Hug-like island growth of Ge on strained vicinal Si(111) surfaces

L. Persichetti, R. Menditto, A. Sgarlata, M. Fanfoni and A. Balzarotti

Dipartimento di Fisica, Università di Roma “Tor Vergata”,
Via della Ricerca Scientifica, I-00133 Roma, Italy.

We examine the structure and the evolution of Ge islands epitaxially grown on vicinal Si(111) surfaces by scanning tunneling microscopy. Contrary to what is observed on the singular surface, three-dimensional Ge nanoislands form directly through the elastic relaxation of step-edge protrusions during the unstable step-flow growth. As the substrate misorientation is increased, the islands undergo a shape transformation which is driven by surface energy minimization and controlled by the miscut angle. Using finite element simulations, we show that the dynamics of islanding observed in the experiment results from the anisotropy of the strain relaxation.

PACS numbers: 68.37.Ef; 62.23.Eg; 68.35.bg; 81.40.Jg
The formation of three-dimensional (3D) nanostructures in the Straks-Krastanov growth of group IV semiconductors is one of the fascinating and complex phenomena related to heteroepitaxy [1–4]. Among the low-index surfaces of Si, the epitaxy of Ge on the Si(111) exhibits a relatively simple behavior consistent with classical nucleation theory. Strain-relieving 3D islands nucleate from fluctuations in the supersaturated wetting layer (WL) and grow as truncated tetrahedra with \{111\} and \{113\} facets [2, 5]. Within the time resolution of scanning tunneling microscopy (STM), Ge nucleation is almost an instantaneous and homogeneous process on the singular Si(111) surface, and only slightly correlated with surface steps. To date, however, only a few experimental results are available regarding highly-stepped vicinal surfaces of Si(111) [6, 7].

In this paper, we show that even a small misorientation of the substrate from the (111) plane affects dramatically the growth dynamics of Ge relative to the flat surface case. The highly anisotropic strain relaxation of Ge triggers the formation of 3D structures directly from step-edge nanoprotusions during the unstable step-flow growth. Snapshots of the islands growth, obtained from STM measurements, reveal an unconventional process in which the island’s shape mimics that of the elastic strain tensor as modeled by finite element (FE) simulations.

Experiments were carried out in an ultrahigh vacuum chamber (p<3x10\(^{-11}\) torr) equipped with a STM microscope. We used Si(111) wafers n-doped with P (\(\rho < 1\Omega cm\)) with nominal azimuthal angle \(\phi=0^\circ\) and polar miscut angle \(\theta\) ranging between 0° and 9.45° towards the [\(\overline{1}12\)] direction. The uncertainty on the offcut angles different from (9.45° ± 0.05°) [(557) surface] was ±0.5°. Samples were cleaned in situ using the following thermal pathway. A flash heating of the substrate to 1523 K was followed by a ramp down to 1333 K during 30 s, a subsequent 2 s quench to 1103 K and a 15 min annealing at 1103 K with slow cool-down to room temperature [8]. To avoid electromigration effects and provide a highly regular array of steps, the d.c. heating current was parallel to the step edges along the [\(\overline{1}10\)] direction [9].

Figure 1 shows the morphological structure of the clean vicinal surfaces obtained with the treatment described above. The typical kink density \(\rho_k\) was in the range of 10\(^{-4}\) per lattice site. The substrates consist of (111) terraces which are (7x7) reconstructed and have decreasing widths as the miscut becomes higher. It is also evident in the left-hand panels of Fig. 1 that, at larger miscuts, the steps are straighter because of the increased step-step interaction [10] and stiffer due to the presence of triple layers [8, 11, 12]. From the high-
resolution STM images displayed in Fig.1, it can be seen that triple steps are dominant on the 9.45° surface [panels (e),(f)], whereas they coexist with single steps at smaller miscuts [panels (c),(d)]. The step-step correlation function of the (557) surface (not shown) gives an almost perfect order at long-range \[13\]. On the mesoscopic scale, the signature of the step mixture is the characteristic behavior of step spacings as a function of the miscut angle, reported in Fig. 3(a). This is consistent with a mixed random phase of single and triple steps, with the density of triples increasing at large miscuts (See caption of Fig. 3 for details).

