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High-density and narrow size-distribution InAs quantum dots formed by a modified two-step growth*

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We develop a modified two-step method of growing high-density and narrow size-distribution InAs/GaAs quantum dots (QDs) by molecular beam epitaxy. In the first step, high-density small InAs QDs are formed by optimizing the continuous deposition amount. In the second step, deposition is carried out with a long growth interruption for every 0.1 InAs monolayer. Atomic force microscope images show that the high-density ($\sim 5.9\times10^{10}\,\mathrm{cm}^{-2}$) good size-uniformity InAs QDs are achieved. The strong intensity and narrow linewidth (27.7 meV) of the photoluminescence spectrum show that the QDs grown in this two-step method have a good optical quality.

Keywords: molecular beam epitaxy, quantum dots, a modified two-step growth
PACC: 7855E, 8115, 7280E

1. Introduction

In recent years, self-assembled InAs/GaAs quantum dots (QDs) grown in the Stranski–Krastanov (SK) mode have aroused a great interest because of their promising applications in long wavelength GaAs-based semiconductor QD lasers.[1−3] For laser applications, InAs QDs must fulfill several requirements, including size uniformity and high surface density. The size inhomogeneity of QDs is a serious drawback for laser applications, for it leads to the great increasing of threshold currents.[4,5] A relatively low density of QDs restricts the ground state saturated gain and maximum power output of a QD laser.[6,7] This problem cannot be solved simply by stacking multilayer QDs, because it can inevitably introduce many InAs/GaAs interfaces and thus increase the number of non-radiative recombination centres.[8] The optimization of a uniform, high-density InAs QD layer is essential for achieving a high-power laser with low threshold current density, room temperature continuous-wave operation, and high characteristic temperature.[9,10]

The typical density of InAs QDs is $2 \sim 4 \times 10^{10}\,\mathrm{cm}^{-2}$ on GaAs in the SK growth mode.[11,12] InAs QDs with densities up to $\sim 1 \times 10^{11}\,\mathrm{cm}^{-2}$ have been fabricated using a continuous high growth rate.[13] However, these QDs have a large size-distribution and poor optical quality, leading to a broad linewidth and low intensity of the photoluminescence (PL) spectrum. Some research groups have obtained InAs QDs with a high density ($\sim 4 \times 10^{11}\,\mathrm{cm}^{-2}$) on AlAs,[14,15] implying that it is possible to use the higher surface roughness of AlAs, which results in a short diffusion length on AlAs compared with that on GaAs. However, the aluminium intermixing towards QDs can cause non-radiative defects, resulting in the deterioration of the optical properties of InAs QDs.[16]

Some methods have been introduced to fabricate InAs QDs with a narrow size-distribution, such as the low growth rate and the growth interruption methods.[11,17] Although the size uniformity of the QDs is improved with these methods, the density is decreased.

Since continuous growth helps to increase QD density and interrupted growth improves the uniformity of QDs, several groups have recently developed a two-step growth method of growing self-assembled InAs QDs by combining the advantages of contin-

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uous and interrupted growths.[12,18] Although these authors claim that the dot density and size uniformity of QDs formed by using a two-step growth method are improved, the density of QDs is still low ($\sim 2.5 \times 10^{10}\,\mathrm{cm}^{-2}$). We modify the two-step growth method by optimizing the continuous deposition amount while fixing the total deposition amount of InAs. Atomic force microscope (AFM) images show that high-density ($\sim 5.9 \times 10^{10}\,\mathrm{cm}^{-2}$) good size-uniformity InAs QDs prepared by using our modified two-step growth method are achieved. An improvement on the PL intensity and the narrow linewidth of the PL spectrum indicate that this modified two-step growth method can greatly enhance the optical quality of the QDs. These results could be applied to the fabrication of QD lasers on GaAs substrate with low threshold current density, high characteristic temperature and high output power.

