Study of the effect of annealing of In(Ga)As quantum dots

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Abstract. The size, structure and chemistry of In(Ga)As quantum dots as-grown by molecular beam epitaxy and after annealing at temperatures of 700 and 750ºC have been studied by scanning transmission electron microscopy. The influence of the annealing temperatures on the degree of interdiffusion is investigated in detail by studying the size change of the quantum dots, and the values of diffusivities and activation energies for interdiffusion are derived.

1. Introduction

InGaAs based quantum dot (QD) devices have been developed for infrared device applications and demonstrated high performance. Post-growth annealing has been used to tune the wavelength of emission of quantum well (QW) lasers and QW photodetectors [1, 2, 3]. In recent years, post-growth annealing processes have also been used in order to improve QD devices. Hence, it is important to study the effects of annealing on the QDs.

2. Experimental

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 QDs, followed by 5nm InGaAs deposition to reduce the strain. Then, 33nm undoped GaAs barrier layers were deposited. Post-growth annealing processes (PAP) were applied to the sample for 5mins at 700ºC or 750ºC, respectively. To study the microstructure of these samples, specimens for cross-sectional transmission electron microscopy (TEM) were prepared by standard methods, glueing together stacks of material, then cutting, grinding, polishing and finally Ar⁺ ion milling them until perforation. We applied annular dark-field (ADF) scanning transmission electron microscopy (STEM) using a JEOL 2010F field-emission transmission electron microscope equipped with a scan unit, providing an electron beam ~0.25nm in size at ~10mrad semi-angle of beam convergence and 0.5mm spherical aberration constant.

3. Results

ADF and bright field (BF) STEM imaging have been used to study the density and the size of the QDs. Some images are shown below, with the growth direction pointing from bottom left to top right. Figure 1 shows an overview of the sample (a) and ADF-STEM images at higher magnification with weak lattice fringe contrast of the QDs of the as-grown sample (b), the sample annealed at 700ºC (c) and 750ºC (d).
Figure 1. ADF overview of the as-grown sample at 120kX (a); ADF at 1500kX magnification of as-grown sample (b), the sample annealed at 700ºC (c) and annealed at 750ºC (d)

4. Discussion

4.1. Size of Quantum Dots

Around 50 QDs have been investigated for each sample using ADF imaging at 1500kX magnification to determine their heights and lengths. 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 [4] we can approximate a chemical map by calculating the square root of the ADF image intensity. Neglecting the subtraction of the background the detector produces when the electron beam is blanked, typically ~25% for the settings we used, will decrease the contrast ratio of InGaAs to GaAs by a few % but not noticeably change the form of intensity profiles evaluated. The heights and the lengths of the QDs have been measured as full widths at half maximum (FWHM) values of
intensity line profiles across these maps. The ratios of length to height (aspect ratios) have also been calculated in order to understand the annealing effect.

![Graph of QD heights before and after annealing at different temperatures.](image1)

**Figure 2.** QD heights (left) and lengths (right) before and after anneal at different temperatures

![Graph of QD aspect ratios before and after annealing at different temperatures.](image2)

**Figure 3.** QD aspect ratios before and after anneal at different temperatures

The histograms demonstrate that interdiffusion mainly occurs with in the (001) growth plane, rather than along the [001] growth direction. Using Gaussian approximations of the profiles we can estimate the diffusivity, $D$, for different anneal temperatures as well as the activation energy, $E$, for interdiffusion along and perpendicular to the growth direction [5].

