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Final Report: Nanopatterned Quantum Dot Lasers for High Speed, High Efficiency, Operation

## ABSTRACT

Quantum dot (QD) active regions hold potential for realizing extremely high performance semiconductor diode lasers. Unfortunately, these unique features of ideal QD active layers have not been fully realized to date. The most successful approach to date of forming QD's is self-assembly under the Stranski–Krastanow (SK) growth mode. However, this approach results in a relatively large distribution of QD sizes, leading to significant inhomogeneous broadening of the spectral gain. SK QDs inherently form on top of a two-dimensional "wetting layer", leading to weak electron and hole confinement to the QD, which results in low gain saturation. Here, we have investigated the use of dense nanoscale diblock copolymer lithography-based nanofabrication and selective quantum dot growth using metalorganic chemical vapor deposition (MOCVD). These methods allowed us to realize quantum dot active regions in which the injected carriers exhibit full three-dimensional nano-scale confinement and elimination of the wetting layer states. The objectives of this project were to develop lasers employing the nano-patterned QD active regions and investigate the characteristics of these novel active regions

# Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

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08/22/2011	3.00	Y. Huang, J. Kirch, L. J. Mawst, Y. Sin, J. H. Park, B. Foran, CC. Liu, P. F. Nealey, T. F. Kuech. Nanopatterned quantum dot active region lasers on InP substrates, Novel In-Plane Semiconductor Lasers X. 23-JAN-11, San Francisco, California, USA. : ,
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08/22/2011	6.00	T. F. Kuech, L. J. Mawst. Patterned InGaAs/InGaAsP/InP quantum dot active lasers using diblock copolymer lithography and selective area MOCVD growth, 2010 23rd Annual Meeting of the IEEE Photonics Society (Formerly LEOS Annual Meeting). 06-NOV-10, Denver, CO, USA. : ,
08/29/2012	7.00	Y. Huang, T. Kim, T. Garrod, L. J. Mawst, S. Xiong, P. F. Nealey, K. Schulte, T. F. Kuech. Nanofabrication of Quantum Dots on InP by In-Situ Etching and Selective Growth, IEEE CLEO 2012. 06-MAY-12, . : ,

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# Names of Personnel receiving masters degrees

NAME Yinggang Huang Total Number: 1

## Names of personnel receiving PHDs

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**Inventions (DD882)** 

#### **Scientific Progress**

See attached file

# **Technology Transfer**

Throughout the program I have interacted closely with Dr. Yongkun Sin at The Aerospace Corporation. They have contributed with TEM measurements of the QDs fabricated in this program.

While Stranski-Krastnow (SK) growth mode self-assembled QDs on GaAs have enjoyed considerable success for device applications, such structures are less well developed on InP substrates. Often elongated structures (i.e. quantum dashes) are observed on InP instead of spherical shaped dots<sup>1</sup>. In addition, MOCVD grown SK QDs generally exhibit a bi-modal size distribution as well as the inherent wetting layer beneath the QD layer. The wetting layer has been identified as a path of leakage current which is an underlying cause for low optical gain and higher device temperature sensitivity, commonly observed in self-assembled OD lasers<sup>2</sup>. An alternate approach to SK QD formation is the use of nanopatterning with diblock copolymers combined with selective MOCVD growth, enabling QD formation over large surface areas intended for device application<sup>3-5</sup>. This approach allows for increased control over the QD size and distribution and elimination of the problematic wetting layer. High OD densities are achieved (> $6x10^{10}$  cm<sup>2</sup>) with a process applicable to large surface coverage, showing promise for implementation into long wavelength ( $\lambda = 1.3-1.5\mu m$ ) sources. In this project, we have demonstrated LT (up to 170K) InPbased laser operation from devices employing patterned QDs (~ 20 nm dia.). Prior reports of patterned QD active lasers on GaAs have shown promising results in terms of low threshold current densities<sup>6</sup>, although little is known concerning InP-based patterned OD active lasers.

