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MOCVD growth and optical characterization of strain–induced quantum dots with InP island stressors

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Abstract

Coherent InP islands, grown by low pressure metal organic chemical vapor deposition (MOCVD), were used to produce quantum dots by strain confinement. Lateral confinement of carriers in a GaAs/AlGaAs quantum well located near the surface was obtained from the inhomogeneous strain produced by the InP islands. The evolution of the InP islands on $Al_{0.3}Ga_{0.7}As$ surfaces with increasing InP coverage at different growth temperatures and substrate orientations was studied using atomic force microscopy. Under certain growth conditions, a fairly uniform distribution of coherent InP islands was obtained which had an average apparent diameter of 140 nm with a standard deviation of 12.2 nm and height of 19.5 $\pm$ 1.1 nm. Lateral confinement depths up to 100 meV were obtained when using the islands as stressors. Photoluminescence from ensembles of the strain-induced dots exhibit multiple peaks, narrow line widths (16 meV) and high efficiency up to room temperature.

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1. Introduction

Semiconductor structures with three-dimensional quantum confinement, commonly called quantum dots (QD), are currently under active investigation to develop a basic understanding of the physics of 0D confined states. Theoretical predictions [1] suggest that the properties unique to the quantum dots will lead to improved practical devices, such as QD lasers. Strain confinement of carriers in a quantum well (QW) was investigated several years ago [2, 3] as a means of fabricating quantum wires and dots. A strained stressor layer under compression is patterned by lithography and etching, allowing it to expand and deform the substrate. The inhomogeneous strain field induced by the patterned stressor results in a localized reduction in the band gap due to deformation potential shifts. The carriers are then confined in a near-surface QW laterally by the strain-induced potential well. The lateral confinement potential for dots

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was found [4] to range from 15 to 60 meV as the size of a compressively strained amorphous carbon stressor increased from 100 to 800 nm. Patterned pseudomorphic InGaAsP [2] and InGaAs [3] layers have also been successfully employed to produce dots with a lateral band-gap modulation of around 14 meV for dot sizes of ~100–200 nm. Typically, only small lateral confinement is obtained with conventional patterned stressors of sufficiently small size to observe quantum-confinement effects. The maximum strain transfer from the stressor to the QW structure is limited due to the adhesion of deposited films or dislocation formation in pseudomorphic layers.

A recent novel approach to strain confinement which provides both a large lateral confinement and a small size relies upon the strain produced by coherent InP islands formed by the Stranski–Krastanow (SK) growth mode. High optical quality strain-induced dots were obtain in strained InGaAs wells [5, 6] with GaAs barriers and in GaAs wells [7] with AlGaAs barriers. The SK-growth mode leads to three-dimensional island formation in a number of highly lattice-mismatched semiconductor systems [8–11]. In the initial stages of SK island growth, islands form and grow without dislocations [8, 9]. Both the islands and the substrate are elastically deformed with strain fields extending a distance [9, 12] of approximately one island diameter into the substrate. These coherent islands have a large mismatch with the substrate (3.7% for InP on GaAs) and, therefore, induce a much larger band-gap shift in the substrate (or QW) than has been obtained with patterned stressors of similar size.

In this work, we have investigated the influence of the growth temperature, InP coverage and substrate orientation on InP island formation and evolution on AlGaAs using atomic force microscopy (AFM). InP island formation on AlGaAs is interesting and important because, as we will show, high-quality strain-induced dots can be produced using these islands in GaAs/AlGaAs quantum wells, which is the prototype QW system. The optical characteristics of the strain-induced quantum dots produced in GaAs/AlGaAs QW by the InP island stressors was studied by low-temperature photoluminescence (PL) spectroscopy.

