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InAs/InP quantum dots emitting in the 1.55 μm wavelength region by inserting submonolayer GaP interlayers

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We report on the growth of InAs quantum dots (QDs) in GaInAsP on InP (100) substrates by chemical-beam epitaxy, with emission wavelength in the 1.55 μm region. Submonolayer coverage of GaP on the GaInAsP buffer before deposition of the InAs QDs results in most efficient suppression of As/P exchange during InAs growth and subsequent growth interruption under arsenic flux. Continuous wavelength tuning from above 1.6 to below 1.5 μm is thus achieved by varying the coverage of the GaP interlayer within the submonolayer range. Temperature dependent photoluminescence reveals distinct zero-dimensional carrier confinement and indicates that the InAs QDs are free of defects and dislocations. © 2004 American Institute of Physics.

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Self-assembled quantum dots (QDs) have been extensively studied in the last decade due to their interest for basic physics research and great potential for optoelectronic device applications. InAs QDs on GaAs and InP substrates are very promising active materials for lasers and semiconductor optical amplifiers used in fiber optical telecommunication systems, provided their emission wavelength is within the 1.31 or 1.55 μm regions. Therefore, for InAs/GaAs QDs efforts are currently concentrated on extending the QD emission wavelength to larger values. On the contrary, methods to obtain shorter wavelengths are desired for InAs QDs in GaInAsP which usually emit at wavelengths above 1.6 μm at room temperature (RT). Postgrowth annealing has been applied to reduce the emission wavelength of InAs QDs on InP (311)B. On InP (100), we have recently introduced a very reproducible method to continuously tune the emission wavelength of InAs QDs in GaInAsP in the 1.55 μm region by inserting ultrathin GaAs interlayers [0.3–2.5 monolayers (MLs)] between the QD layer and the GaInAsP buffer.

In this letter, we exploit the potential of ultrathin submonolayer GaP interlayers being most effective for tuning the emission wavelength of InAs QDs in GaInAsP on InP (100). Both GaAs (Ref. 5) and GaP interlayers reduce the QD emission wavelength due to suppression of As/P exchange reactions and consumption of surface segregated indium. Regarding As/P exchange reactions, besides redshifting the photoluminescence (PL) emission, they are the main cause for rough interfaces and PL line broadening, which is especially relevant for highly strained layers. As/P exchange originates from the relation of bond strengths: The Ga—P bond strength (54.9 kcal/mol) is larger than the Ga—As bond strength (50.1 kcal/mol) while the smaller In—P bond strength (47.3 kcal/mol) is even smaller than the In—As bond strength (48.0 kcal/mol). Therefore, both GaAs and GaP surface termination prevents the substitution of P by As atoms. For GaAs interlayers on GaInAsP, however, As/P exchange is unavoidable at the initial stage of growth for P bound to In. Hence, GaP interlayers are expected to be more efficient in suppressing As/P exchange reactions and, indeed, already submonolayer GaP coverages are found to tune the InAs QD emission wavelength into the 1.55 μm region, less than one-half the necessary amount of GaAs.

The InAs QDs were grown on InP (100) substrates mis-oriented by 2° toward (110) by chemical-beam epitaxy using trimethylindium, triethylgallium, AsH3, and PH3 as precursors. The AsH3 and PH3 gases were thermally decomposed in a high-temperature injector at 900 °C. The InP substrates were indium mounted on Mo blocks, and degassed in the buffer chamber at 200 °C for 30 min before being transferred into the growth chamber. After oxide desorption, a 200 nm InP buffer was grown, followed by 100 nm lattice-matched GaInAsP (λ ⋆ = 1.29 μm) which is a common waveguide core material in InP-based photonic devices. Then, the submonolayer GaP layer with varied coverage and the InAs QDs were grown, which were capped by 100 nm GaInAsP. The same GaP and InAs QD layers were repeated on the sample surface for atomic force microscopy (AFM) measurements. The InAs QDs were formed by 3.2 MLs InAs at a rate of 0.4 ML/s, and 5 s growth interruption under As flux. The growth rate of GaP was 0.155 ML/s and the growth temperature was 500 °C throughout the structures, monitored by an infrared pyrometer. After growth, the samples were cooled down quickly by cutting off the power, and taken out for PL and AFM measurements. For PL, the samples were excited by a Nd:YAG laser with an excitation power density of 256 mW/cm2 in a cryostat at temperatures between 4.8 K and RT. Tapping mode AFM measurements were carried out in air.

