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Low density 1.55 μm InAs/InGaAsP/InP (100) quantum dots enabled by an ultrathin GaAs interlayer

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The authors report the formation of low density InAs/InGaAsP/InP (100) quantum dots (QDs) by metalorganic vapor phase epitaxy enabled by an ultrathin GaAs interlayer. For small InAs amount and low group-V flow rate, the QD density is reduced to below 10 QDs/μm^2. Increasing the group-V flow rate slightly increases the QD density and shifts the QD emission wavelength into the 1.55 μm telecommunication region. Without GaAs interlayer, the QD density is drastically increased. This is attributed to the suppression of As/P exchange during QD growth by the GaAs interlayer avoiding the formation of excess InAs. © *2009 American Institute of Physics*.
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InAs/InP quantum dots (QDs) are ideally suited for applications in fiber based telecommunication systems. One class of applications includes lasers and semiconductor optical amplifiers where excellent performance has been reported.[1–3] These devices require a high QD density to provide gain. The other class of applications requires low density QDs when the device operation is based on single QDs, which have to be isolated such as for single photon sources in quantum information systems. If not employing substrate patterning,[4,5] there are very few reports on the growth of low density InAs QDs on planar InP substrates. Low InAs/InP QD densities have been obtained by the ripening of InAs sticks, which is triggered by the sample cooling under As overpressure in solid-source molecular beam epitaxy[6] and by the reduction of the InAs growth rate and cap-layer growth rate in metalorganic vapor phase epitaxy (MOVPE).[7]

Here we report a different approach for obtaining low density InAs QDs embedded in a lattice-matched InGaAsP matrix on InP (100) by MOVPE, which is based on the insertion of an ultrathin, 1 ML, GaAs interlayer beneath the QDs. Below 10 QDs/μm^2 are obtained for small InAs amount just above the onset of QD formation and low group-V flow rate. Increasing the group-V flow rate tunes the low-temperature QD photoluminescence (PL) emission wavelength into the 1.55 μm telecom region with long PL lifetime without much increasing the QD density. Well-isolated, ultrasharp emission lines from single QDs are easily detected in micro-PL. Without GaAs interlayer a drastic increase of the QD density is observed and the QD density is clearly higher at reduced InAs growth rate even with a GaAs interlayer. Hence, the well-known suppression of As/P exchange during InAs growth by the GaAs interlayer[8,9] enables low density QDs for small InAs amount by avoiding excess InAs formation while strong As/P exchange in the absence of the GaAs interlayer and at high group-V flow rate and low growth rate always causes the QD density to increase.

The samples were grown by low-pressure MOVPE using trimethyl indium, trimethyl gallium, tertiarybutyl arsine (TBA), and tertiarybutyl phosphine as gas sources. The vicinal InP (100) substrates were misoriented by 2° toward (110). The sample structure commenced with 100 nm InP followed by a single layer of InAs QDs with or without a 1 ML GaAs interlayer underneath placed in the center of a lattice-matched quaternary InGaAsP (room temperature bandgap at $\lambda_Q = 1.25$ μm) layer with total thickness of 240 nm. The structure was completed by 100 nm InP, 50 nm InGaAsP, and a layer of InAs QDs with or without GaAs interlayer for assessing the QD morphology by atomic force microscopy (AFM) carried out in tapping mode in air. The growth temperature was 515 °C and the amount of InAs for QD formation was 1.9–2 ML which is just above the onset of QD formation. The InAs growth rate was between 0.15 and 0.70 ML/s. The TBA flow rate was varied between 0.5 and 5 SCCM (SCCM denotes standard cubic centimeter per minute). The growth of the QDs was similar to that detailed in Ref. 8 besides the lower InAs amount. For the PL measurements, a neodymium doped yttrium aluminum garnet laser (532 nm) was used as excitation source with excitation power density of 256 mW/cm^2. The samples were mounted in a He-flow cryostat at a temperature of 20 K. The PL was dispersed by a single monochromator and recorded by a liquid nitrogen cooled InSb detector. For micro-PL, a pulsed laser (pulse width <50 ps) at 750 nm was used. The micro-PL was collected through a microscope objective (numerical aperture=0.5) and coupled to a single-mode fiber to ensure a small collection area of ~2 μm^2. The PL was guided to a superconducting single-photon detector (SSPD)[10] for time-resolved measurements or to a 1 m spectrometer equipped with a liquid nitrogen cooled InGaAs array for high spectral resolution (0.1 nm; 60 μeV) measurements. The SSPD provides a sensitivity over two orders of magnitude higher than that of commercial InGaAs avalanche photodiodes.[11] The output of the SSPD was amplified and sent to the stop input of a correlation card, whose start input was activated by the laser to determine the PL time decay.

