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Reasoning of the self ordering in multilayer quantum dots Part I: Elastic fields

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ABSTRACT

The present work aims to *explain the reasons for* the elastic interactions between quantum dots (QDs) that give rise to their self ordering in different materials with cubic symmetry. This is done by a detailed consideration of the components of the elastic displacements, strains and stresses associated with a dilating QD on a substrate. The interaction between QDs is a result of the anisotropic response of the substrate that is due both to material and geometrical anisotropy. The elastic anisotropy is revealed by a distinction between the soft directions along which the strains are large and the displacements have a short range and the hard directions, along which the strains are small and displacements have a long range. It is shown that the elastic fields and the attractive interactions are dominated by the long range displacements. If a hard direction is normal to the free surface, a single hill-shaped protrusion is formed, which stretches the surface on top of a buried dot and gives rise to favored vertical stacking of QDs. If the normal to the free surface is not a hard direction, a square or a triangular protrusion is formed on the free surface where the hard directions intersect the surface. The formation of favored sites with a square or triangular symmetry gives rise to a staggered vertical stacking and, when the dilatation in these sites is intense enough, may induce lateral ordering of QDs into superlattices. As an outcome of the discussion, a criterion is suggested for the elastic properties of a substrate that can induce the formation of a superlattice.

1. Introduction

The Stranski–Krastanov growth mode is an established way to obtain self ordered arrays of quantum dots (QDs) by successive deposition of layers of the dot and the substrate materials. Due to their nanometric size, QDs exhibit a quantum confinement effect on charge carriers. Due to their ordering, tunneling interaction is possible between the charge carriers in the dots. A combination of these properties makes ordered QDs of much interest for basic science and for electro-optic applications [e.g., 1]. Their pioneering work to determine the experimental conditions and the theoretical fundamentals of the ordering phenomenon was reviewed by Legally and his collaborators [2,3] and Holy and Springholz and their collaborators [4–6]. They found a striking conformance between the resulting locations of elastic energy minima in the wetting layer and the experimentally observed preferred sites in the multilayered QDs. This preference gives rise to vertical and lateral ordering and also to symmetry transitions that depend on the spacer thickness. They emphasized the role of material anisotropy in determining the degree and the symmetry of the ordering. Their work proved that the self-ordering phenomenon can be attributed to substrate-mediated elastic interaction among the QDs. In the next sections the main experimental observations are summarized and compared with results of the calculations of the energy density [2–6].

1.1. Comparison of experimental and calculation results

1.1.1. Vertical correlation

Simple vertical stacking, staggered stacking (or trigonal, face centered cubic-like stacking) of QDs have been observed in different multilayer systems of materials with cubic symmetry. These symmetries depend on the material anisotropy and the crystallographic orientation of the free surface in a systematic way that is summarized in Table 1 and Fig. 1. The anisotropy of materials with cubic symmetry is characterized by the anisotropy factor $A = 2C_{44}/(C_{11} - C_{12})$ where C_{ij} are the elastic moduli. Most metals with a cubic structure, elements from the IV column and semiconducting II–VI compounds (like CdSe) with diamond or zinc blend structures possess $A > 1$. In these materials the elastic constant C_{11} is minimum along <100> directions (soft directions) and maximum along <111> directions (hard directions) (Fig. 3ab). Metals from the V and VI columns and semiconducting III–V compounds (like GaAs) with a rock salt crystal structure possess $A < 1$. In these materials the elastic constant C_{11} is maximum along <100> directions and minimum

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Table 1
Summary of the observed QD correlations. The type of observed correlation depends on material anisotropy and the crystallographic orientation of the free surface. When the stacking order changes due to increasing layer spacing, the observed correlation types are written in an order of increasing layer spacing. The structure observed to form a superlattice is marked by bold letters, the structure only expected to form a superlattice is marked by shadowed letters. Abbreviations: VS—vertical stacking-simple tetragonal, BCT—body centered tetragonal, FCC—face centered cubic (trigonal), R—random.

	$A<1$	$A\sim1$	$A>1$
{100}	VS/R	VS/R	VS/𝔹ℂ𝕋/R
{111}	VS/**FCC**/R	VS/R	VS/R

along <111> directions. In both cases the <110> directions are saddle points.

When the surface normal is an elastically hard direction the QDs are found to grow in vertical stacking. This coincides with the theoretically expected sites of minimum elastic energy that is found vertically above the buried QDs [2–6].

