PHYSICAL REVIEW B, VOLUME 64, 201320(R)

Surface stress-induced island shape transition in Si(001) homoepitaxy

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(Received 6 September 2001; published 5 November 2001)

A low-energy electron microscopy study of two-dimensional Si(001) island shapes near thermal equilibrium on 10×15 μm^2 large single-domain terraces reveals a continuous increase of island aspect ratio and a shape transition from elliptical to ''American-football''-like with increasing island size. The size-dependent island shapes are driven by elastic relaxation caused by the intrinsic surface stress anisotropy present on Si(001). Analysis of the measured elliptical island shapes based on an elastic-model calculation allows a quantitative determination of step energies and of the surface stress anisotropy as a function of temperature.

DOI: 10.1103/PhysRevB.64.201320 PACS number(s): 68.55.Jk

Surface stress is an intrinsic property of a solid surface. It plays an important role not only in determining the static structure and morphology of a surface, but also in controlling the evolution of surface structure and morphology during epitaxial growth. Surface stress is inevitably connected with surface reconstruction, because tilting or stretching of atomic bonds is necessary to reconstruct the surface. For example, the formation of dimer rows on Si(001) causes a large anisotropy in the surface stress tensor, which in turn gives rise to a ground-state stress domain structure consisting of equally populated 1×2 and 2×1 domains.[1,2] Such an interplay between surface stress and structure can lead to a wide range of interesting surface phenomena.[3] Si(001) serves also as the model system for investigations of stress-induced self-organization for fabricating SiGe nanostructures. Consequently, the effects of surface stress in Si(001) have attracted much recent interest.

Men *et al.* first demonstrated that applying a uniaxial stress to the Si(001) surface may drive step movement, favoring one domain over the other.[1] Wavy steps and hilly structures observed on a micrometer scale on highly oriented Si(001) wafers by low-energy electron microscopy (LEEM) have been interpreted as stress-domain structures.[4] However, surface stress has generally been neglected in interpreting thermodynamic properties in two-dimensional (2D, i.e., submonolayer) island growth.

The equilibrium shapes of 2D islands have generally been related to step free energies; consequently, they are temperature dependent but *size independent*.[5–9] Here, we provide direct evidence that the equilibrium shape of 2D Si islands on Si(001) is *size dependent*. The island originally adopts an elliptical equilibrium shape, whose aspect ratio increases continuously with increasing island size, and then transforms into an ''American-football''-like shape beyond a critical size. Both the size-dependent aspect ratio and the elliptical-to-football shape transition are driven by surface stress relaxation. The changing equilibrium aspect ratio of an elliptical island with increasing size but formed at a fixed temperature can be fitted nicely by a recent theoretical model,[10] allowing us to extract quantitative values of not only step free energies but also surface stress anisotropy at different temperatures. Our results demonstrate that caution should be used when deriving step free energies from equilibrium 2D island shapes: The possible influence of stress (if misfit stress or intrinsic surface stress anisotropy is present) must be considered. We show that the intrinsic surface stress anisotropy of Si(001) is temperature dependent, decreasing with increasing temperature from a value of 80 ± 3 meV/Å at 695 °C to 68 ± 4 meV/Å at 855 °C.

Using a LEEM equipped with a Si_2H_6 gas source for Si deposition, we investigate the shape of single 2D epitaxial islands near thermal equilibrium on 10×15 μm^2 large single-domain Si(001) terraces. We first pattern the Si(001) wafers via photolithography, creating mesa structures 20 μm wide and 10 nm high on the surface. After removing the native oxide *in situ*, we deposit Si at step flow conditions (temperature 870 °C) to produce large step-free terraces on top of the mesas.[11] LEEM easily shows when a terrace has no steps. After a step-free terrace forms, we adjust the temperature and Si_2H_6 pressure so that only a single island nucleates preferentially near the middle of the terrace. This island then grows slowly in the LEEM field of view, corresponding to a growth rate of less than 1 monolayer in 10 minutes. During deposition, the base pressure was kept below 3×10^{-8} Pa and the disilane pressure below 5×10^{-6} Pa, to prevent possible sample contamination. Samples were heated by electron bombardment from the rear and the temperature was measured using an optical pyrometer, calibrated via the Si melting point. Data are collected at several different sample temperatures for a fixed very low deposition rate, ensuring that the conditions are as close to equilibrium as possible.

