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Ring-shaped GaAs quantum dot laser grown by droplet epitaxy: Effects of post-growth annealing on structural and optical properties

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Abstract

We investigated the effects of post-growth annealing on structural and optical properties of self-assembled ring-shaped GaAs quantum dots (QDs) by photoluminescence (PL) measurements and cross-sectional high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM). Marginal structural changes of the QDs were observed after the annealing process up to 800 °C while the intensity of PL emission increased drastically. The annealed laser structure with three layers of the ring-shaped QDs showed photo-pumped laser action with clear threshold at 77 K.

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1. Introduction

Semiconductor quantum dot (QD) lasers have attracted considerable attention due to their outstanding performances, such as low-threshold current density and high-temperature characteristics [1,2]. While there have been a lot of efforts on the self-assembled QD lasers in lattice-mismatched systems such as In(Ga)As/GaAs [3,4], the QD–lasers in a lattice-matched GaAs/AlGaAs system have not been well developed. It is simply due to the fact that the extensively used Stranski–Krastanow growth mode has incompatibility with the lattice-matched systems [5]. Meanwhile, we developed droplet epitaxy to fabricate self-assembled QDs [6–8]. Since the technique is based on the self-assembly of liquid metal nanoparticles (droplets) and their crystallization into compound semiconductors, we successfully created self-assembled QDs in the lattice-matched GaAs/AlGaAs system. We also found that the narrow photoluminescence (PL) emission can be obtained by making use of ring-shaped QDs [9,10], which are suitable for application to the QD lasers. To apply these QDs to the lasers, however, there still has been a hurdle which has to be overcome. The droplet epitaxy technique includes low-temperature growth processes for the crystallization of droplets (~200 °C) and the growth of capping layers (~350 °C) [7,8], which degrade the optical qualities. While post-growth annealing processes were normally performed to improve them, the structural changes during the annealing processes have been a matter of concern [11].

In this paper, we report the effects of post-growth rapid-thermal annealing (RTA) on the detailed structural and optical properties of the varied ring-shaped GaAs QDs. By using the RTA process at 800 °C for 4 min, we achieved drastic improvement of the emission intensity without major structural changes and succeeded in laser emission from self-assembled GaAs QDs in a lattice-matched AlGaAs matrix.

2. Experimental procedures

The samples were grown on semi-insulating GaAs (1 0 0) substrates by a conventional solid-source molecular-beam

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epitaxy system. The QD-laser structure consists of: (1) a 1.3 μm thick $Al_{0.58}Ga_{0.42}As$ bottom cladding layer, (2) a 0.28 μm thick $Al_{0.23}Ga_{0.77}As$ active layer with three layers of the ring-shaped GaAs QDs (separated by 20 nm thick $Al_{0.23}Ga_{0.77}As$) [12], and (3) a 1.3 μm thick $Al_{0.58}Ga_{0.42}As$ top cladding layer. The ring-shaped QDs in the active layer were formed by the droplet epitaxy technique. On the $Al_{0.23}Ga_{0.77}As$-$c(4\times4)$ surface, nominally 4.75 monolayers (ML) Ga (1.5 ML/s) was supplied without As_4 flux at 200 °C for the droplet formation. During the Ga supply, the first 1–2 ML of Ga was combined with excess As atoms on the $c(4\times4)$ surface, forming a two-dimensional GaAs layer, and the rest of the Ga formed droplets [7,13]. These droplets were, then, crystallized into the GaAs ring-shaped QDs by supplying an As_4 flux (1×10^{-5} Torr beam equivalent pressure) at the same temperature [9]. The QDs were annealed at 350 °C for 10 min under As_4 flux supply without capping to improve the crystal quality. After annealing, the QDs were capped with a 15-nm-thick AlGaAs capping layer at 350 °C and the rest of AlGaAs layer was grown at 580 °C. After the entire growth sequence, an RTA process was performed (800 °C for 4 min) in an N_2 atmosphere. The sample surface was protected by placing another GaAs substrate on top. The structural properties were studied by a non-contact mode atomic force microscope (AFM) and a cross-sectional high-angle annular dark field scanning transmission electron microscope (HAADF-STEM) [14]. For the PL measurements, the emission excited by an Nd–YAG laser at 532 nm (3 W/cm^2) was detected by a cooled charge-coupled device (CCD) camera.

3. Results and discussion

Fig. 1 shows an AFM image of the surface after crystallization of droplets followed by annealing at 350 °C for 10 min. Well-defined ring-shaped QDs with clear central holes were formed. The formation mechanism of the ring-shaped QDs was attributed to the edge-enhanced crystallization [9,10]. The density, average top diameter, and height of the QDs are 2×10^{10} cm^{-2}, 27, and 2 nm, respectively.

