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Time-resolved photoluminescence of type-II InAs/GaAs quantum dots covered by a thin $GaAs_{1-x}Sb_x$ layer

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We investigate carrier lifetimes of InAs/GaAs quantum dots (QDs) covered by a thin $GaAs_{1-x}Sb_x$ layer by time-resolved photoluminescence (PL). Both the power dependent PL peak shift and the longer decay time confirm the type-II band alignments. Different recombination paths have been identified by temperature dependent measurements. At low temperatures, the long-range recombination with holes trapped in the GaAsSb layer is significant, resulting in non-single-exponential decays. The short-range recombination with holes confined in the band-bending region surrounding the InAs QDs is important at higher temperatures. The variation in decay time across the ground-state and the temporal PL peak redshift further confirm the localization of holes in the GaAsSb layer.

1 Introduction

Tuning the emission wavelength of self-assembled InAs/GaAs QDs to 1.3 µm and 1.55 µm is crucial for developing GaAs-based laser diodes for telecommunication applications [1, 2]. A commonly used approach is to cover the InAs QDs with a thin In(Ga,Al)As strain-reducing layer (SRL) [3]. Recently, InAs QDs covered by $GaAs_{1-x}Sb_x$ SRL have attracted much attention [4-6]. Emission wavelength beyond 1.5 µm and room-temperature continuous-wave operation of a 1.3 µm QD laser have been demonstrated [7].

The $GaAs_{1-x}Sb_x$ covered InAs QDs was believed to exhibit a type-II band lineup as the Sb composition exceeds 14% [5, 6]. Such a type-II band alignment has been examined by power-dependent photoluminescence (PL) and PL excitation (PLE) measurements. In this work, the GaAsSb covered InAs QDs were investigated by TRPL, by which different recombination paths in such type-II QDs were clarified and identified.

2 Experimental

The self-assembled InAs QDs were grown on a GaAs substrate by molecular beam epitaxy (MBE). The QDs were formed by depositing 2.7-ML InAs on a 200 nm GaAs buffer layer and were capped with a 4.5 nm $GaAs_{1-x}Sb_x$ SRL. Two samples with nominal Sb compositions of x = 16 % and 21 % have been grown. A sample with GaAs capped InAs QDs (0 %) was also prepared as a reference of type-I sample. All samples were finally capped by a 50 nm GaAs layer. According to the measurements of atomic force microscopy, uncapped QDs were lens-shaped with an average height of ≈ 8 ± 0.5 nm, a diameter of ≈ 20 nm and a density of 3×10^{10} cm^{-2}. PL was excited by the 488 nm line of an Argon laser and detected by an InGaAs photomultiplier tube. TRPL were performed by a 200 fs Ti: Sapphire laser (780 nm) at 80 MHz or a 50 ps pulse laser diode (405 nm) at 5 MHz. The decay traces were recorded using the time-correlated single photon counting technique with a time resolution of ≈ 150 ps.

3 Results and discussion

PL spectra for the samples with different Sb compositions are displayed in Fig. 1(a). A redshift in emission peak energy was observed with increasing x. The power dependent PL spectra of the 16% sample are shown in Fig. 1(b). Increasing the excitation power (P_{ex}) results in a blueshift in the emission peak energy. As shown in Fig. 1(c), the PL peak energy shifts linearly with $P_{ex}^{1/3}$, which is consistent with the behavior expected for a type-II band alignment [6, 8]. The PL peak thus can be identified as the recombination of the electrons in InAs QDs with the holes in the GaAsSb layer confined by band-bending regions surrounding the QDs. The QD excited-state can be observed at higher powers. Similar power dependence was also observed for the QD excited-state.

