Flexure Structural Scanner of Tip Scan Type for High-speed Scanning Tunneling Microscopy

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Received: 13 December, 2019, Accepted 29 March, 2020, Published 11 April, 2020

Dynamic behaviors of atoms on a material surface are important processes for catalytic reactions and crystal growth. Visualizing their processes will contribute to understanding reaction mechanisms and the process of surface structural formation. Scanning tunneling microscopy (STM) is a powerful tool to investigate the surface structure of solid materials and has been used to observe the atomic structures of various important materials such as metals and semiconductors. However, the scan speed of conventional scanning tunneling microscopes is too slow to capture dynamic processes of surface atoms in real time. In this study, we developed a new scanner for high-speed STM (HS-STM). This scanner is constructed from a flexure structure actuating in the \( x \) and \( y \) directions and a small piezoactuator actuating an STM tip in the \( z \) direction. To enhance the actuation bandwidth, the tip holder was minimized and the flexure was hardened. We successfully imaged atomic structures on both highly oriented pyrolytic graphite under ambient conditions and a Si(111) under ultrahigh vacuum at 1 frame s\(^{-1}\) using the developed HS-STM scanner.

Keywords High-speed scanning tunneling microscopy; Flexure structure; Actuation bandwidth; Atomic resolution

I. INTRODUCTION

Surface adsorption, diffusion, reaction, and nucleation are important processes on solid material surfaces [1]. Scanning tunneling microscopy (STM) is a fundamental tool to visualize these surface structures of conductive materials at the atomic level [2]. However, the scan speed of the typical scanning tunneling microscope is too slow to capture dynamic processes on a material’s surface in real time in situ. Therefore, various efforts have been made to increase the scan speed [3–6]. These efforts have led to fast imaging for Si(111), O/Ru(0001), and highly oriented pyrolytic graphite (HOPG) [3, 7, 8].

Widening the scope of the materials science fields that can be studied by STM requires further improvements in the imaging rate. The scanner that moves the STM tip or sample stage in three dimensions is a critical device that limits the STM imaging rate. This limitation stems from the resonant vibrations of the scanner in the \( x \), \( y \), and \( z \) directions [9]. The \( z \)-directional scanner is particularly important because it is involved in a feedback operation that keeps the tip–sample current constant by moving the tip or sample stage up and down depending on the sample height. This feedback operation is essential for accurately tracing the surface structure and minimizing the disturbance of sample condition. Most conventional high-speed STM (HS-STM) imaging has been conducted without such feedback control. However, in the case of high-speed scanning of rough surfaces such as tall clusters and deep defects, precisely tracing the surface is difficult without height control by feedback. Furthermore, the STM tip can be damaged by contact with the sample. Therefore, a high-speed scanner that can provide fast feedback operation is needed.

A tube-type scanner has been commonly used for conventional STM [10]. This scanner is commercially available and has highly accurate positioning capability; however, low resonant vibration restricts the operating frequency to a few kilohertz. Therefore, high-speed imaging has not been achieved using this scanner. The increase of the resonance frequency of the scanner will lead to an enhancement of the imaging rate. Resonance originates in the piezoactuator itself and in the scanner’s mechanical structure [11]. In the present study, we developed a new tip-scan-type STM scanner using both a tiny piezoactuator with a high reso-
nance frequency and a decoupling flexure structural design with a separately positioned piezoactuator. The size and shape of the tip holder were minimized and improved to increase the resonance frequency. The flexure structure was hardened to increase the $x$- and $y$-directional resonance frequency. We evaluated the actuating performance of the scanner and imaged HOPG in air and Si(111) under ultra-high vacuum (UHV). We succeeded in continuously capturing these atomic structures at 1 frame s$^{-1}$ via constant-height mode with feedback operation. High-speed and high-resolution STM imaging was achieved at atomic scale using this new scanner.

II. EXPERIMENTAL

A custom-built piezoactuator ($2 \times 3 \times 5$ mm$^3$) from NEC Tokin (Japan) was used for $x$ and $y$ actuation. The epoxy-resin exterior covering was removed to prevent outgassing in the case of measurement under UHV conditions. A piezoactuator ($3 \times 3 \times 2$ mm$^3$) of Noliac (Denmark) was used for $z$ actuation. The frequency-response performance of scanner actuation was evaluated using a laser displacement meter (IWATSU, Japan).