2. Experiment

The samples were grown on (100) GaAs by using a molecular beam epitaxy (MBE) system (Varian Gen-II) with an As cracker cell. The growth rates were 1.0ML/s and 0.1ML/s for GaAs and InAs, respectively. A 500nm GaAs buffer layer was first grown at 615°C after oxide desorption. Then the temperature was ramped down to 515°C. Three series of InAs QD samples were grown in the conventional SK growth mode, the repeated growth mode, and by our two-step growth procedure, separately. In the conventional SK growth mode, InAs was continuously deposited to form QDs. In the repeated growth mode, a repeated growth sequence of 0.1 ML InAs was implemented, and then a short growth interruption time (GIT) deposition under a continuous flux of As_2 followed. In the two-step growth process, the first step was that about 2.0 monolayers (ML) of InAs were continuously deposited to form a Gaussian-size-distribution of small coherently strained islands with high density;[19] the second step 2 was that the rest of the InAs was deposited with a 10 s interruption every 0.1 ML to improve the uniformity of the InAs QDs by keeping the total amount of InAs fixed at 2.5 ML. For the PL measurement, a 10 nm GaAs cap layer was grown on the QDs immediately after the post-growth interruption of the InAs QD growth, and then the temperature was raised up to 615°C to grow another 200 nm GaAs cap layer. The measurements of the QDs were performed ex situ by AFM (Nanoscope IIIa) in a contact mode. The PL spectra were detected with a Fourier transform infrared spectrometer operating with a Ge detector.

3. Results and discussion

Figures 1(a)–1(c) show AFM images of InAs QDs on a GaAs substrate grown in the conventional SK growth mode and in a repeated growth sequence of 0.1 ML InAs followed by deposition for different GITs, separately. The total amount of InAs deposited is the same (2.5 ML) in these samples. The dots have a density as high as $1.13 \times 10^{11}\,\mathrm{cm}^{-2}$ when grown continuously (as shown in Fig.1(a)), while the dot density decreases nearly four-fold (only $2.86 \times 10^{10}\ \mathrm{cm}^{-2}$) when the GIT every 0.1 ML is extended to 5 s as shown in Fig.1(c). Figure 1(d) shows the dependence of QD density on GIT. The QD density decreases with the increase of the GIT, due to the enhancement of the diffusion length of the In adatoms during the growth interruption. The average dot diameter and the average height of the QDs both increase when the GIT every 0.1 ML extends from 0 to 5 s as shown in the inset of Fig.1(d). At the same time, the dot size uniformity is improved from the statistical results of AFM images when the GIT extends.

Figure 2 shows the PL spectra of the QDs grown in the conventional SK growth mode and repeated growth mode. The broad PL full width at half maximum (FWHM) at 1.21μm is 66.5 meV, corresponding to that for the sample without growth interruption. The narrow peak (FWHM: 28.6 meV) located at 1.26 μm corresponds to that for the sample with a 5 s GIT every 0.1 ML. It is clear from the narrowing of the PL linewidth that the homogeneity of the QDs is improved as the GIT extends. The peak position is red-shifted, due to the increase of the average size of QDs with the extension of GIT. An improvement on PL intensity with the extension of GIT indicates that the optical quality of the QDs is enhanced greatly, which is perhaps due to the reduction in the misfit dislocations and arsenic antisite related crystal defects in QDs.[17]

