Journal of Physics: Conference Series

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IOP Publishing
Journal of Physics: Conference Series **61** (2007) 889–893
doi:10.1088/1742-6596/61/1/177
International Conference on Nanoscience and Technology (ICN&T 2006)

Materials challenges for devices based on single, self-assembled InGaN quantum dots

Rachel A. Oliver[1], Anas F. Jarjour[2], Abbes Tahraoui[3], Menno J. Kappers[1], Robert A. Taylor[2], Colin J. Humphreys[1]

[1]Department of Materials Science and Metallurgy, University of Cambridge, Pembroke Street, Cambridge CB2 3QZ, UK
[2]Clarendon Laboratory, Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, UK
[3]National Centre for III-V Technologies, Department of Electronic and Electrical Engineering, University of Sheffield, Mappin Street, Sheffield, S1 3JD, UK

E-mail: rao28@cam.ac.uk

Abstract. Builiding on earlier studies of single InGaN quantum dots (QDs), we are considering their potential for use in blue- and green-emitting single photon sources. Envisaging a device based on a resonant cavity light emitting diode, we have studied the effect of growing QDs on an underlying AlN/GaN distributed Bragg reflector, and have shown that enhanced single QD emission may be obtained. Additionally, we have studied the effect of the growth and activation of a p-type cap on an underlying QD layer and have shown that the QDs survive the anneal process.

1. Introduction

The nitride materials system has unique properties which render its quantum structures particularly interesting for both fundamental study and device applications, but also poses more challenges than other III-V materials. Building on our studies of the properties of InGaN quantum dots (QDs) grown by a modified droplet epitaxy technique [1,2], we are considering potential designs for GaN-based single-photon sources. The feasibility of this approach has recently been demonstrated by Santori *et al*. [3], who have performed photon correlation studies of optically-pumped single GaN QDs which emit at ultra-violet wavelengths, and have demonstrated that single-photon emission from these structures is possible. Use of InGaN QDs could provide access to the blue and green spectral regions, providing a good match to commercially-available, highly sensitive, ultra-fast avalanche photodiodes.

In terms of simplicity and practicality [4], an electrically-driven source, based on a single QD has much to offer. A prototype of such a source has been constructed based on an InAs QD in the intrinsic region of a p-i-n diode [4]. We imagine a similar structure based on InGaN QDs, but in order to improve the efficiency of light extraction we envisage the use of a resonant cavity light emitting diode (RC-LED) structure, with the cavity employing $Al_xGa_{1-x}N$/GaN distributed Bragg reflectors (DBRs).

Realisation of such a concept poses a number of challenges to the crystal grower, two of which are considered here: (1) Growth of the $Al_xGa_{1-x}N$/GaN distributed Bragg reflector (DBR) underlying the QD layer may introduce an increased density of defects and a rougher surface morphology due to the strain between the layers. This may effect subsequent QD growth. (2) Under typical conditions for p-type GaN growth, the p-dopant (Mg) forms an electrically inactive complex with hydrogen. Acceptor activation then requires an extended high temperature anneal, which may damage the underlying QDs.

2. Experimental

All samples described here were grown by metal-organic vapour phase epitaxy (MOVPE) in a Thomas Swan 6 x 2", close-coupled showerhead reactor using tri-methyl indium (TMI), tri-methyl gallium (TMG), tri-methyl aluminium (TMA), ammonia, silane, and bis(cyclopentadienyl)magnesium (Cp_2Mg) as precursors, with nitrogen and hydrogen as carrier gases for InGaN and GaN growth respectively. The details of our QD growth methodology have been described elsewhere [1]. As in the earlier study, GaN pseudo-substrates grown on sapphire were employed. Surface morphologies were studied using a Veeco Dimension 3100 atomic force microscope (AFM) in TappingMode™.

