Effect of growth interruption on photoluminescence of self-assembled InAs/GaAs quantum dot heterostructures

Ya-Fen Wu
Department of Electronic Engineering, Ming Chi University of Technology, New Taipei City 24301, Taiwan

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The effects of growth interruption (GI) on self-assembled InAs/GaAs quantum dot (QD) heterostructures have been presented. Samples with different GI times were grown on GaAs substrate by metal-organic chemical vapour epitaxy. Photoreflectance and temperature dependent photoluminescence (PL) spectra from the samples were measured. It is found that increasing GI time during InAs QDs formation induces a larger dot size and a higher QD size uniformity. The carrier dynamics in QD system has been investigated in detail by analyzing the PL spectra. With increasing temperature, thermal redistribution effect among dots is evident for the sample with shorter GI time, leading to a quick red-shift of the PL peak energy and a reduction of the PL line width in mid-temperature range. On the other hand, for sample with longer time, such effect is weak and overcome by the electron-phonon scattering effect, which is consistent with the observed monotonically increasing linewidth of PL spectra with temperature.

Keywords: Quantum dot, Growth interruption, Temperature dependent photoluminescence, Size uniformity

1 Introduction

Self-assembled InAs quantum dots (QDs) formed on single-crystal GaAs substrates have been under intensive investigation for fundamental physics and potential device applications [1-3]. In InAs QD systems, the strong localization of the electronic wave function leads to an atom-like electron density of states and can be used for the realization of novel optoelectronic devices. Various applications such as laser diodes, light-emitting diodes, detectors, and optical amplifiers have been studied intensively [4,5]. However, the self-assembled growth of QDs affords in principle limited control of the structural properties of the dots.

A crucial issue for the realization of room temperature efficient photonic devices is an understanding of the temperature dependence of photoluminescence (PL) for QDs. Temperature dependence of PL emissions has been the subject of extensive studies for clarifying the carrier transferring mechanisms in a randomly distributed QDs system [6,7]. The relaxation dynamics in the zero-dimensional QD systems is expected to differ qualitatively from higher-dimensional systems, since the density of states is a series of δ-functions. The limited number of states available for carriers impairs carrier relaxation towards the ground state [8,9] (“phonon bottleneck” effect). In addition, the finite degeneracy of each QD state leads already to state filling effects when few carriers populate the lowest dot states. Both effects possibly result in inter sublevel relaxation rates that are comparable to inter band recombination rates.

In this work, the thermal activation of carrier dynamics of QD samples with different growth interruption (GI) time by the temperature dependent PL spectra has been investigated. The differences in the PL emission peak and in the PL full-width at half-maximum (FWHM) of the QD samples are well discussed. Lower PL emission energy and narrower FWHM of the PL spectra were observed as a result of longer GI time during QDs formation. The efficiency of carriers redistributed among dots plays an important role in the transferring mechanisms for sample with shorter GI time; while the electron-phonon scattering effect influences the PL spectra evidently for the sample with longer GI time.

2 Experimental Details

The InAs QDs samples studied in this work were grown on GaAs substrate by metal-organic chemical vapour epitaxy using the Stranski-Krastanow growth mode. The heterostructures include a 400 nm Si-doped GaAs buffer layer, an InAs QD active region of 3.3 monolayers (MLs), and a 100 nm undoped GaAs capping layer. The growth rate was 0.1 ML/s and the V/III ratio during the growth of InAs layer was 3. The growth interruption (GI) time introduced during dot formation were 15 s (sample A), 18 s (sample B) and 24 s (sample C), respectively.
The PL measurement was performed in a variable temperature (20-300 K) closed-cycle cryostat under the excitation of the 632.8 nm line of a He-Ne laser with the incident power intensity being 25 mW. Signals from samples were detected by a Ge photodiode using the standard lock-in technique. In PR measurement, the modulation of the built-in electric field is caused by the photo-excited electron-hole pairs created by a mechanically chopped 670 nm line (~5 mW) laser pumping source with a modulating frequency of ~200 Hz. The radiation from a 150 W tungsten-halogen lamp filtered by a 0.25 m monochromator provided the monochromatic light. The reflected light was detected by an InGaAs photodetector. The dc output of photodetector was maintained constant by a servo mechanism of variable neutral density filter. A dual-phase lock-in amplifier was used to measure the detected signals. The entire data acquisition procedure has been performed under computer control. Multiple scans over a given photon energy range was programmed until a desired signal-to-noise level has been attained.