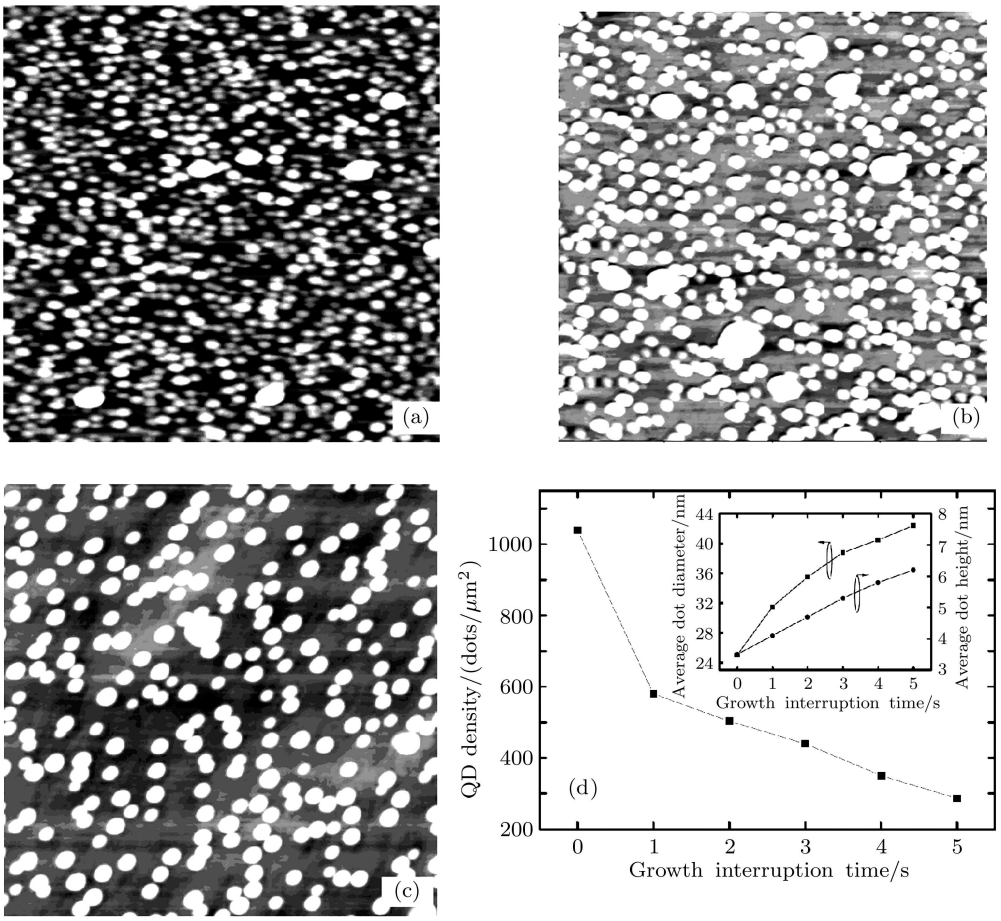


Fig.1. AFM images (1μm×1μm) of InAs QDs grown in the conventional SK mode and repeated growth mode (a–c) and QD density and average size versus GIT (d). The total amount of deposited InAs is 2.5 ML in these samples. (a) Continuous deposition; (b) GIT: 1 s; (c) GIT: 5 s.

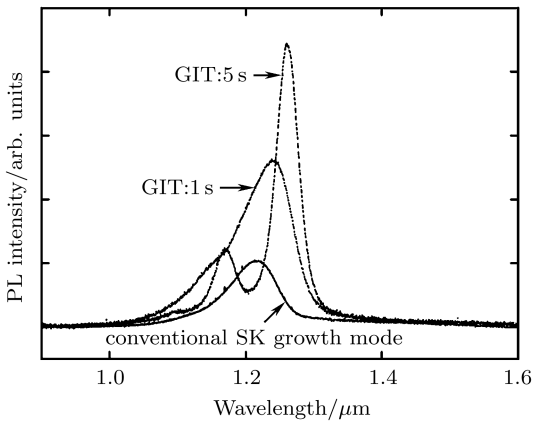


Fig.2. The PL of InAs QDs grown in the conventional SK growth mode and repeated growth mode for different GITs. The deposition amount of InAs is 2.5 ML.

