Curing Quantum Dots using Inductively Coupled Argon Plasma

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ABSTRACT

The crystal quality of InGaAs/GaAs quantum dots (QDs) is substantially improved without redistribution of composition using inductively coupled Ar plasma exposure. An increase in photoluminescence intensity by 1.7 times is observed in the plasma-treated QDs with the peak wavelength unshifted. The bandgap blue-shift subject to the rapid thermal annealing is also suppressed, denoting improved thermal stability. The PL excitation-dependent experiment shows more prominent state-filling phenomenon in the plasma-treated QDs due to higher carrier density by defect density reduction.

Keywords: quantum dot, intermixing, thermal annealing, argon plasma, photoluminescence

1 INTRODUCTION

Great advancement achieved in QD growth using molecular beam epitaxy and metal-organic vapor phase epitaxy (MOVPE) allows QD-based devices to demonstrate superior performances.[1] However, QD structures are highly-strained material systems grown under low temperature such that appreciable amount of defects exist in the materials inherently. Defects may affect device performance such as threshold current due non-radiative recombination and the material's thermal stability [2] as well such that bandgap shift is enhanced in annealing process. 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 bandgap blue-shift. [3-5]

In the previous study of argon inductively coupled plasma (ICP) process for quantum well (QW) intermixing, we have observed significant photoluminescence (PL) enhancement in InGaAsP/InP QW samples [6]. It is further and more clearly evidenced in an experiment using an AlGaAs/GaAs 5-QW structure [7], where the PL peak of a defect-rich QW was recovered in contrast to the PL peaks of the rest QWs. In this paper, we report the effect of the inductively coupled Ar plasma exposure on the crystal quality of an $In_{0.5}Ga_{0.5}As/GaAs$ QD structure fabricated by cycled monolayer deposition. The reduction of low temperature grown-in defects using inductively coupled argon plasma process in the InGaAs/GaAs samples is demonstrated. The optical property and the thermal stability are investigated in 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.

2 EXPERIMENTS

The InGaAs/GaAs QDs structure [8] shown in Fig. 1 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 ($n=2\times10^{18}$ 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 ($n=1\times10^{18}$ cm^{-3}) within a thickness of 10 nm in the center of each layer. On the top is a 600-nm-thick, Si doped ($n=2\times10^{18}$ 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.

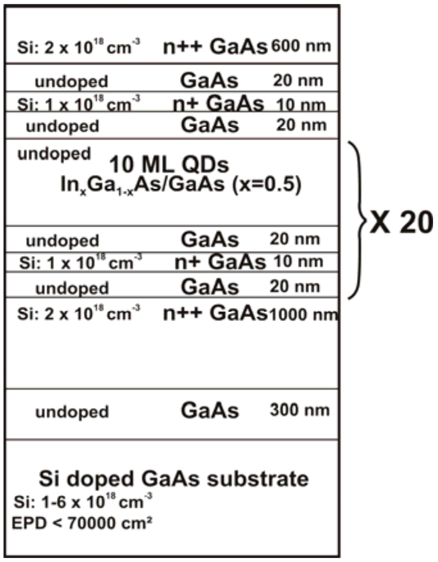

Figure 1: The growth structure of experimental samples

The plasma exposure was carried out using an ICP180 plasma source generator for 1~5 min under the following parameters: 100 sccm Ar flowrate, 60 mTorr chamber pressure, 480 W RF power and 500 W ICP power with a RF-induced DC bias of ~900 V. The annealing was performed at 700~750ºC for 30~120 s under flowing N2 ambient using GaAs proximity caps. PL spectra were measured at 77 K using a green crystal laser (532 nm, ~1 kW/cm2) for excitation, a monochromator and a TE-cooled Ge photodetector associated with a lock-in amplifier.

3 DISCUSSION

The PL spectra from QDs after Ar plasma exposure with the duration varied from 1 min to 5 min with no RTA process are shown in Fig. 2(a). The PL intensity after 1 min exposure is slightly reduced as compared with that of the as-grown one, which can be understood as a result of the roughened surface and created defects in the near-surface region by Ar ions bombardment. However, a significant increase in PL intensity emerges as the duration further increases, and the peak intensity reaches 2.7 times of the original level before saturation for the exposure duration of 3 min.

