Available online at www.sciencedirect.com

SCIENCE DIRECT®

Journal of Crystal Growth 293 (2006) 546–549

JOURNAL OF CRYSTAL GROWTH

www.elsevier.com/locate/jcrysgro

Stress development during annealing of self-assembled InAs/GaAs quantum dots measured in situ with a cantilever beam setup

D.Z. Hu, D.M. Schaadt*, K.H. Ploog

Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5–7, D–10117 Berlin, Germany

Received 22 March 2006; accepted 5 June 2006
Communicated by H. Asahi

Abstract

We have investigated the coarsening of InAs quantum dots (QDs) in real time by measuring the stress evolution using an in situ cantilever beam setup. During deposition of InAs QDs, stress is accumulated, which then relaxes during subsequent annealing. Models based on different mechanisms for Ostwald ripening are fitted to the stress relaxation curves. A model describing ripening limited by diffusion along dot boundaries yields a good fit for annealing at 440 °C. For annealing at 470 °C, the relaxation curve can be fitted very well with a model in which ripening is controlled by attachment/detachment of atoms on the dot surface.

PACS: 68.35.−p; 68.65.Hb; 81.07.Ta; 81.10.Aj

Keywords: A1. Mass transfer; A1. Nanostructures; A1. Ripening; A3. Molecular beam epitaxy; A3. Quantum dots

1. Introduction

Self-organized quantum dots (QDs) have been extensively studied in recent years because of their potential for technological applications [1]. The self-organization occurs by the Stranski–Krastanow (SK) growth mode [2] which applies for heteroepitaxy in systems with lattice mismatch $\geqslant 2\%$ such as Si/Ge (4.1%) and InAs/GaAs (7.2%). Three-dimensional (3D) dislocation-free (so-called coherent) islands form on top of a wetting layer that is composed of the deposited material. For InAs/GaAs (0 0 1), the transition from 2D layer by layer growth to 3D QD growth is found at a thickness of 1.5–1.7 ML [3,4]. Upon post-growth annealing, these coherent islands typically undergo morphological changes with time, known as Ostwald ripening [5]. In the case of InAs/GaAs, ripening from small to large dots (Ostwald ripening) has been reported [6,7]. The fundamental theory of particle coarsening is already well established by Lifshitz and Slezov [8,9] and Wagner [10] and further developed by Vengrenovitch [11,12], where a dot size distribution and a mean dot size as a function of time are derived. In this theory, the coarsening of particles is limited by various possible mechanisms. For ripening of InAs QDs, the following three models have to be considered. The first model, denoted as model 1, describes that coarsening is limited by a kinetic process. In the second and third model, denoted as model 2 and 3, respectively, ripening is controlled by diffusion on the surface (model 2) and along the dot boundaries (model 3). To determine the ripening process of QDs, investigators often try to fit an experimentally obtained island radius distribution to the above-mentioned theoretical models. However, this procedure does not always yield a clear determination of the ripening process. For instance, for annealing of InAs QDs, Krzyzewski and Jones [7], using STM images taken at different annealing times, found that the evolution of the average dot size as a function of time cannot be described by any of the above models.

In this paper, we present a new approach to determine the ripening process by not fitting only the average dot radius but rather by fitting the complete time evolution of the dot radius distribution function to experimental data.

---

*Corresponding author.
E-mail address: dschaadt@pdi-berlin.de (D.M. Schaadt).

0022-0248/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.jcrysgro.2006.06.009

For this purpose, we use an in situ cantilever beam setup to measure the film force evolution during deposition and annealing of InAs QDs grown on GaAs. The stress is then given by the slope in the film force (FF) curves. By extending the above models, we can fit them to FF relaxation curves obtained during the annealing phase of the growth. In combination with AFM images taken at various times during annealing, we are able to determine the involved ripening mechanism.

