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Kinetic calculation analysis of Ga deposition on the morphology evolution of GaAs quantum ring

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Abstract

GaAs Quantum Ring (QR) was gained on GaAs (001) by Droplet Epitaxy (DE), and the microscopic morphology of the GaAs samples was observed by Scanning Tunnel Microscope (STM). The kinetic model of Local Droplet Etching (LDE) was mainly used to study the influence of Ga deposition on the morphology evolution from Ga droplets to GaAs QR. Comparing experimental data with the theoretical value, it can be seen that the increase of Ga deposition will cause the reduction of surface density of Ga droplets, but the volume, height and diameter of Ga droplets will increase. Geometric dimension of GaAs QR increased also with the increase of Ga deposition. In addition, it found that the rate at which the substrate was etched was affected by Ga deposition and As pressure from experiment. The more Ga deposition, the deeper GaAs nano hole. However, GaAs nano hole became shallower under high As pressure. These results are consistent with theoretical calculation analysis. Under high substrate temperature, GaAs double rings finally evolved into a single ring. Above all results have certain guiding significance for the preparation of patterned GaAs substrates and the controlled growth of GaAs QR morphology.

1. Introduction

Low-dimensional semiconductor nanostructures have always attracted increased attention owing to its favorable prospects in the future quantum field. For example, for integrated quantum communications [1], quantum computing [2], quantum optoelectronic devices [3], hybrid quantum networks [4]. In order to develop up-to-date quantum devices, it is essential in the controlled growth of surface morphology of these structures. Currently, the controlled growth of low-dimensional semiconductor nanostructures shape had been accomplished by DE. For example, GaAs/AlGaAs Quantum Dots (QDs) were obtained by high temperature DE, and these QDs show extremely high symmetry [5]. Moreover, the surface shape of QDs was designed by growth kinetics [6]. It reported that the crystallization dynamics of Ga droplets was used to study Ga surface diffusion on GaAs (001) substrate [7], termal annealing has an important influence on InAs QDs formation based on DE [8]. High symmetric QDs were also grown by DE method, and these QDs can be used to apply in telecommunication wavelengths on InP(111) [9]. Quantum Ring (QR) has been favored because of its unique optoelectronic properties and potential application value in photoelectric devices, detectors and other fields [10–12]. In particular, quantum coherence of QR [13] and optical Aharonov–Bohm (AB) effect [14] indicated that QR was a promising nanomaterial in the development of new generation optoelectronic devices. D. Granados and A. Lorke found that the energy band, oscillation intensity, spin polarization and magnetism of QR were controlled by its surface morphology [15, 16], which means that the performance of QR optoelectronic devices will be improved with controlling the morphology growth of QR. There were many kinds of QR preparation methods nowadays [17–19]. Using Molecular Beam Epitaxial (MBE) growth technology, QDs can evolve into QR [20] after a continuous annealing process. The scale of QR was nanometer. Due to its unique rotational symmetry, it
exhibits many interesting properties, such as unusual physical properties and AB effect [14]. Compared with above growth methods, DE is not only a simple and easy method to prepare high-quality QR, but also QR structures prepared by DE technology is not affected by the mismatch of epitaxial material and substrate lattice constant [21]. At present, DE has been used to prepare QR structures with different surface morphology such as single QR [22] and double QR [23]. Although it was confirmed that the height and ring width of Al(Ga)As nanostructures will increase with the increase of Ga deposition, and it also explains that surface density of Ga droplets decreases with increasing Ga deposition, but there is no theoretical calculation and analysis on this conclusion [24].

In the kinetic model of Local Droplet Etching (LDE) proposed by C Heyn [25], when the Ga droplet is not annealed, it is only composed of pure Ga atoms, and its shape is approximated by a segment of a sphere. This model also proposed a calculation equation describing the density and geometric size of Ga droplets. It is also known that GaAs on the substrate surface were activated and then decomposed into Ga and As from the kinetic model of LDE when Ga droplets etched GaAs substrate. As atoms desorbed from GaAs surface by As atoms concentration gradient between the GaAs substrate and Ga droplets and entered into Ga droplets, and some of these As atoms diffused to the edge of Ga droplets and combined with Ga atoms to form GaAs wall. In addition, A Stemmann and C Heyn used LDE method to prepare QR structures on the surface of GaAs and AlGaAs [26], and pointed out that nanoholes density decreased with increasing substrate temperature. As material deposition rate increasing, the outer diameter of a single ring increased accordingly. However, they neither discussed the pore size of QR, nor clearly indicated the influence of Ga deposition on the evolution of QR morphology. In particular, there are few reports on the kinetic calculation and analysis of Ga deposition on the evolution of GaAs QR morphology, so this work is worthy to research and explore.

