BL13XU
Surface and Interface Structures

1. Introduction

The beamline BL13XU is dedicated to revealing the structures of surface layers on solids and thin films at the atomic scale by using X-ray diffraction/scattering (XRD). The techniques users often used are grazing-incidence X-ray diffraction, crystal-truncation-rod (CTR) scattering, reflectivity, nanobeam diffraction, and reciprocal-space mapping in vacuum and air. When a user investigates a surface structure by the above-mentioned XRD, she/he uses an ultrahigh-vacuum (UHV) chamber mounted on a diffractometer. The chamber is equipped with tools for sample preparation and surface analysis in advance. Target materials are widely spread from hard matter (such as a metal and an inorganic material) to soft matter such as an organic semiconductor. Photon energy ranging from 6 to 50 keV is used.

Many users have recently measured diffraction from nanostructures such as atomic wires, nanodots, and ultrathin films. The local structures of device materials such as strains have also been revealed by using a nanobeam. Not only a static structure analysis of a solid surface/interface but also an in situ observation of a dynamic structural response of a surface such as a metal electrode through the imposition of an external field is encouraged.

In addition to the in-vacuum undulator source and standard optics, we offer middle-energy-bandwidth optics using an asymmetric double-crystal monochromator with the Si 111 reflection to meet the growing demand for high photon flux [1]. The monochromator stabilization system and the fast tuning of the incident X-ray energy for anomalous XRD are utilized to meet users' requirements.

In this paper, we report the technical developments and upgrading of the beamline instruments done in FY2021. We note that the second half of the fiscal year was devoted to renewing the beamline optics and relocating the instruments at the end stations, a full picture of which will appear in next year's report.

2. SXRD at ultra-low temperatures

The research on low-dimensional physics has been accelerated by the recent rapid progress in the manipulation of nanometer-scale structures on crystal surfaces, where two-dimensional well-ordered structures can be obtained.

Therefore, understanding the atomic structure of a surface is a starting point for tailoring novel surface materials that meet our requirements and for understanding surface functions such as catalytic reactions. Surface X-ray diffraction (SXRD) is one of the state-of-the-art techniques to determine the constellations of atoms on crystal surfaces, including adsorbates, thin films, and relaxed layers [2].

In low-temperature physics, on the other hand, quantum phases of He atomic layers on graphite at ultra-low temperatures have been attracting much interest over decades as model systems for realizing quantum materials [3], while their structures are still unrevealed. That means SXRD will provide a new opportunity for studying He quantum phases from the firm basis of its atomic constellation if we overcome the difficulties in providing an ultra-low

temperature environment for SXRD [4]. Toward such a new frontier of SXRD, we developed a sample cooling system to reach around 1 K at BL13XU.

A sample holder assembly mounted on the 1 K pot that we developed is shown in Figs. 1(a) and 1(b). The sample stage is capped by a sample chamber with an X-ray window made of a polyimide film adhered to the chamber with stycast. The sample holder assembly is installed in a mother UHV chamber. A UHV environment for a sample is separated from the outer UHV by an indium seal between the stage and the chamber. The sample stage has electronic and gas lines for heating a sample and introducing gases into the chamber, respectively.

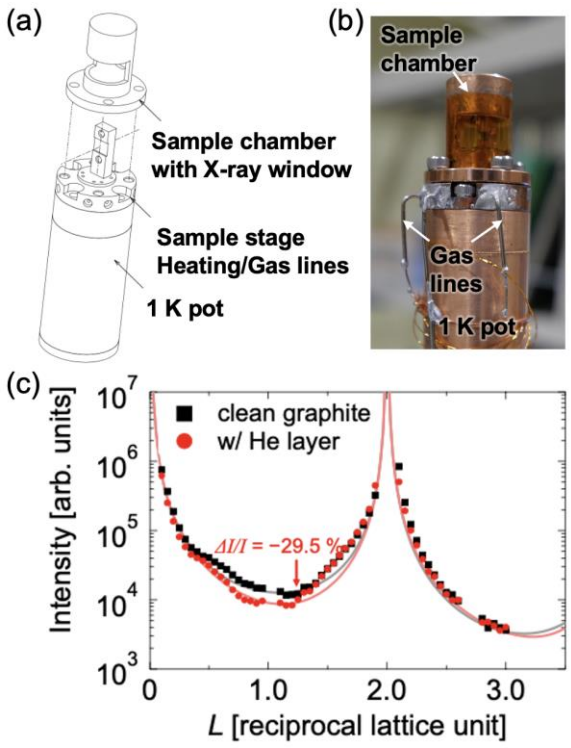

Fig. 1. (a) Drawing and (b) photograph of a sample holder assembly for ultra-low temperatures. (c) CTR intensity profiles from a clean graphite (black symbol) and a He layer on graphite (red symbol) at 1.37 K.

