Initial-Stage Structural Change of Si(100) Surface Induced by Ethylene Gas and Annealing

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The initial stage of the structural change in a clean Si(100)-2 × 1 surface induced by annealing at 640°C and exposure to ethylene gas was studied by reflection high-energy electron diffraction (RHEED) and scanning tunneling microscopy (STM). The RHEED pattern included SiC spots and STM images revealed SiC particles on the surface, which confirmed the carbonization of the Si surface. At a different area of the same sample, the RHEED pattern showed both twice the periodicity of surface spots which indicates a flat surface region and transmitted bulk Si spots which indicates the initial stage of voids. The STM images of the flat region showed a 2 × n (6 ≤ n ≤ 12) reconstruction. The topography of the 2 × n STM image depended on the bias voltage. The 2 × n reconstruction was clearly induced by carbon impurities; STM images were similar to those in previous studies in which structures were formed by various kinds of impurities or contaminations. [DOI: 10.1380/ejssnt.2006.285]

Keywords: Reflection high-energy electron diffraction; Scanning tunneling microscopy; Surface structure, morphology, roughness, and topography; Silicon; Silicon carbide; Alkenes

I. INTRODUCTION

Silicon carbide (SiC) is a wide-gap (ca. 2.35 eV) semiconductor that has potential for technological applications [1–4]. One conventional approach to the growth of 3C-SiC(100) films on Si(100) is chemical vapor deposition (CVD) using exposure to hydrocarbons in tandem with surface annealing [5]. However, three crucial problems with epitaxial SiC formation on the Si(100) surface have been reported.

The first problem is interfacial voids occurring in the growth, observed by transmission electron microscopy (TEM) [6–14]. This phenomenon was discovered by Mogab and Leamy [15] upon cross-sectional observation of a reacted Si sample by scanning electron microscopy (SEM). The activation energy of the Si atom diffusion in bulk was estimated to be 5–7 eV [16–18]. Moreover, it was estimated that the number of Si atoms in the produced SiC film was almost the same as that originally in the voids [19]. Sholtz, et al. [14] showed how to prevent voids in a SiC layer less than 3 nm thick. In order to prevent void formation at thickness greater than 5 nm, it becomes necessary to supply Si atoms in the carbonization process [20] and to add a surfactant such as germanium [10, 21, 22] to stabilize the interface.

The second problem is twin formation [12–14, 23, 24]. Prevention of the twin formation is necessary for the production of high-quality crystalline film. In the case of metals, it is known that crystal growth at low temperatures leads to twin formation. In the case of SiC growth on silicon, it is impossible to eliminate twin formation by annealing alone, since the melting point of silicon is lower than that of SiC. In order to prevent twin formation of 3C-SiC at low temperatures, it is critical to supply a surfactant to prevent the cis-type conformation on the -(Si-C)₃- rings in the crystal.

The third problem is the location of the carbon atoms adsorbed on the Si(100) substrate after the annealing [25, 26]. In the first stage carbonization of a Si(100) surface, 2 × n (6 ≤ n ≤ 12) [27, 28] and c(4 × 4) [27–43] reconstructions have been reported. However, similar 2 × n [44–58] and c(4 × 4) [53–83] reconstructions have been reported without carbonization. It is noteworthy that X-ray photoelectron diffraction (XPD) studies [5, 26, 36–40] have indicated an absence of carbon atoms on the outermost layer of the c(4 × 4) surface induced by carbon. Some investigators showed that carbon, including hydrocarbons and the impurities, plays a crucial role in surface reconstruction of 2 × n [27, 28] and c(4 × 4) [26, 29–42, 57, 76–78, 80, 81], though the carbon concentration on the surface is insufficient for carbon atoms to form a component of the c(4 × 4) structure [78]. Before investigating hydrocarbon impurity-induced structural changes in clean Si(100) surfaces and elucidating problems in epitaxial SiC formation on Si(100) surfaces, study of the structural change in clean Si(100) surfaces at the first stage of carbonization is necessary.

