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Verification of thermal effect produced by irradiation for scanning tunneling microscope combined with brilliant hard X-rays from synchrotron radiation

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ABSTRACT

A scanning tunneling microscope (STM) dedicated to *in-situ* experiments under the irradiation of highly brilliant hard X-rays of synchrotron radiation (SR) can serve a variety of original scientific works. The primary thermal effect by irradiation on the SR-STM system was verified. To separate the thermal effect from the effect by electronic emission or surface photo-voltage (SPV), a geometry was attempted to irradiate deep areas of the sample that lie far from the surface. The results revealed quantitatively that the stripe appearing in the STM image that corresponds to the on–off switching of the X-ray beam is attributed neither to the emission nor SPV, but rather to a thermal effect. Thermal disturbance could be effectively removed from the STM signal by reducing beam size from φ 200 μm to φ 10 μm in diameter. This process provides general clues for increasing the sensitivity of SR-STM for nanoscale chemical analyses.

1. Introduction

Inner-shell excitation of a specific level under scanning tunneling microscopy (STM) observation provides the possibility of analyzing elements or controlling local reactions with the spatial resolution of STM. Thus, an attempt to combine STM with X-rays appears attractive because it contains various possibilities for original and important applications. Although the number of studies combining STM with X-rays has long been quite limited [1–6] in comparison with that combining STM with visible light [7–10], the number of reports on X-ray-based STM has recently increased [11–14].

The difficulties of X-ray-based STM are related to the specific characteristics of X-rays such as small efficiency of excitation, a short lifetime, inconvenience in treatment, among others. To overcome a small efficiency of core-excitation by X-rays, a large intensity of X-rays is required, which simultaneously creates a large disturbance in STM observations. By overcoming the noise and radiation load around the tip [3], an *in-situ* synchrotron radiation (SR)-based STM (SR-STM) system has been developed for research dedicated to chemical analyses at nanometer resolution that is assisted by the element-selective core-hole excitation by energy tunable SR [15,16].

Using this specially designed system, we have successfully obtained STM images with atomic resolution under SR beam irradiation in which element contrast was possible for a semiconductor hetero-interface (Ge nanoislands on a clean Si(111) 7 × 7 surface) by changing the incident photon energy across the Ge absorption edge. By imaging the tip current modulation during an STM scan, the Ge island could be distinguished from the surrounding area in the STM image at a higher incident energy than the Ge absorption edge, due to the selective excitation of the Ge atoms [3]. The element contrast was also obtained for a metal–semiconductor interface (Cu nanoislands on a clean Ge(111) 2 × 8 surface) in the same manner as for the Si–Ge interface [15].

The essence of our methodology is to obtain the tip current modulation as a signal produced by element-specific excitation "under a tunneling condition" [15,16], and to avoid collecting the emitted electrons. This is because electrons are emitted from a wide irradiated area and can damage the spatial resolution of the analysis.

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In this signal acquisition process, the most essential point is how thermal absorption of the irradiated sample primarily affects the STM analysis since it will work as a serious noise. Practically, we have successively removed thermal disruption from the STM signal by reducing the beam size with an accurate alignment among the beam, sample, and tip. However, the thermal effect was not sufficiently estimated in the literature. Since reports on SR-STM have recently increased by attracting wider interests, it may be worth summarizing the estimation of the primary thermal effect in the initial irradiation process by discussing the method to analyze this effect.

From other viewpoints, the interaction of X-ray photons with materials plays an essential role in various fields such as imaging, diffraction and spectroscopy. As a higher photon density of X-rays is generally required for higher throughput of the measurement, the strong irradiation effect caused by brilliant synchrotron radiation must be verified. The strong photon–matter interactions in X-ray region will lead to potential applications in material processing such as crystal growth [17,18]. On the other hand, it is required to investigate the X-ray–matter interaction due to sample damage by X-rays that limits the resolution of structural studies [19]. Since the X-ray-free electron laser (XFEL) facilities have recently started worldwide their operation, it becomes important to take account of the influence, effect, or application of the extremely highly brilliant X-ray source, even if the photon–matter interaction shown in these references [17–19] do not yet relate directly to our elemental contrast. Actually, we have recently discussed about an advanced irradiation effect of X-rays using our SR-STM apparatus [12].

In this report, we provide an estimation of the primary thermal effect by the irradiation of brilliant hard X-rays from SR in ultrahigh vacuum (UHV) using STM dedicated to *in-situ* observation at an SR facility [3]. Also, we report on the method used to judge the thermal effect independently from other effects such as the emission obtained from the STM tip or the surface photo-voltage (SPV) on the sample. Finally, to remove the thermal disturbance from the STM signal, the efficacy of reducing beam size is clearly shown. The method that is presented provides hints as how to improve the sensitivity and accuracy of SR-STM for nanoscale chemical analyses.

