Energy distribution in Ge islands on Si(001): A spectral and site-resolved analysis versus size and morphology

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By a spectral and site-resolved analysis of the energy distribution (both stress and surface originated) at the atomistic scale, calculated by classical molecular dynamics for Ge islands with different morphologies on Si(001), we show how domes actually provide the largest strain release. Moreover, we point out that the usual partition of the total energy into a volume plus a surface contribution also corresponds to two separate spectral energy regions, which are the same in any morphology. However, it turns out that the volume-scaling contribution is quite complex and that the real strain relaxation term corresponds to the lower part of the energy spectrum, not scaling as the volume for small island size.

I. INTRODUCTION

The total energy of partially relaxed three-dimensional islands epitaxially grown on a substrate, with respect to a coherent film with the same number of atoms, N, has been predicted to be composed of several terms scaling as N, $N^{2/3}$, $N^{1/3}$, and $N^{1/3}\ln(N)$, on the basis of elastic continuum theory.[1] It can be summarized as

$$\Delta E = \Delta E_{el} + \Delta E_{facets} + \Delta E_{edges}, \qquad (1)$$

where the first term (negative) is the elastic energy relaxation in the island, which depends on the morphology and includes terms scaling as N, $N^{2/3}$, and $N^{1/3}\ln(N)$. The second and third terms (positive) represent the energy increase due to island surfaces and edges, scaling as $N^{2/3}$ and $N^{1/3}$, respectively.

Still, it is questionable if such a model may really apply to nanometric systems, where the border between the surface and bulk regions is not clear, and the surface itself is mainly composed of stepped facets and atomistic edges.[2–4] Anyhow, several authors[3–5] suggested that the total energy of Ge/Si(001) dislocation-free islands can safely be approximated by a volume plus a surface term, the latter including surface tension and (minor) strain relaxation contributions. This model is actually intriguing, since it resembles classical nucleation theory, where the (negative) volume term competes with a (positive) surface term. Here the former is the difference in elastic energy between the partially relaxed island and the pseudomorphic film, and in place of different phases, we consider the nucleation probability of different morphologies with one single phase. An important point of such analysis is to understand how the strain is released in the island, depending on size and morphology, and to confirm that the total energy is actually partitioned into a volume plus a surface term, independently of how much surface reconstructions can tune the value of the surface tension. This is an easy task for total energy estimations at 0 K with a classical, three-body potential (the Tersoff one, in our case[6]), which can cope with the substrate, the wetting layer (WL), and the island on top, up to few millions atoms. To this end the Ge/Si(001) system is particularly suited, due to the ab-

sence of compositional or electrostatic complications. Obviously, the estimation is obtained after molecular-dynamics- (MD-) simulated annealing, in order to produce a realistic strain distribution in the island and in the substrate.

In this paper we will not accomplish a questionable fitting of several parameters included in Eq. (1) on the total energy for pyramids or domes with different size, an issue which has already been addressed just in the simpler case of {1 0 5} pyramids by a volume plus surface term.[4] On the contrary, we suggest a partitioning of the contributions on the basis of their spectral range in energy and investigate how they scale with size and morphology. In fact, by considering as a real strain-relief term the one provided by all Ge atoms with the elastic energy below the one for a pseudomorphic film (negative term in a nucleation model), the surface contribution is virtually what remains in the higher part of the spectral energy distribution. Still, this does not guarantee that they scale, respectively, as N and $N^{2/3}$, due to the fact that the spectral region in between may include strain release plus subsurface effects of the stepped facets. To this end, a mapping of the spatial distribution at the atomic scale of different spectral contributions is very helpful in assessing their origin, especially when comparing the cross sections for different morphologies.

Finally, we do not attempt estimating the relative stability of pyramids and domes as a function of the number of atoms in the island (i.e., the size), since a quantitative prediction requires the actual facet reconstructions to be taken into account. Unfortunately, the latter are not known but for the {1 0 5} pyramids.[7] However, a comparison of the elastic energy released by different morphologies with respect to the one provided by the (2×8) reconstruction of a stepped pseudomorphic film with the same number of atoms is presented.

