Chapter Four

Studies on growth conditions for area-controlled quantum dots

4.1 Introduction

In the previous chapter, the fabrication of patterned SiO_2/GaAs substrates was introduced. The resulting patterned substrate was covered with SiO_2 mask and only $10 \times 10 \mu m^2$ area on the center has GaAs exposed holes. Figure.4-1 shows the schematic illustrations of a patterned SiO2/GaAs substrate. Considering the growth enhancement within the exposed window from the masked region, the growth conditions of quantum dots on patterned SiO2/GaAs substrates will be quite different from those of non-patterned substrates. In order to grow QDs inside GaAs exposed windows, we investigated the suitable growth conditions for area-controlled dots.

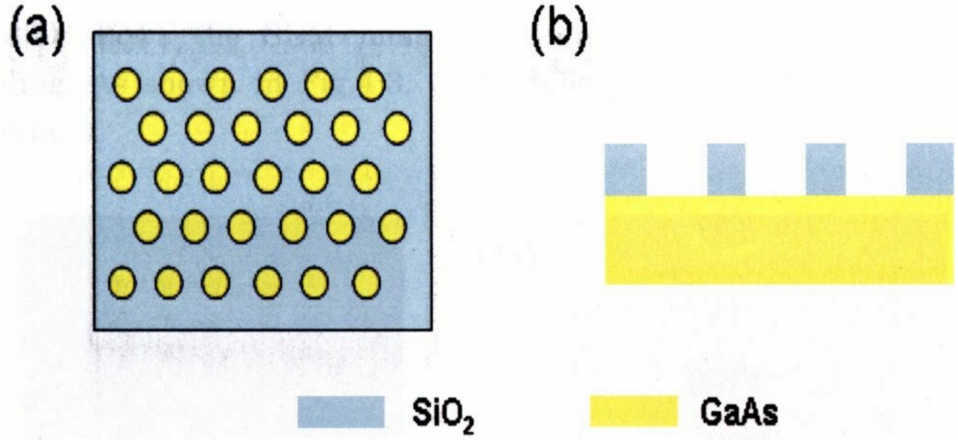

Fig.4-1 Schematic illustrations of a patterned SiO2/GaAs substrate: (a) top view, (b) cross-sectional view

4.2 Growth conditions for area-controlled quantum dots

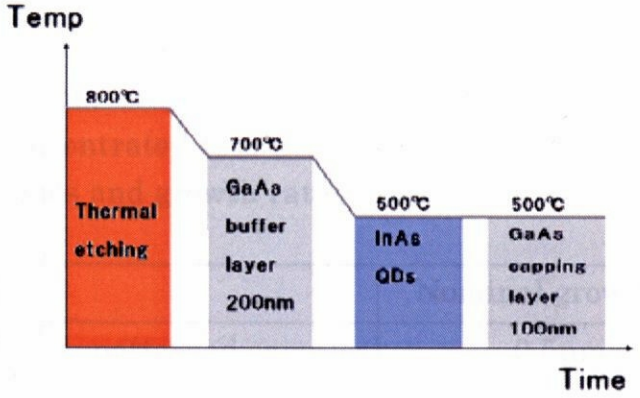

Fig.4-2 Schematic diagram of QD growth procedure

(a) Thermal etching

As introduced in chapter.2, the first step of QD growth is thermal annealing as shown in Fig.4-2. Before deposition of materials, we performed only thermal annealing with a patterned SiO2/GaAs substrate. Annealing was carried out at 800°C for 10 minutes time same as usual. Figure.4-3 (a) shows the SEM image of a patterned sample before thermal annealing, and Fig.4-3(b) shows the SEM image of a patterned sample after thermal annealing. As shown in Fig.4-3, GaAs substrate seems to be etched after annealing.

