Research Paper

Effect of Growth Cycles on the Luminescence Properties of InP/InGaP Quantum Structures Grown Using Migration-Enhanced Epitaxy

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ABSTRACT

The luminescence properties of InP/InGaP quantum structures (Qs) grown using migration-enhanced epitaxy (MEE) with different numbers of repetition of the growth cycles are investigated using temperature-dependent photoluminescence (PL) and time-resolved PL. As the number of MEE repetition cycles increases from 3 to 8, the main PL peak originating from quantum dashes (QDashes) is red-shifted from 1.74 to 1.59 eV, owing to the change in the aspect-ratio of InP QDashes, whereas the PL peak energy originating from InP quantum dots and InGaP wetting layer remains nearly constant. The sample grown with six MEE repetition cycles shows the strongest integrated PL intensity with a comparatively narrow linewidth at room temperature. The six-cycled sample demonstrates an increase in PL decay time and radiative lifetime up to the highest temperature (120 K). These results indicate that the structural and luminescence properties of the InP/InGaP Qs can be controlled by adjusting the number of MEE repetition cycles; the best optical properties are obtained with the six-cycled InP/InGaP Qs sample at a growth temperature of 480 °C.

Keywords: Quantum structure, Photoluminescence, Time-resolved photoluminescence, Migration-enhanced epitaxy

I. Introduction

Low-dimensional semiconductor quantum structures (Qs) such as quantum dots (QDs) and quantum dashes (QDashes) have attracted considerable research attention as prospective materials for use in light-emitting devices, optical amplifiers, solar cells, and transistors [1–3]. InP/InGaP Qs have been studied for manufacturing high-power light sources with an emission wavelength below 850 nm [4–8]. A migration-enhanced epitaxy (MEE) method was employed to enhance the uniformity and alignment of Qs [9–11]. The structural and optical properties of InP/InGaP Qs grown using the MEE method were affected by growth temperature, repetition number of growth cycle, interruption time, InGaP spacer thickness, and others [10–14]. In particular, the structural and luminescence properties of InP/InGaP Qs grown with different numbers of MEE repetition cycles were characterized using atomic force microscopy (AFM), scanning electron microscopy (SEM), photoluminescence (PL), and emission wavelength-dependent time-resolved PL (TRPL) [10,12]. However, merely understanding the luminescence properties and carrier dynamics of InP/InGaP quantum structures (Qs; QDs + QDashes) grown with different numbers of MEE repetition cycles is not sufficient.

In this paper, we investigated the luminescence properties and recombination mechanisms of InP/InGaP Qs as a function of the number of MEE repetition cycles using temperature-dependent PL

and TRPL spectroscopies. As the number of MEE repetition cycles increases from 3 to 8, the main PL peak originated from QDashes was red-shifted, owing to the increase in the size (width and height) of QDashes. The six-cycled sample exhibited the strongest integrated PL intensities and the increase in the PL decay time up to the highest temperature (120 K).

II. Experimental details

InP/InGaP Qs samples were grown on semi-insulating GaAs substrates using a solid source MBE system. Prior to the growth of InP Qs, a 0.1-μm-thick GaAs buffer layer and a 50-nm-thick InGaP spacer layer were deposited on the GaAs substrates at temperatures of 580 °C and 480 °C, respectively. For the growth of InP/InGaP Qs samples, the MEE method wherein In and P₂ sources are alternately supplied with a growth interruption (GI) time was used. The GI time between depositions was provided to obtain the migration time of sources. One cycle in the MEE growth method consisted of In and P₂ sources for a 2 s supply and a GI for 10 s after supplying each source. InP Qs used in this study were grown with different numbers of repetition cycles (3 to 8 cycles) at a temperature of 480 °C. More information for the growth conditions and MEE method can be found in Ref. 10.

PL measurements were employed using a spectrometer (DM500i, Dongwoo Optron Co., Ltd.), a 532-nm diode laser (MGL-FN-532, CNI Laser) as an excitation source, and a charge coupled device detector

(ANDOR DV420-BU2). The samples were mounted in a closed-cycle refrigerator (CCS-150, Janis Research Co., Inc.), which recirculate a fixed volume of helium gas to provide cooling to the sample mounting stages, and temperatures ranging from 10 to 300 K were adjusted by a cryogenic temperature controller (331, Lake Shore Cryotronics, Inc.). In order to verify the recombination dynamics, temperature-dependent TRPL measurement were carried out using a spectrometer (FLS 920, Edinburgh Instruments Ltd.), a picosecond-pulsed diode laser ($\lambda = 375$ nm, pulse width = 90 ps, EPL-375, Edinburgh Instruments Ltd.) as an excitation source, and a micro-channel-plate photomultiplier tube detector using a time-correlated single photon counting system. For the temperature-dependent TRPL measurements, the samples were mounted in a closed-cycle cryostat (GB15, Cryomech Inc.) and a temperature controller (ITC503S, Oxford Instruments) were used for monitoring temperatures (10–300 K).

