BL39XU
Magnetic Materials

1. Introduction

BL39XU is a hard X-ray beamline dedicated to the study of magnetic materials and strongly correlated electron systems. Techniques include X-ray absorption spectroscopy (XAS), X-ray magnetic circular dichroism (XMCD), X-ray emission spectroscopy (XES), and resonant X-ray magnetic scattering. Recent developments have focused on X-ray spectroscopy measurements under multiple extreme and complex conditions, and scanning XAS/XMCD imaging using a nano-focused X-ray beam. These techniques are available for user experiments, and further developments are ongoing.

In FY2019, three projects were undertaken. (1) The vacuum chamber of the XES spectrometer was installed to complete the system for 15 crystals, and three kinds of analyzer crystals were installed to extend the emission energy range. (2) The electromagnet for XMCD measurements under multiple extreme conditions was opened to users. (3) Two-dimensional (2D) XMCD imaging using the on-the-fly method was developed.

2. Experimental station for X-ray spectroscopy under multiple extreme conditions

2-1. X-ray emission spectrometry with multiple analyzer crystals

To study the electronic and magnetic states of strongly correlated electron systems at BL39XU, instrumentations for X-ray spectroscopy under multiple extreme conditions such as high/low temperature, high magnetic field, and high pressure has been progressing [1]. XES is an especially powerful tool to obtain detailed information about electronic states such as valence, spin, and coordination states. Due to the weak emission signals of trace elements, a long time is typically necessary to acquire accurate XES spectra. To overcome this, a high-efficiency XES spectrometer equipped with multiple analyzer crystals has been under development since FY2016.

In FY2019, a vacuum chamber for the XES spectrometer was installed to avoid degradation of the emission X-ray intensity by the X-ray path among sample, analyzer, and detector. Figure 1 shows a photograph of the XES spectrometer. Except for the sample and detector positions, which correspond to the X-ray emission and focal points, respectively, the X-ray path for the emission X-rays, which includes the analyzer crystals, is in a vacuum. The vacuum chamber provides the emission intensity from all 15 analyzer crystals, while

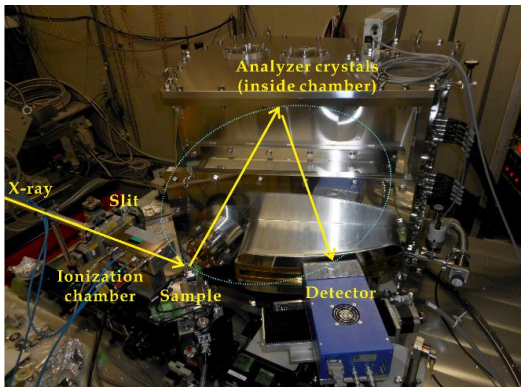

Fig. 1. New vacuum chamber of the X-ray emission spectrometer with 15 analyzer crystals. Three towers, each of which can mount a set of five analyzer crystals, are mounted inside the chamber, whereas the emission point (sample) and focal point (detector) are located outside the chamber.

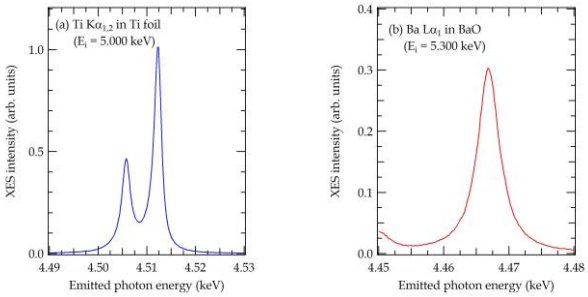

Fig. 2. XES spectra for (a) Ti Kα1,2 emission in Ti foil and (b) Ba Lα1 emission in a BaO pellet using five Ge 400 analyzer crystals.

suppressing X-ray absorption due to air and window materials (Kapton film). Hence, the efficiency for the XES measurements is advancing. Consequently, the XES spectrometer can observe the spectra of a dilute element of Eu 5 ppm involving in Mn compounds.

Five analyzer crystals of Ge 400, Ge 620, and Si 620 were installed to extend the emission X-ray energy. Particularly, the extension of the lower-energy region below 4.8 keV is desired for use of Ti *K*α and Ba *L*α emissions. Figure 2 shows the combination of five Ge 400 crystals with the high-efficiency XES spectra for both Ti *K*α and Ba *L*α emissions.

To use the 15 analyzer crystals, Ge *nnn* and Ge *nn*0 are currently available to select the X-ray emission energy. However, to extend the available X-ray energy range for high-efficiency measurements, we plan to facilitate analyzer crystals sequentially.

Users for performing the XES spectroscopy has gradually increased. In FY2019, about 40% of the beamtime was user occupied. Additionally, the research fields have expanded to include strongly correlated electron systems, chemical analysis, and Earth/environment science. This diversity has led to a demand to select a degree of freedom in emission lines of target elements. To facilitate a wider X-ray range, we plan to amplify different types of analyzer crystals.

2-2. High-field electromagnet with a large sample space

X-ray absorption spectroscopy in the hard X-ray region offers a unique opportunity to investigate the electronic and magnetic states of matter under various environments (strong magnetic field, high/low temperature, high pressure, etc.). The merit of an electromagnet with a large sample space is that it is easier to build an experimental setup that can be combined with various kinds of external fields such as an electric field, electric current, and optical lasers. In FY2018, a new electromagnet, which can combine an additional large sample space and a magnetic field much higher than that previously available, was installed to enhance the advantage of the experimental setup with the existing electromagnet. The electromagnet is already opened to users requiring a magnetic field above 2 T. However, measurements at room temperature are currently limited. The use of the new electromagnet allows an expanded sample environment for X-ray spectroscopy under multiple extreme conditions, extending the advantages to high-sensitivity XMCD measurements at BL39XU. In the near future, a helium-flowing type cryostat will be installed for temperature-dependent measurements.

