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Understanding the elastic relaxation mechanisms of strain in Ge islands on pit-patterned Si(001) substrates

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Abstract

Substrate pre-patterning is a new and effective route for growing ordered arrays of heteroepitaxial nanoislands. Here, by exploiting elasticity theory solved by using finite element methods, we show why islands growing inside pits are better relaxed with respect to the flat-substrate case. Pit pre-patterning is demonstrated to be more important than previously realized, allowing for further degrees of freedom in controlling not only positioning but also shape, strain, and coherence of the growing islands. Our results offer a solid interpretation for the recent experimental results obtained by the group of Professor Günther Bauer.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In a recent letter with the Günther Bauer group [1] we have shown that suitable nanopatterning of a Si(001) substrate with pits (inverted square-base pyramids with inclination of about 8°) gives rise, after deposition of a few Ge monolayers, to the nucleation of a highly ordered array of coherent GeSi islands in the pits. The islands display an impressive uniformity in size and shape, along with a larger volume and aspect ratio (height-to-base ratio), with respect to the case of a flat substrate. In that work, we demonstrated through suitable atomistic and finite element method (FEM) simulations the thermodynamic origin of the experimental observation, on the basis of the enhanced degree of relaxation available for the islands in the pits, with respect to the ones on the flat substrate. However, we did not discuss there the simple reasons for Ge preferentially entering the pits (what is usually, somewhat erroneously, described as being a 'capillarity effect') and why such a relaxation occurs in the pit, contrary to the common intuitive understanding. We address such issues in this paper, along with a quantitative analysis of the energetic crossover between increasing filling of the pit and starting the nucleation of an island on top of it. All the calculations presented in the work are performed using anisotropic elasticity theory, the equilibrium condition of the elastic body being provided by FEM, as implemented in the Comsol Multiphysics package.

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2. Energy relaxation: pits and islands versus wetting layer

During coherent epitaxy of Ge on Si, the former is forced to accommodate the strain resulting from a different lattice parameter—the Si one—originating an in-plane compression that equals the lattice mismatch between these two components (4.21%). In a flat wetting layer (WL), only a tetragonal deformation is possible: an expansion in the z direction (perpendicular to the film surface) takes place in trying to conserve the cell volume, according to the Poisson ratio. Obviously, no relaxation in x and y is allowed by the planar WL geometry. The tetragonal deformation, thus, only partially relieves the strain, and a residual elastic energy density of $\sim$1.4 meV Å^{-3} is stored in the system (as obtained by a simple FEM calculation, using experimental elastic constants).

Let us now compare this very simple relaxation process to the ones for (a) a three-dimensional pit similar to the experimental ones of [1] (i.e. with 8° facet inclination), half-filled with Ge which forms a downward pyramid (DP in the following) and (b) an ideal upward pyramid (UP) on the flat

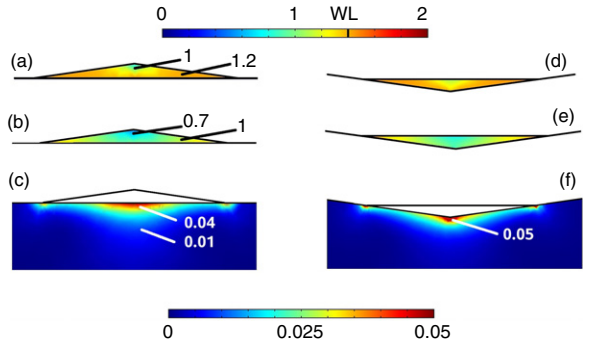

Figure 1. Elastic energy density maps (in meV Å^{-3}) for UP and DP when the substrate is fixed ((a) and (d)) and when it is free to relax ((b) and (e), respectively). Panels (c) and (f) focus on the elastic field in the substrate. The upper color scale refers to panels (a), (b), (d), (e), the lower one to panels (c) and (f). A few values are repeated on the maps to facilitate the interpretation in a black and white version of the figure.