III. Results and discussion

Figure 1(a) displays the room temperature (RT) PL spectra of InP/InGaP QS samples grown with different numbers of repetition of the MEE growth cycle. The PL spectra of all samples include three PL peaks comprising a broad strong peak (P1) and two shoulder peaks that appear below and above P1 (P2 and P3, respectively). Peaks P1 and P2 are attributed to the emissions of InP QDashes and QDs, respectively, and peak P3 is ascribed to the emission of the InGaP wetting layer (WL).

The AFM and SEM images verified the formation of InP QDashes and QDs [10]. The PL peak energies of InP/InGaP QS samples extracted from Gaussian (P2 and P3) or exponentially modified Gaussian (P1) fitting are shown in the inset of Fig. 1(a). With an increase in the number of MEE repetition cycles from 3 to 8, the peak energy of P1 gradually decreases from 1.74 to 1.59 eV and that of P2 slightly changes between 1.50 and 1.53 eV, as seen in the inset of Fig. 1(a). In contrast, the peak energy of P3 remains unchanged as the number of repetition cycles increases. The redshift of P1 can be ascribed to the

change in the aspect-ratio of InP QDashes. In the SEM and AFM images, the height and width of QDashes were seen to be increased because of an increase in the indium amount caused by the increase in the number of repetition cycles while the length of QDashes was shortened [10]. Therefore, an increase in quantum size caused by increasing the number of MEE repetition cycles leads to a decrease in the peak energy of P1.

Figure 1(b) shows the integrated PL intensity and full width at half maximum (FWHM) of PL peaks (P1, P2, and P3) as a function of the number of MEE repetition cycles. The integrated PL intensity of P3 slowly decreases with increasing the number of repetition cycles. On the other hand, the integrated PL intensities of P1 and P2 exhibit an increase and decrease as the number of MEE repetition cycles increases from 3 to 8. The increase in the PL intensities can be explained by the increase in the density of QSs; the decrease in the PL intensities during the repetition cycles of 7 and 8 can be attributed to defects or lattice mismatch between the InP and InGaP layers [10]. As the number of MEE repetition cycles increases from 3 to 8, the increase and decrease in the densities of QDashes and QDs, respectively, were demonstrated by the AFM image [10]. The FWHM of P1 peak is remarkably decreased from 170 to 94 meV as the number of MEE repetition cycles increase from 4 to 6, and then, it becomes slightly narrow up to 86 meV. This FWHM behavior can be explained by the uniform growth of InP/InGaP QSs for MEE repetition cycles from 6 to 8. The 6-cycled sample exhibits the strongest integrated PL intensity and relatively narrow FWHM.

To investigate the PL decay dynamics of InP/InGaP QS samples grown with various MEE cycles, TRPL measurements were performed as a function of temperature from 10–180 K. Figures 2(a)–2(c) show the temperature-dependent PL decay curves of InP/InGaP QS samples grown with 4, 6, and 8 MEE repetition cycles, respectively. The PL decay curves were plotted at the main PL peak (P1) of each sample. All three samples indicate that the PL decays become slower from 10 to 100–120 K, abruptly shortening as the temperature increases further up to 180 K. The PL decay times of the 4-cycled InP/InGaP QS sample are calculated using the bi-exponential function; $I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$, where τ_1 and τ_2 are the fast and slow decay times, respectively, and A_1 and A_2 are the pre-exponential constants. The decay times of the 6- and 8-cycled samples are estimated using a single-exponential function. The fast decay time τ_1 for the 4-cycled sample is dominant at low temperatures, and thus, τ_1 corresponds to the decay time of the QDashes in the sample. The slow decay time τ_2

Figure 1. (Color online) (a) PL spectra of InP/InGaP QS samples grown with different numbers of MEE repetition cycles (3 to 8 cycles) measured at room temperature (RT). The PL emission peak located at intermediate, lower, and higher energies are denoted as P1, P2, and P3, respectively. The inset shows the PL peak energies of InP/InGaP QS samples as a function of the number of MEE repetition cycles. (b) The integrated PL intensities and FWHMs of InP/InGaP QS samples are shown as a function of the number of MEE repetition cycles.

Figure 2. (Color online) Temperature-dependent PL decay curves of InP/InGaP QS samples grown with (a) 4, (b) 6, and (c) 8 MEE repetition cycles at the P1 peak position for each corresponding temperature.

