Transactions of the Materials Research Society of Japan 33[3] 649-652 (2008)

Performance of X-ray Reflectivity and Grazing-Incidence Small-Angle X-ray Scattering Measurement at Beamline BL15 of the Saga Light Source

Kazushi Sumitani, Kotaro Ishiji, Toshihiro Okajima, Yasuharu Hirai,
[1]Kazuhiro Ueda, [1]Akio Yoneyama
Saga Light Source, 8-7 Yayoigaoka, Tosu, Saga 841-0005, Japan
Fax: +81-942-83-5196, sumitani@saga-ls.jp
[1]Advanced Res. Lab., Hitachi, Ltd., Akanuma, Hatoyama, Saitama 350-0395, Japan

Abstract

An x-ray reflectivity measurement system has been installed in the bending-magnet beamline, named BL15, at the Saga Light Source. This beamline provides synchrotron radiation in the energy range from 2.1 to 23 keV for the performance of x-ray diffraction experiments, x-ray reflectivity measurements, etc. To test the performance of this reflectivity measurement system, we have analyzed the interface of thin films with nanometer-scale thickness on semiconductor substrates. The reflectivity of the order of 10^{-7} has been observed for a 50-nm thick film of thermal SiO_2 on a Si wafer. We have fitted the calculated reflectivity profile with a single layer model to the data, and have obtained the value of thickness. In addition, we have performed grazing-incidence small-angle x-ray scattering (GISAXS) experiments on a MnPt thin film.

Key words: synchrotron x-rays, structural analysis, surface and interface, reflectivity, grazing-incidence small-angle x-ray scattering

1. INTRODUCTION

The Saga Light Source (SAGA-LS), in operation since February 2006, is a medium-size facility of synchrotron radiation for material science [1,2]. The electron storage ring operates at 1.4 GeV; the photon energy ranges from extreme ultra-violet light to hard x-rays. Saga prefecture, a local government in Kyushu island of Japan, owns the facility and offers beamtimes to users as a strong experimental tool. The purpose of the facility is not only academic researches but also industrial studies. Nowadays, the beamtimes are provided for users with reasonable cost, simple procedure for proposals, and user-friendly supports. Saga prefecture possesses three beamlines for material processing, for soft x-ray applications, and for hard x-ray experiments.

The beamline named BL15 is a hard x-ray beamline for structural analyses and characterizations of materials [3]. The optical components and the experimental devices are optimized for hard x-ray experiments in the energy range from 2.1 to 23 keV. The methods of x-ray absorption fine structure (XAFS), x-ray fluorescence analysis, imaging, x-ray diffraction (XRD), x-ray reflectivity (XRR), small angle x-ray scattering (SAXS), etc. are available in this station.

In a variety of techniques established in BL15, the x-ray reflectivity measurement is an effective way to investigate a structure of a thin film. In recent industrial field, the solid state property of thin films or multi-layer films which has a thickness of the order of nano-meter becomes important. The reflectivity measurement is a method to analyze film thickness and interface roughness.

In this study, we performed the x-ray reflectivity measurement with a multi-axis diffractometer in BL15. The performance of the beamline including the optical section and the experimental equipment was evaluated for the reflectivity measurement. Furthermore, we developed and demonstrated a grazing-incidence small-angle x-ray scattering (GISAXS) measurement in

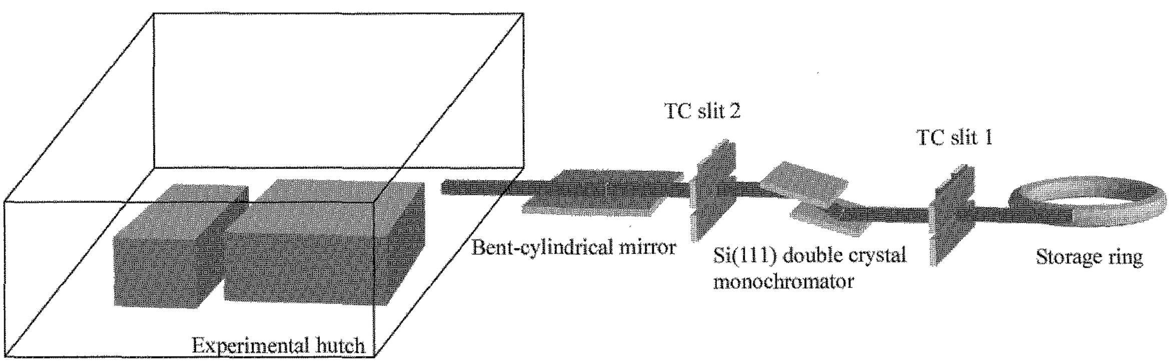


Fig. 1 Apparatus of the beamline BL15 in Saga Light Source.

