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Research on the photoluminescence of spectral broadening by rapid thermal annealing on InAs/GaAs quantum dots

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Abstract: Photoluminescence (PL) test was conducted to investigate the effect of rapid thermal annealing (RTA) on the optical performance of self-assembled InAs/GaAs quantum dots (QDs) at the temperatures of 16 and 300 K. It was found that after RTA treatment, the PL spectrum of the QDs sample had a large blue-shift and significantly broadened at 300 K. Compared with the as-grown InAs QDs sample, the PL spectral width has increased by 44.68 meV in the InAs QDs sample RTA-treated at 800 °C. The excitation power-dependent PL measurements showed that the broadening of the PL peaks of the RTA-treated InAs QDs should be related to the emission of the ground state (GS) of different-sized InAs QDs, the InAs wetting layer (WL) and the $In_{0.15}Ga_{0.85}As$ strain reduction layer (SRL) in the epitaxial InAs/GaAs layers.

Key words: quantum dots; rapid thermal annealing; photoluminescence; spectral width

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1. Introduction

Self-assembled InAs quantum dots (QDs) are usually prepared by heteroepitaxial growth on the GaAs(001) substrate, based on the Stranski–Krastanov (S–K) growth mode, in the molecular beam epitaxy (MBE). Among these InAs/GaAs QDs, three-dimensional carrier confinement can be achieved to realize a discrete energy spectrum with the density of states in the delta-function form. And that the discreteness of electron energy levels in InAs QDs effectively hinders carriers from migrating outside from InAs QDs to the radiation-free center in the GaAs or InGaAs matrix and the wetting layer (WL), and enhances inter-band transition inside QDs, which greatly improves the luminescence of QDs at room temperature[1]. These characteristics conspicuously improve the optoelectronic properties of the QDs heterostructure, laying the foundation for the research and development of a new generation of QDs optoelectronic devices such as laser diodes[2], solar cells[3], photodetectors[4] and optical amplifiers[5] based on the self-assembled QDs.

It is well-known that individual InAs QDs are randomly positioned in the epitaxial layer on the GaAs substrate, and in addition, these QDs have a size uniformity of around 10% due to randomness and statistical fluctuations during MBE growth[6, 7]. The randomness and inhomogeneity of the InAs QDs structural characteristics causes a significantly broadening of the QDs emission spectrum, which is harmful to some of the characteristics of the devices based on InAs QDs (i.e., the threshold of the QDs lasers). However, on the other hand, these characteristics can significantly improve the performance of ultra-wide spectrum devices, such as superluminescent diodes[8, 9]. To further tailor the size distribution and improve the optical performance of QDs, researchers utilized rapid thermal annealing (RTA) characterized by low cost, convenience and powerfulness[10, 11]. For instance, Triki *et al.* stated that the inter-diffusion of In and Ga atoms between InAs QDs and surrounding materials during the process of RTA increased the lateral size of QDs and changed the composition and morphology of InAs/GaAs QDs, thereby forming a shallower confining potential and leading to higher and nearer excitonic energies of QDs[12]. Saravanan *et al.* found that RTA treatment could significantly improve the optical quality and size distribution of QDs in a sample with six layers of InAs/GaAs QDs in the stack and increase the photoluminescence (PL) intensity of QDs by 5.6 times[13]. Adhikary *et al.* showed that defects and dislocations in the sample could be simultaneously eliminated by the post-growth RTA treatment of the InAs QDs[14]. Besides, they found that the composition of QDs and capping layer was effectively changed due to a large inter-diffusion effect during the RTA process, which increased the spectral width of the QD infrared detector by 45% compared with that of the as-grown sample[15]. These studies indicated that investigating the effect of the RTA treatment on self-assembled InAs/GaAs QDs was of great importance for both subsequent regrowth and thermal treatment

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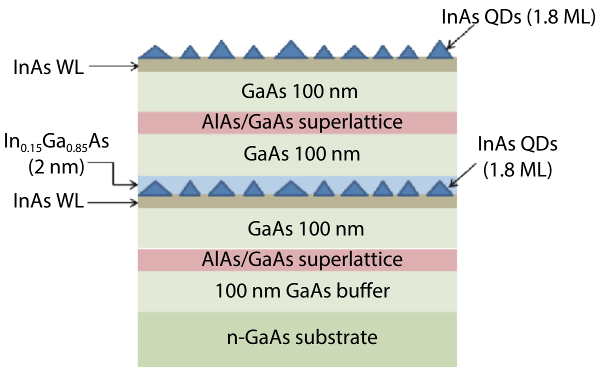

Fig. 1. (Color online) Schematic for the heteroepitaxy structure of InAs/GaAs QDs.

process in the fabrication of devices and the performance improvement of QDs devices[16].

