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Dependence of bimodal size distribution on temperature and optical properties of InAs quantum dots grown on vicinal GaAs (100) substrates by using MOCVD*

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Self-assembled InAs quantum dots (QDs) are grown on vicinal GaAs (100) substrates by using metal-organic chemical vapour deposition (MOCVD). An abnormal temperature dependence of bimodal size distribution of InAs quantum dots is found. As the temperature increases, the density of the small dots grows larger while the density of the large dots turns smaller, which is contrary to the evolution of QDs on exact GaAs (100) substrates. This trend is explained by taking into account the presence of multiatomic steps on the substrates. The optical properties of InAs QDs on vicinal GaAs(100) substrates are also studied by photoluminescence (PL) . It is found that dots on a vicinal substrate have a longer emission wavelength, a narrower PL line width and a much larger PL intensity.

Keywords: self-assembled quantum dots, indium arsenide, bimodal size distribution, MOCVD
PACC: 8115H, 7865P, 7820

1. Introduction

Self assembled quantum dots (QDs) are of great importance for applications in optoelectronic devices due to their δ-like density of states.[1] There has been considerable interest in both the fabrication and the theoretical study of Stranski–Krastanow mode semiconductor quantum dots as potential nanostructures applicable to QD-based devices.[2−8] While InAs/GaAs QD lasers grown by molecular beam expitaxy (MBE) have been shown to possess impressive characteristics such as low threshold current[9] and high characteristic temperature,[10] QDs grown by metal-organic chemical vapour deposition (MOCVD) progress slowly.

In order to fully utilize the superior electronic and optical properties of QDs, efforts have been made to develop methods of position control of QDs, involving substrate lithography prior to growth, and/or selective area growth techniques,[11,12] which usually introduce defects into the QD structures. A simpler approach to achieving lateral ordered InAs/GaAs QDs has been realized by using GaAs (100) vicinal substrates.[13,14] In this case, the self-aligned QDs are formed along the multiatomic step edges on the GaAs (100) vicinal surface. Up to now, a few studies about the optical properties of QDs on misoriented substrates have been reported. It is shown that the misorientation of the substrates leads to a decrease in the FWHM of PL lines and a blue-shift of the emission wavelength.[15]

In this work, InAs quantum dots are grown on vicinal GaAs (100) substrates by using metal-organic chemical vapour deposition. An abnormal temperature dependence of bimodal size distribution of InAs quantum dots is found. The optical properties of QDs on vicinal substrates are studied by PL and also compared with those of dots on exact substrates.

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2. Experimental details

The samples are grown on vicinal GaAs (100) substrates (2 degrees tilted toward (110)) by using low pressure MOCVD, in which the trimethylgallium (TMGa), trimethylindium (TMIn), arsine (AsH_3) are used as source materials. Prior to the growth, the substrates are preannealed at 700 ^{0}C for 3 min in arsine flow for deoxidation. Then a 200nm GaAs buffer layer is grown at 600°C at a growth rate (G_r) of 1 ML/s and V–III ratio (mass flow ratio between precursors of group V and group III) of 25. Under this growth condition, multiatomic steps are formed on the vicinal substrates.[16] The QDs are grown subsequently at an InAs coverage of 1.7 ML, an InAs deposition rate of 0.034 ML/s and a growth temperature ranging from 480° C to 535° C. After the QD formation, a 20s growth interruption (GI) is introduced with the arsine flowing, and then the QDs are either cooled down to room temperature rapidly or overgrown with a final 30nm GaAs cap layer that is grown at 507° C at G_r=1 ML/s. In some cases, dots on GaAs(100) exact substrates are also grown with the two kinds of substrates placed side by side in the same growth chamber.

A Nanoscope Dimension 3100 SPM system with tapping mode in air and An XL30-FEG microscope are used to investigate the QD morphology and to correlate the evolution of size distribution with growth temperature. The PL measurements are carried out in closed-cycle He cryostat under the excitation of 514.5 nm line of Ar^+ laser focused onto a 0.5 mm^2 spot. The luminescence spectra are detected with a Fourier transform infrared spectrometer operating with an InGaAs photodetector.

