Jpn. J. Appl. Phys. Vol. 40 (2001) pp. 1857–1859
Part 1, No. 3B, March 2001

Growth and Electronic Properties of Self-Organized Quantum Rings

Axel LORKE[1,2], R. Johannes LUYKEN[2,3], Jorge M. GARCIA[4] and Pierre M. PETROFF[4]

[1]*Laboratorium für Festkörperphysik, Gerhard-Mercator-Universität, Lotharstr. 17-ME 245, 47048 Duisburg, Germany*
[2]*Sektion Physik und CeNS, LMU München, Geschwister-Scholl-Platz 1, 80539 München, Germany*
[3]*Infineon Technologies, Corporate Research, Otto Hahn Ring 6, 81730 München, Germany*
[4]*Materials Department and QUEST, University of California, Santa Barbara, CA 93106, USA*

(Received September 11, 2000; accepted for publication November 2, 2000)

A method is described which can be used to grow self-organized, nanoscopic InGaAs ring structures on GaAs substrate. Starting from self-organized InAs dots, the crucial step for the ring formation is a short annealing phase after the dots have been covered by a thin GaAs layer. Spectroscopic data are reviewed which show that the ring morphology can be preserved even after the InGaAs islands have been covered by additional cladding layers for the realization of electronically or optically active devices.

KEYWORDS: quantum dots, Aharonov-Bohm effect, rings, spectroscopy, self-organized growth

1. Introduction

The fascination that is associated with the investigation of self-organized semiconductor nanoparticles is in part related to the fact that they can be considered as “artificial atoms”.[1–13] Unlike real atoms, however, semiconductor nano-structures can be subjected to external forces which can compete with the internal confinement. Controllable, external fields can therefore completely rearrange the quantum configuration in such artificial atoms or even change their “atomic number”, i.e. the number of confined carriers. Furthermore, semiconductor nano-structures offer flexibility with respect to the shape of the confining potential. They are therefore ideal model systems, also for studying topological quantum effects, like, *e.g.*, the Aharonov-Bohm effect, which cannot be observed in atomic or molecular structures because of the prohibitively large fields required.

Here, we will review the possibility of fabricating nm-size InGaAs rings in a self-organized fashion, using the Stranski-Krastanov growth mode. These nanorings are coherently strained, dislocation-free and thus well suited for investigating the electronic and optical properties of not simply connected quantum systems in high magnetic fields.[14–23]

2. Growth

The growth (by molecular-beam epitaxy) of the nano-rings starts with depositing, on a GaAs (100) substrate, a GaAs buffer of sufficient thickness to clean and smooth out the surface. Next, the substrate temperature is lowered to 530°C, a temperature which can easily be reproduced by observing, under moderate As flux, the change in surface reconstruction from (2×4) to $c(4 \times 4)$. Following the usual Stranski-Krastanov growth procedure,[3] 1.7 monolayers of InAs are deposited. This is only little more than is necessary to observe the transition from two-dimensional to three-dimensional growth and results in a dilute (density $\approx 1 \times 10^{10}\,\mathrm{cm}^{-2}$), disordered array of quantum dots, 6 nm in height and 20 nm in diameter (Fig. 1(a)).

In order to study their electronic and optical properties, these dots are commonly overgrown by a thick GaAs cladding layer. The only step necessary to induce the transformation from dots to rings (Fig. 1(b)) is a short growth interruption after a thin part (thickness θ ca. 1–4 nm) of the cladding layer has been deposited.[26] In fact, for a direct comparison between rings and dots, samples of *identical* layer sequence can be grown, one with and one without growth interruption.

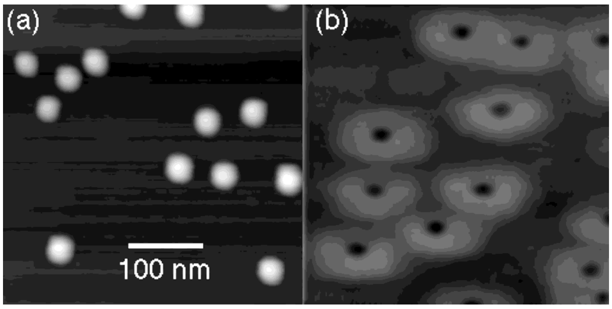

Fig. 1. Atomic force micrographs of self-organized InGaAs nanostructures. (a) shows Stranski-Krastanov-grown InAs dots with a height of approximately 6 nm and a diameter of around 20 nm. In (b), dots similar to the ones shown in (a) were covered with $\theta = 4$ nm of GaAs and annealed for ≈30 s. Ring-shaped islands are formed with a height of ≈2 nm.

