APPLIED PHYSICS LETTERS **95**, 243106 (2009)

Site-controlled InAs quantum dots grown on a 55 nm thick GaAs buffer layer

T. J. Pfau,[1,a)] A. Gushterov,[1] J. P. Reithmaier,[1] I. Cestier,[2] G. Eisenstein,[2] E. Linder,[3] and D. Gershoni[3]
[1]*Technische Physik, Institute of Nanostructure Technologies and Analytics, University of Kassel, Heinrich-Plett-Str. 40, D-34132 Kassel, Germany*
[2]*Electrical Engineering Department, Technion, Haifa 32000, Israel*
[3]*Solid State Institute and Physics Department, Technion, Haifa 32000, Israel*

(Received 19 August 2009; accepted 28 October 2009; published online 14 December 2009)

We present site-controlled low density InAs quantum dots grown by molecular beam epitaxy with a template based overgrowth technique allowing enlarged buffer layers upto 55 nm. Growing a seeding layer of InAs quantum dots in etched holes reduces closing of the holes, so that a second layer of InAs quantum dots can be aligned to the holes after a buffer layer overgrowth. Confocal microphotoluminescence measurements show a significant decrease of the low temperature photoluminescence linewidth of the quantum dots to an average value of ~500 μeV and a minimum width of 460 μeV. This is to be compared to 2 to 4 meV of quantum dots grown on thin buffer layers. This improvement is due to the enlarged distance to residual defects at the overgrown surface. © *2009 American Institute of Physics*. [doi:10.1063/1.3265918]

The combination of InAs quantum dots (QDs) and high-Q resonators like photonic crystals (PCs),[1–5] micropillars,[6,7] and microdisks[8] have been studied in recent years. Such structures serve for cavity quantum electrodynamics experiments and for advanced applications such as single photon sources and threshold-less nanolasers for quantum information processing and cryptography.

To achieve high performance cavities with embedded single QDs, the exact position of the QDs must be known. One common way is to preselect buried QDs by detection of a strain coupled surface dot using scanning electron microscopy[1,2] (SEM) or atomic force microscopy[9] (AFM).

A better approach, which offers flexibility in construction of complex nano devices, is to grow site-controlled QDs (SCQD) on prepatterned substrates.

Both PC cavities and micropillars with SCQD were reported recently.[3,10] The site-controlled growth is realized by the overgrowth of a prepatterned surface. The decisive problem is the short distance of the SCQDs to the residual defects at the interface region, where the dots are formed. The defects attract carriers that compete with the QD potential well and hence reduce not only the emission probability of adjacent SCQD, but also cause a fluctuating charge background. The resulting QDs have rather poor optical properties with emission linewidths of several microelectronvolts [full width at half maximum (FWHM)], in comparison to a few microelectronvolts in conventionally grown self-assembled QDs. To overcome this problem, approaches with thick buffer layers were reported lately. In one case, deeply etched holes on (111)*B* GaAs with a 27 nm buffer layer[3] were used. A second scheme stacked three layers of QDs separated by 10 nm GaAs barriers to increase the total buffer layer thickness to 28 nm.[10]

In this letter, we present a method to increase the buffer layer thickness to 55 nm by using a stack of only two QD layers. This is surprising, because the maximum vertical alignment of InAs QDs based on strain coupling is limited to about 20 nm.[11] Following MBE growth, the SCQDs were characterized by confocal microphotoluminescence (μPL). The emission spectra of single QDs show a minimum luminescence linewidth of 460 μeV (FWHM) due to the large distance to the defect containing interface. These SCQDs are of sufficiently high optical quality to warrant their use within high-Q PhC cavities.

The first step in the process is the definition of marks into poly methyl methacrylate (PMMA) by electron beam lithography on a 300 nm thick GaAs buffer layer. Wet chemical etching with 0.05 mol/l I_2 in potassium oxalate buffer with pH=4 was used to define the marks. Next, hole prepatterning was processed by electron beam lithography of PMMA relatively to the marks. The holes were etched by a solution of 2×10^{-4} mol/l I_2 containing also 6×10^{-4} mol/l KI and 0.5 mol/l potassium oxalate buffer with pH=4, which reduces the under etching below the resist and provides deep holes. The average value of the circular hole depth was 55 ± 5 nm, as measured by AFM and the average diameter was approximately 120 ± 10 nm, measured by SEM. The resist on the samples was removed by acetone for 2 min and by 180 °C hot N-methyl-2-pyrrolidone for 10 min. The sample was mounted in the MBE system and cleaned *in situ* by an atomic hydrogen source from carbon residuals and from oxides.

