The structure of coherent and incoherent InAs/GaAs quantum dots

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ABSTRACT: During the heteroepitaxial growth of semiconductors, misfit-induced strain causes various growth and relaxation phenomena. These include the formation of islands or dots (elastic relaxation of pseudomorphic misfit strain) and the formation of dislocations preferentially at sites of high strain. We have studied strain relaxation in InAs/GaAs quantum dots (QDs). Due to strain-induced renormalization of the surface energies of their facets, an array of QD islands with uniform size and shape can be formed. With increased InAs coverage, incoherent QDs start to form, and the samples with both coherent and incoherent QDs can exhibit bimodal size distributions, with coherent strained QDs that are smaller than incoherent plastically-relaxed QDs. The transition point was determined by both plan-view and cross-sectional transmission electron microscopy (TEM). In the region where two types of QDs coexist, coalescence may occur between adjacent dots and the larger QDs grow at the expense of smaller QDs. By means of both high-resolution transmission electron microscopy (HRTEM) and the measurement of the displacement field of a QD, we have established that misfit dislocations start to form at the QD edge at the beginning of the coherent/incoherent QD transition. Misfit relaxation in large QDs is then accommodated by the generation of misfit dislocation arrays (plastic relaxation), and also by the distortion of lattice planes (elastic strain relaxation).

1. INTRODUCTION

The formation of semiconducting quantum dots in lattice-mismatched heterostructures has been extensively studied in recent years. One of the major problems in the heteroepitaxial structures is that the lattice mismatch between the epitaxial layer and its substrate may introduce misfit dislocations at the interfaces in order to relieve the misfit strain. The phenomenon of strain relaxation in hetroepitaxial InGaAs/GaAs films has been extensively studied earlier (Zou et al 1994). The formation of 3D nano-sized epitaxial islands via the Stranski- Krastanow (S-K) growth mode as the result of strain energy accumulation as well as other complex physical reasons like surface reconstruction and elemental surface segregation has now been employed as the way for self-assembly of the quantum dot structures.

In the self-assembly of InAs/GaAs quantum dots, the 2D-3D transition was observed to happen at the deposition of 1.7-1.8 monolayers (MLs) of InAs, as evidenced by atomic force microscopy (AFM) and scanning tunnelling microscopy (STM) (Joyce et al 1998). When the InAs deposition amount increases, the strain in the QD layer is accumulated. To accommodate or minimise the strain energy in the system, certain strain relief mechanisms should function in general, such as reorganisation of the shape of QDs and the introduction of misfit dislocations. If all the other growth parameters were kept constant, then a growth series with increased InAs coverage from the critical amount will represent complete QD formation and structural evolution process. In this paper, we systematically investigate the structure of coherent and incoherent InAs QDs formed on GaAs(001) at different InAs coverage. The morphology and structure of these QDs are characterised using both plan-view and cross-sectional transmission electron microscopy (TEM).

2. EXPERIMENTAL

The QD samples were grown in a combined MBE-STM growth system that is also equipped with reflection high energy electron diffraction (RHEED) for in *situ* monitoring of growth mode change. The indium, gallium and arsenic cells were calibrated using RHEED oscillation on InAs (001) and GaAs (001) respectively. After initial thermal cleaning at 300 ºC, the native oxide layer was removed under As_2 flux at 620ºC. A 0.6 μm thick GaAs buffer layer was grown at 580 ºC and the substrate temperature reduced to 510 ºC for the deposition of 300Å of GaAs. InAs was then deposited at a growth rate of 0.016 MLs^{-1} and the InAs coverage ranged from 1.75 to 3.8 monolayers.

In similar fashion to our previous TEM study (Zhi et al 2001, 2004), the plan-view and cross-sectional specimens for TEM were produced using conventional sample preparation techniques, involving mechanical thinning followed by ion-milling using Ar^+ at 3-5 keV. Thinned specimens were examined using a JEOL 2010 TEM operating at 200 kV. A geometric phase technique is also applied to analyse the HRTEM images of the QDs in order to measure their displacement fields (Hÿtch et al 2003).

