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GaAs-based long-wavelength InAs quantum dots on multi-step-graded InGaAs metamorphic buffer grown by molecular beam epitaxy

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Abstract
Molecular beam epitaxy growth of GaAs-based long-wavelength metamorphic InAs/InGaAs quantum dots (QDs) is investigated. With optimized multi-step-graded InGaAs metamorphic buffer layers and growth conditions, room temperature 1.46 $\mu$m emission from InAs/In$_{0.15}$Ga$_{0.85}$As QDs is realized, and broad-area laser diodes are fabricated with a very low etch pit defect density of less than 5.0 $\times$ 10$^3$ cm$^{-2}$. The lasers operate under pulsed operation mode at room temperature with a low threshold current density of 146.7 A cm$^{-2}$.

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

1. Introduction

GaAs-based metamorphic InGaAs heterostructures for electronic devices such as high electron mobility transistors have been investigated extensively over the past few decades [1, 2]. In recent years, they have drawn strong research interest for optoelectronic device applications, such as GaAs-based semiconductor lasers operating in the 1.3–1.55 $\mu$m telecom wavelength range [3–8], photodetectors [9] and high-efficiency multi-junction solar cells [10–13]. How to restrain or diminish dislocations in the In$_x$Ga$_{1-x}$As metamorphic buffer layers (MBLs) caused by lattice mismatch between In$_y$Ga$_{1-y}$As and GaAs has been the main problem in growing a high-quality In$_x$Ga$_{1-x}$As layer on In$_y$Ga$_{1-y}$As MBLs to be used as a ‘virtual substrate’ for the succeeding epitaxial growth of active layers. To filter dislocations in the MBLs and to keep them away from active regions, the key issue is to have a good design and to optimize growth parameters of molecular beam epitaxy (MBE) or metal-organic chemical vapour deposition (MOCVD) [3].

The initial method for controlling the indium content $y$ of a metamorphic In$_x$Ga$_{1-x}$As layer is called the single-step-graded (SSG) approach. In this way the In$_y$Ga$_{1-y}$As MBL is first grown at a lower temperature keeping the indium composition $y$ constant and then annealed at a higher temperature to reduce the dislocation density [3, 7]. There were some reports about room temperature (RT) operation 1.4–1.5 $\mu$m InAs quantum dot (QD) lasers grown on SSG MBLs [7, 8, 14]. When the indium content of the SSG In$_y$Ga$_{1-y}$As MBL was increased to 0.25, a metamorphic InAs QD laser realized 1.55 $\mu$m emission but not lasing [15]. It was analysed that the threading dislocations in SSG In$_y$Ga$_{1-y}$As layer would penetrate down into the GaAs substrate if the indium content $y$ was lower than 0.18, but penetrate up into epitaxial layers if the indium content $y$ was higher than 0.28 [16, 17]. These discussions indicated that SSG MB is not the most suitable for higher indium content metamorphic InGaAs structures.

Two modified methods, linearly graded (LG) and multi-step-graded (MSG), are proposed to overcome the disadvantages of the SSG MBL. For the LG case, indium content $y$ of the In$_y$Ga$_{1-y}$As MBLs (0 $\leq$ $y$ $\leq$ $z$) is linearly increased. According to Tersoff’s theory [18], a setback of indium content, defined as $\Delta x = z - x$, should be introduced to eliminate the residual strain in the LG
In$_x$Ga$_{1-x}$As MBL surface, and then the indium content of the following In$_x$Ga$_{1-x}$As ‘virtual substrate’ layer $x$ can be higher than 0.28. RT lasing 1.31–1.55 $\mu$m InGaAs quantum wells (QWs) grown on LG In$_x$Ga$_{1-x}$As MBL structures were realized [5, 6, 19], but 1.55 $\mu$m long wavelength InAs QDs grown on the same LG MBL structures were not successful. The main difficulties are to accurately control the linearly changing and the setback of indium content of the LG MBLs. For the MSG case, the indium content of the In$_y$Ga$_{1-y}$As MBL is increased step by step. In the MSG structures, the final-step In$_y$Ga$_{1-y}$As layer, normally called the ‘overshooting’ layer, has a larger indium content than the following In$_y$Ga$_{1-y}$As layers to balance the possible strain. Based on MSG AlGaAs/Sb MBLs, RT pulsed lasing from the first excited state transition of InAs QDs at 1.3 $\mu$m with a threshold current density of 304 A cm$^{-2}$ has been demonstrated [20]. The MSG structures have also been successfully applied to high-efficiency multi-junction solar cells [11–13].

