Study of annealed InAs/GaAs quantum dot structures

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Abstract. Two similar samples of InAs/GaAs quantum dots and their response to annealing at 700ºC have been studied using scanning transmission electron microscopy. The only difference in the growth of both samples is the inclusion of p-doping by carbon in the GaAs barriers between the quantum dots in one of the wafers. The size and line density of and the change of indium concentration within the quantum dots have been investigated to understand the effect of carbon doping on the diffusion of indium atoms within the structures during the anneal.

1. Introduction

In(Ga)As quantum dots (QDs) emit light at ~1300nm, which can be used for infrared applications such as light emitting diodes (LEDs) and laser diodes. Interdiffusion inside the device due to internal heating can affect the device performance and life time [1]. Recently, carbon doping has been used to reduce the temperature sensitivity of such devices [2-4] but it is unclear how this mechanism works exactly. Therefore, we have studied the change of the microstructure in C-doped and undoped InAs/GaAs QD samples upon annealing.

2. Experimental

Two samples with eight layers of InAs/InGaAs QDs were grown in a molecular beam epitaxy (MBE) system on Si-doped GaAs (100) substrates. Nominally 0.8nm InAs was deposited to form QD layers which was then followed by 5nm InGaAs deposition to reduce the strain. Then, 33nm GaAs barrier layers were deposited (cf. Figure 1). The only difference between the samples is that in one of them the GaAs ~14nm above the QD layers has been p-doped by carbon to a concentration of ~3x10\(^{17}\)/cm\(^3\). A post-growth annealing process (PAP) was applied to both samples for 5mins at 700ºC. In photoluminescence (PL), the carbon-doped sample shows a smaller blue shift of only ~13nm compared to over 50nm for the undoped sample and a stronger PL intensity after annealing (cf. Figure 1). To study the microstructure of these samples, specimens for cross-sectional transmission electron microscopy (TEM) were prepared by standard methods involving glueing, cutting, grinding, polishing and Ar\(^+\) ion milling until perforation. We applied annular dark-field (ADF) STEM using a JEOL 2010F field-emission transmission electron microscope equipped with a scanning unit providing an electron probe of ~0.25nm in size at ~10mrad semi-angle of beam convergence and 0.5nm spherical aberration.

3. Results

Figure 1 sketches the details of the layers and compares the photoluminescence of InAs/InGaAs QDs (with C-doped and undoped GaAs barriers) before and after the annealing process. ADF imaging has
been used at medium magnification (120kX) to determine the spacings between neighbouring QDs and at high magnification (1500kX) to measure the sizes of the QDs (Fig. 3) and the thickness of the InGaAs strain relaxing layer for both samples.

**Figure 1.** Photoluminescence of InAs/InGaAs quantum dots in the C-doped (left) and the undoped sample (right) before and after annealing

**Figure 2.** Overview of the samples before and after anneal: (a1) C-doped sample, (a2) C-doped sample after anneal, (b1) undoped sample, (b2) undoped sample after anneal. The growth direction points towards the top right in all images. Note the high contrast in (a2).
4. Discussion

4.1. Study of quantum dots
The InAs/InGaAs QD sizes and their spacings in C-doped (wafer # 1466) and undoped (wafer # 1467) as-grown samples (1466ag and 1467ag, respectively) and annealed C-doped and undoped samples (1466a and 1467a, respectively) have been measured and compared.

4.1.1. Size of Quantum Dots
Around 50 QDs have been investigated for each sample using ADF imaging at 1500kX magnification to determine the height and length of the QDs. The intensity in ADF imaging is approximately proportional to the square of the average atomic number (“Z-contrast”) if the inner collection angle is very large (high-angle ADF) and the specimen uniformly flat over the field of view. Hence, maps of the chemistry can be obtained for systems such as InGaAs where only two types of atomic species can interdiffuse (here: In and Ga on the group-III sub-lattice). Neglecting contrast reduction due to finite collection angles [5] we can approximate a chemical map by calculating the square root of the ADF image intensity. Then the heights and the lengths of the QDs have been measured as full widths at half maximum (FWHM) of intensity line profiles across these maps. Figures 4 and 5 display the heights and lengths of QDs after annealing for both C-doped and undoped samples.

Figure 3. QD after anneal in the C-doped sample (a) and after anneal in the undoped sample (b). The pseudo colour of the images has been obtained after calculating the square root of the annular dark-field intensity.

Figure 4. QD heights as-grown and after anneal for C-doped (a) and undoped (b) samples.
The heights and lengths of the QDs in the C-doped sample increase during the anneal by 3.2% and 7.8%, respectively. On the other hand, the heights and lengths of the QDs in the undoped sample increase by 1.8% and 11.9%, respectively. This means that the QDs in both samples become larger but undergo shape changes upon annealing, with the QDs in the undoped sample becoming flatter.

