New Types of Unstable Step-Flow Growth on Si(111)-(7 × 7) during Molecular Beam Epitaxy: Scaling and Universality

Hiroo Omi* and Yoshikazu Homma†
NTT Basic Research Laboratories, NTT Corporation, 3-1 Morinosato Wakamiya Atsugi, Kanagawa 243-0198, Japan

Vesselin Tonchev
Institute of Physical Chemistry, Bulgarian Academy of Sciences, 1113 Sofia, Bulgaria

Alberto Pimpinelli
LASMEA, UMR 6602 CNRS/Université Blaise-Pascal—Clermont 2, F-63177 Aubiére Cedex, France
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New types of unstable homoepitaxial growth of vicinal Si(111)-(7 × 7) surfaces are studied using *ex situ* atomic force microscopy. The growth features are two types of step bunching with straight step edges between 700 and 775 °C and one type of simultaneous bunching and meandering at 800 °C. The results of a quantitative size scaling analysis of the straight steps are discussed from the perspective of universality classes in bunching theory.

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Recent advancements in crystal growth techniques, such as molecular beam epitaxy (MBE) and its analogs, have enabled us to create atomically controlled materials in the growth direction on crystalline surfaces. This success in materials engineering has enabled the design of semiconductors that exhibit quantum confinement effects in the growth direction [1]. Current interest in materials design is toward the creation of artificial surface structures with laterally controlled features ranging in size from the atomic scale up to centimeters [2–5]. The main reason is that such structures have good potential as a template for self-assembling nanostructures, such as dots and wires, that exhibit quantum confinement effects.

On vicinal surfaces, it is well known that step-flow growth during MBE can spontaneously produce step patterns as a consequence of the kinetics of atomic motions at step edges and on terraces. Step instabilities, such as bunching and meandering during growth, have been used for pattern formation on semiconductor and metal surfaces, where step kinetics plays important roles [6–9]. The Burton-Cabrera-Frank continuum theory explains well the step instabilities and the resulting pattern formations on surfaces [10]. Of particular importance in the recent development of the theory [11–13] is that scaling analyses enable one to make conclusions about the mechanism leading to step bunching, which has to be fully understood for fine control of the pattern formation. To be specific, the theory is based on the assumption that some symmetry-breaking mechanism creates a nonequilibrium surface current. Then, the scaling properties of the unstable surface are determined uniquely from the dependence of the nonequilibrium surface current on the local surface slope [13]. Thus, determining all the scaling exponents allows one to know the nonequilibrium current, and hopefully the microscopic mechanisms that produce it. This scaling approach has opened up a new way for experiments to characterize the phenomena of surface kinetics, particularly step bunching [14–16]. However, the full power of scaling in investigating bunching has not yet been used in experiments. Instead, most experiments measure just the scaling behavior of one feature such as the scaling of the minimum step distance in the bunch with the bunch size. But just one feature, and thus just one exponent, is not enough for determining the universality class of bunching [10,11]. One noteworthy exception is Mühlberger *et al.*, who measured both the width and the height of ripples (bunches) formed on a Si(001) as a function of time [17].

On vicinal Si(111)-(7 × 7), which is one of the most important semiconductor surfaces [18–20], atomic steps have great potential as templates for nanostructure self-assembly [2,21]. Indeed, surface instabilities induced by direct current (dc) heating a sample (Joule effect) have been thoroughly studied [18,19]. However, step bunching induced by dc heating is an extrinsic effect. Much less is known about intrinsic instabilities during homoepitaxial growth. In fact, it has been reported that homoepitaxial step-flow growth produces step bunching on vicinal Si(111)-(7 × 7) miscut toward the $[\bar{1}\,\bar{1}\,2]$ direction during MBE [15,16,22,23]. However, there has been no systematic investigation of step bunching on Si(111)-(7 × 7) as a function of growth temperature and epitaxial layer thickness, and thus there have been very few scaling analyses of bunching phenomena [15,16].

In this Letter, we study homoepitaxial MBE on vicinal Si(111)-(7 × 7) surfaces miscut towards the $[\bar{1}\,\bar{1}\,2]$ direction. We show that during step-flow growth, three types of step bunching appear, at different growth temperatures: (i) straight edges and $[\bar{1}01]$-oriented kinks at 700 and 725 °C, (ii) microkinks composed of $[\bar{1}\,\bar{1}\,2]$ ledges at 750 and 775 °C, and (iii) considerable meandering at 800 °C. Quantitative size scaling analyses of these step patterns show that each type belongs to a different universality class. We discuss the origins of these step bunches on the basis of the universality classes concept.