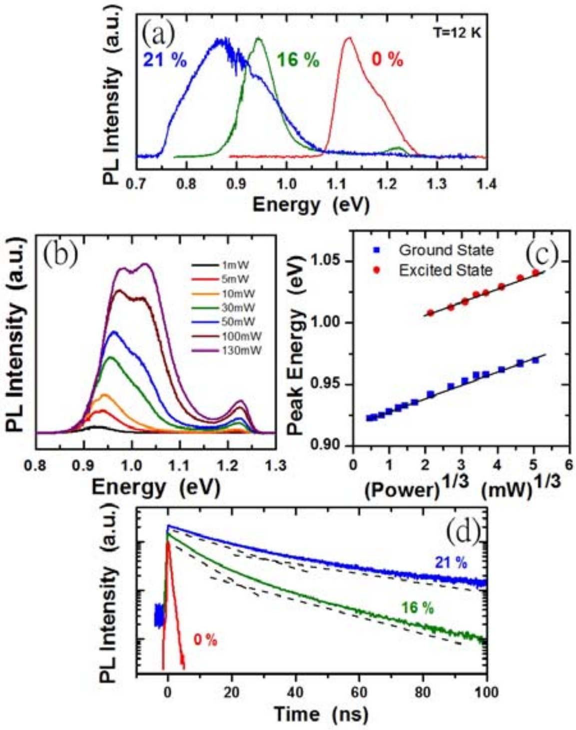

Figure 1 (a) PL spectra for the InAs/GaAs QDs covered by $GaAs_{1-x}Sb_x$ with different x. (b) Power dependent PL spectra of the 16% sample. (c) The ground-state and excited-state peak energies as a function of $P_{ex}^{1/3}$ for the 16% sample. (d) TRPL decay traces for the InAs/GaAs QDs covered by $GaAs_{1-x}Sb_x$ with different Sb compositions x.

The measured TRPL are shown in Fig. 1(d). The decay time of the type-I reference sample is $\tau \approx 0.8 \pm 0.2$ ns, comparable with the reported value of about 1 ns for typical InAs QDs [9]. In contrast, the samples covered by GaAsSb layer exhibit much longer decay time than the reference sample. This phenomenon can be attributed to the reduced overlap between the electron and hole wavefunction due to the type-II band alignment [10]. It is noticed that the decay traces for the GaAsSb covered QDs are non-single-exponential, which can be decomposed into a faster component and a slower tail component. In order to deduce the decay time constant, we fit the decay traces by a double-exponential function: $I(t)=A_1\exp(-t/\tau_1)+ A_2\exp(-t/\tau_2)$. For the 16% sample, the fitted time constants are $\tau_1 = 7.5$ ns and $\tau_2 = 24$ ns, with a relative ratio of $A_2/A_1 = 0.4$. As for the 21% sample, the fitted time constants are longer ($\tau_1 = 8.2$ ns and $\tau_2 = 29$ ns), with a more significant slower decay component ($A_2/A_1 = 0.5$).

The non-single-exponential decay has been observed for various type-II systems [11, 12]. For the GaAsSb covered InAs/GaAs QDs, a time-dependent recombination of non-equilibrium carriers should be considered. After the excitation of carriers, electrons and holes are captured rapidly into the InAs QDs and GaAsSb quantum well (QW) respectively, exhibiting a band-bending in the surrounding. On the other hand, holes are attracted by the nonequilibrium electrons. The induced band-bending tends to confine the holes closer to the InAs QDs and then increases the electron-hole wavefunction overlap. The faster decay component τ_1 can thus be attributed to the short-range radiative recombination of electrons in the QDs with holes in the surrounding band-bending region [11, 12]. As the electrons recombine continuously, the band-bending effect reduces, leading a reduced wavefunction overlap and hence a lower recombination rate. When most of the carriers had recombined, the effect of band-bending becomes less important, leading to a longer decay time τ_2 representing the long-range radiative recombination of the electrons in the QDs with the holes in the GaAsSb QW states.