The general increase in QD size upon annealing is due to interdiffusion of indium between the InAs QDs and the surrounding GaAs barriers. This must reduce the In concentration in the QDs. The drop of In concentration due to interdiffusion increases the bandgap of the QDs from that of InAs towards that of a ternary InGaAs alloy, consequently a blue shift of the ground state (GS) transitions in PL intensity is observed. On the other hand, the slight increase of the heights of the QDs upon anneal reduces the quantum confinement, which would result in a weaker red shift of GS transitions in both samples.

4.1.2. Line density: spacings between nearest quantum dots

More than 300 distances between neighbouring quantum dots have been measured by ADF overview images taken at 50kX, 60kX, 120kX, 150kX, and 200kX magnifications (examples in Fig. 2). Figure 6 compares the histograms of the statistics of both samples before and after the anneal.

Fig. 6(a) shows that new QDs seem to form between existing ones only in the C-doped sample.
4.2. Study of InGaAs barrier layer thickness

We studied the changes in indium concentration in the barrier layers for C-doped and undoped samples before and after anneal qualitatively by comparing the image contrasts, and we used ADF imaging to monitor thickness changes of the InGaAs strain relaxing layer. Table 1 shows the results.

| Table 1. Comparison of InGaAs layer thicknesses (all values in nm) |
|-------------------------|------------------|------------------|------------------|------------------|
|                        | C-doped          | undoped          |                  |                  |
| sample                 | 1466ag           | 1466a            | 1467ag           | 1467a            |
| measurement 1          | 5.59             | 5.09             | 5.61             | 5.55             |
| measurement 2          | 5.66             | 5.10             | 5.59             | 5.60             |
| measurement 3          | 5.34             | 5.25             | 5.35             | 5.49             |
| measurement 4          | 5.58             | 5.11             | 5.70             | 5.60             |
| average                | 5.54             | 5.14             | 5.56             | 5.56             |
| standard deviation     | 0.14             | 0.08             | 0.15             | 0.05             |
| mean error of average  | 0.07             | 0.04             | 0.08             | 0.02             |

The thickness of the InGaAs barriers above the QDs has been measured by ADF-STEM at 1000kX, 1200kX and 1500Kx magnification at four different locations between the quantum dots. According to Table 1, the barrier layer thickness decreases significantly upon anneal, by about 7%, only for the C-doped sample, whereas the thickness stays roughly the same for the undoped sample. Due to the decrease of the thickness of the InGaAs barriers and the conservation of the number of indium atoms, in the C-doped sample In atoms must have moved from the barriers into the QDs, thereby increasing the indium concentration in the QDs upon anneal by lateral segregation, similar to what has been
observed for strained SiGe/Si layers [6, 7]. This also agrees with the much higher ADF contrast in the annealed C-doped sample compared to the undoped annealed sample (cf. figure 3).

5. Conclusion
The annealing process encourages interdiffusion between the InAs QDs and the surrounding (In)GaAs material, resulting in a decrease in indium composition and hence an increase of the bandgap, which shifts GS transitions to the blue. On the other hand, the slightly increased heights of the QDs upon annealing reduce the quantum confinement, leading to a slight red-shift of GS transitions in both samples. The latter effect is, however, much less pronounced than the change in bandgap due to alloying.

The increase of the In concentration in the QDs in the C-doped sample causes a reduction of the bandgap by increased quantum confinement in this material only and thereby counteracts the dominant effect of interdiffusion. As a result, the PL intensity the GS energy of the C-doped sample is less affected and blue-shifted by only 13nm while the undoped sample is blue-shifted by more than 50nm.

Finally, the strong interdiffusion causes the integrated PL intensity of the undoped sample to drop significantly compared to the C-doped sample, while in the C-doped sample the loss of indium due to interdiffusion is partly compensated by the lateral segregation of indium atoms from the InGaAs capping into the QDs.

In effect, C-doping stabilises both the intensity and the energy of optical emission from the quantum dots, however, this may probably be achieved not by directly suppressing interdiffusion but by compensating out-diffusion of indium from the quantum dots by lateral segregation of indium from the InGaAs layers to the quantum dots. This means C-doping influences the lateral diffusivity of indium atoms in such structures. The main question remaining is whether this is due to some of the doping atoms diffusing down to the QD layers rather than attracting Ga vacancies from there. In this case C acceptor atoms on As sites, which should be the most stable configuration in the GaAs lattice [8], could neutralise the charges on Ga vacancies, which would tend to increase further the already large local lattice distortions at the strained layer interfaces, thereby lowering even further the Peierls stress required to be overcome by diffusing atoms.

References
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