Letter to the Editors

InAs quantum dots in InAlAs matrix on (0 0 1)InP substrates grown by molecular beam epitaxy


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Abstract

InAs quantum dots grown on InAlAs lattice-matched to (0 0 1) InP substrates by molecular beam epitaxy are investigated by double-crystal X-ray diffraction, photoluminescence and transmission electron microscopy. The growth process is found to follow the Stranski–Krastanow growth mode. The islands formation is confirmed by the TEM measurements. Strong radiative recombination from the quantum dots and the wetting layer is observed, with room temperature PL emission in the 1.2–1.7 μm region, demonstrating the potential of the InAs/InAlAs QDs for optoelectronic device applications.

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Heteroepitaxial growth of highly strained structures has gained increasing interest as it offers the possibility to fabricate self-organized structures like quantum dots (QDs) and quantum wires without any substrate patterning process. Molecular beam epitaxy (MBE) has been used successfully to fabricate various QDs on GaAs substrates, resulting in emission from the visible for AlInAs/AlGaAs [1] and InP/InGaP [2], to the near-infrared for InGaAs/GaAs [3–6] and for GaSb/GaAs [7]. InP is another very important substrate for today’s semiconductor technologies. Although reports are available on the chemical beam epitaxy (CBE) and metalorganic chemical vapor deposition (MOCVD) grown InGaAs or InAs microstructures on InP [8–11], there are relatively few studies on the formation of InAs QDs on InP substrates by solid source MBE, probably because of the difficulties inherent to the growth of InAlAs ternary alloy buffers which complicate the situation and make it challenging to fabricate QDs with regular shape. The InAs/InAlAs/InP system might be ideal for the...
realization of low threshold-current QD lasers operating at the technologically interesting wavelength region of ~1.55 μm. Therefore, it would be of interest to determine if the strain can be used to produce self-assembled QDs on InP substrates.

In this work, we have grown InAs QDs on InAlAs lattice-matched to (0 0 1)InP substrates. The growth process is found to follow the Stranski–Krastanow growth mode with the onset to three-dimensional growth occurring below 2.5 monolayers (MLs). The island formation is confirmed from the TEM measurements. Strong radiative recombination from the quantum dots and the wetting layer is observed, with room temperature PL emission in the 1.2–1.7 μm region.

The samples were grown on semi-insulating (0 0 1)-oriented InP substrates at 500°C by a Riber 32P MBE system equipped with a 30 kV reflection high-energy electron diffraction (RHEED) system. The substrates were etched with H₂SO₄ : H₂O₂ : H₂O = 7 : 1 : 1 solution for 7 min at 45°C. The In and Al fluxes were calibrated from RHEED intensity oscillation measurements recorded during the growth of In₀.₅₂Al₀.₄₈As. After the desorption of the native oxide layer, a 200 nm In₀.₅₂Al₀.₄₈As layer was first grown as a buffer layer at a rate of 0.7 μm/h and under beam equivalent pressure of 1 × 10⁻⁷ Torr. Following a growth interruption of 60 s, InAs layers of different thicknesses are deposited at a rate of 0.26 ML/s. The nominal InAs layer thickness lies between 2.5 and 10 MLs in different samples. Then the second growth interruption of 60 s is introduced. Finally, a 50 nm In₀.₅₂Al₀.₄₈As cap layer terminates the structural growth.

The structural properties of the samples were investigated by double-crystal X-ray diffraction (DCXD) and transmission electron microscopy (TEM). DCXD is used to examine the degree of lattice matching of the InAlAs buffer layers to the InP substrate, and to optimize the growth parameters to obtain InAlAs buffer layers with flat surface morphology. TEM samples were prepared with mechanical polishing followed by ion milling from the backside. This provided an overview of the QDs confined in the InAlAs matrix.

The optical properties were assessed by Fourier transform photoluminescence (PL) from 15 to 300 K using an Ar ion laser emitting at 513.2 nm. The spectra were dispersed with a IF120HR Fourier transform infrared spectrometer and detected with an InGaAs p–i–n detector. The spectra were not corrected with the response of the detector.

The evolution of the InAs quantum dots is studied in situ with reflection high-energy electron diffraction (RHEED). During the InAlAs growth, a streaky reflection pattern is observed. A superimposed transmission pattern becomes faintly visible during the deposition of the third ML of InAlAs which reveals the transition from 2D to 3D growth, i.e., the conventional Stranski–Krastanow growth mode. The intensity of the spots increases with the amount of In supplied and becomes comparable to the bulky pattern after 6 ML InAs is deposited. The two-dimensional character is quickly recovered after In₀.₅₂Al₀.₄₈As cap layer growth is resumed. The number of monolayers required for the self-organized island formation was well characterized for InGaAs/GaAs [3–6] and AlInAs/AlGaAs [1]. The strain for the InAs pseudomorphic to InP (3.2%) is slightly smaller than the one for In₀.₅₅Ga₀.₄₅As and In₀.₇Ga₀.₃As/ GaAs, in which case ~4 ML would have been required for the island formation [3]. In contrast, here island formation was clearly observed even for the 2.5 ML samples. TEM studies generally confirm this observation. This indicates that the InAs on In₀.₅₂Al₀.₄₈As/InP might have a different equilibrium for the surface, interface, bonds and dislocation energies [12].

