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2D Si island nucleation on the Si(111) surface at initial and late growth stages: On the role of step permeability in pyramidlike growth

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ABSTRACT

Initial and late stages of 2D Si island nucleation and growth (2DNG) on extra-large ($\sim 100\ \mu$m) and medium size (1–10 μm) atomically flat Si(111)-(7 × 7) terraces bordered by step bunches have been studied by *in situ* REM at $T = 600$–$750\ ^\circ$C. At first, the layer-by-layer 2DNG takes place on whole terraces and 2D island concentration dependence on deposition rate R corresponds to critical nucleus size $i = 1$. Continuous 2DNG triggers morphological instabilities: elongated pyramidlike waves and separate pyramids emerge on all terraces at $T \leq 720\ ^\circ$C and $T = 750\ ^\circ$C, respectively. Both instabilities arise due to the imbalance of uphill/downhill adatom currents related with large Ehrlich-Schwöbel (ES) barriers and permeability of straight $[\bar{1}\bar{1}2]$-type step edges. However, the first one is initiated by dominant downhill adatom current to distant sinks: bunches, wave's step edges, and "vacancy" islands emerging on terraces due to 2D island coalescence. As a result, top layer size decreases to the critical terrace width λ where 2DNG takes place. From the analysis of $\lambda \propto R^{-\chi/2}$ scaling at $T = 650\ ^\circ$C, we have found that i increases from $i = 2$ on a three-layer wave to $i = 6$–8 on a six-layer wave. This authenticates the significance of downhill adatom sink to distant steps related to the step permeability. The second instability type at $T > 720\ ^\circ$C is related to the raising of uphill adatom current due to slightly larger ES barrier for step-up attachment comparing to the step-down one ($E_{ES}^{-} \sim 0.9$ eV [Phys. Rev. Lett. 111 (2013) 036105]). This leads to "second layer" 2D nucleation on top layers, which triggers the growth of separate pyramids. Because of small difference between ES barriers, net uphill/downhill adatom currents are nearly equivalent, and therefore layer coverage distributions of both instabilities display similar linear slopes.

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1. Introduction

Advanced epitaxial technologies being used to develop new semiconductor devices should be based on the deep knowledge of surface processes at all growth stages. In order to study adatom diffusion, attachment to steps, and nucleation, the concentration of two-dimensional (2D) islands N_{2D} or critical terrace width λ for 2D island nucleation is often analyzed within the scope of rate-equation approach as function of substrate temperature T and deposition rate R [1,2]:

$$N_{2D}^{-1} \equiv L_s^2 \propto R^{-\chi} \exp \frac{-E_{2D}}{kT}, \tag{1}$$

$$\lambda^2 \propto R^{-\chi} \exp \frac{-E_{2D}}{kT}, \tag{2}$$

where L_s is mean island separation, E_{2D} is the effective activation energy of 2D island nucleation and growth (2DNG), χ is scaling exponent, and k is Boltzmann's constant. Since both L_s and λ are the result of the interaction of diffusing adatoms between each other and with steps, their scaling with R and T contains the information about important thermally activated processes such as adatom diffusion activation energy ϵ_d, critical nucleus size i and dissociation energy ϵ_i, etc. Depending on atomically flat terrace width during experiment, either N_{2D} or λ can be measured, and either Eq. (1) or Eq. (2) can be used, respectively. If terrace size is much wider than λ, the growth proceeds via 2DNG, and its mechanism is studied using the classical rate-equation analysis of N_{2D} scaling [1–7]. If the growth takes place on vicinal surfaces with small step separation, the interaction of adatoms with steps come into play, 2DNG is suppressed, and the growth mode switches to step flow. Under these conditions, the measurement of critical

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terrace size λ is more convenient, and its scaling analysis is used instead [8–12].

Classical theoretical consideration of 2DNG on singular and vicinal surfaces predicts that step permeability (ability of adatoms to cross steps without attachment to step kinks) does not significantly affect N_{2D} and λ scaling [1,2,8,9]. However, large Ehrlich-Schwöbel (ES) barriers and/or step permeability induce destabilizing net *uphill* adatom current that may trigger mound formation on atomically flat terraces [13,14] and govern surface morphology evolution during prolonged growth [15,16]. Recently, Hervieu and Markov have theoretically shown that the step permeability with moderate ES barriers facilitates second layer nucleation atop first-layer 2D islands and decreases its critical size [14]. In this case, the step permeability leads to a multilayer growth and linear coverage distribution of layers [16]. The slope of this distribution weakly depends on deposition time, which is called slope selection. As discussed by Filimonov and Hervieu in Ref. [17], the step permeability depends on step structure and is proportional to the distance between step kinks. Moreover, the kink density dependence on temperature is non-monotonous and can switch step properties between permeable and impermeable.

The STM investigations of the initial Si/Si(111)-(7 × 7) growth stages at $T \lesssim 400$ °C and $R \lesssim 10^{-3}$ ML/s have shown that straight $[\bar{1}\bar{1}2]$-type island edges are permeable, which leads to the nucleation of 2nd and 3rd layer islands atop [18,19]. However, after multilayer island coalescence, further film growth proceeds via layer-by-layer mechanism up to at least 11th layer [20]. In present work we first show that the step permeability becomes even stronger at higher $T = 600–750$ °C and leads to the formation of elongated pyramidlike waves and separate pyramids during prolonged Si growth.

