Nanotechnology **19** (2008) 435710 (5pp) doi:10.1088/0957-4484/19/43/435710

Improved photoluminescence efficiency of patterned quantum dots incorporating a dots-in-the-well structure

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Received 23 May 2008, in final form 28 July 2008
Published 22 September 2008
Online at stacks.iop.org/Nano/19/435710

Abstract
InAs quantum dots embedded in InGaAs quantum well (DWELL: dots-in-the-well) structures grown on nanopatterned GaAs pyramids and planar GaAs(001) surface are comparatively investigated. Photoluminescence (PL) measurements demonstrate that the DWELL structure grown on the GaAs pyramids exhibits a broad quantum well PL band (full width at half-maximum ~ 90 meV) and a higher quantum dot emission efficiency than the DWELL structure grown on the planar GaAs(001) substrate. These properties are attributed to the InGaAs quantum well with distributed thickness profile on the faceted GaAs pyramids, which introduces a tapered energy band structure and enhances carrier capture into the quantum dots.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

InAs quantum dots (QDs) have attracted considerable attention over the past decades due to their unique atom-like discrete density of states and device applications [1, 2]. It has been shown that the structure with self-assembled InAs QDs (SAQDs) sandwiched between InGaAs quantum well (QW) layers grown on GaAs substrate, the so-called dots-in-the-well (DWELL) structure, has many advantages over a structure with the InAs QDs directly grown on GaAs substrate [3, 4]. The InGaAs QW surrounding the QDs reduces the compressive strain in the QDs as well as suppresses the In/Ga intermixing resulting in better crystalline quality [5, 6]. In the DWELL structure, the QD emission efficiency is dramatically higher because of the increased carrier capture rate into QDs for radiative recombination [7–9]. Therefore, the performance of lasers and detectors based on the DWELL structure is significantly improved [10–14]. In this paper, we demonstrate an approach to further improve the emission efficiency of QDs by growing the InAs/InGaAs DWELL structure on nanopatterned GaAs substrate. Photoluminescence (PL) measurements show that this DWELL structure is more efficient on carrier capture due to the tapered energy band structure formed by the distributed inhomogeneous thickness of the InGaAs QW grown on faceted GaAs pyramids.

2. Experiment

The sample growth was carried out using a low-pressure (60 Torr) vertical Thomas-Swan metal-organic chemical-vapor deposition (MOCVD) reactor with trimethylgallium, trimethylindium, and tertiarybutylarsine. For the patterned QD (PQD) sample, the InAs/InGaAs DWELL structure was grown on a semi-insulating GaAs(001) substrate with patterned SiO_2 mask (25 nm thick). This patterning process with interferometric lithography and dry etching results in a two-dimensional array of circular nano-holes of 225 nm ($\pm$10 nm) in diameter with a pitch of 330 nm [15]. Figures 1(a) and (b) show the schematic and the plan-view scanning electron

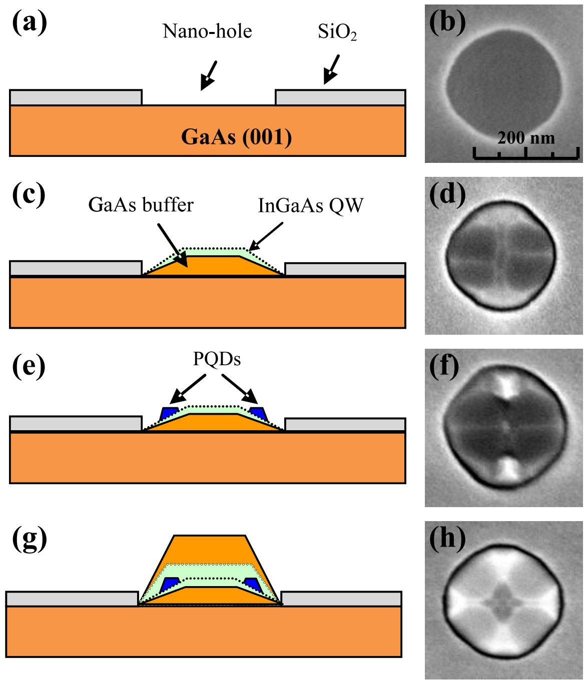

Figure 1. Schematics of sample cross-section profiles and the corresponding plan-view scanning electron microscopy images to show the growth process and structural features for the DWELL structure grown on the patterned substrate [19].

