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Theoretical study of strain-induced step configuration changes on vicinal Si(001) by the growth of Ge

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Abstract. The initial stage of growing Ge on single-domain vicinal Si(001) with a large angle of misorientation has been studied theoretically by the modified Keating model and compared with RHEED and STM experiments. The experimentally observed conversion from the DB step configuration, where all dimer rows are normal to the step edges, to a DA-like step configuration, where dimer rows on one terrace are parallel to the step edges, at Ge coverages larger than 1 ML is identified by our calculations. Our results show that this DA-like step is in fact a pair of steps of single atomic height: SA+SB, with a very wide SA terrace and an SB tooth of about 10 Å. This new step configuration is energetically favoured over DA and DB step configurations.

1. Introduction

Vicinal surfaces of semiconductors have attracted much attention in recent years owing to their scientific and technological importance in device materials, crystal growth, surface chemistry and catalysis. Defect-free epitaxial growth and the fabrications of various low-dimensional structures such as quantum wires and dots can be achieved on these surfaces. Vicinal Si(001) is known to display both single- and double-step structures at thermal equilibrium, depending on the angle of misorientation and/or annealing procedures [1]. The step structure on vicinal Si(001) plays an important role in the growth processes and in determining the physical properties of the materials. It has been shown, for example, that molecular beam epitaxy (MBE) growth of GaAs on the double-step Si(001) substrates can reduce antiphase boundaries significantly [2].

The change in the step configuration from steps of single atomic height to steps of double atomic height was first observed in the MBE growth of Si on the double-domain (single-step) vicinal Si(001) with an angle of misorientation of 0.5° towards the [110] azimuth [4–6]. The growth-induced change in the step configuration was attributed to the anisotropic diffusion of Si. Alternatively, the equilibrium populations of different step configurations can be changed by external strain [7, 8]. In the case of Ge–Si heterostructures, the strain between Ge and Si is known to affect the physical properties of the strained heterostructures and is also responsible for a number of interesting phenomena observed in the pseudomorphic growth process of Ge on Si, such as the Stranski–Krastanov growth mode of Ge on Si, $(2 \times n)$ reconstructions, and the reversal of step morphology upon Ge deposition [9, 10].

Earlier calculations by Chadi [11] have shown that, for fully relaxed vicinal Si(001) with large angles of misorientation, the DB step, where dimer rows on the upper terrace are perpendicular to the step edge, is energetically favoured over the DA step, where dimer

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rows on the upper terrace are parallel to the step edge, at thermal equilibrium. At small angles of misorientation, the single-layer stepped surfaces are more stable. The critical angle is calculated as $\theta_c \approx 2^\circ$ [1]. This has been confirmed by RHEED, LEED and STM experiments. In this paper, we present detailed calculations of different step configurations appearing after the growth of several monolayers of Ge on the vicinal Si(001) substrate with an angle of misorientation of 3.5° and initial single-domain DB steps. We show that the DA-like step configuration, observed by our RHEED and STM experiments at Ge coverages larger than 1 ML, is in fact a pair of single atomic steps: SA+SB, with a wider terrace of SA and a narrower terrace of SB (about 10 Å). This means that strain can change the critical angle.

We observed the conversion of step configurations firstly in the RHEED experiments on vicinal Si(001) by depositing a $Si_{1-x}Ge_x$ layer. The substrates that we used are boron-doped vicinal Si(001) wafers with an angle of misorientation of 3.5° towards to [110] azimuth and less than 0.1° towards the $[\bar{1}10]$ azimuth, as determined by x-ray diffraction. Before growing the $Si_{1-x}Ge_x$ layer, RHEED patterns show that intensities of the half-order beams observed with the primary electron beam along the [110] azimuth are much stronger than those observed at the $[\bar{1}10]$ azimuth. This indicates that the vicinal Si(001) surface has predominantly a double-step structure. After depositing a 10 Å $Si_{0.6}Ge_{0.4}$ layer, the intensities of the half-order beams at the [110] azimuth become weaker, and the half-order beams at the $[\bar{1}10]$ azimuth become stronger. The interchange in the intensities of the two half-order beams demonstrates that the conversion of the surface configuration from (1×2)-domain-dominated steps to (2×1)-domain-dominated steps has occurred [3].

Our STM experiments clearly show the conversion of the step configuration. In the following, we give a simple description of our experimental process and the changes in the surface structures during the growth of Ge. Details will be presented elsewhere.

