J. Synchrotron Rad. (1998). **5**, 554–556

A grazing-incidence reflectometer for BL-39XU at SPring-8

Kenji Sakurai,[a]* Shouji Uehara[b] and Shunji Goto[c]

[a]National Research Institute for Metals, 1-2-1 Sengen, Tsukuba, Ibaraki 305, Japan, [b]Kohzu Seiki Co. Ltd, 2-27-37 Mishuku, Setagaya, Tokyo 154, Japan, and [c]JASRI, SPring-8, Kamigori, Ako-gun, Hyogo 678-12, Japan.
E-mail: sakurai@yuhgiri.nrim.go.jp

(*Received 4 August 1997; accepted 12 November 1997*)

A new grazing-incidence reflectometer has been designed for the X-ray undulator beamline BL-39XU at SPring-8. It will be used for analytical applications, especially ultra-trace-level determination by total reflection X-ray fluorescence (TXRF), nanometer-scale surface topography by specular/non-specular X-ray reflection, and thin-film interface studies by combined measurements of X-ray fluorescence and X-ray reflection. The requirements for the instrument and the solutions in the present design are discussed. A prototype has been developed to test the feasibility of the design. It has been confirmed that the measurements planned at the beamline are achievable with this reflectometer.

Keywords: instrumentation; total reflection; grazing incidence; X-ray fluorescence; TXRF; specular reflection; non-specular reflection; diffuse scattering; ultra-trace-level analysis; surface topography; interface study.

1. Introduction

The recent remarkable progress in total reflection X-ray fluorescence (TXRF) and grazing-incidence X-ray reflectometry is significant for materials analysis, especially for ultra trace level determination and the characterization of thin films (Schwenke & Knoth, 1993; Stoev & Sakurai, 1998). One of the most widely known TXRF applications is the evaluation of surface contamination of semiconductor wafers; according to recent reports, trace determination of the order of 10^9–10^8 atoms cm^{-2} is now possible (Laderman *et al.*, 1995; Wobrauschek *et al.*, 1995). The chemical characterization of trace metals in a small sample, such as a drop of liquid, is also important in biological and environmental analysis (Sakurai, Ida & Shintani, 1997). Chemical shifts of absorption edges are detected by measuring the fluorescence intensity as a function of the incident X-ray energy. A mirror is used as a sample support to improve the signal-to-background ratio, and the chemical state of diluted metals (~0.1 m*M*) in a drop of liquid (~1 µl) can be determined. Both X-ray specular and non-specular (diffuse) reflection are powerful tools in nanometer-scale surface topography (Bowen & Tanner, 1993; de Boer, 1996), and when combined with angle-resolved TXRF measurements they form a unique probe for interface studies by means of the interference effect caused by multiple reflection of X-rays (Sakurai & Iida, 1997).

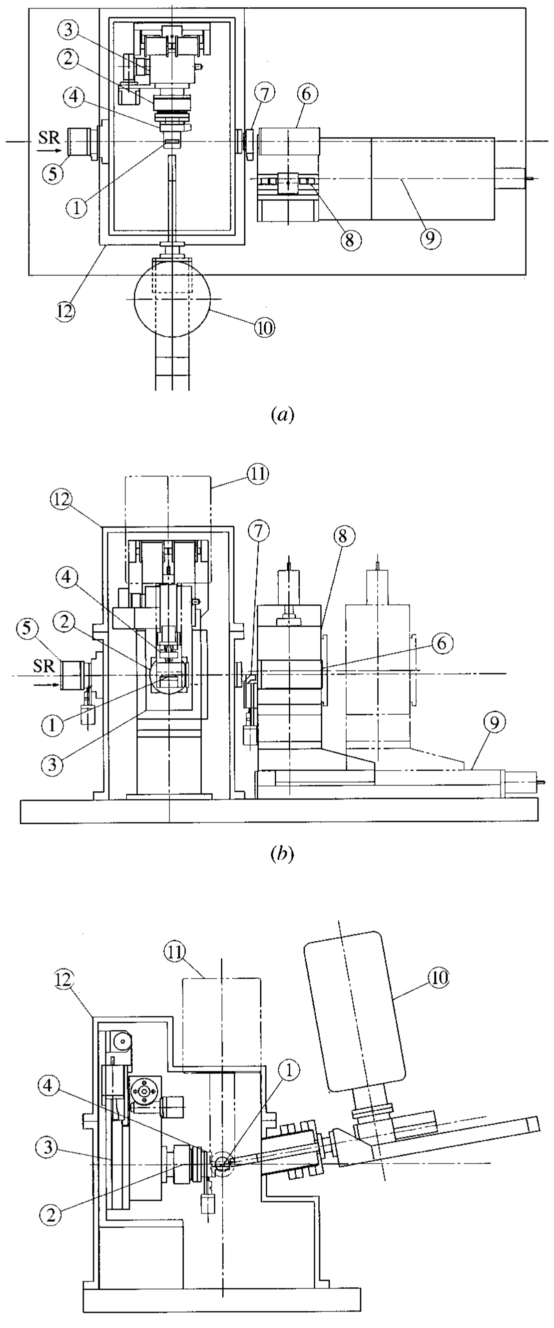
Third-generation synchrotron radiation, especially the extremely high photon flux available from an undulator, is expected to change the basic principles of sample characterization and result in novel techniques and instruments, besides increasing the potential of conventional analytical methods. The BL-39XU beamline (Japan Synchrotron Radiation Research Institute, 1997) at SPring-8 is one of ten public beamlines completed for initial operation in October 1997. This X-ray undulator beamline is equipped with an Si(111) rotated-inclined double-crystal monochromator and a Pt-coated mirror with a vertical axis. We have designed a new X-ray reflectometer for this beamline as part of the activities of the SPring-8 spectrochemical analysis collaborating group. The group is responsible for the development and installation of two different instruments: an X-ray fluorescence spectrometer for micro-imaging (Hayakawa *et al.*, 1997) and this grazing-incidence X-ray reflectometer. In this paper, the detailed design of the reflectometer and some results of the feasibility tests on a prototype are reported.

