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2005 Nanotechnology 16 2775

(http://iopscience.iop.org/0957-4484/16/12/005)

INSTITUTE OF PHYSICS PUBLISHING NANOTECHNOLOGY
Nanotechnology **16** (2005) 2775–2778 doi:10.1088/0957-4484/16/12/005

Growth of low-density InAs/GaAs quantum dots on a substrate with an intentional temperature gradient by molecular beam epitaxy

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Received 30 July 2005, in final form 8 September 2005
Published 11 October 2005
Online at stacks.iop.org/Nano/16/2775

Abstract
InAs was deposited by molecular beam epitaxy (MBE) on a GaAs substrate with an intentional temperature gradient from centre to edge. Two-dimensional (2D) to three-dimensional (3D) morphology evolution was found along the direction in which the substrate temperature was decreasing. Quantum dots (QDs) with density as low as $\sim 8 \times 10^6$ cm^{-2} were formed in some regions. We attribute the morphological evolution to the temperature-dependent desorption of deposited indium and the intermixing between deposited indium and gallium from the buffer.

1. Introduction

Preparations, characterizations and device applications of self-assembled Stranski–Krastanow (SK) quantum dots (QDs) have been studied extensively in recent years because of their unique electronic and optical properties owing to the three-dimensional confinement of electrons and holes [1]. Among the popularly studied QD families, GaAs based In(Ga)As QDs are more attractive than the others, such as In(Ga)As QDs on InP and Ge QDs on Si. High sheet density is essential to most device applications, including laser diodes [2], superluminescent diodes [3], optical amplifiers [4], electro-optical modulators [5] and detectors [6]. Nevertheless, a very low density of QDs will be favourable in some cases. In the basic physical studies, one single QD on a few square microns or larger area is preferred so that information from one QD, rather than that from a QD set including many QDs with different dimensions, can be picked up. In some electronic applications, one tends to attain low-density QDs for facility of single-QD device fabrication [7]. In future optoelectronic device applications for quantum computing and quantum cryptography, single-photon emitters [8] fabricated by low-density QDs are needed. Usually, InAs QDs on GaAs substrate with sheet density in the order of 10^9–10^{11} cm^{-2} can be prepared easily by controlling the growth parameters including growth rate, substrate temperature, arsenic pressure, deposition thickness and growth interruption, by molecular beam epitaxy (MBE) or metal organic vapour phase epitaxy (MOVPE). Lower growth rate, higher substrate temperature, lower arsenic pressure, exact control of InAs deposition amount as well as introduce of growth interruption (either during or at the end of the InAs growth period) are preferred for the growth of low-density InAs/GaAs QDs [9–11]. However, decreasing the density of InAs/GaAs QDs to $\sim 1 \times 10^8$ cm^{-2} or less is rather difficult because the formation of SK self-assembled QDs is a first-order-phase-transition-like process [12]. QD number increases rapidly when the InAs deposition amount reaches the critical thickness, at which the two-dimensional (2D) to three-dimensional (3D) morphology transition happens. It is very difficult in the growth to terminate the supply of indium source at the exact time when the QD density is low enough. In fact, a lack (or excess) of indium supply always happens, which leads to flat 2D InAs layer (or InAs QDs with the density not low enough). So an alternative method, a mask on mid-density QD samples [13], has been used to limit the QD number to be studied. A few QDs in a window with submicron scale can be selected on a sample with the QD density of $\sim 1 \times 10^9$ cm^{-2} by this method. Nevertheless, efforts have been made to grow low-density InAs/GaAs QDs on substrates directly. Density as low as $\sim 1 \times 10^8$ cm^{-2}

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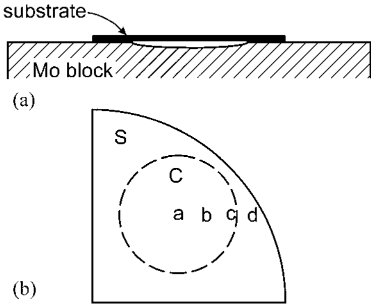


Figure 1. The diagrammatic presentation of the sample pasted on a Mo block that was concaved centrally. (a) Cross-section view, (b) plan view. Letters a, b, c and d indicate positions at which the AFM and PL are measured.

was achieved by means of growth interruption just before the critical thickness was reached [11].