To check the energy levels of the ring-shaped GaAs QDs, we performed simple calculation of the wave function of electrons and holes in the frame work of finite element methods with strain-free, single-band effective-mass approximation in a cylindrical coordinate [15–18]. For the simplicity, the average-sized ring-shaped QD with perfect rotational symmetry was assumed, as shown in Fig. 2(a). A 0.5-nm-thick (corresponding to 1.75 ML GaAs [7]) two-dimensional GaAs layer underneath the QD was also taken into account. In Fig. 2(b), we present energies of calculated principal optical transitions, assuming that the optical transitions with different quantum numbers are negligible [10,19]. The ring-shaped QD has a set of discrete energy states with radial (n) and rotational (l) quantum numbers [19], which prove that this ring-shaped structure is indeed a "*quantum dot*" with three-dimensional quantum confinements.

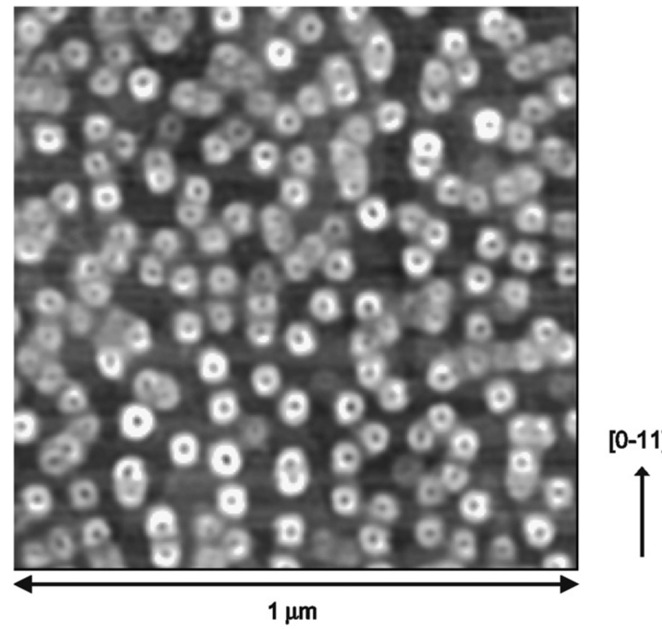

Fig. 1. AFM image of ring-shaped GaAs QDs. The scan area is 1 × 1 μm^2 and the black to white contrast is 4.5 nm.

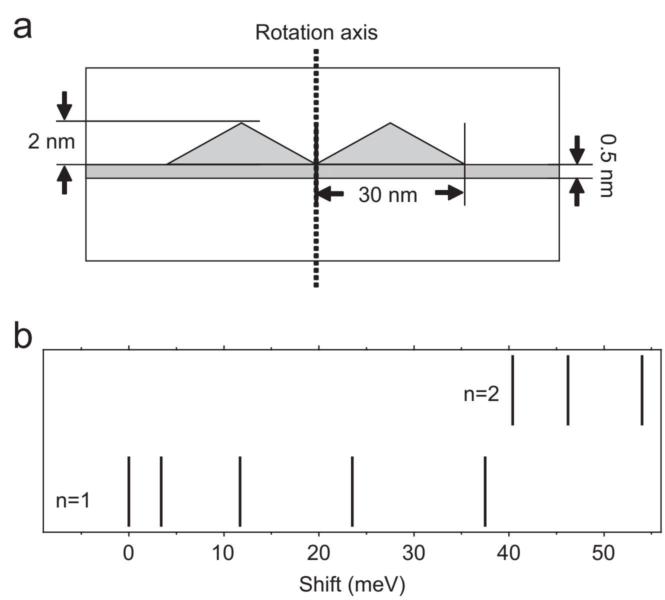

Fig. 2. (a) Schematic illustration of ring-shaped QDs used for the calculation. (b) Calculated principal optical transition energies for $n = 1$ and 2.

The PL emission spectra of the as-grown and RTA samples are shown in Fig. 3. Since the capping layer was grown at 580 °C, the as-grown sample was actually annealed at this temperature. The PL intensity of the RTA sample is 50-times stronger than that of the as-grown sample. In contrast to the increase of the emission intensity, the peak wavelength and width exhibited only

