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Density of InAs/InP(001) quantum dots grown by metal-organic vapor phase epitaxy: Independent effects of InAs and cap-layer growth rates

A. Michon, G. Patriarche, G. Beaudoin, G. Saint-Girons, N. Gogneau, and I. Sagnes^{a)}
*Laboratoire de Photonique et de Nanostructures, LPN/UPR 20-CNRS, route de Nozay,
 91460 Marcoussis, France*

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This letter studies and differentiates the influence of both InAs growth rate and cap-layer growth rate on the density of capped InAs/InP(001) quantum dots (QDs) grown by metal-organic vapor phase epitaxy. The study shows that a decrease of the cap-layer growth rate leads to a reduction of the QD density. This reduction of QD density is of the same order as the kinetic reduction of QD density when decreasing InAs growth rate. Decreasing both InAs and cap-layer growth rates allows one to obtain a low density ($9 \times 10^7/\text{cm}^2$) of QD emitting at around $1.55 \mu\text{m}$. © 2007 American Institute of Physics. [DOI: 10.1063/1.2779101]

Many works have studied the influence of the growth parameters on the density of InAs/GaAs quantum dots (QDs) (see, for example, Ref. 1 and references therein), especially using atomic force microscopy (AFM) on samples grown by molecular beam epitaxy (MBE). The possibility of obtaining high QD density with controlled luminescence properties has led to the fabrication of low threshold and temperature insensitive QD-based lasers emitting at around $1.3 \mu\text{m}$.² Recently, InAs/GaAs QDs have also been successfully used for microphotoluminescence ($\mu\text{-PL}$) and anti-bunching experiments.³ These experiments have become possible thanks to special growth conditions allowing a drastic decrease of the QD density down to about $2 \text{ QD}/\mu\text{m}^2$.

In parallel, InAs/InP QDs have been studied more recently due to their aptitude for $1.55 \mu\text{m}$ emission. Recently, $1.55 \mu\text{m}$ $\mu\text{-PL}$ experiments have been reported for InAs/InP(001) QDs grown by metal-organic vapor phase epitaxy⁴ (MOVPE) or MBE,⁵ showing the interest of these QDs for the realization of telecommunications wavelength single-photon sources. Further studies toward the realization of single-photon sources on InP(001) necessitate a drastic reduction of the QD density. While many works^{6–9} have studied the influence of growth temperature, InAs supply, or group-V pressure on InAs/InP(001) QD density, less works have been reported on the influence of growth rate (GR). Moreover, despite applications require embedded QDs, density are often measured on uncapped samples by AFM. This is prejudicial on one hand because the morphology of uncapped QDs can be changed during cooling from growth to room temperature, thus modifying QD density, and on the other hand because the growth of the cap-layer is liable to modify QD density in the InAs/InP system.¹⁰

In this letter, we present a detailed study of the influence of InAs GR as well as InP cap-layer GR on MOVPE-grown InAs/InP(001) QD density. This study is based on the analysis of transmission electronic microscopy (TEM) on capped samples, thus avoiding change in shape and morphology during the cooling to room temperature on uncapped samples. We report QD density variation over more than two orders of magnitude when varying growth rates. This strong variation

will be discussed by distinguishing the influence of InAs GR and of cap-layer GR on QD density.

The samples were grown by low-pressure MOVPE using H_2 as carrier gas, standard precursor such as trimethylindium (TMI), arsine (AsH_3), and phosphine (PH_3), and a working pressure of 60–70 Torr. A first set of samples was grown using a VEECO D-125 reactor. The QDs are formed by depositing 4 ML (1 ML \approx 0.3 nm) of InAs at 530°C on the InP buffer layer. The QDs are embedded in an InP cap layer. No growth interruption was used after QD growth, and the TMI flow rate was maintained constant after the QD for the growth of the InP cap layer. This standard growth sequence results in proportional InAs and InP GR for each sample.¹¹ Samples P1, P2, P3, P4, and P5 were grown using TMI flow rates leading to InAs GRs of 0.05, 0.2, 0.3, 0.4, and 0.5 ML/s, and corresponding InP cap-layer GRs of 0.04, 0.17, 0.26, 0.35, and 0.44 ML/s, respectively. The arsine flow rates for InAs growth is 6 SCCM (SCCM denotes cubic centimeter per minute at STP) and the phosphine flow rate for InP cap-layer growth is 50 SCCM.

Figure 1 presents TEM plane views in 220 dark field

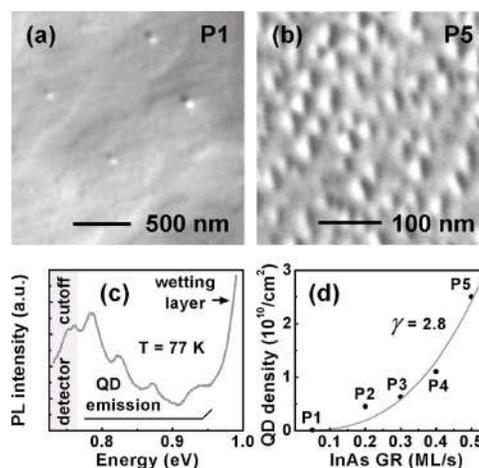


FIG. 1. TEM plane views in 220 dark field conditions of samples P1 (a) and P5 (b). QD densities are about $9 \times 10^7/\text{cm}^2$ for sample P1 (InAs GR = 0.05 ML/s) and $2.5 \times 10^{10}/\text{cm}^2$ for sample P5 (InAs GR = 0.5 ML/s). (c) Low-temperature PL spectrum of sample P1, containing a low QD density ($9 \times 10^7/\text{cm}^2$). (d) QD density measured on TEM-dark field plane views vs InAs GR.

