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Extremely low density self-assembled InAs/GaAs quantum dots

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The self-assembled InAs/GaAs quantum dots (QDs) with extremely low density of 8×10^6 cm⁻² are achieved using higher growth temperature and lower InAs coverage by low-pressure metal-organic chemical vapour deposition (MOVCD). As a result of micro-photoluminescence (micro-PL), for extremely low density of 8×10^6 cm⁻² InAs QDs in the micro-PL measurements at 10 K, only one emission peak has been achieved. It is believed that the InAs QDs have a good potential to realize single photon sources.

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The self-assembled InAs/GaAs quantum dots (QDs) have been studied in the past few years since these structures provide the prospect of low threshold lasers independent of temperature. 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 photon by chemical beam epitaxy (CBE)^[1]. The self-assembled QDs have been of great interest as single photon sources because they are relatively stable, have narrow spectral linewidths and rapid radiative decay rates, and can be integrated into larger fabricated structures as microcavities. In microcavities, collection efficiency of photons is improved due to the spatial limitations, and the radiative lifetime of emitters is also significantly shortened due to the Purcell effect. This improves device performance and makes single photon which can also be applied to quantum information processing^[2].

Therefore, it is important to develop growth procedures to obtain low-density QDs. The low density InAs QDs of $\sim 2.5 \times 10^7 \, \mathrm{cm}^{-2}$ were obtained by a method of combining QDs growth interruption and non-homogeneous InAs deposition through molecular beam epitaxy $(MBE)^{[3]}$. The low-density InAs QDs of 2.7×10^8 cm⁻² were obtained by a growth technique that combines droplet epitaxy and $MBE^{[4]}$. Compared with the MBE system, the QDs grown by metal-organic chemical vapour deposition (MOVCD) have been limited due to the fundamental differences in growth mechanism. Recently, we showed the good optical quality of low-density InAs QDs of $\sim 5 \times 10^8 \text{ cm}^{-2[5,6]}$. InGaAs QDs with extremely low density of 5×10^6 cm⁻² have been obtained by MOVCD by using a set of optimized growth parameters^[7]. The QD size and density strongly depend on the growth rate. With a reduction of the growth rate, the island size increases and the light emission wavelength correspondingly shifts to the longer side. A reduction of the growth rate enhances migration length of the In adatoms. Thus, a reduction of the growth rate leads to a decrease in island density and a corresponding increase in size^[8]. Lower density QDs were achieved using high growth temperature. High growth temperature leads to a reduction of QD density and increase of QD size^[9].

In this paper, we present a method to obtain extremely low density $(8 \times 10^6 \text{ cm}^{-2})$ InAs QDs. In order to obtain QDs with low density, we investigated the QD growth conditions of growth temperature and various InAs coverages. With these low-density QDs, we performed the micro-photoluminescence (micro-PL) measurements at 10 K. The PL measurements proved the good optical quality of low-density QDs. Particularly, for extremely low density QDs in the micro-PL measurements at 10 K, only one emission peak has been achieved without isolating a single QD by electron beam lithography.

All samples were grown on GaAs (001) by lowpressure 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 measure the optical properties of low-density InAs QDs, the QDs were finally capped with a 100-nm-thick GaAs layer at 520 °C using TEG and TBA.

Atomic force microscopy (AFM) was 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 temperature increase. Figure 1 shows the dot densities 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,



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

the QD size distribution is bimodal, with small and bigger dots. The small dots are composed with an average height of ~ 5 nm and diameter 40 nm and the bigger dots with an average height of ~ 6 nm and diameter 80 nm, respectively. The average heights of bigger dots and densities 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.

As shown in Fig. 2, the QD density decreases strongly from 8×10^9 cm⁻² to approximately 8×10^6 cm⁻² as InAs coverage is reduced from 2.3 to 1.9 ML. From 1.9 to 2.3 ML, the diameter of the InAs dots is about 30 nm, and their heights are in the range of 10 – 12 nm. However, for the coverage $\theta = 1.9$ ML, there is a sharp descent of surface density to 8×10^6 cm⁻², as shown in Fig. 3. The achievement of such low densities for low InAs coverage is not surprising. The most striking feature is that the QD size is almost constant with various InAs coverages



Fig. 2. QD densities and heights with various InAs coverages.



Fig. 3. $5\times5~(\mu{\rm m})$ AFM image of InAs QDs with InAs coverage of 1.9 ML. The QDs are labelled by dashed circles.



Fig. 4. Micro-PL spectra with various InAs QD densities of (a) 5×10^8 cm⁻², (b) 8×10^6 cm⁻² at the temperature of 10 K.

because of the large diffusion length of the In adatoms, induced by the combination of low growth rate and high growth temperature. As soon as there is the formation of InAs dots, they subsequently become larger QDs. At higher growth temperature, a greater migration distance leads to a lower QD density.

To investigate the optical properties of low-density InAs QDs, a second series of QD samples were grown under the same conditions except that the QDs were capped by a 100-nm-thick GaAs capping layer. The micro-PL measurements were also performed with a heliumneon laser emitting at 633 nm and detected by a liquidnitrogen cooled InGaAs photodiode array at 10 K.

The micro-PL spectra of low-density QD samples were shown in Fig. 4. As shown in Fig. 4(a), the PL spectra exhibits a few sharp emission peaks. These results show that a few peaks from individual dots have been achieved for the low density of 5×10^8 cm⁻² quantum dots. As shown in Fig. 4(b), only one emission peak has been observed for extremely low density of 8×10^6 cm⁻² quantum dots. The emission peak and the full width at half-maximum (FWHM) of PL spectra are about 1117 nm and 4.9 meV, respectively. As a result of micro-PL, only a few emission peaks from individual dots can be achieved. Particularly, for extremely low density in the micro-PL measurements at 10 K, only one emission peak has been achieved without isolating a single QD by electron beam lithography. It reveals that lower density QDs also have good carrier confinement.

In summary, the results show that it is possible to reach extremely low density QDs of 8×10^6 cm⁻² with higher growth temperature and lower InAs coverage. And we successfully obtain the result of strong single emission peak from QDs, which can only be realized with the assistance of electron beam lithography in other cases. The PL measurements proved the good optical quality of extremely low density InAs QDs. This result confirms that such extremely low density InAs QDs have a good potential to realize single photon sources.

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