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Morphological transformation of $In_yGa_{1-y}As$ islands, fabricated by Stranski–Krastanov growth

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Abstract

A remarkable change in topology occurs when $In_yGa_{1-y}As$ quantum dots, grown by Stranski–Krastanov self-organization, are covered by a thin layer of GaAs. The nano-islands rearrange themselves to form volcano-like islands with distinct, ring-like features. The island topology can be preserved during overgrowth and thus used for the fabrication of ring-shaped quantum structures. The experimental data suggests that two mechanisms, diffusion and dewetting, are driving the transformation from dots to rings. © 2001 Published by Elsevier Science B.V.

Keywords: Dots; Rings; Stranski–Krastanov; Self-organized; Island; Transformation

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1. Introduction

Stranski–Krastanov growth in lattice mismatched semiconductor systems is a simple, yet versatile route for the realization of dislocation free nanometer-sized islands. When incorporated into an electrically or optically active structure, these islands can be used to fabricate the so-called 'quantum dots'. Quantum dots are systems in which electrons, holes, or excitons are confined in all three spatial dimensions on a nanometer scale. Stranski–Krastanov grown ('self-organized') dots have found great use in both basic studies of quantized many-particle systems and in application-oriented model devices [1]. Furthermore, the mechanisms behind the formation of self-organized dots, their morphology and composition, and their ordering have been subjects of considerable interest from a basic materials science view [2,3].

It is commonly assumed that the morphology and composition of self-assembled dots are only little affected by overgrowth. Experimental results obtained from atomic force microscopy, X-ray scattering, transmission electron microscopy, and in situ electron diffraction have been compared without explicitly dividing the data into the two categories 'uncovered' and 'covered' [3–8].

There is mounting evidence, however, that during overgrowth the heteroepitaxial system is not in equilibrium. Even though we believe this to be true quite generally, we will restrict the discussion in the following to the $In_yGa_{1-y}As$ system. In this material system, the non-equilibrium nature of the dot overgrowth manifests itself most prominently in the formation of quantum rings [9].

Here, we will review the formation of quantum rings from partially covered quantum dots, as well as their morphological and electronic properties. We will then discuss possible mechanisms, which might contribute to this striking change in shape, taking into account diffusion, segregation and surface/interface energy differences.

2. From dots to rings

A first systematic study of partially overgrown $In_yGa_{1-y}As$ dots was performed by Garcia et al. [10]. In this study, ring-shaped $In_yGa_{1-y}As$ islands were

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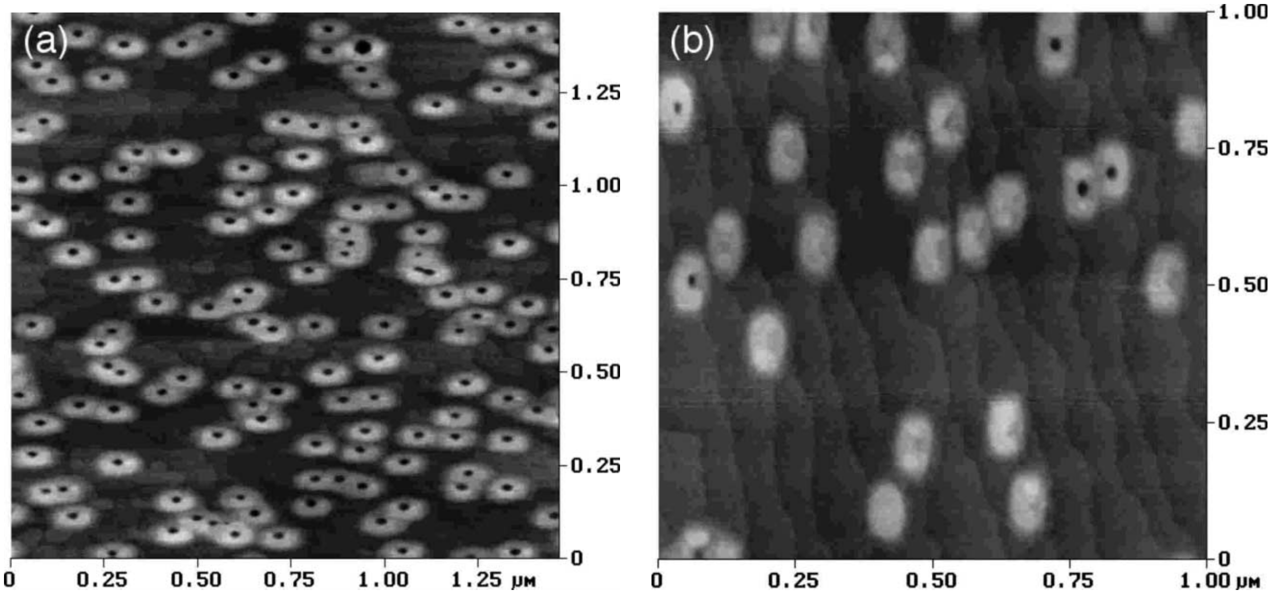