II. RESULTS AND DISCUSSION

We perform several runs starting from one fully strained Ge island (from 8 to 32 nm in size), coherent with the Si(001) substrate. The substrate is 8.1 nm thick and 56×56 nm^2 wide with three pseudomorphic Ge monolayers on top (the WL) and periodic boundary conditions in the (001)
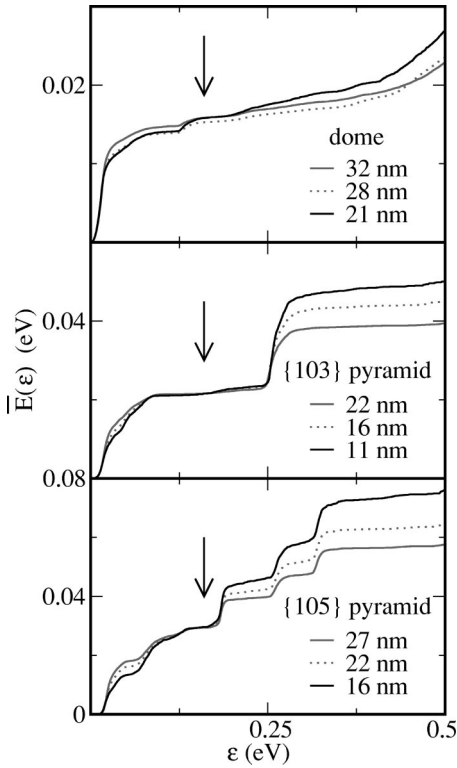
FIG. 1. Partial energy per atoms $\bar{E}$ vs the upper integration value ε [see Eq. (4)] for domes, {1 0 3} pyramids, and {1 0 5} pyramids with different sizes. The arrow indicates the threshold between the bulk and surface contributions (0.160 eV).

plane. The island includes square base pyramids, either bounded by {1 0 5} or {1 0 3} facets. The latter have rarely been observed in experiments,[8] but they are useful in understanding the effects connected to the aspect ratio [height to base ratio (h/l)].[9] Multifaceted domes, bounded by {1 0 5}, {1 1 3}, and {15 3 23} facets,[3] are also considered. The typical aspect ratios of these structures are 0.10, 0.17, and 0.20, respectively. We easily include the (2×8) surface reconstruction of the Ge WL,[10] and in the case of pyramids with {1 0 5} facets some competing reconstructions are considered.[7,11] In particular, the rebonded step (RS) and pair of dimers (PD) reconstructions are compared to the simple
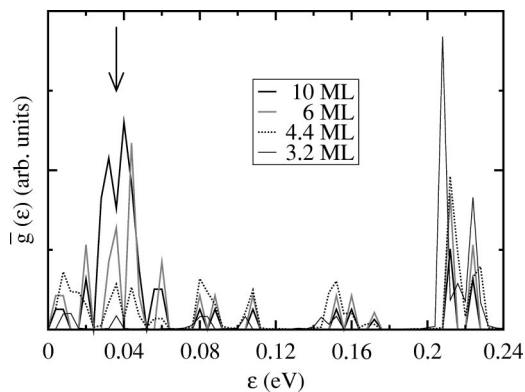

FIG. 2. Normalized distribution in energy of the atomic population for a (2×8) pseudomorphic film with a different thicknesses.
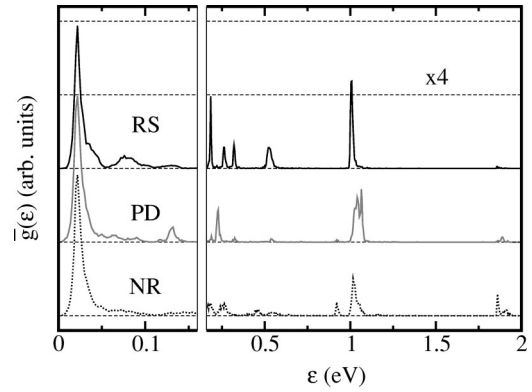

FIG. 3. Normalized distribution in the energy per atom as in Fig. 2 for a 22 nm {1 0 5} pyramid with three different surface reconstructions. See text and Refs. 7 and 11 and for details.