Room temperature photo-luminescence (PL) was performed using a HeCd laser operating at 325 nm. Low temperature (~ 4.2K), spatially-resolved micro-PL measurements were made using 1 ps pulses from a tunable Ti:sapphire laser. Since, we have previously shown that two-photon excitation results in the suppression of the emission from the underlying quantum well to which the QDs are coupled and allows relatively strong QD emission to be observed [5], we have again employed this technique. A 36× achromatic microscope objective lens was used to focus the laser to a spot of ~2 μm, which along with the non-linear excitation allowed the resolution of single QD peaks. The same microscope objective lens was used to collect the PL.

Time-integrated PL spectra were recorded by using a cooled CCD mounted on a 0.3 m monochromator, with 600 or 1200 grooves/mm gratings. An automated mirror allowed the PL to be redirected to another apparatus at the exit of the monochromator in order to perform time-resolved measurements. A time-resolution of ~150 ps was achieved using a time-correlated single photon counting system based on a fast photomultiplier.

3. Results and Discussion

3.1. Quantum dot growth on a distributed Bragg reflector

As an initial foray into the growth of QDs on an underlying DBR, we have grown a simple 20 period AlN/GaN DBR structure on a GaN pseudo-substrate, with 45.5 nm GaN layers, and 51.3 nm AlN layers. Above the final AlN layer, a 91 nm GaN spacer was grown. The DBR was designed to have a maximum reflectivity of ca. 95% at 435 nm. In Figure 1, the morphology of the DBR sample is compared with that of the surface on which the QDs are usually grown – a GaN pseudo-substrate, with a terraced surface. The terraced surface has an rms roughness of 0.35 nm over a 3 μm x 3 μm area. The DBR sample, in comparison, has a much higher rms roughness (3.6 nm), and shows a network of raised features and trenches, rather than flat terraces.

A QD layer was then grown (see [1]) on top of a similar DBR structure, and compared with a QD layer grown on a GaN pseudo-substrate. The results (Figures 2 (a) and (b)) show the formation of small three-dimensional nanostructures in both cases, but other features of the morphology are rather different. For this QD growth route, pits are commonly seen in the InGaN layer underneath the 3D nanostructures, (Figure 2(a)), but for growth on the DBR surface, such pits are not clearly observed (Figure 2(b)) and the density of nanostructures is higher. The rougher morphology of the underlying

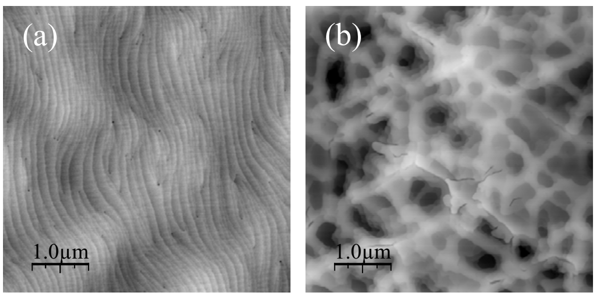

Figure 1. AFM images of (a) a standard GaN pseudo-substrate (image height, h = (image height, h = 3.16 nm) and (b) the surface of the DBR sample. (h = 27.65 nm). QD layers were later grown on sample surface similar to those shown.

