

## Spontaneous Fluctuation between Symmetric and Buckled Dimer Domains of Si(100) at 80 K

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At 80 K, not all the dimers of Si(100) appear buckled in the scanning tunneling microscopy (STM) images but a certain number of the dimers are observed in a symmetric configuration. We report on observations of a two-dimensional spontaneous fluctuation of the symmetric $\leftrightarrow$ buckled dimer domains at some particular locations. We interpret the spontaneous fluctuation to be induced by the competition of several antiphased  $c(4 \times 2)$  buckled domains to expand. The fluctuation of domains was interpreted by two mechanisms: a fast switching between buckled dimer domains; and symmetric dimers induced by migration of P defects.

KEYWORDS: Si(100), dimers, fluctuation, STM, P defect, C defect

### 1. Introduction

At room temperature, most of the dimers of Si(100) appear in a symmetric configuration in the scanning tunneling microscopy (STM) images.<sup>1)</sup> The observed apparent symmetric dimers in the STM images were first considered to support the concept of symmetric dimers. However, many experimental<sup>2)</sup> and theoretical<sup>3)</sup> studies revealed buckled dimers to be more stable than symmetric dimers, and at present, it might be safe to conclude that the concept of buckled dimers is well accepted. Indeed, below 200 K, most of the dimers are observed in a buckled configuration in the STM images.<sup>4)</sup> In order to interpret the apparent symmetric dimers observed by STM at room temperature within the framework of buckled dimers, the concept of flip-flop motion was introduced.<sup>5)</sup> Buckled dimers flip-flop much faster than the scanning rate of STM at room temperature, providing an apparent symmetric dimer in the STM images, while the flip-flop motion is frozen at low temperatures, hence the dimers appear buckled.

As the surface temperature is lowered below the critical temperature ( $\sim 200$  K) of the symmetric $\leftrightarrow$ buckled phase transition, buckled dimer domains nucleate and grow.<sup>6–12)</sup> Defects and steps on the surface critically influence the nucleation and growth of buckled dimer domains in a sophisticated manner, for instance, step edges suppress the formation of buckled dimer domains,<sup>6)</sup> A defects<sup>7)</sup> serve as growth nuclei, and kink sites induce buckling.<sup>8)</sup> What is more important is that there exist two types of  $c(4 \times 2)$  domains which are antiphased with respect to each other. Consequently, the surface does not reconstruct to a single domain, but is divided into many small antiphased domains. Many of the boundaries of the antiphased  $c(4 \times 2)$  buckled dimer domains appear as symmetric dimer domains in the STM images. Therefore, even at temperatures far below 200 K, not all the dimers are buckled, but a certain ratio of dimers appear symmetric which are induced by the defects and boundaries.<sup>9)</sup> The surface is divided into buckled and symmetric dimer domains.

In this article we report on the observation of spontaneous

fluctuations between the symmetric $\leftrightarrow$ buckled dimer domains of Si(100) at 80 K. The spontaneous fluctuation is observed only at some particular regions of the surface. In most cases, the spontaneous symmetric $\leftrightarrow$ buckled transition can be interpreted as a result of competition among several antiphased  $c(4 \times 2)$  buckled domains to expand. During each spontaneous transition, several dimer rows change their configuration in an orchestrated manner: the observed transition is two dimensional. We present two different approaches to explain the observed fluctuation between dimer domains.

### 2. Experiment and Results

Si samples were phosphorus-doped with a conductivity of  $0.005 \Omega\text{cm}$ . After the samples were prebaked at  $\sim 700^\circ\text{C}$  for 12 h, they were flashed once to  $1250^\circ\text{C}$  for 30 s, followed by a slow cooling. The base pressure was kept under  $5 \times 10^{-8}$  Pa during flashing. Electrochemically etched tungsten tips were used for the STM observations.

We begin with an STM image of Si(100) (Fig. 1) to demonstrate the coexistence of symmetric and buckled dimer domains at a low temperature (80 K). In the top-left region, the dimers appear in a symmetric configuration, while in the bottom right region the dimer rows appear as zigzagged chains. The zigzagged chains reflect the antiferromagnetic alignment of buckled dimers, since only the upper or lower atoms of the dimers are observed in STM images depending on whether the filled or empty state is probed (STM images presented in this article are of the empty states). This ordering provides a surface reconstruction of  $c(4 \times 2)$ .