In the present work, structural changes in clean Si(100)-2 × 1 surfaces reacted with ethylene gas were studied with RHEED and STM. More specifically, this investigation focused on the structural change from clean 2 × 1 to 2 × n. Although the reported STM images of a carbon-induced 2 × n structure [27, 28] seem similar to those of structures without carbon induction [47–58], this is not evidence that the structures themselves are the same; differences in bias voltage gave rise to some uncertainty. In fact, our results confirmed the bias energy dependence of STM images.
II. EXPERIMENTS

All the experiments were conducted in an ultrahigh vacuum (UHV) chamber containing an RHEED electron gun (Biemtron, RHG-303), a phosphor screen, and an STM (Omicron, UHV-SPM). The base pressure of the UHV chamber was less than $2 \times 10^{-8}$ Pa. The residual gas in the UHV chamber was checked by mass spectrometer, and more than 98% of the residual gas was hydrogen. Details of the apparatus are reported elsewhere [84, 85].

Si samples were cut from polished p-type Si(100) wafers with a thickness of 0.4 mm and a resistivity of 10-20 Ωcm. After ultrasonic cleaning for 5 minutes in ethanol, a sample was mounted on a molybdenum holder with tantalum clips. In order to prevent metal contamination of the sample, the sample holder contained no stainless steel. No metal materials or particles of any kind ever touched the samples; teflon tweezers were used to pick them up. The mounted sample was loaded into the vacuum chamber, pre-baked at 200°C for 2 hours, and returned to room temperature, and the base pressure of the chamber was less than $1 \times 10^{-8}$ Pa. The sample was then flushed at 1250°C for several seconds several times to obtain a clean surface, as checked by RHEED. The vacuum pressure during this annealing procedure was maintained below $2 \times 10^{-8}$ Pa. The temperature of the sample during the annealing was measured with an infrared radiation thermometer (Chino). After observation of the clean 2 × 1 surface by RHEED and STM, the sample was exposed to ethylene gas for 3 minutes at $5 \times 10^{-5}$ Pa. During this exposure, the sample was annealed at 640°C. After the sample had cooled to room temperature, the surface structure was checked again with RHEED and STM.

The acceleration energy of the RHEED electron gun was 30 keV. The diameter of the RHEED screen was 200 mm, and an ICF253 viewing port for the screen was used in this study. When the acceleration energy was higher than 15 keV, the relative intensity of Kikuchi patterns [86] increases. A rotation sector with the pattern of $\theta = \sqrt{r}$ in the polar coordinates was mounted in front of the screen to compensate for the intensity of the RHEED patterns. The typical rotation speed of the sector was 60 rpm. The distance between the screen and the rotation sector was 100 mm, and the distance between the sector and a digital camera (Olympus, C-3040) was 500 mm. Typical exposure time with the digital camera was 4-8 s at $f = 8 - 10$ with an ISO of 80-100. More details about the rotation sector were described elsewhere [87].

All STM images were taken using a constant current mode. A cut Pt/Ir wire was used for the STM tip. The scanning speed and the tunneling current of all images were 250 nm/s and 20 pA, respectively. It is noted that the vector scanning system allowed us to rotate the scanning direction. No correction of the obtained STM images, including drift compensation, was performed.

III. RESULTS

First, the RHEED pattern and STM image of the clean Si(100)-2 × 1 surface were examined. Figure 1 (a) shows the 2 × 1/1 × 2 double-domain RHEED pattern. The incident direction was [011] and the incident angle almost satisfied the 400 Bragg condition (a); the same conditions except that the incident angle almost satisfies the 200 Bragg condition (b); and STM image of the same surface (c). The electron energy of the RHEED gun was 30 keV. A $\sqrt{r}$ sector was mounted in front of the RHEED screen to compensate for the contrast of the RHEED patterns. The STM image was observed at a sample bias voltage of 1.5 V and a tunnel current of 20 pA, and the image size is 50 nm × 50 nm.
FIG. 2: Si(100) surface after 3 min. exposure to ethylene gas at $5 \times 10^{-5}$ Pa with simultaneous annealing at 640°C: RHEED pattern (a) and STM image (b). The STM image size is 100 nm x 100 nm. The height of the STM image is shown with a gray scale, indexed at the right side of the image. The other experimental parameters of the RHEED pattern and the STM image are the same as those in Fig. 1.

dent angle by 1.0°, almost satisfying the 200 Bragg condition, the intensity of the surface spots was maximized, as shown in Fig. 1(b). Careful observation of the spots along the half Laue zone revealed kinematically forbidden extra spots; the RHEED pattern seems to indicate a $2 \times 2$ structure [88]. Because a slight change in the incident direction continuously moved the extra spots, they were determined not to be bulk spots but surface spots [89]. The extra spots are attributed to double diffraction from the $2 \times 1$ and $1 \times 2$ domains [88]. Figure 1(c) shows a typical STM image of the $2 \times 1/1 \times 2$ double-domain structure with some defects, which is in good agreement with a previous study [90]. The whole area of the sample surface was checked by RHEED, and the obtained RHEED patterns were the same as those shown in Figs. 1(a) and (b).

