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Origin of roughening in epitaxial growth of silicon on Si(0 0 1) and Ge(0 0 1) surfaces

H.J.W. Zandvliet [a,*], E. Zoethout [a], W. Wulfhekel [b], Bene Poelsema [a]

[a] *Faculty of Applied Physics and MESA+ Research Institute, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands*

[b] *Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle/Saale, Germany*

Abstract

It is shown that the formation of (2 × 1) reconstructed islands during the growth of Si on Si(0 0 1) and Ge(0 0 1) surfaces at moderate temperatures and the consequent anisotropic diffusion and sticking lead to roughening. As Si islands nucleate and expand, the dimer rows in neighboring rows need not, in general, align with each other. An anti-phase boundary (APB) will be found if two growing islands meet, but their internal dimer rows are not in the same registry. One type of APB runs perpendicular to the substrate rows, whereas the other runs along the substrate rows. As has been pointed out by Hamers et al. [J. Vac. Sci. Technol. A 8 (1990) 195] this first type of APB is a preferential center for nucleation of next layer islands and thus naturally leads to roughening. Here we show that the other type of APB leads, in combination with the anisotropy in sticking and diffusion of dimers, to the formation of long B-type double layer steps and narrow trenches. We argue that it is the kinetic suppression of filling of these trenches and not, as previously suggested the existence of step edge barrier for the double layer step edge that leads to roughening. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Novel thin film structures synthesized by physical or chemical vapor deposition (CVD) on silicon have found a wide variety of applications in science and technology. Silicon is the material that accounts for more 98% of the sales in the global semiconductor market. The low manufacturing costs and the inherent properties are the key reasons why silicon devices completely dominate the microelectronic market. Compound semiconductors, such as gallium arsenide, are much better suited for optoelectronic devices, such as light-emitting diodes and lasers, but their fabrication costs are much higher. However, by adding another semiconductor (e.g. germanium) to silicon the performance of transistors and circuits can improve significantly. If some silicon atoms are replaced by germanium atoms, one can "engineer" the band gap of the material as well numerous other properties. A simple and straightforward way to replace locally silicon by germanium is to grow a stack of silicon and germanium layers on silicon. These so-called silicon–germanium

* Corresponding author. Tel.: +31-534-89-3091; fax: +31-534-89-1101.

E-mail address: h.j.w.zandvliet@tn.utwente.nl (H.J.W. Zandvliet).

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heterostructures allow the creation of an entirely new class of materials based on modulated band gap engineering and modification of the band properties due to strain.

Smooth and abrupt interfaces between epitaxial layers are often crucial factors in determining the device performance. The smoothness and abruptness of the interface lead to a conflicting demand with respect to the growth conditions. On one hand the growth temperature should be sufficiently low to reduce atom diffusion and hence intermixing of the layers and dopant segregation, while on the other the growth temperature should be sufficiently high to avoid kinetic roughening.

Growth on a crystal usually starts with nucleation: the deposited atoms diffuse on the surface until they meet and form stable nuclei. In the first stage of the growth process the number density of island increases until a saturation value is reached. The newly deposited atoms on the surface are incorporated at the existing nuclei, which grow into two-dimensional (2D) islands. In order to have perfect layer-by-layer growth, the atoms deposited on the growing 2D islands need to descend from the islands before they can nucleate. Ehrlich and Hudda [1] as well as Schwoebel and Shipsey [2] pointed out that due to the reduced coordination of an atom when it hops over an island edge there in general exists an additional energy barrier, the so called Ehrlich–Schwoebel (ES) barrier, for downward diffusion at the island edge. In the absence of an ES barrier for homoepitaxial system one obtains in principle nice layer-by-layer growth. In the presence of a finite ES barrier, however, atoms diffusing on-top of an existing island have a high probability to be reflected at the island edge leading to an increased probability for nucleation on-top of the island and thus resulting in multilayer growth.

