Site-controlled InAs/GaAs quantum dots emitting at telecommunication wavelength

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Abstract: We demonstrate site-controlled InAs/GaAs quantum dot emission at 1.3 µm telecommunication wavelength. The samples were fabricated by molecular beam epitaxy on patterned substrates, which have been prepared by electron beam lithography and wet chemical etching. By embedding a single layer of positioned quantum dots in a strain reducing InGaAs quantum well layer, we successfully shifted the emission band beyond the important telecommunication wavelength of 1.3 µm. Furthermore, the resulting deep carrier confinement allowed us to preserve strong quantum dot luminescence up to room temperature.

Semiconductor quantum dots (QDs) have been established as solid-state sources of non-classical light due to their capability to emit single photons with high purity on demand[1], even under electrical injection[2]. Such sources are in particular desirable for applications in the field of quantum key distribution (QKD)[3]. However, long-distance quantum communication requires emitters matching the absorption and dispersion minimum of optical fibers, i.e. 1.3 µm and 1.55 µm. In fact, first demonstrations of QKD using QD sources in the telecommunication windows have already been demonstrated[4,5]. However, one persisting problem of solid state single photon sources based on QDs is the source brightness, which hardly exceeds 2-4 % for a single QD embedded in bulk material. This can be significantly improved by embedding the QD in an engineered photonic environment, such as a photonic waveguide or a microcavity, where extraction efficiencies in excess of 70 % have been demonstrated[6]. A scalable and parallel fabrication routine of such bright sources, in particular emitting at telecommunication wavelength, is highly desirable, yet a very challenging task. Site-controlled quantum dots (SCQDs) are very promising candidates to realize such a system, since they can be integrated in photonic structures in a scalable manner without the necessity to retrieve individual emitters after

growth. Several approaches were attempted to push the emission band of InAs/GaAs QDs towards the telecommunication wavelength range. Single InAs QDs on GaAs emitting at 1.3 µm were fabricated by using strain reducing layers[7,8] and employing dilute nitrite materials[9]. However, a scalable system with InAs SCQDs at telecommunication wavelength has so far only been shown on InP substrate[10,11].

In this paper we show emission at 1.3 µm from InAs SCQDs on GaAs substrate at room temperature. Our samples are grown via molecular beam epitaxy (MBE) on pre-patterned substrates. The substrates contain an array of nanoholes, which were fabricated by the means of electron beam lithography and wet chemical etching[12]. These substrates were overgrown with a 5 nm GaAs buffer layer, a 3 nm AlAs diffusion barrier and a 4 nm GaAs smoothing layer, as schematically depicted in Fig. 1(a). For the SCQDs a dot-in-a-well (DWELL) structure is used. The InAs SCQDs (1.1 nm InAs) are grown on 1 nm $In_{0.2}Ga_{0.8}As$ and are capped with 5 nm $In_{0.2}Ga_{0.8}As$ and 50 nm GaAs at a substrate temperature of 520°C. The quantum well (QW) serves as a strain reducing layer so that the QD size and the indium content can be enlarged[13]. As QDs are likely to nucleate at crystal defects, the positioning is ensured by the nanoholes.

