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Improved Luminescence Efficiency of InAs Quantum Dots Grown on Atomic Terraced GaAs Surface Prepared with In-situ Chemical Etching

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Observation of the enhanced luminescence efficiency of InAs quantum dots(QDs) grown on atomically controlled GaAs surfaces is reported. With the TDMAAs in-situ surface etching process, formation of atomic steps and terraces on GaAs surfaces were clearly observed. InAs QDs grown on the processed GaAs surfaces showed the clear dependence of QDs size, density and optical characteristics on the surface properties, i.e., the increase of the QDs

height and diameter the decrease of the QDs density. About 6-times enhancement of photoluminescence efficiency which has the peak around 1550-nm wavelength was observed by growing InAs QDs on atomically controlled GaAs surfaces. This is due to the migration enhancement of InAs during growing the QDs.

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1 Introduction

Quantum dots (QDs) have been studied for fundamental physics and many applications such as single electron devices and single photon sources. Up to now, QDs defined by electrostatic potentials applied on surface-patterned electrodes have been used for manipulations of single electron transports like Coulomb blockades. However small energy separations of their QD states requires extremely low-temperature operations below 1K. Higher-temperature operations have been reported by forming direct contact electrodes to smaller sized QDs such as InAs QDs prepared on GaAs surfaces by Stranski - Krastanow (S-K) growth mode [1]. This kind of non-buried (open) InAs QDs are also capable of emitting photons in the 1550-nm optical fiber communication (OFC) band because of release of internal stresses from their open surfaces. In general it is difficult to obtain the long wavelength emissions for embedded InAs QDs unless the strain is compensated [2-4]. Thus the emission wavelength of open InAs QDs is useful for OFC band but their luminescence efficiencies

are lower than those of buried QDs due to the sensitivity to the QDs surface conditions. The combination of the two functions will open the possibilities to control both single electrons and single photons with the same individual QDs. The key issue toward this direction will be the improvement of their luminescence efficiencies.

In this paper, we will demonstrate that luminescence efficiencies of InAs open QDs can be drastically improved by in-situ control of GaAs surfaces with atomic level. Also the QDs size and density were changed by controlling the GaAs surface conditions. About 6-times enhancement of photoluminescence (PL) efficiency around 1550-nm wavelength was observed by growing InAs QDs on atomically controlled GaAs surfaces.

2 Experiments

In this study all QDs were prepared with metal-organic (MO) molecular-beam epitaxy (MBE) system. The MO sources used are triethylgallium (TEGa), triethylindium

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(TEIn) and trisdimethylaminoarsenic (TDMAs) for Ga, In, and As. The epi-ready (001) semi-insulating (SI) GaAs substrates were thermally cleaned at 560°C with the simultaneous injection of 1.0×10^{-4} Pa TDMAs beam equivalent pressure (BEP). A 100-nm-thick GaAs buffer layer was grown with the TEGa and TDMAs supplies at 510°C. Then GaAs surface was treated using TDMAs etching effect[5] on GaAs at 560°C for 0 min. (non-processed), 5 min., 15 min., and 30 min. TDMAs BEP for the surface control process was 1.0×10^{-4} Pa. For the growth of InAs QDs, after the surface control process, the temperature was decreased to 480°C. With the TEIn and TDMAs supplies the formation of InAs QDs was identified by the change of the reflection high-energy electron diffraction (RHEED) patterns from streaky to spotty ones. After noticing the change, the growth was stopped and the substrate temperature was decreased to room temperature with the ramping rate of 30°C/min.

The QDs heights and density were measured with tapping-mode atomic force microscopy (AFM) and the QD diameters were measured with scanning electron microscopy (SEM).

PL measurements were carried out using the second harmonic generation of an yttrium aluminium garnet (YAG) laser at the wavelength of 532 nm. The sample temperature was cooled down to 20 K in a closed-cycle helium cryostat. PL signals were dispersed with a 1-m monochromator through an optical fiber. A liquid-nitrogen-cooled InGaAs detector was used to measure PL spectra.

