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Quantum Dot Formation by Post-Growth Annealing of a Wetting Layer

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1. Introduction

The exact mechanisms in the self-assembly of semiconductor quantum dots (QDs) have not been well understood yet, although considerable effort has been devoted [1]. It is considered that there are two possibilities: consuming the adatoms on the wetting layer and directly consuming the wetting layer [2]. In conventional QD growth, these two are mixed together and may interact with each other. Experimental studies of QD formation without deposition flux is thus required [3]. On GaAs substrates, here we investigate the progression of an InAs wetting layer thinner than but close to the critical thickness, and compare with the conventional QD growth process.

2. Results and Discussions

Our samples were fabricated by molecular beam epitaxy on semi-insulating GaAs (001) substrates. InAs was deposited on a buffer layer of GaAs. Following the stop of InAs deposition, annealing was performed without changing the substrate temperature and the arsenic pressure. It is found that, from an InAs wetting layer thinner than the critical thickness θ_c for conventional transition from two- (2D) to three-dimensional (3D) growth mode, post-growth annealing leads controllably to rather low density of QDs, as shown by the example in Fig. 1. In order to monitor the QD formation process, real-time reflection high energy electron diffraction (RHEED) was used with the substrate fixed such that the pattern along [100] azimuth is recorded, as shown in Fig. 2(a). Focusing on an area corresponding to a bright spot after QD formation as marked in Fig. 2(a), the integrated RHEED intensity versus time can be obtained. It has been confirmed that the RHEED intensity is proportionally correlated with the total QDs volume, so we use the RHEED intensity as a measure of the amount of formed QDs. In Fig. 2(b), scattered symbols demonstrate a series of time-scanned RHEED intensity for InAs/GaAs QD formation at 480 °C with different coverage of InAs deposited at 0.031 ML/s. For the sake of clarity, the zero point of the time axis is taken to be the stop of InAs growth rather than the beginning of InAs deposition. There shown two cases here, QD formation with InAs deposition continuing after, and stopping before 2D-3D transition, which takes place at 53 s. In the former case, RHEED intensity increases rapidly and then saturates, whereas the latter case exhibits a delayed and slowed increase. In fact, there is certainly a saturation behavior even with InAs growth time well shorter than the critical one as long as the post-growth annealing time is sufficiently long.

To understand the above observation, we refer to a mean-field theory advanced by Dobbs *et al.* [4], in which the mass transport between adatoms, precursors and QDs are considered. Extending it to the growth process before 2D-3D transition by taking into account the purely 2D growing wetting layer, we have the rate equations for the amount of adatoms n_1, precursors n_2 and QDs n_3:

$$\begin{aligned} \frac{dn_1}{dt} &= F - \beta n_1 - D(\sigma_2 n_2 + \sigma_3 n_3)n_1 \\ \frac{dn_2}{dt} &= D\sigma_2 n_1 n_2 - \gamma n_2 \qquad , \\ \frac{dn_3}{dt} &= D\sigma_3 n_1 n_3 + \gamma n_2 \end{aligned} \tag{1}$$

where F is the InAs deposition rate, β is a coefficient for 2D growth of the wetting layer, D is the diffusion coefficient of adatoms, σ_2 and σ_3 are the so-called "capture number" for adatoms to be captured by the precursors and QDs, respectively, and γ is the nucleation rate of QDs. Simulations using Eq. (1) well fit the experimental QDs volume versus time, as shown by solid lines in Fig. 2(b). It is clear that the precursors emerging before 2D-3D transition are responsible for the usually observed significant consumption of wetting layer [5]. In conventional case, adatoms diffusing directly into QDs play more and more dominant role as InAs deposition proceeds, while nucleation from precursors is the only dynamics in QD formation from a wetting layer by post-growth annealing. In a good approximation, the latter follows a simple relation:

$$n_3=n_{2s}(1-e^{-\gamma t}), \tag{2}$$

where n_{2s}, describing the saturation QD volume, equals the value of n_2 at growth stop. Fitting the experimental data by Eq. (2), we obtain n_{2s} and γ as shown in Fig. 3. It can be seen that both of n_{2s} and γ exponentially depends on the InAs coverage in the region well below the critical thickness θ_c.

The growth of a wetting layer initiates with a dynamic balance between the pure 2D wetting layer and adatoms. As the precursors grow up, the early amount of adatoms n_1 changes little and that of precursor n_2 keeps initially small, so $dn_2/dt \approx D\sigma_2 F\beta^{-1} n_2$ well before the critical point. This results in exponentially increasing n_2 with the deposition time, equivalently n_{2s} with the coverage $\theta=Ft$ as seen in Fig. 3(a). As to the QD nucleation rate γ, it is considered exponentially depending on the so-called superstress of the wetting layer, $(\theta-\theta_c)/\theta_c$, via QD formation energy [2], which agrees with the result in Fig. 3(b).