In Fig. 1, there exists a C defect at the boundary of the buckled-symmetric dimer domains<sup>13)</sup> indicated by an arrow. When the filled states are probed (not shown), the C defect appears to be two adjacent Si atoms missing along the dimer row direction while it is observed as a bright protrusion in the empty states.<sup>13)</sup> The C defects act as phase shifters in a complete set of buckled dimers,<sup>9)</sup> and frequently the C defects are observed at the boundaries of buckled and symmetric dimer domains. It can be considered that the boundaries and the shape of domains are regulated by these defects.

Spontaneous fluctuations between the symmetric-buckled dimer domains were observed at some particular locations.

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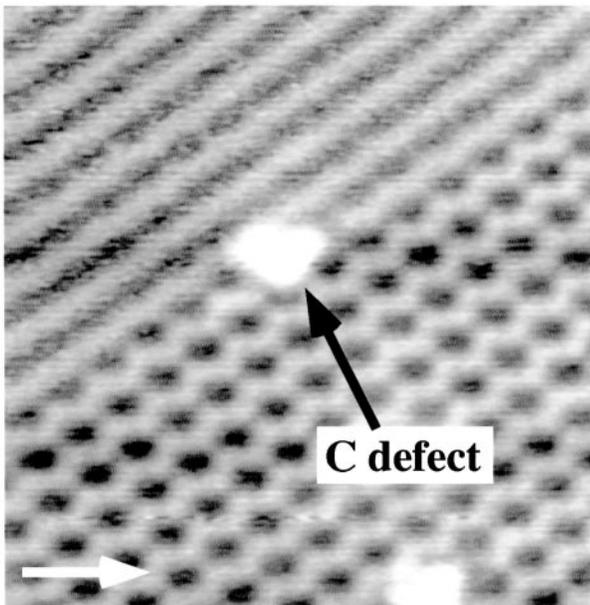


Fig. 1. An STM image of Si(100) at 80 K showing a boundary of a symmetric-buckled dimer domain. The white protrusion in the middle is a C defect. The scan direction is displayed by the arrow. Tunneling conditions: surface bias = +0.8 V; tunneling current = 1 nA. The scanning time is about 35 sec for one image ( $512 \times 512$  pixel). The displayed image shows the whole scanned image.

An example is displayed in Fig. 2. In the initial stage shown in Fig. 2(a), a wide (large) symmetric dimer domain exists in the middle (colored dark). The symmetric dimer domain is surrounded by several buckled dimers domains, and many of the boundaries seem to be regulated by defects, particularly the C defects. There is a step edge and a bounding lower terrace in the bottom-right region which is not shown. We did not observe any kind of fluctuation in domains which extended beyond the step edges. After 4 min, the first transition was observed. As shown in Fig. 2(b), the symmetric dimer domain observed in the initial state was converted into a buckled dimer domain, and a small symmetric dimer domain (colored dark) emerged in the center. Again it appears that this small symmetric dimer domain was regulated by some C defects. As displayed in the set of STM images of Fig. 2, we observed an endless switching of the surface morphology between these two particular configurations of Figs. 2(a) and 2(b) during the period of observation.

Some ambiguity exists concerning the attribution of buckled and symmetric dimers (colored dark) at some locations. When the symmetric  $\leftrightarrow$  buckled boundaries are not regulated by the defects, the configuration of the symmetric  $\leftrightarrow$  buckled dimers seems to change gradually to a buckled/symmetric configuration, making a definite determination of the dimers configuration difficult. This is why the shape of the large and small symmetric dimer domains in Fig. 2 are displayed as if they change their shape after each transition, however we believe that their configurations basically remain unchanged. It must be noted that the spontaneous symmetric  $\leftrightarrow$  buckled phase transition is not observed everywhere but only occurs at some particular regions. The observed endless switching of the surface morphology between the two configurations suggests that the two configurations, however different they are, have a similar total free energy, and thermal fluctuation

between the two configurations is observed as a spontaneous transition.

