CHAPTER FIVE

Formation and optical properties of area-controlled InAs quantum dots

5.1 Introduction

In previous chapter, we preliminarily investigated the growth condition for area-controlled dots. To perform AFM measurement and to increase the efficiency of QD growth, we designed the line patterns. Using these designs, we could know the exact values of dot size, and depth of patterned holes. In this chapter, the formation and optical properties of area-controlled dots are closely studied.

5.2 Formation of area-controlled InAs quantum dots

First, InAs coverage dependence of area-controlled quantum dots was investigated. The diameters and pitches of the patterned holes were 100nm and 280nm, respectively. Area-controlled quantum dots with the growth times of 12.6s, 17.1s, 21.6s, 28.3s were grown at 520 °C. Growth rates were 0.011ML/s. Fig.6-1 shows the AFM measurements of area-controlled quantum dots with various InAs growth times. From the analysis of AFM images shown in Fig.5-1, the size (diameter) and density of area-controlled InAs QDs were estimated at each sample. In this case, the density of area-controlled InAs QDs means the average number of quantum dots inside one patterned hole. The dependence of the size and density of area-controlled InAs QDs on the growth time was shown in Fig.5-2. From 12.6s to 21.6s, the size of quantum dots was almost same and dot density was gradually increased with increasing InAs growth time. At 28.3s, however, dot size became suddenly bigger, and dot density was dramatically decreased. It is considered that some quantum dots were combined into one when the excessive amount of InAs is offered.

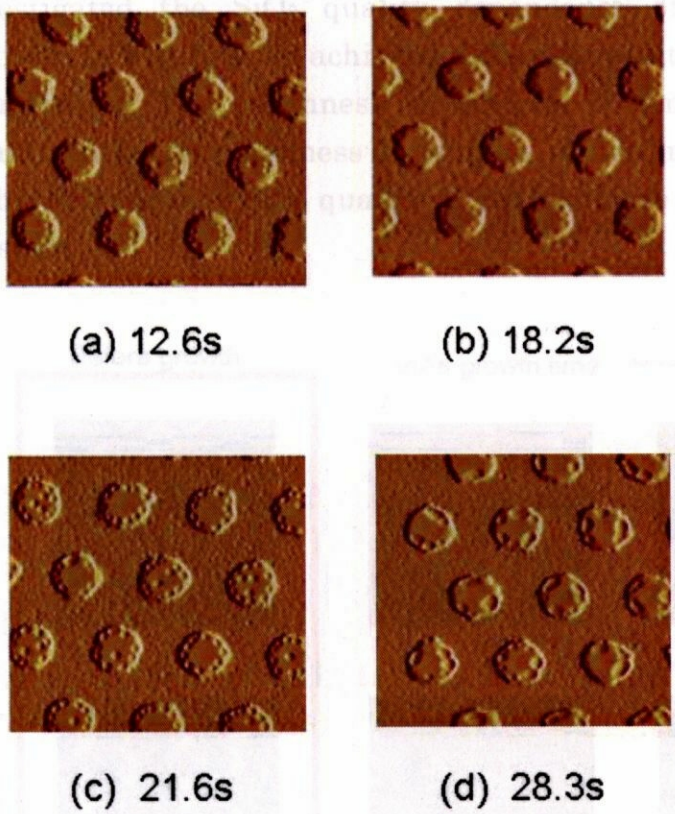


Fig.5-1 1×1 um^2 AFM images of area-controlled quantum dots with various InAs growth times.

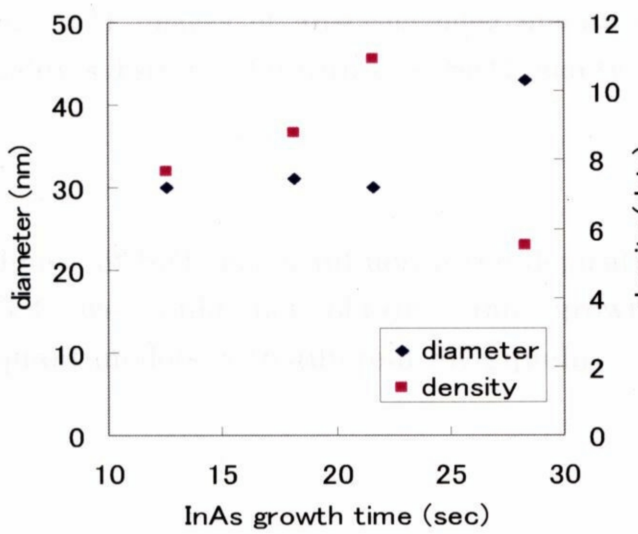


Fig.5-2 The growth time dependence of the size and density of area-controlled InAs QDs

Next, we investigated the SiO_2 quality dependence of quantum dot formation. As shown in Fig. 5-3, we achieved different results with different SiO_2 quality substrates. The roughness of samples shown in Fig.5-3 (b) was 8.995 (Rmax), while the roughness of samples shown in Fig.5-3(a) was 2.798 (Rmax) This difference of SiO_2 quality might be due to the irregularity of sputtering system.