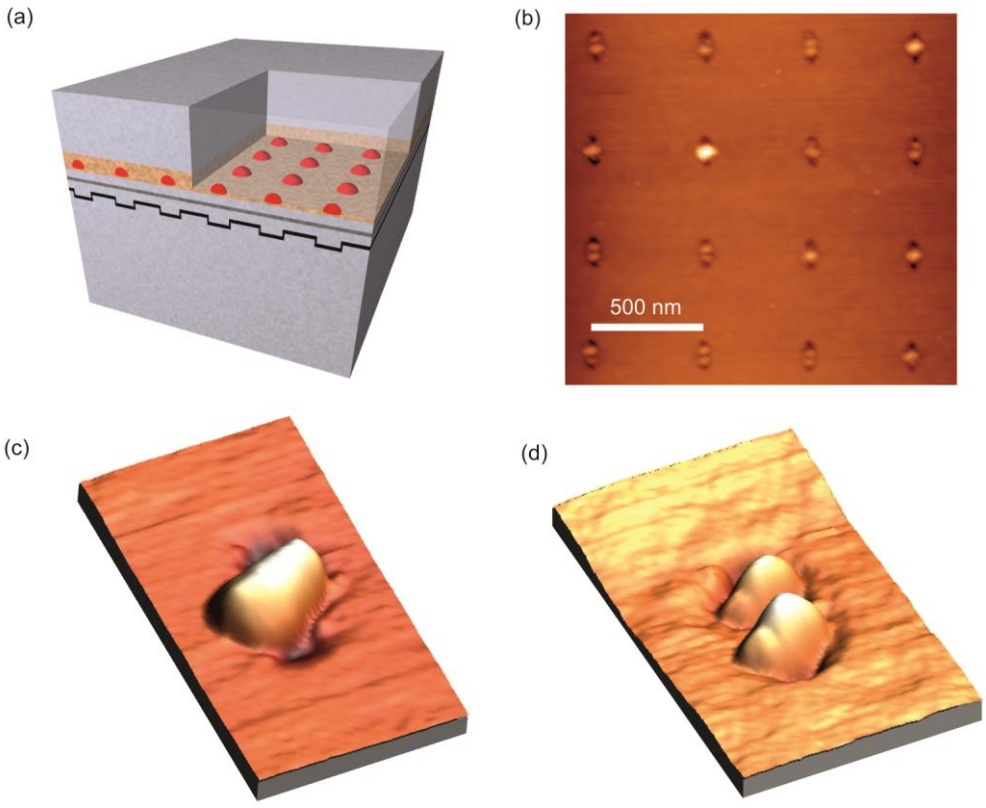

Fig. 1. (a) Schematic drawing of the sample layout. b) AFM image of 4x4 SCQDs with a pitch of 500 nm. c) Detailed AFM image of a single SCQD. (d) Detailed AFM image of a QD molecule.

Fig. 1(b) shows an atomic force microscopy (AFM) image of a sample of uncapped SCQD grown on 1 nm $In_{0.2}Ga_{0.8}As$ with a 500 nm pitch between the SCQDs. The initially circular shaped nanoholes with a diameter of about 100 nm and a depth of 20 nm after wet chemical etching evolve to elongated holes during overgrowth. AFM investigations after the QD growth reveal that the nanoholes are elongated in (011) direction with dimensions of about 120 nm length and 60 nm width. All nanoholes are occupied by well positioned QDs or QD molecules and there are only few interstitial QDs with a very small size. The formation of single SCQDs versus QD molecules strongly depends on the growth conditions, the amount of deposited indium, the growth dynamics and the size and shape of the nanoholes[14,15]. In our specific case we had to find a compromise for the substrate temperature (520°C): Higher substrate temperatures result in a larger migration length during the InAs deposition process, leading to a suppression of molecule formation. However, Indium desorption and inter-diffusion processes with the barrier material become strongly pronounced, leading to a reduction of the indium content which results in a blue-shifted QD emission. Furthermore the elongated shape of the nanoholes facilitates QD molecule formation. These conditions cause QD molecule formation in 55% of the nanoholes in good accordance with previous reports on similar sample layouts and growth conditions[16]. A representative single SCQD is shown in Fig. 1(c) with dimensions of about 60 nm in length, 40 nm in width and 9 nm in height. In Fig. 1(d) an AFM image of a QD molecule is depicted, consisting of two QDs with approximately 35 nm in length, 20 nm in width and 7 nm in height. Both, the site-controlled QDs and the QD molecules are elongated in the (0-11) direction with an angle of about 20°. A detailed analysis of the particular shape, crystalline core and stain for single quantum dots and quantum dot molecules can be found in previous reports[17]. For single quantum dots and molecules a strong similarity in these characteristics was found, resulting in similar photoluminescence spectra[18]. However the smaller size of the QD molecules causes a spectral blue-shift which we believe contributes to the spectral ensemble broadening of the QD emission in our spectra.