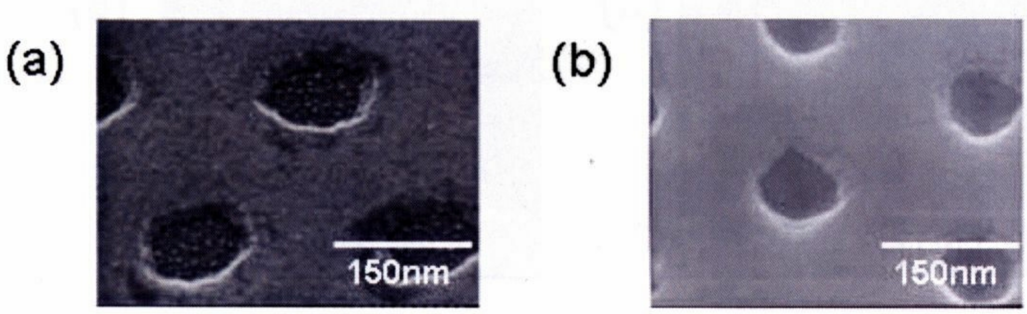

Fig.4-3 SEM images of patterned samples: (a) before thermal annealing, (b) after thermal annealing

(b) GaAs buffer layer

For deposition of GaAs layer, we had to change the growth conditions because the GaAs flow would be concentrated into the GaAs exposed holes due to the growth enhancement within the exposed window from the masked region. Since we couldn't know how much amount of material would be concentrated into the GaAs exposed holes, first we tried to decrease growth times and growth rates as shown in Table.4-1.

	Nominal growth rate	Growth time
Non-patterned samples	0.5nm/s	400s
Patterned ①	0.125nm/s	20s
Patterned ②	0.0625nm/s	5s

Table.4-1 Growth conditions for GaAs buffer layer

Figure.4-4(a) shows the SEM image of *patterned①*. We could confirm that GaAs buffer layers were overgrown although the amount of GaAs layer was very small. It corresponds to the amount for 2.5nm thick GaAs buffer layer in case of non-patterned growth. From this result, we verified that growth enhancement occurred in case of growth with patterned substrates. For *patterned②*, we decreased the amount of GaAs buffer layer again and could succeed to make GaAs exposed area flatter as shown Fig.4-4(b).

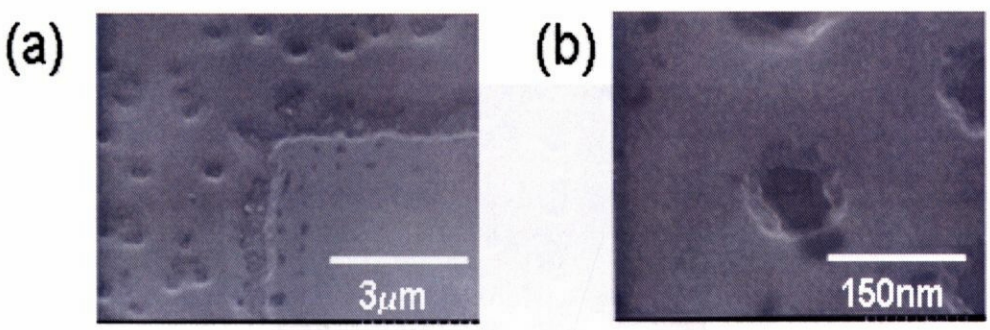

Fig.4-4 SEM images of patterned samples after deposition of GaAs buffer layer:
(a) overgrown, (b) succeedded

(c) InAs QD growth

As mentioned above, growth enhancement within the exposed window from the SiO2 masked area occurred in case of patterned substrates. Thus we also investigated the amount of InAs for making quantum dots inside GaAs exposed holes. As shown in Table.4-2, we decreased InAs growth time dramatically.

	Diameter/pitch (nm)	Growth time (s)
Non-patterned	×	190 (for 2.1ML)
Sample (a)	100/340	6.3
Sample (b)	100/340	9.6
Sample (c)	100/340	13.4
Sample (d)	100/280	13.4
Sample (e)	100/280	20.8

Table.4-2 Design parameters of patterned substrates and InAs growth time

Fig.4-5 shows the SEM image of sample (a). The growth time of sample (a) was 6.3s which corresponds to the 3.3% of 2.1ML in case of non-patterned substrates. We could see the surface of exposed window get arose but no dots existed.