The home-built STM apparatus used in the present study is basically the same as that reported previously [12, 13]. A newly developed high-speed scanner was introduced in this apparatus. Experiments were performed in a chamber under air or under UHV at a base pressure of less than $1.0 \times 10^{-9}$ Torr at room temperature. Clean HOPG surfaces were prepared by cleavage. Clean reconstructed Si(111)-(7×7) surfaces were prepared by a standard method of repeated cycles of flash annealing at 1200°C followed by slow cooling from 900 to 500°C [14]. The STM tip used in this study was a tungsten tip sharpened by an electrochemical polishing method [15].

HS-STM images were analyzed using the WSxM software [16] and the SPIP image analysis software (Image Metrology).

III. RESULTS AND DISCUSSION

A. Development of high-speed STM scanner

Two main modes are available in STM: tip (probe) scan mode and sample-stage scan mode. To achieve stable and precise high-speed scanning, not only actuators but also actuated parts (tip or sample substrate) must possess high mechanical resonance. One possible improvement is to minimize the size of actuated parts to enhance the resonance frequency. However, in the case of the stage scan mode, minimizing the substrate for each measurement and each different sample material is difficult. Therefore, we used the tip scan mode in the present study.

Figure 1(a, b) shows a schematic of the HS-STM scanner developed in this study. The concept of a high-speed atomic force microscopy (HS-AFM) scanner was used to construct this new STM scanner [17]. Two piezoactuators for $x$- and $y$-directional actuation were inserted into hard flexure structures made from a monolithic stainless frame. The $y$ piezoactuator moves the flexure structure with the inserted $x$ piezoactuator in the $y$ direction. The $x$ piezoactuator moves the base of the $z$ piezoactuator in the $x$ direction. The piezoactuator for $z$-directional actuation was placed on the base. A tip holder was placed on the $z$ piezoactuator. A sharpened tungsten tip was glued onto the tip holder. A cylindrical dummy stage was placed on the opposite side as an $x$-directional counterbalance. In this study, to achieve greater mechanical resonance of the $x$ direction, we examined the frequency

Figure 1: Schematic view and photographs of the HS-STM scanner. (a) Side view (left) and diagrammatic perspective view (right) of the HS-STM scanner. The sample substrate on the sample holder approaches the STM tip, and then the STM tip is scanned using the HS-STM scanner. The counterbalance weight for the $x$ direction is placed on the opposite side of the $z$ piezoactuator with the tip holder. (b) Enlarged view of the main parts of the HS-STM scanner. The $x$ and $y$ piezoactuators (green) are confined in a monolithic stainless frame (gray) with a flexure structure shown by dotted rectangles (left view). A tip holder is placed on the $z$ piezoactuator (right view). Upper-right view shows the cuboid-shaped tip holder and the lower-right view shows the cylindrically shaped one. In each figure, green parts are piezoactuators. (c) Photographs of the developed scanner. The colored leads indicate conductive wires for the piezoelectric actuators and the tip current detection.

e-J. Surf. Sci. Nanotechnol. 18, 146–151 (2020) | DOI: 10.1380/ejssnt.2020.146
response of each flexure structure of 0.4 mm and 1.0 mm thickness [dotted rectangles in the left figure of Figure 1(b)]. Furthermore, to achieve high mechanical resonance in the z direction, we compared the performance of tip holders with two different structures: a cuboid-shaped one (7 mm length) and a cylindrically shaped one (2 mm length) with a slot for inserting the STM tip [right in Figure 1(b)]. Figure 1(c) shows photographs of the developed scanner. This scanner was introduced into the STM apparatus.

B. Performance evaluation

We evaluated the frequency response performance of the new STM scanner using a laser displacement meter. Figure 2(a) shows the x-directional frequency response spectra. In the case of the 0.4 mm-thick flexure structure (line 1), a large peak appears at 15 kHz. However, the 1.0 mm-thick flexure structure shows a flat spectrum at frequencies less than 10 kHz and an initial large resonance peak appears at 20 kHz (line 2).