It is the purpose of this paper to point out that the tendency of surfaces to reconstruct during molecular beam epitaxy (MBE) growth can lead to roughening. In order to illustrate our point we have considered the growth of Si on Si(0 0 1) and Ge(0 0 1) surfaces. These surfaces are stabilized by a (2×1) surface reconstruction where the atoms of the outermost layer couple in pairs to form dimers in order to reduce the dangling bonds of the ideally bulk-truncated surface [3]. [1] As Si islands nucleate and grow on (0 0 1) surfaces, the dimer rows in neighboring islands have a probability of 50% to be in the correct registry. An anti-phase boundary (APB) will form if two islands meet, when their internal dimer rows are not aligned. For Si and Ge(0 0 1) surfaces there are in principle two different APBs, one type of APB runs along the substrate dimer rows ($APB_{\parallel}$) and the other runs perpendicular to substrate dimer rows ($APB_{\perp}$). Several authors have pointed out that the latter APB acts as a preferential nucleation site for island growth [4,5]. The first type of APB gives, in combination with the fact that the islands are elongated in the dimer row direction, rise to the formation of trenches and double layer steps (D_B). The preferred shape of the expanding islands (typical aspect ratio from 1:3 up to 1:15) [4] depends on the amount of supersaturation during growth and has therefore a kinetic rather than a thermodynamic origin. But even thermodynamically the preferred shape as can be extracted from the ratio of the experimental determined step edge free energies [6–8] and direct observation [9–11] is elongated along the dimer row direction (aspect ratio 1:2.5). Although the deposition method used here is MBE we think that our findings might also be relevant for the technological important CVD growth method.

2. Experimental

The experiments were conducted in an ultrahigh vacuum (UHV) system equipped with a scanning tunneling microscope (STM). The base pressure of the system is 1×10^{-10} mbar. The Si(0 0 1) samples were outgassed at a temperature of 1000 K for at least one day. In order to obtain an atomically clean surface, the samples were repeatedly flashed at temperatures of 1500–1575 K for several seconds followed by quenching to room temperature.

[1] Recently, we have pointed out that, at least for Ge(0 0 1), the state with the lowest surface free energy at room temperature is a striped $(2 \times 1)/c(4 \times 2)$ pattern rather than simple (2×1) pattern.

The Ge(0 0 1) samples were, after outgassing at 800 K, cleaned by cycles of 800 eV Ar^+ sputtering and annealing at 1100 K. Si was deposited from a resistively heated crystal onto to Ge or Si(0 0 1) surfaces which were kept at a fixed temperature in the range from room temperature to 800 K. After deposition and quenching the samples were transferred in situ to the STM for imaging.

3. Results and discussion

In Fig. 1 2.4 monolayers of Si are deposited on Ge(0 0 1) at a temperature of 525 K. The original substrate consisted of (2×1) and (1×2) terraces separated by monatomic steps. The 2D islands are elongated in a direction along the dimer row direction. The preferred growth anisotropy also persists at coverages above one monolayer. However, as is clear from Fig. 1 the aspect ratio of second layer islands is significantly smaller than first layer islands due to the fact that the dimer row direction rotates by 90° at each monoatomic step. The epitaxial islands nucleate randomly with 50% probability that adjacent islands are in the correct registry, i.e. the dimer rows of both islands are in

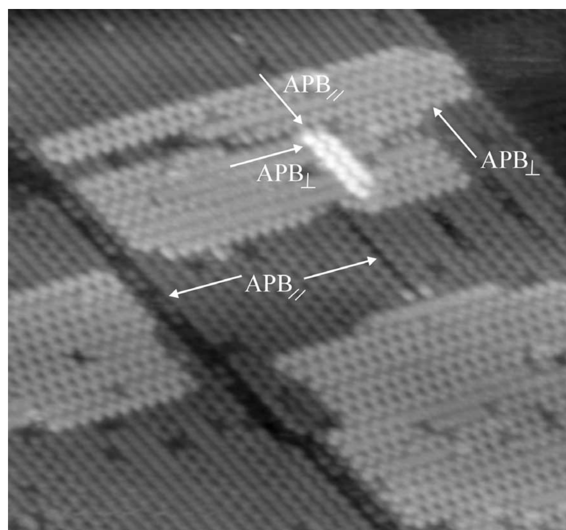

Fig. 1. Ge(0 0 1) surface after deposition of 2.4 ML Si at 525 K. Sample bias −1.6 V and tunneling current 1 nA. Both type of APBs are shown in the figure.

