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Uniform InAs Quantum-dots on vicinal GaAs (100) substrates by pulsed atomic layer epitaxy via metal-organic chemical vapor deposition

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InAs QDs were grown on vicinal GaAs (100) substrate via metal organic chemical vapor deposition. Several different methods were used to investigate the formation of InAs QDs, including conventional continue growth mode and pulsed atomic layer epitaxy (PALE) mode. When the continuous growth mode was used, a bimodal size distribution of QDs with small QDs and large islands is observed and the large InAs islands can be hardly suppressed. When the PALE mode was used, the results show that the InAs QDs with uniform size can be achieved and large InAs islands can be completely suppressed with optimized growth parameters.

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1. Introduction

Self-assembled quantum dots (QDs) have attracted great attention in optoelectronic device applications due to their superior electronic and optical properties such as low threshold currents, good temperature stability and high-speed modulation [1, 2]. To realize all these high performance, it is essential to grow uniform QDs in both size and spatial distribution since the electronic properties of QDs are determined in part by their size, shape and distribution. For InAs QDs, though highly uniform QDs have been obtained by the conventional continuous growth, periodic AsH_3 interruption (PAI) technique and migration-enhanced epitaxy (MEE) [3-5] on exact GaAs substrate, it is still a big challenge to control the position of InAs QDs. Patterned GaAs substrate and vicinal GaAs substrates have been used to achieve the position control. For the former, the processing of patterned GaAs substrate usually requires electron beam lithography and wet chemical etching, furthermore, defects are usually introduced. For the latter, the method is much more straightforward and high density of InAs QDs are found to selectively grow on the step edges of multiatomic steps, however, a bimodal size distribution of QDs with small QDs and large islands is usually observed due to the change of surface migration of adatoms on the multiatomic steps [6-12].

In this work, two different growth methods including conventional continuous growth method and pulsed atomic layer epitaxy (PALE) growth method were used to grow InAs QDs on vicinal GaAs substrate via Straski-Krastanow (S-K) mode. By systematically varying the growth conditions of these two methods, a new approach to achieve uniform InAs QDs on vicinal GaAs (100) substrate has been proposed and demonstrated. The effect of GaAs buffer layer on the InAs QDs was also studied.

2. Experimental procedure

The samples were grown on 2°-off GaAs (1 0 0) substrate in Thomas Swan vertical low-pressure metal organic chemical vapor deposition (MOCVD). Trimethylgallium (TMGa), trimethylindium (TMIn), and AsH_3 were used as source materials. And H_2 was used as carrier gas. The reactor pressure was maintained at 150 mbar during growth. The GaAs substrate was heated to 700°C for oxide desorption. After 10 minutes, the temperature was decreased to grow a 200nm-thick GaAs buffer layer. And the temperature was further decreased to 510°C for InAs QD growth with V/III ratio of 5. After 24s growth interruption at the same temperature, the sample was cooled down to room temperature rapidly under AsH_3 flow. To study the effect of GaAs buffer layer on the InAs QDs, the growth temperature of GaAs buffer layer was varied from 550°C to 650°C with a V/III ratio of 30. For the InAs QDs growth, two growth methods, conventional continuous growth and PALE growth, were used. As far as the PALE growth was concerned, two types of growth mode were tried. The detailed growth schematic is shown in Fig. 1.

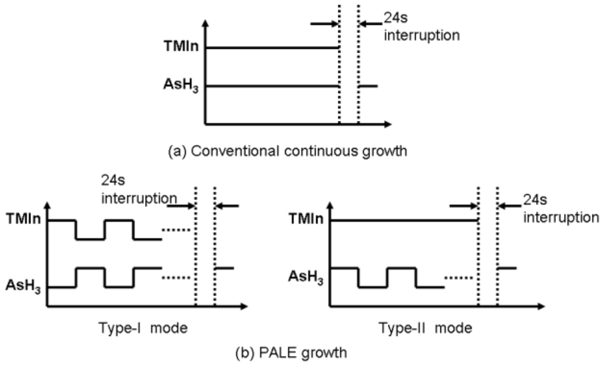

Fig. 1. Growth schematic of InAs QDs with different growth methods: (a) conventional continuous growth; (b) PALE growth with type-I and type-II mode.