After exposing the surface at 640°C to ethylene gas at a pressure of $5 \times 10^{-5}$ Pa for 3 minutes, SiC spots were observed on the RHEED pattern. Figure 2(a) shows the RHEED pattern, in which the intensity of the $2 \times 1$ surface spots decreased, and two SiC spots appeared. Notably, the SiC transmission spots disappeared when the incident angle was increased, a fact which agrees with findings in a study by Miki, et al. [77, 78]. The contrast of the Kikuchi pattern was blurred compared with that of the clean surface, which indicates that electrons emerging from the bulk were scattered by the surface with increasing roughness, though this explanation is not quantitative. In the same area of the RHEED observation, SiC particles with a diameter of several nanometers were observed in the STM image, as shown in Fig. 2(b). The average size of the SiC particles shown in Fig. 2(b) is 15 nm, which is in agreement with our previous study [91].

In a different area, 3 mm distant from the area shown in Fig. 2, no SiC spots were recognized and the contrast of the Kikuchi pattern was clearer. Figure 3(a) shows a typical RHEED pattern, in which both bulk Si spots and 2-times periodic surface spots appeared. Although the transmitted bulk spots indicate that rough regions, corresponding to voids, are in the observed area, the 2-times periodic surface spots and the clear Kikuchi pat-
FIG. 4: Sample bias voltage dependence of $2 \times n$ ($6 \leq n \leq 12$) STM image at the same area as that shown in Fig. 3. The sample bias voltage of each STM image is indicated at the upper left in the corresponding image. The size of each STM image is 50 nm $\times$ 50 nm. The scanning direction of the STM images shown has been rotated 45° compared with those shown in Figs. 1-3, and the crystalline directions are indicated in the upper left image (2 V). The other experimental parameters of the STM image are the same as those in Figs. 1-3. A $2 \times 8$ unit cell is imposed in the figure in the upper left image (2 V).

The STM image shows more than fifty steps run along the [001] direction, and the average step width is 15 nm. Magnified STM images at various sample bias voltages for the same area as shown in Fig. 3, are shown in Fig. 4. At sample bias voltages from 2 V to 1.25 V, stripes with the distance corresponding to the number $n$ of the $2 \times n$ structure can be recognized. When the sample bias voltage was decreased to 1 V, the morphology of the observed image changed. Dark patches appeared on the image, and the breadth of the patches corresponds to the number $n$ of the $2 \times n$ structure. Using our STM apparatus, it was experimentally difficult to obtain STM images at an absolute value of bias voltage lower than 1 V since the tip hits the surface at lower voltages, even decreasing the feedback setting of the tunneling current. At a sample bias voltage from -1 V to -1.25 V, the same dark patch seen at the bias voltage of 1 V was observed. At sample bias voltages from -1.5 V to -2 V, the surface morphology gradually changed and dark lines appeared. Though the direction of these dark lines appeared to be random and differed from those at 2 V, it was confirmed that the pseudo-random features in the STM images had the same $n$-times periodicity as those at 2 V by changing the contrast of the images and also from Fourier-transformed power spectra. From the STM image at the sample bias voltage of 2 V, the frequency of the distances was estimated. Each estimated distance derives from the value $n$. Thus, the frequency of the value $n$ was estimated. Figure 5 shows the histogram of the value $n$. The value $n$ distributes from 6 to 12, and the average is 9. The histogram is in fairly good agreement with findings in a previous study [48].

FIG. 5: Fig. 5. Histogram of the distribution of the spacing corresponding to the Si(100)-$2 \times n$ structure observed in the STM image at the sample bias voltage of 2 V, shown in Fig. 4. The average value of $n$ is 9.
IV. DISCUSSION