An example of a more complicated spontaneous transition of the dimers is displayed in a set of consecutive STM images in Fig. 3. In this case, switching between two particular configurations was not observed, but changes in surface configuration with each transition were observed. Figure 3(a) shows the initial configuration. The white protrusions are the C defects. There are two medium sized symmetric dimer domains colored dark on the left side. Two types of  $c(4 \times 2)$  buckled dimer domains antiphased with respect to each other are hatched with horizontal and vertical lines. After the first spontaneous transition shown in Fig. 3(b), the medium sized symmetric dimer domain at the upper-left region converted into a buckled dimer domain and was absorbed into the buckled dimer domain on the right (hatched with horizontal lines). By the second transition, the medium sized symmetric dimer domain at the bottom-left region extended and the buckled dimer above it was divided into two [Fig. 3(c)]. After 42 min, the final transition occurred and the most dramatic change was observed [Fig. 3(d)]. During the course of the transition, the large buckled dimer domain in the middle converted into a symmetric dimer domain. This transition is quite different from that displayed in Fig. 2 in the sense that the configuration of the surface changes after each transition and does not switch between two particular configurations. During the course of observation, the surface did not revert to its original state. Transitions of this type were observed very rarely.

### 3. Discussion

We propose two different explanations for the observed spontaneous fluctuation. The two explanations are different in the interpretation of the symmetric dimers observed in the STM images at 80 K, a temperature far below the critical temperature (200 K) of the symmetric  $\leftrightarrow$  buckled phase transition.

The first explanation attributes the origin of symmetric dimer domains to fast switching of buckled dimer domains, which we refer to as “symmetric dimers induced by switching of buckled dimer domains” in the following. Two domains of  $c(4 \times 2)$  exist which differ in the phase alignment of the buckled dimers. The site  $(i, j)$  is defined to belong to the  $(\pm)$  phase when  $(-1)^{i+j} S_{i,j} = \pm 1$ , where  $S_{i,j} = \pm 1$  corresponds to the two possible configurations of the buckled dimer. Usually a symmetric dimer domain is sandwiched between two buckled dimer domains which belong to different phases. Therefore, it is possible to consider that the symmetric dimer domain emerges as a result of fast switching between the surrounding  $(+)$  and  $(-)$  phased buckled dimer domain<sup>11,12)</sup> as shown schematically in Fig. 4(a). This concept was introduced from theoretical researches using Monte Carlo simulations to study the dynamical characteristics of the dimers.<sup>10–12)</sup>

Another interpretation of the symmetric dimer domains is that numerous phasons nominated as P defects migrate along the buckled dimer rows much faster than the scanning rate of STM where the dimers appear symmetric.<sup>14,15)</sup> This mechanism is referred to as “symmetric dimers induced by migration of P defects”. In such a case, basically the dimers are buckled, though they appear symmetric in the STM images. The P defects are two adjacent dimers buckled in the same direction (ferromagnetic ordering), and they act as phase defects when located in the antiferromagnetic ordered buck-