case of surface dimerization [equivalent to a (2×1)]. We do not accomplish any reconstruction of {1 0 3} pyramid and dome facets, because they are quite complex and not at all known. For the sake of comparison, we also simulate the same Si substrate covered by a continuous pseudomorphic Ge film with steps on top, representing layer-by-layer growth, including the (2×8) reconstruction.[10]

The actual stress distribution is achieved by a simulated annealing to 600 K, a temperature well below the usual experimental conditions (700–1000 K), still sufficient in relaxing the system and preserving the surface reconstruction. The thermal cycle lasts 140 ps, the heating and cooling rate are 2×10^{13} and 1×10^{13} K/s, respectively, and the equilibration at 600 K is 40 ps long. A final equilibration at 0 K, as long as 10 ps, is sufficient to get convergence on per atom energy to 10^{-6} eV in systems larger than 8×10^{5} atoms. The equilibrium configuration is used to evaluate the energy per atom with respect to the bulk energy (hereafter referred to as atomic excess energy, ε).

Assuming that the edge terms can be neglected (as suggested by Tersoff and Tromp[5]), the total energy is composed of only two contributions: a bulk term and a surface term (the former scaling as N, the latter as $N^{2/3}$). Moreover, by supposing that there is also a separation in the spectral energy distribution $g(\varepsilon)$ between bulk energies (lower side) and surface ones (higher side), a threshold $\tilde{\varepsilon}$ in the excess energy per atom can be found as follows. The bulk energy
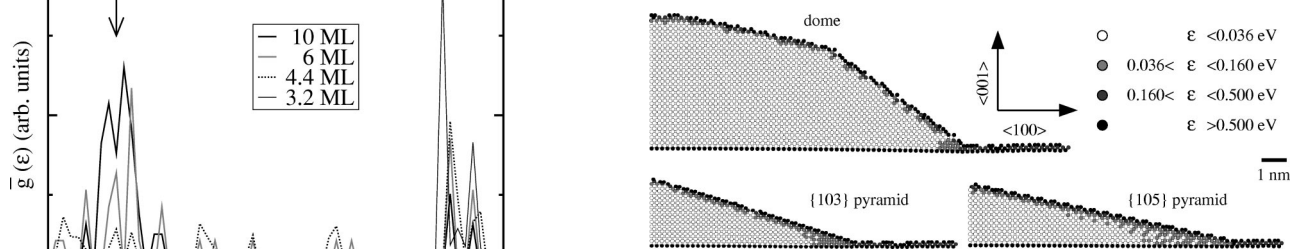

FIG. 4. Energy maps for Ge atoms in the $\langle 010 \rangle$ cross-section view for half a dome (32 nm), a {1 0 3} pyramid (22 nm), and a {1 0 5} pyramid (27 nm). The cross-cut is taken at the middle of the island and the atoms are indicated according to their energy with respect to bulk Ge.
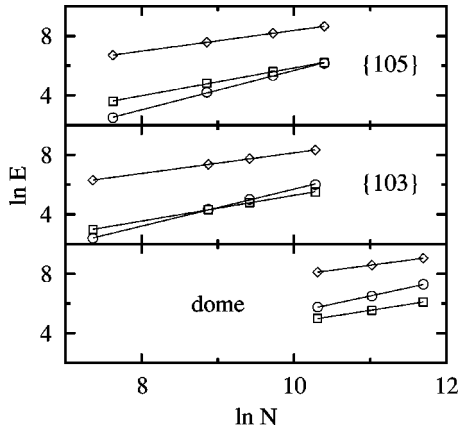
FIG. 5. Fitting of $\ln(E)$ vs $\ln N$ for three morphologies in different energy ranges: $\varepsilon < 0.036$ eV (circles), $0.036 < \varepsilon < 0.160$ eV (squares), and $\varepsilon > 0.160$ eV (diamonds).