In this paper, InAs was deposited on GaAs substrate with gradual-temperature distribution by MBE. Evolution from 2D to 3D morphology transition was found along the direction that the substrate temperature was decreasing. InAs QDs with the density of $\sim 8 \times 10^6$ cm^{-2} were obtained. To the best of our knowledge, this is the lowest density for InAs/GaAs QDs that has been reported.

2. Experiments

The sample was grown by a Riber 32P solid-source MBE machine on a quarter of a 2 inch semi-insulating (SI) GaAs substrate. It was pasted on a Mo block that was concaved centrally, as illustrated in figure 1(a). So gradual temperature distribution existed on the sample surface when the Mo block was heated during the growth. Reflection high-energy electron diffraction (RHEED) was used to check temperature on the substrate surface when the substrate was de-oxided. The temperature at the centre (point a in figure 1) was about 10 °C lower than that close to the edge (point d). First, a GaAs buffer layer, an AlAs/GaAs (3 nm/3 nm) short-period superlattice (SSL) and a 50 nm GaAs layer were grown. Then, the substrate temperature was lowered and 1.7 monolayer (ML) InAs was deposited nominally with a growth rate of 0.03 ML s^{-1}. 1 min growth interruption was processed before the 10 nm GaAs cap layer was deposited. After the temperature was raised, 40 nm GaAs, an AlAs/GaAs (3 nm/3 nm) SSL and 50 nm GaAs were grown. Finally, we lowered the substrate temperature again and grew another InAs layer at the surface with the same growth parameters as that of the inner one for morphology characterization. The substrate was kept on rotating at 50 r min^{-1} throughout the growth. The substrate temperature was about 530 °C at the centre of the sample and about 10 °C higher close to the edge for InAs deposition. So gradual temperature distribution was expected from the centre to the edge.

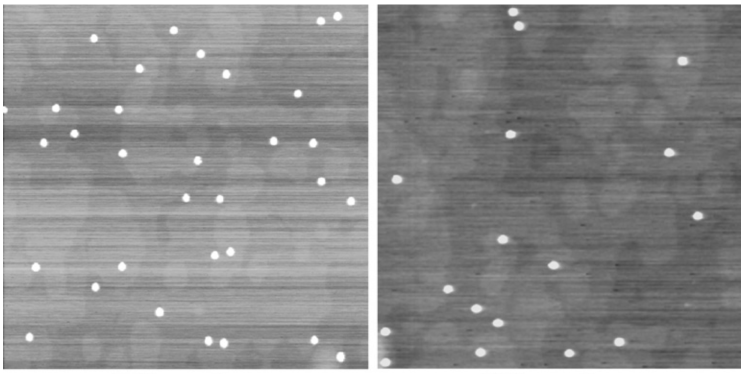
Atomic force microscopy (AFM) measurements were performed in atmosphere in the contact mode with a Solver P47 scanning probe microscope. Room-temperature photoluminescence (PL) spectra were obtained using a He–Ne laser for excitation, and recorded with a 0.58 m grating monochromator and InGaAs diode using standard lock-in techniques.

3. Results and discussion

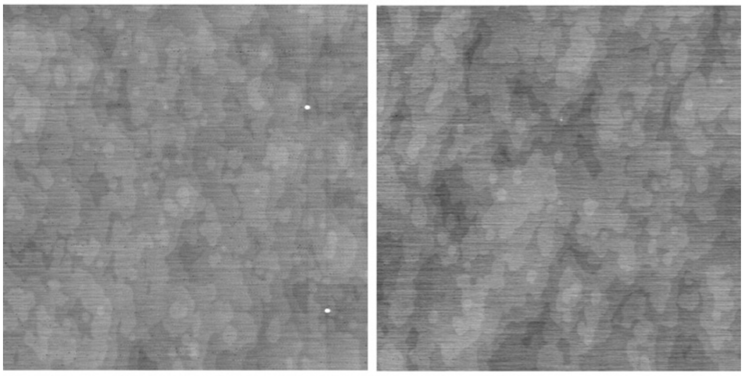
We define the substrate in two parts (see figure 1(b)), the central part C (the part of the substrate above the concavity that has no contact with the Mo block) and the surrounding part S (the part of the substrate pasted on the Mo block using indium). During the growth, the surrounding part S was heated by contact thermal conductance from the Mo block; while the central part was heated by radiation from the concavity of the Mo block and thermal conductance from the surrounding part S. Morphologic evolution is found on the sample from the centre to the edge and is shown in figure 2. InAs/GaAs QDs with the density of $\sim 8 \times 10^8$ cm^{-2} are formed at the centre of the C part (see figure 2(a)). However, near the edge 2D morphology is found (see figure 2(d)). Evolution from 2D flat growth to 3D island growth happens from the edge to the centre. Figure 2(b) shows InAs/GaAs QDs ($\sim 4 \times 10^8$ cm^{-2}) at point b (see figure 1(b)) that is between the centre (point a) and the boundary of the C part. Near the boundary we obtain QDs with the ultra-low density of $\sim 8 \times 10^6$ cm^{-2}, as presented in figure 2(c). To the best of our knowledge, this is the lowest density for InAs/GaAs QDs that have been reported.