The MBE chamber was equipped with a reflection high-energy electron diffraction (RHEED) apparatus. Si was deposited using a 10 kV electron-beam evaporator. Substrates were heated by a W filament placed behind the sample. Substrate temperature was measured by a pyrometer within an uncertainty of 20 °C. The ultimate pressure of the chamber was 4×10^{-11} Torr, and the pressure during Si deposition was below 3×10^{-9} Torr.

The substrates were vicinal Si(111) wafers (B-doped, 20–30 Ω cm) and were miscut 1.0°, 2.0°, 4.0°, or 5.6° to the $[\bar{1}\,\bar{1}\,2]$ direction. The miscut angles were determined to an accuracy of 0.05° by x-ray diffraction. The detailed procedures for obtaining a clean Si(111) surface are described in Ref. [8]. The Si epilayers were deposited at growth rates between 0.5 and 0.7 nm/ min and at a fixed temperature between 550 and 800 °C. During the depositions, we checked that the growing surfaces have (7×7) reconstruction by RHEED. The surface morphology was observed by tapping-mode atomic force microscopy (AFM) in air.

Figures 1(a)–1(d) respectively show representative AFM images of the Si(111)-(7×7) obtained after homoepitaxial growth at substrate temperatures of (a) 650, (b) 700, (c) 750, and (d) 800 °C. In this narrow temperature range the step pattern changed significantly, as seen in the images, and can be characterized into four types: At 650 °C and after 100 nm deposition, the surface is covered mainly with terraces and single bilayer (1 BL = 0.314 nm) steps ($h_{\mathrm{av}} = 1.6 \pm 1.0$ BL), which indicates that the growth proceeded via step-flow growth and that steps do not bunch at this temperature [Fig. 1(a)]. At 700 [Fig. 1(b)] and 750 °C [Fig. 1(c)], the surface steps are of more than 2 BL in height, indicating step bunching. Furthermore, there are clear differences in the edge morphology of the step bunches at different growth temperatures. At 700 °C, most of the bunched steps are composed of $[\bar{1}\,\bar{1}\,2]$-type steps, but there are crossing BL and bunched steps oriented toward the $[\bar{1}01]$ direction on the terraces [Fig. 1(b)]. At 750 °C, on the other hand, the bunched steps are mainly composed of three types of $[\bar{1}\,\bar{1}\,2]$ steps that form macrokinks at the edges as schematically shown in the inset. Also, the BL and bunched steps [arrows in Fig. 1(c)] cross the terraces and most of them form an angle of about 60° with respect to the $[\bar{1}\,\bar{1}\,2]$ direction, as in the case at 700 °C. The ratio of the size of macrokink segment L_{st} to the length of straight part of the step segment L_{mk} was observed to be 0.10 and to stay constant with deposition time below a deposited thickness of 42 nm at 750 °C. With a further increase of the growth temperature to 800 °C, the step pattern changes drastically. The steps not only rapidly bunch, but also meander randomly [Fig. 1(d)] without macrokinks and without steps crossing the terraces. This seems to be similar to the step pattern observed on vicinal Cu surfaces [24]. The main difference is that we found meandering step bunches on Si(111)-(7×7), while Néel *et al.* reported a route to bunching through meandering on vicinal Cu high-index surface [24]. It should be noted that the bunching at 800 °C produced large terraces between neighboring bunches and then small triangular islands nucleated on the large terraces, which was confirmed in close-up AFM images (not shown here). The island nucleation indicates that the growth was limited by the diffusion length of Si adatoms and that the growth mode changes from step flow to island growth mode on large terraces at 800 °C.

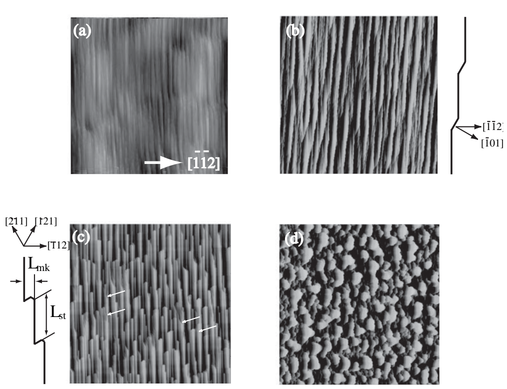

FIG. 1. AFM images of Si(111)-(7×7) after Si depositions of (a) 100 nm at a substrate temperature of 650 °C, (b) 150 nm at 700 °C, (c) 42 nm at 750 °C, and (d) 10 nm at 800 °C. The image size is 14.6 μm × 14.6 μm. The miscut angle is 1.0° from (111) toward the $[\bar{1}\,\bar{1}\,2]$ direction.

