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New Self-Organized Growth Method for InGaAs Quantum Dots on GaAs(001) Using Droplet Epitaxy

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Quantum dot (QD) systems for InGaAs/GaAs without a wetting layer have been fabricated on GaAs (001) surfaces by a new self-organized growth method using droplet epitaxy with highly dense Ga droplets. Droplets of InGa alloy with highly dense Ga droplets have been formed by supplying 1) Ga, 2) In and 3) Ga molecular beams, sequentially. These highly dense Ga droplets have successfully prevented the two-dimensional growth of InGaAs during crystallization under As flux supply. In the plan-view transmission electron microscope image, the InGaAs QDs with the density of $7 \times 10^9\,\mathrm{cm}^{-2}$ are observed. These QDs show a very sharp photoluminescence peak (full width half maximum (FWHM): 21.6 meV) at 946 nm.

KEYWORDS: quantum dots, InGaAs, droplet epitaxy, photoluminescence, transmission electron microscope

1. Introduction

Quantum dots (QDs) have attracted a great deal of attention because of their predicted applications such as in advanced semiconductor lasers with highly monochromatized light and low threshold current density[1)] and enhanced electron mobility devices.[2)] Many groups have reported the fabrication of InGaAs QDs on GaAs by a method of self-organized growth for example Stranski-Krastanov (S-K) mode, caused by the lattice mismatch between the substrate and the overlayer.[3–6)] Although very strong luminescence peaks were observed from the InGaAs S-K dots, the peak widths were very broad.[7,8)] Also, the threshold current densities of the QDs lasers were higher than the predicted values.[9,10)] These might be due to the wide size distribution of the QDs.

On the other hand, for a lattice-matched system, such as InSb/CdTe, GaAs/ZnSe and GaAs/AlGaAs, it has been shown that droplet epitaxy is a promising method for fabricating uniform-size QDs.[11–15)] This droplet epitaxy method is based on V-column-element incorporation into the III-column-element droplets deposited on the III–V (or II–VI) compound semiconductor substrate. Since the size of the III-column-element droplets is uniform, the formation of uniform QDs is expected. So far, we have applied droplet epitaxy to the fabrication of InGaAs QDs in a lattice-mismatched system. However, our preliminary results on the fabrication of InGaAs QDs indicate that the S-termination process, which is useful for preventing two-dimensional growth during crystallization in both the original[13)] and modified droplet epitaxy,[16)] caused the degradation of the optical properties.[17)]

In this paper, we propose a new modified droplet epitaxy method with additional highly dense Ga droplets for the fabrication of uniform InGaAs QDs. The properties of the QDs were investigated using a high-resolution scanning electron microscope (HR-SEM), plan-view transmission electron microscope (TEM), and photoluminescence (PL) measurements.

2. Experimental

GaAs buffer layers with 0.5 μm thickness were grown on GaAs(001) substrates at the substrate temperature of 580°C in a conventional molecular beam epitaxy system with a reflection high-energy electron diffraction (RHEED) system. Flat As-stabilized $c(4 \times 4)$ surfaces were formed by reducing the substrate temperature to 200°C after the growth of the buffer layer. Droplet formation was carried out by the following deposition process on the $c(4 \times 4)$ surfaces at 200°C without As flux (background pressure was kept below 1×10^{-9} Torr): 1) 1.75 ML of Ga (for covering the As-terminated surface with Ga), 2) 2.5 ML of In (for In droplets), and 3) 50 ML of Ga (for highly dense Ga droplets). The deposition rates of In and Ga were set at 3 ML/s and 1.5 ML/s, respectively, determined from the periods of RHEED specular beam intensity oscillation. On the $c(4 \times 4)$ surfaces, there was 1.75 ML of excess As.[18)] Therefore, 1.75 ML of Ga was supplied before droplet formation to prevent the formation of a two-dimensional strained layer which might cause complicated phenomena such as the S-K mode in this method. After the formation of the droplets, As flux was supplied to crystallize the droplets into III–V compound semiconductors. Then, the substrate temperature was raised to 500°C and annealed for 40 min with As flux. Finally, GaAs capping layers were grown by migration-enhanced epitaxy at 500°C,[19)] in order to grow the high-quality capping layers at low growth temperatures.

