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TEM Study on the Growth Characteristics of Self-Assembled InAs/GaAs Quantum Dots

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ABSTRACT

Self-assembled InAs/GaAs quantum dots (QDs) were grown by the atomic layer epitaxy (ALE) and molecular beam epitaxy (MBE) techniques. The structure and the thermal stability of QDs have been studied by high resolution electron microscopy with in-situ heating experiment capability. The ALE and MBE QDs were found to form a hemispherical structure with side facets in the early stage of growth. Upon capping by GaAs layer, however, the apex of QDs changed to a flat one. The ALE QDs have larger size and more regular shape than those of MBE QDs. The QDs collapse due to elevated temperature was observed directly in atomic scale. In situ heating experiment within TEM revealed that the uncapped QDs remained stable up to 580°C. However, at temperature above 600°C, the QDs collapsed due to the diffusion and evaporation of In and As from the QDs. The density of the QDs decreased abruptly by this collapse and most of them disappeared at above 600°C.

Key words : Atomic Layer Epitaxy (ALE), InAs/GaAs Quantum dot, Molecular Beam Epitaxy (MBE), Thermal Stability, Transmission Electron Microscopy (TEM)

INTRODUCTION

Quantum dot (QD) devices have superior optoelectrical properties such as high current gain, low threshold voltage and superior thermal stability (Arakawa & Sakaki, 1982; Marzin et al., 1994). Thus, considerable efforts have been devoted to fabricate QDs lasers (Eaglesham, 1990), advanced memories (Shoji et al., 1995) and infrared photodetectors (Yusa & Sakaki, 1995) by using self-assembled QDs. To understand the optoelectronic properties of QDs devices, information on the structures and growth characteristics of QDs is needed since the quantum effect of QDs originates from nanometer scale structures. In addition, for high quality QD growth, process parameters effects such as growth temperature, growth rate, InAs thickness, etc. on the growth characteristics of QDs should be understood carefully and quantitatively. Growth temperature in particular is very important for the precise control of QD structure, because the diffusion of In and As atoms depends significantly on temperature. Since the surface diffusion of In and As atoms plays a critical role in determining the QD structure, understanding the structural behavior of QDs at high temperature and the thermal stability is important. However, determination of the

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structures and growth characteristics of QDs is not an easy task because of the small size and embedded structure of QDs.

There have been various analyses techniques, such as atomic force microscopy (AFM), scanning tunneling microscopy (STM) (Snyder et al., 1991) and reflection of high energy electron diffraction (RHEED) (Yamaguchi et al., 1996), for the determination of QDs structures. Those techniques, however, suffer varying degrees of imprecision because of the small size of QDs and AFM technique is not capable of resolving their shape owing to well known tip convolution effects. In addition, the structure of QDs changes after capping due to the strain between QDs and matrix (Lian et al., 1998). Those techniques can be applied to imaging of QDs prior to capping matrix. Transmission electron microscopy (TEM) is a unique analysis technique for the determination of the structures of QDs capped by matrix (Miller et al., 2000). The TEM cross-sectional view and plan-view techniques are able to analyze the structure of QDs in various directions, and atomic scale analyses are also possible by high resolution electron microscopy (HREM).

In the present study therefore the structure and thermal stability of InAs QDs grown on GaAs substrate by atomic layer epitaxy (ALE) and molecular beam epitaxy (MBE) deposition techniques have been studied. High resolution electron microscopy (HREM) and high voltage electron microscopy (HVEM) equipped with in-situ heating capability were used in this study. The QDs were found to form a hemispherical structure with side facet in the early stage of growth, and then changed to a flat one upon capping by GaAs. In situ HREM revealed that the uncapped QDs remained stable until 580°C.