On the stepped surfaces of Si(111) vicinals, Ge was deposited by physical vapor deposition at 873 K at a constant flux of 0.1 ML/min. Figures 2(a)-(f) show the morphological evolution induced by Ge deposition in the coverage range 3-5 ML in which nucleation and growth of 3D islands take place on the flat surface. During the growth, the steps initially show a characteristic wriggling which consists of elongated protrusions, originating from extended step-edges [Fig. 2(a,c,e)]. For larger depositions, these initially two-dimensional nanostructures grow across the steps in the step-down direction and become progressively
taller, ultimately acquiring a 3D character [Fig. 2(b,d,f)]. The main difference from the singular surface is that elastic relaxation promotes the transition from a merely 2D growth of step protrusions towards a 3D growth of Ge nanodots. The strain-driven nature of the growth is suggested by the occurrence of the (5x5) reconstruction on the growing (111) facets of the step-edges [inset of Fig. 2(e)], since the (5x5) reconstruction results from a significant Ge/Si intermixing. Furthermore, the (111) facet is the main surface orientation of the 3D islands growing from the propagation of the protrusions. Indeed, the islands have trapezoidal shapes with a dominant (111) facet at their top and a set of steeper lateral facets with {113} orientation, as indicated by the surface orientation map (SOM) in the inset of Fig. 3(c). Since the orientation of the (111) terraces coincides with a dominant low-energy facet [2, 5, 15], the process of island formation is driven by strain and surface energy minimization. Consequently, the protrusions propagate through the steps without disintegrating into other facets, as occurs on vicinal Si(001) substrates [16]. While advancing through the steps, the protrusions grow in height following the misorientation of the substrate. As sketched in Fig. 3(b), the smaller the terrace width, the more pronounced the height of protrusions. Since the average terrace width has a sudden drop between 0° and 1° [Fig. 3(a)], step protrusions spread across many steps and, hence, become effectively 3D. In contrast, on the singular surface, they are confined to the terrace and, thus, remain
two-dimensional. As a result, the formation of 3D Ge islands on the flat (111) surface is not coupled with step meandering but occurs via nucleation and growth on terraces among the steps [Fig. 3(d)]. Moreover, on the vicinal substrates, the 3D islands’ shape is influenced by the distinctive growth mode. Since Ge/Si islands grow from the propagation of the (111) terraces, their height-to-width ratio $r$ is set by the average surface misorientation $\tan(\theta) \approx \theta$ [Fig. 3(b)] [17]. Therefore, Ge/Si islands undergo a shape transformation which is ruled by the preferential (111) faceting and controlled by the miscut angle.

Interestingly, STM images recorded at intermediate stages of growth show that the growth mechanism of 3D islands follows a very peculiar pathway quite different from that occurring on vicinal Si(001) surfaces [18]. The morphology of these islands indicates a highly anisotropic growth which is faster along the rims of the islands [Fig. 4(a-h)]. This growth mode can be understood as a result of the anisotropic elastic relaxation of vicinal surfaces. To this end, the equilibrium distribution of the elastic strain has been obtained, within the continuum elasticity theory, from FE calculations applied to the geometry of the grown islands. The equilibrium strain field within both the island and the substrate is determined by solving the 3D constitutive equations of elasticity for an elastic body under misfit strain [18]. The results of such simulations are displayed in Fig. 4(i). Due to the misoriented substrate, the in-plane-strain maps are spatially nonuniform: the relaxation of the mismatch
strain ($\varepsilon_0 = -4\%$) is higher at the periphery of the islands than in the interior part. Due to the anisotropy of elastic relaxation, growth is promoted along the rims by the effective strain relief, whereas it is hindered in the highly-strained region in the centre of the islands. Therefore, the inward growth of the islands, outlined on the right-hand side of panel (i), effectively minimizes the strain energy. Judging from the agreement between experiment and simulation, strain minimization is likely to be the main driving force for the dynamic of islanding observed on vicinal Si(111) surfaces.

In summary, we have demonstrated that that three-dimensional islanding on vicinal Si(111) substrates occurs directly through the elastic relaxation of step-edge protrusions during unstable step-flow growth of Ge. By simulating the growth process with continuum elastic theory implemented in a finite element framework, we have shown that the unconventional shape evolution of Ge dots is a consequence of the peculiar strain field which takes place in vicinal surfaces. This shape transformation is driven by strain energy minimization and controlled by the miscut angle. This study contributes to a better understanding of the role of elastic strain field in heteroepitaxy and offers insights into the potential role of substrate vicinality for controlling the growth of strained epitaxial nanostructures.

