PHYSICAL REVIEW B VOLUME 57, NUMBER 8 15 FEBRUARY 1998-II

Suppressive influence of steps on a phase transition of the Si(001) surface

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(Received 29 September 1997)

We have studied an order-disorder phase transition of a vicinal Si(001) surface from a 2×1 to a $c(4\times2)$ structure using low-temperature scanning tunneling microscopy (STM). The order parameter for the $c(4\times2)$ structure is derived from the Fourier transform of the atomically resolved STM images in the 65–300 K temperature range. The order parameter on the *A* terrace bounded with an S_A step is found to reach only 0.7, even at 65 K, while the $c(4\times2)$ structure is almost complete on the *B* terrace bounded with an S_B step. In the STM images at 65 K, dimers at the lower side of the S_A step still appear symmetric, resulting in an order-parameter reduction of the *B* terrace. [S0163-1829(98)51308-2]

The atomic details of a Si(001) surface have been widely studied, because of their technological and scientific potential. The Si(001) surface is known to undergo an order-disorder phase transition from a 2×1 to a $c(4\times2)$ structure at cooling.[1,2] In scanning tunneling microscope (STM) images of the 2×1 structure at room temperature, dimers appear to be symmetric due to the time average of the flip-flop motion of asymmetric (buckled) dimers allowed by thermal excitation.[3] However, below $T_c\approx200$ K, the dimers are buckled and arranged in an antiferromagnetic order, resulting in the $c(4\times2)$ structure formation.[1,2,4,5] Detailed studies of the phase transition behavior were made using a low-energy-electron diffraction (LEED) method[1,2] and Monte Carlo (MC) simulations[6–9] based on an Ising-spin model. These studies concluded that this is a second-order phase transition, because no hysteresis was detected between cooling and heating.

Recently, STM observations of the Si(001) surfaces below 200 K have been performed. They have revealed atomic details of the well-ordered $c(4\times2)$ structure[5,10] and the influence of surface defects on the buckling.[4,5,10,11] Smith *et al.*[12] reported that the area of the $c(4\times2)$ structure increased as surface temperature decreased to below about 200 K from their STM images in a temperature range of 127–300 K. However, even at 127 K, only about 40% of the surface was arranged into the $c(4\times2)$ structure because of the high-defect density (about 10%).[12] More recently, the local influence of individual defects, such as *A*-type and *C*-type defects,[13] has been directly observed by low-temperature STM.[10,11]

In addition to the defects, the influence of surface steps must also be studied, since the steps cannot be removed even if the surface is highly oriented in the [001] direction. In particular, the alternate configuration of S_A and S_B steps[14] on the vicinal Si(001) surface miscut towards the [110] direction is very interesting because these steps divide the surface into two types of terraces, *A* and *B*, where dimer rows run parallel and perpendicular to the steps, respectively (see Fig. 1). These terraces are known to have an anisotropic stress due to the dimerization of the first-layer atoms.[15] Moreover, different structures of the S_A and S_B steps also induce the local stress on these terraces. We therefore expect different behavior in the phase transition between the *A* and *B* terraces, due to the anisotropic stress.

In this paper, we use low-temperature STM to study the influence of steps on the buckling and the related phase transition behavior of a vicinal Si(001) surface. We find that the $c(4\times2)$ structure is incomplete at the lower side of the *A* steps, even at 65 K. On narrow terraces of around 15 nm, the influence of the S_A step results in a reduction of the order parameter to about 0.7 at 65 K.

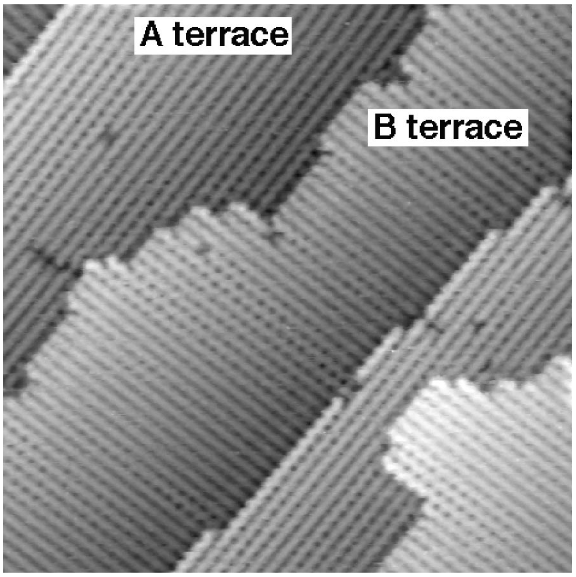

FIG. 1. Filled-state STM image on the vicinal Si(001) surface at 196 K extending over $29\times29\ \mathrm{nm}^2$. The *A* and *B* terraces, where dimer rows run parallel and perpendicular to the steps, alternately exist. The partial ordering of the buckled dimers appears near the upper side of the S_A steps, the kinks, and the *A*-type defects.