BL15. GISAXS is a hot topic to study the surface and the interface characteristics using synchrotron x-rays.

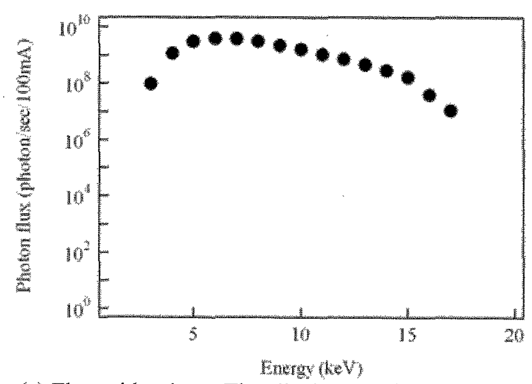
2. BEAMLINE APPARATUS

Apparatuses of BL15 are shown in Fig. 1. The x-ray beam from the storage ring is monochromated by a Si(111) double-crystal monochromator. A bent-cylindrical mirror follows the monochromator for eliminating higher order harmonics. The mirror also plays a role for focusing x-rays in the experimental hutch. The focusing point is optimized on the position of the detector for SAXS.

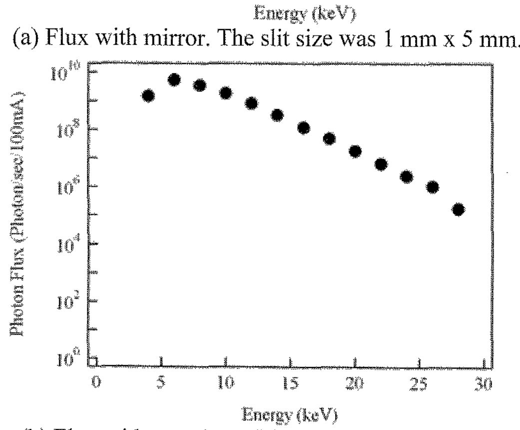
In the experimental station, we have established a series of slits, goniometers and detectors. For adjusting to many types of experiments, each device is mounted on sliding stages. For example, we use the diffractometer at the end of the table. In case of a GISAXS measurement, however, we move the diffractometer to the middle of the table. We can take a camera length enough for the GISAXS measurement. All equipments are arranged on the ground of such flexibility.

The photon flux was estimated at the sample position in the experimental hutch (Fig. 2). When the focusing mirror was used, the maximum flux of 4 x 10^9 photons/s was achieved at the photon energy of 6 keV in the area of 1 mm x 5 mm. Below 10 keV, we obtained the flux more than 10^9 photons/s.

The mirror is designed to cut off x-ray photons above 14.2 keV. We can obtain a direct beam from the monochromator by removing the mirror from the beam axis. Without the focusing mirror, the incident beam with a wide cross section of 5 mm x 50 mm was obtained. This wide beam is convenient for imaging experiments such as phase contrast imaging and topography. In a 5 mm x 5 mm area, the maximum flux of 5 x 10^9 photons/s was obtained at 6 keV. Even in the energy range of 20~23 keV, we obtained at least 5 x 10^6 photons/s.



(a) Flux with mirror. The slit size was 1 mm x 5 mm.



(b) Flux without mirror. The slit size was 5 mm x 5 mm.

Fig. 2 Photon flux at the sample position in the experimental station.

3. REFLECTIVITY MEASUREMENT

We performed the reflectivity measurement of a SiO_2 thin film grown on a Si substrate prepared by the conventional thermal process. The designed thickness of the SiO_2 layer was 50 nm. For evaluating the layer thickness, we applied a measurement of spectroscopic ellipsometry. The thickness of the SiO_2 film obtained was at 50.9 nm.