On an as-grown sample that was capped with 5 nm $In_{0.2}Ga_{0.8}As$ and 50 nm GaAs, the photoluminescence (PL) of the SCQD ensemble was measured with an InGaAs charge-coupled device detector under above bandgap excitation (532 nm). For each single spectrum the temperature of the sample in the cryostat was raised from 4 K up to 290 K. In the waterfall plot of Fig. 2(a) the temperature dependent PL spectra are depicted. The PL peak intensity of the SCQDs is about a factor of 3 smaller than the peak intensity of self-assembled In(Ga)As QDs in the same spectral range at cryogenic temperatures. We attribute this to a reduced quantum efficiency caused by non-radiative carrier escape, as previously reported on similar structures emitting at 950 nm[19].

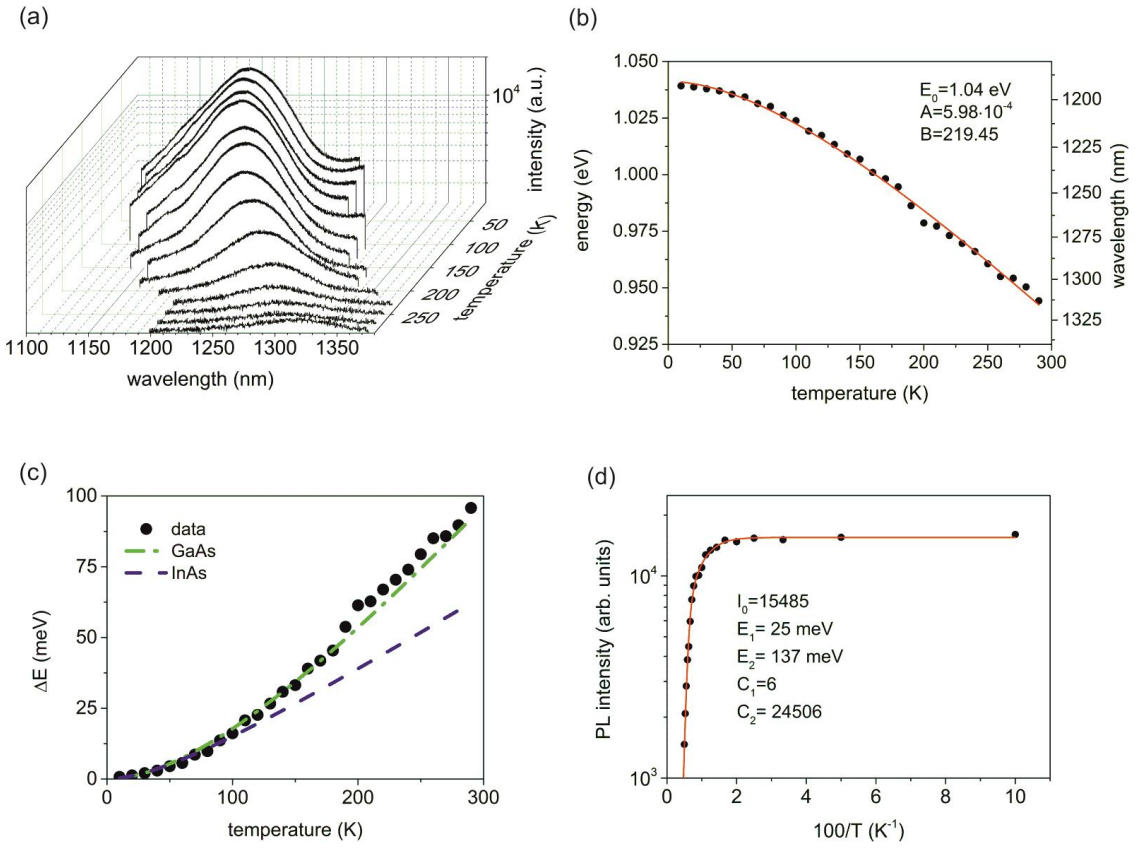

Fig. 2. (a) Waterfall plot of the temperature dependent PL spectra. (b) Varshni fit of the temperature dependent peak-wavelength. (c) Relative change of the emission energy ΔE of the measured data, GaAs bulk and InAs bulk material. (d) Arrhenius plot of the temperature dependent PL intensity.