3. Conclusions

We studied InAs/GaAs QDs formation by post-growth annealing of an InAs wetting layer thinner than the critical thickness for 2D-3D growth mode transition. Based on a mean-field theory, the time evolution of total QDs volume, first increasing and finally saturating, is well explained simply by QD nucleation from precursors. The exponential coverage dependences of the saturation QDs volume and the QD nucleation rate are also understandable within the frame of this theory. Extending to conventional QD growth, it is suggested that the precursors emerging before 2D-3D transition are responsible for the usually observed significant consumption of wetting layer.

References

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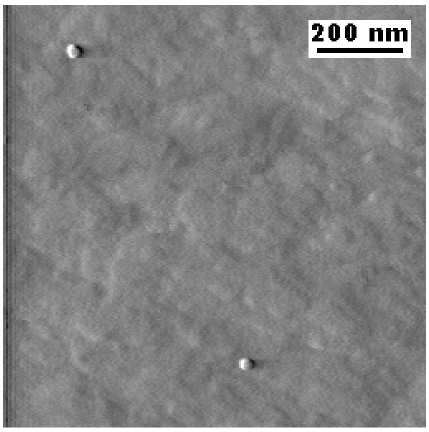

Fig. 1 The atomic-force microscope image of an InAs/GaAs QD sample prepared by depositing 1.49 ML of InAs at 480 °C followed by 60 s of post-growth annealing.

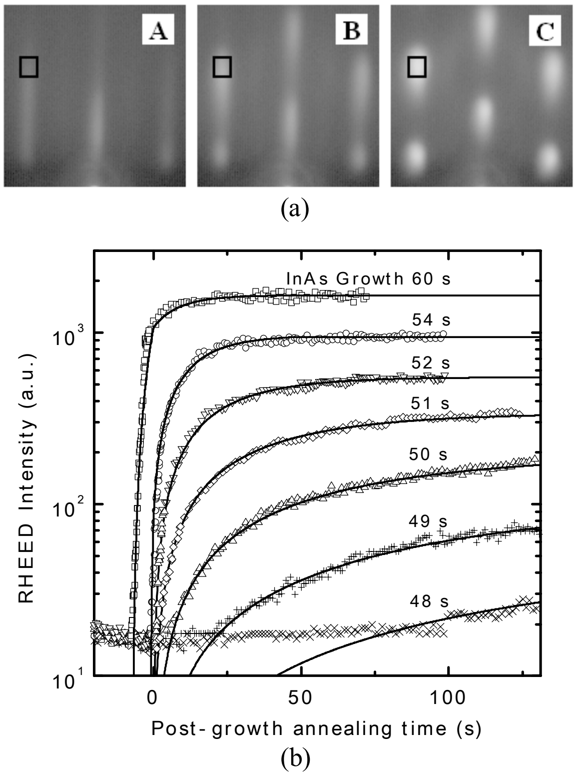

Fig. 2 (a) Typical RHEED images before (A), at the beginning of (B) and after (C) the formation of QDs; (b) RHEED intensity evolution with the process of QD formation. The solid curves are the simulated results according to Eq. (1).

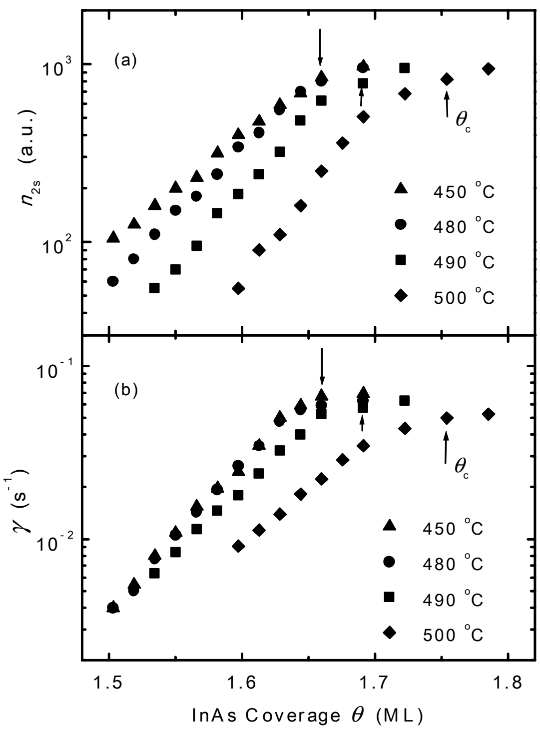

Fig. 3 InAs coverage dependence of n_{2s} and γ obtained by fitting the RHEED data with Eq.(2).