Optical Properties of InAs Quantum Dots

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InAs quantum dots grown on GaAs substrate were investigated by optical spectroscopy. We particularly emphasized on the photoluminescence intensity, the stability of the photoluminescence intensity versus temperatures and wavelength of the InAs dot emission at various thermal treatments and different structures. We found that hydrogen can strongly passivate nonradiative centers without causing any structure degradation, and both n - and p -type modulation doping can reduce the decrease in the photoluminescence intensity when the sample temperature increases from the helium temperature to room temperature. The emission wavelength and the efficiency of the InAs quantum dots can also be manipulated by choosing proper materials of cap layer.

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1. Introduction

A quantum box is an ideal structure to explore quantum confinement, radiative recombination, carrier relaxation and many other fundamental physical phenomena, since the electronic structures can be theoretically predicted. Therefore, research on quantum dots (QDs) has become an extremely active field, not only due to the fundamental physics, but also because there are great potentials for opto-electronic applications of the dot structures, such as $1.3\ \mu\text{m}$ lasers in telecommunication. It has been shown that quantum dot lasers can operate at

very low threshold current density and can exhibit high stability with temperature. The properties of QDs, such as the electronic structures, emission wavelength, relaxation and decay times have been reported [1–7], as well as the InAs dot applications to e.g. laser devices [8, 9]. Experimental results show that the shape and size of InAs dots strongly depend on the growth conditions such as substrate temperature, growth rate, capping layer and nominal InAs thickness [1, 10]. Since the confined energies of electrons and holes in dots are determined by the shape, size and strain distribution, the emitting wavelength from InAs dots can be tuned by changing the growth conditions. In order to fabricate the InAs dots laser at the emission wavelength of 1.3 μm , it is not only needed to know what influences the emission wavelength, but it is also important to obtain high efficiency of the radiative recombination from the InAs QDs. A high efficiency of radiative recombination leads to a high optical gain. Furthermore, it is desired to obtain large value of the so-called T_0 which quantifies the temperature stability of semiconductor lasers.

The aim of this work is to investigate how the optical properties of the InAs dots can be manipulated, such as how the wavelength of the InAs QDs emission can be tuned, how the efficiency and the stability of the photoluminescence intensity from the InAs QDs can be improved. The post-growth treatments have also been carried out to study whether nonradiative centers can be suppressed.

2. Samples and experiment

Samples used in this investigation were grown in an EPI930 solid source molecular beam epitaxy system on semi-insulating (001) GaAs substrates, with a substrate temperature of 510°C and a nominal InAs thickness either 3 monolayer (ML) or 3.5 ML. A 50 nm GaAs buffer layer was first grown at a temperature of 580°C, then the growth was interrupted and the substrate temperature was ramped down to 510°C. The InAs QDs were formed at a growth rate of 0.1 $\mu\text{m}/\text{h}$. Three sets of samples were grown for this study. The first set of samples was directly grown on GaAs buffer, the difference is the thickness and In concentration of the $\text{In}_x\text{Ga}_{1-x}\text{As}$ insertion layer between the InAs layer and the final 200 nm GaAs cap layer. The second set of samples was that the InAs QDs layer directly grown on GaAs buffer layer, then 200 nm GaAs was followed. The only difference is that the 10 nm doping layer in GaAs barrier with 10 nm GaAs space layer was introduced. The InAs layer for the third set of samples was grown on $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ buffer layer, while the different thickness of the $\text{In}_x\text{Al}_{1-x}\text{As}$ plus 3 nm $\text{In}_x\text{Ga}_{1-x}\text{As}$ layers were followed, and finally a 200 nm $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ layer was grown. All samples used here are summarized in Table, and the structures are illustrated in Fig. 1.

The size and density of InAs dots were characterized by AFM measurements without capping layer. Figure 2 shows a typical AFM image picture for the InAs QDs grown on GaAs buffer. The lateral size of QDs is about 45 nm and the height is about 8 nm. The dot density is about $2.5 \times 10^{10}/\text{cm}^2$.

TABLE

The samples used in this study, A, B, and C represent the first, second, and third sets of samples, respectively, as illustrated in Fig. 2, L represents the thickness of InGaAs or InAlAs layer, x represents indium concentration, and t is the interrupting time.

Samples	L	x	t	Doping concentration	Ref. no. of sample
A1	0 ML	0	0	0	MBE318
A2	10 ML	0.40	0	0	MBE336
A3	7 ML	0.40	0	0	MBE348
A4	30 ML	0.20	0	0	MBE334
A5	0 ML	0	0	0	MBE491
A6	0 ML	0	5 min		MBE492
B1			0	0	MBE444
B2			0	$n = 5 \times 10^{18}/\text{cm}^3$	MBE503
B3			0	$p = 1 \times 10^{19}/\text{cm}^3$	MBE505
C1	0 ML	0	0	0	MBE495
C2	2.4 ML	0.17	0	0	MBE496
C3	4.8 ML	0.17	0	0	MBE498

Fig. 1. Schematic pictures of the samples used in this investigation.

