Phase Transition between $c(4 \times 2)$ and $p(2 \times 2)$ Structures of the Si(100) Surface at 6 K Caused by the Fluctuation of Phase Defects on Dimer Rows due to Dimer Flip-Flop Motion

Hidemi Shigekawa[*1], Koji Miyake, Masahiko Ishida, Kenji Hata, Haruhiro Oigawa,
Yasuo Nannichi, Ryozo Yoshizaki[*2], Akira Kawazu[*3], Takeru Abe[*4],
Takehiro Ozawa[1] and Toshihiko Nagamura[1]

*Institute of Materials Science and Center for TARA, Tsukuba Advanced Research Alliance,
University of Tsukuba, Tsukuba 305, Japan*
[1]*Unisoku Co., Ltd., Hirakata 573-01, Japan*

Dimers of the Si(100) surface previously observed at 120–140 K, which partially appeared to be symmetric, were frozen at 6 K, and an atomically resolved structural change between $c(4 \times 2)$ and $p(2 \times 2)$ arrangements of the Si(100) surface was observed for the first time by scanning tunneling microscopy. The observed change was due to the fluctuation of phase defects of a new type on dimer rows, which have a structure similar to that of the type-C defect. Dimer flip-flop motion at the phase boundary which induces the apparent phase transition, and creation and annihilation of the anti-phase domain were clearly observed.

KEYWORDS: STM, Si(100), surface reconstruction, dimer, $c(4 \times 2)$, $p(2 \times 2)$.

It is well established that pairs of top-layer atoms form dimers on the Si(100) surface. As Hamers *et al.* showed by scanning tunneling microscopy (STM),[1] dimers give rise to the (2×1) structure. According to recent low-temperature STM observations conducted at ~ 140 K for a surface with low defect density of $\sim 1\%$, the $c(4 \times 2)$ structure was observed to be the most stable among the buckled dimer structures.[2] At the boundaries of the anti-phase domains induced by type-C defects,[3] a symmetric dimer structure was observed instead of the $p(2 \times 2)$ arrangement. Since the type-C defect acts as a phase shifter, when it exists in the $c(4 \times 2)$ arrangement area, an out-of-phase structure is induced at one side of the defect. At ~ 140 K, the out-of-phase contributions from two adjacent dimer rows cancel the buckling in dimers, which results in the formation of an apparently symmetric dimer region. The observed apparently symmetric dimer structure was understood to be the result of rapid dimer flip-flop motion caused by the instability of the $p(2 \times 2)$ arrangement of a dimer row with respect to the two adjacent dimer rows at the observation temperature (140 K).[2, 4]

In the lower-temperature STM work at 120 K, the $p(2 \times 2)$ arrangement was observed at some boundaries between two adjacent $c(4 \times 2)$ domains, and it was attributed to the high defect density ($\sim 8\%$) of the surface. However, 20% of the unbuckled dimer area, indicating the rapid dimer flip-flop motion, still existed at 120 K.[5]

Therefore, in order to clarify the characteristics of the dimer flip-flop motion and related structural changes between the $p(2 \times 2)$ and $c(4 \times 2)$ arrangements on the Si(100) surface, it is necessary to analyze the dimer fea-

tures on a low-defect-density surface on an atomic scale at temperatures sufficiently low to freeze the dimer flip-flop motion.

In this work, we present the results of the first direct STM observation of the dynamics of the Si(100) surface dimers at low temperature; i.e., dimer flip-flop motion and related structural changes between the $c(4 \times 2)$ and $p(2 \times 2)$ arrangements at 6 K.

Experiments were performed using a low-temperature STM that allowed observation with atomic resolution at ~ 6 K in ultrahigh vacuum (5×10^{-9} Pa). A phosphorus-doped ($0.005 \,\Omega \cdot$ cm) Si(100) sample surface was heated to $\sim 1200°$C after prebaking for one day, and cooled very slowly to reduce the density of surface defects. The base pressure was $\sim 1.0 \times 10^{-8}$ Pa, and the pressure during the heat treatment was kept below $\sim 5 \times 10^{-8}$ Pa.

