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# Real time analysis of self-assembled InAs/GaAs quantum dot growth by probing reflection high-energy electron diffraction chevron image

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Self-assembling process of InAs/GaAs quantum dots has been investigated by analyzing reflection high-energy electron diffraction chevron images reflecting the crystal facet structure surrounding the island. The chevron image shows dramatic changes during the island formation. From the temporal evolution of the chevron tail structure, the self-assembling process has been found to consist of four steps. The initial islands do not show distinct facet structures. Then, the island surface is covered by high-index facets, and this is followed by the formation of stable low-index facets. Finally, the flow of In atoms from the islands occurs, which contributes to flatten the wetting layer. Furthermore, we have investigated the island shape evolution during the GaAs capping layer growth by using the same real-time analysis technique. © 2008 American Institute of Physics.

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## I. INTRODUCTION

Self-assembled InAs/GaAs quantum dot (QD) grown by Stranski–Krastanov (SK) mode has been attracting great interest because of excellent improvement of the devices performance owing to the atomlike density of states. This unique characteristic realizes novel high-performance optoelectronic devices, such as direct modulation lasers, broadband and high-gain optical amplifiers, and high-speed wavelength converters.<sup>1</sup> The device performance strongly depends on the QD parameters, such as the size, density, and uniformity, which can be controlled by performing precise growth. It is well known that the SK growth mode utilizes the strain energy derived from the large InAs–GaAs lattice mismatch. The strain energy accumulates as the growth proceeds. Then the growth mode changes from the two-dimensional layer growth into the three-dimensional island growth when the InA growth exceeds a critical thickness.

Several approaches have been performed to investigate the self-assembling process in the SK growth by using atomic force microscopy (AFM) (Refs. 2 and 3) and scanning tunneling microscopy (STM).<sup>4–6</sup> In the report of the STM observation by Xu *et al.*,<sup>4</sup> island facet structures appeared in various growth stages have been shown. The islands are predominantly covered by high-index facets in the initial growth. Then, as the growth proceeds, low-index facets appear at the under part of the island, while the top part of the island is covered by high-index facets. However, it is noted that these are *ex situ* characterizations being impossible to reveal the temporal evolution of the island shape. On the other hand, Tsukamoto *et al.*<sup>7</sup> have observed detailed surface-phase transitions in the two-dimensional wetting layer (WL) by using an *in situ* STM system and found formation of precursor islands for QDs. Unfortunately, it is difficult to observe the following rapid assembling process by the *in situ* STM because of the finite scanning time.

It is widely known that reflection high-energy electron diffraction (RHEED) is a powerful *in situ* characterization tool to investigate the epitaxial growth surface. The RHEED is quite sensitive to the crystallographic surface structures in real time. When starting the island formation, the RHEED pattern dramatically changes from the streak image reflecting the two-dimensional surface into the diffraction spot image. In particular, the diffraction spot shows a chevron structure depending on the electron-beam incident azimuth. The chevron structure originates from the island surface facets.<sup>8–10</sup> Especially, the side-wall facets that are parallel to the electron-beam incident azimuth dominantly contribute to the formation of chevron structure.<sup>8</sup> The chevron structures for several island shapes have been simulated by Hanada *et al.*<sup>9</sup> and Feltrin and Freundlich.<sup>10</sup> They have demonstrated that the side-wall facet orientation can be directly extracted from the angle between chevron intensity tails. The RHEED chevron structure includes information concerning the QD structure. If the QD structure is simple, such as a pyramidal shape, the analysis of the chevron angle is not difficult as simulated in Ref. 9 that enables us to inform the facet structure. However, generally, the QD structure is not simple as reported in Ref. 4. Furthermore, the faceted structure changes as the assemble growth proceeds. When the QD is surrounded by complicated facets, the chevron tail structure is a superposition of the signals coming from those facets, and there is no one-to-one relation between the tail angle and a certain facet orientation. Therefore, the change in the chevron tail structure corresponds to a transition of the facets dominantly contributed to the diffraction signal. In this work, we focused on the chevron shape diffraction, and the detailed island shape transitions in the self-assembling process were studied using real-time probing of the dynamic evolution of the chevron tail structure. This probing technique is quite easy way to investigate the self-assembling process in real time. In addition, QD shape evolution during the GaAs capping layer growth was studied by using the same technique.