2. Experimental procedure

The samples were grown by low-pressure metal organic chemical vapor deposition (MOCVD) at 75 Torr utilizing arsine, phosphine, triethylgallium (TEG), trimethylaluminum (TMA) and trimethylindium (TMI) as the precursors. A single QW structure consisting of a 50 nm GaAs buffer, 100 nm AlGaAs inner barrier, a 2–5 nm GaAs QW and 2–15 nm AlGaAs outer barrier was first deposited at 750°C. The aluminum mole fraction in the AlGaAs barrier layers was 0.3–0.35. After the growth of the QW, the sample was cooled with an arsine overpressure of 0.15–0.3 Torr to the InP deposition temperature. InP was deposited at temperatures of 580–680°C, V/III ratios of 215–430 and a growth rate of 0.56 monolayers (ML)/s at a TMI flow rate of 4.2 μmol/min. Before the growth of InP, a 5 s hydrogen purge was used to flush arsine from the reactor. InP deposition times ranged from 2 to 24 s. The samples were then cooled under a phosphine overpressure of 0.3 Torr. Island morphology on GaAs(1 0 0) $\pm$ 0.5° direct and viscinal substrates with miscuts of 2° and 4° was examined by AFM in air with the tip in contact mode.

PL spectroscopy was performed over the temperature range of 10–300 K in a variable temperature dewar. The 488 nm line of a CW argon-ion laser was used as the excitation source. Laser power was varied between 1 μW and 50 mW and focused to a nominal spot size of ~100 μm.

3. Results and discussion

A series of samples grown on a (1 0 0)-oriented QW with increasing amounts of InP deposited at 620°C was used to study island development as a function of InP coverage. AFM images for five samples with nominal InP coverages of 3.4–13.4 ML are shown in Fig. 1. With 3.4 ML of InP (Fig. 1a), the InP has not yet undergone the 2D layer to 3D island transformation. The average roughness (~3Å) of this surface is comparable to that measured on the starting substrate surface. With only 1 ML additional InP (Fig. 1b), 3D island formation has occurred at 4.5 ML. The threshold thickness for 2D to 3D growth depends upon

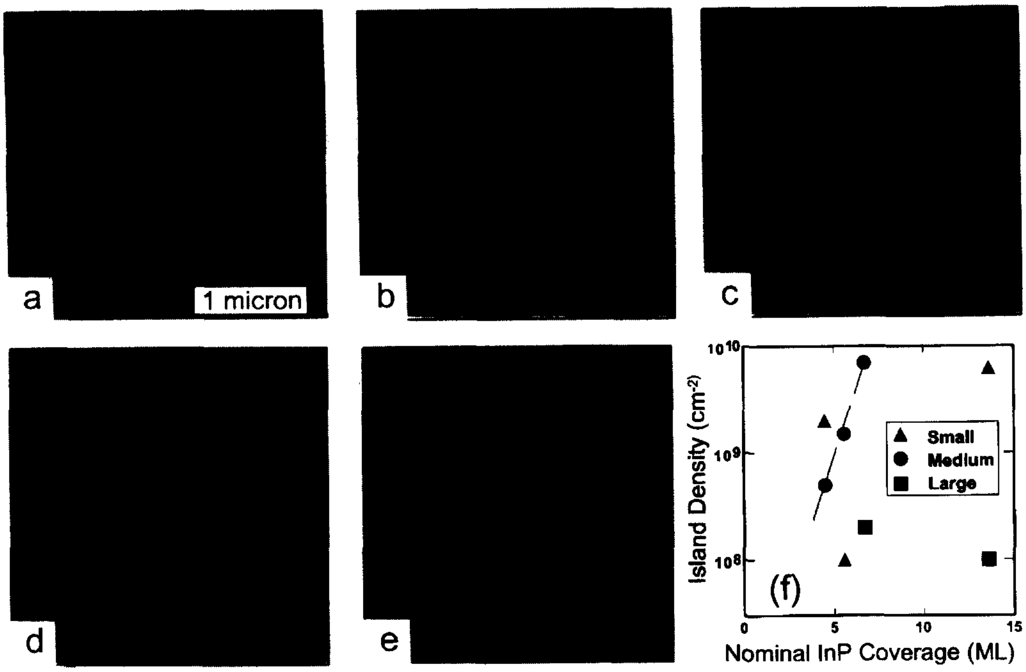

Fig. 1. AFM images of InP island evolution as a function of increasing nominal InP coverage for InP deposited at 620°C: (a) 3.4 ML, (b) 4.5 ML, (c) 5.6 ML, (d) 6.7 ML, and (e) 13.4 ML. The island density vs. InP coverage is shown in (f) for three distinctive size groups: small (diameter, $d < 100$ nm), medium ($d = 100$–150 nm) and large ($d > 150$ nm). Three-dimensional islands form at approximately 4 ML InP deposition for these growth conditions. Island size ripening is observed between 4.5 and 5.6 ML, and larger, faceted islands form at higher coverages.

growth parameters such as temperature, deposition rate and substrate orientation [13]. InP 3D growth has been previously found to occur on (1 0 0) surfaces at 1.8 ML at 580°C for GaInP/GaAs [14], at approximately 1 ML at 650°C for GaInP/GaAs [15], and at 2.5 ML at 650°C for InP grown on GaAs [13].

