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Enhanced optical properties of InAs/InAlGaAs/InP quantum dots grown by metal-organic chemical vapor deposition using a double-cap technique

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ABSTRACT

The effects of a double-cap procedure on the optical properties of an InAs/InAlGaAs quantum dots (QDs) system grown by metal-organic chemical vapor deposition (MOCVD) have been investigated by atomic force microscopy (AFM) and room temperature photoluminescence (RT-PL) spectroscopy. An optimized QD growth condition has been achieved, with an areal density of 4.6×10^{10} cm^{-2}. It was found that the thickness and lattice constant of the high temperature second cap layer (SCL) were crucial for improving the integrated PL intensity and line-width of the 1.55 µm emission from the InAs/InAlGaAs QD system grown on a semi-insulating InP (100) substrate. With fine-tuned SCL thickness and lattice constant, the optical performance of the five-stack QDs was enhanced. The improvements can be attributed to the smooth growth front, observed from the AFM images, and the well-balanced stress engineering.

1. Introduction

Self-assembled InAs quantum dots (QDs) by Stranski–Krastanow (SK) growth mode have been drawing intense attention due to their unique properties and great potential in photonic device applications, especially QD semiconductor lasers [1,2]. Quantum dot lasers offer advantages, including low threshold current density with less temperature sensitivity and higher differential gain, over their conventional quantum well laser counterparts [3]. So far, great efforts have been devoted to the InAs/GaAs system [4,5], operating in the 1.3 µm region. InP-based QDs have been playing a dominant role in the 1.55 µm long wavelength laser application, driven by optical fiber communications [6]. Research on the InAs/InP system is limited, due to a lower lattice mismatch (3.2% for InAs/InP and 7.2% for InAs/GaAs) and challenging chemical reaction at the growth interface during the As/P exchange [7].

Currently, InGaAsP and InAlGaAs are two mainstream matrixes in a quantum dot laser structure [8]. For the InAs/InGaAsP material system, the As/P exchange reaction occurring on top of the QDs [9], while for InAs in the InAlGaAs matrix, phase separation are issues in the QDs growth [10]. Compared with the InGaAsP quaternary alloy, InAlGaAs provides a larger conduction band-offset, leading to enhanced differential gain and better temperature characteristics [11]. To date, studies regarding InAs/InAlGaAs have mainly been dominated by molecular beam epitaxy (MBE) [12,13]. In contrast, InAs QDs with InAlGaAs as a cap layer grown by metal-organic chemical vapor deposition (MOCVD) have not yet been incorporated and reported in high performance, long wavelength quantum dot lasers [14]. Nevertheless, MOCVD remains an attractive growth choice for photonic devices in its ability of growing diverse compound semiconductors and scalability to high volume production.

To optimize the optical properties of InAs QDs embedded in an InAlGaAs matrix, a uniform dot size distribution is required, since QDs with large size inhomogeneity will inevitably result in a broad PL line-width (typically about 120 meV at room temperature) [15]. In this work, an InAlGaAs double-cap procedure has been shown to dramatically decrease dot height dispersion, reduce phase separation and provide more flexibility in tuning the emission wavelength of InAs QDs to 1.55 µm. A thin low temperature InAlGaAs first cap layer (FCL) was deposited with a thickness smaller than the average quantum dot height, followed by a growth interruption and temperature ramp-up for QDs annealing. During the annealing, the unprotected top parts with excess height are flushed away and a more uniform QD height thus can be achieved. Finally, a high-temperature second cap layer (SCL) was deposited as a separation layer between each quantum dot layer in the stacked InAs/InAlGaAs structure.

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In this paper, the effects of the double cap layer parameters on the optical properties of multi-stack InAs/InAlGaAs QDs grown on a semi-insulating (SI) InP (001) substrate by MOCVD were investigated by room temperature photoluminescence (RT-PL) spectroscopy and atomic force microscopy (AFM). By increasing the InAlGaAs SCL thickness from 20 to 50 nm, the PL peak intensity increased significantly by more than three times. This is attributed to the smaller kinetic roughness before the subsequent QD layer growth, as confirmed by the AFM images. Moreover, with slight decrease of the lattice constant of the InAlGaAs SCL to make it smaller than that of the InP substrate, PL peak intensity enhancement related to the stress compensation in the structure was also observed.