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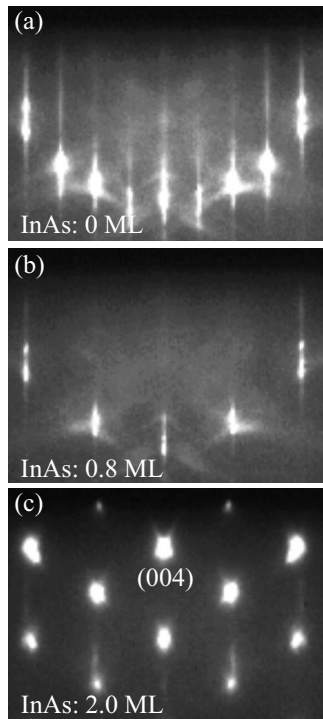


FIG. 1. RHEED patterns from (a) GaAs  $c(4 \times 4)$  reconstructed surface, (b) InGaAs WL, and (c) QDs surface.

## II. EXPERIMENT

Self-assembled InAs QDs were grown on a GaAs(001) substrate using molecular beam epitaxy (MBE). InAs was deposited at  $450^\circ\text{C}$  after growing a GaAs buffer layer. The deposition rate of InAs was  $0.012\text{ ML/s}$ . The As pressures used were  $3.0 \times 10^{-6}$  and  $8.0 \times 10^{-6}$  Torr. Real-time probing of the self-assembling process has been performed by using a conventional RHEED system equipped with the MBE. The electron-beam incident azimuth was set to the  $[-110]$ . The RHEED images were taken by using 12 bit color scale charge coupled device camera.

## III. RESULTS AND DISCUSSION

The RHEED pattern shows quite highly sensitive changes depending on the SK growth, as shown in Fig. 1. As reported in many literatures, the InAs deposition changes a streak pattern [Fig. 1(a)] of the initial GaAs  $c(4 \times 4)$  surface into a diffused pattern [Fig. 1(b)] of the InGaAs WL. Finally, the chevron diffraction pattern representing the three-dimensional island formation appears, as shown in Fig. 1(c). After the island formation, the chevron pattern keeps changing, while the islands are growing up.

### A. Four growth steps of QD self-assembling process

We recorded the RHEED diffraction spot intensity as a function of the InAs deposition thickness. To extract information of the self-assembling process, we first focus on both the peak intensity at the (004) diffraction spot and its integrated intensity covering the whole chevron images. Figure 2 shows results obtained at the As pressure of  $8.0 \times 10^{-6}$  Torr. With increasing the InAs thickness, the

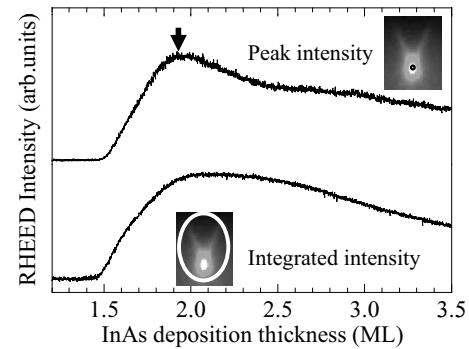


FIG. 2. (004) diffraction peak intensity and the integrated intensity including chevron tails, as shown in the inset, as a function of InAs deposition thickness. As pressure was  $8.0 \times 10^{-6}$  Torr.

RHEED intensity increases rapidly at about 1.5 ML indicating the beginning of three-dimensional island growth. It is found that the peak intensity profile is different from the one of the integrated intensity. The peak intensity profile shows a clear peak structure as indicated by an arrow. After that, the intensity decreases rapidly. On the other hand, the integrated intensity decreases gradually after reaching the maximum. These are due to the appearance of the chevron tails at the diffraction spot, so that the islands are considered to be covered by crystallographic facets.

