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Ultrahigh-Vacuum Facility for High-Resolution Grazing-Angle X-ray Diffraction at a Vertical Wiggler Source of Synchrotron Radiation

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A versatile ultrahigh-vacuum chamber has been designed for grazing-angle X-ray standing-wave and diffraction experiments at the vertical wiggler source of the Photon Factory. Unlike at other sources, the vertically polarized X-rays from the wiggler favour the use of a horizontal scattering geometry. The X-ray chamber is equipped with a hemispherical beryllium window, which allows any scattering angle to be attained and secondary emissions to be measured. The chamber is of a compact design, sitting on a precision rotary table which is rotated for scans. Samples are introduced from a portable vessel. The whole procedure can be performed in a vacuum better than 10^{-7} Pa. The system has successfully been applied to a grazing-angle X-ray standing-wave experiment, which determined the dimer bond length and the domain structure of Si(001) surfaces deposited with monolayer arsenic.

Keywords: ultrahigh-vacuum chambers; grazing-angle X-ray standing waves; surface X-ray diffraction; arsenic-deposited Si(001) surfaces; vertical wigglers.

1. Introduction

Diffraction and scattering of X-rays in grazing-angle geometry at synchrotron radiation sources are powerful tools for investigating atomic structures of surfaces. A number of diffractometers with ultrahigh-vacuum chambers have been designed for surface studies. These can be classified into two types: facilities with growth devices, such as effusion cells and evaporators, and the associated surface characterization devices, and those without. Those which do not have growth devices are compact, but only allow the investigation of rather stable surfaces, which are prepared in a separate vacuum chamber and transported to the X-ray chamber. This type of chamber was first used by Eisenberger & Marra (1981) in a study of a Ge(001) 2×1 surface. Actually, this was not so compact since two sputter-ion pumps were used. A rather compact chamber, 15 kg in weight, was designed and used to measure an InSb(111) 2×2 surface by Bohr *et al.* (1985).

Facilities with growth devices can be further classified into two types: those which include a feedthrough mechanism, and those that do not. A facility without the feedthrough mechanism rotates the whole chamber and has a fixed sample position relative to the chamber. This has the advantage that *in situ* measurements can be readily made. However, the goniometer is heavy and large in proportion to the vacuum chamber, making the entire system considerably large. A typical system was designed by Brennan & Eisenberger (1984). The alternative approach allows the feedthrough mechanism in a facility to adjust the sample orientation in a vacuum very precisely. This means that the equipment can be smaller and more general. One example is a design by Fuoss & Robinson (1984). The feedthrough uses bellows and a differentially pumped rotating seal. A sample can be carried to the preparation and analysis devices in the system and shifted to a position near a beryllium window for X-ray measurements. The basis of this design has been adopted on many different beamlines at conventional wigglers and bending-magnet synchrotron sources for measurements using the vertical scattering geometry (Norris *et al.*, 1986/1987; Vlieg *et al.*, 1987; Sauvage-Simkin *et al.*, 1989; Ferrer & Comin, 1995; Takahasi *et al.*, 1996). The techniques used are chiefly surface X-ray diffraction (SXD) and crystal-truncation rod measurements, as reviewed by Feidenhans'l (1989) and Robinson (1991). At the same time, ultrahigh-vacuum facilities dedicated to X-ray standing-wave (XSW) measurements were designed at the DORIS (Funke & Materlik, 1985) and NSLS (Zegenhagen & Patel, 1990) synchrotron sources. Both facilities are equipped with sample preparation devices and measurements are performed by changing the pass energy of monochromators driven by piezo mechanisms. The application of these instruments and others has been reviewed by Zegenhagen

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(1993). There is no facility, however, to the authors' knowledge, that allows both SXD and XSW measurements, except the baby chamber designed by Bohr *et al.* (1985).

Sakata & Hashizume (1995*a*) fabricated a compact chamber for grazing-angle XSW (GAXSW) experiments on beamline 14B at the Photon Factory using vertically polarized X-rays and horizontal scattering geometry. The facility achieved a high accuracy and stability; it was successfully applied to the determination of the in-plane position and the ordering of an arsenic-adsorbed Si(111) surface (Sakata *et al.*, 1993; Sakata & Hashizume, 1995*b*). This success encouraged us to design a new chamber enabling both GAXSW and SXD measurements on this same beamline. The combination of SXD and GAXSW is useful for detailed studies of surface structures with a high crystalline perfection. GAXSW allows model-independent analysis of the structure of foreign surface atoms, the results of which would make the interpretation of SXD data easier and more accurate. This paper reports a versatile ultrahigh-vacuum X-ray chamber designed to this end, as well as the associated sample transportation and introduction system which is used for sample transfer between a molecular-beam-epitaxy facility at our institute and the X-ray chamber at the Photon Factory. We have already reported an outline of this design, before a performance test was conducted (Sakata *et al.*, 1995).