3 Results and discussion

Figures 1(a) and 1(b) show the AFM images (1 $\mu\text{m} \times 1 \mu\text{m}$) of the non-processed GaAs surface and the one with the in-situ etching for 30 min, respectively. The vertical scale is 3 nm. Although atomic steps were not observed for the non-processed GaAs, they were clearly observed for the processed GaAs surface. The extension of the atomic terrace length was observed for GaAs surfaces which were processed for the longer time. These results are attributed to the TDMAs etching effect [5] of the GaAs buffer layer surface. Figures 2(a) and 2(b) show the AFM images (1 $\mu\text{m} \times 1 \mu\text{m}$) of the InAs QDs grown on the non-processed surface and processed one for 30 min., respectively. The vertical scale is 15 nm. The atomic terraces were observed for all the samples on the InAs wetting layer surface including the non-processed GaAs sample. The longer atomic terrace length was observed for the longer-time-processed samples which will be the reflection of the initial GaAs surfaces. Figure 3(a) shows the process-time dependences of the QDs height and diameter and fig. 3(b) shows the ones of the QDs density and the total volume. The QDs volume was estimated assuming that the QD shape is a cone and QDs total volume (effective thickness) is defined by the QDs density times the QD volume. The drastic change of QDs size and density were observed. The reduction of the average QDs density and the total volume from $1.2 \times$

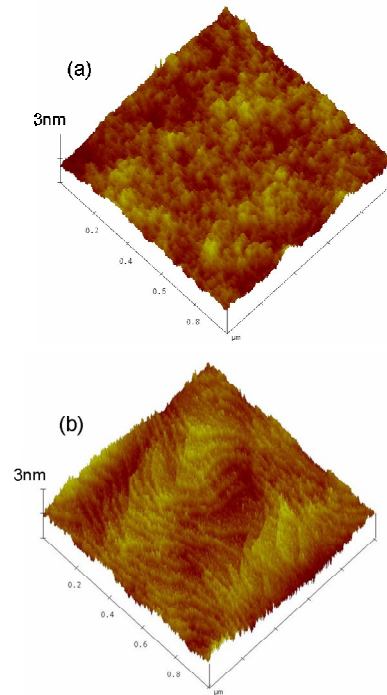


Figure 1 AFM images (1 $\mu\text{m} \times 1 \mu\text{m}$) of the non-processed GaAs (a) and processed for 30 min. GaAs surface, respectively. The vertical scale is 3.0 nm.

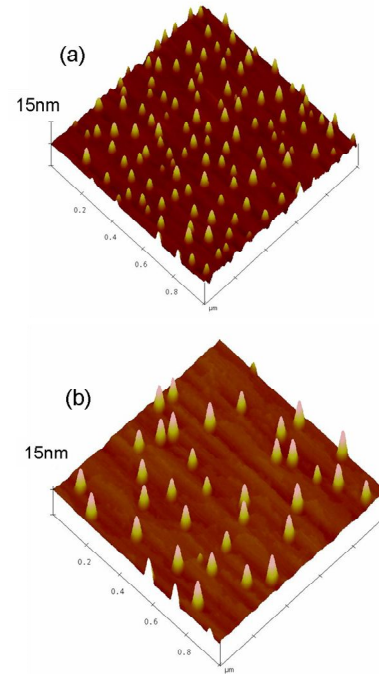


Figure 2 AFM images (1 $\mu\text{m} \times 1 \mu\text{m}$) of the InAs QDs grown on the non-processed GaAs (a) and processed for 30 min. GaAs surface, respectively. The vertical scale is 15.0 nm.