This upgrade should inspire research in materials science, magnetism, spintronics, and applications for industrial products. Potential applications include the development of high-performance permanent magnets with very little use of rare earth elements such as Dy, elucidation of the functions of

strongly correlated materials, development of spintronics and magnetic storage devices operating under external electric and magnetic fields, and studies on peculiar magnetism emerging under high pressure.

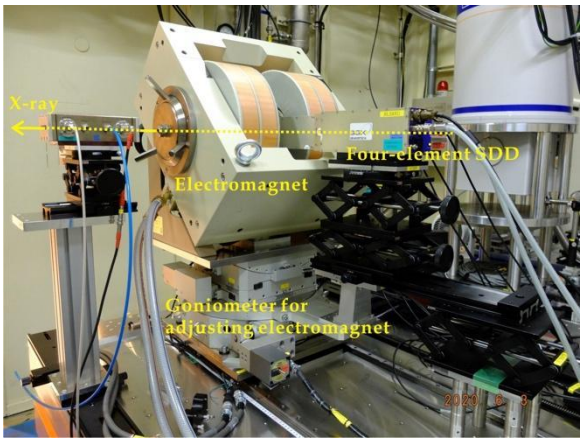

Fig.3. New electromagnet for X-ray spectroscopy experiments under extreme conditions installed in the experimental hutch 1 of BL39XU.

3. Experimental station for X-ray nanospectroscopy

Since FY2011, a scanning hard X-ray nanoprobe has been developed for XAFS/XMCD microscopy at the X-ray nanospectroscopy station in experimental hutch 2 [2]. Kirkpatric and Baez (KB) mirror optics are used to generate a circularly polarized and focused X-ray beam with a typical spot size of 100 nm × 100 nm between 5 keV and 16 keV.

An X-ray magnetic tomography technique was developed [3] to observe the three-dimensional (3D) magnetic domain structure in ferromagnetic samples. However, a long time is required to obtain a 2D XMCD image due to the conventional step-scan method. In FY2019, a 2D XMCD scanning microscopy was developed using the on-the-fly method [4]. Specifically, a multi-channel gate circuit synchronized with fast switching of X-ray polarity was developed. If the helicity switching is very fast compared to the position movement and sampling time, then the XMCD signal can be considered as an acquisition at the same position, even if the sample position is moving.

Figure 4 shows an example of a 2D XMCD image around Fe *K*-edge in $Y_3Fe_5O_{12}$ film using a four-element silicon drift detector. Here, the beam size at the sample position was 130 nm (horizontal) × 170 nm (vertical), and the estimated photon flux was 7 × 10^{11} photons/s at 7.113 keV. Using a helicity-switching frequency of 37 Hz, a scanning speed of 10 μm/s, and a sampling rate of 500 ms/point, the acquisition time in this area was about 16 min. Compared to the conventional step-scanning method, which required 60 min, this is a significant time reduction. Even with the shorter time, the magnetic domain structure was clearly observed. Moreover, the currently developed on-the-fly technique is compatible with 2D XMCD imaging in the fluorescence detection mode and will be useful

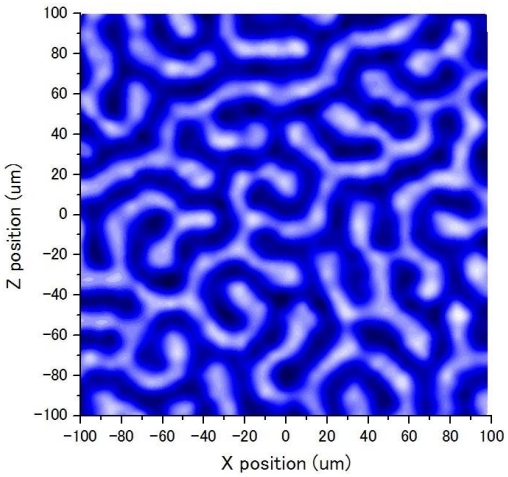

Fig. 4. 2D XMCD image at the Fe K-edge (7.113 keV) in $Y_3Fe_5O_{12}$ film. Field of view is 200 μm × 200 μm (40×40 pixels).

to study magnetic thin films and micro-magnetic devices in dynamic processes [5].

Naomi Kawamura, Hitoshi Osawa, Masaichiro Mizumaki, and Motohiro Suzuki

Spectroscopic Analysis Group II, Spectroscopy and Imaging Division, Center for Synchrotron Radiation Research, JASRI

References:

[1] N. Kawamura *et al.*, *J. Synchrotron Rad.* **16**, 730 (2009).

[2] M. Suzuki, H. Yumoto, T. Koyama, H. Yamazaki, T. Takeuchi, N. Kawamura, M. Mizumaki, H. Osawa, Y. Kondo, J. Ariake, A. Yasui, Y. Kotani, N. Tsuji, T. Nakamura, S. Hirosawa, K. T. Yamada, S. Kim, K. J. Kim, M. Ishibashi, T. Ono, and H. Ohashi, *Synchrotron Radiation News* **33**, 4 (2020).

[3] M. Suzuki *et al.*, *Appl. Phys. Express* **11**, 036601 (2018).

[4] H. Osawa *et al.*, *The 33th Annual Meeting of Japan Synchrotron Radiation (JSR2020)* (2020).

[5] N. Kikuchi *et al.*, *Jpn. J. Appl. Phys.* **59**, SEED03 (2020).