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Growth of Space Ordered 1.3μm InAs Quantum Dots on GaAs(100) Vicinal Substrates by MOCVD*

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Abstract: Space ordered 1.3μm self-assembled InAs QDs are grown on GaAs(100) vicinal substrates by MOCVD. Photoluminescence measurements show that the dots on vicinal substrates have a much higher PL intensity and a narrower FWHM than those of dots on exact substrates, which indicates better material quality. To obtain 1.3μm emissions of InAs QDs, the role of the so called InGaAs strain cap layer (SCL) and the strain buffer layer (SBL) in the strain relaxation process in quantum dots is studied. While the use of SBL results only in a small change of emission wavelength, SCL can extend the QD's emission over 1.3μm due to the effective strain reducing effect of SCL.

Key words: InAs; quantum dots; MOCVD

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1 Introduction

Self-assembled InAs/GaAs quantum dots (QDs) are intensively studied due to their δ-like density of states[1~6]. Laser structures containing QDs as active material are expected to have high differential gain, low threshold current, and high characteristic temperature[7], which would make it possible that the InP based devices will be replaced by the less expensive GaAs based devices. For optical fiber communication, the pursuit of 1.3μm or 1.5μm QD lasers is of great importance. However, emission wavelength from self-assembled InAs QDs is typically 1.0μm. A widely used method to extend the InAs/GaAs QDs to 1.3μm and beyond is to use the strain reducing layer to place the InAs dots in or below an InGaAs matrix[3]. To fully utilize their superior electronic and optical properties, spatial ordering of QDs is desired. A certain degree of lateral ordering of InAs QDs can be achieved by using GaAs (100) misoriented substrates[8]. A few works about the optical properties of QDs on misoriented substrates have been reported. It was shown that the misorientation of the substrates led to a decrease of the FWHM of photoluminescence (PL) lines and a blue shift of the emission wavelength[9].

In this paper, InAs QDs were grown on GaAs (100) vicinal substrates by MOCVD. Space ordered QDs in lines were obtained with good PL properties. To obtain a 1.3μm wavelength emission, the role of the matrix in the strain relaxation process in quantum dots was studied. The InGaAs layer was put above or below

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the dot as a strain cap layer (SCL) or a strain buffer layer (SBL), respectively. Our PL results show that the SCL is more effective in shifting the emission peak of quantum dot than SBL.

2 Experiment

All the QD samples were grown via a horizontal low-pressure MOCVD reactor (AIXTRON-200) at a total pressure of 600Pa, with Pd-diffused H_2 as carrier gas. Trimethylgallium (TMGa), trimethylindium (TMIn), arsine (AsH_3) were used as source materials. As shown in Fig. 1, two sets of samples were prepared, including InAs QDs put above an InGaAs SBL and below an InGaAs SCL with different In composition. After the growth of the 200nm GaAs buffer layer on (100) 2°off (110) Si-doped GaAs substrate, either InAs QD or InGaAs SBL was deposited first, followed by the deposition of InGaAs SCL or InAs QD layer for the two kinds of samples, respectively. Finally, the samples were covered with 50nm GaAs. The growth temperatures were 600 ℃ for the GaAs buffer, and 507 ℃ for the InAs QDs and SCL/SBL and GaAs cap layer. The V/III ratio are 25 for GaAs and 5 for InAs QDs growth and InGaAs SBL (SCL) growth. We use the growth rate of 0.034ML/s and total thickness of 1.7ML during InAs growth. In order to examine the quantum dot emission peak shift with respect to the InGaAs matrix, the $In_xGa_{1-x}As$ SCL and SBL were grown with the In composition varying from $x = 0$ to $x = 0.25$. For AFM and SEM measurements, the samples were cooled down rapidly after the formation of QDs.

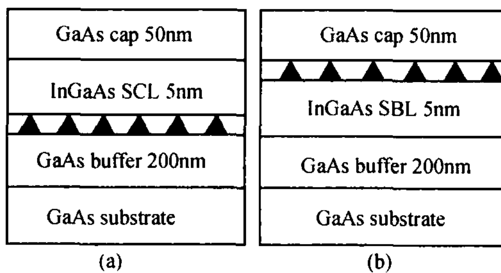

Fig. 1　Schematic of InAs QD structures with SCL (a) and SBL (b)

The SEM measurements were performed using a XL30-FEG microscope at 20kV. A nanoscope dimension 3100 SPM system with a tapping mode in air was used for AFM measurements. The PL measurements were carried out in closed-cycle He cryostat under the excitation of 514.5nm line of Ar^+ laser focused onto a $0.5mm^2$ spot. The luminescence spectra were detected with a Fourier transform infrared spectrometer operating with an InGaAs photodetector.

