Electron microscopy of the strain on the Si(111)\(7 \times 7\) surface induced by the STM tip

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(March 22, 2022)

The Si(111)\(7 \times 7\) surface was observed by reflection electron microscopy (REM) and scanning tunneling microscopy (STM) simultaneously in an ultra-high vacuum electron microscope. The distance between the STM tip and the Si surface was detected from the REM image, which showed the real and the mirror image of the tip. We approached the tip to the surface or retracted from the surface by a piezo drive to observe the strain induced on the Si(111)\(7 \times 7\) surface as a function of the tip-surface distance. This investigation was done with and without the bias voltage between the tip and the substrate. With bias voltage of 1.0 V on the sample, the tip was approached to 1.6 nm above the sample surface for the tunneling current of 0.8 nA, no detectable order of strain (\(\sim 10^{-4}\)) was induced on the sample surface. When the bias decreased within the range of \(-0.3 \text{ V} \sim +0.5 \text{ V}\), the surface was compressed over the Si surface area of 100 nm. Without the bias voltage, tensile and compressive strain was detected as the tip-surface distance changed from attractive to the repulsive interaction regime. The strain field extended over 50 nm \(\sim 140 \text{ nm}\), and the force became neutral at the tip-substrate distance of 0.45 nm.

I. INTRODUCTION

Scanning tunneling microscope (STM) and atomic force microscopy (AFM) has been used not only as a mean of microscopy, but also as a tool for nanofabrication, nanoprobeing, or manipulation of atoms and molecules on surfaces. The techniques attract much interest in the fields of nano-devices and molecular electronics. In these techniques modification by tips or the migration of surface atoms caused by high electric field are utilized. Dynamical processes occurring at the gap between the tip and the substrate surface is important to be understood.

A combination of a STM with an electron microscope have been devised in some research groups to see the tip and the substrate surface "in-situ" simultaneously. Those pioneer works have successfully revealed the tip-substrate gap and the tip-surface distance. More recently a plastic change of the tip apex was observed during STM operation. Furthermore, STM tip was used to make a gold nanowire between the tip and the substrate.

A significant elastic deformation of the tip and the surface is noticed under STM operation or AFM operation. Theoretical study on the jump-to-contact demonstrated that attractive interaction between the tip and the substrate causes straining of the tip and substrate, which sometimes provoke the atom transfer. Although such interaction at atomic level was investigated from force measurement by AFM contact size and the tip-surface distance, have no ways to be detected.

We devised a STM holder attachable to our ultra-high vacuum (UHV) electron microscopy to observe the tip-substrate contact by reflection electron microscopy (REM). Although the deformation of the tip and the
surface was small, REM images were sensitive enough for detecting strains of the order of $10^{-4}$.

Here, we report REM observation of the Si(111)$7 \times 7$ surface strained by a tungsten tip, as the tip approaches to the surface. The strain of the order of $10^{-4}$ was first observed to have extended over a circular area of about 100 nm.

II. EXPERIMENTAL

A. Design of REM holder

The experiments were performed in the UHV electron microscope (JEM-2000FXV) whose pressure is better than $5 \times 10^{-7}$ Pa. The REM-STM specimen holder (Fig.1) was devised to fit into the narrow gap (3.5 mm) of the objective pole piece. The Si(111) crystal (0.02 Ωcm, n-type) was flash cleaned at 1200°C by passing the DC current directly through the Si crystal. Tungsten STM tip was sharpened by chemical etching, and preheated for cleaning in an UHV chamber before REM-STM experiment. The STM tip was approached to the substrate by a mechanical drive (2 mm) and a stack piezo (6 µm), and STM image is obtainable by a tube piezo scanner (1 µm). The tip motion was observed directly by transmission electron microscopy (TEM).

B. STM tip

Apices of tungsten tips were observed by high-resolution TEM and electron diffraction pattern. They always had the (110) plane vertical to the tip axis direction and had a curvature of 2 - 6 nm. Because of the preheating before putting the tip into the UHV electron microscope, we have not seen heavy contaminations covering over the tip apices. After several STM scans on the Si(111)$7 \times 7$ surface, the apices were found often to have been scraped. The scanning was usually done at sample bias of $V_s < 2$ V, and the tunneling current of $0.3 \text{nA} < I_t < 2.0 \text{nA}$. The scraped tips were terminated with the (110) plane, which was as wide as 10 - 300 nm. Such apices of STM tips were used in the following experiments. We are allowed observing STM images of the Si(111)$7 \times 7$ surface at atomic resolution, when the truncated tip had an adatom cluster on its top.