This task reported GaAs QR was grown in the MBE system. The volume, height and diameter of Ga droplets were calculated through the kinetic model of LDE, and studied the pore size of QR and the nanoholes depth. Average diffusion distance of Ga and As atoms was described by Einstein’s equation, and analyzing QR width. In addition, experimental data onto various components in QR are compared with their theoretical calculation values, and the effect of Ga deposition on the evolution of GaAs QR morphology is mainly studied. Through in-depth discussion on the kinetic model of LDE, Ga deposition and As pressure was studied when analyzing substrate etching rate respectively. Relevant conclusions can not only verify kinetic theory of LDE, but also help to understand the microscopic mechanism of GaAs QR morphology evolution more deeply. It may help to understand the controllable growth of QR morphology.

### 2. Experimental procedure

In this experiment, the sample is a GaAs (001) substrate with Si doping concentration ($N_D = 1.49 \times 10^{18} \text{cm}^{-3}$). Each sample was grown in the MBE system equipped with ultra-high vacuum growth chamber (vacuum background $2.0 \times 10^{-10} \text{Pa}$). Before the experiment, Beam Equivalent Pressure (BEP) of each source was calibrated respectively, and substrate temperature ($T_{\text{sub}}$) was also calibrated by Zhou [27] reporting the calibration method of $T_{\text{sub}}$. Each sample was deoxidized at 580 °C for 10 min. Setting Ga sources temperature of 1080 °C, As BEP of $1.6 \times 10^{-3} \text{Pa}$, and $T_{\text{sub}}$ of 550 °C, a 500 nm GaAs buffer layer was grown at a rate of 0.32 ML s$^{-1}$, and then in situ annealing for 1 h ensured the sample surface to become atomic level smooth. In order to determine the critical formation of Ga droplets on the sample surface and calculate the height, volume and diameter of Ga droplets, keeping substrate temperature at 550 °C, 0.65 ML and 1.30 ML Ga were deposited on GaAs samples surface on which GaAs buffer layer has been grown, respectively. After quenching, these samples were sent to STM for characterization.

In order to explore the kinetic effect of Ga deposition on the controllable growth of GaAs QR morphology, $T_{\text{sub}}$ was reduced to 390 °C. The samples were divided into three experimental groups. These samples in each experimental group were grown with a GaAs buffer layer and made samples surface to become atomic level smooth, and then 5.00 ML, 10.00 ML, and 10.00 ML Ga were deposited on GaAs samples surface, respectively. Each sample of experimental groups was crystallized by the following table 1. To study the growth morphology of QRs at low substrate temperature and low As pressure, we set the crystallization conditions as references [28, 29]. Therefore, experimental parameters were setting as follows: the crystallization time of 20 s, As BEP of $4.0 \times 10^{-3} \text{Pa}$. Here we increase the substrate temperature in the second step to overcome the barrier effect generated by the first ring. The morphology of QRs was observed by changing Ga deposition.

All samples need to be quickly quenched at room temperature, and then immediately sent to STM for observation.
3. Results and discussion

a. Kinetic calculation of volume, height and diameter of Ga droplets
As shown in figure 1 (a), when Ga deposition was at 0.65 ML, its corresponding STM image showed that there are many multi-layers long island structures on GaAs sample surface, these structures were formed because of the migration and bonding of Ga atoms on sample surface in a high-energy state at 550 °C. In figure 1 (b), Ga deposition is at 1.30 ML, clear and independent Ga droplets appeared on GaAs sample surface. According to experimental statistics, the average height of Ga droplets $H_{\text{exp}}$ is about 5.00 nm, and the average diameter $D_{\text{exp}}$ is about 36.00 nm. Surface density of Ga droplets is about $1.90 \times 10^9 \text{cm}^{-2}$.