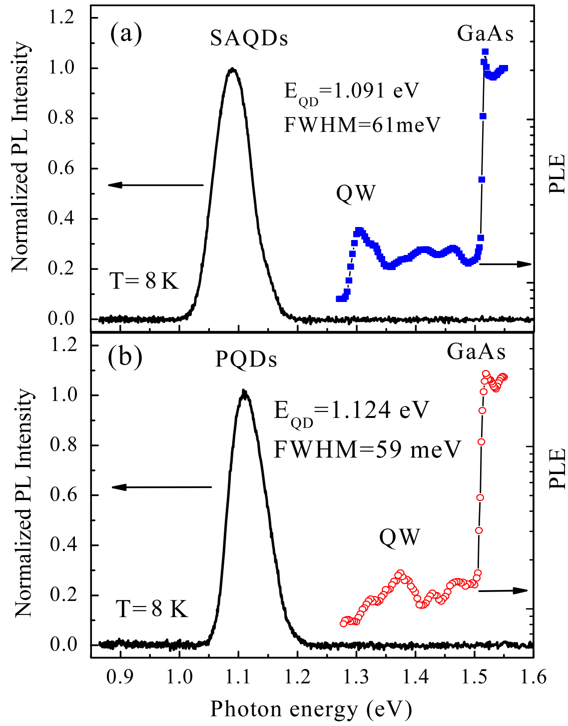

Figure 2. Normalized low temperature ($T = 8$ K) PL spectra and PLE spectra for both the PQD and SAQD samples.

microscopy images of one nano-hole, respectively. After the growth of 1 nm GaAs buffer at 700 °C, the reactor temperature is reduced to 510 °C and 2 nm of $In_{0.15}Ga_{0.85}As$ is subsequently deposited. As shown in figures 1(c) and (d), due to the special growth mode on patterned substrate [16–18], GaAs pyramids are formed in the nano-holes with a unique set of facets, resulting in distributed thickness profiles for both the GaAs buffer and the InGaAs layer. The growth continues at 510 °C for the deposition of 2 ML of InAs to obtain PQDs nucleated on selective facets [19], as shown in figures 1(e) and (f). After that, these InAs PQDs are capped with 3 nm (planar thickness) of $In_{0.15}Ga_{0.85}As$ and 3 nm (planar thickness) of GaAs also at 510 °C to complete the DWELL structure. As shown in figures 1(g) and (h), the QW in this PQD DWELL structure has reduced thickness near the edge of the nano-hole.

As a comparison, one SAQD DWELL sample was grown on a planar GaAs(001) substrate under similar growth conditions. InAs QDs are formed under the Stranski–Krastanow growth mode as a result of strain relaxation after the deposition of 2.5 ML of InAs atop an $In_{0.15}Ga_{0.85}As$ layer of 2.8 nm thickness. These SAQDs are then capped with 4.2 nm of $In_{0.15}Ga_{0.85}As$ and 80 nm of GaAs. This QW structure is designed in such a way that the total QW thickness of the SAQD DWELL sample is close to the thickness of QW at the center part of the PQD DWELL structure. Due to the planar growth mode, this sample has abrupt GaAs/$In_{0.15}Ga_{0.85}As$ /GaAs interfaces and the $In_{0.15}Ga_{0.85}As$ QW has a uniform thickness. Because of the difference in growth mode, the PQD sample has a QD density of 2×10^9 cm^{-2}, compared to the SAQD sample's QD density of 5×10^{10} cm^{-2}.