Photoluminescence (PL) measurements were carried out at room temperature for all spectra that were shown in this study. A double grating monochromator and a liquid nitrogen cooled Ge-detector were used to disperse and detect the InAs QDs emission. An Ar^+ laser and an Ar^+ laser pumped tunable sapphire:Ti solid state laser were used as the excitation sources.

Fig. 2. $1 \times 1 \mu\text{m}$ AFM image of a sample grown under the same condition as the samples used in this investigation, but without a cap layer.

Parts of samples were treated by dc-hydrogen plasma. The samples were mounted on a heater block which can control the sample temperature of T_0 with 2°C accuracy, and placed 10 cm downstream from the plasma with a bias voltage of 300 V. The pressure of the plasma was 1.8 mbar. The duration of H-plasma treatment was t_0 . The two temperature (T_0) of 150°C and 250°C were used and the duration time of treatment (t_0) was 15, 30, 60, and 120 minutes, respectively.

3. Results and discussions

It has been concluded that it is difficult to realize $1.3 \mu\text{m}$ emission by directly capping the InAs QDs with a GaAs layer [1]. However, the emission wavelength from InAs dots can be tuned by inserting an $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer between the InAs dots and the GaAs cap layer [10], since the InGaAs layer can strongly suppress the In segregation from the InAs dots, resulting in a much less decrease in the InAs QDs height. In Fig. 3 we show the effect of the InGaAs insertion. At a constant In concentration, the wavelength of the InAs dot emission shows a red-shift with increasing the thickness of the insertion. Both the thickness and indium concentration influence the wavelength of the InAs dot emission. By adjusting the thickness and indium concentration, we can easily achieve $1.3 \mu\text{m}$ emission, for example, as shown from the PL spectra of samples A3 and A4 in Fig. 3. The InAs PL spectra in Fig. 3 show two transition peaks, which involve the first (low energy peak) and second (high energy peak) confined electron states in the QDs. With increasing the excitation power, the relative PL intensity of the transition involving the second electron state increases due to increase in electron population of this level.

Fig. 3. PL spectra for samples A1, A2, A3, and A4, measured at room temperature with the excitation power of 150 mW and an excitation wavelength of 514.5 nm.

The emission wavelength of the InAs QDs can be manipulated as discussed above. However, the intensity of the InAs QDs after the InGaAs insertion does not significantly increase, in many cases the PL intensity decreases as can be seen in Fig. 3. In order to enhance the optical gain, it is desired to obtain a high PL efficiency for laser structures. It is known that nonradiative centers in the GaAs region and the interface defects between InAs/GaAs or InAs/InGaAs can strongly reduce the efficiency of the radiative recombination from the InAs QDs. It is difficult to eliminate the nonradiative centers in GaAs region by simply modifying the structures. However, the interface defects may be suppressed by introducing a tunneling barrier. To illustrate the existence of the interface defects, Fig. 4 shows the PL spectra from two samples A5 and A6. The two samples have

Fig. 4. PL spectra for samples A5 and A6, measured at room temperature with the excitation power of 150 mW and an excitation wavelength of 514.5 nm.

exactly geometric structure and growth conditions, except that sample A6 contains an interruption between the InAs and the GaAs cap layer. The interrupting time is 5 minutes. When the interruption time becomes longer, there is a high probability to create more interface defects, and consequently the PL intensity of the InAs QDs will decrease. This fact is clearly shown in Fig. 4. The PL intensity of the InAs QDs with 5 minutes interruption is about a factor of 20 lower than the one without the interruption. In order to suppress the interface defects we may introduce a barrier between the InAs layer and GaAs layer such as InAlAs. In this case, the carriers generated by photon-excitation from the GaAs region in the top side of the InAs layer will be selectively stopped to interact with the interface defects by InAlAs, since the first few monolayers of InAlAs will only cover the region without any InAs QDs (i.e. only InAs wetting layer region), but not InAs dots part, at least the thickness which covers the InAs QDs region is much smaller than one which covers the InAs wetting layer region. Therefore the carriers can still be trapped in the InAs dots region, but the carriers can be sufficiently stopped to be trapped in the region without InAs QDs. Taking into account that the InAs QDs only cover 10 to 20% of whole surface and much slower carrier trapping in the InAs QDs than in the InAs wetting layer region, the intensity of the InAs QDs emission will be strongly enhanced due to the suppression of the interaction between the photon generated carriers and the interface defects, as shown in Fig. 5. The enhancement is strong, about factor of 100 when a 4.8 ML InAlAs insertion was introduced in the case shown here.

Fig. 5. PL spectra for samples C1, C2, and C3, measured at room temperature with the excitation power of 150 mW and an excitation wavelength of 514.5 nm.