Figure 3. (Color online) (a) Normalized integrated PL intensities and (b) PL decay times of P1 in InP/InGaP QS samples grown with 4, 6, and 8 MEE repetition cycles as a function of temperature. The temperature-dependent FWHMs and radiative lifetimes of P1 for InP/InGaP QS samples are displayed in the insets of (a) and (b), respectively.

for the 4-cycled sample may be attributed to carrier redistribution from higher energy states (smaller QDs) to lower energy states (larger QDs) via the WL [12,15,16]. The decay times τ of the 6- and 8-cycled samples also correspond with the PL decay times of the QDashes in InP/InGaP QS samples.

The temperature-dependent integrated PL intensities of P1 in InP/InGaP QS samples grown with different numbers of MEE repetition cycles are shown in Fig. 3(a). The integrated PL intensities of all three samples are almost constant up to ~100 K, while decreasing rapidly with an increase in temperature up to 300 K. The PL intensity of the 6-cycled sample is larger than that of the other samples for all temperatures. The temperature-dependent FWHMs of P1 in InP/InGaP QS samples are shown in the inset of Fig. 3(a). As temperature increases from 10 to 140 K, the FWHM of the 4-cycled sample becomes narrower from 64 to 52 meV, while beginning to broaden with further increase in temperature up to 300 K. In contrast with the 4-cycled sample, the 6- and 8-cycled samples exhibit a gradual broadening of the FWHM with increasing temperature from 10 to 300 K, indicating the formation of enlarged and/or elongated QDash structures caused by agglomerated QDs with increasing number of MEE repetition cycles.

The FWHM narrowing in the 4-cycled sample is attributed to an increase in the carrier redistributions from smaller QDs to larger QDs with increasing temperature [14]. The decrease in PL intensity and the broadening of FWHM with increasing temperature are attributed to the increase in the non-radiative recombination process and thermal expansion of the carrier distribution function, respectively [17].

Figure 3(b) shows the temperature-dependent PL decay times of InP/InGaP QS samples estimated with the PL decay curves shown in Fig. 2. The PL decay times for all three samples are increased up to 100–120 K, and then decreased at higher temperatures. The PL decay time of the 6-cycled sample is increased up to 120 K, which is the highest temperature among samples. The radiative (τ_R) and non-radiative (τ_{NR}) lifetimes were calculated with $\eta(T) = \tau_{PL}(T)/\tau_R \approx I(T)/I_0$, $1/\tau_{PL} = 1/\tau_R + 1/\tau_{NR}$, where $\eta(T)$, $I(T)$, and $\tau_{PL}(T)$ are the internal quantum efficiency, integrated PL intensity, and PL decay time, respectively, as

a function of temperature. We assumed $\eta(10\text{ K}) = 1$ and $I(10\text{ K}) = I_0$, and the temperature-dependent integrated PL intensity and PL decay time were used for $I(T)$ and $\tau_{PL}(T)$, respectively [18,19]. In the inset of Fig. 3(b), the estimated radiative lifetimes for all three samples are shown as a function of temperature. As can be seen, the radiative lifetimes for all three samples increase continuously with increasing temperature, which is a typical characteristic of semiconductors. The comparison between the decay time and radiative lifetime indicates that the PL decay times at low temperatures (≤ 100 –120 K) are strongly dependent on radiative recombination. At high temperatures (> 100 –120 K), the decrease in the PL decay time can be attributed to an increase in the non-radiative recombination process. The increase in the decay time at low temperature regions can be explained by several mechanisms, such as thermally activated migration of carriers from WL and InGaP barriers, carrier redistribution among the QDs, and reduced wave function overlap caused by carrier expansion.

IV. Conclusions

The luminescence properties of InP/InGaP QS samples grown with different numbers of repetition of MEE growth cycles were investigated using PL and TRPL spectroscopies as a function of temperature. With an increase in the number of MEE repetition cycles from 3 to 8, the main PL peak originated from QDashes was red-shifted; this was attributed to the increase in the size (width and height) of QDashes. The 6-cycled sample exhibited the strongest integrated PL intensities among the samples at all temperatures ranging from 10 to 300 K. The temperature-dependent PL decay times of InP/InGaP QS samples grown with 4-, 6-, and 8-cycled MEE method showed a typical characteristic of QS semiconductors such, as an increase and decrease in decay time with increasing temperature. The 6-cycled sample demonstrates the strongest PL intensity and the increase in the PL decay time up to the highest temperature (120 K), indicating the optimum number of repetition cycles in the MEE growth at 480 °C for the formation of InP/InGaP QDs.

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