The fact that giving the material extra time to rearrange has such dramatic consequences on the island morphology is an interesting finding in itself, since it indicates that the overgrowth of InAs with GaAs *is not an equilibrium process*.

A conclusive model of the ring formation has not yet been developed. A possible scenario, however, is given in Fig. 2. Here, we assume that the GaAs deposited after the dot formation only covers the sides of the dots, so that for thin GaAs deposition, the InAs remains exposed at the top of the islands (Fig. 2(a)). This is supported by the fact that ring formation no longer takes place when the thickness θ of the GaAs layer becomes comparable to the height of the dots (see also Fig. 3). During the annealing period, indium, which at 530°C is much more mobile on the surface than gallium, will diffuse outward and leave a void at the former location of the InAs island (Fig. 2(b)). Just as a nanoscopic volcano erupting, the outdiffusing material, which is possibly alloyed with the surrounding substrate to form less mobile InGaAs, creates a nanoisland of ring shape (Fig. 2(c)).

It is worth noting that in their original paper,[27] Stranski and Krastanov discussed a liquid-on-liquid type model which included neither strain nor diffusion. Similarly, the ring formation in the InGaAs system might be closely related to the physics of *dewetting*. In dewetting experiments on completely unrelated material combinations, ring-shaped voids of striking similarity have been observed.[28]

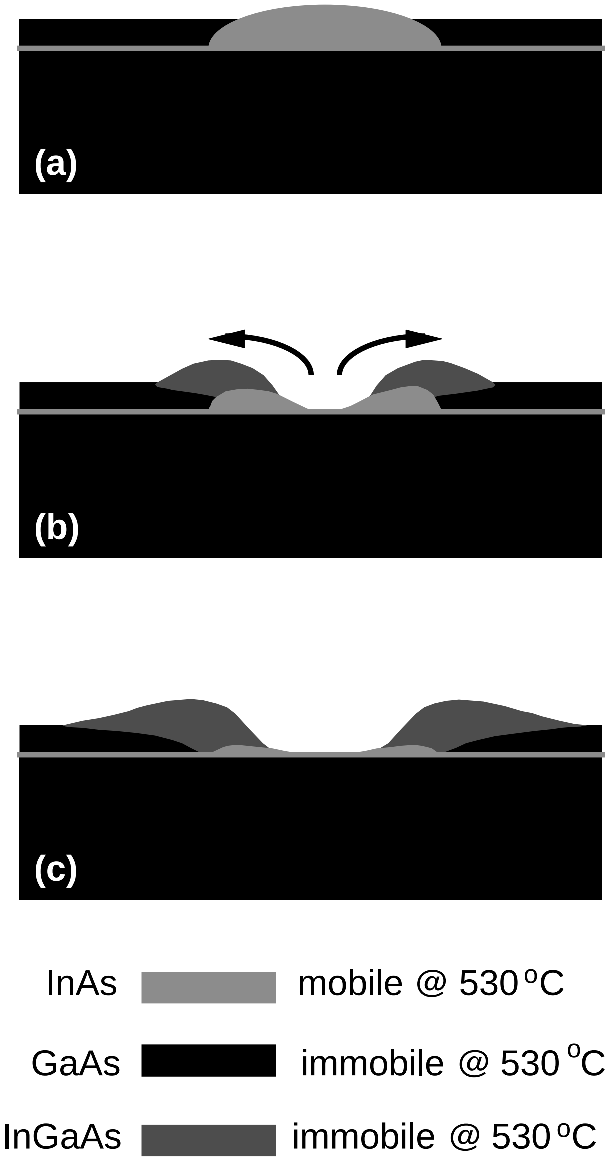

Fig. 2. Possible scenario for the self-organized ring formation: (a) InAs dots are partly covered by a thin layer of GaAs. (b) during the annealing time, In diffuses away from its original location and forms a volcano-like structure on the surface (c).

3. Spectroscopy

Knowing that the coverage of InAs islands with GaAs can so profoundly change their morphology, it is of great interest to know how far the ring shape can be preserved when the samples are further covered for the fabrication of optically[29] or electronically[30] active devices.

