Controlled growth mechanism of ring-like In(Ga)As quantum Dot pairs on GaAs ring-ring-disk nanostructures templates

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Abstract
The ring-like In(Ga)As quantum dot pairs (QDPs) is prepared on GaAs ring-ring-disk (R-R-D) nanostructures template by droplet epitaxy (DE) method. The surface morphology of GaAs samples are characterized with Scanning Tunneling Microscope (STM), and studying local controlled growth mechanism of quantum dots (QDs) on GaAs R-R-D nanostructures. STM images show that In(Ga)As QDPs self-assembled a kind of complicated structure with one quantum dot (QD) pair in the center of nanostructures, other QDPs appearing both double rings and disk areas of original GaAs R-R-D nanostructures distributed like a exquisite ring. These In(Ga)As QDPs are uniformly arranged along the [110] direction of GaAs(001) surface. Combining Stranski-Krastanov (S-K) growth mode and the tensile effect of strain field in the [110] direction on sample surface, the nucleation position and distribution shape of ring-like In(Ga)As QDPs is locally controllable. It is found that high uniformly QDPs can be locally controlled by the original GaAs R-R-D nanostructures templates. These results can provide certain guiding significance for the controllable growth of QD by DE method.

1. Introduction

Low-dimensional semiconductor nanostructures always have attracted great attention due to their wide application and infinite potential prospect in many fields of quantum communication [1], quantum computing [2], quantum lasers [3], hybrid quantum networks [4], ion detection [5] and cell imaging [6]. The most commonly studied low-dimensional semiconductor nanostructures mainly include quantum well, quantum ring and QD. In particular, QD has become an ideal choice for the design of a new generation of optoelectronic devices and development of information technology because of their unique properties such as quantum size effect [7], quantum tunneling effect [8], surface effect [9] and Coulomb blockade effect [10]. When the morphology of QD changes, its properties also changes. For example, the emission spectrum of QD can be controlled by changing QD size [11]. Besides, the controlled growth of QD was always a hot topic in the current research area of low-dimensional nanostructures material.

In order to achieve the controllable growth of QD, many methods have been tried to accomplish this goal [12–17]. Compared with above these technologies, droplet epitaxy (DE) method has become one of the current mainstream technologies because it is not limited by the lattice mismatch between substrate material and epitaxial deposition material [18]. In the process of preparing QD by DE, changing experimental conditions will obtain different morphological QDs [19–22]. For example, it is reported that temperature and annealing have an important effect on the QD growth [23]. Changing annealing process will make QDs evolve into different hetero-structures [24]. Based on molecular beam epitaxy (MBE) technology, the structure, composition, strain and optical properties of QD can be designed [25]. The use of DE technology can obtain highly symmetrical
quantum dots [26]. In addition, it was confirmed that the controlled growth of quantum dots can be achieved by changing experimental parameters during the growth process of QD, which hugely further expands its development in optoelectronic devices [27–33]. Although DE method has been proved to be very successful in the controlled growth of QD, there is still some works worth to do.

In this study, we reported on the use of GaAs R-R-D nanostructures template formed by DE for In(Ga)As QDPs based on the S-K growth mode. Here we established a step density model to explain the nucleation sequence of deposition materials on GaAs R-R-D nanostructures, and introduced a strain field in the [110] direction of GaAs substrate to illustrate the controlled growth mechanism of In(Ga)As QDPs.

2. Experimental details

All samples in our study were GaAs(001) substrates with Si doped concentration of N_D = 1.49 × 10^{18} \text{cm}^{-3}. Sample surfaces were monitored with a reflection high energy electron diffraction (RHEED) system, and MBE system was equipped with a highly accurate solid-source valve. Before experiment, the beam equivalent pressure (BEP) of each source was measured, and substrate temperature was precisely calibrated. The oxide on each sample was beforehand desorbed at 580 °C for 10 min, setting Ga source temperature of 1060 °C, As BEP of 1.06 × 10^{-3} \text{Pa}. A 500 nm GaAs buffer was grown by the deposition rate of 0.3 ML s^{-1}. Then a 1 h in situ annealing took place, and ensuring sample surface to become atomically flat. Substrate temperature was gradually decreased to 540 °C. Closing As valve and Keeping Ga source temperature constant, substrate temperature was slowly decreased to 270 °C, 10 Ml Ga were deposited on the sample surface by the rate of 0.3 ML s^{-1}. Then opening As valve, its value was regulated with As BEP of 6.5 × 10^{-4} \text{Pa}. As BEP time was 20 s and immediately closed As valve. Next, substrate temperature was increased to 330 °C, opening As valve, As BEP value was the same as the previous step, keep it for 20 s, again closed it. Finally, the substrate temperature was again increased to 370 °C, repeating above As pressure crystallization step, but the time was maintained for 20 min to assure that Ga droplets were fully crystallized. After each sample was quenched to room temperature, all samples were transferred to ultra-high vacuum STM to characterize the surface topography.