Using a large terrace with a single island nucleating in the middle of the terrace far from the terrace edges not only allows us to observe islands that grow up to several micrometers in diameter, but also allows us to exclude possible stress-induced island–island and island–step (terrace edge) interactions that can influence island shapes. Previous studies of Si island growth on Si(001) have either employed low-temperature molecular beam epitaxy in which island shapes were affected by slow kinetics[5,6] or limited the islands to less than 100 nm in diameter before they interacted with terrace edges or neighboring islands.[7,12] Consequently, these early experiments are inappropriate for the analysis we perform below.

Figure 1 displays different stages of a single island growing slowly on a rectangular 10×15 μm^2 large mesa terrace

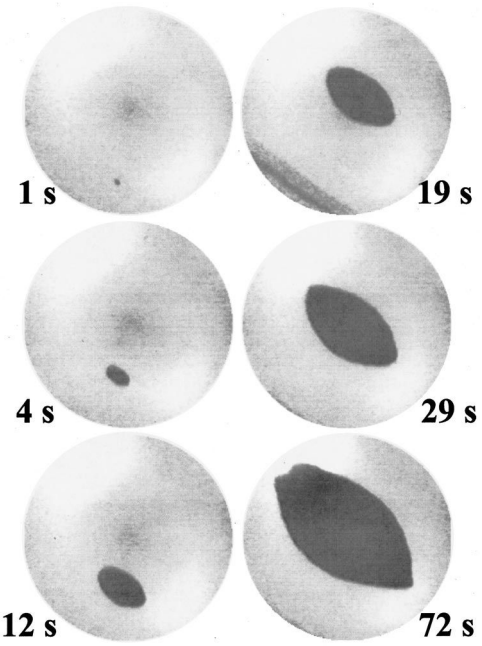

FIG. 1. Evolution of 2D Si island size and shape on an extremely large (10×15 μm^2) single-domain Si(001) terrace during very slow, near-equilibrium, chemical beam epitaxy of Si at 855 °C. The time after observing island nucleation is given in seconds. The island shape evolves with increasing island size, from initially elliptical to ''American-football''-like and eventually with 2D faceting (swallow tail at $t=72$ s) for island diameters larger than 6 μm. The field of view is 9 μm. The frame at $t=19$ s shows part of one long mesa edge (gray area at lower left); the base terrace extends beyond the field of view in all other frames. Inhomogeneities in the image (bright area at the corner of the terrace) are due to imperfect focus and inhomogeneities in the channel plate.