3. Results and discussion

Figures 1(a) and 1(c) show the SEM images of surface InAs QDs on misoriented (100) GaAs and exact (100) GaAs grown in the same growth run at 507° C, at a G_r of 0.034 ML/s, InAs coverage of 1.7 ML, and V–III ratio of 5, respectively. The corresponding size histograms are shown in Figs.1(b) and 1(d), respectively. As can be seen from Fig.1, while dots on an exact substrate show only one dominant size, QDs on misoriented GaAs are formed in lines and show a clear bimodal size distribution with two groups of large and small dots. The densities of the large and small QDs

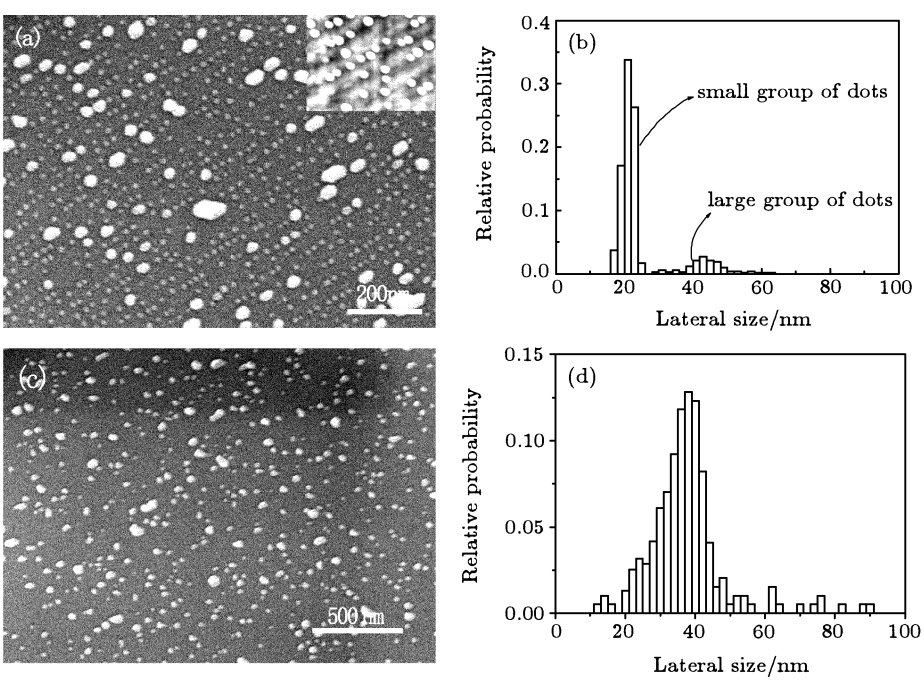


Fig.1. SEM images of QDs on the misoriented (100) GaAs(a) and exact (100) GaAs substrates (c), with corresponding size histogram shown in (b) and (d), respectively. The dots are formed at 507°C by 1.7 ML of InAs and G_r=0.034 ML/s and V–III ratio =5.

are $0.91\times10^{10}\,\mathrm{cm}^{-2}$ and $4.05\times10^{10}\,\mathrm{cm}^{-2}$, respectively, with a total of $4.96\times10^{10}\,\mathrm{cm}^{-2}$, greater than that of dots on the exact substrate, which is $1.21\times10^{10}\,\mathrm{cm}^{-2}$. The formation of dots in lines is due to QDs nucleating preferably along the multiatomic step edges on misoriented substrate,[13] as shown clearly in the inset of Fig.1(a). The higher QD density results from smaller critical nuclei for stable island growth when dots nucleate on the steps.[14] The bimodal size distribution of QDs on the misoriented surface can be related to the reduced surface diffusion during the MOCVD growth. The small QDs are not in thermal equilibrium and will subsequently develop into the energetically favourable large QDs. However, on the misoriented substrates, migration of adatoms on the surface is prohibited due to the energy barrier formed at the step kinks,[17] which slow down the transformation of the small QDs into the energetically favourable large dots and in such a case a bimodal size distribution appears.

