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Lateral wavelength control of InAs/InGaAsP/InP (100) quantum dots in the 1.55 μm region by selective-area metal organic vapor-phase epitaxy

D. Zhou, S. Anantathanasarn, P. J. van Veldhoven, F. W. M. van Otten, T. J. Eijkemans, T. de Vries, E. Smalbrugge, and R. Nötzel^{a)}

COBRA Inter-University Research Institute, Eindhoven University of Technology,
5600 MB Eindhoven, The Netherlands

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We report lateral wavelength control of InAs quantum dots (QDs) embedded in InGaAsP on InP (100) substrates by selective-area metal organic vapor-phase epitaxy (SA MOVPE). The technologically important 1.55 μm telecommunications wavelength region is assessed by the combination of ultrathin GaAs interlayers beneath the QDs with proper SiN_x mask design. Atomic force microscopy and microphotoluminescence reveal evolution of the QDs formed by 2 ML InAs as a function of growth rate enhancement with pronounced height and density increase, resulting in a wide wavelength tuning range of 110 nm. Saturation of QD formation is observed for 3 ML InAs supply producing a much smaller tuning range of only 25 nm which is supported by the increasing GaAs interlayer thickness. Hence, two regimes are identified allowing either wide wavelength tuning or wavelength stability of QDs in the 1.55 μm region offering complementary applications of the monolithic integration of optoelectronic devices by SA MOVPE. © 2006 American Institute of Physics. [DOI: 10.1063/1.2398796]

I. INTRODUCTION

Selective-area metal organic vapor-phase epitaxy (SA MOVPE) is widely used for the monolithic integration of optoelectronic devices in a single growth step. It is based on lateral transport of the precursors from nongrowing dielectric masked areas through vapor phase and surface diffusion to unmasked areas, leading to local growth rate enhancement determined by the ratio of mask width to opening.^{1–4} For bulk layers the growth rate enhancement has been intensively studied by measuring thickness and composition, directly related to the diffusion lengths and surface reactivities of the precursors.⁵ Applied to quantum structures, the growth rate enhancement allows lateral tuning of the band gap energy through the quantum size effect which has been successfully demonstrated for quantum wells (QWs) (Refs. 6 and 7) and quantum dots (QDs).^{8,9} For applications in photonic devices operating in the technologically important 1.55 μm telecommunications wavelength region, however, the lateral wavelength control of QDs remains a challenge.

We report lateral wavelength control of InAs/InGaAsP/InP (100) QDs in the 1.55 μm wavelength region by combining SA MOVPE with the insertion of ultrathin GaAs interlayers underneath the QDs. As a function of the GaAs interlayer thickness, the QD emission wavelength is reduced into the 1.55 μm region^{10,11} while the lateral growth rate enhancement of InAs in SA MOVPE increases the QD emission wavelength. Atomic force microscopy (AFM) and microphotoluminescence (micro-PL) reveal two regimes. For small InAs supply [2 ML (monolayers) InAs together with 1 ML GaAs], a wide wavelength tuning range

of 110 nm from 1450 to 1560 nm is obtained together with an increase of the QD height and density, indicating the evolution of QDs as a function of InAs growth rate enhancement. For larger InAs supply (3 ML InAs together with 1.5 ML GaAs) a much smaller tuning range of 25 nm from 1460 to 1485 nm is observed due to saturation of QD formation, supported by the increasing GaAs interlayer thickness. These two regimes enable complementary applications of the monolithic integration of optoelectronic devices where either wide wavelength tuning or wavelength stability of QDs is beneficial in SA MOVPE.

II. EXPERIMENTAL DETAILS

On the InP (100) substrates, misoriented by 2° towards (110), 100 nm SiN_x was deposited by plasma-enhanced chemical-vapor deposition (PECVD) and patterned by conventional photolithography and reactive ion etching (RIE). A scheme of the 2 mm long triangular mask with maximum width of 150 μm and 10 μm wide open stripe along [011] is shown in Fig. 1(a). After cleaning in oxygen plasma and diluted phosphoric acid, the masked substrates were loaded into the MOVPE reactor. Trimethyl-indium (TMI), trimethyl-gallium (TMG), tertiarybutyl-arsine (TBA), and tertiarybutyl-phosphine (TBP) were used as precursors. The reactor pressure was 100 mbars. Growth commenced with 20 nm InP and 20 nm lattice-matched $\text{Q}_{1.25}$ InGaAsP, which is a standard waveguide core material in photonic devices, at 580 $^\circ\text{C}$ before the substrate temperature was lowered to 500 $^\circ\text{C}$ for deposition of the GaAs interlayer and InAs QDs. The InAs (GaAs) supply was 2 (1) ML and 3 (1.5) ML in different samples. The TBA flow rate for InAs QD formation was 1 SCCM (SCCM denotes cubic centimeter per minute at STP).¹⁰ The InAs QDs were capped by

^{a)}Author to whom correspondence should be addressed; electronic mail: r.noetzel@tue.nl

FIG. 1. (Color online) (a) Scheme of the triangular-shaped mask pattern. (b) Growth rate enhancement factor as a function of mask width.

