Modelling Ge/Si quantum dots using finite element analysis and atomistic simulation

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Abstract. Finite Element Analysis (FEA) and atomistic simulations are used to model Ge/Si quantum dots. The three dimensional non-uniform composition profile in Ge(Si)/Si(001) quantum dots is calculated using atomistic modelling. The results are compared to experimental data from the literature. FEA is used to model the contact angle dependence of the strain energy of the QD. An equation is fitted to the modelled dataset describing the strain energy of a uniformly alloyed, pyramid shaped Ge/Si quantum dot as a function of the contact angle.

1. Introduction

During the layered growth of lattice-mismatched semiconductors, a strain-induced transition from planar growth to 3D island growth (Stranski-Krastanow growth [1]) can occur, leading to the formation of quantum dots (QDs). Their small size (nm in dimension) leads to quantum confinement of the electrons, resulting in electronic and optical properties which show promise for use in a wide range of devices ranging from semiconductor lasers to quantum computers. These properties of the QDs are controlled by their size, shape and composition. The growth of Ge(Si) on Si(001) has been extensively studied because of its importance as a model system for the growth of other, more complicated, lattice mismatched semiconductors (e.g. [2]).

Whereas the shape and size of Ge(Si)/Si(001) QDs can be determined from AFM [3] and TEM [4] to a high precision, an accurate measurement of the composition and composition distribution remains unsolved. Measurements show a significant non-uniformity in the composition profile [4] but although several details of the composition profile have been resolved, a coherent model of the alloying profile, taking into account all different driving forces, remains elusive.

For a complete understanding of the QD formation process it is necessary to take into account the non-uniformity of the composition profile. In this paper an atomistic modeling approach is presented that accounts for the non-uniformity of the composition profile. The atomistic model forms the basis for an analysis of the energetics of the QD.

For large QDs or assemblies of QDs an atomistic treatment may not be feasible because of the large number of atoms involved in such systems. Therefore finite element analysis (FEA) is outlined as an alternative method to model the QD energetics.
2. Atomistic modelling of the QD alloying

To obtain an accurate representation of the energetics of a non-uniformly alloyed QD, the atomistic model has to resemble the shape, size and non-uniform composition profile of a real QD as closely as possible. In the case of Ge/Si quantum dots, there are two distinct shapes the QD can assume which are well defined in terms of the crystallographic planes which they contain [2].

Initially, the QDs grow having a pyramid shape with facets along {105} crystallographic planes. At a certain critical size, islands transform to a more complicated domes shape by incorporating steeper facets. We have previously published the results of atomistic modelling for pyramid shaped QDs [5]. In this paper we focus on the modelling of dome shaped QDs.

The size of coherent Ge/Si islands (i.e. not containing dislocations) has been found to vary strongly depending on the average composition of the island. Coherent islands with diameters between 20nm and 200nm are possible. For their use as QDs, smaller islands are preferred. In this work the size of the QDs studied in the atomistic simulations was limited by the computational resources available. Therefore the atomistic models studied here have a diameter between 8-16nm. It is however possible to extrapolate the results obtained from small QDs to larger QDs found experimentally (cf. Figure 1).

The aim in this work is to calculate the quasi-equilibrium composition profile in Ge$_x$Si$_{1-x}$ (with $x$ representing the Ge content) QDs. It is important to point out that the present simulation does not account for kinetic effects on the composition profile such as the anisotropic surface diffusion due to surface reconstruction and the difference in the diffusion lengths of Ge and Si, effects which may lead to experimental results which deviate from the composition profile presented here. However, these deviations from the quasi-equilibrium composition profile are expected to be small for sufficiently high growth temperatures and slow growth rates.

In the calculations, a uniform starting composition profile is assumed and subsequently the energy of the system is minimized computationally. The system sizes and time-scales involved in the formation of the composition profile are too large to employ classical molecular dynamics or ab-initio modelling; therefore the approach used here is based on a combination of atomistic total energy calculations and a Metropolis Monte – Carlo process which provides the mechanism for the reorganization of the alloy within the QD. The starting composition in the QD is assumed to be a homogeneous alloy. This assumption implies that the modelling either is simulating a system in which the QD grows with a composition as deposited, and then undergoes diffusion/segregation post-growth, or is simply a starting model as a basis for energy minimization.

![Figure 1](image1.png)  
**Figure 1.** The composition profile obtained from our modelling for a Ge$_{0.66}$Si$_{0.33}$ QD is scaled up and compared to an experimental measurement [4].

![Figure 2](image2.png)  
**Figure 2.** A quarter of a Ge$_{0.5}$Si$_{0.5}$ QD is shown (a) from the top, (b) from the side and (c) from the bottom. The local composition is shown.