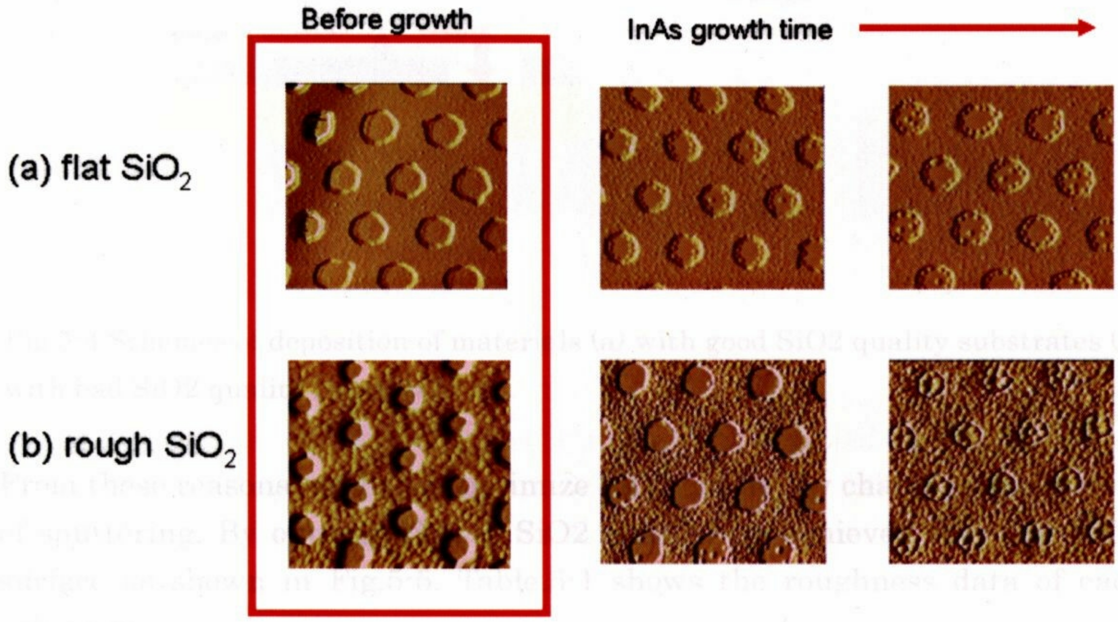


Fig.5-3 1×1um2 AFM images of area-controlled quantum dots grown with different SiO2 quality substrates. (a) with good SiO2 quality (b) with bad SiO2 quality

Since the roughness of SiO_2 mask influence the deposition of material as shown in Fig.5-4, we could not obtain same growth conditions for area-controlled quantum dots with different SiO_2 quality.

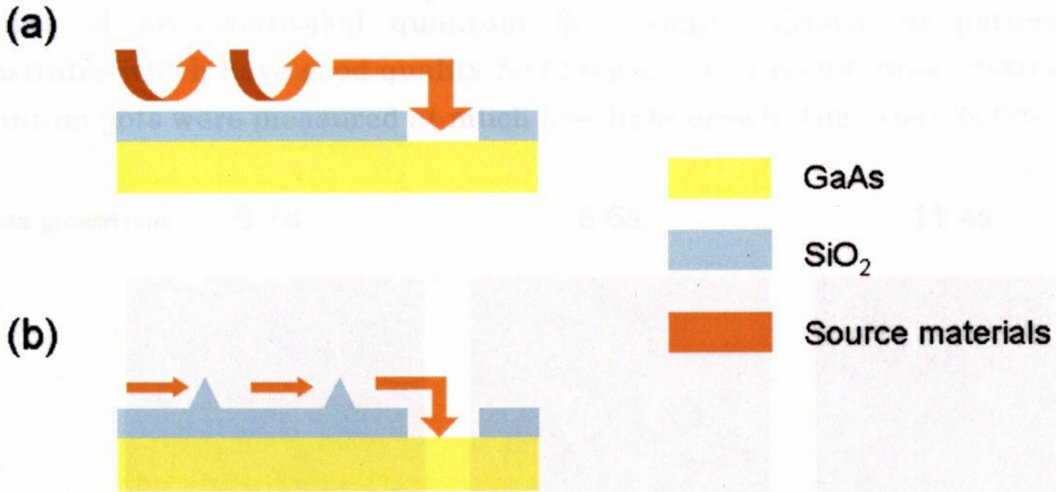
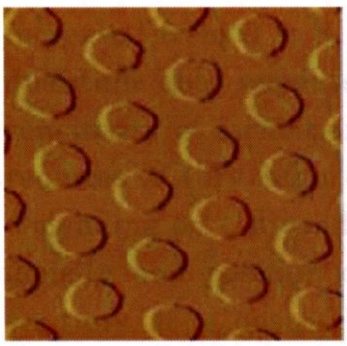


Fig.5-4 Schemes of deposition of materials (a) with good SiO2 quality substrates (b) with bad SiO2 quality substrates

From these reasons, we had to optimize SiO2 quality by changing the factors of sputtering. By optimization of SiO2 quality, we achieved very flat SiO2 surface as shown in Fig.5-5. Table.5-1 shows the roughness data of each substrate.



