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Effects of Interdiffusion on Luminescence of InAs/GaAs Quantum Dots Covered by InGaAs Overgrowth Layer*

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Abstract: The effects of postgrowth rapid thermal annealing have been studied on the optical properties of 3-nm-height InAs/GaAs quantum dots covered by 3-nm-thick $In_xGa_{1-x}As$ ($x=0$, 0.1 and 0.2) overgrowth layer. At a higher annealing temperature ($T\geqslant$ 750℃), the photoluminescence peak of InGaAs layer has been observed at the lower-energy side of InAs quantum-dot peak. In addition, a similar blueshift in photoluminescence (PL) emission energy is observed for all samples when the annealing temperature increases from 650 to 850℃. However, the trend of photoluminescence linewidth towards narrowing is totally different for InAs quantum dots with different In mole fraction in InGaAs overgrowth layer. The results suggest that the intermixing in the lateral direction plays an important role in obtaining a better understanding of the modification of optical properties induced by the rapid thermal annealing.

Key words: rapid thermal annealing; InAs quantum dots; InGaAs overgrowth layer

PACC: 8100; 8115; 8140G

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1 Introduction

The postgrowth thermal treatment has been recently used to modify the optical properties and tune the intersubband energy spacing of self-assembled quantum dots (QDs)[1—9]. The strong narrowing of photoluminescence (PL) linewidth, large blueshift in the intersubband transition energy and the significant reduction in intersublevel spacing energy due to the rapid thermal annealing (RTA) have been observed[1—8]. The experimental results indicate that the postgrowth thermal treatment provides a promising method for the application of quantum-dot structure in semiconductor QD lasers and detectors. The structural and optical properties of InAs/GaAs QDs subjected to RTA have been widely investigated by using the transmission electron microscopy and PL techniques. Although the strong narrowing and significant blueshift of QD luminescence emission can be attributed to the In-Ga interdiffusion between InAs QDs and GaAs barrier layer, the detailed mechanisms are still unclear. In addition, the investigation into the intermixing in lateral direction is rarely reported. In the final analysis, such question is to understand the optical changes of QDs subjected RTA.

In this paper, a QD structure is proposed to estimate the effects of intermixing in the lateral

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direction. The 3nm-height InAs/GaAs QDs covered by 3-nm-thick $In_xGa_{1-x}As$ (x= 0, 0.1, 0.2 and 0.3) overgrowth layer have been fabricated. The atomic force microscopy measurement indicates that the surface topograph of InAs islands covered by $In_xGa_{1-x}As$ ($x \leq 0.2$) becomes flatter. Because the thickness of InGaAs overgrowth layers equals the QD's height, the lateral intermixing is suppressed during the RTA process and the InGaAs overgrowth layer has few influences on the intermixing in the vertical direction. Therefore, the influence of lateral intermixing on the QD optical characteristics can be studied with these samples. Our experimental results indicate that the In-Ga intermixing in lateral direction has a strong influence on the reduction of PL linewidth but a weak effect on the blueshift in OD emission energy. In addition, the photoluminescence peak of InGaAs layer at lower-energy side of InAs quantum-dot PL peak has been observed as the samples are annealed at a higher temperature.

2 Experimental Process

Samples studied here were grown on the GaAs (100) substrates by the conventional solid source molecular beam epitaxy system (MBE) with a Riber 32P machine. The growth order of the sample structure is a 500nm GaAs buffer layer, a 2.0ML InAs QDs layer, a 3nm $In_xGa_{1-x}As$ (x= 0 for sample A, 0.1 for sample B, 0.2 for sample C, and 0.3 for sample D), and 100nm GaAs cap layer. The InAs and InGaAs layers were grown at 500℃ while the GaAs buffer and cap layers at 580℃. The formation of 3D islands was signaled by the abrupt change from a streaky reflection high-energy election diffraction pattern to a spotty one. After deposition of 100nm SiO_2, the samples were subjected to RTA in an argon ambient for 60s within the temperature range from 650 to 850℃. In a closed-cycle He cryostat and under the excitation of 514.5nm line of an Ar^+ laser, the SiO_2 layers were removed after we dipped the samples in HF solution for PL measurements. The luminescence spectra were detected by using a Fourier transform infrared spectrometer and an InGaAs photodetector.