The multi-axis diffractometer was used in the measurement at 8 keV (Fig. 3). The beam was focused by the mirror on the sample. The measured beam size was 0.1 mm x 1 mm, which was defined by the slit in front of the diffractometer. The detector was NaI scintillation counter. To observe a wide dynamic range with NaI, a series of Al attenuator was used for the adjustment of the incident x-ray flux.

Figure 4 shows the reflectivity curve obtained in the measurement. The clear oscillation was seen according to the thickness of the SiO_2 layer. The oscillation was observed up to $2\theta=8°$; the reflectivity decreased to the order of 10^{-7}. This indicates the advantage of the brilliant synchrotron radiation source compared with a laboratory x-ray source. The measurement of the curve took almost two hours in which the diffractometer and sample alignment times did not included.

At first, we examined a Fourier transform of the reflectivity in the analysis. In this method, the information of the layer was obtained without constructing the layer model [4,5]. For the extraction of the oscillation, the reflectivity was normalized by the averaged curve derived by the polynomial equation fitting in the logarithmic plot. As a result, the strong peak was obtained at the thickness of 48 nm (Fig. 5). This result obtained from the profile of the Fourier transform indicates that the SiO_2 layer thickness is about 48 nm.

In the next step, we performed a least-squares fitting analysis. We constructed a single layer model with the SiO_2 layer of 48 nm as a starting point of the procedure. As a result, the thickness of the SiO_2 layer obtained by

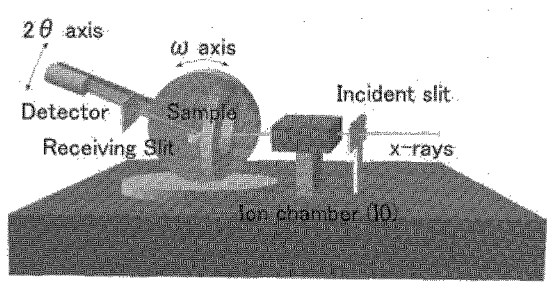


Fig. 3 Setup for the reflectivity measurement. The size of the incident slit was 0.1 mm x 1 mm. The reflectivity measurement was performed using ω and 2θ axes. The receiving slit was set to accept the entire reflection beam.