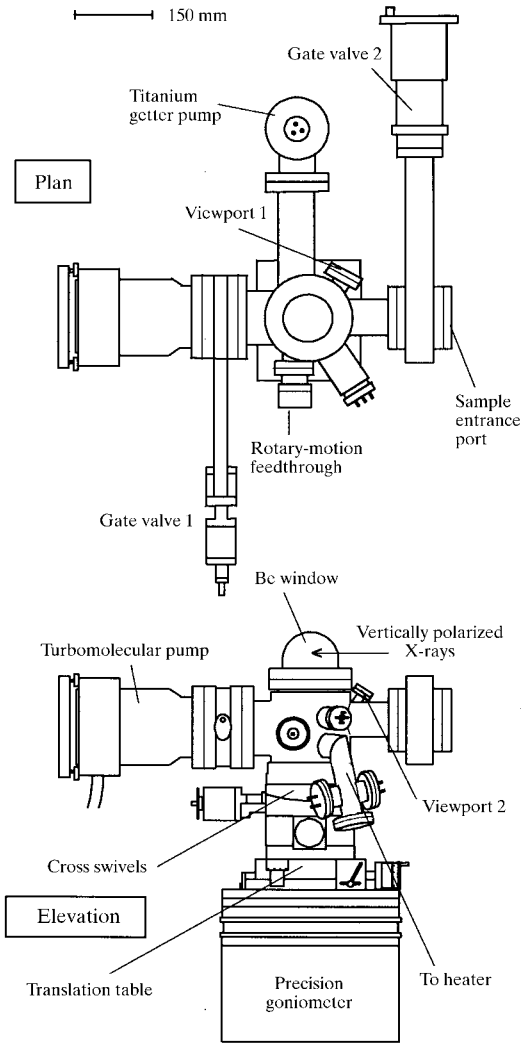

Figure 1
Ultrahigh-vacuum X-ray chamber designed for grazing-angle X-ray standing-wave and surface X-ray diffraction measurements at a vertical-wiggler synchrotron source.

2. Design of an ultrahigh-vacuum X-ray chamber and a sample transfer system

Two prerequisites of the ultrahigh-vacuum X-ray chamber, which can be utilized for SXD and GAXSW measurements, are given in (*a*) and (*b*) below. Also included are the design options we chose to fulfil these conditions.

(*a*) It is desirable to measure intensities from as many reflections as possible for SXD. In addition, we need to collect fluorescence X-rays near a sample surface for GAXSW measurements. A small hemispherical beryllium dome has been adopted as a window for X-rays to satisfy these two requirements. Bohr *et al.* (1985) used a cylindrical beryllium window in their portable X-ray chamber.

(*b*) GAXSW measurements demand accurate rotation of a sample near Bragg positions. To achieve this, a small X-ray chamber is mounted on a precision rotary table and the whole chamber is rotated around the vertical axis of the table.

The main body of the X-ray chamber is 130 mm in inner diameter and 195 mm in height (Fig. 1). The hemispherical Be dome with an extended base, 104 mm in diameter and 0.5 mm in thickness, is attached to the CF-152 conflat flange at the top of the body. A sample is located at the centre of the Be dome almost in the horizontal plane with its surface upward. Incident and diffracted X-rays are approximately in the horizontal plane as well. The X-ray chamber is evacuated to 1×10^{-8} Pa with a $300\ \mathrm{l\ s^{-1}}$ magnetically levitated turbomolecular pump and a $300\ \mathrm{l\ s^{-1}}$ titanium getter pump. The turbomolecular pump has small vibration compared with standard types, and is much lighter than a sputter-ion pump with the same speed of evacuation. Reduced vibration is important in GAXSW measurements, which require very precise angular control. The two pumps are located below the X-ray path so as not to block the beam. A $250\ \mathrm{l\ min^{-1}}$ rotary pump, which is connected to the turbomolecular pump *via* a 4 m flexible tube, is placed on the roof of the beamline hutch (not shown in Fig. 1). An all-metal gate valve is attached at the entrance flange for sample introduction while a viton gate valve is used in front of the turbomolecular pump. The entire chamber is bakeable to 393 K. The pumps and the gate valves are arranged in well balanced positions. The X-ray chamber has a mass of 45 kg including the pumps and the gate valves. Two viewports help us to introduce and remove samples.