The temperature dependent redshift of the peak-wavelength follows the empirical Varshni equation[20] $E(T) = E_0 - (A \cdot T^2) \cdot (B + T)^{-1}$ as shown in Fig. 2(b). The data can be well reproduced with $E_0 = 1.04\,eV$ (corresponding to the emission energy at 0 K), $A = 5.98 \cdot 10^{-4}$ and $B = 219.45$. At a temperature of 270 K the emission wavelength passes 1.3 µm what constitutes the first experimental measurement of SCQD emission at telecom wavelength on this material system. Fig. 2(c) shows the relative temperature dependent emission energy of the measured data, GaAs bulk and InAs bulk material referring to the particular band gap at 0 K. The redshift of the data follows the shape of the branch of GaAs. The intensity decrease of the QD emission at higher temperatures due to thermal quenching can be analyzed by calculating the activation energies. A simple rate model[21] with two activation energies is used to fit the experimental data:

$$I(T) = \frac{I_0}{1 + C_1 \cdot e^{-\frac{E_1}{k_b \cdot T}} + C_2 \cdot e^{-\frac{E_2}{k_b \cdot T}}} \qquad (1)$$

In Eqn. (1) the temperature dependent intensity $I(T)$ is described by I_0, the extrapolated intensity at 0 K, two activation energies E_1 and E_2 and two parameters C_1 and C_2, which represent the strength of the two loss channels. The Arrhenius plot in Fig. 2(d) reveals the activation energies $E_1 = 25\,meV$ for lower temperatures and $E_2 = 137\,meV$ for the high temperature regime. These values are comparable with values in previous reports on self-assembled InAs QDs in DWELL structures with values of 25 - 80 meV[22-26] for low temperatures and 170 - 289 meV[24-26] for high temperatures. The lower activation energy E_1 can be attributed to nonradiative recombination of excitons in QDs due to defects in close proximity[23]. Such a defect can be introduced via the etched surface below the SCQDs and is a possible reason that E_1 ranges at the lower end of the reported values. For the high temperature region the dominating loss channel is a reduced exciton capturing in the QDs, which is limited by thermal escape from the surrounding $In_{0.2}Ga_{0.8}As$ QW into GaAs bulk[26]. A simple calculation shows that E_2 nearly matches the difference of the binding energies $\Delta E = 145\,meV$ for a 6 nm $In_{0.2}Ga_{0.8}As$ QW (1.358 eV) and the GaAs bulk exciton (1.503 eV) at 80 K.

In conclusion, we described the fabrication and morphological analysis of site-controlled InAs QDs on GaAs substrate in a DWELL structure. This design allows an enlargement of the QD size and the indium content. Temperature dependent photoluminescence measurements were performed followed by a detailed analysis of the loss channels. For the first time PL emission of site-controlled InAs QDs at a telecommunication wavelength of 1.3 µm could be shown on this material system. This result is an important step towards a scalable system of InAs QDs for the use in semiconductor devices aiming for the application in telecommunication systems.

Acknowledgments

This work was financially supported by the German Ministry of Education and Research (BMBF) via the project QuaHL-Rep and by the State of Bavaria.

References

[1] P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, Lidong Zhang, E. Hu, A. Imamoglu; Science 290, 2282-2285 (2002)

[2] T. Heindel, C. Schneider, M. Lermer, S. H. Kwon, T. Braun, S. Reitzenstein, S. Höfling, M. Kamp, and A. Forchel; Appl. Phys. Lett. 96, 011107 (2010)

[3] E. Waks, K. Inoue, C. Santori, D. Fattal, J. Vuckovic, G. S. Solomon and Y. Yamamoto; Nature 420, 762 (2002)

[4] P. M. Intallura, M. B. Ward, O. Z. Karimov, Z. L. Yuan, P. See, A. J. Shields, P. Atkinson, and D. A. Ritchie; Appl.Phys. Lett. 91, 161103 (2007)

[5] J. Tatebayashi, M. Nishioka, and Y. Arakawa; Appl. Phys. Lett. 78, 3469 (2001)