By using the sample holder assembly, we performed a structure analysis of a He atomic layer on graphite, which is a very important target as a low-dimensional quantum model system. Figure 3(c) shows a typical result of SXRD from a He atomic layer on graphite at 1.37 K. One can see approximately 30% differences in intensity between CTR scatterings with and without the He layer, which is clear evidence that we succeeded in observing the He layer by synchrotron X-rays [5]. Our system including the sample holder assembly can accelerate structural surface science at ultra-low temperatures using synchrotron XRD.

3. Development for SAXS measurements on nanobeam X-ray diffraction system

In the fourth hatch of BL13XU, a nanobeam X-ray diffraction system is operated for analyzing the local distortion of crystals. The X-ray beam is focused below 1 μm by a Fresnel zone plate (FZP) and compound refractive lenses (CRLs) depending on the X-ray energy [6–8]. The main object of the investigation is well-oriented crystalline materials such as functional thin films and device materials. In addition, requests for small-angle X-ray scattering (SAXS) measurements combined with an X-ray nanobeam have been increasing recently. In 2021, we have realized SAXS measurements on the nanobeam X-ray diffraction system by designing additional components for SAXS measurements.

The schematic of the SAXS measurement system is shown in Fig. 2. X-rays from the storage ring are tuned by a Si(111) double-crystal monochromator, followed by the total reflection mirrors to eliminate higher-order harmonics. The

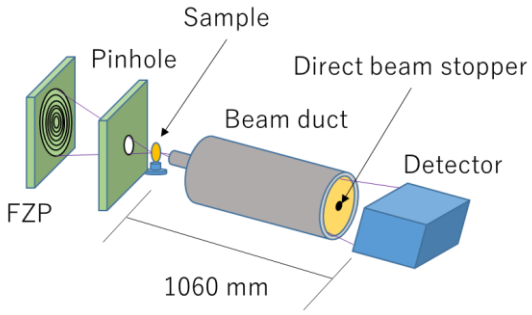

Fig. 2. Schematic of SAXS measurements on nanobeam X-ray diffraction system.

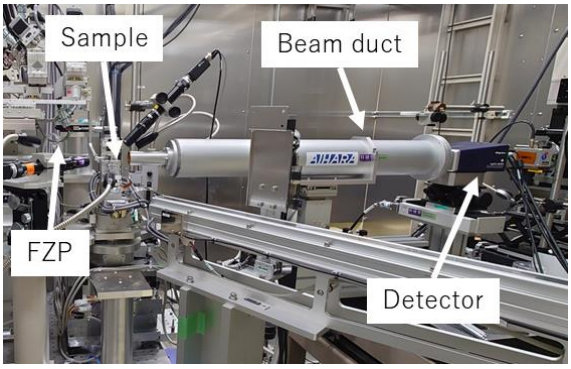

Fig. 3. Picture of SAXS components.

beam is focused by FZP or CRLs on the sample. We prepared a vacuum beam duct with a direct beam stopper and a detector stage for SAXS measurements (Fig. 3). The length of the beam duct is 1 m. The duct has windows with capton films on both the sample and the detector side. A direct beam stopper of lead of 3 mmφ size and 1 mm thickness is fixed on the window of the detector side. The beam duct that is evacuated by the dry pump is mounted on an XZ motorized stage for adjusting accurately the position of the stopper on the optical axis. The detector, HyPix-3000 by Rigaku Co., is mounted on the detector stage after the beam duct. The distance between the sample and the detector is fixed at 1060 mm. In this system, the angular acceptance values by the detector area are 4.1 degrees in the horizontal direction and 2.0 degrees in the vertical direction. The angular resolution is 0.006 degrees.

With this equipment, the structural analysis of noncrystalline materials such as soft materials, nanoparticles, and biomaterials has become possible. The expanding application of the X-ray nanobeam is expected to contribute to the acceleration of material science and development.

Tajiri Hiroo and Sumitani Kazushi
Diffraction and Scattering Division, JASRI

References:

[1] Tajiri, H. *et al.* (2019). *J. Synchrotron Rad.* **26**, 750.
[2] Tajiri, H. (2020). *Jpn. J. Appl. Phys.* **59**, 020503.
[3] Saunders, J. (2020). *J. Low Temp. Phys.* **201**, 615.
[4] Yamaguchi, A. Tajiri, H. *et al.* (2022). *J. Low Temp. Phys.* **208**, 441.
[5] Kumashita, A. Tajiri, H. *et al. JJAP Conf. Proc.* accepted.
[6] Imai, Y. Sumitani, K. & Kimura, S. (2019). *AIP Conf. Proc.* **2054**, 050004.
[7] Sumitani, K. Imai, Y. & Kimura, S. (2018). *Microsc. Microanal.* **24** (Suppl 2), 302.
[8] Sumitani, K. Imai, Y. & Kimura, S. (2018). *J. Instrum*. **13**, C09002.