A. SiC particle formation

In this section we discuss the formation of SiC particles. First, the adsorption of ethylene molecules on the Si(100) surface is discussed. Yoshinobu, et al. [92, 93] studied the adsorbed state of ethylene on the Si(100) surface with electron energy loss spectroscopy (EELS), low-energy electron diffraction (LEED), and Auger electron spectroscopy (AES). According to their work, chemisorption of the unsaturated hydrocarbons occurs even at the relatively low temperature of 80 K. By heating at 650 K, ca. 40% of ethylene is desorbed, while the remainder is decomposed to CH, CH\textsubscript{2}, and SiH species [92, 93]. By heating up to 1000 K, the H adatoms are completely desorbed, and SiC is formed on the Si(100) [92, 93], outcome of which is in good agreement with results of our previous study [8]. Based on the temperature programmed desorption (TPD) study by Clemen, et al. [94], the activation energy of ethylene desorption is 38 kcal/mol, and for the di-\textalpha-ethylene-Si\textsubscript{2} complex, each Si-C bond has a strength of ca. 73 kcal/mol. On the other hand, Bozzo, et al. [95] studied the reaction of Si(100) with ethylene using a molecular beam source with X-ray photoelectron spectroscopy (XPS), EELS, and AES. Based on studies of the characteristic bulk- and surface-plasmon features in the SiC thin film, they showed a surface aggregation from bulk Si on the top layer of the growing SiC film [95]. Their result is in good agreement with the photoelectron diffraction (PED) studies [5, 26, 36–40]. Shi-momura, et al. [96] used STM and PED to study the ethylene-chemisorbed Si(100) surface without annealing during and after chemisorption. At ethylene coverage of ca. 0.5 ML, p(2×2) and c(4×2) structures were observed by STM, and the PED results shows that the chemisorbed carbon atoms are on the outermost surface [96].

Butz and Lüth [32] observed the SiC nuclei at a mono-layer of carbon by STM. The diameter of the nuclei ranged from 5 to 20 nm [32], which is in agreement with our previous study [8]. De Crescenzi, et al. [97] showed STM images of the aggregated SiC particles at a higher exposure of acetylene (1.0 – 2.5 × 10\textsuperscript{15} Torr for 10 min), combined with annealing at 650\textdegree C during exposure. The size of the particles ranged from 30 to 40 nm [97]; they were larger than those in our previous study [91].

In our previous study [91], we observed aggregates comprised of SiC particles with an average size of 17 nm. The aggregate indicates that SiC particles migrate to the surface during the annealing process. The phenomenon of particle diffusion during bulk solidification is generally known as 'separation theory' [98].

Kitabatake [99] simulated the heteroepitaxial growth of SiC on Si(100), elucidating the mechanisms of carbonization of 3C-SiC/Si(100) as the shrinkage of the [110] row of Si lattice atoms with C adatoms. The typical cross-sectional transmission electron microscope (XTEM) image supported the simulation results [99]. However, not all the SiC/Si(100) interfaces are ordered, as shown in the XTEM studies. Scholz, et al. [14] showed the disorder of the interface over several atomic layers. Moreover, the effect of two-dimensional shrinkage on epitaxial growth should be considered [100]. Not the hard-sphere model, but a model considering the covalent chemical bonds of Si-C and Si-Si would be necessary to explain the growth mechanism.

B. 2×n reconstruction

Next, we will discuss the 2×n structure shown in Fig. 4. It has been reported by several researchers that Ni impurities induce the 2×n structure [44, 46, 48, 52]. It is noted that such Ni contamination also occurs on the Si(111) surface, leading to the √19 × √19 structure [89, 101]. However, the 2×n defective structure of the Si(100) surface has also been prepared by various other methods: rapid quenching [45, 50], annealing after ion bombardment [49], thermal deposition of oxide layers [51, 53], homoepitaxial growth [47, 54, 55], exposure to 100 L (1.3 × 10\textsuperscript{-5} Pa for 1000 s) of ethylene at 700\textdegree C [41], annealing alone between 590 and 700\textdegree C for hours [56], and Ge deposition [58]. In the case of the carbon-induced 2×n reconstruction, based on XPD [5, 26, 36–40], EELS [95] and temperature-programmed desorption (TPD) [94, 102] studies revealed an absence of carbon atoms on the top layer after annealing. Consequently, we suppose in this paper that the 2×n structure model is composed solely of silicon atoms.

Some induced 2×n structural models have been proposed. Aruga and Murata [44] proposed an ordered missing-dimer defect model. Martin, et al. [45] explained the 2×n structure by the ordering of excess missing-dimer defects. Niehus, et al. [46] explained the 2×n STM image by a complex missing dimer model with one and multiple dimer vacancies. Natori, et al. [103] investigated the stability of the (2×n)-ordered missing-dimer structure and the ordering of missing dimers, concluding that the (2×n)-ordered missing-dimer structure is more stable than the (2×1)-ordered surface without missing dimers only if a compressive strain larger than 0.5% is applied parallel to the dimer row. We applied these models to our study. However, we could not explain the bias dependence of the STM images by those models. More theoretical studies might be necessary to explain the STM result shown in Fig. 4.