3. RESULTS AND DISCUSSIONS

The formation of QDs was observed by TEM in the QD samples with the InAs coverage larger than 1.7ML. Figure 1a shows a plan-view bright-field TEM image of the QD sample with 1.75 ML InAs coverage. The image was taken under Laue diffraction conditions and along the [001] direction. The detailed structure of the above QDs formed at 1.75 ML indium coverage was then observed by the cross-sectional HRTEM lattice image, as illustrated in Fig. 1b. The defect-free coherent QD was found to form at a surface step with a height difference of about two MLs. Although from TEM observation it is not clear when and how the observed step is formed, the surface steps produced by the 2D growth mode earlier at lower InAs coverage facilitate QD formation by creating more nucleation sites.

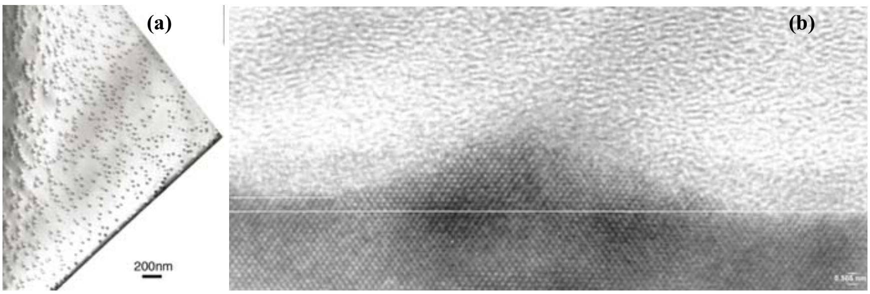

Fig. 1. (a) [001] zone axis bright-field plan-view TEM image of uncapped QDs grown with 1.75 ML InAs deposited at a growth rate of 0.016 MLs^{-1}. (b) [110] zone axis HRTEM image of an uncapped QD grown with 1.75ML InAs deposited at a growth rate of 0.016 MLs^{-1}.

Figure 2a shows a typical on-zone plan view TEM image taken from the QD sample with 2.2ML of InAs coverage, which shows the uniform, dense, coherent QDs present on the surface. These QDs are uniformly distributed and isolated from each other. The QD density was found to increase with the InAs coverage and reaches a maximum with an InAs coverage of 2.2ML. Both plan view and cross-sectional TEM images from these samples also show the increase of QD diameter with the InAs coverage increasing from 1.75ML to 2.2ML. From the above TEM images, only one type of QD, i.e. coherent strained QDs, was observed from the sample with InAs coverage ranging from 1.75 to 2.2ML. It was also observed that these coherent InAs QDs have a uniform size distribution. For InAs coverages over 2.4 ML, however, it was found that the QDs adopt a bimodal distribution of size and the QD number density begins to decrease. This bimodal QD distribution can be observed by plan-view TEM, as shown in Fig. 2b. Two types of QD can then be distinguished for the QD samples with InAs coverages over 2.4 ML, i.e. small coherent strained QDs and large plastically relaxed QDs. The relaxed QDs can be easily identified by the presence of moiré fringes inside them on plan-view TEM images.

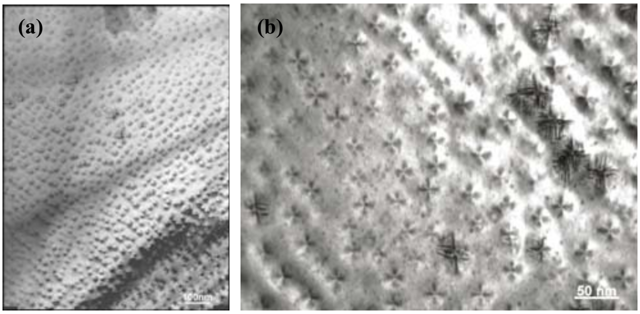

Fig. 2. (a) [001]-zone bright-field TEM image of uncapped QDs grown with 2.2ML InAs deposited at a growth rate of 0.016 MLs^{-1}. (b) [001] plan-view bright field TEM image of uncapped QDs with 2.7ML of InAs deposition.

