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Xu, S. J.; Wang, X. C.; Chua, S. J.; Wang, C. H.; Fan, Weijun; Jiang, J.; Xie, X. G.

1998

Xu, S. J., Wang, X. C., Chua, S. J., Wang, C. H., Fan, W., Jiang, J., & Xie, X. G. (1998). Effects of rapid thermal annealing on structure and luminescence of self-assembled InAs/GaAs quantum dots. Applied Physics Letters, 72, 3335.

https://hdl.handle.net/10356/100261

https://doi.org/10.1063/1.121595

Effects of rapid thermal annealing on structure and luminescence of self-assembled InAs/GaAs quantum dots

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(Received 23 January 1998; accepted for publication 22 April 1998)

Postgrowth rapid thermal annealing was used to modify the structural and optical properties of the self-assembled InAs quantum dots grown on GaAs substrates by molecular beam epitaxy. It is found that significant narrowing of the luminescence linewidth (from 78.9 to 20.5 meV) from the InAs dot layer occurs together with about 260 meV blueshift at annealing temperatures up to 850 °C. Observation of high-resolution transmission electron microscopy shows the existence of the dots under lower annealing temperatures but disappearance of the dots annealed at 850 °C. The excited-state-filling experiments for the samples show that the luminescence of the samples annealed at 850 °C exhibits quantum well-like behavior. Comparing with the reference quantum well, we demonstrate significant enhancement of the interdiffusion in the dot layer. © *1998 American Institute of Physics.* [S0003-6951(98)03025-3]

The electronic and structural properties of self-assembled quantum dots (QDs) directly grown by the Stranski–Krastanow (S–K) method have attracted much attention due to interest in both their fundamental physics and potential device applications over the past few years.[1–6] The defect-free dots can be formed in lattice mismatched heterostructures by the S–K growth mode. However, the size distribution in the dots as evidenced from high-resolution transmission electron microscopy results in inhomogeneous broadening of the photoluminescence emission of the dots. Although better device performance such as higher differential gain, lower threshold current density of quantum dot lasers than that of quantum well (QW) lasers has been expected due to the δ-like density of states of the QDs, the size distribution of the self-assembled QDs continues to negate these advantages of the QD lasers and constitutes a technical barrier for the development of the QD lasers. Therefore, the need to control the size uniformity in QDs is a major goal. To this end, it is important to understand the detailed mechanisms of the nucleation and growth processes in the spontaneous formation of the QDs.[7,8] On the other hand, the post-growth thermal treatment has been recently used to modify the structural and optical properties of the self-assembled QDs.[9–11] Leon *et al.*[9] and Malik *et al.*[11] have reported the significant narrowing and blueshift of the luminescence emission of the self-assembled QDs by using the rapid thermal annealing (RTA) technique. However, the mechanisms resulting in the significant narrowing and blueshift of the QD luminescence are not clear. In this letter, we studied the influence of RTA on the structural and optical properties of the InAs/GaAs QDs with photoluminescence (PL) and high-resolution transmission electron microscopy (HRTEM) techniques. A large blueshift (about 260 meV) and significant narrowing (from 78.9 to 20.5 meV) of the PL peak of the InAs QDs annealed up to 850 °C were observed. An obvious increase in the integrated PL intensity was also observed for the samples annealed at temperatures up to 750 °C. However, at higher annealing temperatures a large decrease in the PL intensity was observed, implying a degradation in material quality. The TEM observation shows that the QDs still exist at lower annealing temperatures while the QDs disappear and combine with the wetting layer into a QW-like structure at an annealing temperature of 850 °C. Moreover, some dislocations were also observed in the sample annealed at 850 °C. These dislocations induce a large decrease in the emission intensity. Compared with the reference QW layer, the strongly enhanced interdiffusion of the QD layer is demonstrated.

The InAs quantum dots studied in the present work were grown by molecular beam epitaxy (MBE) on semi-insulating GaAs (100) substrate without rotation. The growth order of the whole structure is a 300 nm thick GaAs buffer layer, a 3 nm $In_{0.17}Ga_{0.83}As$ layer as a reference QW, a 100 nm GaAs barrier layer, a 6 Å (about 1.7 monolayers thick) InAs QDs layer, and a final 30 nm thick GaAs cap layer. The samples cut from the central region of the wafer were capped with about 200 nm thick SiO_2 and subjected to RTA treatment in a nitrogen ambient at temperatures ranging from 650 to 850 °C for 50 s. The SiO_2 film was then removed by dipping the samples in HF solution.