3. Results and discussion

Continuous-wave PL measurements were performed with a frequency-doubled ($\lambda = 532$ nm) Nd-YAG laser as the excitation source. The PL emission was dispersed by a 0.5 m spectrometer and collected using a liquid-nitrogen-cooled InGaAs detector array. The PL spectra for the PQD and SAQD samples were first measured at low temperature (8 K) with a low excitation intensity of 0.1 W cm^{-2}. We estimate this pump intensity produces, on average, less than one carrier per QD in the PQD sample during the carriers' lifetime (∼1 ns). For both samples only the emission from the QD ground state is observed as shown in figure 2. The PQD sample has a PL peak at 1.124 eV with a linewidth (full width at half-maximum, FWHM) of 59 meV while the SAQD sample has a PL peak at 1.091 eV with a FWHM of 61 meV. Emission from the QW and GaAs barrier is absent in both PL spectra at this low excitation intensity. With the $\lambda = 532$ nm light excitation, the photo-excited carriers are mainly generated in the GaAs matrix, which serves as the carrier reservoir. The absence of GaAs and QW PL signal indicates that the carrier transfer from the GaAs matrix through the InGaAs QW, into the InAs QDs is faster compared to the lifetime of carriers in the QDs.

To prove the existence of the QW, PL excitation (PLE) spectra were measured at 8 K using a continuous-wave Ti:Sapphire laser with a constant excitation intensity of 0.3 W cm^{-2} that is also estimated to provide less than one photo-excited carrier per QD on average. As shown in figure 2(a), the PLE spectrum of the SAQD sample shows a pronounced excitation resonance at ∼1.308 eV, which is close to the PL location (∼1.331 eV) of the exciton recombination in the $In_{0.15}Ga_{0.85}As$ QW of 7 nm thickness, calculated from the eight-band $k \cdot p$ model [20]. Therefore, it is attributed to the ground state optical absorption in the InGaAs QW. The resonance enhancement at the excitation energy of 1.308 eV reveals a strong carrier transfer from QW to SAQDs. Compared to the SAQD sample, PLE spectrum of the PQD sample shows a broad excitation resonance band from 1.300 to 1.410 eV, indicating an inhomogeneous QW characteristic that is attributed to the distributed thickness of the InGaAs QW in the PQD sample.

To further investigate the optical properties of these two samples, PL spectra are measured under elevated excitation intensity (0.01–10^3 W cm^{-2}) at 8 K. For both samples, at low excitation intensities from 0.01 to 50 W cm^{-2}, the QW PL band is absent whereas the PL from the QD ground state is the only feature. As the excitation intensity increases, at first the QD excited states and then the prominent InGaAs QW peak appears. In order to better illustrate the spectral contents, line-shape analysis of the PL spectrum obtained with an excitation intensity of 500 W cm^{-2} is presented in figures 3(a) and (b). For both samples, the spectrum is well described as a convolution of four Gaussian peaks, which are the ground state (E0), the first excited state (E1), and the second excited state (E2) transitions of the QD ensemble, together with the peak from the QW. The line-shape analysis of the PL spectra also confirms that the PQD sample has a very broad QW PL signal (FWHM ~90 meV) compared to the SAQD sample (FWHM ~35 meV).

