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Nanometer-scale GaAs ring structure grown by droplet epitaxy

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Abstract

Nanometer-scale GaAs ring structure is self-assembly realized by droplet epitaxy in a lattice-matched system. By changing the As_4 flux intensity during the crystallization of Ga droplets into GaAs, balance between the crystallization inside and at the edge of the droplets is changed, resulting in the shape control from dot to ring. The ring structure exhibits clear photoluminescence emission up to room temperature. Droplet Epitaxy is a promising growth method not only for quantum dots but also for quantum rings, with high structural and optical qualities in lattice-matched systems.

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1. Introduction

The fabrication of quantum-confined nanostructures by self-assembling growth methods has been intensively investigated in the last decade for basic physics and device applications [1–3]. Among them, semiconductor quantum rings have attracted a great deal of attention because fascinating properties have been predicted and demonstrated [4,5]. Recently, InGaAs or GaSb ring structures have been self-assembly realized on GaAs in strained systems by modifying the capping layer growth process or changing the amount of deposited GaSb [6–8]. In these systems, however, the real structures are complex due to the heavy intermixing of the materials [6]. Moreover, it has been pointed out that the electrons and/or the holes might localize in particular in-plane directions due to the strain and piezoelectric potential in these strained systems [9]. From this viewpoint, well-defined strain-free rings are desirable for studies of the physics of semiconductor quantum rings. For the growth of high-quality self-assembled nanostructures in lattice-matched systems, droplet epitaxy is a promising method [1,10]. While droplet epitaxy has been used only for quantum dot (QD) formation, this method has potential for the growth of more complex nanostructures [11,12].

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In this paper, we report the formation of well-defined nanometer-scale GaAs ring structure on AlGaAs by droplet epitaxy in a lattice-matched system. By changing the As_4 flux intensities during crystallization, the shapes of the GaAs nanostructures can be controlled from dot to ring, which is attributed to the balance of two processes during crystallization. This ring structure exhibits clear photoluminescence (PL) emission up to room temperature (RT).

2. Experimental procedure

The samples were grown by conventional solid-source molecular beam epitaxy (MBE) on semi-insulating GaAs (1 0 0) substrates. For the precise control of As_4 flux intensity, a valved cell was used for the As source. After the growth of a 400 nm thick $Al_{0.3}Ga_{0.7}As$ buffer layer at 580 °C, As-stabilized $c(4\times4)$ surfaces were formed by reducing the substrate temperature to 200 °C. For the formation of GaAs dot or ring structures, droplet epitaxy comprised the following sequences [12,13]:

(1) Supply of nominally 3.75 ML Ga (0.5 ML/s) on the $c(4\times4)$ surface at 200 °C without As_4 flux (the background pressure was kept below 4×10^{-9} Torr). On the $c(4\times4)$ surfaces, there is 1.75 ML of excess As. The first 1.75 ML of Ga changes into two-dimensional GaAs layer and the rest of 2 ML Ga forms droplets [12].
(2) Crystallization of the droplets into GaAs by supply of As_4 flux (2×10^{-4} or 1×10^{-5} Torr beam equivalent pressure (BEP)) at 200 °C.
(3) Annealing at 350 °C for 10 min under As_4 flux supply.
(4) Growth of a 50 nm thick $Al_{0.3}Ga_{0.7}As$ capping layer by migration-enhanced epitaxy [14] at 350 °C.
(5) Annealing at 650 °C for 10 min under As_4 flux.
(6) Growth of 50 nm thick $Al_{0.3}Ga_{0.7}As$ and 20 nm thick GaAs layers at 580 °C.

For structural characterization, the surface morphology of the samples after annealing at 350 °C (sequence (3)) was observed by non-contact mode atomic force microscope (AFM) in air. For PL measurements, rapid thermal annealing (RTA) (750 °C for 5 min) was performed after the entire growth sequences (1–6), which drastically improves the PL intensity of the GaAs nanostructure (factor ~100) [13]. PL peak energy change caused by the RTA process is less than 15 meV, indicating small structural changes such as an intermixing [13]. The 514 nm line of an Ar^+ laser was used as an excitation source with an excitation power density of 1 or 10 W/cm^2. The PL signal was dispersed by a single monochromator and detected by a photomultiplier tube.

3. Results and discussion

Fig. 1 shows the AFM image of the surface after the supply of 3.75 ML Ga on the $c(4\times4)$ surface at 200 °C. Many hemispherical-shaped Ga droplets are formed on the surface with a density of $\sim4\times10^9$/cm^2. The average base size and height are ~25 and ~7 nm, respectively. The size distribution of the droplets is ~20%.