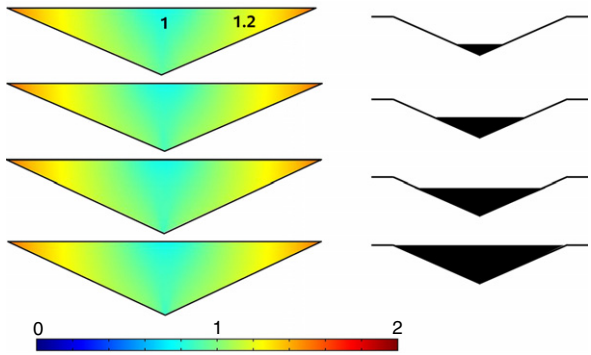

Figure 2. Elastic energy density maps (in meV Å^{-3}) for a downward pyramid progressively filling the pit. From top to bottom, full filling is reached while the energy map is not altered, i.e., the elastic field is self-similar. A few values are repeated on the maps to facilitate the interpretation in a black and white version of the paper. Note the clear higher degree of relaxation with respect to a flat wetting layer (elastic energy density of ∼1.4 meV Å^{-3}).

Table 1. Elastic energy densities W in meV Å^{-3} computed for Ge filling a 8° sidewall pit (DP), for the same geometry but reversed upside down and placed on a flat substrate (UP), and for a wetting layer. W_{Si}, W_{Ge}, W_{Ge+Si}, are respectively the elastic energy contributions due to the Si and Ge portions separately, and their sum.

Geometry	W_{Ge} (Si fixed)	W_{Ge} (Si free)	W_{Si} (Si free)	W_{Ge+Si} (Si free)
UP	1.3713	1.1213	0.1150	1.2364
WL	1.3963	1.3963	0.0000	1.3963
DP	1.3985	1.1564	0.1197	1.2684

substrate with the same facet inclination of the pit. The geometry of systems (a) and (b) can be easily inferred from figure 1. In table 1 we report the elastic energy density (W) for the three geometries, as divided into the contributions of the Ge portion (W_{Ge}) and of the Si substrate (W_{Si}). The latter is obtained by dividing the total Si elastic energy by the volume of the stressor, i.e. the Ge amount. In the same table we also report the results of calculations where the Si substrate was kept frozen, since this configuration allows us to indicate the pure relaxation mechanism generated by having zero stress component perpendicular to the free facets, with no stress redistribution between Ge and the Si substrate [2].

Both the Ge and the total (Ge + Si) elastic energy densities (W_{Ge+Si}) for the DP are found to be lower than in the case of a flat wetting layer, as can be seen in table 1. By keeping the Si frozen, we obtain very similar results in terms of elastic energy relaxation for DP and WL (first column in table 1). Intuitively, the horizontal free surface of the DP allows for (almost) the same tetragonal relaxation of the WL. In addition to that, however, the DP Ge can also relax by stressing the surrounding Si substrate. For instance, a compressive strain ε_{xx} (x being the direction parallel to the film surface in figure 1) up to +0.7% is induced close to the pit apex. This is made possible by the tilting of the Ge/Si interface (pit facet inclination) and represents the basic mechanism [3] occurring also in the case of (nearly flat) Ge islands on a flat substrate (UP), as shown in the second column of table 1. Notice the very similar amounts of substrate deformation in the UP and DP (third column in table 1). The elastic energy maps of figure 1 provide further evidence for the symmetric effect acting both in DP and in UP. The fundamental role played by substrate deformation in determining strain relaxation in three-dimensional islands (already active at early growth stages, i.e. low aspect ratios) was predicted also by analytical theories [3, 4], indicating that the lateral expansion at free facets should be regarded as a correction, while it is often seen in the literature as the main mechanism allowing for a reduced elastic energy.

In this section we have shown that pit filling offers a better elastic relaxation channel with respect to WL thickening, providing an additional thermodynamic reason for Ge preferentially filling the pits, with respect to the commonly quoted 'capillarity effect', which is based on the reduction of the total (free) surface energy. Calculations were performed for half-filled pits. Let us now show that the degree of filling is unimportant.

3. Ge relaxation versus pit filling

Let us consider a pit partially filled with Ge. If the pit is not completely full, then the Ge free surface intersects the Si sidewalls at a certain angle (172° in our pit geometry). This angle disappears at full pit filling, where sidewalls are no longer present. Since this situation corresponds to a breaking of the geometric self-similarity, we want to quantify this elastic effect, with the aim of understanding to what extent the results found in section 2 can be used beyond the assumption of a half-filled pit.