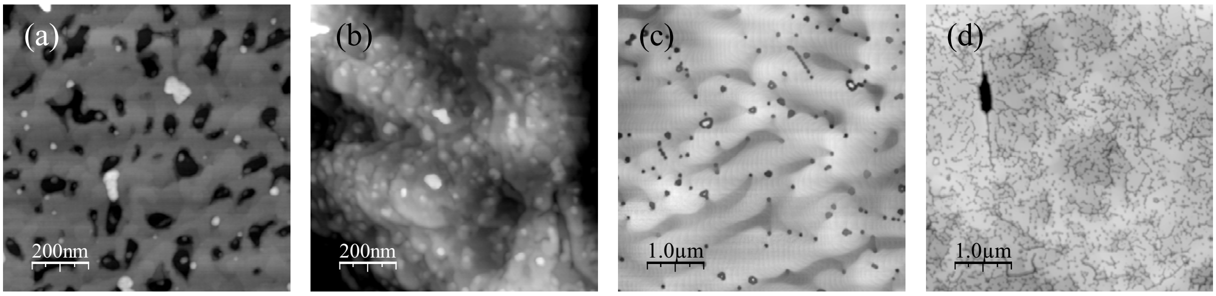

Figure 2. AFM images of (a) an InGaN QD epilayer grown on a standard GaN pseudo-substrate (h = 11.75 nm), (b) an InGaN QD epilayer grown on underlying AlN/GaN DBR (h = 21.49 nm), (c) a similar sample to that in (a) capped with 91 nm GaN (h = 10.48 nm) and (d) a similar sample to that in (b) capped with 91 nm GaN (h = 167.19 nm).

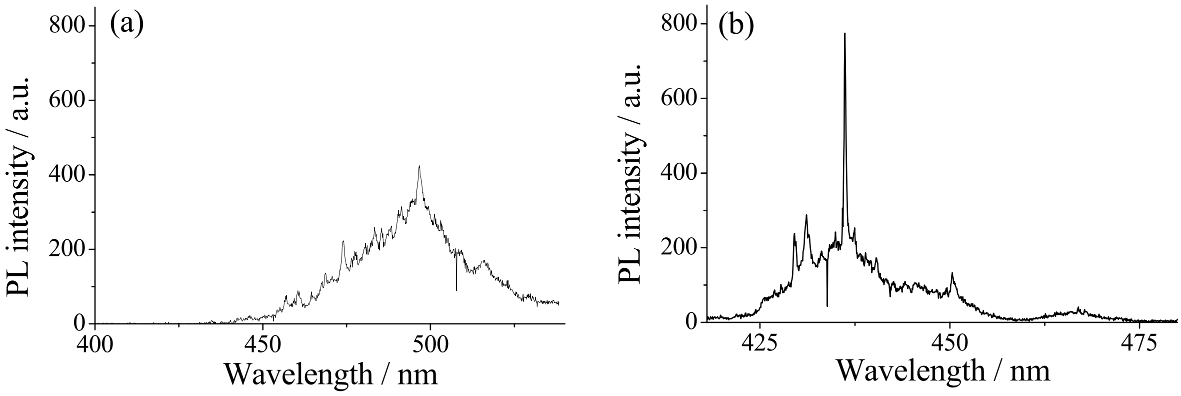

Figure 3. Low temperature micro-PL spectra for (a) capped QD sample grown on a standard GaN pseudo-substrate and (b) capped QD sample grown atop a DBR structure.

material is also evident in this case. The observed changes in the QD layer may be due to the rough and defected nature of the surface on which the QD layer was grown, which could result in changes in the number of sticking and nucleation sites, and lead to a change in the indium incorporation.

Another pair of samples was then grown, based on those shown in Figures 2 (a) and (b), in which a 91 nm GaN cap was added on top of the QD layer. The resulting morphologies are shown in Figures 2 (c) and (d). The sample grown on a GaN pseudo-substrate (Figure 2(c)) exhibits a wavy terraced surface with some pits or raised defects. The rms roughness over a 5 μm x 5 μm area is 1.2 nm. The sample grown on the DBR, on the other hand, is much rougher, with a web of interlinking small and large trenches, giving an rms roughness of 9.5 nm.

Room temperature PL measurements on the samples in Figures 2 (c) and (d) showed that the use of the underlying DBR led to a blue-shift of the luminescence peak from ca. 490 nm to ca. 445 nm, and to an increase in peak intensity by about a factor of 8. Subsequent low temperature micro-PL (Figure 3) revealed sharp luminescence peaks characteristic of single QDs for both samples, but the ratio of the QD signal to the background signal (arising from the non-uniform, quantum well-like layer) was at least a factor of 3 greater for the sample with the DBR. The observed differences in PL between the two samples are thought to be due to a combination of the increased extraction efficiency at shorter wavelengths due to the presence of the DBR and to a change in the indium content and morphology of the QD layer due to its being grown on a rough surface. Additionally, the strain in the DBR is partially relaxed by crack formation and the subsequent QD layer thus undergoes different strains and piezo-electric fields compared to the sample grown on the GaN pseudo-substrate. Nonetheless it is encouraging that enhanced QD emission may successfully be observed from the sample with the DBR.

