Ordering of Epitaxial Quantum Dots on Nanomembranes

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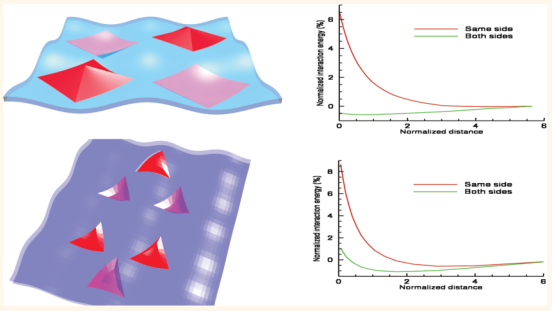
ABSTRACT



Semiconductor nanomembranes (NMs) provide fascinating opportunities to create unique structures and electronic devices owing to their mechanical flexibility. A fascinating question is whether the growth mediated by such flexibility can lead to the formation of ordered epitaxial surface nanostructures. By using computational modeling, we investigate the energetics of ordering of SiGe quantum dots (QDs) on both Si(001) and Si(111) NMs. We calculate the interaction energies for quantum dots grown on one side and on both sides of the NM and assembled in a square lattice for the Si(001) surface and in a hexagonal lattice for the Si(111) surface. Our calculations show that for QDs grown on the Si(001) NM the interaction energy possesses a minimum at a well-defined spacing only when the QDs are positioned on both sides in a square array and aligned along the [110] direction. The predicted QD ordering, spacing, and other features are in excellent agreement with recent experimental results. For QDs grown on the Si(111) NM, our calculations predict that ordered QDs can be achieved for both one-side and both-side growth, albeit with different QD spacings. The present work suggests that semiconductor NMs are a fascinating template for the self-assembled growth of ordered QDs.

KEYWORDS: flexible materials · nanomembranes · quantum dots · interaction energy · semiconductors

The quest for flexible materials, with useful electronic and optical properties, has recently attracted a great deal of attention since the introduction of semiconductor nanomembranes (sNM).[1] Because these membranes usually have thicknesses on the order of a few tens of nanometers and a lateral size of hundreds of nanometers, their small bending stiffness allows significant lateral deflection, reducing the risk of fracture.[2] Because the electronic, thermal, and optical properties of the membranes are directly related to their mechanical deformation,[3−5] these novel structures also provide an additional degree of freedom to control their properties by using strain engineering. Thus it is of great importance to understand, predict, and control the deformation of sNM, which represents a key issue of current research.

Based on sNM, a number of device applications have already been proved possible. For example, Ko *et al.*[6] have shown that the characteristics of the *I*−*V* curve of a transistor can be engineered using a nanomembrane as gate element, and the *I*−*V* curve can be tuned by choosing membranes of different thicknesses.[6] It was shown by Feng *et al.*[7] that laser illumination of the Si NM gate element induced unusual *I*−*V* curves, highlighting interesting direct applications in photovoltaics. Moreover, when an InGaAs NM is rolled up to form microcavities, this structure constitutes a laser, where the active region is an InGaAs quantum dot and the membrane acts as the photonic microcavity.[8] Last but not least, hybrid NMs made of metal/oxide multilayers have shown the capability to self-assemble into rolled nanocavities, as a highly efficient energy-storage nanocapacitor element.[9]

The use of nanomembranes to grow epitaxial quantum dots (QDs) is particularly appealing, as each quantum dot represents a local source of deformation, caused by the stress field originated from the lattice mismatch between substrate and quantum dot. In fact, epitaxial growth of silicon−germanium quantum dots was proved to be feasible on free-standing silicon NMs.[2] In such growth, both surfaces of the membrane are available for nucleation of quantum dots,[3] as opposed to the classic scenario of nucleation on a flat, semi-infinite substrate.[10]

Very recently, experiments have been carried out to investigate the ordering of

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SiGe quantum dots on Si(001) NMs.[2,11] In these experiments, growth of SiGe quantum dots was carried out using chemical vapor deposition (CVD) on a Si(001) NM with a thickness of 30 nm. Growth was stopped when the dots adopted the typical shape of {105} pyramids,[12] therefore preventing the shape transition to domes.[13] It was observed that the quantum dots tend to self-assemble in a square lattice oriented along the [110] direction, with a distance between islands comparable to the size of one island. Such self-assembled growth allows the formation of a regular, reproducible, and deterministic array of QDs on sNM, providing opportunities to create unusual optical and electronic devices.