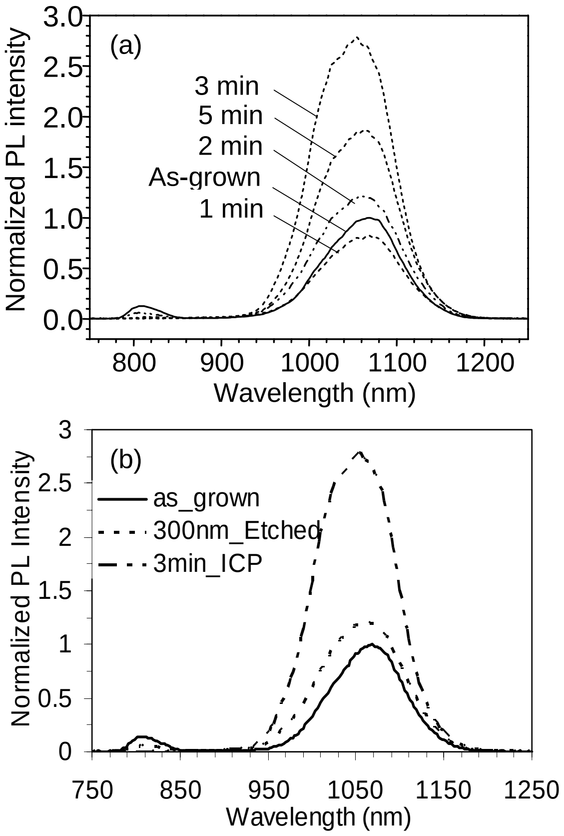

Figure 2: (a) PL spectra normalized to the as-grown sample taken from the as-grown and plasma-treated samples, and (b) PL spectra of an as-grown, a 300 nm chemically wet-etched, and a 3 minute

The sputtering effect in the plasma process may cause the reduction of thickness of the top GaAs layer. This might be one of the factors for the observation of PL intensity enhancement. To emulate the effect of thickness reduction, we etched 300 nm away chemically from an as-grown structure and compared its PL signal with the as-grown and the 3-min plasma exposed samples shown in Fig. 2(b)), since 3-min plasma exposure will sputter away the GaAs layer for ~300 nm in thickness. Compared with the as-grown sample, the 300nm-etched sample shows a 20% increase in PL, whereas the plasma exposed sample shows over a factor of two in PL intensity improvement. Therefore, the major effect for PL intensity enhancement observed in Fig. 2(a) is not due to the thickness reduction.

Previous investigations have demonstrated the increase in PL intensity using Ar plasma exposure due to annihilation of grown-in defects in QWs. [6,7] In this sample structure, QD layers were grown at much lower temperature (~515°C) than the optimal required for growing high quality lattice, an appreciable amount of LT grown-in defects present in the structure and may behave as non-radiative 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(a), denoting no change in the composition profile of QDs.

By the reduction of grown-in defects in the QD structure, the crystal quality is improved, and therefore better thermal stability. Good thermal stability is important when net differential bandgap shift is pursued in selective-area intermixing. In this investigation, the bandgap shift versus the plasma exposure duration was plotted in Fig. 3 using samples exposed to Ar plasma with different durations and annealed at 750ºC for 60 s. It shows that the bandgap shift decreases with increasing Ar plasma exposure duration, and a maximum bandgap shift reduction of 20 nm is obtained.

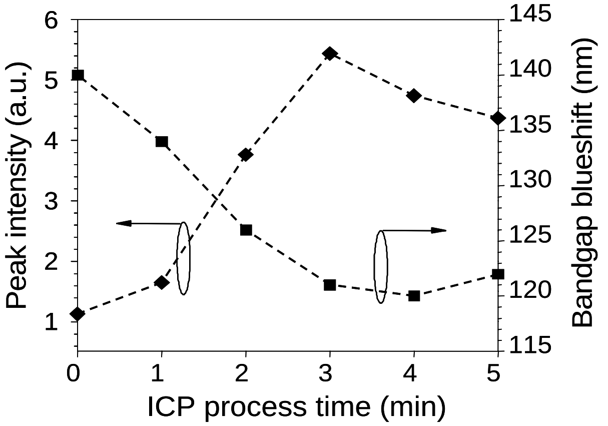

Figure 3: PL peak intensity and bandgap blueshift versus ICP process time. PL intensity was sampled after annealing for 60 s at 750ºC and is normalized to the PL peak of the as-grown sample.

The trend of PL intensity versus the plasma exposure duration is similar to that from unannealed samples used for Fig. 2, except that the PL peak intensity after annealing is higher. The highest PL peak intensity reaches up to 5.5 times of the original PL peak intensity. 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 only about 10% increase in PL peak intensity, denoting that the conventional RTA process is not as efficient as the Ar plasma exposure in annihilating grown-in defects in the crystal.