Substrate temperature was gradually increased to 490 °C and In source temperature of 850 °C. 2.0 Ml In was deposited on GaAs R-R-D nanostructures template by the rate of 0.1 ML s^{-1}. As BEP was adjusted to 9.0 × 10^{-4} \text{Pa} for 3 min. Surface topography of each sample was then imaged by STM.

3. Results and discussion

3.1. Step density analysis of each component in ring-like GaAs R-R-D nanostructures

GaAs R-R-D nanostructures were formed on GaAs substrate surface by depositing 10.0 Ml Ga. Figure 1 shows a set of STM images of GaAs R-R-D nanostructures template. Figure 1(a) is a STM image of GaAs R-R-D nanostructures template obtained in our experiment. Although some GaAs nanostructures on the sample surface are stacked on top of each other, they still maintain R-R-D shape. Figure 1(b) and figure 1(c) are two-dimensional (2D) and three-dimensional (3D) STM images of a single GaAs R-R-D nanostructure, respectively. This kind of R-R-D structure was considered that the result of Ga droplet evolution after three times of As pressure crystallization [34]. In detail, Ga was deposited on GaAs substrate surface then to form Ga droplets. Firstly, Ga deposited on GaAs samples to from a GaAs thin film. Owing to system minimum surface energy, this film began to break into many small droplets with unstable energy state, and then several neighbor small droplets were aggregated into bigger droplets with stable energy state. Each droplet undergoes As BEP crystallization to form one ring structure, after three times As BEP crystallization to form three rings. Since the third crystallization time was longer, a disk structure is obtained after the third crystallization. Obviously, we have observed that 3D GaAs R-R-D nanostructure in figure 1(c) is damaged, which is mainly a result of the anisotropic diffusion of Ga on the substrate surface during the preparing of GaAs R-R-D template.

The height and diameter of GaAs nanostructures in figure 1(a) are counted, and figure 2(a) is the height and diameter statistics of GaAs R-R-D nanostructures. In figure 2(a), the average depth of nanohole is about 1.55 nm, the average height of double rings structures is about 6.5nm, the height of disc structure is between 2.0 ~ 3.0 nm, and the valley height between double rings is about 4.0nm. The average diameters of nanohole, double rings, and disc structure are approximately 20.0 nm, 50.0 nm, and 100.0 nm, respectively. Its surface density is 4 × 10^{10} \text{cm}^{-2}. Figure 2(b) is the linear analysis of a single GaAs R-R-D nanostructures in the [110] direction, and figure 2(c) is micro-element model of step density analysis of each component in GaAs R-R-D nanostructures.

As mentioned above, deposition material will preferentially choose to nucleate and grow in high step density areas on GaAs nanostructures [35]. In order to analyze the nucleation sequence of each area on GaAs R-R-D nanostructures, it is necessary to know the step density of each area on this nanostructure. It defines that the...
change in the number of steps per unit length in the radial direction as the step density in this area. As shown in figure 2(c), the ring is made up of many steps. In the left half of GaAs ring, take a micro-element area on GaAs R-R-D nanostructures. Describe the number of steps in this area, and the radial length is in nm. The density is in \( \text{nm}^{-1} \). The radial step density formula (1) in the micro-element region is

\[
\rho = \frac{\partial N}{\partial x} \tag{1}
\]

Designate these steps height on GaAs R-R-D nanostructures is equal to 1 nm per unit length. It can be inferred that the number of steps in these regions of radial length is equal to the ring height values. As shown in figure 2(c), when the micro-element area is infinitely small, the radial step density in the micro-element area on
GaAs R-R-D nanostructure can be represented by the area slope, that is, the slope is

\[ k = \frac{dh}{dx} \]

\( dh \) represents the height of micro-element area, \( dx \) represents the radial length of micro-element region. Above formula (2) only discusses step density. We want to know the number of steps, it must consider the ring symmetry. From above conclusions, the larger slope of area represents the greater step density on GaAs R-R-D nanostructures, deposited material will preferentially nucleate in this area of the greatest step density. Combining formulas (1) and (2), for GaAs R-R-D nanostructures in our experiment, the slopes order of each region should be arranged from large to small as nanohole, double rings, disc structure and substrate surface flat area. Therefore, the nucleation sequence of deposited material on GaAs R-R-D nanostructures is nanoholes, double rings, disc structure and substrate surface flat area.