Figure 2 shows the SEM images of surface QDs on vicinal GaAs (100) substrates at different temperatures at a G_{r} of 0.034 ML/s, InAs coverage of 1.7 ML, and V–III ratio of 5. Figures 2(a), 2(b), 2(c) and 2(d) correspond to growth temperatures of 480, 500, 520, and 535°C, respectively. The resulting QD densities of the large and the small groups of dots are plotted as a function of temperature in Fig.3(a). We can see that by increasing the growth temperature, the density of the large dots decreases, while the density of the small dots first grows larger when temperature is below 520°C, and then suddenly decreases at 535°C. The trends are in contrast to those in early reports. Since the surface diffusion length shows a strong temperature dependence, it has been shown that, at a low temperature, high-density small QDs are formed due to shorter diffusion length, while at high temperatures, a greater migration distance leads to a lower QD density of larger QDs.[2] Porsche *et al* used temperature to control the bimodal size distribution of InP/InGaP QDs on the exact (001) plane and found that by increasing the temperature, the number of small dots reduced and the quantity of large dots increased due to the enhanced surface diffusion.[7] Our seemingly surprising results can be explained by taking into account the presence of the multiatomic steps on the substrates. At low temperatures (below 520°C), the energy barrier at the multiatomic kinks can limit the migration of adatoms within two adjacent steps. Since the nucleation on steps is energetically favourable, multiatomic steps can bring on the formation of smaller critical nuclei for island growth,[14] resulting in the formation of islands with small average diameters and high densities, as observed in our QD growth. As the temperature increases, the surface adatom mobility is enhanced, which offers the adatoms more chances to reach the multiatomic steps and nucleate there on a large number of nucleation sites to form more small dots. In a given InAs deposition thickness, the fast increase in number of small dots consumes the deposited InAs rapidly, leading to a decrease in number of the large dots as shown in Fig.3, which would otherwise also increase with temperature. However, at 535°C, the mobility of the adatoms is increased so that they can migrate across the barrier at the steps easily and the effect of step edges is neglectable. QDs nucleate and grow in a mode similar to that on exact substrates, which leads to a decrease in density of small dots and increase in size of the large dots. Moreover, the In evaporation can also cause the number of QDs to decrease at high temperatures.[2] The change of lateral dot size with temperature is shown in Fig 3(b). Though the resolution (1.5 nm) of the FEG SEM is relatively low compared with that of AFM or TEM, we can measure the average sizes of our dots in an enough accuracy, which are typically over 20 nm and 40 nm for the small and large groups of dots, respectively. As can be seen, the size of the small group of dots increases slightly with the increase of growth temperature, while the size of the large dots decreases before 520°C, which can be attributed to the consumption of InAs by the rapid increasing of small dots as discussed above.

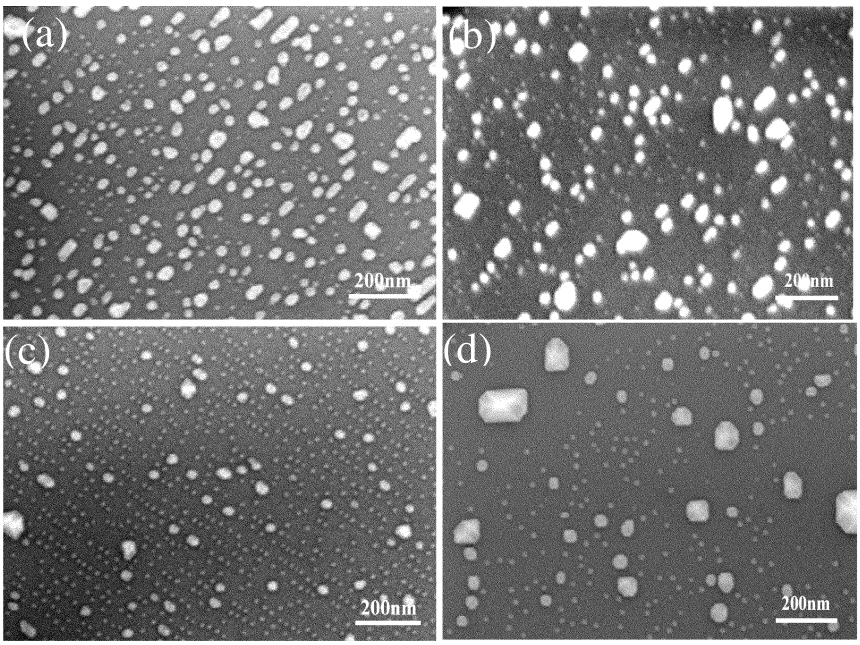


Fig.2. SEM images showing the effect of growth temperature. (a) T=480 °C, (b) T=500°C, (c) T=520°C, (d) T=535°C. All samples are formed at 1.7 ML of InAs and G_r=0.034 ML/s and V–III ratio =5.

