Dense Arrays of Semiconductor Quantum Dots with controlled Positions Grown in Inverted Pyramids

E. Pelucchi [1)] , S. Watanabe [1)] , B. Dwir [1)] , K. Leifer [1)] , E. Kapon [1)]

1) Laboratory of Physics of Nanostructures, Institute of Quantum Electronics and Photonics,Swiss Federal Institute of Technology Lausanne (EPFL), CH−1015 Lausanne, Switzerland

The possibility of tailoring the photonic modes of semiconductor emitters placed in a cavity by smart engineering of cavity geometry and/or active coupling has been the subject of intense research in the last few years. This has been the leading concept of a number of experimental and theoretical achievements, which, on the other hand, show a number of differences on many important aspects. Within these results it is worth mentioning photonic molecules [1], where the active material is a simple InGaAs quantum well, and the more complex photonic heterostructures/crystal formed by arrays of coupled vertical cavity surface emitting lasers (VCSELs)[2]. More recently photonic crystal (defect) microcavities with self assembled randomly nucleated InAs quantum dots (QD) have been realized [3] for cavity quantum electro dynamic experiments, and a new design of a photonic crystal microcavity for quantum electrodynamics with a single InAs quantum dot has been proposed [4]. The goal, among others, is to achieve single dot lasing, exploiting the theoretical high Q factor and small mode volume of the cavity.

Nevertheless designing active photonic heterostructures with InAs self−assembled Stranski−Krastanow (SK) QDs has faced so far the difficult problems of controlling the quantum dot nucleation sites and of the poor uniformity of geometrical (and hence optical) characteristics. Attempts of improving positioning and uniformity of SK QDs are still far from giving satisfactory results when both good geometrical and optical quality is sought [5].

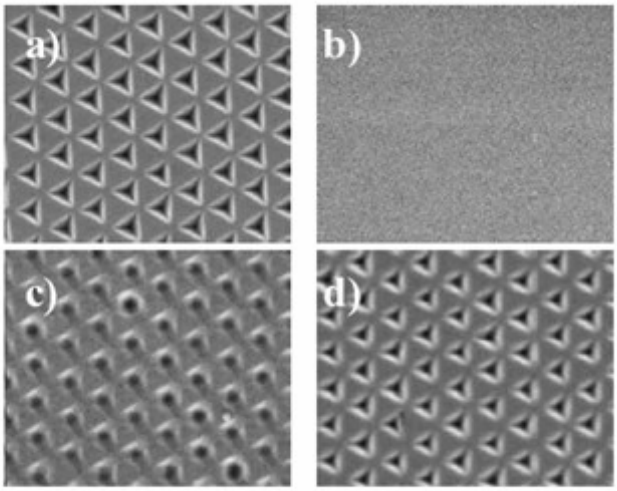

Fig. 1 SEM images of a) a 300nm-pitch substrate before growth. b) Surface after deoxidation under AsH_3 flux at substrate temperatures around 730°C, c) Surface after deoxidation under AsH_3 flux at substrate temperatures around 620°C, d) Surface after deoxidation under AsH_3 flux at substrate temperatures around 530°C.

For these purposes a promising approach is the fabrication of GaAs/AlGaAs and InGaAs/AlGaAs quantum dots by organometallic chemical vapour deposition (OMCVD) on non−planar substrates. This technique has been studied extensively in micron−size pyramidal recess patterns where it allows excellent control over dot position and dimensions, accurate engineering of the QD energy levels, and very good homogeneity of emission energies [6]. These characteristics make already these QDs of interest in applications such as quantum computing and cryptography. Similar control on QD features, combined with higher dot surface density (i.e., with periodicities down to optical wavelengths scale, 200−300 nm in III−V semiconductors) would make possible applications such as efficient QD lasers or photonic crystals/heterostructures. We report here on the growth and optical properties of pyramidal QD arrays with pitch as small as a few hundreds of nm, as a first and basic step toward the implementation of such concepts.

Prior to growth, triangular arrays of pyramidal pits with 300nm and 500nm pitch were etched into {111}B GaAs substrates (misoriented 2° toward [01−1]) using electron−beam lithography and wet etching. To obtain high resolution and etch resistance, PMMA resist and a SiO_2 intermediate mask were used. Following deoxidation, OMCVD growth of a multilayer GaAs/AlGaAs heterostructure was performed, using growth conditions such as deoxidation and growth temperatures, growth rates and V/III ratio, which had to be chosen quite differently from those employed with micron−size pyramids [6].

