Photoluminescence studies of GaAs quantum dots grown by droplet epitaxy

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Abstract

We have investigated post-annealing effects of photoluminescence (PL) properties in GaAs/AlGaAs QDs fabricated by modified droplet epitaxy. The annealing temperatures were changed between 520°C and 760°C. The PL intensity of QDs increased drastically with the increase of annealing temperature. The PL intensity of QDs after the annealing at 760°C was enhanced by two orders of magnitude as compared to that of before post-annealing. This sample showed a distinct PL peak even at the room temperature. With the increase of annealing temperatures, the peak energy shifted from 1.646 to 1.749 eV, continuously. These effects may be caused by improving the crystallinity of QDs systems and the size reduction and/or changing the composition of QDs by the post-annealing.

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1. Introduction

Recently, various fabrication methods for semiconductor quantum dots (QDs) structures have been reported [1–11]. Because of high structural crystallinity, high density and simple growth procedures, self-organized QDs grown by the Stranski–Krastanov (S–K) growth mode [4] have become extremely attractive in the case of a lattice-mismatched system such as InAs/GaAs system. On the other hand, in a lattice-matched system such as GaAs/AlGaAs, only a method termed “droplet epitaxy” was proposed for the self-organized formation of QDs [1–3]. But unfortunately, a strong photoluminescence (PL) from QDs grown by original droplet epitaxy was not observed.

Recently, we have proposed a novel modified droplet epitaxy method using a high As flux irradiation at a low substrate temperature for the self-organized formation of high-quality GaAs/AlGaAs QDs system, and observed the clearly PL of the QDs [5]. In this paper, for the purpose of further improvement of PL intensity, we investigated post-annealing effects of the GaAs/AlGaAs QDs system fabricated by this method.
2. Experiment

The process was carried out with a Riber-32P molecular-beam epitaxy (MBE) system with elemental sources and an EPI-valved As cracking source. Although only uncracked As was used in this study, the valve enabled us to rapidly irradiate high As$_4$ flux. After the desorption of native oxides on a GaAs (001) wafer, a 300 nm thick GaAs buffer layer and a 500 nm thick Al$_{0.3}$Ga$_{0.7}$As barrier layer were grown at 580°C. Then the substrate temperature was reduced to 180°C and the As valve was closed. Ga droplets were deposited by the Ga-molecular beam supply without an As flux. The Ga flux was equivalent to the GaAs growth rate of 0.5 monolayers (MLs)/s. The supplied total Ga amounts was 2.75 or 4.75 MLs. Next an As$_4$ molecular-beam with the flux of $2 \times 10^{-4}$ Torr was irradiated on the surface. After the complete change of reflection high-energy electron diffraction (RHEED) pattern from halo to transmission spots, an Al$_{0.3}$Ga$_{0.7}$As barrier layer with a 10 nm thickness was grown by migration-enhanced epitaxy (MEE) [12] at the same temperature to prevent the two-dimensional regrowth of naked GaAs microcrystals [5]. Then the samples were again heated to 580°C and a 90 nm thick Al$_{0.3}$Ga$_{0.7}$As barrier layer and a 10 nm thick GaAs cap layer were also grown by ordinary MBE. Small pieces cut from the sample grown from supplied Ga total amount of 4.75 MLs were reloaded into the growth chamber, and annealed under As$_4$ atmosphere of $1.5 \times 10^{-5}$ Torr at the temperatures ranged from 520°C to 760°C for 60 min. The surface and cross section of the buried sample were observed with a field-emission-type high-resolution scanning electron microscope (HRSEM). For PL measurements of samples, an Ar-ion laser was used as an excitation source and the spectra were observed by a GaAs photodetector through a spectrometer. The laser beam diameter was about 0.8 mm and its power was 2 mW.