### 3.2. Locally controllable growth of In(Ga)As R-R-D QDPs

InAs was deposited on GaAs R-R-D nanostructures template. Figure 3 shows a set of STM images of novel In(Ga)As QDPs obtained with InAs deposition amount of 2.0ML. As shown in figure 3(a), a kind of new QD structure appears on GaAs(001) surface. There is one QD pair in this new structure center, and many QD pairs in the nearby area are uniformly distributed around the primeval same center. Combining the 2D and 3D STM images of a single new In(Ga)As QDPs in figures 3(b) and (c), figure 3(c) clearly shows that it is a composite structure with a clear pair of QD in the center. Approximately 16 QDPs are in a circular ring shape and are evenly distributed on the edge of original nanohole. QDPs are also distributed in the area outside the 16 QDPs, and their shape is also roughly ring-shaped. Therefore, this paper named this composite structure as ring-like In(Ga)As QDPs. Surface density of this composite structure is \( 4 \times 10^{10} \text{ cm}^{-2} \), which shows that the number of composite structures are exactly the same as the number of primitive GaAs R-R-D nanostructures. Observing closely at figure 3(c), these QDPs in this composite structure are generally arranged along the [1 1 0] direction on GaAs (001) surface. The formation of the ring-like In(Ga)As QDPs will be explained in the following sub-regions, and the formation mechanism of arrangement direction of QDPs in the composite structure will be explained.

### 3.3. Local controllable growth of ring-like In(Ga)As QDPs

According to above conclusions, the nucleation sequence of InAs on the GaAs R-R-D nanostructures template is from nanohole, double rings to disk area. In the nanohole region of GaAs R-R-D nanostructures, the appearance of a single In(Ga)As QDPs was attributed to that InAs materials were deposited in the nanohole with nucleation growth by the S-K growth mode. Specifically, as InAs materials entering nanohole, those materials first gradually filled GaAs nanoholes on substrate surface by film growth mode, then GaAs nanoholes gradually become shallower. When InAs deposition amount increases to 1.7ML, InAs films have reached a critical thickness in those holes, at this time, the growth mode of InAs deposition materials was transformed from thermodynamic

![Image](image.png)
equilibrium to non-equilibrium. InAs epitaxial layer began to break, InAs growth mode changed from layered to island, In\(_\text{Ga}_\)As QDPs began to appear on the sample surface, as InAs material continues to be deposited on GaAs (001) surface, and two In\(_\text{Ga}_\)As QDPs appeared in the center of initial GaAs R-R-D nanostructures, then experiment samples were annealed, the bottom of two In\(_\text{Ga}_\)As QDPs are connected by the material flowing from In\(_\text{Ga}_\)As QDPs top surface, finally, a single In\(_\text{Ga}_\)As QDP was formed [36].

For ring-like In\(_\text{Ga}_\)As QDPs appearing in original double rings position of GaAs R-R-D nanostructures, figure 4 was used to describe the formation process of In\(_\text{Ga}_\)As QDPs appearing at this position. When InAs was deposited on GaAs double rings surface, with InAs deposition amount gradually increasing, in figure 4(a), the height of In\(_\text{Ga}_\)As double rings slowly increase by the layered growth mode. As InAs deposition amount measuring up a critical value, as shown in figure 4(b), just observing these QDs on each ring, its shape is very similar to a ring chain. According to Z. M. Wang principle in preparing the In\(_\text{Ga}_\)As QD chains [37], strain can drive deposited materials transport. When the amount of InAs deposited materials continues to increase, InAs deposition amount increases to 1.7ML, InAs have reached a critical thickness on double rings area, these In\(_\text{Ga}_\)As rings begin to break into many ring segments under the influence of buried layer effect of bottom materials in the original GaAs rings of GaAs R-R-D nanostructures. Continuously to increase the amount of InAs deposited materials, it was considered that InAs materials have the same growth rate in each direction. A perfect center-symmetric dot-pair structure was formed under this condition, and its shape is similar to a ring. However, the strain effect caused by the lattice mismatch between InAs deposited material and GaAs substrate material forces broken ring segments to link together. Eventually this strain effect promotes the distribution of In\(_\text{Ga}_\)As QDs in the [110] direction of GaAs substrate. For the disk structure, the growth trend and distribution shape of QDs is the same as double rings. It makes In\(_\text{Ga}_\)As QDs to grow faster in the dominant growth direction, and these In\(_\text{Ga}_\)As QDs become narrow and long along the [110] direction of GaAs substrate. Correspondingly the shape of perpendicular direction of In\(_\text{Ga}_\)As QDs looks like shorter. As shown in figure 4(c), these QDs grow faster in the [1\(-\)10] direction so that the bottoms of In\(_\text{Ga}_\)As QDs formed in the double rings are connected together in the [1\(-\)10] direction. In the end, In\(_\text{Ga}_\)As QDPs appeared in original double rings of GaAs R-R-D nanostructures.

