Growth Kinetics Effects on Self-Assembled InAs/InP Quantum Dots

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(Dated: April 14, 2018)

A systematic manipulation of the morphology and the optical emission properties of MOVPE grown ensembles of InAs/InP quantum dots is demonstrated by changing the growth kinetics parameters. Under non-equilibrium conditions of a comparatively higher growth rate and low growth temperature, the quantum dot density, their average size and hence the peak emission wavelength can be tuned by changing efficiency of the surface diffusion (determined by the growth temperature) relative to the growth flux. We further observe that the distribution of quantum dot heights, for samples grown under varying conditions, if normalized to the mean height, can be nearly collapsed onto a single Gaussian curve.

PACS numbers: 68.65.Hb, 78.67.Hc, 81.07.Ta, 68.37.Ps, 81.15.Gh

Strained heteroepitaxy beyond the critical thickness can lead to spontaneous generation of three dimensional nanoclusters via the Strunski-Krastanov growth route.

In this letter, we have studied metal-organic vapor phase epitaxy (MOVPE) grown InAs/InP quantum dots. In a large number of previous studies on this system, the actual morphology and/or the emission properties of quantum dot ensembles have been found to be dependent on both the specific details of the material parameters and growth conditions (e. g., substrate miscut\(^{[4]}\), long ranged surface stresses\(^{[3]}\), matrix material\(^{[3]}\), material flux and partial pressures during growth\(^{[4]}\), annealing\(^{[10]}\) and growth interruptions times\(^{[1]}\)) as well as on the generic growth parameters\(^{[12]}\), like as on the generic growth parameters\(^{[12, 13, 14]}\) like the growth temperature, materials flux and coverage. Since the dependence of the quantum dots’ properties on the latter set of growth kinetics parameters is expected to provide a more system independent insight into the problem of self-assembly, we have also followed this approach. Apart from the above mentioned references on the InAs/InP system, the approach also allows us to relate our work to other studies carried out in the same spirit but on different material systems. These include Monte Carlo simulations by Meixner, et al.\(^{[15]}\), a rate equation based model for growth\(^{[16]}\) and theoretical and experimental observations on InP/GaAs by Johansson and Seifert\(^{[17, 18]}\) and many studies on the InAs/GaAs system, among which the one by Dubrovskii, et al.\(^{[19]}\) is quite substantial. Furthermore, we demonstrate a very simple (phenomenological) scaling collapse of the heights distribution data onto a single Gaussian curve. The present study therefore attempts to demonstrate in a qualitative sense that the quantum dot density and the average size but also their size dispersion may be understood and therefore predicted on the basis of the three most basic growth parameters—coverage, growth rate and growth temperature—provided the growth is carried out under far from equilibrium conditions.

MOVPE growth was carried out on n+ doped (001) InP substrates using trimethyl indium and arsine as group III and V sources in a horizontal reactor at a pressure of 100 torr with hydrogen as the carrier gas. InAs layers were grown at a relatively low temperature of 430-450°C. Prior to InAs deposition, an InP buffer layer was grown, first ∼500 Å at 625°C and then with temperature continuously ramped down to the InAs growth temperature and finally another 500 Å at the stable temperature. To avoid switching transients, the indium flux for the buffer, the InAs, and the cap layers was kept the same. For a given set of growth conditions, a pair of samples was grown with identically deposited InAs layer in two growth runs. In the first case, the sample was immediately cooled and taken out of the reactor after InAs deposition itself to enable a study of surface morphology and in the second case, an InP cap layer was grown for samples used for photoluminescence (PL) study. For these samples, about 50 Å InP cap was deposited at the InAs deposition temperature to minimise further ripening during the subsequent growth of the remainder of the cap at higher temperature. The uncapped dots were characterized by Nanoscope atomic force microscope in contact mode. PL spectra were recorded at ∼25K with a 0.67 meters McPherson grating monochromator and 325 nm helium-cadmium laser as the excitation source at a

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FIG. 1: (left column) ~ 500nm×500nm AFM surface scan images, (middle column) heights histograms and (right column) the 25K PL spectra (on the corresponding capped samples) grown at different growth rates (GR) and growth temperatures (T) and growth durations (D). Growth rate G0 corresponds to approximately 2.5 ML/s. Peaks around 1eV in (b), (c) and (f) are due to wetting layer. Notice that the morphology and PL (a) and (d), and in (e) and (g) are qualitatively similar.