The electronic topology inside InGaAs rings, embedded in a GaAs/AlGaAs field-effect structure, was determined by capacitance–voltage (C–V) and far-infrared (FIR) spectroscopy.[31]

Figure 3 demonstrates the strong influence that the growth interruption has on the single-electron charging spectrum of InGaAs islands. Trace A shows the C–V spectrum of a ring sample with $\theta = 1$ nm. Two single-electron charging peaks (arrows) can be identified just before the onset of a large increase in capacitance, caused by the formation of a two-dimensional electron gas in the field-effect transistor structure.[10,13] For comparison, the well-investigated C–V characteristic of a typical dot sample is shown in C. Here, six single-electron charging events can be identified, which are grouped according to the quantum dot shell structure. For details on intraband quantum dot spectroscopy, see refs. 10–13.

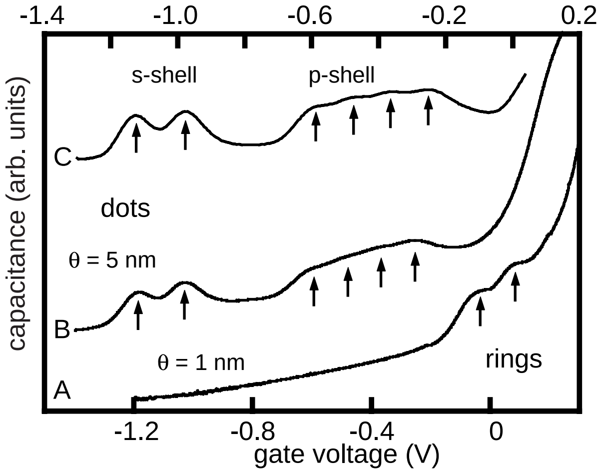

Fig. 3. Capacitance-voltage traces of self-organized InGaAs nanostructures (traces A, B: lower x-axis, trace C: upper axis). Each maximum (arrows) corresponds to the additional loading of a single electron per dot/ring. Compared to dots, in rings the loading of the first electron is shifted to higher gate voltages (corresponding to higher energies). This is caused by the reduced thickness of the rings (cf. Fig. 1). The $\theta = 5$ nm sample, which otherwise was grown identical to the $\theta = 1$ nm sample, shows that when the GaAs cladding layer is sufficiently thick, no ring formation takes place.

It can be seen in Fig. 3 that the C–V profile of a sample with $\theta = 5$ nm (trace B), that was otherwise grown identical to the $\theta = 1$ nm sample, exhibits no change in the electronic properties compared to the dot sample (trace C). This indicates that 5 nm of GaAs is sufficient to cover the InAs dots, so that no outdiffusion and ring formation can occur (see the discussion above).

Figure 3 only shows that the electronic states in the $\theta = 1$ nm sample are *different* from those in the unannealed dots. The *ring* shape can be verified by investigating their Aharonov-Bohm-type magnetic properties. One manifestation of the Aharonov-Bohm effect is the stepwise increase of the ground-states' angular momentum with increasing magnetic field as more and more flux quanta penetrate the interior of the ring.[15–17] This increase is intimately related to the so-called persistent currents in meso- and nanoscopic rings.[14,18–22] As discussed *e.g.* in ref. 32 these ground-state transitions take place in not simply connected fermionic systems but not in (simply connected) dots.

The ground-state energy of the single electron state in the ring sample can be determined by plotting the position of the lowest charging peak (cf. Fig. 3) as a function of the magnetic field. The result is shown in Fig. 4, (solid data points). Also shown (solid lines) is a calculation[31] of the single-particle energy in a ring with a confining potential[33] $U(r) = \frac{1}{2}m^*\omega_0^2(r - R_0)^2$, where $2R_0 = 28$ nm is the diameter and $\hbar\omega_0 = 12$ meV the characteristic energy of the quantum wire loop. Here, we have used an effective mass $m^* = 0.07m_e$, a value obtained from spectroscopic investigations of InAs quantum dots.[12,13]

The comparison between the data and the calculations, together with the evaluation of independent data from FIR spec-

