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Fabrication of InGaAs quantum dots on GaAs(0 0 1) by droplet epitaxy

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Abstract

We have proposed a new self-organized growth method for InGaAs quantum dots (QDs) using droplet epitaxy with highly dense Ga droplets. During the crystallization process of InGaAs in droplet epitaxy, the highly dense Ga droplets effectively prevented the two-dimensional growth of InGaAs. Phase-separation during annealing for the InAs–GaAs system resulted in the formation of high-quality InGaAs QDs in the upper part of the sample with a flat surface. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Quantum dot (QD) lasers have attracted much attention, since excellent properties such as low threshold current density are expected [1]. So far, many groups have reported InGaAs QDs grown on GaAs by the Stranski–Krastanov (S–K) growth mode [2–4]. However, the threshold current densities of QD lasers fabricated by the S–K mode are higher than the predicted value, [5,6] probably due to the large size distribution of QDs.

We have recently proposed a new self-organized growth method for InGaAs quantum dots (QDs) without a wetting layer on GaAs(0 0 1) substrates using droplet epitaxy with highly dense Ga droplets [7]. The highly dense Ga droplets successfully prevented the two-dimensional growth of InGaAs during crystallization under the As flux supply. These QDs showed very narrow photoluminescence peaks (linewidth: 21.6 meV) at 946 nm due to the uniform size of InGaAs QDs, which mainly originated from the size uniformity of In droplets. The size of the InGaAs QDs estimated from the Bohr radius of the exciton is smaller than that estimated from the droplet size [7]. This result has not been explained by only the restriction effect of highly dense Ga droplets for the two-dimensional growth

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of InGaAs. In this paper we have investigated the detailed growth mechanism in this method, especially in the annealing process.

2. Experimental procedure

The molecular beam epitaxy (MBE) system used in this work was a conventional system (RIBER 32P) with reflection high-energy electron diffraction (RHEED). Elemental Ga, In and As were used as the molecular beam source materials. GaAs buffer layers with 0.5 μm thickness were grown on GaAs(0 0 1) substrates at the substrates temperature of 580°C. Flat As-adsorbed c(4 × 4) surfaces were formed by reducing the substrate temperature to 200°C. Droplet formation was carried out by the following deposition processes on the c(4 × 4) surfaces at 200°C without As flux (back ground pressure was kept under 1×10^{-9} Torr): (1) 1.75 MLs of Ga (for compensating the 1.75 MLs excess As on GaAs-c(4 × 4) surface [8]), (2) 2.5 MLs of In (for In droplets), and (3) 50 MLs of Ga (for highly dense Ga droplets). For the droplet formation, the deposition rate of In and Ga were set at 3 and 1.5 MLs/s, respectively, that were determined from the periods of RHEED specular beam intensity oscillation. After the formation of the droplets, As flux was supplied for the crystallization of the droplets to III–V compound semiconductors as in the original droplet epitaxy [9]. Then, the substrate temperature was raised up to 500°C and annealed with As flux until the RHEED pattern was changed to streaky (for 40 min). Finally, 0.1 μm GaAs capping layers were grown by migration-enhanced epitaxy [10] at 500°C. For the PL measurement, an Ar^+ ion laser