As a first step to clarifying the origin of these various morphological evolutions, we plotted surface width ϖ (rms) of the 1.0°-miscut Si(111) in Fig. 2(a) as a function of growth thickness θ after deposition at temperatures of 650, 700, 750, and 800 °C. The error bars represent the standard deviation from the mean. The lines are least-square fits to the data, which provide the scaling relationship $\varpi \sim \theta^{\beta_r}$. The roughness exponent β_r nearly vanishes at 650 °C, but it is 0.25 ± 0.07 at 700 °C, 0.50 ± 0.09 at 750 °C, and 1.20 ± 0.02 at 800 °C. This indicates that the Si(111)-(7×7) surfaces evolved differently during step-flow growth at different temperatures. To further assess the details of the temperature dependence of the growth dynamics, similar experiments were performed at 675, 725, and 775 °C. The exponent β_r obtained by *ex situ* AFM observations is plotted in Fig. 2(b). This figure clearly shows that the growing surface is stable below 675 °C, but it starts to become unstable above 700 °C. Furthermore, we can see three types of unstable surfaces between 700 and 800 °C: one where β_r nearly equals 1/4 (type I bunching), one where it nearly equals 1/2 between 750 and 775 °C (type II bunching), and one where $\beta_r = 1.20 \pm 0.02$ at 800 °C (type III bunching and meandering). Surprisingly, the transition regions between these regimes are about 25 °C wide, which is quite narrow. Indeed, the

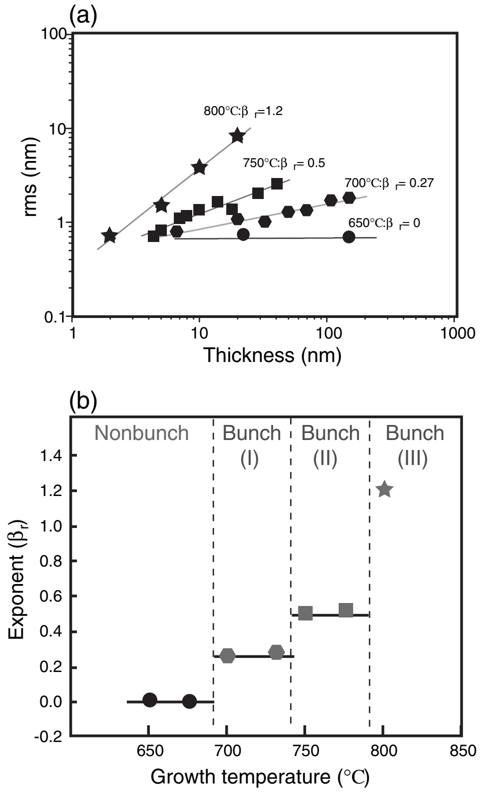

FIG. 2. (a) Evolution of rms roughness during homoepitaxial growth on vicinal Si(111)-(7 × 7) miscut toward the $[\bar{1}\,\bar{1}\,2]$ direction at different growth temperatures. (b) Time exponents of rms roughness as a function of growth temperature between 650 and 800 °C. The step bunching can be categorized into three types (I, II, and III) with respect to the growth temperature.

exponent appears to increase abruptly with increasing growth temperature and stays constant in between the rather sharp transitions around 700, 750, 775, and 800 °C. This seems to imply that the bunching mechanism during growth is different in the various regimes, as has been previously argued for the current-induced step bunching on Si(111) [25].

In addition to examining the 1.0°-miscut sample, we also studied the morphological dynamics at 750 °C during growth for a 2° miscut and found that β_r is 0.49 ± 0.03 at 750 °C, which is nearly equal to that for the 1° miscut [Fig. 1(c)]. This indicates that the growth dynamics is independent of the miscut angle.

To clarify the stability of wafers at different temperatures and with different miscut angles towards bunching, we performed experiments on substrates with 2°, 4°, and 5.6° miscuts toward the $[\bar{1}\,\bar{1}\,2]$ directions. Figure 3 shows the morphological diagram of the as-grown homoepitaxial growth on the Si(111) miscut toward the $[\bar{1}\,\bar{1}\,2]$ directions. One can see that increasing the initial terrace width (decreasing the miscut angle) increases the stability of the step train. This is in contrast with the theoretical results on growth by the inverse Schwöbel effect, where it was found

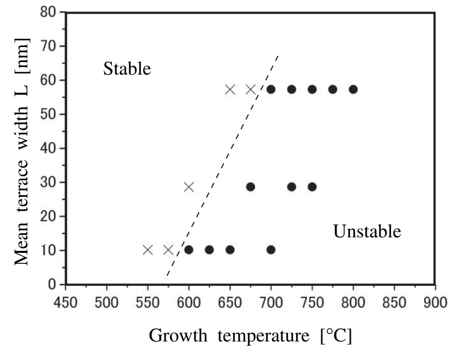

FIG. 3. Morphological diagram of the as-grown vicinal Si(111) as a function of miscut angles and growth temperatures. The dotted line shows the transitions between stable and unstable growth on the vicinal Si(111).

that increasing the initial step distance (decreasing the initial surface slope) makes the growing step system unstable [12]. In what follows, our results are given for a given miscut angle, and one should remember that the temperature regions change with changing miscut angle.