### 3 Results and Discussion

Figure 1 shows an image for sample with GI=15 s (sample A) taken by high-resolution transmission electron microscopy (TEM), it is observed that a lens-shaped QD sample was fabricated. From the TEM image, the average dot density of sample A is estimated to be $1.3 \times 10^{10}$ cm$^{-2}$, and the average dot diameter of the sample is 19±0.5 nm. The normalized PL spectra of the samples measured at 20 K are shown in Fig. 2. All of the spectra are dominated by a pronounced three-peak emission from the InAs QDs. The PL signals are well reproduced by a convolution of three Gaussian-shaped peaks based on the line shape analysis. Emissions from transition level of ground state, the first excited state, and the second excited state are observed, denoted as $E_0$, $E_1$, and $E_2$, respectively. This may be due to the hindered intersublevel relaxation\cite{10,11}. For sample A, the $E_0$, $E_1$, and $E_2$ transitions are 1.032, 1.091 and 1.171 eV, respectively. For sample B, the corresponding values are 1.025, 1.089 and 1.167 eV, respectively. Lowest values of 1.014, 1.085 and 1.159 eV are observed for sample C with the longest GI time. Additional emission peaks at about 1.35 and 1.5 eV observed for sample A and sample B are from the wetting layer and the GaAs capping layer, respectively. Increase in dot size produces stress in the material which may affect the PL emission. Taking into account the quantum-size effect on the peak energy, the excitons localized in smaller dots exhibit higher energy emission, while the excitons localized in larger dots exhibit lower energy emission. The decreasing emission energy of the samples with increasing GI time implies that the dot size becomes larger as the GI time increases. Increase in dot size may produce stress in the material which should induce a blue shift in the PL emission. The observed red-shift emission energy of the samples with increasing GI time implies that the strain effect is unapparent in our samples.

During the InAs QDs formation, increasing the GI time not only induces a larger dot size, but also induces a higher dot uniformity\cite{12,13} of QDs. The FWHMs of the $E_0$, $E_1$, and $E_2$ transition levels for sample A are 34.93, 70.08, and 166.15 meV, respectively. For sample B and sample C, the corresponding values are 32.31, 44.8, 160.63 meV, and 30.25, 35.82, 152.06 meV, respectively. The value of PL linewidth is determined mainly by size fluctuation of the InAs islands at low temperature\cite{14,15}. The size fluctuation indicates the homogeneity of the QD size distribution, causing a distribution of the

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Fig. 1 — Plan-view TEM images for InAs/GaAs quantum dots of sample A with GI=15 s

Fig. 2 — Normalized PL spectra for samples A, B and C at 20 K. The growth interruption time for samples A, B and C are 15 s, 18 s, and 24 s, respectively
density of states over a wide energy range, resulting in a spectrally broad luminescence. Therefore, the measured narrowest FWHMs for sample C with longest GI time are consistent with its best size uniformity among the samples. More evidence can be seen from the weakest peak intensity of the second excited state ($E_2$) and the unclear wetting layer and capping layer emissions for sample C, which are attributed to the relaxed phonon bottleneck effect and the induced higher relaxation rate, resulting from its better QDs uniformity.

To further examine the emission states, PR measurements were performed on sample B and sample C, as shown in Fig. 3. The contactless mode of PR is non-destructive as PL is but as absorption-like they appear to be complementary to the emission-like techniques. They can probe various higher order states in the structure, which are not accessible in emission. In PR measurements, modulation of the built-in electric field in the sample is caused by photo-excited electron-hole pairs created by the pump source. This procedure results in sharp derivative-like spectral features in the region of inter band transitions. The derivative nature of PR spectra suppresses uninteresting background effects and greatly enhances the precision of inter band transition energies. The PR spectra marked with arrows shown in Fig. 3 are consistent with the corresponding emission peaks of PL spectra at 300 K. More complicated band-shapes are observed from PR spectra, which we attribute to the higher quantized energy states in InAs active layers.

The temperature dependent peak position and FWHM of sample A are shown in Fig. 4. Observing our experimental data, the red-shift of peak energy is greater than that of the InAs bulk band gap and the FWHM first decreases and then increases with temperature. In semiconductors, the variation of the peak position with temperature can be attributed to the effect of dilation of lattice and electron-lattice interaction. This effect is related by the thermodynamic identities to the influence of electron-hole pairs on the lattice vibration frequencies. The electron-hole pair softens the lattice vibration of the crystal when it is excited across a band gap. Theoretical treatments show that these effects should follow the Varshni relation:

$$E_g(T) = E_g(0) - \frac{\alpha T^2}{T + \beta} \quad \text{…(1)}$$

where $E_g(T)$ is the emission energy at temperature $T$, $E_g(0)$ the energy gap at 0 K, and $\alpha$ and $\beta$ are Varshni's fitting parameters. In our calculation, the values of $E_g(0)$, $\alpha$, and $\beta$ for InAs are 0.417 eV, 0.276 eV/K, and 93 K, respectively. The fast red-shift of PL

![Fig. 3 — PR and PL spectra of InAs QD samples at 300 K. The arrows designate the energy states observed from PR measurement](image)

![Fig. 4 — Experimental temperature dependent (a) peak energies, and (b) FWHMs of $E_0$ (ground state), $E_1$ (the first excited state), and $E_2$ (the second excited state) for sample A. The dashed line in (a) is calculated according to the Varshni law using the parameters of InAs and shifted along the energy axis](image)
energy and the anomalous decrease of line width with increasing temperature have been attributed to the thermally enhanced carrier relaxation among dots due to carrier thermionic emission and lateral transition through the wetting layer. At low temperatures, carriers are randomly distributed among the potential minima and there is no significant thermal emission of carriers out of the QDs. With elevated temperatures, carriers may be thermally activated outside the small dots into the wetting layer and then preferentially relax into large dots with relatively low-localized energy states. Thus, the peak energy experiences a quick red-shift because the emitting QDs are characterized by energetically lower states. At the same time, the size distribution of QDs taking part in the emission becomes narrower and the FWHM follows the same trend and decreases as the temperature increases. As the temperature increases further (>200 K), both the electron-phonon scattering and thermal broadening become important, and the FWHM increases with temperature.

