Shape transformation of Si$_{1-x}$Ge$_x$ structures on ultra clean Si(5 5 7) and Si(5 5 12) surfaces

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Abstract. We report growth of Ge nano/microstructures on ultra clean, high vicinal silicon surfaces of Si(5 5 7) and Si(5 5 12) under two substrate heating conditions: direct current (DC) and radiative heating (RH). These were grown under ultra high vacuum conditions while keeping the substrate at a temperature of 600°C. The results for 10 monolayer (ML) and 12 ML thick Ge deposited on the above surfaces show spherical island structures for RH conditions while aligned trapezoidal structures were observed under DC conditions of heating. We find that the longer side of trapezoid structures are along <110> irrespective of DC current direction.

In the case of 10 ML Ge deposited on Si(5 5 7), elongated Si$_x$Ge$_{1-x}$ nanostructures with an average length of $\approx 300$ nm and a length/width ratio of $\approx 3.3$ have been formed along the step edges. Under similar conditions for 10 ML Ge growth on Si(5 5 12), we found aligned Si$_x$Ge$_{1-x}$ trapezoidal microstructures of length $\approx 6.25$ $\mu$m and an aspect ratio of $\approx 3.0$. Scanning transmission electron microscopy (STEM) measurements showed the mixing of Ge and Si at the interface and throughout the over-layer. Detailed electron microscopy studies (scanning electron microscopy (SEM) and STEM) reveal the structural aspects of these microstructures.

1. Introduction

The use of high index silicon surfaces as substrates can enhance the formation of aligned nanostructures with self-assembled growth [1–5], 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 and vicinal 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. These self-assembled nano-structures can be used as templates for the growth of low dimensional semiconductor and metallic nanostructures with controlled size and periodicity. In the past, the growth of Ge nanostructures on low index silicon substrates (such as Si(100), Si(111)) has been studied extensively. However, less attention has been paid to Ge growth on
high-index Si substrates. Hence, the study of self-assembly of Ge on high-index silicon surfaces would be an important field of research in obtaining various types of nanostructures that may be useful in optoelectronic devices. The lattice mismatch between Ge and Si (≈4.2%) is an important property of such quantum devices, as it determines the maximum thickness of smooth films (quantum wells) and controls band structures, mobility of carriers, and quantum confinement potential.

Study of Ge growth on high index surfaces, such as Si(5 5 7), Si(5 5 12) surfaces, is an area where very limited research work has been carried out [1–5]. Among the high index silicon surfaces oriented between (001) and (111), Si(5 5 12) exhibits a relatively stable reconstruction having 1D symmetry with a (110) mirror plane [5, 6]. Si(5 5 12) is oriented 30.5° away from (001) towards (111) with one-dimensional periodicity over a large unit cell [5]. Si(5 5 7) has a vicinal angle of 9.45° from (111) towards (112) [7]. The highly corrugated triple step structure of the Si(5 5 7) surface is easier to detect than other long-range surface reconstructions. In both the Si(5 5 12) and (5 5 7), the step edges are parallel to the <110> direction.

In this paper, we report the comparative study of aligned Si-Ge structures formed at the interface of ultra clean reconstructed Si(5 5 12) and Si(5 5 7) surfaces and Ge, where the substrates were heated by direct current (DC) and radiative heating (RH) during deposition of Ge. The size of the aligned Si-Ge Structures depends on the substrate orientation and over-layer thickness.

2. Experimental Details

The Ge depositions on Si(5 5 7) and Si(5 5 12) were carried out under ultrahigh vacuum (UHV) condition in a custom-designed molecular beam epitaxy (MBE) system with a base pressure of ≈2.5 × 10^{-10} mbar in the growth chamber. The substrates used were of size 8 × 3 mm² cut from p-type (boron doped) single crystal Si wafers with a resistivity of 10 to 15 Ω cm. Atomically clean, reconstructed surfaces were prepared by following the usual thermal cleaning procedure: the samples were first degassed at about 600 °C for 12–14 h followed by a repeated flashing for 30 s at ≈1250 °C. The reconstructed surfaces were confirmed by in-situ reflection high energy electron diffraction (RHEED). Ge was deposited to various thicknesses (3.0 – 12.0 ML) at a typical deposition rate of 0.6 ML min⁻¹ (one ML corresponds to 7.8 × 10^{14} atoms cm⁻² for a Si(111) surface). The substrates were kept under three substrate heating conditions: (i) at room temperature, (ii) at 600°C where RH is achieved through a filament underneath, and (iii) at 600 °C but with DC heating achieved by passing a direct current. In the second case, the sample was annealed at a temperature of 600 °C for 15 min after deposition using RH, and for case (iii) the annealing was done by passing the direct current through the sample with a power of 7.95 W (1.5 A and 5.3 V) corresponding to a temperature of 600 °C . The post-growth characterization of the samples was carried out ex-situ by field emission gun scanning electron microscopy (FEGSEM) and scanning transmission electron microscopy (STEM) based x-ray energy dispersive spectrometry (EDS). The FEGSEM measurements were carried out with 20 kV electrons using a Neon 40 cross-beam system (M/S Carl Zeiss GmbH) at Institute of Physics, Bhubaneswar, India. STEM and TEM measurements were carried out with 300 keV electrons using a Cs-corrected FEI Titan 80/300 system at University of Bremen, Germany. The cross-sectional TEM (XTEM) specimen was prepared for 12 ML Ge grown on Si(5 5 12) under DC heating conditions. The preparation of the specimen to electron transparency was achieved by focused ion beam milling followed by low-energy-ion milling.