A 5 nm GaAs buffer layer was grown at a rate of 1 μm/h with a V/III-ratio of 23 (beam equivalent pressure ratio). An InAs QD layer with a nominal thickness of 0.9 nm was grown next at 530 °C at a rate of 160 nm/h and a V/III-ratio of about 77. The structure was covered by 50 nm GaAs on top of which 0.9 nm InAs QDs were deposited. The AFM image in Fig. 1 reveals, that the holes have not vanished despite of the 55 nm GaAs and that every hole is occupied at least by one QD. The hole diameter is

---

[a)] Author to whom correspondence should be addressed. Electronic mail: pfau@ina.uni-kassel.de. Tel.: +49-561-804-4248. FAX: +49-561-804-4136.

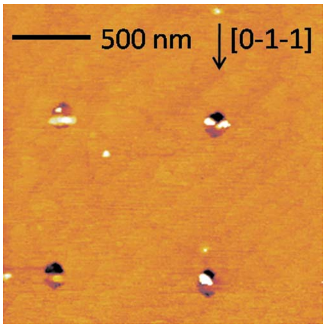

FIG. 1. (Color online) InAs QDs after growth of 55 nm GaAs buffer layer. The holes have a 1 µm spacing (2×2 µm^2).

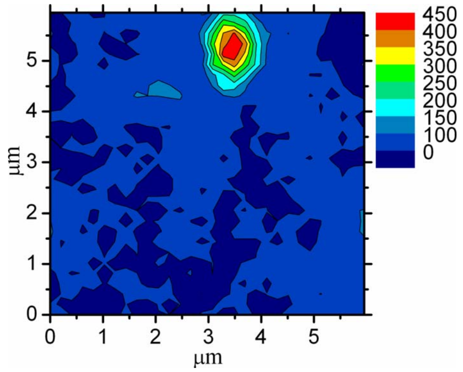

FIG. 3. (Color online) Intensity map of a 6×6 µm^2 area integrated at the energy of 1.32 eV.

187±9 nm in [0-1-1]-direction and 137±13 nm in [0-11]-direction. The hole depth is difficult to measure because of the QDs but is in most of the holes between 7 and 8 nm. Further measurements show, that most of the holes (75%) contain two dots, 20% one dot and 5% three dots. Although the double occupancy of flat but very wide holes is not unusual and especially observed at flat, wet chemically etched holes,[12] the occurrence of additional dots between the holes and the high amount of double occupied holes point out, that the growth conditions were not perfect. Further overgrowth tests with 55 nm thick GaAs layers result in very flat holes while holes are fully closed at a GaAs layer thickness exceeding 70 nm (not shown).

Normally, the overgrowth of holes takes place very fast.[13] In 1994, Xie[14] reported that around an InAs QD the layer thickness of a GaAs/AlGaAs quantum well structure is reduced. He attributed this effect to the strain of the InAs island. We think that the hole conservation after growth of 55 nm GaAs on our sample is traced back to the very big hole depth of 55 nm and to the reduction of GaAs rate by island induced strain. First, the InAs island induced strain reduces the hole filling effect and therefore the hole depth remains constant or increases despite of increasing GaAs buffer thickness around the hole. With increasing buffer layer thickness, the QD gets also overgrown and consequently the effect of the strain is reduced resulting in a flattened growth front.

The sample used for optical characterization differed slightly from the one described above which was used for morphology studies. After prepatterning, a 5 nm GaAs layer was grown followed by 0.8 nm InAs, 50 nm GaAs, 0.9 nm InAs, 75 nm GaAs. On top, 0.8 nm InAs layers were grown for morphological low density check. Figure 2 displays schematically the cross section of the sample. The smaller nominal InAs thickness of the first QD layer should result in smaller dots and therefore a shorter emission wavelength. The GaAs growth rate was 435 nm/h at 530 °C and a V/III-ratio of 44, the InAs rate was 165 nm/h at a V/III-ratio of 73.

The optical properties of the SCQDs were characterized using µPL in a confocally configured system. A microscope objective was used to focus the laser pump beam to a spot size of less than 3 µm and the photoluminescence (PL) signal emitted from the sample was spatially filtered by a pinhole with a diameter of 75 µm and detected using a charge coupled device detector placed at the output of a 0.1 nm resolution monochromator. The sample can be moved in the cryogenic chamber by piezoelectric motors allowing for PL mapping with a spatial resolution better than 200 nm.