In this paper, the RTA treatment was performed on the InAs/GaAs QDs at temperatures 750, 800, 850, and 900 °C, respectively. PL measurements was conducted on the as-grown and RTA-treated samples to investigate the effect of RTA on the optical quality of InAs QDs. The dependences of PL peak energy, PL spectral width and integrated PL intensity on annealing temperature used in RTA experiments were demonstrated and analyzed respectively. These results indicated that annealing temperature had a significant influence on the optical quality of InAs/GaAs QDs. In addition, it was demonstrated that the RTA treatment at 800 °C could significantly enlarge the PL spectral width of QDs, which greatly facilitated the fabrication of devices (i.e., superluminescent diodes[17]) that required a wide emission spectrum. The power dependence of PL revealed that the increase of the PL spectral width should be related to the ground state (GS) emission from the different-sized QDs, the WL as well as the strain reduction layer (SRL) in the MBE epitaxial layer.

2. Experimental

The InAs/GaAs QDs sample was grown on the semi-insulated GaAs (001) substrate by a Riber 32P solid source MBE system. Firstly, a 100 nm GaAs buffer layer and bottom barrier layer were successively grown on the GaAs substrate at 580 °C. Next, the temperature of the substrate was quickly cooled down to 500 °C to deposit 1.8 monolayer (ML) InAs QDs, which were covered by $In_{0.15}Ga_{0.85}As$ SRL with a thickness of 2 nm. Then, a 100 nm GaAs upper barrier layer was grown at 580 °C. 10-cycle 2 nm AlAs/2 nm GaAs superlattices (SLS) were grown before and after the deposition of the GaAs barrier layer to prevent photo-generated charge carriers from diffusing to the substrate to reach the surface. Finally, a 100 nm GaAs layer and an uncapped QDs layer identical to the buried layer were grown on the AlAs/GaAs SLS. Fig. 1 shows the schematic of InAs/GaAs QDs sample heteroepitaxy structure.

Our samples, cut from the central region of the as-grown epitaxial layer were subjected to RTA treatment in AccuThermo AW 610 system for 60 s in a nitrogen atmosphere at the annealing temperature T_{RTA} = 750, 800, 850 and 900 °C, respectively. During the RTA process, a GaAs wafer was used to cover these samples in order to prevent the samples from degradation due to arsenic volatilization from the surface at high temperature.

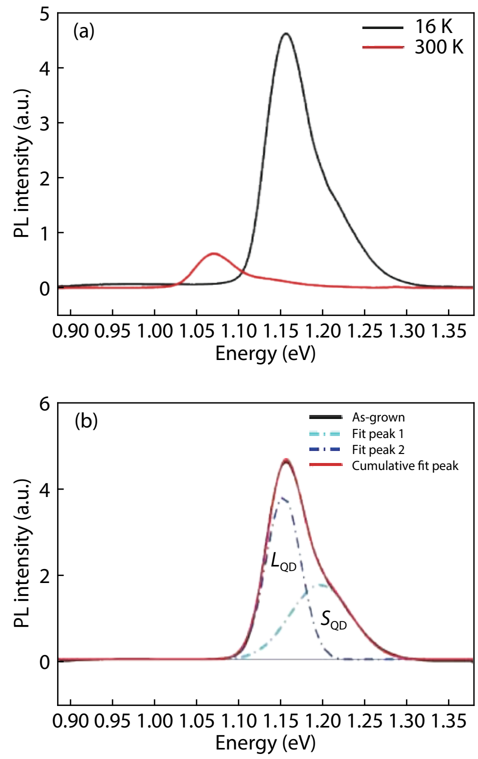

Fig. 2. (Color online) (a) The PL spectrum of the as-grown QDs sample under 14 mW of excitation power at 16 and 300 K respectively. (b) The Gaussian fitting diagram of the PL spectrum of the as-grown QDs sample at 16 K, with the dash-dot lines showing the Gaussian fitting of different emission peaks.