3 Results and discussion

Figures 2(a) and (b) show the SEM image and the corresponding lateral size histogram of the InAs QDs formed on GaAs substrates without a cap layer respectively. As can be seen, QDs were formed in lines and exhibited a clear bimodal size distribution with two groups of large or small dots. The density of the large and small QDs is $0.91 \times 10^{10} cm^{-2}$ and $4.05 \times 10^{10} cm^{-2}$, respectively. In contrast, the SEM image shown in Fig. 2(c) and the corresponding lateral size histogram in Fig. 2(d) reveal that InAs QDs on exact GaAs (100) substrates prepared in the same growth run had only one dominant size and were randomly formed with a density of $1.21 \times 10^{10} cm^{-2}$, which is much smaller than that of the dots on vicinal substrates. The formation of dots in lines is due to QDs preferably nucleate along the multiatomic step edges on the vicinal substrate[10], as shown clearly in the AFM image in the inset of Fig. 2(a). According to the research work of Leon *et al*.[8], surfaces with multiatomic steps enable the formation of smaller critical nuclei for stable island growth, since nucleation on steps is energetically favorable. The effects of smaller critical nuclei for stable nucleus formation result in smaller average diameters and higher island densities when step nucleation (heterogeneous nucleation) is predominant, which is the case when the adatom diffusion lengths are larger than the step spacing. In thermaldynamic equilibrium, only the larger QDs are expected due to

their lower total energy consisting of strain, surface, and boundary energy. The bimodal size distribution of QDs on the vicinal surface can be related to the reduced surface diffusion during MOCVD growth. The small QDs formed on step edges are not in thermal equilibrium and will subsequently develop into the energetically favorable large QDs. However, on the vicinal substrates, migration of adatoms on the surface is prohibited due to the energy barrier formed at the step kinks[11], which slows down the transformation of the small QDs into the energetically favorable large dots, and in such cases a bimodal size distribution appears.

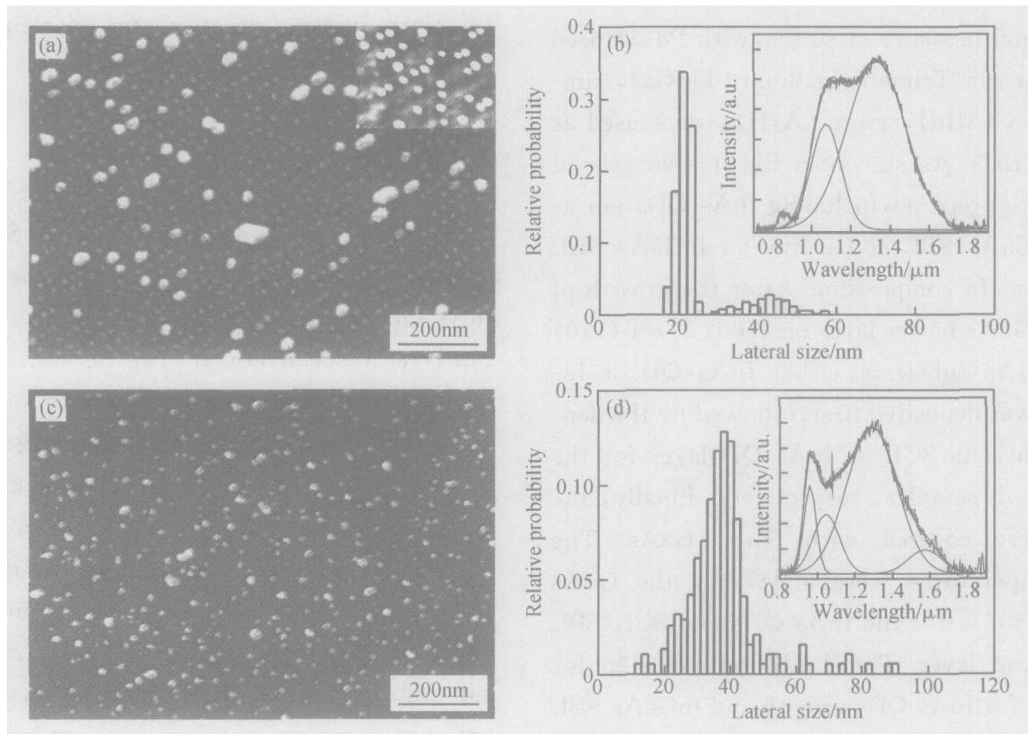

Fig. 2 SEM images (a) and the corresponding size histograms (b) of QDs on vicinal GaAs(100) substrates The SEM image and the corresponding size histograms of surface QDs on exact substrates is shown in (c) and (d). Inset of (a): a 300nm × 250nm AFM image showing the dots formed on step edges. Inset of (b) and (d) show the 77K PL spectra of surface dots on vicinal substrates and exact substrates, respectively.

