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CONCENTRATION PROFILES IN HETEROEPITAXIAL NANOISLANDS

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Wide experimental evidence for non-uniform Ge concentration profiles in SiGe islands calls for the development of theoretical methods able to investigate the local distribution which allows for the best elastic-energy relaxation. Here, after reviewing a fast computational technique recently introduced in the literature [New J. Phys. **10**, 083039 (2008)], we apply it to the study of intermixed three-dimensional pyramids of different aspect ratios. The local Ge distribution minimizing the elastic energy is found, emphasizing the driving force for Si enrichment close to the island base edges.

1. Introduction

Deposition of Ge on Si(001) leads to Stranski-Krastanow (SK) growth: the formation of a thin wetting layer (WL) is indeed followed by the creation of three-dimensional (3D) islands. While building 3D structures requires an energetic cost related to the formation of extra exposed surfaces, it also allows for a better mismatch-strain relaxation (Ge has a lattice parameter ~4% larger than Si), the latter term dominating at large enough volumes. In general, the higher is the aspect ratio the stronger is the volumetric energy relaxation [1], so that one expects, by increasing the amount of deposited material, a transition from flat film to shallow islands, and then a transformation towards steeper geometries. Although this general trend is indeed observed [2], some interesting additional effects take place, making the description of the whole SK growth mode more complex. Si atoms coming from the original substrate can indeed mix with the deposited Ge, leading to alloyed SiGe islands characterized by a lower effective misfit and, as a consequence, by a lower elastic energy. Several experimental observations (exploiting X-rays diffraction and/or selective etching) allowed to characterize the local distribution of Ge inside the islands, pointing out strong deviations from uniform distributions (for very recent references, see Refs. [3-6]). The process of SiGe mixing depends on the growth parameters. For example, high temperatures T are well known to promote a higher average Si content. At the same time, one expects the entropic contribution to push towards more uniform distributions at higher T, but one should always consider that the system is not able to explore the full phase space since in the T-range of interest ([500,700]°C), bulk diffusion is frozen. Because of the interplay between several different contributions, it is important to establish at least some limiting cases. If from one side one can see a uniform SiGe distribution as representative of a system where entropy plays a more important role with respect to strain relaxation, at the other extreme it is interesting to look at the distribution $c_{\min}(x, y, z)$ which minimizes the elastic energy.

In this paper we shall explain how it is possible to determine theoretically $c_{\min}(x, y, z)$, giving a few technical details concerning a recently introduced method which exploits Monte Carlo within a Finite Element Methods approach (MC-FEM). After applying such MC-FEM method to intermixed Ge pyramids of different aspect ratios, we shall show how the steepness of the island facets influences the SiGe distribution.

2. Minimizing the elastic energy for a given shape and average Ge concentration

A very direct approach for finding the Ge distribution $c_{\min}(x, y, z)$ which minimizes the elastic energy of an island of a given shape is given by Metropolis-like MC simulations within an atomistic approach [7,8]. If the empirical potential used to describe Si-Si, Si-Ge, and Ge-Ge interaction provides a good description of the elastic constants, one can reliably find $c_{\min}(x, y, z)$ by randomly picking pairs made of one Ge and one Si atom, exchange their type, minimize the energy of the system, and accept the exchange if it lowers the elastic load. Such simulations are however extremely CPU-time consuming. We have therefore devised a similar method which however exploits fast continuum elasticity theory calculations. In our MC-FEM technique two different meshes are defined (see Fig. 1). A fine one, used to solve the equilibrium condition of the elastic body (island + substrate), and a coarser one which is exploited to change the Ge concentration profile. The procedure can be conveniently started by considering a uniform Ge distribution $c(x,y,z)=\bar{c}, 0\le\bar{c}\le 1$. If for islands made of pure Ge the initial (prior to relaxation) internal strain condition reads $\varepsilon_{xx} = \varepsilon_{yy} = -0.04$ (x and y being perpendicular directions in the growth plane), for uniformly alloyed islands it is sufficient to set $\varepsilon_{xx} = \varepsilon_{yy} = -0.04\times\bar{c}$. Any FEM solver (all calculations reported here were