Figure 1 shows STM images acquired through continuous scanning at ~ 6 K. The surface was scanned in the constant current mode from left to right, and from top to bottom, at a scan rate of 9 Hz. The sample voltage V_s and tunneling current I_t were set at +1.5 V and 1.5 nA, respectively. Positive sample bias voltage was used to highlight the contrast between type-C and other defects.[3] Type-C defects appear as bright in Fig. 1. Each image shows the change taking place during a scan of about one minute.

As shown in Fig. 1, the $c(4 \times 2)$ and $p(2 \times 2)$ arrangement areas coexist. They appear as red and blue, respectively in each image. In addition, as shown in Figs. 1(a) to 1(c), the observed surface is remarkably dynamic; a transition between the two phases is occurring. The observed structural change is caused by the dimer flip-flop motion at the phase boundaries on dimer rows as will be shown later. This is the first atomically resolved observation of dimer flip-flop motion and fluctuation between the two phases.

Figure 2 shows magnified images of the same area in the upper terrace of Figs. 1(a) and 1(b). Fluctuation between the two arrangements is clearly shown here. The dimer rows which rearranged with respect to their two

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[*1]Corresponding author, e-mail address: hidemi@mat.ims.tsukuba.ac.jp
[*2]Permanent address: Institute of Applied Phys., and Cryogenics Center, University of Tsukuba, Tsukuba 305, Japan.
[*3]Permanent address: Department of Applied Phys., University of Tokyo, Bunkyo-ku, Tokyo 113, Japan.
[*4]On leave from Depertment of Material Science, Ryukoku University, Otsu, Shiga 520-21, Japan.
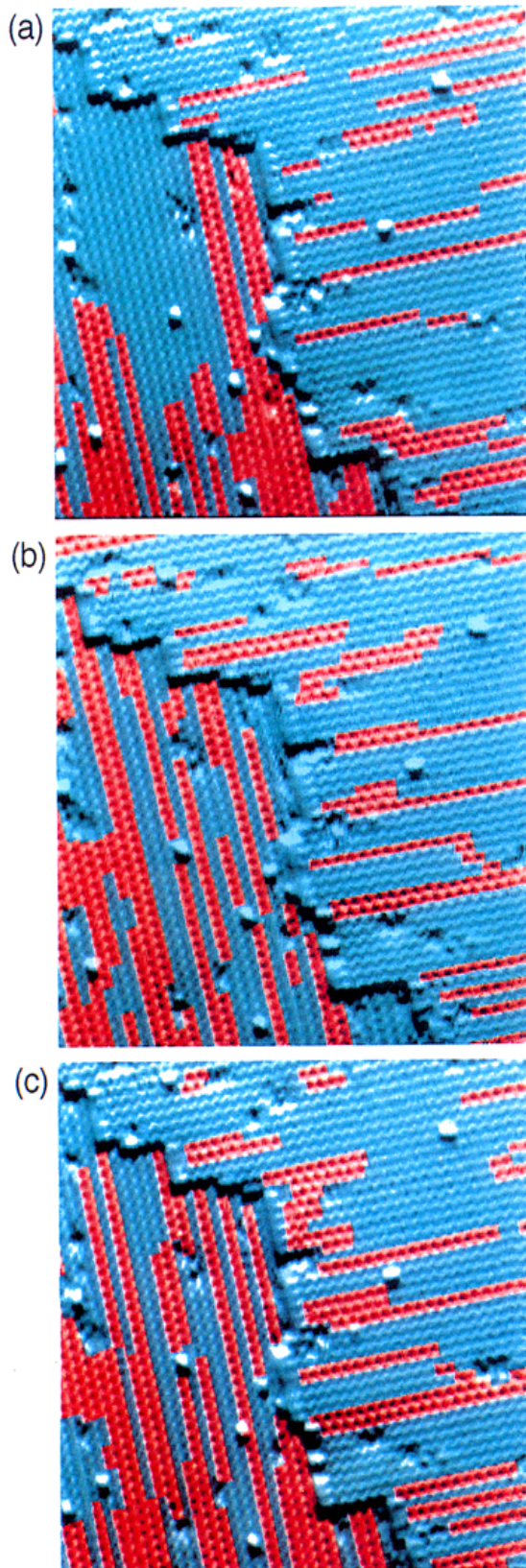
(a)