3. Results and Discussion

Here we present results for the 10 ML and 12 ML Ge film thickness. For the case of RT deposition of 10 ML thick Ge on Si(5 5 12), the average island diameter was found to be 504 ± 38 nm (data not shown). Similar random structures are observed for 10 ML Ge on Si(5 5 7) under RT conditions. Figure 1(a) and 1(b) show the morphology for 10 ML Ge on Si(5 5 12) and Si(5 5 7), respectively, at a
substrate temperature of 600°C followed by a post-anneal of 15 min at the same temperature with RH heating. In this case, a distribution of bigger circular Ge islands surrounded by a large number of smaller islands was observed. This type of distribution is consistent with Ostwald ripening. Figure 1(c) shows the typical particle size histogram for 10 ML Ge/Si(5 5 12)-RH depicting a bimodal size distribution with an average value for the size of the smaller islands of 15.8 ± 1.5 nm and for the bigger islands of 54.0±7.0 nm. In figure 1(d), a bimodal distribution of particle size is shown for the spherical Ge islands grown on Si(5 5 7) in RH condition as well. In this case, the number of bigger islands is also significant, unlike in the case of Si(5 5 12). The average values for the size of the smaller islands and bigger islands are 12.8 ± 0.5 nm and 37.2 ± 2.1 nm, respectively.

Figure 1. (a) and (c) SEM image and islands size histogram of 10 ML Ge/Si(5512) at 600 °C – RH. (b) and (d) SEM image and island size histogram of Ge/Si(557) at 600°C-RH

Figure 2. (a) and (c) SEM image and histogram/length of DC heated 10 ML Ge/Si(5512) at 600 °C. (b) and (d) SEM micrograph and histogram (length) of Ge/Si(557) at 600 °C-DC

We have shown circular shapes of Ge structures for the RH conditions in figures 1(a) and (b), but for the case of DC heating, we found a different shape evolution of the islands [8]. Figure 2(a) and (b) depict the formation of ordered trapezoidal structures for both Ge/ Si(5 5 12) and Ge/Si(5 5 7) cases in which direct current (DC) was passed through the sample. The histogram for length of the trapezoidal structures is shown in figure 2(c) and (d). The length of the aligned structures for Ge/Si(5 5 12) and Ge/Si(5 5 7) systems are (6.2 ± 0.3) μm and (297 ± 11.2) nm, respectively. Though the reason for the larger island structures is not yet understood, our results show that substrate orientation indeed makes a difference in size of self-assembled structures. Interestingly, similar length/width ratio ([≈3.1 for Ge/Si(5 5 12) and ≈3.3 for Ge/Si(5 5 7)] were found. The elongated trapezoidal structures are formed along the step edges irrespective of the DC current direction. In all the cases in which the direction of DC current was changed, we have found that the trapezoidal structures aligned in the step direction <110> only [8].

In the following, we present the results on the composition determination of Si$_x$Ge$_{1-x}$ layer using STEM-EDS measurements with 300 keV electron beams. STEM measurements were carried out on 12 ML Ge deposited on Si(5 5 12) substrate under DC heating conditions. Figure 3(a) shows a STEM image taken from this specimen. As shown in Figure 2(a), the SiGe trapezoidal structures are of an average length of 6.2 μm. Figure 3(a) shows a cross-section image of this island, which is a small part of the bigger island. For making a cross-sectional FIB specimen, two kinds of protective Pt depositions were made (one using e-beam deposition and a larger thick one with FIB based gas injection system). Pt shown in Fig. 3(a) is deposited by gas injection with e-beam. The energy...
dispersive spectrum in (b) shows Ge and Si signals from the region that is shown with a dot on the line XY in Figure 3(a). Figure 3(c) depicts the integrated and normalized (to maximum value of Si and Ge signals, respectively) intensity of Si and Ge signals obtained at various points along the line XY (300 nm length, shown in figure 3 (a)). The intensity line profile confirms the variable composition of Si and Ge in the island layer, in agreement with our ion scattering measurements [8]. Figure 3(d) shows a high resolution STEM image. From this figure it is clear that the film of Si\(_x\)Ge\(_{1-x}\) is crystalline in nature and grown epitaxially on Si (5 5 12). These results show that it is possible to make aligned structures of variable sizes and graded composition. In general SiGe alloy layers are formed by either co-deposition of Si and Ge or by depositing buffer-layers of Si before the Ge deposition and followed by vacuum annealing.

4. Summary
In this work, we presented a one-step procedure to form a graded composition of Si\(_x\)Ge\(_{1-x}\) epitaxially grown on clean high index surfaces. Trapezoidal structures of similar length/width ratio (≈ 3.1 – 3.3) were observed. The longer side of the trapezoidal structures is ≈ 21 times larger in case of Si(5 5 12) compared to Si(5 5 7).

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