3. Results and Discussion

Figures 1(a) and 1(b) show the surface morphologies before and after the supply of 50 ML of Ga, respectively. Numerous hemispherical In droplets are observed in Fig. 1(a). The density of the In droplets is $3 \times 10^9\,\mathrm{cm}^{-2}$. The average base size and the size distribution of the In droplets are 60 nm and 20%, respectively. After the supply of 50 ML of Ga, the size of the droplets increases and the density of droplets also changes to $2 \times 10^{10}\,\mathrm{cm}^{-2}$, as shown in Fig. 1(b). Near the initially deposited In droplets, some of the deposited Ga atoms might be incorporated into the In droplets, forming InGa alloy droplets, and other Ga atoms among the In droplets might form highly dense Ga droplets. This is due to the difference between the surface migration lengths for In and for Ga, which is confirmed by the fact that the density of Ga droplets without In is higher than that of In droplets formed under the

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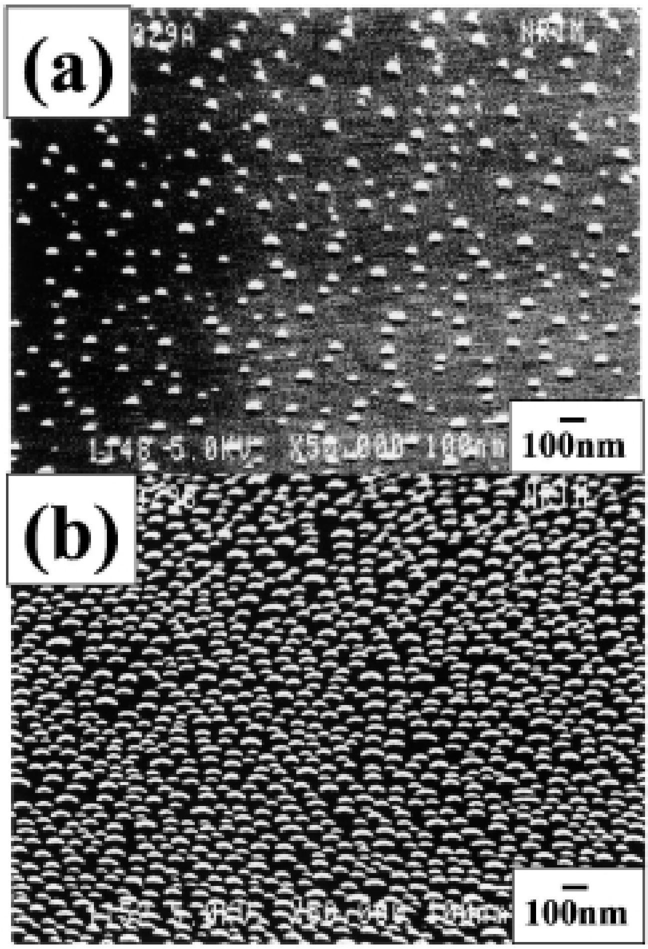

Fig. 1. HR-SEM image of droplets (a) before and (b) after the supply of 50 ML of Ga.