	Rms (Rq)	Rmax
Flat substrate	0.229	2.798
Rough substrate	1.913	8.995
Optimized substrate	0.133	1.746

Table.5-1 Roughness data of each substrate

Fig.5-5 1×1um² AFM image of a patterned SiO2/GaAs substrate with very flat SiO2 surface.

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With these substrates, we have grown area-controlled quantum dots under the same growth condition as previous samples. Fig.5-6 shows the AFM images of area-controlled quantum dot samples grown on patterned substrates which have good quality SiO2 masks. As a result, area-controlled quantum dots were measured at much less InAs growth time than before.

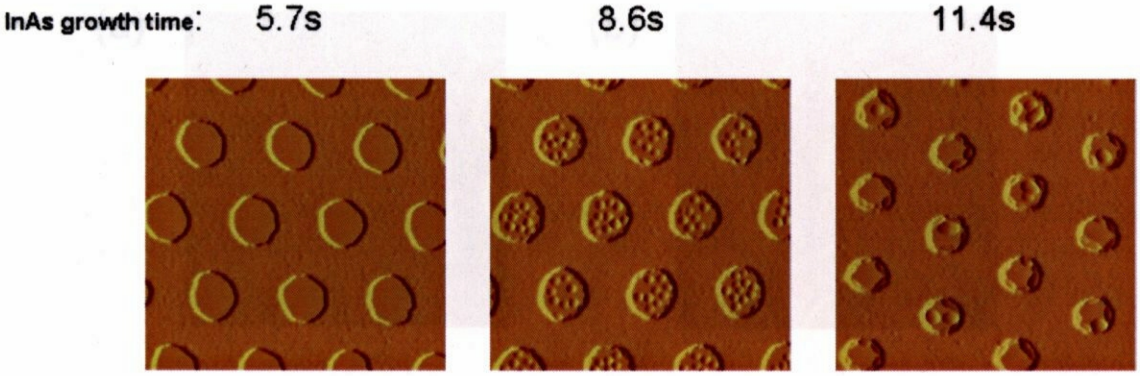

Fig.5-6 1×1um² AFM images of area-controlled quantum dot samples grown on patterned substrates which have good quality SiO2 masks

One of the main issues for formation of area-controlled quantum dots is difficulty in control over quantum dot size and density. As shown in Fig.5-6, the formation of area-controlled quantum dots is very sensitive to InAs coverage because growth time became 20 times lower than non-patterned growth due to the growth enhancement. For instance, when InAs growth time is increased from 5.7s to 8.6s, which corresponds to a difference of only 2.9s, the number of area-controlled quantum dots in one GaAs exposed hole, increased from 0 to 8. And by adding another 2.8s, huge (diameter>50nm) quantum dots appears which are assumed as coalesced QDs. This high sensitivity of formation of area-controlled quantum dots on InAs coverage requires a very high control of growth, while growth conditions changes depending on the conditions of MOCVD system. Another main issue is the density and position of area-controlled quantum dots. For single photon emitters, it is desirable that each GaAs exposed holes have only one dot which located on the center. In our case, however, usually more than 6 dots exist in one hole and most QDs located on the edge. When we tried to decrease the density using lower InAs coverage, the number of GaAs exposed holes which have no quantum dots is increased and the size of quantum dots get smaller as shown in Fig.5-7(a). In the process of optimizing growth conditions, we found the promising solution to resolve the second issue.

Fig.5-7(b) shows the AFM image of low density area-controlled quantum dots. There are 1～4 dots inside one hole, and the position of quantum dots become closer to the center.

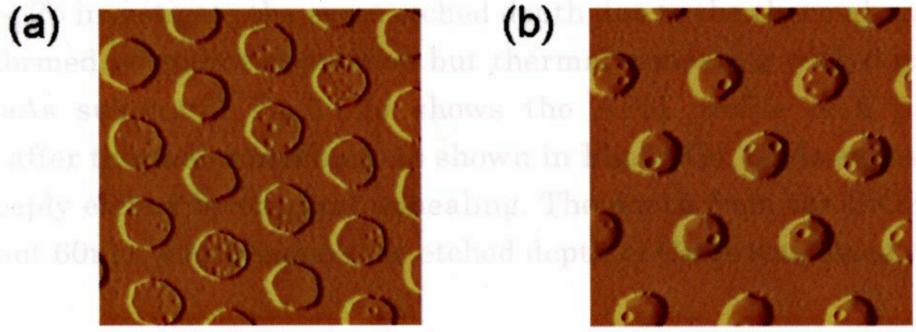

Fig.5-7 1×1um2 AFM images of area-controlled quantum dot samples which have (a) many small(<3nm) dots or no dots (b) only a few big (about 12nm) dots

In order to investigate the morphology of these low density area-controlled quantum dots, we used the section analysis of AFM. Figure.5-8(a) indicates the AFM image of area-controlled quantum dots which is focused on the only one hole. And the AFM profile of this hole is shown in Fig.5-8(b). As shown in Fig.5-8(b), a quantum dot was grown on the deeper GaAs plane (the depth of GaAs hole is about 37nm), while previous quantum dots were grown on the much less deeper GaAs plane (depth of GaAs hole: about 17nm) as mentioned in chapter4.