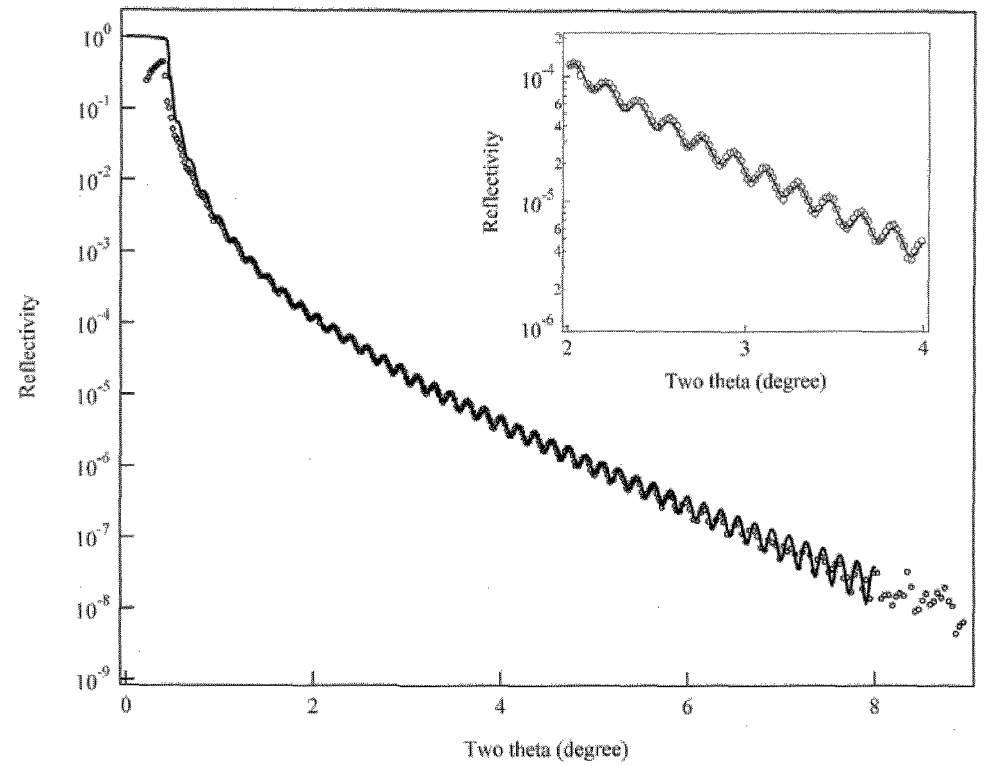


Fig. 4 Reflectivity from the thermal SiO_2/Si. The open circle and the solid line indicate the observed and calculated reflectivity. The inset figure shows the magnification of the reflectivity curve.

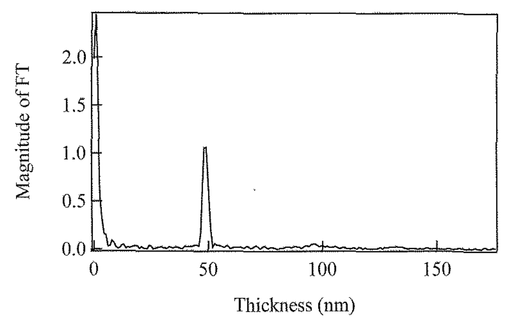


Fig. 5 Fourier transform of the reflectivity.

the fitting was 49.0 ± 0.15 nm, which was roughly consistent with the thickness estimated by the ellipsometry. The values of roughness of the SiO_2 surface and the SiO_2/Si interface were 0.3 nm and 0.1 nm, respectively, which indicates that the interface was abrupt. By comparing the calculated and obtained profiles, it is obvious that the single layer SiO_2 model well explains the reflectivity. According to these results, we conclude that the reflectivity measurement at BL15 is an effective method to analyze film thickness and interface roughness.

4. GISAXS MEASUREMENT

In this study, we also tried a GISAXS measurement in BL15. GISAXS is considered as a promising tool to investigate a state of surface and interface in the range of nano-scale [6].

The sample was a MnPt thin film, which have been used as a material for a giant magnetic resistance (GMR) sensor. The film was grown on a glass substrate with the thickness of about 20 nm.

The experimental setup is shown in Fig. 6. The sample was mounted on the diffractometer, which enabled the x-ray incident angle controlled precisely. The incident beam size was 0.8 mm x 0.7 mm formed by the 1st slit. The 2nd slit was set so that the scattered x-rays from the blade of the 1st slit were eliminated. The incident angle was 0.35 degrees, which was greater than the critical angle of the MnPt film. The scattered x-rays passed through the vacuum path and were detected by the imaging plate or the CCD camera with the image intensifier. The beam stopper was put in front of the detector to shut out the direct beam and specular reflection. The camera length was 1770 mm.

The scattering pattern is shown in Fig. 7. Broad peaks were observed at the side of the specular reflection, q_y=0.5 nm^{-1} and q_z=1.0 nm^{-1}. These scatterings indicate that the film has periodic structure in the in-plane direction. The period is 12.5 nm calculated by $L=2\pi/q_y$. We also found peaks at q_z=3.0 nm^{-1}. The peaks at higher q_z reflect the three-dimensional structure of the particles. From this result, we conclude that it is possible to perform a GISAXS measurement for investigating the nano-scale structure in BL15.

The ring patterns clearly seen in the figure were diffraction peaks from the beam stopper produced by the specular reflection. Although the beam stopper was necessary for blocking the direct and specular reflected beam, the stopper itself was seen as a scatter. For further analysis, we have to optimize the shape and the size of

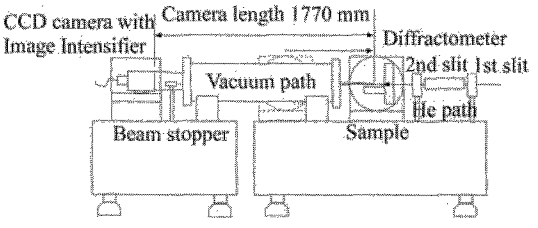


Fig. 6 Experimental setup of the GISAXS measurement.