A sequence of FEM simulations were carried out by taking the same pit as considered before, with Ge reaching four different heights: 25%, 50%, 75%, and 100% of the maximum. For each simulation the elastic energy density was computed, keeping all the other parameters (such as boundary conditions and mesh element size) unaltered. The comparison of the elastic energy maps for these systems is reported in the left side of figure 2, where the DPs are drawn with the same size

(expanded with respect to the actual dimensions reported on the right, for a better comparison): the behavior is fully self-similar. It is interesting to focus our attention on the case where the Ge upper surface comes closer to the edges of the pit. Here, the geometric self-similarity is lost, but the elastic field is not altered even in this case, at least within the accuracy ($\sim 10^{-4}$ meV Å^{-3}) of our calculations. In the following, we shall therefore safely consider a self-similar behavior, using the elastic energy density calculated in the previous section independently of the degree of filling.

4. Island nucleation on the flat substrate versus that in the pit

Since pits provide better relaxation than the WL, one might ask whether island formation in the pit is still thermodynamically favored, and whether it eventually takes place for realistic pit fillings. In order to compute such critical Ge volume, one must take into account not only the elastic energy relaxation (which is proportional to the volume V, as seen above), but also the cost associated with additional surface creation. Here we shall assume that all the free facets have the same surface energy of 62.5 meV Å^{-2}. Actually, *ab initio* calculations indicate a common value of (or very close ones around) 62.5 meV Å^{-2} for the surface energy of several facet orientations [5, 6]. The dependence of the surface energy on the local strain [6], instead, could play some minor role, but it is here neglected for simplicity. For the case of a pit patterning with the same pit geometry as for our case, it has been experimentally demonstrated [7] that the early stages of Ge deposition result in a pit sidewall decoration consisting of a regular faceting with {105} stripes and (001) terraces. Since the surface energies for these facets can be safely taken equal [6], we fixed the value for the surface covered by the island in the pit to be also equal to 62.5 meVÅ^{-2}. {105} pyramids are the first islands to appear on both flat [8] and our pit-patterned substrates (8° facet inclination) [1, 7]. We here consider such islands (11° facet inclination), placed over the DP partially filling a 8° faceted pit. For simplicity we shall consider only the situation sketched at the top of figure 3, with the base of the {105} island coinciding with the upper surface of the DP. In the following, we shall indicate with DP + 105UP the region including both the DP and the {105} UP pyramid. The total energy of the two configurations, for a Ge volume V, can be schematically computed using the simple formula

$$E_{\mathrm{DP}} = \left(\frac{3}{\alpha_{1,1,10}}\right)^{\frac{2}{3}} \left(\gamma - 2C_{1,1,10}\gamma\right) V^{\frac{2}{3}} + \left(W_{\mathrm{Ge}}^{\mathrm{DP}} + W_{\mathrm{Si}}^{\mathrm{DP}} - W_{\mathrm{WL}}\right) V$$

$$E_{\mathrm{DP+105UP}} = \left(\frac{3}{\alpha_{1,1,10} + \alpha_{105}}\right)^{\frac{2}{3}} \left(2C_{105}\gamma - 2C_{1,1,10}\gamma\right) V^{\frac{2}{3}} + \left(W_{\mathrm{Ge}}^{\mathrm{UP11}} + W_{\mathrm{Si}}^{\mathrm{UP11}} - W_{\mathrm{WL}}\right) V$$

where γ is 62.5 meV Å^{-2}. Here $\alpha_{1,1,10}$ is the aspect ratio of a {1,1,10} pyramid (0.0707), and α_{105} is the aspect ratio of a {105} pyramid (0.1). The geometric parameters C read $C_{1,1,10} = \sqrt{1/4 + \alpha_{1,1,10}^2}$ and $C_{105} = \sqrt{1/4 + \alpha_{105}^2}$.