In a pioneering study, Huang *et al.*[3] have performed two-dimensional calculations of lattice deformation for constant island size and distance, showing that the basic mechanism of island−island interaction is by superposition of the strain fields arising from each island and propagating through the NM thickness.[3] The ordering of the dots and the corresponding deformation of a Si(001) NM were studied using a three-dimensional model by Kim-Lee *et al.*[2] It was shown that ordering of QDs on NM (001) surfaces is significantly affected by the NM thickness. Since growth can also be performed on other NM surfaces, such as (111), it is fascinating to know whether ordering of QDs can occur on surfaces other than a (001) surface.

In the present work, by using finite element method (FEM) calculations, we study Ge quantum dot interaction in a Si(001) NM as a function of the distance between QDs and show why ordering is achieved when islands are allowed to nucleate on both sides of the membrane, while it is absent when islands nucleate on only one side. With the confirmation that our results are consistent with previous calculations and experimental evidence, we move our investigation to Si(111) NMs and predict the ordering occurs on both one side and both sides of such nanomembranes. Because, to the best of our knowledge, experiments on quantum dot nucleation on Si(111) NMs have not been performed yet, our calculations represent a prediction, guiding experimentalists to control the quantum dot positioning and NM deformation in an ordered fashion.

RESULTS AND DISCUSSION

We begin with the experiments of Kim-Lee *et al.*[2] and consider a Si(001) NM with a thickness of 30 nm, where SiGe quantum dots of lateral size of 70 nm, with the shape of a {105} pyramid, nucleated on both sides of the membrane. Under these conditions, ordering of quantum dots is observed along the [110] direction, forming a square lattice,[2] with the island−island spacing approximately equal to one island base size.[2] Our goal is to understand why the ordering occurs and why it occurs at such lattice spacing. To reach this goal, we

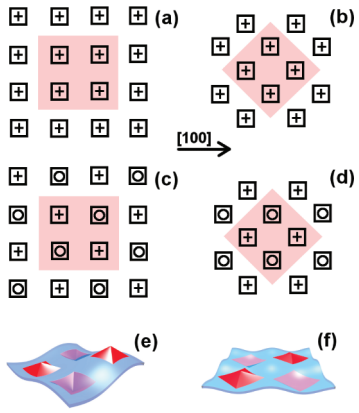

Figure 1. Schematic representation of the simulation cell considered for Si(001) nanomembranes, when islands are positioned on the same side (a, b) or on both sides (c, d) of the membrane and positioned along the [100] direction (a and c) or along the [110] direction (b and d). In each panel, the shape and size of the simulation cell is depicted in pink; the remaining islands are the images of the ones in the simulation cell as a consequence of the periodic boundary conditions applied. Each island is positioned on the side of the membrane facing the viewer or on the opposite side, depending on if it contains a circle (viewer's side) or a cross (opposite side). (e and f) Three-dimensional perspective representation of a deformed membrane with quantum dots on both sides for the [100] and [110] lattice orientations, respectively.

calculate the interaction energy between islands, arising from the superposition of the individual stress field generated by each quantum dot. Because each field propagates through the membrane, resulting in the overall membrane deformation, the calculation of the interaction energy is able to represent well the role of the sNM in allowing quantum dot interaction, as compared with the case of a thick substrate.[14]

We study the experiments in ref 2 by using the same membrane thickness and island size as in the experiments and begin by investigating the island−island interaction along the [100] direction and by placing the quantum dots in a square lattice. Within the simulation cell, islands were positioned on the membrane, either on the same side (Figure 1a) or on both sides (Figure 1b) of it. In the case of islands placed on both sides, each island was positioned such that its four first neighbors are located on the other side of the membrane. Periodic boundary conditions were applied to the lateral sides of the simulation cell, so that the actual simulation consists of an infinite array of quantum dots with a regular lattice.

Systematic FEM calculations were performed, where in each calculation the quantum dots were set at a specific spacing, and the corresponding total strain energy of the system was computed. In each FEM calculation, the island composition was considered 100% Ge for simplicity, and anisotropic elastic constants were implemented for both the Ge and Si lattice. After the total strain energy was computed, the QD spacing between dots was increased by a small amount, and another FEM calculation was performed.

