Shape evolution of MBE grown Si$_{1-x}$Ge$_x$ structures on high-index Si(5 5 12) surfaces: a temperature dependent study

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Abstract
The morphological evolution and the effect of growth temperature on size, orientation and composition of molecular beam epitaxy grown Ge–Si islands on Si(5 5 12) surfaces have been investigated in the temperature range from room temperature to 800 °C. Two modes of substrate heating, i.e. radiative heating (RH) and direct current heating (DH) have been used. The post-growth characterization was carried out ex situ by scanning electron microscopy, cross-sectional transmission electron microscopy and Rutherford backscattering spectrometry. In the RH case, we found spherical island structures at 600 °C with a bimodal distribution and upon increasing temperature, the structures got faceted at 700 °C. At 800 °C thick (∼122 nm) dome-like structures are formed bounded by facets. While in the case of dc heating, after the optimum critical temperature 600 °C, well aligned trapezoidal Si$_{1-x}$Ge$_x$ structures with a graded composition starts forming along the step edges. Interestingly, these aligned structures have been found only around 600 °C, neither at low temperature nor at higher temperatures.

(Some figures may appear in colour only in the online journal)

1. Introduction
Self-assembly of strain-induced islands in heteroepitaxial systems is a promising route to use nanoscale structures as quantum dots in opto-electronic devices. Among the different strained material combinations investigated so far, the Ge/Si system is often considered as a prototype for understanding fundamental properties of heteroepitaxial growth. In general, Ge prefers to adsorb on the surface, since the Ge dangling-bond energy is lower than that of Si [1, 2]. However, theoretically, it is expected that the tensile stress induced due to reconstruction makes Ge atoms diffuse into the silicon subsurface and into the bulk [3]. Hence, evolution of crystallographic facets of strained Ge–Si structures and diffusion of Ge into the Si substrate depend on the kind of the substrate and nature of reconstructions [4].

Faceting is a fundamental concern in the crystal growth process. It is observed when the system is allowed to minimize its free energy (thermodynamic equilibrium) [6–8]. The faceting phenomenon not only depends on material properties but also on the sample configurations and process conditions (pressure, growth temperature, composition, etc). Faceting can be used in order to improve electrical performance as in folded devices or minimized and suppressed as in silicon on nothing (SON) technology [9].

The growth of Ge nanostructures on low index silicon surfaces has been studied by many groups [5, 10–12] in the last two decades. More recently, the study of self-organization in the Ge/Si system has been extended to high-index surfaces [13–16]. However, the kinetic and thermodynamic factors determining the shape of the self-assembled Ge–Si structures are not yet well studied.

A high-index surface is misoriented from the low index plane by a small vicinal angle, relative to low index surfaces. The ultra-clean reconstructed surface shows periodic steps and terraces. The anisotropic surface reconstruction on high-index silicon surfaces makes them potentially significant substrates for the growth of ordered nanostructures. The use of such matrices as templates can enhance the formation of aligned nanostructures with self-assembled growth, which is not so easily achievable with lithography tools. Due to its natural process and damage-free features, structural self-organization
on high-index surfaces has received much attention in the formation of coherent 1D and 2D semiconductor/metal nanostructures. High-index surfaces are found to have reconstructions into regular hill (step) and valley (terrace) structures with periods ranging from several nanometers to about one hundred nanometers. The periodicity of such structures depends mainly on the substrate orientation and miscut or vicinal angles. The width of the terraces and step height can easily be modulated with temperature and coverage. These anisotropic silicon surfaces having alternating terraces and atomic steps can be used to form aligned one-dimensional nanostructures [17–19]. Among the high-index silicon surfaces oriented between (0 0 1) and (1 1 1), Si(5 5 12) exhibits relatively stable reconstruction having 1D symmetry with a (1 1 0) mirror plane. The Si(5 5 12) surface is oriented 30.5° away from (0 0 1) towards (1 1 1) and offers a stable 2 × 1 surface reconstruction with one-dimensional periodicity over a large unit cell [17, 20].

In this paper, we investigate the morphological evolution of strained Ge–Si islands on high-index Si(5 5 12) surfaces as a function of substrate temperature in two modes of heating condition, i.e. by RH and direct current heating.

2. Experimental details

The experiments discussed in the following were performed in an MBE chamber under ultra-high vacuum(UHV), at a base pressure of ∼2.5 × 10⁻¹⁰ mbar. Si(5 5 12) of size 8 × 3 mm² were prepared through cutting from commercially available p-type boron doped wafers (of resistivity of 10–15 Ω cm). Substrates were degassed at 600 °C for about 12 h followed by repeated flashing (with direct current (dc) heating) for 30 s at a temperature of 1250 °C to remove the native oxide layer to obtain a clean and well-reconstructed surface. The reconstruction has been confirmed with in situ reflection high energy electron diffraction (RHEED). The temperature was monitored with an infrared pyrometer calibrated with a thermocouple attached to the sample holder. Two series of samples have been prepared. 10 ML Ge deposition on Si(5 5 12) from RT to 800 °C, with two modes of heating, namely, (i) heating is achieved through a filament underneath (radiative heating: RH). (ii) heating achieved by passing a dc through the sample. The post-growth characterization of the samples was carried out ex situ by field emission gun based scanning electron microscopy (SEM), transmission electron microscope (TEM) and MeV ion scattering (Rutherford backscattering spectrometry (RBS)). To compare with growth on a high-index surface, we also carried out 10 ML thick Ge growth on a Si(1 1 1) substrate with a resistivity of 0.5–30 Ω cm. The base pressure inside the MBE chamber was ∼3.5 × 10⁻¹⁰ mbar. A dc of 1.5 A (5.7 V) as applied along the ⟨112⟩ direction. The post-growth characterization of the samples was carried out ex situ by field emission gun based SEM.