2. Design considerations

The design policy for the reflectometer is based on the scientific programme planned at the beamline. So far, a number of highly sophisticated instruments have been developed for grazing-incidence X-ray experiments, especially for surface diffraction techniques. Some have been equipped with many stages for multi-purpose measurements and others have been designed for use in an ultra-high vacuum to study well defined surfaces. The present reflectometer is a much simpler, but nevertheless carefully designed, instrument for ultra trace level analysis and realistic surface/interface study by combined measurements of TXRF and specular/non-specular X-ray reflection.

Figs. 1 and 2 show the X-ray reflectometer schematically. A high degree of accuracy and precision in angular scanning are essential. In the present design, a single-axis goniometer [(2) in Figs. 1 and 2] with 0.005 arcsec resolution (Ishikawa *et al.*, 1991) is used to rotate the sample. The resolution is chosen for conventional X-ray reflection/scattering experiments and for future grazing-incidence X-ray interference experiments using a partially coherent beam. For detector (6) scanning, an independent translational stage (8) is used instead of the second axis of the goniometer. Since the angle of incidence is shallow (typically of the order of mrad) straight motion gives exact angular change and high resolution is easily achievable because of the rather long distance from the goniometer centre (250–500 mm). The distance is controlled by the stage (9) and can be read by a magnetic scale. A crystal analyser is not employed to enhance the resolution because a distance of 500 mm can provide good enough resolution for our experiments. Precise alignment capability is extremely important for the grazing-incidence X-ray reflectometer; the goniometer rotation centre and sample surface must be set on the exact centre of the incident beam. Two linear stages with 0.25 µm resolution are used for this alignment; one, (3), for positioning the goniometer and the other, (4), for the sample (1). Although the automatic scan range of the goniometer is limited to 6°, it is designed to allow manual 180° rotation. The beam centre can be detected easily, *e.g.* by scanning a very thin wire, and the rotation centre position can be corrected so that the difference between the 0 and 180° positions becomes zero. For reliable positioning of the sample surface, a direct beam stopper [a knife edge, (7)] is required just in front of the reflection detector. At maximum reflection intensity the sample surface is exactly on the beam centre and the angle of incidence is just below the critical angle.

This reflectometer is also intended for ultra trace level analysis by TXRF. It is important to obtain a good signal-to-scatteringbackground ratio and also to reduce fluorescent X-rays from the environment (outside the sample). In the present design, a low-pressure chamber (12) is prepared to reduce air scattering and to avoid contamination from airborne particles. An oil-free pump is used for evacuation. A Teflon surface finish is added to every part around the sample and the X-ray fluorescence detector [(10) or (11)]. The sample holder is fixed on a small goniometer which allows manual orientation adjustment to avoid possible diffraction by a single-crystal sample. The distance between the sample and the detector (10) can be changed from 0 to 345 mm. The detector is equipped with a collimator to limit the viewing area, which is useful in avoiding the influence of scattering at the edges of the sample. It is also essential to be able to vary the sample orientation by switching the rotation axis between horizontal and vertical, as wet, liquid or small samples need horizontal axis geometry (Fig. 1), but vertical axis geometry (Fig. 2) is typically advantageous in the analysis of semiconductor wafers, as it makes full use of the linear polarization of undulator radiation to reduce the scattering further. In horizontal axis geometry, the fluorescence detector is usually set at 10° inclination ('side view' geometry) as shown in Fig. 1(*c*), but a 'top view' layout using a direction-convertible detector is also possible. This is sometimes important when collecting fluorescence signals from extremely small particles on the substrate. The upper part of the chamber can be removed so that different chambers or equipment can be connected. Initially, the roof is a transparent plastic plate to allow internal inspection of the chamber. It is also possible to introduce gas (He, dry N_2 *etc.*) for biological and materials research.



