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Influence of annealing on the formation of InGaAs quantum dots in GaAs matrix during metal organic chemical vapor deposition

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Abstract. Structure investigations of InGaAs quantum dots (QD) grown by metal organic chemical vapor deposition (MOCVD) on GaAs substrate at various growth conditions were carried out by transmission electron microscopy. Incomplete covering of InGaAs QD layer by a thin GaAs spacer grown at 480°C followed by annealing at 600°C is shown to result in removal of developed defect structure observed in unannealed samples. The growth of three InGaAs QD layers separated by GaAs spacers using the same treatment algorithm for each InGaAs QD layer led to perfect QD system formation.

Introduction

During the last several years self-organized nano-sized domains (so-called quantum dots) have been attracting much attention [1, 2]. In particular, QD present the ultimate spatial confinement of charge carriers and to date have been applied as an active media in modern semiconductor devices, i.g. lasers [1]. Currently molecular beam epitaxy is widely used to grow structures with InAs or InGaAs QDs on GaAs substrate [1]. The MOCVD growth process for QDs in InAs/GaAs system has not been developed yet [2]. The aim of this work is to specify the influence of the growth conditions on the formation of InGaAs QDs during the growth on GaAs substrate by MOCVD.

1. Specimens and methods

The structures under study were grown by MOCVD using equipment with a horizontal resistively heated reactor at low pressure (76 Torr). Three kinds of structures with a single InGaAs QD layer and two kinds of structures with three InGaAs QD layers separated by GaAs spacers were grown. The AlGaAs and GaAs buffer layers were subsequently grown at 480°C on GaAs substrate (001) for all structure kinds. To grow the sample with the single QD sheet InGaAs layer was deposited for 3 sec to be transformed into QDs and cover AlGaAs and GaAs layers of 15 nm thickness each were then grown at 480°C. The main difference between the first and second or third types of structures is that the InGaAs QD layer was partially overgrown by 5 nm GaAs layer followed by annealing at 600°C and that the deposition of the AlGaAs and GaAs cover layers was performed at 600°C. The difference between the second and the third types of structures is in an annealing time of 10 or 20 min, respectively. To grow the sample with three QD sheets three InGaAs layers separated by 120 nm GaAs spacers were deposited at 480°C for 3 sec each, annealed and covered at 600°C by AlGaAs and GaAs layers of 15 nm thickness each. The main difference in the growth procedure for this structure and the second one is that the annealing at 600°C for the second structure was repeated three times after incomplete 5 nm GaAs covering of each InGaAs QD layer. Investigations were carried out using transmission electron

microscope (TEM) Philips EM-420 operating at 100 kV accelerating voltage. Specimens were prepared both in (001) plan view and (110) cross section. The conventional technique of TEM specimen preparation including mechanical treatment (grinding and polishing) and final sputtering by Ar^+ ions with an energy of 4 keV at 14° ion beam tilt from the sample surface was used.

2. Results and discussion

To obtain a distinct contrast of epitaxial layers of semiconductor heterostructures with zinc-blend lattice the dark-field (DF) technique of image formation with operating reflection (002) of which the amplitude is proportional to the difference between atom scattering factors of A and B sublattices is widely used. Analyzing (001) plan-view bright-field (BF) and (110) cross-section dark-field images obtained in two-beam conditions with operating reflections (220) and (002), respectively, allowed us to evidence QDs presence by the characteristic contrast. Along with QDs, three-dimensional (3D) dislocated islands showing characteristic moiré pattern were also detected in the sample with the single InGaAs QD layer grown without annealing. Using the value of GaAs (220) interplanar distance and the average island moiré period the average lattice constant of 3D islands was estimated to be 5.98 Å. Taking into account a residual strain in the 3D islands the obtained value is believed to correspond to the InAs lattice constant. The 3D island density is $\sim 10^8 - 10^9 \text{ cm}^{-2}$. The island lateral size varies from 50 to 100 nm. At the same time no developed defects were observed in the structure with the annealed InGaAs QD layer. Table 1 shows the results of image analysis including average QD density, lateral size and height for the samples with the single InGaAs QD layer. Thus annealing of InGaAs QD layer partly covered by 5 nm

Table 1. Data obtained from TEM images of InGaAs QD layer.

The structure type	Average QD density, cm^{-2}	Average QD lateral size, nm	Average QD height, nm
Unannealed	1.3×10^{10}	17	11
Annealed (600°C)			
$t_{\text{ann}} = 10 \text{ min}$	2.3×10^{10}	20	9
Annealed (600°C)			
$t_{\text{ann}} = 20 \text{ min}$	2.4×10^{10}	20	7

