Research Paper

Temperature-dependent Luminescence Properties of Digital-alloy In(Ga_{1-z}Al_z)As

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Abstract The optical properties of the digital-alloy (In_{0.53}Ga_{0.47}As)_{1-z}/(In_{0.52}Al_{0.48}As)_z grown by molecular beam epitaxy as a function of composition z ($z = 0.4, 0.6$, and 0.8) have been studied using temperature-dependent photoluminescence (PL) and time-resolved PL (TRPL) spectroscopy. As the composition z increases from 0.4 to 0.8, the PL peak energy of the digital-alloy In(Ga_{1-z}Al_z)As is blueshifted, which is explained by the enhanced quantization energy due to the reduced well width. The decrease in the PL intensity and the broaden FWHM with increasing z are interpreted as being due to the increased Al contents in the digital-alloy In(Ga_{1-z}Al_z)As because of the intermixing of Ga and Al in interface of InGaAs well and InAlAs barrier. The PL decay time at 10 K decreases with increasing z , which can be explained by the easier carrier escape from InGaAs wells due to the enhanced quantized energies because of the decreased InGaAs well width as z increases. The emission energy and luminescence properties of the digital-alloy (InGaAs)_{1-z}/(InAlAs)_z can be controlled by adjusting composition z .

Keywords: Digital-alloy, InGaAlAs, Photoluminescence, Time-resolved photoluminescence

I. Introduction

InGaAlAs quaternary alloys have been of much interest for optoelectronic devices operating at 1.0 to 1.65 μm [1-5]. The digital-alloy technique has been used to grow ternary or quaternary alloys by molecular-beam epitaxy (MBE). This technique overcomes intrinsic problems of the conventional MBE growth such as change of cell temperature, growth interruption, and additional source cell, etc. [6,7]. Defects such as alloy disorder due to an intermixing of Ga and Al atoms at the interfaces between InGaAs well and InAlAs barrier can be created in the digital-alloy InGaAlAs grown with (In_{0.53}Ga_{0.47}As)_{1-z}/(In_{0.52}Al_{0.48}As)_z short-period superlattices (SPSs) [8,9]. In order to reduce defects, which act as non-radiative recombination centers, many efforts such as rapid thermal annealing process and adjusting the thickness of InGaAs well and/or InAlAs barrier have been proposed [8-10].

Temperature- (T-) dependent optical processes give rise to various thermal dynamics of carriers in semiconductors such as carrier redistribution between energy states and/or carrier escape from active region to barrier [11,12] and can calculate the interband energy spacing by analyzing the T-dependent integrated photoluminescence (PL) intensity [13]. In this paper, the luminescence properties of digital-

alloy In(Ga_{1-z}Al_z)As grown by MBE have been investigated by using a T-dependent PL as a function of composition z in digital-alloy (In_{0.53}Ga_{0.47}As)_{1-z}/(In_{0.52}Al_{0.48}As)_z ($z = 0.4, 0.6$, and 0.8). At low (high) temperature region, the thermal activation energies are extracted to be 3.87 (95.2), 8.89 (86.4), and 15.97 (70.5) meV for the sample with $z = 0.4, 0.6$, and 0.8 , respectively. In order to better understand the recombination dynamics of the digital-alloy InGaAlAs, T-dependent time-resolved PL (TRPL) have been also carried out as a function of temperature.

II. Experiments

The digital-alloy In(Ga_{1-z}Al_z)As samples were grown on n-InP substrate by MBE system. A 100 nm-thick In_{0.52}Al_{0.48}As cladding layer was deposited on top of InP substrate at 510°C. The digital-alloy In(Ga_{1-z}Al_z)As layers, which are the lattice-matched to InP substrate were grown at 510°C and followed the growth of 25 nm-thick In_{0.52}Al_{0.48}As capping layers at the same temperature. The composition z of In(Ga_{1-z}Al_z)As layers are varied from 0.4 to 0.8. The thickness of In_{0.53}Ga_{0.47}As (In_{0.52}Al_{0.48}As) layer for $z = 0.4, 0.6$, and 0.8 are 9.8 (6.6), 6.6 (9.8), and 3.75 (15) Å, respectively. The interface and superlattice periodicity of digital-alloy InGaAlAs were clearly observed by cross-sectional transmission electron microscopy [14]. More detailed information for the growth conditions of the samples can be found in Refs. 6 and 7.

