BL13XU
Surface and Interface Structures

1. Introduction

BL13XU is dedicated to revealing the structures of the surface layers on solids and thin films at the atomic scale using X-ray diffraction/scattering (XRD). Typically, users employ grazing-incidence X-ray diffraction, crystal-truncation-rod (CTR) scattering, reflectivity, microbeam diffraction, and reciprocal-space mapping in a vacuum as well as in air. Investigations of the surface structure by the aforementioned XRD are performed using an ultrahigh-vacuum (UHV) chamber mounted on a diffractometer. The chamber is equipped in advance with tools for sample preparation and surface analysis. Target materials are widely spread from hard matter (e.g., metal or an inorganic material) to soft matter (e.g., organic semiconductor). The photon energy in the range from 6 to 50 keV is used.

Many users have recently measured the diffraction from nanostructures such as atomic wires, nanodots, and ultra-thin films. Local structures of device materials such as strain have been revealed using a microbeam. Not only a static structure analysis of a solid surface/interface, but also *in situ* observations of a dynamic structural response of a surface such as a metal electrode through the imposition of an external field are possible.

In addition to the in-vacuum undulator source and the standard optics, the middle energy–bandwidth optics are available using an asymmetric double-crystal monochromator with the Si 111 reflection to supply the growing demand for a high photon flux [1]. A monochromator stabilization system and fast tuning of the incident X-ray energy for anomalous XRD are utilized to meet user's requirements. This report details the technical developments and upgrades of the beamline instruments in FY2019.

2. Coherent scattering for surface and interface

The rapid progress in the manipulation of nanometer-scale structures on crystal surfaces, where two-dimensional well-ordered structures can be obtained, has accelerated research on low-dimensional physics. Since an atomic constellation is strongly related to its electronic structures, surfaces exhibit intrinsic physical and chemical properties, which differ from those of crystals due to their low dimensionality. Therefore, understanding the atomic structure of a surface is the starting point for tailoring novel surface materials to meet specific requirements and to elucidate surface functions such as catalytic reactions. Surface X-ray diffraction (SXRD) is a state-of-the-art technique to determine the constellations of atoms on crystal surfaces, including adsorbates, thin films, and relaxed layers [2].

In addition, hierarchical structures of surfaces from an atomic to a mesoscopic scale have recently received interest for a deeper understanding of corrosion, plating, self-assembly, etc., where surfaces play an important role intrinsically. Hence, a mesoscopic probe in combination with SXRD should provide new opportunities for studying surface phenomena from the firm basis of its atomic constellation and morphology. Toward this goal, BL13XU was adapted for coherent scattering.

Figure 1(a) shows coherent scattering from a 0.5-

μm-thick Ta test chart with a mesh structure of 1-μm-linewidth and 2-μm pitch using a Hypix-9000 detector (RIGAKU) with 8-keV X-rays. The test-chart image was successfully reconstructed from coherent scattering with a resolution finer than hundreds of nm (Fig. 1(b)), which is consistent with scanning electron microscopy (Fig. 1(c)).

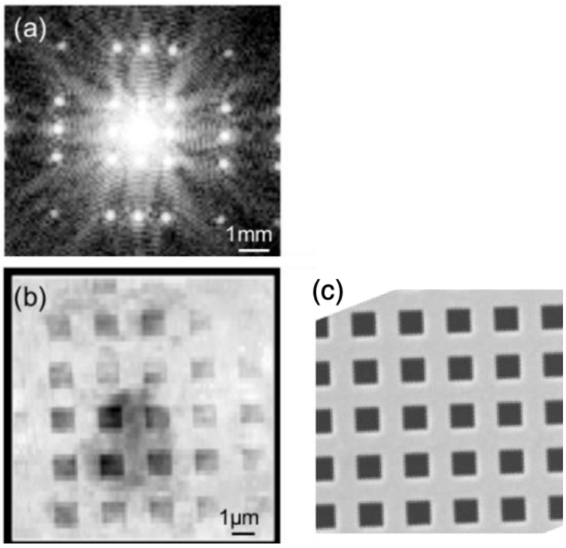

Fig. 1. (a) Coherent scattering from a Ta test chart observed at BL13XU. (b) Reconstructed image of (a). (c) Scanning electron microscopic image of the test chart.