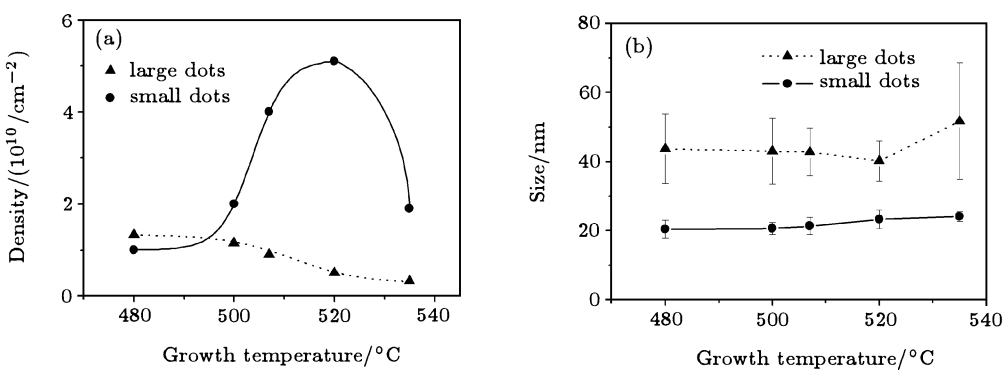


Fig.3. (a) Dot density as a function of growth temperature; (b) The change of dot size with temperature.

Next, taking the dots shown in Fig.1 for example, we study the effect of substrate misorientation on the optical properties of both the surface and buried QDs. To measure the PL of the surface QDs is difficult because of the surface nonradiative recombination, which decreases the PL intensity by 3–4 orders of magnitude as compared with the capped dots.[18] While the latter can be observed at room temperature easily, the PL of surface QDs is only observed at low temperatures. Figure 4(a) shows the 77 K PL of surface QDs on vicinal substrates, which can be decomposed into two Gaussian peaks. Since the PL spectra show that they are independent of excitation power, 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 irregularly shaped large dots which may be dislocated. Figure 4(b) shows the 77 K PL spectra of surface QDs on exact substrates. As can be seen, the spectrum can be decomposed into at least four Gaussian peaks. While the peak at about 1.29eV can be attributed to the emission of wetting layer, the other three are attributed to groups of dots with different sizes, reflecting the wide size distribution of dots on exact substrates.

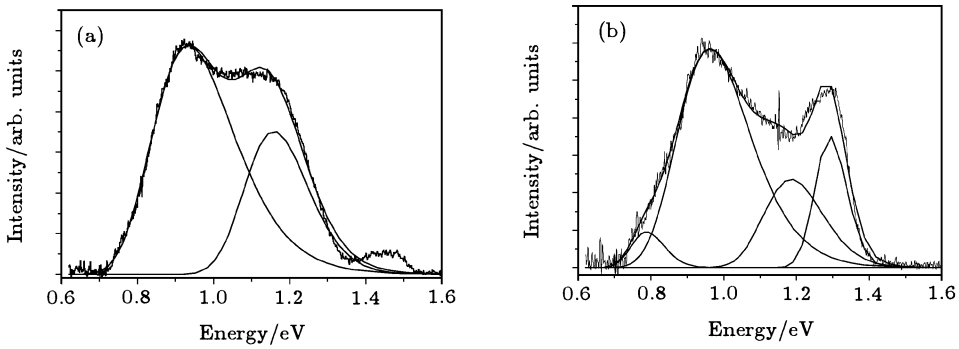


Fig.4. 77K PL spectra of surface QDs shown in Fig.1. Fig.4(a) is for dots on vicinal substrate and Fig.4(b) is for dots on exact substrates.

Contrary to QDs with cap layers that usually show state filling effect in PL measurements, no emission from excited states is observed from our surface dots. Two possible reasons are explained below. First, as discussed in Ref.[2], there may exist only one bound state for quantum dots with no cap layers. Then, the strong nonradiative recombination at the surfaces of QDs without cap layers consumes a large number of carriers.[18] So a very large excitation power (out of the range of our laser source) is needed in order to be able to observe the state filling effect.