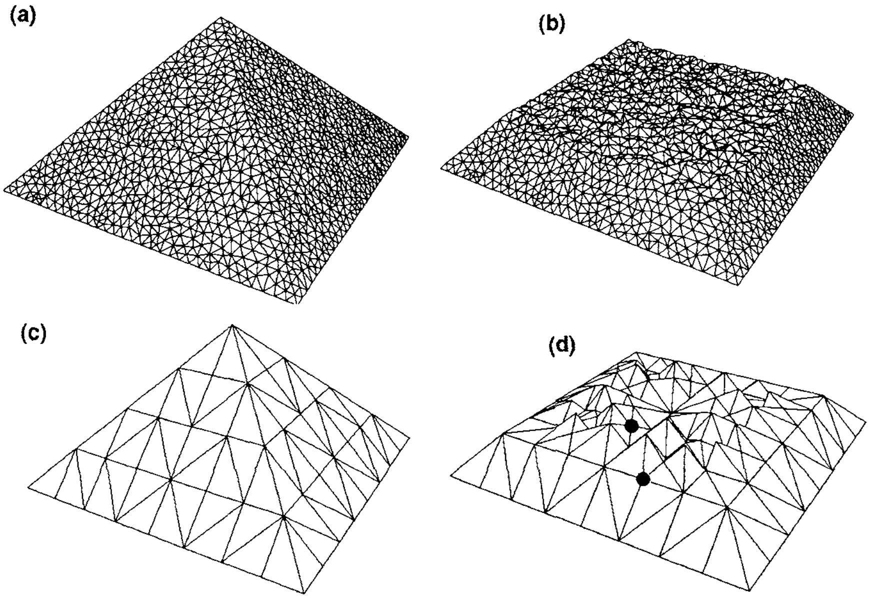

Figure 1. Two different meshes used in our MC-FEM calculations. Panels (a) and (b) display the fine mesh used to solve the elastic problem, while the coarser mesh among which concentration changes are attempted (for example, involving the nodes indicated with filled circles in panel (d)) is shown in panels (c) and (d). Panels (a) and (c) highlight nodes at the island surface, while in (b) and (d) a cross-cut shows also some internal nodes. The Si substrate below the islands is not shown.

performed using the Comsol Multiphysics commercial code) can then be used to find the proper equilibrium condition, and to evalutate the elastic energy E.

We then randomly pick two nodes of the coarse mesh and we make opposite random changes in the concentration value, suitably rescaling values in neighboring nodes to make sure that the average Ge concentration in the island is unchanged. As usual in FEM, values outside the nodes are computed by linear interpolation. The Ge distribution is now a non-uniform function $c(x, y, z)$, leading to an initial internal strain condition $\varepsilon_{xx}(x, y, z) = \varepsilon_{yy}(x, y, z) = -0.04\times c(x, y, z)$. The elastic problem is solved, and a new value of elastic energy (in the whole system, *i.e.* including the Si substrate) is found. If it is lower than the initially estimated E, the exchange is accepted and the procedure is iterated until convergence is reached. Considering that a mesh of $\sim10^4$ nodes (see Fig. 1a and Fig. 1b) is needed to properly solve the elastic problem, the use of a coarser mesh (Fig. 1c,d) to control the concentration profile is fundamental in order to reduce the number of required iterations. We verified that 10^2 nodes are sufficient to accurately describe typical Ge distributions within 3D islands. Notice that at each step the elastic problem is solved based on the last accepted

Ge distribution. With this respect, the method is fully self-consistent. Further details on MC-FEM can be found in Refs. [9,10]. In Ref. [10] the method was also extended to treat entropic contributions at finite temperatures. Here, however, we shall focus on elastic-energy minimization only. Let us now apply the method to islands of different height-to-base aspect ratios, quantifying the effect of non-uniform concentration profiles on the elastic energy stored in 3D islands.

3. Extra-relaxation provided by non-uniform concentration profiles

Here we shall analyze $c_{\min}(x, y, z)$ for 3D pyramidal islands of different aspect ratio, as obtained exploiting the MC-FEM procedure. Shapes corresponding to (105), (113), (101), and (111) square-based pyramids are considered. The average Ge composition is fixed to the value $\bar{c} = 0.6$, while the substrate is made of pure Si. Results are displayed in Fig. 2.

It is clear that for all the considered islands, Ge tends to accumulate towards the top (light areas), leaving Si-rich regions (dark) close to the base edges. The reason for this behavior can be immediately understood by looking at the left column of Fig. 3, where the elastic energy (density) of the islands is plotted in the case of a uniform distribution. Si tends to be accumulated precisely where the elastic energy is higher, *i.e.* close to the base edges. Having a smaller lattice parameter, the presence of silicon helps releasing the compressive field. By enhancing the aspect ratio the driving force leading to deviations from uniformity becomes stronger. Indeed, islands with an aspect ratio of 0.7 show a pronounced segregation between Ge and Si.

It is interesting to quantify the extra-relaxation energy provided by having Ge inside the islands distributed following $c_{\min}(x, y, z)$ and not uniformly.

