Development of a Scanning Tunneling Microscope for *In Situ* Experiments with a Synchrotron Radiation X-ray Microbeam

An attempt to combine scanning tunneling microscopy (STM) with X-rays appears attractive because it contains various possibilities for original and important applications. Inner-shell excitation of a specific level under STM observation provides a possibility to analyze elements or to control local reactions with the spatial resolution of STM. However, the number of studies to combine STM with X-rays is quite limited. This may be because of difficulties specific to X-rays. To overcome a small efficiency of core-excitation by X-rays, we installed an STM system at beamline **BL19LXU** that is providing the world's-highest brilliance of X-rays from a 27 m long undulator, and focused the beam two-dimensionally to increase the photon density. To solve problems derived from the high brilliance (thermal and electrical noise, damage around the STM scanner, instability of the system such as thermal drift, etc.), we developed a special STM system dedicated to the *in situ* observation under the irradiation of X-rays from synchrotron radiation (SR)[1].

Our strategy is not to collect the emitted electrons that come from a wide irradiated area and can damage the spatial resolution of the analysis. It is essential to acquire the tunnelling signal to obtain the locality, thus we need to measure a tip current modulation caused by core-excitation under the tunneling condition, by avoiding collection of the widely emitted electrons. The technical point is how to adjust the incident X-rays as small as possible at the observation point of STM in order to avoid the excessive irradiation. The *in situ* STM measurements were enabled by developing an accurate alignment system (Fig. 1): the small X-ray beam of ϕ 10 μm in diameter was aligned at the STM observation point with accuracy of 1 μm in ultra-high-vacuum. A system to monitor the STM tip and sample in the X-rays was developed to realize a prompt alignment. In order to obtain high signal to noise (S/N) ratio and record a small signal, we developed a detection system of the tunnelling signal using an optical chopper and lock-in amplifier [2]. Also a special tip coated by an insulator (Fig. 2) was developed using focused ion beam (FIB) etching to remove the electrons coming from wide irradiated area [3].

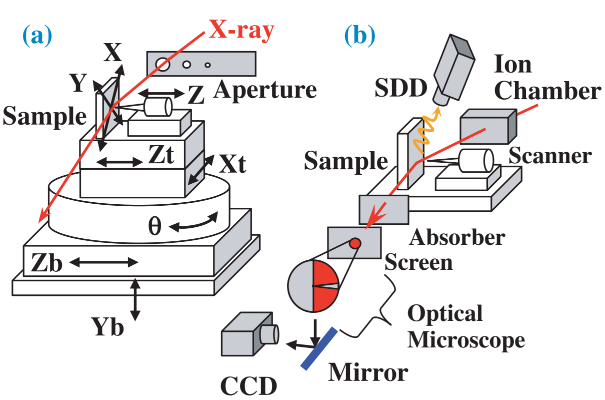

Fig.1. Schematic view of **(a)** alignment and **(b)** monitoring system. Under a conventional STM scanner, an independent four-axis stage is set, whereas the height Yb is controlled by an air dumper.

Despite the noisy condition and radiation load around the tip, STM images were successfully obtained with atomic resolution. After thermal noise was effectively removed, the tip current modulation was successfully obtained on Ge nanoislands on a clean Si (111) surface, by changing the incident photon energy across the Ge absorption edge. Figure 3 shows the topographic (left) and beam-induced current images (right), which were obtained simultaneously, at higher (11.114 keV) and lower (11.090 keV) energies than the Ge absorption edge. The tip current image at higher photon energy shows the Ge island darker than the surrounding area. The emitted electrons cannot explain the decrease in current on the Ge island, because the emitted electrons must increase the current on the Ge island, in which the Ge atoms are richer than in the surrounding area. The decrease of the tip current on the Ge island was not observed at lower photon energy, which indicates that the dark Ge island is not an effect of the modulated tunneling gap that is based on the tip motion. The above results indicate that the difference between on and out of the Ge island was detected across the absorption edge with spatial resolution of STM. In consideration of the cross-sectional profile in Fig. 3(e), the spatial resolution in this analysis is estimated to be ~ 4 nm.

After the first successful elemental identification at a nanometer scale for a semiconductor hetero-interface (Si(111)-Ge nanoisland), we succeeded in obtaining a second result stably on another sample, which is composed of metal and a semiconductor (Cu nano-domain on a Ge(111) clean surface) [4]. The spatial resolution was found to be ~2.5 nm by the same estimation as shown in Fig. 3(e). These results mean that the "single nanometer scale" elemental analysis was successfully achieved for "two systems" that have essentially different composition from each other, even though both of them have "extremely dilute" surface atomic concentration (total amount of the deposited element is ~0.3 monolayer for both).