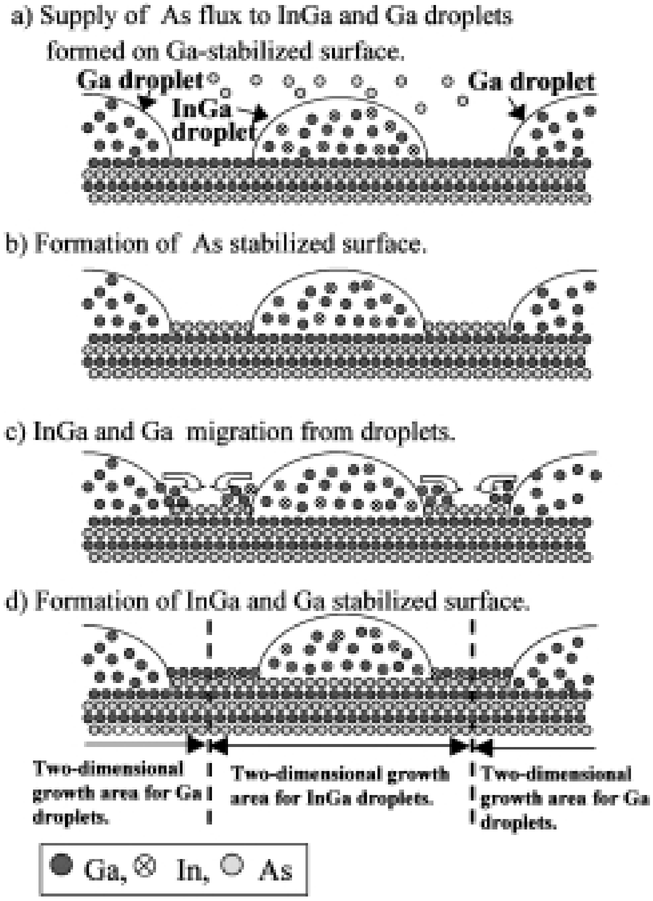

Fig. 2. The schematic model of InGa and Ga droplets during As molecular beam supply for crystallization.

same conditions as mentioned above.

After the crystallization and annealing process, not the S-K mode but a flat surface was observed. At this time, InGaAs QDs must be formed in this surface because of the restriction effect of the highly dense Ga droplets. Figure 2 shows the schematic model of the crystallization for InGa and Ga droplets during As molecular beam supply. The As flux is supplied to the InGa and Ga droplets formed on a Ga-stabilized surface (Fig. 2(a)). The impinging As atoms cover the entire area of surface (b). When the surface is covered with As, it is possible for In and Ga atoms to detach from the droplets and attach themselves to the nearest As atoms (c). Then, the surface will be completely covered with In or Ga (d). Processes (a)–(d) continue until the disappearance of droplets. It is worth pointing out that the highly dense Ga droplets restrict the area of two-dimensional diffusion of the InGa atoms from the droplets, forming the InGaAs QDs. For comparison, the same procedure, except for the supply of 50 ML Ga, was performed. In this case, the S-K mode, which indicates the two-dimensional growth of InAs, was observed. A flat surface was observed after annealing which is necessary to obtain a strong PL emission from the QDs, as mentioned below.

Figure 3 shows the bright-field plan-view TEM image of the uncapped samples with 50 ML of Ga after the crystallization and the annealing process. Strong contrast originating from the strain field of InGaAs QDs with the density of $7 \times 10^9\ \mathrm{cm}^{-2}$ was observed in this image. The density value agrees well with the density of In droplets, as shown in Fig. 1(a), indicating that the origin of InGaAs QDs is InGa droplets.

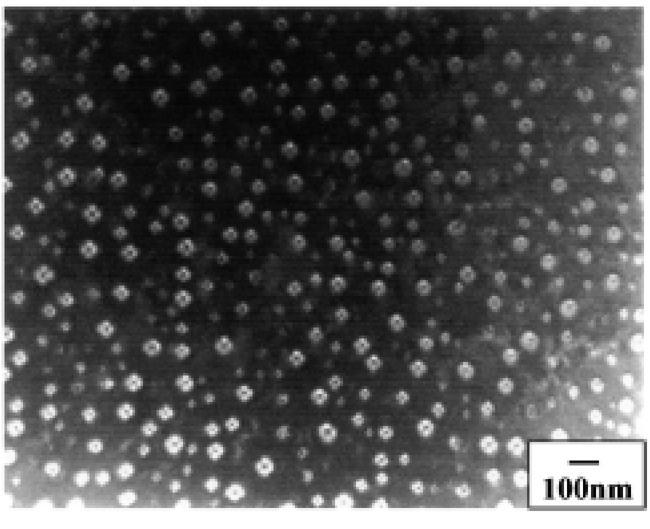

Fig. 3. Plan-view TEM image of InGaAs QDs after the crystallization and annealing process.