3 Experimental Discussion

Figure 1 shows the 0.5μm × 0.5μm AFM image of the self-assembled 2.0ML InAs QDs grown on GaAs (100) substrate. The statistical analysis indicates that the average lateral size is

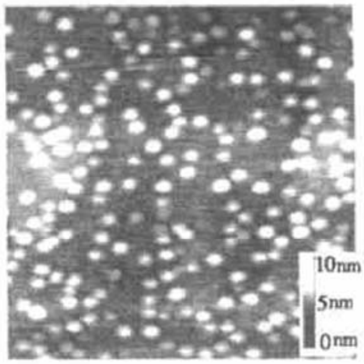

FIG. 1 0.5μm × 0.5μm Atomic Force Microscope Image of 2.0 Monolayer InAs QDs Grown on GaAs (100) Substrate

18.0±1.6nm and the height is 3.0±0.3nm. The dot density of samples is approximately 3.8×10^{10} cm^{-2}. Another AFM image (not shown here) shows that 2.0ML InAs QDs covered by 3nm $In_{0.2}$-$Ga_{0.8}As$ is almost flattened. Figure 2 shows the low-temperature PL spectra (at 77K) of sample C having been unannealed or annealed at different temperature (700—850℃). The postgrowth RTA treatment leads to a blueshift (up to 140meV) in the emission energy and a narrowing of the full width at half maximum (FWHM) from 35meV of the as-grown sample to 17meV of the samples annealed at 850℃. The changes induced by RTA are similar to those in InAs/GaAs QDs that has been reported. However, there is a lower-energy PL shoulder at 1.28 eV for sample C when it is annealed at 750℃, which has not been observed for In(Ga)As QDs subjected to RTA. Moreover, the increment in annealing temperature induces a redshift in the emission energy of lower peak.

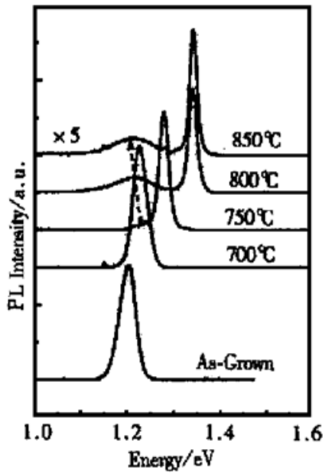

FIG. 2 Low-Temperature PL Spectra (at 77K) Obtained from InAs QDs with InGaAs Overgrowth Layer (Sample C) Unannealed and Annealed at Different Temperature (700—850℃) for 60s

A low-energy peak has also been detected in the PL spectra of InAs QDs covered by $In_{0.1}Ga_{0.9}$-As and $In_{0.3}Ga_{0.7}As$ layer. Figure 3 shows the emission from InAs/GaAs QDs with InGaAs overgrowth layer annealed at 800℃. However, no broad PL peak has been observed at the lower-energy side of InAs emission peak for sample A.

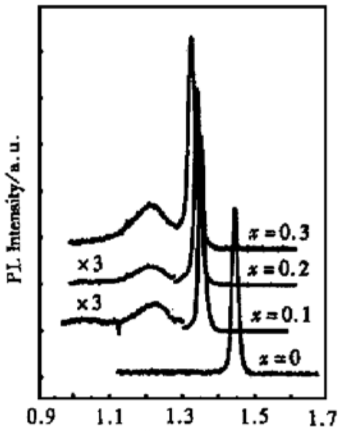

FIG. 3 Low-Temperature PL Spectra (at 77K) Obtained from InAs QDs with $In_xGa_{1-x}As$ (x = 0, 0.1, 0.2 and 0.3) Overgrowth Layer Annealed at 800℃ for 60s

These results indicate that the broad luminescence peak cannot be attributed to the InAs QDs, but because of the redshift in the PL energy with the RTA temperature increasing, shown in Figure 2[1—8]. It is believed that this peak corresponds to the emission of InGaAs overgrowth layer. The band gap of InGaAs overgrowth layer is larger than that of InAs QDs among the as-grown samples. Thus, the PL peak of InGaAs has not been detected in unannealed samples. For InAs/GaAs QDs, the RTA treatment results in a larger blueshift of QD peak energy. However, the effects of RTA on the optical properties of quantum wells (QWs) are smaller than those of QDs. The blueshift induced by postgrowth thermal treatment in PL energy of QWs cannot be clearly observed at a low annealing temperature ($T <$ 800℃)[2,8]. Therefore, with the annealing temperature increasing, the band gap of InAs QDs become larger than that of InGaAs quantum well. With the annealing temperature further increasing, the interdiffusion of In-Ga will lead to the increment in In mole fraction in the InGaAs overgrowth layer as well as the redshift of PL energy.