From the excitation-intensity-dependent PL spectra, the integrated PL intensities are extracted and plotted in figure 3(c). The PQD sample and SAQD sample have different slopes in the low excitation regime. The relationship between the integrated PL intensity of the ground state QD PL emission and the excitation intensity can be described by:

$$I_{\mathrm{PL}} = \eta P^{\alpha},$$

where I_{PL} is the integrated PL intensity and P the excitation intensity, η and α fitting coefficients [21]. By fitting the experimental data, the coefficient α approximately equal to unity in both case, indicating that the main contributing mechanism is the exciton recombination and the carrier losses due to nonradiative recombination are small for both samples [22]. In particular, η is closely related to the PL efficiency. We obtain $\eta = 0.049$ for the PQD sample and $\eta = 0.019$ for the SAQD sample. The larger η value of the PQD sample indicates that this sample has better PL emission efficiency than the SAQD sample in the low excitation regime [21]. Furthermore, as shown by the integrated PL intensity ratio in figure 3(c), the integrated PL intensity of the SAQD sample is about 2–12 times higher than that of the PQD sample when the excitation intensity increases from 0.1 to 100 W cm^{-2}. Considering the 25 times higher QD density of the SAQD sample, we estimate the PL efficiency of the PQD sample to be about 2–12 times higher than the SAQD sample. The higher PL efficiency of the PQD sample may be attributed to several sources, including the different efficiency on excitation photon absorption, photo-excited carrier capture,

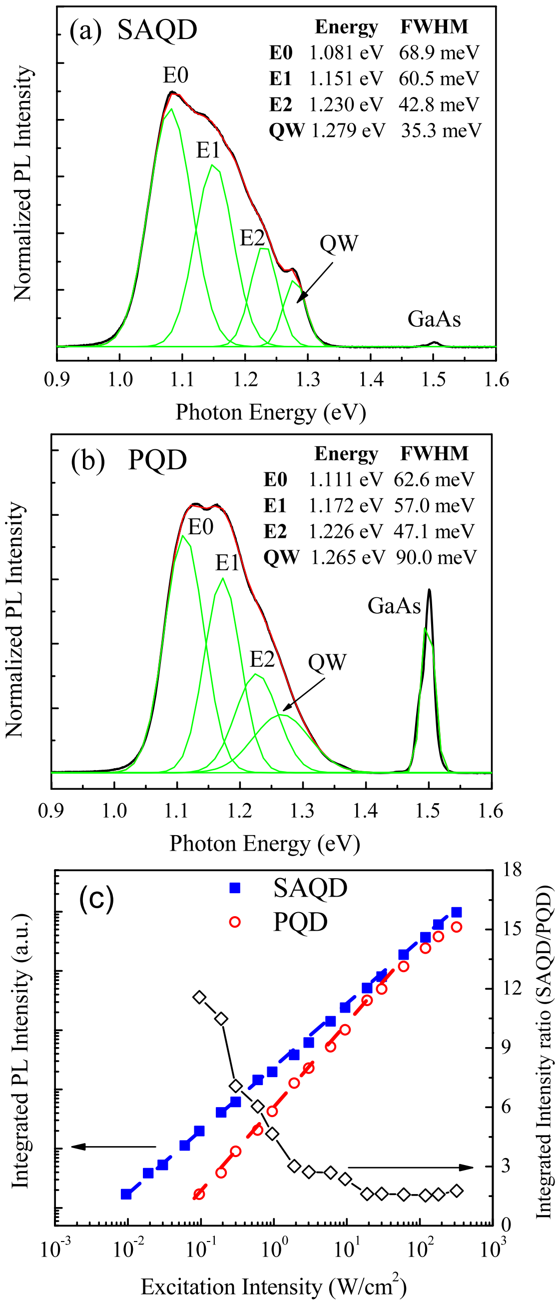

Figure 3. (a) and (b) are the line-shape analysis of the normalized PL spectrum obtained with an excitation intensity of 500 W cm^{-2} for the SAQD and PQD sample, respectively. (c) shows the integrated PL intensities and the intensity ratio (SAQD sample / PQD sample) as a function of the laser excitation intensity.

and PL photon extraction. To verify the main contribution to the improved PL efficiency, the QD PL intensities of the two samples are compared while both of them are resonantly excited at the QW peak wavelength under the same laser pump power. In this case, the photo-excited carriers are most generated inside the InGaAs QW and then transfer into the QDs. After excluding the QD density effect, the PQD sample has stronger PL signal than the SAQD sample, and it is hence concluded that the PQD DWELL structure is more efficient on carrier capture.