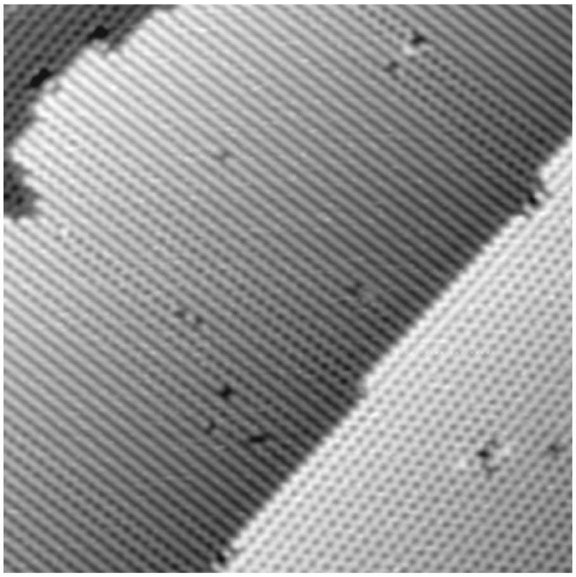

FIG. 2. Filled-state STM image on the vicinal Si(001) surface at 177 K extending over $33\times33\ \mathrm{nm}^2$. Most of the A terraces exhibit the $c(4\times2)$ ordering, while the ordering was restricted to rows near the kinks of the S_A steps and the A-type defects on the B terrace

We used silicon wafer (Sb doped with $0.05-0.09\ \Omega$ cm) miscut towards the [110] direction by 0.5°. The silicon sample was cleaned by flashing at 1450 K after being degassed for 20 h at 850 K in an ultrahigh vacuum chamber. The sample was slowly cooled to room temperature (3 K/s) to obtain a regular step distribution, and then transferred to a low-temperature STM. The sample was maintained for over 6 h at each temperature (65–300 K), then STM observations were made. After each observation, the sample was cleaned by flashing for use at the next temperature. All filled-state STM images were obtained by a constant-current mode of 100 pA with a sample voltage of -1.5 V.

The vicinal surface has the two types of striped terraces, A and B, with an average width of about 15 nm each, as seen in Figs. 1, 2, and 3. The surface defects remained, but were estimated to be as low as 1% of the surface area, most of which appear as the A-type defects. The A-type defects induce characteristic buckling of dimers near the defects at around $T_c\approx200$ K, but their influence disappears at 78 K, as described elsewhere.[11] Although C-type defects have been reported to disrupt the $c(4\times2)$ ordering even well below T_c,[9,10,16] they were rarely found in our sample.

In our STM images of the room-temperature surface (not shown), symmetric-appearing dimers formed the 2×1 structure. We confirmed previous observations[3,13,17] that dimer rows at the upper edge of the S_A steps were buckled in addition to those which meet with kinks of the S_A or S_B steps. In the STM image of Fig. 1, around $T_c\approx200$ K, the buckled-dimer rows near the S_A step edges and the kinks were ordered in the $c(4\times2)$ structure. In addition, the buckling of dimers occurred preferentially around the A-type defects.[11]

At 177 K, about 20 K below T_c, the buckled dimers were arranged in an orderly $c(4\times2)$ phase on the S_A terrace, as shown in Fig. 2. However, the buckling of dimers on the B terrace was restricted to rows near the A-type defects and the kinks. A comparison of Figs. 1 and 2 shows that the symmetric-appearing dimers extended vertically from the ledge of the straight S_A steps. Even at 65 K, the dimers still appear to be symmetric at the lower side of the S_A steps in Fig. 3. Thus, these STM images show that the $c(4\times2)$ ordering proceeded differently on the A and B terraces. Furthermore, the ledge of the straight S_A steps inhibited dimer buckling on the B terrace.