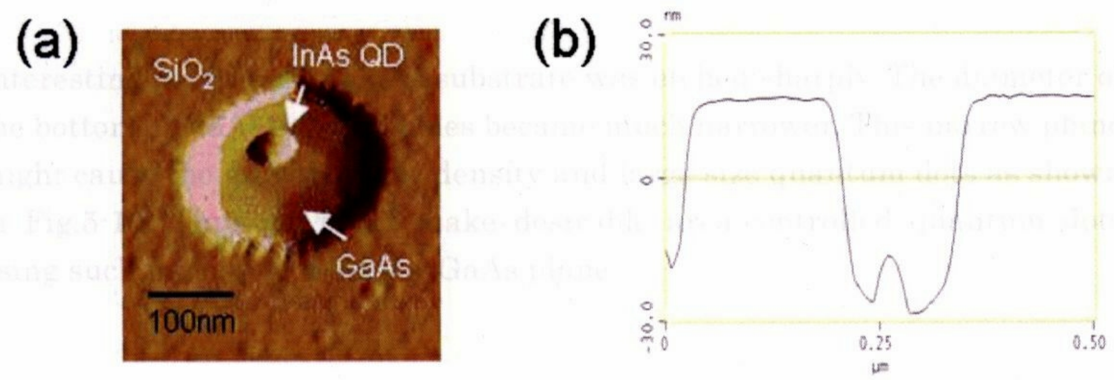

Fig.5-8 (a) 1×1um2 AFM image of a single QD grown in a GaAs exposed hole (b) section profile of (a)

With this AFM profile, we could confirm that GaAs exposed holes were etched. And it turns out to be caused by thermal annealing. Thermal annealing was performed at the first stage of growth for deoxidization. Under the atmosphere of TBA, the samples were annealed at 800℃ for 10 minutes. To investigate the exact etched depth due to the thermal annealing, we performed no source deposition but thermal annealing with a patterned SiO2/GaAs substrate. Fig.5-9(b) shows the AFM profile of a patterned sample after thermal annealing. As shown in Fig.5-9(b), GaAs exposed holes were deeply etched by thermal annealing. The depth from the SiO2 surface was about 60nm, which means the etched depth of GaAs was about 35nm.

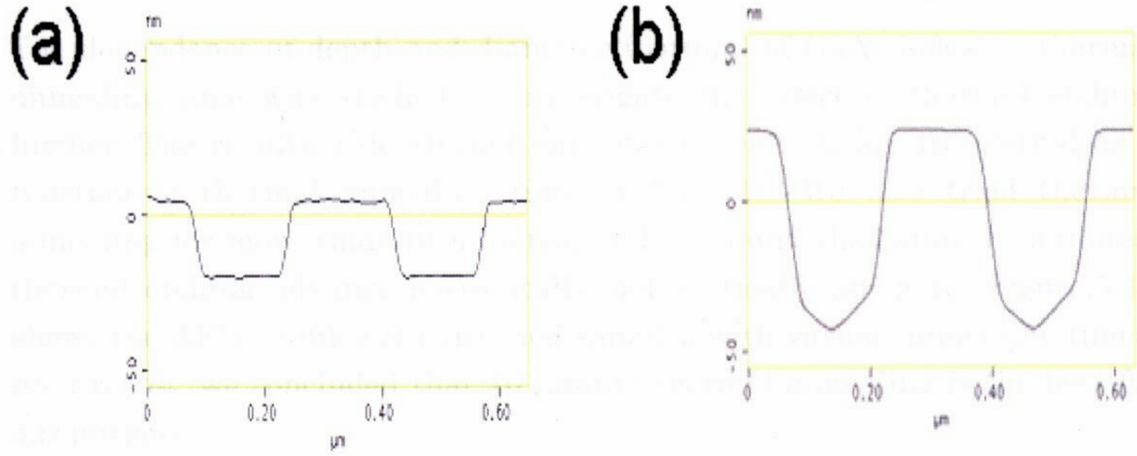

Fig.5-9 AFM profiles of patterned substrates (a) before growth (b) after thermal annealing

Interesting point is that GaAs substrate was etched sharply. The diameter of the bottom of GaAs exposed holes became much narrower. This narrow plane might cause the growth of low density and large size quantum dots as shown in Fig.5-10. Thus we could make desirable area-controlled quantum dots using such narrow and deeper GaAs plane.

