# Three dimensional strain distribution during stacking of self-assembled InGaAs/GaAs quantum dot layers

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## Abstract

We investigated theoretically the distribution of lattice distortion in three dimension around self-assembled InGaAs/GaAs quantum dot (QD) and its influence on vertical QD stacking. Based on the finite element method, we computed the strain distribution around pyramidal QD so that the total strain energy takes the minimal value. Mechanism of QD alignment in growth direction is discussed taking into account the effect of indium hole formed in stacked wetting layer just above underlying QD. Assuming that the QD in the stacked layer is grown at the position where the lattice distortion is minimal, we estimated the critical thickness of spacer layer between QD layers to make QDs align in growth direction, that agrees well with previously reported values in experiments.

Keywords: A3 Quantum dots, B1 Nanomaterials, B2 Semiconducting III-V materials, B2 Semiconducting gallium arsenide, B2 Semiconducting indium compounds / PACS code 81.07.Ta Quantum dots

## I. Introduction

The self-assembled InGaAs/GaAs quantum dot (QD) is one of the most promising materials for the devices of future optical telecommunication and quantum computation [1 - 5]. To meet the requirements of the device developments, high controllability of QD properties such as size, composition, uniformity, numerical density and positioning has been eagerly investigated. For the purpose, various growth techniques have been examined, e.g., usage of substrate orientation, alternative precursors, strain reducing layer, and addition of surfactant [6 - 9]. It is a well-known and important technique to make QDs align in growth direction by stacking two QD layers closely [10]. Using the vertical self-alignment phenomenon, control of emission wavelength and reduction of emission linewidth have been achieved [11, 12]. The phenomenon was affected by the growth temperature, growth rate, and spacer layer thickness between QD layers [13 - 16]. On the other hand, analysis of the QD growth process has also been promoted. Early study revealed that atom migration caused depressions in GaAs capping layer above QDs during growth [10]. The mass transportation of indium atoms has been believed to form partial wetting layer on the surface of capping layer [17, 18]. Recently, the surface depression has come to be observed directly in molecular beam epitaxy system combined with in-situ scanning tunneling microscope [19]. Hole and valley structures were observed on the surface of capping layer just above QDs when the growth rate was low [19, 20]. The vertical self-alignment process must be dominated by these surface structure, but the detail has not been well investigated, yet.

In this paper, we investigated theoretically the distribution of lattice distortion in stacked self-assembled InGaAs/GaAs QD structure. We computed the lattice distortion in three dimension using the finite element method, and estimated the influence of indium hole on the strain distribution in the stacked wetting layer as a function of distance between QD layers. We discuss the probability of QD alignment in growth direction assuming that QD is formed at the position where the lattice distortion is minimal.

## II. Model of Calculation

We assumed a pyramidal InGaAs QD structure with wetting layer grown on GaAs substrate and capped by GaAs layer. Figure 1(a) shows schematic of the basic model in the calculation. The structure with single QD is illustrated. Considering the symmetry of the structure, we computed the strain distribution of one fourth of the structure as shown in the figure. The origin of axes was set at the center of bottom of the wetting layer. In the calculation, we assumed that the indium composition is common among QD and wetting layer. To apply the three-dimensional finite element method, we regarded the structure to be continuous elasticity, and divided it into cubes composed of six trigonal pyramidal voxels [21]. Length of side of the cube was set to be 0.75 nm. We computed strain distribution in the structure so that the strain energy takes the minimal value. Figure 1(b) indicates the model of the calculation with stacked wetting layer before QD is formed on the stacked layer. We compared the case when the indium hole exists just above QD and not. The hole is sized as the same as the base of underlying QD. The thickness of spacer layer between the layers is indexed by t. We assumed that the structure is capped by GaAs layer for the convenience of computation. Thus, the hole is filled by GaAs. The assumption of GaAs capping layer will make the absolute value of strain to be underestimated, but the influence of the indium hole will appear in the results of calculation.

## III. Results and Discussions

The three-dimensional distribution of lattice distortion with single QD is shown in Fig. 2, that compares the calculated distribution of direct strain in z axis in x-y plane, $\varepsilon_{zz}$, at various depth. The difference of $\varepsilon_{zz}$ between its maximum and minimum, $\Delta\varepsilon_{zz}$, is also indicated in the figure. In this calculation, we assumed that QD indium composition is 0.5 and that base length and height of QD are 24 nm and 6 nm, respectively. The wetting layer thickness was set to be 0.75 nm. We can see that the shape of strain distribution changes much depending on z value. Under the wetting layer ($z < 0$ nm), strain field expands to the circumference of QD bottom. In the plane slicing QD ($0$ nm $< z < 6$ nm), the outline of the maximum strain indicates about the border between QD and surrounding GaAs. As z increases from the bottom to the top of QD, the size of QD cross section decreases and the outline shrinks to the center. The center at QD top has the strongest strain field. In the GaAs capping layer above the QD top ($z > 6$ nm), $\varepsilon_{zz}$ decreases as z increases.