20 nm InGaAsP ($\lambda_0 = 1.25 \mu\text{m}$; $Q = 1.25$), followed by 20 nm InP and a second 20 nm InGaAsP layer on which growth of the GaAs interlayer and InAs QDs was repeated for structural analysis by AFM. The growth rates were 0.75 ML/s for InAs and 0.21 ML/s for GaAs at 500 °C. The optical properties were assessed by micro-PL with a spatial resolution of 2–3 μm using the 632.8 nm line of a He–Ne laser with a power of 10 μW as excitation source. The PL was dispersed by a 1 m single monochromator and detected by a cooled InGaAs charge-coupled device with detection limit at 1.6 μm. The growth rate enhancement was measured by a surface profiler. Growth rate enhancement, AFM, and PL measurements were taken from the center of the 10 μm wide open stripe. Hence, mainly the growth rate enhancement effects related to vapor-phase diffusion are addressed, rather than those due to surface diffusion, leading to additional growth rate enhancement close to the mask edge, decaying on a micrometer length scale.

III. GROWTH RATE ENHANCEMENT

Figure 1(b) depicts the growth rate enhancement factor within the 10 μm wide open stripe as a function of the mask width determined by measuring the layer thickness. The growth rate enhancement factor linearly scales with the mask width. Note that the growth rate enhancement is unity only after a few micrometers outside the masked area. In different experiments no significant dependence of the growth rate enhancement on the orientation of the mask is observed. For mask widths larger than 80 μm strong roughening of the surface morphology occurs. This indicates the onset of large compositional changes in the $Q = 1.25$ InGaAsP layer due to different diffusion lengths and surface reactivities of the

FIG. 2. (Color online) AFM images of the 2 ML InAs QDs with 1 ML GaAs interlayer for mask widths of (a) 0, (b) 10, (c) 20, (d) 40, (e) 60, and (f) 80 μm. The height contrast is 10 nm.

group-III precursors (the group-V precursor supply is assumed to be not affected by the mask⁵) although, in the case of TMI and TMG, the growth rate enhancement of InAs is only slightly larger compared to that of GaAs.¹² For the analysis of the structural and optical properties of the InAs QDs we focus on mask widths less than 80 μm.

IV. QDS FORMED BY SMALL INAS SUPPLY

Figure 2 shows the AFM images of the QDs formed by 2 ML InAs and 1 ML GaAs interlayer in unmasked areas for different mask widths. This combination of InAs and GaAs supply is chosen to shift the QD emission wavelength (see Fig. 3) in the 1.55 μm region.^{10,11} Without GaAs interlayer, the QD emission wavelength is beyond 1.6 μm due to As/P exchange during InAs growth producing too large QDs. Hence, the role of the GaAs interlayer is to suppress As/P

FIG. 3. (Color online) Normalized micro-PL spectra taken at 10 K of the 2 ML InAs QDs with 1 ML GaAs interlayer for different mask widths.

exchange, continuously reducing the QD height and emission wavelength as a function of GaAs thickness. With increasing mask width, i.e., growth rate enhancement, the average QD height increases from ~ 3.5 to 5.5 nm and the QD density increases from $1 \times 10^{10} \text{ cm}^{-2}$ in unmasked areas to $4 \times 10^{10} \text{ cm}^{-2}$ for $80 \mu\text{m}$ mask width. This reveals strong evolution of the QD morphology as a function of InAs amount from underdeveloped QDs in unmasked areas to fully developed QDs in the masked areas.

