

## Strain induced stabilization of stepped Si and Ge surfaces near (001)

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We report on calculations of the formation energies of several [100] and [110] oriented step structures on biaxially stressed Si and Ge (001) surfaces. It is shown that a novel rebonded [100] oriented single-height step is strongly stabilized by compressive strain compared to most well-known step structures. We propose that the side walls of “hut”-shaped quantum dots observed in recent experiments on SiGe/Si films are made up of these steps. Our calculations provide an explanation for the nucleationless growth of shallow mounds, with steps along the [100] and [110] directions in low- and high-misfit films, respectively, and for the stability of the (105) facets under compressive strain. © 2002 American Institute of Physics. [DOI: 10.1063/1.1491611]

Strain induced self-assembly is actively being pursued as a technique for the fabrication of nanoscale electronic devices and memories that have the potential to bring higher speed to information processing and higher areal and volumetric capacity to information storage. In the past few years, significant advances have been made on both the technological and fundamental aspects of self assembly. On the technological side, it has become possible to prepare regular spatial arrays of nanostructures.

Several basic issues concerning the physical mechanisms involved in the different stages of the formation and morphological evolution of the nanostructures are only now becoming clear. The initial stages of epitaxial nanostructure formation remains least well understood. Of particular interest here, recent experimental work<sup>1–4</sup> has revealed that (105) oriented quantum dots in SiGe/Si films grow from shallow precursor mounds whose sidewalls are made up of widely spaced steps that are oriented in the [100] or [110] directions for low or high Ge compositions, respectively. These observations cannot be understood on the basis of a competition between surface energy increase involved in creating the walls of the dots and the strain energy reduction through elastic relaxation. Such a competition would lead to a nucleation barrier for the formation of the islands, while the experiments clearly show that the stepped mounds emerge as a natural instability without any such barrier.<sup>1,2</sup>

Recently we have demonstrated that nucleationless growth of stepped mounds can be understood by including the physics of surface steps, in particular the dependence of their formation energy on strain and their interactions.<sup>5</sup> In this letter, we calculate the parameters that characterize step formation and interactions in order to understand the stabilization of [100] or [110] steps on (2 × 1) reconstructed Si and Ge (001) surfaces at different levels of biaxial strain. The strain dependence of the step formation energy is determined by the disposition of the atomic bonds in the vicinity of the step-edge; if there is significant rebonding such that these bonds are stretched (compressed) relative to the bonds in the bulk material, a compressive (tensile) mismatch stress tends

to lower their formation energy.<sup>6</sup> The interactions between steps include the repulsive dipolar interactions,<sup>7</sup> which depend on the local step density, and the nonlocal attractive interactions due to the force monopoles induced by the mismatch strain.<sup>8</sup> The surface energy of a stepped surface that is oriented at a small angle  $\theta$  with respect to the (001) direction can be written by including the step formation and dipolar interaction energies as

$$\gamma(\theta, \epsilon) = \gamma_0(\epsilon) \cos \theta + (\beta_1 + \tilde{\beta}_1 \epsilon) |\sin \theta| + \beta_3 \frac{|\sin \theta|^3}{\cos^2 \theta}, \quad (1)$$

where  $\epsilon$  is the biaxial surface strain,  $\gamma_0$  is the surface energy of the (001) surface,  $\beta_1$  is the formation energy of a step,  $\tilde{\beta}_1$  is a measure of the sensitivity of the formation energy of a step to strain, and  $\beta_3$  characterizes the strength of the step-step dipolar interactions. Since the attractive monopole induced interactions favor a stepped surface over a flat surface, stepped mounds can grow without any nucleation barrier if the surface energy of their sidewalls becomes lower than that of the (001) surface.<sup>5</sup> It can be seen from Eq. (1) that this indeed happens for stepped surfaces with orientations in the range  $|\theta| < \theta_c$ , where  $\theta_c \approx \sqrt{-\beta(\epsilon)/\beta_3}$ , if the condition  $\beta(\epsilon) \equiv \beta_1 + \tilde{\beta}_1 \epsilon < 0$  is satisfied. In the present work we show by means of atomistic simulations that this condition is satisfied for the mismatch strains of interest.