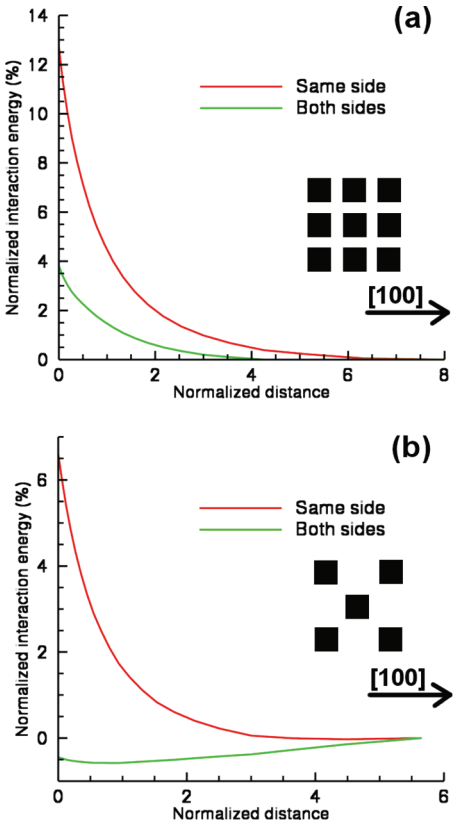

Figure 2. Plot of the interaction energy for islands positioned on the Si(001) membrane and all located on the same side (red line) or on both sides (green line) of the membrane. (a) Interaction energy for the case of islands aligned along the [100] direction. (b) Interaction energy of islands aligned along the [110] direction. The reciprocal positioning of the islands is schematically represented in the insets. The interaction energy is computed from the total strain energy of the system using eq 1 in the text.

More details on the computation of the strain energy are reported in the Methods section. When the distance between the edges of two different dots reached ~500 nm, we noticed that the total strain energy was no longer noticeably affected by a change in island distance. Therefore, we consider the total energy of this configuration as the total energy $E_{TOT}(\infty)$ when the islands are positioned infinitely apart. Finally, the interaction energy was computed with the formula

$$E_{int}(d)(\%) = \left(\frac{E_{TOT}(d)}{E_{TOT}(\infty)} - 1\right) \times 100 \qquad (1)$$

The results are compiled in Figure 2 for the case of islands on one side (Figure 2a) or on both sides (Figure 2b) of the membrane, where the island spacing was normalized to the island base size. Because the interaction energy is positive and monotonically decreasing, the island−island interaction is repulsive for the cases of islands on the same and on both sides of the membrane. Therefore, we do not expect island ordering along the [100] direction.

Next, we investigate the possibility of ordering along the [110] crystallographic direction. Following the same

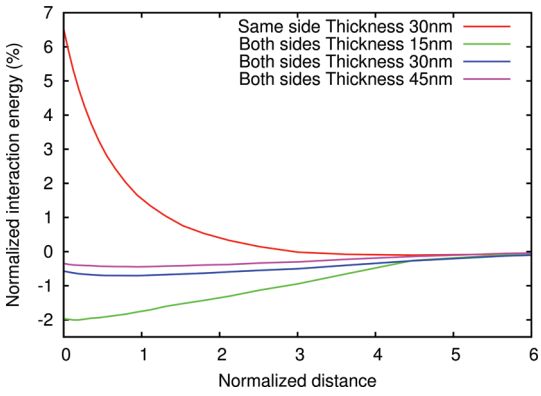

Figure 3. Behavior of the interaction energy for islands positioned on both sides of the membrane and aligned along the [110] direction, as a function of the NM thickness. For the NM with a thickness of 15 nm (green line), the interaction is attractive with a minimum at about 0.2 base size, suggesting that very thin membranes can promote island coalescence. On the other hand, the NM with a thickness of 45 nm (purple line) shows a minimum close to the one calculated for 30 nm thickness, while the reduced strength of the interaction is consistent with the fact that, with very thick membranes, the situation should approach that of a semi-infinite Si(001) substrate without ordering. The interaction energy is computed from the total strain energy of the system using eq 1 in the text.

procedure as outlined before, the interaction energy was computed by considering islands of the same size and shape and membrane of the same thickness as before. The interaction energy is reported in Figure 2b. It is seen that, while the interaction remains repulsive for islands on the same side of the membrane, it becomes attractive when islands are positioned on both sides and shows a minimum at a well-defined island−island distance. Because a minimum in the interaction energy reflects a minimum in the total elastic energy of the system, we expect that island ordering occurs in this configuration. Notice that a criterion based on the strain energy alone, as opposed to the full calculation of the free energy of the system,[15] is sufficient to explain the ordering of QDs. In fact, at volumes far from nucleation, the strain contribution to the total energy dominates over the other terms.[15] The validity of this criterion in explaining the experimental results has also been assessed for the case of SiGe QD positioning on patterned substrates.[16−18] We find the change of behavior from the [100] to the [110] direction very interesting, because it is solely due to the different reciprocal positioning of the islands across the membrane. Moreover, it is noticed that the dot ordering observed on the Si(001) NM is in contrast to the growth of QDs on a flat thick Si(001) substrate, in which dot ordering was not observed.[19,20]

