BL09XU
HAXPES

1. Introduction

BL09XU is an X-ray beamline with a 32-mm-period standard linear undulator. Until FY2020, the beamline had been shared between nuclear resonant scattering (NRS) and hard X-ray photoelectron spectroscopy (HAXPES). In FY2021, BL09XU was reorganized as a beamline dedicated to HAXPES, and the name of the beamline was changed to “HAXPES”.

HAXPES measurements require many optical instruments such as high-resolution monochromators (HRMs) and X-ray phase retarders (XPRs) to determine detailed chemical bonding states, as well as focusing techniques to perform efficient measurements in microscale areas [1]. Until the end of FY2020, BL09XU and BL47XU had been partially used for HAXPES, and the applications covered a wide range of research studies from basic physical properties to industrial development using these advanced optical and focusing instruments [2,3]. Sharing beamline components, however, causes problems such as the instability of beam performance and significant beamtime loss. To overcome these problems, BL09XU was reorganized as a beamline dedicated to HAXPES by integrating the HAXPES activities of the two beamlines in FY2021. In this upgrade, all optics except for a liquid-nitrogen-cooled double-crystal monochromator with Si 111 reflection were upgraded to state-of-the-art ones specialized for HAXPES experiments for conducting more advanced HAXPES applications, such as resonant HAXPES (r-HAXPES) and three-dimensional (3D) spatial-resolved chemical bonding analysis.

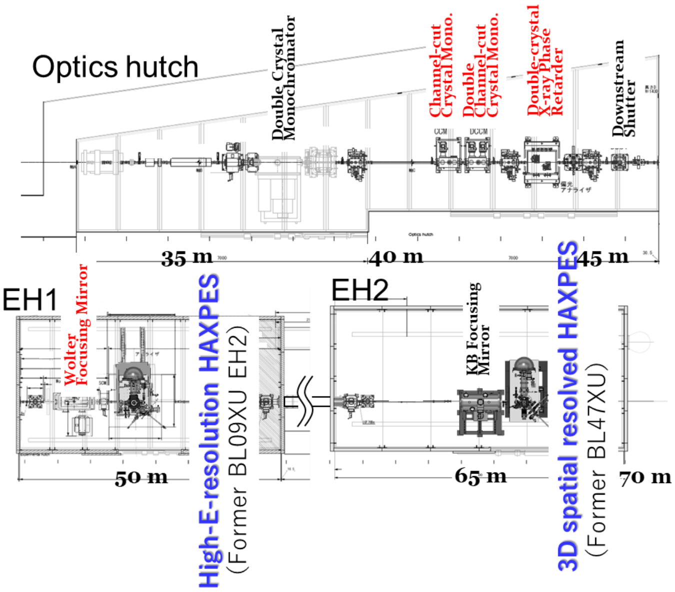

Fig. 1. Beamline layout of new BL09XU. All optical components except for the focusing system are located in OH. Two sets of photoelectron analyzers and focusing mirrors with different specifications are installed in EH1 and EH2.

2. Beamline layout and optical components

The beamline layout is shown in Fig. 1. The key point is that all the optical components except for the focusing system are located upstream of a downstream shutter in the optics hutch (OH) to ensure a constant heat load to the optics devices, resulting in a highly stable beam performance, even when entering the experimental hutch (EH) to, for example, exchange samples.

In OH, HRMs, including a channel-cut crystal monochromator (CCM) and two double-CCMs (DCCMs), and a double-crystal XPR (DXPR) are installed. The CCM is used for high-energy-resolution analysis at discrete photon energies of 6, 8, and 10 keV. For example, the energy resolution ΔE is ~34 meV at 7.94 keV using the Si 444 reflection. On the other hand, the DCCMs with the low-index reflections of Si 220 or 311 provide a high-intensity beam with a moderate ΔE below 300 meV in a wide energy range of 4.9–12 keV. In addition, a fixed-exit condition is fulfilled for the DCCMs. The DCCMs are used for most applications including resonant HAXPES. The DXPR using two diamond crystals with a thickness of 0.2 mm in the 220 Laue geometry provides a high degree of polarization above 0.9 in a wide energy range of 5.9–9.5 keV.

3. HAXPES instruments and focusing mirrors

In EHs, we installed two pairs of HAXPES instruments and focusing mirrors that match the performance of the analyzers. The two HAXPES instruments have the same SCIENTA OMICRON R4000 analyzers, but different features: The EH1 analyzer acquires photoelectrons with kinetic energies up to 12 keV, allowing the analysis of electronic states from surfaces to deeper buried interfaces, as well as r-HAXPES applications of 5d transition elements with absorption edges in the 10–12 keV range. On the other hand, a wide-acceptance-angle objective lens, originally developed at SPring-8, with a photoelectron detection angle of ±32° is mounted in front of the EH2 analyzer [2,3]. This analyzer is suitable for dissecting the depth information of chemical bonding states from the sample surface to the bulk, since it captures photoelectrons over a wide angular range simultaneously.