(b)

(c)

Fig. 1. STM images of Si(100) surface obtained continuously at ∼ 6 K ($V_s = 1.5$ V, $I_s = 1.5$ nA). Each area in the images shows the change taking place during a scan of about one minute ((a) → (b) → (c)). The areas with the $c(4 \times 2)$ and $p(2 \times 2)$ arrangements appear as red and blue, respectively.
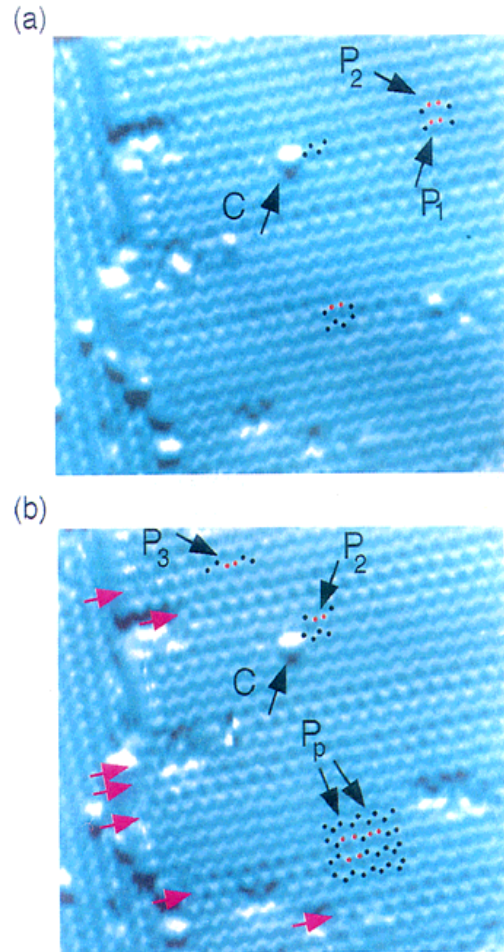
(a)

(b)

Fig. 2. STM images of Si(100) surface obtained continuously at ∼ 6 K ($V_s = 1.5$ V, $I_s = 1.5$ nA). Each area in the images shows the change taking place during a scan of about one minute. Type-P defects are indicated by red dots. A type-C defect is indicated by C. The dimer rows which fluctuated between the $c(4 \times 2)$ to $p(2 \times 2)$ arrangements are indicated by red arrows.

adjacent dimer rows are indicated by red arrows. As is indicated by red dots in Fig. 2, there exist some defects on dimer rows which have a structure similar to that of the type-C defect. However, their contrast resembles that of other dimers, as opposed to that of the type-C defect. We call the new type of defect at the boundary the type-P defect. From a comparison of the two images, type-P defects were found to move during a scan, which resulted in a structural change in the arrangement of the dimer rows with respect to their two adjacent dimer rows. Therefore, the observed structural change is considered to be caused by the movement of type-P defects along dimer rows at 6 K. In fact, as shown in Figs. 1 and 2, the structural change between the $c(4 \times 2)$ and $p(2 \times 2)$ arrangements seem to occur one-dimensionally along the dimer rows rather than two-dimensionally. In addition, dimer rows appeared to change their phase independently of the arrangement of the dimer rows with respect to their two adjacent dimer rows, whether it be $c(4 \times 2)$ or $p(2 \times 2)$, which resulted in nearly equivalent change observed between the $c(4 \times 2)$ and $p(2 \times 2)$ arrangement areas.