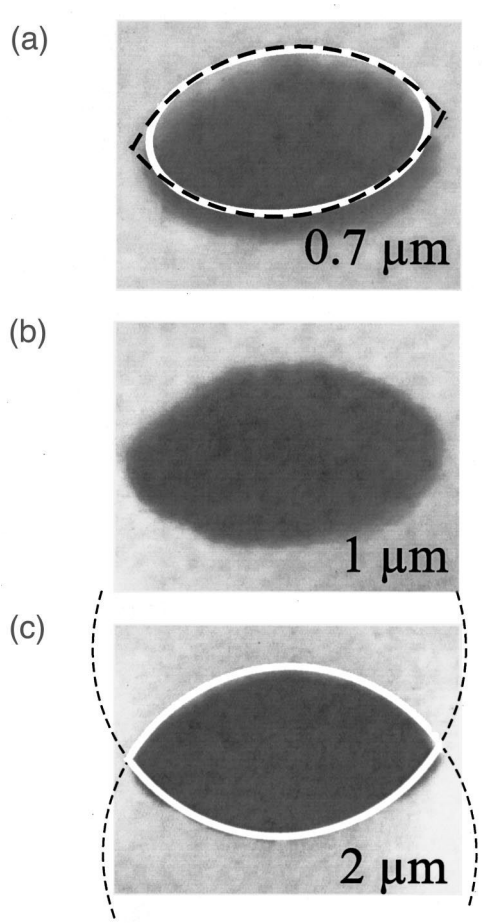

FIG. 2. Island shape transition with island size depicted for three stages: (a) elliptical shape, (b) intermediate shape, (c) football shape. Images are scaled to compensate for island size. The elliptical outline enclosing the island in (a) is constructed using the elliptical function. The football outlines, depicted as dashed lines in (a) and (b), are constructed as an intersection of two circles.

at 855 °C in low resolution (field of view is 9 μm). For each frame the time t after island nucleation is given in seconds. The island appears as black because the LEEM images are taken in dark-field mode, using a diffraction spot of the 2×1 superstructure of the base terrace (white area). The island shape evolves in several different stages. At $t=1$ s only a black dot is visible at the lower corner of the image. The island shape appears as elliptical at $t=4$ s and $t=12$ s. Sharp tips develop at the far ends of the island at $t=19$ s, leading to an ''American-football''-like shape at $t=29$ s. At $t=72$ s, one sharp tip splits into two rounded corners.

To illustrate the island shape evolution, Fig. 2 shows images scaled to compensate for island size. An island size of 1.4 μm [major axis, see panel (a)] has an elliptical shape, as indicated by the perfect elliptical fit, the white contour enclosing the island. When the island size becomes three times larger [panel (c)], it is better fit by the intersection of two circles (football shape). Panel (b) shows an intermediate state between the elliptical-to-football shape transition.

In general, the shape of 2D islands reflects the dependence of step free energy on step orientation, just as the shape of isolated 3D crystallites reflects the dependence of surface free energy on facet orientation.[13] For Si islands on Si(001), the anisotropic shape has conventionally been attributed solely to the different step energies of two orthogonal orientations,[5–8] namely S_A and S_B steps.[14] However, step energies are island size independent, which would lead to a size-independent island aspect ratio. Therefore, the size dependence of the island shape must originate from an additional free-energy contribution. The stress relaxation energy provides the appropriate behavior.[10]

To analyze island shape evolution quantitatively, we determine the island shape aspect ratios and island areas using image analysis software. Both low- and high-resolution images have been used to capture island sizes over a wide range from 0.1 to 3.0 μm in diameter. Figure 3 shows the island aspect ratio vs island area at two sample temperatures, 695 °C and 855 °C, respectively, with islands remaining in a perfect elliptical shape up to 1.5 μm^2. Both sets of experimental data have been fitted, using a theoretical model[10] that includes stress relaxation energy in addition to step free energy in determining the equilibrium shape of islands as a function of island size.

As a 2D island forms on Si(001), the dimer rows in the island are rotated by 90° with respect to the substrate dimer rows. Consequently, the anisotropic surface stress introduces

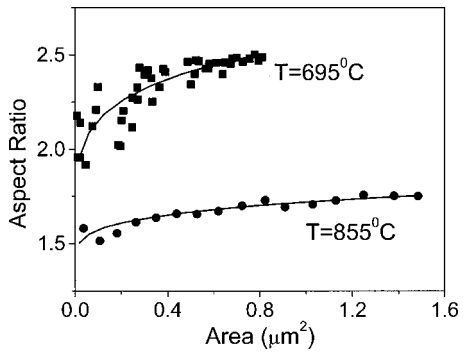

FIG. 3. Dependence of 2D island shape aspect ratio (major to minor axis) of a single island grown on an extremely large single-domain terrace. Experimental data at temperatures of 855 °C and 695 °C are given as circles and squares, respectively. The solid lines are fits to the data.