GaAs spacer leads to improving crystal quality and changing QD shape, i.e. decreasing QD height and increasing QD lateral size. To explain the obtained data one needs to analyze the growth process of material lattice mismatched to the substrate. If the sum of layer surface energy and interface energy does not exceed substrate surface energy the deposition of small InGaAs quantity on the GaAs (001) surface leads to the planar strained InGaAs layer formation. The subsequent material deposition results in an increase of a layer elastic strain energy. To decrease this energy the layer can spontaneously transform to an array of three-dimensional islands on the residual wetting layer (Stranski–Krastanow (SK) growth mode). These islands have the pyramidal shape [1] and is known to be QD. But actually three-dimensional dislocated islands significantly exceeding QDs in size also appear. The reasons for these islands to occur are not clear yet but the substrate surface undulations and the presence of other surface peculiarities which are difficult to avoid in MOCVD may be the nucleus of dislocated islands. The presence of dislocations in islands may cause an enhanced material accumulation on them. The average thickness of the GaAs spacer used for overgrowth of QDs is about 5 nm which is insufficient for complete covering of

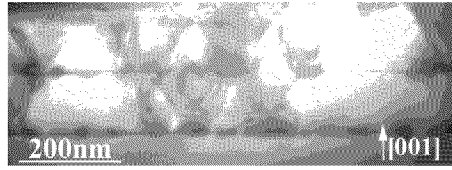


Fig. 1. The (110) cross-section DF image obtained in two beam conditions with the (002) operating reflection from the structure with three annealed InGaAs QD layers.

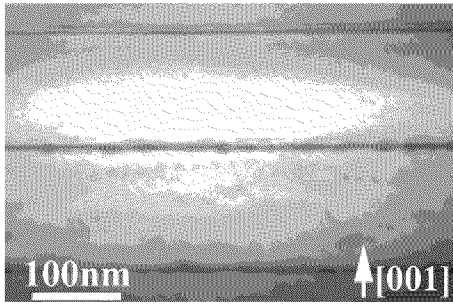


Fig. 2. The (110) cross-section DF image obtained in two beam conditions with (002) operating reflection from the structure with three InGaAs QD layers with intermediate annealings.

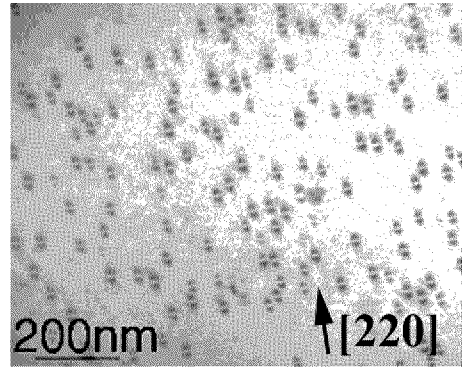


Fig. 3. The (001) plan-view BF image obtained in two beam conditions with (220) operating reflection from the structure with three InGaAs QD layers with intermediate annealings.

QDs having ~ 11 nm height according to our observations. Under annealing with growth interruption In migrates from the top of pyramidal QD and distributes on the GaAs surface because of a high surface migration ratio of In. The changing QD shape is due to In migration and diffusion processes. Indium redistribution within an annealed layer is also the reason for dislocation disappearing on the interface of wetting layer and an island.

TEM study of the structure with three InGaAs QD layers separated by GaAs spacers and post-growth annealing revealed the developed defects presented all over the InGaAs QD layer-GaAs spacer system (Fig. 1). Three-dimensional dislocated islands were the origin of these defects starting with the first InGaAs QD layer. The presence of QDs was confirmed by the characteristic contrast which presented at the TEM images. The QD height is about 13 nm. Considerably decreasing QD density from the bottom layer up to the top one as well as a failure of planar growth are observed in (110) cross-section images. It is believed to be due to the influence of defects in the bottom InGaAs QD layer which accumulate the subsequent InGaAs deposit. The observed defects propagate all three layers and suppress In redistribution within each layer to eliminate defects under post-growth annealing. The InGaAs QD sheets showing dark contrast separated by 125 nm GaAs layers were observed in the (110) cross section image of the structure grown with intermediate annealings of each QD layer (Fig. 2). The characteristic contrast from QD presents in the images of each layer. The average QD density, lateral size and height calculated for the upper layer from the (001) plan-view image (Fig. 3) are $1.5 \times 10^{10} \text{ cm}^{-2}$, 19 nm, 10 nm respectively. One

can observe thin layers with dark contrast at the 10–11 nm distance from the QD sheets. These thin layers are believed to be InAs layers (Fig. 2). Comparing the distance from QD sheet to the observed thin InAs layer, the QD height and the GaAs spacer thickness specified by the growth regime we consider the changing QD shape and the thin InAs layer appearance to be the effect the In redistribution from QD tops onto the GaAs spacer surface during annealing after incomplete QD covering by GaAs spacer.

3. Conclusion

To summarize the main results we can formulate the following conclusions:

- In the case of InGaAs deposition at 480°C without an additional treatment at elevated temperature both perfect QDs and large three-dimensional dislocated islands are formed;
- Intermediate annealing after incomplete covering of QDs by GaAs spacer leads to defect elimination;
- The developed defect structure exists in the system consisting of three InGaAs QD sheets separated by GaAs spacers where annealing was applied after the deposition of all three periods;
- Applying growth-annealing procedure for each QD sheet results in perfect QD system formation with maintaining crystal quality of the whole structure.

Thus the intermediate annealings during InGaAs QD growth in GaAs matrix by MOCVD method improve the QDs and whole structure quality and can be introduced in QD-InAs/GaAs laser structure fabricating process.

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