Fig. 1. Atomic force micrographs of self-organized $In_yGa_{1-y}As$ rings. The thickness of the GaAs cover layer is 4 nm. The crystal direction in which the islands are elongated is [1 − 10]. The depth of the center hole is estimated to be around 1–3 nm, the rim is approximately 2 nm high. In (b), not all islands exhibit a well-developed center hole.

observed on samples with moderate densities of self-assembled $In_yGa_{1-y}As$ dots and coverages of few nanometers. Redistribution of material during overgrowth [11] had been observed before. Also, the surface morphology of partially capped samples used for ballistic electron emission microscopy [12] had suggested an island shape with a depression in the center. Nevertheless, the reproducibility and homogeneity of the rings came as a surprise. Further studies [13–16] substantiated these initial findings and suggested that the rings could be used for the investigation of the electronic and optical properties of nanometer-sized, few-particle systems with a complex (i.e. not simply connected) geometry.

The growth of the rings, by molecular beam epitaxy, proceeds as follows: on a clean and smooth GaAs (001) surface, around 1.7 monolayers of InAs are deposited at a substrate temperature of 530 °C, resulting in the formation of a low-density, random array of self-organized quantum dots with a diameter of 20 nm and a height of around 6 nm [6]. By in situ electron diffraction (RHEED) it can be observed that the mobility of the deposited material is such that the redistribution necessary for dot formation happens on a time scale between a few seconds to a few ten seconds. The dots are then covered by a thin (few nanometers) GaAs layer and annealed under As flux between 30 s and 1 min at 530 °C. Samples that are then removed from the growth chamber for ex situ atomic force microscopy (AFM) characterization exhibit a typical island morphology as shown in Fig. 1(a). Their shape resembles a nanoscopic volcano, with a diameter of the inner hole ('chimney') of about 20 nm, an outer diameter between 60 and 140 nm and a maximum height (at a diameter of ≈ 30 nm) of about 2 nm. On some samples, not all island have a well-defined depression in their center, as seen in Fig. 1(b). We attribute this to the fact that these samples have been removed immediately after the capping layer had been deposited, so that enough time was not allowed for the rings to completely form (we estimate the maximum cooling rate in our system to be ≈ 5 °C s^{-1}). The fact that islands with well developed holes and islands without any depression in their center coexist, suggests that the hole formation is an abrupt process rather than a smooth transition (see also the discussion below).

One important conclusion can immediately be drawn from the observed shape change on partially capped islands: *the overgrowth of InAs dots with GaAs is a non-equilibrium process*. The fact that a simple growth interruption will so dramatically alter the morphology clearly shows this. This has important consequences for the growth of embedded island in general. Experimental parameters, such as growth temperature, deposition rate and As overpressure are of great importance for the size, shape and composition of the quantum dots. Therefore, great care has to be taken when comparing electronic, optical and other properties of dots grown under slightly different conditions. Furthermore, the change in shape will be an obstacle for the realization of strongly coupled, i.e. narrowly spaced, multiple quantum dots. When the GaAs layer that is intended to separate the dots is thin and the growth is interrupted or drastically slowed down (as is common practice during the growth of the next dot layer), the original shape of the islands in the first layer might be corrupted.