The model of the hydrogen-adsorbed surface can be considered a candidate for the 2×n structure; considering that hydrogen gas was found to be the major residual gas in our study, we cannot ignore this possibility. Boland [104] reviewed the hydrogen-terminated silicon surface. In his review, however, no 2×n structure of the hydrogen-adsorbed Si(100) surface was indicated. Maeng and Kim [57] prepared a 2×n structure by exposure to 100 L (5 × 10\textsuperscript{-7} Torr for 200 s) of hydrogen that contains atomic hydrogen by cracking with a hot W filament (ca. 1500\textdegree C), and subsequent annealing of the sample at 600\textdegree C. However, it is not clear that exposure to atomic hydrogen is indispensable to preparing the 2×n structure.

Our 2×n STM images were compared with those of several previous studies. Kim, et al. [27, 28] reported filled-state STM images of a 2×n structure prepared by exposure to hydrocarbon molecules, and their images are different from ours in Fig. 4; dimer rows aligned well in their images but not in ours. This is due to differences in the 'recipes'; they annealed the sample immediately after
exposure whereas we annealed during the exposure. Ikeda and Nagashima [41] prepared their sample by almost the same recipe as ours, and their $2 \times n$ STM image at 2.0 V is in agreement with ours. Hoeven, et al. [47] reported an image at -2 V that is in agreement with ours. Our STM image at 2 V is also in agreement with that reported by Sakurai, et al. [48], though they did not show the bias voltage of their images. Although the histogram they obtained was in fairly good agreement with ours, we could not observe the sub-peak at $n = 11$. The difference may be due to the sample preparation; they annealed the sample at 1200°C [48]. Feil, et al. [49], Wei, et al. [51] Lin [55], Lin and Wu [56], and Maeng and Kim [57] observed a $2 \times n$ STM image at -2 V, and their images are in agreement with those reported by Kim, et al. [27, 28]. Our STM image at 2 V is in agreement with that reported by Johnson, et al. [53], though they did not show the bias voltage of their images. Zhang, et al. [54] reported a $2 \times n$ STM image at -2 V, but they also reported that the surface contained a $c(4 \times 4)$ structure in the magnified image at a sample bias voltage of -1.4 V [54]. We checked our STM image at -1.5 V, but no $c(4 \times 4)$ periodicity was recognized. Consequently, although our $2 \times n$ STM images at 2 V and -2 V are in agreement with those reported by some researchers, no comparable data at the absolute bias voltage less than 2 V was found in the previous studies. Further investigation is necessary to conclude whether our $2 \times n$ STM images taken from the sample prepared by annealing during ethylene exposure are the same as those prepared by different methods.

C. Void formation

The formation of voids during the carbonization on silicon surface is discussed on this section. Mogab and Leamy [15] described the initial stage of void formation on Si(100) surfaces. According to their findings, the initial stage of the void formation is as follows. SiC nuclei grow more rapidly laterally than vertically with the Si reactant supplied from adjacent unreacted surface regions. At the onset of the reaction, the area fraction of unreacted Si greatly exceeds that of SiC, and impinging ethylene molecules adsorb mainly on unreacted Si. The Si reactant is removed uniformly over the surface as growth proceeds. Continued lateral growth leads to eventual impingement and coalescence of adjacent nuclei. During this growth process, the SiC surface expands at the expense of the unreacted Si surface. Consequently, adsorption of ethylene molecules occurs to an increasingly greater extent on SiC rather than on unreacted Si. Since the diffusion of Si through the region of SiC particles is quite slow, the main sources of Si for further growth become the occluded regions of unreacted Si. These in turn experience an increased demand for Si, as they must now supply a much larger area than during the initial stages of the growth. Compliance with this demand leads to void formation [15]. This description implies the idea of the prevention of void formation.

Sholz, et al. [14] introduced a method of preventing void formation. They used propane gas for carbonization instead of ethylene, and found optimal conditions for preventing the formation of micropipes and voids at the SiC / Si interface. However, they mentioned in the last sentence of their paper that in the cases in which out-diffusion defects such as micropipes and voids are prevented, the density of $\{111\}$ stacking faults and twins in the SiC layer becomes particularly high [14]. In other words, their method contributes to the second problem noted in the introduction of this paper.