Samples were mounted in an enclosed circulating Helium cryostat, which allowed for tuning the temperature from 16 to 300 K. Temperature-dependent PL measurements were performed on all the samples using a 633 nm excitation wavelength under 14 mW of power. Then, the power-dependent PL measurement was performed, with an Nd:YAG laser of 532 nm at 300 K, on the annealed samples, as well as the as-grown sample.

3. Results and discussion

Fig. 2(a) demonstrates the PL spectrum of InAs/GaAs QDs in the as-grown sample recorded at the temperatures of 16 and 300 K, respectively, under 14 mW of excitation power. As shown in the figure, the full width at half maximum (FWHM) of these two PL spectra were 133.35 meV (16 K) and 196.18 meV (300 K), respectively. As usual, the PL intensity significantly decreased with the increase of temperature from 16 to 300 K, accompanied with the peak energy shifting from 1.155 eV at 16 K to 1.068 eV at 300 K. Besides, the PL spectrum of the as-grown InAs QDs at both 16 and 300 K was apparently asymmetric, with an obvious shoulder at the high-energy side of the PL peak.

Fig. 2(b) shows a Gaussian fitting diagram of QDs PL spectrum at 16 K. As illustrated by the dash-dot lines in Fig. 2(b), the asymmetric PL spectrum of QDs at 16 K can be well fitted using two Gaussian type functions with peaks at 1.153 and 1.197 eV respectively. In particular, the spacing between both Gaussian peaks was estimated to be around 44 meV, which was significantly smaller than the typical value (60–80 meV) of the energy separation between the GS and first ex-

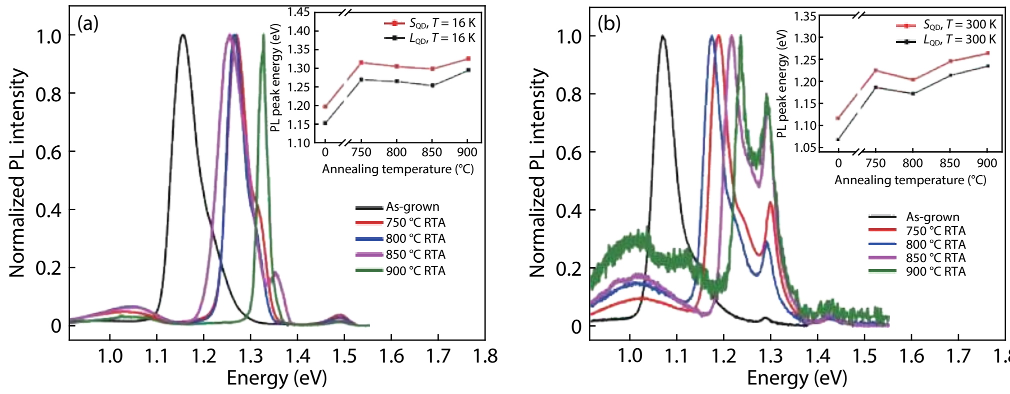

Fig. 3. (Color online) The normalized PL spectra of as-grown QDs and QDs annealed at various temperatures obtained at (a) 16 K and (b) 300 K under an excitation power of 14 mW. Inset: The PL peak energy of QDs at (a) T = 16 K and (b) T = 300 K with the change of annealing temperature, with the red curve representing small QDs and the black curve representing large QDs.

cited state of an InAs QD in a population of InAs QDs[18]. Therefore, the PL spectrum of the as-grown sample could be appropriately described by the GS emission of two QDs groups with two different mean QDs sizes.

