

Nucleation of Ge quantum dots on the C-alloyed Si(001) surface

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Abstract

Carbon pre-deposition onto the bare Si(001) surface has been shown to alter the (2×1) surface structure by formation of $c(4 \times 4)$ reconstructed domains containing a high C-concentration. Here we studied by ultra-high vacuum scanning tunneling microscopy the effect of this restructured surface on the initial stages of Ge nucleation by molecular beam epitaxy. Ge is found to form three-dimensional (3D) islands already at sub-monolayer coverage, resulting in a Volmer–Weber growth mode. Strain effects repel Ge adatoms from the C-rich domains, leading to enhanced Ge island formation on the C-free surface regions in between the $c(4 \times 4)$ areas. At a low growth temperature of 350°C, very small three-dimensional islands (3–5 nm in diameter, height 3–4 ML) with a density of nearly $1 \times 10^{12} \text{ cm}^{-2}$ are obtained for only 0.5 ML of Ge. At higher substrate temperatures of approximately 500°C this three-dimensional growth mode is less pronounced, but still evident. The initially nucleated three-dimensional islands define the positions of the larger quantum dots at higher Ge coverage, that exhibit enhanced photoluminescence (PL) properties. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Ge quantum dots; Molecular beam epitaxy; In situ scanning tunneling microscopy

1. Introduction

It is a long term goal of many research teams to develop Si-based structures, which permit the monolithic integration of electronic and optical devices. Adding Ge to the Si crystal, opens several routes towards this aim. An enhancement of radiative recombination of excited carriers was observed in Si/Ge superlattices, originating in the quasi direct band gap as a result of zone folding [1]. Lately, a lot of attention

was paid to the growth and investigation of the structural [2,3] and optical [4,5] properties of Ge islands embedded in Si. On the bare Si surface Ge islands are formed by the Stranski–Krastanov growth mode [2]. Their size and nucleation density depends on deposition temperature and rate. At temperatures below 450°C, where small islands are obtained, the optical properties of Si/Ge structures are generally deteriorated due to an enhanced incorporation of point defects [6]. Theoretical calculations indicate that extremely small islands and a pronounced confinement of the carriers are required to enhance the optical transition probability in Si-based nanostructures [7]. A considerable reduction of the Ge island size at moderate growth temperatures (in the range of 500°C) was

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observed by growing Ge islands on a C-alloyed Si surface [8]. These C-induced Ge islands exhibit enhanced PL intensity [8].

In earlier studies the C-alloyed Si(001) surface was analysed in detail by in situ ultra-high vacuum scanning tunneling microscopy (UHV-STM) [9,10]. It was established that this surface contains C-rich areas exhibiting a $c(4 \times 4)$ reconstruction and areas with very little or no C content showing a 2×1 reconstruction of Si(001) surface with buckled dimers. The inhomogeneous distribution of the C induces an undulating strain field on the Si(001) surface. In this study we used the UHV-STM to get insights of the initial stages of the Ge nucleation on this C-alloyed Si surface.

The samples were prepared by molecular beam epitaxy (MBE) using e-gun evaporation of Si and Ge. Prior to growth they were cleaned by a wet chemical etch and baked in the MBE chamber at 900°C for 30 min. First a 100-nm thick Si buffer layer was deposited at 700°C. For the C and Ge deposition the temperature was reduced to the range between 350 and 520°C. Si was evaporated at a rate of 0.1 nm/s. Carbon was sublimated from a hot pyrolytic filament. Subsequently, Ge was evaporated at a rate of 0.01 nm/s. After the deposition process the samples were transferred under UHV into the STM chamber. A detailed description of the STM system is given elsewhere [11].

