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Red luminescence from strain-induced GaInP quantum dots

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The strain of self-organized InP islands is used to induced quantum dots in near-surface GaInP/AlGaInP quantum wells. To obtain quantum dot luminescence in a widely tunable wavelength range of 630–700 nm, the composition and thickness of the GaInP quantum well is varied. The effect of different cap layer materials, i.e., GaAs, AlGaAs, GaInP, and AlGaInP on the InP island formation and quantum dot luminescence properties is investigated. The luminescence intensity ratio of the quantum dot peak to the quantum well peak is found to be highest when a GaAs cap is used.

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Recently, visible luminescence has been reported from overgrown AlInAs (see Refs. 1 and 2) and InAs (see Ref. 2) quantum dots (QDs) in Al(Ga)As matrix. The luminescence peak wavelength of 645 nm was achieved in InAlAs/AlAs QDs.² In addition to the overgrown dots, QD structures with high optical quality can be fabricated by modulating the band gap of a near-surface quantum well (QW) by a stressor. The stressors can be either defined by lithographic means³ or formed by self-organized growth.⁴ Photoluminescence (PL) spectra of GaInAs QDs induced by self-organized InP stressors exhibited clearly resolved luminescence peaks from the QD ground state and up to four excited states.⁵ Calculated strain distribution, confinement potential, and energy levels in the structure corresponded well with the measured PL spectra.⁶ Also time-resolved PL⁷ and magnetoluminescence⁸ measurements were performed on the GaInAs QDs.

The formation of InP islands on GaAs and GaInP layers by coherent Stranski–Krastanow growth mode⁹ has been investigated in several reports.^{10–14} Highly uniform, coherently strained islands having no dislocations¹¹ were observed on both materials. The strain arising from the large lattice mismatch is accommodated by both the islands and the substrate.⁹ The calculations⁶ showed that the strain induced by an InP island on the underlying QW is tensile below the island and compressive under the edges of the island, thus creating a cylindrical, nearly parabolic confinement potential in the QW plane. In this letter the use of self-organized InP islands to create strain-induced QDs into GaInP/AlGaInP QWs is reported. It is demonstrated that the QD luminescence wavelength can be tuned in a range of at least 630–700 nm by varying the QW composition and thickness.

The samples were grown in a horizontal metalorganic vapor phase epitaxy reactor at atmospheric pressure on epitaxially semi-insulating $\pm 0.5^\circ$ (100) GaAs substrates. The source materials were trimethylaluminum, trimethylgallium, trimethylindium, tertiarybutylarsine, and tertiarybutylphosphine. A 100-nm-thick GaAs buffer layer, a 150-nm-thick $(\text{Al}_{0.40}\text{Ga}_{0.60})_{0.52}\text{In}_{0.48}\text{P}$ lower barrier layer, a $\text{Ga}_{1-x}\text{In}_x\text{P}$ QW and a 5-nm-thick $(\text{Al}_{0.60}\text{Ga}_{0.40})_{0.52}\text{In}_{0.48}\text{P}$ top barrier layer were grown at 680 °C with a V/III ratio of 25 for GaAs and

80 for the phosphides. Larger aluminum concentration was used in the upper barrier to confine carriers more efficiently into the near-surface QW. To investigate the effects of a thin lattice-matched cap layer, introduced between the top barrier layer and the islands,¹⁰ four samples having a GaAs, $\text{Al}_{0.30}\text{Ga}_{0.70}\text{As}$, $\text{Ga}_{0.51}\text{In}_{0.49}\text{P}$, and $(\text{Al}_{0.60}\text{Ga}_{0.40})_{0.52}\text{In}_{0.48}\text{P}$ cap were grown. The thickness of the cap layers was 6 ML, i.e., 1.7 nm. The arsenide and phosphide cap layers were grown at 650 and 680 °C, respectively. Depending on the cap layer material, nominally 1.5 or 2.4 ML of InP was deposited at 650 °C using TMIIn molar flow of 7.9 $\mu\text{mol/min}$ and a V/III ratio of 100. Atomic force microscopy (AFM) was used to measure the height and density of the InP islands. The optical properties of the samples were investigated by low-temperature PL measurements. The PL spectra were recorded by a photomultiplier tube using standard lock-in techniques. The 488.0 nm line of an argon ion laser was used for excitation and the laser beam was focused into a spot having a diameter of about 200 μm .