To avoid planarization of the recess patterns, low temperature (~530−550ºC) had to be used for deoxidation [7]. Figure 1a) shows a scanning electron microscopy (SEM) image of a 300nm−pitch substrate before growth. Figure 1b) shows the same surface after deoxidation under AsH_3 flux at substrate temperatures around 730ºC. Evidently surface mass transport completely planarized the substrate [8]. We stress that these deoxidation conditions are routinely used in our laboratory for growth on V−groove gratings with comparable pitch etched into {001} substrates, where no such major effect can be observed. When triangular arrays of inverted pyramids with several micron pitch are deoxidized at comparable temperatures only a modest rounding at the bottom of the pyramidal structure can be observed. Figure 1c), d) show respectively a substrate deoxidized at ~620 and at ~530ºC. Quite clearly only in the last case mass transport did not significantly perturb the original pyramidal pattern.

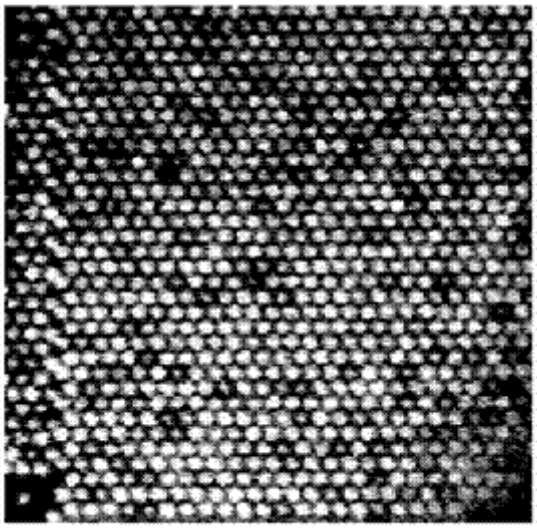

Fig. 2 Wavelength dispersive cathodoluminescence image of sample 945, centred at dot emission (1.55 eV) with a collection window of 7meV. The pattern pitch is 500 nm, while the apparatus spatial resolution is around 200 nm. Nominal dot thickness is 0.25 nm.

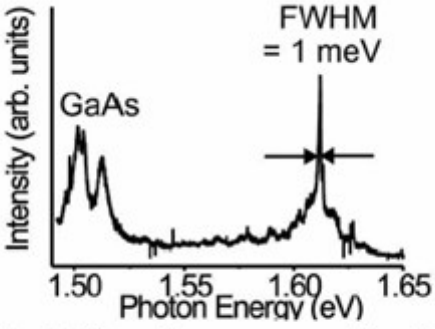

Fig. 3 Micro-PL spectrum at T = 10 K from a 300 nm pitch QD array sample.

After deoxidation, temperature is ramped up during GaAs buffer deposition at very high V/III ratio (> 5000) to minimize mass transport and at low nominal growth rate (0.006 nm/s, as estimated by growth on a planar (001) GaAs wafer) to reach temperatures >620ºC before AlGaAs barrier deposition at a lower (800) V/III ratio. Nominal AlGaAs growth rates in a range between 0.02 nm/s and 0.04 nm/s had to be employed, since mass transport from planar (non−growth) areas caused the effective growth rate in the recess patterns to be more than 10 times larger. GaAs dot deposition (with fraction of nm nominal thickness) was performed under similar conditions. Cladding layers with 55% Al content were also added before and after the $Al_{0.3}Ga_{0.7}As$ barriers of the GaAs QD to improve carrier collection in the dot. To further increase the photoluminescence (PL) efficiency, a post−growth substrate removal was used, which in some samples involved the insertion of a high−Al−content layer in the structure (back−etching) [6].

The dense QD arrays were characterized structurally by scanning emission microscopy, atomic force microscopy and transmission electron microscopy. The micrographs show that good surface morphology can be obtained after growth, and that thickened GaAs areas are obtained at the bottom of the pyramids, indicating the formation of QDs (not shown).

Optical characterization was performed using a low−temperature microscopic PL set−up, cathodoluminescence (CL) spectroscopy and wavelength−dispersive imaging.

Figure 2 shows a wavelength dispersive cathodoluminescence image of sample 945 obtained in a back−etched geometry, centred at an energy corresponding to the dot emission (1.55 eV), measured at 10K. The energetic collection window was 7 meV. The triangular pattern pitch is 500 nm, whereas spatial resolution is around 200 nm, enough to clearly distinguish the signal of each individual quantum dot. Good uniformity of the emission pattern is clearly shown, while only a few dots show to be damaged (less than 1% on the whole sample). The distribution of emission energy in the range of 10−15 meV within a 15−µm excitation region. Therefore in Fig. 2 some dots show a less intense signal, because only the tail of their emission falls in between the 7 meV of the energetic collection window.