MATERIALS AND METHODS

InAs QDs were grown on semi-insulating GaAs (001) substrates in a molecular beam epitaxy system by means of ALE and MBE techniques. In order to analyze the effect of capping on the change of QD structure, 2 period stacked QDs were grown and TEM analyses were performed. After QDs were formed in the first period, a 120 nm thick GaAs capping layer was deposited after which the second QDs were grown without capping. The growth characteristics and thermal stability of QDs were studied using a 200 kV HR-TEM (JEM-2010F) and 1.25 MV HVEM at the Korea Basic Science Institute (KBSI). The thermal stability of QDs was evaluated using a heating specimen holder in the HVEM at a temperature range of 25 ~ 600°C. TEM specimens were prepared by mechanically polishing and dimpling down to a thickness less than 5 μm and then ion milling by Ar^+ on precision ion polishing system (GATAN, PIPS). The analyses on compositional variation were carried out by means of energy dispersive spectroscopy (OXFORD, ISIS). TEM images were recorded on image plates with 0.25 μm pixel size. Through Fast Fourier Transformation (FFT) and inverse FFT processes, the QDs structure HREM resolution was improved enough to measure the size of QDs using digital intensity profiling.

RESULTS AND DISCUSSION

1. Structures of ALE and MBE QDs

Compressive strain is usually induced on the InAs QDs and tensile strain to the GaAs matrix in InAs and GaAs heteroepitaxy systems with 7.2% lattice misfits. This strain induces a dark contrast on the QDs during TEM observations using diffraction contrast as well as phase contrast. Fig. 2 shows cross-sectional TEM bright field (BF) images of ALE (a) and MBE QDs (b), respectively. The HREM images of uncapped QDs grown by ALE (c) and MBE growth (d), and capped QDs by ALE (e) and MBE growth (f), respectively. The 2 period stacked QDs were grown successively without any defects. Spindleshaped strong dark contrast was observed on and beneath the wetting layer due to strain formed along the wetting layer. The ALE QD height was identified as ~5 nm and the lateral width was ~23 nm along the [$\bar{1}$10] direction and MBE QD height and width were ~4 nm and ~18 nm, respectively. The QDs were distributed randomly and the average spacing between QDs was about 30~50 nm. The HREM images also revealed that the uncapped QD showed a round apex structure with side facet, while the capped QD had a flat apex structure. The QD shape change was possible because of the compressive strain induced to the QD by the GaAs matrix followed by In atom diffusion during high temperature growth (Ledentsov et al., 1996). HREM images, observed within [110] zone and [100] zone, confirmed that the shape and size of QDs were nearly identical, suggesting an axial symmetrical structure.

2. In situ heating observation of QDs

In order to observe the structural behavior of QD at high temperature and to evaluate thermal stability, in situ heating TEM analysis was performed within a HVEM. Fig. 3 shows [110] zone HREM images with FFT patterns of ALE QD structure as temperature increased. Fig. 3(a) shows the HREM of uncapped QD with FFT pattern taken from QD at RT and (b), (c), (d) show the HREM images at 300, 450 and 470°C, respectively. The QD height and diameter were 5.7 nm and