The islands on the samples of Fig. 1b–Fig. 1e were classified into three size groups: small, diameter $d < \sim 100$ nm, medium, $100 < d < 150$ nm and large, $d > \sim 150$ nm, which are similar to those reported by Reaves et al. [12]. Transmission electron microscopy studies [12] of InP islands found that the small and medium-size islands are coherent with the substrate, while the faceted large islands may contain dislocations.The areal densities of the three island types are plotted vs. ML coverage in Fig. 1f. We should point out that the island diameters reported here are obtained directly from AFM images and have not been corrected for tip-convolution effects, so the actual island size may be overestimated by as much as a factor of 2 [13]. For the sample with 4.5 ML (Fig. 1b), the size distribution is bimodal, with average diameters of 70 and 140 nm for the small and medium-size islands, respectively. At 5.6 ML, the size distribution is peaked around 140 nm with a standard deviation of 15 nm. Further InP deposition to 6.7 ML increases the density of the medium-size islands to 8×10^9 cm^{-2}. Additionally, large-sized islands with a faceted shape have formed with a density of 2×10^8 cm^{-2}. As the coverage is increased to 13.4 ML, we find that the large islands have grown rapidly with little change in their density, while the size of the medium islands have decreased significantly to approximately 85 nm. Once the large, partially relaxed islands form, they grow much faster than the smaller coherent islands

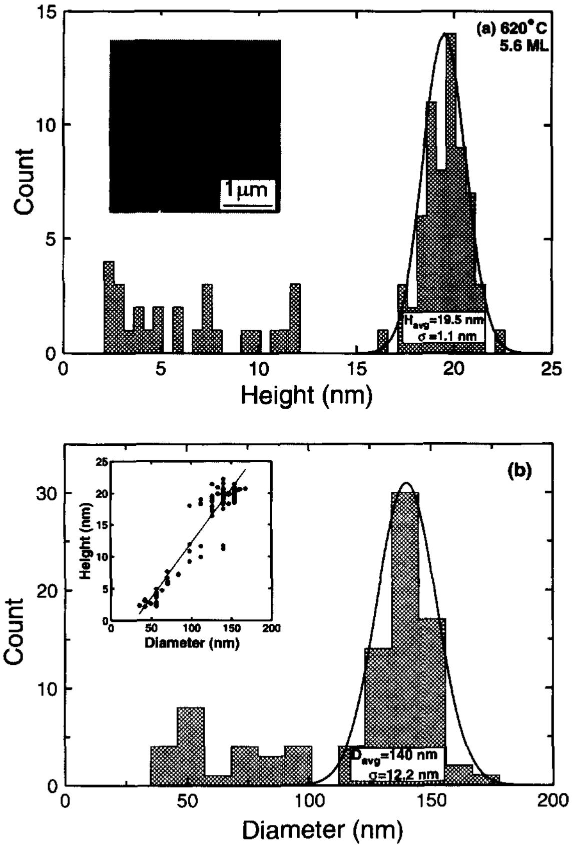

Fig. 2. (a) Island height distribution and (b) diameter distribution for 5.6 ML InP grown at 620°C on a (1 0 0)4° → (1 1 1)B quantum well. At 620°C, the size distribution is quite narrow. The inset in (a) shows an AFM image ($2 \times 2\ \mu m^2$) of the islands. The inset in (b) shows a linear relation between island height and diameter. The nonzero offset in the diameter is due to AFM tip-convolution effects.

