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RESEARCH ARTICLE

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Evolution of InAs islands in the Stranski-Krastanow mode of InAs/GaAs(001) fabricated using molecular beam epitaxy

Abstract Based on step-by-step observation using atomic force microscope, two distinctive successive phases were distinguished in accordance with evolution of the three-dimensional InAs islands during the Stranski-Krastanow mode of the InAs/GaAs(001) system fabricated using molecular-beam epitaxy. The initial phase is consistent with a power law, and the latter phase is a comparatively gradual one.

Keywords In As/GaAs dots, SK mode

PACS numbers 81.07.T, 81.16.Dn, 64.70.Nd

1 Introduction

The two- to three-dimensional (2D-3D) growth mode transition, or Stranski-Krastanow (SK) mode, is a basic phenomenon for heteroepitaxial growth. Generally, in the SK mode for a heterostructure, the epitaxial growth is two-dimensional at the outset. After a deposition of a few monolayers (ML) on the substrate, the growth mode is transformed into a three-dimensional growth mode. The InAs/GaAs (001) fabricated using molecular beam epitaxy (MBE) is a model system in which the self-assembled quantum dots are fabricated via the SK mode, and there have been many experiments as well as theoretical studies on the subject. In spite of much effort to investigate the SK mode for MBE InAs/GaAs(001), its detailed nature still remains poorly understood.

In an early time, it was established that under conventional MBE growth conditions, the critical InAs coverage (θ_c) for the 2D-3D transition is ~1.6 ML, beyond which the 3D InAs islands nucleate and the growth mode is transformed from two- to three-dimensional [1, 2]. After nucleation, the number density of the 3D InAs islands (N) increase sharply from quite few to a magnitude of a few 10^{10}/cm^2 within an InAs coverage (θ) increment of ~0.2 ML. This rapidly increasing period in island density within the 0.2-ML increment in InAs deposition is generally regarded as the 2D-3D transition process for MBE InAs/GaAs(001). The 2D-3D transition is not the conventional SK mechanism, during which a large amount of InAs material is transferred between the 2D InAs wetting layer and the ensemble of 3D InAs islands [3－6]. Leonard *et al*. [1] pointed out that during the 2D-3D transition the number density N of 3D InAs islands increases with InAs coverage θ according to a power law $N \propto (\theta - \theta_c)^{\alpha}$. In discussing their observation on variation of the average size of 3D InAs islands with θ, Kobayashi *et al*. [7] implicitly divided the 2D-3D transition process into two successive stages. In their initial stage of the 2D-3D transition, N is low and the 3D islands were isolated from each other, and these islands grow at the same rate. In the latter phase, the size-limiting effect [7, 8] sets in, and InAs material was transported between the islands.

In this work, the 2D-3D transition process in MBE InAs/GaAs(001) was investigated step-by-step using atomic force microscope (AFM). It was observed that the evolution of the 3D InAs islands in both the total volume V and number density N with InAs coverage θ can be divided into two distinctive phases during the 2D-3D transition. In the first phase, both V and N increase rapidly according to a power law; while in the second phase, the increase in V as well as in N with θ is much more gradual.

2 Experimental

The sample for an AFM observation was fabricated by depositing 2.0 ML of InAs on GaAs(001) substrate at substrate

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temperature $T = 500$ °C under the conventional MBE conditions for the self-assembly of InAs QD: As-rich and 0.1 ML/s growth rate. The substrate temperature was calibrated by the de-oxidation of the surface of GaAs (001) substrate at 580 °C, and was monitored by the infra-red pyrometer. The GaAs (001) substrate was not rotating during the InAs deposition to obtain an InAs film with the thickness varying from 1.5 to 3.0 ML across the 5.5-cm diameter in the [110] direction on the substrate due to the inhomogeneous indium flux. The AFM observation was focused on the regions of the sample surface where θ is in the range of 1.65—1.81 ML, which corresponds to the 2D-3D transition of the SK mode. The AFM measurement was made every 0.25 mm along the [110] diameter of the substrate with InAs increasing by 0.0068 ML in each step. The exact shape of the InAs islands is unknown, especially in their initial growth process, but their base shape is nearly rotationally symmetrical in the AFM images. The total volume V of the ensemble of the 3D InAs islands is assumed to be proportional to $N \times w^2 \times h$, where N, w and h are the number density, average width and height, respectively, of the 3D InAs islands obtained by statistically averaging on the AFM images of $1\times1\ \mu\text{m}^2$ in size.

