

Optical properties and structure of InAs quantum dots in near-infrared band

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Abstract The InAs quantum dots (QDs) grown by molecular beam epitaxy (MBE) are studied as a function of growth temperature at a specific InAs coverage of 2.7 ML. The QDs density is significantly reduced from 8.0×10^{10} to $5.0 \times 10^9 \text{ cm}^{-2}$ as the growth temperature increases from 480°C to 520°C, while the average QDs diameter and height becomes larger. The effects of the growth temperature on the evolution of bimodal QDs are investigated by combining atomic force microscopy (AFM) and photoluminescence (PL). Results show that the formation of the bimodal QDs depends on the growth temperature: at a growth temperature of 480°C, large QDs result from the small QDs coalition; at a growth temperature of 535°C, the indium desorption and InAs segregation result in the formation of small QDs.

Keywords quantum dot (QD), molecular beam epitaxy (MBE), atomic force microscopy (AFM), photoluminescence (PL)

1 Introduction

Self-organized semiconductor structures with reduced dimensionality are currently attracting strong interest due to their potential device advantages. The size, distribution and density of quantum dots (QDs) can influence the optical and electrical properties of the QDs device, so it is important to understand the effects of growth parameters and formation mechanisms on the growth of bimodal size distribution QDs [1–5].

QDs with bimodal size distribution have been widely investigated due to their distinctive features in comparison with single modal distribution QDs. There are two main

photoluminescence (PL) peaks in PL spectra of the bimodal distribution QDs. Although the phenomena of the carrier escape from QDs, carrier loss in QDs and carrier capture have been studied based on the variable temperature PL, the effects of the growth temperature on the formation and properties of QDs need to be further investigated.

In this paper, the self-organized bimodal InAs QDs prepared on GaAs(100) substrate are analyzed by combining atomic force microscopy (AFM) and PL. The effects of growth temperature on the size, distribution and density of QDs are investigated. The formation mechanism of bimodal QDs is analyzed and the effects of vertical and transversal confinement on optical properties of QDs are also discussed.

2 Experiments

The InAs QDs were grown on semi-insulating GaAs(100) wafers in a Riber 32 compact 21 solid source molecular beam epitaxy (MBE) system equipped with a valved cracker cell for arsenic and reflection high-energy electron diffraction (RHEED) for in situ monitoring of the growth process. The growth temperature was calibrated by infrared pyrometer. After desorbing the surface oxide at 580°C, 100 nm GaAs buffer layer was grown and deposited, and a single layer of quantum dots was grown by depositing the 2.7 ML thickness InAs at different growth temperatures (520°C and 480°C). The 15 nm GaAs cap layer was then deposited. The RHEED pattern shows a transformation of a typical three-dimensional (3D) growth mode from streaky to well-developed diffraction spots. The same structures without cap layer were grown for topographic investigation with atomic force microscopy (AFM). Measurement of the distribution and topographic images of the uncapped dots were performed ex situ with Nanoscope III AFM, and the PL measurements were performed in closed-cycle N₂

cryostat under the excitation of 514.5 nm line of an Ar⁺ laser. Its luminescence spectra were detected with a Fourier transform infrared spectrometer operating with an InGaAs photo detector.

3 Results and discussion

Figure 1 shows the analysis results of the QDs morphology by AFM. It can be observed that the height of the

QDs is bimodal distribution in samples A₁ and A₂. The height distribution of QDs in sample A₁ is mainly concentrated at near 3.4 and 5.3 nm, and in sample A₂ at near 3.9 and 5.4 nm. The size of large QDs in sample A₁ is similar to that in sample A₂, while the size of small QDs in sample A₂ is a little larger than that in sample A₁. For diameter distribution, the diameters of QDs increase slightly with an increase of growth temperature. According to the growth dynamics, the surface diffusion length of the atom can be expressed as [6]