2. Experimental

The *in-situ* SR-STM system has already been installed for research on nanometer-scale chemical analyses. The details of the apparatus are presented in our past reports [3,15,16,20].

2.1. Light source

To overcome a small efficiency of core-excitation by X-rays, we used SPring-8, which is an SR facility that provides highly brilliant X-rays. Furthermore, we installed the STM system at the beamline BL19LXU [21] that can provide the highest brilliance of X-rays from a 27 m long undulator, and focused the beam in two-dimensional to increase 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.), a special STM system was developed [3]. To avoid excessive heat load by brilliant X-ray irradiation, incident X-rays as small as possible (10 μm in diameter) were used under a condition of total reflection (grazing incident angle of the X-rays was ∼0.15°) that can effectively reduce the penetration depth of the X-rays into the sample. Beam size can be changed by changing the aperture size to estimate the thermal effect, and also, conveniently, to achieve the alignment gradually and effectively, step by step (800 μm, 200 μm, 100 μm, 50 μm, and 10 μm).

2.2. STM

Next, an essential point is the stability and accuracy of the experimental system. This is because the signal (beam-induced tip current modulation) is unstable and the yield varied easily (in a range of more than ±50%), and is affected by the delicate alignment of the incident X-rays in a range of a few microns. This instability must be overcome to pursue elemental analysis. By considering the experimental scheme (including the alignment system and stability of an active dumper) and by attempting various STM conditions under X-ray irradiation [3,15,16], the signal to noise (S/N) ratio was found to be a clue to obtain successful results. To effectively obtain local information with a high spatial resolution, it is essential to reduce the noise, including the electrons emitted from a wide irradiated area. Therefore, a specially designed insulator-coat STM tip was developed [20], which was originally made from tungsten wire (φ 0.3 mm) by electrochemical etching in NaOH solution (2 mol/l).

The sample and tip on the STM stage were controlled relative to the X-rays with an accuracy of ∼1 μm in UHV, which was possible by a specially fabricated alignment system composed of a multi-axis stage and also a monitoring system for the alignment [3]. The alignment was made using a silhouette of the sample and tip in the X-ray beam (Fig. 1). Fig. 2 shows the silhouettes obtained with an aperture of diameter 800 μm under low (a) and high (b) magnifications, and with an aperture of diameter 10 μm under a high magnification (c). This system enables the alignment of the X-ray beam of diameter 10 μm to the STM observation point.

The STM controller and scanner were based on a conventional STM system (JEOL Co., Ltd, Tokyo, Japan). The sample stage was set in a UHV chamber with a base pressure of 1.5×10^{-8} Pa. For sample surface, we used a clean Si(111) 7 × 7 surface (n-type, 4–7 Ω cm) from both viewpoints of standard and well-defined rigid clean surface.

3. Results and discussion

Despite a noisy condition in the experimental hutch of the SR facility and a large radiation load around the probe tip, STM images were successfully obtained with atomic resolution. First, it is necessary to estimate the influence of damage or noise caused by the X-ray irradiation on the STM observation. Actually, an X-ray beam of maximum intensity without the aperture breaks the monitor screen. However, such extensive damage was decreased by limiting the beam size to less than 200 μm in diameter. Fig. 3(a) shows an STM image of a clean Si(111) 7 × 7 surface in the constant-current mode (0.3 nA) with on–off switching of the X-rays where the inset shows a silhouette in the beam of diameter 200 μm. The energy of the incident beam was 16.5 keV at an incident angle of 1.5° from the sample surface.

The bright and dark bands in the stripe correspond to the on and off states of the beam, respectively. Since the images were obtained

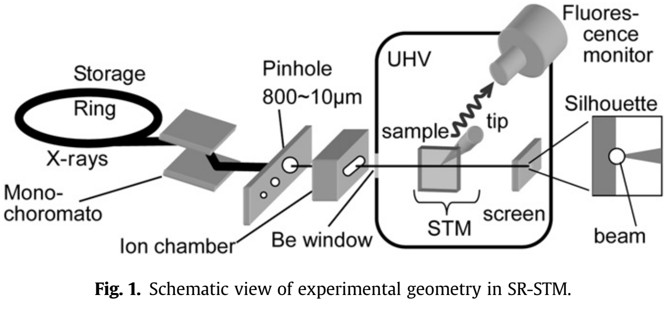

Fig. 1. Schematic view of experimental geometry in SR-STM.