When the surface normal is an elastically soft direction, a splitting of the energy minima in the wetting layer is found in the calculations [7]. On {111} surfaces of materials with $A<1$, like PbSe/PbEuTe, the energy minimum is split into three minima, thus in a superlattice an *ABCABC* dot stacking is formed (Fig. 1c). Each layer consists of a 2D hexagonally ordered dots separated by the spacer layers. The overall dot arrangement is similar to the atoms stacking in face-centered cubic lattices except that the ratio between the lateral dot spacing and the vertical spacer thickness is variable, thus the whole dot arrangement resembles in general to a trigonal superlattice of dots [8–10]. On {100} surfaces of materials with $A>1$ the energy minimum is found by calculations to split into four energy minima above each buried dot, thus an *ABAB* body centered tetragonal stacking is expected (Fig. 1b). However, only an anticorrelated interlayer dot arrangement has been found in II–VI and III–V compounds like CdSe/ZnSe [11–13], CdTe/ZnTe [14], InGaAs/GaAs [15] (and also in InAs /AlInAs quantum wire superlattices on InP [16]). In Ge/Si [17–19] and InAs/GaAs [20,21] systems, mostly a nearly perfect vertical dot alignment in columns has been observed for small spacer thickness (Fig. 1a). This is attributed to their smaller elastic anisotropy.

The angle α between the surface normal and the orientation of the energy minima varies systematically with the anisotropy. For materials with $A<1$ on {111} planes, the splitting occurs for $A<0.6$, and below this value α varies linearly with the reciprocal value of A [5]. For materials with $A>1$ on {100} surfaces the splitting of the energy minima occurs if the anisotropy exceeds a critical value of $A>1.4$, and beyond this value α varies linearly with A. The observed angles vary between 16° to 41° and lies within 3° from the prediction for $A<1$ materials and within 12° from the prediction for $A>1$ materials. The energy minima gradually fade away when the surface normal is tilted from the soft direction [6,9,10].

1.1.2. Lateral correlation and size transitions

In materials with $A<1$ like the PbSe/PbEuTe system with {111} free surface, the QDs grow first with vertical order. With increasing spacer thickness they show better order and they grow larger in size [19]. In the intermediate spacer thickness range, between 40 and 55 nm, a transition to self-organized *fcc*-like 3D lattices of dots with tunable lattice constant is observed [8,9]. The dots are intermediate in size and the in-plane dot separation scales linearly with the spacer thickness [8–10,22–25]. With larger spacer thickness the dots are randomly distributed in the lateral (as well as in the vertical) directions and are very small [9,10,21]. This has been attributed without details, to the small size of the dots and to the changes in the strain fields from the near-field to the far field behavior [6]. With increasing number of deposited layers the size uniformity and lateral correlation improve, and the thickness of a wetting layer for initiation of island formation decreases [9,10,19,21].

For materials with $A>1$, a similar transition from a vertical dot alignment to an anticorrelated stacking was predicted with increasing spacer thickness. This prediction is found to be in good agreement with the experimental observations [13,24]. However, the tendency for lateral ordering of the surface islands is weak and the expected body centered tetragonal dot lattice is not observed, only an anticorrelated dot stacking. Also the experimentally derived interlayer correlation angles of these structures are found to be larger than those theoretically expected [10–13]. This failure to form a superlattice is explained by the calculation of the elastic energy density by the separation of the four side minima only by a weak saddle point. The so formed sites for new dots, are not sharply favored and a 3D lattice cannot form. In an interesting analogy, ordered QDs of Ge were grown on Si induced by the stress fields of dislocations [26].

1.2. The theoretical knowledge

Three types of problems in elasticity are of interest to understand the ordering of QDS: (a) a second phase particle in an infinite matrix, (b) a particle embedded below a free surface and (c) a particle on a free surface. The situation in which the uniformity of an infinite elastic medium is disturbed by a region within which the lattice parameters or elastic constants have changed attracted interest since the early 1900s. On 1957 Eshelby suggested a general solution for both problems by using Green functions to represent a distribution of tractions on the interface between the matrix and the second phase [27]. His approach has been extended to anisotropic materials by Mura [28] and Khachaturyan [29]

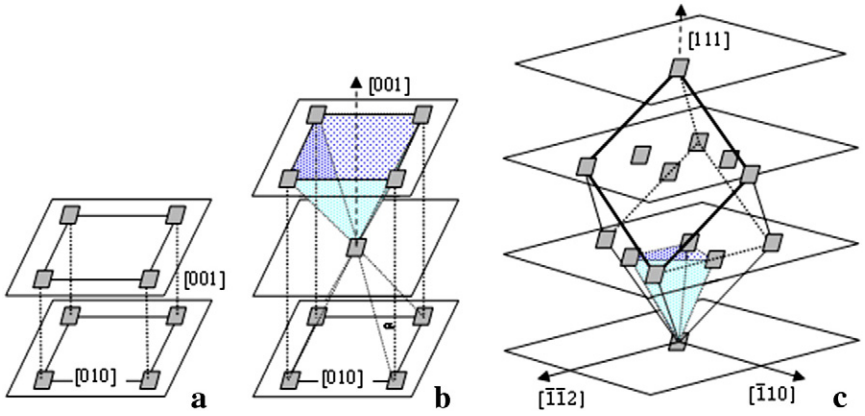

Fig. 1. Illustration of the three types of ordering of QDs in materials with cubic symmetry: (a) simple vertical correlation, (b) anticorrelated stacking, that tends to form a BCT superlattice, and (c) anticorrelated stacking that has been observed to form a trigonal, FCC like superlattice.