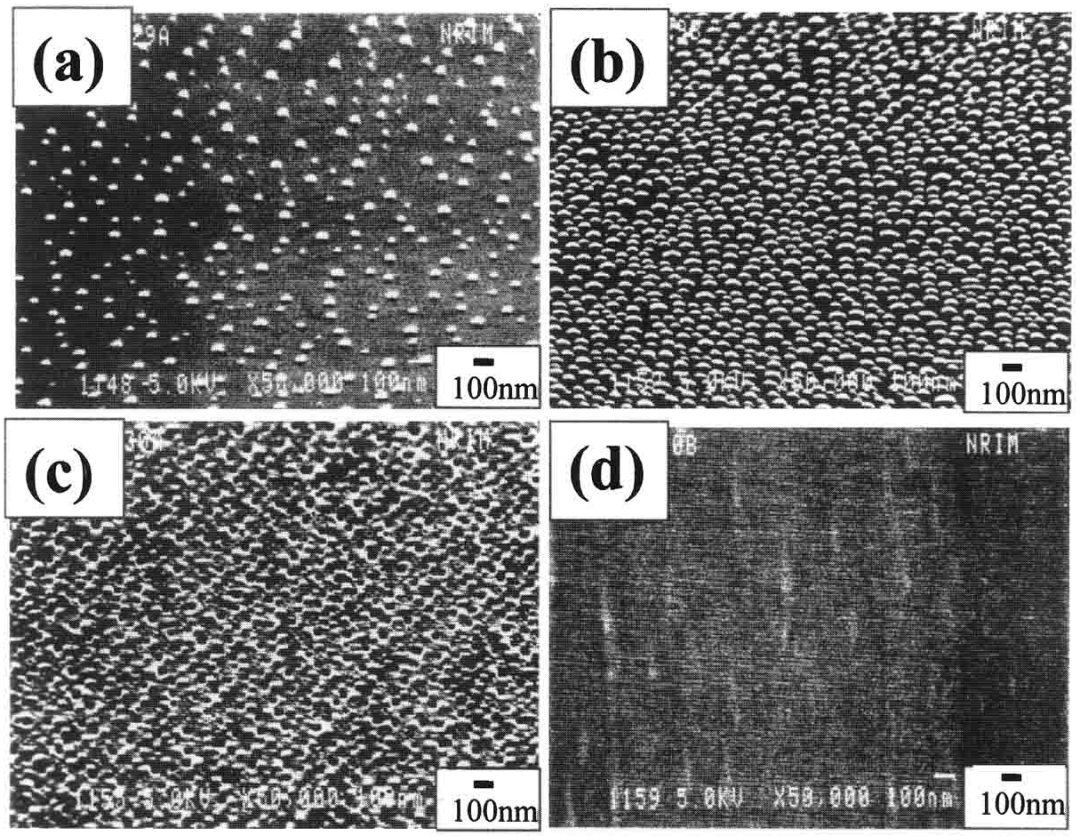

Fig. 1. HR-SEM images of the samples: (a) and (b) are images of droplets before and after the supply of 50 MLs Ga, respectively; (c) and (d) are the images before and after the annealing process, respectively.

was used as an excitation source at the wavelength of 488 nm with the power density of 10 mW cm^{-2}, and the data were obtained by a cooled InGaAs photo-detector through a spectrometer.

3. Results and discussion

Fig. 1 shows the high-resolution scanning electron microscope (HR-SEM) image of the samples; (a), (b), (c) and (d) are images before the supply of 50 MLs Ga, after the supply of 50 MLs Ga, before the annealing process, and after the annealing process, respectively. The InGa droplets with highly dense Ga droplets were formed by the supply of 1.75 MLs Ga, 2.5 MLs In (a), and 50 MLs Ga (b), sequentially. Comparing the density of the droplets before and after the supply of 50 MLs Ga, it increased from 3×10^9 to 2×10^{10} cm^{-2}. Provided the In atom rearrangement does not occur during the 50 MLs Ga deposition, 15% of the deposited Ga might be incorporated in the initially deposited In droplets, forming $In_{0.25}Ga_{0.75}$ droplets with the density of 3×10^9 cm^{-2}. After the As flux was supplied to these droplets at 200°C, the RHEED pattern changed from halo to spotty, which indicated the crystallization of the droplets. During the procedure, highly dense Ga droplets restricted the two-dimensional growth area for InGaAs [7]. From the HR-SEM observation of this sample, crater-like structures were observed, as shown in Fig. 1c. This phenomenon can be explained in terms of As diffusion in liquid droplets. When the InGa or Ga droplets are exposed to As flux, GaAs or InGaAs is formed at the periphery of the droplets predominantly. At the bottom of the droplets, the As flux was supplied through the liquid droplets [8]. However, at the periphery of the droplets, As flux was directly supplied, as shown in Fig. 2a. Since the diffusion rate of the As in the droplets might be slow, the growth rate at the bottom of the droplets is slower than that at the periphery of the droplets, forming crater-like structures shown in Fig. 2b. After annealing, the crater-like structures become flat in order to reduce the surface energy, shown in Fig. 1d.

Figs. 3a, b and c show the PL spectra measured at 20 K for the samples before and after annealing

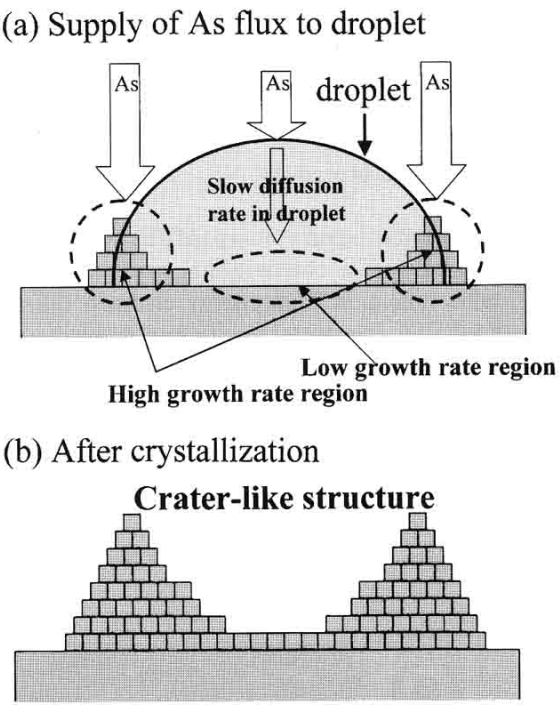

Fig. 2. Schematic illustration of the InGaAs crystallization process of the droplets.

without capping layers and after the growth of a capping layer, respectively. Before the annealing, only the luminescence from the substrates was observed at 818, 830 and 920 nm, which probably corresponded to a GaAs bulk transition, a transition related to carbon impurities, and other impurities, respectively. After the annealing, the weak luminescence from the QDs appeared at 1000 nm. From these results, it is confirmed that the crystalline properties were improved and/or the InGaAs QDs were formed. The PL intensity from the capped sample is about 100 times stronger than that of the uncapped sample. This fact means that the QDs are formed at the upper part of the flat layer with nearly bare surface, because the surface states of QDs might weaken the PL intensity. Moreover, the PL peak energy shifted from 1000 nm (1.24 eV) to 946 nm (1.31 eV) after capping, which was due to the compressive strain in the QDs caused by the GaAs capping layer [11]. The size of the QDs estimated from magneto-PL measurements of the