In our STM experiments we used Si(001) substrates with the same angles of misorientation as those used in RHEED experiments. Before growing Ge, the substrates display the one-domain DB step configuration. At 0.8 ML Ge coverage, although the DB step configuration remained unchanged, the dimer rows on the surface almost exclusively consist of buckled dimers. The buckling in adjacent dimer rows is in and out of phase, leading to (2×2) and $c(2 \times 4)$ reconstructions [11]. As the Ge coverage increases to above 1 ML, the DB configuration begins to show significant changes. There appear the dimer-vacancy (DV) lines in the regions where the $(2 \times n)$ reconstruction is fully developed, where n is the distance between two adjacent dimer vacancies on a given dimer row. The DV lines can serve as a guide to identify the dimer row orientation and the step configuration. At the 2.3 ML Ge coverage, the DV lines in the fully developed $(2 \times n)$ reconstruction are clearly evident from the STM image. In more than 60% of the imaged surface area, the DV lines run perpendicular to the step edges, indicating that, in these areas of $(2 \times n)$ reconstruction, the dimer row orientation is parallel to the step edge. This is a new step configuration and looks like a DA step at first glance but is not. In the following we refer to this DA-like step configuration as the modified DA step (MDA), because both have similar structures. We also find some regions with a very wide terrace; DV lines are at the centre of the terrace and parallel to the step edges. As the Ge coverage is further increased, the population of the MDA relative to DB increases. At 3.7 ML Ge coverage, nearly 90% of the surface area shows MDA and the number of DV lines increases as shown in figure 1. After observing the STM image in this figure carefully, we can find that, on each terrace of the MDA, the surface shows $(2 \times n)$ reconstructions with n mainly at 6 and next at 7. Besides this, each MDA step is decorated with an approximately 10 Å SB tooth (here the term 'tooth' refers to the narrow SB terrace, as shown in figure 2). In other words, the

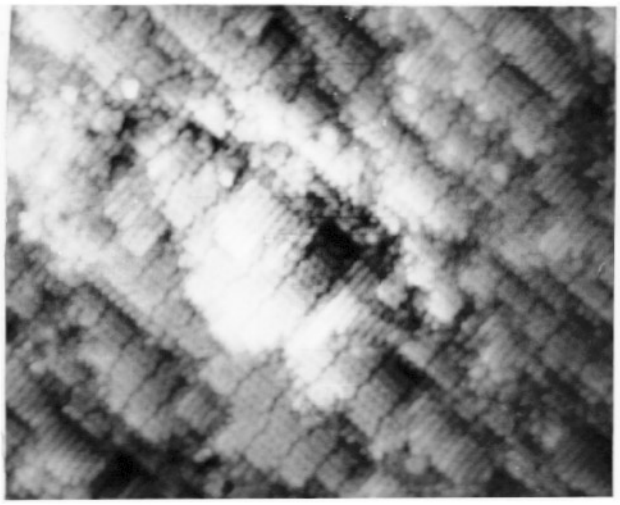

Figure 1. STM image of the vicinal Si(001) surface at 3.7 ML Ge coverage (scanned area, 490 Å × 440 Å; $V_s = -2.0$ V; $I = 33$ pA).

MDA step in fact consists of a pair of single atomic steps: SA+SB, with a wider SA terrace and a narrower SB terrace.

The above phenomena are very interesting and important. It is apparently necessary to understand them quantitatively. Our following calculations explain how the compressive strain induced by growing Ge drives the above processes.

2. The modified Keating model

The modified Keating model was firstly used by Tersoff to explain the (2×8) $Ge_3Si(001)$ reconstruction [13]. Here we use the same model to explain our experimental results. In this model, the elastic energy of the system is written as

$$E_{el} = \sum_i \left(\sum_{j=1}^{4} \frac{\alpha_{ij}}{a_{ij}} \left(\boldsymbol{x}_{ij}^2 - \tfrac{3}{16} a_{ij}^2 \right)^2 + \sum_{j,k>i}^{4} \frac{2\beta_i}{a_{ij} a_{ik}} \left(\boldsymbol{x}_{ij} \cdot \boldsymbol{x}_{ik} - \tfrac{3}{16} a_{ij} a_{ik} \cos \theta_i \right)^2 \right). \tag{1}$$

Here $\boldsymbol{x}_{ij}$ is the vector connecting atoms i and j. Each pure material is described by three parameters: its lattice constant a_i and its elastic parameters α_i and β_i. As the β_i-values are almost the same for Si and Ge, we use the same value for them. For Si–Si and Ge–Ge bonds, we take values of a_{ij} and α_{ij} from their elemental values [14]. For Si–Ge bonds, we take the geometric mean of their elemental values: $a_{ij} = (a_i a_j)^{1/2}$ and $\alpha_{ij} = (\alpha_i \alpha_j)^{1/2}$. For four-coordinated atoms, we take θ_i as the tetrahedral bond angle $\cos^{-1}(-\frac{1}{3})$; for three-coordinated atoms at the surface we take θ_i as $\cos^{-1}(-0.48)$, considering that these atoms have a tendency towards sp^2 bonding.