(a) c(4x2)   **(b) p(2x2)**

(c) C+P   **(d) P+P**
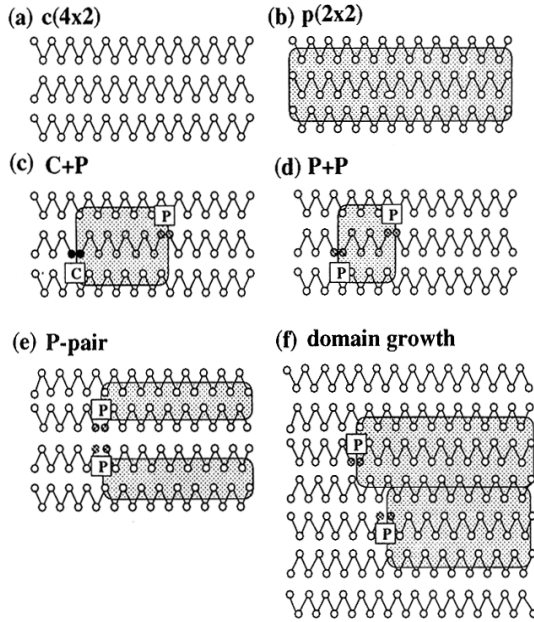
(e) P-pair   **(f) domain growth**

Fig. 3. Schematic structural models for the Si(100) surface: (a) $c(4 \times 2)$, (b) $p(2 \times 2)$, (c) type C+P, (d) type P+P, (e) a pair of type P, and (f) $p(2 \times 2)$ domain growth. Only the upper Si atoms of the buckled dimers are illustrated using circles. Lines between the circles are drawn in order to compare the phase shift, and $p(2 \times 2)$ arrangement areas are dotted.

The tip effect must be considered; however, since each dimer was scanned 15 to 20 times in order to obtain the image, we conclude that if the effect had played an important role in the observed change, the image would have been less stable. The change was observed on both upper and lower terraces in Fig. 1, where dimer rows were parallel and perpendicular to the scanning direction. No relationship between the scan direction and the dimer row direction is observed.

Figure 3 shows schematic structural models for the Si(100) surface with type-P defects. Only the upper Si atoms of the buckled dimers are illustrated using circles. Lines between the circles are drawn in order to compare the phase shift, and $p(2 \times 2)$ arrangement areas are dotted.

Since the type-C defect acts as a phase shifter, when it exists in the $c(4 \times 2)$ arrangement area, an out-of-phase structure with a type P defect on the other end, is induced at one side of the defect. An anti-phase domain induced by a type-C defect was observed to be unstable at 120 K–140 K. The instability of one anti-phase domain in a $c(4 \times 2)$ arrangement area (Fig. 3(c)) is assumed to be the reason behind the observation of apparently symmetric dimers at 140 K.[2, 4] However, if the dimer flip-flop motion is frozen at low temperature, the $p(2 \times 2)$ phase is expected to appear as shown in Fig. 3(c). In such a case, the other end of the anti-phase boundary has a type-P defect whose structure is similar to that of the type-C defect. The type-P defect is not necessarily fixed in position like the type-C defect; it may move along the dimer row. In any case, when a type-C defect exists, an out-of-phase structure is induced at one side of the defect. An example of a type-C defect which induced a

type-P defect is shown in Fig. 2(a).

Figure 3(d) shows a model of a pair of type-P defects created on a dimer row. Thereby, an anti-phase domain similar to that caused by the type-C defect is formed. In this case, since both ends of the domain are formed by type-P defects, both boundaries may fluctuate and cause the observed structural change. A significant difference between these two models is that the pair of type-P defects in the latter model can be annihilated when they meet each other. An example of pair formation of the type-P defect is shown in Fig. 2(b) as P_{p}. Type-P defects may be introduced at step edges, an example of which is shown in Fig. 2(b) as P_3.