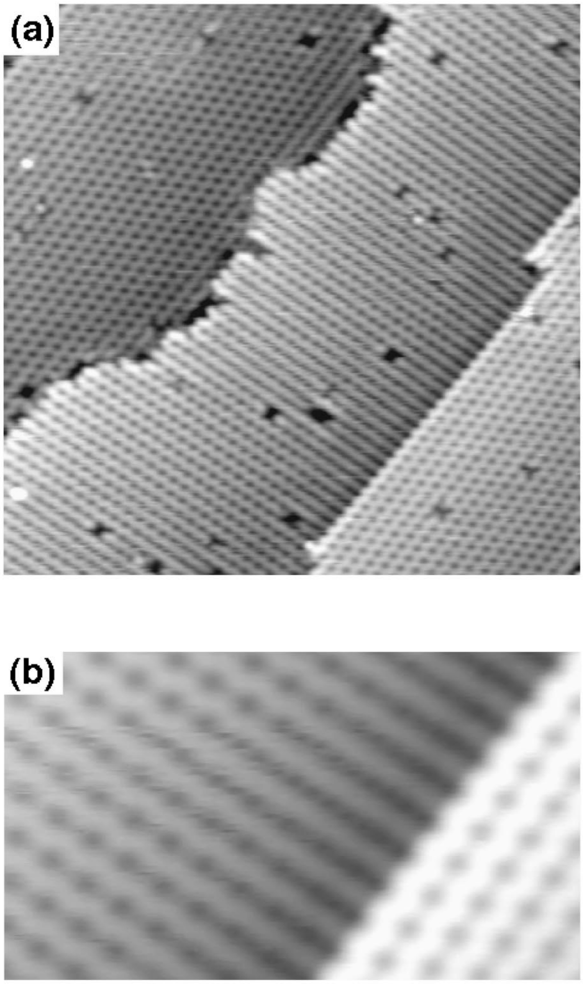

FIG. 3. Filled-state STM image on the vicinal Si(001) surface at 65 K extending over (a) $29\times29\ \mathrm{nm}^2$ and (b) $9\times5\ \mathrm{nm}^2$. The $c(4\times2)$ ordering proceeded on both the B terrace and the A terrace, in contrast with Figs. 1 and 2. However, the symmetric-appearing dimers still remained at the lower side of the straight S_A steps on the B terrace.

In order to see the different ordering on the A and B terraces, we performed a Fourier transform[18] of our STM images. We measured the intensity ratio of the (1/4,1/2) reflection spot to the (1/2,0) reflection, and subtracted the diffuse backgrounds from the reflection intensity, similarly with the LEED analysis.[2] The intensity ratio corresponds to the relative area of the $c(4\times2)$ structure. Assuming that each dimer has a scattering power of f_{nm} at the (n,m) site of the 2×1 unit cell on the surface, the intensity of the $(h/4,k/2)$ reflection is given by

$$I(h/4,k/2)=\left[\sum_{n,m} f_{\mathrm{nm}}\exp\{\pi i(hn+km)\}\right]^2.$$

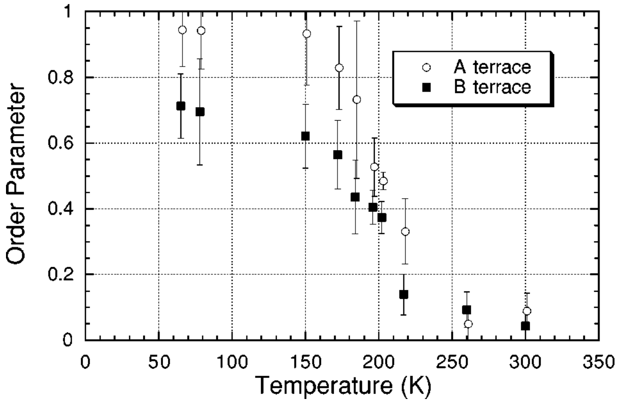

FIG. 4. Temperature dependence of our defined order parameter for the $c(4\times2)$ structure derived from the Fourier transform of the STM images. We employed 5 or 6 STM images of $30\times30-60\times60\ \mathrm{nm}^2$ to obtain the order parameter. Circles indicate the *A* terrace and squares indicate the *B* terrace. The standard deviation is shown as the error bar.

If the scattering power is assumed to be f_+ and f_- for the two different orientations of the buckled dimers and $f_0=(f_++f_-)/2$ for the symmetric-appearing dimers, then the intensity of the (1/2,0) reflection for the complete $c(4\times2)$ phase is calculated as

$$I_0(1/2,0)=4f_0^2,$$

and that of the (1/4,1/2) reflection is

$$I_0(1/4,1/2)=4(f_+-f_-)^2.$$

For the partially ordered phase, the intensity of the (1/2,0) reflection is still $I_0(1/2,0)=4f_0^2$, and is independent of the degree of the $c(4\times2)$ ordering, while the intensity of the (1/4,1/2) reflection is

$$I(1/4,1/2)=\left[\sum_{n,m}\delta(f_{\mathrm{nm}})\exp\{\pi i(n+m)\}\right]^2,$$