The PL measurements were performed using a standard phase lock-in technique. The excitation light is the 488 nm line of an Ar^+ laser. The luminescence signal is dispersed by a 0.75 m monochromator, and detected with a liquid nitrogen cooled Ge detector. The samples were mounted on the cold finger of a close-cycle helium cryostat with temperatures

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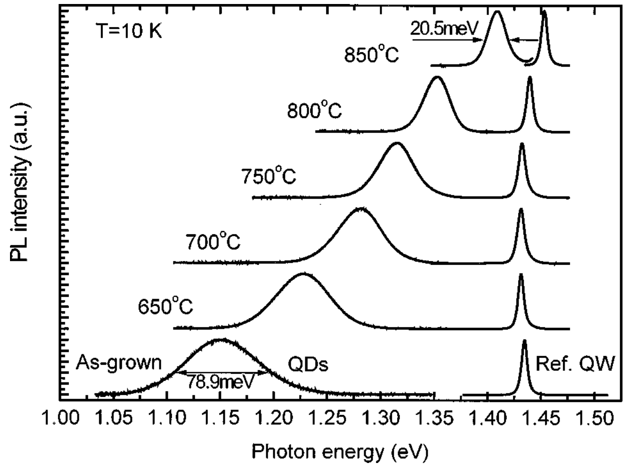

FIG. 1. 10 K photoluminescence spectra taken from the samples as-grown and annealed at different temperatures. The emission peaks at lower energy are from the QDs layers while the emission peaks at higher energy are from the reference QW layers.

varying from 4 to 300 K. The HRTEM observation was performed in Philips CM200/FEG.

Figure 1 shows the 10 K PL spectra measured from the samples annealed at different temperatures. The broad luminescence peak at lower energy is from the InAs dots while the narrow one at the higher energy is from the reference $In_{0.17}Ga_{0.83}As$ QW. For clarity, all spectra in Fig. 1 were normalized. For the QDs, a large blueshift (up to 260 meV) in the PL peak position and a significant narrowing of the PL linewidth from 78.9 meV of the as-grown sample to 20.5 meV of the sample annealed at 850 °C can be seen. In addition, there is an increase in the integrated PL intensity of the samples annealed at temperatures up to 750 °C, as shown in Fig. 2. These results are in agreement with those reported by Malik *et al.*[11] However, some new results have been obtained in the present work, such as our TEM observation, which demonstrates that the QDs disappear in the sample annealed at 850 °C. Moreover, no excited-state emissions can be seen even at high excitation power for the samples annealed at 850 °C. This is also evidence of the disappearance of the QDs at high annealing temperature.

Both the size and compositional changes of the QDs are thought to be responsible for the blueshift and the linewidth

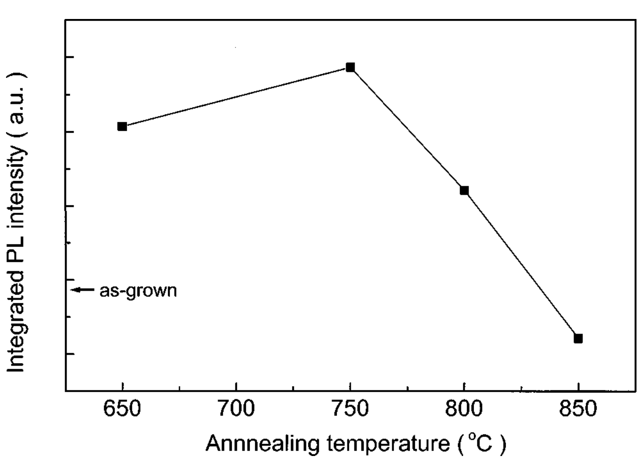

FIG. 2. The integrated PL intensity varies as a function of annealing temperatures.