the correct registry. When two growing islands meet, there will be a 50% probability that they will form an APB. As shown in Fig. 1 two types of APBs, labeled ($APB_\perp$ and $APB_\parallel$) can be found. As has been pointed out by Hamers et al. [4] and Bedrossian and Kaxiras [5] the $APB_\perp$ type of APB acts as a preferential site for nucleation of islands in the next layer and thus leads to roughening. The role of the other type of APB with respect to the evolution of the growth morphology has, however, not been addressed in detail. It is clear that the rotation of the dimer row direction at each monatomic step in combination with the growth anisotropy leads to the formation of double layer steps (D_B steps) [12]. Those D_B steps have a rebonded character, i.e. at the upper part of the steps there is a row of rebonded atoms. Even after deposition of several monolayers deep trenches persist [12]. Initially we attributed this remarkable feature, which results in substantial roughening to the existence of an ES barrier at these D_B steps which hinders mass transport across the steps and therefore leads to multilayer growth. However, a more detailed analysis of the data reveals that if islands nucleate in the third layer it is almost exclusively on APBs. This in combination with the strong diffusional anisotropy [13–18] implies that atoms deposited on the second layer high islands can in principle diffuse across the D_B steps. Below we will demonstrate that this means that the step edge barrier of a D_B step must be smaller than 0.2 eV for Si on Ge(0 0 1) as well as for Si on pure Si(0 0 1). At a certain stage the expanding two atomic layers high islands will meet. If there exists an APB between the islands this will lead to formation of long narrow trenches. Filling of these narrow trenches can easily be kinetically hindered at these temperatures because the surface needs to locally lift the reconstruction at several atomic levels near the D_B steps. Moreover, also the two D_B steps, which are out-of-phase with each other, need to be annihilated (for two in-phase D_B steps this is not a problem!).

The diffusion of Si dimers on Si and Ge(0 0 1) is strongly anisotropic [13–19]. The second layer islands are initially elongated along the dimer row direction, but as soon as the island edges touch, the islands edges of the island underneath the top

layer island start to expand in a direction perpendicular to the dimer rows and becomes more compact (see Fig. 1). Therefore, it is quite appealing and well justified to use a simple one-dimensional model in order to determine the ES barrier of a D_B step. We consider here the diffusion of dimers on the second layer islands, which we model as a one-dimensional array of lattice sites ranging from position 0 to position m. The barrier to hop from site to site is E and τ is the mean dwell time on a certain site n $(0 < n < m)$

$$\tau = \frac{1}{2v} = \frac{1}{2v_0 e^{-E/kT}} \tag{1}$$

where $v = v_0 e^{-E/kT}$ is the hop rate and v_0 the attempt frequency. At the edges of the interval $[0, m]$ the barrier is assumed to be higher by an amount E_{ES} $(s = \exp(-E_{ES}/kT))$. The first exit time for a dimer located at site n $(0 < n < m)$ on $t = 0$ to leave the interval $[1, m-1]$ is given by

$$T_1(n) = \frac{(m-n)n}{2v} = (m-n)n\tau \tag{2}$$

At site 0 and m the mean dwell time is given by

$$\tau_0 = \frac{1}{v(1+s)} = \frac{2\tau}{(1+s)} \tag{3}$$

Subsequently the dimer can now either escape or return. The escape and return probabilities, p_{escape} and p_{return}, are given by

$$p_{escape} = \frac{s}{s+1} \quad \text{and} \quad p_{return} = \frac{1}{s+1} \tag{4}$$

Thus after starting at $t = 0$ on site n the dimer will do its first attempt to escape after a time $T_1^*(n)$ given by

$$T_1^*(n) = (m-n)n\tau + \tau_0 \tag{5}$$

The probability to escape is $s/(s+1)$.

The second attempt will take place after $T_2^*(n)$

$$\begin{aligned} T_2^*(n) &= T_1^*(n) + T_1(1) + \tau_0 \\ &= T_1^*(n) + (m-1)\tau + \tau_0 \end{aligned} \tag{6}$$

The probability to escape after $T_2^*(n)$ is $(1/(s+1))(s/(s+1))$.