Using the cross-sectional HRTEM images, we have observed that misfit dislocations are actually generated from the edge of the QDs (Figs. 3a and 3b). This observation can be further confirmed by the measurements of the displacement field around a QD, as shown in Figure 3c.

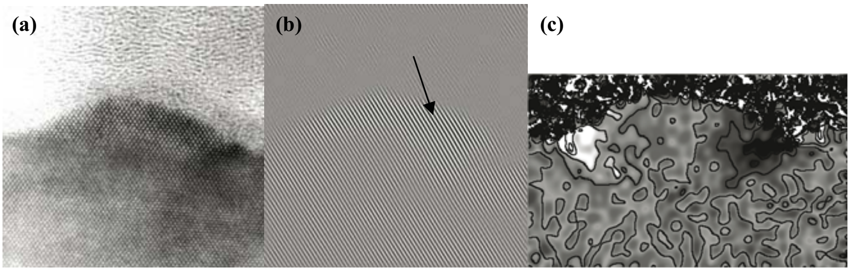

Fig. 3. (a) [110] cross-sectional HRTEM image of a smaller InAs QD with 2.7 ML InAs deposition. (b) reconstructed image by inverse Fourier transformation using the sideband $\bar{1}1\bar{1}$, a misfit dislocation is indicated by arrow, (c) a lattice rotation contour map measured for the QD shows two dislocations formed at the QD edges.

For the samples with InAs coverages in the range of 2.7 to 3.8ML, the fraction of relaxed QDs increases with increasing InAs coverage. The dimensions of the relaxed QDs are 30-50nm. The distances between contiguous moiré fringes are found to be smaller for larger incoherent QDs. For relaxed QDs with base sizes over 30 nm, the lattice distortion in the top region of the QD and the generation of misfit dislocations close to the InAs/GaAs interface could be observed directly in cross-sectional HRTEM images. Figure 4 is a [110] cross-sectional HRTEM image taken near the edge of a large QD in a 2.7ML InAs coverage sample, showing a misfit dislocation. In this case, the misfit dislocation can be identified as a 60º dislocation with **b** = $\mathbf{a}_{QDlayer}/2$ <101>, as shown in the Burgers vector analysis on the lattice image of Fig. 4.

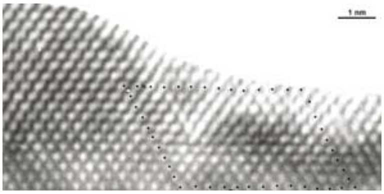

Fig. 4. [110] cross-sectional HRTEM image of an unburied QD. The black dotted line shows the determination of Burgers vector.

For higher InAs coverage, such as 3.8ML (not shown here), cross-sectional lattice images of large relaxed QDs show the introduction of an array of misfit dislocations close to the interface with the GaAs substrate. These misfit defects can be identified as 90º edge dislocations, using a similar Burgers vector analysis. In this high-lattice-mismatch system, the morphological transition takes place at the critical thickness from two-dimensional layer-by-layer growth to three-dimensional island nucleation, believed to be induced by strain with consideration of the surface condition of the materials in the system. Here, with increasing InAs coverage during MBE growth, the accumulated strain is first relieved by coherent island formation and then by dislocation generation at the island edges (e.g. 60º dislocation generation at the QD side edges). As more InAs is deposited, adjacent QDs may coalesce and a variety of defect types may be created.

4. CONCLUSIONS

This study shows that InAs/GaAs QDs grown at low InAs coverage are coherent. They are small in size (~13-20 nm) and have a uniform size and shape distribution, in a dense arrangement (10^{10}-$10^{11}cm^{-2}$). With increasing InAs coverage, incoherent QDs start to form, and samples with both coherent and incoherent QDs usually exhibit bimodal size distributions, where coherently strained QDs are smaller than incoherent plastically-relaxed QDs. Where the two types of QD coexist, Ostwald ripening may occur, where large QDs grow at the expense of small QDs. Misfit relaxation in large QDs is accommodated by the generation of misfit dislocations, and also by the distortion of lattice planes (elastic strain relaxation).

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