Our calculations and the experiments in ref 2 show agreement that is not only qualitative but also quantitative. In fact, the calculated minimum in Figure 2b occurs at ~0.75 times the pyramid's base size, which is in good agreement with the experimental spacing reported in refs 2 and 11. In the present calculations, the NM and

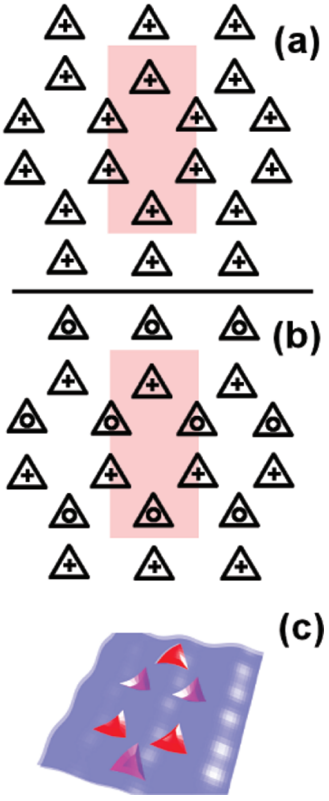

Figure 4. Top-view representation of the positioning of triangular-shaped islands in a hexagonal lattice, for the case of islands positioned on one side of the membrane (a) or on both sides (b). A circle inside the island indicates that the island is placed on the side of the membrane facing the viewer, while a cross indicates that the island is located on the opposite surface of the membrane. The actual simulation cell is depicted in pink; the remaining islands are the result of the periodic boundary conditions applied to the simulation cell. (c) Representation of the deformed membrane in perspective view for the case of islands positioned on both sides of the membrane. The picture shows a region slightly bigger than the actual simulation cell for illustrative purposes.

islands follow a linear elastic relation. Therefore, if the system geometries are changed in proportion, the relation between the normalized interaction energy and the normalized island distance should remain unchanged. However, if the system geometries are not changed in proportion, for example, the island size and shape are fixed while the NM thickness is varied, the change in MN thickness will affect the island assembly. To further understand the effect of NM thickness, we performed more calculations by varying the NM thickness and leaving all other parameters unchanged. Results for the three thicknesses, that is, 15, 30, and 45 nm, are shown in Figure 3. It is seen that there is a strong island interaction for the 15 nm NM. However, since the energy minimum occurs at a distance of approximately 0.2 unit base length, a thin NM could lead to island coalescence, which might result in a less dense packing array of larger islands. On the other hand, the interaction for the 45 nm NM exhibits a shallow minimum approximately at the same island distance as that computed for the 30 nm NM. This

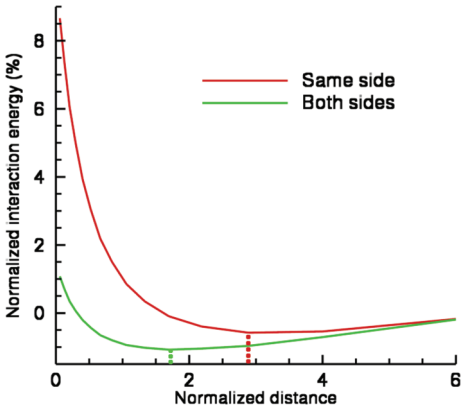

Figure 5. Plot of the interaction energy for the case of triangular-shaped islands positioned on a Si(111) nanomembrane. The two plots correspond to the case of islands located on the same side (red line) and on both sides (green line) of the nanomembrane, respectively. For each configuration, the position of the minimum is highlighted with a dashed line.

indicates that the island array for the 45 nm NM should have similar characteristics to that of the 30 nm NM, but with a reduced driving force for ordering. It is expected that at a very large thickness the island assembly will approach that of a semi-infinite substrate, without ordering of islands.[19,20] Following the above-mentioned agreement between calculations and experiments, we decided to move on and focus our attention on Si(111) NM, where the exposed surfaces have the (111) orientation.

In this case, we consider islands with a triangular base and flat top, typical of the Ge/Si(111) heteroepitaxial system.[21,22] We keep the NM thickness constant at 30 nm and use an island base length of 70 nm, consistent with the previous case. Because the island shape is triangular, we drop the consideration of a square lattice, in favor of a hexagonal lattice as shown in Figure 4. This is because the hexagonal lattice, among the five possible Bravais lattices in two dimensions, is the only one that allows each lattice unit to have three nearest neighbors, consistent with the number of sides of the quantum dot shape.[23] Moreover, it allows the positioning of islands on one side (Figure 4a) and both sides (Figure 4b and c) of the membrane in an ordered fashion, such that each island always has three nearest neighbors.