We performed the same estimation of strain distribution assuming various diameter, composition, and height of QD. The estimated in-plain differences of direct strain in x and z axis, $\Delta\varepsilon_{xx}$ and $\Delta\varepsilon_{zz}$, are shown in Fig. 3 as a function of z. In Fig. 3(a), we see that $\Delta\varepsilon_{xx}$ changes smoothly and becomes zero at about $z = 24$ nm. Though the maximum value of $\Delta\varepsilon_{xx}$ varies with structural parameters, the expansion limit in z direction is almost common. In Fig. 3(b), we can see that the behavior of the strain in z axis is different from that in x axis. There are the two peaks corresponding to the position of QD top and wetting layer. We see in all cases that $\Delta\varepsilon_{zz}$ takes the minimum at around $z = 16$ nm and becomes very small at over the z value though both the peak position of $\Delta\varepsilon_{zz}$ and its maximum value depend on structural parameters.

We next investigated the stain distribution with stacked layer. We based on the model of QD stacking process shown in Fig. 4. The mass transportation on QD causes retardation of GaAs coverage during the growth of spacer layer, and forms wetting layer having indium holes on the underlying QDs [13, 14]. The stacked QD is formed at the indium holes. In the following calculation, we modeled the situation of Fig. 4(c) by Fig. 1(b). We are interested in the critical spacer layer thickness with which the hole forming causes vertical QD alignment. We then simulated how the strain distribution in the stacked layer depends on the spacer layer thickness between QD layers.

Figure 5 compares the in-plane distribution of direct strain in z direction, $\varepsilon_{zz}$, at the bottom of the stacked wetting layer between two cases with and without indium hole. The difference of $\varepsilon_{zz}$ between its maximum and minimum, $\Delta\varepsilon_{zz}$, is also indicated in the figure. Here, we assumed QD diameter of 24 nm, QD height of 6 nm, and QD indium composition of 0.5. Thickness of both wetting layers was 0.75 nm. As shown in Fig. 5(a), without indium hole, $\varepsilon_{zz}$ takes the largest value at the center above the QD top, and $\Delta\varepsilon_{zz}$ reduces as the spacer layer thickness, t, increases. With indium hole, the strain is reduced at the hole and the shape of strain distribution changes much depending on t. In Fig. 5(b), the center of the layer has rather small strain compared with the surrounding area when $t > 10.5$ nm. On the other hand, the most strained area is the center when $t < 10.5$ nm though weakly-strained area exists around.

Figure 6 indicates the maximum of in-plane direct strain in z axis, $\varepsilon_{zz}(\max)$, and the difference to the minimum, $\Delta\varepsilon_{zz}$, as a function of t. We see that $\varepsilon_{zz}(\max)$ is always smaller with indium hole, but the effect of the hole is almost negligible when $t > 14$ nm. Spacer layer thickness has much effect on $\Delta\varepsilon_{zz}$. Without indium hole, $\Delta\varepsilon_{zz}$ decreases simply as t increases. When we assume indium hole, $\Delta\varepsilon_{zz}$ takes the minimal value at $t = 10.5$ nm. The minimum point is the border where the strain at the center area competes with the strain at the surrounding area (see Fig. 5(b)). We noted that the two lines of $\Delta\varepsilon_{zz}$ in cases with and without hole cross each other at about t = 11.5 nm.

In the following, we discuss the QD stacking process based on the results of our calculation. If we assume that QD is grown on the area where the surface strain is minimal, our results predict where QD will be formed in stacked layer. Figure 5(a) suggests that QD will not grow at above underlying QD without indium hole, and in the case, QD will not align in growth direction. With indium hole, when the wetting layer separation is large, the weakly-strained area exists at above underlying QD as seen in Fig. 5(b). Even when the wetting layer separation is small, there is still weakly-strained area around above QD, that will lead to the growth nucleation at the area. Ultimately, QD position will be correlated in growth direction with indium hole.