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The PL and TRPL measurements were carried out using an FLS 920 spectrometer (Edinburgh Instruments). T-dependent luminescence measurements in the range of 10 to 300 K were performed using a helium closed-cycle cryostat. The PL and TRPL spectra of the digital-alloy $\text{In}(\text{Ga}_{1-z}\text{Al}_z)\text{As}$ samples excited by using a 532-nm continuous-wave laser and a 634-nm pulsed diode laser (pulse width = 93 ps), respectively, were measured using a near-infrared photomultiplier tube detector. The PL decay profiles were measured by using a time-correlated single photon counting system.

III. Results and Discussion

Figure 1(a) shows the PL spectra of the digital-alloy $(\text{In}_{0.53}\text{Ga}_{0.47}\text{As})_{1-z}/(\text{In}_{0.52}\text{Al}_{0.48}\text{As})_z$ samples measured at 10 K. The PL peak energy for the $(\text{In}_{0.53}\text{Ga}_{0.47}\text{As})_{1-z}/(\text{In}_{0.52}\text{Al}_{0.48}\text{As})_z$ samples with composition z of 0.4, 0.6, and 0.8 is 1.117, 1.252, and 1.409 eV, respectively. As z increases from 0.4 to 0.8, the PL peak energy at 10 K is blueshifted about 292 meV. This blue-shift is attributed to the enhanced quantization energy caused by the decrease of well width from 9.8 to 3.75 Å with increasing z from 0.4 to 0.8, respectively. The weak PL peak around 1.441 eV in Figure 1(a) comes from the InP substrate [6]. As seen in Figure 1(a), the sample with $z = 0.6$ exhibits the strongest PL intensity, which indicates the enhanced carrier confinement into unit well regions due to reduced unit well width and increased InAlAs barrier's thickness. The sample with $z = 0.8$ exhibits slightly reduced PL intensity compared with that of sample with $z = 0.6$. Figure 1(b) shows the full width at half maximum (FWHM) of the PL spectrum for the samples. The sample with $z = 0.6$ shows the narrowest FWHM (27.0 meV), while the sample with $z = 0.8$ exhibits much broad FWHM (36.4 meV). The decreased PL intensity and broadened FWHM of the sample with $z = 0.8$ compared with those of the sample with $z = 0.6$ can be attributed to the poor crystalline quality due to the high aluminum (Al) contents. With z increases from 0.6 to 0.8, the Al contents in the digital-alloy InGaAlAs are increased due to an intermixing of Ga and Al in interface of InGaAs wells and InAlAs barriers [15].

Figure 2(a) shows the integrated PL intensities of the digital-alloy samples as a function of inverse temperature. The T-dependent integrated PL intensities give the thermal activation energy of carriers by fitting them to an Arrhenius equation with $I(T) = I_0/[1 + \sum A_i \exp(-E_i/k_B T)]$, where I_0 is the PL intensity at 0 K, A_i is the fitting coefficient, E_i is the thermal activation energy, and k_B is the Boltzmann constant [16]. The activation energies E_1 and E_2 are calculated by fitting the PL data at low temperature ($T \leq 80$ K) and high temperature range ($80 \text{ K} < T \leq 300 \text{ K}$), respectively. The calculated activation energies E_1 and E_2 are shown in inset of Figure 2(a) as a function of composition z in digital-alloy $(\text{In}_{0.53}\text{Ga}_{0.47}\text{As})_{1-z}/(\text{In}_{0.52}\text{Al}_{0.48}\text{As})_z$. The estimated

Figure 1. (a) PL spectra and (b) full width at half maximum of digital-alloy $(\text{In}_{0.53}\text{Ga}_{0.47}\text{As})_{1-z}/(\text{In}_{0.52}\text{Al}_{0.48}\text{As})_z$ samples with composition z of 0.4, 0.6, and 0.8 measured at 10 K.

Figure 2. (a) Temperature-dependent integrated PL intensities of the digital-alloy InGaAlAs as a function of inverse temperature and the schematic band diagrams for the sample with (b) $z = 0.4$ and (c) $z = 0.8$. The inset in (a) shows the activation energies E_1 and E_2 as a function of z estimated from the temperature-dependent integrated PL intensities.