We also observed the Si(100) surface at 80 K, where type-P defects were found to exist.[6, 7] However, when two type-P defects exist in neighboring rows, they form a pair, which maintains the $c(4 \times 2)$ structure on both sides of the defects and between the two rows, as shown schematically in Fig. 3(e). Therefore, the $p(2 \times 2)$ arrangement could not be enlarged at 80 K, and the observed surface had a structure similar to that observed at 120–140 K. An anti-phase domain of a dimer row which forms a $p(2 \times 2)$ arrangement with the two adjacent rows (Figs. 3(c) and 3(d)) is considered to be unstable at 80 K. Two type-P defects in neighboring rows may interact to form a pair that reduces the area of the complete $p(2 \times 2)$ arrangement (Fig. 3(e)).

On the other hand, phase change in a dimer row, i.e., fluctuation of type-P defects, occurred almost independently of the arrangement of the dimer row with respect to the two adjacent dimer rows, whether it be $c(4 \times 2)$ or $p(2 \times 2)$. In fact, two type-P defects, P_1 and P_2, on the two neighboring dimer rows moved while maintaining the $p(2 \times 2)$ arrangement between the two dimer rows, as shown in Figs. 2(a) and 2(b). Change in the phase of dimers near the type-C defect caused by P_1 is indicated by black dots in Figs. 2(a) and 2(b). They might have formed a pair as shown in Fig. 3(e) at 80 K. The weak interaction between two type-P defects in neighboring rows may be due to the fact that the $p(2 \times 2)$ arrangement at the boundaries is stable at 6 K. Thereby, reduction of the $p(2 \times 2)$ arrangement area by pair formation of type-P defects shown in Fig. 3(e) is not necessary at 6 K. Comparison of the properties of the type-P defects observed at 6 K and 80 K will be made elsewhere in detail.[6–8]

Giving consideration to the obtained results, the observed structural change between $c(4 \times 2)$ and $p(2 \times 2)$ arrangement is due to the independent movement of type-P defects along dimer rows at 6 K. When one dimer row changes its buckling phase in a $c(4 \times 2)$ arrangement area, the area between that row and its two neighboring rows changes in phase from $c(4 \times 2)$ to $p(2 \times 2)$, as shown in Figs. 3(c) and 3(d). Therefore, when anti-phase domains are formed in alternating dimer rows, the $p(2 \times 2)$ arrangement area will be enlarged in the direction perpendicular to the dimer rows (Fig. 3(f)).

A possible explanation for the stability of the $p(2 \times 2)$ arrangement at 6 K is that the barrier height for the transition from the $p(2 \times 2)$ phase to the $c(4 \times 2)$ phase, which is very low,[9] almost zero at ~ 100 K, works more effective at 6 K. As shown in Fig. 1, the area with $p(2 \times 2)$

arrangement was comparable to that with $c(4 \times 2)$ arrangement. Considering the fact that the energy difference between the $c(4 \times 2)$ and $p(2 \times 2)$ reconstructions obtained by theoretical calculations is only on the order of $1 \, \text{meV}$ per dimer,[10] in order to clarify the stability of the $p(2 \times 2)$ arrangements at $6 \, \text{K}$, more detailed analysis is necessary.

In conclusion, the apparently symmetric dimers observed at high temperatures were frozen at $6 \, \text{K}$, and $p(2 \times 2)$ structure was observed to be more stable compared to that obtained at $120 \, \text{K}$–$140 \, \text{K}$. A change between the $c(4 \times 2)$ and $p(2 \times 2)$ arrangements was caused by the fluctuation of type-P defects, and when antiphase domains were formed in alternating dimer rows, it was enlarged in the direction perpendicular to the dimer rows, which resulted in the observed structural change.

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