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# Study of site controlled quantum dot formation on focused ion beam patterned GaAs substrate

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Abstract. In this project, we combine Ga<sup>+</sup> focused ion beam (FIB) patterning and selfassembly of InGaAs quantum dots in order to produce regular quantum dot (QD) arrays. The GaAs substrate is patterned before transporting into molecular beam epitaxy growth chamber to do anneal and overgrowth. The thickness of deposited InAs is precisely controlled. Quantum dots are expected to nucleate at specific locations, where the ion beam has previously implanted gallium atoms. The properties of the quantum dots formed are related to the FIB beam parameters, which include accelerating voltage, probe current, dwell time and pitch. We studied and statistically analyzed the relationship, focusing on their diameter, height and distribution. Scanning electronic microscope (SEM) and atomic force microscope (AFM) images have been used to analyze the patterns and QDs before and after overgrowth. Micro-PL study was performed to test the QD opto-electronic property. Scanning transmission electron microscope (STEM) cross sectional analysis and X-ray mapping is performed.

#### **1. Introduction**

Quantum dots (QDs) are small artificial crystal structures, whose electronic properties depend strongly on the size. One of the dominating fabrication techniques is Stranski-Krastanov growth by molecular beam epitaxy (MBE). It is a top-down approach, which is a quick and easy way to grow QD on a large area. But the inherently random position of nucleation puts limits on down-sizing devices nowadays. If only a few quantum dots are included in a device, the location and uniformity of quantum dots will have a strong influence on the reproducibility of the electronic and opto-electronic performance across multiple devices. The bottom-up growth by patterning the substrate before overgrowth is an alternative approach which can produce regular QDs on patterned substrates with better position control and homogeneity [1].

We studied InGaAs quantum dots overgrowth on Ga<sup>+</sup> focused ion beam (FIB) patterned GaAs (001) substrate. QDs are expected to nucleate at specified locations structure by the ion beam. The aim is to study how patterns fabricated with different ion beam parameters could lead to different nucleation results.

#### 2. Patterning and overgrowth

The patterns were fabricated in a JEOL 6500F scanning electronic microscope (SEM) equipped with an Orsay Physics Ga+ FIB column, which makes it a dual-beam SEM/FIB instrument. A Raith lithography system was used to design and fabricate the patterns. The patterning ion beam had different parameters including accelerating voltage, probe current, dwell time and pitch. After FIB

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patterning, in-situ secondary electron images (SEI) were captured at each individual patterned region. Before transferring the patterned substrate into the MBE chamber for quantum dot growth, atomic force microscope (AFM) images were also captured in tapping (non-contact) mode. During overgrowth, the sample was degassed at 400°C for 1 hour and then annealed under an arsenic flux at 580°C for 30 minutes, during which the surface was monitored using reflection high-energy electron diffraction (RHEED). A clear (2x4) reconstruction was observed after this process, which is indicative of a smooth and relatively oxide-free surface. Then a 20nm GaAs layer was grown at 580°C and a growth rate of 0.5ML/s. The thickness of this layer was deemed sufficient to reduce the effect of any surface ion damage and yet not to fill in the FIB-generated patterns. Finally, InAs quantum dots were grown at 520°C and a growth rate of 0.02ML/s by depositing around 1.5 monolayers of material. The amount of deposited material was judged to ensure that quantum dots nucleated mainly at patterned locations and not on the flat surface between or outside the patterned regions [2]. The finished sample was imaged again by SEM and AFM respectively. Two pairs of SEM and AFM images are shown below in figure 1 and figure 2. The images clearly indicate island structures were formed during overgrowth, at specified locations where ion beam had scanned before.



Figure 1. Secondary electron images captured before (a) and after (b) overgrowth. The patterning ion beam parameters are accelerating voltage of 20kV, probe current of 4pA and dwell time 20ms.



Figure 2. AFM images captured at the same region as fig. 1, before (a) and after (b) overgrowth.

Apart from the island structures shown above, two other types of structure formed, rings and holes. Figure 3 shows the relationship between the patterning ion beam parameters and the structures produced. The result shows that short dwell times and small aperture sizes (low doses) produced dots while long dwell times and big aperture sizes (high doses) produced rings or holes. The height and diameter of islands are measured by analysing AFM images. The measurements were collected from three patterns formed at 20keV ion energy and 5pA probe current with three different dwell times (1ms, 5ms and 20ms), as well as from one pattern formed at 20keV ion energy but with the probe current increased to 40pA at a dwell time of 1ms. From the left part of figure 4 can be seen that the island heights are almost constant, covering a narrow range from 12nm to 25nm, with values slightly increasing with pitch. The diameter scales with the square root of the pitch, as can be seen from the right part of figure 4, and is also increasing with the dwell time and probe current, which is not so obvious from the diagram. The result confirms the possibility of controlling the nucleated quantum dot size. However, it is clearly difficult to create islands under 100nm in diameter and below 10nm in height, which will be a prerequisite for strong quantum confinement effects in optical spectra.



**Figure 3.** Different ion beam parameters and the resulting structures formed over the patterns. The ion dose is increasing from bottom left to top right.



Figure 4. Height and diameter of islands vs. pitch

## 3. Micro photoluminescence and TEM cross sectional analysis

Micro photoluminescence ( $\mu$ -PL) studies were performed at room temperature. The exciting laser (green) had a diameter of ~5 $\mu$ m, a wavelength of 532nm and 1-2mW power.  $\mu$ -PL measurements were performed scanning the laser beam along a straight line from positions outside the patterns, where QDs had randomly nucleated on the surface, across the entire patterned area with a step size of 5 $\mu$ m. The spectra are not as sharp as expected [3]. There are two rather broad peaks in the spectra at ~878nm (1.41eV) and ~1152nm (1.08eV). They are related to the underlying GaAs substrate and the InGaAs QD signal. From the latter peak wavelength we can estimate the compositional ratio of Ga:In atoms as 74:26 (i.e. In<sub>0.26</sub>Ga<sub>0.74</sub>As). The FIB patterned area shows higher signals for both peaks, where the signal from the GaAs substrate increases more rapidly. The enhanced optical contributions from both the pure GaAs and the InGaAs islands may be explained by indium atoms from the wetting layer formed upon epitaxy having been more efficiently integrated into the islands when the surface has previously been FIB patterned, confirming the formation of QDs.



**Figure 5.** Set of experimental  $\mu$ -PL spectra on and off the pattern region (a). The intensities at two wavelengths, 878nm (GaAs) and 1152nm (InGaAs), were extracted at each scan position (b).

STEM cross sectional analysis was performed on a FIB prepared membrane which has a row of island embedded. Annular dark-field and bright-field STEM imaging and X-ray maps of GaK, AsK and InL are shown in figure 6. All images are background subtracted with the maximum element counts listed below. We calculate the In composition from their relative intensity ratios. The X-ray maps indicate a peak indium concentration of  $x \approx 0.16$  near the top of the islands.



**Figure 6.** X-ray elemental maps of Ga (a), As (b) and In (c); (d) is an indium concentration map. (e) is an ADF image of the cross sectioned islands; (f) shows concentration profiles from the top of the island to the substrate, calculated from the marked area shown in (d).

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