However, the very rough top surface of the capped sample is unsuitable for subsequent growth of a top reflector, and development of a more sophisticated DBR structure is clearly required.

3.2. Quantum dot growth with a p-type cap layer

To investigate the effect of p-type capping on the QD layer, a p-i-n structure with a QD layer in the i-region has been grown. The sample was grown on an n-type GaN pseudo-substrate doped with 2 x 10^{18} cm^{-3} Si. Onto a similar n-type connecting layer, 50 nm of unintentionally-doped (u.i.d.) GaN was grown, followed by a QD layer, capped with a further 50 nm of u.i.d. GaN. A ca. 50 nm thick p-cap was then grown at 1000 °C, using molar gas flows of 355 μmol/minute TMG, 446 mmol/minute NH_3, and 1.6 μmol/minute Cp_2Mg, with H_2 as the carrier gas. The sample was then cooled to 780 °C and annealed for 1200 s in 20 slm N_2. A control p-type layer consisting of 500 nm of p-type GaN grown under similar conditions on a highly resistive substrate underwent Hall probe measurements which revealed a carrier density of ca. 1 x 10^{17} cm^{-3}. From secondary ion mass spectrometry data we estimate a Mg impurity concentration of ca. 3 x 10^{19} cm^{-3}.

Room temperature PL spectra of the control p-type layer (Figure 4) show a faint, defect-related, blue emission peaking at ca. 440 nm. In contrast, the p-i-n structure shows 17 times higher peak PL at a longer wavelength of ca. 455 nm under the same excitation power. Hence, the emission from the p-i-n structure may be largely attributed to the InGaN layer. For some InGaN/GaN structures, capping with p-type material results in a loss of optical output [6], but that is not the case for the QD layer. To investigate whether the QDs survive the annealing process, low temperature micro-PL was performed on the p-i-n structure. Using two-photon excitation, no emission was observed from the control p-type layer. Sharp peaks indicating the presence of individual QDs were observed in spectra from the p-i-n sample. As has been previously demonstrated for QDs without a p-type cap [5], using two-photon excitation it is possible to achieve a high QD-signal to background ratio. In the case of the p-i-n structure, the background may arise from two sources: the quantum well layer on which the QDs rest, and the defect-related emission from the p-cap. By tuning the excitation wavelength, it is possible to almost completely suppress this background, as is shown in Figure 5.

Further characterization of the QDs in the p-i-n structure was achieved using time-resolved PL (TRPL). In earlier studies of InGaN QDs without a p-type cap, we have usually only been able to measure the TRPL signal from QDs emitting at wavelengths below ca. 445 nm, and have observed lifetimes in the range 1 – 6 ns. For the p-i-n sample, we have been able to measure the TRPL from QDs with rather longer emission wavelengths (up to ca. 475 nm), and have seen significantly longer lifetimes, greater than 100 ns in some cases. In order to check that these long-lived emissions do not

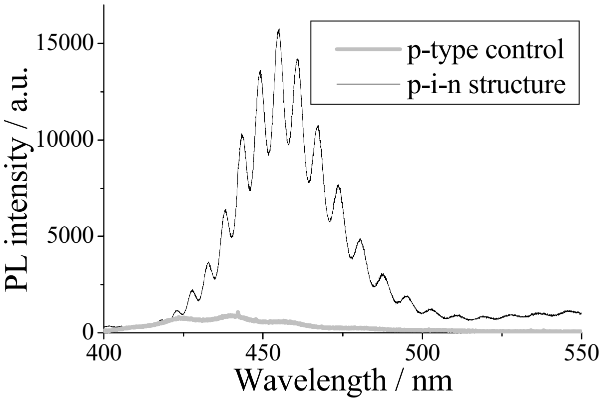

Figure 4. Room temperature PL spectra from the p-type control layer and the p-i-n structure under the same excitation power. Note the brighter emission at longer wavelength from the p-i-n structure compared to the control.