E_1 is 3.87, 8.89, and 16.0 meV and the E_2 is 95.2, 86.4, and 70.5 meV for the sample with $z = 0.4$, 0.6, and 0.8, respectively. The E_1 is attributed to the carrier confinement energy at potential fluctuations in digital-alloy InGaAlAs, and the E_2 is ascribed to the energy difference between the

Figure 3. (a) PL peak energies and (b) FWHM of the digital-alloy InGaAlAs samples as a function of temperature. The solid lines in (a) are estimated using Varshni's type equation with $E_g(T) = E_g(0) - 4.0 \times 10^{-4} T^2 / (T + 226)$, where $E_g(0)$ is the PL peak energy of each samples at 10 K.

Figure 4. The PL decay curves of the digital-alloy InGaAlAs samples measured at (a) 10 K and (b) 140 K taken at the PL peak energy for each sample.

ground states of the InGaAs well and InAlAs barrier. The increase of E_1 with increasing z from 0.4 to 0.8 is due to larger potential fluctuations caused by higher Al contents.

The schematic band diagrams of the digital-alloy InGaAlAs samples with $z = 0.4$ and 0.8 are represented in Figure 2(b) and 2(c), respectively. The bandgap energies of $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ and $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$ layers at 300 K are 0.75 and 1.45 eV, respectively, and the valence-band and conduction-band offset are 0.2 and 0.5 eV, respectively [17]. The ground state (GS) transition energy and activation energy of the sample with $z = 0.4$ are 1.039 eV and 95 meV, respectively, shown in Figure 2(b) while the GS energy (1.335 eV) and activation energy (70 meV) of the sample with $z = 0.8$ are shown in Figure 2(c). These transition and carrier activation energies were obtained by the T-dependent PL measurement. Note that the energy separation between the GS of the InGaAs well and continuum states of the InAlAs barrier in the conduction band decreases from 316 to 45 meV with

increasing z from 0.4 to 0.8, respectively, as shown in Figure 2(b) and 2(c). As z increases from 0.4 to 0.8, the InGaAs well width decreases, and thus the quantized energy in the well increases with increasing z . As a result, the carrier activation energy E_2 decreases.

T-dependent PL peak energies of the digital-alloy InGaAlAs samples are calculated by fitting each set of PL spectra with Gaussian or exponentially modified Gaussian peak, and the results are shown in Figure 3(a). The solid lines are calculated using Varshni's type equation with $E_g(T) = E_g(0) - \alpha T^2 / (T + \beta)$, where $\alpha = 4.0 \times 10^{-4}$ eV/K, and $\beta = 226$ K, and $E_g(0)$ is used the PL peak energy of each sample measured at 10 K [18,19]. The calculated curves for the InGaAlAs samples with $z = 0.4$ and 0.6 agree well with the PL data as shown in Figure 3(a) while the PL peak energies of the sample with $z = 0.8$ also agrees well with the calculated curve except for the intermediate temperature range ($100 \text{ K} < T < 250 \text{ K}$). The fast bandgap shrinkage of the sample with $z = 0.8$ is ascribe to larger potential fluctuations and thermally easier escape of carriers from InGaAs wells due to the shallow potential barriers as shown in Figure 2(c). Figure 3(b) displays the FWHM of the digital-alloy InGaAlAs samples as a function of temperature. The FWHM of the samples increases with increasing temperature. The sample with $z = 0.8$ exhibits much broader FWHM compared with the other two samples, which is attributed to larger potential fluctuations and weak carrier confinement.