We have previously shown in Ref. [12] that Si growth on 2–5 μm Si(111)-(7 × 7) atomically flat terraces between step bunches at $T \approx 600–750$ °C starts as classical layer-by-layer 2DNG and then switches to periodic 2DNG on a λ-width terrace formed due to preferential *downhill* adatom attachment. We have studied $\lambda^2(R, T)$ dependences and found that step kinetics is limited by ~0.9 eV ES barrier for adatom attachment to descending steps linked with impeded double kink nucleation at the straight $[\bar{1}\bar{1}2]$-type step edges. In present study, we used extra-large (~100 μm) singular terraces and moderately large (1–10 μm) flat terraces on the step-bunched surface to compare 2DNG at a constrained and "infinite" terraces within $T = 600–750$ °C range. We have analyzed in detail N_{2D} and λ^2 dependences on R, T on the step-bunched surface to understand the influence of adatom sink to steps bordering pyramidlike waves on their formation.

2. Experimental details

In situ experiments were carried out in an ultrahigh vacuum reflection electron microscope (UHV REM) equipped with a Si evaporator. We used specimens with dimensions 8 × 1.1 × 0.3 mm^3 cut from *n*-type (0.3 Ω cm) Si(111) wafers with the miscut angle about 0.5°. The details of the UHV REM technique and sample preparation were described elsewhere [21]. The important feature of the REM technique is ~50 times image compression along beam incidence that distorts images of atomic steps and 2D islands. Therefore, we used atomic-force microscopy (AFM) to analyze exact shape of steps and 2D islands as well as their distribution on the Si(111) surface.

Moderately large (up to 10 μm) atomically flat terraces required for $N_{2D}(R)$ study on the Si(111)-(7 × 7) surface were produced in UHV REM using step bunching at $T = 1050–1300$ °C [22]. After the step-bunched surface preparation, T was reduced first rapidly (~400 K/s) to $T \approx 830$ °C and then slowly (~0.1 K/s) to 790 °C, which resulted in the formation of large-scale (~10–100 μm^2) superstructural (7 × 7) domains. This allowed us to exclude the impact of domain walls and atomic steps on 2D island nucleation and to analyze $N_{2D}(R)$ dependences at initial Si growth stages.

To produce extra-large atomically flat round terraces of ~100 μm diameter on sample surface, we used ion etching followed by high temperature AC resistive annealing in UHV REM at $T > 1300$ °C [23]. We used extra-large terraces to investigate 2DNG and surface morphology evolution during prolonged Si/Si(111)-(7 × 7) growth.

3. Results and discussion

3.1. 2D island nucleation on the step-bunched Si(111)-(7 × 7) surface

All stages of the Si growth on the step-bunched surface observed by *in situ* UHV REM technique at 600 °C are presented in Fig. 1 including the transformation of a regular step train on the initial vicinal Si(111) surface (Fig. 1a) to the step bunches at $T \sim 1200$ °C followed by sample cooling to $T = 600$ °C (Fig. 1b). One can see that during prolonged Si growth clear multilayer structure similar to a pyramidlike wave is formed (Fig. 1c–e). The waviness of steps (thin lines) and bunches (black strips) on pre-deposition

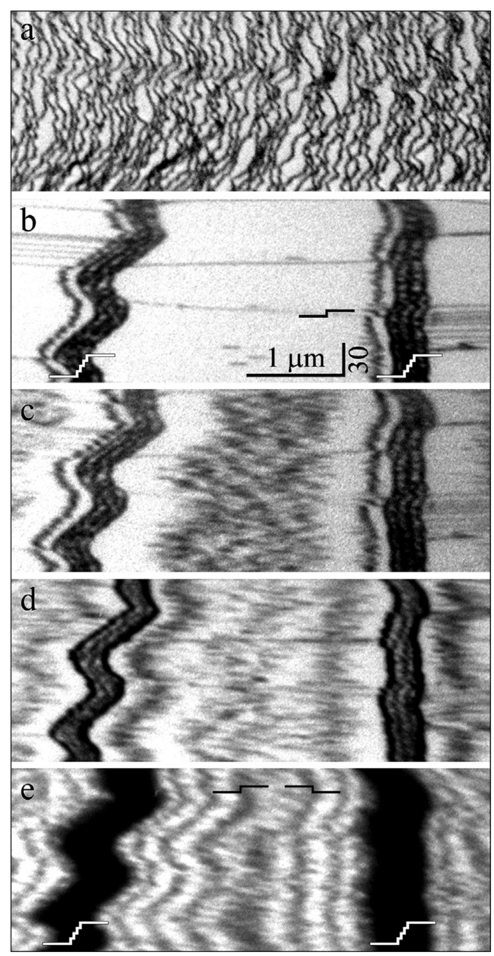

Fig. 1. *In situ* REM images of (a) an initial vicinal Si(111) surface at 1050 °C and (b) flat terraces between two step bunches at 600 °C before Si deposition. (c)–(e) Successive stages of pyramidlike wave formation during Si deposition at 600 °C: (c) 0.12 ML, (d) 1 ML, and (e) after prolonged Si deposition (Θ ~ 20 ML). A real-time *in situ* movie of pyramidlike wave's periodic evolution during Si growth is available on the World Wide Web as a supplementary material to this paper.

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