Photoluminescence Studies of In$_{0.5}$Ga$_{0.5}$As/GaAs Quantum Dots

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Abstract

In this paper, we present an experiment study of the photoluminescence properties of self-assembled In$_{0.5}$Ga$_{0.5}$As/GaAs quantum dots (QDs). High quality In$_{0.5}$Ga$_{0.5}$As QDs were grown by using Metalorganic Chemical Vapor Deposition (MOCVD). The low temperature photoluminescence (PL) spectrum of the In$_{0.5}$Ga$_{0.5}$As quantum dots surrounded by GaAs exhibit an emission peak at 1.137 to 1.182 eV and a narrow linewidth (FWHM). The PL peak energy and intensity are varied with the excitation powers. Room temperature PL shows slightly lower peak energy with a broader spectrum linewidth. The distribution of the peak energy, peak intensity, and FWHM for the whole sample was obtained by using the PL mapping method.

Keywords: In$_{0.5}$Ga$_{0.5}$As quantum dots, MOCVD, photoluminescence.

Introduction

Self-assembled InGaAs QDs, epitaxially grown on GaAs, have attracted considerable attention in the recent decade, because of their high quality and promising applications in electronic and optoelectronic devices, such as, single electron transistors [1], lasers [2,3], optical memories [4], and infrared photodetectors [5]. The self-assembly or self-organization process is defined as the spontaneous organization of various components (molecules or nanoparticles) into a single, ordered aggregate. The organization process is made into a desired structure via physical, chemical, or biochemical interactive processes, including electrostatic & surface forces, hydrophobic, hydrophilic chemical interactions. All these processes are very selective and reject defects, and thus the resulted structure is in high degree of perfection [6].

The self-assembled QDs grown by the Stranski-Krastanov (SK) growth mode [7,8,9] have become extremely attractive because of high structural crystallinity, high density, discrete artificial atom-like energy levels, thermally stable states, a d-function-like density of states, and their simple growth procedures. An epitaxial growth of materials with a large lattice mismatch (in our case is InGaAs on GaAs) is unstable, and at a critical coverage the strain leads to a spontaneous formation of a uniform array of 3D defect-free islands (QDs) on a narrow wetting layer (WL). A large lattice mismatch (> 1.8%) between the epilayer and substrate is a necessary condition for SK-growth. It provides the thermodynamic driving force for the transition from 2D planar epitaxial growth of the epilayer to growth of the epilayer as 3D islands, the QDs, due to the more efficient lateral strain relaxation of 3D islands in comparison to the same material in a pseudomorphic 2D layer. The resulting densities, sizes and shapes of grown QDs are primarily determined by the growth conditions.

Photoluminescence spectroscopy is a very sensitive tool for investigating both intrinsic electronic transitions and electronic transitions at impurities and defects in semiconductor [10]. PL is used to detect an optical transition from an excited electronic state to a lower electronic state, usually the ground state. In the case of InGaAs QDs buried in GaAs, electron-hole pairs are formed by photon excitation. Since the InGaAs QDs band gap is smaller than the surrounding material (GaAs), so electrons will tend to "fall" into the dot to reach a lower-energy configuration. Recombination of
excitons will often produce a photon, in which the energy of photon tells us the band structure of the electron and hole.

Here, we report our investigation on the photoluminescence (PL) properties of self-assembled In_{0.5}Ga_{0.5}As/GaAs quantum dots. The QDs described here are the lens-shaped structures spontaneously formed on InGaAs WL and surrounded by the GaAs barriers. The PL characterizations of QDs were done on ensemble of QDs. In homogeneous broadening effect due to size and shape dispersion was clearly seen in the PL spectra obtained. PL measurements were carried out in order to study the interband transitions and thus the electronic band structures of InGaAs QDs. Effect of excitation power and measurement temperature on the PL properties were studied.

Materials and Methods

The QD structures in this study were grown on semi-insulating GaAs (001) substrates by Aixtron 200/4 metal-organic chemical vapor deposition (MOCVD) reactor. Arsenic (AsH₃), trimethylgallium (TMGa) and trimethylindium (TMIn) were used as sources for arsenic, gallium and indium respectively. In_{0.5}Ga_{0.5}As QDs were grown on top of a 400nm GaAs buffer layer and terminated by a 100nm GaAs capping layer. First, the GaAs buffer layer was grown at 650°C, and then a growth interrupt was done to reduce the temperature to 550°C before growing the InGaAs QDs. Immediately after the deposition of QDs, the GaAs growth (capping layer) was resumed while the temperature was ramped up to 650°C.

The PL measurements were performed at both low temperature and room temperature. For the low temperature PL measured at 4K, the 514.5nm line of an Argon laser was used as an excitation source. The luminescence signal was detected with a liquid nitrogen cooled Ge detector. The samples were mounted on the cold finger of a closed-cycle helium cryostat during the measurement. The room temperature PL and PL mapping measurement were carried out by using the Accent RPM 200 Rapid PL mapper. Argon laser with 532nm wavelength and 5.6mW was used as the excitation source. InGaAs photodetector is used to measure the room temperature PL signal.

Results and Discussion

In order to study the effect of the excitation power on the PL properties of In_{0.5}Ga_{0.5}As QDs, we have measured the PL spectral in a range of excitation power from 10mW to 40mW at 10mW intervals. Figure 1 shows the low temperature (4K) PL spectra of the investigated QD system measured at different excitation powers. Comparison of the peak wavelength, peak energy, intensity and full-width at half-maximum (FWHM) of these PL spectra is summarized in Table 1. All the PL spectra obtained have a symmetric Gaussian shape with a FWHM greater than 91meV. The broad behavior of the PL peak indicates that the PL spectra were inhomogeneously broadened due to averaging over QD ensembles with significant fluctuations in size, shape, composition and interdot separation.
Table 1: Summary of maximum intensity, peak wavelength, peak energy, FWHM of In$_{0.5}$Ga$_{0.5}$As QDs measured at different excitation power.

