Self organized defect free InAs/GaAs and InAs/InGaAs/GaAs quantum dots with high lateral density grown by MOCVD

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Abstract

We report on the growth of InAs/GaAs and InAs/InGaAs/GaAs quantum dots (QDs) by metalorganic chemical vapor deposition (MOCVD). High density, defect-free InAs/GaAs quantum dots can only be grown in a narrow growth parameter window. The optimum thickness range of ~1.65 monolayers (MLs) has to obeyed within ±10% in order to obtain defect-free high density (10^11 cm^-2) QDs. During the growth interruption after the InAs deposition, the AsH₃ flux also has to be switched off in order to avoid the formation of incoherent clusters. Under optimized conditions, high quality QD stacks with various separation layer thickness have been obtained. A reduction of the inhomogeneous broadening and an increase in efficiency of the room temperature luminescence is observed when the QDs are covered with a thin ternary In₀.₃Ga₀.₇As layer before the deposition of the GaAs cap layer. © 1998 Elsevier Science B.V.

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1. Introduction

Rapid progress in the growth of self-organized In(Ga)As/GaAs quantum dots by molecular beam epitaxy (MBE) in the Stranski–Krastanow growth mode has led to device quality structures enabling the fabrication of QD lasers in the recent years [1–6]. In contrast, MOCVD grown material suffers from the formation of large, dislocated clusters so far. In this study, we present the MOCVD growth recipe for the deposition of high density, dislocation-free single and multiple sheets of InAs and combined InAs/InGaAs QDs. The optical properties of these structures are comparable to or better than those grown by MBE.

2. Experimental

All samples were grown in an AIXTRON-200 MOCVD system equipped with a rotating susceptor, at 20 mbar total pressure with TMIn, TMGa and pure AsH₃ as source gases on exactly oriented (001) (±0.1°) GaAs substrates. After deposition of the GaAs buffer layer at 640°C, the growth was stopped and the temperature was lowered to 480–505°C for the InAs QD growth. After the QD deposition, a growth interruption (GRI) of 4–14 s was applied with both the group-III and group-V precursors...
switched off. Subsequently, the QDs were overgrown with a GaAs cap layer at the same temperature. For stacked QDs the cap layer growth was interrupted after the deposition of 4–18 nm of GaAs and the QD growth procedure was repeated up to 5 times. For a second type of samples, a thin layer of In$_{0.3}$Ga$_{0.7}$As was inserted between the InAs QDs and GaAs cap layer, applying a second GRI after the InGaAs deposition.

Photoluminescence (PL) measurements were performed at room temperature (RT). For excitation, the 514 nm line of an Ar$^+$ laser with an excitation density of 500 W cm$^{-2}$ was used. The luminescence light was detected by an LN$_2$-cooled Ge diode. Transmission electron micrographs were taken in an JEOL JEM 4000 microscope (400 kV). The cross section images were made under (200) and (220) dark field imaging conditions.

3. Results and discussion

The dependence of the RT luminescence on the nominal amount of InAs deposited is shown in Fig. 1. For an InAs thickness of $D_0 = 1.5$ ML, a peak of the 2-dimensional wetting layer (WL) at 1.35 eV and a GaAs related luminescence around 1.43 eV are observed. When the thickness is increased by 7% an additional peak, which is attributed to the QDs, appears at 1.15 eV and dominates the spectrum. Maximum PL intensity of the QD peak is found for $D_{\text{optimum}} = 1.11D_0$. When the layer thickness is further increased by 10%, the QD peak drops in intensity and starts to broaden, indicating degradation due to plastic strain relaxation. Thus we conclude that the optimum thickness range is defined within $D_{\text{optimum}} = 1.65$ ML $\pm 10\%$.