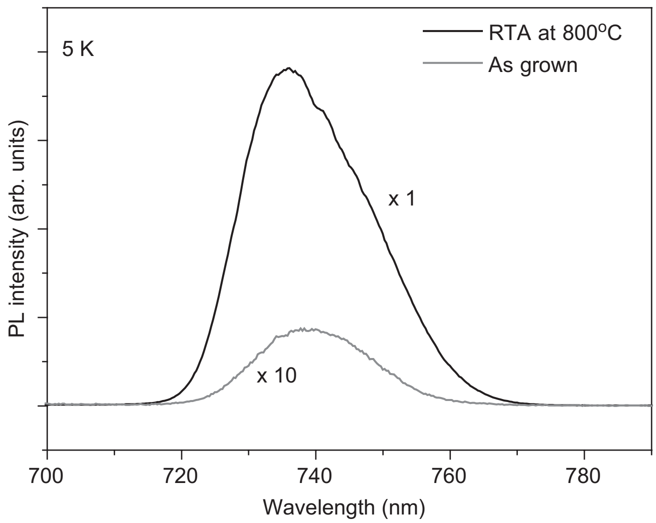

Fig. 3. PL spectra of the as-grown (grey line) and RTA (black line) samples measured at 5 K.

small changes. The wavelength changes from 739 to 738 nm and peak width changes from 38 to 43 meV after the RTA process. These results suggest that the RTA process drastically improves the optical quality of the GaAs QDs and AlGaAs matrix. However, the structural changes induced by the process, such as the intermixing, are relatively small compared with the previous report [11]. This finding is consistent with the small interdiffusion coefficient of Al and Ga up to 800 °C as reported previously [20]. Also, rather short annealing time (4 min) may suppress the major structural changes. To verify the origin of the improvement of optical qualities, further studies are necessary.

To confirm the above discussion, we performed HAADF-STEM measurements of the as-grown and RTA samples. For this study, we additionally prepared large-sized GaAs ring structures (Fig. 4(a)) in AlGaAs matrix with high Al composition ($x = 0.45$) in order to gain a high contrast in the images [21]. Fig. 4(b) shows the HAADF-STEM image of the as-grown sample. A GaAs nanostructure was formed on top of a 0.6-nm-thick two-dimensional layer. A clear dent is visible at the top of the nanostructure, indicating that the ring structure was varied in an AlGaAs matrix with keeping its original shape. In the case of the RTA sample (annealed at 800 °C), we also observed almost the same structure (Fig. 4(c)). A high contrast is still visible at the interface of the QDs and matrix. To roughly estimate the amount of the intermixing, we compared the thicknesses of the two-dimensional layer before and after the RTA process. After the RTA process, the thickness slightly increased from 0.6 to 0.9 nm (Fig. 4(b) and (c)). Hence, the RTA process (800 °C for 4 min) causes only monolayer-order intermixing at the interface between the QDs and matrix so that the original structure is almost unchanged.

For the laser emission experiments, the sample after the RTA process was cleaved into a cavity length of

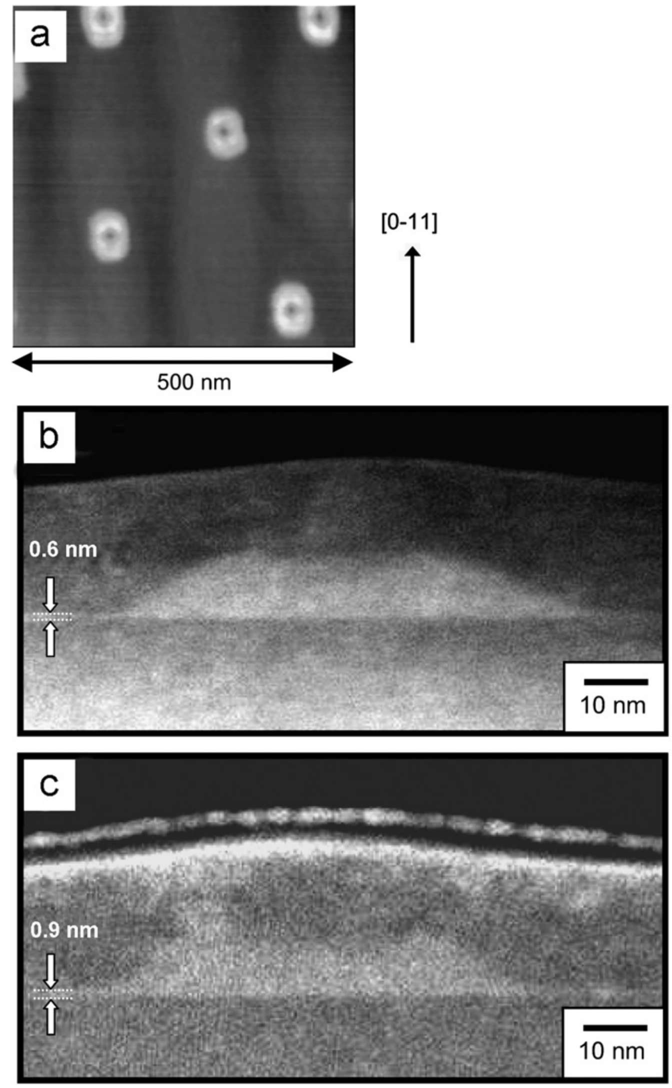

Fig. 4. (a) AFM image of GaAs ring structures. The scan area is $500 \times 500\,\text{nm}^2$ and the black to white contrast is 18 nm. (b), (c) Cross-sectional HAADF-STEM images of the GaAs/AlGaAs ring structures: (b) as-grown and (c) RTA samples.