Figure 2(b) shows the z-directional frequency response spectra of the new STM scanner. In the case of the new STM scanner with a cuboid-shaped tip holder (line 1), some small peaks appear at approximately 10−30 kHz and large peaks appear at 100 kHz. By comparison, the new STM scanner with a cylindrical holder shows a flat spectrum at frequencies less than 100 kHz and an initial large resonance peak appears at 200 kHz (line 2). These results indicate that the asymmetric and vertically long structure of the cuboid holder cause unstable vibrations at low frequencies. Although we can more readily fix the STM tip to the cuboid holder than to the cylindrical holder, the operation bandwidth of the cuboid one is lower than that of the cylindrical one.

To compare the performance of our new scanner and a conventional scanner, we measured the frequency-response spectrum of the z direction for a tube scanner [Figure 2(c)]. Some small peaks appear at approximately 5−10 kHz, and large peaks appear at approximately 10−30 kHz. These results show that the z-directional actuation bandwidth of our new STM scanner with a cylindrical tip holder is more than ten times greater than that of the conventional tube scanner. We achieved a resonance frequency of 20 kHz in the x direction and 200 kHz in the z direction with the new scanner.

C. High-speed imaging

To demonstrate the performance of the new scanner for HS-STM, we performed STM imaging of solid material samples [HOPG and Si(111)]. These measurements were initially started at a slow scan speed to obtain high-resolution images. We then increased the scan speed in a step-by-step manner.

We observed a freshly cleaved HOPG surface under air using the newly developed STM scanner. Figure 3 shows successive images in an HS-STM movie captured at an imaging rate of 1 frame s⁻¹ (Movie S1 in Supplementary Material). The hexagonal lattice structure composed of carbon atoms can be seen in each STM image. The lattice constant of the atomic structure in HOPG is 2.5 Å. This movie indicates that the spatial resolution of our new HS-STM is greater than 2.5 Å. The atomic structure of the HOPG surface could be continuously visualized for longer than 2 min. We also successfully captured atomic structures at a higher imaging rate of 2 frames s⁻¹; however, the spatial resolution was lower than that achieved at 1 frame s⁻¹ (data not shown).

Next, we observed clean reconstructed Si(111)-(7×7) surfaces under UHV. Figure 4(a) shows successive images from an HS-STM movie captured at an imaging rate of 1 frame s⁻¹ (Movie S2 in Supplementary Material). The (7×7) dimer-atom-stacking-fault (DAS) structure on the Si(111)
surface was visualized in each STM image. Some atomic defects are observed in the \((7 \times 7)\) DAS structure. The atomic structure with the defects could be stably observed for more than 1 min. Figure 4(b) shows a cross-sectional view of the HS-STM images at 8 s shown in Figure 4(a). In the case of high-speed imaging without feedback control of the tip-sample distance, parachuting is often observed such that the STM tip completely detaches from the sample surface at a steeply inclined region of the sample [18]. In our imaging with feedback control, obvious parachuting is not observed on the defect in the HS-STM image and the atomic structure is clearly traced on the Si(111) surface. Furthermore, we analyzed the position of Si atoms in successive HS-STM images [Figure 4(c)]. A shift of the scan area caused by drift is not observed even for high-speed imaging. These results demonstrate that HS-STM with our new scanner can stably and precisely visualize the atomic structure in high temporal resolution.

We present the HS-STM movies captured at 1 frame s\(^{-1}\) in this paper (Supplementary Material). However, the mechanical performance of the newly developed scanner has the potential to achieve even higher imaging rates. Therefore, we continue to attempt faster imaging as part of our ongoing work. In addition to such efforts, the performance of the other devices involved in STM control, such as the current amplifier and feedback circuit, should also be enhanced. Furthermore, high-quality STM probe tips are important for high-speed imaging with high spatial resolution at the atomic level. Such improvements will lead to the realization of video-rate STM imaging in the future.

IV. CONCLUSIONS

In this study, we developed a new scanner for HS-STM imaging. The actuation bandwidth of this scanner was drastically increased compared with that of a conventional tube scanner. We successfully visualized an atomic structure at an imaging rate of 1 frame s\(^{-1}\). This technique will be useful for studying dynamic processes on material surfaces at the atomic level.

Acknowledgments

This work was supported by JSPS KAKENHI Grant numbers 15K18517 and 17KT0024 (to H.Y.), and 19H05789 and 18K19023 (to M.A.), PRESTO/JST, Foundation Advanced Technology Institute, Multidisciplinary Research Laboratory System of Osaka University, and the Osaka University Program for the Support of Networking among Present and Future Researchers (to H.Y.).