2. Experimental details

All experiments were performed in a III–V semiconductor molecular beam epitaxy chamber equipped with a cantilever beam setup. Epi-ready GaAs (0 0 1) substrates of $25 \times 5 \times 0.15\,\mathrm{mm}^3$ in size were clamped on one side only to allow the other free end to bend. During deposition, the deflection of the free end was measured in real time by a capacitance technique combined with lock-in-assisted signal detection. Using a modified form of Stoney's equation [13], the FF, defined as the force per film width, was calculated from the substrate bending. The GaAs substrates were coated with a 500 nm thick GaAs buffer layer after oxide desorption at 605 °C. Uncapped InAs QDs were then grown at 440 and 470 °C by depositing nominally ~2.0 ML InAs. To monitor the annealing behavior of the QDs, the films were kept after deposition at the growth temperature for a given time and then rapidly quenched to room temperature for AFM studies. To obtain a FF curve, the substrate bending was measured during growth of the InAs QD layer and after growth during annealing for a fixed time of 10 min.

3. Results and discussion

Fig. 1 shows the evolution of InAs FF curves measured during growth at 440 and 470 °C followed by annealing for 10 min. Upon opening of the In shutter, a linear increase in FF to ~2.3 N/m is observed up to a thickness of ~1.6 ML. With further InAs deposition, the FF increase becomes slower and proceeds with a small slope up to a value of 2.5 N/m. This is due to the change from 2D layer by layer growth to 3D island formation and growth. During annealing, the FF decreases from 2.5 to 1.76 and 1.01 N/m for 440 and 470 °C, respectively. AFM images clearly show that ripening occurs during annealing. As shown in Figs. 2 and 3, before annealing, QDs are small and their density is high. With annealing time, the size and density of big dots increase while almost all small dots disappear (d in Fig. 2 and b in Fig. 3). A closer look at the AFM images reveals pyramid-shaped dots. During ripening, a constant dot height to radius ratio of about 0.2 is measured, yielding an opening of about 11°. This indicates that the dots form initially as pyramids and then grow by increasing the side facets, yielding a 3D coarsening.

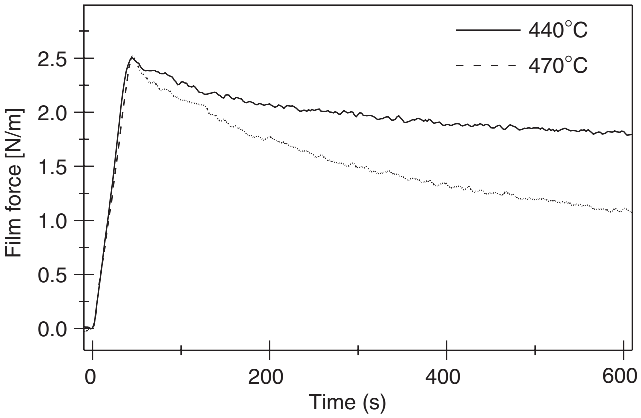

Fig. 1. Film force measured during InAs deposition for 45 s and followed by post-growth annealing for 10 min at 440 and at 470 °C, respectively.

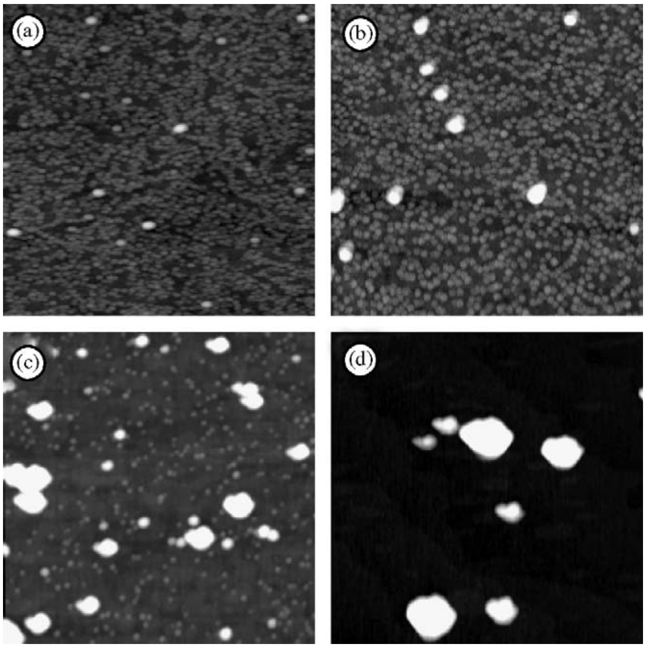

Fig. 2. $2 \times 2\,\mu\mathrm{m}^2$ AFM images of InAs QDs deposited for 45 s at 440 °C, taken (a) before annealing, (b) after annealing for 1 min, (c) 5 min, and (d) 10 min. The z-scale in all images is 10 nm.