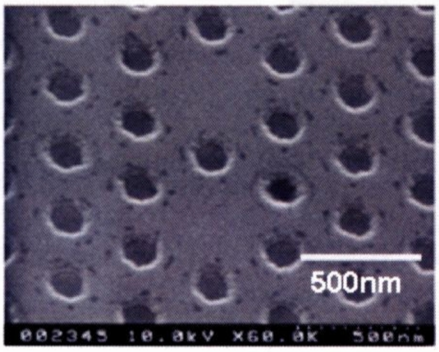

Fig.4-5 SEM image of sample (a)

Next, we increased the growth time to 9.6s (sample (b)). Fig.14 shows the SEM image of sample (b). Most GaAs exposed holes have no dots as shown in Fig.4-6(a), but we could find only one hole which have quantum dots. (Fig.4-6 (b)) Since this hole was the edge of the pattern and upper hole was failed to etch, InAs flow might be concentrated into this hole and thus quantum dots were grown. There are 5 dots inside the hole and their sizes were very small as shown in Fig.4-6(c). (Diameter: about 10nm)

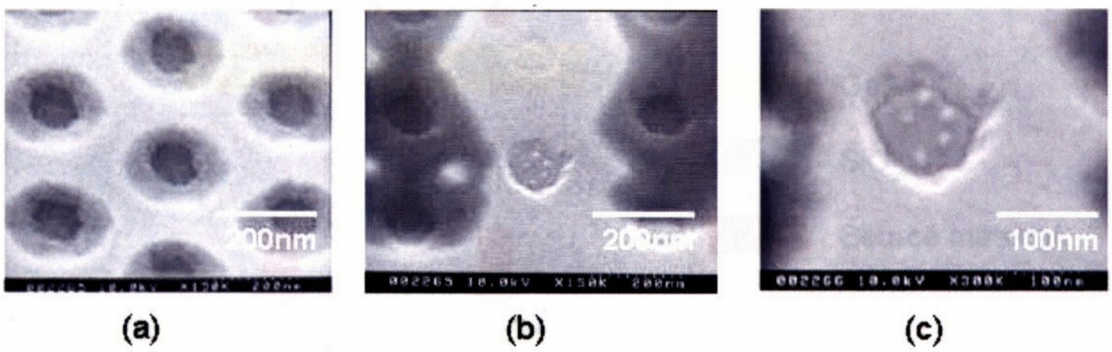

Fig.4-6 SEM image of sample (b)

In order to grow quantum dots inside every GaAs exposed holes, we increased the growth time to 13.4s. Sample (c) and sample (d) were grown with these conditions and their pitches were 340nm, 280nm, respectively. Figure.4-7 shows the SEM images of sample (c) and sample (d). The interesting point is that we have quantum in case of sample (c), but we don't have any quantum dots in case of sample (d).

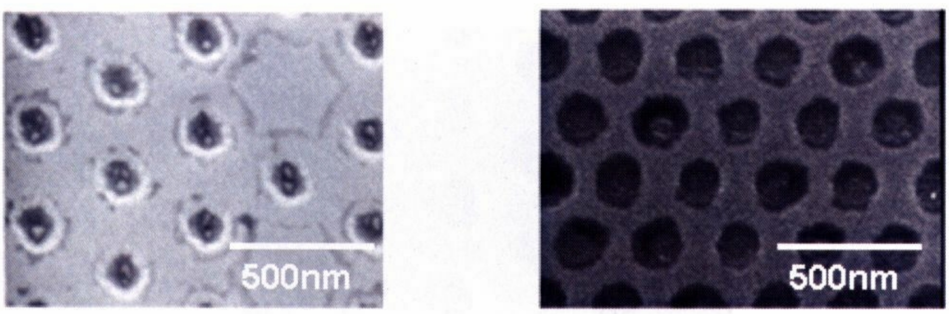

Fig.4-7 SEM images of sample (c) and sample (d)

It can be considered because sample (c) has less patterned GaAs exposed holes than sample (d). As shown in Fig.4-8, when the area of GaAs exposed region get smaller, the concentration of source material become higher. Thus, source material might be more concentrated in case of sample (c) and it might affect the formation of area-controlled quantum dots.