In order to get more insight into the mechanisms of type-I and type-II step bunching, we performed further scaling analyses of the obtained bunches. The step bunch formation is a result of a fine balance between the destabilizing effect of the diffusion kinetics and the stabilizing effect of the interstep repulsions. Thus, the exponents in the scaling relations for the step bunches are expected to be a result of microscopic characteristics of the system. A scaling hypothesis connecting the exponents describing the surface stabilization and destabilization from one side with the observable exponents from the size and time scaling of the step bunches from the other side has been proposed [11] and recently improved [12]. The size and time scaling of the step bunching on the Si(111) are, respectively, shown in Figs. 4(a) and 4(b) for type I and in Figs. 4(c) and 4(d) for the type II. From the scaling relationship of $BW \sim BH^{1/\alpha}$, $BH \sim \theta^{\beta_h}$, and $BW \sim \theta^{1/z}$, we obtained $1/\alpha = 0.61 \pm 0.03$, $\beta_h = 0.43 \pm 0.04$, $1/z = 1/(a/\beta_h) = 0.26 \pm 0.04$ at 700 °C, and $1/\alpha = 0.54 \pm 0.08$, $\beta_h = 0.49 \pm 0.09$, $1/z = 0.26 \pm 0.08$ at 750 °C, where BW is the bunched width and BH the height of a bunched step. A quantitative scaling analysis of type III was not performed because a theory of step bunching and meandering has not yet been established as pointed out by Néel *et al.* [24]. A most noteworthy result is that the exponent $1/z$ is the same for bunching types I and II. Using the continuum theory of Ref. [11] together with the slope-dependent surface mobility [12], one can easily show that an exponent $z = 4$ is common to all different destabilizing mechanisms if the interstep repulsions are characterized by the canonical power $n = 2$. Then, $1/\alpha = 3/5$ and $\beta_h = 5/12 = 0.417$ imply that the nonequilibrium current varies as $J \sim m^{-2}$ as a function of the local slope m. The values $1/\alpha = 1/2$ and $\beta_h = 1/2$ imply instead $J \sim m^{-1}$. These

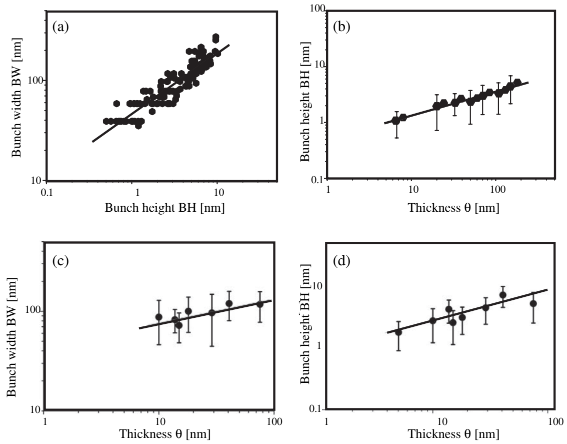

FIG. 4. Size and time scaling of step bunching during MBE on vicinal Si(111)-(7×7) miscut toward the $[\bar{1}\,\bar{1}\,2]$ direction at substrate temperatures of (a) 700 °C and (b) 750 °C.

behaviors have been associated with step-edge diffusion and inverse Schwöbel effect, respectively, in Ref. [11], as the microscopic sources of step bunching.

In conclusion, we have systematically studied morphologies of Si(111)-(7×7) miscuts towards the $[\bar{1}\,\bar{1}\,2]$ direction during homoepitaxial step-flow growth at substrate temperatures between 600 and 800 °C using *ex situ* AFM. We found three types of unstable growth between 700 and 800 °C. The growing surface is stable in step bunching between 600 and 775 °C , but is stable in both step bunching and meandering at 800 °C. Size- and time-scaling analyses showed that these types of step bunching belong to different universality classes characterized by slow attachment and detachment at the steps. The instability appears to be driven by microscopic mechanisms acting as an effective step-edge diffusion and as an effective inverse Schwöbel effect for type I and type II bunching, respectively. We hope that these findings help us to obtain spontaneously controlled atomic step patterns for nanostructure self-assembly on silicon surfaces.

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*Corresponding author.
Electronic address: homi@will.brl.ntt.co.jp
†Present address: Department of Physics, Tokyo University of Science, Kagurazaka 1-3, Shinjuku-ku, Tokyo 162-8601, Japan.

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