Modified two-dimensional to three-dimensional growth transition process in multistacked self-organized quantum dots

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This report proposes ideas that enlighten the modified two-dimensional to three-dimensional transition process in multistacked self-organized quantum dots. Actually, it is shown that the driving force for both the vertical correlation and the reduction of the critical thickness experimentally observed in multistacked self-organized quantum dots does not merely result from the nonuniform strain distribution (induced by the buried dots) as usually accepted, but rather from the elastic interaction between this strain distribution and the strained surface islands. This has been shown by calculating (within continuous elasticity framework) the strain distribution in the case of Ge islands in a Si matrix for which recent experiments are available, but the result applies to other multistacked self-organized quantum dot systems such as InAs/GaAs or InN/GaN.

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It is currently well accepted that, when growing multilayer arrays of coherently strained islands, the strain distribution due to the layer of buried dots favors vertical alignment organization.[1–5] All these very informative previous studies have considered the variations of the ''strain energy density'' at the surface of the spacer layer, in other words, after burying the first dot layer and before depositing the second dot layer.

Recent experiments[7,6] on Ge/Si(001) quantum dots in multilayer structure first confirm the dot vertical autoorganization by transmission electron microscopy (TEM) measurements, and second clearly demonstrate that the critical thickness in the second layer is significantly lower than that in the first layer. They also clearly pointed out that by varying systematically the thickness of the spacer layer from 8 nm to 150 nm, the thinner the spacer layer, the earlier the two-dimensional to three-dimensional (2D-3D) transition occurs, and the better the vertical correlation.

In this report we propose an explanation for such a behavior. Three reasons can be invoked.

(i) Some segregation of Ge towards the surface during the burying process that provides additional available Ge in the second layer.

(ii) The roughening of the surface that occurs during the deposition of the spacer layer.

(iii) The effect of the strain distribution on the nucleation of 3D islands at the surface of the spacer layer.

The first potential cause, even if partly efficient, appears not to be sufficient for explaining such a wide range of critical thickness variations [from 4 monolayers (ML's) for the thickest spacers to less than 1 ML for the thinnest ones]. The second hypothesis will not be considered here, as it appears that in those experiments the roughness seems to be limited. One has to recall that this is not the case when one deposits SiGe alloys instead of pure Ge layers.[8,9] Both effects of Ge segregation in the spacer layer and roughness enhancement cannot be neglected in the case of low-misfit alloy heteroepitaxy. However, as we are interested here in pure semiconductor heteroepitaxy, where these effects will be significantly lowered, we have chosen to focus here on what we suppose to be the main key parameter, the buried-dot-induced strain distribution.

In order to learn a bit more about the effective role of this strain distribution, we have chosen to calculate it within linear continuous elasticity framework, by making use of the 3D finite difference method,[10] taking into account the actual shape of buried islands and the anisotropic character of the spacer layer material. Because Ge/Si is a representative case of high mismatched heteroepitaxy, we have considered without any loss of generality a square periodic array of buried Ge-truncated square-based pyramids in a Si matrix. The period is 200 nm, the dot basis is 100 nm wide, and the dot height 6 nm, which corresponds to the observed data. We used octahaedral meshes typically 3 nm large, and the Si thick substrate was simulated by imposing a ''frozen to bulk value'' bottom layer more than 300 nm below the first Ge box layer. Whereas the elastic energy density on the Si surface is clearly defined, it is interesting to define an ''elastic energy density on the surface from the germanium point of view'' as the local elastic energy density of a thin, fully wetting Ge layer deposited on the surface. The resulting elastic energy densities on the surface given in Fig. 1 present, for a thin spacer layer, four optimal locations for germanium deposition on the surface (which correspond to the more strained areas for the silicon).[11] This agrees with the results of Ref. 5, which were obtained by neglecting the actual shape of the buried dot and considering isolated dots. Due to the interaction of buried dots, one no longer observes invariant contours when using reduced coordinates x/h_b and y/h_b (where h_b is the spacer layer thickness).

Once we have this strain distribution, it is useful to go further and to study how it influences the deposition of the second Ge layer. For this, we have calculated the total (relaxed) elastic energy of a system made of those periodically arranged buried Ge dots on which is deposited a Ge monolayer plus a small ''box'' (typically a square-based 2D platelet, one or two ML's high, 1 or 2 nm wide).[12] We have performed several calculations for different spacer layer thicknesses and with moving of the small deposited surface box all over the period. The main information one gets from