3. Optical and electronic properties

Apart from the morphology, it is also of great interest to investigate the electronic and optical properties of self-organized quantum rings [9,17]. The not simply connected shape makes them promising candidates for studying flux-related magnetic properties of quantum systems in the nanometer-size range. These experiments also help addressing materials-related questions, such as: (i) Will it be possible to maintain the island shape during further coverage, e.g. with GaAs? (ii) And if so, will the configuration of the island translate into a ring-shaped, attractive potential for carriers in the grown structure? The first question is of particular importance in view of the conclusion drawn above, namely that the coverage of dots is a non-equilibrium process that can lead to a considerable redistribution of material. The same could be true for the coverage of the ring islands and thus their shape might be destroyed during capping. The second question will give some insight into the composition of the volcanoes, since only an In-rich ring will lead to an attractive potential.

Capacitance, far-infrared and optical spectroscopy were carried out in order to investigate the carrier confinement in samples with overgrown volcano islands [9,17,18]. Here, the response to an external magnetic field [19] is of particular interest, since Aharonov–Bohm-type effects are features that will clearly distinguish multiple connected quantum systems (rings) from simply connected ones (dots).

Assuming a diameter of a few ten nanometers, one flux quantum penetrating the interior of the ring will correspond to an external magnetic field B of a few tesla. Experimentally, at a magnetic field around 8 T, the electronic excitation spectrum—in the far-infrared spectral range—indeed undergoes a dramatic change, with new resonances appearing and others disappearing [9,15]. Model calculations show that the observed spectral features are in agreement with the electronic properties of rings of ≈ 28 nm diameter [9,20]. At around 8 T, the single-particle electronic ground state in the rings will change from zero angular momentum to angular momentum $L = -1$. In consequence, the allowed dipole transitions will change and thus the observed excitation spectrum. In the mesoscopic picture, the non-vanishing ground state angular momentum corresponds to the occurrence of persistent currents when a flux quantum penetrates the interior of the ring [21].

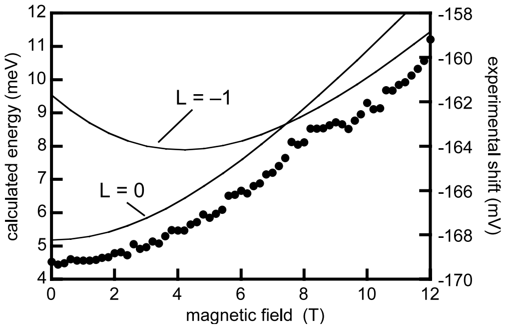

Fig. 2. Calculated single-particle ground state energy (solid lines) in a quantum ring of 28 nm diameter. Data points give the voltage of the lowest charging peak in the CV spectra (after Ref. [9]).

The magnetic field induced transition of the ground state angular momentum inferred from the far-infrared spectroscopic data can be confirmed by capacitance–voltage (CV) measurements as a complementary spectroscopic technique. While the far-infrared data gives information on the energies of the allowed transitions between electronic states, capacitance spectroscopy allows for a direct examination of the ground state energies themselves. This is shown in Fig. 2, where the position of the lowest charging peak observed in the CV spectra (which can be translated into the single-electron ground state energy) is plotted as a function of B. A clear cusp in the magnetic field dispersion is seen at $B \approx 8$ T. This is in good agreement with the critical field obtained from far-infrared spectroscopy. Moreover, the ground state dispersion in the entire field range can well be accounted for when using the ring parameters obtained from the far-infrared investigations (solid lines in Fig. 2).