It has been reported that step bunching occurs in GaInP and AlGaInP layers at certain growth conditions,¹⁵ causing surface roughness and thickness variation in QWs. Therefore, the surface roughness of the QW structure grown at various temperatures was investigated by AFM. In the temperature range of 680–720 °C the lowest average step height of 2 nm was obtained at 680 °C. Next, the formation of InP islands on different cap layer materials was studied using a 9-nm-thick tensile $\text{Ga}_{0.57}\text{In}_{0.43}\text{P}$ QW in the structure. Figure 1 shows the AFM images of the samples with a (a) GaAs, (b) AlGaAs, (c) GaInP, and (d) AlGaInP cap. Three types of InP islands are observed in the images. The smallest islands, present in all the samples, are 1–6 nm high and can be best seen in Fig. 1(c). The strain field of these islands is too weak to produce QDs into the underlying QW. The second type consists of uniform islands with an average height of 15–29 nm depending on the cap material. The large islands constituting the third type have a nonuniform size distribution and are noncoherently strained because of dislocations.¹¹ On GaInP and GaAs surfaces the uniform islands are created at the threshold coverage and the large islands appear only after the coverage is further increased. However, on AlGaAs and AlGaInP both uniform and large islands are formed at the

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FIG. 1. Atomic force microscopic images taken from samples having a cap layer of (a) GaAs, (b) $\text{Al}_{0.30}\text{Ga}_{0.70}\text{As}$, (c) $\text{Ga}_{0.51}\text{In}_{0.49}\text{P}$, and (d) $(\text{Al}_{0.60}\text{Ga}_{0.40})_{0.52}\text{In}_{0.48}\text{P}$. The InP coverage is 2.4 ML on GaAs and AlGaAs and 1.5 ML on GaInP and AlGaInP. The scan size is $5 \times 5 \mu\text{m}^2$ in all the images.

threshold coverage. The optimum amount of InP to produce uniform islands on the GaAs cap was 2.4 ML. On thicker layers of GaAs the optimum amount has been found to be 3 ML.¹⁴ When this amount was deposited on a thin GaAs cap layer, large islands were also formed. The optimum coverage on the GaInP cap was 1.5 ML, which agrees with the results of island formation on thicker GaInP layers.¹¹ The same InP coverage as on GaAs and GaInP was used on AlGaAs and AlGaInP caps, respectively. The apparently different lateral dimensions of the uniform islands in Fig. 1 may originate from the variation of the AFM tip shape.

Table I shows the average height (h) and the areal density (ρ) of the uniform islands on each cap material. On GaAs, AlGaAs, and GaInP the island height is almost the same, 25–29 nm, whereas on AlGaInP it is 15 nm. The height of 27 nm on a thin GaAs cap is somewhat larger than 24 nm measured on a thicker GaAs layer at 650 °C.¹⁴ However, the height of the uniform InP islands has been observed to depend on the growth temperature.¹⁴ In this work InP was deposited just after the temperature was lowered from 680 to 650 °C. Therefore, the surface temperature of the substrate may be higher, resulting in a larger island height. When the InP coverage of 3 ML was used, the same areal density of

TABLE I. Average height (h) and areal density (ρ) of the uniform InP islands on different cap materials calculated from AFM images. Also shown is the redshift of the QD peak from the QW peak (ΔE), the FWHM of the QD peak, and the intensity ratio of the QD peak to the QW peak (R) obtained from low-temperature PL measurements.

