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$_{0.52}$Ga$_{0.47}$As)$_{1-x}$/(In$_{0.52}$Al$_{0.48}$As)$_x$ 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 broadened 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)$_{1-x}$/(InAlAs)$_x$ 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 mm [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$_{0.52}$Ga$_{0.47}$As)$_{1-x}$/(In$_{0.52}$Al$_{0.48}$As)$_x$, 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$_{0.52}$Ga$_{0.47}$As)$_{1-x}$/(In$_{0.52}$Al$_{0.48}$As)$_x$ (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$_{0.52}$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.
The PL and TRPL measurements were carried out using an FLS 920 spectrometer (Edinburgh Instruments). Temperature-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 In(Ga$_{1-z}$Al$_z$)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 (In$_{0.53}$Ga$_{0.47}$As)$_{1-z}$/(In$_{0.52}$Al$_{0.48}$As)$_z$ samples measured at 10 K. The PL peak energy for the (In$_{0.53}$Ga$_{0.47}$As)$_{1-z}$/(In$_{0.52}$Al$_{0.48}$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 ($80 < T \leq 300$ 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 (In$_{0.53}$Ga$_{0.47}$As)$_{1-z}$/(In$_{0.52}$Al$_{0.48}$As)$_z$. The estimated $E_1$ 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
the conduction band decreases from 316 to 45 meV with InGaAs well and continuum states of the InAlAs barrier in Figure 2(c). These transition and carrier activation energies were obtained by the T-dependent PL measurement. The energy separation between the GS of the 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 InGaAs samples with $z = 0.4$ and 0.8 are represented in Figure 2(b) and 2(c), respectively. The bandgap energies of In$_{0.5}$Ga$_{0.5}$As and In$_{0.52}$Al$_{0.48}$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, and $E_{1}$ and $E_{2}$ are the fast and slow decay times, $\alpha$ and $\beta$ 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 at 10 K decreases from 1.20 to 0.88 ns while the slow decay time at 140 K decreases from 0.71 to 0.61 ns. 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 potential fluctuations as shown in Figure 2(c). Figure 3(b) displays the FWHM of the digital-alloy InGaAs 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 InGaAs 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(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. $\tau_{1}$ and $\tau_{2}$ 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 at 10 K decreases from 1.20 to 0.88 ns while the slow decay time at 140 K increases from 0.71 to 1.32. On the other hand, the decay time $\tau_{2}$ 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 $\tau_{1}$ and $\tau_{2}$ 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.
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 ($\tau_R$) and nonradiative ($\tau_{NR}$) lifetimes can be estimated by using the relationships of $\eta(T) = \frac{\tau_{PL}(T)}{\tau_R} \approx \frac{I(T)/I_0}{1}$, and $1/\tau_{PL}(T) = 1/\tau_R(T) + 1/\tau_{NR}(T)$, where $\eta$ is the internal quantum efficiency, $I$ is the integrated PL intensity, and $\tau_{PL}$ is 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 $\tau_R$ and the $T$-dependent nonradiative decay time $\tau_{NR}$ are plotted in the inset of Figure 5(b). The $\tau_R$ of the samples with $z = 0.6$ and 0.8 increase continuously from 10 to 140 K while the $\tau_R$ of the sample with $z = 0.4$ increases up to 40 K and then shorten at higher temperatures. On the other hand, the $\tau_{NR}$ for all three samples decreases steadily with increasing temperature, a typical property of semiconductors [20-22]. As seen in Figure 5, the $\tau_R$ 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 $\tau_{NR}$. 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 $z$ ($z = 0.4, 0.6, 0.8$) 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 ($T \leq 80$ K) increases while the $E_2$ at high temperatures ($80 \text{ K} < T \leq 300$ 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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