PL spectra have been measured along line a–d. The spectra at points a and b are similar. Figure 3(a) shows the PL spectrum at point a. Up to the fourth excited-state transition can be found for QDs. By fitting the transition peaks with a Gaussian function, we get the transition energies for the ground-state transition and the four excited-state transitions. They are 1.022, 1.078, 1.133, 1.189 and 1.243 eV, respectively, showing equivalent energy spaces between these peaks. The strong PL peak below the GaAs bandgap comes from the wetting layer (WL). At point d (see figure 3(b)), only peaks from WL and GaAs are observed. The PL spectrum at c (not shown) is similar to the spectrum at d. At point c no PL signal from QDs is found because of the very low dot density.

Note that no other growth parameter is variable except for the substrate temperature. So we attribute the evolution from the 2D flat morphology to the 3D island morphology to the inhomogeneous substrate temperature. Substrate temperature is an important role that affects the growth and the consequent morphology of InAs on GaAs. It has been found that the critical coverage (or critical time) for the formation of QDs is strongly affected by temperature-dependent In–Ga intermixing [14] and desorption of deposited In [15]. Below 520 °C, the small increase of the critical time with the substrate temperature was attributed to the intermixing between the deposited In and Ga from the buffer [14]. Above 520 °C, fast increase in the critical time was dominated mainly by the desorption process [15]. In this work, the substrate temperature is higher than 530 °C. So it seems that the temperature-dependent In desorption should be mainly responsible for the morphological evolution from points a to d. However, the contribution of the In–Ga intermixing should not be ignored absolutely, though it is minor. In addition, comparison of substrate temperature between different works should be careful because temperature determination may be various in different experiments. So we attribute the morphological evolution with the temperature to



(a) 2 µm × 2 µm (b) 2 µm × 2 µm



(c) 5 µm × 5 µm (d) 5 µm × 5 µm

Figure 2. AFM images obtained at (a) point a, (b) point b, (c) point c and (d) point d.

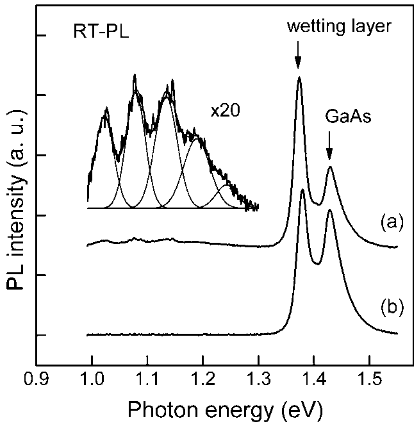


Figure 3. PL spectra recorded (a) at the centre of the sample (point a) and (b) close to the edge of the sample (point d). Inset is a magnification of spectrum (a) in the photon energy range of 1.0–1.3 eV and its Gaussian fit.

the combination of desorption of deposited In and intermixing between deposited In and Ga from buffer.

4. Conclusion

InAs was deposited on a GaAs substrate with inhomogeneous substrate temperature. The inhomogeneous temperature distribution was realized by pasting the substrate on a Mo block heater with a concavity on its surface. 2D to 3D morphology evolution was found along the direction in which the temperature decreased. In the low-temperature region, QDs with the density of $\sim 8 \times 10^8$ cm^{-2} were formed. In the high-temperature region, however, flat 2D morphology was found. QDs with very low density ($\sim 8 \times 10^6$ cm^{-2}) were obtained at some mid-temperature region. The morphological evolution is attributed to the temperature-dependent desorption of deposited In and intermixing between deposited In and Ga from buffer.

Acknowledgments

This work was supported by Special Funds for the Major State Basic Research Project of China (Nos G2000068303 and 2002CB311905), the National Natural Science Foundation of China (Nos 90201033, 60306010 and 90301007), and the National High Technology Research and Development Program of China (No 2002AA311070).

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