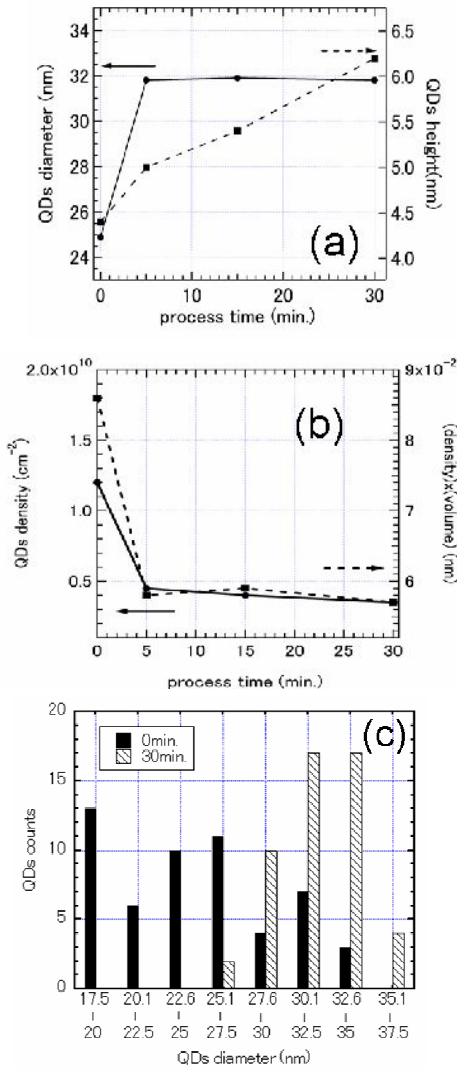


Figure 3 (a) InAs QDs diameter and height dependence on processed time and (b) shows the InAs QDs density and density times volume dependence on processed time, respectively. The InAs shape was assumed the cone. (c) shows the QDs diameter distribution for non-processed and 30min. processed QDs.

10^{10}cm^{-2} to $3.5 \times 10^9\text{cm}^{-2}$ and from 0.086nm to 0.059nm, respectively, were observed. On the other hand, the increase of the QDs height and diameter from 4.4nm to 6.2nm and 24.9nm to 31.8nm, respectively, were observed. These results show that each QD size was increased, while the QDs total volume was decreased. In addition to the QDs size changes, the QD diameter distribution change was observed. Figure 3(c) shows the QDs diameter distribution of non-processed QDs and processed for 30min., respectively. QD diameter was measured from 50 QDs. Although non-processed QDs show the broad distribution of the diameter, the processed QDs show the improved uniformity of the size distribution.

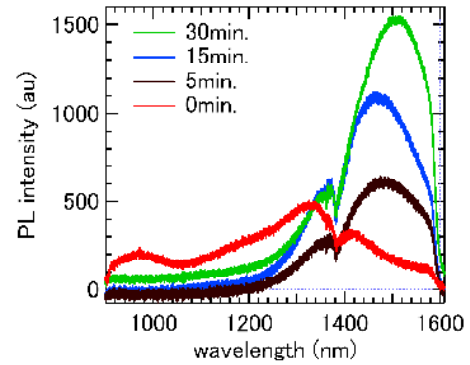


Figure 4 PL spectra of InAs QDs grown on differently processed GaAs, 0min., 5min., 15min. and 30min

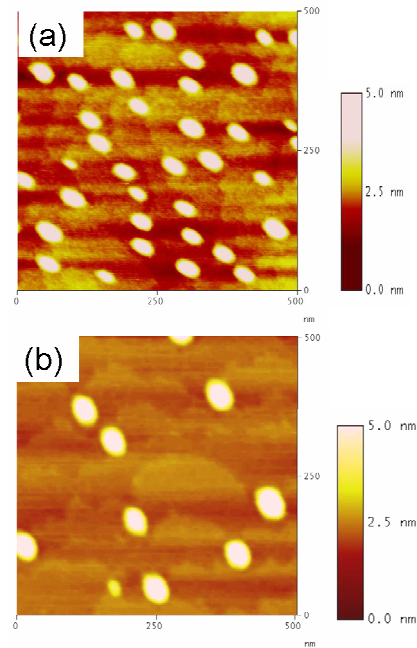


Figure 5 AFM images (500nm x 500nm) of the InAs QDs grown on the non-processed GaAs (a) and processed for 30min. GaAs surface, respectively.