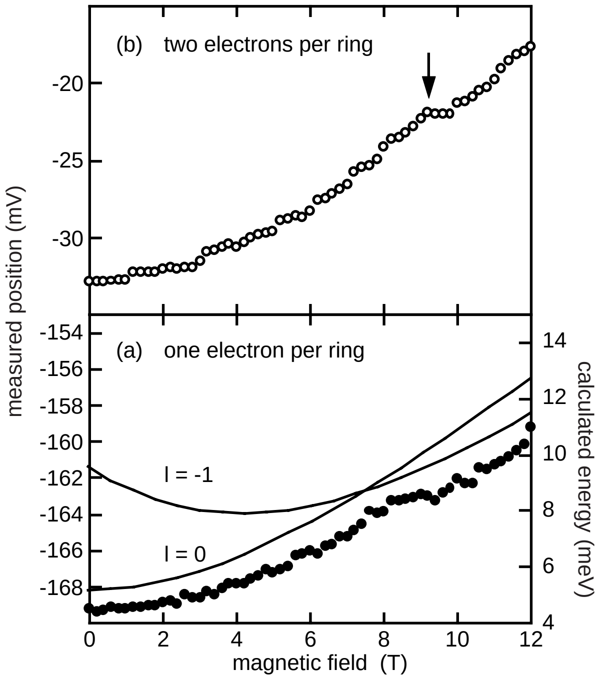

Fig. 4. (a) magnetic-field-induced shift of the single electron charging peak. At ≈8 T, a change in slope can be identified in the magnetic field dispersion. This corresponds to a change from a zero angular momentum ($\ell = 0$) ground state to a ground state with $\ell = -1$. Solid lines display the results of a single-particle calculation of the ground-state energy in a potential $U(r) = \frac{1}{2}m^*\omega_0^2(r - R_0)^2$, with $R_0 = 14$ nm and $\hbar\omega_0 = 12$ meV. (b) magnetic-field-induced shift of the second charging peak. A possible ground state transition at ≈ 9 T is indicated by the arrow.

troscopy31) shows that at a magnetic field B of approximately 8 T, i.e., when roughly one flux quantum penetrates the interior of the ring, a ground state transition takes place (arrow) from a zero angular momentum ($\ell = 0$) state to one with $\ell = -1$.

The small electronic ring diameter of about 30 nm at first seems surprising, considering the large outer diameter of the islands in Fig. 1. Analysis of the island height profile, however, shows that, as in a real volcano, the highest elevation is close to the inner hole, which is roughly 20 nm in diameter. Since the electronic states are expected to be confined to the parts where the InGaAs island is the thickest, the height profile and the electronic determination of the ring diameter are in satisfactory agreement.

Figure 4 also displays the magnetic-field-induced shift of the 2-electron ground state (open symbols). Here, again, a kink can be seen in the data (arrow), however, at a somewhat larger magnetic field $B \approx 9$ T. This is in agreement with recent calculations by Emperador *et al.*,34) which also predict a ground state-transition at $B \approx 3$ T. Even though some structure is present in the data at this field, the present experimental results cannot unambiguously confirm this prediction.

In conclusion, we have presented structural and spectroscopic data which show that it is possible to fabricate ring-shaped electronic structures, using the now well-established Stranski-Krastanov growth procedure.

We would like to thank A. O. Govorov, T. Chakraborty, J. P. Kotthaus, M. Barranco, and V. Gudmundsson for valuable discussions. Financial support by the German Science Foundation (DFG) is gratefully acknowledged.