The origin of void formation is open to debate. Kim, et al. [105] proposed a different formation mechanism of SiC and voids. They speculated that the voids originate from oxygen defects existing in the bulk Si wafer. However, it is difficult to prove this idea. The SiC formation process differs from that on the surface with the oxide. Though the natural oxide thin film completely desorbs at more than 1000°C [53, 106], this is the case of the surface oxides. The oxide interferes with the carbonization process, and the interference so complicates the process that it could not be adequately discussed in this paper.

In a previous paper we reported on surface and cross-sectional SEM images of voids formed on Si(111) surfaces.
[107]. We found particle nuclei, averaging 100 nm in size, at the center in every void. Considering the mechanism of the void formation discussed in the previous papers [13, 15], we concluded that the particle nuclei serve as lids to the voids, stopping the diffusion from the voids to the surface. Further investigation of the nuclei will elucidate the formation mechanism of voids though we could not address that issue in this study.

Some researchers have reported photoluminescence in SiC thin films on silicon surfaces [108–110]. In their papers, however, the existence of voids was not mentioned. Further investigation to determine whether the photoluminescence derived from SiC or voids is necessary to clarify the mechanism.

D. Reaction scheme of the carbonization

Kusunoki, et al. [8] proposed a reaction scheme for the carbonization on the silicon surface. Based on the above discussion, a new reaction scheme has been proposed. Figure 6 shows a reaction scheme of the carbonization of Si(100) surface with ethylene gas. With the exposure of ethylene gas to an Si(100) clean surface as shown in Fig. 6(a), most of the ethylene molecules scatter but some adsorb [8]. The adsorbed molecules decompose [8] and the surface is inhomogeneously covered with hydrocarbons (-CH$_4$) and seeds of SiC particles, which is schematically shown in Fig. 6(b). During the anneal at 640°C, Si atoms diffuse from unreacted area and/or pits [15] as shown in Fig. 6(b), so that the area becomes voids (domain α) as shown in Fig. 6(c). At the flat area near the voids without the seeds of SiC particles (domain β), the Si atoms from the voids in domain α pass over the domain α, which induces the diffusion of carbon atoms into subsurface and results in $2 \times n$ reconstruction. At the area with seeds of SiC particles (domain γ), the supply of the Si atoms is adequate to the exposure of the ethylene gas, so that SiC particles are grown on the domain γ. It is noted that the size of the SiC particles depends not only on the annealing temperature but also on the flux of the ethylene gas [91]. This scheme indicates that the reaction on the surface is not homogeneous, and the interaction between the domains should be considered to elucidate the reaction mechanism.

Kitabatake [99] simulated the heteroepitaxial growth of SiC on Si(100), elucidating the mechanisms of carbonization of 3C-SiC/Si(100) as the shrinkage of the [110] row of the Si lattice atoms with C adatoms. The typical cross-sectional transmission electron microscope (XTEM) images supported the simulation results [99]. However, not all the SiC/Si(100) interfaces are ordered, as shown in the XTEM studies. Sholz, et al. [14] the disorder of the interface over several atomic layers. Moreover, the effect of two-dimensional shrinkage on epitaxial growth should be considered [100]. Not the hard-sphere model, but a model considering the covalent chemical bonds of Si-C and Si-Si would be necessary to explain the growth mechanism.

V. CONCLUSION

The initial stage of the structural change in a clean Si(100)-2 $\times$ 1 surface induced by annealing at 640°C and exposure to ethylene gas has been studied by RHEED and STM. Three types of domains were observed. In domain α, transmitted bulk Si spots were observed with RHEED. The domain size was so small that the actual RHEED pattern showed the mixed pattern of transmitted bulk Si spots and twice the periodicity of surface spots attributed to the flat area near the voids. The domain α was too rough to observe with STM. The domain α is assigned to voids. In domain β, which is a region near domain α, STM images showed a flat surface with a $2 \times n$ ($6 \leq n \leq 12$) reconstruction. The STM images of the $2 \times n$ reconstructed surface depend on the bias voltage. The $2 \times n$ reconstruction was clearly induced by carbon impurities; the STM images in this study were similar to those in previous studies in which structures were formed by various kinds of impurities or contaminations. In domain γ, which is a region distant from domain α, the RHEED pattern included SiC spots and STM images revealed SiC particles on the surface. Considering the results and the previous studies, an inhomogeneous surface reaction scheme has been proposed.

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