The second way to suppress the nonradiative channels is the post-growth treatments. It is generally known that hydrogen can passivate shallow impurities, deep levels and defects. Some of them can be nonradiative centers, such as deep level charge defects and dangling bonds. We have treated the samples by hydrogen

Fig. 6. PL spectra for samples A2, measured at room temperature with two different excitation powers at an excitation wavelength 800 nm: (a) 130 mW, (b) 8 mW.

plasma. Figure 6 shows a typical result before and after the hydrogen treatments. The PL spectra in Fig. 6 are from sample A2 at two different excitation powers. The results clearly show an enhancement of the total PL intensity from the InAs QDs after the H-plasma treatment. The H-plasma treatment at such low temperature does not cause any structural modification since there is no energy shift of the InAs dots emission in Fig. 6. The enhancement of the total PL intensity of the InAs dots depends on the excitation power. At lower excitation power, we can see that the enhancement is stronger. The enhancement of the PL intensity is due to the H-passivation of nonradiative channels in the structures, since the thermal annealing in argon gas environment of the same samples does not show any change of the PL intensity. Furthermore, the H-passivation of those nonradiative channels can become active again by normal thermal annealing at temperatures above 400°C. The results show that the enhancement of the PL intensity after H-plasma treatment depends on the excitation power, from about a factor of 12 at the low excitation (about 3 mW) to about a factor of 2 at the highest excitation power (about 130 mW) used in this study. By carefully analyzing the dependence of the integrated PL intensity, we can conclude that the nonradiative centers in GaAs region are a main factor which causes the different enhancement at different excitation, since the number of photogenerated carriers will decrease faster when the carriers diffuse a long distance if the concentration of nonradiative centers is higher. The integrated PL intensity of the InAs QD emission versus temperatures and duration of the hydrogen treatment is summarized in Fig. 7 for sample A2. The results show a maximum enhancement at the duration time around 15 minutes, and the results with the sample treated at 150°C is better than one treated at 250°C. With increasing the duration time, the PL intensity starts to decrease,

Fig. 7. Integrated PL intensity of the InAs QDs from sample A2 versus the duration time of the hydrogen treatment at two temperatures. PL spectra were measured at room temperature with excitation power of 15 mW and excitation wavelength of 514.5 nm.

which is caused by surface damage during the hydrogen plasma bombardment. The surface damage is proved by the X-ray measurements, which show the graduate disappearing of the fringes with increasing the duration time. All samples show a similar result as discussed above.

So far, we have shown two ways to improve the radiative efficiency of the InAs QDs, and we have shown how to manipulate the emission wavelength of the InAs QDs. Now we see how the T_0 , which quantifies the temperature stability of semiconductor lasers, can be improved. Figure 8 shows the temperature dependence of the integrated PL intensity of samples B1, B2, and B3, which are the undoped, n -type doped and p -type doped InAs QDs structures as illustrated in Fig. 1, respectively. The intensity is normalized to the spectra measured at the lowest temperature in the figure. The results clearly show that the decrease in the PL intensity versus temperatures is much slower for the modulation-doped structures. The decrease in the PL intensity with increasing the temperature is probably mainly because of the increasing contribution of nonradiative recombination and the carrier escape due to the thermalization. The calculated data (lines in the figure) shows the effect of thermalization due to a simple two-level model. The carrier concentrations in the ground level (n_1) and the first excited level (n_2) have the following relation:

$$n_1 + n_2 = n,$$

$$\frac{n_2}{n_1} = \exp\left(-\frac{\Delta E}{kT}\right),$$

$$n_1 = \frac{n}{1 + \exp(-\Delta E/kT)}, \quad (1)$$

Fig. 8. The relative PL intensity of the InAs QDs from samples B1, B2 and B3 versus temperatures, measured with excitation power of 30 mW and excitation wavelength of 514.5 nm. The lines show the calculated results from Eq. (1).

where n is the total carrier concentration, and ΔE is the energy separation between the two levels. The lines in Fig. 8 are plotted as $\frac{n-1}{n}$ for ΔE equal to 5 meV (corresponding to n -type doping, since there are electrons in the InAs dots, the escape of the photon-generated carriers will be mainly due to hole level, which is in the order of 5 meV), 60 meV (corresponding to p -type doping, since there are holes in the InAs dots, the escape of the photon-generated carriers will be mainly due to electron level, which is in the order of 60 meV), and the effects from both electrons and holes (corresponding to undoped structure). If there is only thermalization effect, the undoped and n -type doped structures will show a similar result. From Fig. 8 we can conclude that the nonradiative channels mainly determine the temperature dependence of the PL intensity. Furthermore, the modulation doping can also suppress in a large degree the nonradiative centers, probably due to the change of Fermi level. The results also show that the structure with the p -type doping shows a better result, i.e. not only the nonradiative channels can be suppressed, but also the carrier escape due to thermalization can be reduced.

In summary, we have presented a detailed study on the InAs QDs grown on GaAs substrates. We show how the emission wavelength, the efficiency of the radiative recombination and temperature dependence of the PL intensity from the InAs QDs can be manipulated by the post-growth treatment and using proper structures. These factors are the key parameters to fabricate high quality QD laser.

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