3. Results and discussion

Fig. 2 shows the AFM images of the InAs QDs on GaAs buffer grown at temperature 550°C, 600°C and 650°C respectively. Here, InAs QDs were grown for 8 seconds with the thickness around 2ML using the conventional continuous growth method. The AFM images reveal that the InAs QDs density changes with the GaAs buffer layer growth temperature and bimodal size distribution exists for all three samples. The respective number of QDs and large islands is also shown in Fig. 2. It is obvious that the InAs QDs on GaAs buffer layer grown at 600°C shows the best uniformity with less large InAs islands.

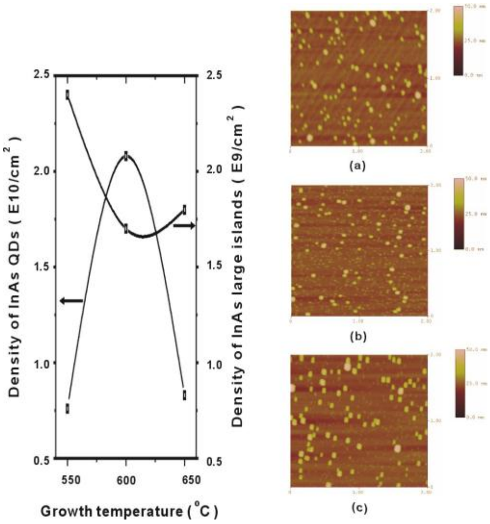

Fig. 2. InAs QDs on GaAs buffer layer grown at (a) 650°C; (b) 600°C; (c) 550°C.

To clarify the described difference of InAs QDs on GaAs buffer layer grown at different temperatures, the surface morphology of these GaAs buffer layers were studied. We noticed that very smooth surface with fairly faint step features were obtained at 550°C, while multiatomic step features presented at 600°C and 650°C due to the Ga adatoms migration on vicinal GaAs (1 0 0) surface [13], though the atomic step features can barely seen in Fig. 2(b). Furthermore, AFM images reveal that the step height and terrace width are larger for the GaAs layer grown at 650°C. This behavior is consistent with the observation of Shinohara [14]. Naturally, the above described difference of InAs QDs can be related with the appearance of the multiatomic step features of GaAs buffers. On one side, the step edges as energetically favorable nucleation sites can facilitate the formation of large amount of small InAs QDs. On the other side, the step kinks usually serving as energy barriers can prohibit the migration of the adatoms and precipitate the formation of large InAs islands, finally leading to the bimodal distribution of dot sizes. For the GaAs buffer layer grown at 650°C has the highest step and thus more energy are needed for the adatoms to transform from one terrace to another, therefore, the size difference between the large dots and small dots is most distinct in these three samples. With smaller step height and terrace width, as the GaAs buffer grown at 600°C, the favorable nucleation sites increase and the necessary energy for the transformation of adatoms is less, therefore, the density of InAs QDs are higher while the size distribution gets more uniform. However, for the GaAs buffer grown at 550°C, the surface is getting similar to the exact GaAs buffer layer, the nucleation sites become random, and under this InAs growth condition, large islands increase.

In order to obtain ordered InAs QDs with uniform size on vicinal GaAs(100) substrate, experiments with systematically varied growth parameters such as temperature, V/III ratio have been conducted, but the results show that it is extremely difficult to completely avoid the bimodal size distribution of InAs QDs due to the above discussed multiatomic step effect. We realized that one possible method to obtain uniform and ordered InAs QDs is improving the migration and diffusion length of In adatoms via the PALE growth, which has been used as an effective way to enhance the migration of Al adatoms during the growth of AlN epilayer [15].

For the PALE growth of InAs QDs, the GaAs buffer layer was grown at 600°C and the type-I and type-II PALE growth modes (Fig. 1) were used for comparison. As the schematic shows, for type-I, TMIn and AsH_3 were alternatively supplied in 1s duration respectively (2s/cycle) and the total cycles were changed to get the best results. Fig. 3 shows the AFM image of the representative result via type-I mode. Very large islands and no obvious QDs can be clearly observed, which may be due to the interruption of AsH_3 resulting in the insufficient nucleation sites. It can be concluded that using type-I mode in our system may result in ineffective nuclei and thus no fully coverage of InAs QDs.