where $\delta(f_{\mathrm{nm}})$ is $(f_+-f_-)/2$ or $(-f_++f_-)/2$ for the buckled dimers and zero for the symmetric-appearing dimers. Since $I(1/4,1/2)$ is zero in the region of the 2×1 and $p(2\times2)$ structures, $I(1/4,1/2)$ almost reflects the $c(4\times2)$ area. Thus, the intensity ratio, $I(1/4,1/2)/I(1/2,0)$, gives the relative areas of the $c(4\times2)$ phase, independent of the black-white contrast and the scan size of the original STM images. Because the order parameter of the complete ordered phase is expected to reach 1.0, the intensity ratio was, then, normalized by $I_0(1/4,1/2)/I_0(1/2,0)$ to give the order parameter. In the STM images, it is difficult to distinguish the areas of the $c(4\times2)$ and 2×1 phases, because the intermediate regions, i.e., slightly buckled dimers, exist between the two phases. Therefore, we found that the relative area of the $c(4\times2)$ phase directly derived from the STM images is somewhat smaller than the order parameter, due to the intermediate regions.

In Fig. 4, we plot the temperature dependence of the order parameter, defined as the normalized $I(1/4,1/2)/I(1/2,0)$. The critical temperature T_c, the midpoint of the transition region, was estimated to be about 200 K on both *A* and *B* terraces. However, this figure shows that the ordering proceeded more slowly on the *B* terrace than on the *A* terrace. Moreover, even at 65 K, the ordering occurred on only about 70% of the *B* terrace, while the rest was comprised of symmetric-appearing dimers. As mentioned above, the symmetric-appearing dimers were found on rows extending from the ledge of the straight S_A steps on the *B* terrace in Figs. 2 and 3.

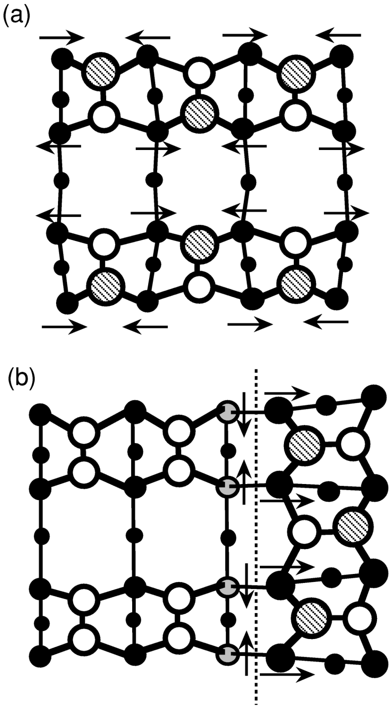

FIG. 5. Top view of (a) the $c(4\times2)$ structure and (b) the S_A step. Hatched atoms indicate buckled atoms. The $c(4\times2)$ structure is stabilized by displacement of the second-layer atoms, as denoted by arrows in (a). Larger circles indicate the upper side of the step in (b). Second-layer edge atoms at the upper and lower side of the step edge interact with each other through the lattice strain shown by arrows. The lattice strain induces the buckling of dimers at the upper side of the step, even at room temperature. At the lower side, the lattice strain of the second-layer edge atoms (shaded circles) inhibited dimer buckling, as shown in Figs. 1, 2, and 3.

The symmetric-appearing dimers of the *B* terrace seem to originate in the lattice strain on the edge of the S_A step. Basically, the buckling of dimers is stabilized by the small displacement of the second-layer atoms, as shown in Fig. 5(a).[11,19] At the S_A step, the second-layer atoms on both the upper and lower terrace edges interact with each other through the dimerization of the first-layer atoms, as illustrated in Fig. 5(b). The lattice strain of the second-layer atoms at the step edge can either reduce or raise the energy barrier height between the two different orientations of the buckled dimers. Even at room temperature, the lattice strain causes the upper-edge dimers along the S_A steps to buckle[3] due to an increase in the barrier height. However, at the lower side of the S_A steps, the lattice strain of the second-layer edge atoms, denoted by shaded circles in Fig. 5(b), seems to reduce the barrier height, because these atoms are fixed by the step edge. The reduction in the barrier height allowed the flip-flop motion of the lower-edge dimers of the straight S_A steps even at 65 K, and the influence was extended to 5–15 dimers along dimer rows, as shown in Fig. 3.

As a result, the local influence of the S_A steps smeared out the transition of the B terrace, compared to that of the A terrace, as shown in Fig. 4.

In summary, we directly observed the suppressive influence of the S_A steps and their related phase transition behavior on the vicinal Si(001) surface in a temperature range of 65–300 K using low-temperature STM. In addition, the order parameter, which was derived from the Fourier transform of the images, revealed the different phase transition characteristics between the A and B terraces. The strain field of the second-layer atoms at the S_A steps inhibited the dimer buckling at their lower side even at 65 K, and the order parameter reached only about 0.7 on the B terrace with an average width of about 15 nm.

We would like to thank M. Okamoto for useful discussions. We also thank Q. Ru for several comments about the Fourier transform of the STM images.

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