BL09XU
Nuclear Resonant Scattering

1. Introduction

BL09XU, which is known as the Nuclear Resonant Scattering beamline [1], is an X-ray beamline with a 32-mm-period standard linear undulator dedicated to investigating a broad range of physical science questions. It is mainly used for nuclear resonant scattering (NRS) and Hard X-ray Photoelectron Spectroscopy (HAXPES). Intense X-rays between 4.91 keV and 80 keV are obtained in the experimental hutch using a double-crystal liquid nitrogen–cooled monochromator operating at the Si (111) reflection for E<38 KeV and the Si (333) reflection for higher energies.

2. NRS

NRS is resonant scattering using the transition between the ground state and an excited state in nuclei. The resonance energy width is typically in the μeV–neV range, which is much narrower than meV–eV scale of the energy level of electrons. Techniques currently conducted in the beamline using NRS include: (1) synchrotron Mössbauer spectroscopy, (2) nuclear inelastic scattering (NIS), (3) quasi-elastic scattering (QES), and (4) nuclear excitation for fundamental physics.

In FY2019, the main beamline improvements included (1) upgrading the spectrometer for QES using time-domain NRS and (2) the installation of two closed-cycle cryostats. One is for the samples and the other is for the analyzer in energy-domain synchrotron Mössbauer spectroscopy.

2-1. Upgrade of the spectrometer for QES using time-domain NRS

QES is a unique tool for studying the dynamics of soft matter. One barrier for more widespread use was the signal rates. Hence, a new spectrometer with improved specifications, which was constructed in collaboration with Dr. Saito of Kyoto University in FY2018 [2], was tested in FY2019 to obtain higher resonant signals. The spectrometer is composed of a high-resolution monochromator (HRM), a cryostat, and APD detectors. Compared to the original 3.5-meV HRM, the measured resonant counts after the new 6-meV HRM increased by 50%. Figure 1 shows the new cryostat and APD detectors. The shorter outer diameter of this He-flow cryostat allows wider solid angles to be covered with less background scattering due to the thinner Kapton window. In the new system, the aluminum mylar films are replaced by two transparent Kapton windows, which enable the direct measurement of sample vibration in the cryostat by laser. The vibration is small enough for QES measurements. One APD detector has two-

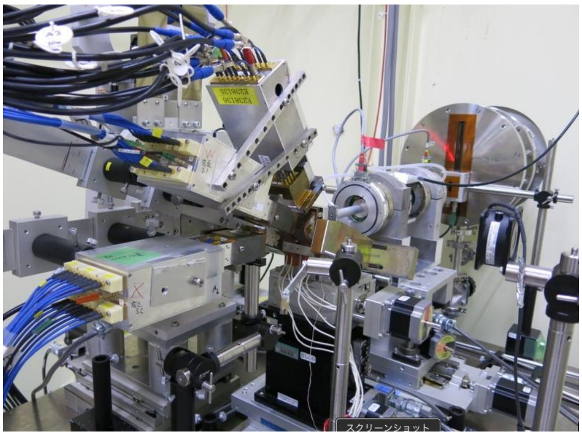

Fig. 1. Upgraded spectrometer for QES using time-domain NRS.

layered 8-channel APD elements, and the detectors are set with a smaller dead space to cover the sequential q-range.

2-2. Installation of low-vibration closed-cycle cryostat systems

Energy-domain Mössbauer spectroscopy is a robust technique, which is particularly interesting for high-energy nuclear resonances (e.g., ^{61}Ni at 67.4 keV, ^{174}Yb at 76.5 keV). To realize a suitable energy resolution and a higher efficiency, both the sample and analyzer must have low vibrations and low temperatures. However, the recent supply problem of liquid He in Japan (increased cost and reduced availability) has raised the urgency to replace the flow-type cryostat with a closed-cycle one. Therefore, two pulse-tube closed-cycle cryostat systems with lower vibrations were installed. A damping mechanism was also installed by the company that supplied the cryostats to reduce the vibration. These systems will be tested in FY2020.

