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2002 Nanotechnology 13 576

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Selective growth and photoluminescence studies of InAs self-organized quantum dot arrays on patterned GaAs(001) substrates

K W Sun^{1,4}, J C Wu², B C Lee³ and C P Lee³

¹ Department of Physics, National Dong Hwa University, No 1 Da Hseuh Rd, Sec. 2, Shoufeng, Hualien 974, Taiwan, Republic of China

² Department of Physics, National Changhua University of Education, Changhua, Taiwan, Republic of China

³ Department of Electronics Engineering and Institute of Electronics, National Chiao Tung University, Shin Chu, Taiwan, Republic of China

E-mail: kwsun@mail.ndhu.edu.tw

Received 19 February 2002, in final form 6 August 2002

Published 30 August 2002

Online at stacks.iop.org/Nano/13/576

Abstract

In this paper, we present experimental results on the selective growth of InAs self-organized quantum dots on patterned substrates using electron-beam lithography and molecular beam epitaxy. Higher dot densities were found on the patterned substrate with a particular pattern orientation compared to the densities of dots grown on the non-patterned area. Good quality of the quantum dot arrays and long-range ordering was also achieved. We have also studied the luminescence spectra of these quantum dots. Dots grown on patterned substrates do indeed show different luminescence characteristics compared to dots on non-patterned surfaces.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

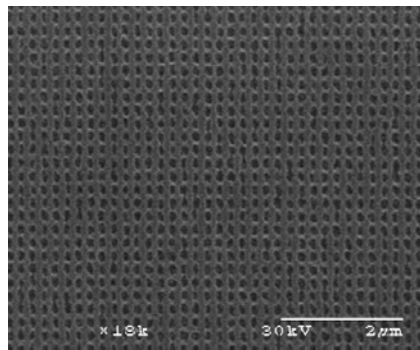
Quantum dots have been one of the most fascinating areas of semiconductor physics over the past few years. With the demonstration of the high optical efficiency of self-assembled formations of quantum dots, there ensued a marked increase in the research on applications such as in optoelectronic devices based on quantum dot structures. Bimberg *et al* [1] achieved improved operation of a quantum dot laser by increasing the density of the quantum dot structures, stacking successive strain-aligned rows of quantum dots and thereby achieving vertical as well as lateral coupling of the quantum dots.

On the other hand, there has been increasing interest in the study of self-organized quantum dot (SOQD) formation on patterned substrates, designed to improve position control. Selective formation of InAs SOQDs on oxide-patterned GaAs substrates using chemical beam epitaxy (CBE) to spatially con-

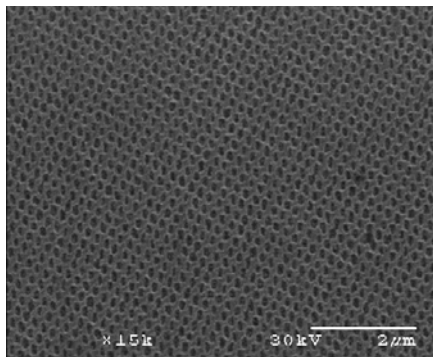
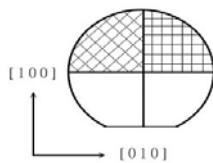
trol the positioning and alignment of SOQDs has been reported in [2–5]. Several groups have fabricated InAs self-assembled quantum dots with molecular beam epitaxy on GaAs substrates which were patterned via contact photolithography or holography followed by wet chemical etching [6–9]. InAs islands grown by CBE on e-beam lithography-patterned GaAs substrates were reported by Jeppesen *et al* [10]. More recently, Lee *et al* [11] have produced long-range ordering of self-assembled QDs using a periodic subsurface stressor lattice.

In this paper, we report the formation of InAs SOQD arrays on an e-beam lithography-patterned GaAs(001) substrate via molecular beam epitaxy. We have also studied the growth behaviour and optical properties of SOQDs grown on chessboard-like patterns with two different orientations with respect to the [100] direction. Technical details and experimental results for the dots formed on the substrate with and without patterns will be compared and discussed. Under photoexcitation, InAs quantum dots on one of the patterned substrates emit stronger luminescence than the others.