<table>
<thead>
<tr>
<th>Excitation Power</th>
<th>Maximum Intensity</th>
<th>Peak Wavelength (nm)</th>
<th>PL peak, $E_{\text{max}}$ (eV)</th>
<th>FWHM (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10mW</td>
<td>2.57875</td>
<td>1053.2</td>
<td>1.177</td>
<td>146.9</td>
</tr>
<tr>
<td>20mW</td>
<td>8.81487</td>
<td>1048.7</td>
<td>1.182</td>
<td>113.9</td>
</tr>
<tr>
<td>30mW</td>
<td>10.4676</td>
<td>1068.2</td>
<td>1.161</td>
<td>91.0</td>
</tr>
<tr>
<td>40mW</td>
<td>2.80951</td>
<td>1090.7</td>
<td>1.137</td>
<td>96.0</td>
</tr>
</tbody>
</table>

Figure 2: Low temperature PL spectra of In$_{0.5}$Ga$_{0.5}$As QDs measured at different excitation power (10mW to 40mW).

The low temperature PL spectra recorded with laser excitation power of 10mW shows broad luminescence peak which can be divided into two bands: one at 1.177eV and another lower intensity emission at 1.24eV (as shown in the inset of Fig. 1). The PL peak is first slightly blueshifted to 1.182eV when the excitation power was increased to 20mW, but then redshifted to 1.137eV with the further increase in excitation power. The peak intensity increases with excitation power, except for the PL spectra of 40mW excitation power which shows a striking decrease in peak intensity. When the excitation power increases, there will be higher excited population and thus higher recombination possibility, which causes the increase in luminescence intensity. The PL spectrum intensity of the 40mW excitation power is lower than that of 30mW, due to the saturation of signal which has been attained under a power of 30mW. At the same time, the FWHM of the spectra decreases with the excitation power, which suggests that the volume of QDs is so small that it is necessary to excite the sample at a very small excitation power.

It has been shown [11] that the In$_{0.5}$Ga$_{0.5}$As QDs emit at 1050nm, which is match with the luminescence observed in our study. Since the bulk GaAs has its band gap energy at 1.43eV, the luminescence observed here is not from the surrounding GaAs layers. Dominant luminescence peaks (1.137 - 1.182eV) observed at all excitation power are corresponding to interband transitions from the ground electronic subband to the ground heavy-hole band ($E_{1}\text{-HH}_1$). The luminescence peak energy is above the band gap of bulk In$_{0.5}$Ga$_{0.5}$As, which is 0.806eV, which is attributed to quantum confinement effects. The electronic confinement within nanometer size crystallites leads to a blueshift of the band gap of the In$_{0.5}$Ga$_{0.5}$As QDs from those of bulk. In molecular terminology, this
corresponds to the widening of the energy gap between the highest occupied molecular orbital and the lowest unoccupied molecular orbital as the size decreases. There have been suggested also that the effect of phonon bottleneck, state-filling, and segregated inhomogeneous broadening, can give rise to higher energy emission peak [12].

Figure 3: Room temperature PL spectra of In0.5Ga0.5As QDs measured at different excitation power (10mW to 40mW)

Figure 3 shows the PL spectra of In0.5Ga0.5As QDs measured at room temperature. The room temperature PL shows a peak at 1.014eV with the FWHM of 105.7meV. As the values for the energy gaps of the InAs QDs decrease with increasing temperature, the PL peak corresponding to carrier recombination shifts to lower energy. The redshift of peak energy and decrease of linewidth compared with the low temperature PL of 10mW excitation (similar excitation power) can be explained by the thermally enhanced carrier relaxation between dots due to carrier thermionic emission and/or carrier transport through the WL.

The PL line shape of the QDs at low temperature is strongly affected by the inhomogeneous distribution of the dots size and consists of a superposition of the emissions from many different-sized dots. When the temperature is high enough (room temperature), since the effect of the electron-phonon scattering becomes dominant, the PL linewidth starts to increase with increasing temperature.

Figure 4: PL mapping results of In0.5Ga0.5As QDs: PL peak wavelength (left), peak intensity (middle), and FWHM (right).
In order to study about the dot distribution and uniformity of the In_{0.5}Ga_{0.5}As QDs samples, the PL mapping measurement was done. The PL mapping results are shown in Figure 4. The mapping result shows an even distribution of emission peak (1165 to 1189nm) and FWHM (82.2 to 98.4nm) throughout the whole sample. However, the peak intensity has a broader distribution.

Conclusion

In summary, we have investigated the luminescence properties of In_{0.5}Ga_{0.5}As quantum dots grown on 400nm GaAs buffer layer at 550°C by using MOCVD. The reported dots show emission peak at 1.137 to 1.182eV for low temperature PL, and 1.014eV for room temperature PL. For the low temperature PL, an increase in PL intensity and a reduction in FWHM were observed for increasing laser excitation power. The low temperature and room temperature PL properties were compared. A redshift of peak energy and increased in spectrum linewidth was observed for the room temperature PL. The average PL linewidth from ensembles of dots are still broad and should be narrower. Hence, in future, the fluctuations in size and shape of nanometer scale islands formed in the SK growth mode have to be further reduced in order to produce QDs with superior electronic performance at room temperature.

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References