$L = 2200\,\mu\text{m}$. The width of the cavity was defined by the excitation laser spot ($W = 20\,\mu\text{m}$). A pulsed Nd–YAG laser was used for excitation (λ: 532 nm, pulse width: 5 ns, repetition rate: 4 kHz). The excitation light focused in a stripe shape ($2000\,\mu\text{m} \times 20\,\mu\text{m}$) was applied to the top surface of the cooled sample (77 K) by using two cylindrical lenses. The emission from the cleaved edge was gathered and detected by a CCD camera through a spectrometer (spectral resolution: 5 nm). Fig. 5(a) shows emission spectra with different excitation powers. On increasing the pump power, the spontaneous emission from the QDs increased. Around a pump power of 10 mW, stimulated emission was observed. In the plot of integrated emission intensity (Fig. 5(b)), we can observe a clear threshold that means the onset of lasing. In a high-resolution measurement (the inset of Fig. 5(a)), several quasi-periodic narrow lines related to the longitudinal modes were observed (spectral

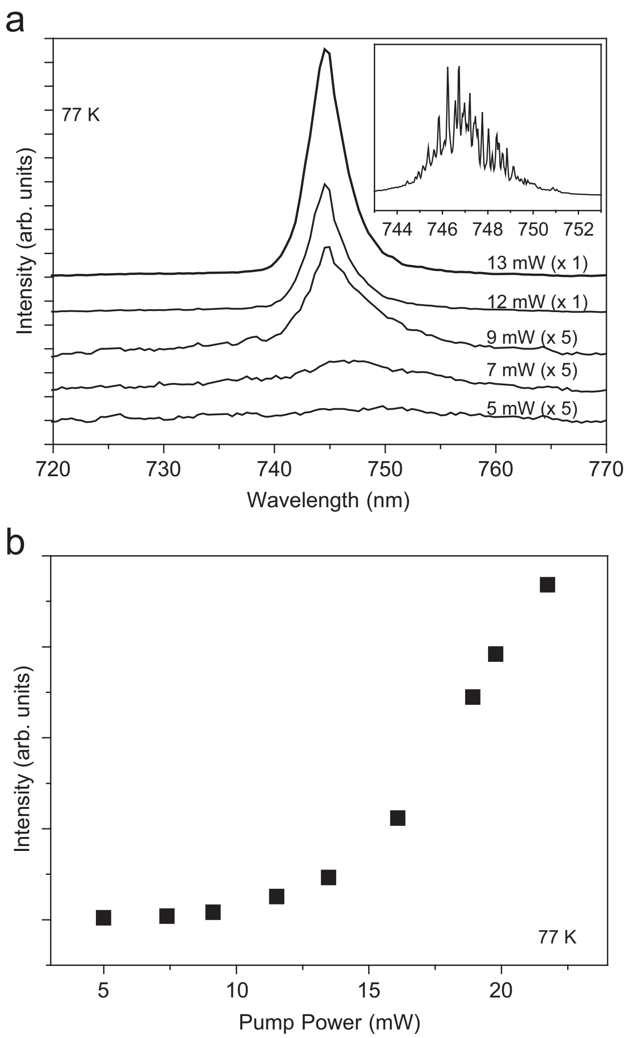

Fig. 5. (a) Emission spectra with various excitation powers at 77 K. The inset shows a high-resolution spectrum above the threshold. (b) Integrated emission intensity plotted as a function of excitation power.

resolution: 0.1 nm), supporting that the emission is caused by the laser action. Since the laser emission wavelength is almost the same as that of the spontaneous emission, the lasing may occur mainly in the ground state of the QDs.

The successful laser action suggests that the low optical qualities induced by the low-temperature processes are sufficiently recovered by the RTA process and that the device-quality self-assembled QDs are realized in a lattice-matched system.

4. Conclusions

We studied an RTA (800 °C for 4 min) effect on structural and optical properties of ring-shaped GaAs QDs grown by droplet epitaxy. While the RTA process caused only marginal structural changes, the PL emission intensity drastically increased. The annealed laser structure with three stacked ring-shaped QDs exhibited photo-pumped laser action with clear threshold at 77 K. These results suggest that the device-quality self-assembled GaAs QDs are realized in a lattice-matched system. With further optimization of growth processes, droplet epitaxy will be a promising method to realize the QD lasers in the various lattice-matched compound semiconductor systems.

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