1) L. Goldstein, F. Glas, J. Y. Marzin, M. N. Charasse and G. Le Roux: Appl. Phys. Lett. **47** (1985) 1099.
2) Q. Xie, A. Madhukar, P. Chen and N. P. Kobayashi: Phys. Rev. Lett. **75** (1995) 2542.
3) D. Leonard, M. Krishnamurthy, C. M. Reaves, S. P. Denbaars and P. M. Petroff: Appl. Phys. Lett. **63** (1993) 3203.
4) J.-Y. Marzin, J. M. Gerard, A. Izrael, D. Barrier and G. Bastard: Phys. Rev. Lett. **73** (1994) 716.
5) M. Grundmann, J. Christen, N. N. Ledentsov, J. Böhrer, D. Bimberg, S. S. Ruvimov, P. Werner, U. Richter, U. Gösele, J. Heydenreich, V. M. Ustinov, A. Yu. Egorov, A. E. Zhukov, P. S. Kop'ev and Zh. I. Alferov: Phys. Rev. Lett. **74** (1995) 4043.
6) S. Fafard, K. Hinzer, S. Raymond, M. Dion, J. McCaffrey, Y. Feng and S. Charbonneau: Science **274** (1996) 1350.
7) R. J. Warburton, C. S. Dürr, K. Karrai, J. P. Kotthaus, G. Medeiros-Ribeiro and P. M. Petroff: Phys. Rev. Lett. **79** (1997) 5282.
8) L. Landin, M. S. Miller, M.-E. Pistol, C. E. Pryor and L. Samuelson: Science **280** (1998) 262.
9) M. Bayer, O. Stern, P. Hawrylak, S. Fafard and A. Forchel: Nature **405** (2000) 923 and references therein.
10) H. Drexler, D. Leonard, W. Hansen, J. P. Kotthaus and P. M. Petroff: Phys. Rev. Lett. **73** (1994) 2252.
11) G. Medeiros-Ribeiro, D. Leonard and P. M. Petroff: Appl. Phys. Lett. **66** (1995) 1767.
12) M. Fricke, A. Lorke A, J. P. Kotthaus, G. Medeiros-Ribeiro and P. M. Petroff: Europhys. Lett. **36** (1996) 197.
13) B. T. Miller, W. Hansen, S. Manus, R. J. Luyken, A. Lorke, J. P. Kotthaus, S. Huant, G. Medeiros-Ribeiro and P. M. Petroff: Phys. Rev. B **56** (1997) 6764.
14) M. Büttiker, Y. Imry and R. Landauer: Phys. Lett. A **96** (1983) 365.
15) A. G. Aronov and Yu. V. Sharvin: Rev. Mod. Phys. **59** (1987) 755.
16) T. Chakraborty and P. Pietiläinen: Phys. Rev. B **50** (1994) 8460.
17) L. Wendler and V. M. Fomin: Phys. Stat. Sol. (b) **191** (1995) 409.
18) L. P. Lévy, G. Dolan, J. Dunsmuir and H. Bouchiat: Phys. Rev. Lett. **64** (1990) 2074.
19) V. Chandrasekhar, R. A. Webb, M. J. Brady, M. B. Ketchen, W. J. Gallagher and A. Kleinsasser: Phys. Rev. Lett. **67** (1991) 3578.
20) D. Mailly, C. Chapelier and A. Benoit: Phys. Rev. Lett. **70** (1993) 2020.
21) A. F. Morpurgo, J. P. Heida, T. M. Klapwijk, B. J. van Wees and G. Borghs: Phys. Rev. Lett. **80** (1998) 1050.
22) R. Schuster, E. Buks, M. Heiblum, D. Mahalu, V. Umansky and H. Shtrikman: Nature **385** (1997) 417.
23) A. Emperador, M. Barranco, E. Lipparini, M. Pi and Ll. Serra: Phys. Rev. B **59** (1999) 15301.
24) I. Magnúsdóttir and V. Gudmundsson: Phys. Rev. B **60** (1999) 16591.
25) L. Wendler, V. M. Fomin, A. V. Chaplik and A. O. Govorov: Phys. Rev. B **54** (1996) 4794. and references therein.
26) J. M. García, G. Medeiros-Ribeiro, K. Schmidt, T. Ngo, J. L. Feng, A. Lorke, J. Kotthaus and P. M. Petroff: Appl. Phys. Lett. **71** (1997) 2014.
27) I. N. Stranski and L. Krastanow: Akad. Wiss. Lit. Wien, Math.-Natur. Kl. IIb **146** (1938) 797.
28) see, *e.g.*: S. Herminghaus, K. Jacobs, K. Mecke, J. Bischof, A. Fery, M. Ibn-Elhaj and S. Schlagowski: Science **282** (1998) 5390 and references therein.
29) H. Pettersson, R. J. Warburton, A. Lorke, K. Karrai, J. P. Kotthaus, J. M. Garcia and P. M. Petroff: Physica E **6** (2000) 510.
30) A. Lorke and R. J. Luyken: Physica B **256** (1998) 424.
31) A. Lorke, R. J. Luyken, A. O. Govorov, J. P. Kotthaus, J. M. Garcia and P. M. Petroff: Phys. Rev. Lett. **84** (2000) 2223.
32) R. Benoist and W. Zwerger: Z. Phys. B **103** (1997) 377.
33) V. Halonen, P. Pietiläinen and T. Chakraborty: Europhys. Lett. **33** (1996) 377.
34) A. Emperador, M. Pi, M. Barranco and A. Lorke: Phys. Rev. B **62** (2000) 4573.