$$E_b(\tilde{\varepsilon}) = \int_0^{\tilde{\varepsilon}} w g(w) dw \qquad (2)$$

should be proportional to N, whereas the surface energy

$$E_s(\tilde{\varepsilon}) = \int_{\tilde{\varepsilon}}^{\infty} w g(w) dw \qquad (3)$$

should scale as $N^{2/3}$. In order to estimate $\tilde{\varepsilon}$, the partial energy per atom,

$$\bar{E}(\varepsilon) = \frac{\int_0^{\varepsilon} w g(w) dw}{N} \qquad (4)$$

can be plotted for different dot sizes. The threshold for the surface contribution should be indicated by a progressive splitting of degenerate lines, with the one for the smallest island on top and the one for the largest one at the bottom. In Fig. 1 we plot $\bar{E}(\varepsilon)$ for domes (top panel), {1 0 3} pyramids (middle panel), and RS-reconstructed {1 0 5} pyramids (bottom panel) with different sizes. Here we limit the spectral range of ε to 0.5 eV, as beyond that value the curves do not display any further information. We see that the lines start separating at about 0.160 eV for any morphology (arrow in the panels). In the case of the dome, the trend after 0.160 eV is slightly ambiguous, as small variations in $g(\varepsilon)$ are present, because the simulated islands of different sizes are not exactly self-similar, due to the complex shape constraint imposed by the different facets.

Therefore, at excess energies larger than 0.160 eV surface-tension contributions should play a relevant role. However, at energies lower than 0.160 eV the "bulk lines" do not superimpose, especially for the {1 0 5} pyramids, suggesting that this spectral region should be divided in two: one that scales at a higher power than N (low-energy side) and one that scales at a lower power of N (high-energy side). Actually, by calculating the normalized distribution in energy for a (2×8)-reconstructed stepped pseudomorphic film of increasing thickness (Fig. 2) we see that true surface features (decreasing in intensity with thickness) appear above 0.2 eV, and subsurface contributions related to the elastic effects of the (2×8) reconstruction are located below 0.160–0.180 eV (decreasing with thickness only above 4.5 ML). A progressive blowup of the features at 0.036 eV appears, which corresponds to the excess elastic energy of a Ge atom in a film with no free surfaces, compressed to the Si lattice parameter in the (001) plane and relaxed along the $\langle 001 \rangle$ direction. Therefore, we consider as the unambiguous strain release contribution only the one provided in the spectral region below 0.036 eV.

In Fig. 3 we report the normalized distribution $\bar{g}(\varepsilon)$ [i.e., $g(\varepsilon)/N$] for one 22 nm {1 0 5} pyramid, with different facet reconstruction (RS and PD models) (Refs. 7 and 11) and without any reconstruction (NR), except for the spontaneous surface dimerization which occurred during the thermal cycle. Note that the low-energy peak below 0.036 eV provides a larger strain relief than the stepped pseudomorphic film, preserving its shape and intensity independently of the surface reconstruction. However, the surface effects (mainly located above 0.160 eV) play a role also in the energy region between 0.036 and 0.160 eV, depending on the different reconstructions. This is confirmed by mapping the spatial distribution of the atoms pertaining to the different spectral regions. In Fig. 4 we report in a cross-cut along $\langle 010 \rangle$ for half the dome the {1 0 3} pyramid and the RS {1 0 5} pyramid.[12] We see that in each of the three cases the main part of the island does display a strain relaxation with respect to a thick Ge film (open circles). These atoms contribute to the first term in Eq. (1). Figure 4 indicates also that the most energetic atoms (dark gray and black dots) are in the second and first layers at the surface, respectively, the latter being consistent to nearly one Ge broken bond (0.962 eV in our model). Finally, a sizable number of atoms (light gray circles) cannot be sharply identified as surface or bulk atoms since they are located below the surface and in the narrow region close to the base edges of the island.

An estimation of the actual power of N, hereafter indicated as α, by which the total energy E scale in the different

TABLE I. Exponent of the scaling law N^{α} for the total energy in different regions of the excess energy spectrum, as reported for different island morphologies (see text). The aspect ratio h/l is also reported.

