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Structural Characteristics on InAs Quantum Dots multi-stacked on GaAs(100) Substrates

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Abstract: The InAs self-assembled quantum dots (SAQDs) were grown on a GaAs(100) substrate using a molecular beam epitaxy (MBE) technique. The InAs QDs were multi-stacked to have various layer structures of 1, 3, 6, 10, 15 and 20 layers, where the thickness of the GaAs spacer and InAs QD layer were 20 monolayers (MLs) and 2 MLs, respectively. The nanostructured feature was characterized by photoluminescence (PL) and scanning transmission electron microscopy (STEM). It was found that the highest PL intensity was obtained from the specimen with 6 stacking layers and the energy of the PL peak was split with increasing the number of stacking layers. The STEM investigation exhibited that the quantum dots in the 6 stacking layer structure were well aligned in vertical columns without any defect generation, whereas the volcano-like defects were formed vertically along the growth direction over 10 periods of InAs stacking layers.

Keywords: self-assembled quantum dots, molecular beam epitaxy, photoluminescence, scanning transmission electron microscopy, volcano-like defect

초 록: 분자선에피택시법에 의하여 GaAs(100) 기판 위에 InAs 자발형성양자점을 성장하였다. InAs 양자점은 1, 3, 6, 10, 15 및 20층 등으로 다양하게 적층되어졌고, GaAs 층과 InAs 양자점 층은 각각 20 MLs와 2 MLs의 두께를 갖도록 하였다. 이 양자점의 나노구조적 특성은 PL과 STEM을 사용하여 분석하였다. 가장 높은 PL 강도는 6층의 적층구조를 갖는 시편에서 나타났고 PL 피크의 에너지가 적층회수가 증가함에 따라 분리됨을 알 수 있었다. STEM 분석결과, 6층의 적층구조에서는 결함이 거의 없이 수직으로 형성된 구조를 보여준 반면에 10층 이상의 적층구조를 가질 때 그 성장 방향에 따라 volcano형상을 갖는 결함이 수직하게 성장되어졌다.

1. Introduction

InAs/GaAs heterostructures (lattice mismatch of 7%) are of practical interest because of their optoelectronic application such as quantum dot (QD) lasers, QD spectrometer, QD infrared photodetector, spectral hole burning memory, single electron tunneling devices and so on.[1-4] Self-assembled QDs using the Stranski-Krastanow (S-K) growth mode have the advantage of high density but it has been known that the control in size, density and position of QDs is difficult. Therefore, it is necessary to control the growth condition of QDs to have high density, size uniformity and position for device applications. Multilayered InAs QDs are useful for laser applications because the increased effective number of dots increases the modal gain and helps to overcome gain saturations.[5] Although the InAs QDs were grown in vertical columns, unpredictable structural defects and the stoppage of QDs' formation were frequently found. For instance, it can be seen that surface depressions are formed above InAs islands.[6-8] A volcano-like defect due to the gradient of surface chemical potential was found.[9-10] In addition, long stacking faults (LSF), which are originated from the interface and extended to the surface, and short stacking faults (SSF), which are originated at the interface but ended inside the film, were also reported.[11] Therefore this study is aimed to find the optimized stacking condition for QDs layers using a MBE. Scanning transmission electron microscopy (STEM) and photoluminescence (PL) measurement were used for characterization of the QDs grown.

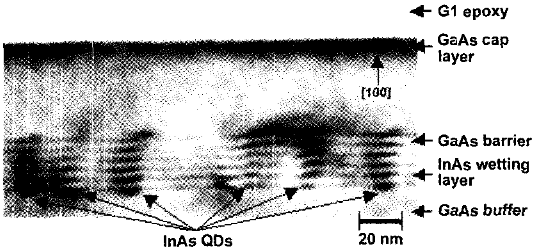

Fig. 1. Cross-sectional STEM micrograph of vertically aligned InAs QDs structure formed on GaAs(100) substrate.