During the formation of Ga droplets, in order to reduce the energy of Ga droplets system, many small Ga droplets will be aggregate into a stable and large Ga droplets nearby by Wulf surface energy minimization principle and Ostwald maturation theory. The shape of large Ga droplets is similar to a segment of a sphere, which is consistent with the kinetic model of LDE proposed by C Heny [25]. Equation (1) was used to calculate the average number of Ga atoms in Ga droplets.

$$N_{\text{Ga}} = \frac{(Q - 1)}{(n/n_0)}$$

Equation (1)

$N_{\text{Ga}}$ represents the average number of Ga atoms. $Q$ represents Ga deposition. $n$ represents the surface density of Ga droplets, its unit is $\text{cm}^{-2}$. $n_0 = 6.25 \times 10^{14} \text{cm}^{-2}$ [25]. $Q = 1.30$ ML and $n = 1.90 \times 10^9 \text{cm}^{-2}$.

![Figure 1. STM images of Ga deposition in (a) 0.65 ML, (b) 1.30 ML, respectively.](image)

![Figure 2. The kinetic model of GaAs substrate etched by Ga droplet.](image)

### Table 1. Two steps of crystallization treatment in each experiment sample.

| Ga deposition/ML | Time/s | As BEP/Pa | $T_{\text{sub}}$/°C | The first step | The second step |
|-----------------|--------|-----------|---------------------|----------------|----------------|
|                 |        |           |                     |                |                |
| 5.00            | 20     | $4.0 \times 10^{-4}$ | 280                 |                |                |
| 10.00           | 20     | $4.0 \times 10^{-4}$ | 280                 |                |                |
| 10.00           | 20     | $4.0 \times 10^{-4}$ | 340                 |                |                |

![Table 1](image)
were substituted into equation (1), \( N_{\text{Ga}} \approx 9.9 \times 10^4 \). According to the kinetic model of LDE, Ga droplets were simply made up of pure Ga atoms without annealing. The volume of Ga droplet was approximately equal to the product of the average number of Ga atoms in a droplet and the volume of single Ga atom. The volume of a single Ga atom is about \( 1.96 \times 10^{-29} \text{m}^3 \). The volume of Ga droplet was described by equation (2).

\[
V_{\text{Ga,droplet}} = 1.96 \times 10^{-29} N_{\text{Ga}}
\]

Here \( V_{\text{Ga,droplet}} \) represents the volume of Ga droplet, its unit is \( \text{m}^3 \). \( N_{\text{Ga}} \approx 9.9 \times 10^4 \) was substituted into equation (2), \( V_{\text{Ga,droplet}} \approx 1.94 \times 10^{-24} \text{m}^3 \). The height and diameter of Ga droplet were described by equations (3) and (4) [16].

\[
H_{\text{cal,droplet}} = \left( V_{\text{Ga,droplet}} / 13.25 \right)^{1/3}
\]

\[
D_{\text{cal,droplet}} = 2H_{\text{cal,droplet}} / \alpha
\]

Where \( H_{\text{cal,droplet}} \) and \( D_{\text{cal,droplet}} \) represents the theoretical values of height and diameter of Ga droplet, respectively. \( \alpha = 0.35 \) [25]. When Ga deposition is 1.30 Ml, \( H_{\text{cal,droplet}} = 5.24 \text{ nm}, \ D_{\text{cal,droplet}} = 30.00 \text{ nm} \). \( H_{\text{exp}} < H_{\text{cal,droplet}}, \ D_{\text{exp}} > D_{\text{cal,droplet}} \). There was an incredulous error between experimental data and theoretical
calculation value. The reason for this error may be that Ga droplet has started to etch GaAs substrate. We used figure 2 to illustrate this process. As shown in figure 2, the amount of Ga atoms was gradually decreased because Ga droplet continuously etched downwards GaAs substrate, so Ga droplet slowly became flat and its height gradually decreases. On the other hand, part of As atoms desorbed from GaAs substrate and Ga atoms combined to form a GaAs wall at the edge of original Ga droplet [25]. According to equations (1)–(4), it can be seen that surface density of Ga droplets has an important effect on the number of Ga atoms, the height and diameter of Ga droplets. This conclusion has important implications for the templates etched droplets and the controllable growth of QR.

b. The pore size of GaAs QR
According to the kinetic model of LDE, GaAs wall was formed at the edge of Ga droplet. The diameter of Ga droplets was approximately equal to the pore size of GaAs QR in this calculation. The pore size of QR can be described by equation (5).

\[ D_{Ga,QR} = 2.41(V_{Ga,droplet})^{1/3} \]  