The 77K PL spectrum of surface QDs on vicinal substrates is shown in the inset of Fig. 2(b), which can be decomposed into two peaks with Gaussian shape. Since the PL spectra show no excitation power dependence, the two peaks are attributed to the large and small groups of dots, respectively. This indicates that most of the two groups of dots are coherently strained, though there are some irregular shaped large dots which may be dislocated, as shown in Fig. 2(a). The inset of Fig. 2(d) shows the 77K PL spectrum of surface QDs on exact substrates. As can be seen, the spectrum can be decomposed into at least four peaks with Gaussian shape. While the peak at about 0.96μm can be attributed to the emission of the wetting layer, the other three are attributed to groups of dots with different sizes, reflecting the wide size distribution of dots on exact substrates.

Figure 3 shows the room temperature PL spectra of InAs QDs with InGaAs SCL of different In compositions. Different from the PL spectra of surface dots which exhibit a multi-peak feature, those of the capped QDs show only one dominant emission peak. We attribute this change of PL spectra to the strain-enhanced In/Ga interdiffusion of dots during the overgrowth of the GaAs cap lay-

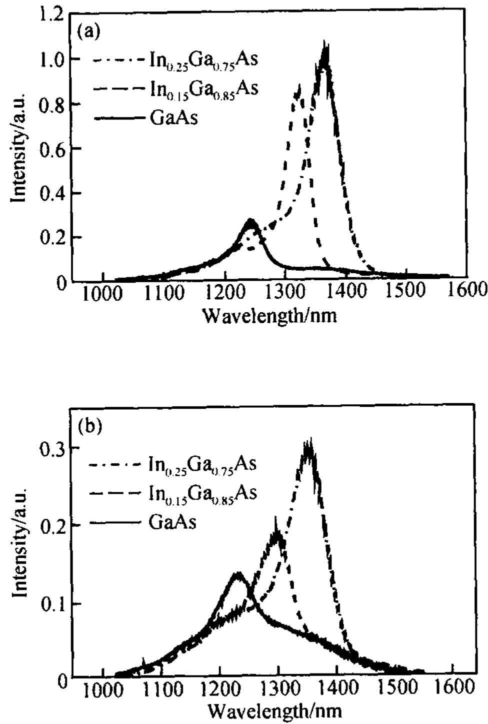

Fig.3 PL spectra of InAs QDs with InGaAs SCL of different In compositions (a) Dots on vicinal substrates; (b) Dots on exact substrates All the data are normalized to the PL intensity of dots on vicinal substrate with $In_{0.25}Ga_{0.75}As$ SCL.

er. There is residual strain in and around the dots. Compared to the small dots, the large dots have larger strains[4] and therefore they obtain a stronger interdiffusion of In/Ga atoms during GaAs overgrowth. A decrease of dot size difference between the two groups of dots during the cap layer growth is then expected, with the two emission peaks merging into one. The PL results are summarized in Table 1. As can be seen, compared with dots grown on exact substrates, dots on vicinal substrates have narrower FWHM, which is similar to the report in Ref. [9]. What is different from the results in Ref. [9], however, is that dots on vicinal substrates also have a much larger PL intensity, which indicates better material quality and is favorable for device use. This can also be attributed to the presence of steps on the substrates. The energy barrier at the steps reduces the reservoir of atoms available for dot growth, thus reducing the dot's possibility of growing so large that dislocation forms. The strong binding effect of step edges leads to the size saturation of the dots, resulting in a narrower size distribution relative to dots on exact substrates. This is evident in our SEM measurements. The lateral size dispersion is 13% for dots on vicinal substrates, smaller than that of dots on exact substrates, which is 25%. As reported in Ref. [12], the presence of very large dots containing dislocations can influence the PL emission intensity due to the nonradiative recombination in the vicinity of these defects. As can be seen from Fig.2, the number of the very large irregular shaped dots on exact substrates is much larger than that of the dots on vicinal substrates, making the PL intensity of the dots on exact substrates much lower.

Table 1 Dependence of PL intensity and FWHM on the substrates and cap layer used

Parameter		GaAs	$In_{0.15}Ga_{0.85}As$	$In_{0.25}Ga_{0.75}As$
Intensity /a.u.	Exact	0.1359	0.1898	0.2974
	Vicinal	0.259	0.8518	1
FWHM /nm	Exact	123	82	81
	Vicinal	59	46	63

As shown in Fig.3, for dots on both the two kinds of substrates, the PL peaks are shifted toward longer wavelengths by increasing the In composition of $In_xGa_{1-x}A$ SCL. Taking dots on vicinal substrates as an example, the emission wavelength is 1.246μm when the QDs are directly capped with GaAs and increases to 1.321μm when capped by $In_{0.15}Ga_{0.85}As$ SCL. Further increase is observed to be 1.368μm as $In_{0.25}Ga_{0.75}As$ SCL is used. In the figures, with the increasing of In in the SCL, the PL intensity became stronger. Compared to Ref. [5], our data show that we can get long emission wavelengths while maintaining good material quality within a wider range of In composition in InGaAs. The extension of emission wavelengths for QDs is believed to be dominated by the compressive hydrostatic strain[3]. The relaxation of compressive strain in InAs QDs increases as more In is added into the InGaAs SCL. The increase of PL in-

tensity indicates that the relaxation of compressive strain in InAs QDs is dominant and no dislocation is formed within the range of our In composition.