⁴ Author to whom any correspondence should be addressed.



Pattern A



Pattern B

Figure 1. A schematic diagram of the patterns defined on the GaAs substrate and SEM images of the pattern A (stripes are oriented at 0° and 90° to $[100]$) and pattern B (stripes are oriented at 45° and 135° to $[100]$) after wet chemical etching.

2. Experimental results and discussion

In the formation of patterned substrates, two chessboard-like patterns with sizes of about $80 \mu\text{m} \times 80 \mu\text{m}$ were first defined on a flat (001) GaAs substrate via e-beam photolithography followed by the wet chemical etching using $\text{H}_2\text{SO}_4:\text{H}_2\text{O}_2:\text{H}_2\text{O} = 1:8:80$. In figure 1 we have shown a schematic diagram of the grid pattern defined on the GaAs substrate. The GaAs substrate was first covered with photoresist, and then exposed to an electron beam to define the patterns. The two chessboard-like patterns were composed of crossed stripes oriented at angles of 0° , 90° (pattern A) and 45° , 135° (pattern B) with respect to the $[100]$ direction. On both pattern A and pattern B, those stripes have a pitch size of $0.1 \mu\text{m}$. The e-beam-defined patterns were etched into the substrate, resulting in pits with area of about $150 \text{ nm} \times 100 \text{ nm}$ and depth about 15 nm . The pits on pattern A have their edges parallel to $[100]$, $[010]$. For pits on pattern B, their edges are parallel to $[\bar{1}10]$, $[110]$. Figure 1 shows the scanning electron microscope (SEM) images of the defined pattern after the wet

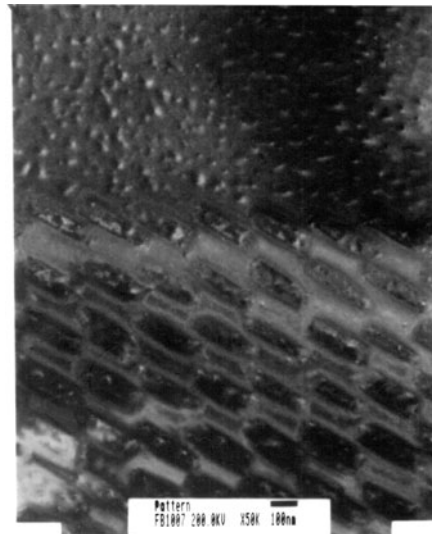


Figure 2. The TEM image of the uncapped SOQDs on the non-patterned area (in the upper half of the figure) and pattern B (in the lower half part).

chemical etching. The pits were rectangular in shape since the etching processes were anisotropic on the two perpendicular facets inside the pits. This resulted in different thicknesses of the walls surrounding the pits. For example, on pattern A, the walls parallel to $[010]$ are thinner than the walls along $[100]$. Even though pits of the two chessboard-like patterns were similar in shape, the TEM images showed that they were bounded by different facets. From the tilt angles in TEM images, we estimated that the facets bounding the pits on pattern A were (101) , $(\bar{1}01)$, (011) and $(0\bar{1}1)$, while the pits on pattern B were bounded by (113) , $(\bar{1}\bar{1}3)$, $(\bar{1}13)$ and $(1\bar{1}3)$.

Before introducing the patterned substrate into the molecular beam epitaxy (MBE) system, the wafer was cleaned in solvents and a thin layer of GaAs was removed from the top using wet chemical etching. The regrowth process started with thermal desorption of the oxide layer at 610°C . The molecular beam epitaxial growth sequence consisted of two monolayers of InAs appearing with the growth rate of 0.05 ML s^{-1} at 520°C after the growth of a 10 nm buffer layer at 570°C on the patterned substrate. The SOQDs were formed on both patterned (A and B) and non-patterned areas, under the same growth conditions. The transmission electron microscope (TEM) image of the SOQDs formed on the non-patterned area is shown in the upper part of figure 2. We have also shown the atomic force microscope (AFM) images taken from the centre of pattern A in figure 3. The AFM image in figure 3 looks slightly different from the SEM image of the e-beam-defined pattern shown in figure 1. The reason is that the wet chemical etching process operative before introducing the patterned sample into the MBE chamber has effected further thinning of the stripes parallel to $[010]$ on pattern A. Therefore, the image of the stripes parallel to $[010]$ is not evident in figure 3. Nevertheless, the inset in figure 3 shows that only a single row of dots was formed on stripes parallel to $[100]$. Those stripes have top widths of about 50 nm . The dots that appeared on the stripes have an average base width of about 30 nm and are more uniform in size than dots formed on the