\( D_{Ga,QR} \) represents the pore size of GaAs QR. It can be seen that surface densities of GaAs QR are \( 5.75 \times 10^{10} \) cm\(^{-2} \), \( 5.0 \times 10^{10} \) cm\(^{-2} \) and \( 4.75 \times 10^{10} \) cm\(^{-2} \) from figures 3(a)–(c), respectively. The experimental data of average pore size of GaAs QR are 19.26 nm, 28.32 nm and 29.14 nm, respectively. Above surface densities of GaAs QR were substituted into equations (1) and (5), respectively, their corresponding theoretical calculation of pore diameters of GaAs QR were 22.85 nm, 31.36 nm and 31.92 nm, respectively. The experimental data was slightly smaller than theoretical value, the reason for this result was that GaAs wall shrank and grown inward.

c. The depth of nanohole
According to the research conclusion of DE etching nanoholes proposed by zhiming W, Ga droplets etched many nanoholes on GaAs substrate, on the other hand, GaAs wall was formed on the nanohole edge [28].

When the substrate temperature, As pressure and crystallization time are kept unchanged, figure 4 showed the relationship curve of nanohole depth in GaAs QR with Ga deposition. The Ga deposition was 1.0 ML, 5.0 ML and 10.0 ML, respectively, and the average depth of corresponding nanoholes was 0 nm, 1.0 nm, 4.0 nm, respectively. The nanohole depth is 0 at 1.0 ML because the first monolayer Ga was consumed for transforming GaAs surface structure from As-rich to Ga-rich. Based on above conditions, we established the following equation (6) to illustrate the relationship between nanohole depth and Ga deposition.

\[ d_{nanohole} \approx 0.04(Q_{Ga} - 1)^2 \]  

Where \( d_{nanohole} \) represents the nanohole depth, \( Q_{Ga} \) represents Ga deposition. As shown in figure 4, it is known that the nanoholes would become deeper as increasing Ga deposition. According to the kinetic model of LDE, it was concluded that substrate etching rate was controlled by the relative concentration of As atoms in Ga droplets. Equation (7) was used to described the relationship between substrate etching rate and the relative concentration of As atoms.
\[ R_{\text{etch}} = (C_{As, \text{max}} - C_{As})(2k_B T/h) \exp \left[ -\frac{E_g}{(k_B T)} \right] \]  

(7)

The applicable range of equation (7) was that As atoms concentration was less than the maximum solubility [30]. Where \( R_{\text{etch}} \) represents the substrate etching rate, \( C_{As, \text{max}} = 6.1 \times 10^{-16} \exp \left( \frac{T}{K} / 60 \right) \), \( C_{As, \text{max}} \) represents the maximum solubility of As atoms in Ga droplet, \( C_{As} \) represents the instantaneous concentration of As atoms in Ga droplet, \( C_{As} = \frac{N_{As, \text{droplet}}}{(N_{Ga, \text{droplet}} + N_{As, \text{droplet}})} \), and Boltzmann’s constant \( k_B \), the temperature \( T \), Planck’s constant \( h \), the activation energy \( E_g \). According to equation (7), when the temperature was a constant, substrate etching rate was only controlled by the instantaneous concentration of As atoms. Obviously, assuming that the number of As atoms in Ga droplets was a certain value, the increase of the number of Ga atoms caused the substrate etching rate to increase, and nanoholes etched by Ga droplets on GaAs surface will be deeper. On the other hand, the substrate etching rate will decrease with only increasing the number of As atoms. The number of Ga atoms in Ga droplet will increase with increasing Ga deposition from equation (1), so nanohole will become deeper with only increasing Ga deposition. Besides, \( R_{\text{etch}} \) will decrease because As atoms amount in Ga droplets increase in the condition of high As pressure. Nanohole will become shallower under high As pressure. These results play an important role in the production of patterned GaAs substrates and the growth of low-dimensional ordered semiconductor nanostructures.