2. Experimental details

All samples in the study were grown on semi-insulating InP (001) substrates in an Aixtron AIX-200/4 horizontal MOCVD system equipped with a Laytec EpiRAS for in-situ growth spectral reflectance measurement. Trimethylindium (TMIn), trimethylaluminum (TMAl), and triethylgallium (TEGa) were used as the group III precursors, and tertiarybutylarsine (TBA) and phosphine (PH_3) were used as the group V precursors. The reactor pressure during the growth was maintained at 100 mbar. Initially, a 60 nm InP buffer was deposited at 600 °C on the InP substrate, followed by five stacks of InAs QDs/InAlGaAs double cap layers (DCLs). A 2 nm $In_{0.4}Ga_{0.6}As$ wetting layer was grown at 600 °C prior to the InAs QD growth; the substrate was then cooled down to 500 °C in the TBA ambient to prevent surface decomposition. Afterwards, a 3.6 monolayer (ML) of InAs was deposited, with a growth rate of 0.12 nm/s and a V/III mole ratio of 0.4. A growth interruption of 5 s was introduced subsequently with TBA flux to allow the Indium adatoms to migrate on the surface and form the dots. The quantum dots were then capped by an InAlGaAs double-cap procedure. More specifically, the first $In_{0.8-x}Al_{0.2}Ga_xAs$ cap layer was deposited at the same growth temperature as the QDs (500 °C), with a thickness of 1.2 nm to keep the QD emission wavelength in the 1.55 μm region. After that, the sample was heated up to a higher temperature of 600 °C in five minutes, with a flow of TBA to desorb the uncapped QD parts and flatten the growth front before the SCL deposition. Finally, a high temperature $In_{0.71-x}Al_{0.29}Ga_xAs$ was grown. The thickness of the SCL is to be optimized. The schematic diagram of the complete structure is shown in Fig. 1.

3. Results and discussion

The growth conditions for the InGaAs wetting layer and InAs QDs were optimized and examined by AFM. Fig. 2 shows a typical 1 μm × 1 μm AFM image of the QD surface, and the QD density is about $4.6 \times 10^{10}\ cm^{-2}$, with a 3.6 ML InAs supply. The average diameter and height of the closely packed QDs are estimated to be 42 and 3.8 nm, respectively. The majority of QDs are preferentially orientated along the $[1\ \bar{1}\ 0]$ direction, caused by greater adatom surface diffusion along the $[1\ \ 1^{-}\ \ 0]$ direction for As-stabilized surfaces [16]. Very few QDs are elongated into Qdash-like geometries, due to the smooth growth front of the strained $In_{0.4}Ga_{0.6}As$ wetting layer [15].

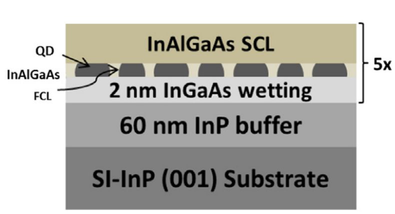

Fig. 1. Schematic diagram of five-stack InAs QDs embedded in an InAlGaAs double-cap matrix.

The effect of the SCL thickness on the optical properties of stacked InAs/InAlGaAs QDs was first investigated by RT-PL, with excitation by a 300 mW all-solid-state 671 nm red laser. Valuable information about the optical properties can be extracted from the PL spectrum: the PL intensity reflects the optical quality, and the line-width indicates the size and shape distribution of the QDs [17]. The central wavelength (peak energy) is sensitive to the effective QD heights, determined by the thickness of the FCL. The FCLs of the four samples included in this study were fixed at 1.2 nm $In_{0.52}Al_{0.2}Ga_{0.28}As$, and the elemental compositions of the SCLs were kept as $In_{0.51}Al_{0.29}Ga_{0.2}As$, while the thicknesses of the SCLs varied from 20 to 50 nm. The composition of Indium in the InAlGaAs alloy was determined by X-ray diffraction (XRD) fitting, and the Aluminum composition was calculated based on RT-PL [11]. Fig. 3 shows the room temperature PL spectra of the four samples. It is noted that the PL intensity increased sharply when increasing SCL thickness from 30 to 40 nm, with the central wavelengths close to 1.6 μm. In the inset of Fig. 3, the normalized integrated PL intensity and full-width at half-maximum (FWHM) dependences on the SCL thickness are presented. For the five-stack InAs/InAlGaAs QDs, 50 nm InAlGaAs separation provides the best optical performance, with the highest integrated PL intensity (3.3 times over the 20 nm cap layer), as well as the lowest FWHM value (71 meV). This can be attributed to two factors: first, due to the thicker InAlGaAs cap layer, more carriers are generated, enhancing the absorption of external excitation. Moreover, a thicker high-

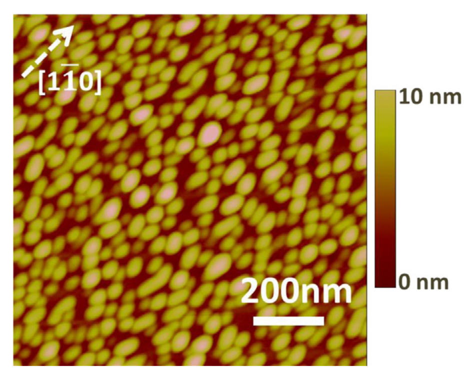

Fig. 2. Typical $1 \times 1\ \mu m^2$ AFM image of 3.6 ML InAs QDs grown on 2 nm $In_{0.4}Ga_{0.6}As$ wetting layer.