The total energy should include the energy of dangling bonds at the surface. For Si, we take $E_{DB} = 1.0$ eV; for Ge, $E_{DB} = 0.8$ eV [13]. Growing Ge on Si(001) can form missing dimer rows (DV line), which cause rebonded missing dimers (RMDs) in the second layer. Thus, forming each RMD can eliminate two dangling bonds. Further, it can lead to some enhanced π bonding within two dimers neighbouring the RMD [13]. So the total energy

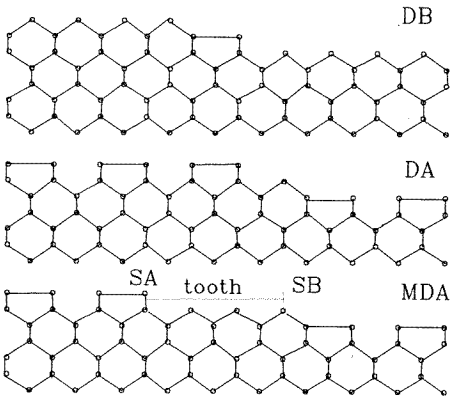

Figure 2. Side view of the atomic structure of steps of DB, DA and MDA on vicinal Si(001) surfaces. A [$\bar{1}$10] projection is shown.

of the system is

$$E_{tot} = E_{el} - \sum_{RMD} \left(2E_{DB} + 2\ \Delta E_{\pi}\right) \tag{2}$$

where ΔE_{π} is the magnitude of the enhancement of the π bonding in each neighbouring dimer. Tersoff estimated $\Delta E_{\pi} = 0.2$ eV to form the (2×8) $Ge_3Si(001)$ reconstruction.

3. Results and discussion

In this study, we do not consider intermixing between Ge layers and Si layers, as this would make the problem too complicated to deal with. So for the initial vicinal Si(001) with one domain of DB steps, growing 2 ML of Ge, we still obtain a DB step configuration; growing 3 ML of Ge, we obtain a DA step configuration; then on a little increase in the coverage of Ge, we obtain the MDA step configuration; growing 4 ML of Ge, we obtain the DB step configuration again. Figure 2 shows the atomic structures of DB, DA and MDA steps.

Our calculations are carried out on vicinal surfaces and are realized by constructing a series of ascending flat terraces along the y [110] axis with the normal along the z [001] direction and the step edges along the x [$\bar{1}$10] direction. We impose periodic boundary conditions just as Tze Wing Poon *et al* [15] did in studying Si steps. For DA and MDA step configurations, we consider two cases: $n = 6$ and $n = 8$ for the distance between the neighbouring DV lines. We take cells including 12–13 atomic layers which are allowed to relax and two bottom layers which are fixed at their ideal bulk positions during relaxing. All calculations are iterated until the force on each atom is less than 1 mRyd au^{-1}.

Firstly, we have calculated the surface energies of (2×1), (2×6) and (2×8) $Ge_3Si(001)$ reconstructions. The results are 1.370 eV/a^2, 1.356 eV/a^2 and 1.330 eV/a^2, respectively. Here $a = 3.85$ Å is the lattice constant on the (2×1) reconstructed Si(001) surface. These values are in agreement with those calculated by Tersoff. Secondly, we calculate the average surface energies of the vicinal $Ge_2Si(001)$ and $Ge_4Si(001)$ with an angle of misorientation of 3.5°. Their values are 1.330 eV/a^2 and 1.490 eV/a^2, respectively. If we impose a DV line at the centre of the terrace along the step edge, then we obtain their values as 1.314 eV/a^2 and 1.368 eV/a^2, respectively. This result can explain why DV lines appear at the centre of the wide terraces along the step edges at a Ge coverage of 2.3 ML. Thirdly,