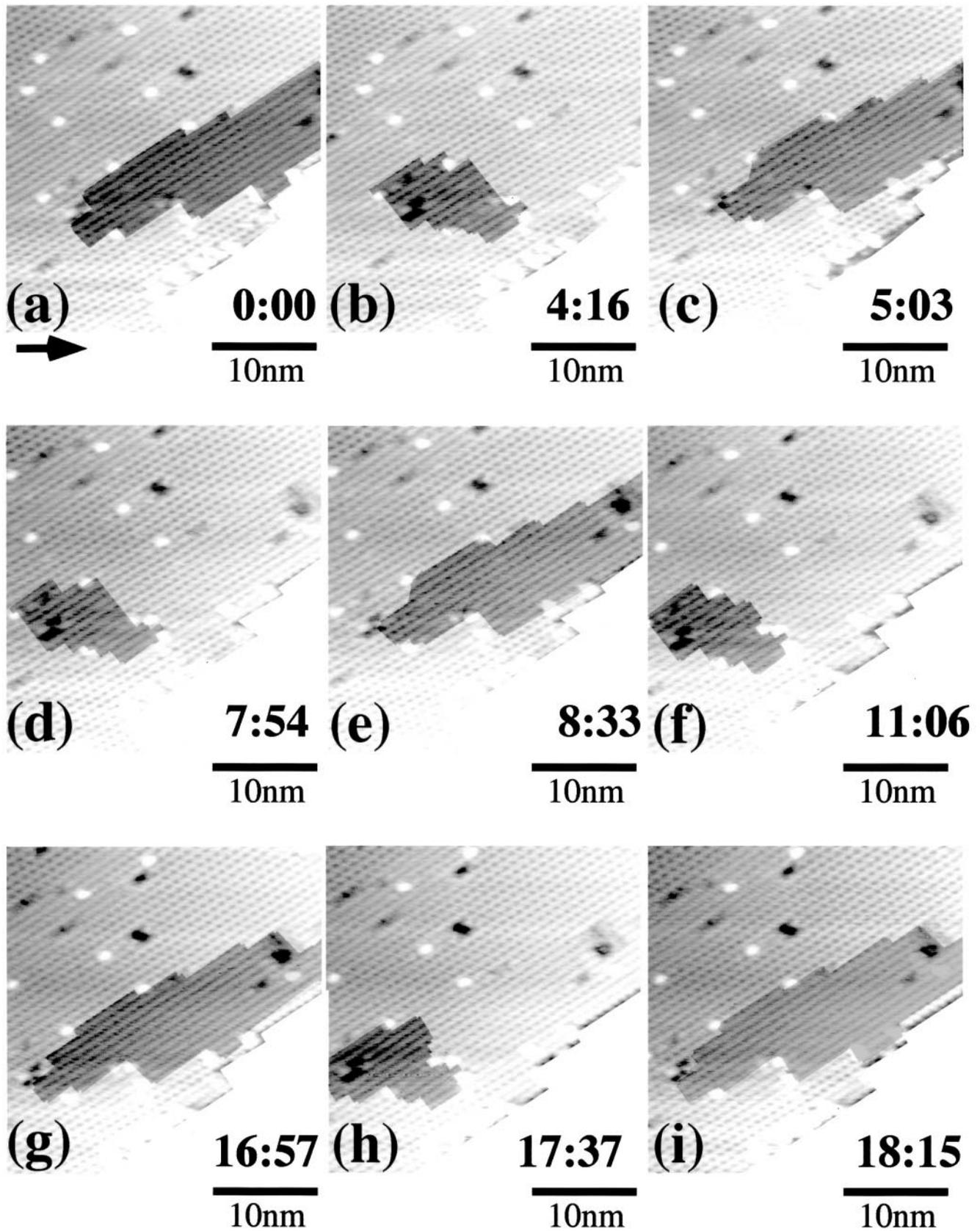


Fig. 2. A set of consecutive STM images of Si(100) at 80 K showing a spontaneous fluctuation of symmetric-buckled dimer domains. The dark colored regions represent apparent symmetric dimer domains. White protrusions are the C defects. Tunneling conditions: surface bias = +0.8 V; tunneling current = 1 nA. Scanning scale = 30 nm. The arrow shows the scanning direction. The scanning rate is about 35 s for one image. The time displayed indicates the duration in minutes and seconds from the end of scanning of Fig. 2(a) to the end of scanning of each image.