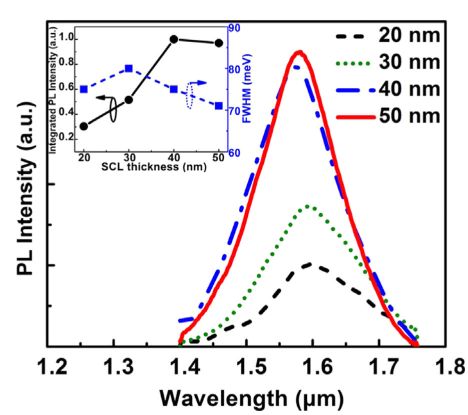

Fig. 3. RT-PL spectra of five stacks of InAs QDs with different SCL thicknesses. The inset implies normalized integrated PL intensity and FWHM as functions of the SCL thickness.

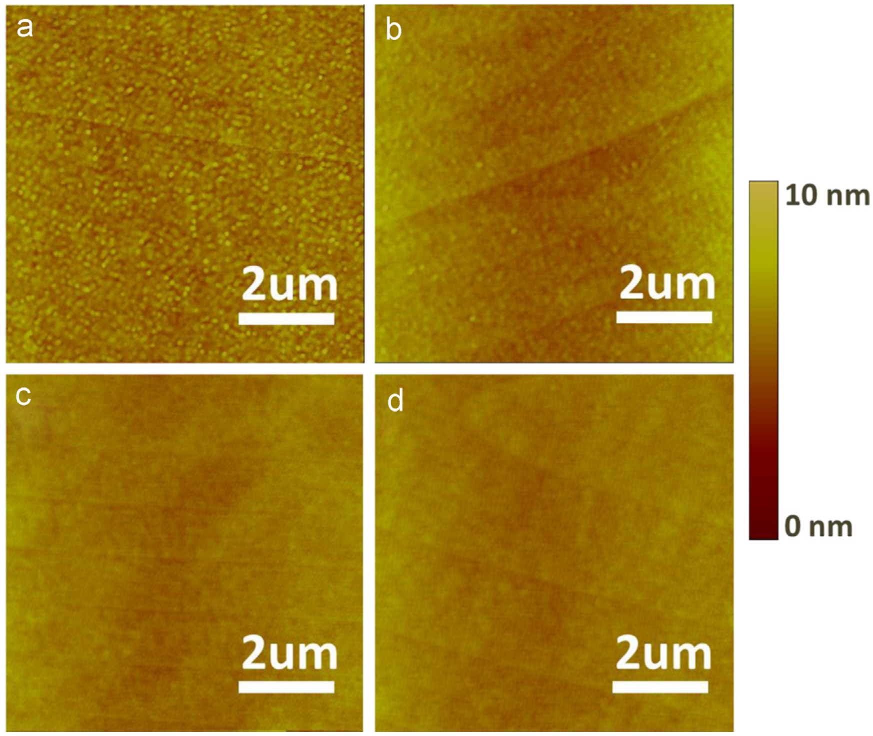

Fig. 4. $10 \times 10\ \mu m^2$ AFM images of five-stack InAs/InAlGaAs with the SCL thickness of: (a) 20 nm, (b) 30 nm, (c) 40 nm, and (d) 50 nm. The RMS roughness values are 0.38 nm, 0.44 nm, 0.28 nm and 0.25 nm.