To summarize this section we conclude that the spectroscopic data from two complementary spectroscopic techniques together with detailed model calculations clearly show that the ring shape of the self-organized islands shown in Fig. 1(a) can be preserved during overgrowth. Furthermore, the topology of the islands translates into a ring-shaped, attractive potential for carriers in the grown structure, which confirms the conjecture that the volcano islands are In-rich.

4. Possible mechanisms

We will now turn to the discussion of different mechanisms that could account for the transformation from partially capped dots to ring shaped islands. A scenario based on In diffusion, is outlined in Fig. 3. In Fig. 3(a) a possible configuration of the island structure, immediately after the deposition of the GaAs capping layer, is shown [22]. Here we have assumed that the GaAs capping layer does not cover the islands as continuous ‘blanket’ and that the tops of the InAs dots remain uncovered. This is supported by the fact that the strain relaxation of the InAs at the top of the

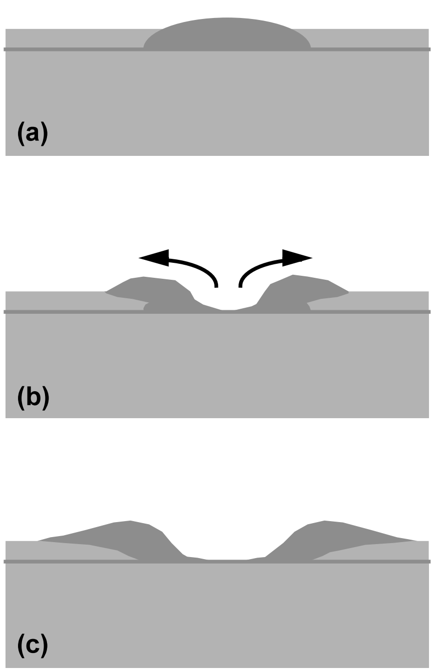

Fig. 3. Model for a diffusion-driven transformation from dots to rings. During annealing, the In-rich material (dark gray) diffused away from the original location of the dot, whereas the less mobile Ga-rich material (light gray) remains mostly unchanged. A possible $In_yGa_{1-y}As$ alloy formation is not shown for simplicity (after Ref. [22]).

islands makes them unfavorable locations for GaAs growth. Furthermore, spectroscopic data of samples with different capping layer thicknesses support this picture [22].

As we have mentioned above, at the chosen growth temperature of 530 °C[1] the In is quite mobile on the surface. The Ga atoms on the other hand diffuse only little after they have been incorporated into the crystal lattice. Consequently, as shown in Fig. 3(b), the In atoms can diffuse out of the partially capped islands onto the surface of the deposited GaAs. This outward diffusion might be enhanced by the chemical forces that promote the In surface segregation that has been observed in the InAs/GaAs system [23]. As schematically depicted in Fig. 3(c), after an appropriate annealing time the outward diffused In will form an In-rich $In_yGa_{1-y}As$ rim around the void at the former location of the dot. Together, the rim and the void can account for the observed volcano shape of the islands.

The fact that diffusion plays an important role in the formation of the rings can be deduced from their elongated shape which reflects the fact that indium diffuses much faster along $[1-10]$ than in the 110-direction.

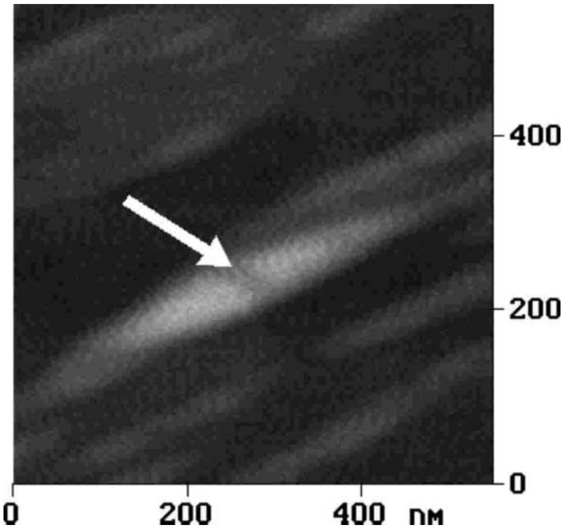

Fig. 4. Atomic force micrograph of an extremely elongated island, showing the influence of anisotropic diffusion.