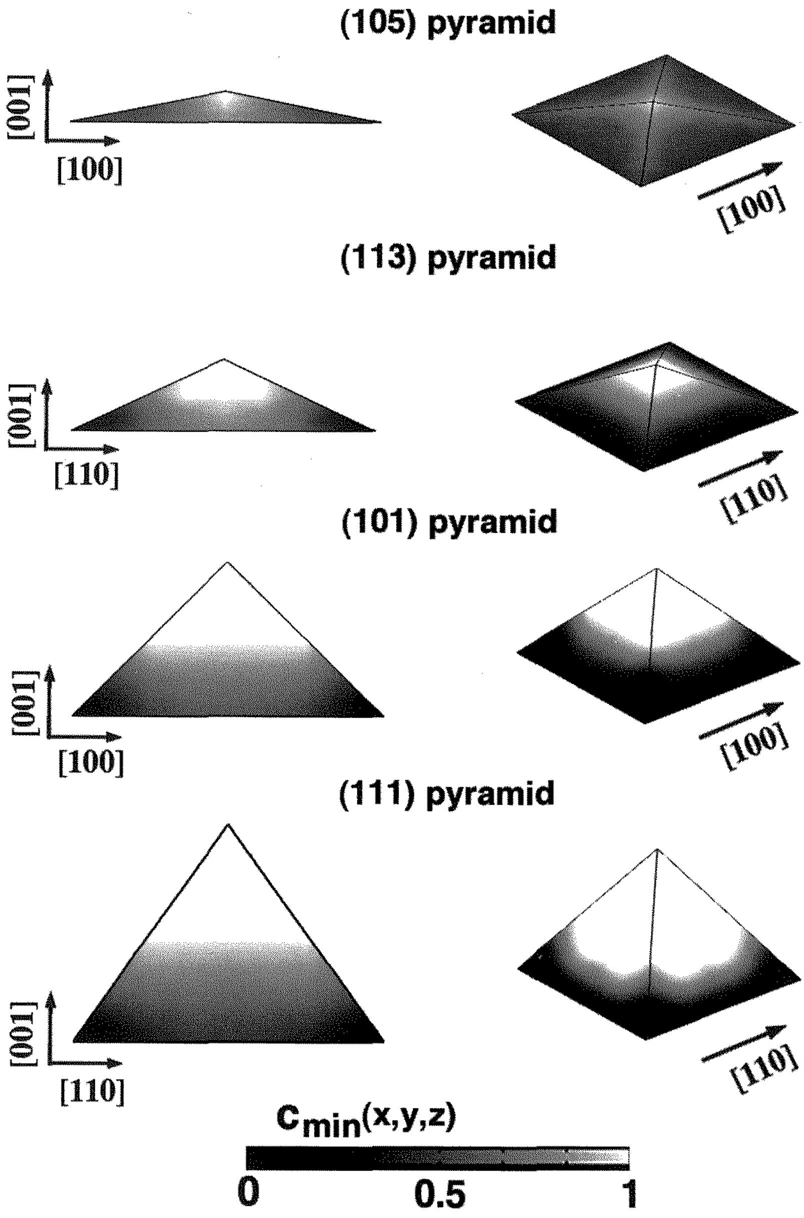

Figure 2. Concentration profile minimizing the elastic energy for different pyramids. Left column: cross-sectional view. Right column: perspective view. The aspect ratio of (105), (113), (101), and (111) pyramids is 0.1, 0.23, 0.5, and 0.707, respectively.

