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Improving crystal quality of InGaAs/GaAs quantum dots by inductively coupled Ar plasma

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The crystal quality of InGaAs/GaAs quantum dots (QDs) is substantially improved without redistribution of composition using inductively coupled Ar plasma exposure. After plasma exposure, the QDs exhibit an increase in photoluminescence intensity by a factor of 1.7 while keeping the peak wavelength unshifted, and the band gap blueshift after rapid thermal annealing is suppressed, denoting an improvement in thermal stability. The time-resolved photoluminescence shows an increase in carrier lifetime from 735 to 1140 ps by plasma exposure, indicating the mechanism of grown-in defects reduction in the QD regions. © 2006 American Institute of Physics.

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Continuous advancement in the growth techniques of quantum-dot (QD) structures has enabled practical QD-based devices with superior performance, such as low laser threshold, high quantum efficiency, low temperature-dependent characteristics, etc.¹ However, typical growth at low temperature (LT) leads to a certain amount of nonradiative grown-in defects in QD regions, which limit performances of QD devices and enhance thermal interdiffusion. The interdiffusion effect is significant particularly in QD structures because of the large interface area to volume ratio between QDs and the surrounding barriers compared to quantum-well (QW) structures. In order to eliminate the grown-in defects and improve QD performances, thermal treatment has been used during overgrowth and at postgrowth level with the penalty of notable band gap blueshift.^{2–4} In the view of monolithic integration utilizing the selective QD intermixing,^{5,6} such low thermal stability limits the net differential band gap shift due to the high temperature required for activating intermixing. In either application, it is important to improve the crystal quality and the thermal stability of QD structures, such that the thermal induced band gap modification can be minimized.

In this letter, we report the effect of the inductively coupled Ar plasma exposure on the crystal quality of an InGaAs/GaAs QD structure fabricated by cycled monolayer deposition. The optical property and the thermal stability are investigated with samples processed by plasma exposure and rapid thermal annealing (RTA). Low temperature photoluminescence (PL) is used to study the modification of QD optical properties and the change in QD thermal stability. Time-resolved photoluminescence (TRPL) is further used to assess the role of Ar plasma exposure to the improved crystal quality.

The InGaAs/GaAs QDs structure used in this work is a QD infrared photodetector structure grown by molecular beam epitaxy on a Si doped $n+(100)$ -oriented GaAs substrate. A 300-nm-thick undoped GaAs buffer layer was first grown and subsequently a 1000-nm-thick, Si doped ($n=2 \times 10^{18} \text{ cm}^{-3}$) GaAs bottom contact layer. 20 stacks of InGaAs QD layers were then consecutively grown, separated by 50-nm-thick GaAs layers which were doped with Si ($n=1 \times 10^{18} \text{ cm}^{-3}$) within a thickness of 10 nm in the center of each layer. On the top is a 600-nm-thick, Si doped ($n=2 \times 10^{18} \text{ cm}^{-3}$) GaAs contact layer. For each QD layer, five pairs of alternating InAs and GaAs monolayers were grown under a constant As flux with interruption after each monolayer in order to stabilize the surface. The growth temperature was 515 °C for QD layers and 600 °C for other layers.

The plasma exposure was carried out using an ICP180 plasma source generator for 1–5 min under the following parameters: 100 SCCM (SCCM denotes standard cubic centimeters per minute at STP) Ar flow rate, 60 mTorr chamber pressure, 480 W rf power, and 500 W inductively coupled plasma (ICP) power with a rf-induced dc bias of ~ 900 V. The annealing was performed at 700–750 °C for 30–120 s under flowing N_2 ambient using GaAs proximity caps. PL spectra were measured at 77 K using a green crystal laser (532 nm, $\sim 1 \text{ kW/cm}^2$) for excitation, a monochromator, and a thermoelectric (TE)-cooled Ge photodetector associated with a lock-in amplifier. TRPL measurements were carried out using a femtosecond self-mode locked Ti-sapphire laser pumped by a 532 nm green crystal laser. The excitation wavelength was selected at 800 nm with an output power of 15 mW. The repetition rate was 82 MHz with a pulse width of 150 fs. The detection wavelength was selected at the PL peak wavelength of the ground state emission from the QD samples. The time resolution of the TRPL system is 30 ps.

Figure 1 presents the evolution of PL spectra from QDs after Ar plasma exposure with the duration varied from

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FIG. 1. (Color online) PL spectra at 77 K taken from (a) as-grown sample, and samples exposed only to Ar plasma for (b) 1 min, (c) 2 min, and (d) 3 min, respectively. The curve of PL peak intensity vs the plasma exposure time is given in the inset.