The diblock copolymer nanopatterning process consists of a series of pattern-transfers from a dense array of nano-sized holes in a diblock copolymer thin film to a dielectric template mask, allowing the patterned access to the InP substrate for selective growth of the QDs.  $SiN_x$  (10 nm) / InP(100) substrates were employed using the diblock copolymer process and selective MOCVD growth, as reported previously.<sup>5,7</sup> Fig. 1 shows a top view SEM image after the selective growth (610°C) of InGaAsP Q1.15µm (2nm)/ In<sub>0.53</sub>Ga<sub>0.47</sub>As (2nm)/InGaAsP Q1.15µm (2nm) QDs on top of the nanopatterned InGaAsP/n:InP waveguide/cladding layer, forming the bottom half of a device structure. A schematic diagram of the QD laser structure is shown in Fig. 2. We have reported on the RT PL emission from such structures in ref. 7. After QD growth, the dielectric mask is removed followed by the regrowth of the top InGaAsP Q1.15µm SCH waveguide region and p: InP cladding layer at 620°C to complete the laser device structure. We found that the OD emission wavelength blueshifts by 200nm after the upper waveguide/cladding regrowth and laser oscillation is not observed<sup>12</sup>. However, by employing a lower temperature (550°C) SCH and cladding layer regrowth to bury the QD active region, we have been able to largely suppress the blue shift. Ridge-waveguide lasers were then fabricated and the electroluminescence (EL) spectrum was measured at LT (20K), as shown in Fig. 3. Laser operation (J<sub>th</sub>~1.9KA/cm<sup>2</sup> at 20K) is observed from devices with 50 µm-wide stripes and 3.6 mm-long cavities up to temperatures of 170K. Based on the shorter emission wavelength ( $\sim$ 1.3 µm) observed here, compared with that of the LT PL spectral peak (~1.4 µm), it appears that lasing may occur on a higher energy (excited state) OD transition. Further improvements in OD growth and pre-etching are expected to lead to ground state emission with lower current densities. Multi-segmented devices have also been fabricated for amplified spontaneous emission (ASE) measurements to allow extraction of the optical spectral gain and overall radiative efficiency, which could provide further insights into the operation of these devices. However, since only low temperature operation was achieved, it was not possible to collect sufficient data from the multi-segmented devices.

Cross-sectional TEM studies, Fig. 4, of the nanopatterned QD active regions confirm full three-dimensional confinement through the absence of a wetting layer. Since the QD formation

does not rely on strain, as for self-assembled QDs, the strain of the  $In_xGa_{1-x}As$  QDs can be varied over a large range from tensile-strain to compressive strain. Note that the composition of the  $In_xGa_{1-x}As$  layers used in the QDs is determined from planar layer structures, so there is uncertainty in the actual composition within the QDs, due to the selective growth occurring only within the nanosized openings of the dielectric mask. For the InAs QDs, the InAs layers were in the range of 1.5-2.5nm thick and surrounded by nominally lattice-matched InGaAs layers, as shown in Fig. 5. Low temperature photoluminescence measurements indicate that the emission wavelength of the QDs ranges from 1.4 µm for tensile-strained QDs to 2.1µm for compressively strained (InAs) QDs (Fig. 3). Shorter emission wavelengths are expected by implementing larger tensile strain, potentially accessing the 1.3 µm wavelength region. Longer emission wavelength into the mid-IR spectral region may be possible through the use of patterned formation of InAs QDs on materials with larger lattice constant, such as metamorphic buffers layers.

#### Optimization on dielectric mask and RIE condition for Multiple QD stacks

*Park et al*'s demonstration in 1997 on BCP lithography opened a way to fabricate nano-sized uniform array in the absence of the wetting layer<sup>8</sup>. Also, *Li et al* showed that it is possible to fabricate periodic dense arrays of hole pattern can be achieved using cylinder-forming block copolymer with a high degree of pore-size uniformity through the pattern transfer procedures by the selective MOCVD growth<sup>9</sup>.

