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Reasons for self ordering in multilayer quantum dots Part II: Interaction energy

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ABSTRACT

Two ways to calculate the elastic interaction between quantum dots (QDs) in the framework of linear elasticity are introduced and shown to vary in a similar way as the hydrostatic pressure. It is shown that the hydrostatic stress is the potential for the elastic interaction energy. The approach was used to estimate quantitatively the interaction energy between QDs in material systems that may form vertical order, anticorrelated order and FCC superlattice. The vertical interaction energy is very small compared with the thermal energy, nevertheless it is just enough to induce vertical ordering of QDs between layers. The attractive *lateral* interaction is an order of magnitude smaller than the vertical interaction, therefore ordering of multilatered QDs is made possible only by the *vertical* interactions. Several experimental observations are explained based on this understanding. The *lateral* interactions are small due to the small stresses set in the substrate after relief of the misfit strains in the free space. The *vertical* interactions are larger due to the large tensile stresses set up in the thin layer of matrix that separates an embedded QD from the free surface.

1. Introduction

Growth of quantum dots (QDs) in multilayered semiconductors is observed to yield different types of arrays ranging from random arrays, vertically stacked arrays and vertically staggered arrays. The last arrays may be laterally random or ordered in FCC-like superlattices. Ordering has been shown to be the result of substrate mediated elastic interactions with dots in the underlying layer. The elastic fields associated with QDs have been analyzed by many researchers who used different methods [1–4]: analytic continuum approach [e.g., 5–12], numeric continuum approach (finite difference [e.g., 13,14] or finite element [e.g., 15–17] methods) and atomistic approach [e.g., 18–21]. In the analytical continuum approach, the QDs are treated as inclusions in a matrix. The elastic fields due to the lattice mismatch between the QDs and the matrix are obtained by integrating the Green's function over the interface of the inclusions and the matrix. Only limited geometries are solvable and homogeneity of the elastic constants is usually assumed. The finite difference and finite element approaches overcome these limitations. The atomistic approach describes the strain energy in terms of the potentials between the atoms and the strain fields are obtained by minimizing the potential energy. Due to the large number of atoms required in the analysis, this approach is computationally limited to a single QD. Comparison between the approaches shows that the continuum approaches reasonably approximate the atomistic calculations [18].

Following Tersoff et al. [5], Holy et al. [4,7] and other authors considered a three layer situation consisting of a QD layer, a cupping layer, made of the material of the substrate and a wetting layer, made of the material of the dots. They calculated the density of the elastic energy in the wetting layer on the free surface and proposed that the favored sites for nucleation and growth of new dots in the Stransky Krastanov mode are those where the elastic energy density is minimal. Ignoring the lateral interactions they found that when the elastic anisotropy factor $A \equiv 2C_{44}/(C_{11}–C_{12}) > 1.6$ (C_{ij} are the elastic constants) and the substrate surface is the (001) crystallographic plane, an embedded dot creates favored sites that are expected to form a body centered tetragonal (BCT) superlattice. When $A < 0.6$ and the substrate surface is the (111) plane, trigonal or FCC-like superlattices are expected. In the other cases vertical stacking without lateral order is expected. A remarkable resemblance between the predictions and the experimental observations is found in different materials, except for the BCT superlattice that was not observed [1–4].

Several groups have calculated the elastic energy density in the wetting layer [1,4,5] and in the cupping layer [13–16]. Apparently only Liu et al. [15] admitted that the preferred sites for nucleation of new dots are those where the density of elastic energy in the *cupping layer* is *maximal* and those sites coincide with sites of *minimum* elastic energy density in the *wetting layer*. This difference can be explained in a straightforward way by the considerations in ref. [22]. A buried dot with lattice parameter larger than that of the substrate, generates tensile stresses in the cupping layer, made of the substrate material. The larger the tensile stresses in the cupping layer — the larger the density of the elastic energy there. On the other hand, on top of the

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cupping layer rests the wetting layer, made of the dot material. It is severely compressed in the lateral directions, due to the epitaxial relation with the substrate. Thus the minimum elastic energy density in the wetting layer is found when it is relatively *relaxed* from the compressive epitaxial stresses by the tensile stresses due to the underlying QD. These sites can be expected to be favored for nucleation of new QDs and it was indeed observed. While these are qualitative arguments, the actual energy gain of the new dot at a favored site should be *directly* calculated. This gain is defined as the interaction energy between the dots [23].