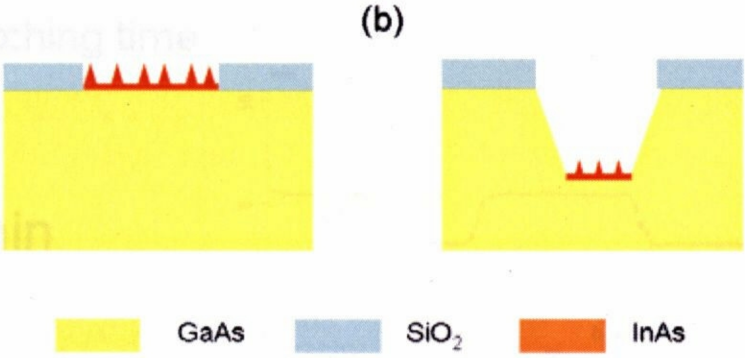

Fig.5-10 Schemes of area-controlled quantum dot (a) without thermal annealing effect (b) with thermal annealing effect

The dependence of depth and diameter (bottom) of GaAs holes on thermal annealing time was studied to investigate the effect of thermal etching further. The resulting depth and diameter of GaAs holes are plotted as a function of thermal annealing time in Fig.5-11. We also tried thermal annealing for more than 10 minutes, and we found that after 10 minutes, thermal etching advance horizontally, not vertically anymore. Figure.5-12 shows the AFM profiles of patterned samples with various annealing times. As a result, we concluded that 10 minute thermal annealing is the best for our purpose.

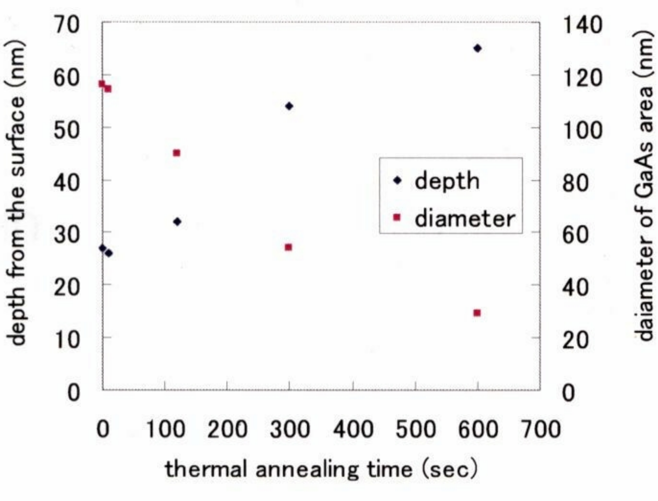

Fig.5-11 data plots from AFM measurements of depth and diameter of GaAs holes as a function of thermal annealing time