Detailed self-assembling process can be studied by analyzing the dynamic evolution of the chevron tails. We monitored the RHEED intensity line profile at a position as indicated by the red bold line shown in Fig. 3. Utilizing the intensity profile, the change in the chevron angle  $\theta$  was evaluated. Figure 4(a) displays the chevron angle evolution as a function of the InAs deposition thickness at the As pressure of  $3.0 \times 10^{-6}$  Torr. It is found that the chevron angle shows a series of dramatic changes. The self-assembling growth process can be divided into four steps, as indicated in Fig. 4(a). In the initial growth step A, a round shape spot without showing distinct chevron tails was observed, and this attributes to the appearance of multifacets.<sup>9</sup> Then, chevron tails appear clearly in the next growth step B, which means the formation of specific orientation facets on the island surfaces. Remarkable assembly of islands proceeds in this step, and the chevron angle increases gradually as guided by black arrows in Fig. 4(a). This behavior continues up to the InAs deposition thickness of 2.0 ML. Although, as I mentioned above, the RHEED chevron angle does not identify a unique facet, the increase in the chevron angle can safely be considered to be due to a transition of the dominantly contributed facets changing from the high-index one to the low-index

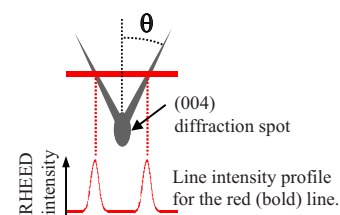


FIG. 3. (Color online) Illustration of a chevron structure. Chevron angle  $\theta$  is evaluated by an intensity profile, as indicated by a red bold line.

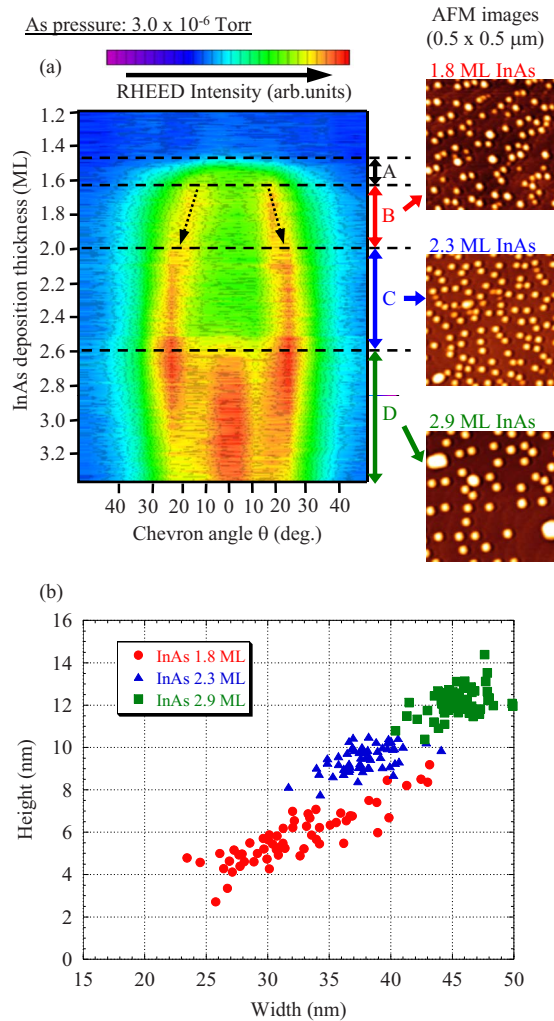


FIG. 4. (Color online) (a) Dynamic evolution of chevron angle  $\theta$  as a function of InAs thickness deposited at the As pressure of  $3.0 \times 10^{-6}$  Torr. AFM images were taken from each growth step. Island size distribution at each growth step is shown in (b).

one. Recently, we have succeeded in observing the three-dimensional structure of an embedded single QD.<sup>11</sup> Also, another report, such as Ref. 4, demonstrates clear faceted structures of uncapped QDs. According to these results, the QD is known to be an island with twofold symmetry with high-index facets on the base with fourfold symmetry with low-index facets. The low-index facets, such as  $\{110\}$  and  $\{111\}$ , should appear after forming the high-index facets, such as  $\{317\}$  and  $\{137\}$ , because the low-index facets are stable.<sup>4</sup> These considerations agree well with our results. During this facet evolution, we can confirm a remarkable island size distribution, as indicated by red symbols in Fig. 4(b); small islands covered by high-index facets and large islands containing low-index facets coexist. These results suggest that the growth step B is a stage where the island volume increases. In the next growth step C, the chevron angle becomes constant, where the islands are considered to be covered by stable low-index facets. Since the stable low-index facets are difficult to incorporate many In atoms, the assembling process comes to be self-limited, so that highly uniform island growth can be achieved in this growth step, as indicated by blue symbols in Fig. 4(b). In the last growth