3 Results

Successive AFM images reveal that the 3D InAs islands with a height above 1 nm begin to appear at $\theta \sim 1.65$ ML, and their number density N increases steadily from below $10^9/\text{cm}^2$ to $\sim 7\times10^{10}/\text{cm}^2$ at $\theta \sim 1.81$ ML, beyond which N increases more gradually to $\sim(8–9)\times10^{10}/\text{cm}^2$ until $\theta \sim 2.2$ ML, then island coalescence occurs and large dislocated islands begin to appear. Such behavior is similar to what are usually reported in literature. Figure 1 (a) is the log-log plot of island number density N against extra InAs deposition $(\theta - \theta_c)$ with $\theta_c = 1.65$ ML. It can be seen from the figure that, approximately, the evolution of N with InAs coverage θ in the 2D-3D transition can be divided into two successive phases with a transition region around $(\theta - \theta_c) \sim 0.05$ ML and $N \sim (1–2)\times10^{10}/\text{cm}^2$. The very initial phase proceeds drastically with the increase of InAs deposition, and the second phase is more gradual. The experimental data can be fitted with two straight lines of different slope, implying that the two phases in the 2D-3D transition are consistent with the different power law $N \propto (\theta - \theta_c)^{\alpha_i}$ with i = 1 and 2, respectively. The critical exponent for the first phase $\alpha_1 = 1.80$ is close to the value of 1.76 as pointed by Leonard *et al*. [1], while the exponent for the second phase is much smaller, $\alpha_2 = 0.60$. The distinction between the two successive phases in the 2D-3D transition is more clearly demonstrated in the evolution of total island volume V with $(\theta - \theta_c)$, as shown in Fig.1 (b). In this figure, the first phase is also consistent but with a power law with $\alpha = 1.94$, while the second phase proceeds much more gradually and the total volume increases with a very small rate. It should be noted that in the estimation of $V(\propto N \times w^2 \times h)$, some constant factor is inevitably missing, and the estimated value is not precise. However, the general trend of the variation in V with θ should be true. The large portion ~70 % of the total increment in V in the 2D-3D transition occurs in the first phase with the InAs deposition extending from 1.65 ML to 1.72 ML. In this period, the nominal InAs coverage θ increases by only ~ 0.07 ML, while the total island volume V increment is ~ 0.3 ML. In the second phase, from $\theta \sim 1.72$ ML up to $\theta \sim 1.81$ ML, V increases by only ~ 0.15 ML, roughly consistent with the amount of material from the InAs impinging flux. It is generally assumed that the transferring of InAs material between the 2D InAs wetting layer and the ensemble of 3D InAs islands extends over the whole 2D-3D transition [6]. However, the observation in this work shows that the material transferring occurs mainly in the very initial period of the 2D-3D transition, while the later stage is basically a conventional growth process. In plotting the total island volume as a function of InAs coverage for the same system, Placidi *et al*. [6] obtained a linear relation. Obviously, our result is inconsistent with their's, and the discrepancy needs further investigation.

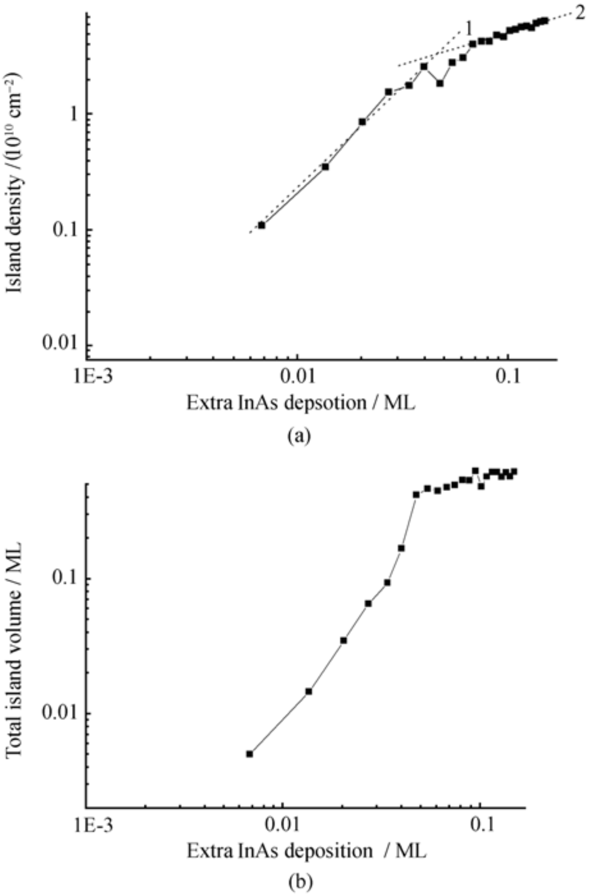

Fig. 1 Variations in both the island density **(a)**, and total island volume **(b)** against extra InAs deposition $(\theta - \theta_c)$ with $\theta_c = 1.65$ ML in the 2D-3D transition of MBE InAs/GaAs (001).

