TEM studies of multilayered In$_{0.33}$Ga$_{0.67}$As quantum dots

DV Sridhara Rao$^1$, K Muraleedharan$^1$, R Balamuralikrishnan$^1$, R Muralidharan$^2$, T Srinivasan$^2$ and D Banerjee$^3$

$^1$Defence Metallurgical Research Laboratory, Kanchanbagh Po, Hyderabad-58, India.
$^2$Solidstate Physics Laboratory, Timarpur, Lucknow Road, Delhi-54, India
$^3$Defence Research and Development Organisation, New Delhi, India.

E-mail: vsrd2@cam.ac.uk, dvsridhararao@gmail.com

Abstract. We have employed TEM techniques to investigate vertical alignment in multilayered In$_{0.33}$Ga$_{0.67}$As quantum dots (QDs) as a function of deposition thickness. Our experimental results are discussed in the context of an analytical model reported in the literature, in terms of the so-called pairing probability, which can assess the quality of vertical alignment.

1. Introduction
In$_x$Ga$_{1-x}$As quantum dots (QDs) find applications in optoelectronic devices. In$_x$Ga$_{1-x}$As grows on GaAs as a two-dimensional (2D) layer up to a transition thickness ($t_c$), and further growth results in a 2D-3D (island) morphological transition with a wetting layer. This is known as Stranski-Krastanov growth [1]. This growth mode is attractive as it results in self-assembled islands. However, many applications require the growth of QDs with large areal density, which is usually not realizable in the case of single-layered QDs. This can be overcome by the growth of multilayered dots, i.e. by repeated growth of a coherent layer of In$_x$Ga$_{1-x}$As QDs followed by a thin GaAs spacer layer. The tensile strain field due to the buried dots transmits through the spacer layers, resulting in preferential growth of dots on top of the buried dots, thereby leading to their vertical alignment [2].

In this work, we have used TEM techniques to investigate vertical alignment in multilayered In$_{0.33}$Ga$_{0.67}$As quantum dots (QDs) as a function of deposition thickness. Our results suggest that the dots are neither prominent nor vertically aligned when the deposition thickness is just above $t_c$. However, for deposition thickness ~ 1.5$t_c$, the dots are well formed with good vertical alignment. Our experimental results are discussed in the context of an analytical model reported in the literature, in terms of the pairing probability ($P$) [3], which can be used to determine the quality of vertical alignment.

2. Experimental
The (001) epi-ready GaAs substrates were loaded in the growth chamber of a RIBER 2300 R&D molecular beam epitaxy (MBE) system. After desorption of the native oxide at 580°C, a buffer structure consisting of a semi-insulating GaAs (0.5 µm thick) layer and an Al$_{0.24}$Ga$_{0.76}$As/GaAs superlattice (20 periods, thickness of one period is 10 nm) was grown by using Ga, In and As solid sources and under arsenic overpressure (or beam equivalent pressure, BEP ratio of about 20). The multilayered QD structure consisting of 20 periods of In$_{0.33}$Ga$_{0.67}$As layers with 20 nm thick GaAs spacers was grown at 500°C, without growth interruption. During growth, in-situ reflection-high
energy electron diffraction (RHEED) was used to establish that the transition thickness of $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$ layers was 12 monolayers (MLs) (~3.47 nm). Two different samples were grown with a nominal thickness of 13 MLs (just above $t_c$, case-A) and another with 17 MLs (~1.5 $t_c$, case-B) respectively. The latter structure was capped with a 5 nm thick GaAs, whereas the 13 MLs sample was left uncapped. The samples were cooled to room temperature under arsenic overpressure.

Cross-sectional TEM specimens were prepared by argon ion beam thinning using a Gatan Duo Mill (Model 600) with a liquid nitrogen cold stage. The samples were investigated using a Philips EM430T analytical TEM (operated at 250 kV) and a Tecnai 20 UT high-resolution TEM (HRTEM, 200 kV). The specimen preparation procedure ensured that the foils were near <110> zone axis at zero tilt conditions. This zone contained the direction of growth (i.e., [001]). As a result, the multilayers could be investigated in the edge-on orientation with respect to the electron beam. Also, (002) composition-sensitive two-beam dark field (DF) imaging was used for the estimation of the structural parameters (height and width) of dots [4].

3. Results and Discussion
In case-A, the cross-sectional TEM studies (Fig. 1A) revealed that the buried dots are not well formed. However, in case-B, the QDs are formed prominently (Fig. 1B), with a wetting layer, confirming the S-K growth mode. The wetting layer thickness is 3.5±0.3 nm, which is, within measurement errors, close to the transition thickness of 3.47 nm (corresponding to 12 MLs). The extra material deposited beyond the transition thickness has contributed to the growth of the dots to a bigger size. Also, the QDs are coherent with the GaAs spacers (Fig. 1C). Further, the QDs are well aligned along the growth direction, and they resemble the shape of a plano-convex lens. The strain effects are dominant in the <110> on-zone imaging condition; hence the structural parameters of the QDs are estimated by using the (002) two-beam dark (DF) field imaging condition. Due to the composition sensitive nature of this imaging condition, $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$ appears darker than GaAs (Figs. 1A,1B). In addition, a contrast variation has been observed within each dot. This can be attributed to a shape factor [5] caused by the plano-convex morphology of the dot, due to which the electron beam passes through varying thicknesses of $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$ and the embedding GaAs, and also from one phase to the other, as we move from the top of the dot to its base. Beanland [5] has also shown that the actual size of the dots is the region enclosed within the black contrast periphery and its outer edge has to be used for the estimation of the height ($h$) and diameter ($D$) of the dots.