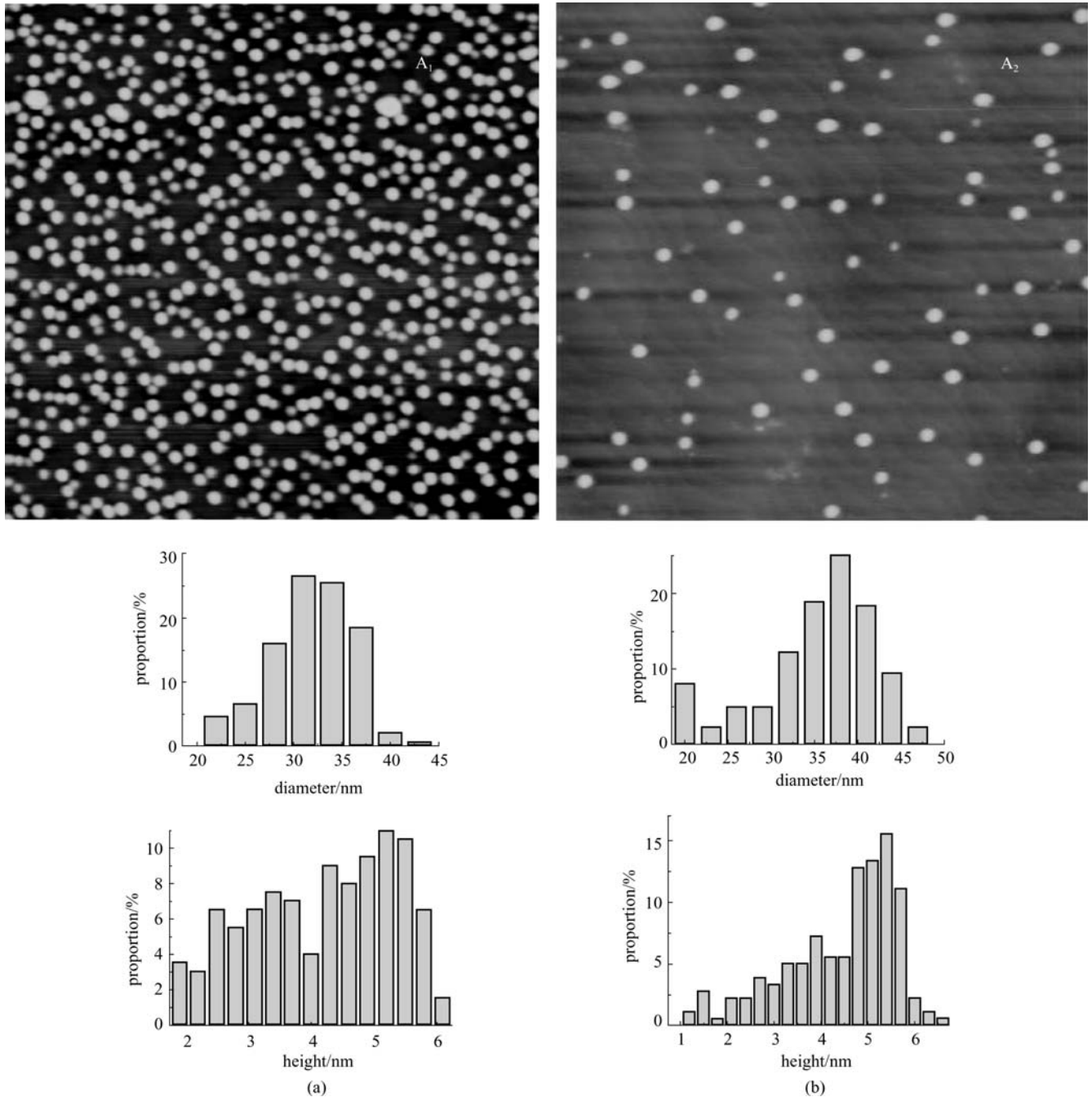


Fig. 1 AFM images (1 $\mu\text{m} \times 1 \mu\text{m}$) and size distribution of InAs QDs grown at different growth temperatures of 480°C (A₁) and 520°C (A₂)

$$\lambda = \sqrt{\tau D}, \quad (1)$$

where τ is the atom effective diffusion time, and D is the diffusion constant, defined as [7,8]

$$D = D_0 \exp[-E/(kT)], \quad (2)$$

where T is growth temperature, E is the barrier energy of atom mobility, and D_0 is a constant. Equations (1) and (2) indicate that the diffusion length increases as the diffusion constant D increases, and the diffusion length decreases as the growth temperature is lowered. An increase in the atom diffusion length, the nucleation sites and the density of QDs reduces, and as a result of this the size of the QDs will increase.

According to the self-confinement effects of QD, an optimized value at a certain growth condition exists in strain-induced self-organized growth of QDs [9,10]. As indicated in the further analysis of graph A₁ in Fig. 1, three types of QDs, displayed in Fig. 2, are investigated. It can be noted that the combination of the nearest neighbor QDs results in either the formation of large-sized QDs or the coalition of QDs. The lower the growth temperature is, the shorter the atom diffusion length, and the larger the nucleation probability will be. Therefore, when the density of QDs becomes larger, the combination of QDs occurs more easily. The atom diffusion length is small at 480°C growth temperature, which means some InAs material cannot transform into QDs with the completely released strain energy due to the short diffusion time, which results in the formation of the bimodal size distribution QDs [11].

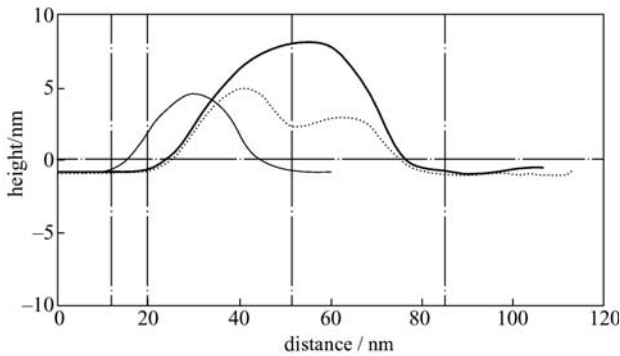


Fig. 2 AFM images of single QD profile grown at the temperature of 480°C

Based on the above formula, the density and nucleation probability of QDs decrease when the atom diffusion length increases. Thus, it is unlikely that combinations of QDs will take place. Although the same bimodal distribution of QDs is obtained at growth temperatures of 480°C and 520°C, the formation mechanism is obviously different. In segregation and InAs desorption have a great impact on the growth process of QDs at 520°C growth temperature. The