^{a)}Electronic mail: isabelle.sagnes@lpn.cnrs.fr

imaging conditions of samples P1 and P5. In these imaging conditions, QD appears as a strong black-white contrast. Sample P5 [Fig. 1(b)], grown at a high GR, presents a high QD density of about $2.5 \times 10^{10}/\text{cm}^2$. On the opposite, sample P1 [Fig. 1(a)], grown at a lower GR, presents a QD density as low as $9 \times 10^7/\text{cm}^2$. This density, about 20 times lower than that of precedents works,⁵ is the lowest value reported for InAs/InP(001) QDs. The low temperature (77 K) PL spectra of sample P1 [Fig. 1(c)] using a conventional photoluminescence setup (described in Ref. 10) with a cooled Ge-detector (cutoff at around 0.77 eV) present emission from QD at around 0.8 eV ($1.55 \mu\text{m}$). The structuring of the spectra originates from the emission of different QD families with height varying by monolayer step.¹⁰ The realization of low density QD emitting at around $1.55 \mu\text{m}$ is one of the first step toward the realization of single-photon sources.

The QD density deduced from TEM plane views is plotted in Fig. 1(d) as a function of InAs GR. QD density regularly increases with GR. The increase of QD density is correlated to a reduction of the QD size: for example, average lateral sizes are 42.9 nm for sample P1 and 23.1 nm for sample P5. The increase of QD density is often explained by kinetic effects: while GR increases, adatom density on the substrate increases, resulting in a rapid nucleation and in a reduced diffusion length. This behavior of QD density and size is similar to what is observed when the growth temperature⁶ is decreased. Nevertheless, such a strong increase of the QD density (by a factor of 250) with increasing GR (by a factor of 10) has never been reported for InAs/InP(001) QDs.

Various kinetic growth models quantitatively describe the evolution of QD density with GR. In these models, the QD density varies such as GR^γ with $\gamma=1/2$ (Ref. 12), $\gamma=1$ (Ref. 13), or $\gamma=3/2$ (Ref. 14). Surprisingly, the power law fitting of our data gives $\gamma \approx 2.8$, which is undoubtedly higher than all the reported values, inducing that effects other than kinetics may occur and contribute to the strong QD density variation.

Our previous studies¹⁰ have shown that InAs/InP QD density can slightly be decreased during the QD cap-layer growth. An optical study has also shown that cap-layer growth effects become more important with lower GR. Therefore, one can expect that cap-layer GR also modifies QD density. In order to verify this assumption, we have to grow QD sample with an independent control of InAs GR and cap-layer GR. This implies change on TMI flow rate between InAs and cap-layer growth. Moreover, growth interruption is forbidden in order to avoid strong modification of QD morphology. Consequently, TMI flow rate has to be changed directly between InAs and cap-layer growth without growth interruption, requiring two different TMI sources and special configuration of the gas panel of the MOVPE system.

A second set of sample was grown in an EMCORE D-180 vertical-reactor MOVPE system where the gas panel was adapted to allow change on TMI flow rate between two consecutive layers. For this set of sample, the QDs were formed by depositing 2.4 ML of InAs at 510°C on the buffer layer. The arsine flow rate for InAs growth is 10 SCCM and the phosphine flow rate for InP cap-layer growth is 100 SCCM. Reference sample (D0) is grown using identical TMI flow rate for InAs and cap-layer growth. This provides an InAs GR of 0.4 ML/s and a cap-layer GR of 0.2 ML/s. Samples D1–D4 are grown using a special growth

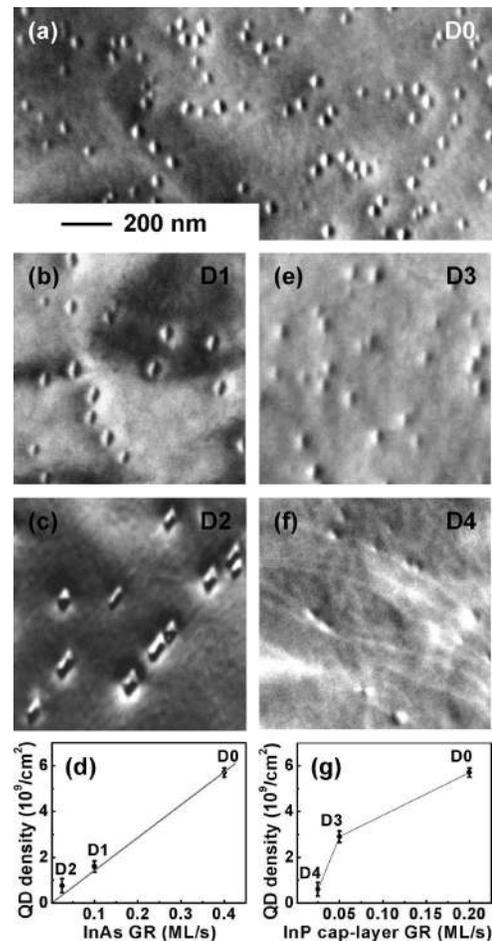


FIG. 2. TEM plane views in 220 dark field conditions of samples D0–D4. Variation of QD density vs (d) InAs GR and (g) cap-layer GR.