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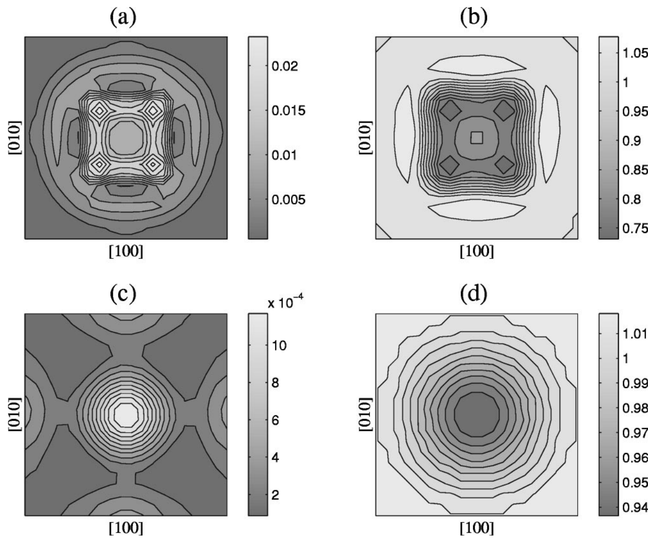

FIG. 1. Variations of the Si or Ge elastic energy density (EED) at the surface of a buried array of Ge dots in Si (see text for details). The contours are given over one period for two different spacer layer thicknesses. EED's are normalized to the energy density in a homogeneously biaxially strained Ge layer on Si. (a) : Si EED, $h_b = 15$ nm, (b) : Ge EED, $h_b = 15$ nm, (c) : Si EED, $h_b = 60$ nm, (d) : Ge EED, $h_b = 60$ nm.

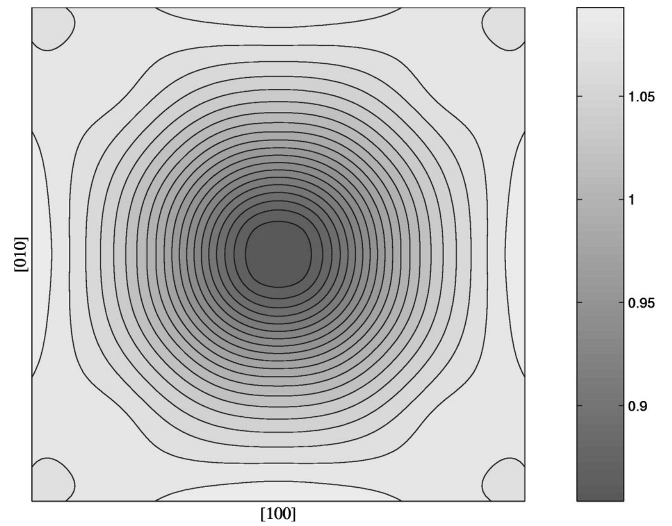

FIG. 2. Variations of the normalized reduced energy of a 2D Ge platelet at the surface of a buried array of Ge dots in Si, for a spacer layer of 21 nm. The platelet considered here is square based, $\simeq 60$ nm wide and 2 ML (i.e., 0.3 nm) high.

this study is that, as long as the box is small (typically lower than 10 nm), it relaxes so well that the the energy of this box varies less than 1/1000 when the box moves all over the surface. In order to be sensitive to the strain distribution at the surface, the box needs to be wide enough. This point contradicts the rather frequently assumed idea that the buried-dot-induced strain distribution at the surface creates *nucleation sites* at the surface: very small atom clusters will not be sensitive to the strain modulation of the surface as they elastically relax too much and thus their elastic contribution is too weak for modifying significantly the strain distribution. Table I displays the variations of the reduced energy of an about 60 nm wide platelet as the spacer thickness varies. Energies are normalized to the biaxial energy so that a value lower than 1 indicates a platetet less strained than a full 2D layer and a value greater than 1 corresponds to an unstable location (of course the surface energy has to be added for determining the stability on the platelet). For such a ''wide'' platelet, one no longer observes four equivalent minima around the ''vertical correlation'' (called the ''center''), as can be viewed in Fig. 2. As a matter of fact, the optimal location of the platelet is above the buried dots. This indicates that the surface strain distribution due to the buried-dot array is strong enough to ''organize'' the deposition of 2D large enough platelets during the 2D growth mode of the second Ge layer. As long as the platelet widens and covers more and more of the surface, the elastic energy gain at optimal position vanishes (one tends to the full wetting layer value given in the last line of Table I). Figure 3 clarifies this a bit more: the energy difference between the optimal loca-

TABLE I. Normalized reduced energy of a 2D Ge platelet at the surface of a buried array of Ge dots in Si located at just above a buried dot (center) or between the dots (corner), for several spacer layer thicknesses h_b (in nm). The energies correspond to the average energy per atom in the platelet, and normalized to the value of a perfectly biaxially strained 2D Ge layer on a uniform [100] Si substrate, that is, typically 37 meV/atom. The platelet considered here is square based, 61.25 nm wide, and 2 ML (i.e., 0.3 nm) high.