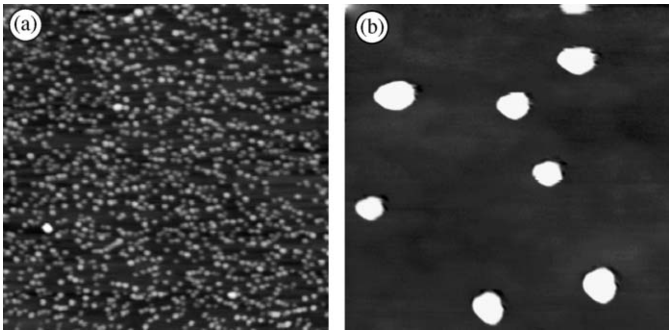

Fig. 3. $2 \times 2\,\mu\mathrm{m}^2$ AFM images of InAs QDs deposited for 45 s at 470 °C, taken (a) before annealing and (b) after annealing for 10 min. The z-scale in all images is 10 nm.

In modelling the FF curve during ripening of the QDs, in a first approximation, we assume a constant surface tension σ for the dots with the total surface area of the dots A_{dots} changing with time. For such a situation, the FF(t) is given by

$$\mathrm{FF}(t) = \sigma * A_{\text{dots}}(t)/A_{\text{subs}}, \tag{1}$$

where A_{subs} is the constant surface area of the substrate. Based on the above discussion of the dot shape and dot size evolution during ripening, 3D coarsening must be considered and we therefore employ the model developed by Vengrenovitch for ripening of 3D particles [11]. According to this model, at a given time t, the maximum radius of the particles r_g is given by

$$r_{g_n}(t) = a_n(t + b_n)^{1/(n+1)} \quad \text{with } b_n = r_{0g}^{n+1}/a_n, \tag{2}$$

where r_{0g} and a_n are constants. For $n = 1$, the ripening will be controlled by attachment or detachment of atoms on the dot surface, for $n = 2$, the mechanism for QDs ripening is limited by surface diffusion of atoms while for $n = 3$ the ripening is limited by diffusion of atoms along grain boundaries. Assuming that the total volume of all dots remains constant during the ripening process, the distribution of particle sizes r at time t is then calculated as

$$f_n(r,t) = \mathrm{const}_n \int_0^{r_{g_n}(t)} \frac{g_n(r/r_{g_n}(t))}{r_{g_n}(t)^4}\,\mathrm{d}r \tag{3}$$

with distribution functions $g_n(u)$ derived by Wagner [10], Lifshitz and Slezov [8,9] and Vengrenovitch [11]. The overall dot surface area $A_{\text{dots}_n}(t)$ then is

$$A_{\text{dots}_n}(t) \propto \pi \int_0^{r_{g_n}(t)} r^2 f_n(r,t)\,\mathrm{d}r. \tag{4}$$

During calculation for each model a_n is normalized. We choose different b_n to fit the experimental curves and get the value of b_n according to the minimum of relative errors.

Our calculation results are shown in Fig. 4. For annealing at 440 °C, model 3 yields the best fit to the experimental curve. However, the other two models also closely fit the experimental data. It is therefore difficult to clearly distinguish which mechanism really limits the ripening process of dots or if a combination of two or more mechanisms governs the process. On the other hand, for annealing at 470 °C, model 1 fits the experimental data very well, indicating that the kinetic process, in which attachment/detachment of atoms on the dot surface is the limiting step, is the mechanism for dot ripening. Considering that diffusion is typically a limiting factor at lower temperature and kinetic processes are the limiting factors at higher temperatures, our results appear reasonable. Due to the employed fitting of the experimental data with a model which incorporates the whole time evolution of the dot distribution function rather than only an average dot radius, our results are more compelling in determining the ripening process compared to previously published work.