Crystallization mechanisms of GaAs nanostructures from these droplets are roughly classified into two processes, as schematically illustrated in Fig. 2. The first one is GaAs growth inside the

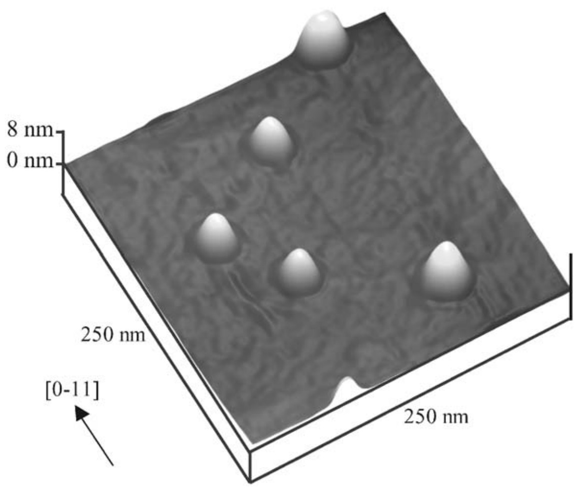

Fig. 1. AFM image of the surface after the supply of 3.75 ML Ga to the $c(4\times4)$ surface at 200 °C. The scan field is 250 nm × 250 nm.

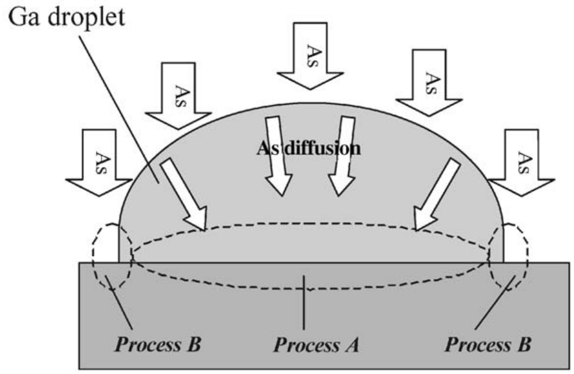

Fig. 2. Schematic illustration during crystallization of Ga droplet into GaAs.

droplets by As atom diffusion into the droplets (process A) [1,10]. As atoms attached on the droplet surface diffuse into the droplets. When these As atoms reach the interface between the droplets and GaAs (or AlGaAs), the As and Ga atoms change into epitaxial GaAs with some probabilities. The second one is GaAs growth at the edge of the droplets (process B). Since both Ga (from the droplets) and As (from the flux) atoms are directly supplied to GaAs (or AlGaAs) surfaces, efficient crystallization is expected at this area [15]. The two processes are correlated to each other and final shapes of the nanostructures are determined by the balance of them. Although more quantitative discussion is necessary for deeper understanding and further development, it is obvious that the shapes of the GaAs nanostructures can be simply controlled by crystallization parameters, i.e, substrate temperatures and As_4 flux intensities. In this study, we fix the substrate temperature and control the As_4 flux intensities for the realization of well-defined ring structures.

By crystallizing the droplets with high As_4 flux supply (2×10^{-4} Torr BEP) for reference, well-defined pyramidal-shaped GaAs QDs are formed with almost the same density of the original droplets ($\sim5\times10^{9}/\mathrm{cm}^2$), shown in Fig. 3(a) [12]. The base sizes in $[0\,\bar{1}\,1]$ and $[0\,1\,1]$ are 50 and 35 nm, respectively, and the height is 7.5 nm. The anisotropic shape might be due to the migration difference of Ga atoms during crystallization

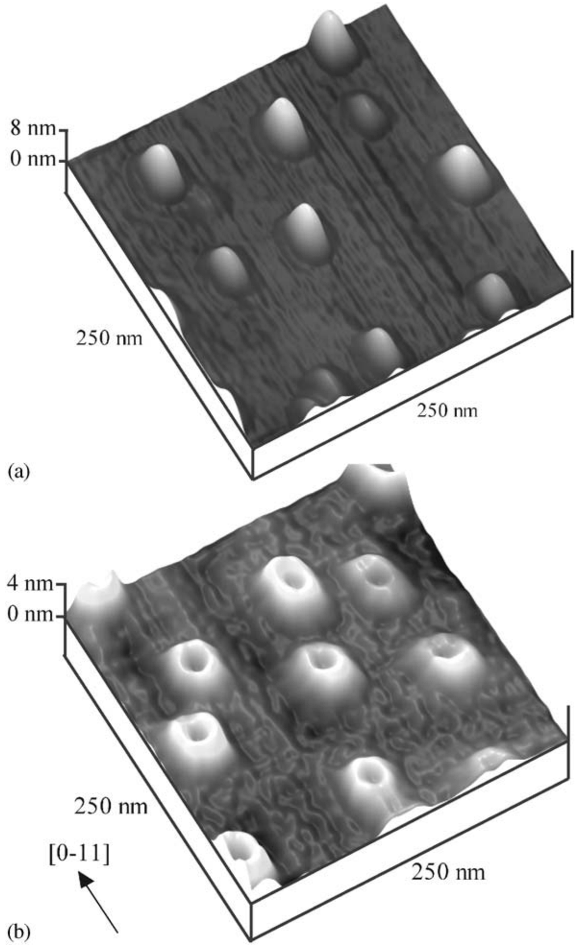

Fig. 3. AFM images of the surfaces after crystallization and annealing at 350 °C. As_4 flux intensities for crystallization are (a) 2×10^4 Torr BEP and (b) 1×10^5 Torr BEP. The scan field is 250 nm × 250 nm.