In this experiment using InGaAs/GaAs QD structure, we did not observed the intermixing enhancement by ICP plasma process in contrast to the large enhancement obtained in InGaAs/InP QW structures [10]. This is because that the interdiffusion is subjective to the collective effect of the near-surface defects and the grown-in defects. On one hand, ICP plasma process introduces a certain amount of defects in the near-surface region, and on the other hand, it reduces a certain amount of grown-in defects in the active region several hundred nanometers under the surface. Whether intermixing is enhanced or retarded depends on the net effect of these two facts. The reduction of grown-in defects in the QD structure is more prominent than the introduction of surface defects due to the presence of large quantity of low-temperature grown-in defects in the QD structure. It is noticed that the bandgap shift reduction decreases for long plasma exposure time, and in consistence, the PL intensity enhancement decreases as well. This suggests that the effect of the surface defect generation on intermixing comes into play as the surface defects have accumulated to a certain amount.

The bandgap shift reduction denotes better thermal stability of crystal due to the improvement of crystal quality. A maximum bandgap shift reduction of 20 nm is obtained with respect to the shift obtained from the annealed-only sample. This shift reduction was obtained with the presence of near-surface defects in the annealing process. It can be expected that better results can be obtained if the near-surface defects is removed by wet-etching the near-surface region away chemically before the annealing is done.

An excitation-dependent PL experiment was also done in an as-grown and a plasma-treated samples. The PL spectra as shown in Fig. 4. Under low level excitation, the PL spectra show only one peak which is corresponding to the ground-state interband transition. As the excitation level increases, more peaks appear due to the state-filling effect, and the additional peaks present due to interband transitions between the excited states in the conduction band and the valence band. The peaks for excited-state transitions grow stronger than the ground-state transition when the electron and hole densities are sufficiently high. In the as-grown sample (see Fig. 4(a)), the peak for the ground-state transition is always higher than the other peaks, although the emergence of these peaks can be observed from the spectra during the range of the PL excitation in the experiment. However, in the plasma treated sample (see Fig. 4(b)), the peak of the lowest excited-state transition emerges and becomes higher than the peak of the ground-state transition. This shows that higher electron and hole densities are achieved in the plasma treated sample under the same level of PL excitation. This is because of less consumption rate of electrons and holes from the non-radiative recombination, implying that the non-radiative defects have been reduced.

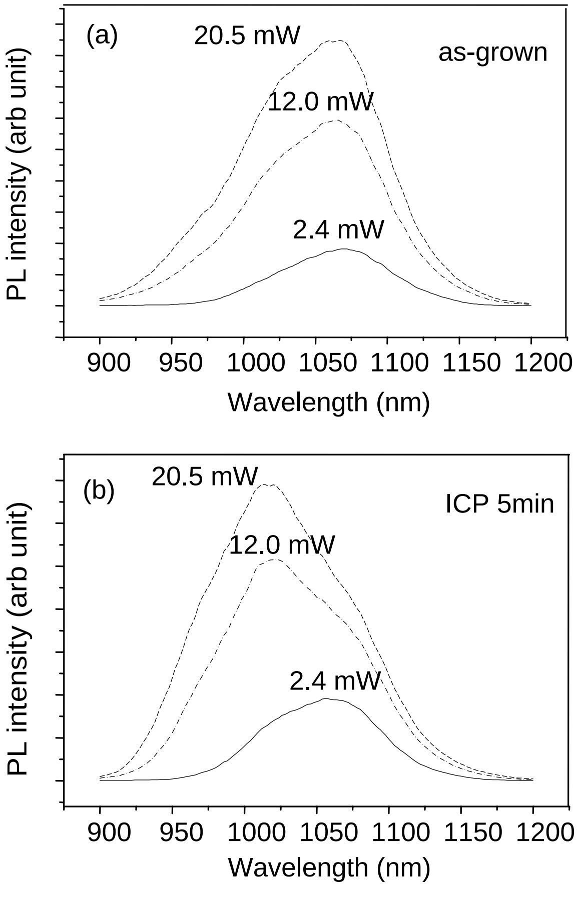

Figure 4: PL spectra under different powers of PL excitation source from (a) an as-grown sample and (b) a sample being treated by ICP for 5 min.

4 SUMMARY

In summary, we have demonstrated the improvement of crystal quality of InGaAs/GaAs QDs using inductively coupled Ar plasma exposure. PL intensity enhancement is observed immediately after plasma exposure without discernible bandgap shift. The thermal stability is also improved and suppression of bandgap shift can be clearly observed. 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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