Figure 3 shows a micro PL spectrum of a 300 nm pitch QD array measured at an excitation power of 45 µW; the spectrum represents the emission of more than 10 dots.

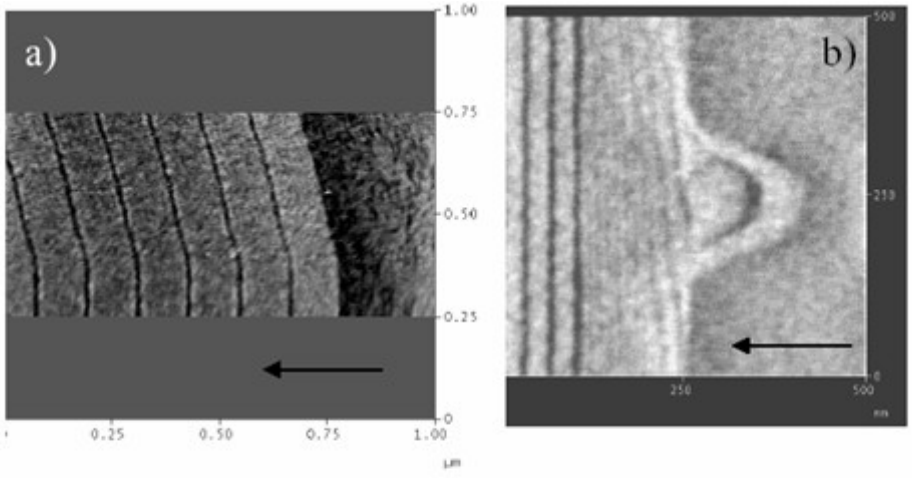

Fig. 4 a) Atomic force microscope cross-sectional image of an AlGaAs layers with GaAs markers grown by OMCVD on a (111)B GaAs surface. The arrow indicates growth direction. The visible layer bending is an artifact and should be ignored. b) Similar structure grown on a patterned substrate.

A QD spectral peak at 1.61 eV with full width half maximum (FWHM) of 1 meV is observed in this case. We did

not observe any higher energy spectral peaks, which would be expected due to recombination at the surrounding barriers of the QD. This can be explained by efficient carrier transport to and capture by the dots. Unfortunately this high uniformity is observed only in some specific regions of the sample, in other the micro PL spectrum shows more than one peak, spaced by ~1 meV or less and with comparable FWHM. We do consider this a promising indication that with a further improvement of the patterning process such homogeneity will be attainable on areas of several tens of microns. One of the limitations of the use of patterned (111)B GaAs substrates is the difficulty in obtaining good two−dimensional (2D) growth over the planar portions of the substrate. In fact, at the growth temperatures at which we obtain our QDs, no regular 2D growth takes place on the (111)B surface. Such planar (111)B growth would allow for example the growth of GaAs/AlGaAs Bragg mirrors in order to incorporate the pyramidal QDs in an optical microcavity. Good quality OMCVD growth on (111)B GaAs substrates of AlGaAs has been reported in the literature at very high temperatures (at least ~900 °C) [9], unattainable in our growth apparatus. Moreover, very little information can be found in the literature on such (111)B growths. We have thus started the optimisation of GaAs/AlGaAs growth on our (111)B GaAs substrates. In Figure 4a) we show cross sectional atomic force microscope image of the resulting heterostructure. The sample consists of an AlGaAs layer with GaAs markers, grown at substrate temperature ~810°C, using high V/III ratio (800) and low growth rate (~0.15 micron per hour), i.e. conditions very close to those used for our QDs with the only exception of the growth temperature. Fig. 4 b) shows a similar structure with GaAs markers grown on a patterned substrate. The marker period was chosen to be quite short at the beginning of the growth to monitor dot formation in the inverted pyramids and than increased once planarization was achieved to take into account the differences between the effective growth rates in the pyramids and on the planar areas. Clear planarization and 2D growth are obtained on the (111)B surface, opening the way for epitaxy of the structure mentioned above.
Work is underway to check the effect of significantly lower V/III ratios and lower/higher growth rates on this type of samples, to reduce the pitch of our QD arrays to 200nm, thus producing QD arrays of densities similar to Stranski−Krastanow QDs, and finally incorporating them into photonic crystal and laser structures.

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