(*a*)

(*b*)

(*c*)

Figure 1
Design drawings for the reflectometer (horizontal axis geometry): (*a*) plan view; (*b*) side view (without fluorescence detector); (*c*) section view. (1) sample; (2) goniometer; (3) stage for positioning the goniometer centre; (4) stage for positioning the sample surface; (5) incident X-ray monitor with entrance slit; (6) reflection detector with receiving slit; (7) direct beam stopper and the stage; (8) stage for reflection detector scan; (9) stage for adjusting angular resolution; (10) fluorescence detector; (11) direction-convertible fluorescence detector; (12) vacuum chamber.

In the present X-ray reflectometer, a high-resolution X-ray fluorescence detector is needed. Initially, an Si(Li) detector was used, but in the future a superconductor tunnelling junction detector (Kurakado *et al.*, 1997) of a crystal analyser may be used instead. However, for X-ray reflection a wide dynamic range is important and an ionization chamber or an APD detector is used. The incident intensity is monitored by an ionization chamber [(5) in Figs. 1 and 2].

3. The prototype and feasibility tests

The prototype reflectometer was developed at the National Research Institute for Metals, Tsukuba, Japan. No vacuum chamber was prepared because the main aim was to test the mechanical motion of the reflectometer and the precision and feasibility of real measurements. Only the vertical axis geometry (see Fig. 2) was tested with the Si(Li) detector in the 'side view' position, but, in principle, the specification of the reflectometer is very close to the design for the beamline discussed above. The alignment procedure was as follows. The reflectometer was connected to a laboratory X-ray source (45 kV, 80 mA, 30 μm × 3 mm focus) and an Si(111) channel-cut monochromator was installed on the tube wall. The size of the entrance and receiving

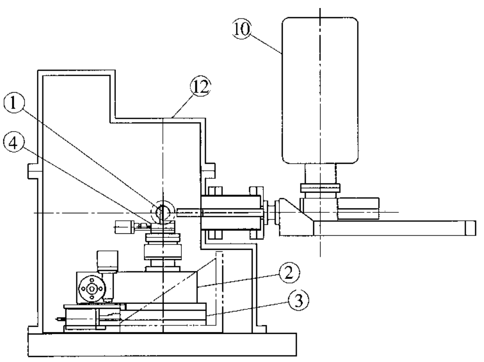


Figure 2
Design drawings for the reflectometer (vertical axis geometry, section view). For key see Fig. 1.

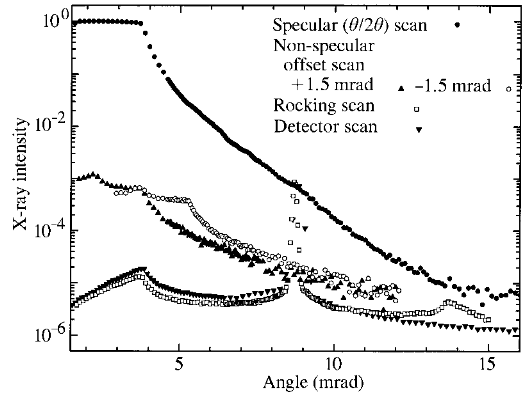


Figure 3
Specular and non-specular reflection for synthetic quartz mirror material using different scans [specular ($\theta/2\theta$), quasi-longitudinal (offset), transverse (rocking) and radial (detector) scans] using the prototype reflectometer with a laboratory source.

slits was 40 µm (width) × 10 mm (height). The distance between the goniometer centre and the receiving slit was set to 285 mm. A YAP:Ce scintillation detector was used for X-ray reflection measurements.