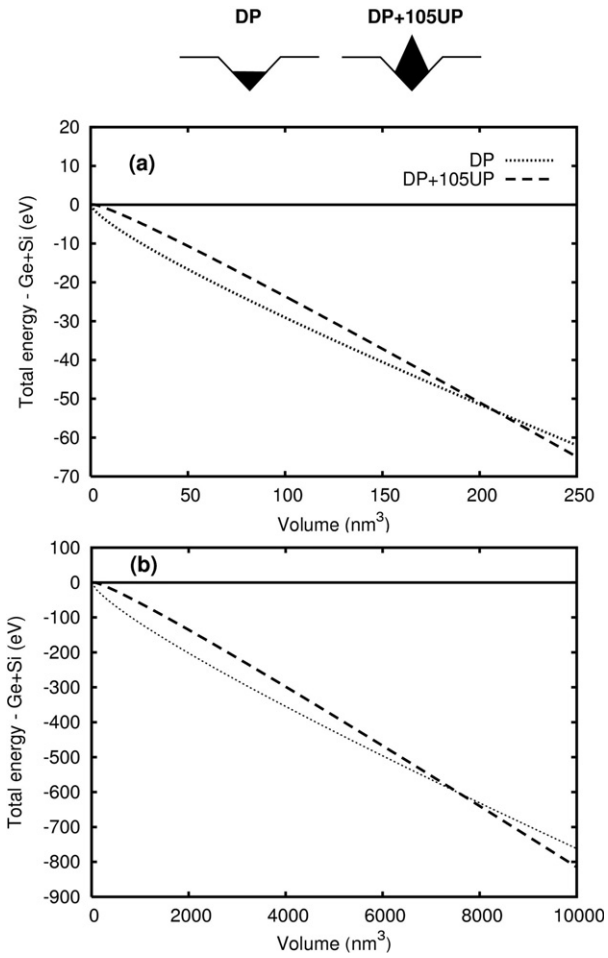

Figure 3. Total energy calculations made for a downward pyramid (DP) filling the pit and a 11° facet tilted island placed in the pit (DP + 105UP). The critical volume for shape transition from DP to DP + 105UP is the crossing point of the two curves. In panel (a) energy is computed for a concentration of 100%Ge, while in (b) this value is set to 50%.

As can be inferred from figure 3(a), where V is the total volume occupied by Ge, the energy of the DP is always negative, i.e., it is always favored with respect to the wetting layer. In fact, the surface energy change is negative: the exposed surface is smaller than the covered one. At very small volumes, a clear trend proportional to $V^{2/3}$ can be noticed. This effect is often called 'capillarity', and it is important to explain only the initial stages of pit versus WL filling observed in the experiments [7]. However, a change to linear behavior in V can be seen at larger volumes, indicating the dominant role of the extra relaxation term highlighted in the previous sections. Let us now look at the curve relative to the pyramid growing on the pit, DP + 105UP. For small volumes, its energy is higher than the DP one, because of the additional surfaces exposed. By increasing the volume, the DP energy is lowered, but the DP + 105UP's one becomes even lower since the DP and the 105UP relaxation mechanisms act at the same time. The crossing of the two curves indicates the critical volume for island nucleation in the pit. Except for extremely small volumes (not visible in figure 3), the DP + 105UP curve is lower than the one for the WL. This

behavior can be readily explained. The lower pit inclination with respect to the 105UP pyramid generates extra exposed surface when covering an empty pit with a 105UP. Therefore, as V goes to zero, the energy becomes positive. Since the extra exposed surface is very small (pit and {105} pyramids have almost equal inclinations), however, it is sufficient to reach volumes of about 5 nm^3, for obtaining a total energy lower than the WL one, as a result of the extra relaxation provided by the pit. In summary, both pit filling and island nucleation on pits are predicted to be preferential with respect to WL thickening, thus explaining the highly selective island nucleation revealed by the experiments of [1, 7, 9]. Clearly, reported values for the critical volumes are much smaller than the ones expected in actual experiments, where a certain degree of Si/Ge intermixing in the islands is expected, lowering the strain load and thus enlarging transition volumes. For a quantitative comparison, by repeating the same calculations for a uniform Ge content of 50% (see figure 3(b)), we can see that the qualitative behavior of the curves is unchanged, but the crossing points are considerably shifted towards higher values, resulting in a critical volume for island nucleation in pits of $\sim 10^4$ nm^3, i.e. of the same order as is seen experimentally [7].

5. Conclusions

Pit patterning substrates to obtain ordered arrays of deposited nanostructures is a new and powerful method, which deserves a careful understanding of the thermodynamic and kinetic effects acting at the island/pit interface. The results of [1] and of the present paper show that the influence of a pit is far reaching, allowing for a better strain relaxation. While here we have focused our attention on the pit geometry considered in [1, 7], many results are clearly dependent on the actual pit geometry and density [10]. While a systematic investigation of pit geometry relaxation is still ongoing in our group, it is clear that the possibility of fine-tuning the elastic state of the deposited material, by building suitable pits, opens up new intriguing perspectives in terms of specific pre-patterning used to obtain a specific result. Kinetic limitations (such as barriers opposing material flow inside steep pits) could however appear, limiting full exploitation of the effects discussed in this paper.

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