Growth and characterization of InAs quantum dots with low-density and long emission wavelength

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The growth parameters affecting the deposition of self-assembled InAs quantum dots (QDs) on GaAs substrate by low-pressure metal-organic chemical vapor deposition (MOCVD) are reported. The low-density InAs QDs ($\sim 5 \times 10^8$ cm^{-2}) are achieved using high growth temperature and low InAs coverage. Photoluminescence (PL) measurements show the good optical quality of low-density QDs. At room temperature, the ground state peak wavelength of PL spectrum and full-width at half-maximum (FWHM) are 1361 nm and 23 meV (35 nm), respectively, which are obtained as the GaAs capping layer grown using triethylgallium (TEG) and tertiallybutylarsine (TBA). The PL spectra exhibit three emission peaks at 1361, 1280, and 1204 nm, which correspond to the ground state, the first excited state, and the second excited state of the QDs, respectively.

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Self-assembled InAs/GaAs quantum dots (QDs) have been studied in the past years since these structures provided the prospect of temperature-independent low threshold lasers. The development of quantum protocols for optical transmission of cryptographic keys has given a strong impetus to attain low-density QDs for reliable sources of single photons[1,2]. Therefore, it is important to develop growth procedures to obtain low-density QDs. The QD density of $\sim 2.5 \times 10^7$ cm^{-2} was obtained by combining QD growth interruption with non-homogeneous InAs deposition by molecular beam epitaxy (MBE)[3]. The low-density QDs of 2.7×10^8 cm^{-2} were obtained by a growth technique that combines droplet epitaxy and MBE[4]. By using a set of optimized growth parameters, QDs with density as low as 5×10^6 cm^{-2} have been obtained by metal-organic chemical vapour deposition (MOCVD)[5]. However, most studies were concentrated on the wavelength range of less than 1.0 μm, while fiber-based quantum cryptography requires emission wavelengths matching the 1.3- or 1.55-μm transmission windows of optical fibers. It is hard to reach 1.3-μm emission wavelength due to the difficulty of growing sufficiently large and thick QDs. Recently, low-density QDs of $(2-3)\times 10^8$ cm^{-2} have been obtained, and 1.3-μm emission wavelength have been achieved using an ultralow InAs growth rate and capping with an InGaAs layer[6]. The island size and density strongly depend on the growth rate. With a reduction in growth rate, the island size increases and the light emission wavelength correspondingly shifts to the longer side. A reduction in growth rate enhances migration length of the In adatoms. In order to minimize strain and surface energy, the In atoms incorporate into existing dots instead of forming new dots. Thus, a reduction in growth rate leads to a decrease in island density and a corresponding increase in size[7].

In this paper we present a method to obtain low-density InAs QDs ($\sim 5 \times 10^8$ cm^{-2}) and ground state emission beyond 1.3 μm at room temperature. In order to obtain QDs with low-density and long emission wavelength, we investigate the QD growth conditions of growth temperature and various InAs coverages.

All samples used in the study were grown on GaAs (001) by low-pressure MOCVD system, using trimethylindium (TMI), trimethylgallium (TMG), triethylgallium (TEG) as the group III sources, and tertiallybutylarsine (TBA) as the group V source. The total background pressure during the growth was 76 torr. After oxide desorption at 800 °C, a 200-nm GaAs layer was deposited at 700 °C using TMG and TBA. Then the substrate was cooled down and the subsequent InAs QDs were deposited at 480 – 540 °C using TMI and TBA. The growth rate for InAs was 0.01 ML/s (ML: monolayer). To achieve long wavelength emission beyond 1.3 μm, the QDs were finally capped with a 100-nm-thick GaAs layer at 520 °C using TEG and TBA, respectively. The use of TEG in place of TMG as a gallium source during the deposition of a GaAs capping layer cannot only shift the emission to longer wavelengths, but also narrow the full-width at half-maximum (FWHM) of photoluminescence (PL) due to the much greater growth rate of GaAs using TEG and TBA compared with TMG and TBA[8,9]. In order to achieve lower QD density, the InAs QDs were deposited at high growth temperature (520 °C), in comparison, the substrate temperature in Ref. [5] was lowered to 500 °C for the QD layer growth.