In the first part of the present work [22] we discussed qualitatively the elastic interactions among dots in multilayers. In the present part we rigorously prove two ways to calculate the elastic interactions in the framework of linear elasticity. From them we deduce a third simple estimate of the elastic interaction, obtainable from the solution of the elastic fields of a single dot and show that it is a better estimate than the elastic energy density in the cupping or the wetting layers. The three methods are applied and assessed numerically, to quantitatively determine the interaction energy in two types of systems where ordered arrays are observed: simple vertical stacking and vertically staggered stacking. Then the failure to form the expected BCT superlattice (in A > 1 materials with {001} free surface) is quantitatively compared with the FCC superlattice (in A < 1 materials with {111} free surface), which is experimentally observed. Finally the lateral interaction between dots in the same layer is calculated in order to quantitatively determine its role in the ordering of the QDs.

2. Theory of elastic interactions

2.1. The elastic interaction energy

We follow the derivation of Eshelby in refs. [23,24] of the interaction energy between two stress sources in infinite solid and adopt it to a solid with free surfaces. Suppose that in a body with volume V_o, enclosed by external surface Σ_o there are two systems of internal stresses that are due to misfit strains ε_{ij}^{T}: A whose sources lie entirely within a surface Σ_A and B whose sources lie entirely outside Σ_A (Fig. 1). Both may be embedded inside the body or attached to its surface.

Let E_A and E_B denote the total elastic energy when A or B exists alone in the body. Then the total energy when they exist together is, within the framework of linear elasticity:

$$E_{tot} = \frac{1}{2}\int_{Vo} \sigma_{ij}\varepsilon_{ij} dV = \frac{1}{2}\int_{Vo}\left(\sigma_{ij}^{A}+\sigma_{ij}^{B}\right)\left(\varepsilon_{ij}^{A}+\varepsilon_{ij}^{B}\right)dV. \tag{1}$$

Eq. (1) can be written as:

$$E_{tot} = E_A + E_B + E_{Int} \tag{2}$$

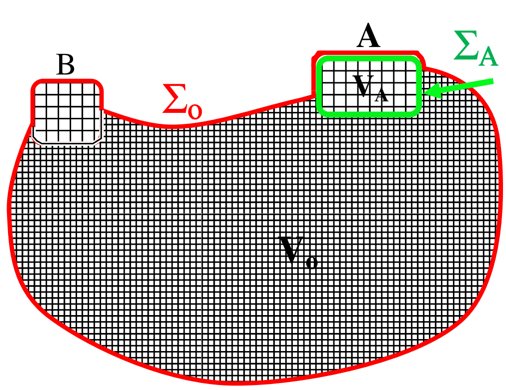

Fig. 1. A schematic illustration of a matrix and two stress sources embedded in or attached to the surface of the matrix.

where,

$$E_{int} = \frac{1}{2}\int_{Vo}\left(\sigma_{ij}^{A}\varepsilon_{ij}^{B}+\sigma_{ij}^{B}\varepsilon_{ij}^{A}\right)dV \tag{3}$$

is the interaction energy between the stress sources A and B. σ_{ij}^{k} and ε_{ij}^{k} are respectively the elastic stresses and strains that are due to source k alone. Now define $u_i(r)$, the components of the displacement of each point r in the material, relative to its position before the misfit appeared between regions A and B and the matrix. The symmetrized matrix of the derivatives of the displacements due to A: $\frac{1}{2}\left(u_{i,j}^{A}+u_{j,i}^{A}\right)$ (the subscript, j indicates derivative with respect to x_j and repeated indices are summed over x, y and z) is the elastic strain tensor ε_{ij}^{A}, everywhere except in the region A. In the region A the elastic strains are measured relative to *the stress free state of the material A*. Namely, the elastic strains *due to* A are:

$$\varepsilon_{ij}^{el,A}(r) = \begin{cases} \frac{1}{2}\left(u_{i,j}^{A}-u_{j,i}^{A}\right) & \textit{outside } \Sigma_A \\ \frac{1}{2}\left(u_{i,j}^{A}-u_{j,i}^{A}\right)-\varepsilon_{ij}^{T,A} & \textit{inside } \Sigma_A \end{cases} \tag{4}$$

In a similar way, the elastic strains *due to* B are:

$$\varepsilon_{ij}^{el,B}(r) = \begin{cases} \frac{1}{2}\left(u_{i,j}^{B}-u_{j,i}^{B}\right) & \textit{outside B, in particular inside } \Sigma_A \\ \frac{1}{2}\left(u_{i,j}^{B}-u_{j,i}^{B}\right)-\varepsilon_{ij}^{T,A} & \textit{inside B} \end{cases} \tag{5}$$

The strains measured relative to the stress free state of the matrix:

$$\varepsilon_{kl}^{C,k} = \frac{1}{2}\left(u_{i,j}^{k}-u_{j,i}^{k}\right) \tag{6}$$

were termed by Eshelby the constraint strain.

Returning to Eq. (3), one finds that the two terms in the brackets are equal by the reciprocity theorem ($\sigma_{ij}^{A}\varepsilon_{ij}^{B}=C_{ijkl}\varepsilon_{kl}^{A}\varepsilon_{ij}^{B}=C_{klij}\varepsilon_{kl}^{A}\varepsilon_{ij}^{B}=\sigma_{kl}^{B}\varepsilon_{kl}^{A}$ due to Hooke's law and the symmetry of the elastic constants tensor).

Eshelby divided the integral in Eq. (3) into an integral inside Σ_A and an integral outside Σ_A. Each integral was written only in terms of the constraint stains (derivatives of $u_i(r)$):

$$E_{int} = \int_{inside\ \Sigma A}\sigma_{ij}^{A}u_{i,j}^{B}dV + \int_{outside\ \Sigma A}\sigma_{ij}^{B}u_{i,j}^{A}dV \tag{7}$$

To express the first term he wrote the sum of the derivatives of $\sigma_{ij}u_i$ and applied the equilibrium equations $\sigma_{ij,j}=0$:

$$\left(\sigma_{ij}u_i\right)_{,j} = \sigma_{ij,j}u_i + \sigma_{ij}u_{i,j} = \sigma_{ij}u_{i,j} \tag{8}$$

Application of Gauss's theorem on Eq. (8) converts the first term in Eq. (7) into $\int_{\Sigma A}\sigma_{ij}^{A}u_i^{B}dS_j$. Similarly the second term becomes: $\int_{\Sigma o}\sigma_{ij}^{B}u_i^{A}dS_j-\int_{\Sigma A}\sigma_{ij}^{B}u_i^{A}dS_j$, the minus sign in the second integral is due to n_j being oriented outward normal to Σ_A. The first integral over Σ_o vanishes due to the boundary conditions on the free surface Σ_o: $\sigma_{ij}^{B}n_j=0$. Combining these results into Eq. (7) one is left with two terms:

$$E_{int} = \int_{\Sigma A}\left(\sigma_{ij}^{A}u_i^{B}-\sigma_{ij}^{B}u_i^{A}\right)dS_j \tag{9}$$

The surface Σ_A can now be taken as a surface enclosing an island A. Thus Eq. (9) expresses the interaction energy between A and B in the form of an integral over a surface surrounding only *one* of the systems of the internal stresses.