Next, we will discuss the relative changes in optical properties induced by RTA for InAs/GaAs QDs with different In-mole-fraction InGaAs overgrowth layer. Figure 4 shows the annealing temperature dependences of the peak positions of

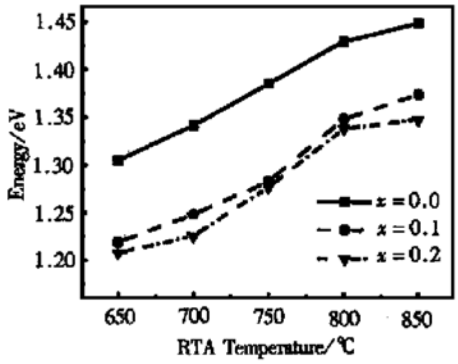

FIG. 4 Annealing Temperature Dependence of PL Energy of Sample A, B and C

PL spectra for sample A, B, and C. It can be clearly seen that the trends of blueshift in emission energy are similar for all samples. However, the reduction of PL linewidth depends strongly on the composition of InGaAs overgrowth layer, as is shown in Fig. 5. It decreases with the In mole fraction in the InGaAs overgrowth layer increasing. At the annealing temperature of 850℃, the FWHM of sample C becomes the largest. This unexpected result is because of the smallest PL linewidth of sample C among the as-grown samples.

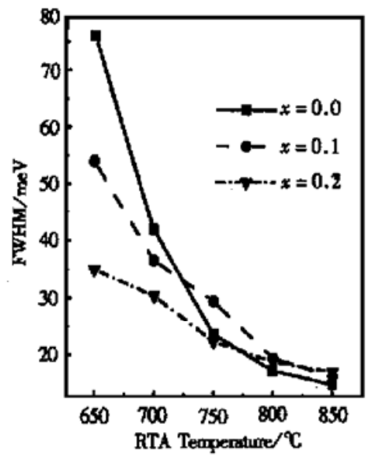

FIG. 5 Annealing Temperature Dependence of FWHM of PL Peaks of Sample A, B and C

Figure 6 (a) and (b) shows the cross-sectional TEM pictures of InAs QDs covered by GaAs and 3nm $In_{0.2}Ga_{0.8}As$ strain-reduced layer, respectively. For InAs QDs covered by GaAs layer, the dark contrast in Fig. 6 (a) expands from InAs QDs to the GaAs cap layer. The dark contrast around the InAs QDs results from the strain induced in GaAs cap and buffer layer. In comparison with Fig. 6 (a), the dark contrasts in $In_{0.2}Ga_{0.8}As$ overgrowth and buffer layers in Fig. 6 (b) are weaker, which suggests that the overgrowth InGaAs layer grown on the InAs dots can reduce the strain in the growth direction of QDs. As a result, the peak position of the latter is much lower than that of the former.

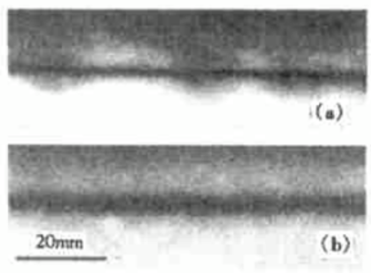

FIG. 6 Cross-Sectional TEM Images of 2.0 ML InAs QDs Covered by (a) GaAs Cap Layer and (b) 3nm-Thick $In_{0.2}Ga_{0.8}As$ Overgrowth Layer and GaAs Cap Layer