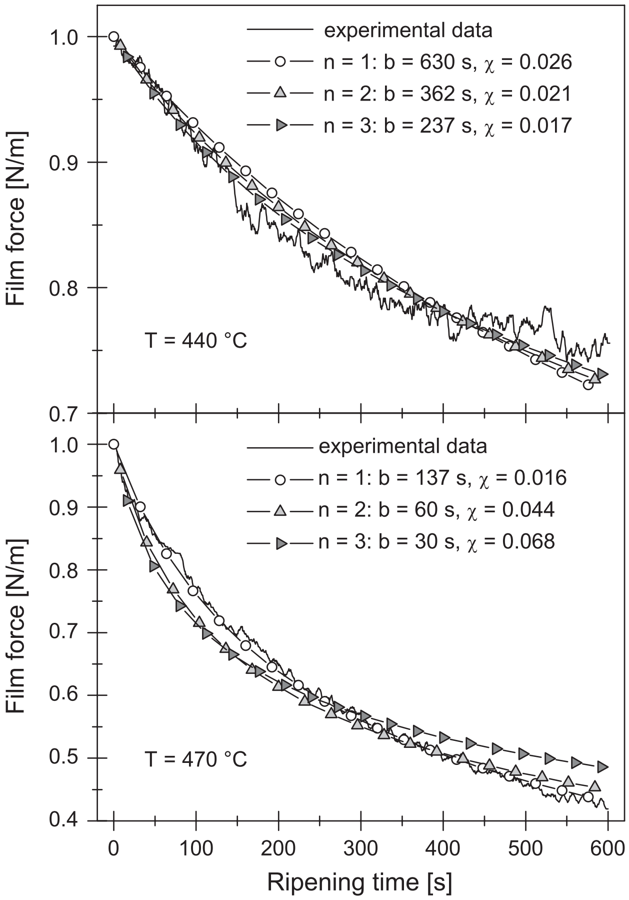

Fig. 4. Fitted curves to film force curve measured as a function of time during InAs post-growth annealing for 10 min at 440 and at 470 °C, respectively.

4. Conclusions

We have studied the ripening of InAs QDs grown on GaAs (0 0 1) substrates by measuring in situ the stress during growth and subsequent annealing. A model based on Ostwald ripening was developed that allows us to fit the relaxation curves and determine which mechanism plays the main role. We find that, for annealing InAs/GaAs (0 0 1) at 440 °C, it is difficult to determine the ripening process clearly, but still a model describing ripening controlled by atom diffusion along dot boundaries fits the experimental data best. On the other hand, for annealing at 470 °C, we find that the ripening process is clearly limited by atom attachment/detachment on the dot surface.

Acknowledgements

The authors thank Ms. Krauss and R. Koch for experimental assistance. The work was supported by the Deutsche Forschungsgemeinschaft (Sfb 296).

References

[1] See, e.g., M. Grundmann, Physica E (Amsterdam) 5, 167 (2000), and references therein.

[2] I.N. Stranski, L. Krastanow, Sitzungsber, Akad. Wiss. Wien, Math.-Naturwiss. Klasse 146 (1937) 797.

[3] M. Moison, F. Houzay, F. Barthe, L. Leeprince, E. Andre, O. Vatel, Appl. Phys. Lett. 64 (1994) 196.

[4] J.Y. Marzin, J.M. Gerard, A. Izrael, D. Barrier, G. Bastard, Phys. Rev. Lett. 73 (1994) 716.

[5] W. Ostwald, Z. Phys. Chem. (Leipzig) 34 (1990) 495.

[6] K. Poetschke, L. Mueller-Kirsch, R. Heitz, R.L. Sellin, U.W. Pohl, D. Bimberg, N. Zakharov, P. Werner, Physica E 21 (2004) 606.

[7] T.J. Krzyzewski, T.S. Jones, J. Appl. Phys. 96 (2004) 668.

[8] I.M. Lifshitz, V.V. Slezov, Soviet Phys. JETP 35 (8) (1959) 331.

[9] I.M. Lifshitz, V.V. Slezov, J. Phys. Chem. Solids 19 (1959) 35.

[10] C. Wagner, Z. Electrochem. 65 (1961) 581.

[11] R.D. Vengrenovitch, Acta Metall. 30 (1982) 1079.

[12] R.D. Vengrenovich, Yu.V. Gudyma, S.V. Yarema, Semiconductors 35 (2001) 1378.

[13] G.G. Stoney, Proc. R. Soc. London A 32 (1909) 172.