Fig. 1 shows the experimental DCXD pattern of the sample containing 4.5 ML InAs. Breaking the continuity of the In₀.₅₂Al₀.₄₈As lattice parameter by the InAs produces a dephasing phenomenon between the X-ray wave fields diffracted by the buffer and cap layer which are coherently related by the Bloch wave criterion. The beating between these two fields modulates the reflectivity, giving rise to Pendellösung fringes. Any interface imperfection or composition fluctuation as well as any strain relaxation would have caused a strong damping of the fringes. Our findings, thus, show that any relaxation in these In₀.₅₂Al₀.₄₈As/InAs/In₀.₅₂Al₀.₄₈As structures does not occur. The typical lattice-mismatch of our samples is < ± 1 × 10⁻³.
Nominal 2.5, 4.5, 5 and 6 ML thick InAs layers are prepared for TEM observation. The plan-view TEM micrographs of the samples with 2.5, 4.5 and 6 ML InAs are shown in Fig. 2a–c, respectively. The quantum dots are clearly evident through the strain contrasts as previously observed for the case of InGaAs/GaAs quantum dots [3–6]. The size and density of QDs increase with the InAs thickness. Though the growth conditions are all the same, the distribution of the QDs in these samples are strikingly different. The 4.5 ML samples shown in Fig. 2b reveal a high density \( (2.5 \times 10^{10} \text{ cm}^{-2}) \) of dots oriented along the \( \langle 110 \rangle \) direction, similar to the findings for MOCVD and MBE grown InGaAs/GaAs QDs [13,14], while the dots in the 6 ML sample seems to be arranged randomly (Fig. 2c). Details analysis for this will be discussed elsewhere. Two different sizes of InAs islands appear as seen in Fig. 2c, i.e., small island about 30 nm in diameter or less, and large islands, greater than 40 nm. Most of the quantum dots are round-shaped, and the aerial coverage of the quantum dots is about 20%. Fig. 2d shows the cross-sectional TEM micrographs of 4.5 ML sample. The island height is about 5 nm.

Using the plan-view TEM results, statistics of the QD size can be extracted. For the samples with 6 ML InAs, a mean diameter of 39 nm with standard deviation of 6 nm is obtained. For the
coherent Stranski–Krastanow growth mode, the best island uniformity has been found to be for thickness just slightly larger than the critical thickness to induce the island formation. Therefore, more uniform QDs are likely to be achieved by optimizing the growth conditions, and also by carefully monitoring the surface morphology during the growth. This is confirmed by our 2.5 ML sample (Fig. 2a) in which very uniform QDs with the mean island diameter of 28 ± 2 nm are observed.

The InAs QDs confined by In$_{0.52}$Al$_{0.48}$As give room temperature luminescence when the thickness of InAs is below 6 ML. The relatively weak luminescence intensity for thicker samples is attributed to the partial strain relaxation which degrades luminescence significantly. We have plotted in Fig. 3 the PL spectra of the samples with 5 ML InAs measured at different temperatures between 15 and 300 K. No luminescence from the In$_{0.52}$Al$_{0.48}$As buffer (expected at ~1.5 eV at 15 K) is detected. So the PL at longer wavelength can unambiguously be attributed to the InAs layer. The broad PL peak located around 0.8 eV (in the 1.2–1.7 μm region) is considered to be originated from the InAs quantum dots, while PL peaks above 1.0 eV are attributed to the luminescences from the InAs wetting layer. The shoulders of the wetting layer-related peak are originated from surface areas with one monolayer variation in the InAs quantum well width [8,15]. The mean energy splitting between the shoulders and the main peak is about 50 meV, which is in good agreement with previous observation [15,16]. The fairly large PL linewidths of the QDs-related peak is probably because the large size fluctuation of the QDs. It should also be noted that the response of the InGaAs detector drops off abruptly near 0.75 eV, and therefore the line shape of the PL spectra at low-energy edge is distorted.

In summary, we have fabricated InAs QDs in InAlAs matrix lattice-matched to InP(0 0 1) by MBE. The QDs formation is confirmed from the TEM as well as the PL measurements. The luminescence from both QDs and wetting layer are observed. In addition, we have demonstrated room temperature luminescences from these QDs, which is important for application in optoelectronic devices, especially in QD lasers. The growth of InP-based QD lasers is under current investigation.

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References