In the [110] direction, we will only consider the double-height B-type (DB) steps;<sup>9</sup> these steps are known to be lower in energy than the combination of alternating single-height A-type (SA) and B-type (SB) steps<sup>9</sup> for vicinal angles that are more than about 1°–2°. In the [100] direction we consider two kinds of steps: (1) zigzag SA + SB<sup>[100]</sup> steps proposed by Chadi<sup>9</sup> and experimentally observed by Wu *et al.*<sup>10</sup> and (2) straight steps on which the dimer rows of the adjoining terraces arrive at the steps at an angle of 45°. If we focus attention on the family of (10*n*) surfaces to which the (105) surface belongs, there are two distinct low-energy step structures that can be obtained by removing every other atom from the step-edge in order to reduce the number of dangling bonds, as shown in Fig. 1. The key difference between the two structures is that in one of them, which we call single-height rebonded (SR), there are atoms at the edge of the step

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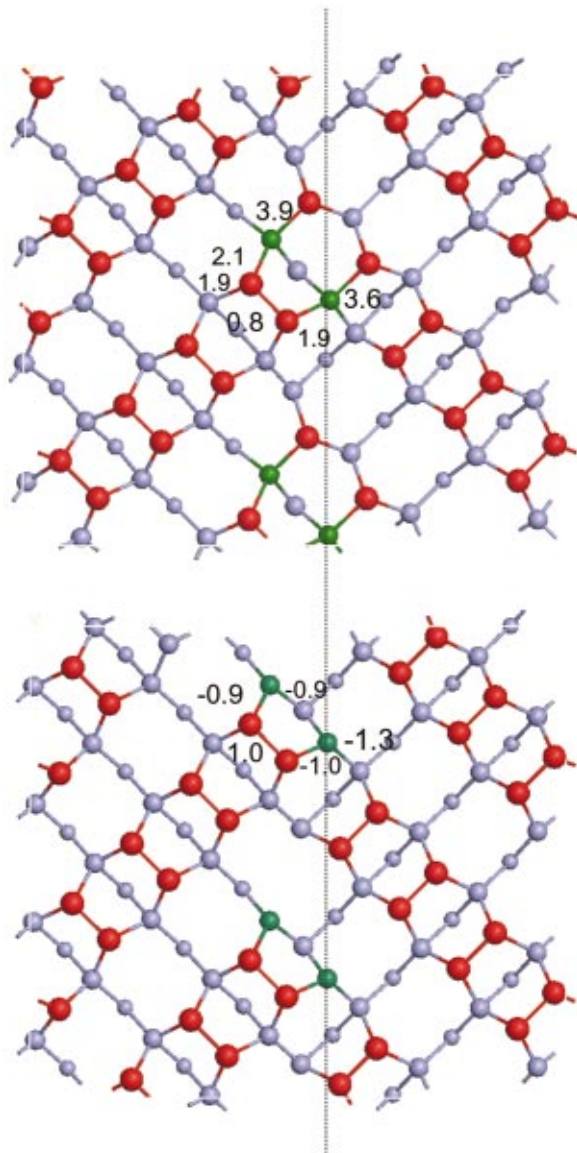


FIG. 1. (Color) The structure of the [100] oriented SR step (top) and the SU step (bottom) on a (2×1) reconstructed (001) surface. The dashed line denotes the edge of the step, where the dimer orientation undergoes a 90° rotation. Atoms are colored according to the number of dangling bonds (*b*) before surface and step-edge reconstructions: red=2*b*, green=1*b*, blue=0*b*. The numbers represent the stretching (in %) of some of the bonds relative to the bond length in the bulk for the Si(109) surface relaxed using the T3 potential. Note that the rebonding of the atoms across the step edge leads to stretching of several bonds in the SR step.

bonded to atoms on the adjoining terraces, while this type of rebonding is absent in the single-unrebonded (SU) step (refer to Fig. 1). The structure of the (105) facet made up of SU and SR steps correspond to the reconstructions for this sur-