[6] J. Claudon, J. Bleuse, N. S. Malik, M. Bazin, P. Jaffrennou, N. Gregersen, C. Sauvan, P. Lalanne and J.-M. Gérard; Nature Phot. 4, 174 - 177 (2010)

[7] B. Alloing, C. Zinoni, V. Zwiller, L. H. Li, C. Monat, M. Gobet, G. Buchs, A. Fiore, E. Pelucchi and E. Kapon; Appl. Phys. Lett. 86, 101908 (2005)

[8] M. B. Ward, O. Z. Karimov, D. C. Unitt, Z. L. Yuan, P. See, D. G. Gevaux, A. J. Shields, P. Atkinson and D. A. Ritchie; Appl. Phys. Lett. 86, 201111 (2005)

[9] M. Strauss, S. Höfling and A. Forchel; Nanotechnology, 20, 505601 (2009)

[10] M. E. Reimer, D. Dalacu, J. Lapointe, P. J. Poole, D. Kim, G. C. Aers, W. R. McKinnon and R. L. Williams; Appl. Phys. Lett. 94, 011108 (2009)

[11] H. Z. Song, T. Usuki, S. Hirose, K. Takemoto, Y. Nakata, N. Yokoyama and Y. Sakuma; Appl. Phys. Lett. 86, 113118 (2005)

[12] C. Schneider, A. Huggenberger, T. Sünner, T. Heindel, M. Strauß, S. Göpfert, P. Weinmann, S. Reitzenstein, L. Worschech, M. Kamp, S. Höfling and A. Forchel; Nanotechnology 20, 434012 (2009)

[13] K. Nishi, H. Saito, S. Sugou and J.-S. Lee; Appl. Phys. Lett. 74, 1111 (1999)

[14] R. Songmuang, S. Kiravittaya, and O. G. Schmidt; Appl. Phys. Lett. 82, 17 (2003)

[15] C. Schneider, A. Huggenberger, M. Gschrey, P. Gold, S. Rodt, A. Forchel, S. Reitzenstein, S. Höfling, and M. Kamp; Phys. Stat. Sol. A 209, No. 12, 2379–2386 (2012)

[16] P. Atkinson, S. Kiravittaya, M. Benyoucef, A. Rastelli, and O. G. Schmidt; Appl. Phys. Lett. 93, 101908 (2008)

[17] B. Krause, T. H. Metzger, A. Rastelli, R. Songmuang, S. Kiravittaya, and O. G. Schmidt; Phys. Rev. B 72, 085339 (2005)

[18] O.G. Schmidt, S. Kiravittaya, Y. Nakamura, H. Heidemeyer, R. Songmuang, C. Müller, N.Y. Jin-Phillipp, K. Eberl, H. Wawra, S. Christiansen, H. Gräbeldinger, H. Schweizer; Surface Science 514, 10–18 (2002)

[19] F. Albert, S. Stobbe, C. Schneider, T. Heindel, S. Reitzenstein, S. Höfling, P. Lodahl, L. Worschech, and A. Forchel; Appl. Phys. Lett. **96**, 151102 (2010)

[20] Y.P. Varshni; Physica 34, 149-154 (1967)

[21] D. Bimberg, M. Sondergeld and E. Grobe; Phys. Rev. B 4, 3451–3455 (1971)

[22] K. Mukai, N. Ohtsuka, and M. Sugawara; Appl. Phys. Lett. 70, 2416 (1997)

[23] R. Chen, H. Y. Liu, and H. D. Sun; J. Appl. Phys. 107, 013513 (2010)

[24] L. Seravalli, P. Frigeri, M. Minelli, P. Allegri, V. Avanzini, and S. Franchi; Appl. Phys. Lett. 87, 063101 (2005)

[25] L. Chen, V.G. Stoleru, D. Pan, E. Towe; J. Crystal Growth 242, 263–269 (2002)

[26] J. L. Casas Espinola, M. Dybic, S. Ostapenko, T. V. Torchynska and G. Polupan; Journal of Physics: Conference Series 61, 180–184 (2007)