Cap material	h (nm)	ρ (cm^{-2})	ΔE (meV)	FWHM (meV)	R
GaAs	27	5×10^8	59	22	1.5
AlGaAs	25	3×10^8	54	23	0.17
GaInP	29	4×10^8	62	25	0.04
AlGaInP	15	5×10^8	48	23	0.1

FIG. 2. Photoluminescence spectra of the samples having a $\text{Ga}_x\text{In}_{1-x}\text{P}$ QW with composition and thickness of (a) $x=0.58$ and 5 nm, (b) $x=0.55$ and 9 nm, and (c) $x=0.43$ and 8 nm, respectively. The inset shows the schematic structure of the sample with a GaAs cap layer (not in scale).

the uniform islands ($1 \times 10^9 \text{cm}^{-2}$) as observed on thicker GaAs was obtained.¹⁴ The density of the large islands on AlGaAs and AlGaInP is 1×10^6 and $4 \times 10^7 \text{cm}^{-2}$, respectively. The height of the large islands in the Figs. 1(b) and 1(d) are 180 and 40–110 nm, respectively.

Photoluminescence spectra of the samples with different cap materials (not shown here) exhibit three peaks in the visible part of the spectrum. At 580 nm is a peak originating from the lower barrier material $(\text{Al}_{0.40}\text{Ga}_{0.60})_{0.52}\text{In}_{0.48}\text{P}$. The transition of the carriers confined in the 9-nm-thick $\text{Ga}_{0.57}\text{In}_{0.43}\text{P}$ QW and in the QDs are at about 640 and 660 nm, respectively. The 660 nm peak was not observed in a reference sample grown without the InP islands. Table I shows results determined from the PL spectra. The redshift (ΔE) of the QD peak from the QW peak increases with the InP island height, suggesting that the higher islands produce larger strain under the islands. The full width at half maximum (FWHM) of the QD peak is typically 1 meV larger than the FWHM of the QW peak (average 22 meV). This indicates that the main factor determining the QD peak width is the QW peak width as was observed also in GaInAs QD samples.⁵ A minimum peak width of 46 meV at 660 nm has been reported for the overgrown $\text{In}(\text{Al})\text{As}/\text{Al}(\text{Ga})\text{As}$ QDs.² The luminescence intensity ratio of the QD peak to the QW peak (R) is highest when the cap material is GaAs. However, the absolute luminescence intensity is highest when InP is deposited directly on the AlGaInP surface. The suppression of the PL intensity in the sample with a GaAs cap may result from a disadvantageous Fermi level pinning on the surface or from nonradiative recombination at the interface of AlGaInP and GaAs.

Figure 2 shows three low-temperature PL spectra from the samples with a GaAs cap and various QW compositions and thicknesses. The QD luminescence wavelength was tuned in the range of 630–700 nm using both tensile and compressive QWs. The schematic structure of the sample is

FIG. 3. Photoluminescence spectra from a sample with a 9-nm-thick $\text{Ga}_{0.45}\text{In}_{0.55}\text{P}$ QW and a GaAs cap layer measured at the excitation intensity of (a) 1 mW, (b) 10 mW, (c) 25 mW, and (d) 100 mW.

shown in the inset of Fig. 2. The spectra in Fig. 2 show also that the peak intensity ratio depends on the QW thickness. The QD peak is more intense in the samples with a thicker QW.

Figure 3 shows PL spectra from a sample having a 9-nm-thick compressive $\text{Ga}_{0.45}\text{In}_{0.55}\text{P}$ QW and a GaAs cap measured with different excitation intensities. With increasing excitation the high-energy side of the QD peak rises and the intensity of the QD peak with respect to the QW peak is suppressed. This behavior is attributed to state filling and consequent luminescence from the excited states in the dots. Excited-state-luminescence peaks cannot be resolved because of the large peak width. Thickness variation due to step bunching and long-range ordering of GaInP in the GaInP/AlGaInP QW can cause spatial variation in the radiative transition energy of the QW.¹⁶ This results in a large QW peak width and also in a slight blueshift (5 meV in Fig. 3) of the QW peak with increasing excitation.

In summary, the strain of self-organized InP islands is used to induce QDs in GaInP/AlGaInP QWs by locally modifying the QW band gap. The QD luminescence wavelength is tuned in the range of 630–700 nm. High intensity ratio of the QD peak to the QW peak is obtained by using a GaAs cap layer between the top AlGaInP barrier and the InP islands. The width of the QD PL peak is mainly determined by the QW peak width. Luminescence in the high-energy side of the QD peak with increasing excitation is associated with state filling in the QDs.

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