As shown in Fig.6, when transferring the pattern onto the dielectric mask, reactive ion etching has been used other than wet etching in order to avoid its inherent undercutting and its heavy dependence on the location on the wafer, which potentially increase the chance of resulting in larger and non-uniform QD arrays. However, it was also observed that RIE creates significant amount of plasma damage on the surface of the sample, which will, in return, increase the number of non-radiative recombination centers on the surface. Therefore, in order to avoid both undercutting and the surface damage, it is necessary to have the minimum etching time as well as faster etching rate while this process has to be done in the controllable range of time. Also, the dielectric has to be completely removed selectively by wet etchant in a short time after the QD growth. Thus, to address these issues, the processes involving the dielectric mask deposition and RIE conditions were optimized.

Even though J. *Park et* al has successfully reported uniform and dense QD arrays employing dilock copolymer lithography, their photoluminescence at room temperature was relatively low<sup>10</sup>. To increase the PL intensity and optical gain, multiple stacks of QD grown by SK growth mode has been utilized<sup>11</sup>. However, unlike the case of SK growth mode, the number of stack is limited by the dielectric mask thickness for this selective growth. In this section, the optimization on dielectric and the following RIE is discussed to have thicker dielectric hard mask.

It is known that  $SiN_x$  is a significantly better diffusion and chemical barrier than  $SiO_x$ . Therefore,  $SiN_x$  deposited by PECVD has been chosen as a dielectric mask. Also, in order to achieve the faster etching rate in both HF-based solution and RIE using CF4 plasma, the flow rate ratio of NH<sub>3</sub>

and SiH<sub>4</sub> was increased so that the resulting SiN<sub>x</sub> is N-rich. It was checked that the etching rate of N-rich SiN<sub>x</sub> is faster than Si-rich SiN<sub>x</sub> under the same RIE condition, which agrees with the reports by *Williams et al*<sup>12,13</sup>. Low r.f. power was also employed to increase the uniformity in the thickness.

r.f. Power	NH3/SiH4 ratio	Temperature	<b>Deposition Rate</b>
25W	6	250°C	0.054nm/s
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In order to prevent the polymer mask from being attacked during RIE, pure CF4 plasma was used to transfer the pattern onto the  $SiN_x$  hard mask. During this process, low pressure and gas flow were chosen so that the undercut of  $SiN_x$  is minimized. Also, the moderate amount of r.f. power was used to avoid the erosion of polymer and to result in shorter yet controllable etching time.

r.f. Power	Pressure	CF4 Gas flow
100W	10mTorr	10sccm

<sup>&</sup>lt;Table 2: Optimized RIE parameters>

In order to have multiple stacks of QDs for higher luminescence and gain, it is desirable to have thicker  $SiN_x$  layers. Previously, we employed only about 10nm of  $SiN_x$ , resulting in uniform and dense single-stack QD arrays. Using the optimized recipes for  $SiN_x$  and RIE, it became possible to have uniform and dense QD arrays with thicker (20nm)  $SiN_x$ , with the etching time of 17 second. This thickness allowed us to achieve up to three-stack QD structure. Specific details will be discussed in the next section.

Stacked QD arrays as a gain medium for laser diode require them to be grown on the lower part of the separate confinement heterostructure(SCH) as shown in Fig 7. 1.5µm thick n: InP(5 ×  $10^{17}$  cm<sup>-3</sup>) and 150nm thick InGaAsP( $\lambda_g = 1.15\mu$ m) lattice-matched to InP were grown at 630°C by MOCVD on the InP substrate as a laser base with precursors of TMIn, TMGa, AsH<sub>3</sub>, and PH<sub>3</sub>. Then, SiN<sub>x</sub> was deposited for 460 second, using the optimized parameters. After 20nm thick SiN<sub>x</sub> deposition, the sample was annealed in the MOCVD reactor under AsH<sub>3</sub> and PH<sub>3</sub> flow for 30 min at 630°C, as it was reported that annealing prevents SiN<sub>x</sub> from being cracked during QD growth and improves the adhesion<sup>10</sup>. Then, 50nm thick cylinder-forming BCP was patterned on the laser base. In order to find the optimized RIE time, the pattern-transfer onto SiNx mask has been carried out by RIE for 17sec. After having wet treatment in diluted HCl and acetic acid to recover plasma damage for 30 sec, three stack InGaAs QD structures consisting of 3 x InGaAsP/InGaAs/InGaAsP (2nm each) was grown at 630°C. Right before the QD growth, *in-situ* etching targeting 2.5nm was done using CBr4 to remove the remaining plasma damage<sup>14, 15</sup>. After QD growth, the SiN<sub>x</sub> hard mask was removed by 6:1 buffered oxide etchant (BOE) for 30 second.

