Scanning-Tunneling-Microscopy Study of Single-Domain Si(001) Surfaces Grown by Molecular-Beam Epitaxy

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Initial stages of Si epitaxial growth on vicinal Si(001) substrates were investigated using scanning tunneling microscopy. For a growth temperature of about 750 K it was found that initial growth occurs almost exclusively at one of the two nonequivalent types of step edge. This leads to the formation of a single-domain surface with an array of evenly spaced straight steps with biatomic height. This structure can be preserved by quenching the sample to room temperature.

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The interest in the use of silicon molecular-beam epitaxy (Si MBE) on Si(001) surfaces for the production of novel electronic devices has motivated many studies of the initial stages of growth and the influence of the preceding cleaning treatment. One topic of interest has been the preparation of single-domain surfaces, where the steps between adjacent terraces have a height of an even number of monolayers (ML). This is important in reducing the amount of antiphase boundaries in III-V films grown on Si substrates.[1] It has been reported in the literature that a prolonged annealing of samples with a misorientation less than 0.2° at about 1250 K may result in a single-domain surface.[2-5] On samples with a misorientation of more than about 4° towards [110], biatomic height steps were found after cleaning and annealing at about 1150 K.[6-8] During MBE on a sample with a misorientation of 0.5° towards [110], a single-domain reflection high-energy electron diffraction pattern was observed.[9,10] However, Sakamoto *et al.*[10] mentioned that this single-domain surface is not stable. When growth is terminated and the sample is kept at the growth temperature of about 750 K, this surface changes back into a two-domain surface within a few minutes.[10]

We present scanning-tunneling-microscope (STM) images of epitaxially grown single-domain Si(001) surfaces that were quenched to room temperature immediately after termination of growth.

The STM uses a mechanical approach[11] and is described in more detail elsewhere.[12,13] It is mounted in an ultrahigh-vacuum chamber with a base pressure of 1×10^{-8} Pa, together with a low-energy electron diffraction (LEED) system. Si can be evaporated *in situ* from resistively heated Si strips at a growth rate of about 1 ML/min, as calibrated with Rutherford backscattering spectrometry on carbon substrates, and at a pressure better than 2×10^{-7} Pa. The 5×20-mm^2 substrates were cut from commercially available wafers (Wacker, floating zone, *n* type, 1 Ω cm) and ultrasonically rinsed in ethanol before loading into the vacuum chamber. The misorientation, as determined by x-ray diffraction, was 0.52° in the [110] and 0.09° in the [$\bar{1}$10] directions, respectively. Inside the chamber the samples were heated resistively. Temperatures were measured with an infrared pyrometer with an accuracy of 50 K. After outgassing the sample and the holder for several hours, the sample was thermally cleaned at 1450 K for 2 min and cooled at an average rate of 25 K/min. Si films have been deposited on these clean substrates at 750 K. Immediately after deposition the sample was quenched to room temperature and transferred to the microscope. STM images were taken with a negative sample bias of 2 V and a tunneling current of 0.5 nA.

As discussed elsewhere,[14] we found that annealing at about 1450 K is necessary to obtain a clean surface with a regular step distribution reflecting the macroscopic misorientation of the sample. In Fig. 1 an STM image of a clean surface with four monatomic steps is shown. The step-down direction is from the upper right to the lower left. The orientation of the dimer rows rotates over 90° at every step, as expected for a monatomic step. It can be seen that the shape of the step depends on the

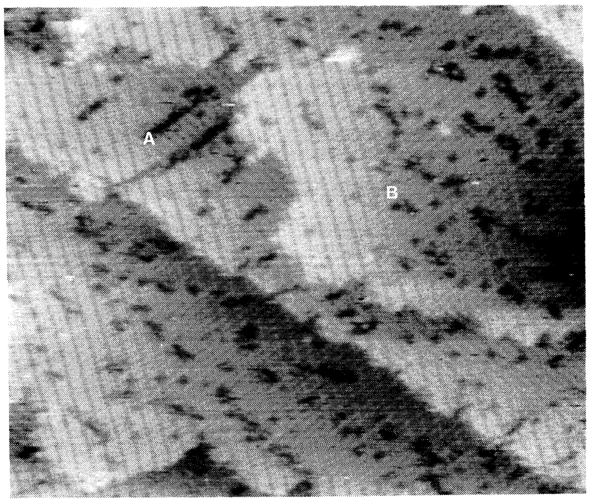

FIG. 1. 50×45-nm^2 area of clean Si(001) surface.

corresponding dimer-row orientation: The steps parallel with the dimer rows in the upper terrace are straight whereas the others are ragged. Following Chadi's notation,[15] we call these two inequivalent terraces and steps type *A* and type *B*, respectively, as indicated in the figure.