The sample was excited by a 632 nm continuous-wave He-Ne laser at a power of 400–500 nW. Individual QDs were optically addressed by integrating a scan over a single energy window. Figure 3 shows an exemplary intensity mapping of a 6×6 µm^2 area, integrated at the energy of 1.32 eV. A single QD was isolated by filtering this particular energy. Figure 4 shows the measured spectrum emitted by this specific QD indicating a clear single peak near 1.32 eV. The zoom spectrum shown in the inset of Fig. 4 reveals a narrow linewidth of about 460 µeV. This is the narrowest value achieved in this experiment. Other QDs on the same sample had slightly wider widths with an average of 500 µeV and one dot with a width as large as 580 µeV. While these linewidths are broad compared to that of conventional self organized QDs, they definitely show a marked narrowing compared with previous SCQDs thereby demonstrating the

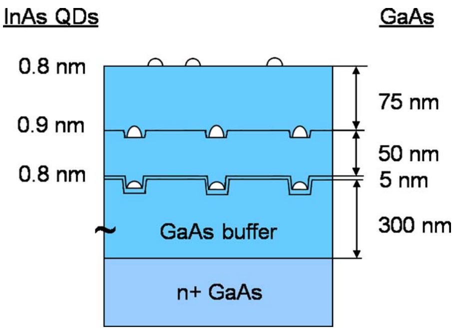

FIG. 2. (Color online) Schematic cross section of the sample analyzed by photoluminescence measurements. The first and the second QD layer are site-controlled, whereas the third QD layer is distributed randomly.

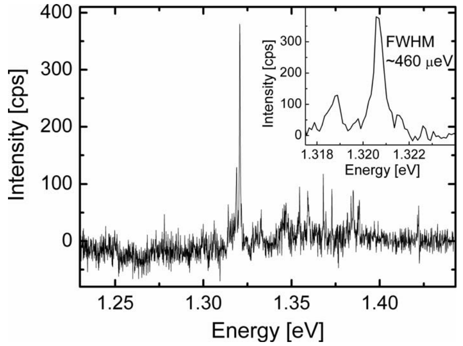

FIG. 4. Cryogenic temperature photoluminescence spectrum of the QD selected in Fig. 3. The inset shows a zoom of the spectrum, revealing a linewidth of about 460 µeV.

advantage of the buffering procedure introduced here.

In summary, site-controlled growth of InAs QDs on prepatterned surfaces covered with a thick buffer layer of 55 nm GaAs could be established by using an initial seeding layer of InAs QDs in wet-chemically etched holes. A stack of only two QD layers reduces the problems to distinguish the dots from the different layers optically. The increased buffer layer thickness results in dots of high optical quality with a minimum linewidth of 460 μeV, which is lower than commonly observed in SCQDs.

This work was supported by the European Commission through the projects “QPhoton” and “Gospel.”

[1] A. Badolato, K. Hennessy, M. Atatüre, J. Dreiser, E. Hu, P. M. Petroff, and A. Imamoglu, Science **308**, 1158 (2005).

[2] K. Hennessy, A. Badolato, P. M. Petroff, and E. Hu, Photonics Nanostruct. Fundam. Appl. **2**, 65 (2004).

[3] P. Gallo, M. Felici, B. Dwir, K. A. Atlasov, K. F. Karlsson, A. Rudra, A. Mohan, G. Biasiol, L. Sorba, and E. Kapon, Appl. Phys. Lett. **92**, 263101 (2008).

[4] T. Sünner, S. Schneider, M. Strauß, A. Huggenberger, D. Wiener, S. Höfling, M. Kamp, and A. Forchel, Opt. Lett. **33**, 1759 (2008).

[5] S. M. Thorn, M. T. Rakher, H. Kim, J. Gudat, W. T. M. Irvine, P. M. Petroff, and D. Bouwmeester, Appl. Phys. Lett. **94**, 111115 (2009).

[6] C. Santori, D. Fattal, J. Vučkovlć, G. S. Solomon, and Y. Yamamoto, Nature (London) **419**, 594 (2002).

[7] J. P. Reithmaier, G. Sęk, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. V. Keldysh, V. D. Kulakovskll, T. L. Reinecke, and A. Forchel, Nature (London) **432**, 197 (2004).

[8] J. P. Reithmaier, Semicond. Sci. Technol. **23**, 123001 (2008).

[9] K. Hennessy, A. Badolato, M. Winger, D. Gerace, M. Atatüre, S. Gulde, S. Fält, E. L. Hu, and A. Imamoglu, Nature **445**, 896 (2007).

[10] C. Schneider, T. Heindel, A. Huggenberger, P. Weinmann, C. Kistner, M. Kamp, S. Reitzenstein, S. Höfling, and A. Forchel, Appl. Phys. Lett. **94**, 111111 (2009).

[11] Q. Xie, A. Madhukar, P. Chen, and N. Kobayashi, Phys. Rev. Lett. **75**, 2542 (1995).

[12] P. Atkinson, O. G. Schmidt, S. Bremner, and D. A. Ritchie, C. R. Phys. **9**, 788 (2008).

[13] H. Heidemeyer, C. Müller, and O. G. Schmidt, J. Cryst. Growth **261**, 444 (2004).

[14] Q. Xie, P. Chen, and A. Madhukar, Appl. Phys. Lett. **65**, 2051 (1994).