This work was supported in part by the Queensland Government (Australia) through the NIRAP project "Solar Powered Nanosensors".
References

[1] J. Stangl, V. Holý, and G. Bauer, Rev. Mod. Phys. 76, 725 (2004).
[2] B. Voigtländer, Surf. Sci. Rep. 43, 127 (2001).
[3] I. Berbezier and A. Ronda, Surf. Sci. Rep. 64, 47 (2009).
[4] F. Ratto and F. Rosei, Mater. Sci. Eng. R 70, 243 (2010).
[5] G. Capellini, N. Motta, A. Sgarlata, and R. Calarco, Sol. State Comm. 112, 145 (1999).
[6] I. Berbezier, B. Gallas, L. Lapena, J. Fernandez, J. Derrien, and B. Joyce J. Vac. Sci. Technol. B. 16, 1582 (1998).
[7] Z. Xu, Y. Zhang, R. L. Headrick, H. Zhou, L. Zhou, and T. Fukamachi, Phys. Rev. B 75, 233310 (2007).
[8] A. Kirakosian, R. Bennewitz, J. N. Crain, Th. Fauster, J. -L. Lin, D. Y. Petrovykh, and F. J. Himpsel, Appl. Phys. Lett. 79, 1608 (2001).
[9] S. Yoshida, T. Sekiguchi, and K. M. Itoh, Appl. Phys. Lett. 87, 031903 (2005).
[10] X. -S. Wang, J. L. Goldberg, N. C. Bartelt, T. L. Einstein, and E. D. Williams, Phys. Rev. Lett. 65, 2430 (1990).
[11] J. Wei, X. S. Wang, J. L. Goldberg, N. C. Bartelt, and E. D. Williams, Phys. Rev. Lett. 68, 3885 (1992).
[12] M. K. Kim, D. H. Oh, J. Baik, C. Jeon, I. Song, J. H. Nam, S. H. Woo, C. Y. Park, and J. R. Ahn, Phys. Rev. B 81, 085312 (2010).
[13] J. L. -Lin, D. Y. Petrovykh, J. Viernow, F. K. Men, D. J. Seo, and F. J. Himpsel, J. Appl. Phys. 84, 255 (1998).
[14] The position of each spot represents the local normal orientation relative to the (111) plane, while the intensity gives the relative amount of surface having this orientation.
[15] P. Müller and R. Kern, Surf. Sci. 457, 229 (2000).
[16] H. Lichtenberger, M. Mühlberger, and F. Schäffler, Appl. Phys. Lett. 86, 131919 (2005).
[17] From simple geometric arguments, it follows that the ratio between the island height and the projection of the dominant (111) facet on the vicinal plane is given by \( \tan(\theta) \). The aspect ratio
Figure Captions

Fig. 1: (color online). STM images of clean vicinal Si(111) surfaces. (a,b) 1.5°-miscut surface. (c,d) 5°-miscut surface. In panel (d) the coexistence of single- and triple-height steps is highlighted. (e,f) 9.5°-miscut surface. In panel (f) triple steps are evidenced. The arrows indicate the \([\overline{1}1\overline{2}]\) direction.

Fig. 2: (color online). STM images: (a,b) 1.5°-miscut surface after deposition of (a) 3.6 ML and (b) 4.0 ML of Ge. (c,d) 5°-miscut surface after deposition of (c) 3.9 ML and (d) 5.0 ML of Ge. (e,f) 9.5°-miscut surface after deposition of (e) 3.8 ML and (f) 5.0 ML of Ge. The arrows indicate the \([\overline{1}1\overline{2}]\) direction.

Fig. 3: (color online). (a) Measured average-terrace widths of vicinal Si(111) surfaces. The continuous line represents the expected terrace-width dependence for a mixture of single- and triple-height steps given by \([n_s(\theta) + 3n_t(\theta)(\tan\theta)^{-1}h]\), where \(h = 0.31\) nm is the height of a single step and \(n_s, n_t\) are the density of single- and triple-height steps, taken from Ref. \([11]\). Triple steps (shown by the STM image in the inset) increase the average step-separation compared to a pure single-height phase (dashed curve). (b) Island aspect ratio as a function of miscut angle. The dashed line is the average surface misorientation \(\tan(\theta)\). (c) STM image of a Ge islands on the 5°-miscut Si(111) surface after deposition of 4.5 ML of Ge. In the inset, the corresponding SOM is displayed. (d) STM image of a Ge islands on the singular Si(111) surface after deposition of 4.0 ML of Ge. The arrows indicate the \([\overline{1}1\overline{2}]\) direction.

Fig. 4: (color online). (a-h) STM images of different stages of Ge island formation on the 1.5°-miscut Si(111) surface. The \([\overline{1}1\overline{2}]\) direction is indicated by arrows. (i) FE simulations of the in-plane strain tensor \(\varepsilon\) for 3D models of Ge islands based on the experimental geometry extracted from STM images. The white arrows indicate the direction of the island growth observed in the experiment.