Atomic force microscopy (AFM) images of 1×1 (μm) size were used to measure the surface morphology and the density of InAs QDs on a GaAs surface with various InAs coverages. Structural properties of InAs QDs in the samples were characterized by measuring uncapped GaAs layer.

In general, increasing growth temperature of QDs is often used to decrease the QD density because of the diffusion of In adatoms on the surface with increasing temperature. Figure 1 shows the dot density of the samples with InAs coverage of 2.3 ML grown at different temperatures. At the temperature $T = 480$ °C, the density is $\sim 3\times 10^{10}$ cm^{-2}, with an average height of ~ 9 nm. At $T = 520$ °C, the density is $\sim 8 \times 10^9$ cm^{-2}, with an average height of ~ 11 nm. However, at $T = 540$ °C, the QD size distribution is bimodal, with small and bigger

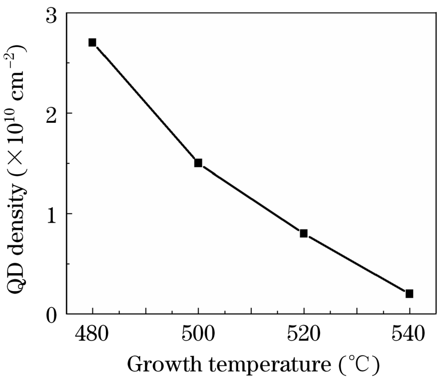

Fig. 1. InAs QD densities of the samples grown at different temperatures.

dots. The small dots assemble with an average height of ~ 15 nm and a density of $\sim 2 \times 10^9$ cm^{-2} and the bigger dots with an average height of ~ 6 nm and a density of $\sim 5 \times 10^7$ cm^{-2}, respectively. The average height of bigger dots and density are decreased due to the desorption of In adatoms at higher growth temperature. Therefore, the InAs QDs were deposited at $T = 520$ °C with lower InAs coverages to achieve low QD density.

Figure 2 shows the AFM images of InAs/GaAs QD layers grown with various InAs coverages between 1.95 and 2.10 ML. Figure 3 shows the dependence of QD density and island height on the InAs coverage. For the coverage $\theta = 1.95$ ML, the QDs are composed of a bimodal distribution, with small dots of average height of ~ 1.5 nm and density of $\sim 1.3 \times 10^{10}$ cm^{-2}. For $\theta = 2.00$ ML, the QD characteristics change significantly, the QD size distribution is also bimodal, with small and bigger dots. However, the densities of small and bigger dots decreases and increases, respectively. In fact, the small dots are observed at $\theta = 1.95$ ML, but the small dots are not observed any more for higher InAs coverage ($\theta > 2.0$ ML). The QD size distribution becomes unimodal with big coherent InAs dots with height in the range of $10-12$ nm, as shown in Fig. 3. The density of the big coherent InAs dots increases gradually with the increase of InAs coverage, from 5×10^8 to 2.9×10^9 cm^{-2} when θ changes from 1.95 to 2.10 ML. The achievement of such low densities for low InAs coverage is not so surprising, the most striking feature is that the QD size is almost constant for higher InAs coverage ($\theta > 2.0$ ML), with the height ranging from 10 to 12 nm. As soon as there is the formation of InAs dots, large dots are formed. This is possible because of the large diffusion length of In adatoms, induced by the combination of low growth rate and high growth temperature. We also note the coalescence of dots at $\theta > 2.05$ ML, with the appearance of giant dots ($\sim 10^7$ cm^{-2}, not shown) with height up to 15 nm and diameter up to 140 nm, which is attributed to the giant dots which contain crystal defects such as dislocations.