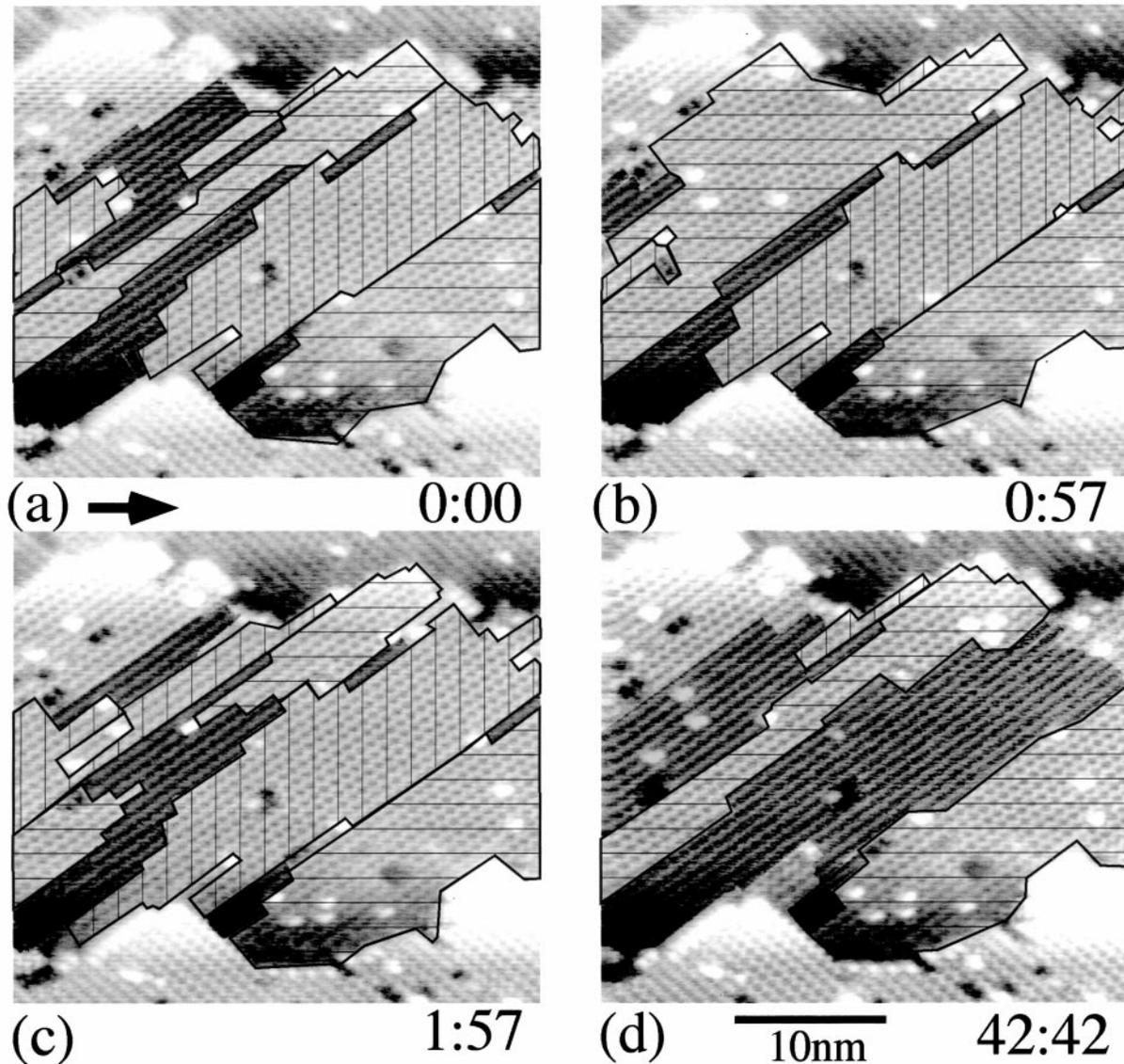


Fig. 3. Another example of a set of STM images of Si(100) showing a spontaneous fluctuation of symmetric-buckled dimer domains. Tunneling conditions: surface bias = +0.6 V; tunneling current = 1.5 nA. Scanning scale = 30 nm. The arrow shows the scanning direction. The scanning rate is about 35 s for one image. The time displayed indicates the duration in minutes and seconds from the end of scanning of Fig. 3(a) to the end of scanning of each image.

led dimers in the  $c(4 \times 2)$  phase as shown schematically in Fig. 4(b). The P defects are movable and do not appear as bright protrusions when probing the empty states. Existence of the P defects was first directly confirmed by STM observations at 6 K.<sup>16)</sup> At 80 K, P defects migrate much faster than the scanning rate of STM. Therefore, areas where the P defects exist and migrate should appear as symmetric dimers in the STM images as shown in Fig. 4(b). At 80 K, symmetric dimer domains can be regarded as a “sea” of these phasons imprisoned in a particular area by the C defects and the surrounding antiphased  $c(4 \times 2)$  buckled dimer domains.