Redistribution processes in QDs strongly affect the temperature dependence of the PL spectra for sample A. To illustrate the thermal induced behaviour of these samples, the excited state emission peaks of the PL spectra from 20 to 300 K are shown in Fig. 5. It can be seen that a similar but weaker thermal trend of peak energy is observed for sample B and sample C. The temperature dependence of the peak energy becomes weaker as the GI time is increasing. We calculate the amount of red-shift of peak energy in the temperature range 20-300 K for all the samples and the values are listed in Table 1. Referring to the calculated results, all of the red-shifts of peak energy are larger than that of bulk InAs (~58.95 meV). Sample C exhibits the smallest value, which is owing to the higher QD uniformity and larger dot size of it. The carriers in larger dots have a lower excited state energy corresponding to a higher binding energy. Thus, fewer carriers can be thermalized with increasing temperatures. The thermal redistribution effect of carriers among dots is unobvious and the amount of quick red-shift of peak emission is smaller.

Figure 6 shows the temperature dependent PL FWHMs of the $E_2$ transition level for the samples. It is noticeable that the measured result of sample C is significantly different from the other samples. The broadening of the PL spectra increases monotonically with temperature, no reduction is observed as the temperature increases. Due to the lowest PL intensity of the excited state, fewer carriers exist in the state, and the thermal redistribution of carriers via wetting layer is unobvious. Besides, the higher size uniformity of this sample also makes the redistribution effect among QDs becomes indistinct. The quick red-shift of peak energy shown in Fig. 4, confirms the weak redistribution effect, whereas the increase of line width with temperature implies that the electron-phonon scattering is dominant in the PL spectra. It is found that the thermal-activated redistribution and electron-phonon scattering effects both exist in the sample; however, for sample with better size

![Fig. 5 — Temperature dependent peak energies of the second excited state of InAs QD the samples. The solid line is calculated according to the Varshni law using the parameters of InAs and shifted along the energy axis](image1)

![Fig. 6 — Temperature dependent FWHMs of the second excited state for samples A, B and C](image2)
uniformity the repopulation of carriers is indistinct and overcome by the thermal enhanced electron-phonon scattering\textsuperscript{13,19}.

To identify the mechanism of the PL quenching, the integrated PL intensity of the second excited state for the samples has been calculated over a broad temperature range, as shown in Fig. 7. The curves of the samples can be theoretically fitted\textsuperscript{20} using the following Eq.(2):

\[
I_{PL}(T) = \frac{1}{1 + \alpha \exp\left(-\frac{E_a}{kT}\right)} \quad \ldots(2)
\]

where \( I_{PL}(T) \) is the integrated PL intensity at temperature \( T \), \( \alpha \) is a constant, \( kT \) is the thermal energy, and \( E_a \) is the thermal activation energy. From the fittings, we find that the activation energies are 95.91, 97.56, and 146.23 meV for samples A, B and C, respectively. The energy difference between the \( E_2 \) transition level and the GaAs barrier is about 310 meV for all of the samples, significantly larger than the obtained activation energy. We suggest that PL quenching mechanism may be related to the defect state formed at the InAs/GaAs interface because the heteroepilayers grown in three-dimensional mode generally contain some density of defects. Furthermore, the activation energy extracted from the plot also has a size-dependent behaviour. Since the activation energy is correlated with exciton confinement ability, the higher activation energy of sample C implies a deeper confining potential, consistent with its larger dot size. The experimental results demonstrated that sample with longer GI time has a better carrier confinement that can increase radiative recombination rate.

4 Conclusions

In the present paper, the PL spectra for the samples with different GI time over a broad temperature range have been studied. The different temperature dependent behaviour of the PL emission peak and FWHM of the samples are well discussed. Increasing the GI times leading to a larger dot size and a higher dot uniformity of QDs, which are confirmed by the lower PL emission energy and narrower FWHM of the PL spectra. The effect of thermal emission and recapture of carriers among dots is evident for the sample with shorter GI time. On the other side, owing to the better size uniformity for sample with longer GI time, the carrier redistribution is indistinct and overcome by the thermal enhanced electron-phonon scattering. Finally, the activation energy of the samples has been calculated to analyze the thermal quenching mechanism. The higher activation energy of longer-GI sample implies a deeper confining potential and better carrier confinement that can increase radiative recombination rate. The detailed investigation of carrier dynamics in QD system is of particular significance to the design of QD structures and is expected to lead to improved optoelectronic devices.

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