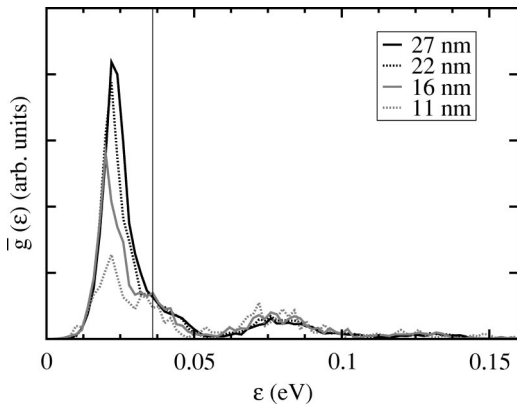|	h/l	0–0.036 eV	0.036–0.160 eV	0–0.160 eV	0.160 eV–∞
Dome	0.20	1.10	0.78	1.02	0.66
{103} pyramid	0.17	1.25	0.85	1.04	0.69
{105} pyramid	0.10	1.35	0.93	1.08	0.70

FIG. 6. Normalized distribution of the energy per atom in $\{1\ 0\ 5\}$ pyramids with different sizes for a spectral region corresponding to the energy contribution scaling as the number of atoms. The vertical line indicates the excess energy of a pseudomorphic Ge film without free surfaces (0.036 eV).

spectral regions ($\varepsilon < 0.036$ eV, $0.036 < \varepsilon < 0.160$ eV, and $\varepsilon > 0.160$ eV) can be attempted by fitting a plot of $\ln(E)$ vs $\ln(N)$ (as reported in Fig. 5). In Table I, we see that for $\varepsilon < 0.160$ eV and $\varepsilon > 0.160$ eV the total energy contributions nearly scale as a bulk ($\alpha = 1$) and a surface ($\alpha = 0.67$) term for any morphology, particularly well for domes. Still, for $\varepsilon < 0.160$ eV two separate spectral regions behave differently, since the contribution below 0.036 eV (pure relaxation) scales at a higher power of N ($\alpha \cong 1.10–1.35$) while for $0.036 < \varepsilon < 0.160$ eV it scales in between a surface and a bulk term ($\alpha \cong 0.78–0.93$). Notably, this effect is much more relevant for morphologies with smaller aspect ratio. In particular, Fig. 6 clearly shows that for the $\{1\ 0\ 5\}$ pyramids the fraction of atoms below 0.036 eV increases with pyramid dimension, while the reverse occurs for $\varepsilon > 0.036$ eV. Thus, a spectral intensity transfer between these two regions occurs (decreasingly) with size. This can be understood in terms of Eq. (1) by considering that, by increasing the dimension of smaller pyramids, the fraction of relaxed atoms ($\varepsilon < 0.036$ eV) is not constant, because the edge and surface contributions are progressively decreasing their effects on
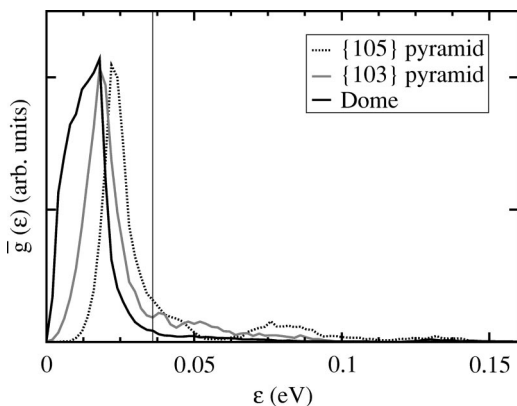

FIG. 7. The same as Fig. 6 for a dome (32 nm, solid black line), a $\{1\ 0\ 3\}$ pyramid (22 nm, solid gray line), and a $\{1\ 0\ 5\}$ pyramid (27 nm, dashed gray line).
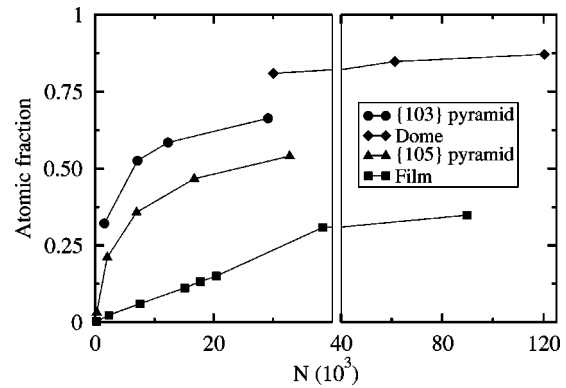