C. REM imaging of the tip apex

Ray diagram of the REM is illustrated in Fig.3. The Si(111) substrate is placed to the REM-STM holder, whose surface is inclined by an angle $\theta_0$ from the objective lens axis. Then the incident electron beam is reflected specularly to the surface, propagating along the
objective lens axis to give the REM image on a fluorescent screen. The image is projection of the sample surface, being foreshortened by a factor of \( \sin \theta_0 \) in the direction of the propagating beam. For the specular beam of the 444 Bragg reflection of the Si, the image is foreshortened by a factor of \( 1/73 (\theta_{444} = 1.36 \times 10^{-2} \text{ rad}) \) at the accelerating voltage of 200 kV.

When the tip is approached to the surface, the real and its mirror image of the tip apex appear on the REM image, as shown in Fig. 4(a). The distance between these apices is \( (1 + \cos 2\theta_0)d_{\text{REM}} \), where \( d_{\text{REM}} \) is the gap between the tip and the reflection plane of the Si(111) surface. Although no RHEED calculation had predicted the position of the reflection plane, which should locate between the adatom and the stacking-fault layer of the Si(111)\(7\times7\) surface.

D. Strain contrast in REM image

Provided that the surface has no strain, the specular beam changes its intensity, \( I_0 \), for the incident beam angle, \( \theta_0 \), as illustrated in Fig. 3(a). The rocking curve of the specular beam, a relation of intensity and incident angle, has sharp peaks at the Bragg reflection conditions (\( \theta_0 = \theta_B \)). The Bragg width, \( \Delta \theta_B \), which was the maximum half width of the specular reflected beam intensity peak, of the 444 Bragg reflection of the Si(111) crystal was calculated to be \( 7.2 \times 10^{-4} \text{ rad} \) on the dynamical Bethe theory (100 kV accelerating voltage was assumed in the calculation). The Bragg width at 200 kV accelerating voltage is of the same order as \( 10^{-4} \text{ rad} \). When the incident beam angle changes by an amount of the Bragg width from the angle of Bragg condition, the REM image changes from bright to dark. Based on this criterion, we understand strain contrast in the REM image. When a compressive force is exerted on the surface, surface lattice strains as shown in Fig. 3(b). Provided that the incident beam satisfies the Bragg condition for the unstrained surface, the incident beam does not satisfy the Bragg condition for the strained area. Thus, the strained area gives dark contrast, except the central area (see Fig. 3(b)). When the incident angle is smaller (larger) than the Bragg angle by the Bragg width for the unstrained area, the dark contrast appears only on one side of the strained area.

III. RESULTS AND DISCUSSION

A. Tip-approach with bias voltage

The real and mirror images of an STM tip appear in the REM image, as reproduced in Fig. 3(a). The appearance of the lattice fringes of the 7\(\times7\) surface along the vertical direction, the [110] direction, proves cleanliness of the surface. The tip is kept at a constant height from
the surface, while the STM tip is biased by 1.0 V and tunneling current of 0.8 nA. The real and the mirror image gave the tip-surface distance of $d_{REM} = 1.6 \pm 0.5$ nm. By further reduction of the voltage ($-0.3 \text{ V} < V_s < 0.5 \text{ V}$, and $I_t = 0.8 \text{ nA}$), the tip approached so close to the surface that the separation between the real and mirror tip images could not be resolved. In these bias voltages, a dark horizontal line appeared between the two tip images, as shown in Fig.4(b). The dark line extended over an area of 120 nm. This dark line image is due to compressive strain of the Si(111)$7 \times 7$ surface induced by the tip. As explained before, contrast analysis confirmed the compressive strain.

Figure 5 shows REM image of a compressive strain, where $V_s = +0.5 \text{ V}$ and $I_t = 0.8 \text{ nA}$. The grazing angle of the incident electron beam increases from (a) to (c), passes the Bragg condition in (b). The strain contrast underneath the tip is dark-bright in (a), dark-dark in (b) and bright-dark in (c). This change indicates the compressive strain. The strain of the order of $10^{-4}$ extends over 120 nm, as seen from the length of the dark line in Fig.5.