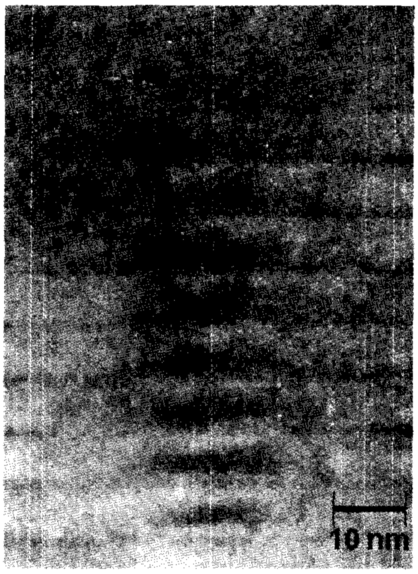

Fig. 2. High resolution STEM image of multilayered InAs quantum dots.

2. Experimental Procedures

The samples were grown by the V8OH molecular beam epitaxy (MBE) system. The substrates were cleaned using trichloroethylene (TCE), acetone, methanol and etched using sulfuric acid etchant (H_2SO_4:H_2O_2:H_2O=8:1:1) before inserting in the growth chamber of the MBE system. The structure is consisted of a 500 nm thick GaAs buffer layer, and then a 2 MLs of InAs/a 20 MLs thick GaAs layer superlattice, finally a 74 nm GaAs cap layer was grown. The InAs QD layers also had different periods of 1, 3, 6, 10, 15 and 20. The growth temperature was 580°C for the buffer GaAs layer and lowered to 500°C for the InAs and GaAs superlattice layer. The growth rates were kept at 1 ML/sec and 0.14 ML/sec for GaAs and InAs, respectively. In the case of InAs QD layer, the growth of alternative 0.14 ML of InAs and interruption of 5 sec was performed. The formation of QDs was verified by changing from streaky to spotty RHEED patterns.[12] Microscopic observation was performed using a high-voltage Philips CM30 (300 kV) electron microscope. Photoluminescence (PL) data were obtained using an Ar^+ laser (λ=514 nm). The luminescence was detected by a liquid nitrogen cooled Ge detector. For measuring temperature-dependent PL data, all PL measurements were performed at 16-300 K, with a pumping power of 0.2 W for the Ar^+ laser.

3. Results and Discussion

Fig. 1 shows the cross-sectional STEM image of 6 periods of InAs islands with no associated dislocation generation which was grown vertically along the growth direction. The origin of vertical ordering of QDs is due to the QDs in upper layer preferably aligned on the place where the strain induced by the QDs in lower layer is strongly localized. Since the heavier In atoms than Ga atoms have the increased scattering amplitude caused by incident beam, InAs QD is visible darker contrast than GaAs spacer. The observed InAs QDs are shown in the verically aligned lens-shaped STEM contrast. Atomic force microscopy (AFM) measurements indicate that the dot size is 18 nm in width to the in-plane direction and also show that the in-plane dot size does not change when island layers are added.[13]

The cross-sectional STEM image shown in Fig. 2 represents that InAs islands have vertically aligned up to the 8th layer in 10 periods of InAs layers and then entirely stopped. Although the island is approximately 4-5 nm in height, it is difficult to measure accurately. Because at the dot peaks only a small number of InAs atoms represent in cross section to contribute to the

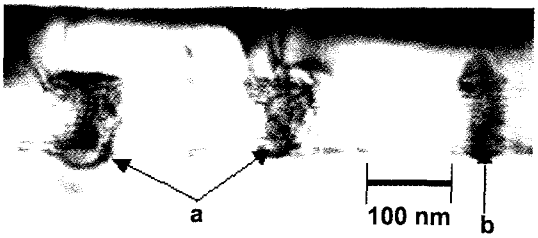

Fig. 3. Cross-sectional STEM micrograph of vertically grown volcano-like defects formed on GaAs (100) substrate.

contrast, and besides, the observed cross-section might not cross the dot centers.[14] The kinetic processes giving the vertically self-organized growth behavior are also represented appropriately by other researchers.[15]

Fig. 3 shows the cross-sectional STEM image of volcano-like defects vertically grown in 10 periods of InAs layers formed on GaAs(100) substrate. There are irregularities in the grown multilayered InAs structure, indicated by arrow "a" and the formed defects are increased with increasing the layers. It is seen that the incomplete volcano-like defect is generated between the defects arrowed by "a". The formation of complete volcano-like defects is marked by "b".[9-10] The volcano-like defect started to form through the sinusoidal profile of small amplitude due to the gradient in surface chemical potential and then gradually evolved into a cusp-like morphology.[9-10] The generation of abnormal defect is likely to be originated from the growth parameters as growth temperature, deposition rate, misfit strain, and stress concentration.[16] It is observed that the formation of islands is stopped as the growth proceeds. On the whole the formation of islands was inhibited because of the relaxation of strain field effect due to larger generated volcano-like defects.