Interpretation of the observed transition of the dimer domains shown in Fig. 2 by the two approaches is a simple and straightforward procedure. In Fig. 5 we show the schematics of the large [Fig. 5(a)] and small [Fig. 5(b)] symmetric dimer domains of Fig. 2 with the surrounding buckled dimer domains. Careful analysis of the phase of the buckled dimers reveals that there exist two antiphased buckled dimer domains [(A) hatched with horizontal lines], and [(B) hatched with ver-

tical lines] surrounding the symmetric dimers domain. The boundary of the two buckled dimer domains is observed as symmetric dimer domains [regions (C) and (D)] which is regulated by the surrounding C defects.

We interpret that the observed spontaneous transition is induced by the competition of the two buckled dimer domains (A) and (B) to expand. The power balance between the two domains should be delicate enough to fluctuate either by perturbation of the scanning tip or thermal fluctuation. From the standpoint of symmetric dimers induced by fast switching of buckled dimers, the fluctuation of the symmetric dimer domains represents a change in the region switching between domains (A) and (B). A transition from the configuration of Fig. 5(a) to Fig. 5(b) means that the buckled dimer domain (A) has expanded. The transition observed in Fig. 3 can be explained in a similar manner. From the standpoint of symmetric dimers induced by migration of P defects, some P defects migrate along the dimer rows in regions (C) and (D). The observed spontaneous transition is a result of a change of

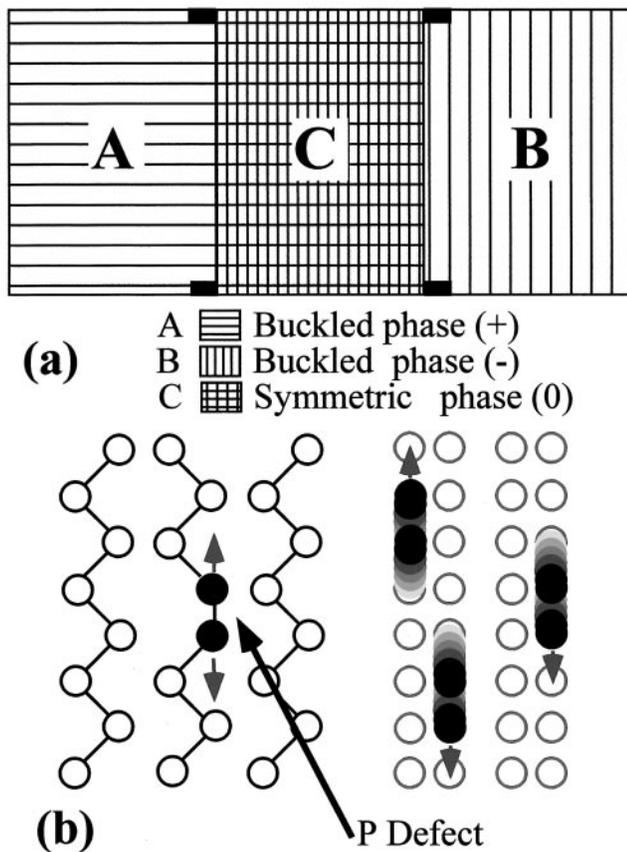


Fig. 4. (a) A schematic showing apparent symmetric dimer domains caused by a fast switching between two  $c(4 \times 2)$  antiphased buckled dimer domains. Domains (A) and (B) represent buckled dimer domains in the (+) and (-) phase, respectively. The boundaries of domain (C) in between (A) and (B) are regulated by the defects represented by the black boxes in the corners. The buckled dimers in domain (C) switching fast between the (+) and (-) phases, and as a result observed as symmetric dimers by STM. (b) A stick and ball schematic showing the structure of the P defects (black circles). The circles represent the down (up) atoms of the buckled dimers. The P defect is composed of two buckled dimers aligned in the same direction. It is highly mobile as indicated by the arrows. Accompanied with the migration of the P defect, the adjacent buckled dimer flip-flops to the other configuration. The schematic on left side shows many P defects migrating along the dimer rows resulting in an apparent symmetric configuration of dimers in the STM images.

