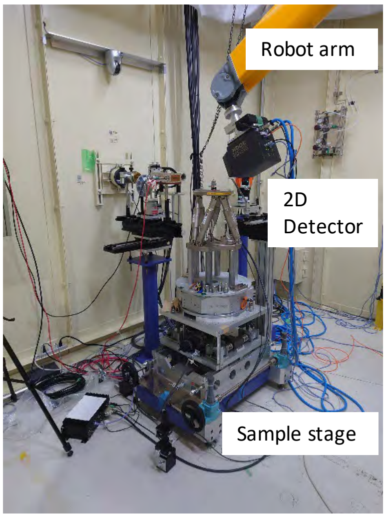

Fig. 1. Diffraction measurement multipurpose frame.

The sample stage and other optical components were relocated from other beamlines at the beginning. To achieve effective user experiments in this hutch, however, we had to customize and add several parts to the components.

First, we modified the base of the sample stage. With the original composition, the maximum sample height from the top plate of the hexapod was less than 180 mm. This small height restricted the dimensions of the sample environment devices. We developed a lower base and expanded the sample height to 380 mm.

Second, we developed alignment tools for the sample stage and the area detector on the robot arm. For precise diffraction measurement, the positions of the sample stage and the detector must be aligned exactly against the X-ray beam. We developed an alignment method using a laser tracker to define a laboratory coordinate frame on which the sample position is set as the origin and the X-ray axis is one of the coordinate axes.

The tools for the alignment of the sample stage are shown in Fig. 2. A pair of pillars is set at the front and rear of the sample position. On the top of the pillars, knife edges are combined in horizontal and vertical directions. The corners of these edges must be adjusted to identify the X-ray beam position. After the alignment of the sample stage, the knife edges are replaced with the reflectors for the laser tracker to read the beam position.

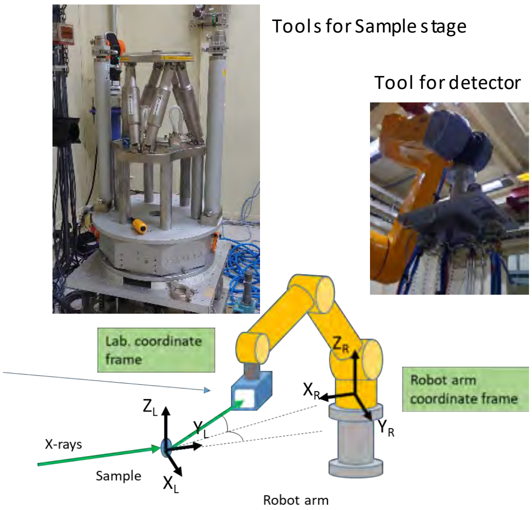

Fig. 2. Alignment tools for sample stage and detector. A schematic of the coordinate system is also shown below.

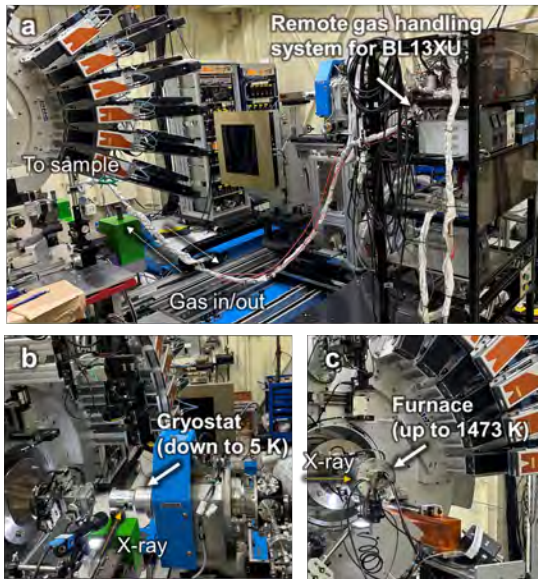

Fig. 3. Photographs of new sample environment system: (a) gas handling system, (b) cryostat, and (c) furnace at EH3.

The tools for the alignment of the detector were designed in the same manner. The tools have the same weight and center of gravity as PILATUS X 300k. The knife edge position is identified as the center of the receiving surface of the area detector. We can sample the positions of the detector surface center in laboratory coordinates and robot arm coordinates and calculate translation using the tool. After the procedure, the detector position is precisely controlled in the laboratory coordinates.

With the use of these tools, the appropriate alignment and the definition of the laboratory coordinate system become possible, which enable highly precise diffraction measurements. The tools also enable quick setup in user experiments.

3. High-resolution powder diffractometer

A high-resolution powder diffractometer [2] was introduced at EH3 in FY2022, allowing for higher throughput measurements and *in situ* and operando experiments and ensuring a large sample space of 600 mm × 600 mm and a weight capacity of about 500 kg. The 2θ axis is equipped with six sets of two-dimensional CdTe (LAMBDA 750k) detectors with high efficiency for high-energy X-rays, enabling very quick measurement of high-energy X-rays exceeding 60 keV. It also extends data acquisition beyond a Q-range of 20 Å^{-1} for pair distribution function (PDF) analysis. Currently, it is available for X-ray energies from 16 to 72 keV and has been used under various sample conditions. In the sample environment, nitrogen gas low-temperature/high-temperature blower devices are usually installed, allowing for temperatures ranging from 90 to 1100 K. By combining this with an automatic sample exchanger, automatic measurements of up to 100 samples, including variable temperatures, are possible.

In FY2023, to further develop the sample environments, a remote gas handling system, a cryostat for powder diffraction, and an electric furnace, as shown in Fig. 3, were commissioned. The gas handling system was developed for BL13XU by improving the system initially designed for BL02B2 [3]. This gas handling system can control absolute pressures from 1 Pa to 130 kPa and is compatible with various gas species. By using this system and a gas cell for capillaries, powder X-ray diffraction in the transmission configuration can be measured in synchronization with LAMBDA detectors. With nitrogen gas low-temperature/high-temperature blower devices, high-resolution powder diffraction data can be obtained under gas and vapor pressures at temperatures ranging from 90 to 1100 K. It is also capable of millisecond time-resolved measurements during gas adsorption and reaction processes. In addition to commissioning the gas handling system, various temperature control devices were also commissioned for temperature regions not achievable by nitrogen gas blower devices. For low-temperature measurements, a closed-cycle cryostat, which is a customized modification of an HE05 (ULVAC Cryogenics, Inc.), allows measurements at temperatures ranging from 5 to 400 K. A copper sample holder inside the cryostat enables low-temperature experiments simply by mounting a capillary. During the measurement, the ω axis oscillates to make the Debye–Scherrer ring uniform. For high-temperature measurements, an electric furnace (HTK1200, Anton Paar) capable of heating capillary samples up to 1473 K at a maximum rate of 50 K/min is available. Usually, powder samples filled in quartz glass capillaries are loaded on a 6-axis spinner on the diffractometer for measurement. We actively continue to develop the measurement system to realize a wide variety of *in situ*/operando observations.

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