Figs. 3(a) and 3(b) present the normalized PL spectrum of as-grown and RTA-treated QDs samples with T_{RTA} = 750, 800, 850 and 900 °C respectively at the temperatures of 16 and 300 K. As shown in both figures, all annealed QDs samples exhibited a distinct blue-shift in the PL spectrum (measured at the temperatures of 16 and 300 K) compared with as-grown QDs. Fig. 3(a) shows that in the PL spectrum at T = 16 K the main peak position of the QDs annealed at 750 °C was observed to be at 1.269 eV, which was blue shifted by 113 meV compared with that of the as-grown QDs (1.156 eV). However, the PL peak in the QDs annealed at T = 800 and 850 °C were blue shifted by 108.37 and 97.23 meV, respectively, which was slightly less than the RTA-induced blue-shift in the PL spectrum of the QDs annealed at 750 °C. Meanwhile, it can be observed that the sample annealed at 900 °C presented the largest blue-shift (169 meV). This behavior was completely different from other reports in the literature, to our knowledge, that the RTA-induced blue-shift increased significantly with the increase of annealing temperature[16, 19]. The strain-reducing effect could be enhanced since an appropriate annealing treatment could improve the crystal performance of InGaAs SRL[20]. It was speculated that the above experimental phenomenon might be caused by the inter-diffusion of In and Ga atoms between InAs QDs and surrounding materials during the RTA process or the reduction of strain between QDs and GaAs barrier layers after RTA. The inter-diffusion of In and Ga atoms from InAs QDs to SRL dominated for as-grown QDs annealed at 750 °C and was also possible for samples annealed at higher annealing temperatures (800 and 850 °C), while the inter-diffusion rate might be relatively lower than that of the interface between QDs and GaAs barrier[20, 21]. Therefore, the strain-reducing effect of samples annealed at 800 and 850 °C might be relatively dominant, which resulted in the obvious red-shift of PL spectrum peak compared with the samples annealed at 750 °C. In addition, the blue-shift of as-grown QDs annealed above 850 °C could be attributed to the inter-diffusion of In and Ga atoms among InAs QDs, $In_{0.15}Ga_{0.85}As$ SRL and GaAs barrier layer. However, RTA was relatively complicated due to the inhomogeneous size distribution of as-grown QDs, which could not be simply ascribed to one of the above mechanisms.

Furthermore, the PL peak energy of different-sized QDs was shown as a function of the RTA temperature in the insets of Figs. 3(a) and 3(b), and the similar variation trends were observed with the increase of annealing temperature.

In addition, the peak shape of the PL spectrum of the QDs sample subjected to RTA presented a slight modification. At 16 K, the asymmetry of the PL spectrum peak shape of samples annealed below 850 °C decreased with the increase of annealing temperature. The peak shape of the PL spectrum became sharper with annealing temperature increased to 900 °C. What is more, it was found that the PL spectrum of QDs annealed at 850 °C produced a new spectral peak at 1.354 eV compared with that of as-grown QDs. Moreover, it can be seen that the PL spectrum was significantly broadened when the temperature of PL measurements rose to 300 K. Therefore, the PL spectra in Fig. 3 were fitted by Gaussian fitting, and the curves of PL spectral width and integrated PL intensity versus annealing temperature were plotted as shown in Fig. 4.

Fig. 4(a) showed the PL spectral width of the RTA-treated QDs with the change of annealing temperature, measured at both T = 16 and 300 K. At 16 K, the PL spectral width of the QDs sample annealed at up to 750 °C was greater than that of the as-grown sample, which implied that RTA did not always induce the narrowing of PL spectral peaks. The PL spectral width of the samples annealed at 800 °C saw a sharp decline, which indicated that RTA treated InAs/GaAs QDs at an annealing temperature of 800 °C had relatively excellent crystal quality. With the further increase of annealing temperature, the PL spectral width increased, then decreased and obtained the minimum value at 900 °C. At 300 K, it can be observed that the PL spectral width changed from 216.4 meV to 240.86 meV with the increase of annealing temperature from 750 to 800 °C. Compared with as-grown QDs, QDs saw an increase of 44.68 meV in PL spectral width after being annealed at 800 °C. This result indicated that the electron–phonon scattering effect and thermal distribution had a significant impact on annealed samples with different QDs densities, leading to the significant broadening of PL spectrum[19, 22, 23]. Besides, it can be observed that further increasing annealing temperature gave rise to the shrinkage of the PL spectrum.