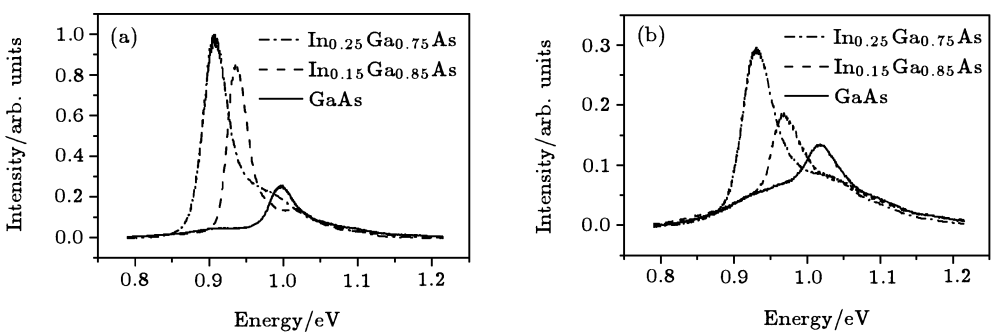


Fig.5. Room temperature PL spectra of InAs QDs shown in Fig.1 with or without InGaAs SRL. Fig.5(a) is for dots on misoriented substrates and Fig.5(b) is for dots on exact substrates. All the data are normalized to the PL intensity of dots on a misoriented substrate with $In_{0.25}Ga_{0.75}As$ SRL.

Figure 5 shows the room temperature PL spectra of samples with 2 nm InGaAs strain reducing layers of different In composition sandwiched between the dots and the GaAs cap layer. The PL results are summarized in Table 1. As can be seen, for dots on vicinal substrates, the PL spectra of capped QDs show only one dominant emission peak though the InAs dots exhibit a bimodal size distribution. The emission peaks of samples with vicinal substrates can be attributed to the large group of dots. The In/Ga intermixing during the cap layer growth will lead to a shallower confining potential and a wider band gap of QDs. Since the small dots have a higher ratio of interface-area to volume than the larger dots, intermixing will have a stronger effect on the smaller dots. This makes it easier for the carriers to be thermally excited out of the small dots and recombined in the large dots. Note that though the large group of dots on misoriented substrates has a small density compared with dots on the exact GaAs, their PL spectra have a much larger intensity. This can also be attributed to the presence of multiatomic steps on the misoriented GaAs substrates. The strong binding effect of the step edges can limit the possible sizes of QDs.[19] Thus the number of very large dots, in which dislocations may exist and act as nonradiative centres, can be reduced. As can be seen from both Fig.1(c) and Fig.1(d), there are a number of very large dots (with lateral size being larger than 50 nm) on the exact substrates. On the contrary, only a few very large dots are formed on misoriented substrates, as shown in Figs.1(a) and 1(b). Thus the number of nonradiative centres is smaller for dots on

misoriented substrates, leading to an increase in radiative recombination. As shown in Table 1, similar to the results in Ref.[15], PL spectra on misoriented substrates have a narrower FWHM. In contrast to the results in Ref.[15] which show a blue shift of PL maximum, however, our dots on misoriented substrates can produce emission at long wavelength compared with dots on exact substrates. The bimodal size distribution of our dots on misoriented substrates makes it possible that one group of dots is a little bit larger than dots on exact substrates. From the PL spectra in Fig.5, we cannot observe the luminescence from InGaAs layers. This indicates that the carriers generated in InGaAs layer can be efficiently injected into InAs quantum dots.[20]

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

	cap layer	GaAs	$In_{0.15}Ga_{0.85}As$	$In_{0.25}Ga_{0.75}As$
normalized PL intensity	exact	0.14	0.19	0.30
	misoriented	0.26	0.85	1
FWHM/nm	exact	123	82	81
	misoriented	59	46	63

4. Conclusions

Due to the presence of multiatomic steps on the GaAs surfaces, the InAs QD bimodal size distribution demonstrates an abnormal dependence on temperature. With other parameters fixed the density of small dots increases while the density of the large dots decreases as the growth temperature increases. The optical properties of InAs QDs on the GaAs (100) vicinal surface are also studied. It is found that dots on the vicinal substrate have a longer wavelength emission, a narrower PL line width and a much larger PL intensity.

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