The relationship between the vertical QD alignment and the spacer layer thickness is revealed in Fig. 6. When the spacer layer is thicker than 14 nm where the reduction of strain owing to indium hole becomes negligible, the indium hole will be diminished with surface migration of atoms during growth of the spacer layer. Considering that the modulation of strained area reaches to about 25 nm in height in the spacer layer as shown in Fig. 3, we suppose that the critical thickness for the existence of indium hole will take the value between 14 and 25 nm. Growth condition will determine the actual value. Note that the transmission limit of strain modulation does not depend much on QD structure. With spacer layer thicker than 25 nm, QD will grow at random in position on growth surface. These estimated values of thickness agree well with the critical spacer layer thickness for vertical QD alignment in previously reported experiments.

When the spacer layer is thinner than 11.5 nm where both the maximum strain and in-plane strain variation become smaller with indium hole, the hole will exist just before QD is formed at the surface. In this case, QD will align in the growth direction. This value also agrees well with the values in experiments. For instance, it was reported that the indium holes were formed with 10 nm GaAs cap layer when the growth rate was low [20]. Considering that our calculation models static process, it is reasonable that our results well describe the phenomena at low growth rate. It was also reported that the correlation of QD position in growth direction was as high as 95 % when the spacer layer thickness is smaller than 11 nm [15].

When the spacer layer thickness is set to be intermediate value between above two critical values (i.e., 11.5 nm < t < 14 nm), the indium hole will have a tendency to form, but actual surface dynamics such as migration, nucleation and intermixing will dominate the surface morphology. In experiment, it was reported that the depth of valley on capped QD was largest with 10-nm cap layer and drastically reduced as the cap layer thickness increased to 17 nm [19].

It should be noted that if the indium hole is filled out to form uniform flat wetting layer at the beginning of growth nucleation, there is no reason for vertical QD alignment. The actual growth will not proceed in a such way since it raises total strain energy. As one explanation, we suppose that the hole will be buried by atoms having smaller indium composition than existing wetting layer. The low indium area has high potentiality to form QD after the amount of source exceeds the elastic limit of layer-by-layer growth. Surface intermixing during growth will affect this process and wipe out the trace of indium hole easily since the wetting layer is monolayers in thickness.

## IV. Summary

Based on the finite element method, we investigated theoretically the three dimensional lattice distortion around the self-assembled InGaAs/GaAs QD and its influence on the stacking of QD layers. Mechanism of vertical QD alignment was discussed taking into account the influence of indium hole in stacked wetting layer. Assuming that the QD in the stacked layer is grown at the position where the lattice distortion is minimal, we estimated the critical thickness of spacer layer between QD layers to make QDs align in growth direction, that agrees well with previously reported values in experiments.

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Figure captions

Figure 1 Schematic of pyramidal QD assumed in the calculation; (a) single QD, and (b) QD with stacked wetting layer having indium hole above the QD.

Figure 2 Strain distribution in one fourth of x-y plane sliced at several z positions. Here we assume that QD has base length of 24 nm, height of 6 nm and indium composition of 0.5.

Figure 3 Difference of direct strain in x-y plane as a function of z; (a) in x axis, $\Delta\varepsilon_{xx}$, and (b) in z axis, $\Delta\varepsilon_{zz}$. As indicated in the figure, we compare the cases when diameter (R), indium composition (x), and height (h) of QD vary.

Figure 4 Schematic illustration of vertical QD alignment process with retardation of GaAs coverage and following indium hole formation.

Figure 5 Strain distribution in one fourth of x-y plane sliced at the bottom of stacked wetting layer: (a) without an indium hole and (b) with an indium hole.

Figure 6 The maximum of direct strain in z axis, $\varepsilon_{zz}$(max), and its difference to the minimum, $\Delta\varepsilon_{zz}$, in x-y plane at the bottom of stacked wetting layer as a function of spacer layer thickness, t.

