Growth and annealing of InAs quantum dots on pre-structured GaAs substrates

M. Helfrich[1], D. Z. Hu[1], J. Hendrickson[2], M. Gehl[2], D. Rülke[1], R. Gröger[3], D. Litvinov[4], S. Linden[5], M. Wegener[1], D. Gerthsen[4], T. Schimmel[3], M. Hetterich[1], H. Kalt[1], G. Khitrova[2], H. M. Gibbs[2], and D. M. Schaadt[1]

[1] *DFG-Center for Functional Nanostructures (CFN) and Institut für Angewandte Physik, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany*
[2] *College of Optical Sciences, University of Arizona, Tucson, AZ 85721, USA*
[3] *Institute of Nanotechnology (INT) and Institut für Angewandte Physik, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany*
[4] *Laboratorium für Elektronenmikroskopie, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany*
[5] *Physikalisches Institut, University of Bonn, 53113 Bonn, Germany*

Semiconductor quantum dots (QDs) have attracted a lot of attention due to their unique properties in the past two decades. Not only do they exhibit a delta-function-like density of states, which makes them interesting for laser applications, but moreover they can generate entangled photon pairs and act as single photon sources and are thus promising candidates for quantum information schemes. Early on, QDs were grown by self-assembly. Properties, such as dot sizes or dot densities, can be altered by choosing the proper growth parameters. However, this alone does not provide any control over quantum dot positions, since self-assembly is inherently a random process. With regard to quantum information applications it is essential to have an ability to define arbitrary device architectures, i.e. to control the location of each individual QD with the prospect of scalability. But not solely applications generate a desire to control QD nucleation sites. Experiments with single QDs to study their physical properties, especially coupling to cavities, require physical access to single dots which is generally accompanied by a tedious search for the right dot.

Techniques to precisely position QDs have therefore been elaborated in the past decade. Top-down techniques such as electron beam lithography (EBL), local oxidation or mechanical nano-indentation have proven to be viable in order to define QD nucleation sites [1-3]. Common to those approaches is the creation of small holes on the substrate surface which leads to selective QD nucleation at the desired locations. Pre-structuring is commonly performed *ex-situ* and usually involves several process steps. Besides intended surface manipulation, contamination can occur. Therefore, great care has to be taken with regard to surface cleanliness prior to regrowth in order to inhibit unintended QD nucleation caused by defects. In addition, the optical properties are very sensitive to defects as well, so that site-selective QDs have inferior optical quality compared to self-assembled ones. The main reason is attributed to a change of morphology at the hole site. The defects originate from the regrowth interface [4]. This obstacle is often circumvented by growing QD stacks consisting of a QD seed layer and an as large as possible spacer layer [5]. Reducing the dot density and controlling the occupation number of QDs per site was, however, not improved that way alone.

A different approach to access the aforementioned quantities makes use of the fact that QDs undergo morphological changes during annealing. At lower temperatures they tend

to ripen whereas they dissolve at higher temperatures [6, 7]. By choosing the right annealing conditions it should therefore be possible to control, to some extent, the final QD size as well as the QD distribution and the occupation number per site. Annealing studies on unstructured substrates have already confirmed an increase in QD size uniformity [8, 9].
InAs QDs are grown by molecular beam epitaxy (MBE) on pre-structured (100) GaAs substrates. Small holes of about 70 nm in diameter are defined on the substrate surface by EBL and wet chemical etching.

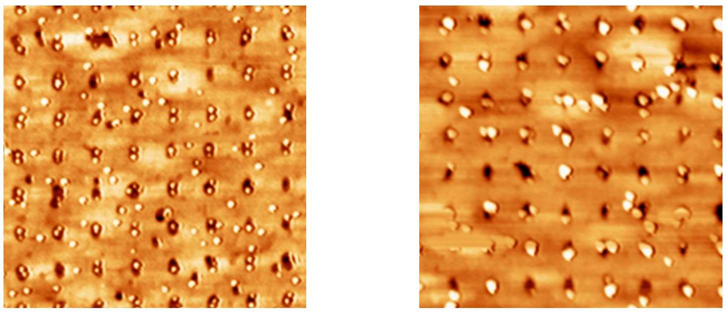

Fig. 1: AFM images (2 µm x 2 µm) of two samples with InAs QDs grown on patterned GaAs substrates; as grown (left), annealed (right).

In this study we investigate the effect of *in-situ* annealing, which is performed right after QD growth, on the QD distribution and occupation number per site as well as the QD density on the pre-structured substrate. Initially, where mainly two QDs occupy a hole we see a morphological transition after annealing resulting in single QDs per hole, i.e. two dots merge into one, as is observed in Figure 1. Since the original two QDs are close to each other inside the hole they easily coalesce to form one large dot during the annealing process. The annealed QDs exhibit an increase in size compared to the original dots. However, preliminary micro-photoluminescence (µ-PL) measurements reveal no distinct difference between as grown and annealed QDs.
In addition, we investigate the origin of defects which create unintentional holes besides the defined holes. It is found that the holes form at the regrowth interface and can act as QD nucleation sites as well. The substrate preparation process is therefore analysed carefully in order to identify the reasons for the defects to occur. The morphology of the QDs is characterised by atomic force microscopy (AFM) and transmission electron microscopy. The optical properties are analysed by µ-PL spectroscopy.

[1] S. Jeppesen et al., *Appl. Phys. Lett.* **68**, 2228 (1996)
[2] O. G. Schmidt et al., *Surf. Sc.* **514**, 10 (2002)
[3] P. Atkinson et al., *Physica E* **32**, 21 (2006)
[4] P. Atkinson et al., *C. R. Physique* **9**, 788 (2008)
[5] T. J. Pfau et al., *Appl. Phys. Lett.* **95**, 243106 (2009)
[6] H. Lee et al., *Appl. Phys. Lett.* **71**, 2325 (1997)
[7] C. Heyn, *Phys. Rev. B* **66**, 075307 (2002)
[8] D. M. Schaadt et al., *Appl. Phys. A - Mat. Sci. Proc.* **83**, 267 (2006)
[9] D. Z. Hu et al., *J. Crystal Growth* **312**, 447 (2010)