Figure 2 shows a 100×100-nm^2 scan of a surface on which approximately 0.5 ML of Si has been grown. The type-*A* terrace is almost completely covered by the type-*B* terrace and in most areas the type-*B* steps coalesced with the *A* steps, creating an array of straight steps with a biatomic height. Large-area scans taken on several areas of the same sample showed that this single-domain structure extended over the whole surface. This was confirmed by LEED images showing a single-domain pattern. The fact that the deposited 0.5 ML of Si has fully covered the type-*A* terrace implies that growth occurs almost exclusively at the type-*B* step edge, i.e., at the end of the dimer rows. This preferential growth effect is illustrated in Fig. 3, showing a 120 $\times135$-nm^2 scan of a Si(001) surface after deposition of about 0.2 ML. A quantitative determination of the type-*A* and -*B* terrace areas indicated that the fraction of the surface covered by type-*B* areas increased about 20%. This fact and the observation that no type-*B* islands exist imply that the deposited material has been incorporated almost completely at the type-*B* step edge.

This effect may be ascribed to a different reactivity at the two inequivalent step edges, caused by the different bonding geometries. This can be correlated to a difference in step formation energy for the two types of steps. In a simple picture, where long-range effects are neglected, a difference in formation energies must be due to a difference in binding energies between adjacent dimer pairs, depending on their relative orientation. The step with the dimers oriented in the energetically most favorable orientation would be the most stable one[16] and have the lowest formation energy. Using semiempirical tight-binding-based total-energy calculations, Chadi[15] has found that the formation energy for a type-*A* step is significantly lower than for a type-*B* step. If we neglect the long-range effects, this would imply that the binding energy is larger in the direction perpendicular to the dimer bond. This is consistent with our observation that growth occurs preferentially at the ends of dimer rows.

Note that during the formation of this single-domain structure diffusion over a step edge must occur: Since all material is concentrated on a single terrace the material deposited on the other terrace must have moved over a terrace edge and must have diffused over up to the entire terrace width.

It was suggested by Stoyanov[17] that the existence of an easy diffusion direction on Si(001) might induce a splitting of the terrace width distribution at certain growth temperatures. During initial growth at these temperatures nucleation of islands would only occur on the terraces with the easy diffusion direction parallel to the step edge whereas on the other step the deposited material would get incorporated at the step edge. This would cause the terrace on which no islands develop to grow faster. However, on images from samples with about 0.2 ML of Si deposited (see Fig. 3), no islands were observed on the slowly growing terrace. Thus, we have to conclude that this effect is of minor importance under the growth conditions in our work. This is consistent with results of Lagally *et al.*[18] on growth at tem-

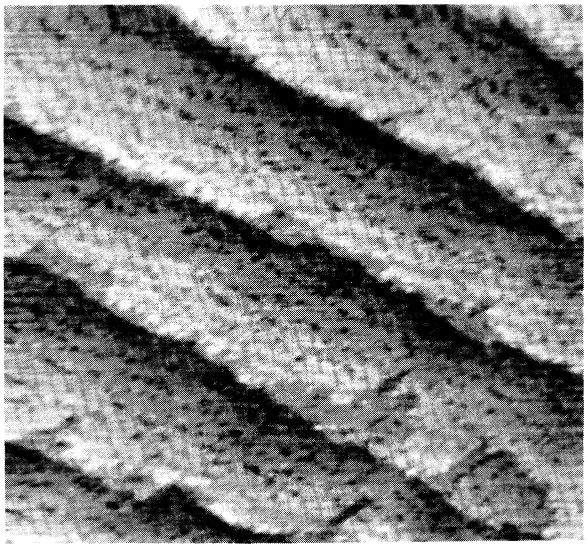

FIG. 2. 100×100-nm^2 area of Si(001) surface after deposition of 0.5 ML of Si.