Figure 1(a) shows the normalized PL spectra (4.8 K) of the InAs QDs with GaP interlayers of different coverage (0–1.09 MLs) inserted between the InAs QD layer and the GaInAsP buffer. A drastic blueshift of the InAs QD PL line from 1556 to 1485 nm is already observed for 0.31 MLs GaP. The PL line continuously shifts to shorter wavelength with increase of the GaP coverage reaching 1388 nm for 1.09 MLs. The PL linewidths are between 40 and 50 meV except for the sample with 1.09 MLs GaP interlayer, exhibiting a slightly broader linewidth of 57 meV. The PL effi-

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ciency is comparable for all samples. Hence, despite of the high tensile strain there is no degradation of the PL efficiency and linewidth for GaP coverages in the submonolayer range. These GaP coverages are less than one-half of those necessary for GaAs interlayers to realize the same wavelength shift, as shown in Fig. 1(b) for comparison. This confirms the higher efficiency of GaP to suppress the As/P exchange reactions. At RT, the PL peak wavelength reaches 1.55 μm for GaP coverages between 0.6 and 0.8 MLs, and a wide wavelength range to below 1.50 μm is covered when the GaP coverage is increased to 1 ML. The PL linewidths are about 70 meV for all samples and only for the highest GaP coverage above 1 ML the PL efficiency is slightly reduced. PL spectra taken at RT of the InAs QDs with 0.62, 0.78, and 1.09 MLs GaP interlayers inserted between the InAs QDs and the GaInAsP buffer lattice matched to InP are shown in Fig. 2.

The influence of the thin GaP interlayer on the morphology of the InAs QDs is shown by the AFM images in Fig. 3. The InAs QDs formed directly on GaInAsP [Fig. 3(a)] have an average height of 7–8 nm. With increasing GaP coverage, the InAs QD height is continuously reduced to 4–5 nm for 1.09 MLs GaP [Figs. 3(b)–3(f)]. This is a direct consequence of the strong suppression of As/P exchange which is known to raise the aspect ratio of InAs/InP QDs (Ref. 8) and red-shift the PL emission. Given the binary compound bond strengths, P bound to Ga is hardly substituted by As to reduce the As/P exchange reaction rate during InAs QD growth on the GaInAsP surface as a function of the GaP coverage in the submonolayer range. With regard to the observed blueshift of the QD PL emission, although other factors may contribute like: (i) Consumption of surface segregated In by the GaP interlayer on the quaternary buffer to form GaInAsP, (ii) substitution of As by P on the GaInAsP buffer layer surface during the initial stage of GaP growth to partially compensate the subsequent As/P exchange during InAs QD growth, and (iii) enhanced Ga/In intermixing in the QDs in the presence of the GaP layer, its main cause is assigned to the QD height reduction due to the suppression of As/P exchange during InAs QD growth on the GaP terminated GaInAsP surface.

To study the confinement of carriers in the InAs QDs, temperature dependent PL measurements are carried out between 4.8 K and RT for the QD sample with 0.78 MLs GaP interlayer. The dependence of the QD PL peak energy and linewidth as a function of temperature is shown in Fig. 4(a). The PL peak energy and linewidth exhibit a behavior which is very similar to that observed for InAs/GaAs QDs, originating from zero-dimensional carrier confinement in inhomogeneous QD ensembles. At temperatures below 50 K, the PL peak energy is almost constant at 0.876 eV, due to carrier localization in the InAs QDs. A pronounced low-energy shift of the PL peak then occurs in the temperature range from 50 to 150 K, which is much steeper than that of the InAs band-gap energy derived from the empirical Varshni law of the form $E_g(T) = E_g(T=0) - \alpha T^2/(T+\beta)$ with the fitting parameters of InAs, $\alpha=0.276$ meV/K and $\beta=93$ K. The influence of the thin GaP interlayer on the morphology of the InAs QDs is shown by the AFM images in Fig. 3. The InAs QDs formed directly on GaInAsP [Fig. 3(a)] have an average height of 7–8 nm. With increasing GaP coverage, the InAs QD height is continuously reduced to 4–5 nm for 1.09 MLs GaP [Figs. 3(b)–3(f)]. This is a direct consequence of the strong suppression of As/P exchange which is known to raise the aspect ratio of InAs/InP QDs (Ref. 8) and red-shift the PL emission. Given the binary compound bond strengths, P bound to Ga is hardly substituted by As to reduce the As/P exchange reaction rate during InAs QD growth on the GaInAsP surface as a function of the GaP coverage in the submonolayer range. With regard to the observed blueshift of the QD PL emission, although other factors may contribute like: (i) Consumption of surface segregated In by the GaP interlayer on the quaternary buffer to form GaInAsP, (ii) substitution of As by P on the GaInAsP buffer layer surface during the initial stage of GaP growth to partially compensate the subsequent As/P exchange during InAs QD growth, and (iii) enhanced Ga/In intermixing in the QDs in the presence of the GaP layer, its main cause is assigned to the QD height reduction due to the suppression of As/P exchange during InAs QD growth on the GaP terminated GaInAsP surface.