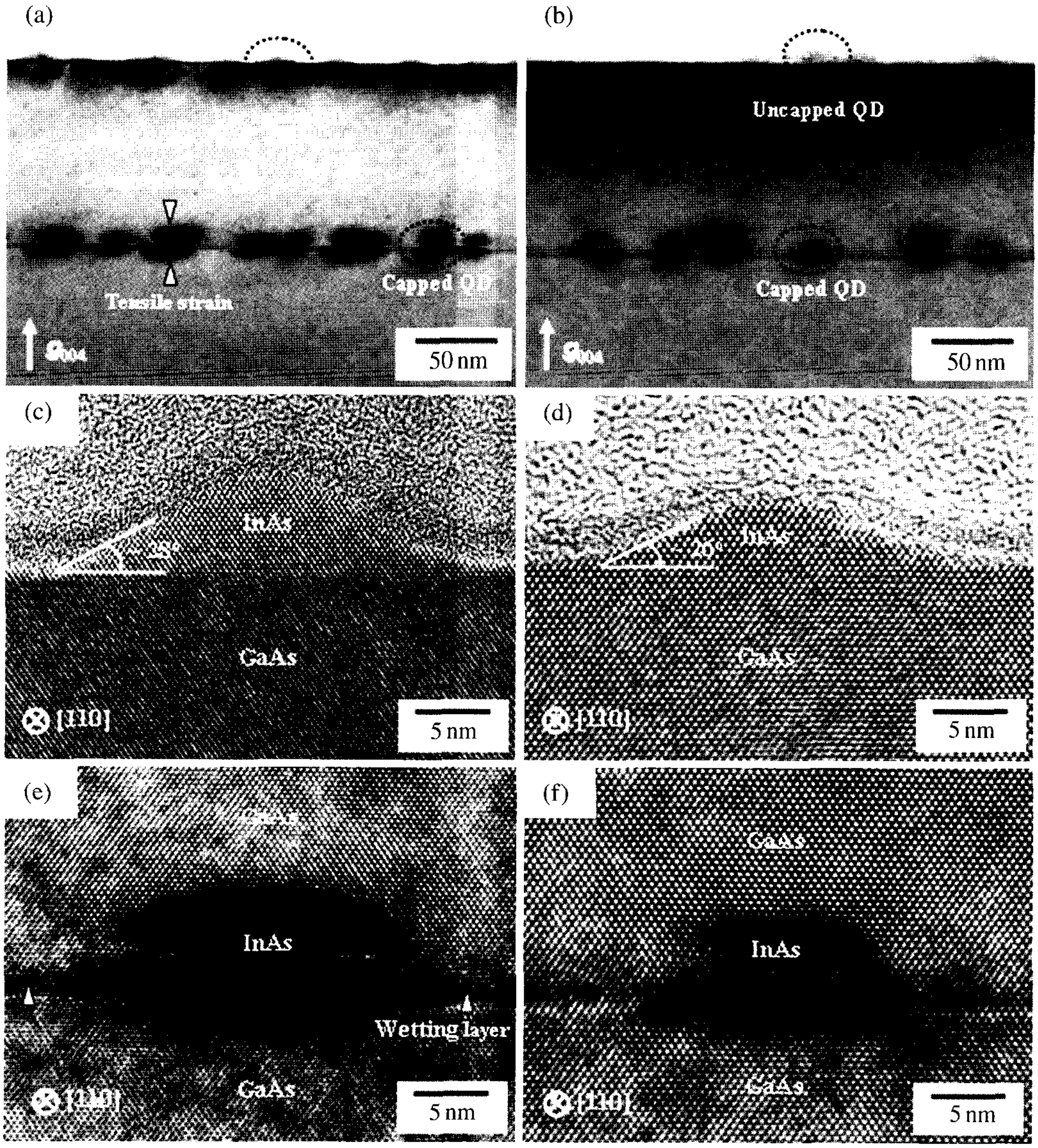

Fig. 1. Cross-section TEM micrographs showing the microstructure of InAs/GaAs QDs: low magnification bright field (BF) images under g_{004} two beam condition by ALE growth (a) and MBE growth (b). HREM images of uncapped ALE QD (c) and MBE QD (d), capped ALE QD (e) and MBE QD (f) in the [110] zone, respectively.

25.8 nm at RT. The change of QD size was small until 300°C. However, the height and diameter decreased to 2.0 nm and 19.5 nm at 450°C and crystalline defects occurred at the interface between QD and substrate. The HREM images and FFT patterns show the degradation of crystallity and the development of amorphization as temperature increased. Fig. 3 (d) shows that amorphization formed at the interface developed at 470°C. As the

