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Nucleation and evolution of $Si_{1-x}Ge_x$ islands on Si(001)

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Abstract

In this study, we report a systematic investigation of the metastable morphologies of $Si_{1-x}Ge_x$ layers obtained by the interplay of kinetics and thermodynamics during growth on Si(001). We show that three main growth regimes can be distinguished as a function of the misfit and of the deposited thickness. They correspond to three equilibrium steady state morphologies that consist of (105)-facetted hut islands, huts and domes in co-existence, and a bimodal size distribution of domes, respectively. The shape transitions between these states are attributed to different levels of relaxation.

Keywords: Self-organisation; Si; Ge; Islands; Molecular beam epitaxy; 2D–3D growth transition; Stress relaxation

1. Introduction

During the past 10 years, intensive research has been produced in the field of self-organisation growth of Ge quantum dots on Si(001). It has been mainly motivated by the goal of the microelectronics industry to integrate optoelectronic devices based on nanostructures (quantum dots, wires or wells) onto Si chips. Even if this goal is far from being achieved as yet (mainly because of the large lateral sizes of islands and of the random nature of island nucleation), impressive progress has been obtained in the understanding and control of the 2D–3D growth transition. In fact, the major interest of this system is now more fundamental, lying in the growth and relaxation mechanisms of strained epitaxial layers. Indeed, the deposition of Ge on Si(001), commonly described as a classic Stranski–Krastanov (SK) process, has recently been shown to be considerably more complex than the simplified SK growth scheme. Briefly, Ge deposition evolution can be summarized as follows [1–3]: in a first stage, growth proceeds in a layer-by-layer mode, characterised by a fast increase of the ML step density. 3D islands start to form just after this surface roughening, probably using the micro-roughness as nucleation centres. As coverage continues to increase, different temperature-dependent metastable morphologies occur (hut clusters, domes, ...), which finally undergo a phase transition towards very large (≥ 1 μm) dislocated islands [4]. However, in this scheme of growth, the origin of the first stage of roughening is still under debate (kinetic- or stress-driven instability). In addition, the origin of the bimodal size distribution of islands and the transition from one size to the other is a matter of controversy. Moreover, the evolution of the $Si_{1-x}Ge_x$ undulation (obtained at low misfit) is also much more complex than thermodynamics predicts [5], because these metastable morphologies are even more sensitive to kinetic effects and present a situation far from equilib-

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rium. Finally, experimental results seem to diverge to some extent, probably because of the lack of reproducibility of the growth parameters among research groups [6].

In this paper we report a systematic investigation of $Si_{1-x}Ge_x$ island morphologies ($0.15 < x < 0.5$) during growth and annealing. We concentrate on the metastable morphologies that occur in between the 2D–3D growth transition (with the exception of dislocation-induced morphologies). We show that while kinetic undulations form at low misfit regime, well-separated islands of different shapes form at higher misfit regimes. We explain the stabilisation of these different shapes by the different levels of elastic relaxation observed in each type of island. For instance, relaxation levels of approximately 0.2 and 0.8% (slight variations are also found as a function of the initial stress) are attributed to hut and dome shapes respectively. Equilibrium steady state morphologies obtained after ~70 h annealing at 550°C remain as huts and domes, with similar levels of relaxation.

2. Experimental

$Si_{1-x}Ge_x$ layers were grown in a Riber molecular beam epitaxy (MBE) system with a base pressure of $\sim 10^{-11}$ torr. Si and Ge were deposited from an electron beam evaporator and an effusion Knudsen cell, respectively. Si/Ge flux ratios were adjusted to obtain $Si_{1-x}Ge_x$, with x between 0.15 and 0.5. The deposition rate of Si was maintained constant, at approximately 0.3 Å/s. The growth temperature used was 550°C.

Low resistivity nominally singular Si(001) wafers (miscut < 0.5°) were chemically cleaned ex situ and protected with an oxide cap layer during the final step. Subsequent in situ cleaning consisted of thermal desorption of the oxide layer at a temperature of approximately 900°C. A 50-nm thick Si buffer layer was systematically grown to achieve a reproducible surface, whose cleanliness was qualitatively checked by the RHEED intensity of the (2×1) reconstruction streaks.

In situ control of the growth mode and stress relaxation was achieved using real-time acquisition of RHEED patterns along either the $\langle 100 \rangle$ or $\langle 110 \rangle$ azimuth.