The chamber, placed on crossed swivels which cover an angular range of $\pm 10^{\circ}$, is mounted on a precision rotary table (Fig. 1). The swivels and the rotary table are driven by five-phase pulse motors and are rotated by 1.21, 1.03 mrad

and 2.42 μrad per 100 pulses, respectively. The centre of rotation of the swivels is positioned 10 mm below the sample surface. A rotation of the sample changes the surface height by 1.5 μm. This height change can be accommodated by moving vertical slits to account for the offsets. This is necessary because the different reflections require different swivel angles. The uniformity of the incident beam allows the slits to be moved with a negligible flux variation at the sample. No slit control is made in practice, however, because a fairly large vertical aperture is used.

Surface X-ray diffraction measurements on a conventional four-circle diffractometer are made using the φ-scan mode at a fixed incident angle. A rotation around the precision rotary table axis in our instrument is a good approximation to a φ scan if the scan range is as small as several degrees. For wider-angle scans at a fixed incident angle, we can simulate the φ scan using a combined rotation of the table and swivel axes.

When setting up, the axis of the rotary table and the centre of rotation of the crossed swivels must be aligned. A translation table is used for this purpose, which moves the chamber in one direction in the horizontal plane. Movement in the same plane, but in a perpendicular direction, is allowed by a small amount of clearance in the lower swivel fixing screws.

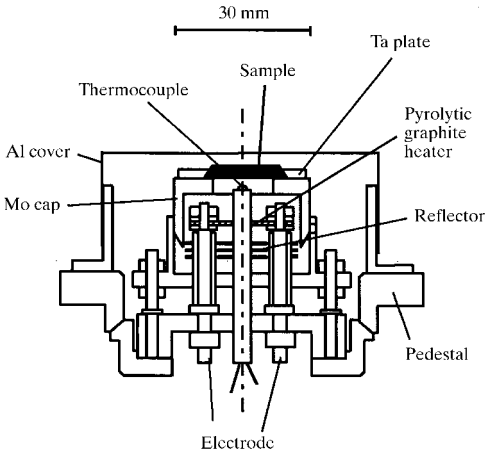

Figure 2
Sample holder with heater. The Al cover is removed when heating is not conducted during X-ray measurements.

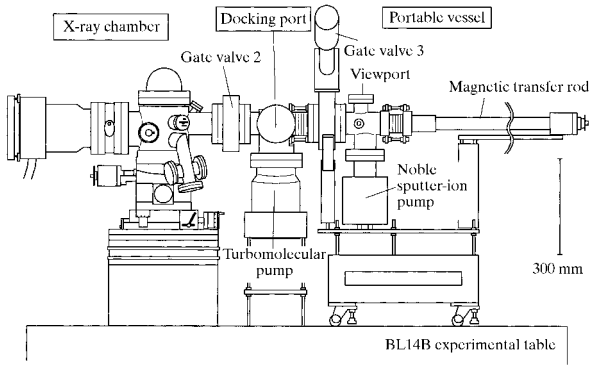

Figure 3
An X-ray ultrahigh-vacuum chamber, a docking port and a portable vessel assembled on the experimental table of beamline 14B at the Photon Factory for sample introduction or removal.

Fig. 2 shows a sample holder, which receives a molybdenum cap with a central bore (14 mm diameter) into a bayonet socket. The sample, about 16 × 18 × 2 mm, has a pair of bevelled edges. Two tantalum plates clamp the sample down to the outer surface of the Mo cap. Their upper surfaces are lower than the sample surface so that the X-rays are not blocked. The Mo cap has two sets of three lugs on its side surface; the lower three mate to the bayonet slots on the holder and the upper three mate to the transfer rod. The sample can be heated from the back by a pyrolytic graphite ohmic heater located 7.2 mm behind. Pyrolytic boron nitride coats the heater to suppress outgassing. The design allows the sample temperature to reach 1373 K with a maximum 700 W load applied to the heater. Using a 70 W load, a sample-surface temperature of 1103 K was measured with a pyrometer through viewport 1, while the sample was in the introduction position. A 15 μm-thick aluminium cover is inserted between the sample holder and the Be window to keep the inside surface of the window clean while a heating measurement is conducted. We do not use the cover for surface X-ray diffraction measurements since it may be polycrystalline and produce powder diffraction.