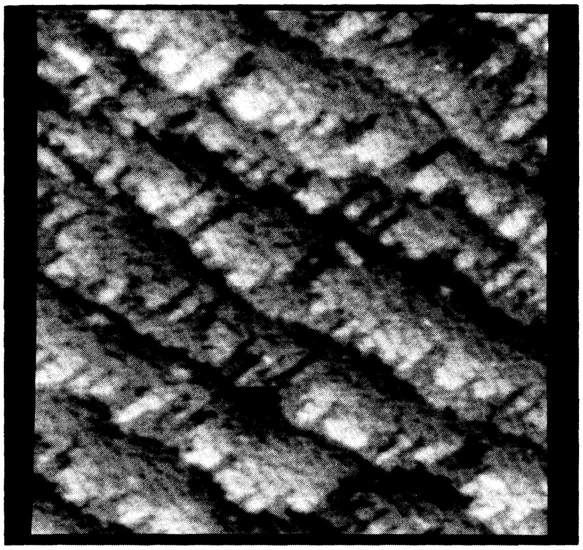

FIG. 3. 120×135-nm^2 area of Si(001) surface after deposition of 0.2 ML of Si.

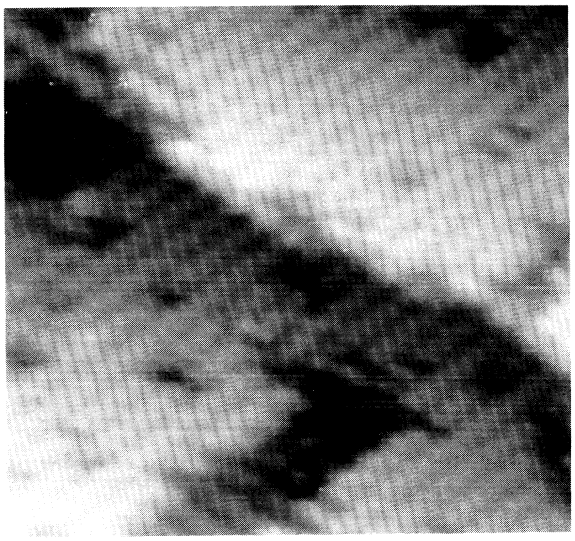

FIG. 4. $14\times14\text{-nm}^2$ area of Si(001) surface after deposition of 0.5 ML of Si showing the double step in more detail.

peratures below 570 K.

The single-domain growth effect can only be due to a difference in energy gain for the two nonequivalent edges, favoring growth at the ends of dimer rows. At the few kinks in the type-*A* steps (see Fig. 1) equivalent end positions exist. Due to the much higher density of these positions at the type-*B* step edges growth will mainly occur there. During an anneal at the growth temperature the surface may come back to a two-domain surface[10] by incorporating the excess material at the kinks in the type-*A* steps.

In the bottom right-hand corner of Fig. 2 a few type-*A* areas can still be seen. Three different explanations can be given for this observation. Firstly, the growth might locally be blocked due to a contamination. Secondly, this structure may be due to a too slow quenching of the sample. Since the single-domain structure is not stable at the growth temperature, some areas might already be partially changed back into a two-domain structure. Thirdly, it is possible that the amount of Si deposited is somewhat less than 0.5 ML. However, in images from samples with about 1.5 ML of Si deposited the type-*A* areas were still observed.

Figure 4 shows a detailed image of a biatomic step with resolved separate dimers. Note that the dimer rows are oriented along the same direction on both sides of the step. The structure of parts of the double step shown in this image strongly resembles the structure of the biatomic step on a vicinal surface shown by Wierenga, Kubby, and Griffith,[7] which was found by Chadi[15] to be energetically more favorable than a combination of two monatomic steps. However, in our case this biatomic step structure does not exist on the whole step length.

In summary, STM images show that at temperatures around 750 K growth occurs preferentially at one type of step edge, resulting in a single-domain surface with a periodic parallel array of straight biatomic steps. Anisotropic diffusion plays an underlying role. The single-domain surface can be preserved by quenching to room temperature.