The above observation of the QD sizes and densities will be summarized as follows: The increase of the QD sizes on the atomically terraced GaAs surfaces prepared with the in-situ etching process shown in Fig. 3(a) will be attributed to the enhanced surface migration on the terraced surface [6]. The enhanced surface migration on the terrace surface will result in the edge growth of the QDs [7] and also increase the probability of coalescence of QDs, which eventually will result in the increase of the average QD size. This will naturally lead to the decrease of the QD density shown in Fig. 3(b) and the improved homogeneity of the QD size distribution shown in Fig. 3(c). The decrease of the averaged total QD volume shown in Fig. 3(b) indicates the increased desorption rate of the In adatoms during the enhanced migration on the terraced surfaces.

Figure 4 shows the PL spectra of InAs QDs grown on non-processed, 5min. 15min. and 30min. processed surfaces. The sharp spectral dip observed at around 1400nm is not intrinsic but due to the OH absorption in the optical fiber used for the measurements. Although broader PL spectra extending to the shorter wavelength were observed for the non-processed QDs, red-shifted narrower emission peaks at the wavelength of ~ 1500 nm were observed for the QDs grown on the processed GaAs. In addition to the emission peak red-shift, the enhancement of the PL intensity was observed. The PL intensity at around 1500nm was enhanced about 6-times by the in-situ processing of the GaAs surfaces.

It is well-known that the emission wavelength is depend on the QD size and smaller QDs emit shorter wavelength[8] due to the confinement size effect. The broader PL spectrum extending to the shorter wavelength observed with the non-processed QDs shown in Fig. 4 corresponds the larger inhomogeneity of the QD diameter distribution extending to the smaller diameter range shown in Fig. 3(c). PL intensity in usual cases will be proportional to the QDs density. However the maximum improvement of the PL intensities by ~ 6 -times at around the wavelength of 1500nm shown in Fig. 4 was observed from the sample with the reduced QD density by $\sim 1/3$ or the reduced QD volume by $\sim 1/5$ prepared with the longer processed time of 30 min. This indicates that the enhancement of the PL efficiencies is due to the improvement of the QDs optical quality.

The expanded view of the AFM images is show in Fig. 5 to examine the relation between the grown QDs and the surface terraces and atomic steps. Figures 5(a) and 5(b) show the InAs QD samples grown on the non-processed GaAs surface and the one in-situ etched for 30 min., respectively. In Fig. 5(b), most of the QDs shows the growth initiation from atomic step edges. Although no terraced structure was observed on the non-processed GaAs surface, it is observed after the growth of InAs as shown in Fig. 5(a), i.e., the growth of the InAs wetting layer changed the surface to the terraced one. InAs QDs grown on this surface is smaller and some of the QDs are situated on the terraced surface isolated from the atomic steps. This will be related to the grown strained layer which changes into the preferential growth of islands on the terrace surface [6]. The surface migration on the initial GaAs surface without terraced structures as well as the suggested modification of the surface migration processes of adatoms will be related to the larger QD size distribution and may result in the different optical quality of the grown InAs QDs. QDs were grown on the step edges and on the terraces. On the other hand, the processed QDs were grown at the step edges. As shown above the processed GaAs have longer atomic terrace. Then it was expected that the enhancement migration was occur before growth the QDs. These phenomena agree with the QD size and density change. It is important that the presence of atomic steps and longer terraces influence the QDs size, density and luminescence efficiencies. It is expected that the more high luminescence efficiencies may

be obtained from QDs grown on the GaAs surface which have longer terrace.

4 Conclusions

We demonstrated the enhancement of the luminescence efficiency of InAs open QDs when they were grown on the surface prepared into the atomically controlled GaAs surface with in-situ chemical process. The in-situ TDMAAs etching of GaAs surfaces showed clearly longer atomic steps. The increased QD size and reduction of the density and diameter distribution were observed for processed QDs. These changes influence the emission wavelength and PL efficiency of InAs QDs, the red-shift of the emission wavelength and sharper PL spectra. The enhancement of the PL intensity was observed due to the improvement of the QDs quality. These changes were due to the enhancement of the migration effect on the terraced structure. Although the processed QDs were grown at the step edge, non-processed QDs were grown on the terrace and at the step edge. It is concluded that the preparation and control of GaAs surfaces with atomic-level configuration is important to realize the high optical quality of the grown InAs QDs.

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