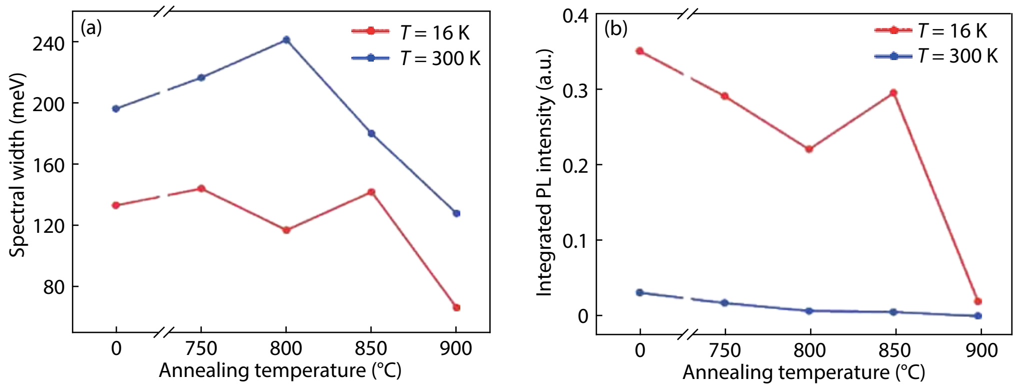

Fig. 4. (Color online) (a) The PL spectral width of QDs at $T = 16$ K (denoted by red curve) and $T = 300$ K (denoted by blue curve) with the change of annealing temperature. (b) The integrated PL intensity at $T = 16$ K (denoted by red curve) and $T = 300$ K (denoted by blue curve) with the change of annealing temperature.

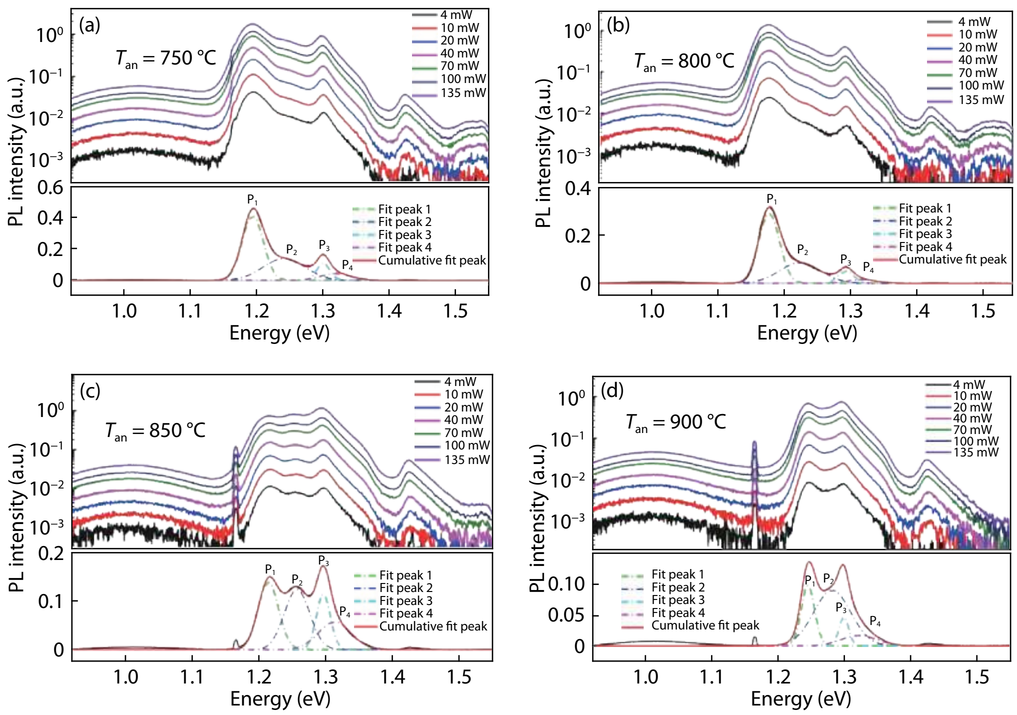

Fig. 5. (Color online) The top half of the figure shows the power-dependent PL spectrum of samples annealed at (a) 750, (b) 800, (c) 850, and (d) 900 °C recorded at 300 K. The PL spectrum of the annealed sample under an excitation power of 40 mW was extracted and Gaussian fitting was performed, as shown in the lower part of the figure.

When the RTA temperature rose to 900 °C, PL spectral width would be reduced to 128.09 meV. This could be explained by the fact that the inter-diffusion between the In-Ga atoms, which greatly improved the size distribution and composition fluctuation of QDs[10]. On the other hand, it might also be caused by the QDs gradually transformed into a quantum well (QW) structure, because the partial QDs dissolved in the surrounding WL at the higher annealing temperature.