h_b	21	30	60	90
Center	0.854	0.887	0.925	0.966
Corner	1.082	1.072	0.996	0.992
Full wetting	1.000	0.987	1.001	0.999

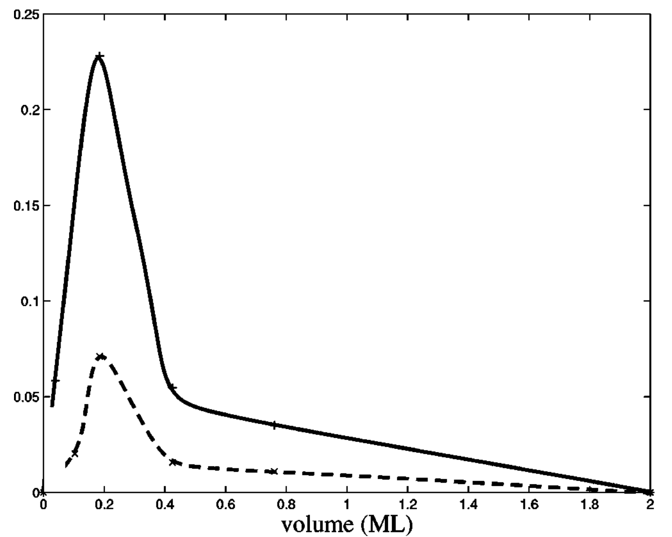

FIG. 3. Variations, versus the platelet size, of the energy difference between the optimally located and the less favorably located 2D Ge platelet (2 ML high) at the surface of a buried array of Ge dots in Si, for a spacer layer of 21 nm (full line) and 60 nm (dotted line).

tion and the less favorable one depends on both the size of the platelet and the spacer layer thickness. Moreover, the size corresponding to the strongest vertical organization (curves maxima in Fig. 3) also depends on the height of the platelet.

The calculation we have just described can be summarized by two points. First, during the 2D growth mode platelets will preferentially locate in vertical correlation with the buried dots. When the growth turns to the 3D mode, as the transition is rather abrupt, one can consider that the first nucleating islands will be located where the 2D platelets present in the 2D growth mode were[13] (available Ge atoms gathering preferentially there, in order to form 3D islands). Consequently the vertical correlation between two successive dot layers stems from the correlation between the 2D surface platelets and buried dots. Second, and certainly the main point, the *elastic interaction* between the surface islands and the buried dots (intrinsically included in our calculation) appears to be more efficient than the buried-dot-induced strain distribution itself. In other words, one could say that in the vertical correlation phenomenon, the surface dots do not simply undergo the strain distribution but play an active role as their own relaxation enhances the energy distribution variations. We have checked that one can find surface dots (thicker than 1 or 2 ML's) and spacer thicknesses for which the elastic energy gain in optimal position is more than 30%.

Let us now turn to the two-dimensional to three-dimensional critical thickness. It is well known that in heteroepitaxy near equilibrium conditions (low growth rate, high enough growth temperature), one passes from 2D growth to 3D growth when the total energy of the dots $W = W_{el} + W_s$ (elastic +surface) becomes lower than the total energy of the 2D wetting layer. As the surface is larger in dots than in 2D layers, this requires the dots to be wide enough so that the surface part in reduced energy (which corresponds to positive energy) becomes smaller than the elastic part (which is negative as the island relax their elastic energy more than the 2D layer). For having wide enough dots one needs to have deposited enough material quantity. For the first Ge layer, this occurs at typically 4 ML.[7] For the second layer, experiments[6] show that the critical thickness keeps 4 ML for h greater than 150 nm, but decreases to respectively 3.4, 2.4, and 1.4 ML for $h = 85$, 55, and 25 nm. This phenomenon can simply be explained by making use of elastic energy arguments as follows.

The reduced energy (energy per deposited atom) of the islands deposited on a uniform substrate is written as

$$W_{red} - W_{red\quad 2D} = -A_{elas} + \frac{A_s}{h} \tag{1}$$

as the elastic energy is proportional to the volume of the island, whereas the surface energies to the surface, A_{elas} and A_s, only depend on the shape of the island, which we keep unchanged in the following.

For the first Ge layer, at the critical thickness around 4 ML, a wetting layer of at least 1 ML is observed, which is due to the fact that Ge surface tension is lower than the Si surface tension. This means that, once the first wetting ML has been deposited, islands containing about 3 ML are more stable than the 3 ML high 2D layer.