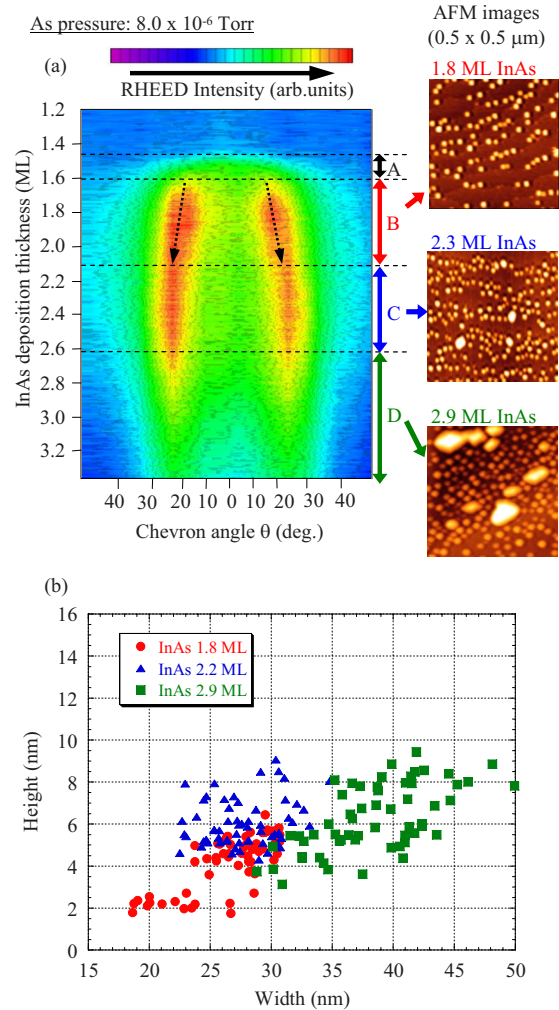


FIG. 5. (Color online) (a) Dynamic evolution of chevron angle  $\theta$  as a function of InAs thickness deposited at the As pressure of  $8.0 \times 10^{-6}$  Torr. AFM images were taken from each growth step. Island size distribution at each growth step is shown in (b).

step D, a streak diffraction appears in between the chevron tails, and the chevron signal intensity becomes weak gradually. In addition, the island density has been observed to be decreased, compared to the density in the last growth step. These results suggest that the In atoms tend to flow from the islands and the excess In atoms are incorporated to the WL. Then the WL surface becomes atomically smooth, which is the origin of the streak image. The In diffusion from the islands also helps the formation of giant islands. The explained results in this section make clear the relationship between the island formation process and the chevron structure evolution.

## B. Effect of As pressure on QD self-assembling process

The QD growth can be engineered by controlling the growth parameters, such as deposition rate, growth temperature, and As pressure. All these parameters affect the In migration length, and so this is a key factor of the island formation. To observe the In diffusion effects on the self-assembling process, we investigated the As pressure dependence of the chevron structure evolution. Figure 5(a)



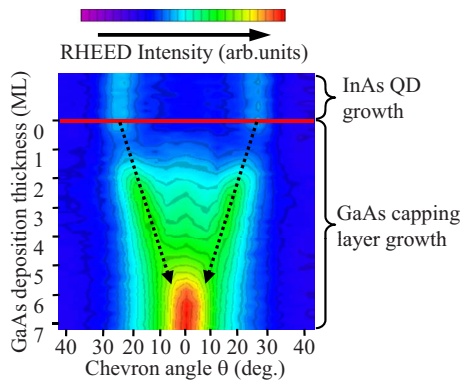


FIG. 6. (Color online) Dynamic evolution of the chevron angle  $\theta$  during the GaAs capping layer growth as a function of GaAs deposition.

displays the chevron angle evolution at the As pressure of  $8.0 \times 10^{-6}$  Torr. The four growth steps, as mentioned in Sec. II, can be also confirmed in the high As pressure. However, due to a change in the In migration length, the chevron angle evolution increases slowly in the growth step B, compared to the change in the As pressure of  $3.0 \times 10^{-6}$  Torr. This means that the formation speed of the stable low-index facets becomes slow because of the suppression in the In diffusion. The short In migration length causes a nonuniform size distribution even in the growth step C, as shown in Fig. 5(b). Compared to the results in the As pressure of  $3.0 \times 10^{-6}$  Torr, an obviously different behavior appears in the growth step D. It is found that the streak image does not appear clearly, which means that the suppressed In diffusion at the high As pressure prevents the WL from forming an atomically flat surface.