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FIG. 1. (a) Normalized PL spectra taken at 4.8 K of the InAs QDs in lattice-matched GaInAsP on InP (100) with submonolayer GaP interlayers between the InAs QDs and the GaInAsP buffer. (b) Dependence of the PL peak wavelength of the InAs QDs on the GaP and GaAs interlayer coverages for comparison. (The data for the GaAs interlayers are taken from Ref. 5.)

FIG. 2. PL spectra taken at RT of the InAs QDs with 0.62, 0.78, and 1.09 MLs GaP interlayers inserted between the InAs QDs and the GaInAsP buffer lattice matched to InP.

FIG. 3. AFM images of the InAs QDs on GaInAsP lattice matched to InP. Submonolayer GaP interlayers are grown on the GaInAsP buffer before InAs QD deposition. The GaP coverage is (a) 0, (b) 0.31, (c) 0.47, (d) 0.62, (e) 0.78, and (f) 1.09 MLs. The black-to-white height contrast is 10 nm in all images.

FIG. 4. (a) Temperature dependence of the QD PL peak energy and linewidth for the QD sample with 0.78 MLs GaP interlayer. The PL peak energy and linewidth exhibit a behavior which is very similar to that observed for InAs/GaAs QDs, originating from zero-dimensional carrier confinement in inhomogeneous QD ensembles. At temperatures below 50 K, the PL peak energy is almost constant at 0.876 eV, due to carrier localization in the InAs QDs. A pronounced low-energy shift of the PL peak then occurs in the temperature range from 50 to 150 K, which is much steeper than that of the InAs band-gap energy derived from the empirical Varshni law of the form $E_g(T) = E_g(T=0) - \alpha T^2/(T+\beta)$ with the fitting parameters of InAs, $\alpha=0.276$ meV/K and $\beta=93$ K.
This is attributed to thermally activated carrier redistribution, preferentially from small to large QDs. That is also reflected in the PL linewidth, which is almost constant below 50 K, and then undergoes a distinct minimum around 120 K. With a further increase of the temperature, the PL linewidth increases due to equilibration of the carrier distribution in large and small QDs when the probability of carrier escape from the large QDs increases. The PL peak energy then follows the shift of the InAs band-gap energy.

The integrated PL intensity of the InAs QDs is almost preserved up to around 150 K and decreases exponentially at higher temperatures due to thermal quenching, as shown in Fig. 4(b). The activation energy ($E_a$) of the PL quenching can be extracted by fitting the integrated PL intensity to $C/(1+Ae^{-E_a/(k_BT)})$, where $k_B$ is the Boltzmann constant, and $C$ and $A$ are fitting parameters. $T$ denotes the temperature. The thermal activation energy amounts to 148±8 meV which is very close to the difference (143 meV) of the GaInAsP barrier band-gap energy and the InAs QD emission energy. Thus, we conclude that the PL quenching is due to thermionic emission of carriers from the InAs QDs into the GaInAsP barrier, and that, for GaP coverage in the submonolayer range, the InAs QD layer is free of defects and dislocations which may provide nonradiative recombination channels.11

In summary, we have demonstrated that submonolayer GaP interlayers are effective to continuously tune the emission wavelength of InAs QDs embedded in lattice-matched GaInAsP on InP (100). A wide wavelength range from above 1.6 to below 1.5 μm at RT is reproducibly accessed by solely varying the GaP interlayer coverage in the submonolayer range, which is very promising for InAs QD applications in InP-based optoelectronic devices operating in the 1.55 μm wavelength region. The continuous wavelength tuning is attributed to a drastic suppression of As/P exchange reactions on the GaInAsP buffer layer surface. Temperature dependent PL measurements clearly reveal zero-dimensional carrier confinement in the InAs QDs which are free of defects and dislocations.

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