Calculations were performed using the same methodology as described above, but taking into account the different orientation of the anisotropic elasticity constants, in such a way that the (111) direction is now normal to the NM surfaces. Results are compiled in Figure 5 for both cases, where the islands are positioned on the same side or on both sides of the NM. The figure shows that the interaction energy has a minimum for islands positioned on both sides. Therefore, island ordering is predicted in such configuration, similar to the case of Si(001) NM, although with a different lattice spacing, that is, ~1.75 units of base size (Figure 5 green curve). Surprisingly, however, even

for the case of islands on the same side, the interaction energy is not always repulsive, but also shows a minimum. This minimum falls at ~2.4 times the island base size. Therefore, our calculations suggest that ordering in a hexagonal lattice is possible for islands both on the same side and on both sides of the NM, although with two different characteristic spacings. We interpret the ordering of the islands on the same side of the Si(111) NM as a consequence of the triangular island shape and hexagonal lattice. In fact, for a hexagonal island lattice with a triangular island base, each vertex of an island faces an edge of a neighboring island. While the edge's deformation is mainly along the edge direction, the deformation of the vertex is mainly outward toward the island base. These two deformations mitigate each other, giving rise to the minimum energy observed here. It is noted that to a certain extent the ordering of Ge quantum dots on the (111) surface of a bulk Si substrate was also observed.[21,24] Furthermore, we notice that while growth on opposite sides leads to a closer packing array of islands, growth on one side only allows for a more "open" island lattice. This finding might have interesting consequences in terms of tuning the optimal island spacing, and therefore NM electronic properties, for device applications.

CONCLUSIONS

In conclusion, we have investigated the energetics of Si(001) and Si(111) nanomembranes when Ge quantum dots are positioned on their surfaces and studied the optimal ordering that quantum dots can achieve on the surfaces. For the Si(001) NM, by performing FEM calculations using quantum dots positioned on one side of the NM and on both sides, we find that the interaction energy shows a minimum for islands on both sides of the membrane and aligned along the [110] direction, but does not show a minimum in other configurations; we expect ordering to occur only in this configuration. This ordering and other features are consistent with the experimental evidence. Gaining support from this comparison, we next studied the Si(111) NM and considered quantum dots of triangular shape positioned in a hexagonal lattice. For this case, our FEM calculations of interaction energy have shown that ordering is expected not only for islands on both sides on the NM but also for islands placed only on one side of the NM, albeit with a different spacing. Therefore, our present work suggests that semiconductor nanomembranes can be highly attractive for the positioning of quantum dots in an ordered fashion. Because an ordered array of quantum dots corresponds to a regularly deformed membrane, the local electronic, thermal, and optical properties of the membrane will correspondingly show a regular pattern, with promising implications for the use of these semiconducting flexible materials in next-generation nanoelectronic and optoelectronic devices.

METHODS

The calculations of the total strain energy of the system, as a function of the distance between quantum dots, were performed in the following way. The simulation cell, including the membrane and the quantum dots, was created using the finite elements preprocessor software Patran. The same software was later used to create the finite elements mesh for both the membrane and the quantum dots. Accuracy tests were performed to ensure that the mesh size was sufficiently refined to avoid numerical errors. After the preprocessing, the elasticity problem was solved using the software Abaqus Standard. The periodic boundary conditions, allowing to replicate the simulation cell in the in-plane directions, were implemented in the form of equations on the displacements on the nodes, such that the displacement at a boundary node was set to equal the displacement of the node on the corresponding opposite lateral side of the membrane. The lattice mismatch between Ge and Si was implemented by applying an initial stress to the Ge lattice, such that the Ge lattice is initially compressed at the lattice parameter of Si. After the strain was computed, the corresponding elastic energy was extracted using the built-in function of Abaqus CAE. Because the calculation of the elastic energy was needed for each value of the distance between quantum dots, the steps described above were enclosed into a loop, where at each step the quantum dot distance was set. Therefore, each step in the loop corresponded to a Patran call to create the geometry, an Abaqus call to solve the elasticity problem, and an Abaqus call to calculate the total strain energy. Because many different configurations were considered (including membrane orientation, quantum dot positioning, quantum dot shape), one loop was designed per each configuration.

Conflict of Interest: The authors declare no competing financial interest.

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