Figures 3(a)–3(c) show the AFM images of the InAs QDs grown by using the modified two-step growth method, with the continuous deposition amount varied but the total deposition amount fixed at 2.5 ML. While the density of QDs is only $2.5 \times 10^{10}\,\mathrm{cm}^{-2}$ when the continuous deposition amount is 1.9 ML (shown in Fig.3(a)), high density InAs QDs (over $5.9 \times 10^{10}\,\mathrm{cm}^{-2}$) is successfully fabricated when the continuous deposition amount is at least 2.0 ML. Figure 3(d) shows the dependence of density of QDs on continuous deposition amount. The density of QDs increases with the increase of continuous deposition amount.

Figure 4 shows the room temperature PL spectra of the InAs QDs grown by using our two-step growth method. The narrow PL linewidth (27.7 meV) indicates that the uniformity of the InAs QDs prepared by using the two-step growth method is improved compared with that in the conventional SK growth mode. Tang *et al*[18] suggested that atoms should be precluded from reaching the large QDs because of the strain energy barrier on the edge of the QDs, so the larger dots grew more slowly than the smaller dots. The smaller QDs might grow faster due to the growth interruption. Therefore, the dot size and the dot size uniformity are improved in the two-step growth mode.

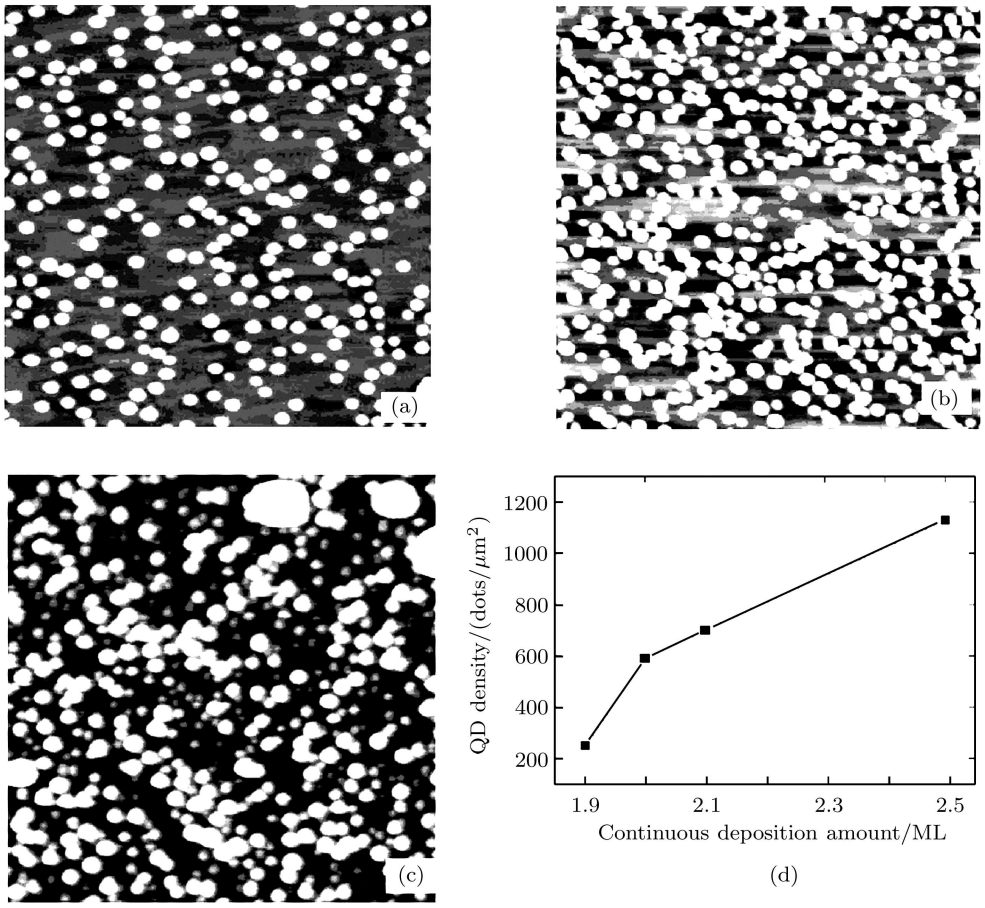


Fig.3. AFM images (1μ m$\times 1\mu$m) of InAs QDs by using the modified two-step growth method (a–c) and QD density versus continuous deposition amount (d). The total deposition amount is the same (2.5 ML). (a) 1.9 ML (Step 1) +0.6 ML (Step 2), (b) 2.0 ML (Step 1) +0.5 ML (Step 2), (c) 2.1 ML (Step 1) +0.4 ML (Step 2).