In contrast to the conditions of SCL, with the increase of In composition of the InGaAs SBL, only a small change of QD's emission wavelength was observed, as shown in Fig. 4(a). With $In_{0.15}Ga_{0.85}As$ SBL, QD's PL peak is only about 10nm longer than that of dots grown directly on GaAs. Our results show that the InGaAs is more effective in manipulating the emission wavelength as it is placed above the QD layer. This can be first attributed to the larger contact area between the QDs and the SCL compared to the case of having the SBL beneath the dots. Thus the SCL is more effective in

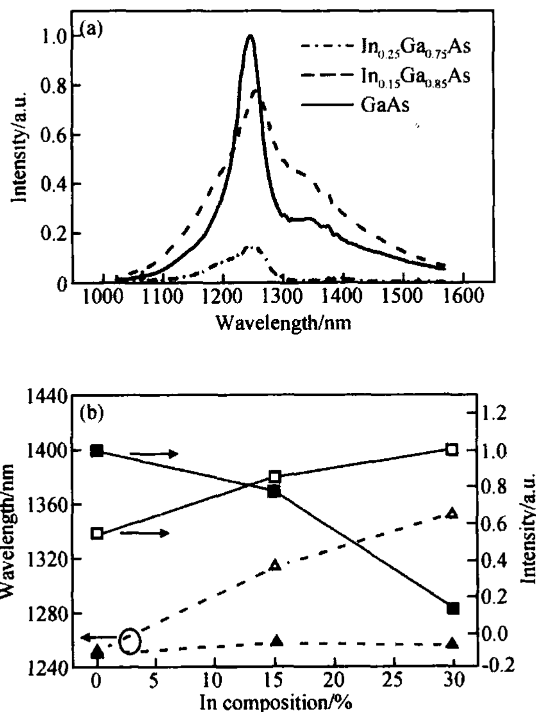

Fig. 4 (a) PL spectra of InAs QDs on vicinal substrates with InGaAs SBL of different In compositions; (b) Dependence of PL intensity and peak position on different In compositions (filled symbol for dots with SBL, open symbols for dots with SCL)

reducing the strain in the InAs QDs. Secondly, as reported by Liu *et al.*[13], though the density of dots increases with the increase of In composition in InGaAs SBL, the height of the dots decreases, which will blueshift the emission wavelength. Contrary to the PL results of dots with SCL shown above, the PL intensities of QDs with SBL decrease quickly with the increase of In composition in SBL, as shown in Fig. 4(b). This further indicates the more effective strain reducing effect of SCL than that of SBL. With the increase of In in SBL, the rapid buildup of strain in the QD system leads to the fast increase of the nonradiative recombination centers, which results in the decrease of PL intensity.

4 Conclusion

1.3μm self-assembled InAs QDs with space ordering were grown on GaAs(100) vicinal substrates. It is shown that they have a better material quality than that of dots on exact substrates. The role of the InGaAs strain cap layer and the strain buffer layer in the strain relaxation process in quantum dots was studied. While SCL can extend the QD's emission effectively over 1.3μm, the use of SBL resulted in only a small change of emission wavelength with a rapid decrease of material quality.

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用 MOCVD 在偏(100) GaAs 基片上生长空间规则排列的 1. 3μm InAs 量子点*

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摘要: 用 MOCVD 技术在偏(100) GaAs 衬底上生长了发光波长在 1. 3μm 的线状空间规则排列 InAs 量子点. 光致发光实验表明 ,相对于正(100)衬底 ,偏(100) GaAs 衬底上生长的 InAs 量子点具有更好的材料质量 ,光谱有更大的强度和更窄的线宽. 为了得到发光波长为 1. 3μm 的量子点 ,对比研究了不同 In 含量的 InGaAs 应力缓冲层(SBL) 和应力盖层(SCL)的应力缓冲作用. 结果表明 ,增加 SCL 中 In 含量能有效延伸量子点发光波长到 1. 3μm ,但是随着 SBL 中 In 的增加 ,发光波长变化不明显 ,并且材料质量明显下降.

关键词: InAs; 量子点; MOCVD

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