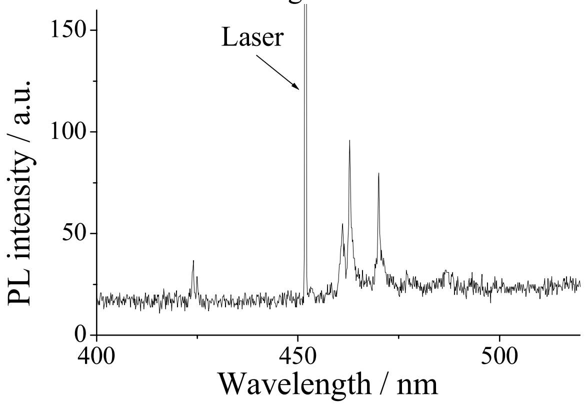

Figure 5. Low temperature micro-PL spectrum from the p-i-n diode structure for an excitation wavelength, λ of 900 nm and an excitation power, P of 60 mW. λ and P have been optimized to minimize the background signal intensity.

arise from defect states due to the presence of Mg in the cap layer, or to fields inherent to the p-i-n structure, we have performed similar TRPL on a sample which has undergone the same growth and heat treatment schedule but with no Mg admitted to the reactor. In this case, we are also able to measure the lifetime of QDs at long wavelengths (up to 505 nm) and again see lifetimes in excess of 100 ns. These QDs which emit at longer wavelengths should be either larger or contain more indium than those emitting at shorter wavelengths. The long lifetimes observed are then consistent with increased strain leading to an increased piezoelectric field and hence a smaller overlap of the electron and hole wavefunctions. It is currently unclear whether the QDs themselves have actually changed during the p-cap growth and annealing, or whether optimisation of the two-photon excitation methodology may also be contributing to these measurements.

Although these PL data, which illustrate that the QDs are not destroyed by the p-capping process, are encouraging, our final goal is to build a device which exhibits not only *photo*- but also *electro*-luminescence. Thus far, we have observed electroluminescence at room temperature. With appropriate processing, the p-i-n structure functions as a light emitting diode, emitting blue light. A forward voltage of 6.5 V gives a current of 20 mA. We plan to perform low temperature electroluminescence measurements on this sample in the future.

4. Conclusions

We have explored two crystal growth challenges relevant to the construction of an InGaN QD single photon source. We have successfully grown a QD layer on top of an AlN/GaN DBR and demonstrated that, compared to a sample with no DBR, brighter luminescence can be seen from single QDs, with a better QD signal to background ratio. Whilst this approach appears promising for future development of a cavity structure, currently the surface roughness of the capped QD layer grown on the DBR is rather high. Significant effort may be required to develop a more appropriate DBR, especially if strong coupling is to be achieved.

We have also considered the impact of the growth and activation of a p-type cap on an underlying QD layer, as a precursor to studying QD electroluminescence. We have shown that the QDs are not destroyed by the heating to which they are subjected, and that using annealed samples QDs with emission at longer wavelengths may be studied by TRPL, revealing long exciton lifetimes. Although these are only preliminary steps towards operational devices based on real QDs, the data presented here appear promising, since no substantial road-blocks have been discovered.

Acknowledgements

This research is part of the QIP IRC (www.qipirc.org) supported by EPSRC (GR/S82176/01). Growth studies are EPSRC funded under GR/S49391/01. We also acknowledge funding from the Clarendon Fund and the ORS scheme (AFJ), and from Peterhouse, Cambridge (RAO).

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