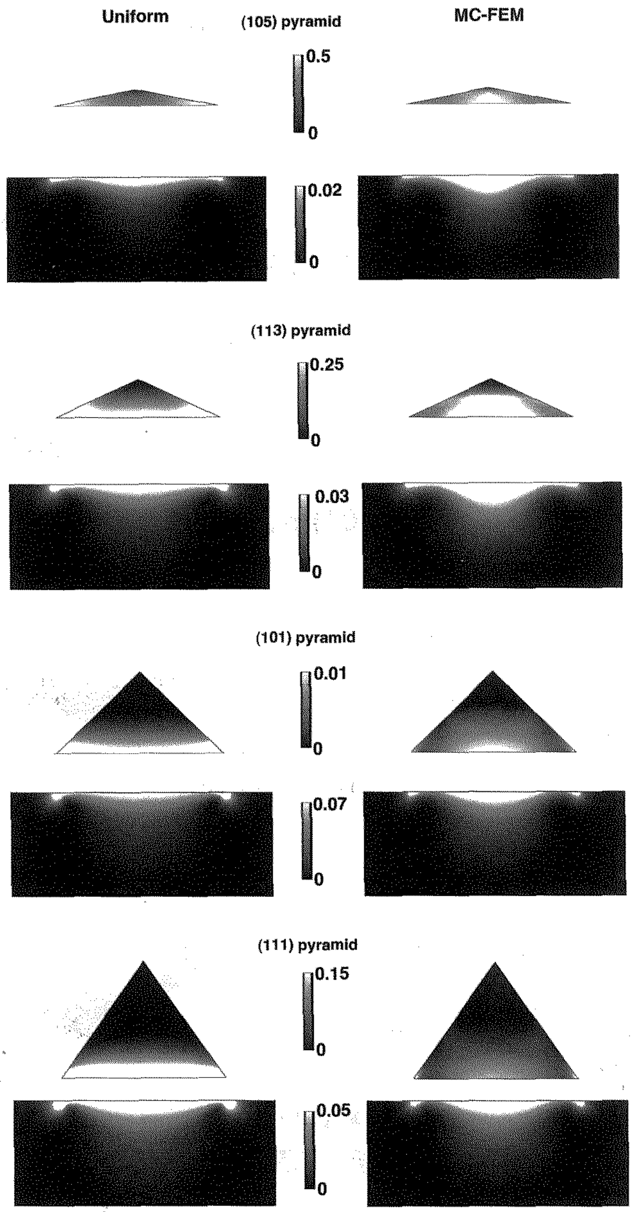

Figure 3. Elastic energy density in eV/nm^3 for the various islands, displayed in a cross-sectional view. Left column: uniform distribution. Right column: after MC-FEM. Notice that a different scale is used for islands and substrate. In the figure only, we inserted a void region between island and substrate in order to facilitate the distinction between the island and the substrate behavior.

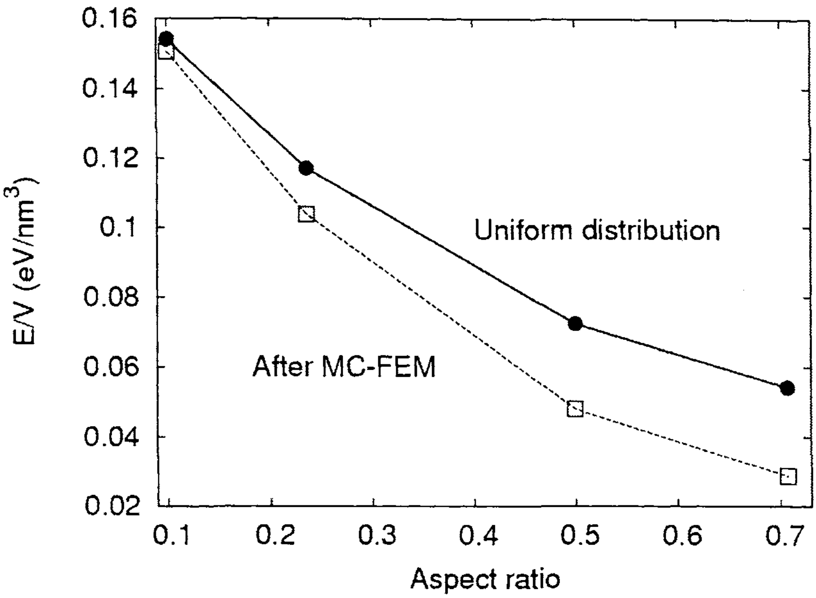

Figure 4. Elastic energy density as a function of the aspect ratio for an average Ge concentration of 0.6. Continuous line with full circles: uniform Ge distribution. Dashed line with empty boxes: Ge distribution minimizing the elastic energy, as obtained using MC-FEM. The reported quantity is computed by dividing the elastic energy of the full system (substrate + island) by the volume of the island.

In Fig. 4 we report the elastic energy stored in the system (normalized over the island volume) as a function of the island aspect ratio, for both a uniform Ge distribution and $c_{\min}(x, y, z)$ provided by MC-FEM. Let us first consider the simple case of a uniform distribution. By moving from shallow to steep islands, the elastic energy clearly decreases owed to the strongest relaxation allowed for by the larger component of the surface normal in the growth plane. Despite the better relaxation, however, the MC-FEM curves show that the driving force pushing the Ge distribution away from uniformity grows with the aspect ratio. Fig. 3 reveals the important role played also by the deformation extending in the Si substrate. At high aspect ratios, indeed, the non-uniform Ge distribution, strongly lowers the elastic energy stored below the island. This effect can be interpreted in view of the pronounced Si/Ge segregation (Fig. 2). While a Ge-rich island acts as a tensile stressor on the silicon below [1], if the base becomes strongly enriched in Si the effect tends to vanish.

4. Conclusions

The Ge distribution minimizing the elastic energy in 3D islands was calculated for pyramidal islands of various aspect ratio, exploiting the recent MC-FEM method. Results showed that the driving force leading to non-uniform profiles is stronger for steeper aspect ratios. It is important to emphasize that if, on general grounds, $c_{\min}(x, y, z)$ should be regarded only as a relevant theoretical limit, important features of the analyzed distributions, such as Ge-rich island top and Si-rich edges were confirmed by actual experiments [3,6].

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