3. Results and discussion

Surface morphologies before the growth of AlGaAs overlayer were observed by a HRSEM, as shown in Fig. 1. Total amounts of supplied Ga were 2.75 MLs (a) and 4.75 MLs (b). Numerous pyramidal-shaped microcrystals were observed. The typical base size of microcrystals were 10 nm $\times$ 15 nm (a) and 16 nm $\times$ 20 nm (b). The both density was almost same as $\sim 1 \times 10^{10}$ cm$^{-2}$. Fig. 2 shows PL spectra of the buried microcrystals before post-annealing. Distinct peaks were observed around 1.78 eV (a) and 1.65 eV (b), and indicate the clear blue shift of 0.13 eV with the decreasing size of the microcrystals. Then it is clear

Fig. 1. Surface morphologies before growth of AlGaAs overlayer observed by a HRSEM. Total amounts of supplied Ga were 2.75 MLs (a) and 4.75 MLs (b).
that these peaks originated from GaAs QDs, and also this was confirmed by the magneto-photo luminescence [13]. The full-widths at half maximum were 86 meV (a) and 103 meV (b). The peak shape of (a) seems a gauss one, indicating that the size distribution of QDs remains the original size distribution of Ga droplets even after overgrowth. In the case of (b), there were a sharp shoulder on lower energy side and a broad one on higher. Almost flat surface appeared during heating process after a first 10 nm AlGaAs overgrowth. If the height of QDs is larger than this thickness, additional height of QDs might be diffused by heating process, appearing rectangular cross-sectional shape dots. Then, majority dots height became same. Therefore, the sharp shoulder might appear on lower energy side by the limitation of the confinement for perpendicular direction. The broad one on higher energy side might reflect the droplets distribution as same as the case of (a).

Fig. 3 shows the post-annealing temperature dependence of the PL spectra in the GaAs/AlGaAs QDs grown from the supplied total Ga amounts of 4.75 MLs measured at 20 K. These PL properties remarkably depend on the annealing temperatures. The integrated PL intensity, peak energy and full-widths at half maximum of these spectra are summarized in Figs. 4(a)–(c), respectively. The PL intensities increase drastically with increasing the annealing temperature. The intensity after the annealing at 760°C was enhanced by two orders of magnitude as compared to that of before post-annealing. Additionally, in the case of this sample, the strong emission of the QDs is successfully observed even at room temperature as shown in Fig. 5. The FWHM increased with the annealing temperature.

Fig. 6(a) and (b) show HRSEM images of a cross section of the buried QD after stain etching for the samples before and after post-annealing at 680°C for 60 min, respectively. It is revealed that the base size and density of the finally buried QDs before post-annealing were almost the same as those before the growth of the overlayer. However, the size of the QDs after post-annealing became smaller than that of the QDs before post-annealing.

During the post-annealing process, the inter-diffusion of Ga and Al between GaAs QDs and
AlGaAs barrier proceeded. The effective size of the QDs decreased and/or GaAs QDs changed to AlGaAs QDs. As results, the blue shifts of the PL peaks were observed. It is reported that the interdiffusion of Ga and Al began to become significant for temperatures above 800°C for hetero-interface of GaAs/AlGaAs grown by normal MBE growth [14]. In our case, even by the post-annealing at 520°C, the blue shift of this QDs was obviously observed, indicating the interdiffusion in the QD structure. In this droplet epitaxy, it is necessary that GaAs QDs and the upper thin AlGaAs barrier layer were grown at 180°C [5]. Generally, it is well known [15] that excess As are incorporated into low temperature MBE growth layer. By this excess As, the interdiffusion may be easily promoted under the post-annealing processes, improving the crystallinity and increasing the PL intensity of the GaAs/AlGaAs QD systems. Also the interdiffusion may result in reducing the size and/or changing the composition of QDs, shifting the PL peak to higher energy. The size distribution of QDs may become large by reducing the average size of QDs with the increase of annealing temperature. By this droplet epitaxy with the post-annealing process, the high-quality self-organized GaAs/AlGaAs QDs structure was successfully grown, resulting the intense PL even at room temperature.

4. Conclusions

We have investigated the post-annealing effects of the PL properties in GaAs/AlGaAs QDs fabricated by modified Droplet Epitaxy. The annealing temperatures were changed between 520°C and 760°C. The PL intensity of QDs increased drastically with the increase of annealing...
temperature. The sample annealed at 760°C for 60 min showed the PL peak even at the room temperature. The peak energy shifted to higher energy with increasing the annealing temperatures. These effects may be caused by improving the crystallinity of QDs systems and the size reduction and/or changing the composition of QDs by the annealing.

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References