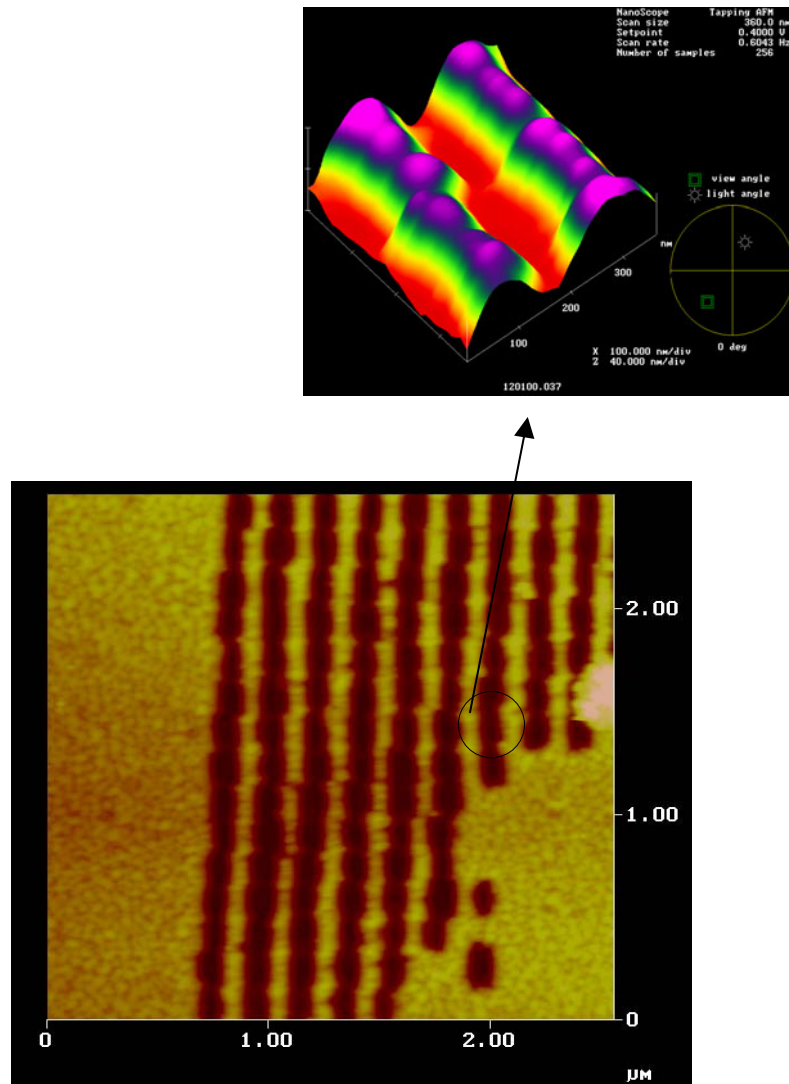


Figure 3. The AFM images of SOQDs grown on the pattern A. The inset shows a single row of dots that appeared on the stripes.

non-patterned area. Note that no dot was found on a stripe parallel to [010] due to its much narrower top width.

The AFM image taken from the inside of the pits on pattern A is also given in figure 4. The inset in figure 4 indicates that there is also a single row of dots formed inside the pits. The position of those dots that appeared inside the pits looks asymmetric with respect to the centre of the pits. The TEM image of QDs on pattern A as shown in figure 5 is also consistent with results from the AFM images.