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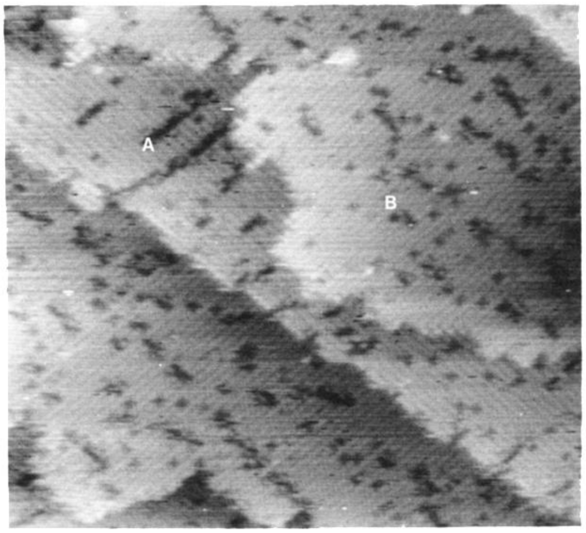

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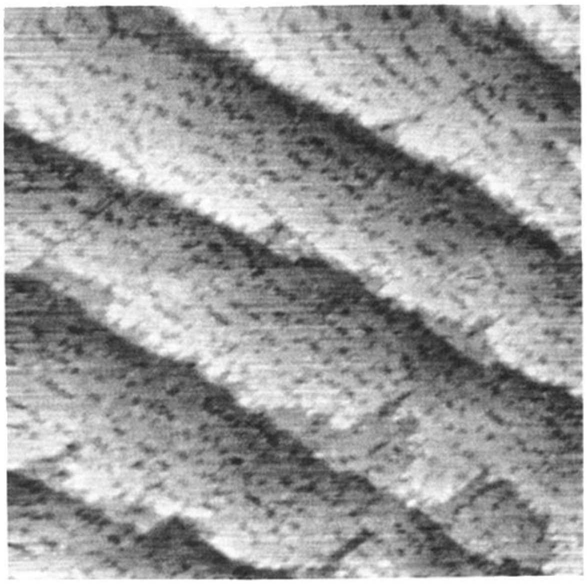

FIG. 2. $100\times100\text{-nm}^2$ area of Si(001) surface after deposition of 0.5 ML of Si.

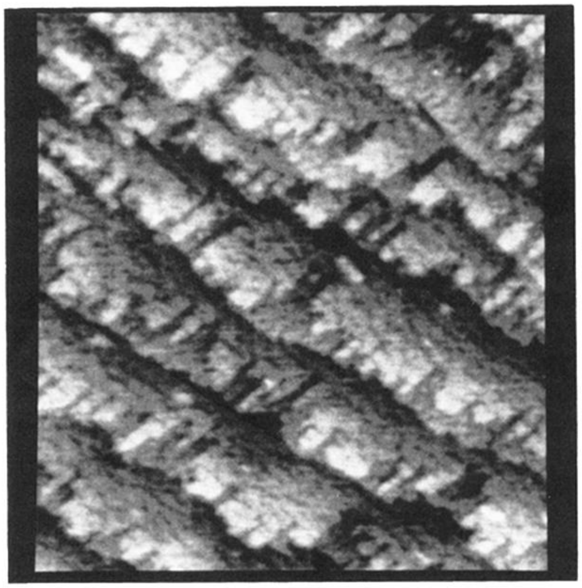

FIG. 3. $120 \times 135\text{-nm}^2$ area of Si(001) surface after deposition of 0.2 ML of Si.

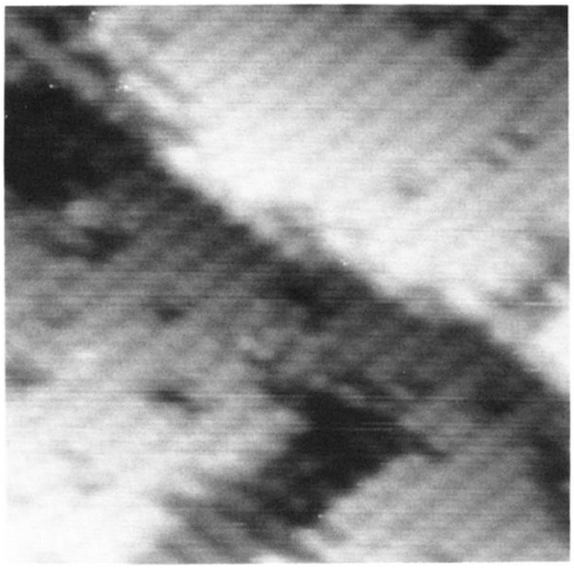

FIG. 4. 14×14-nm^2 area of Si(001) surface after deposition of 0.5 ML of Si showing the double step in more detail.