Here we do not pretend to calculate the critical thickness, as the surface energy is not well known, but simply to estimate how much the strain-field interaction between the buried and the new surface dots modifies this critical thickness. For the deposition of the first island layer we have calculated the reduced elastic energy for three islands, which are truncated square-based pyramid with [101] facets, of height h, and a ratio (basis)/$h \simeq 130$. Such islands can be viewed as an intermediate situation between the 2D platelet and the much sharper stable dot [ratio (basis)/$h \simeq 7$ in Ref. 7]. The results are given in Table II. Obviously as the shape is unchanged, one gets one single value 0.959 (expressed in 2D reduced elastic energy) from which one deduces $A_{elas} = 0.041$. Assuming that $h = 7$ (or equivalently 3.28 ML in the island) corresponds roughly to the transition between the 2D and 3D growth mode, one gets an estimation of A_s. From this estimation one deduces the balance given in Eq. (1). For a positive balance the system is kept 2D and one passes to a 3D growth mode for a negative balance.

If one now considers the deposition of the second layer, Eq. (1) is no longer valid as the reduction of the elastic energy in the island (compared to the elastic energy of a 2D layer) now depends on the size and the location of the island, and also of the spacer layer thickness, as demonstrated in the first part of this report (whereas the surface energy is not

TABLE II. For a given shape of the transition island (see text), calculated elastic energies for three different volumes of this island deposited either on a uniform Si substrate ($E_{1st\quad layer}$) or on a array of buried islands ($E_{2nd\quad layer}$) with a varying spacer layer thickness h_b. The balance energy, obtained by making use of an estimation of the surface energy, indicates whether, for the corresponding Ge deposition, the growth is kept 2D (balance >0) or passes to 3D (balance <0); all energies are reduced energies and normalized to the reduced elastic energy of a biaxially strained 2D Ge layer on Si substrate.

h (ML)	5	6	7
Volume of the island (equivalent ML's)	1.29	2.06	3.28
$E_{1st\quad layer}$	0.959	0.959	0.959
Estimated reduced surface energy	0.057	0.048	0.041
Balance for the first layer deposition	>0	>0	$\simeq 0$
$E_{2nd\quad layer}/h_b$ (nm)	0.836/24	0.889/54	0.951/84
Balance for the second layer deposition	<0	<0	<0

significantly modified). Table II also displays the calculated reduced elastic energy for three islands located in their *optimal* location (that is, vertically above the buried island) for three different spacer thicknesses. The corresponding energy balance clearly establishes that, for a 54 nm (24 nm) thick spacer layer, a 3D growth mode will appear with 1 ML(2 ML's) less Ge deposit than for a spacer layer 84 nm thick. Consequently, it appears that the elastic interaction between the buried dots and the deposited islands that reduces the elastic part of the total energy of the system strongly lowers the critical thickness for the second Ge dot layer deposition, compared to the first Ge dot layer. The calculation shows qualitative agreement with the observed critical thickness reduction and its variations with the spacer thickness.

Here we have simply focused on the main mechanism, which is related to the strain distribution in the system, it is clear that atomic segregation and roughening of the growth front will also contribute to the modification of the growth process, and these phenomena warrant careful studying from a theoretical point of view. This idea goes beyond the topic of this report and will be discussed later with the authors of Ref. 6.

Here we have considered Ge dots in Si matrices, but the study applies to every semiconductor multistacked self-organized quantum dots: the mechanism does not depend on the considered materials. Only the numerical values depend mainly on the mismatch between dots and substrate and also on substrate orientation. However, only for very low mismatches and alloy materials do additional mechanisms have to be taken into account.

In order to summarize this study, two main points have been demonstrated.

(i) The surface strain distribution that comes from the buried-dot layer is not sufficient to create so-called nucleation sites for isolated atoms or small clusters, but is very efficient for organizing large enough 2D or 3D islands on the surface (for adequate spacer layer thicknesses). This efficiency is activated by the elastic interaction between the deposited island and the buried dots. In other words, the strain field induced by the surface island is the releasing point for vertical organization of quantum dots.

(ii) This elastic interaction is capable of lowering the elastic energy of 3D nucleating island so efficiently that the critical thickness can decrease by several monolayers.

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[11] The dot-induced strain distribution present at the surface can modify the hopping barriers and thus adatom diffusion (as shown recently for metals in Phys. Rev. B **55**, 6750 (1997) and also in Ref. 1 for semiconductors). Therefore, the approach we focus on in this report (based on stress interaction rather than surface mobility) is an alternative for the classical surface strain effect.
[12] Such a calculation requires the use of a more refined mesh (at the surface the meshes have to be 0.15 nm large), and a minimal size of the box is required as one regards the numerical relative precision for 1×10^{-5} of the total elastic energy. From a practical point of view, the minimization of the elastic energy has required the use of preconditioned conjugate gradient technique.
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