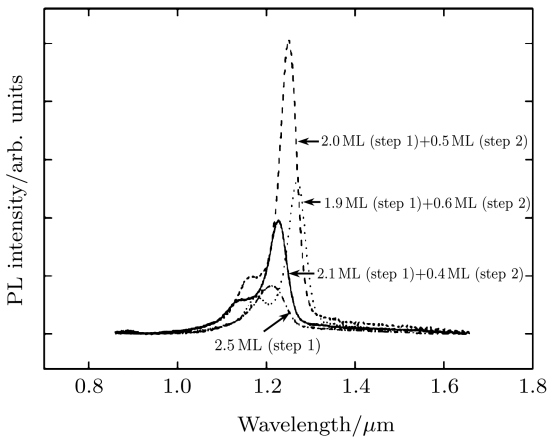


Fig.4. Room temperature PL of two-step growing InAs QDs. The total deposition amount is kept the same (2.5 ML).

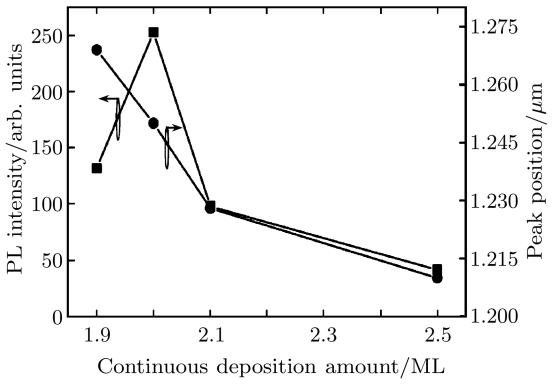


Fig.5. Dependences of density of QDs and PL intensity on continuous deposition amount.

Figure 5 shows the dependences of QD PL intensity and peak position on continuous deposition amount. It is clear that the largest intensity and narrow PL linewidth (only 27.7 meV) are obtained from the sample with the deposition of 2.0 ML InAs in step 1 and 0.5 ML InAs in step 2. The PL intensity of the sample with 2.0 ML of InAs grown in step 1 and 0.5 ML of InAs in step 2 is almost twice that of the sample with 1.9ML of InAs grown in step 1 and 0.6 ML

of InAs in step 2, due to their different QD densities as shown in Fig.5. Although the QD density is over $7.0 \times 10^{10}\,\mathrm{cm}^{-2}$ when the continuous deposition is 2.1 ML, the PL intensity is greatly decreased. Obviously the decrease of PL intensity does not originate from the QD density but it is due to the occurrence of dislocations, which greatly affects the optical quality of the QDs. Previous experimental results[20,21] indicate that dislocations can occur when the continuous deposition amount well exceeds 2.0 ML, which is not in full agreement with our measurements. The peak position is red-shifted with the decrease of the continuous deposition amount, which we attribute to the increase of average size of QDs.

4. Conclusions

We have systematically studied the effect of growth interruption on the structural and optical properties of InAs/GaAs QDs grown by MBE. A modified two-step method of growing InAs QDs is developed. InAs is first continuously deposited to form high-density, small-size QDs in step 1, followed by deposition with a 10 s GIT every 0.1 ML InAs. AFM images indicate that high-density ($\sim 5.9 \times 10^{10}\,\mathrm{cm}^{-2}$) good size-uniformity InAs QDs are achieved. The narrowing of the PL linewidth (27.7 meV) implies that the QDs have much better uniformity. An improvement on PL intensity shows that the structural and optical qualities of the QDs are enhanced greatly.

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