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Coalescence of domes and superdomes at a low growth rate or during annealing: Towards the formation of flat-top superdomes

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ABSTRACT

We investigate the morphology of islands obtained by epitaxial growth of Ge on Si(001). During a low growth rate or during annealing and depending of the quench method, we observe the formation of a new type of superdomes, which exhibit additional {15 3 23} facets, an increase of their {001} and {105} facet area on their top and a smaller aspect ratio compared to known superdomes. This flat-top superdome shape results from the complex evolution of domes and superdomes: coalescence, intermixing and ripening. Dome or superdome coalescence which results in the newly observed shape appears as a dominant pathway towards dislocation nucleation for the investigated temperature range.

1. Introduction

Self-assembled semiconductor nanostructures are the subject of intense investigations since they present potential applications in future nanoscale devices [1] such as quantum-dot lasers and memories. The physical properties of such nanostructures depend on their size, shape, strain and composition, which thus have to be fully understood. The growth of Ge on Si(001) is one of the most interesting system due to its model character and its potential applications. It follows the Stranki-Krastanow mode, which has been extensively investigated [2]. The growth is characterized by the formation of a pseudomorphic wetting-layer up to 3–4 deposited Ge monolayers (ML). By increasing the Ge coverage, the strain energy stored in the wetting-layer partially relaxes via the formation of square pyramids having {105} facets which then evolve upon further deposition into dome-shaped islands with {105}, {113} and {15 3 23} facets, and the top (001) facet [3,4]. Finally, the dome islands may evolve into islands called 'barns' with additional {111} and {20 4 23} facets [5,6]. They are still of coherent nature and precede the appearance of large, dislocated islands named 'superdomes' [7], exposing the same facets as barns, but with different relative sizes. The growth and properties of coherent islands during the pyramid-to-dome and dome-to-barn transitions have been studied extensively (see Refs. [3,8–15] as examples), while the growth and evolution of dislocated islands have been less analyzed [7,16–23]. In 2003, Capellini et al. [24] investigated the role of the growth rate on the occurrence of superdomes. These authors observed that the density of superdomes increases with decreasing growth rate. Recently, a selective etching method was used to reveal the evolution and morphological age of superdomes [23]. By using this technique, island coalescence has been determined as the dominant pathway towards dislocation nucleation at low temperatures while at higher temperatures, anomalous coarsening is effective. Other experimental works [25,26] have revealed the remarkable lateral motion of islands leading to asymmetric intermixing and nonuniform composition profile of islands during post-growth annealing. These phenomena have recently been explained theoretically by using thermodynamic driving forces [27], which showed that the morphological and compositional evolution of islands are strongly coupled. Furthermore, the critical size and shape for dislocation nucleation in SiGe dome-shaped islands have been calculated by using a realistic three-dimensional model [28].

In this paper, we report the formation of superdomes with a new shape. They are observed at low growth temperatures (773–923 K) and at low growth rates (here, ~0.006 ML/s) and result from the complex evolution of domes or superdomes: coarsening, inter-

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mixing and ripening. These results provide new insight into mechanisms governing shape transformations of incoherent faceted semiconductor islands.

2. Experimental

The samples were grown by molecular-beam epitaxy (MBE) in a devoted ultra-high vacuum chamber equipped with large beryllium windows and coupled to a surface diffractometer for X-ray diffraction, on the BM32 synchrotron beamline at the ESRF, Grenoble [29]. The Si(001) substrates were deoxidized by annealing at 1200 K until a sharp, 2×1 reconstructed, reflection high energy electron diffraction (RHEED) pattern was observed. Germanium was deposited with a Knudsen cell whose slow deposition rate (170 s for one Ge monolayer (ML)) was *in situ* calibrated using both a quartz microbalance and X-ray reflectivity. Ge was deposited monolayer after monolayer at three growth temperatures (823 K, 873 K and 923 K). Ten to eleven monolayers (ML) of Ge were deposited. After completion of each monolayer, the samples were immediately cooled to 723 K for subsequent X-ray analysis (not shown here) and to avoid possible annealing effects [30,31]. Another sample was grown at 823 K with 8 ML of Ge deposited and with growth interruption after completion of each monolayer. In order to investigate annealing effects, it was then kept at 823 K for one day before cooling to room temperature. The surface morphology of all these samples was next investigated *ex situ* by means of atomic force microscopy (AFM) in tapping mode.