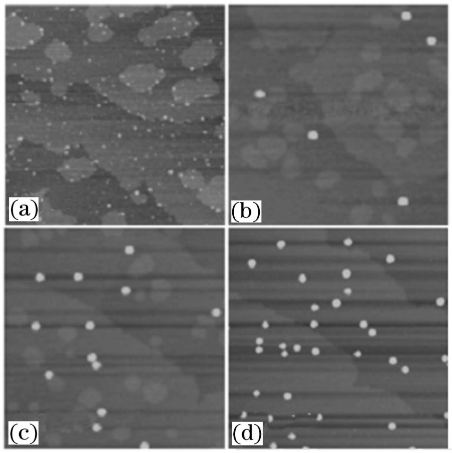

Fig. 2. 1×1 (μm) AFM images of InAs QDs on a GaAs surface with various InAs coverages of (a) 1.95, (b) 2.00, (c) 2.05, and (d) 2.10 ML.

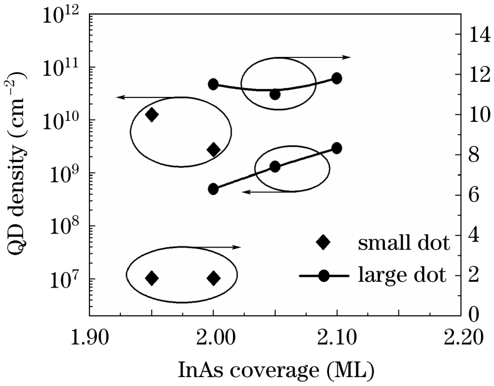

Fig. 3. QD densities and heights with various InAs coverages.

To investigate the optical properties of low-density InAs QDs, a second QD sample was grown under the same conditions except that the QDs were capped by 100-nm-thick GaAs capping layer. PL characteristics were measured with a semiconductor laser of excitation wavelength of 670 nm and detected by a liquid-nitrogen cooled InGaAs photodiode array at room temperature (RT). Figure 4 shows the PL spectra of low-density (5×10^8 cm^{-2}) QD sample measured at different excited powers. The PL spectra exhibit three emission peaks at 1361, 1280, and 1204 nm, which correspond to the ground state (GS), the first excited state (ES1), and the second excited state (ES2) of the QDs, respectively. The results of PL measurements at RT show a red shift of PL peak emission wavelength as the growth rate is decreased, and the wavelength reaches 1310 nm at RT at the lowest growth rate (0.0012 ML/s)[6].

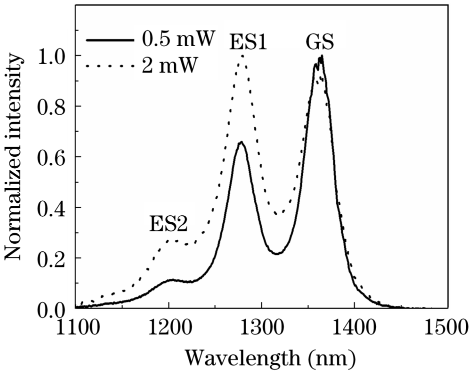

Fig. 4. PL spectra of low-density ($\sim 10^8$ cm^{-2}) QD samples at different excitation powers.

The experimental results at different excitation powers reveal that the PL intensity ratio between GS and ES1 peaks changes accordingly. For high excitation power, emission intensity is clearly visible from both GS and ES1 peaks, though the ES1 peak is dominant. At lower excitation power, the GS peak is dominant, the FWHM of the GS peak is equal to 23 meV.

In conclusion, we have shown that it is possible to obtain low-density QDs with emission wavelength beyond 1.3 μm by GaAs capping layer using TEG and TBA. The PL spectra exhibit three emission peaks at 1361, 1280, and 1204 nm, corresponding to three states of the QDs, respectively. QDs with lower density ($\sim 5 \times 10^8$ cm^{-2}) are achieved using high growth temperature and low InAs coverage. PL measurements prove the good optical quality of the low-density InAs QDs.

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