In contrast to the results on pattern A, the TEM image of the dots on pattern B (as shown in figure 6) shows that the dots were only grown inside the pits and no dots can be found on the top of stripes with orientation parallel to $[\bar{1}10]$. This is due to the undercut edges of the etched sidewalls of the pits, which have prevented the dots from being grown on the top of the stripes [7]. The estimated density of the SOQDs formed on pattern A is about $3 \times 10^{10} \text{ cm}^{-2}$ and is much higher than the densities of those on pattern B (about $4 \times 10^9 \text{ cm}^{-2}$) and the non-patterned area (about the same as for pattern B).

We have studied and compared the photoluminescence spectra from the SOQDs grown on pattern A, pattern B and the

non-patterned area after the samples were capped with 50 nm GaAs. In the photoluminescence experiments, the sample was excited with an argon-ion laser operated at 514.5 nm. The laser was focused to a spot size of approximately $60 \mu\text{m}$ in diameter (to avoid excitation outside the patterned area) with an average power density of about 5 kW cm^{-2} . The sample was kept in a closed-cycle refrigerator at about 15 K. The image of the sample was first magnified in order to direct the laser beam onto the individual patterned areas through a periscope arrangement behind the entrance slit of the spectrometer. The luminescence was then collected and analysed with a combination of a 0.6 m Triplemate spectrometer and a liquid-nitrogen-cooled CCD camera. In figure 7, we have shown the photoluminescence spectra of the SOQD sample taken from three different areas of the sample: pattern A, the non-patterned area and pattern B. The SOQDs grown on pattern A emitted the strongest PL intensity among the three (about four times stronger than that from the other areas). We attribute this to the improvement of the dot quality and the higher density of SOQDs formed in this area. It also indicated that our patterned surface has recovered during the regrowth from the processing damage.

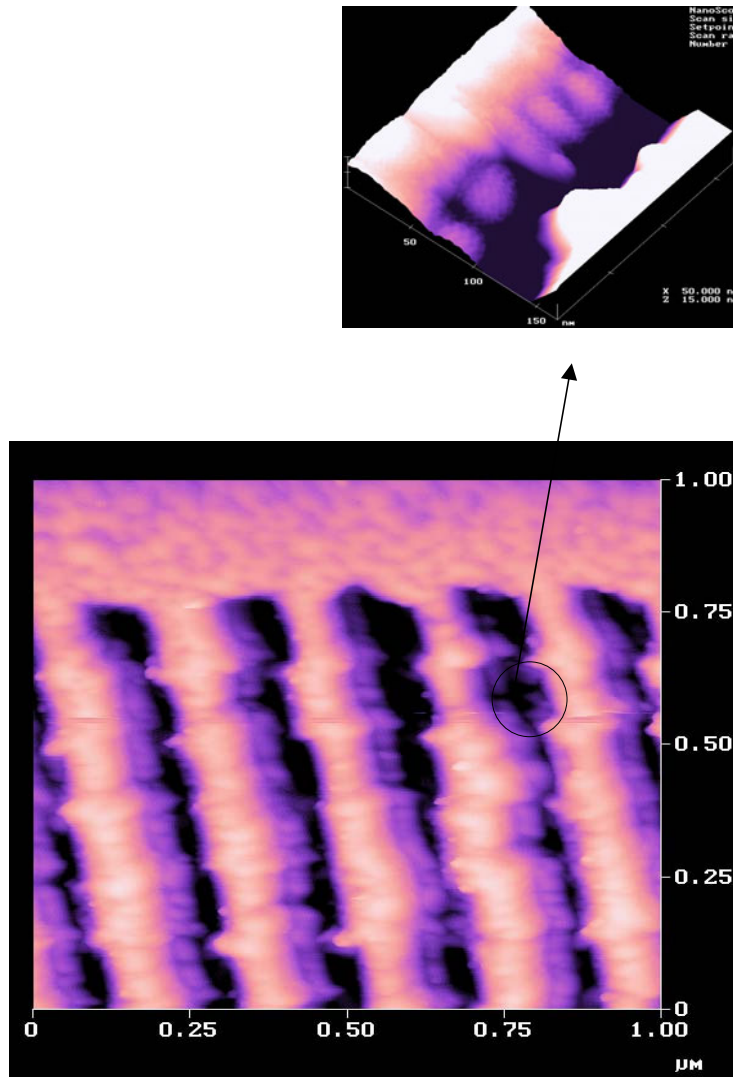


Figure 4. The AFM images of SOQDs inside the pits of pattern A. The inset also shows one single row of dots sited against one of the sidewalls.