The identification of the two successive phases in the 2D-3D transition as described above is also demonstrated in the evolution of the average size of islands. Figure 2 shows the variation in both the average height *h* and average width *w* of the 3D InAs islands with InAs coverage θ during the 2D-3D transition. It can be seen from the figure that the average height *h* increases gradually from ~1 nm at first and then reaches maximum value of ~5 nm at the point with $\theta \sim$ 1.7 ML and $N \sim 2\times10^{10}/\text{cm}^2$, which are coincident with the separating point between the first phase and the second phase in the 2D-3D transition. After reaching the peak, the average height of islands decreases, and finally approaches an asymptotic constant value of $h \sim 3$ nm. Such a trend in variation of *h* is consistent with the observations mad by Kobayashi *et al*. [7] and Ebiko *et al*. [9]. However, the island density at which the peak happens reported in Refs. [7] and [9] is different from each other, and the datum in the latter is ~ $3\times10^{10}/\text{cm}^2$, while in the former, the datum is much lower by an order of magnitude. Such a distinction in the average height peak position may be due to the substrate temperature for the deposition of InAs used respectively by these authors. Obviously, our peak position for the average height is more close to that of Ebiko *et al*. [9]. The variation in average width *w* is more complicated than that of *h*. The value of *w* is about 30 nm at the beginning of the 2D-3D transition and decreases to a minimum of ~24.5 nm at $\theta \sim 1.68$ ML, and then sharply increases with θ to a maximum of ~32 nm at the same position as that of *h*, $\theta \sim 1.7$ ML and $N \sim 2\times10^{10}/\text{cm}^2$, and then rapidly decreases to a gradually varying process, staggering toward lower values. It may be noted that, as shown in Fig. 2, at the outset of the 2D-3D transition, the InAs islands are low in height and large in width, and therefore, the formation of 3D InAs islands may start from the 2D InAs islands with comparatively large lateral dimension, as suggested by Priester and Lannoo [10].

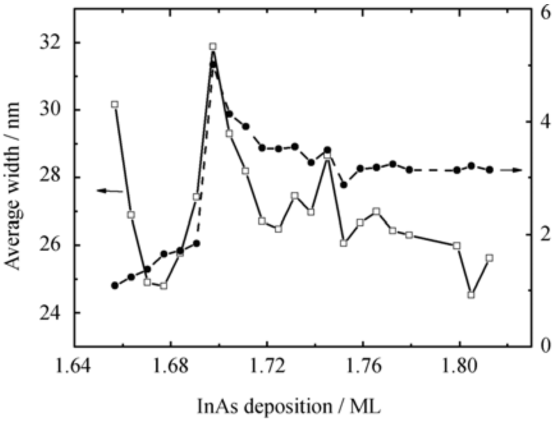

Fig. 2 Variations of the average island height and width with InAs deposited.

AFM observation showed that both the height and width distributions of the 3D InAs islands in the two successive phases during the 2D-3D transition are also different from each other. Figure 3 (a) and (d) are the AFM snapshots at $\theta \sim$ 1.67 and ~1.74 ML, respectively, representative for the first and second phase in the 2D-3D transition. Figure 3 (b) and (c) are the typical histograms of the island height and width, respectively, for the first phase. It can be seen that the average island height peaks at the small-size end, and decreases toward the larger sizes, while the width distribution is scattered. Figure 3(e) and (f) are the height and width distributions, respectively, and they are typical and representative for the second phase, and both are nearly of a Gaussian, distinctively different from the corresponding distribution in the first phase.