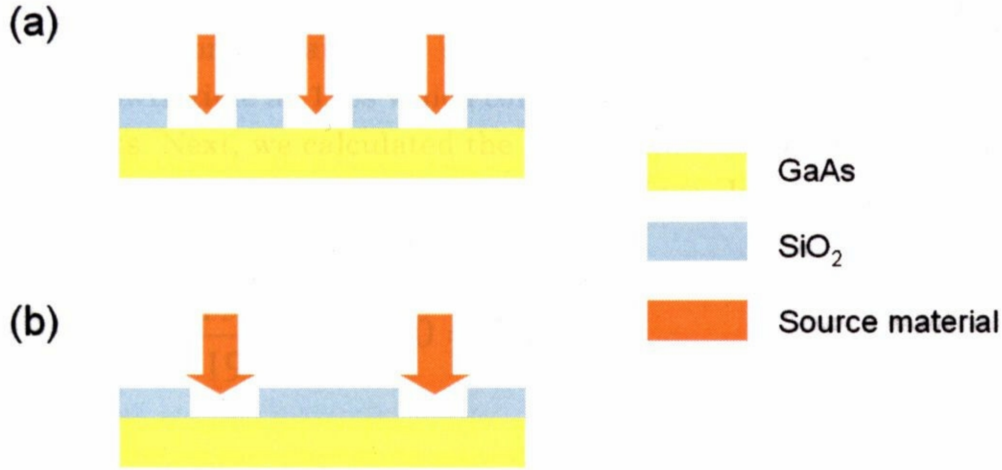

Fig.4-7 Schematic illustrations of deposition of materials: (a) in case of more GaAs exposed holes, (b) in case of less GaAs exposed holes

For sample (e), InAs growth time was increased to 20.8s in order to grow quantum dots inside 280nm pitch GaAs exposed holes. As shown in Fig.4-8, we achieved quantum dots inside every hole. Since we increased InAs growth time drastically, the size of dots was large. (Diameter is about 50nm)

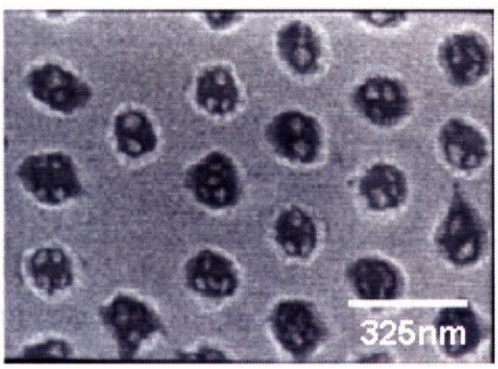

Fig.4-7 SEM image of sample (e)

4.3 Optimization of pattern design

In the previous section, we preliminarily investigate the growth conditions for area-controlled dots. We achieved area-controlled quantum dots when InAs growth time was 13.4s for the substrate of which hole pitch was 340nm, and when InAs growth time was 20.8s for the substrate of which hole pitch was 280nm. From these results, we carried out rough calculations for estimating the concentration of source materials. First, we assumed the standard growth time as 190s which corresponds to 2.1ML for non-patterned quantum dots. Next, we calculated the growth time ratio between patterned samples and non-patterned samples. For instance, In case of 100/340, the ratio is calculated as:

Pat/non-pat: $$\frac{13.4\,\text{sec}}{190\,\text{sec}} \times 100 = 7\%$$

Then we also calculated the ratio between the areas of whole GaAs exposed holes and that of entire patterned area (we refer it 'filling factor'). When diameter is 100nm and pitch is 340nm, filling factor is calculated as:

Filling factor: $$\frac{50nm \times 50nm \times 3.14}{340nm \times 340nm} \times 100 = 6.8\%$$

All the values are shown in Table.4-3. We can see that the ratio of pat/non-pat is similar to the filling factor.

	Diameter/pitch (nm)	Growth time (s)	Pat/non-pat (%)	Filling factor (%)
Non-patterned	×	190 (for 2.1ML)	100	100
Sample (c)	100/340	13.4	7	6.8
Sample (e)	100/280	20.8	11	10

Table.4-3 Ratios between pat/non-pat and filling factors

According to these results, it can be considered that only nearby SiO_2 region affect the formation of area-controlled quantum dots. Owing to this fact, we changed the pattern designs as shown in Fig.4-8, in order to perform further analysis. Instead of only 10×10μm² patterned area in one sample, we made a line structure by repeating 10×10μm² patterns. Such line structure allows us to perform AFM analysis and also to measure PL luminescence much easily. Using AFM, we can achieve the exact thickness and size of materials inside patterned holes.