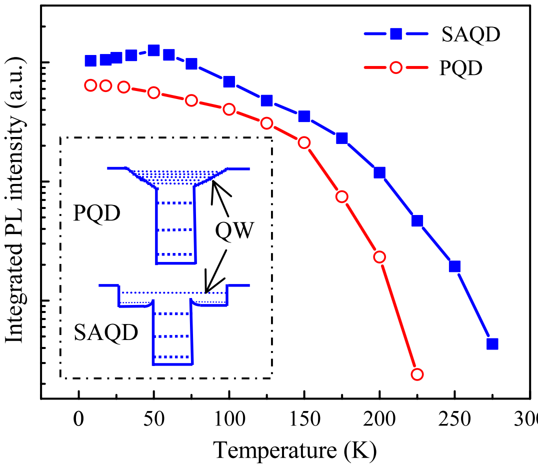

Figure 4. Integrated PL intensities from both the SAQD and PQD samples as a function of temperature. The inset shows the schematic of the conduction band structures for both samples.

Finally, for the temperature-dependent characteristics of the PL, the excitation laser intensity is set to 3 W cm^{-2} in the low excitation regime to ensure that only the QD ground state is predominantly excited. In figure 4, the integrated PL intensity is plotted as a function of temperature, which reveals two characteristic regions for the PQD sample. From 8 to 150 K the integrated PL intensity decreases slowly with temperature, demonstrating the strong three-dimensional confinement and larger exciton binding energy (∼12 meV) of the PQDs. Above 150 K, the integrated PL intensity decreases rapidly with increasing temperature, indicating accelerated carrier thermal escape from PQDs [23]. The SAQD sample shows different temperature-dependent PL behavior. From 8 to 50 K the integrated PL intensity increases slowly with temperature, and above 50 K it decreases with increasing temperature.

The increase of integrated PL intensity for the SAQD sample in the range of 8–50 K can be explained by the strain field between InAs QDs and InGaAs QW, which creates a potential barrier at the interface [24, 25] as shown by the schematics of the conduction band structure in the inset of figure 4. Due to this potential barrier, some carriers are localized in the trap centers in the QW at the low temperature and obtain thermal energy to be captured into InAs SAQDs as the temperature increases. On the other hand, in the PQD sample the QW with distributed thickness profile induces a tapered conduction band structure, which reduces the localization of photo-excited carriers in the QW and subsequently enhances the carrier capture into PQDs [14]. This helps to explain the higher PL efficiency of the PQD sample compared to the SAQD sample. The PQD DWELL structure is also beneficial to device applications based on a single QD, such as single QD sensor, detector, and single photon emitter. In those cases, the PQD DWELL structure provides not only high emission efficiency, but also the capability to engineer and individually address single QD.

4. Conclusions

In conclusion, we systematically investigate the InAs/InGaAs DWELL structures grown on patterned GaAs pyramids and planar GaAs(001) substrate. The PLE spectra reveal that the PQD DWELL structure has a broad QW resonance peak. Excitation intensity dependent PL measurements also show a broad QW PL band (FWHM ∼90 meV) and better PL emission efficiency for the PQD sample. These properties are attributed to the InGaAs QW structure with distributed inhomogeneous thickness due to the special growth mode on the faceted GaAs pyramid, which helps to increase the carrier capture into QDs. This research demonstrates that growth on the patterned substrate is a useful approach to further increase the carrier capture efficiency into the QDs with the InAs/InGaAs DWELL structure.

Acknowledgments

The authors gratefully acknowledge the financial support of this work by NSF of IGERT (through Grant DGE-0504276) and DARPA (through Grant HR 0011-05-1-0006, and W911NF-07-1-0323).

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