The tip-surface distance, $d_{REM}$, was measured as a function of the bias voltage ($I_t$ is kept constant), and plotted in Fig.4. The observed $d_{REM}$ vs. bias relationship in Fig.4 do not accord with the previous one that was deduced from the conductance oscillation due to the tunneling barrier resonance. $d_{REM}$ decreases steeply to zero as the positive bias decreases to $+0.5 \text{ V}$, or as the negative bias increases to $-0.3 \text{ V}$. The bias voltages that $d_{REM}$ goes to zero are close to the valence and the conduction band edge. When the bias is close to the band edge as in the case of Fig.6, the tip is almost touching to the sample surface. The tip had no mechanical contact, because of repulsive interaction. No trace of mechanical contact was seen on the Si(111)$7 \times 7$ surface after the retraction of the tip from the surface, indeed.

### B. Strain of the Si(111) induced by the tip without bias voltage

The Si(111)$7 \times 7$ surface was also found to be strained by the tip, when no bias voltage was applied. The tip was approached to and retracted from the surface by the tube piezo scanner. REM images for the tip motion was recorded on a videotape, and analyzed in detail. Figure 7 is a series of REM images, each of which show the real (upper side) and mirror (lower side) image of the tip apex. The $7 \times 7$ lattice fringes of the Si(111) surface were appearing always. As the tip approaches from (a) to (d), a dark horizontal line comes out in (b), disappears in (c), and reappears in (d). On the way back from (d) to (g), the line contrast changes reversibly. The tip-substrate gap distance was measured in reference to the $d_{REM}$ in Fig.7(a). The $d_{REM}$ in Fig.7(a) was measured directly from the REM to be 1.25 nm. Further
approach of the tip did not allow accurate measurement of $d_{\text{REM}}$ value, so that the gap distance was estimated by 

$$d = d_{\text{REM}} - \Delta d_{\text{piezo}}$$

where $\Delta d_{\text{piezo}}$ is the elongation of the tube piezo scanner. The gap distance, then, is 0.9 nm, 0.4 nm, 0.15 nm, 0.45 nm, and 0.85 nm for (b) - (f), respectively. In (g), the gap distance became $d_{\text{REM}} = 1.7$ nm, which was measured directly from the REM image. The length of the dark lines in REM images in Fig.7 (and other series of tip approach) were measured as a function of the gap distance, $d$, and plotted in Fig.8. The strain for $0.15 \text{ nm} < d < 0.4 \text{ nm}$ (Fig.7(d)) is compressive, while that $d > 0.45 \text{ nm}$ is tensile (Fig.7(b) and (f)). Neither attractive nor repulsive force works at the gap distance of $0.45 \text{ nm} \pm 0.03 \text{ nm}$ (Fig.7(c) and (e)). Looking the length of the dark lines (the area having strained more than $10^{-4} \text{ rad}$) in Fig.8, the range of the strain field is found to be extremely wide. The maximum strain field for the attractive interaction extends over the area of 50 nm at the gap distance of 0.8 nm. For repulsive interaction regime, the range extended even more than 100 nm for $d < 0.2 \text{ nm}$. We calculated the range of the compressive strain field, following the classical elastic approach by assuming various radiiuses of a flat topped tip and a compressive force. However, no reasonable radius or force could explain the magnitude of the strain field seen in the REM image.

In the experiments, we did not observe for our tip to jump-to-contact with the Si surface, since our tip is rigid enough. On the other hand, we observed jump-to-detach motion of our tip while withdrawal of the tip. On the withdrawal, the tensile strain contrast reaches to its maximum in Fig.7(f) at $d = 0.85 \text{ nm}$. Its contrast is kept constant by further withdrawal (0.05 nm) of the tip, but it disappears suddenly (within one frame of the VTR recording: time after 30 ms). In this jump-to-detach motion, the attractive force changed from its maximum to zero, and the gap distance had changed from $d = 0.9 \text{ nm}$ to $d_{\text{REM}} = 1.7 \text{ nm}$ (Fig.7(g)). The reason is not clear. The tip apex had no mechanical contact with the substrate during the approach and withdrawal process, if the tip-surface distance was larger than 0.15 nm. When we push the tip to a distance closer than 0.1 nm, we began to see scratch mark on the Si surface after the tip withdrawal. The tip also strained greatly. From these observations, mechanical contact between the tip and the surface begins at the gap distances larger than 0.15 nm.