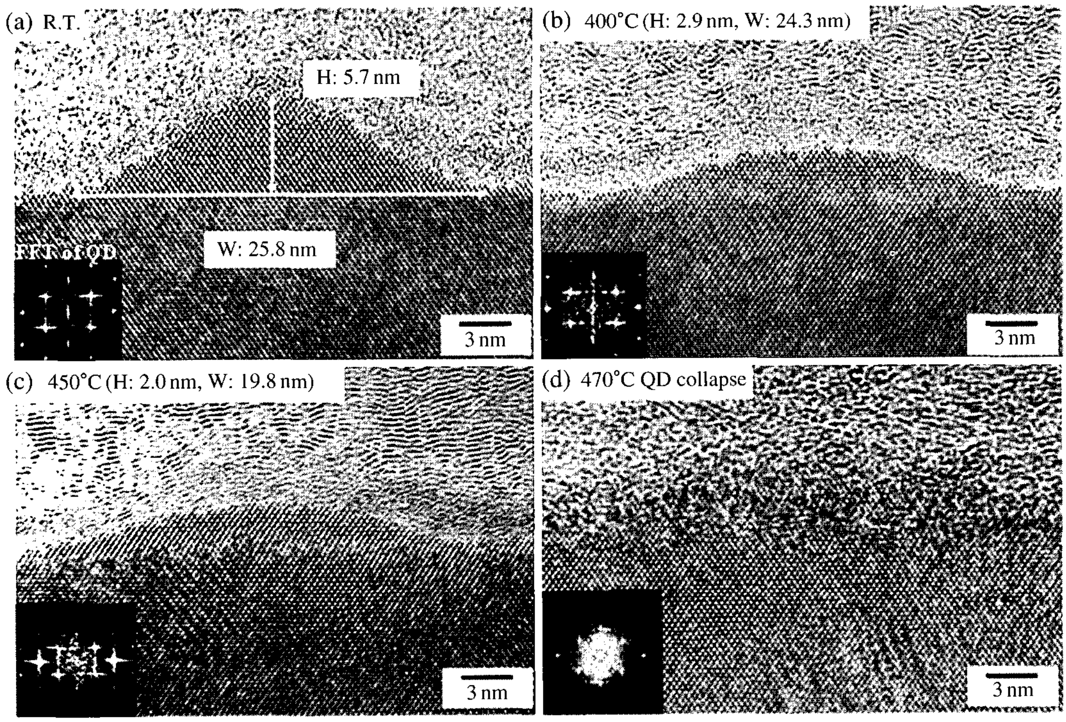

Fig. 2. In situ heating TEM observation for the evaluation of QD thermal stability: HREM images of uncapped QD with a height of 5.7 nm and a width of 25.8 nm at R.T. (a), high temperature HREM images of QDs at 300°C (b), 450°C (c) and 470°C (d), respectively.

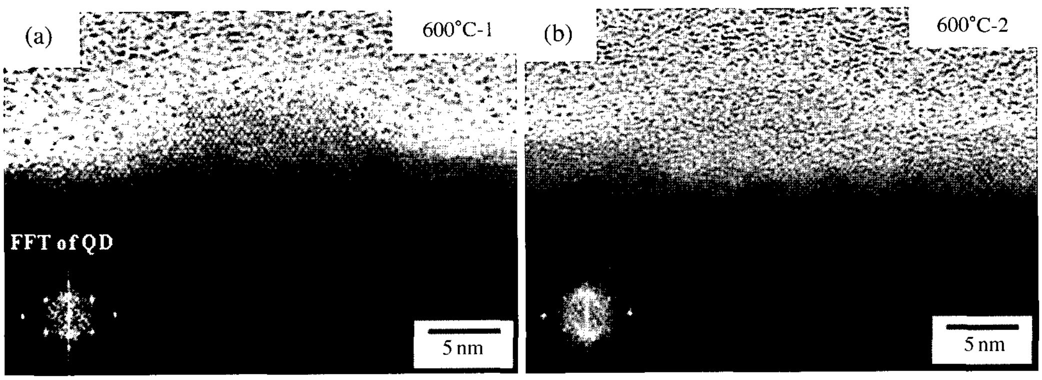

Fig. 3. In situ heating HREM images of ALE QD under minimized e-beam irradiation at 600°C: QD truncation (a) and QD collapse by InAs diffusion (b), respectively.

result of amorphization, the QD collapsed below the QD growth temperature of 480°C. The electron beams which was accelerated by the high voltage of 1.25 MV induced irradiation damage such as amorphization and evaporation during high temperature observation. In commonly used intermediate TEM acceleration voltages ranging from 100 kV to 400 kV, electron irradiation damage of semiconductors is not a major problem. However, a 1.25 MeV high energy electron beam can induce such irradiation damage, especially at the high temperatures.

In order to exclude the effects of electron beams, observations were performed in the thick region of specimen and the electron gun shutter was closed during increasing temperature. As a result of minimized e-beam condition, the ALE QDs remained stable until 580°C and electron beam irradiation defects did not form. The QD structure remained stable. However, the upper structure of QD changed to a flat one at 600°C. After flattening, InAs molecules diffused from QD to substrate and the QD collapsed. Fig. 3(a) shows the flattening of QD upper structure and Fig. 3(b) shows the collapse of QD by InAs diffusion at 600°C. The density of QDs decreased abruptly by this collapse and most of them disappeared at 600°C.