segregation length of the In atom can be expressed as

$$\lambda = -\frac{d}{\ln R}, \quad (3)$$

where d is the lattice constant, and R is the segregation rate. The relationship between the growth temperature and segregation length can be expressed as [12,13]

$$R = \frac{1}{\exp[E_s/(kT)] + 1}, \quad (4)$$

where E_s is segregation energy, and the desorption rate can be expressed as

$$M \propto N \exp\left(-\frac{E_{\text{des}}}{kT}\right), \quad (5)$$

where N is surface density of atom, E_{des} is desorption energy, and T is InAs QDs growth temperature.

Equations (3) and (4) indicate that the segregation rate and segregation length increase when the growth temperature increases. The concentration profiles of the In atom in QDs will be a gradient distribution, and increase gradually. The strain energy between the segregation InAs and GaAs substrate is smaller than that between stoichiometric materials, which will result in an increase in the size of QDs, and the accumulation of more In atom on the surface of QDs. Equation (5) shows that with an increase in growth temperature, the higher the surface density of In atom is, the more serious the In segregation and desorption is, which contributes to the formation of bimodal distribution [13], and even multimodal distribution of QDs. This is mainly due to the fact that the process occurring in the interval between when the In atom is desorbed from QDs and the In atom is adsorbed by QDs is dynamic. The high-quality QDs need to be prepared at a proper growth temperature, which can prevent many In atoms from being desorbed. Only in this way can QDs with better order and consistency be obtained.

Figure 3 shows the PL spectra of samples A₁ and A₂ at 77 K temperature. The emission wavelength of QDs PL depends on the size of QDs. The PL spectra peak of sample A₁ locates at 1.099 and 1.197 μm , and the full width at half maximum (FWHM) is 0.046 and 0.117 eV, respectively. The low energy PL spectra peak corresponds to the ground state transition of large QDs; the high energy band edge to the wetting layer PL spectra; and 1.676 eV to GaAs bulk material PL spectra. The high energy PL spectra peak possibly includes the first excitation state of large QDs and the ground state transition of small QDs. The PL spectra intensity of QDs indicates the high proportion of large QDs in bimodal QDs distribution. The low energy peak value in sample A₂ is almost the same as that in sample A₁. As for sample A₂, the high energy peak has a redshift and PL has a narrow linewidth. The above analysis shows that the high

growth temperature causes small QDs to increase in size and keeps the large-sized QDs unchanged. From the narrow FWHM in sample A₂, it can be noticed that there is also an improvement in the consistency of QDs. The PL results correspond well with the AFM experimental data. The relationship between the confinement energy and size of QDs can be expressed as [14]

$$E \propto L^{-2}. \quad (6)$$

The PL spectra with bimodal distribution correspond to the vertical size distribution of QDs, which proves that vertical confinement plays a major role in the PL spectra of QDs.

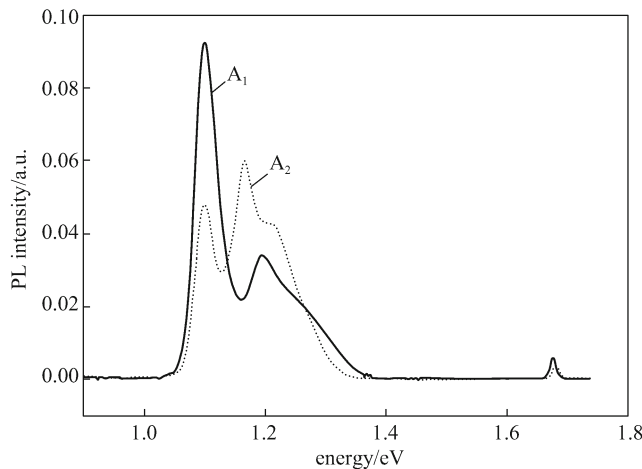


Fig. 3 PL spectra of sample A₁ and A₂ at 77 K

4 Conclusions

The self-organized InAs quantum dots (QDs) grown on GaAs(100) substrate by MBE are studied at 480°C and 520°C growth temperature. The effects of growth temperature on the evolution of bimodal QDs are investigated by combining the AFM and PL. The density of the QDs reduces at 520°C growth temperature, while the size of the QDs becomes larger. The strain energy cannot be completely released at the low growth temperature since the diffusion time is too short. The combination of the nearest neighbor QDs results in either the formation of large-sized QDs or the coalition of QDs, which is the main reason for bimodal QDs formation at low growth temperature. Indium desorption and In segregation can result in the formation of bimodal, even multi-modal, distribution QDs at high growth temperature. The PL results are in accordance with the AFM experimental data for the samples prepared at different growth temperatures, and the crucial role of vertical confinement in QDs PL is also proved.

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