T-dependent TRPL measurements have been performed to explore the carrier dynamics in the digital-alloy InGaAlAs samples. The PL decay curves measured at 10 and 140 K are plotted in Figure 4(a) and (b), respectively. As the composition z increases from 0.4 to 0.8, the PL decay becomes faster at 10 K while the decay becomes slower at 140 K. The decrease of PL decay time with increasing z at 10 K can be explained by the easier carrier escape from the InGaAs well due to the enhanced quantized energies as shown in Figure 2(b) and 2(c). The PL decay curves are fitted by a two-exponential function, $I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$, in order to estimate decay times. τ_1 and τ_2 are the fast and slow decay times, respectively, and A_1 and A_2 are the contribution of the corresponding parts to the total PL intensity. Table 1 shows the estimated PL decay times and amplitudes of the samples measured at 10 and 140 K. As z increases from 0.4 to 0.8, the fast decay time τ_1 at 10 K decreases from 1.20 to 0.88 ns while the τ_1 at 140 K increases from 0.71 to 1.32. On the other hand, the decay time τ_2 for all samples are pretty similar as shown in Table 1. In previous emission-photon energy-dependent decay times and excitation-power-dependent PL studies, the τ_1 and τ_2 are related to the exciton transition and donor-acceptor pair transition, respectively [10].

The decay times of the digital-alloy samples were measured at the PL peak as a function of temperature

Table 1. Estimated PL decay times (τ_1 , τ_2) for the digital-alloy InGaAlAs samples with composition z of 0.4, 0.6, and 0.8. The PL decays at 10 and 140 K were measured at the PL peak energy at each temperature.

composition z	τ_1 (ns)		τ_2 (ns)	
	10 K	140 K	10 K	140 K
0.4	1.20	0.71	6.20	7.40
0.6	1.01	0.83	5.90	6.30
0.8	0.88	13.32	5.70	6.01

Figure 5. Temperature-dependent (a) PL decay time τ_1 and (b) radiative lifetime τ_R of the digital-alloy InGaAlAs samples. The inset in (b) shows the temperature-dependent nonradiative lifetime τ_{NR} of the digital-alloy InGaAlAs samples. Each PL decay times are obtained at the PL peak energy for each temperature.

shown in Figure 5(a). The PL decay times of the samples with $z=0.4$ and 0.6 increase up to 30 and 60 K, respectively, and then decrease with increasing temperature further, while the PL decay of the sample with $z=0.8$ increases up to 70 K and then is constant up to 140 K. T-dependent radiative (τ_R) and nonradiative (τ_{NR}) lifetimes can be estimated by using the relationships of $\eta(T) = \tau_{PL}(T)/\tau_R \approx I(T)/I_0$, and $1/\tau_{PL}(T) = 1/\tau_R(T) + 1/\tau_{NR}(T)$, where η is the internal quantum efficiency, I is the integrated PL intensity, and τ_{PL} the PL decay time. $\eta(10\text{ K}) = 1$ are assumed by using $I(10\text{ K}) = I_0$. Figure 5(b) shows the T-dependent radiative decay time τ_R and the T-dependent nonradiative

decay time τ_{NR} are plotted in the inset of Figure 5(b). The τ_R of the samples with $z=0.6$ and 0.8 increase continuously from 10 to 140 K while the τ_R of the sample with $z=0.4$ increases up to 40 K and then shorten at higher temperatures. On the other hand, the τ_{NR} for all three samples decreases steadily with increasing temperature, a typical property of semiconductors [20-22]. As seen in Figure 5, the τ_R at low temperatures ($T \leq 30, 60$, and 70 K for the samples with $z=0.4, 0.6$, and 0.8 , respectively) well represents the PL decay times while at higher temperatures the PL decay times are mainly described by the τ_{NR} . The increase in the PL decay time at low temperatures are attributed to the thermally-activated carriers from potential fluctuations.

IV. Conclusions

The temperature-dependent PL and TRPL spectra of the digital-alloy InGaAlAs grown with $(\text{In}_{0.53}\text{Ga}_{0.47}\text{As})_{1-z}/(\text{In}_{0.52}\text{Al}_{0.48}\text{As})_z$ short-period superlattices as a function of composition z ($z=0.4, 0.6$, and 0.8) have been investigated at temperature ranging from 10-300 K. As z increases from 0.4 to 0.8, the activation energy E_1 estimated at low temperatures ($T \leq 80\text{ K}$) increases while the E_2 at high temperatures ($80\text{ K} < T \leq 300\text{ K}$) reduces. The increased E_1 is attributed to larger potential fluctuations due to high Al contents with increasing z , and the reduced E_2 is ascribed to the shallow potential barrier caused by the increased quantized energy due to the narrow quantum well width as z increases. The emission energy and luminescence properties are controlled with adjusting the well and barrier widths of digital-alloy InGaAlAs.

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