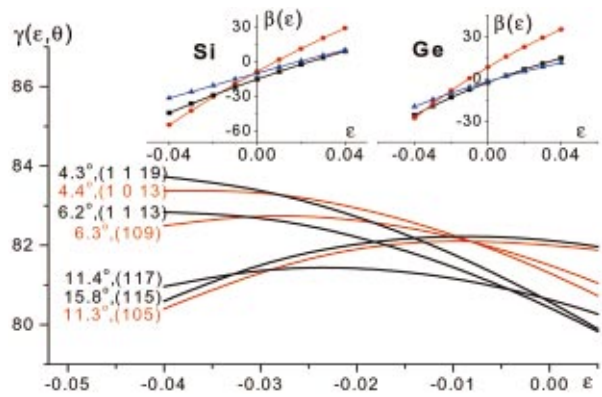


FIG. 2. (Color) Surface energy of stepped surfaces (in meV/Å<sup>2</sup>) consisting of [100]SR (red) and [110]DB (black) steps as a function of the biaxial strain  $\epsilon$  for Ge, computed with the T3 potential. The vicinal angles and surface orientations are indicated in the figure. The insets show the strain dependent formation energy  $\beta(\epsilon)$  (in meV/Å<sup>2</sup>) for three types of steps, DB(black squares), SR(red circles), and SA+SB<sup>[100]</sup> (blue triangles) in Si and Ge.

face proposed by Mo *et al.*<sup>11</sup> and Kohr and Das Sarma,<sup>12</sup> respectively.

We determine the step formation and interaction energies using the following procedure: the surface energies of various stepped surfaces are first determined as a function of applied biaxial strain for both the (10*n*) and (11*n*) families as shown in Fig. 2. The step formation and interaction energies are then extracted from the surface energy  $\gamma(\epsilon, \theta)$  using Eq. (1). The strain dependent step formation energies for various step structures calculated using the Tersoff (T3)<sup>13</sup> potential are shown in Fig. 2 for Si and Ge.<sup>14</sup> In all cases, the step formation energies can be reasonably approximated by a linear relation assumed in Eq. (1) with small deviations that can be attributed to nonlinear effects, particularly at larger strains. Calculation of step energies using the Stillinger–Weber (S–W)<sup>15</sup> potential also showed almost linear dependence of the formation energies with strain. The parameters  $\beta_1$  and  $\bar{\beta}_1$ , obtained by using a linear fit to  $\beta(\epsilon)$ , along with the dipolar interaction strength  $\beta_3$  are given in Table I.

If we focus attention on the [100] oriented steps in Si, we find that the formation energies of the SR steps are lower than those for the SU and SA+SB<sup>[100]</sup> steps for compressive mismatch strains. This can be understood by analyzing the bond lengths at the step-edge (refer to Fig. 1); since there are several stretched bonds in the SR structure, we expect its energy to be lower in compressive strains than the SU structure. Although the zigzag SA+SB<sup>[100]</sup> steps are energetically lower than the SR steps in the absence of strain, Fig. 2 shows

TABLE I. Step formation energies  $\beta_1$ , their derivatives with respect to strain  $\bar{\beta}_1$ , and the step–step dipolar interaction coefficient  $\beta_3$  for Si and Ge (with values for Ge enclosed in parentheses) calculated using the Stillinger–Weber and Tersoff (T3) potentials. In order to allow a direct comparison of the formation energies of different step structures,  $\beta_1$  is measured relative to the formation energy of the DB step calculated using the T3 potential. All quantities are expressed in meV/Å<sup>2</sup>.