FIG. 8. Fraction of atoms with energy per atom lower than 0.036 eV as a function of the number of atoms in the island for the three morphologies. The values for a stepped pseudomorphic film are also reported.

the core region. In particularly, from Fig. 4 we also note that the spectral intensity transfer from the $0.036 < \varepsilon < 0.160$ eV fraction originates nearby the interface with the WL, close to the base edges.

The aspect ratio of the island deeply influences the excess energy distribution. In fact, it is qualitatively understood that the steeper the morphology, the larger the strain relief in the island.[9] This issue is quantitatively confirmed in Fig. 7, where the normalized distribution $\bar{g}(\varepsilon)$ in the low-energy size is reported for islands larger than 30×10^3 atoms, with different morphologies. Note that the position of the maximum shifts towards low energy with increasing the aspect ratio and the shape of the dome distribution is different, with a larger fraction of relaxed atoms and negligible surface structures. Accordingly, we report in Fig. 8 the fraction of atoms which have ε lower than 0.036 eV versus N. Here, for the sake of comparison, we also display the values computed for a Ge pseudomorphic film on top the WL. Starting from 30×10^3 atoms the hierarchy in stress relaxation among different morphologies is very clear: the dome has a larger fraction of relaxed atoms. The initial trend with size of pyramids is very steep, suggesting that the measurement of the distribution of the in-plane lattice parameter for tiny islands should be broader than the one for the larger domes, which displays a nearly constant fraction of relaxed atoms.

The reason why the pseudomorphic film displays an increasing fraction of relaxed atoms is related to the (2×8) stress-relieving reconstruction, as also reported in Fig. 2. Such an effect reaches saturation when the film is sufficiently thick that the lower layers cannot take advantage of the (2×8) surface reconstruction and the fraction of atoms below 0.036 eV eventually vanish for $N \to \infty$.

III. CONCLUSIONS

The results of our atomistic simulations indicate that a partition of the total energy into a volume-scaling plus a surface-scaling contributions for three dimensional epitaxial Ge islands on Si(001) is possible, particularly on the basis of a different origin in the spectrum of the energy per atom. In fact, the total energy provided by Ge atoms below 0.160 eV

is proportional to the total number of atoms, N, whereas the one originated by Ge atoms above 0.160 eV scales as $N^{2/3}$. Remarkably, the threshold separating such elastic contributions from the surface-tension one is the same in any morphology, stepped film included. However, strain release with respect to a pseudomorphic stepped film is strictly provided only by the fraction of atoms below 0.036 eV, which scale as N^{α}, with α larger than 1, especially for pyramids of small size. The atoms contributing to the spectral energy region $0.036 < \varepsilon < 0.160$ eV are understood to be located in a subsurface region, or close to the island base, and the spectral intensity transfer to the $\varepsilon < 0.036$ eV region with size can be considered equivalent (still not the same) to the effect of edge and surface terms in the first term of Eq. (1). Despite a quantitative comparison of the total energy provided by the {1 0 5} pyramids and the domes being not possible, due to the missing information on the surface reconstruction for the

dome {1 1 3} and {15 3 23} facets, the analysis of the atomic fraction with energy per atom below the one for an infinite pseudomorphic film (true relaxation) indicates that the dome morphology by far releases a larger amount of strain. Finally, the inhomogeneous and size-dependent nature of the strain in the {1 0 5} pyramids indicates that a naive nucleation picture including a (negative) volume term, representing the average strain relaxation, plus a (positive) surface-tension contribution cannot be straightforwardly applied.

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[12] See EPAPS Document No. E-PRBMD0-66-235408 for a color scale version of the figure. A direct link to this document may be found in the online article's HTML reference section. The document may also be reached via the EPAPS homepage (http://www.aip.org/pubservs/epaps.html) or from ftp.aip.org in the directory /epaps/. See the EPAPS homepage for more information.