The relationship between full width at half maximum (FWHM) and standard deviation $\sigma$ is:

$$\text{FWHM} = 2(2 \ln 2)^{1/2} \sigma \approx 2.355 \sigma$$  \hspace{1cm} (1)

The diffusion lengths $l_i$ at temperatures $T_i$ ($T_1 = 973K$, $T_2 = 1023K$) can then be calculated using $\sigma_i$:

$$l_i = \sqrt{2(\sigma_i^2 - \sigma_0^2)}$$  \hspace{1cm} (2)

Perpendicular to the growth direction we measure $\sigma_0 = 7.79\text{nm}$, $\sigma_1 = 8.60\text{nm}$ and $\sigma_2 = 9.30\text{nm}$ for as-grown, annealed at 700°C and 750°C, respectively. The diffusion length, $l_i$, during anneal for a time $t$ is related to the diffusivity, $D_i$, by:

$$l_i = (2D_i t)^{1/2}$$  \hspace{1cm} (3)

From equations (2) and (3), we can evaluate the diffusivity, $D_i$, at temperature $T_i$ by:

$$D_i = \frac{\sigma_i^2 - \sigma_0^2}{t}$$  \hspace{1cm} (4)

Thus, in the (001) plane, $D_1 = 5.0 \times 10^{-20} \text{ m}^2\text{s}^{-1}$ for annealing at 700°C and $D_2 = 9.1 \times 10^{-20} \text{ m}^2\text{s}^{-1}$ for 750°C. Furthermore, the diffusivity, $D$, can also be expressed in terms of the diffusion constant, $D_0$, by:

$$D_i = D_0 \exp \left(-\frac{E}{kT_i} \right)$$  \hspace{1cm} (5)

where $E$ is the activation energy, $k$ the Boltzmann constant and $T$ the anneal temperature. From this,

$$\frac{D_1}{D_2} = \exp \left(\frac{E(T_1 - T_2)}{kT_1T_2} \right)$$  \hspace{1cm} (6)
Thus, the activation energy is:

$$E = \frac{kT_2 \ln \frac{D_1}{D_2}}{T_1 - T_2}$$

(7)

Within the (001) plane the activation energy for lateral interdiffusion of QDs is calculated as $E_{\text{lat}}=1.0\text{eV}$. Along the growth direction, the In(Ga)As-on-GaAs interface is rather abrupt and the chemical profile may be approximated by just one side of a Gaussian curve, extending into the InGaAs overlayer so that we can evaluate $\sigma$ by using the half width at half maximum (HWHM) instead:

$$\text{HWHM} = (2 \ln 2)^{1/2} \sigma$$

(8)

From this we obtain $\sigma_0=7.02\text{nm}$, $\sigma_1=7.14\text{nm}$ and $\sigma_2=7.39\text{nm}$ for as-grown, anneal at 700°C and 750°C, respectively. Equation (4) gives $D_1=0.61\times10^{-20}\text{m}^2\text{s}^{-1}$ for annealing at 700°C and $D_2=1.8\times10^{-20}\text{m}^2\text{s}^{-1}$ for 750°C. Using equation (7), the activation energy for vertical interdiffusion can be calculated as $E_{\text{vert}}=1.9\text{eV}$, which is about twice as large compared to $E_{\text{lat}}$ and only marginally lower than an earlier measurement of activation energy for vertical interdiffusion in low pressure chemical vapour deposited SiGe/Si layers [6].

4.2. Line density: spacings between nearest quantum dots

More than 300 distances between neighbouring quantum dots have been investigated by ADF and BF STEM overview images taken at lower magnifications of 50kX, 60kX, 120kX, 150kX and 200kX. Figure 4 compares the histograms for the as-grown and annealed samples. At 750°C some of the quantum dots exhibit so strong interdiffusion with the surrounding InGaAs that they disappear from the statistics, causing an additional peak at about twice to three times the original period.

![Figure 4. Spacing between neighbouring quantum dots](image)

5. Conclusion

GaAs/InAs/InGaAs QDs of as-grown and annealed, nominally undoped materials have been studied. The post-growth annealing process causes interdiffusion of In and Ga atoms. The interdiffusion mainly happened within the (001) plane rather than along the [001] growth direction. Diffusivities and activation energies have been calculated along both directions. Results show the diffusivity in the (001) plane, perpendicular to the growth direction, is 5 to 8 times higher than along [001] and the activation energy is halved for lateral interdiffusion. The spacings between neighbouring QDs show that due to interdiffusion some QDs disappear.

References

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