Morphological characterization was performed by atomic force microscopy (AFM) in air using an autoprobe CP (from Park Scientific Instruments) in non-contact mode.

Lateral elastic relaxation ($\Delta\varepsilon_{xx}$) was deduced from the measurement of the (220) bulk peak diffraction obtained by grazing incidence X-ray diffraction (GIXRD).

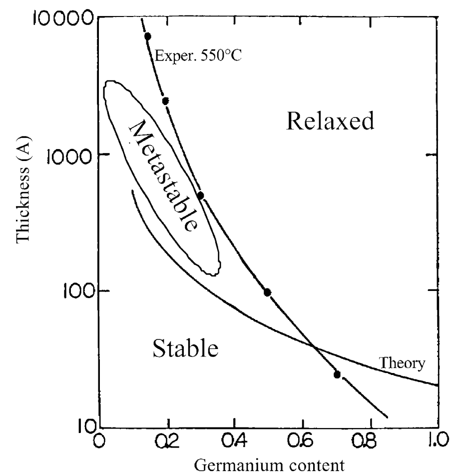

Fig. 1. Schematic view of the different states (strained, metastable and relaxed) of $Si_{1-x}Ge_x$ alloys as a function of the thickness and of the Ge concentration.

The $Si_{1-x}Ge_x$ deposited thickness was adjusted to maintain the structures in metastable states (Fig. 1), just below the critical thickness of dislocation nucleation (h_{cr}), but higher than the critical thickness for islanding (H_{cr}).

3. Results

The evolution of $Si_{1-x}Ge_x$ metastable morphologies can be broken into three main regimes, which are summarized as a function of the two most relevant experimental parameters (the deposited thickness h and the misfit ε), on the phase diagram given in Fig. 2. This 'kinetic phase diagram' has been drawn from the morphologies of as-grown layers that are stabilised by both thermodynamic and kinetic parameters in our growth conditions. In the following, we mainly concentrate on the layer morphologies obtained in regimes I and II, and on the transition from regime I to II. We only briefly summarise the results obtained in regime III.

In regime III, a bimodal size distribution of domes is observed. For instance, Fig. 3 presents two groups of islands with $L_1 \sim 1.3$ μm and $L_2 \sim 0.7$ μm, which were obtained for $h = 15$ nm and $\varepsilon = 2.1\%$. They correspond to dislocated and coherent states, respectively. Hut islands were never observed in this regime since they will be no more detailed dislocated islands are not relevant to this paper.

In regime I, ripple-like islands (or undulations) that exhibit a broad distribution in size and in shape are observed at low misfits and low thicknesses. At increasing h, the ripple-like islands become larger and elon-

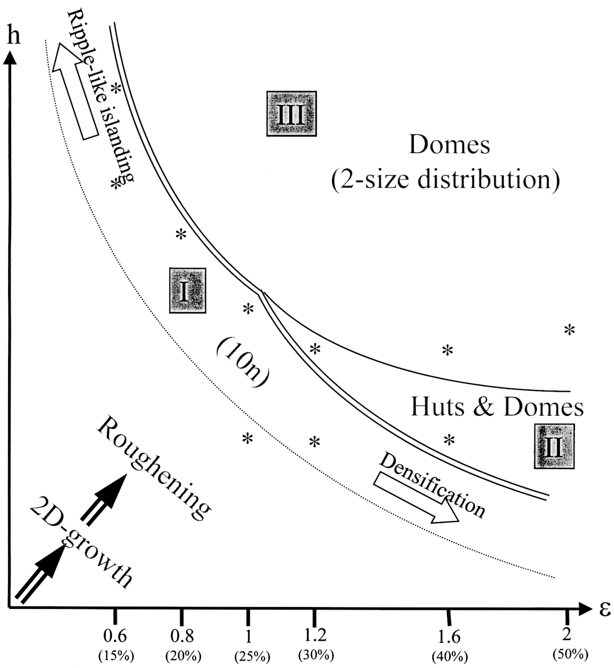

Fig. 2. Schematic 'kinetic phase diagram' representing the three main growth regimes (at constant growth temperature ~550°C). Every regime induces a typical morphology of the as-grown $Si_{1-x}Ge_x$ layers. Stars represent some of the experimental data presented in the results.

gate in chains or in square patterns oriented along [100] and [010] (Fig. 4). They present very small aspect ratios ($h/L \sim 0.03$) and side angles of approximately ~5° [instead of 11.3° for (105) facets]. No visible facet can be detected on the side of the undulations, even if small ($10n$) facets could be suspected. Previous results have shown that such undulations are fully strained [7] (no $\Delta\varepsilon_{xx}$ detectable by convergent beam electron diffraction). It was also suggested that both the onset and evolution of this roughening process are predominantly driven by kinetics.