**Figure1**

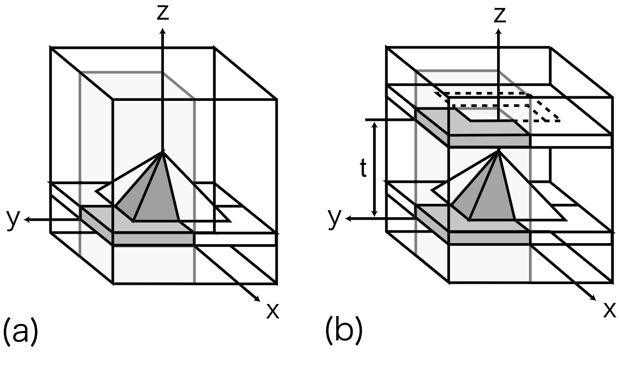


Figure 1  Mukai_JCG

**Figure2**

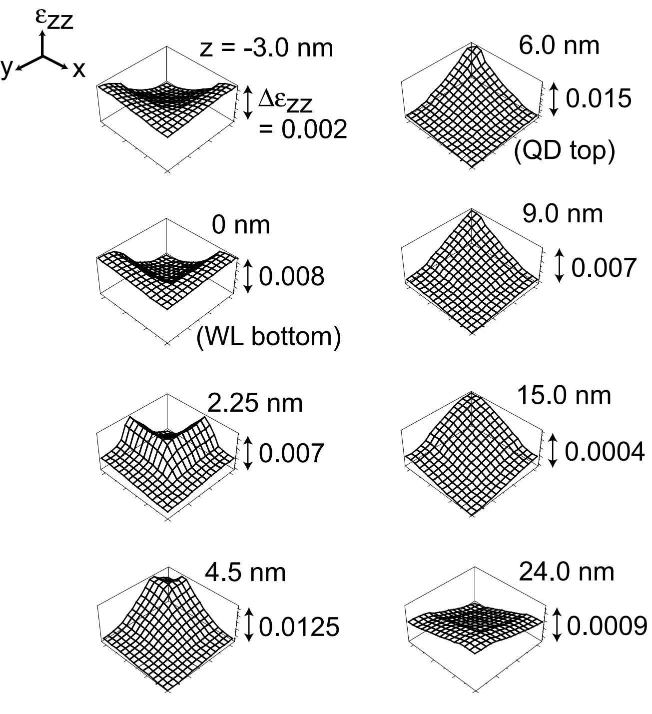
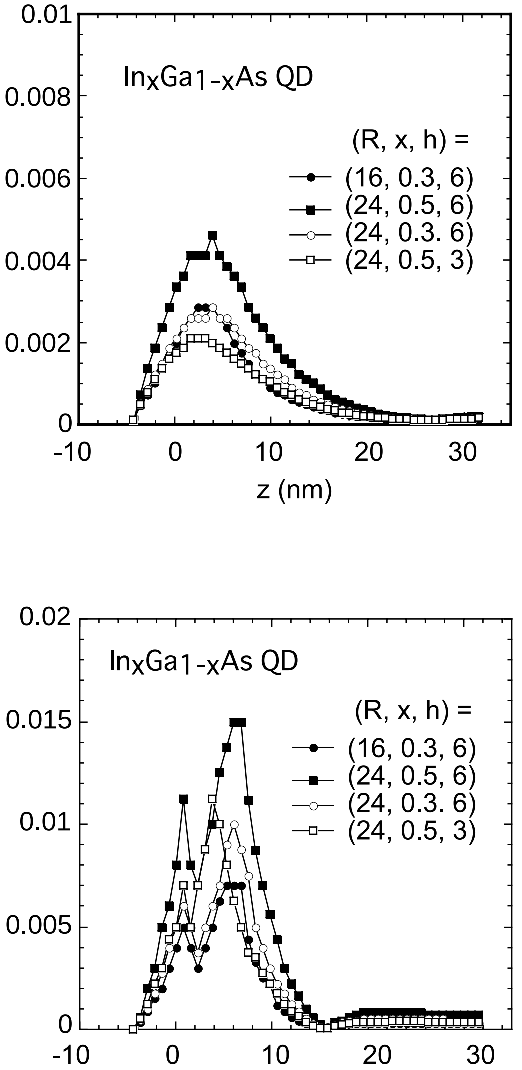


Figure 2  K. Mukai_JCG

**Figure3**



(a)

(b)