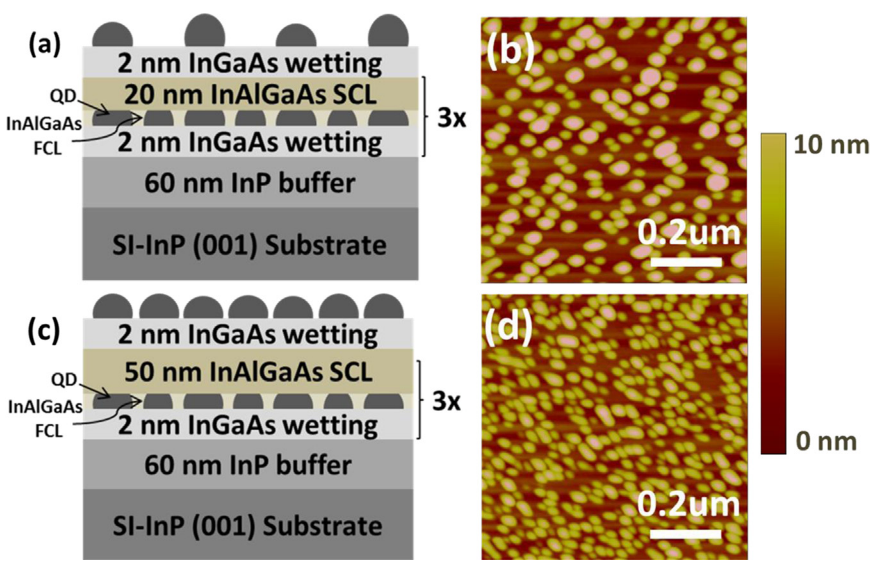

Fig. 5. Schematic diagrams and AFM images of a single InAs QDs layer grown on the topmost of three-stack InAs/InAlGaAs structures with a spacer thickness of (a and b) 20 nm and (c and d) 50 nm.

temperature InAlGaAs cap layer results in a smoother growth front and better surface morphology, as seen from the surface morphology evolution of the four samples in Fig. 4. The surface morphologies of these samples are quite different: for the 20 nm InAlGaAs separation, large amounts of small clusters could be observed on the surface, which were considered a reflection of defects beneath [18]. However, for the 50 nm separation sample, the surface was quite smooth, without any small clusters. Furthermore, the root-mean-square (RMS) roughness of the 50 nm cap sample (0.25 nm, as shown in Fig. 4(d)) was smaller than that of the 20 nm cap sample (0.38 nm, as depicted in Fig. 4(a)). This result proves a smaller kinetic roughness after the SCL was grown thicker. Meanwhile, the integrated PL intensity saturated when the SCL thickness was further increased from 40 to 50 nm.

To further investigate the influence of these small clusters on the evolution process of stacked QDs, two incomplete InAs/InAlGaAs structures were grown with different cap layer thicknesses (20 nm and 50 nm). Both of these samples were deposited with three stacks of InAs/InAlGaAs first, and then a single layer of InAs QDs was grown in-situ on the top for AFM comparison. The uncapped structures are described in Fig. 5(a) and (c). As shown in the AFM images (Fig. 5(b) and (d)), the surface density of InAs QDs is enhanced markedly from 2.0×10^{10} to 4.3×10^{10} cm^{-2} when the SCL thickness was increased from 20 to 50 nm. Furthermore, the lateral size and height of the QDs became more uniform with a

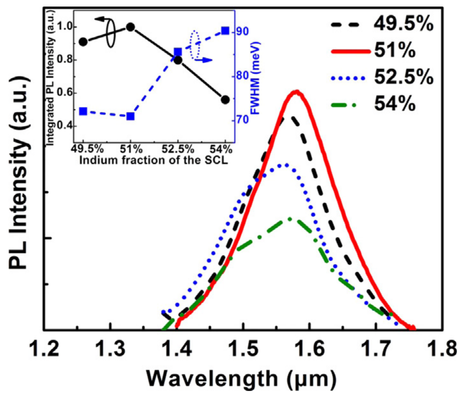

Fig. 6. PL spectra taken at room temperature of the five-stack InAs QDs with different Indium compositions in the InAlGaAs SCL. The inset shows the evolutions of normalized integrated PL intensity and line-width with Indium fraction of the SCL.

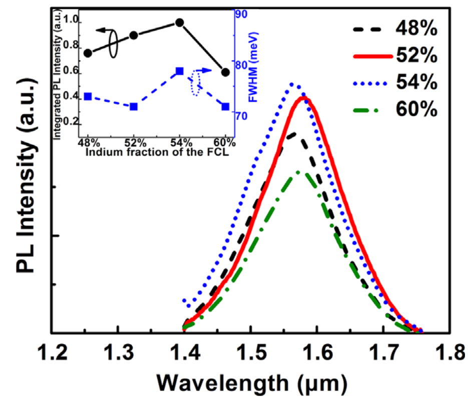

Fig. 7. RT-PL spectra of five-stack InAs QDs with different Indium compositions in low temperature FCL. The inset shows the normalized integrated PL intensity and line-width as functions of Indium fraction in the FCL.