Fig. 3 shows the specular and non-specular (diffuse) reflection from synthetic quartz, which is commercially available as mirror material ($\lambda/20$, $\lambda = 632.8$ nm; 2 arcsec). While specular reflectivity is sensitive to changes in the electron density with depth, non-specular reflection provides information in the lateral direction. Every scan was carried out successfully, and it was found that the experimental data obtained give reasonable surface parameters such as roughness, correlation length and fractal dimension (Sakurai & Stoev, 1998). In this case, the surface is expected to be randomly rough, but the step structure and initial or intermediate stages of island growth of thin films constitute other interesting models. Furthermore, when the experiment is extended to thin films, the most interesting and important target is the interface, and different models are required for analysis. The prototype reflectometer worked well in preliminary studies.

The advantage of an undulator radiation beamline for TXRF measurements is clear. By using the interference effects in the X-ray fluorescence intensity, it is possible to enhance the segregated trace impurities localized at a specific interface (Sakurai & Iida, 1997); this will be one of the main future programmes at the beamline. Another interesting possibility is the separation of true roughness (physical shape) and the degree of chemical gradation for each interface. Although it is difficult to distinguish them by only X-ray specular reflection, which detects changes in electron density, the combined measurement of non-specular X-ray reflection and fluorescent X-rays from the interface is promising as non-specular X-ray reflection is observed when the interface is physically rough, and does not depend on chemical gradation (Sinha *et al.*, 1988), while X-ray fluorescence is sensitive to chemical composition, even at the trace level. When measurements were attempted using the limited intensity of a laboratory source (Sakurai & Stoev, 1998), it was confirmed that the present design fits well with the planned experiments.

4. Conclusions

A grazing-incidence X-ray reflectometer has been designed for analytical use at BL-39XU, SPring-8. The feasibility of the design has been successfully confirmed by testing the prototype. We are now considering additional designs for sample handling and transfer. Since ultra trace level element studies are the main objectives of this beamline, possible contamination prior to the measurements can be a big problem. To achieve much better detection limits than with a second-generation synchrotron radiation source, a clean, versatile instrument will be needed. Controlling small quantities of liquid drop samples is also essential and we plan to install an instrument for producing drops inside the chamber. One of the advantages of the present reflectometer is that it is relatively easy to add or change equipment to suit each experiment.

The authors are grateful to all members of the SPring-8 spectrochemical collaboration group for valuable discussions. Our thanks also go to Dr K. N. Stoev and Dr L. Orteaga at the National Research Institute for Metals for useful comments. Part of the work, including the development of the reflectometer prototype, received financial support from the Science and Technology Agency, Japan, through the nuclear energy research program.

References

Boer, D. K. G. de (1996). *Phys. Rev. B*, **53**, 6048–6064.
Bowen, D. K. & Tanner, B. K. (1993). *Nanotechnology*, **4**, 175–182.
Hayakawa, S., Goto, S., Shoji, T., Yamada, E. & Gohshi, Y. (1997). *J. Synchrotron Rad.* **5**, 1114–1116.
Ishikawa, T., Hirano, K. & Kikuta, S. (1991). *Nucl. Instrum. Methods*, A**308**, 356–362.
Japan Synchrotron Radiation Research Institute (1997). *SPring-8 Beamline Handbook.* Version 1.1, p. 20.
Kurakado, M., Ohsawa, D., Katano, R., Ito, S. & Isozumi, Y. (1997). *Rev. Sci. Instrum.* **68**, 3685–3696.
Laderman, S. S., Fischer-Colbrie, A., Shimazaki, A., Miyazaki, K., Brennan, S., Takaura, N., Pianetta, P. & Kortright, J. B. (1995). *Anal. Sci.* **11**, 515–518.
Sakurai, K. & Iida, A. (1997). *Adv. X-ray Anal.* **39**, 695–700.
Sakurai, K., Iida, A. & Shintani, H. (1997). *J. Phys. IV (France)*, C**2**, 713–714.
Sakurai, K. & Stoev, K. (1998). In preparation.
Schwenke, H. & Knoth, J. (1993). *Handbook of X-ray Spectrometry*, edited by R. E. Van Grieken & A. A. Markowicz, p. 464. New York: Marcel Dekker.
Sinha, S. K., Sirota, E. B., Garoff, S. & Stanley, H. B. (1988). *Phys. Rev. B*, **38**, 2297–2311.
Stoev, K. N. & Sakurai, K. (1998). *Spectrochim. Acta B.* In the press.
Wobrauschek, P., Kregsamer, P., Ladisich, W., Streli, Ch., Pahle, S., Fabry, L., Garbe, S., Haller, M., Knöchel, A. & Radtke, M. (1995). *Nucl. Instrum. Methods*, A**363**, 619–620.