The ith attempt to escape occurs after

$$T_i^*(n) = T_1^*(n) + (i-1)[(m-1)\tau + \tau_0] \tag{7}$$

In order to get an impression of the typical time interval between successive escape attempts we consider an island with length $m = 50$ and we deposit the dimer at site $n = 25$, the first escape attempt takes place after a time lapse of $625\tau + \tau_0$ s, the second escape attempt $49\tau + \tau_0$ s later than the first attempt, the third attempt again $49\tau + \tau_0$ s later and so on.

The probability to escape after a time lapse $T_i^*(n)$ is $(1/(s+1))^{i-1}(s/(s+1))$

The average escape time $T_{av}(n)$ for a dimer initially located at site n to leave the interval $[0, m]$ is

$$T_{av}(n) = \sum_{i=1}^{\infty} \left[T_1^*(n) + ((m-1)\tau + \tau_0)(i-1)\right] \times \frac{s}{s+1}\left(\frac{1}{s+1}\right)^{i-1} \tag{8}$$

which reduces to

$$T_{av}(n) = [T_1(n) - (m-1)\tau] + \frac{s+1}{s}(m-1)\tau + \frac{2\tau}{s} \tag{9}$$

The average first exit time for a dimer situated on the interval $[1, m-1]$ is

$$\begin{aligned} \bar{T}_1 &= \frac{1}{m-1}\int_{n=1}^{n=m-1} (m-n)n\tau \mathrm{d}n \\ &= \frac{\tau}{6}\left(m^2 + m - 5 - \frac{1}{m-1}\right) \end{aligned} \tag{10}$$

and hence

$$T_{av} = \frac{\tau}{6}\left[m^2 - 5m + 1 - \frac{1}{m-1}\right] + \frac{s+1}{s}(m-1)\tau + \frac{2\tau}{s} \tag{11}$$

The number of hops, $N_{av} = T_{av}/\tau$, is given by

$$N_{av} = \frac{1}{6}\left[m^2 - 5m + 1 - \frac{1}{m-1}\right] + \frac{s+1}{s}(m-1) + \frac{2}{s} \tag{12}$$

In the absence of an ES barrier (i.e., $s = 1$) one finds

$$N_{av}(s=1) = \frac{1}{6}\left(m^2 - 5m + 1 - \frac{1}{(m-1)}\right) + 2(m-1) + 2 = \frac{1}{6}\left(m^2 + 7m + 1 - \frac{1}{m-1}\right)$$

Let us consider the growth of Si on Ge(0 0 1) at 525 K. A typical growth rate of about 1 ML per minute results after two minutes of deposition (coverage is ~2 ML) into elongated Si islands with a long axis of about $160a_0$ ($a_0 = 3.84$ Å). If growth proceeds second layer islands are formed on-top of those islands. As pointed out in the text double layer step are formed rapidly. Interestingly, no islands are formed on-top of these second layer islands (besides APB nucleated islands). This implies that the material deposited on-top of these islands (typical length of about $50a_0$) does typically overcome the ES barrier at 525 K. Using the above derived equations and assuming that the monolayer steps do not exhibit a ES barrier and that the dimers diffuse preferentially along the substrate rows one can derive, using Eq. (12), that the ES barrier of a double-layer step should be smaller than 0.2 eV, i.e. $s > 0.014$ at 525 K (for the system Si/Si(0 0 1) one also finds the ES barrier of D_B step is smaller than 0.2 eV moreover, we have analyzed several STM images at various growth temperatures, but we always arrived at the conclusion that the ES barrier is smaller than 0.2 eV.) Therefore, much of the material deposited on-top of the second layer islands will eventually diffuse across the D_B steps into the trenches. Interestingly these trenches are not filled. Clearly the lifting of the (2×1) reconstruction and the annihilation of the two stable out-of-phase D_B steps provides a too high activation energy.

4. Conclusions

In summary, the formation of APBs during the epitaxial growth on Si and Ge(0 0 1) at moderate temperatures leads to roughening of the growth front. As known already one type of APB is a preferential site for nucleation. The other type of APB leads to the formation of long double layer steps and trenches. At moderate growth temperatures (400–700 K) the filling of these trenches, which are bounded by very stable D_B steps, is kinetically suppressed. The filling of these trenches is difficult because it requires a local lifting of the surface reconstruction and annihilation of the stable D_B steps. The ES step edge barrier for the D_B (estimated to be smaller than 0.2 eV) is not a key factor for the onset of roughening.

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