Compared with SSG MBLs, MSG MBLs can realize a higher indium content, and it is easier to accurately control the indium content of each In$_x$Ga$_{1-x}$As step layer when compared with LG MBLs. However, there has been no report on GaAs-based 1.55 $\mu$m wavelength range QD lasers using MSG InGaAs MBLs.

In this work, InAs/InGaAs QDs based on MSG In$_x$Ga$_{1-x}$As MBLs grown by MBE are investigated. The optical property and crystal quality of the InAs QDs dependent on the indium content of the overshooting layer and on QD growth conditions are analysed. A broad-area InAs/In$_{0.15}$Ga$_{0.85}$As QD laser operating at 1.47 $\mu$m is realized, with a threshold current density of 146.7 A cm$^{-2}$ under RT pulsed mode.

2. Experiments

Samples are grown in a Veeco Mod Gen-II solid source MBE system. The epitaxial structures are illustrated in figure 1: a 300 nm GaAs buffer, MSG In$_y$Ga$_{1-y}$As MBLs, a 200 nm In$_{0.15}$Ga$_{0.85}$As layer as ‘virtual substrate’, a 2 nm GaAs layer, two InAs/InGaAs QDs layers and a final uncapped InAs/InGaAs QDs layer.

The MSG MBLs are designed by increasing the indium content by 0.02 for each 200 nm step layer. The growth temperatures are 500 °C at the beginning and then lineally decreased to 390 °C. Between every two steps, a 6 min growth interruption is introduced for changing the temperatures of indium and gallium cells to accurately control molecular beam flux. In order to avoid indium desorption, a 1.5 nm AlAs layer is grown on the top of each InGaAs step layer. The 2 nm GaAs cover layer is grown after the 200 nm In$_{0.15}$Ga$_{0.85}$As layer is deposited to avoid indium desorption during the substrate temperature change from 390 to 510 °C.

The two-layer InAs QD structures are specially designed as shown in figure 1(a). The 5 nm-thick In$_{0.33}$Ga$_{0.67}$As layer acts as a strain-reducing layer and it improves the optical quality of the QDs [21]. The 2 nm GaAs layer sandwiched between the In$_{0.33}$Ga$_{0.67}$As layer and the 30 nm In$_{0.15}$Ga$_{0.85}$As layer acts as a carrier tunnelling injection barrier to reduce hot-carrier effects [7, 22]. The second 2 nm GaAs layer grown on top of the 30 nm In$_{0.15}$Ga$_{0.85}$As layer assures that every InAs QD layer is grown under the same conditions as the first one. The final uncapped InAs QD layer is for surface morphology characterization.

The sample morphology is measured by a Nanonavi Esweep S-II atomic force microscope (AFM) in contact mode. The microstructure and crystal quality of epitaxial layers are evaluated by a Philips Tecnai F20 field emission gun transmission electron microscope (FEG-TEM). Photoluminescence (PL) spectra are performed at RT using a Nicolet FTIR760 Fourier transform spectrometer with a liquid nitride cooled Ge detector. The excitation light is the 632.8 nm line from a He–Ne Laser and the power is about 11 mW.

3. Results and discussion

3.1. Optimization of MSG MBLs and growth conditions of InAs QDs

One of the effective ways to optimize the metamorphic layers particularly the indium content of the overshooting In$_y$Ga$_{1-y}$As layers is to compare the optical quality of the metamorphic InAs QDs. In$_y$Ga$_{1-y}$As layers with four $z$ values (0.15, 0.17, 0.18 and 0.20 for samples A1, A2, A3 and A4, respectively) are grown and thus there are four values of the indium content setback $\Delta x (= z - 0.15)$: 0, 0.02, 0.03 and 0.05, respectively). 2.9 ML InAs QDs are grown at 510 °C for these four samples.