2. Results and discussion

Fig. 1 shows four STM images of C-induced Ge dots. The Ge was deposited at 520°C (Fig. 1a,c) and at 350°C (Fig. 1d). Prior to Ge deposition, 0.1 ML of C was deposited at 520°C. The Ge coverage was increased from 0.5 ML (Fig. 1a,d) to 1 ML (Fig. 1b) and to 2.5 ML (Fig. 1c). At 520°C the Ge growth appears to be dominated by step flow on a first glance, the image is stepped downwards from the right to the left (Fig. 1a). Looking in more detail, it is found that the S_A steps are predominately formed by a 2×1 reconstruction exhibiting buckled dimers and missing dimer rows, which is indicative for Ge deposition. In contrast the S_B steps are often formed by rows showing a $c(4 \times 4)$ reconstruction induced by the C-deposition [9]. Generally, quite large areas are $c(4 \times 4)$ reconstructed, indicating that the Ge is repelled by the C-rich areas and Ge starts to nucleate on the areas which had the 2×1 reconstruction of Si. The C at surface lattice sites reduces the lattice constant in the $c(4 \times 4)$ areas, thus the bigger Ge atoms are easier attached to the surface in areas exhibiting the 2×1 Si reconstruction, i.e. areas free of C or at least with a very low C concentration.

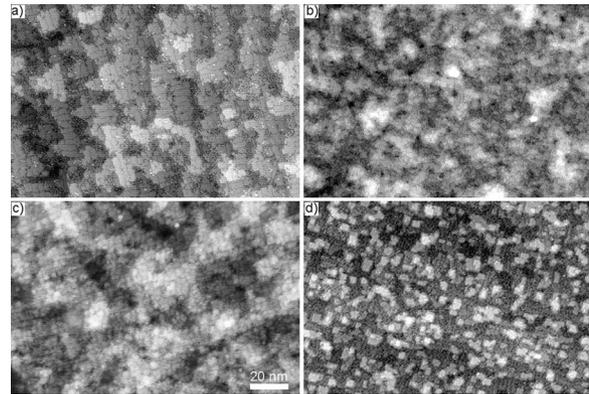


Fig. 1. STM images of C-induced Ge quantum dots on a Si(001) surface, taken after the deposition at 520°C of: (a) 0.1 ML C and 0.5 ML Ge; (b) 0.1 ML C and 1 ML Ge; and (c) 0.1 ML C and 2.5 ML Ge; (d) is obtained after deposition of 0.1 ML C at 520°C and 0.5 ML Ge at 350°C.

After the deposition of 1 ML Ge (Fig. 1b) still substantial surface portions are covered with the C-induced $c(4 \times 4)$ reconstruction. Consequently, Ge does not wet the entire surface. Ge island growth is more pronounced and the RMS roughness has increased considerably to 0.17 ± 0.02 nm when compared to the deposition of 0.5 ML Ge (RMS roughness: 0.10 ± 0.02 nm). The island density remains constant at a value of $3 \times 10^{11} \text{ cm}^{-2}$. A continuation of Ge supply to a coverage of 2.5 ML (Fig. 1c) leads to a further increase in RMS roughness. However, the $c(4 \times 4)$ reconstructed areas are no longer visible indicating that the Ge islands grew in diameter, overlapping the C-rich areas. It is expected that the C also starts to redistribute and to intermix with the underlying Si and the Ge top-layer. The Ge islands have typical diameters of 10–15 nm and a height of 7 ML. They are not formed by the Stranski–Krastanov growth mode since no wetting layer develops. The islands are created by a modified Volmer–Weber three-dimensional growth mode on a surface with undulating strain fields.

At 350°C, where the adatom mobility is reduced and interdiffusion is suppressed, this mode becomes more pronounced. At 0.5 ML Ge coverage (Fig. 1d) the island density increases to approximately $9.5 \times 10^{11} \text{ cm}^{-2}$ and islands already pile up to 3–4 ML in height (RMS roughness 0.15 ± 0.02 nm). Noticeably, the islands have predominantly a rectangular, almost quadratic shape, in contrast to the elongated shape of nucleation centers found for low temperature MBE on 2×1 reconstructed surfaces [12]. This might be attributed to the C-induced restriction of the area available for nucleation and to the fact that this area contains only buckled Si dimers. Another interesting observation is that many islands are placed at step edges, forming the border between adjacent terraces in

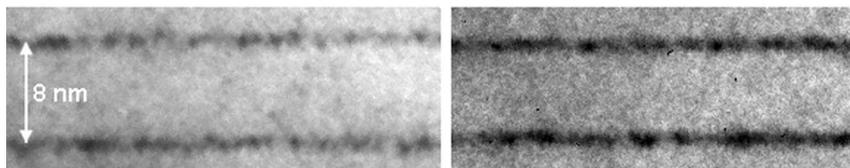


Fig. 2. Cross-sectional TEM images of C-induced Ge dots embedded in Si: (a) 0.2 ML of C and 0.4 ML of Ge; (b) 0.2 ML of C and 0.8 ML of Ge deposited at 460°C.

those places, where the $c(4 \times 4)$ reconstruction does not reach the step edge.