sequence with different TMI flow rates for InAs and InP cap-layer growth. For the growth of samples D1 and D2, InP cap-layer GRs are identical than for sample D0, but InAs GRs are decreased to 0.1 ML/s (sample D1) and 0.025 ML/s (sample D2). For the growth of samples D3 and D4, InAs GRs are identical than for sample D0, but InP cap-layer GRs are decreased to 0.05 ML/s (sample D3) and 0.025 ML/s (sample D4). Principally due to the difficulty in accurately measuring the growth temperature in MOVPE, growth conditions of the two MOVPE systems are not directly comparable. However, a lot of characterizations (not shown here) show that QD from the second set of sample present very similar properties than QD of the set of sample presented former.

Figure 2 presents TEM plane views of samples D0–D4 using 220 dark field imaging conditions. All the samples contain QDs. When decreasing only InAs GR (samples D0, D1, and D2), the QD density decreases, as observed for the first set of sample. We have measured QD density on dark field TEM planes views. As observed on the previous set of sample, QD density decreases when decreasing InAs GR. QD density versus InAs GR is plotted in Fig. 2(d). QD density follows the linear law ($\gamma=1$) observed on uncapped samples in different material systems and described by the simple kinetic model of Ref. 13. We observe a small deviation from the linear law for small density due to a larger contribution of the nucleation on atomic step, neglected by the model.

Samples D3 and D4 contain QD similar than QD of sample D0 but are embedded with lower cap-layer GR than for D0. TEM plane views of samples D3 and D4 present QD with a poor contrast. As can be seen in Fig. 2(g), the QD density decreases when decreasing cap-layer growth rate. Our previous studies¹⁰ have shown that reducing cap-layer GR leads to a decrease of the QD height. This effect is shown to be more important for low cap-layer GR. Therefore, we expect that the reduced cap-layer GR for QD of samples D3 and D4 reduces their height to smaller value than for QD of sample D0. This is verified by the analysis of TEM plane views: the poorer contrast on TEM view in 220 dark field conditions of samples D3 and D4 indicates that QDs induce lower strain fields due to reduced height with respect to QDs of the reference sample D0. This evidences an influence of the cap-layer growth rate. The reduction of QD density originates from the desorption of the smallest QD among the height distribution, while taller QD partially resists to the cap-layer growth. We have also grown samples similar to samples D3 and D4 but with lower cap-layer GR. Photoluminescence spectra of these samples grown with very small cap-layer GR (not presented here) show that they do not contain QD. This indicates that for very small cap-layer GR, all QDs are totally desorbed during the cap-layer growth.

Sample D0 presents a QD density of about $5.7 \times 10^9/\text{cm}^2$. We can compare the effects on QD density of (i) a reduction of the InAs GR by a factor of 4, and (ii) a decrease of the cap-layer GR by a factor of 4. In the case of the reduction of the InAs GR (i), the density is reduced by a factor of about 3.6 (samples D0 and D1), and in the case of the reduction of the cap-layer GR (ii), the density is reduced by a factor of about 2.0 (samples D0 and D3). The reduction of QD density when decreasing cap-layer GR is of the same order of magnitude as the reduction observed when decreasing InAs GR. As a result, when decreasing simultaneously both InAs and cap-layer GRs, the combination of these two effects leads to the unusual QD density reduction observed on the first set of sample.

In summary, we have studied the influence of both InAs GR and cap-layer GR on the density of InAs/InP(001) QDs grown by MOVPE. As a first conclusion, we have shown that reducing simultaneously InAs and cap-layer GR by a factor of 10, QD density decreases by a factor of 250. This strong QD density reduction originates, on one hand, from

kinetic effects when decreasing InAs GR and, on the other hand, from the desorption of smaller QDs at lower cap-layer GR. The combination of these two effects allows one to obtain a very low density ($9 \times 10^7/\text{cm}^2$) of QDs emitting at around $1.55 \mu\text{m}$. This density is the lowest reported for QDs emitting at around $1.55 \mu\text{m}$. As a second conclusion, we have shown that cap-layer growth conditions can modify QD density. In particular, we have pointed out that decreasing exclusively InAs GR or exclusively cap-layer GR can lead to reductions of QD density of the same order of magnitude. From our point of view, the dispersion of the results reported on the growth of InAs/InP QD could partly originate from difference in cap-layer growth conditions.

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