3. Results and discussions

In this report, we will emphasize on the shape transformations of Ge–Si structures on clean Si(5 5 12) surfaces as a function of substrate temperature in two modes of heating condition, i.e. by RH and direct current heating.
Figure 2. Histogram of (a) island size in 10 ML Ge/Si(5 5 12) system at RT and (b) bimodal size distribution in 10 ML Ge/Si(5 5 12) under RH condition.

Figure 3. Cross-sectional TEM images of 10 ML Ge/Si(5 5 12) at (a) 600 °C–RH (the marked line is to show a single island) (b) faceted island at 700 °C–RH (c) faceted dome structures at 800 °C–RH. (d) showing the epitaxial nature of faceted Ge–Si structures.

3.1. Shape evolution under RH condition

Figure 1 shows SEM micrographs for 10 ML Ge growth on the Si(5 5 12) surface at various substrate temperatures under RH condition. Figure 1(a) shows the FEGSEM image for the Ge film deposited, while keeping the substrate at room temperature (RT). The island size distribution of the random structures is given in figure 2(a). The mean size of the islands is found to be 504 ± 38 nm in diameter. Figure 1(b) depicts the morphology for the Ge film deposited at a substrate temperature $T = 600$ °C under RH condition. In this case,
a distribution of bigger spherical Ge islands surrounded by a large number of smaller islands was observed, showing a kind of Ostwald ripening process with a bimodal distribution of islands (figure 2(b)). The mean value for the size of the smaller islands was found to be 15.8±1.5 nm, while the bigger islands are found to have an average size of 54.0±7.0 nm. For the 700 °C deposition case, the mean size of the island increases at the expense of decreasing the number of smaller islands. Daruka et al [21] showed the shape evolution of islands to various faceted structures in terms of surface energy minimization with increasing volume. Normally, the chemical potential of an island decreases continuously with size, due to the smaller surface-to-volume ratio. As a result, material diffuses from smaller to larger islands. In this coarsening or ‘Ostwald ripening’, large islands grow while small islands shrink and disappear. Above a lower transition volume, the Ge–Si island is stable and has become faceted, as seen in figure 1(c). The mean size of the faceted island is 150±15 nm. To obtain a clearer view of these facet structures, cross-sectional transmission electron microscopy (X-TEM) analysis has been carried out. Figure 3 depicts the interfacial morphology of these Ge structures grown on Si(5 5 12) under RH conditions. In figure 3(a), the X-TEM image of spherical islands formed at $T = 700$ °C–RH has been displayed. Figure 3(b) shows a cross-sectional view of faceted island structure formed in the $T = 700$ °C case, where the thickness of the island is 42.6±2.3 nm. A set of Ge/Si(5 5 12) sample has been prepared keeping $T = 800$ °C. Upon increasing the temperature, it causes the formation of only larger dome-like structure with disappearance of smaller islands. Figure 1(d) displays the Ge–Si dome-like structures bounded by facets with average size of 630.5±16.4 nm. The thickness of the faceted dome structure is found from the cross-sectional TEM view in figure 3(c), which is 122±5.6 nm. We have also calculated the angles between the faceted planes and determined the orientation of facet planes as per the study carried out by Marks [22]. In the case of $T = 700$ °C–RH, the faceted island is bounded by the $[111]$ and $[100]$ planes and the faceted dome structure in the $T = 800$ °C–RH case are bounded by $[111]$ and $[311]$ set of planes. Figure 3(d) confirms that the faceted Ge–Si structures are epitaxial in nature.

Chaparro et al [23] showed that higher growth temperature activates additional pathways for the Ge islands to relieve their strain via Ge/Si intermixing. Si–Ge alloying causes the formation of quite large hut clusters for $T > 600$ °C. Furthermore, they observe that there is an increase in the mean dome cluster size with increasing temperature. RBS experiment has been performed on all the three sets of samples, i.e. Ge/Si(5 5 12) at $T = 600$, 700 and 800 °C to investigate the possible intermixing of Si–Ge. It is evident from figure 4 that there is Ge diffusion towards silicon for $T = 700$ °C and $T = 800$ °C. The simulated composition of Ge and Si are tabulated in table 1. Ge–Si alloying may also be attributed in the strain relief mechanism in forming the larger facets structures minimizing the surface energy.