between $[0\,\bar{1}\,1]$ and $[0\,1\,1]$ [16] and/or difference between (1 1 1)A and (1 1 1)B facets [10]. In the case of the high As_4 flux supply, both processes A and B might have significant roles to keep the original droplet shape since a great number of As atoms attach on the droplet surface.

For the formation of a well-defined ring structure, the As_4 flux intensity is reduced to 1×10^{-5} Torr BEP, which probably enhances crystallization by process B rather than by process A. After crystallization, nanometer-scale GaAs ring structures are observed (Fig. 3(b)). The density of the ring structure is the same as that of the original droplets ($\sim 5\times10^9/\mathrm{cm}^2$). The typical outside and top diameters of the ring along $[0\,\bar{1}\,1]$ (along [0 1 1]) are ~51 nm (~45 nm) and ~25 nm (~22 nm), respectively. The height of the ring is ~1.5 nm. The top diameter of the ring structure is almost the same as the base size of the original droplets, indicating that crystallization is enhanced at the edge of the original droplets. This is consistent with the enhancement of process B.

PL spectra of the capped GaAs ring structure at low temperature (excitation density of $1\,\mathrm{W/cm}^2$) and RT (excitation density of $10\,\mathrm{W/cm}^2$) are shown in Fig. 4(a) and (b). At 5 K, PL emission from the ring structure is centered at 719 nm (1.72 eV) with peak width of 63 meV. PL lines around 625 and 830 nm stem from AlGaAs layer and GaAs bulk, respectively (Fig. 4(a)). PL emission from the ring structure is clearly visible up to RT (Fig. 4(b)). PL line width slightly increases to 75 meV, which might be due to the thermal excitation to the excited states. These results suggest that unstrained GaAs ring structure with high structural and optical qualities are realized by droplet epitaxy.

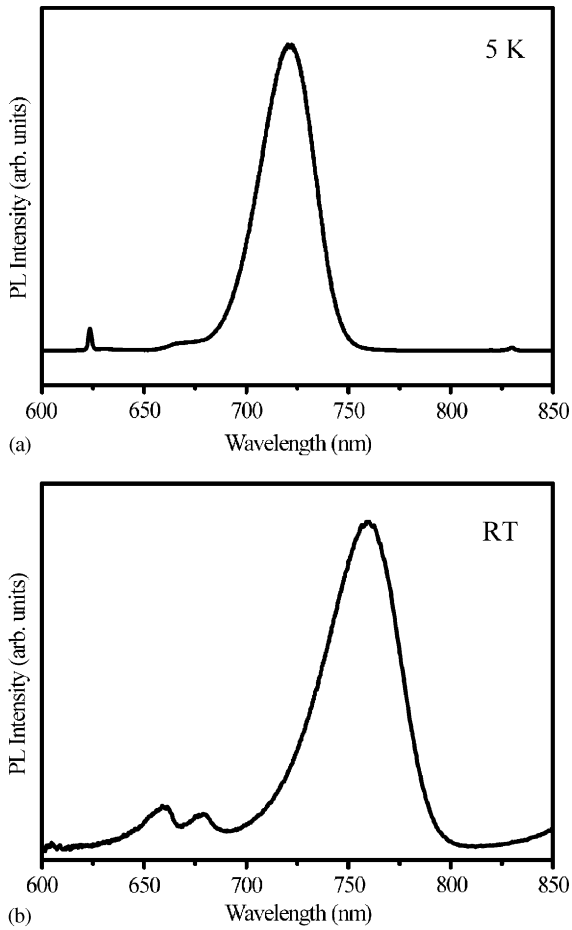

Fig. 4. PL spectra of the capped GaAs ring structure at (a) 5 K with the excitation density of $1\,\mathrm{W/cm}^2$ and (b) RT with the excitation density of $10\,\mathrm{W/cm}^2$.

4. Conclusion

Well-defined nanometer-scale GaAs ring structure was formed by droplet epitaxy in a lattice-matched system. By adjusting the As_4 flux intensity, crystallization was enhanced at the edge of the original droplets, resulting in the formation of a well-defined ring structure. The ring structure exhibits strong PL emission up to RT, indicating the high qualities. Although further studies of crystallization processes of GaAs nanostructures from Ga droplets are necessary, droplet epitaxy is highly promising not only for dot but also for ring formation in lattice-matched systems, which is ideal for studies of the physics of quantum rings.

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