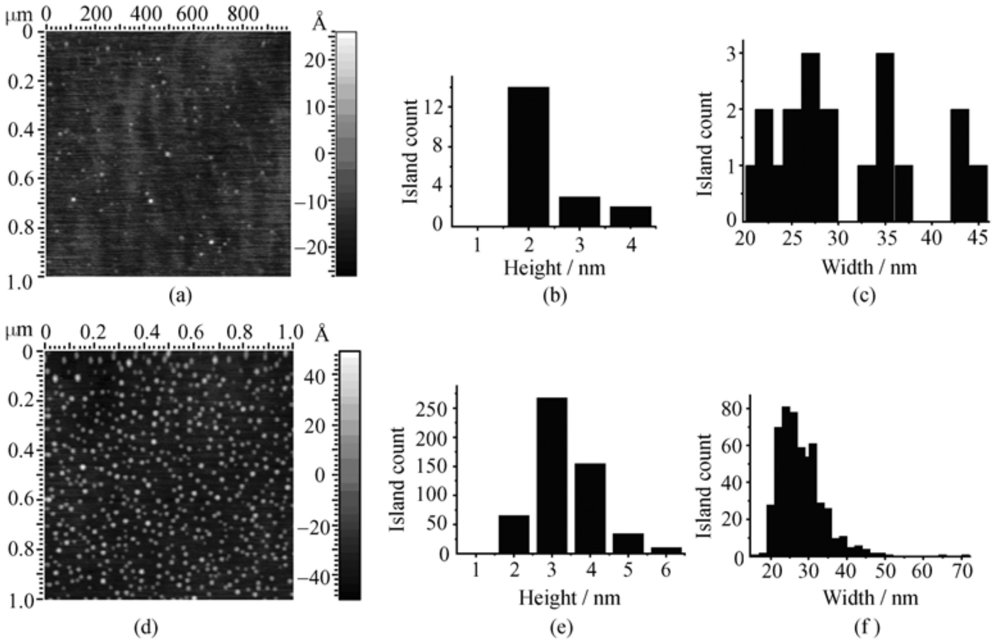

Fig. 3 **(a)**, **(b)** and **(c)** are the AFM image, histograms of island height and width $\theta \sim 1.67$ ML, respectively; **(d)**, **(e)**, and **(f)** are that of at $\theta \sim 1.74$ ML.

4 Discussion

Our observations described above identified two successive phases in the evolution of 3D InAs islands during the 2D-3D transition. The first phase in the very initial period of the 2D-3D transition is consistent with a power law and this implies that the nucleation and growth of 3D InAs islands in the first stage may be either a dynamical growth process or a second order phase transition in the sense of thermodynamics. The 2D-3D transition is “critical” as usually referred to in the literature. In addition, the first phase is not a conventional growth process during which the transferring of InAs material between the 2D InAs wetting layer and the 3D InAs island ensemble occurs drastically. Therefore, the first phase in the 2D-3D transition may be more likely a critical phenomenon [11] in the second order phase transition. In analogy, the total island volume is the order parameter in the second order transition, and the controlling parameter is θ, instead of temperature as in the ordering of a solid solution. It is generally regarded that the nucleation of stable 3D InAs islands has to overcome an energy barrier with its critical stable size exceeding at least a few hundred atoms [12, 13]. However, if the formation of 3D InAs islands begin with a critical phenomenon of the second order transition, there should be no such an energy barrier. Of course, this is an important topic that needs further investigation.

The mechanism for the subdivision into two successive phases of the 2D-3D transition in the MBE InAs/GaAs system may be similar to that as suggested by Kobayashi *et al.* [4]. In the first phase, the number density of the 3D InAs islands is very low and they nucleate and grow, isolated from each other due to the comparatively large distance, and the local thermodynamic equilibrium is established for each InAs island among its side facets and with its surrounding wetting layer [14]. The first phase may be terminated when the island density exceeds a critical value, $\sim 2\times10^{10}/\text{cm}^2$ in this work, and the elastic interaction among the 3D islands sets in, and the gradual increasing stage begins. In the second stage of the 2D-3D transition, individual islands interact with each other via the strain field in the wetting layer, and the material was transported between them, and a global thermodynamic equilibrium may be established to some extent [12]. There may be a thermodynamically favored value for the island size in the second phase, and resembles a Gaussian distribution, as shown in Fig. 3 (e) and (f). In this stage, the total island volume increases in a comparatively low rate and the amount of material required for the formation and growth of the InAs islands may be mainly provided by impinging InAs flux.

5 Conclusions

Two successive phases with the distinctive behaviors in the evolution of the 3D islands were identified in the SK growth mode for MBE InAs/GaAs(001). The first phase is consistent with a power law and similar to the second order phase transition in thermodynamics, during which the material transferring between the 2D InAs wetting layer and the 3D island system occurs drastically. In the second phase, the total island volume increases gradually at a rate roughly consistent with the InAs impinging flux.

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