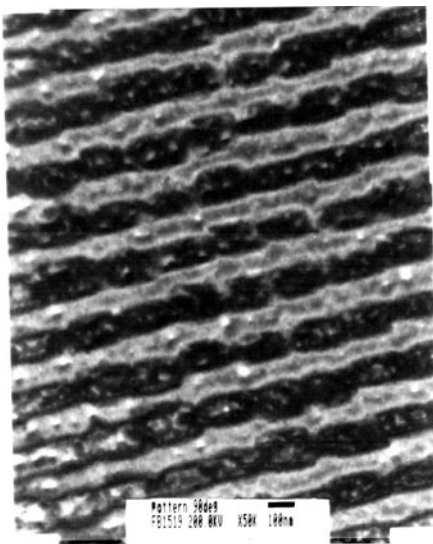


Figure 5. The TEM image of SOQDs on pattern A.

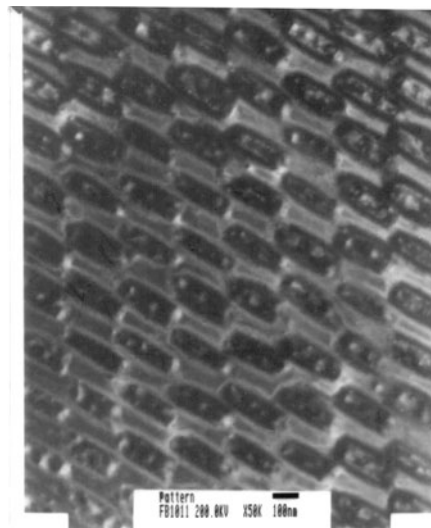


Figure 6. The TEM image of SOQDs on pattern B. The dots only appeared inside the pits.

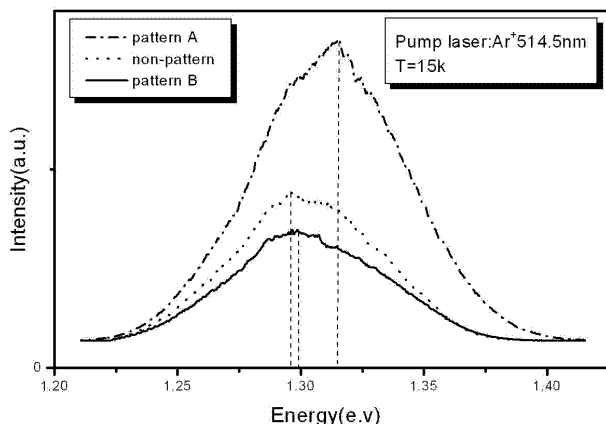


Figure 7. 15 K photoluminescence spectra of the three different areas on the sample: pattern A, non-patterned and pattern B. All of the spectra were taken using a liquid-nitrogen-cooled CCD camera under the same excitation conditions. The integration times for acquiring the three different spectra were the same.

The peak of the luminescence also showed a significant blue-shifting (about 25 meV) compared to the luminescence signal from the dots on the non-patterned area. This indicates that quantum dots formed on pattern A are different in size and composition compared to the dots on pattern B and on the non-patterned area. For SOQDs grown on pattern B, the luminescence intensity and peak position are not very different from those for dots on the non-patterned area.

3. Summary

In summary, we have grown SOQDs via molecular beam epitaxy on patterned GaAs(001) substrates prepared by e-beam lithography and wet chemical etching. Our TEM and AFM images show ordering of SOQDs formed on the

stripes and inside the pits. From our luminescence studies, we found that the SOQDs grown on pattern A gave the strongest luminescence intensity among the three areas that we have investigated. We attribute this to the higher density, ordering and improvement in quality of the dots.

Acknowledgment

This work was supported by the National Science Council of the Republic of China under contract grant no NSC 89-2112-M-259-021 and NSC 90-2215-E-259-005.

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