From Fig. 1, it is evident that for similar coverage but by changing the growth rate and growth temperature, it is possible to change the quantum dots’ density by over an order of magnitude. The corresponding peak PL emission wavelength is also seen to change from ~ 0.65eV in Fig. 1(f) to the more usual ~ 0.8eV in Fig. 1(a, d, e and g). At lower coverage, corresponding to intermediate stage of growth, we observe that the distribution is bimodal for comparatively smaller growth rates (Fig. 1(b, c)). This intermediate stage bimodality (Fig. 1(b), (c)) is suppressed by making the growth more non-equilibrium, by either lowering the growth temperature as in Fig. 1(a), or by increasing the growth flux as in Fig. 1(d). This behavior has also been observed in previous studies on InAs/GaAs but also needs to be contrasted with the more complex trend observed for InP/GaAs samples.

Furthermore, we observe that Fig. 1(a and d) and Fig. 1(e and g) are qualitatively more similar to each other than they are to Fig. 1(b) and Fig. 1(f) respectively. This indicates that (1) a smaller growth rate (and an enhanced growth temperature) yields larger dots with a smaller areal density and (2) that the effect of a smaller growth rate can be compensated by a larger growth flux. Specifically, our observation of point (2) is qualitatively very similar to the expectations in a recent growth simulation by Meixner, et al. (Fig. 7 in reference [15]). The simplest models for self-assembled cluster growth [15, 16] are developed in analogy with the submonolayer deposition [22] with the assumption that the later stage of self-assembly is largely dictated by the kinetic processes occurring at the surface. Then the average quantum dot density is dictated by how efficiently the preexisting material can diffuse and find an equilibrium site before more fresh material arrives on the surface. Quantitatively, this takes the form of a scaling relation [15, 22], where the mean island density depends only on the ratio of the growth flux and surface diffusion efficiency. The largeness of this dimensionless ratio may also be taken to be the measure of departure from equilibrium.
Around the mean (the height dispersion by a single Gaussian curve centred to unity in Fig. 2). Very approximately, we may describe mean height and normalized the area under the curve other curves in the figure scaled down by a factor of three for clarity of comparison with Fig. 1 except that the curve corresponding to Fig. 1 (a) is a Gaussian function. Inset shows the same histograms as in Solid line shows a fit of the average of these data points to the respective histograms (Fig. 1) scaled by the average height.

FIG. 2: Probability distribution function constructed from the respective histograms (Fig. 1) scaled by the average height. Solid line shows a fit of the average of these data points to a Gaussian function. Inset shows the same histograms as in Fig. 1 except that the curve corresponding to Fig. 1 (a) is scaled down by a factor of three for clarity of comparison with other curves in the figure.

mean height and normalized the area under the curve to unity in Fig. 2. Very approximately, we may describe the height dispersion by a single Gaussian curve centred around the mean \( \langle h \rangle / \langle h \rangle = 0.98 \) with a full width at half maximum of 0.43. These values provide a rough but very useful estimate of the expected size dispersion (since area \( \propto h^2 \)) in terms of the average size of dots. Since the average dot-sizes themselves may be written in terms of growth kinetic parameters, such a prescription can, in principle lead toward a first principles prediction of the inhomogeneous broadening in terms of a few growth parameters, especially because the primary confinement occurs along the height of the quantum dots due to the large aspect ratios (\( \sim 6 - 10 \)).

While the peak energy and the modality of the size distribution can be correlated with the PL spectra, a direct correlation between the size dispersion and the low temperature PL emission linewidth is not always seen in the high density samples, Fig. 1 (a and e). This is presumably because of the strong interdot coupling effects. This may be understood as a combination of two effects (1) an overlap between the dots can lead to an excess ‘bandwidth’ over and above the energy spread associated with quantum dots’ size dispersion (2) despite a band formation (which would typically imply a narrower linewidth due to the transfer of carriers to the lowest available state in the density of states continuum), at low temperature, the strong potential fluctuations localize the excitons and they are not easily transferred to the lowest possible energy. A temperature dependent PL study that shows a qualitative difference between the temperature dependent emission properties of moderate and high density dots and which partially supports this hypothesis will be presented elsewhere.

Conclusions: MOVPE grown InAs/InP quantum dots ensembles grown at different growth rates and temperature were studied for their morphological and optical properties. We observed that the growth was largely kinetically determined with the surface diffusion being the most prominent process within the space of (deliberately highly non-equilibrium) growth parameters studied. The bimodality in quantum dots sizes and the PL emission peaks could be controlled by changing the growth conditions. It was also established, both in morphology and in optical properties, that the effect of lowering the growth temperature is qualitatively similar to the that of increasing the growth rate at a higher growth temperature. For dots with a unimodal distribution, the distribution of heights normalized by the average height were shown to be quite similar for samples with widely varying average heights (from 6nm to 12 nm). We thank J. John and Sandip Ghosh for their help with the AFM and PL measurements and Sandeep Krishna for his help with the development of the image processing software.

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