Figure 1 shows the AFM images of the 2 ML InAs QDs with 1 ML GaAs interlayer for TBA flow rates of (a) 0.5 SCCM, (b) 3 SCCM, and (c) 5 SCCM. The InAs growth rate is 0.70 ML/s. The average QD density in (a) is 8.5×10^8 cm^{-2}. The surface between the InAs QDs is shallowly modulated with a modulation height around 1.0 nm, prob-

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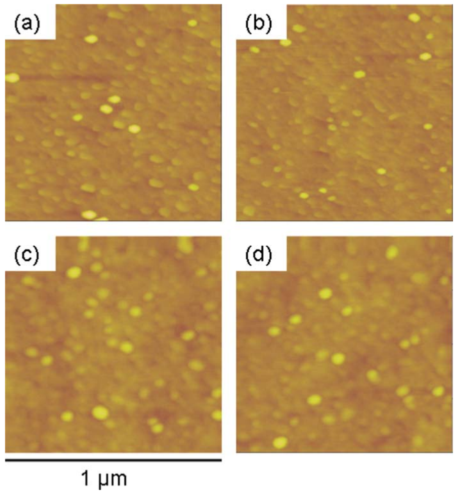

FIG. 1. (Color online) AFM images of 2 ML InAs QDs with 1 ML GaAs interlayer for TBA flow rates of (a) 0.5 SCCM, (b) 3 SCCM, and (c) 5 SCCM. (d) AFM image of 1.9 ML InAs QDs with 1 ML GaAs interlayer for TBA flow rate of 5 SCCM. The InAs growth rate is 0.70 ML/s. AFM scan fields are 1×1 μm^2 and the height contrast is 15 nm.

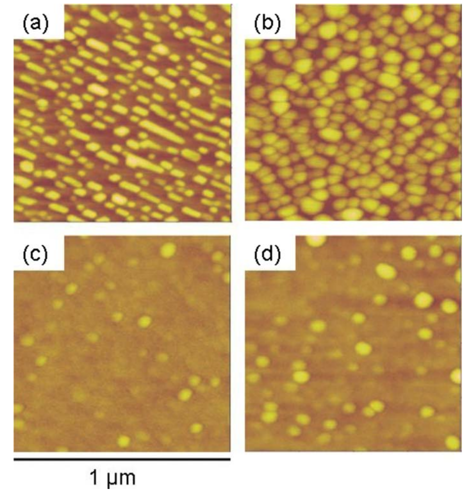

FIG. 3. (Color online) AFM images of 2 ML InAs nanostructures without GaAs interlayer for TBA flow rates of (a) 0.5 SCCM and (b) 3 SCCM. The InAs growth rate is 0.70 ML/s. [(c) and (d)] AFM images of 2 ML InAs QDs with 1 ML GaAs interlayer for reduced InAs growth rate of (c) 0.35 ML/s and (d) 0.15 ML/s. AFM scan fields are 1×1 μm^2 and the height contrast is 15 nm.

ably two-dimensional islands precursors of QDs, which is distinctly smaller than the average QD height of 5.4 nm. For a slightly reduced InAs amount of 1.8 ML, no QDs form and for larger InAs amount the QD density increases. For increasing TBA flow rate, the average density of the QDs increases from 1.0×10^{9} cm^{-2} for 3 SCCM to 1.6×10^{9} cm^{-2} for 5 SCCM, together with their average size.

The PL spectra of these QDs buried in InGaAsP are shown in Fig. 2. The PL emission peak is at 1384 nm for 0.5 SCCM TBA flow rate and redshifts to 1536 nm when the TBA flow rate is increased to 3 SCCM and to 1550 nm for the TBA flow rate of 5 SCCM. This is in agreement with the increase in size due to larger As/P exchange with increasing TBA flow rate. The larger PL linewidth for higher TBA flow rate is attributed to increasing QD size fluctuations for larger As/P exchange, similar to that reported in Ref. 8 for QDs formed by 3 ML InAs. For the TBA flow rate of 5 SCCM the InAs amount has been reduced to 1.9 ML, still forming QDs. Compared to the 2 ML InAs QDs, this reduces the average QD density from 1.6 to 1.3×10^{9} cm^{-2}, see Fig. 1(d), but

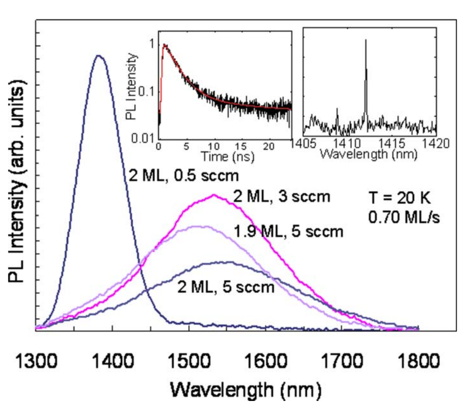

FIG. 2. (Color online) PL spectra taken at 20 K of 2 ML InAs QDs with 1 ML GaAs interlayer for TBA flow rates of 0.5, 3, and 5 SCCM and PL spectrum of 1.9 ML InAs QDs with 1 ML GaAs interlayer for TBA flow rate of 5 SCCM. The InAs growth rate is 0.70 ML/s. Insets: Time resolved PL (left) and micro-PL spectrum (right) of 2 ML InAs QDs with 1 ML GaAs interlayer for TBA flow rate of 3 SCCM.

blueshifts the QD PL peak emission wavelength from 1550 to 1520 nm, shown in Fig. 2. This demonstrates that the target of low QD density plus emission in the 1.55 μm wavelength region demands a very delicate balance between smallest possible InAs amount and TBA flow rate.