	SR		DB		SU	SA+SB <sup>[100]</sup>
	S–W	T3	S–W	T3	T3	T3
$\beta_1$	19.9(29.9)	5.8(10.1)	12.1(14.8)	0.0(0.0)	46.3(38.6)	5.98(0.96)
$\bar{\beta}_1$	741(738)	1046(781)	440(459)	667(501)	−93(−89)	523(385)
$\beta_3$	169(186)	217(143)	239(189)	307(229)	2(11)	585(382)

that a modest amount of compressive strain (0.3% in Si) stabilizes the SR steps over these steps. Table I also shows that while the formation energies of the SR steps on an unstrained surface are larger than the corresponding values for the DB steps, the strain sensitivity  $\tilde{\beta}_1$  for the SR steps is about 50% larger in both Si and Ge. This implies that the SR steps are preferred over the DB steps at large values of compressive strain, with one key difference between Si and Ge. Using both the S–W and T3 potentials, we find that the formation energies of the SR steps fall below the energies for DB steps when strains become more than about 1% and about 4% in Si and Ge, respectively (refer to the insets in Fig. 2 and Table I).

The above observations have important implications for the growth of stepped islands observed in recent experiments.<sup>1–4</sup> If we assume that the results for Si are indicative of the trends in low-misfit films ( $\epsilon \approx 1\% - 2\%$ ), it is clear that the [100] SR steps should be observed during early stages of island growth since their formation energies are lowest among all the cases that we have considered. While we are not aware of a direct experimental observation confirming this prediction, our picture is consistent with the experiments of Sutter and Lagally<sup>1</sup> who find that in SiGe films with 25% Ge content, the stepped mounds are made up of [100] oriented monolayer steps. Experiments that can identify the surface structure of these stepped mounds will be invaluable in resolving the issue. In distinct contrast to the low-misfit films, experiments on pure Ge<sup>3</sup> and Si-capped Ge<sup>4</sup> islands show that the stepped mounds are made up of [110] oriented steps.<sup>16</sup> These observations can also be understood on the basis of the formation energy of the SR and DB steps in Ge shown in Fig. 2. Here, the SR steps do not become favorable until mismatch strain becomes close to 4% and 5% in the T3 and S–W potentials respectively; since the strains in these mounds should be less than the mismatch between Si and Ge (4.2%), the observation of [110] oriented steps are in agreement with our calculations.

While a negative step creation energy would lead to formation of surfaces with closely spaced steps, repulsive interactions increase as the spacing between the steps become smaller. This competition leads to an optimum slope, which can be determined using Eq. (1) as  $\theta^* \approx \sqrt{-\beta(\epsilon)/3\beta_3}$ .<sup>5</sup> The existence of such a slope is indeed confirmed by the surface energies for Ge shown in Fig. 2, where at 4% strain the (105) and (115) surfaces are seen to be the optimum facets in the (10*n*) and (11*n*) directions respectively. In the case of the (11*n*) surfaces, the (115) surface corresponds to closest spacing of DB steps (the (114) and (113) surfaces are reconstructed so that the steps lose their identity). In the presence of compressive strains, the (105) surface with SR steps has a smaller surface energy than both the high index (10*n*) surfaces shown in Fig. 2 and the low index (103) surface. It can also be seen from Fig. 2 that the surface energy of the (105) surface in Ge lies below the energy of the (115) surface throughout the range of interest, while it falls below the energy of the (117) at a strain of about 3%. In the case of Si, we

find a similar trend with the (105) surface falling below the (117) surface close to 1%. These observations indicate that the (105) facet made up of SR steps is stabilized by compressive strains in both low- and high-misfit SiGe films. On the other hand, the (105) surface with the SU steps proposed by Mo *et al.* is stabilized by tensile rather than compressive stresses since the bonds at the step-edge are in compression (refer to Fig. 1). While the results in the present work are obtained using empirical potentials, we have verified by means of *ab initio* simulations that the atoms at the edges of the SR steps in the (105) surface are indeed in a stretched state. The details of these calculations will be published elsewhere.

In summary, the nucleationless growth of stepped mounds can be understood on the basis of strain dependence of [100] and [110] oriented steps on (001) surfaces. The re-bonding at the edge plays an important role in stabilizing stepped surfaces in the presence of strain. The competition between repulsive step interactions and strain induced lowering of step formation energies leads to optimum low-energy orientations such as (105). Further work that analyzes the effect of surface segregation on step formation energies will be invaluable in gaining deeper insight into early stages of quantum dot formation in SiGe and other alloys.

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