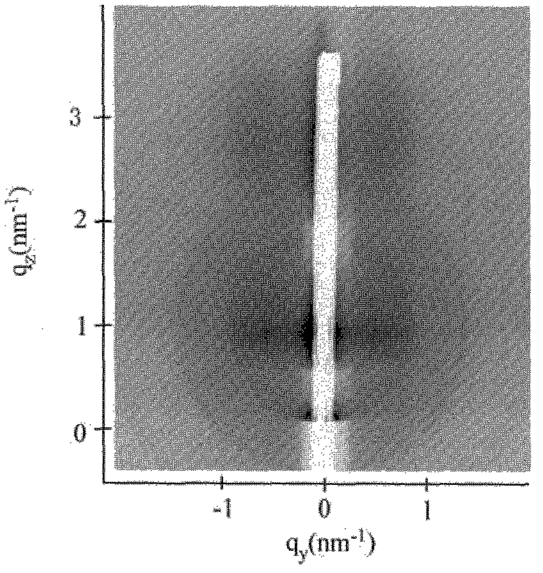


Fig. 7 GISAXS pattern from the MnPt thin film.

the stopper for the GISAXS measurement.

5. CONCLUSION

We demonstrated a performance of BL15 at Saga Light Source for an x-ray reflectivity measurement. In this study, the reflectivity ranging from 1 to 10^{-7} was measured with a SiO_2/Si sample. The thickness of SiO_2 film was 49.0 ± 0.15 nm, which was obtained by a least-squares analysis with a single layer model. We conclude that the optical and mechanical properties of the beamline BL15 are suitable for the analysis of thin films by the x-ray reflectivity measurement. Furthermore, we tried a GISAXS measurement. We obtained a scattering pattern from the periodic structure in a MnPt thin film. The details of the measurement system have to be refined for the GISAXS measurement to reduce the background. However, we confirm that the beamline has a potential to perform the GISAXS measurements.

The applications of the synchrotron x-rays to the surface and interface analysis have some advantages against a laboratory x-ray source. One is the tunability of the wavelength. By using an anomalous dispersion effect in XRD and SAXS measurements, element specific analyses can be performed. Moreover, a time-resolved analysis are possible using a multiwavelength-dispersive reflectometer with white X-rays [7].

Another advantage is brought by microbeam optics. With a recent development of the optical components as Kirkpatrick-Baez (KB) mirrors or a Fresnel zone plate, the beam size of the order of several tens nm was achieved with a reasonable photon flux. The use of microbeam will open the way to the local structural analysis of surface and interface [8].

Moreover, many experiments can be applied for the structural analysis of surfaces and interfaces such as XAFS, x-ray fluorescence analysis, XRD etc., within the same beamtime. By the complementary combination of these methods, we intend to estimate the structure and properties of thin films quickly and effectively.

Reference

[1] T. Tomimasu, S. Koda, Y. Iwasaki, H. Ohgaki, M. Yasumoto, T. Kitsuka, Y. Yamatsu, M. Mori, Y. Ochiai, Proc. PAC 2003, 902-904 (2003).
[2] Y. Iwasaki, S. Koda, T. Tomimasu, H. Ohgaki, M. Yasumoto, Y. Yamatsu, T. Kitsuka, Y. Hashiguchi, Y. Ochiai, Proc. PAC 2003, 3270-3272 (2003).
[3] T. Okajima, Y. Chikaura, M. Tabata, H. Hashimoto, Y. Soejima, K. Hara, N. Hiramatsu, Nucl. Inst. and Meth. B, 238, 185-188 (2005).
[4] K. Sakurai, A. Iida, Adv. In X-ray Anal., 35, 813-818 (1991).
[5] K. Sakurai, A. Iida, Jpn. J. Appl. Phys., 31, L113-L115 (1992).
[6] J. R. Levine, J. B. Cohen, Y. W. Chung, P. Georgopoulos, J. Appl. Cryst., 22, 528-532 (1989).
[7] T. Matsushita, Y. Niwa, Y. Inada, M. Nomura, M. Ishii, K. Sakurai, Appl. Phys. Lett. 92, (2008) 024103.
[8] K. Sakurai, M. Mizusawa, M. Ishii, S. Kobayashi, Y. Imai, J. Phys. Conference Series 83 (2007) 012001.

(Recieved December 10, 2007 ; Accepted February 23, 2008)