A monolithic Wolter mirror was adopted for the focusing system of EH1. The characteristics of this mirror are a few adjustment axes and a high tolerance to pitching error. These features simplify the refocusing procedure after the experiments at EH2 and make the beam conditions stable after focusing adjustment. A Kirkpatrick–Baez (KB) focusing mirror system is used for the apparatus at EH2 [3]. This system has a pair of long mirrors, providing a high-intensity beam with a flux of ~6×10^{12} photons/s. The focusing size is variable with the aperture size of the front-end slit at the upstream of the beamline: 1 μm vertically and 1 to 11 μm horizontally. In particular, by reducing the horizontal width of the front-end slit, the horizontal focusing size is reduced to 1 μm.

4. Upgrade of instrument control system

The control method of instruments was switched to the “BL774” system [4]. Recently, there has been increasing needs to perform advanced measurements by linking and controlling multiple instruments, such as r-HAXPES analyses and automatic measurements with sample exchange. The BL774 system allows users to control devices with different manufacturers and drive mechanisms, such as motor-driven devices and piezo stages, in a unified manner without knowing the details of the instrument.

5. Advances of resonant HAXPES application (EH1)

The r-HAXPES experiment provides a unique opportunity to analyze electronic structures with enhanced element and valence selectivities by achieving incident X-ray energy scanning near the X-ray absorption edge. In particular, the 2p-5d r-HAXPES of rare-earth compounds allows us to experimentally obtain the Coulomb repulsion U_{fd} between 4f and 5d electrons at a rare-earth site [5,6], which plays an important role in the quantum critical phenomena of valence fluctuation [7]. We developed a r-HAXPES technique in BL09XU before upgrade in collaboration with Partner User (PU) members. The conventional measurement using a single CCM, however, had a problem in that the height of the beam emitted from the CCM is changed by $d\cos\theta$ in energy scanning, where d is the distance between the blades of the CCM and θ is the Bragg angle, and is available only in a limited sweep range of photon energies of about 100 eV. If the fixed-exit condition is not satisfied, r-HAXPES analyses are very difficult on samples with few flat surfaces or spatially inhomogeneous materials. In particular, for strongly correlated electron materials including rare-earth compounds, the fracture or cleavage process is indispensable to expose clean surfaces, and the exposed surfaces have almost no flat areas.

With the upgrade, a DCCM was introduced and a fixed-exit condition is realized during energy scanning, resulting in a stable photoelectron emission intensity in the r-HAXPES analysis. This allows us to target samples that were previously difficult to analyze. The target of r-HAXPES measurement has expanded to industrial materials such as photocatalytic and steel materials, not only rare-earth compounds. The r-HAXPES analysis of a microstructure in these materials has become a target for future applications.

6. Advances of three-dimensional electronic state analysis (EH2)

By combining a 1 μm focused beam, sample scanning, and HAXPES measurement, scanning photoelectron imaging becomes possible. Figure 2(b) shows the Ta 3d intensity distribution for an X-ray chart sample (Fig. 2(a)) when the sample is scanned in the 2D direction along the sample surface. The Ta 3d intensity is obtained in a chart pattern, which correctly reflects the sample structure.

In this manner, by defining a measurement point with 2D mapping in the surface direction and performing an angle-resolved measurement at that point, 3D spatially resolved chemical bonding states are obtained. The chemical bonding state analysis in

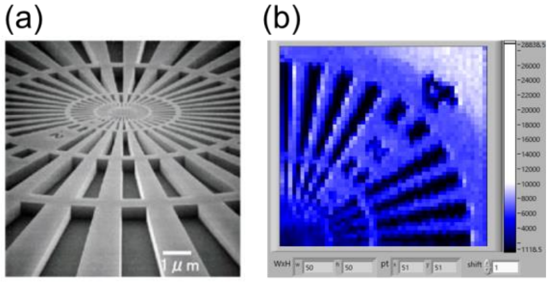

Fig. 2. (a) Scanning electron microscopy image of an X-ray chart sample (XRESO-50HC produced by NTT-AT). (b) Photoelectron intensity map at Ta 3d core level.

the depth direction with nanometer resolution in a local region on the micrometer scale is unique in the world. A similar 1 μm focused beam was available at BL47XU [2,3], but the photoelectron intensity at EH2 in BL09XU after upgrade is about 10 times higher than in BL47XU, enabling analysis in a shorter time and with a higher signal-to-noise ratio. In recent years, user needs for local electronic structure analysis have been increasing in a wide range of fields, such as grain boundary materials in composite materials and applied electric devices, and the EH2 instrument is capable of meeting these needs.

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