50 nm cap layer. This is ascribed to the smoother surface morphology and smaller kinetic roughness as the thickness of InAlGaAs spacer layer increases, prior to the subsequent QD layer growth. For the 20 nm cap layer, the relatively rough capping surface with a higher density of small clusters formed more favorable nucleation sites to decrease the QDs density and increase the size and height of the InAs QDs in the next stack. Therefore, as the stack number increases, the QDs get higher and become more inclined to form coalesced islands (see Fig. 2 and Fig. 5(b)) with the 20 nm cap layer. It has also been observed that a smaller density of QDs will result in a degraded PL intensity and enlarged FWHM in the stacked InAs QDs [17].

Another important growth parameter that impacts the optical properties of QDs is the lattice constant of the $In_{0.71-x}Al_{0.29}Ga_xAs$ SCL. To examine this, four samples with different SCL lattice constant were compared. The FCLs were maintained at 1.2 nm $In_{0.52}Al_{0.2}Ga_{0.28}As$, and the SCLs thicknesses were fixed at the optimized 50 nm. Fig. 6 presents the PL spectra of QDs with different SCL lattice constants, controlled by varying the Indium fraction in the InAlGaAs alloy. From the inset of Fig. 6, it can be seen that with an $In_{0.51}Al_{0.29}Ga_{0.2}As$ separation SCL, the integrated PL intensity of five-stack InAs/InAlGaAs reaches the highest, along with the smallest line-width of 71 meV. The emission efficiency enhancement can be explained by strain balancing or strain compensation by the SCL [19,20]. A lower Indium composition (too smaller lattice constant) of the InAlGaAs SCL will over-balance the tensile strain generated from the InAs quantum dots underneath; thus the accumulated compressive stress along the sample will be more inclined to generate dislocations and other defects [21]. Similarly, for a certain thickness of the SCL, if the lattice constant of the InAlGaAs is larger than that of InP, namely, the Indium fraction is larger than 53%, it is more difficult to compensate the tensile strain in multi-stack QDs. The imposed strain leads to an obvious broad FWHM value and weak PL intensity, as depicted in the inset of Fig. 6. A 50 nm $In_{0.51}Al_{0.29}Ga_{0.2}As$ was found to be the optimum in preventing the accumulation of internal excessive stress, reaching an ideal strain balancing situation.

Finally, based on the optimized growth parameters of the high-temperature SCL, the influence of the low temperature FCL was further investigated. Fig. 7 demonstrates the PL spectra as a function of the FCL lattice constant, by tuning the Indium fraction of the FCL. Compared with the SCL, the impact of FCL lattice constant is relatively weak due to the much smaller thickness. However, the data still clearly indicates the role of stress engineering in the optical performance improvement. From the inset of Fig. 7, it can be seen that further increase of the Indium composition to 60% resulted in a sharp decrease in integrated PL intensity. Besides this, although the integrated PL intensity reaches its maximum when the FCL is nearly lattice-matched to the InP substrate ($In_{0.54}Al_{0.2}Ga_{0.26}As$), the FWHM is actually the largest (79 meV). This is due to the compromised well-balanced stress from the combined role of the double cap layers. Further reducing the lattice constant of the FCL to be slightly smaller than that of the InP substrate ($In_{0.52}Al_{0.2}Ga_{0.28}As$), the strain-compensation condition can be better satisfied [20], with the lowest line-width (71 meV), as well as a decent integrated intensity (90% of the strongest integrated PL intensity). Moreover, since the FCL in this procedure is thin enough (1.2 nm), the negative effects of phase separation and non-radiative defect centers are far more trivial than with a single layer of low temperature InAlGaAs cap in the MOCVD growth. This is also a remarkable advantage of adopting the double-cap procedure for stacked InAs/InAlGaAs growth by MOCVD.

4. Conclusions

In conclusion, the effects of InAlGaAs double cap layers on the optical performance of a stacked InAs/InAlGaAs structure on semi-insulating InP (001) substrates grown by MOCVD were thoroughly investigated. It was found that by increasing the SCL thickness from 20 to 50 nm, the integrated PL intensity increased by a factor of 3.3, which could be ascribed to a smoother growth front and better surface morphology before the subsequent QDs growth. In addition, the impact of the lattice constants of the double cap layers was also optimized, and the optical properties of multi-stack QDs were improved by strain compensation. Finally, the growth condition for the multi-stack InAs/InAlGaAs QDs system was successfully optimized, with an improved integrated PL intensity and a small line-width of 71 meV. The central wavelength was located at 1.58 μm at room temperature. The results demonstrated here pave the way to realizing high performance 1.55 μm InAs QD lasers based on the InAs/InAlGaAs/InP system.

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