In the left-side inset in Fig. 2, the time resolved PL measurement of the 2 ML InAs QDs with 1 ML GaAs interlayer for 3 SCCM TBA flow rate is shown, using a 1400 nm high pass filter. The time decay is fitted by the convolution of a biexponential curve with the time response of the experimental setup. The deduced lifetimes are 2.2 and 10.6 ns. 2.2 ns is related to the lifetime of the QDs, which is in the range of values observed for similar QDs,[12,13] revealing high optical quality. The lifetime of 10.6 ns could be related to the coupling between the bright and dark exciton states.[14] In micro-PL, shown in the right-side inset in Fig. 2, well-isolated ultrasharp lines with resolution limited linewidth of 0.1 nm (60 μeV) are easily detected without the need of mesa etching[15] or metal masks.[16]

In Fig. 3, the AFM images of the 2 ML InAs nanostructures without GaAs interlayer for TBA flow rates of (a) 0.5 SCCM and (b) 3 SCCM are shown. The InAs growth rate is 0.70 ML/s. At the lowest TBA flow rate of 0.5 SCCM, dense dashlike structures are formed with an average density of 2.0×10^{10} cm^{-2}, similar to those formed by 3 ML InAs.[8] For the higher TBA flow rate of 3 SCCM, dotlike structures form, however, with high average density of 2.6×10^{10} cm^{-2}, large size, and redshifted PL peak at 1642 nm, again comparable to those formed by 3 ML InAs.[8] The shape transition in itself is interesting, however, beyond the scope of this paper.

In Fig. 3(c) and 3(d), the AFM images of the 2 ML InAs QDs with 1 ML GaAs interlayer formed at the reduced growth rates of (c) 0.35 ML/s and (d) 0.15 ML/s are shown. The TBA flow rate is 3 SCCM. Clearly the average QD density is increased from 1.8×10^{9} cm^{-2} for 0.35 ML/s to 2.9×10^{9} cm^{-2} for 0.15 ML/s compared to that obtained at 0.70 ML/s of 1.0×10^{9} cm^{-2} and the average QD size becomes larger. Accordingly the PL peak is redshifted from

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1536 nm for 0.70 ML/s to 1554 nm for 0.35 ML/s and to 1614 nm for 0.15 ML/s. Hence, even in the presence of the GaAs interlayer, the increasing residual As/P exchange at lower InAs growth rate which is increasing the QD density and size outweighs the larger In adatom surface migration length to reduce the QD density and increase the size, which is commonly observed for InAs QDs on GaAs where of course no group-V exchange is present.[17] This is opposite to the reduction of the QD density with reduction of the growth rate of 4 ML InAs QDs directly on InP as reported in Ref. 7.

All these observations are in line with the suppression of As/P exchange by the GaAs interlayer allowing the formation of low density 1.55 μm InAs/InGaAsP/InP (100) QDs. Without GaAs interlayer high density and large InAs dashes or dots with long PL wavelength form due to unavoidable formation of excess InAs in the presence of strong As/P exchange. In the samples with GaAs interlayer the QD density and size are small for InAs amounts just above the onset of QD formation and for low TBA flow rate. The QD density and size increase only slightly with increasing TBA flow rate and clearly with reduction of the InAs growth rate due to increasing residual As/P exchange and, for not too low InAs growth rate, the PL redshifts into the 1.55 μm telecom wavelength region.

In conclusion, we have achieved the formation of low density InAs QDs embedded in a lattice-matched InGaAsP matrix on InP (100) substrates by MOVPE. The low QD density is enabled by the insertion of an ultrathin GaAs interlayer beneath the QDs. For small InAs amount at the onset of QD formation and low group-V flow rate the density of the QDs is below 10 QDs/μm^2 and only slightly increases with the group-V flow rate together with the QD size, shifting the QD emission wavelength into the 1.55 μm telecommunication range. Without GaAs interlayer the QD density and size are drastically increased and clearly increased at reduced InAs growth rate even with a GaAs interlayer. This is attributed to the efficient suppression of As/P exchange during QD growth by the GaAs interlayer at not too low growth rate avoiding the formation of excess InAs.

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