An extreme example is given in Fig. 4, where the length along the $[1-10]$-direction has increased to 600 nm. We attribute the strongly elongated island shape on this surface to the fact that the sample was grown on a substrate holder with a large thermal mass which did not allow the sample to cool down fast enough to preserve the ring morphology. The hole in the center is hardly discernible (see arrow), but it clearly does not reflect the strong anisotropy of the outer edge of the island. This is in agreement with the assumption in Fig. 3 that the GaAs of the covering layer hardly diffuses during the formation of the rings. An image of islands with similar properties (strongly elongated outer edge, almost circular inner hole) can be found in reference [13].

Even though the scheme outlined in Fig. 3 is able to account for a number of experimental observations, there is strong evidence that diffusion alone is not sufficient to explain the phenomenon of ring formation. As mentioned above, the coexistence of islands with and without center holes (cf. Fig. 1(b)) suggests an abrupt process rather than a continuous hole formation by diffusion. Also, the almost sharp outer island edge seen in Fig. 1 seems to be incompatible with a diffusive picture. Note that in the direction of fast diffusion the island edges are much more washed out in Fig. 4. Finally, the strongly different aspect ratios seen in Figs. 1 and 4 indicate that an additional mechanism contributes to the ring formation. For a purely diffusive process, the aspect ratio of the islands would be given by the different diffusion constants along 110 and $[1-10]$ and would thus be constant over time.

There are striking similarities between the present, ring-shaped $In_yGa_{1-y}As$ islands and structures observed in dewetting processes [24]. This leads us to conclude that

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[1] It should be pointed out here that a large uncertainty is associated with the growth temperature, particularly when comparing different growth chambers. For reproducibility, we chose 530 °C as the temperature at which the GaAs (2×4) reconstruction changes to $c(4 \times 4)$ under moderate As flux.

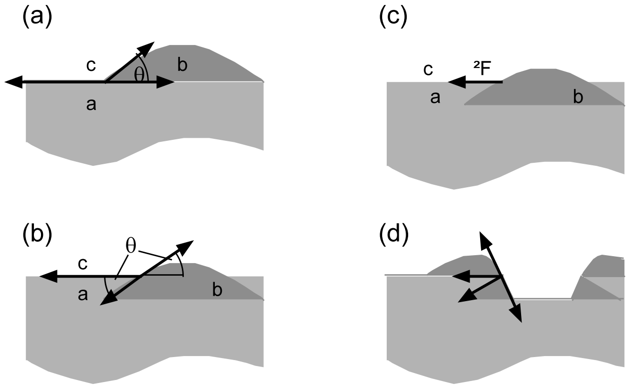

Fig. 5. Model of ring formation, promoted by a wetting droplet instability. See text for details (after Ref. [25]).

surface and interface forces (which are already important ingredients in the understanding of the Stranski–Krastanov mechanism of dot formation) also play a role in the transformation from dots to rings in partially capped islands.

The argument is outlined in Fig. 5 [25]. In a purely solid-on-liquid picture, the wetting angle θ of uncapped islands is given by the balance of forces at the foot of the island,

$$\gamma_{\mathrm{ac}} = \gamma_{\mathrm{bc}} \cos(\theta) + \gamma_{\mathrm{ab}} \tag{1}$$

Here, γ_{ij} is the force at the interface between materials i and j, which in the present system are a = GaAs, b = InAs, and c = As-vapor. For partially capped islands, the corresponding relation would read

$$\gamma_{\mathrm{ac}} = \gamma_{\mathrm{bc}} \cos(\theta) - \gamma_{\mathrm{ab}} \cos(\theta), \tag{2}$$

which is obviously incompatible with Eq. (1). The configuration in Fig. 5(b) therefore leaves an unbalanced net outward force $\Delta F = \gamma_{\mathrm{ab}}(1 + \cos\theta)$ (Fig. 5(c)), which promotes the disintegration of the partially capped islands and the formation of the center hole. Fig. 5(d) schematically illustrates that the balance of forces might indeed be better fulfilled in volcano-shaped islands.