3. Results and discussion

Fig. 1 shows AFM images of samples obtained upon deposition of 10–11 ML Ge at 823 K, 873 K and 923 K. A bimodal growth is observed with the coexistence of domes, possible barns (small islands with a diameter of ~100 nm), and superdomes (large islands with a size of ~400–600 nm) which are observed for all growth temperatures. With increasing temperature, the island density clearly decreases for the small islands (domes, barns) (Fig. 1d–f) and increases for superdomes as an effect of surface diffusion and anomalous coarsening which are enhanced at high temperatures. To discriminate between coherent islands and dislocated superdomes, a study of the island volume and island shape has been performed. Most islands which present {111} facets are no longer regularly shaped and some island volumes are even higher than 1×10^7 nm^3, which is larger than the critical volume ($\sim 0.4 \times 10^6$ nm^3) for the transition from coherent to dislocated islands [9] on flat (001) substrates. This confirms the presence of dislocated superdomes on each sample. Superdomes present much larger diameters than domes: as observed by LeGoues et al. [16], once a dislocation has been introduced, dislocated islands become more relaxed and are thus preferred with respect to domes for Ge attachment. Superdomes then grow much faster than the surrounding domes.

Fig. 2 shows the morphology of two types of superdome islands, named "flat-top superdome" (Fig. 2a–c) and "*usual* superdome" (Fig. 2d–f) and observed on all samples. In the following, the "flat-top superdome" designation will be explained. First, we have analyzed their facets using the procedure given in Refs. [32,33] by performing facet orientation plots (see Fig. 2c and f). The facet orientation plots have been restricted to the island surface without including the substrate. The conventional {105}, {113} and {15 3 23} facets of dome islands [34,35] appear for both "flat-top superdome" and "*usual* superdome" islands. Additional spots can be recognized as arising from steeper facets. They correspond to facets usually observed at the base of superdome islands [7]: {111} and {20 4 23} facets. The superdome, named "*usual* superdome", has the conventional superdome shape showing these five facets: {105} shallow facets at the top, medium steepness facets ({113}, {15 3 23}) and steep facets ({111}, {20 4 23}) at the bottom of the island. The other superdome-island, named "flat-top superdome" (Fig. 2a–c) shows the conventional superdome facets: {105}, {113}, {15 3 23}, {111} and {20 4 23} facets but their arrangement is different. The island surface shows unusual concave areas labelled by a question mark '?' in Fig. 2b. The facets in theses regions appear as surface indentation in the middle of the usual {15 3 23} facets. They can also be indexed as {15 3 23} facets. But they have an orientation different from the usual {15 3 23} facets that they indent.

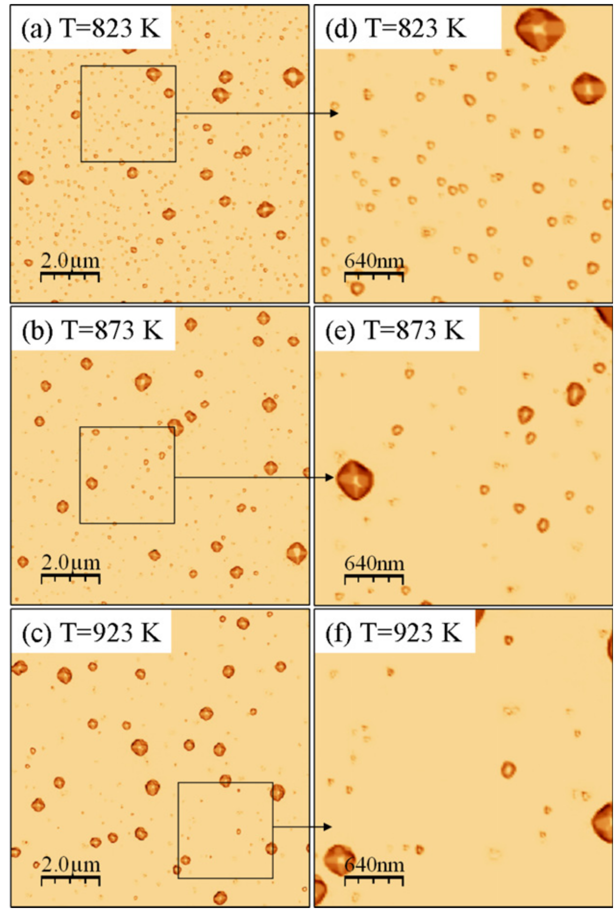

Fig. 1. AFM images of samples obtained after deposition of 10–11 ML of Ge at 823 K (a, d), 873 K (b, e) and 923 K (c, f). Figures on the right side correspond to zooms.

Indeed, to clearly determine the nature of these concave facets, we measured the facet contact angles by using AFM. From the knowledge of the crystallographic orientation of the line defined by the intersection of the facet plane and the substrate plane, this contact angle allows the facets of Ge superdomes to be indexed. A cross-sectional line scan through the island center along the [100] direction and passing along the intersection of the two additional facets was performed and showed a contact angle of $(33 \pm 1)^\circ$ (see Fig. 3b). The possible additional facets which appear along the [100] direction can be either {15 3 23} or {42023} facets as no other additional spots are observed on the facet plot (Fig. 2c). They correspond to contact angles of 33° or 41.5°, respectively. The measured contact angle thus corresponds to {15 3 23} facets. We can thus conclude that the two additional facets with the same size in the left-hand corner of Figs. 2b and 3a are additional {15 3 23} facets. A similar observation can be performed on the opposite side of the superdome, where, contrary to the other side,