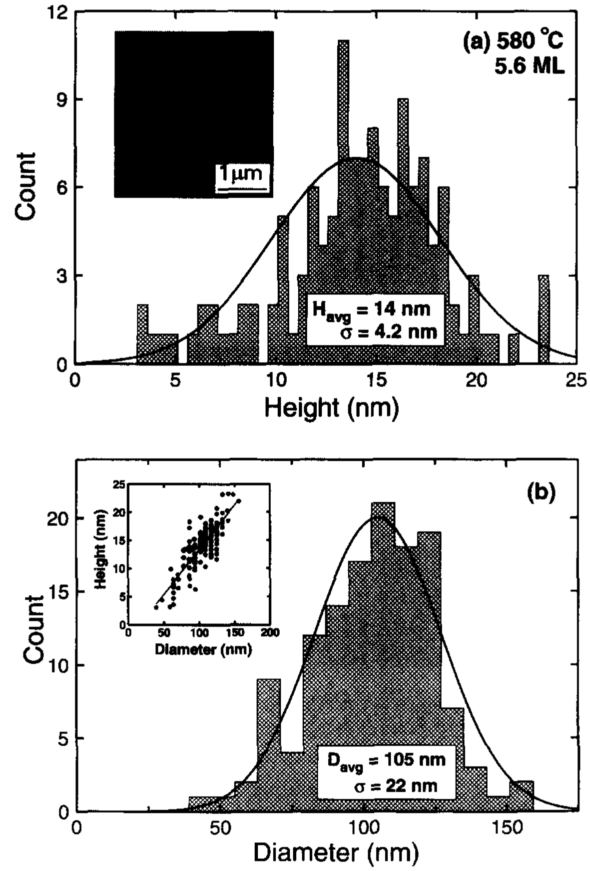

Fig. 3. (a) Island height distribution and (b) diameter distribution for 5.6 ML InP grown at 580°C on a (1 0 0)4° → (1 1 1)B quantum well. The inset in (a) shows an AFM image ($2 \times 2\ \mu m^2$) of the islands. The inset in (b) shows a linear relation between island height and diameter.

due to strain energy considerations [16, 17]; therefore, the large islands act as an effective sink for supplied adatoms, and also for previously deposited InP which causes the size reduction of the coherent islands. Qualitatively, similar island development has been observed for InP islands on GaInP [12] or GaAs [13] and Ge islands on Si [16].

A series of samples with 5.6 ML of InP was grown at 580, 620 and 680°C to investigate the influence of InP growth temperature on InP island size and density. The island density increased as the deposition temperature decreased as expected from reduced adatom diffusion at lower growth temperatures. The densities were $3.3 \times 10^9\ cm^{-2}$ at 580°C, $2.3 \times 10^9\ cm^{-2}$ at 620°C, and $< 2.5 \times 10^7\ cm^{-2}$ at 680°C. In Figs. 2 and 3, we plot the AFM determined island height and diameter histograms for InP deposited on (1 0 0)4° → (1 $\bar{1}$ 1)B QW samples at 620 and 580°C, respectively. Islands grown simultaneously on (1 0 0)2° → (1 1 0) QW samples had similar size statistics, but with about 40% lower density. The 620°C sample has a distinct narrow size distribution (<9%) of symmetric medium-sized islands, as summarized in Fig. 2. The coherent island size of ~140 nm obtained under these growth conditions does not easily increase due to an energetic barrier for misfit dislocation formation in the islands [17, 18]. This growth behavior tends to self-limit the maximum coherent island size and helps to achieve uniform size distributions. A narrow size distribution is important in obtaining narrow luminescence line width from ensembles of strain-induced dots. At 580°C, the islands are elongated in the $(0\,\bar{1}\,1)$ direction and their size distribution is somewhat broader and the average height and diameter are smaller than the islands grown at 620°C. These results indicate that the island size uniformity can be controlled effectively by the growth temperature for a given InP coverage. A linear relation between height and diameter is found, which indicates that both dimensions evolve uniformly during growth.