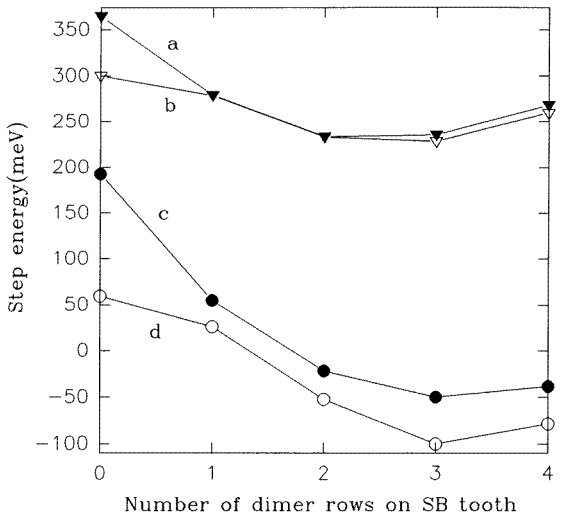

Figure 3. Step energies of four different kinds of MDA step: (a) $n = 8$, $W_{SA} = 9.5a$; (b) $n = 8$, $W_{SA} = 11.5a$; (c) $n = 6$, $W_{SA} = 9.5a$; (d) $n = 6$, $W_{SA} = 11.5a$. For (a) and (b), step energies are calculated by taking (2×8) $Ge_3Si(001)$ as the reference; for (c) and (d), (2×6) $Ge_3Si(001)$ is taken as the reference.

we have calculated step formation energies of DA and MDA. We have considered two values of n: $n = 6$ and $n = 8$. For each n, we further consider two different widths of SA terrace in DA and MDA steps: $W_{SA} = 9.5a$ and $11.5a$. These two values are the main widths of the SA terrace in figure 1. We fix the width of the SA terrace and change the width of the SB tooth. For $n = 6$, the step energy of each step configuration is calculated by taking the (2×6) $Ge_3Si(001)$ reconstruction as the reference; for $n = 8$, we take the (2×8) $Ge_3Si(001)$ reconstruction as the reference. Our calculations show that, for two step configurations with different n and the same widths of the SA terrace and the SB tooth, the $n = 6$ step configuration has a larger absolute average surface energy than the $n = 8$ step configuration. This is because we have chosen $\Delta E_\pi = 0.2$ eV, which means that the (2×8) $Ge_3Si(001)$ reconstruction is more stable than the (2×6) reconstruction. If we double this parameter, the (2×6) $Ge_3Si(001)$ reconstruction becomes more stable. Here we still take $\Delta E_\pi = 0.2$ eV [13], as the modified Keating model is just a simple model; it does not describe the bondings of the surface completely. Figure 3 presents our calculated results for different MDA step configurations. A MDA with zero width of the SB tooth is just a DA. We can see that the DA has a larger step formation energy than other MDA step configurations for four cases: in figure 3(a), $n = 8$ and $W_{SA} = 9.5a$; in figure 3(b), $n = 8$ and $W_{SA} = 11.5a$; in figure 3(c), $n = 6$ and $W_{SA} = 9.5a$; in figure 3(d), $n = 6$ and $W_{SA} = 11.5a$. For two MDA step configurations with the same n and same width of SB tooth the one with the wider SA terrace has the smaller step formation energy. When the number of dimer rows on the SB tooth is equal to 3 or so, the step has the minimum formation energy.

Xie *et al* [16] have even calculated the change in the step formation energy on Si(001) under compressive strain. They found that the step formation energies of SB and DB increase rapidly, while the value of SA changes little. Increasing the width of the SB tooth can increase the energy of the SB terrace but decrease the energy of the SB step.

The combined effects mean that the MDA has the minimum step formation energy at three dimer rows on the SB tooth. This result is in agreement with the widths of most SB teeth in figure 1.

Although considering a little more than 3 ML Ge coverage in calculations on MDAs is not completely in agreement with the 3.7 ML Ge coverage in figure 1, we think our results are at least qualitatively right. The STM image in figure 1 shows that MDA steps are very rough and have a large fluctuation in the width of terraces. Considering all these factors is too complicated. Similarly, although we cannot calculate the surface energy of the DB step growing 3 ML Ge directly, our calculated large surface energy of the DB step in the growth of 4 ML Ge can explain why DB steps do not exist in the STM image in figure 1. It is known that, on vicinal Si(001) with a small angle of misorientation, the SB step is rougher than the SA step. After growing 1.6 ML Ge, Wu *et al* [10] found that the SA step can become rougher than the SB step. We think that this can explain why the STM image in figure 1 is so rough. The statistics of the roughness are not considered here and will be studied further.

4. Summary

In summary, we have studied the initial-stage growth of Ge on single-domain vicinal Si(001) theoretically by the modified Keating model. We confirmed that the experimentally observed DA-like step configuration is in fact of a pair of single atomic steps: SA+SB, with a very wide SA terrace and a 10 Å SB tooth. This new step configuration is energetically favoured over DA and DB step configurations at Ge coverages larger than 3 ML. We also found that DV lines can be formed at the centre of the terraces along the step edges at 2 ML Ge coverage.

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