The sample was prepared in our home institute (TITech) using a molecular-beam-epitaxy (MBE) facility (Sakata *et al.*, 1990). A growing surface structure is monitored using reflection high-energy electron diffraction (RHEED), low-energy electron diffraction (LEED) and Auger electron devices. The base pressure of the MBE chamber is

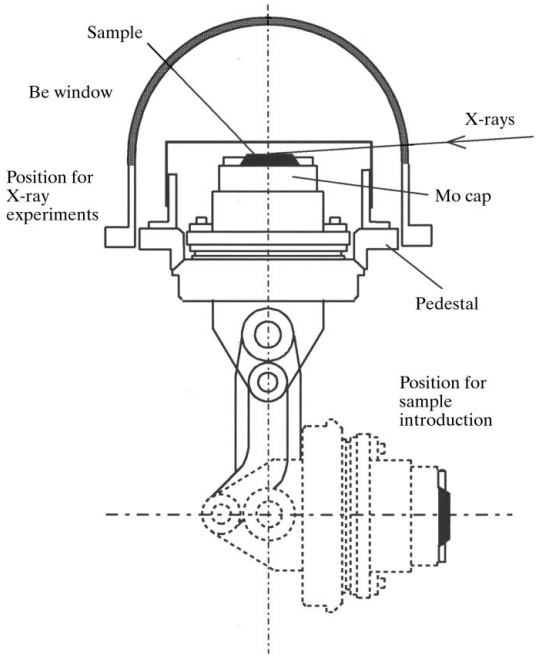

Figure 4
Cam mechanism for transfer of a sample between the two positions in the X-ray chamber.

1×10^{-8} Pa. A prepared sample is loaded into a small UHV portable chamber equipped with a 680 mm magnetic transfer rod. This vessel is 1040 mm in total length and 15 kg in weight and the chamber is evacuated to a pressure of 1×10^{-7} Pa by a 16 l s^{-1} noble sputter-ion pump, which is powered by a car battery. The sample-mounted Mo cap is in the bayonet socket at the end of the transfer rod. Two plates, which are located at the far end of a linear-motion feedthrough inside the vessel, support the cap during transportation by car. The sample is introduced from the portable vessel to the X-ray chamber *via* a docking port on the experimental table of the beamline at the Photon Factory, as shown in Fig. 3. During this operation the sample is sideways with the surface in a vertical position and the pressure remains better than 1×10^{-7} Pa. The all-metal gate valve is closed after introduction and then the X-ray chamber is separated from the other chambers. A cam mechanism, driven by a manually operated rotary-motion feedthrough, changes the sample orientation by 90° as it lifts the sample to the level of the X-ray beam (Fig. 4).

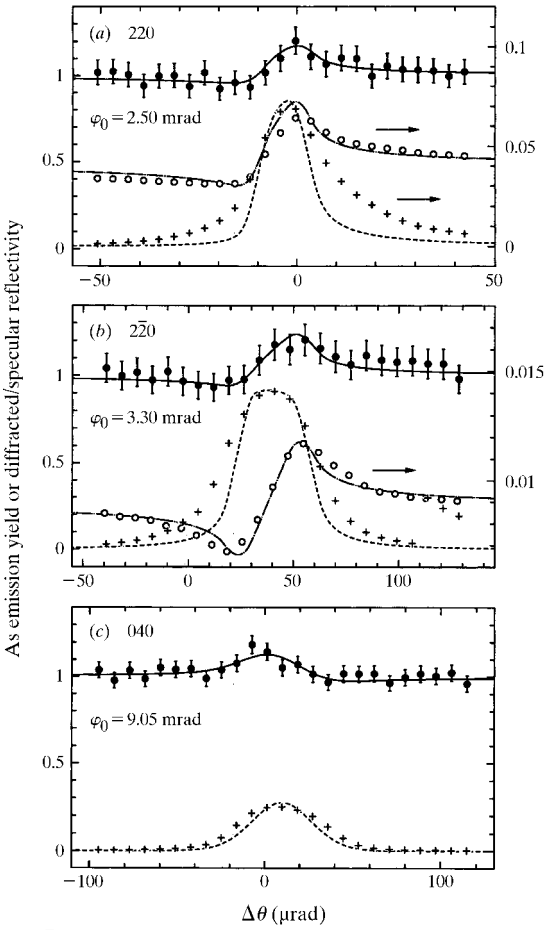

Figure 5
As fluorescence signals (closed circles), specular fluxes (open circles) and diffracted fluxes (crosses) observed from an As/Si(001) sample in grazing-angle diffraction for (*a*) 220, (*b*) 2$\bar{2}$0 and (*c*) 040 reflections in ultrahigh vacuum. Average As emission counts in 500 s are 650 in (*a*), 500 in (*b*) and 1400 in (*c*). The glancing-incidence angles used are indicated (φ_0). Lines show least-squares fits.

The pedestal reproducibly locates the sample in the X-ray position.