The formation of defect-free QD ensembles in MOCVD critically depends on the AsH$_3$ flux during the QD growth [7] and growth interruption after the QD deposition. In Fig. 2, the PL spectra of three InAs QD single layer samples which were grown under identical conditions, except for the AsH$_3$ flux variation introduced during the GRI, are shown. For all samples, the total GRI time (TMIn switched off) was 14 s. Obviously, the maximum PL efficiency is obtained when the AsH$_3$ flux is switched off for the first 12 s during the GRI. In contrast, when the sample surface is stabilized under AsH$_3$ as is common practice, the QD luminescence drops by more than two orders of magnitude (continuous line). AFM investigations of this sample (not shown here) exhibit a high density ($10^9$ cm$^{-2}$) of large clusters, which are known to be dislocated from TEM measurements [8]. In the intermediate case, when the AsH$_3$ is switched off only for the first three seconds of the GRI, the QD peak intensity is reduced by less than a factor of two as compared to the optimum sample. From this behaviour, it becomes clear that the AsH$_3$ flux during the first stage after InAs deposition is most critical. The presence of AsH$_3$ in all cases leads to a reduction of the QD peak intensity accompanied by a redshift of the peak position and a reduction of its linewidth. Such a behaviour is...
not reported for molecular beam epitaxy of InAs QDs. Therefore we propose that the presence of atomic hydrogen in the MOCVD reactor, due to AsH₃ decomposition, destabilizes the InAs layer. As a consequence, an enhanced surface mobility of the In atoms leads to the formation of incoherent clusters, larger QDs and a reduction of the QD density as compared to the hydrogen-free case.

Under optimized conditions, a defect-free ensemble of QDs with a lateral density of $1.5 \times 10^{11}$ cm⁻² is obtained, as shown in the plan view TEM micrograph in Fig. 3a. The lateral QD size is 9–12 nm thus corresponding to an areal fill factor of ~ 12%. The absence of macroscopic defects allows for the growth of high quality QD stacks. Fig. 3b shows a (200) cross-section TEM image of a three fold stack with 4 nm GaAs separation layer thickness ($d_{sep}$).

![Fig. 3. Plan-view TEM micrograph of a single sheet QD sample (a) and cross-section images of stacked QDs with 4 nm (b) and 18 nm (c) separation layer thicknesses.](image)

The QD luminescence efficiency can be further enhanced by the deposition of a thin layer of InGaAs directly onto the InAs QD layer after the first growth interruption. In Fig. 4, RT luminescence spectra of binary InAs QDs (Fig. 4a) and combined InAs/In₀.₃Ga₀.₇ As QDs (Fig. 4b–e) samples are shown. Both the QD and WL peaks are redshifted, as compared to the InAs sample, upon deposition of 1 nm of In₀.₃Ga₀.₇ As when no second GRI is applied before the GaAs cap layer growth (Fig. 4b). The QD peak intensity is enhanced by 40% and the linewidth is reduced by 10%. The introduction of a second GRI (group-III and group-V precursors switched off)
of 2.5 s–8.5 s duration after the In$_{0.3}$Ga$_{0.7}$As deposition results in a continuous blueshift of the QD luminescence, but does not alter the WL energy significantly (Fig. 4b–e). The GRI also enhances the luminescence efficiency and reduces the QDs peak width, which is minimum for $t_{\text{GRI}} = 2.5–4.5$ s. For an In$_{0.3}$Ga$_{0.7}$As layer thickness of 1.5 nm, we achieved a QD luminescence enhancement of up to 300% as compared to the pure binary InAs QDs. The high optical quality of these binary/ternary QDs allows for the fabrication of low-threshold QD lasers as described elsewhere [9]. The strong impact of the second GRI on the QDs energy and on the inhomogeneous broadening is not expected. We propose a model involving local intermixing of the InAs QDs with the In$_{0.3}$Ga$_{0.7}$As layer resulting in an increase of the effective QD size and a reduction of the In content in the QDs. Such an intermixing leads to a blue shift of the QD energy and also to a decrease of the inhomogeneous broadening of the QD ensemble, as already observed for the interdiffusion of InGaAs QDs with the surrounding GaAs barriers during thermal annealing [10,11].

4. Summary

In summary, we have grown device quality InAs/GaAs QDs with high lateral densities by MOCVD. When the AsH$_3$ flux is switched off during the growth interruption, defect-free single and stacked QD sheets are obtained. The impact of the AsH$_3$ flux on QD and defect formation is attributed to the role of atomic hydrogen during MOCVD growth. For stacked QDs with a small separation layer thickness ($d_{\text{sep}} = 4$ nm), we observe complete vertical ordering. In contrast, the QDs in subsequent sheets are independent of each other for $d_{\text{sep}} = 18$ nm. The optical quality of the InAs QDs is further improved by the subsequent deposition of an additional ternary InGaAs layer and the introduction of a second growth interruption.

References