Compared figures 3(b) and (c), the nanoholes in figure 3(c) become shallower mainly due to high temperature causing deposition material at the top of inner ring near the nanohole edge to flow and fill these nanoholes, and these nanoholes gradually become shallower [31]. It indicates that temperature has an important influence on the nanoholes depth in QR.

d. The ring width of GaAs QR

Assuming that Ga atoms in Ga droplets diffuse in its diffusion direction by one-dimensional random walk theory proposed by Einstein. The average diffusion distance of Ga atoms were described by equation (8).

\[ Z_{avg} = (2\delta t)^{1/2} \]  

(8)

Where \( Z_{avg} \) represents the average diffusion distance of Ga atoms, \( \delta \) represents the surface diffusion coefficient of Ga atoms, its unit is \( \text{cm}^2 \text{ s}^{-1} \). \( t \) was diffusion time, its unit is s. The surface diffusion coefficient of atoms proposed by Nishizawa [32] was described by equation (9).

\[ \delta_{\text{surf}} \approx 7.0 \times 10^{-6} \exp \left[ -\frac{56}{R} \left( \frac{T_m}{T} \right) \right] \]  

(9)

Here \( R \approx 8.314 \text{ J/mol} \), \( T_m \) represents the melting point of a single element, its unit is K. \( T \) represents GaAs substrate temperature, and its unit is also K. Since atoms including Ga and As diffused on GaAs substrate surface, the average surface diffusion coefficient of two kinds of atoms is described by equation (10) [33].

\[ \delta_{\text{surf avg}} = 0.5\delta_{\text{Ga,surf}} + 0.5\delta_{\text{As,surf}} \]  

(10)

The experimental data of average width of double ring in figure 3(a) is about 25.0 nm. When \( T_{sub} \) is 280 °C and 340 °C, respectively. Diffusion time was 40 s. The above data are substituted into equations (8)–(10), the value of \( \delta_{\text{surf avg}} \) is between 9.0 × 10^{-9} and 1.2 × 10^{-8} \text{ cm}^2 \text{ s}^{-1}, and the average diffusion distance of atoms on the sample surface is about 20.0 to 30.0 nm. Other experimental conditions was remained unchanged, the average width of double rings sharply increased to about 80.0 nm with increasing Ga deposition from 5.0 ML to 10.0 ML. This result showed double rings become wider with increasing Ga deposition, it is attributed to big Ga droplets formed by increasing Ga deposition has more initial potential energy, which causes Ga atoms in big Ga droplet to diffuse farther on GaAs sample surface under the same experimental conditions.

e. The height of each ring in GaAs QR

The experimental data of average height of double rings in GaAs QR are showed in figures 3(a1)–(c1). As shown in figure 3(a1), the inner ring is about 3.0 nm, and the outer ring is 2.5 nm. The inner ring of 6.5 nm and outer ring of 4.0 nm are shown in figure 3(b1). Through analyzing above experimental data, the average height of double rings in GaAs QR all increase with the increase of Ga deposition. While in figure 3(c1), a single ring of 4.0 nm appeared on GaAs surface. Comparing figures 3(b1) and (c1), we can infer that high temperature can make double ring become a single ring. The appearance of single ring is considered that high temperature makes double rings evolve into single ring. When Ga is deposited on the GaAs sample surface, double rings can be obtained after two As pressure crystallization. However, the deposition material on the top of the first ring began to flow downwards and to cover the second ring with substrate temperature increasing during the second As pressure crystallization in figure 3(c1) [29]. As the crystallization process continuing, the height of inner ring decreased, and the height of outer ring continued to increase. When the heights between inner ring and outer ring are the same, a GaAs single ring appears on the sample surface after two crystallization.
4. Conclusions

GaAs QR were formed on GaAs sample surface based on DE method, and various components of GaAs QR are calculated by the kinetic model of LDE. The theoretical value perfectly explains the experimental data. For the height of Ga droplets on GaAs sample surface, \( H_{\text{exp}} < H_{\text{cal,droplets}} \). \( D_{\text{exp}} > D_{\text{cal,droplet}} \). These results are mainly because Ga atoms in Ga droplets have begun to diffuse during the measurement and cause the height of Ga droplet to decrease, while GaAs wall appeared on the edge of Ga droplet leads to the diameter of Ga droplet become larger, so the experimental data of Ga droplet diameter is larger than the theoretical value of Ga droplet diameter. On the other hand, the surface density of Ga droplets will decrease with the increase of Ga deposition. While the volume, height and diameter of Ga droplets, the pore size, nanohole depth, ring width and height in GaAs QR will all increase with the increase of Ga deposition. In addition to theoretical analysis found that nanoholes in GaAs QR will become shallower under high As pressure. High temperature will lead to double rings evolve into a single ring. Above conclusions are helpful for the preparation of patterned GaAs templates based on DE and the controlled growth of GaAs QR.

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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