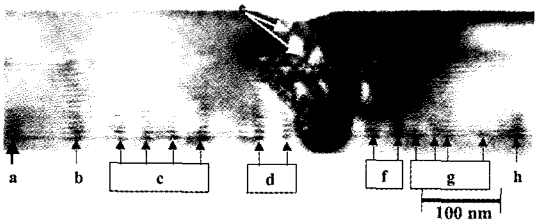

Fig. 4. Cross-sectional STEM micrograph of InAs QDs with partially vertical columned structure and vertically grown defect of asymmetrical down triangle shape formed on GaAs(100) substrate.

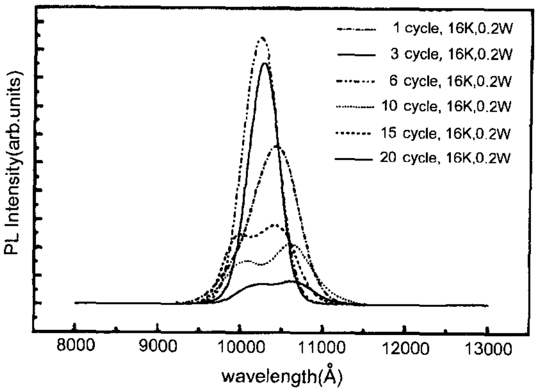

Fig. 5. Relationship of relative intensity vs stacking cycle.

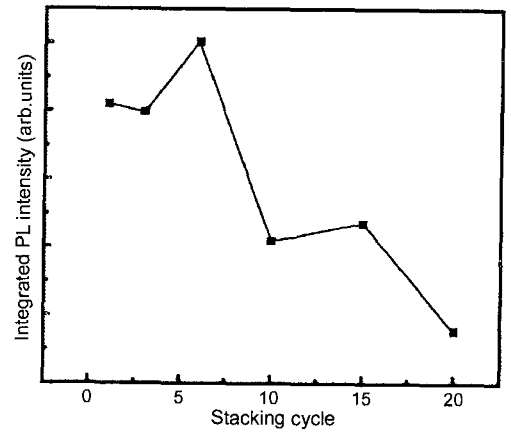

Fig. 6. Relationship of integrated PL intensity vs stacking cycle.

Fig. 4 shows the cross-sectional STEM micrograph to be coexistent with vertically columned InAs QDs structure and vertically grown defect of asymmetrical down triangle shape (ADTS). In the case of b, InAs islands were grown up to 15 periods. However, in the case of a, c, g and h, the formation of vertically columned islands was only grown up to 5-7 periods. Therefore, we found that the QDs were well vertically formed up to 6-7 layers in 15-stacked structure. Region e presents the threading dislocation into GaAs cap layer due to the strain field of defect edges, and hence the cap layer was depressed. Interestingly, InAs SAQDs/GaAs superlattice structure (regions d and f) can be seen obviously outside the defect of ADTS at which SAQDs were only formed below the 6-7 periods of the layers though. Regions d and f also present that the stoppage of QDs' formation results from defects.

Fig. 5 shows PL spectra with various stacking cycle at 16 K. It is observed the 6 periods of InAs stacking layers provide the highest peak and the narrowest full width at half maximum (FWHM). In the PL spectra it is also noted that the degradation of PL peak intensity over 10 periods of InAs stacking layers seems to be resulted from defects formation which is elucidated by the STEM image.

From the dependence of PL integrated intensity on stacking cycle shown in the Fig. 6, the 6 periods of InAs stacking layers result in the increased effective number of dots.

4. Conclusions

The defect generation due to lattice mismatch of InAs/GaAs and the increase of layer at multilayered InAs QDs structure was investigated. It was shown that vertically aligned islands were grown up to 6-7 periods in each multi-stacked structure. However, volcano-like defects due to the gradient of surface chemical potential and asymmetrical down triangle shaped defects were observed when the stacking cycle were over 6-7 periods. Therefore, it is essential that the defects creation decrease as well as the effective number of dots increase by increasing stacking cycle.

Acknowledgments

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