Embedding these islands in Si leads to slight intermixing of the Ge with the Si cap layer. Generally high-resolution transmission electron microscopy (TEM) reveals an increase in islands height. Fig. 2 shows two cross-sectional TEM micrographs of samples containing multiple layers of C-induced Ge dots fabricated with 0.2 ML of C and 0.4 ML (a) and 0.8 ML (b) of Ge at a deposition temperature of 460°C. Both samples show a high density of small Ge islands, indicated by the strain fields (dark areas in Fig. 2). They become more pronounced with the increase in the nominal Ge coverage. The contrast visible in the areas between the Ge dots is attributed to the C-rich areas.

Fig. 3 depicts three low temperature (1.8 K) photoluminescence (PL) spectra of Ge dots induced by 0.1, 0.2 and 0.3 ML of C. The Ge deposition was constant at 2 ML. Each sample contains 10 dot layers separated by 8-nm wide Si spacer layers. Besides the broad PL of the dots the spectra contain narrow lines that are due to the Si substrate. The PL signal related to the dots is shifted by 180 meV towards lower energies by increasing the C deposition from 0.1 to 0.3 ML. This shift is predominantly attributed to an increase in dot height leading to increase in the hole-confinement energy. As

illustrated in the schematics in Fig. 3, the increase in C deposition leads to a higher portion of the surface covered with the $c(4 \times 4)$ reconstruction, leaving smaller areas for the nucleation of Ge dot. Consequently, the dots shrink in diameter and grow in height for identical Ge coverage.

3. Conclusion

The detailed structural analysis of the nucleation mechanism of Ge dots on the C-alloyed Si(001) surface using STM as well as TEM reveals that the Ge dots nucleate in the areas between the $c(4 \times 4)$ patches. The shape of the dots can be modified by the growth temperature and the amount of C deposited prior to the Ge deposition. At low growth temperatures as well as at increased C coverage the dots grow in height and shrink in diameter leading to an increase in the confinement energy.

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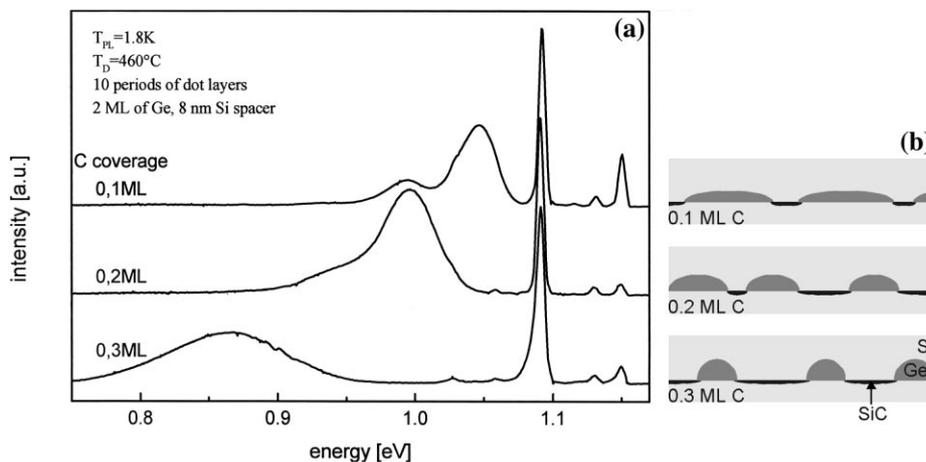


Fig. 3. 1.8 K PL spectra of Ge dots induced by 0.1, 0.2 and 0.3 ML of C. The schematics illustrate the effect of the variation of the C deposition on the shape of the Ge dots.

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