Fig. 4 also shows [110] zone HREM images of MBE QD at RT (a), 480°C (b), 580°C (c) and 600°C (d), respectively. The result of the thermal stability of MBE QD coincided with the case of ALE QD. The QDs remained stable until 580°C as shown in the in situ HREM images. However, QD apex changed to a flat one and the QD height decreased as temperature increased at above 580°C. After flattening, InAs molecules diffused from the QDs to the substrate and the QDs collapsed at 600°C as shown in Fig. 4(d). F. Heinrichdorff and D. Bimberg et al. have already reported that the optical properties of InGaAs/GaAs QDs remained constant until an annealing temperature of 580°C (Heinrichdorff et al., 1998). The photoluminescence spectra however shifted to a higher energy region above 610°C due to the intermixing of In and Ga atoms (Heinrichdorff et al., 1998). Therefore, it is confirmed that the irradiation of electron beams does not significantly affect the collapse of QDs. Nanobeam EDS revealed that the ratio of As to In and Ga, measured as approximately 48 : 52 at RT, decreased slightly to 46 :

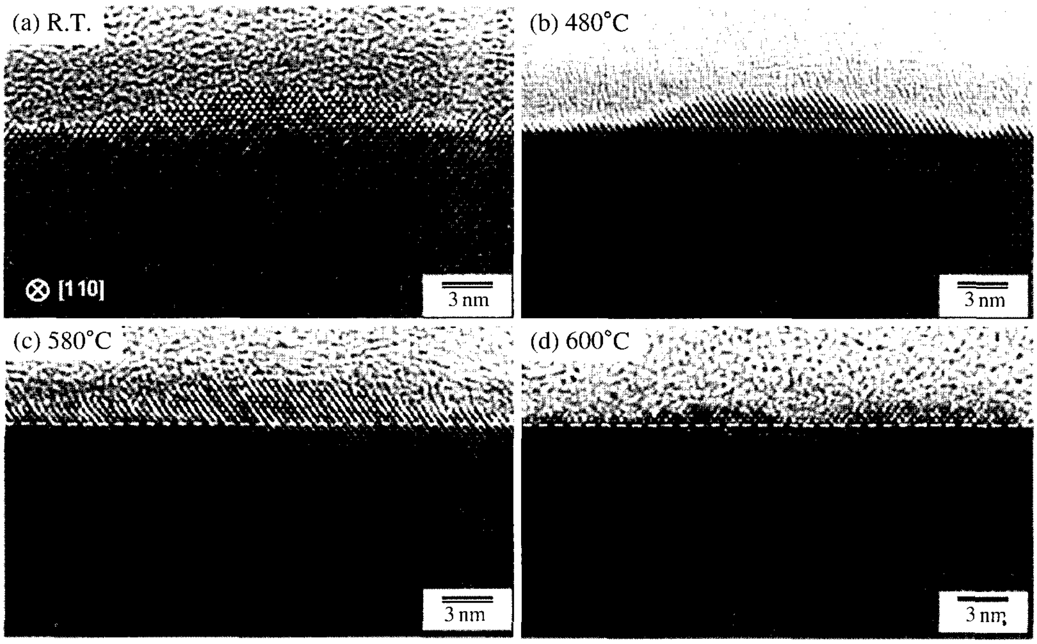

Fig. 4. In situ heating TEM observation for the evaluation of the thermal stability of MBE QD: The HREM images of uncapped MBE QD under [110] zone at R.T. (a), 480°C (b), 580°C (c) and 600°C (d), respectively.

54 in the QD region after heating. The QDs collapsed due to the diffusion and evaporation of In and As from the QDs above 600°C.

SUMMARY

Self-assembled InAs/GaAs quantum dots (QDs) were grown by the ALE and MBE deposition techniques and the structure and thermal stability of QDs have been studied. The ALE and MBE QDs were found to form a hemispherical structure with side facets in the early stages of growth. Upon capping by GaAs layer, however, the apex of QDs changed to a flat one. The ALE QD has larger size and more regular shape than those of MBE QD because of the lower vapor pressure and higher diffusivity of In compared to the InAs in MBE growth. In order to observe the structural behavior of QDs at high temperature and to evaluate thermal stability, in situ heating TEM analysis was performed in the HVEM. The uncapped ALE and MBE QDs remained stable until 580°C. At temperature above 600°C, the QDs collapsed due to the diffusion and evaporation of In and As from the QDs. The density of the QDs decreased abruptly by this collapse and most of them disappeared at above 600°C.

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