To grow the uniform and homogeneous capping layer after the  $SiN_x$  removal, it is important to completely remove it for a short time using a wet etchant. Since both concentrated HF and phosphoric based acid attack InP-based semiconductor material, BOE was selected, which is known as an excellent selective etchant for SiNx. After SiN<sub>x</sub> removal process, scanning electron microscopic images were taken to check the size of QD grown as well as SiN<sub>x</sub> removal as shown in Fig. 4.

It can be clearly seen that with RIE time of 17 second, the pattern of the BCP was well transferred onto the SiN<sub>x</sub> hard mask, and the subsequent growth resulted in the uniformly arrayed QDs with the diameter of ~33nm. To check the reproducibility, *this process has been repeated multiple times*, and the results remained the same, confirming its reproducibility. The density of QDs was found to be ~ $6.1 \times 10^{10}$  cm<sup>-2</sup>, which is comparable with J. *Park*'s result<sup>10</sup>. Also, it was confirmed that 6:1 BOE can selectively etch away SiN<sub>x</sub> within 30sec.

Shown in Fig. 5 is a high-resolution transmission electron microscopic image of the cross section of a QD stack in the array. These image were obtained from our collaborators (Dr. Yongkun Sin) at The Aerospace Corporation. Fig. 5 (b) shows the periodic twinning plane developed during the QD growth, which were reported in the context of nanowire(NW) growth in the literature, originated from the coexistence of zinc blende(ZB) and wurtzite(WZ) crystal structure when grown epitaxially in a close-packed direction<sup>16-19</sup>. Fig. 6 shows a HR-STEM image of a QD stack in which, again, stacking faults are indicated. In reference 19, Shtrikman *et al* demonstrated employing highly terraced (311)B substrate resulted in stacking-faults-free zinc blende GaAs nanowires. Therefore, a similar approach may be employed in the future for the stacked QD materials. Also, for the better interface before the regrowth of QDs as well as subsequent capping layers, effectiveness of *in-situ* etching on InGaAsP and InP layer using CBr<sub>4</sub> will need to be verified and optimized.

## Photoluminescence (PL) Study of Stacked QDs

The optical characteristics of QDs grown were investigated by PL measurement at various temperatures. The PL measurements were carried out using a continuous-wave Ar-ion pump laser at a wavelength of 514nm. The data was collected using a grating monochrometer and a cooled germanium detector. Low temperature measurements carried out with the sample mounted in a continuous-flow liquid-helium cryostat and cooled to 15K. Also, the capping layer was not grown on the QD layer. To observe the characteristics of the QDs, multiple quantum wells, which were grown without the patterning, were grown on the same laser base at the same condition as the growth of the stacked QDs.

Shown in Fig. 7 are PL spectra measured from 15K to 265K. At 15K, the peak around 1.5 $\mu$ m is observed, which is believed to originate from the QDs, while the peak from MQW structure is positioned around 1.4 $\mu$ m. The difference may come from the quantum size effect, i.e., the stacked QDs are confirmed to be thicker than what is targeted due to the limited surface migration inside the cylindrical hole on the SiN<sub>x</sub> mask. This is evident from the HR-TEM image in Fig. 5 as the height of the stacked QD is measured to be ~29nm, significantly thicker than the intended thickness. This increased thickness is a result of growth rate enhancement present from the selective area growth. We anticipate that after optimizing the QD thicknesses based on the TEM images, further samples could be fabricated and capped to allow for room temperature PL characterization of the QDs.