We thus conclude that the formation of the rings is promoted by at least two different mechanisms. One is the diffusion of atoms out of the In-rich dots onto the Ga-rich surface. The other is dewetting, promoted by the imbalance of surface and interface forces acting upon the partially capped islands.

5. Summary

In summary, we have investigated the morphological transformation from partially capped InAs/GaAs self-assembled dots to ring-shaped islands. There is strong evidence that the center hole of the rings is at the former location of the dots and that the volcano-shaped rim surrounding the center hole consists of In-rich material, which has been transported outward from the original dot location. We conclude that the driving mechanisms for this material transport are diffusion and wetting droplet instability. The rings-shaped islands can be overgrown and incorporated into active devices in which the not simply connected topology is preserved and reflected in the magnetic-flux dependence of the electrical and optical properties.

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References

[1] For a review, see e.g.: D. Bimberg, M. Grundmann, N.N. Ledentsov, Quantum Dot Heterostructures, Wiley, Chichester, 1992.
[2] J. Tersoff, C. Teichert, M. Lagally, Phys. Rev. Lett. 76 (1996) 1675.
[3] Q. Xie, A. Madhukar, P. Chen, N.P. Kobayashi, Phys. Rev. Lett. 75 (1995) 2542.
[4] T. Walther, A.G. Cullis, D.J. Norris, M. Hopkinson, Phys. Rev. Lett. 86 (2001) 2381.
[5] W. Wu, J.R. Tucker, G.S. Solomon, J.S. Harris Jr., Appl. Phys. Lett. 61 (1992) 2814.
[6] D. Leonard, et al., Appl. Phys. Lett. 63 (1993) 3203.
[7] I. Kegel, et al., Phys. Rev. Lett. 85 (2000) 1694.
[8] Y. Nabetani, T. Ishikawa, S. Noda, A. Sakai, J. Appl. Phys. 76 (1994) 347.
[9] A. Lorke, et al., Phys. Rev. Lett. 84 (2000) 2223.
[10] J.M. Garcia, et al., Appl. Phys. Lett. 71 (1997) 2014.
[11] Q. Xie, P. Chen, A. Madhukar, Appl. Phys. Lett. 65 (1994) 2051.
[12] M.E. Rubin, et al., Phys. Rev. Lett. 77 (1996) 5268.
[13] I. Kamiya, I. Tanaka, H. Sakaki, J. Cryst. Growth 201/201 (1999) 1146.
[14] J.-S. Lee, H.-W. Ren, S. Sugou, Y. Masumoto, J. Appl. Phys. 84 (1998) 6686.
[15] A. Lorke, R.J. Luyken, Physica B 256–258 (1998) 424.
[16] J.M. Garcia, et al., Appl. Phys. Lett. 72 (1998) 3172.
[17] R.J. Warburton, et al., Nature 402 (2000) 926.
[18] H. Pettersson, et al., Physica E 6 (2000) 510.
[19] T. Chakraborty, P. Pietilainen, Phys. Rev. B 50 (1994) 8460.
[20] A. Emperador, M. Pi, M. Barranco, A. Lorke, Phys. Rev. B 62 (2000) 4573.
[21] M. Büttiker, Y. Imry, R. Landauer, Phys. Lett. A 96 (1983) 365.
[22] A. Lorke, R.J. Luyken, J.M. Garcia, P.M. Petroff, Jpn. J. Appl. Phys. 40 (2000) 1857.
[23] See, e.g.: O. Brandt, et al., Appl. Phys. Lett. 61 (1992) 2814.
[24] See, e.g.: S. Herminghaus, et al., Science 282 (1998) 5390 (and references therein).
[25] R. Blossey, A. Lorke, submitted for publication.