RT PL spectra of the four samples are shown in figure 2. Their peak wavelengths are centred at 1.46 $\mu$m showing very tiny shifts. When $\Delta x$ is increased from 0 to 0.05, the PL intensities first increase to a maximum at $\Delta x = 0.03$ and then decrease. This value ( $\Delta x = 0.03$) was also used for InGaAs/InGaP multi-junction solar cells with similar MSG structures [10, 12]. This indicates that the In$_{0.15}$Ga$_{0.85}$As virtual substrate grown on the In$_{0.18}$Ga$_{0.82}$As overshooting layer has the best quality. This is possibly due to the fact that the residual strain in the overshooting layer originated from

![Figure 1. Schematic diagrams of (a) sample structure and (b) MSG In$_x$Ga$_{1-x}$As metamorphic buffer structures. The indium content $x$ is increased by 0.02 per step.](Image)
its two sides (the underlying In$_x$Ga$_{1-x}$As graded layer and the upper In$_{0.15}$Ga$_{0.85}$As virtual substrate) reaches a balance, so that the In$_{0.18}$Ga$_{0.82}$As overshooting layer has the smallest ‘net strain’. This strain balance is principally similar to the situation in LG MBL structures [18]. However, according to Torsell’s theory [18], the strain-relax mechanism in MSG MBLs is not exactly the same as that in LG MBLs, because the indium content setback values for MSG MBLs (0.03 with grading slope 0.01 $\mu$m$^{-1}$) and for LG MBLs (0.047 with the same grading slope) are different.

After the optimization of MBLs, the growth conditions of InAs QDs should be carefully optimized. Taking sample A3’s growth conditions as reference, the indium content setback is fixed at $\Delta x = 0.03$, but a lower growth temperature and an increased InAs coverage [23–25] are used to enhance the QD density. Finally, sample A5 is grown with 3.0 ML InAs QDs grown at 500 $^\circ$C. Figure 3 shows a comparison of the results between samples A3 and A5. The QD density is improved from 1.3 $\times$ 10$^{10}$ cm$^{-2}$ to 2.0 $\times$ 10$^{10}$ cm$^{-2}$; the PL intensity is enhanced 1.65 times and the FWHM is reduced from 33.4 to 27.9 meV. These results indicate that the optical quality of the InAs QD is effectively improved by changing the growth conditions.

3.2. Metamorphic InAs/In$_{0.15}$Ga$_{0.85}$As QD laser diode

3.2.1. Epitaxial structure. To further test the application feasibilities of the MSG MBL structures for devices, MSG metamorphic InAs/InGaAs QD laser diodes are grown using the growth technique as mentioned above. The laser structure consists of Si-doped 1.8 $\mu$m MSG InGaAs MBLs (setback $\Delta x = 0.03$), a Si-doped 200 nm In$_{0.15}$Ga$_{0.85}$As layer, a Si-doped 1.8 $\mu$m In$_{0.15}$Al$_{0.35}$Ga$_{0.65}$As bottom cladding layer, an undoped six-layer QD active region (3.0 ML grown at 500 $^\circ$C), a Be-doped 1.8 $\mu$m In$_{0.15}$Al$_{0.35}$Ga$_{0.65}$As up cladding layer, a heavily Be-doped 250 nm In$_{0.15}$Ga$_{0.85}$As and a 10 nm GaAs contact layer. TEM cross-sectional images of the laser structure are shown in figures 4(a) and (b). The broad-area laser diode (250 $\times$ 600 $\mu$m$^2$) was fabricated using standard processes.

In figures 4(a) and (b), dislocations can be seen in MSG InGaAs MBLs but do not penetrate up into the In$_{0.15}$Al$_{0.35}$Ga$_{0.65}$As cladding layer nor into the InAs QD active region. More detailed TEM images of the MSG InGaAs MBLs and InAs QD active region are shown in figures 4(c) and (d). It can be seen that dislocation density in the top two steps of the MSG InGaAs MBLs is much lower than that in the lower steps. This phenomenon is in good agreement with previous reports [26]. The low dislocation density in the top part of the MSG InGaAs MBLs indicates that the In$_{0.18}$Ga$_{0.82}$As overshooting layer has an appropriate composition to compensate for the residual strain. This goes well with the analyses from the PL spectra of samples A1, A2, A3 and A4.

Figure 4(d) shows similar density distribution for each QD layer. The uniform size distribution as seen in the lower four QD layers goes well with the narrow FWHM (28 meV) of sample A5 with a two-layer QD active region. However the QD size of the lower four layers is slightly larger than that of the upper two layers as confirmed by high-resolution TEM observations. This may be caused by surface undulation during epitaxial growth [7].