Fig. 4(b) shows the integrated PL intensity of PL peak emission with the change of annealing temperature. The integrated PL intensity of RTA-treated samples at an annealing temperature of 750 °C and above saw a slight drop at 300 K, which was caused by the weakening of carrier confinement ability and the thermal excitation of more photo-generated carriers in QDs into the InAs WL or GaAs barrier layer. At low temperature (16 K), however, the integrated PL intensity was enhanced as samples were annealed at 850 °C, which might result from the reduction of non-radiative recombination and the contribution of lower thermal energy to increasing the integrated PL intensity of annealed QDs. The integrated PL intensity of QDs annealed at 900 °C was greatly attenuated owing to the partial dissolution of QDs and the degradation of the crystal quality of samples at high temperatures.

As mentioned above, the PL spectrum of the QDs subjected to RTA treatment showed a significant inhomogeneous broadening at 300 K, especially for the sample RTA-treated at 800 °C. And that, there are new peaks on the PL high energy side of all samples as shown in Figs. 3(b). To identify the source of the PL spectrum and explain its broadening, a power-dependent PL experiment was performed on all annealed samples. The upper half of Figs. 5(a)–5(d) demonstrates the PL spectrum of four annealed samples at 300 K as a function of excitation power from 4 to 135 mW. For data analysis, the PL spectrum of all annealed samples under the excitation power of 40 mW was extracted and Gaussian fitting was performed for data analysis. As shown in the lower half of Figs. 5(a)–5(d), the PL spectrum of four annealed samples was composed of four spectral peaks, namely P_1, P_2, P_3 and P_4.

Figs. 5(a) and 5(b) show that the peak intensities of the four spectral peaks P_1, P_2, P_3 and P_4 of RTA treated samples at an annealing temperature of 750 and 800 °C almost all decreased at a constant ratio. All spectral peaks existed even under the lowest excitation power, suggesting that these peaks were not derived from the luminescence of the excited state of QDs. In the PL spectrum, the first two peaks (P_1 and P_2) in the low-energy region showed the size distribution of QDs corresponding to the GS luminescence of QDs with different sizes. The increase of test temperature to 300 K resulted in the thermal excitation of some carriers in QDs into the WL where radiative or non-radiative recombination occurred. Therefore, the third peak (P_3) in the high-energy region was the luminescence peak of the QDs WL, which had a relatively small FWHM. Besides, it was suggested the fourth peak (P_4) at the high-energy side of the spectrum might be generated from the $In_{0.15}Ga_{0.85}As$ SRL. Thus, it can be seen that the broadening of the PL spectrum of QDs annealed at 750 and 800 °C was associated with the GS emission of different-sized QDs and the emission of the WL and SRL.

Figs. 5(c) and 5(d) are the PL spectrum and Gaussian fitting diagrams of InAs/GaAs QDs annealed at 850 and 900 °C, respectively. Based on Gaussian fitting, four peaks can be clearly defined on the high-energy side of PL spectrum, while the low energy side (1.165 eV) of the PL spectrum is related to the laser. For this kind of QD sample annealed at a higher temperature (850 or 900 °C), the lateral sizes of the QDs increased with increasing RTA temperature and the partial QDs dissolved into the surrounding WL, resulting in a modified WL and a gradual conversion of InAs QDs to an InGaAs QW structure[24]. From Figs. 5(c) and 5(d), it could be found that the GS PL peak tended to be saturated and the PL intensity of the third peak (P_3) was considerably enhanced with the increasing of the excitation power. In addition, the intensity ratio of QDs GS to P_3 decreased slightly with further increase of excitation power, which indicated that P_3 might be derived from the luminescence of QW formed by modified InAs WL and $In_{0.15}Ga_{0.85}As$ SRL. In the same way, this result further illustrated that the broadening of the PL spectrum was associated with the GS emission of different-sized QDs and the emission of the modified InAs WL and $In_{0.15}Ga_{0.85}As$ SRL caused by high-temperature RTA.

4. Conclusions

In summary, PL measurements were performed to investigate the effect of RTA treatment on the optical performance of self-assembled InAs/GaAs QDs. It was found that the variation in the PL blue-shift of InAs/GaAs QDs as a function of annealing temperature was associated with the reduction of strain between QDs and GaAs barrier layers after RTA and the inter-diffusion of In and Ga atoms at specific annealing temperatures. In addition, the PL spectrum could be broadened effectively by RTA at 300 K. The PL spectral width of the QDs sample annealed at 800 °C was 240.86 meV, which was obviously higher than that of the as-grown sample (196.18 meV). This feature of annealed samples is very applicable to optical coherence tomography. Therefore, this study broadened the PL spectrum of InAs/GaAs QDs by RTA.

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