stress discontinuities, i.e., force monopoles along the island step edges. The total free energy of an elliptical island can be calculated as

$$F_{total}=F_{step}+F_{stress}=\int_0^{2\pi}F_s(\theta)r(\theta)d\theta + \frac{1}{2}\int\int \mathbf{u}[\mathbf{r}_1,\mathbf{F}(\mathbf{r}_2)]\cdot\mathbf{F}(\mathbf{r}_1)d\mathbf{r}_1 d\mathbf{r}_2, \qquad (1)$$

where $F_s(\theta)$ is the step free energy at the angle θ in reference to the S_A step orientation, $r(\theta)$ is the radius, and $\mathbf{u}[\mathbf{r}_1,\mathbf{F}(\mathbf{r}_2)]$ is the displacement at point $\mathbf{r}_1$ induced by the force $\mathbf{F}$ at point $\mathbf{r}_2$. The step free energy, $F_s(\theta)$ can be calculated from the step energies (E_A and E_B) and corner energy (E_c) at zero temperature, $F_s(\theta)=F_A\cos\theta+F_B\sin\theta+F_c\sin 2\theta$, with $F_{A,B}=E_{A,B}-TS_{A,B}$ and $F_C=E_C-kT\ln 2$. T is the temperature and k is the Boltzmann constant. $S_{A,B}$, the step entropy at T, can be calculated from E_A, E_B, and E_C, using a solid-on-solid model.[15] The stress relaxation energy, F_{stress}, however, has to be integrated numerically, because it cannot be solved analytically for an elliptical shape.

We use a trial-and-error fitting procedure. For each set of trial values of E_A, E_B, E_c, and surface stress anisotropy, $\Delta\sigma$, the optimal island aspect ratio for a given island size is determined by minimizing the island total energy, in which the step free energies are calculated analytically while the stress energies are calculated numerically. The calculated curve of aspect ratio vs island size (area) is in turn compared to the experimental data. The trial values of E_A, E_B, E_c, and $\Delta\sigma$ are then adjusted until the theoretical curves converge to the experimental data with the smallest deviation, i.e., the sum of the differences between the two at each experimental data point. Although there are four parameters, the fitting is in fact quite robust and step energies and stress anisotropies are fit independently to different physical characteristics of the data. The average vertical positions of the data (i.e., the aspect ratio) in Fig. 3 are controlled by the step free-energy ratios. By fitting simultaneously to the two sets of data at different temperatures, the individual step energies (E_A and E_B) can be determined. The curvature of the data in Fig. 3 (i.e., the dependence of aspect ratio on island size) is controlled by the ratio of step energy and strain energy, and hence by the stress anisotropy for the given step energies. Hence, the stress anisotropies at different temperatures are determined by fitting separately to the obviously different curvatures of the two sets of data.

Our calculations following this scheme yield step energies $E_A=25\pm3$ meV, $E_B=53\pm6$ meV, and $E_C=56\pm8$ meV at zero temperature, and surface stress anisotropies, $\Delta\sigma=68\pm3$ meV/Å at $T=855$ °C and $\Delta\sigma=80\pm4$ meV/Å at $T=695$ °C. The error bars are estimated by varying the trial parameters to the largest extent possible for the calculated curves to remain within the upper and lower bounds of the experimental data. The step energies agree very well with previous experimental values determined from measurements of step meandering or fluctuations,[7,15] in which the effect of surface stress is expected to be less significant. The surface stress anisotropy increases with decreasing temperature, possibly because of the different dynamic buckling states of surface dimers at different temperatures. First-principles calculations show that the surface stress anisotropy depends strongly on dimer buckling.[16] The quantitative values of stress anisotropy we obtain also agree well with previous experiments in a similar temperature range.[17]