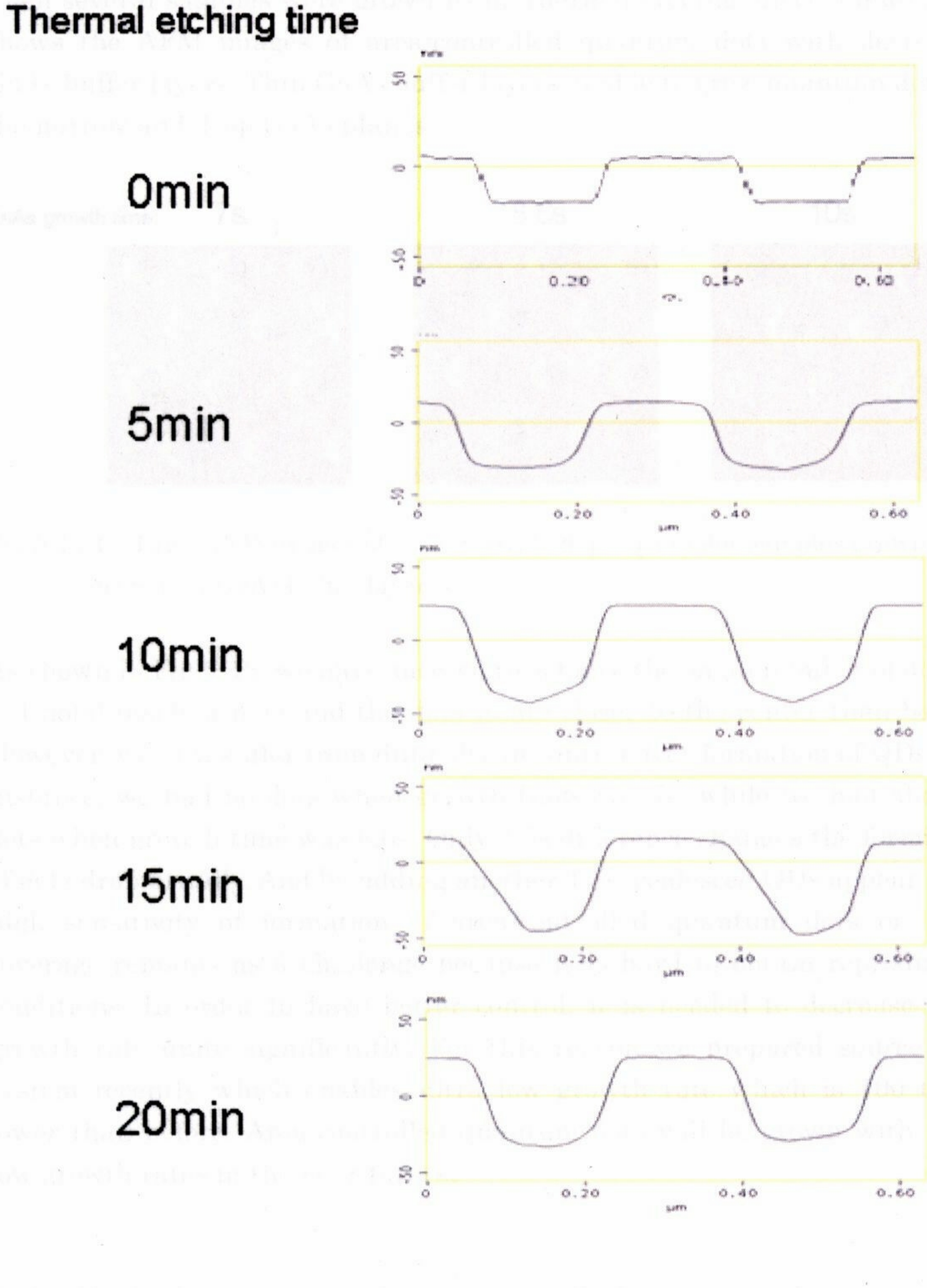

Fig.5-12 AFM profiles of patterned samples with various annealing times

Then several samples were grown using thermal etching effect. Figure.5-13 shows the AFM images of area-controlled quantum dots with decreased GaAs buffer layers. Thin GaAs buffer layers enable to grow quantum dots on the narrow and deep GaAs planes.

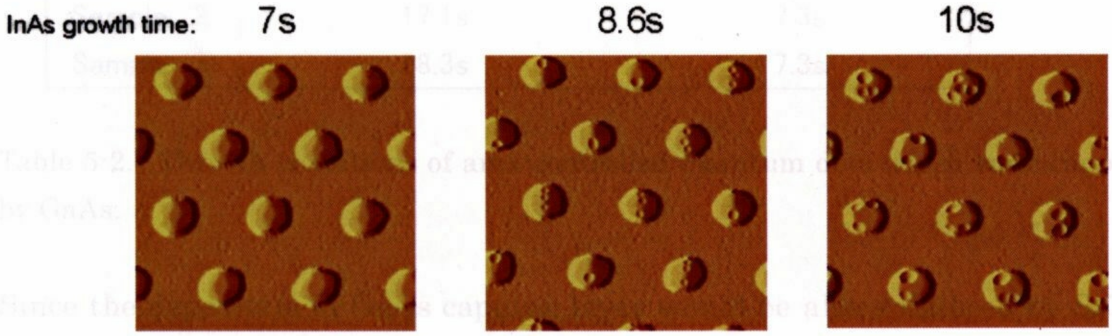

Fig.5-13 1×1um² AFM images of area-controlled quantum dot samples grown with decreased GaAs buffer layer

As shown in Fig.5-13, we have succeed to achieve the small number of dots (1 ~4 dots) inside a hole and the dots locate closer to the center than before. However, we still suffer from difficulty in control over formation of QDs. For instance, we had no dots when growth time was 7s, while we had about 2 dots when growth time was 8.6s. Only 1.5s difference changes the formation of dots dramatically. And by adding another 1.5s, coalesced QDs appear. This high sensitivity of formation of area-controlled quantum dots on InAs coverage remains as a challenge because it is hard to obtain reproducible conditions. In order to have better control, it is needed to decrease InAs growth rate quite significantly. For this reason we prepared source flow system recently which enables ultra low growth rate which is 100 times lower than before. Area-controlled quantum dots will be grown with ultra low growth rates in the near future.

5.2 Optical properties of area-controlled quantum dots

Next, area-controlled quantum dots which were capped by GaAs were grown to investigate the PL properties. First, we made three kinds of capped samples without using thermal etching effect. Table 5-2 shows the growth conditions for three samples. For all samples, InAs growth and GaAs capping were performed at 520℃ and GaAs BL growth time was 5sec

	InAs growth time	GaAs capping time
Sample ①	17.1s	10.9s
Sample ②	17.1s	7.3s
Sample ③	28.3s	7.3s

Table 5-2. Growth conditions of area-controlled quantum dots which were capped by GaAs.

Since the deposition of GaAs capping layer would be also enhanced by SiO2 mask, we decreased the amount of GaAs for capping. In order to see the difference due to the thickness of GaAs capping, we prepared two samples which have same InAs coverage and different GaAs thickness. (sample① and sample②) And we also prepared two different InAs coverage samples .(sample② and sample③) Fig.5-14 shows the AFM images of two different InAs coverage samples.

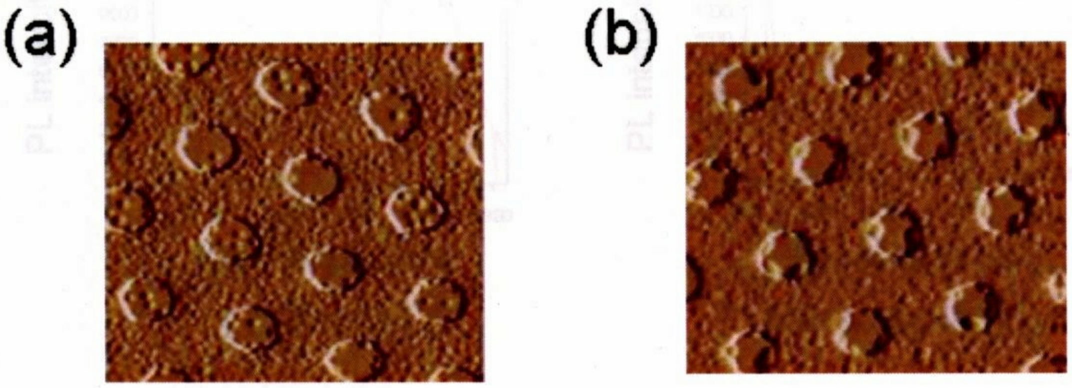

Fig.5-14 AFM images of two different InAs coverage samples. InAs growth time were (a) 17.1s (b) 28.3s, respectively.