To obtain this quasi-equilibrium profile, the energy of the system is minimized in two steps: (i) while maintaining the homogeneous composition distribution of the QD, and the pure Si of the substrate, the top two atom layers of the substrate and the total contents of the island are relaxed using a conjugate gradient type algorithm implemented in the OXON package [6]. The interaction between
the atoms is described by the Tersoff potential [7], and periodic boundary conditions are applied to the substrate. (ii) following this relaxation, diffusion of atoms within the QD is allowed. Diffusion of atoms into the substrate is not allowed because this would result in the QD dissolving and Ge being dispersed in the Si bulk forming a dilute alloy. In experimental growth this does not occur because of kinetic limitations. To model the diffusion, an atom is randomly chosen and the atoms surrounding it, within a cut-off radius of 0.48 nm, are identified. One of these surrounding atoms, with a label different from the first atom, is then chosen at random, and the labels of this atom and the first atom are exchanged. The energy of the new configuration is calculated, and a Metropolis Monte-Carlo process is employed to decide which configuration to accept. The temperature for the Monte Carlo process was set at 900K, a typical growth temperature for GeSi/Si(001). This process is continued to convergence: it was found that, for the system described, the process converges after about 3*N steps where N corresponds to the number of atoms in the QD.

Figure 1 shows the resulting composition profile for a dome shaped Ge_{0.66}Si_{0.33} QD, averaged on a plane cutting through the centre of the QD. Good agreement is found with experimental results by Liao et al [4]. Figure 2 shows the composition profile of a Ge_{0.66}Si_{0.33} QD in three dimensions. This reveals that there is significant detail hidden in the composition profile which is not revealed by 1D or 2D measurements which average the composition over one direction. The modelling shows for example a significant enrichment of the surface with Ge and a non-uniform enrichment of Si at the QD-substrate interface, both of which cannot be deduced from the data shown in Figure 1.

3. The strain energy as a function of the contact angle using finite element analysis

Finite element analysis (FEA) has previously been used to calculate the strain field of dome shaped QDs and to perform TEM image simulations that correspond well to experimental results [8]. The commercial FEA package STRAND7 was used for the calculations in this work. STRAND7 allows the user to define a thermal expansion coefficient $\alpha$ for each material. Furthermore the temperature can be defined at each node, which determines the expansion in combination with $\alpha$. This can be used to introduce a strain term into the FEA, which is equivalent to the strain due to the lattice mismatch between Si and Ge in the atomistic simulations into the FEA. To clarify this, two examples are given:

(i) A pure Ge QD on a Si substrate: The lattice mismatch between Ge and Si is 4% and therefore $\alpha$ is set to be 0.04 for Ge and 0 for Si. Because the material in the QD is pure Ge QD, all node ‘temperatures’ are set to 1.

(ii) A non-uniform composition profile: To transfer the composition profile from the atomistic simulations the local Ge content near each node is mapped to node ‘temperature’ in an interval between 0 and 1 with 0 being pure Si and 1 being pure Ge. This number is equivalent to the node temperatures in the FEA.

The finite element software calculates the strain energy density at each brick. This value can be plotted and represents the strain energy distribution. In this work we assumed a pyramid shaped island and studied the strain energy density as a function of the contact angle. It is well known that the average strain energy density decreases of an island decreases with steeper contact angles. Balanced against an increase of the surface energy, this leads to the shape transformation from shallow facets to steeper facets during the growth of QDs [2]. The trend of decreasing strain energy density with increasing contact angle is reproduced well by the finite element calculations (cf. Figure 3) for pure Ge islands on a Si substrate. We have fitted an equation to the graph in Figure 3:

$$E_{\text{strain}} = K \cdot \alpha^{-0.85}$$  \hspace{1cm} (1)

where K is $4\times10^7$ and the angle $\alpha$ is measured in radian. In the case of uniformly alloyed islands with a Ge content of c, we have shown previously [11], that energy of the alloyed island ($E_{\text{alloyed}}$) is given by

$$E_{\text{alloyed}} = E_{\text{Ge}} \cdot c^2$$  \hspace{1cm} (2)

where $E_{\text{Ge}}$ is the energy of a pure Ge island of the same shape and size. Therefore, for a pyramid shaped, uniformly alloyed, Ge/Si island the strain energy is given by:

$$E_{\text{strain}} = K \cdot \alpha^{-0.85} \cdot c^2$$  \hspace{1cm} (3)
4. Summary and future work

Atomistic simulations have been performed to calculate the non-uniform alloying profile in dome shaped Ge/Si QDs. The composition profile was shown to be rather complex and not a simple linear distribution as sometimes assumed in the literature. FEA was employed as a useful tool to study strain effects in uniformly alloyed QDs. The strain energy of uniformly alloyed pyramid shaped QDs was studied as a function of the contact angle. An equation for the strain energy of a uniformly alloyed pyramid shaped QD was derived.

During growth, an island will arrange its facets so as to minimize its total energy, which is given by the sum of the strain energy and the surface energy. In most relevant semiconductor systems, there are only a few crystallographic planes which occur as facets of QDs. Using equation (3), the composition of a quantum dot could be determined from the measurement of the facet lengths and their crystallographic orientations, if the surface energy on all these planes was known accurately. This would be valuable to the experimentalist because, while the shape of a QD can be determined fairly routinely using e.g. scanning tunneling microscopy, it is notoriously difficult to determine the average composition of an island. We therefore suggest that further efforts are required to determine possible surface reconstructions and surface energies of surfaces occurring as facets in QDs.

References
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Figure 3. The strain energy density of a pure Ge, pyramid shaped QD shown as a function of the contact angle between the facet and the substrate.