We have used growth conditions of very low supersaturation to ensure the island shapes are as close to equilibrium as possible, as suggested by previous studies.[6,7] By studying the Si island growth shape on Si(001) as a function of deposition rate and temperature, Swiech and Bauer[6] have suggested that equilibrium island shapes can be obtained at a growth rate of 0.2 ML/min at 530 °C. Our growth rate is twice slower (0.1 ML/min) and the temperature is several hundred degrees higher. Several observations also support the conclusion that the islands in our experiments are close to equilibrium:

(1) On Si(001), anisotropic adatom sticking coefficients to island edges can lead to anisotropic island growth shapes.[5] Such kinetic effects elongate the islands along the dimer rows.[5] Thermodynamically, the islands are also elongated along the dimer-row direction because of the lower S_A step energy than S_B.[5] Therefore, a kinetics-influenced island shape in Si(001) always has a larger aspect ratio than the equilibrium shape. In contrast, the measured island aspect ratio in Fig. 3 is smaller than the step free-energy ratio in a certain range of island sizes, which is explained perfectly by the equilibrium stress model.[10]

(2) We have done both growth and evaporation (at >900 °C) measurements on the same island, and the dependence of island shape on island size in both cases is the same.

(3) The excellent and robust fit (based on an equilibrium theory) to the experimental data and the very good agreement between the fitting parameters and the previous results from independent measurements further support quantitatively the conclusion of equilibrium.

(4) The elliptical-to-football shape transition can be explained by the equilibrium theory (Wulff construction), as discussed below.

The aspect ratio of elliptical islands increases continuously with increasing island size. The transition from elliptical to football shape is then a natural extension of this size dependence. The equilibrium island shape can be constructed from the step free energies, using the Wulff construction.[6,13] If the island anisotropy gets so large that parts of the free-energy diagram no longer contribute to the equilibrium shape, then tip development, i.e., an elliptical-to-football shape transition, will occur.[6,13] The tips, associated with the forbidden angles in the equilibrium shape, are energetically metastable and will then split, decomposing into stable (2D) facets of angle-allowed orientation,[13] as shown in Fig. 1. (To the best of our knowledge, no kinetic effect has been shown to induce such tip development.) Conceptually, one could combine the stress energy into ''effective'' step energies[18] that would be island-size-dependent. Based on our results, the ratio of ''effective'' step energies would then increase with increasing island size, leading to increasing island aspect ratio and eventually to the elliptical-to-football transition. It is possible to derive the ''effective'' step energies from the measured island shapes, using the inverse Wulff construction.[6,13] However, the stress energy contained in the ''effective'' step energies can only be determined separately after knowing the island shape as a function of island size at constant temperature, as we have done. There is no mechanism to decompose stress energy into individual step energies of different orientations.

Recently, Hannon *et al.*[9] have observed an elliptical-to-football shape transition of Si islands on highly boron-doped Si(001) as the temperature decreases. They attributed the transition to the anomalous temperature dependence of step free energies caused by boron segregation to steps in addition to entropy. In contrast, we propose a distinctively different physical mechanism. We observe the elliptical-to-football shape transition under the physical conditions of *changing island size at a fixed temperature*. Because step energies do not change at a fixed temperature, the driving force for our island size-induced transition must come from a different source, namely the strain energy, which does change with size. A stress-modified step energy has also been suggested to explain step morphologies on Si(001),[18] supporting our model.

In conclusion, we have demonstrated the influence of surface stress on island shape in Si homoepitaxy, an influence that has not been recognized earlier but seems obvious in retrospect. In addition to the step free energies, the elastic relaxation energy induced by the intrinsic surface stress anisotropy contributes significantly to defining the equilibrium island shape. Because the elastic relaxation energy is island size dependent (as is physically quite reasonable), the island aspect ratio continuously increases with increasing island size, leading eventually to an elliptical-to-football shape transition. By fitting the experimental data with theory, we derive quantitative values of (temperature-independent) step energies and of the intrinsic surface stress anisotropy of Si(001) as a function of temperature. We show that the intrinsic surface stress anisotropy of Si(001) decreases with increasing temperature. The analysis presented here should be generally applicable to quantitative studies of island shapes in those systems in which misfit strain or surface stress anisotropy is present.