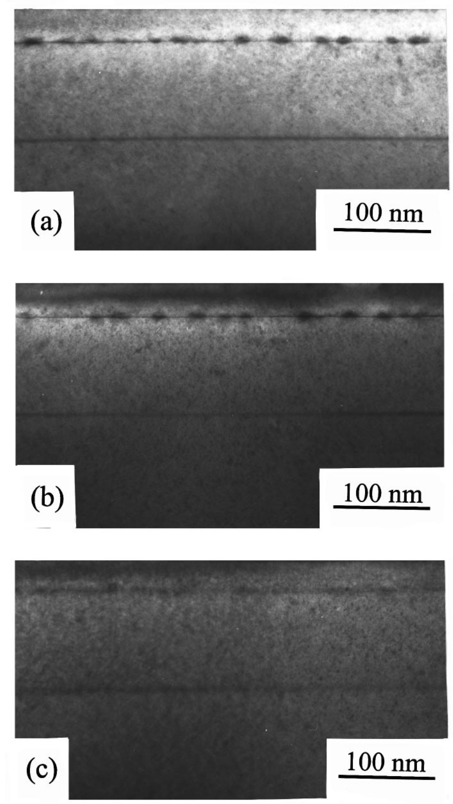

FIG. 3. The cross-section TEM images of the samples (a) as-grown, (b) annealed at 750 °C, and (c) annealed at 850 °C.

narrowing of their PL peaks.[7,9–12] The interdiffusion of the In and Ga atoms at the interface between the QD and the GaAs barrier results in a change in the size and the composition of the QDs during annealing. As for the enhancement of the PL intensity of the samples annealed up to 750 °C, it is believed to be due to the reduction of the nonradiative recombination centers by RTA.[13] On the other hand, too high an annealing temperature will degrade the structural quality of the samples, and thus induce a dramatic decrease in the PL intensity of the samples. This conclusion is further confirmed by our TEM observation.

Comparing the spectral variation between the QDs and the reference QW, the effect of RTA on the luminescent properties for the QW is much smaller than for the QDs. As seen in Fig. 1, almost no blueshift in emission energy of the QW takes place when the annealing temperatures are below 800 °C, indicating weak interdiffusion. Of course, like the QDs, an improvement in the emission intensity of the QW is still observed. Only when the annealing temperature was increased to 800 °C and above does clear blue shift in emission energy occur. This is in agreement with the previous observation[14] that the larger diffusion coefficients of the InGaAs/GaAs QWs occur in the annealing temperature range of 850–950 °C. It was found that high annealing temperatures also result in a decrease in the PL intensity of the QW.

It is known that there is a direct correlation between structural and optical properties of the self-assembled QDs.[7,15] Figure 3 shows the cross-section transmission electron microscope (TEM) images of the samples (a) as-grown, (b) annealed at 750 °C, and (c) annealed at 850 °C. From Fig. 3 it can be seen that the QDs still exist after annealing at

750 °C. But the QDs are difficult to be resolved for the sample annealed at 850 °C, implying the disappearance of the QDs. The strong intermixing at high annealing temperature destroys the dots. No similar observation was reported previously,[9–11] which may be due to lower annealing temperatures or shorter annealing time in their experiments. Figure 3(c) also shows that the layer containing the QDs transfers into a two-dimensional layer structure. The thickness of this ''new'' two-dimensional layer is even larger than that of the reference QW layer, which indicates the significantly enhanced interdiffusion of the QD layer during RTA. Moreover, the dislocations were observed in the sample annealed at 850 °C (not shown here). These results demonstrate that too high an annealing temperature cannot only destroy the self-assembled QDs but also degrade the material quality. This observation is consistent with the decrease in the luminescence intensity of the samples annealed at higher temperatures.

Finally, we would like to mention that the evolution of the strain in the QD layer during thermal annealing is an open problem, although the lattice mismatch strain is a controlling factor in the formation of self-assembled QDs in the heterostructure material systems. Further investigation into the effect of strain during RTA is ongoing.

In conclusion, we have studied the effect of RTA on the structural and optical properties of the InAs self-assembled QDs by using TEM and PL techniques. Large blueshift in emission energy and significant linewidth narrowing of the luminescence peaks have been observed for the annealed samples. They are attributed to the enhanced interdiffusion of the atoms in the QD layer resulting in changes in size and composition of the QDs. Our results demonstrates that RTA at suitable temperatures can result in a significant improvement of the structural and optical qualities of the self-assembled QDs. However, too high an annealing temperature can degrade the quality of the QDs and even destroy the QDs.

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