In Fig. 4, we present a series of luminescence spectra measured at 10 K with excitation powers of 1 μW–1 mW for a sample with a 2.5 nm QW and 10 nm $Al_{0.34}Ga_{0.66}As$ outer barrier. A nominal 5.6 ML of InP was deposited at 620°C resulting in primarily medium-size InP islands (density $\sim 1 \times 10^9$ cm^{-2}) and some $\sim$200 nm large islands at a much lower density ($\sim 4 \times 10^7$ cm^{-2}). At very low excitation powers, we see two clearly resolved, narrow peaks which are red-shifted from the QW peak by approximately 50 and 100 meV, respectively. The separation between these two peaks (50 meV) is too large to assign them to the ground and first excited state dot transitions based upon calculations [19] of energy levels in similar-size InP island strain-induced dots. We tentatively ascribe the two QD peaks seen in the 1 and 10 μW spectra to emission from dots formed by the medium and large islands. The large islands, which may be partially relaxed, would induce a larger strain in the QW and result in a larger red-shift (100 meV) compared to the medium-size islands. The peak associated with the large islands also has a narrower FWHM of 16 meV. This peak saturates as the laser pump power is increased because of the low density of the large islands in this sample. The QD emission red-shifted by $\sim$50 meV is due to the strain-induced dots from the medium-size islands which have a high density ($\sim 1 \times 10^9$ cm^{-2}) and consequently does not saturate with increasing pump power. Emission from dots formed by different-sized island stressors has also been observed previously [5, 7]. The broad emission in the spectral range 1.55–1.60 eV is believed to originate from the InP islands [20] on the surface as it is observed in other samples. The dot and well emission was clearly seen at 300 K. The QD emission intensity at 300 K was $\sim\frac{1}{5}$ the intensity of the QW for the sample of Fig. 4.

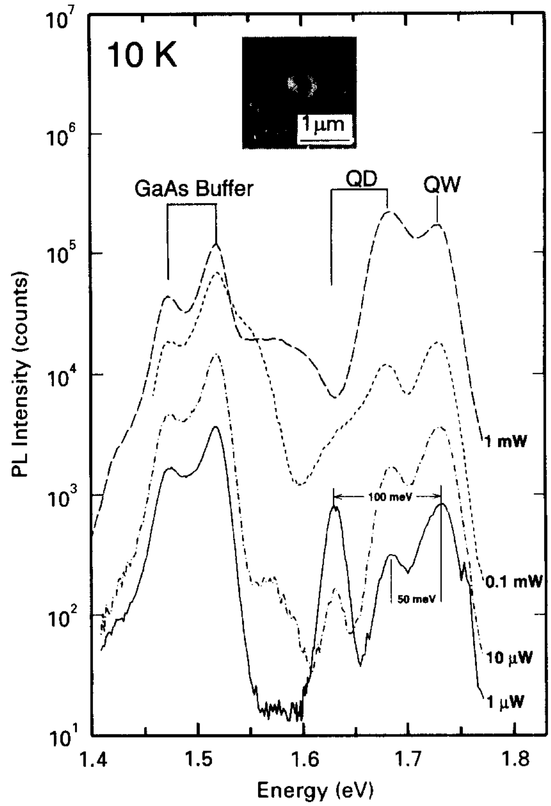

Fig. 4. Laser power dependence of the 10 K photoluminescence of a sample with 5.6 ML InP deposited at 620°C. (Note that this is a different sample from the one presented in Fig. 2.) The AFM image (2×2 μm^2) shown in the inset shows many medium and a single large-size islands. At the lowest powers of 1 μW ($\sim$10 mW/cm^2) and 10 μW, clearly resolved peaks at 1.685 and 1.63 eV (FWHM = 16 meV) are observed, and are believed to originate from the dots associated with medium and large-sized islands, respectively.

4. Summary

Coherent Stranski–Krastanow InP islands employed as stressors were found to produce high-quality strain-induced dots in GaAs/AlGaAs QW with lateral confinement of up to 100 meV. The islanding behavior of InP grown on AlGaAs with $x = 0.3$–0.35 was found to be qualitatively similar to that of InP deposited on GaAs and $Ga_{0.51}In_{0.49}P$. Under particular growth conditions, a fairly uniform size distribution was obtained with an average diameter of 140 ± 12.2 nm and an average height of 19.5 ± 1.1 nm. Strain-induced quantum dots produced in the underlying QW with these stressor islands had intense and narrow luminescence peaks. This QD fabrication technique is applicable to other QW systems and also offers the possibility of obtaining smaller QD sizes with large strain confinement by using smaller SK islands.

Acknowledgements

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