3. HAXPES

The HAXPES station at BL09XU was opened for public use in FY2014. The advantages over BL47XU are its high flux and energy tunability [3]. The high-flux micro-focus beam with a size of about 5 μm (vertical) × 13 μm (horizontal) is achieved by a long Kirkpatrick-Baez (K-B) focusing mirror with a length of about 1 m. Compared to that at BL47XU, its intensity realizes a 30 times higher photoelectron detection efficiency. The high-flux beam allows spectra with a high energy resolution ΔE of about 100 meV to be acquired and a diamond phase retarder to be used. Hence, changing the beam polarization can measure the magnetic circular dichroism of HAXPES spectra. Consequently, spintronic materials, for example, can be investigated.

In FY2017, an energy-tunable system was developed to realize resonant HAXPES (r-HAXPES) measurements in collaboration with Partner User (PU). This upgrade was part of the PU program of SPring-8 called, “Construction of composite measurement technology of resonant hard X-ray photoemission and X-ray absorption spectroscopies, for elucidating quantum critical phenomena of strongly correlated electron systems”. Selective utilization of Si 311 and Si 333 channel-cut monochromators (CCMs) allows r-HAXPES spectra to be measured with ΔE<300 meV in the incident photon energy range of 4.91–9 keV. Unfortunately, the aging analyzer began to frequently experience troubles inside the HAXPES analyzer such as insulation failure. In FY2019, a new analyzer was installed.

3-1. High-resolution electron analyzer with a high withstand voltage and strengthened correction function of photoelectron trajectory

The HAXPES analyzer was more than 10 years old, and it recently began experiencing troubles due to its age. At the end of FY2018, a new analyzer was introduced to improve the performance of HAXPES measurements. The features of the new analyzer are an increased withstand voltage from 9 kV to 12 kV and a strengthened correction function of the photoelectron trajectory.

The 12 kV withstand voltage is expected to significantly expand the range of samples used in r-HAXPES experiments. Currently, the main targets of r-HAXPES measurements are 4f rare-earth compounds. In particular, the clarification of the mechanism on the quantum critical phenomenon

(QCP) is a hot topic, and r-HAXPES is of great interest because it can realize a direct and quantitative estimate of the Coulomb interactions between the 4f and 5d electrons in the rare-earth site (U_{fd}), which play important roles in the QCP. However, the usefulness of the r-HAXPES is not limited to only fundamental research on strongly correlated compounds. It is also practical for the development of applied materials such as steel and photocatalysts. Consequently, its application is gradually expanding, and the new analyzer should support r-HAXPES measurements at the L_3-edge of 5d transition elements, which constitute strongly correlated spintronics materials that should be the foundation of next-generation information technology (IT) devices and other advanced device materials.

The strengthening of the correction function of the photoelectron trajectory is an indispensable part of the development of the HAXPES technique in an external magnetic field. Under a magnetic field, the Lorentz force bends the photoelectron trajectory, making it difficult to detect photoelectrons. However, applying a corrective electric field to the deflecting lens can detect photoelectrons influenced by the magnetic field. Therefore, strengthening the corrected electric field has realized an efficient photoelectron detection. Currently, HAXPES measurements at 0.2 T has been achieved, and combining the use of high-energy photoelectrons with a kinetic energy > 9 keV should make it possible to conduct a HAXPES measurements at higher external magnetic fields.

The new analyzer not only resolves problems such as insulation failure, but also should expand the type of samples that can be measured by HAXPES. In addition, a high-speed charge-coupled device (CCD) detector equipped with this analyzer should dramatically improve the photoelectron detection efficiency. As a result, the measurement time is estimated to be halved, and the linearity of the detection improved.

Yoshitaka Yoda[*1], Akira Yasui[*2], and Yasumasa Takagi[*2]

[*1] Precision Spectroscopy Division, JASRI

[*2] Spectroscopic Analysis Group II, Spectroscopy and Imaging Division, JASRI

References:

[1] Y. Yoda et al., Nucl. Instrum. Methods A, 467-468, 715 (2001).

[2] Y. Yoda et al., SPring-8 Annual Report 2018, 32 (2019).

[3] E. Ikenaga et al., Synchrotron Radiation News 31, 10 (2018).