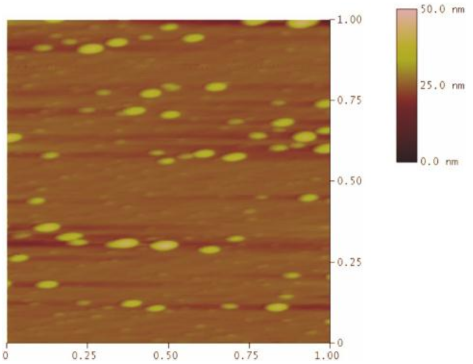

Fig. 3. AFM image of InAs QDs via PALE growth type-I mode.

With regard to the type-II mode, TMIn was supplied during the entire InAs QD growth process, while AsH_3 was supplied intermittently. To get the uniform InAs QDs, we systematically varied the close and open intervals of AsH_3 supply. Table 1 lists the experimental details of AsH_3 supply and the total growth time of the InAs QDs. The AFM images of these four samples are shown in Fig. 4. The AFM image of sample A exhibits ordered InAs QDs but still with bimodal size distribution. Comparing sample A with that in Fig. 2(b), effective nucleation sites increase by the enhancement of the migration of In adatoms. When the close intervals of AsH_3 supply was increased from 1s (sample A) to 2s (sample B), the large islands get larger and total density decreases, indicating that the longer close intervals facilitate the coalesce of the InAs QDs, which is different from the formation of large islands via conventional continuous growth. Taking the coverage effect into account for sample C and D, we set the close interval as 1s but reduce the open intervals by periods. The results show that in sample C, the large InAs islands are significantly suppressed and the QDs are well aligned along the multiatomic steps. The further decrease of the growth time results in the InAs QDs with uniform size (sample D). However, we also notice that the QDs are not well aligned. This can be attributed to the relatively long interruption time, 24s, for the relatively less InAs coverage, which always means smaller QDs. Since the small QDs are not in thermal equilibrium and tend to develop into energetically favorable large QDs, the original smaller QDs in sample D are more likely to form the large QDs during long interruption time and thus lose the order.

Table 1. Experimental details and results of AsH_3 supply and the total growth time of the InAs QDs via type-II mode.

Sample	Total growth time (s)	Period #1		Period #2		Period #3		InAs coverage (ML)	InAs QDs density ($/cm^2$)	InAs large islands density ($/cm^2$)
		Open interval (s)	Close interval (s)	Open interval (s)	Close interval (s)	Open interval (s)	Close interval (s)			
A	9	2	1	2	1	2	1	1.5	$5x10^{10}$	$5x10^{9}$
B	12	2	2	2	2	2	2	1.5	$1.5x10^{10}$	$2.3x10^{9}$
C	7.5	2	1	1.8	1	0.7	1	1.2	$3.6x10^{10}$	$7x10^{8}$
D	5.8	2	1	1.8	1	/	/	1.0	$1.3x10^{10}$	0

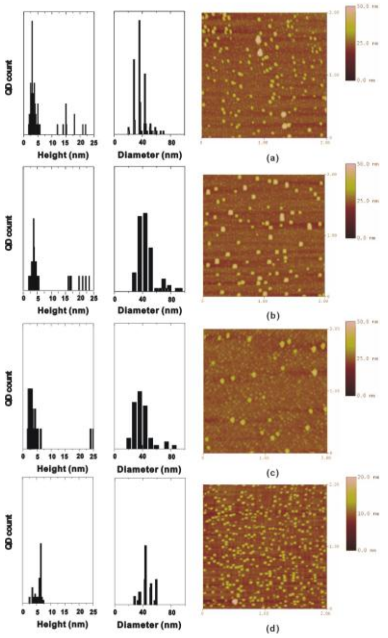

Fig. 4. AFM images and size distribution of InAs QDs of samples A-D respectively.

4. Conclusion

In summary, we have grown InAs QDs on 2°-off GaAs (1 0 0) substrate in vertical MOCVD reactor via the conventional continuous growth and PALE growth. As what the other works state, it is extremely hard to grow InAs QDs with uniform size on the vicinal GaAs substrate due to the multiatomic step effect. In our work, we found that because of the atomic step features of the GaAs buffer layer on vicinal GaAs substrate can be manipulated by growth temperature, the density and distribution of subsequently grown InAs QDs can be changed. By optimizing the growth conditions of the PALE growth method, we have also demonstrated that the large InAs islands can be completely suppressed, and InAs QDs with uniform size can be achieved due to the enhancement of the migration of the adatoms. Our investigation may suggest an easier and straightforward way to control the size and position of the InAs QDs.

Acknowledgments

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