3. Performance test

The system was used for data collection from an arsenic-adsorbed Si(001) surface on beamline 14B at the Photon Factory synchrotron source. Vertically polarized light from the 5 T vertical wiggler was monochromated by the double Si(111) beamline monochromator. The Photon Factory storage ring was injected with 2.5 GeV positrons at a 350 mA current.

The sample surface was mis-cut by 4° away from the (001) surface towards the [1$\bar{1}$0] surface. When As is deposited on such a surface, area fractions M_1 and M_2 of two superlattice domains depend on growth conditions, as reported by Bringans *et al.* (1991). A sample was prepared in TITech using our MBE facility. RHEED and LEED observations revealed mixed 1×2 and 2×1 domains, in which As—As bonds are perpendicular and parallel to step edges, respectively, with slightly brighter 2×1 spots. We transferred the sample using the portable UHV vessel over a ~100 km distance to the X-ray chamber placed at beamline 14B.

An Si(220) monochromator was used in the beamline hutch for GAXSW measurements. The critical angle for total external reflection was 1.83 mrad for the wavelength of 0.73 Å used. Fig. 5 shows the observed (and normalized) As emission, together with the specular and diffracted intensities. All three spectra were recorded simultaneously as the sample was scanned across the Bragg positions ($\Delta\theta$ = 0) for the 220, 2$\bar{2}$0 and 040 reflections. The sample emission was resolved with an energy-dispersive high-purity germanium detector, while NaI detectors were used to record specular and diffracted intensities. The net As fluorescence integrated over a small energy range of about 400 eV around the 10.5 keV As *K* line was 1–3 photons s^{-1} when the illuminated area on the sample surface was 1 mm in width and about 16 mm in length.

Expressions for As emission include the structure parameters, M_1, M_2, and the As—As bond length, L (Sakata *et al.*, 1995). Results of least-squares fits of the As emission data in Fig. 5 indicate L = 2.55 (5) Å, M_1 = 0.36 (11) and M_2 = 0.48 (11), with the remaining atoms being randomly distributed (Sakata *et al.*, 1997). The calculations take into account the beam divergence angles, $\delta\theta$ = 11.5 μrad for the 220 and 2$\bar{2}$0 reflections, non-dispersive with the monochromator, and $\delta\theta$ = 38.5 μrad for the 040 reflection, dispersive with the monochromator. The As—As bond length obtained is in agreement with the documented value (Jedrecy *et al.*, 1990), while the area fractions differ from the reported value (Bringans *et al.*, 1991). Growth conditions of the arsenic monolayers, such as substrate temperature, may account for this difference.

Owing to the limited synchrotron beam time, SXD measurements had to be performed in a separate run using a newly prepared sample. An Si(111) monochromator was

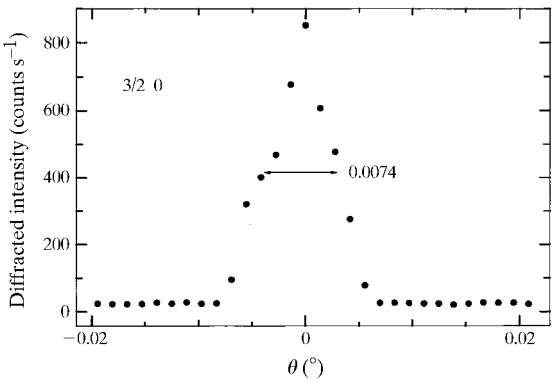

Figure 6
Rocking curve from a fractional-order reflection of an As/Si(001) sample in ultrahigh vacuum at a grazing angle of 15.9 mrad.

employed in the beamline hutch for this experiment. Using a wavelength of 1.19 Å, the critical angle was 2.98 mrad. Fig. 6 shows a rocking curve for the 3/2 0 in-plane peak, expressed using a surface lattice system, observed from an arsenic-deposited Si(001) surface, at a glancing angle of 15.9 mrad. The reflection has indices 3/2 3/2 0, expressed in a bulk cubic lattice system. It originates from the reconstructed Si(001):As surface. We note the following features of the rocking curve: the profile is not Lorentzian in shape due to strain or disorder which was not apparent in the LEED observation; the angular width (0.0074°) corresponds to a correlation length of 3200 Å, which indicates that the surface has a large domain. The sample was carried in vacuum back to our molecular-beam-epitaxy facility after the X-ray measurements. Observed LEED patterns were similar to those observed before the measurements.

In conclusion, we have built a compact and stable system for grazing-angle X-ray standing-wave and surface X-ray diffraction experiments with samples in an ultrahigh-vacuum environment.

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