Fig.5-14(a) corresponds to the sample① and ② and Fig.5-14(b) corresponds to the sample③. For all samples, the depth of GaAs holes was about 17nm. Micro PL spectra of these samples are shown in Fig.5-15. PL measurements were carried out at 7K using Ti-Sa laser and single- grating monochromator.

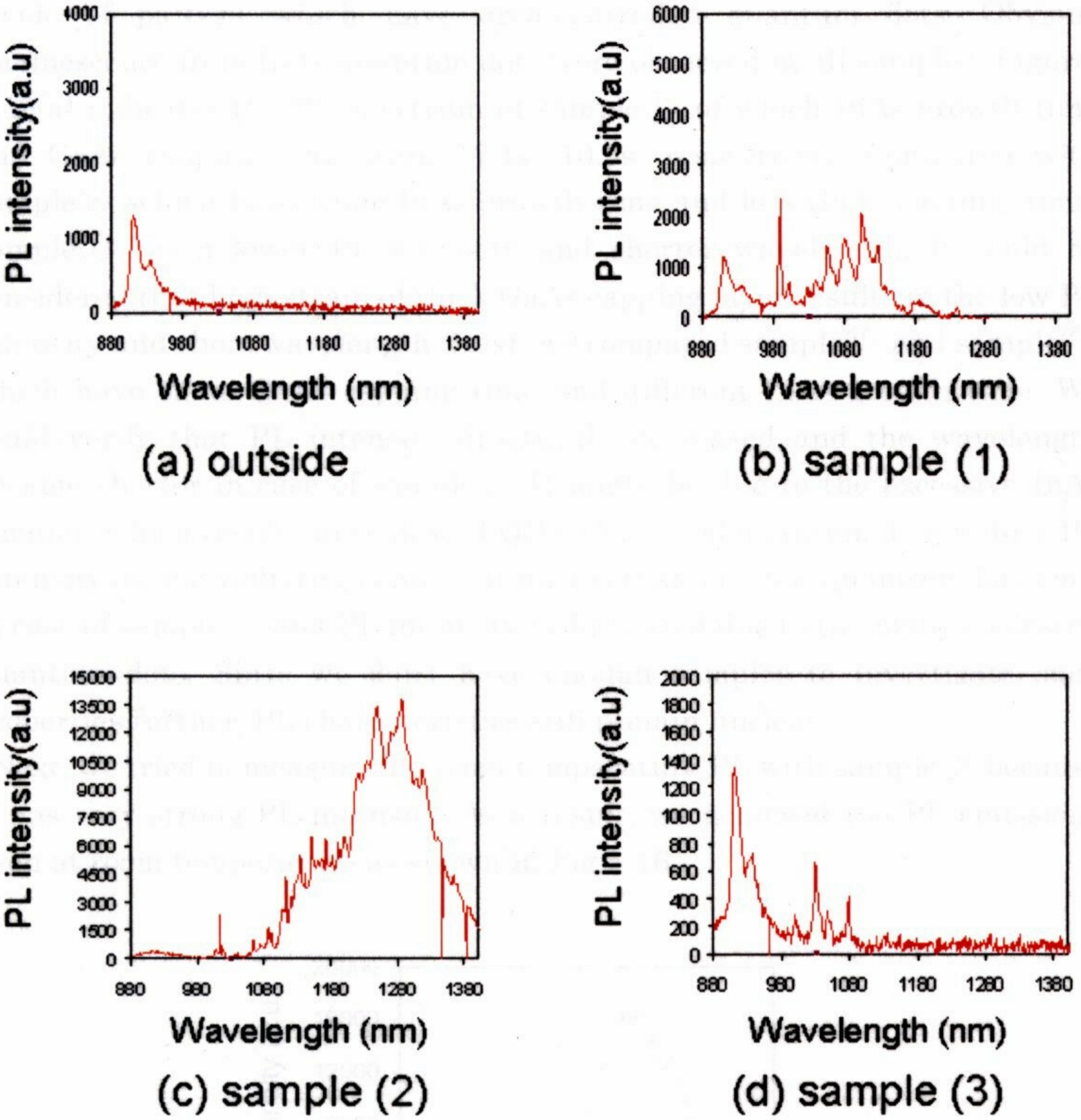

Fig.5-15 low temperature micro PL spectra of area-controlled quantum dots: (a) PL spectrum of outside of pattern, (b) PL spectrum of sample (1), (c) PL spectrum of sample (2), (d) PL spectrum of sample (3)