The growth mechanism of In\(_\text{Ga}_\)As QDs appearing on the disk in the original GaAs R-R-D nanostructures is based on the combined effect between the S-K growth mode and annealing. Specifically, InAs materials was first deposited on the surface of the disk by the layered growth mode, as the amount of deposited material

![Figure 4](image_url). The formation process of In\(_\text{Ga}_\)As QDPs at the position of double rings, (a) InAs is formed on the surface of the substrate according to the S-K growth mode of In\(_\text{Ga}_\)As QDPs model, (b) In\(_\text{Ga}_\)As QDPs under ideal conditions, (c) In\(_\text{Ga}_\)As QDPs under strain field conditions.
continues to increase, it is grown by the island-shaped growth mode. After annealing it, the nearest two QDs are connected by In(Ga)As materials flowing from the top to form a QDP [37].

3.4. The controllable growth mechanism of In(Ga)As QDPs on the [1 1 0] direction of GaAs (001) surface

Figure 3(c) shows that all In(Ga)As QDPs in this kind of composite structure arranged along the [1 1 0] direction of GaAs (001) surface. This reason is mainly that the growth rate of InAs along the [1 1 0] direction of GaAs substrate during the nucleation growth process is the fastest than that of other directions. For InAs materials growth rate in the [1 1 0] direction is the best than other directions, this effect was regarded as a tensile strain field in this direction. The strain field plays a role of stretching those In(Ga)As QDs and further promoting the connection of these QDs in the growth process. That is, as the amount of InAs deposition increases, In(Ga)As QDs change from a circular shape to a long and narrow island under the action of this strain field, with annealing it, and the adjacent two In(Ga)As QDs are connected by the In(Ga)As materials flowing from the QD top and formed a dot pair. However, the [1 1 0] direction of these dot pairs is controlled by the arrangement of the strain field along the [1 1 0] direction. Therefore, In(Ga)As QDPs on the GaAs surface unanimous point to the [1 1 0] direction eventually.

During the formation of ring-like In(Ga)As QDPs, the step density of each part of GaAs R-R-D nanostructures is proportional to the slope of every region. On GaAs R-R-D template, nanohole step density is the highest, double rings part is the second, and disc area is smaller. InAs will gradually nucleate and grow on GaAs R-R-D nanostructures in the order of nanoholes, double rings, disks, and flat areas on GaAs(001) surface. The surface density of ring-like In(Ga)As QDPs on the sample is equal to that of GaAs R-R-D nanostructures on the template. In(Ga)As QDPs in the composite structure is ring-shaped, it was mainly because In(Ga)As QDPs were localized growth by the ring-like GaAs R-R-D nanostructures in GaAs template. The arrangement of these QDPs along the [1 1 0] direction of GaAs sample surface is the same as the strain field in the [1 1 0] direction GaAs sample surface. In summary, this paper confirms GaAs R-R-D nanostructures template obtained by the DE method can achieve the local controllable growth of surface density, nucleation position, distribution morphology and other apparent characteristics of QD.

4. Conclusion

In conclusion, ring-like In(Ga)As QDPs were formed on GaAs R-R-D nanostructures template with InAs deposited on GaAs sample surface in the MBE system based on DE method. The nucleation and growth sequence of InAs is controlled by the number of step density of each component in GaAs R-R-D nanostructures. The distribution shape of QDs is also controlled by the nanostructure shape of original template. Dominant growth of deposition material in the [1 1 0] direction determines the final alignment of the In(Ga)As QDs on GaAs substrate. These In(Ga)As QDPs point to the [1 1 0] direction of GaAs sample surface, it was mainly because the strain field in the [1 1 0] direction during the nucleation process controls In(Ga)As QDs arrangement direction. If this strain field is overcome, the distribution shape of In(Ga)As QDs will become symmetrical with the circle center. The above research results can provide a certain experimental and theoretical basis for the controlled growth and manufacture optoelectronic devices of QDs.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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