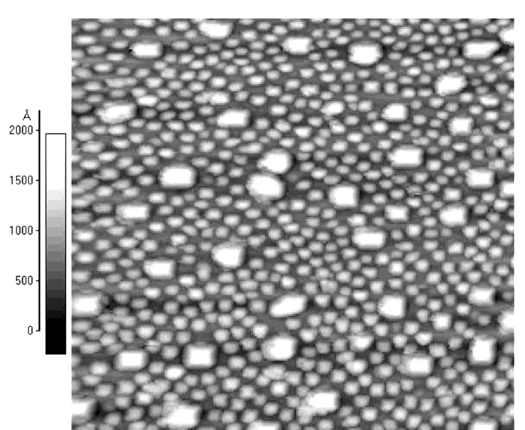

Fig. 3. AFM image of the bimodal size distribution of domes obtained in regime III for $Si_{0.5}Ge_{0.5}$ ($h = 15$ nm).

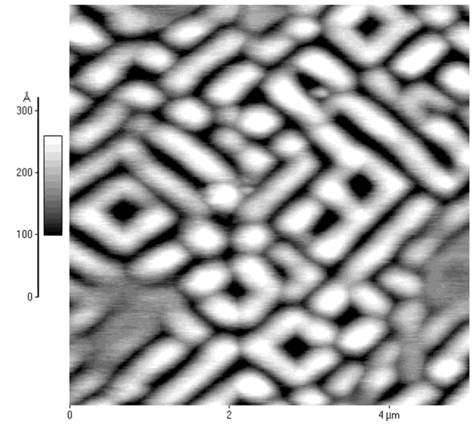

Fig. 4. AFM image of ripple-like islands elongated along ⟨100⟩ and ⟨010⟩ directions obtained in regime I for $Si_{0.85}Ge_{0.15}$ ($h = 100$ nm).

We must first remark that the increase in island shape anisotropy induced by increasing deposited thickness (in agreement with [8]) cannot yet be explained, even if alignment of islands along [100] and [010] portions of mono-atomic steps seems to be the most likely phenomenon.

At a higher misfit ($\varepsilon \sim 1.2\%$, $x \sim 0.3$), square isotropic mounds (with $L \sim 100$ nm) nucleate, superimposed on a rough surface. These islands, usually called 'huts', adopt a pyramidal shape, with the four bases oriented along ⟨100⟩ directions and the four facets approximately corresponding to (105) facets. GIXRD analysis proves the absence of elastic relaxation in such 'hut' islands [9]. At this stage, the shape evolution with the thickness and with the misfit differs. Indeed, at increasing thickness, impingement of neighbouring compact hut-islands occurs. This produces elongated hut islands, which exhibit a partial elastic-stress relaxation of $\Delta\varepsilon_{xx} \sim 0.2\%$ (Fig. 5). In contrast, at increasing misfit, new square islands nucleate between the previous ones (far from each other), until the surface is completely filled.

In order to determine the stability of the 'hut' island

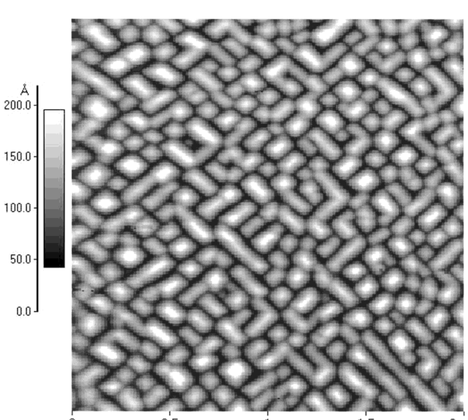

Fig. 5. AFM image of 'hut' islands elongated along ⟨100⟩ and ⟨010⟩ directions obtained in regime I for $Si_{0.7}Ge_{0.3}$ ($h = 10$ nm).