### C. QD shape transition during the GaAs capping layer growth

For practical optoelectronic QD applications, generally, the islands are covered by GaAs. However, since the capping layer growth on InAs QDs is also lattice mismatched heteroepitaxial growth, the large lattice mismatch gives rise to the significant In segregation and the resultant In–Ga intermixing. These phenomena dramatically change the island shape and size,<sup>11</sup> which is known to modify the optical properties remarkably. It is very important to understand what occurs in the islands during the GaAs growth. Here, we performed the *in situ* analysis of the island transition during the GaAs capping layer growth by using the same real-time chevron analysis technique. The GaAs capping layer was grown at 450 °C just after growing InAs QDs of 2.3 ML at the As pressure of  $3.0 \times 10^{-6}$  Torr. The 2.3 ML growth of InAs realizes highly uniform islands in the growth step C, as shown in Fig. 4. The deposition rate of GaAs was 0.5 ML/s.

Figure 6 displays the chevron angle evolution as a function of the GaAs deposition thickness. The red line in Fig. 6 indicates the beginning of the GaAs growth. When starting the GaAs growth on InAs QDs, the chevron angle decreases continuously. This means that the island facets change from low-index facets into high-index facets continuously, which is the reverse process of the island formation, as mentioned in Sec. III A. At the beginning of this process, we found a

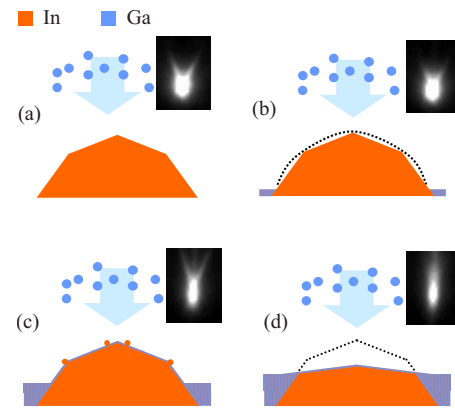


FIG. 7. (Color online) Model of the island shape transition during the GaAs capping layer growth.

transient decrease in the chevron signal intensity around 1 ML GaAs deposition. Such transient phenomenon has been observed even when we change the growth conditions that suggests formation of unfaceted islands. This characteristic transient phase disappears rapidly. The following clear chevron signal predicts that the unfaceted surface comes off the islands and then the facet planes dramatically appear again.

According to these results, we suppose a possible model in Fig. 7. The as-deposited QD structure in Fig. 7(a) is illustrated based on the results presented in Ref. 4. When GaAs starts to deposit on the QD, an unfaceted surface, as drawn by a dotted line in Fig. 7(b), is formed. Detailed structure of the unfaceted surface is not clear. Soon this unfaceted surface comes off the islands, and then, the faceted surface appears again. In the next step, we confirmed a gradual shift of the chevron angle. The angle approaches zero when the deposited thickness approaches about 6 ML ( $\sim 1.7$  nm). This thickness is very thin as compared to the as-deposited QD height, as shown in Figs. 4(b) and 5(b). Therefore, the top of the QD is considered to be truncated.<sup>12–14</sup> Since the clear chevron tail signal is observed during this process (Fig. 6), the growth front consists of the facet planes, as illustrated in Fig. 7(c). In segregation preferentially occurs in the directions with relatively low surface energies. Especially, this is evident to the (001) plane of the topmost surface of the island. Therefore, the decrease in the island height is supposed to accompany the change in the average orientation of the island surface [Fig. 7(d)].

## IV. SUMMARY

We have studied the self-assembling process of InAs/GaAs QD in detail by using RHEED chevron probing technique. It had been understood from the dynamic evolution of the chevron structure that the QD growth consists of four significant steps. According to the feature observed in each growth step, these steps can be called *nucleation*, *assembling*, *self-limited*, and *dissolving steps*. These four steps appear, even when the growth conditions are changed. Furthermore, we have demonstrated the QD shape evolution during the GaAs capping layer growth on InAs QDs by using the same analysis technique. From these results, the real-time analysis of the RHEED chevron structure is a powerful tech-

nique to engineer desirable QDs for practical device applications.

## ACKNOWLEDGMENTS

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