![Figure 1](image.png)

**Figure 1.** (002) Dark field TEM micrographs of multilayered quantum dots corresponding to a $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$ deposition thickness of (A) 13 MLs (case-A) and (B) 17 MLs (case-B). The lattice resolution image of a buried dot is shown in figure C.
We have quantified the vertical alignment of the QDs along the growth direction, using the analytical model proposed by Xie Qianghua et al. [3]. In this model, developed for the InAs/GaAs system, the authors quantified the vertical alignment using the concept of pairing probability ($P$). The factors considered are: dot size ($d$), inter-dot spacing ($l$), thickness of spacer layer ($Z_s$), and the elastic properties (such as misfit and elastic constants) of the InAs and GaAs layers. The average dot size is given by the diameter of an equivalent sphere that has the same volume as that of the plano-convex lens of height $h$ and diameter $D$. For an ideal case (i.e. perfect vertical alignment), $P = 1$. It decreases with the deterioration of the alignment. Based on the value of $P$, the authors have classified the alignment as being strongly correlated ($P \sim 0.9$ or higher), moderately correlated ($P \sim 0.4-0.9$), or uncorrelated ($P < 0.4$).

**Table 1.** Material parameters used for estimation of pairing probability. For ternary alloy, the respective parameters were estimated using Vegard’s law.

| Parameter                          | GaAs | InAs | In$_{0.33}$Ga$_{0.67}$As |
|------------------------------------|------|------|--------------------------|
| Young's Modulus ($E$, GPa)         | 86.96| 51.42| 75.23                    |
| $C_{11}$ (GPa)                     | 118.8| 83.4 | 107.12                   |
| $C_{12}$ (GPa)                     | 53.8 | 45.4 | 51.03                    |
| $C_{44}$ (GPa)                     | 59.4 | 39.5 | 52.83                    |
| Bulk Modulus ($B$, GPa)            | 76.28| 57.13| 69.96                    |
| Poisson’s ratio ($\gamma$)         | 0.31 | 0.35 | 0.323                    |
| Lattice mismatch (with GaAs)       | -    | 7.2% | 2.3%                     |
| Lattice parameter ($a$, nm)        | 0.6058| 0.5653| 0.5787                   |
| (Rounded-off to fourth decimal)    |      |      |                          |

*Reference [6]  
*Bulk Modulus=$E/3(1-2\gamma)$

We have calculated the pairing probability for QD structures (cases A, B) using the structural parameters obtained from our experiments. The QDs in the top layer were not considered in the estimations, as the capping affects the structural parameters. In case-A, though the buried dots are not well formed, we have nevertheless attempted to estimate the required structural parameters. The average dot height ($h$) and diameter ($D$) are estimated to be about 3 nm and 17 nm leading to an average dot size of 8.78 nm. The average interdot spacing ($l$) has been measured to be 70 nm. In case-B, the average dot height ($h$) and diameter ($D$) of the buried dots are estimated to be 9 nm and 42 nm leading to an average dot size of 23.3 nm. The average interdot spacing ($l$) has been measured to be 58 nm. Ternary material parameters, such as lattice parameter and bulk modulus, were calculated using Vegard’s law from the reported data for InAs and GaAs (Table 1). Similar to the analysis of Qianghua Xie et al. [3], the pairing probability as a function of the spacer thickness has been plotted for the two cases (Fig. 2). At a spacer thickness of 20 nm (our experiments), $P$ has been estimated to be about 0.25 and 0.92 respectively, indicating that uncorrelated and strongly correlated conditions are prevalent in the respective cases. These predictions are in good agreement with our experimental observations (Figs. 1A and 1B), indicating that the model continues to hold for QDs with lower indium content.
Figure 2. Pairing probability ($P$) as a function of spacer thickness of In$_{0.33}$Ga$_{0.67}$As dots at a deposition thickness: $\sim t_c$ (13 MLs, corresponding to figure 1A) and $\sim 1.5t_c$ (17 MLs, corresponding to figure 1B).

4. Summary
We have investigated the formation of In$_{0.33}$Ga$_{0.67}$As multilayered dots with GaAs spacers. The dots are well formed with good vertical alignment for a deposition thickness close to 1.5 times the transition thickness. In contrast, the dots are not prominently formed and relatively poorly aligned for the deposition thickness just above the transition thickness. In both cases, the dots are predominantly coherent. Further, considerable agreement on the vertical alignment of dots has been found between our experiments on In$_{0.33}$Ga$_{0.67}$As quantum dots and the analytical model of Qianghua Xie et al. [3] developed for InAs quantum dots.

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