Figure 4 shows the PL spectrum of the GaAs-capped InGaAs QDs sample measured at 20 K. For the PL measurement, Ar^+ ion laser light of $10\ \mathrm{mW{\cdot}cm^{-2}}$ at 488 nm was used as an excitation source and the data were obtained by means

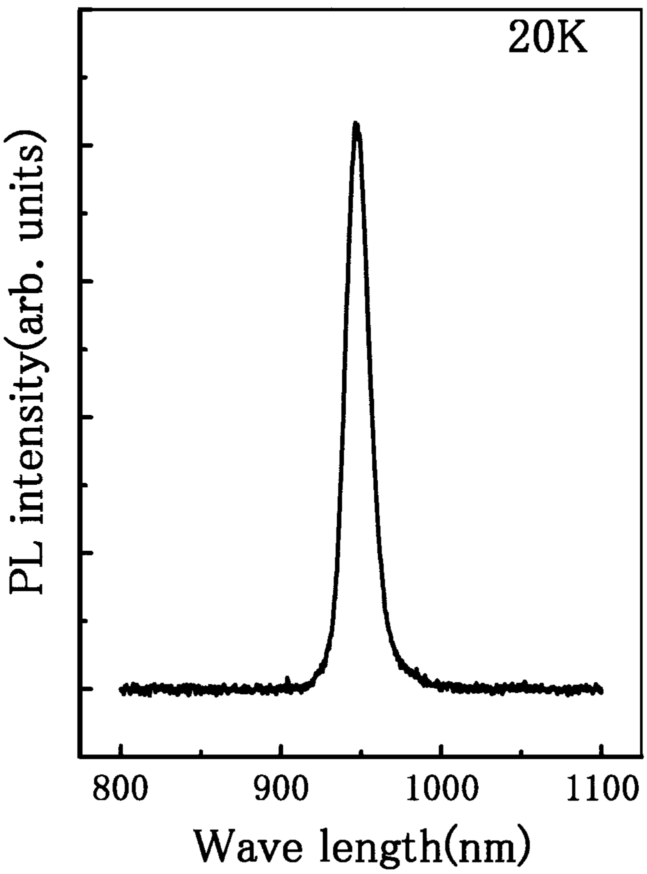

Fig. 4. Photoluminescence spectrum for capped InGaAs QDs at 20 K.

of a cooled InGaAs photodetector in a spectrometer. In this spectrum, a sharp peak (FWHM: 21.6 meV) is observed at 946 nm (1.31 eV). This marked sharpness of the PL peak can be attributed to the size uniformity of the InGaAs QDs, which was originated mainly from the size uniformity of In droplets. Luminescence from the wetting layer was not observed, even with high-power laser excitation up to 800 mW$\cdot$cm^{-2}.

On comparing the PL spectra of the samples before and after annealing without the capping layer, only the annealed sample showed the luminescence from the QDs. This result indicates the necessity of the annealing process for the formation of high-quality QDs. Moreover, it should be noted that phase separation of InAs–GaAs might be enhanced during annealing. The Bohr radius of the exciton in the QDs estimated from the magneto-PL[20] was about 7 nm, which is smaller than the size of initially deposited InGa droplets. This suggests that the segregation of the In atoms during annealing exerts some effect on the formation of uniform-size QDs. The exact mechanism of the segregation and the In composition of the QDs were not clarified in this work. Further detailed investigation of the annealing process is necessary.[21]

4. Conclusion

InGaAs QDs were grown by droplet epitaxy with highly dense Ga droplets. These highly dense Ga droplets effectively prevent 2-dimensional growth of InGaAs, forming InGaAs QDs in the restricted regions. These InGaAs QDs showed sharp luminescence (FWHM: 21.6 meV) at 946 nm. These results suggest that droplet epitaxy with highly dense Ga droplets is a very useful method for the fabrication of uniform InGaAs QDs of high quality.

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