This work was supported by NSF, Grants No. DMR-9632527 and DMR-9304912, DOE, Grant No. DE-FG02-00ER45816, and the Alexander von Humboldt Foundation.

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[1] F.K. Men, W.E. Packard, and M.B. Webb, Phys. Rev. Lett. **61**, 2469 (1988).

[2] O.L. Alerhand, D. Vanderbilt, R.D. Meade, and J.D. Joannopoulos, Phys. Rev. Lett. **61**, 1973 (1988).

[3] Feng Liu and M.G. Lagally, Phys. Rev. Lett. **76**, 3156 (1996).

[4] J. Tersoff and E. Pehlke, Phys. Rev. Lett. **68**, 816 (1992); R.M. Tromp and M.C. Reuter, *ibid.* **68**, 820 (1992); R.M. Tromp and M.C. Reuter, Phys. Rev. B **47**, 7598 (1993).

[5] Y.W. Mo, J. Kleiner, M.B. Webb, and M.G. Lagally, Phys. Rev. Lett. **73**, 553 (1991).

[6] W. Swiech and E. Bauer, Surf. Sci. **255**, 219 (1991).

[7] N.C. Bartelt, R.M. Tromp, and E.D. Williams, Phys. Rev. Lett. **73**, 1656 (1994).

[8] A. Pimpinelli and J.J. Metois, Phys. Rev. Lett. **72**, 3566 (1994).

[9] J.B. Hannon, N.C. Bartelt, B.S. Swartzentruber, J.C. Hamilton, and G.L. Kellogg, Phys. Rev. Lett. **79**, 4226 (1997).

[10] Adam Li, Feng Liu, and M.G. Lagally, Phys. Rev. Lett. **85**, 1922 (2000).

[11] S. Tanaka, C.C. Umbach, J.M. Blakely, R.M. Tromp, and M. Mankos, Appl. Phys. Lett. **69**, 1235 (1996). In this work, large terraces on pre-patterned Si(001) surfaces were created by sublimation. The process is similar for growth, J. B. Maxson, Ph.D. Thesis, 2001; V. Zielasek, J. B. Maxson, and P. Sutter (to be published).

[12] W. Theis, N.C. Bartelt, and R.M. Tromp, Phys. Rev. Lett. **75**, 3328 (1995); N.C. Bartelt, W. Theis, and R.M. Tromp, Phys. Rev. B **54**, 11 741 (1996).

[13] For a review see M. Wortis, in *Chemistry and Physics of Solid Surfaces*, edited by R. Vanselow and R. Howe (Springer, Berlin, 1989), Vol. 7, p. 367.

[14] J.D. Chadi, Phys. Rev. Lett. **59**, 1691 (1987).

[15] B.S. Swartzentruber, Y.W. Mo, R. Kariotis, M.G. Lagally, and M.B. Webb, Phys. Rev. Lett. **65**, 1913 (1990); H.J.W. Zandvliet, H.B. Elswijk, E.J. van Leonen, and D. Dijkkamp, Phys. Rev. B **45**, 5965 (1992).

[16] A. Garcia and J.E. Northrup, Phys. Rev. B **48**, 17 350 (1993); J. Dabrowski, E. Pehlke, and M. Scheffler, *ibid.* **49**, 4790 (1994).

[17] M.B. Webb, F.K. Men, B.S. Swartzentruber, R. Kariotis, and M.G. Lagally, Surf. Sci. **242**, 23 (1991).

[18] J.P. Pelz, C. Ebner, D.E. Jones, Y. Hong, E. Bauer, and I.S.T. Tsong, Phys. Rev. Lett. **81,** 5473 (1998).