The evolution of the QD morphology results in a strong shift of the PL spectra to longer wavelengths with increasing mask width, as shown in Fig. 3. The redshift of the PL peak emission wavelength is as large as 110 nm , from 1450 to 1560 nm , for the mask width of $60 \mu\text{m}$. The peak at the short wavelength side, most evident for small mask widths, is attributed to emission from the wetting layer, which is expected to be most pronounced for low QD density. Excited state emission is excluded since the intensity ratio changes only marginally with excitation power. The PL intensity of the QDs drops by a factor of 3 when the mask width increases to $60 \mu\text{m}$. This is attributed to defects formed due to increasing strain accumulation in the InAs QDs and GaAs interlayer, similar to unmasked areas where a decrease of the PL efficiency occurs for InAs supply above 4 ML and GaAs interlayer thickness larger than 2 ML,^{10,11} and/or reduced quality of the Q 1.25 InGaAsP barrier layers due to compositional changes.

V. QDS FORMED BY LARGER INAS SUPPLY

The AFM images for different mask widths of the InAs QDs formed by 3 ML InAs and 1.5 ML GaAs interlayers in unmasked areas are shown in Fig. 4. This combination of InAs and GaAs supplies is chosen to obtain a similar QD peak emission wavelength in the unmasked areas for comparison (see Figs. 3 and 5). Independent on the mask width, the QDs are well developed, and the average QD height of $3.6\text{--}3.8 \text{ nm}$ and density of $(3\text{--}4.5) \times 10^{10} \text{ cm}^{-2}$ merely change. This indicates saturation of the QD formation with a self-limiting QD size in all areas which is reflected in a relatively stable PL peak emission wavelength as a function of mask width, shown in Fig. 5. The PL peak emission wavelength increases by only 25 nm , from 1460 to 1485 nm , for $60 \mu\text{m}$ mask width. A comparison of the PL peak emission wavelength shift as a function of mask width is shown in Fig. 6 for the QDs formed by 2 and 3 ML InAs in unmasked areas. Also for the 3 ML InAs QDs the PL intensity decreases by a factor of 3 for $60 \mu\text{m}$ mask width.

The stability of the QD morphology and emission wavelength in the regime of saturated QDs is supported by the increase of the GaAs interlayer thickness, compensating the QD height and wavelength increase with InAs amount for increasing mask width. Increasing the InAs supply from 3 to 3.5 ML on unmasked substrates results in a slight QD height increase and PL redshift of 20 nm , independent on the GaAs interlayer thickness.^{10,11} On the masked substrate, the corresponding growth rate enhancement of 1.17 assumed for InAs produces a reduced PL red shift of only 12 nm , indicating the effect of the simultaneously increasing GaAs interlayer

FIG. 4. (Color online) AFM images of the 3 ML InAs QDs with 1.5 ML GaAs interlayer for mask widths of (a) 0, (b) 20, and (c) $40 \mu\text{m}$. The height contrast is 10 nm .

thickness. However, the growth rate enhancement of InAs is certainly larger than that of GaAs which is demonstrated by the fact that a thickness increase of 1.17 of the GaAs interlayer on the unmasked substrate produces a PL blueshift of 31 nm .^{10,11} Unfortunately, there is no reference without GaAs interlayer since InAs quantum dashes are formed for submonolayer GaAs coverage.¹⁰ For the QDs formed by 2 ML InAs on the masked substrate the effect of increasing GaAs interlayer thickness is not visible due to the large wavelength shift upon QD evolution with increasing InAs amount. In the saturated regime the QD emission wavelength is stabilized by the interplay between increasing InAs supply and GaAs interlayer thickness.

FIG. 5. (Color online) Normalized micro-PL spectra taken at 10 K of the 3 ML InAs QDs with 1.5 ML GaAs interlayer for different mask widths.

FIG. 6. (Color online) PL peak emission wavelength at 10 K of the 2 and 3 ML InAs QDs as a function of mask width.

VI. CONCLUSIONS

We have investigated the lateral wavelength control of InAs/InGaAsP/InP (100) QDs by SA MOVPE. Through the combination of ultrathin GaAs interlayers beneath the QDs with proper dielectric mask design the technologically important 1.55 μm wavelength region is covered. Micro-PL and AFM reveal a wide wavelength tuning range of 110 nm from 1450 to 1560 nm for small InAs supply (2 ML InAs together with 1 ML GaAs) together with a QD height and density increase due to the evolution of QDs as a function of

growth rate enhancement. For larger InAs supply, (3 ML InAs together with 1.5 ML GaAs) a much smaller tuning range of 25 nm from 1460 to 1485 nm is obtained due to saturation of QD formation and increasing GaAs interlayer thickness. Complementary applications are offered by the two regimes allowing either wide wavelength tuning or wavelength stability in SA MOVPE of InAs/InP QDs in the 1.55 μm tele communications wavelength region for integrated photonic devices.

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