#### Conclusion

Three stacked QD arrays were successfully grown based on (100) InP substrate employing the pattern-transfer technique using BCP. It was found that the optimization on the parameters in PECVD SiN<sub>x</sub> and the subsequent RIE are crucial for the reliable selective growth of the stacked QD arrays. TEM images show that the stacked QD arrays were selectively grown on the nano-sized pattern while the stacking fault and twinned plane were observed, which were reported in the context of NW growth in the literature. To overcome these issues, the optimization on the *insitu* etching using CBr<sub>4</sub> will be necessary to make the surface, before the regrowth, better, removing the line defects, which might be generated during pattern-transfer. Also the use of high index (311)B substrates may be employed to reduce stacking faults, since it has been reported that (311)B substrate can suppress the generation of WZ structure in the NW structure. At low temperature, PL emission, likely from the stacked QD, were observed although obtaining room temperature PL can only be expected after capping the stacked QDs. Also, in order to better control the thickness of the QD within a hole in SiN<sub>x</sub> pattern, it will be necessary to more accurately determine the growth rate within the holes in the nano-pattern.

#### Key findings from this project:

- 1. We demonstrated an alternate approach to Stranski–Krastanow (SK) QD formation involving the use of nanopatterning with diblock copolymers (DBC) combined with selective MOCVD growth, enabling QD formation over large surface areas intended for device application. This approach allowed for increased control over the QD size and distribution and elimination of the problematic wetting layer associated with SK QDs.
- 2. Compared with self-assembled QDs, the nanopatterning and selective growth process provides for a greater degree of control over the compositional range within the QDs, allowing for *tensile-strained*, lattice-matched, or compressively-strained QDs. Tensile-strained (InGaAs) and compressively-strained (InAs) QDs on InP were fabricated and exhibited luminescence near 1.4 and 2.1 microns respectively. We are not aware of any other reports of tensile-strained QDs.
- 3. The *first* nano-patterned QD lasers were fabricated in this program. Lasing was observed only at low temperatures, presumably a result of non-radiative carrier recombination originating from fabrication induced surface states within the QD active region. Additional work is require to reduce non-radiative surface recombination in the QD active region, which is expected to lead to improved performance.
- 4. In order to further improve the optical properties (i.e. reduce non-radiative recombination) of the QD active region, in-situ etching using CBr<sub>4</sub> prior to the QD deposition showed improved luminescence from the QDs. These techniques need to now be implemented for QD active devices.
- 5. An optimized DBC process coupled with thicker nitride masking material allowed us to demonstrate *for the first time* stacked QDs using the selective growth process. TEM imaging reveals that stacking faults are present in the QDs, similar to that observed in nanowires fabricated by selective growth epitaxy using a dielectric mask.

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Fig 1. Top view SEM image of  $InGaAsP(Q1.15)/In_{0.53}Ga_{0.47}As/InGaAsP(Q1.15)$  QDs after selective MOCVD growth.





# Fig 2. Schematic diagram of QD active laser structure

Fig 3. Low temperature, 20K EL spectrum of InGaAsP (Q1.15)/  $In_{0.53}Ga_{0.47}As/InGaAsP$ (Q1.15) QD laser with current density of 1.94 KA/cm<sup>2</sup>.



Fig 4. TEM cross-sectional image of the buried QD active region, demonstrating QDs with no wetting layer present.



Fig. 5 LT (30K) PL spectrum of tensile-strained, lattice-matched, and compressive-strained In(Ga)As QDs (2nm In<sub>0.3</sub>GaAs QD, 2nm In<sub>0.53</sub>GaAs QD, and InAs QDs thicknesses of 1.5nm and 2.5nm). Pre-cleaning procedure (HCL etching) prior to InAs QD growth was employed. PL intensities are normalized.



Fig. 6. Selective Growth of QD arrays using Cylinder-forming Diblock Copolymer



Fig 7. Laser Base Structure with three stack QD array







Fig. 8 Top view of Scanning Electron Microscopic Images for QDs grown with various RIE time



Fig. 9 Cross sectional HR-TEM of a three stacked QD



Fig. 10 HR-STEM of a three stacked QD



Fig. 11 Temperature dependent PL measurement from (a) three stacked QDs grown with SiNx mask (b) triple QWs grown without SiNx mask on the laser base