As a next step, an application of coherent scattering for real surfaces and thin films has been commissioned in combination with SXRD. This work was partly supported by JSPS KAKENHI (Grant No. 18H03479) and JASRI President Fund (R18-05).

3. High-resolution micro/nano-beam X-ray diffraction

Micro/nano-beam X-ray diffraction (XRD) is an excellent method to accurately reveal local lattice distortions in materials such as semiconductor thin films or electric devices. In the experimental hutch 4 (EH4) at BL13XU, a micro/nano-beam XRD system is arranged with a Fresnel zone plate (FZP) and compound refractive lenses (CRLs) as focusing devices (Fig. 2) [3, 4]. Previously, a CCD camera and a pixel array detector were used. However, an advanced detector system, which exhibits features such as a wide active area, wide dynamic range, high sensitivity, and low noise, is desired for innovative applications with smaller distortions and operando measurements. In FY2019, a two-dimensional hybrid pixel array detector, HyPix-3000, produced by Rigaku was introduced.

HyPix-3000 is a two-dimensional photon-counting detector. Its sensor element is made of silicon. The detector has 775 × 385 pixels with a pixel size of 100 μm × 100 μm, which results in a wide active area of 77.5 mm × 38.5 mm. The maximum counting rate of the pixels exceeds 1 × 10^6 count/sec. Each pixel is equipped with two pairs of comparator and digital counter. By applying these counting circuits as high and low thresholds, the energy window is applied to eliminate noise due to fluorescence and cosmic rays diving into the detector. Additionally, combining two counters or sequential measurements without any dead time by alternatively switching counters can realize a wide dynamic range.

To mount HyPix-3000, the two-theta arm on the micro/nano-beam XRD system was remodeled. The camera distance can be varied from 120 mm to 1000 mm from the sample by the translation stage on the arm. At the position furthest from the sample, the detector covers a two-theta range of about 4.4° with a high angular resolution of 0.0057°. At the position closest to the sample, the wide range of the two-theta angle by about 35.7° is observed simultaneously with the resolution of 0.047°.

With this detector system, micro/nano-beam XRD can be applied not only to single crystals and epitaxial films but also to materials in the polycrystalline or powder form. The detector also helps reduce the measurement time while realizing a high sensitivity and low noise. Hence, it contributes to the more effective use of the beam time.

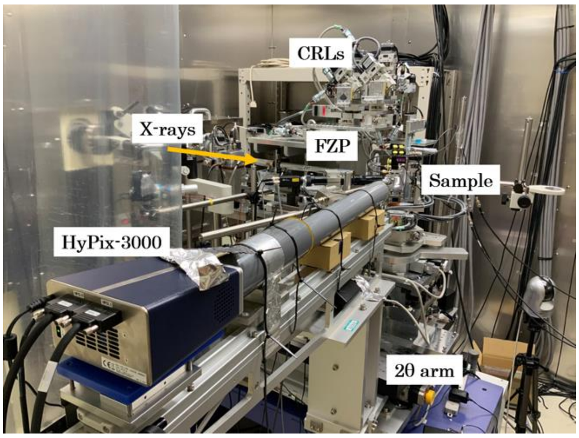

Fig. 2. Micro/nano-beam XRD system in EH4 at BL13XU.

Hiroo Tajiri and Kazushi Sumitani
Diffraction and Scattering Group II, 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] Imai, Y. et al. (2019). *AIP Conf. Proc.* 2054, 050004.
[4] Sumitani, K. et al. (2018). *Microsc. Microanalysis*,. 24 (Suppl2), 302.