Figure 3 K. Mukai_JCG

**Figure4**

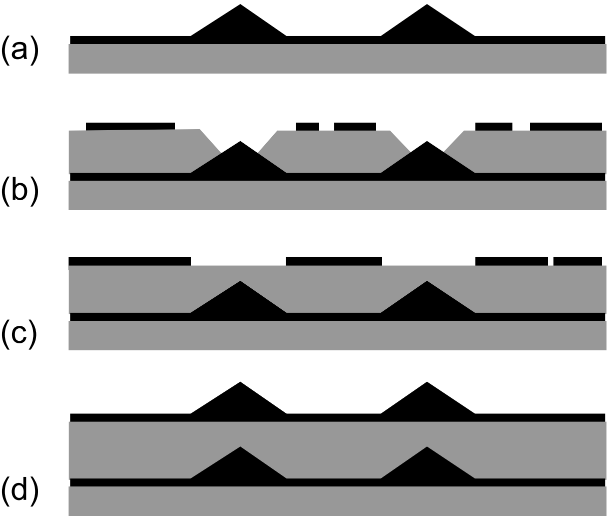


Figure 4 K. Mukai_JCG

**Figure5**

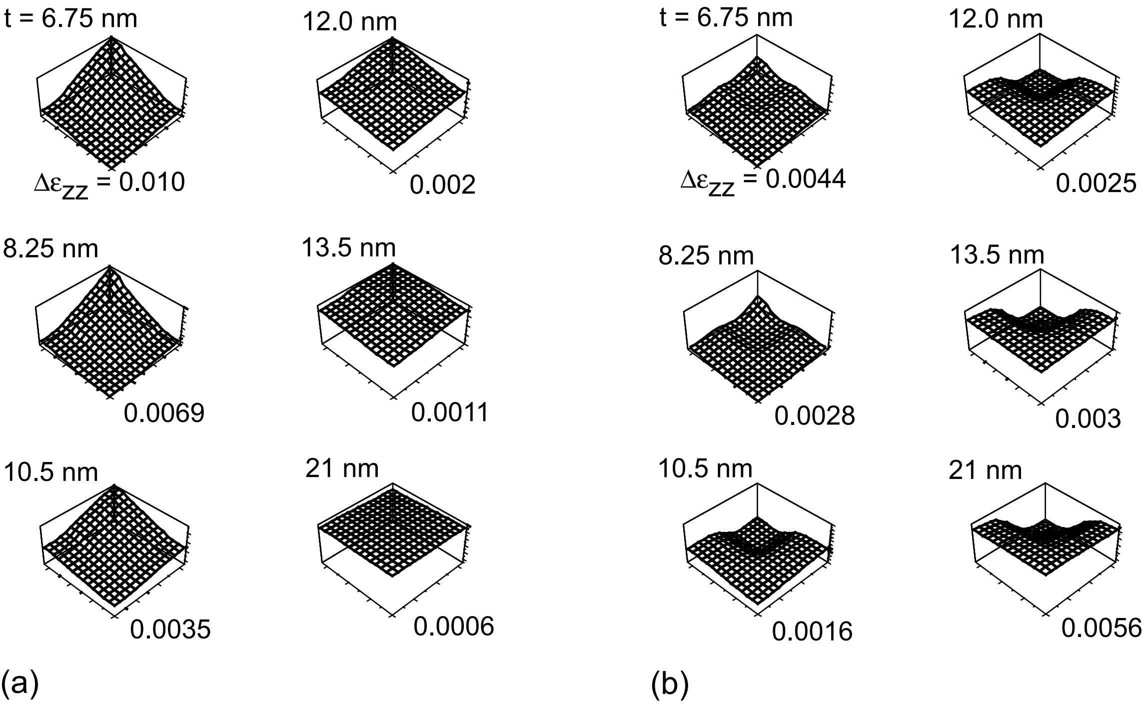


Figure 5 K. Mukai_JCG

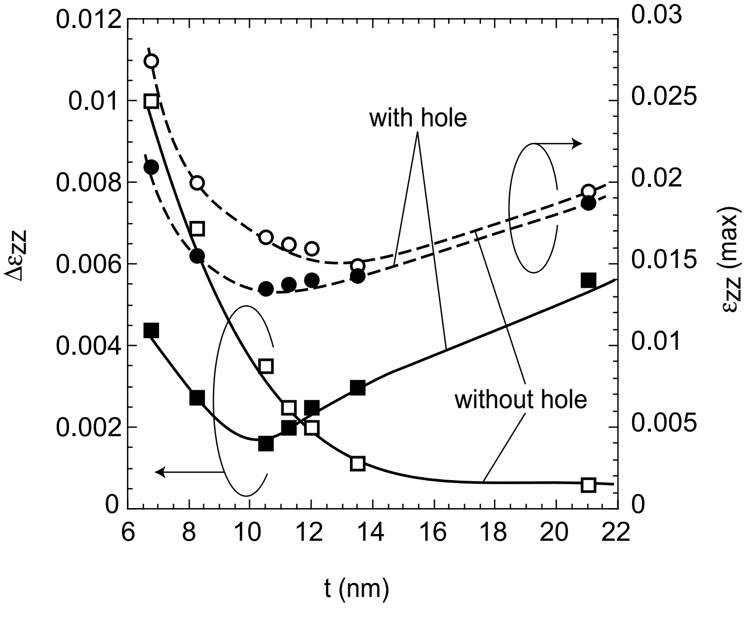


Figure 6 K. Mukai_JCG