Appendix

HS-STM movies of the HOPG and Si(111)-(7 \times 7) surfaces are available in Supplementary Material at https://doi.org/10.1380/ejssnt.2020.146.

Note

This paper was presented at the 12th International Symposium on Atomic Level Characterizations for New Materials and Devices ’19 (ALC ’19), in conjunction with the 22nd International Conference on Secondary Ion Mass Spectrometry (SIMS-22), Miyako Messe, Kyoto, Japan, 20–25 October, 2019.

Figure 3: Successive HS-STM images of an HOPG surface under air at room temperature. These images were acquired at 1 frame s\(^{-1}\). The sample bias voltage \((V_s)\) and tip current \((I_t)\) were 0.5 V and 2.6 nA, respectively. The scan area was 2 nm \times 2 nm. The pixel size was 200 \times 200. Vertical brightness range: 4 Å. Atomic arrangements of carbon on the HOPG surface (white circles) were superimposed in the STM image at 0 s. See Movie S1 in Supplementary Material.
References

[1] G. A. Somorjai and Y. M. Li, Proc. Natl. Acad. Sci. U.S.A. 108, 917 (2011).
[2] G. Binnig and H. Rohrer, Surf. Sci. 126, 236 (1983).
[3] R. Curtis, T. Mitsui, and E. Ganz, Rev. Sci. Instrum. 68, 2790 (1997).
[4] C. Dri, F. Esch, C. Africh, and G. Comelli, Meas. Sci. Technol. 23, 055402 (2012).
[5] M. Wilms, M. Schmidt, G. Bermes, and K. Wandelt, Rev. Sci. Instrum. 69, 2696 (1998).
[6] D. Croft and S. Devasia, Rev. Sci. Instrum. 70, 4600 (1999).
[7] J. Winterlin, J. Trost, S. Renisch, R. Schuster, T. Zambelli, and G. Ertl, Surf. Sci. 394, 159 (1997).
[8] Q. Li and Q. Lu, Rev. Sci. Instrum. 82, 053705 (2011).
[9] C. Y. Nakakura, V. M. Phanse, G. Zheng, G. Bannon, E. I. Altman, and K. P. Lee, Rev. Sci. Instrum. 69, 3251 (1998).
[10] G. Binnig and D. P. E. Smith, Rev. Sci. Instrum. 57, 1688 (1986).
[11] N. Kodera, H. Yamashita, and T. Ando, Rev. Sci. Instrum. 76, 053708 (2005).
[12] D. Katsube, H. Yamashita, S. Abo, and M. Abe, Beilstein J. Nanotechnol. 9, 686 (2018).
[13] S. Ojima, D. Katsube, H. Yamashita, Y. Miyato, S. Abo, and M. Abe, Jpn. J. Appl. Phys. 58, S11A10 (2019).
[14] A. Yurtsever, M. Abe, S. Morita, and Y. Sugimoto, Appl. Phys. Lett. 111, 233102 (2017).
[15] Z. Q. Yu, C. M. Wang, Y. Du, S. Thevuthasan, and I. Lyubimetsky, Ultramicroscopy 108, 873 (2008).
[16] I. Horcas, R. Fernández, J. M. Gómez-Rodriguez, J. Colchero, J. Gómez-Herrero, and A. M. Baro, Rev. Sci. Instrum. 78, 013705 (2007).
[17] T. Ando, T. Uchihashi, N. Kodera, D. Yamamoto, A. Miyagi, M. Taniguchi, and H. Yamashita, Pflügers Arch. 456, 211 (2008).

Figure 4: HS-STM observation of Si(111) under UHV at room temperature. (a) Successive HS-STM images of the Si(111) atomic structure. These images were acquired at 1 frame s⁻¹, Vₜ and Iₜ were 1.0 V and 0.9 nA, respectively. The scan area was 8 nm × 8 nm. The pixel size was 64 × 64. Vertical brightness range: 1.5 Å. See Movie S2 in Supplementary Material. (b) Cross-sectional profile on the black line shown in the HS-STM image at 8 s. (c) Trajectories of three Si atoms in successive HS-STM images. The position of the gravity center in each atom was analyzed and plotted in a two-dimensional graph.
[18] T. Ando, T. Uchihashi, and T. Fukuma, Prog. Surf. Sci. 83, 337 (2008).

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