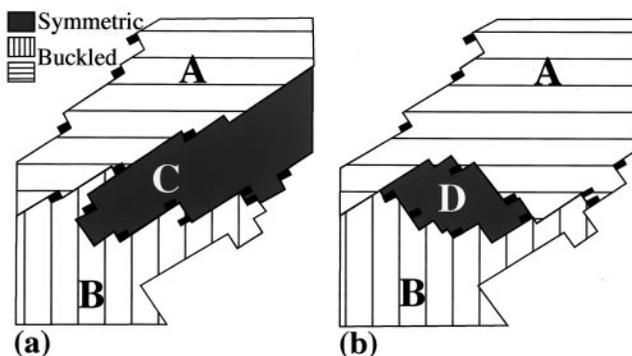


Fig. 5. Schematics showing the configuration of dimers and the location of the C defects at the boundaries of the surface area of Fig. 2. The C defects at the boundaries are represented as dark spots. (a) Schematic showing the configuration of the large symmetric dimer domain, and (b) the small dimer domain. Regions hatched with horizontal and vertical lines are antiphased  $c(4 \times 2)$  buckled dimer domains, while regions colored black represent symmetric dimer domains.

from the standpoint of symmetric dimers induced by switching, because individual switching between buckled dimer domains must occur very fast (if not, symmetric dimer domains would not be observed at low temperatures in the first place) and should be two-dimensional. On the other hand, from the standpoint of symmetric dimers induced by migration of P defects, this aspect is more complicated to explain. We assume that a strong interaction exists among the symmetric dimer rows, the origin of which we attribute to a strong attractive interaction between the P defects in adjacent symmetric dimer rows. Indeed, at 80 K, it is reported that the P defects in adjacent dimer rows have a strong tendency to form pairs.<sup>17)</sup> We interpret that the observed two-dimensional transition is induced by a concord migration of the P defects which originates from the strong attractive interaction.

Further work is required to give a complete atomic scale description of the transition and to combine the two different mechanisms presented here.

#### 4. Conclusion

To conclude, we have observed a spontaneous fluctuation between buckled and apparent symmetric dimer domains on Si(100) at 80 K by using a scanning tunneling microscope. We interpret that most of the fluctuations are induced by a sophisticated competition among the neighbouring antiphased the area in which the P defects migrate with ease. A transition from the configuration of Fig. 5(a) to Fig. 5(b) means that the P defects in region (C) are confined to the small region (D) accompanied with the expansion of region (A).

Defects seriously influence the configurations of the dimer domains and thus the transitions. Configuration of dimers on surfaces with low densities of C defects (1%~ similar to our experiments) has been studied by Monte Carlo simulations.<sup>10-12)</sup> Defects on the surface were demonstrated to significantly influence the configuration of the dimers in the STM images. What is more important, Nakamura *et al.* have shown that the influence of a combination of several defects is much stronger than the sum of the influence of isolated defects.<sup>11)</sup> When several defects are located in a particular configuration (in their calculations four C defects at the corners of a rectangle), the influence of the defects extends to the adjacent dimer rows and the regions surrounded by defects show a strong tendency to appear as symmetric dimers in the simulated STM images.<sup>11)</sup> We interpret that the middle region of (D) where dimers always remain symmetric is such a location.

Some commonly observed aspects of the transition provide important implications on the interaction between dimers and defects. It is important to note that the intervals between transitions are on the time scale of a few minutes, while the transition is accomplished instantly when compared to the time scale of scanning of STM (~msec.). We call it an avalanche type transition; the rate limiting process is to start the transition and once the transition is triggered, the accompanied transformation of dimers is quickly accomplished (which makes it difficult to analyze the details of the transition by STM). In the course of each transition, many dimers change their configuration in an orchestrated manner in a very short time, while once the transition is accomplished they remain in the same configuration for a long time. Moreover, the transition extends to the adjacent dimer rows: the transition is two-dimensional. It is very simple to understand this aspect

$c(4 \times 2)$  domains to expand, where the boundaries of the domains are observed as symmetric dimer domains. We present two different approaches to explain the observed fluctuation between domains: a fast switching between buckled dimer domains and symmetric dimer domains induced by migration of P defects.

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