The present REM-STM observation of the Si(111)7×7 surface by a tungsten tip, thus, has revealed the strain range of 50 nm at gap distance of 0.8 nm. Such strain field might cause potential gradient of the surface to excite migration of adsorbed atoms or of the surface atoms. The absolute value of the gap distances is supposed to be overestimated, since the gap distances (Fig.5) in STM condition are larger than the previous report. Any way, the strain field of the substrate was detected first in this experiment. The strain changed tensile to compressive in relation to the attractive and repulsive force from the tip, respectively.
Atomic process such as jump-to-contact has not seen in the REM-STM experiment. This might be poor resolution of the REM image, and should be done by TEM-STM in future. TEM-STM, however, is only sensitive to the strain of the order of $10^{-3}$. Detection of the surface strain becomes possible by REM imaging of the surface.

IV. CONCLUSION

By a combination of STM with UHV electron microscope, the Si(111)7 \times 7 surface was observed simultaneously in REM and STM. The tip apex could be imaged in REM images of the specularly reflected electron beam. From the real and the mirror images of the tip, we knew the tip-surface distance directly. The tip was approached to and retracted from the sample surface with or without bias voltage being applied between the tip and the sample surface. The tensile strain was induced on the Si surface when the gap distance is $0.45 \text{ nm} < d < 0.85 \text{ nm}$. The surface strain turns into compressive for $0.15 \text{ nm} < d < 0.45 \text{ nm}$. Mechanical contact of the tip to the sample surface occur for $d < 0.10 \text{ nm}$. The range of the strain field, the area strained more than $10^{-4}$, is 50 nm in the attractive force regime, while it exceeds 100 nm in the repulsive interaction regime.

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FIG. 1. Design features of the STM holder for our electron microscope.

FIG. 2. Schematic illustration of REM imaging. An electron beam with an incident angle, \( \theta_0 \), is specularly reflected by a substrate surface. The REM image is foreshortened by the factor of \( \sin \theta_0 \). It is just like the projected image from the virtual incidence. When the tip is approached to the surface, a true and the mirror images of the tip are seen on the REM image of the surface. The distance between the two tip apices images is given by \( (1 + \cos 2\theta_0) d_{REM} \).

FIG. 3. (a) Schematic illustration for the beam reflection on the surface and specular reflection intensity, \( I \), as a function of incidence angle, \( \theta_0 \). The intensity at the Bragg condition, \( \theta_0 = \theta_B \), has sharp peak with the Bragg width, \( \Delta \theta_B \). (b) Schematic illustration for the changes of incident angle on the compressive strained surface and the specular reflection intensity distribution. Two dark lines contrast at \( \theta_0 = \theta_B \) change to bright-dark (dark-bright) contrast under condition that incident angle, \( \theta_0 \), is larger (smaller) than the Bragg angle, \( \theta_B \).
FIG. 4. (a) shows the REM image of a tungsten tip and a Si(111) surface when the tip is approached to the surface in a constant current mode of STM ($V_s = +1.0\text{ V}$, $I_t = 0.8\text{ nA}$ and). The gap distance between the tip and the reflection plane in the substrate surface, $d_{REM}$, is estimated to be 1.6 nm. (b) is the strained surface image of the Si indicated by a horizontal dark line contrast between the two tip images as reducing the sample bias to $-0.3\text{ V} < V_s < +0.5\text{ V}$. The dark contrast length implies the strain of the order of $10^{-4}$ extended over the 120 nm diameter area.

FIG. 5. REM images of the strained surface below the tip at $V_s = +0.5\text{ V}$, $I_t = 0.8\text{ nA}$ taken (a) under out of Bragg condition of $\theta_0 < \theta_0$, (b) for Si (444) Bragg condition of $\theta_0 = \theta_B$ and (c) under out of Bragg condition of $\theta_0 > \theta_B$. Changes of the line contrast between the true and mirror tip images are noted.

FIG. 6. The $V_s$ vs $d$ plot obtained by REM-STM observation. The circles and open triangles refer to different tunneling currents, $I_t = 0.35\text{ nA}$ and 0.8 nA, respectively.

FIG. 7. REM images of the straining on a Si surface induced by a tip without applying sample bias voltage, which were taken at (a) $d_{REM} = 1.2\text{ nm}$, (b) $d = 0.8\text{ nm}$, (c) $d = 0.4\text{ nm}$ and (d) $d = 0.1\text{ nm}$ in a tip approaching process and at (e) $d = 0.45\text{ nm}$, (e) $d = 0.85\text{ nm}$ and (f) $d_{REM} = 1.7\text{ nm}$ in a tip withdrawing process.

FIG. 8. The contrast length of the strained surface in fig.7(b) - (f) plotted for the tip-surface distance, $d$. The length of the compressive (attractive) straining is shown by positive (negative) value.
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