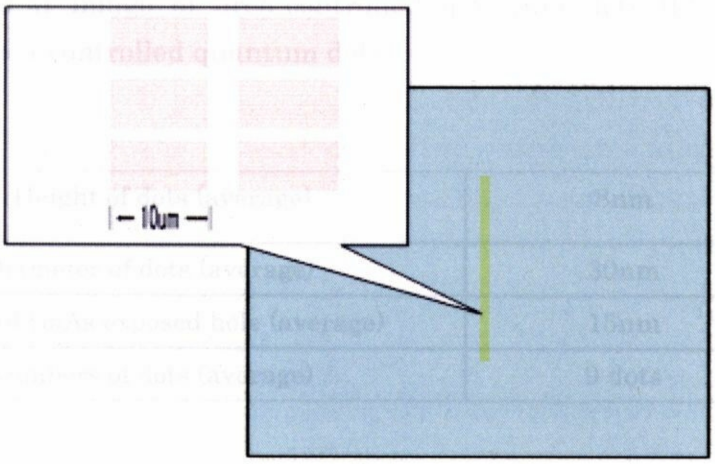

Fig.4-8 Schematic illustration of new designed pattern

With new designed substrates, we fabricated area-controlled quantum dots and measured by AFM. These new substrates were achieved by same fabrication procedures as we used before. Fig.4-9 shows the first AFM image and AFM profile of area-controlled dots. The diameter and pitch of the patterns were 100nm, 280nm, respectively. We used 12.6s as InAs growth time and other growth conditions were same as before. As shown in Fig.4-9(b), we could measure the dot size and the depth of GaAs exposed hole. (table.4-4)

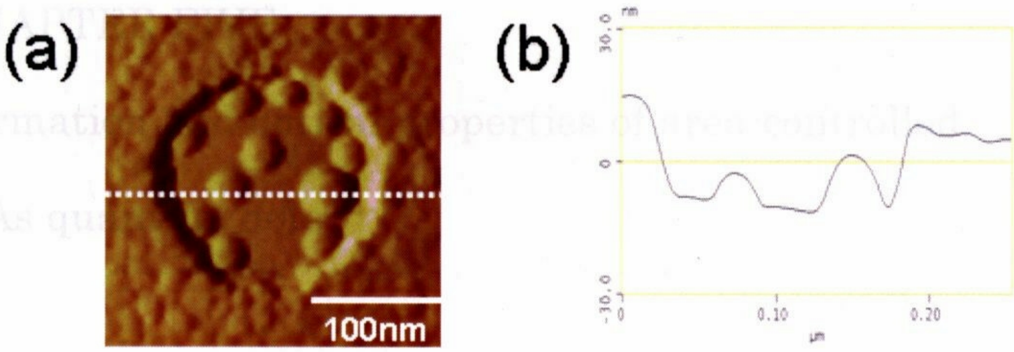

Fig.4-9 (a) AFM image of area-controlled quantum dots (b) AFM profile of area-controlled quantum dots

Height of dots (average)	8nm
Diameter of dots (average)	30nm
Depth of GaAs exposed hole (average)	15nm
Numbers of dots (average)	9 dots

Table.4-4 various data of area-controlled quantum dots

4.4 conclusion remarks

In this chapter, we preliminary studied the growth conditions for area-controlled quantum dots. Because of the growth enhancement within the exposed window from the masked region, the growth conditions of quantum dots on patterned SiO_2/GaAs substrates was quite different from those of non-patterned substrates and thus we had to decrease the depositing amount of materials. We found that only nearby SiO_2 region affect the formation of area-controlled quantum dots. Owing to this fact, we changed the pattern designs in order to perform further analysis. Instead of only $10\times10\mu m^2$ patterned area in one sample, we made a line structure by repeating $10\times10\mu m^2$ patterns. Such line structure allows us to perform AFM analysis and also to measure PL luminescence much easily. Specific investigation about the formation and optical properties of area-controlled quantum dots will be described in Chapter.5.