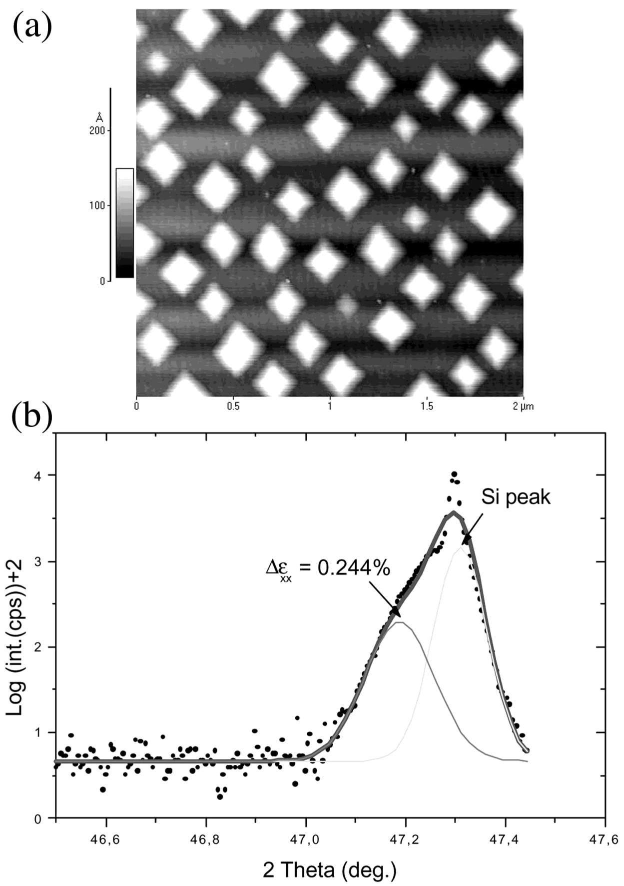

Fig. 6. (a) AFM image and (b) GIXRD spectrum of 'hut' islands stabilised during 18-h annealing at 550°C of a $Si_{0.75}Ge_{0.25}$ ($h = 10$ nm). An elastic relaxation $\Delta\varepsilon_{xx} \sim 0.25\%$ was estimated.

shape, we have followed their morphological evolution during an annealing at 550°C. Starting from a ripple-like as-grown surface, fully strained 'hut' islands form, after 1.5 h of annealing (no elastic relaxation was detected at this stage). After 18 h of annealing, islands evolve towards large isolated 'huts' that exhibit well-defined

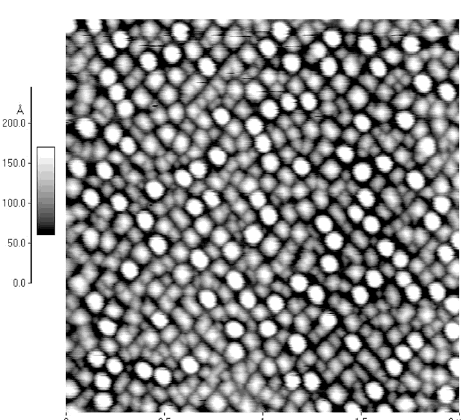

Fig. 7. AFM image of the coexistence of 'huts' and 'domes' obtained in regime II for $Si_{0.6}Ge_{0.4}$ ($h = 5$ nm).