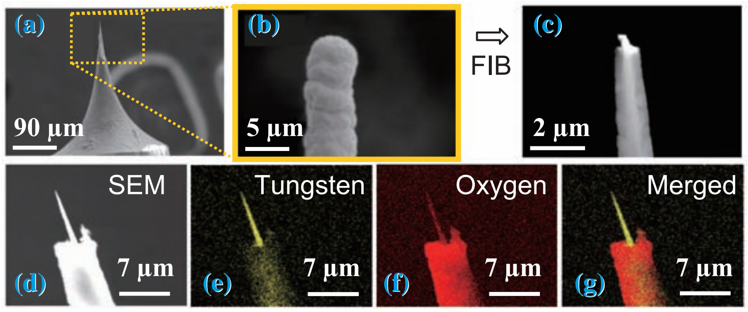

Fig. 2. **(a)**~**(g)** SEM images of the fabricated insulator coat tips. [upper] SEM images before **(a,b)** and after **(c)** FIB etching. [lower] SEM **(d)** and EDX images to distinguish the tungsten core **(e)** and coated SiO_2 film **(f)**. Merged image is shown **(g)**.

Practically, this STM system was found to serve as a realistic tool for element-identification. For example, our STM system enables clear recognition of a boundary between Cu and Ge nano-domains on a coarse surface terrace [5], which was roughly treated and rather common than well-defined surfaces. Such boundary is generally difficult to distinguish by the conventional STM because of the roughness. This SR-based STM study will also provide new information on basic science by investigation on an interaction of surface atoms and the hard X-rays. Also this system will serve to local reaction control with STM resolution.

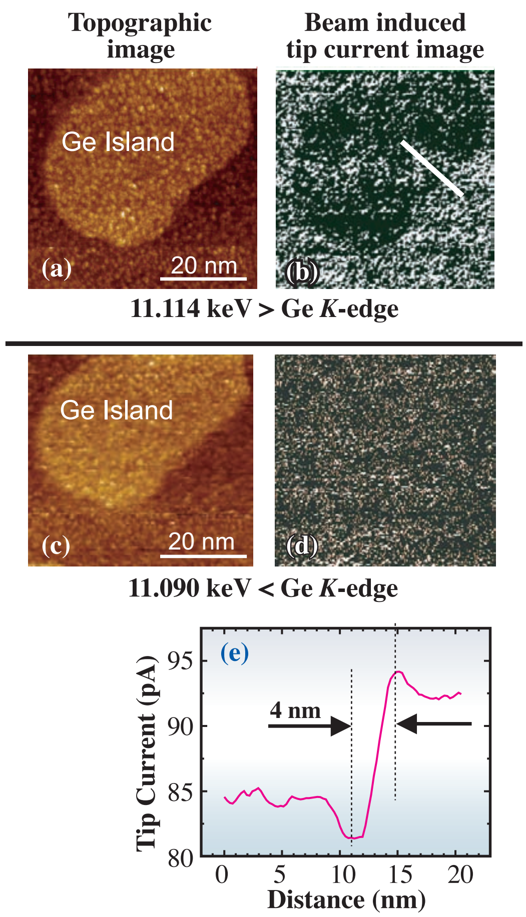

Fig. 3. Energy dependence of tip current images **(b, d)** and simultaneously obtained conventional topographic STM images **(a, c)**. (50 × 50 nm^2, -2 V, 0.5 nA for all). Cross-sectional profile along white line in **(b)** is shown in **(e)**.

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References

[1] *A. Saito, J. Maruyama, K. Manabe, K. Kitamoto, K. Takahashi, K. Takami, Y. Tanaka, D. Miwa, M. Yabashi, M. Ishii, Y. Takagi, M. Akai-Kasaya, S. Shin, T. Ishikawa, Y. Kuwahara, M. Aono: J. Synchrotron Rad.* ***13*** *(2006) 216; Chap. 27 in "Handbook of Surface Phys. and Engineer.," Maruzen Inc., Tokyo (2007).*
[2] A. Saito *et al.*: Jpn. J. Appl. Phys. **45** (2006) 1913.
[3] A. Saito *et al.*: Surf. Sci. **601** (2007) 5294.
[4] A. Saito *et al.*: J. Surf. Sci. Soc. Jpn. **28** (2007) 453; A. Saito *et al.*: Surf. Interface Anal. **40** (2008) 1033.
[5] A. Saito *et al.*: Chap. 4.2 in "Nano-Imaging," NTS Inc., Tokyo (2008) p.278.