Figure.5-15(a) shows the PL spectrum of outside of pattern. Since there is no InAs quantum outside of pattern, only very weak luminescence from the wetting layer was observed. Figure.5-15(b)(c)(d) show the PL spectra of inside of pattern which have area-controlled quantum dots. Obvious luminescence from InAs quantum dots were observed in all samples. Figure. 5-15(a) indicates the PL spectrum of sample①, of which InAs growth time and GaAs capping time were 17.1s, 10.9s, respectively. Compared with sample②, which have same InAs growth time and less GaAs capping time, sample① has a lower PL intensity and shorter wavelength. It could be considered that high strain of thick GaAs capping layer results in the low PL intensity and short wavelength. Next, we compared sample② and sample③, which have same GaAs capping time and different InAs growth time. We could verify that PL intensity drastically decreased and the wavelength became shorter in case of sample③. It might be due to the excessive InAs amount, which results in coalesced QDs. Coalesced quantum dots reduce PL intensity as non-radiative centers. It means that only few quantum dots emit in case of sample③, and PL intensity is decreased due to the many coalesced quantum dots. Since we don't have enough samples to investigate such properties further, PL characteristics still remain unclear.

Next, we tried to measure the room temperature PL with sample② because it has very strong PL intensity. As a result, we achieved the PL emissions even at room temperature as shown in Fig.5-16.

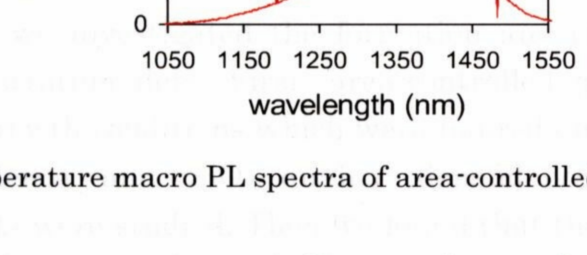

Fig.5-16 room temperature macro PL spectra of area-controlled QDs

Emission at room temperature indicates the good optical quality of area-controlled quantum dots. So far, room temperature photoluminescence from area-controlled quantum dots has barely been achieved because of damaged interface due to the patterning process. This good optical quality indicates that our area-controlled quantum dots have great potential to be positioned around the center of photonic crystals.

Next, PL measurements of area-controlled quantum dots which were grown by using thermal etching effect were performed. Figure.5-17(a) shows the micro PL spectra at 7K and Fig.5-17(b) shows the macro PL spectra at room temperature. As seen from Fig.5-17, strong luminescence from the quantum dots was observed both at ultra low temperature and at room temperature. It is considered that the thermal etching yields the removal of damaged interface and thus enables us to grow quantum dots with good optical quality.

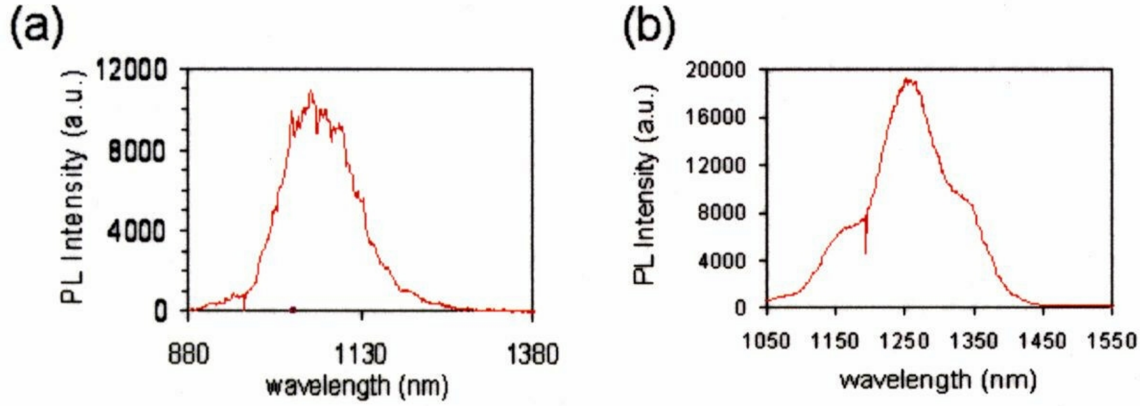

Fig.5-17 Ultra low temperature micro PL spectra of area-controlled QDs which were grown by using thermal etching effect

6.4 Conclusion remarks

In this chapter, we investigated the formation and optical properties of area-controlled quantum dots. First, area-controlled quantum dots were grown by using growth conditions which were figured out in chapter.4. InAs coverage dependence and SiO_2 quality dependence of formation of area-quantum dots were studied. Then we found that thermal etching effect allows us to make low density quantum dots which were desirable for combination with photonic crystals. Next, PL characteristics of area-controlled quantum dots were investigated. For all samples, we could achieve emissions from quantum dots, and we succeed to obtain the PL emissions even at room temperature. It is considered that thermal etching yields the removal of damaged interface and thus enables us to grow quantum dots with good optical quality.