![Figure 4. Experimental RBS spectrum from 10 ML Ge/Si (5 5 12) at $T = 600$, 700 and 800 °C in the RH case.](image)

| Layer number | Ge composition | Si composition | Thickness ($1 \times 10^{15}$ atoms cm$^{-2}$) |
|-------------|----------------|----------------|----------------------------------|
| 1           | 0.6000         | 0.4000         | 0.100                            |
| 2           | 0.0600         | 0.9400         | 100.0                            |
| 3           | 0.0075         | 0.9925         | 250.0                            |
| 4           | 0.0070         | 0.9930         | 300.0                            |
| 5           | 0.0050         | 0.9950         | 350.0                            |
| 6           | 0.0020         | 0.9980         | 400.0                            |
| 7           | 0              | 1              | Bulk                             |

3.2. Shape evolution under DH condition

We have discussed the temperature dependent shape evolution of Ge–Si islands under the RH condition. In this section, we investigate the morphological change with increasing temperature under the dc heated condition. Here, the dc current direction was parallel to the step direction, i.e. (1 1 0). Figure 5 is a SEM micrograph of Ge–Si structures at various temperatures in the DH case. When substrate temperature $T = 400$ °C (where the applied current was 1.1 A and voltage 5.8 V), the Ge–Si islands start nucleating to form random bigger islands with the reduction in smaller island density. The mean size of the bigger islands is 645.0±22.0 nm. On further increasing temperature to 800 °C (applied 1.8 A and 5.2 V) and above, the Ge–Si islands start evaporating and hence the island density decreases. The signal of Ge diffusion in this case can be seen from the RBS spectrum in figure 6. The interesting shape transition occurs at $T = 600$ °C by passing the dc through the sample with 1.5 A and 5.3 V. Here, well aligned Si$_{1-x}$Ge$_x$ trapezoidal microstrutures of average length 6.25±0.27 μm and aspect ratio 3.13±0.3 form along the step edge (1 1 0) (figure 5(c)). In figure 6, the RBS measurement has revealed the intermixing of Ge and Si. Additionally, the inward diffusion of Ge (channels 1250–1350) into bulk Si and the outward diffusion of Si (channels 900–940) can be clearly seen from figure 6. The RBS simulation has been carried out treating the specimen as a multilayer with variation of Si and Ge...
concentrations in each layer (table 2). We found a graded composition of Ge–Si in these structures. The synchrotron-based high resolution x-ray diffraction (HRXRD) also showed the presence of the graded Si$_{1-x}$Ge$_x$ composition from the interface to the top of the trapezoid structures (data not shown).

Under the DH condition, dc current acted as an additional factor in stimulating the motion of germanium adatoms towards the edges of steps. In [24], Rönspies et al showed that steps in a high-index surface, apparently do not interrupt the conducting path, but due to local scattering at step edge increase the local resistivity by more than one order of magnitude. Hence, the local temperature along the step direction is more and stimulates the Ge–Si diffusion along the steps making the structures well elongated.

Table 2. Parameters obtained on fitting the experimental RBS data for Ge/Si(5 5 12) at $T = 600^\circ$C–DH, using the SIMNRA simulation code.

| Layer number | Ge composition | Si composition | Thickness ($1 \times 10^{15}$ atoms cm$^{-2}$) |
|--------------|----------------|----------------|---------------------------------------------|
| 1            | 0.7500         | 0.2500         | 0.75                                        |
| 2            | 0.0750         | 0.9250         | 2.0                                         |
| 3            | 0.0070         | 0.9930         | 150.0                                       |
| 4            | 0.0050         | 0.9950         | 250.0                                       |
| 5            | 0.0040         | 0.9960         | 350.0                                       |
| 6            | 0.0020         | 0.9980         | 500.0                                       |
| 7            | 0.0010         | 0.9990         | 3500.0                                      |
| 8            | 0              | 1              | Bulk                                        |

Figure 5. SEM micrographs of 10 ML Ge/Si(5 5 12) at (a) RT (b) 400 $^\circ$C–DH (c) 600 $^\circ$C–DH (d) 800 $^\circ$C–DH.

Figure 6. Experimental RBS spectrum from 10 ML Ge/Si (5 5 12) at $T = 400, 600$ and 800 $^\circ$C in the DH case.

To see the effect of dc on low index silicon surfaces, we performed similar growth and characterization for the Ge/Si(1 1 1) system. Figure 7 depicts the SEM micrograph for 10 ML deposited Ge film on the Si(1 1 1) surface under...
DH condition. In this case, the dc direction was applied along (1 1 2). Here irregular structures are formed with no proper alignment.

4. Conclusion

In summary, we report the shape evolution of MBE grown Si$_{1-x}$Ge$_x$ islands on reconstructed high-index Si(5 5 12) surfaces, as a function of growth temperature. Also, the mode of heating (i.e., RH and DH) plays a vital role in the shape transformations. We show that a self-assembled growth at optimum temperature leads to interesting shape transformations, namely, spherical islands to faceted dome structures in the case of RH condition and to elongated trapezoidal structures for the DH case. We have also observed the intermixing of Si–Ge in the larger faceted dome structures and aligned trapezoidal structures. We look forward to carrying out a spectroscopic analysis along the step edges and of the aligned Si$_{1-x}$Ge$_x$ structures as well.

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