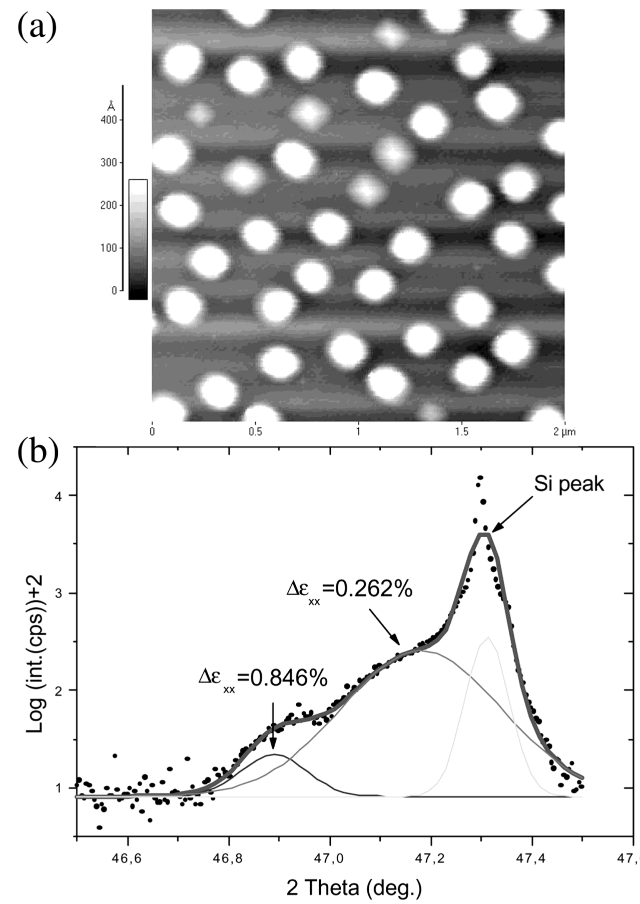

Fig. 8. (a) AFM image and (b) GIXRD spectrum of 'hut' and 'domes' stabilised during the h annealing at 550°C of a $Si_{0.6}Ge_{0.4}$ ($h = 5$ nm). Elastic relaxation of $\Delta\varepsilon_{xx} \sim 0.85\%$ and $\Delta\varepsilon_{xx} = 0.25\%$ were estimated for 'domes' and 'huts' respectively.

(105) facets (Fig. 6a). The elastic stress relaxation measured ($\Delta\varepsilon_{xx} \sim 0.25\%$) is attributed to the pyramidal 'hut' shape (Fig. 6b), even if small $\Delta\varepsilon_{xx}$ variations are detected as a function of the initial stress. No further evolution of the morphology and of the relaxation occurs during the following 46 h of annealing.

This proves that 'huts' represent an equilibrium steady state, probably stabilised by the residual stress in the islands. Similar results were reported in [10]. We suggest that (105) facets could participate to the equilibrium shape of $Si_{1-x}Ge_x$ under stress.

In regime II, co-existence of 'huts' and of 'domes' is observed (see, for instance, Fig. 7 at $h = 5$ nm and $\varepsilon = 1.6\%$). The two islands groups are characterised by different aspect ratios (~ 0.15 and ~ 0.04 for 'domes' and 'huts', respectively) and different shapes: 'domes' mainly present a round shape with facets approximately corresponding to {113} planes. Several other side orientations were found at the dome surface, but they could not be accurately determined by AFM. The respective proportion of each group of islands varies with the experimental conditions: 'domes' are favoured in the higher stress regime, while 'huts' are favoured in the lower stress regime. A considerably higher level of elastic relaxation was measured in 'domes'. For instance, the as-grown $Si_{1-x}Ge_x$ layers with $h=5$ nm and $\varepsilon=2.1\%$ presented a $\Delta\varepsilon_{xx}\sim 0.78\%$.

The evolution with annealing (at 550°C over 18 h) of the as-grown square islands obtained at $\varepsilon=1.6\%$ and $h=5$ nm also shows a 'hut'/'dome' island shape-transition, which is again accompanied by an increased level of relaxation ($\Delta\varepsilon_{xx}\sim 0.85\%$ for domes) (Fig. 8). When the transformation is complete, the highly faceted domes are randomly distributed on the flat surface. This suggests that the 'hut'/'dome' transition is mainly driven by stress relaxation. At high misfit, (105) facets are destabilised by the nearly total elastic stress relaxation. This leads to the onset of the (113) and (111) facets formation, that are present in the equilibrium shape of bulk Si (without stress).

4. Conclusion

The detailed morphological evolution of islands obtained in three distinct growth regimes was examined. In regime III, a bimodal size distribution of domes, elastically and plastically (dislocated) relaxed was observed. In regime I, flat undulations were observed, which evolved towards ripple-like islands and densely packed (105)-facetted 'huts', with increasing h and ε, respectively. The equilibrium steady state consists of large faceted 'hut' islands elastically relaxed ($\Delta\varepsilon_{xx}\sim$ 0.25%). In regime II, a bimodal size distribution of islands consisting of coherent 'huts' and 'domes' was observed. The transition from 'huts' to 'domes' is attributed to the different range of elastic relaxation in every group of islands (approximately $\Delta\varepsilon_{xx}\sim 0.2\%$ and $\sim 0.8\%$, respectively). This suggests that the pyramidal 'hut' shape [and the (105) facets] is stabilised by stress.

Acknowledgements

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