Since the height of QDs is much smaller than their lateral size, the blueshift in PL peak energy induced by RTA mainly results from the change in vertical direction[6]. As for our samples, the thickness of InGaAs overgrowth layer is equal to that of the InAs QDs. The In-Ga intermixing in vertical direction is very alike for all the samples. Therefore, the trends of blueshift in PL energy shown in Fig. 3 are similar for all samples. On the other hand, the relative changes in PL linewidth can be explained in terms of the influence of lateral intermixing between InAs QDs and barrier layer. For the InAs QDs covered by GaAs, the In-Ga intermixing in lateral direction is larger than that in vertical direction, due to the anisotropic strain distribution at the interface[7,8,10]. Therefore, the relative quantum effects in lateral direction of QDs become weaker when the QD samples are annealed at a higher temperature. However, the lateral intermixing can be suppressed for the samples covered by InGaAs, resulting in the relative increment in quantum effects in lateral direction. For samples subjected to RTA, the effects of lateral size distribution on the PL linewidth will be enhanced with the increase of the In mole fraction in overgrowth layer. Therefore, the changes of InAs/GaAs QD PL linewidth induced by RTA will decrease with the increase of the In mole fraction in InGaAs overgrowth layer.

4 Conclusion

In conclusion, the effects of lateral intermixing between QDs and barrier layer have been investigated by the RTA treatment of InAs/GaAs QDs covered by InGaAs overgrowth layer. Similar trend of blueshift in PL QD energy is found in all samples. However, the changes of PL linewidth decrease with the increase of the In mole fraction in the overgrowth layer, as can be attributed to the suppression of lateral intermixing. In addition, at a higher annealing temperature ($T \geqslant 750$℃), the photoluminescence peak of InGaAs layer has been observed at the lower-energy side of the InAs quantum-dot photoluminescence peak.

References

[1] C. Lobo, R. Leon, S. FafardandP. G. Piva, Appl. Phys. Lett., 1998, **72**: 2850.

[2] S. J. Xu, X. C. Wang, S. J. Chua, C. H. Wang, W. J. Fan, J. Jiang and X. G. Xie, Appl. Phys. Lett., 1998, **72**: 3335.

[3] Q. W. Mo, T. W. Fan, Q. Gong, J. Wu, Z. G. Wang and Y. Q. Bai, Appl. Phys. Lett., 1998, **73**: 3518.

[4] R. Leon, Yong Kim, C. Jagadish, M. Gal, J. Zou and D. J. H. Cockayne, Appl. Phys. Lett., 1998, **69**: 1888.

[5] F. Heinrichsdorff, M. Grundmann, O. Stier, A. Krost and D. Bimberg, J. Cryst. Growth, 1998, **195**: 540.

[6] S. Fafard and C. Ni. Allen, Appl. Phys. Lett., 1999, **75**: 2347.

[7] T. M. Hsu, Y. S. Lan, W.-H. Chang, N. T. Yeh and J.-I. Appl. Phys. Lett., 2000, **76**: 691.

[8] X. C. Wang, S. J. Xu, S. J. Chua, Z. H. Zhang, W. J. Fan, C. H. Wang, J. Jiang and X. G. Xie, J. Appl. Phys., 1999, **86**: 2687.

[9] S. Malik, C. Roberts, R. Murray and M. Pate, Appl. Phys. Lett., 1997. **71**: 1987.

[10] A. O. Kosogov, P. Werner, U. Gosele, N. N. Ledentsov, D. Bimberg, V. M. Ustinov, A. Yu. Egorov, A. E. Zhukov, P. S. Kop´ev, N. A. Bert and Zh. I. Alferov, Appl. Phys. Lett., 1996, **69**: 3072.

快速热退火对带有 InGaAs 盖层的 InAs/GaAs 量子点发光特性的影响*

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摘要: 系统地研究了快速热退火对带有 3nm $In_xGa_{1-x}As$ (x= 0, 0.1, 0.2) 盖层的 3nm 高的 InAs/GaAs 量子点发光特性的影响. 随着退火温度从 650℃上升到 850℃, 量子点发光峰位的蓝移趋势是相似的. 但是, 量子点发光峰的半高宽随退火温度的变化趋势明显依赖于 InGaAs 盖层的组分. 实验结果表明 In-Ga 在界面的横向扩散在量子点退火过程中起了重要的作用. 另外, 我们在较高的退火温度下观测到了 InGaAs 的发光峰.

关键词: InAs/GaAs 量子点; InGaAs 盖层; 快速热退火

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