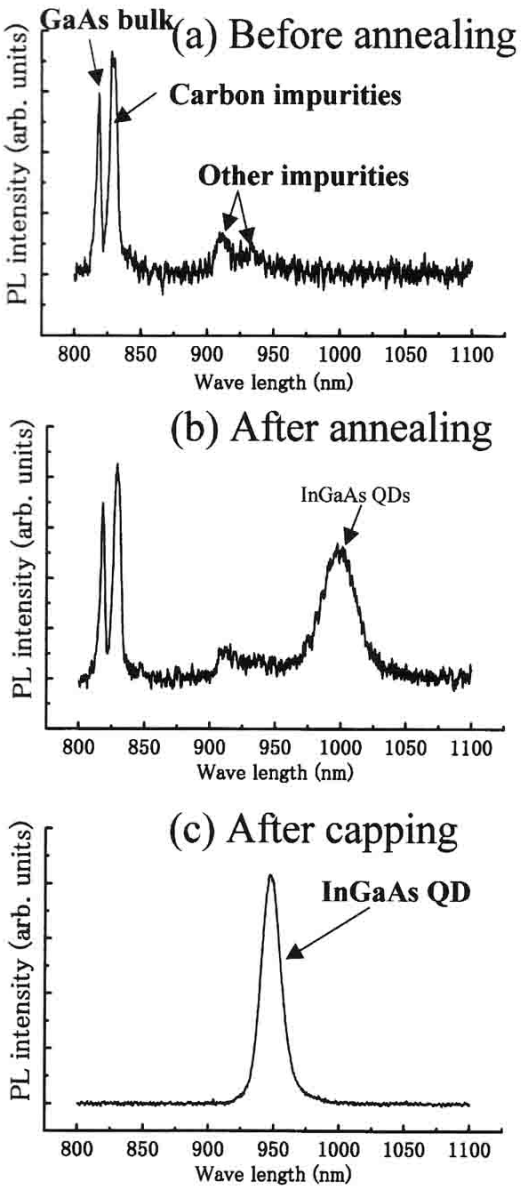

Fig. 3. PL spectra of the samples: (a) before annealing; (b) after annealing; (c) after the growth of a capping layer.

QDs [12] was about 10 and 4 nm in plane and in vertical directions, respectively.

From these results, the formation mechanism for InGaAs QDs was understood, as shown in Fig. 4 schematically. First, the highly dense Ga droplets restrict the two-dimensional growth of the InGaAs

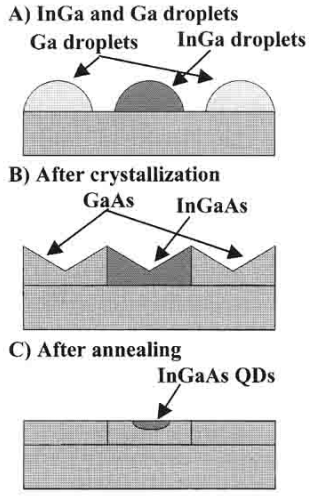

Fig. 4. Schematic illustration of the SPEED method for the fabrication of InGaAs QDs: (a) InGa droplets with highly dense Ga droplets; (b) after the crystallization of droplets by the As flux supply; (c) after annealing.

(a), (b). Second, during the annealing at 500°C, the surface becomes flat to reduce the surface energy. At this time, the phase-separation effect must be enhanced because the composition of InGaAs is in the miscibility gap for the InAs–GaAs system at the annealing temperature of 500°C [13]. The volume of each crystallized InGaAs region is calculated to be 7×10^4 nm^3, which is estimated from a product of the thickness of 52.5 MLs and the inverse of the density for the droplets (2×10^{10} cm^{-2}). However, the volume of a QD is roughly estimated to be 4×10^2 nm^3 from the Bohr radius of the exciton, which was deduced from the magneto-PL measurements [12]. This discrepancy suggests that the segregation of the InGaAs occurs during annealing caused by phase-separation. Also from the PL measurements, it is confirmed that the high-quality QDs were formed due to the annealing process. Because the detailed mechanism of segregation is not clear, quantitative discussion of In content of the QDs cannot be done in this paper. However, this phase-separation mechanism is the essential part of this method. Therefore, we refer to this modified droplet epitaxy method as separated-phase enhanced epitaxy with droplets (SPEED).

4. Conclusions

InGaAs QDs were grown by droplet epitaxy with highly dense Ga droplets by a method termed SPEED. From the HR-SEM and PL measurements, it is indicated that the phase-separation for InAs–GaAs might be enhanced, forming InGaAs QDs in a flat surface, during the annealing process. These QDs show strong PL with narrow linewidth. These results indicate that the SPEED method is a promising method for the fabrication of InGaAs QDs with high quality.

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