1 to 5 min with no RTA process. The PL peak intensity versus the duration is also given in the inset. The plot in the inset shows an overall trend of increasing PL peak intensity within 3 min exposure, with the PL peak intensity reaching a maximum 2.7 times of the original level at 3 min exposure. The PL intensity does not show an increase in the first minute, mainly resulting from the roughened surface due to Ar ion bombardment. Previous investigations have demonstrated the increase in PL intensity using Ar plasma exposure owing to reduction of grown-in defects in QWs.^{7,8} In this sample structure, QD layers were grown at much lower temperature ($\sim 515^\circ\text{C}$) than the optimal required for growing a high quality lattice, an appreciable amount of LT grown-in defects present in the structure may behave as nonradiative recombination centers.⁹ The enhancement in PL intensity indicates the reduction of LT grown-in defects around the QD regions. It should be further noted that no discernible PL peak wavelength shift is involved in this process as seen in

FIG. 2. (Color online) PL spectra at 77 K from (a) as-grown, (b) control, and (c) 3-min-exposed and annealed samples. The control sample was annealed without being exposed to plasma beforehand. Annealing was carried out at 750°C for 60 s. The inset shows the band gap shifts and the PL peak intensities against the plasma exposure durations measured after annealing.

FIG. 3. (Color online) (a) 77 K PL peak shifts and (b) normalized PL peak intensities after consecutive annealing cycles at 700°C for (i) as-grown and (ii) 3-min-exposed samples.

Fig. 1, denoting no change in the composition profile of QDs.

A natural inference of the reduction of grown-in defects in the QD structure, or the improvement of the crystal quality, is the improvement of the thermal stability, which is fairly important when the net differential band gap shift is pursued in selective intermixing. The band gap shift versus the plasma exposure duration was investigated using samples exposed to Ar plasma with different durations and annealed at 750°C for 60 s. The inset in Fig. 2 shows that the band gap shift decreases with increasing Ar plasma exposure duration, and a maximum band gap shift suppression of 20 nm is obtained. This observation suggests the improvement of QD crystal quality and thus the thermal stability.

The trend of PL intensity versus the plasma exposure duration is similar to that shown in the inset of Fig. 1, except that the PL peak intensity after annealing is higher. This is because PL spectra of QDs are less diverse after intermixing such that the summed PL peak becomes narrower but higher. Comparison between the spectra of the annealed-only and the as-grown samples shown in Fig. 2 shows insignificant change in PL peak intensity, denoting that the conventional RTA process is not as efficient as the Ar plasma exposure in diminishing grown-in defects in the crystal.

While diminishing the grown-in defects in the active region microns under the surface, Ar plasma exposure also introduces a certain amount of point defects in the near surface regions. Whether intermixing can be enhanced or retarded is determined by the net effect of these two facts. Previous study in InGaAs/InP QW structures showed large intermixing enhancement by Ar plasma exposure,¹⁰ whereas in this study of the InGaAs/GaAs QD structure, the reduc-

FIG. 4. (Color online) TRPL spectra at 100 K for (a) as-grown and (b) 3-min-exposed samples. No annealing was performed. The lines indicate the best fittings from the experimental data.

tion of grown-in defects is more prominent due to the large quantity of original LT grown-in defects. The band gap shift decreases almost linearly within the first 3 min of plasma exposure. However, this trend gradually vanishes beyond 3 min of exposure because the defects introduced near the surface of the top layer keep accumulating while the grown-in defects have been sufficiently reduced.

The improved crystal quality of the QD structure can be more clearly seen in Fig. 3. Two samples, one as grown and the other experienced 3 min plasma exposure underwent repetitive annealing cycles at 700 °C with each cycle of 30 s, whereas the band gap shift and PL peak intensity after each cycle were collected. The consecutive annealing cycles cause a monotonic increase in band gap shift for both samples. The consistent band gap shift reduction occurs in the plasma exposed sample, e.g., a suppressed band gap shift of 26 nm between 3-min-exposed and the control samples. Significant PL peak intensity enhancement is maintained after each annealing cycle in the plasma exposed sample, compared to the low PL intensity observed in the control sample.

The presence of defects in III-V semiconductor materials can be assessed by the lifetime deduced from TRPL measurements.¹¹ To verify that the grown-in defect density is reduced by the plasma exposure, the TRPL of a 3-min-exposed sample without annealing was measured and

compared with that of an as-grown one (Fig. 4). To deduce the TRPL lifetime, the experimental data were fitted using the following formula:

$$I_{\text{PL}}(t) = a_0 + a_1 e^{(t-t_0)/\tau_d} + a_2 e^{(t-t_0)/\tau_d},$$

where τ_r and τ_d are the PL rise time and decay time, denoting the effective carrier capture time and the effective carrier lifetime in QDs, respectively. The TRPL lifetimes for the as-grown and exposed QDs are 735 and 1140 ps, respectively. The increased lifetime after plasma exposure clearly confirms the reduction of defects in the region of LT-grown QDs to guarantee more carriers for radiative recombination leading to enhanced PL peak intensities.

In summary, we have demonstrated the improvement of crystal quality of InGaAs/GaAs QDs using inductively coupled Ar plasma exposure. The PL intensity can be enhanced by 1.7 times immediately after plasma exposure without discernible band gap shift. The thermal stability is also improved and suppression of band gap shift can be clearly observed. The carrier lifetime is increased by 55%, indicating the reduction of grown-in defects as nonradiative recombination centers. This result indicates a more efficient approach to improving the crystal quality of QD structures than the conventional RTA process, which is beneficial to QD-based applications.

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