3.2.2. Dislocation study. Etch pit density (EPD) of the metamorphic InAs/In$_{0.15}$Ga$_{0.85}$As QD laser structure is studied using a modified A-B etchant solution with an etching rate of 2.2 $\mu$m min$^{-1}$ at 60 $^\circ$C [27, 28]. Four different etching depths...
Figure 4. TEM images of different regions of the metamorphic InAs/In$_{0.15}$Ga$_{0.85}$As QD laser diode. (a) Region I: MSG MBLs; region II: bottom cladding layer; (b) region II: bottom cladding layer; region III: six-layer InAs QD active region; region IV: upper cladding layer; region V: heavily doped contact layer; (c) MSG MBLs; (d) six-layer InAs QD active region.

(2.2, 3.0, 4.3 and 7.0 µm) are confirmed by cross-sectional scanning electron microscope images. From figure 5, it can be seen that dislocation density of the entire laser structure is less than 5.0 × 10⁴ cm$^{-2}$, slightly lower than that of the GaAs substrate. This value is better than the results of In$_{0.35}$Ga$_{0.65}$As on a four-step-graded InGaAs metamorphic buffer which has a 1.0 × 10⁴ cm$^{-2}$ dislocation density [16].

The PL spectra of the samples at different etching depths are shown in figure 6. The unetched sample has one peak at 1.04 µm which originates from the heavily Be-doped In$_{0.15}$Ga$_{0.85}$As contact layer. After etching away the top 2.2 µm layers, a peak at 1.47 µm originating from the InAs QDs can be seen. The peak FWHM of 33.1 meV is larger than that of sample A5 (27.9 meV) due to QD size distribution from the top two to the bottom four QD layers which was proved by high-resolution TEM observation in figure 4(d). No signal from InAs QDs is detected for the unetched sample due to free-carrier absorption at 1.47 µm from the heavily doped layers. When the etching depth is increased to 4.3 µm, a 1.03 µm strong peak from the 200 nm In$_{0.15}$Ga$_{0.85}$As on the MSG InGaAs MB layer is observed. Compared with the unetched samples, this peak is slightly blue shifted with a higher intensity. This is due to the band-tail effect resulting from the heavy Be doping in the In$_{0.15}$Ga$_{0.85}$As contact layer.

Figure 6. RT PL spectra of the metamorphic InAs/In$_{0.15}$Ga$_{0.85}$As QD laser structures with different etching depths.

3.2.3. Lasing characterization. The broad-area metamorphic InAs QD laser diode is tested using a pulsed current source with a repetition rate of 1 kHz and a pulse length of 1 µs. The $V$–$I$ and $P$–$I$ curves are shown in figure 7. The good rectifying behaviour of the laser is proved by the $V$–$I$ curve, and RT pulsed lasing is realized with a low threshold current density of 146.7 A cm$^{-2}$. This is the first report of InAs QD lasers based on MSG InGaAs MBLs. The performance of longer lasing wavelength and lower threshold current density is better than that of the previous reports on 1.3 µm InAs QD lasers based on MSG AlGaAsSb MBLs [20]. It has been noted that 1.46 µm metamorphic InAs QD lasers have already been reported [7, 8, 14]; however, the laser performance is expected to be improved by increasing the QD density or by introducing thermal annealing of the In$_{0.15}$Ga$_{0.85}$As space layer in the active region [7], and the lasing wavelength can be extended...
to 1.55 µm by increasing the indium content of the InGaAs virtual substrate.

4. Conclusion

Using multi-step-graded InGaAs MBLs, GaAs-based long-wavelength InAs quantum dots grown on In$_{0.15}$Ga$_{0.85}$As virtual substrates are studied. The optimized indium content setback value for the growth of multi-step-graded InGaAs MBLs is 0.03. Etch pit density in the InAs quantum dot active layers is less than $5.0 \times 10^3$ cm$^{-2}$. Finally, a 250 × 600 µm$^2$ broad-area laser diode has realized pulsed lasing with a low threshold current density of 146.7 A cm$^{-2}$ at room temperature. These results show that multi-step-graded InGaAs metamorphic buffer is a useful alternative structure for GaAs-based telecom wavelength optoelectronic devices.

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