To further study the non-single-exponential decay and to clarify the two decay components, temperature dependent PL measurements have been performed. The results for the 16% sample are displayed in Fig. 2(a). We found that the PL peak exhibits a so-called S-shaped energy shift with temperature. The peak energy as a function of temperature for the 16% and reference samples are displayed in Fig. 2(b). It blueshifts in the range of $T = 30$ - 100 K and then redshifts again for $T \geq 100$ K. This is a typical feature of carrier localization effect and has been observed in many alloy systems, such as InGaN/GaN and GaInNAs/GaAs QWs [13, 14]. Because the S-shape feature was not observed in the reference InAs/GaAs QDs, the localization state must be in the GaAsSb layer, where alloy fluctuations and/or Sb clustering may occur. At low temperatures, the holes are trapped by these localization states. As the temperature was increased from 30 to 100 K, these trapped holes gain thermal energy and begin to delocalize into the GaAsSb QW state, leading to the blueshift observed in PL spectra. As the temperature was increased further, most of the trapped holes have been delocalized into the GaAsSb QW so that the peak redshift as the typical Varshni behavior.

Figure 2(c) shows the temperature dependent TRPL. The observed non-single-exponential decays can be compared with the S-shaped feature showed in the temperature dependent PL spectra. We found that the slower tail disappears at $T \geq 100$ K. It can be inferred that the slow decay component is possible to arise from the long-range recombination with holes trapped by the localized states in GaAsSb QW. At low temperatures, the localized holes are

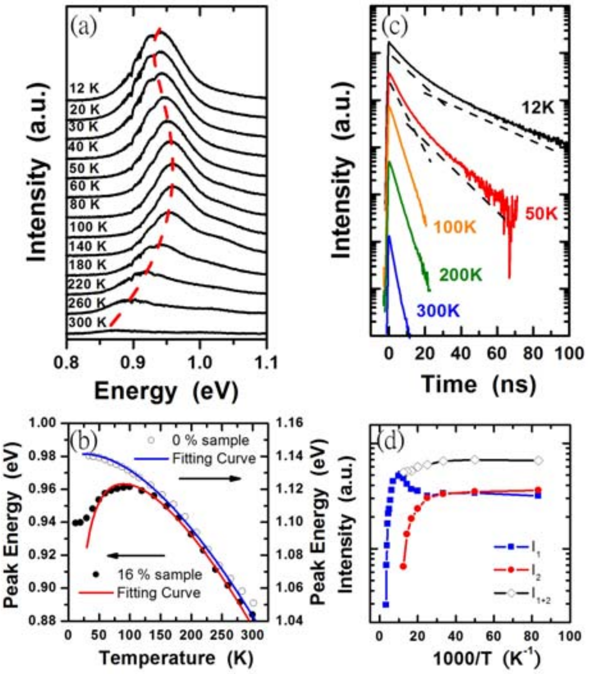

Figure 2 (a) Temperature evolution of the PL spectra for the 16% sample. (b) The PL peak energies as a function T for the 16% and the reference samples. Lines are fitting curves. (c) Temperature evolution of the decay traces for the 16% sample. (d) Arrhenius plot of the time-integrated intensities for the faster (I_1) and the slower (I_2) decay components.

less mobile and unable to be attracted into the band-bending region. Therefore, the slower component is more significant at low temperatures. With increasing temperature, these holes are delocalized and become able to be attracted into the band-bending region. This explains why the decay traces become single-exponential at $T \geq 100$ K and are dominated by the faster component.

The hole localization energy can be quantitatively estimated from the S-shaped energy shift. By using the Varshni-type relation with localization effect [15]: $E(T) = E(0) - \alpha T^2/(\beta + T) - \sigma^2/kT$, with parameters E_0 = 0.998 eV, $\alpha = 11\times10^{-4}$ eV/K, β = 600 K and a localization energy $\sigma \approx 14$ meV, a best fit can be obtained, which is shown in Fig. 2(b). An Arrhenius plot of the time-integrated intensities of the faster and the slower decay components obtained from the TRPL traces as a function of temperature is displayed in Fig. 2(d). In the range of T = 30-100 K, I_2 decreases, while I_1 increases, in a way that the total intensity $I_2 + I_1$ remains nearly constant. Such a intensity change can be attributed to carrier transfer processes. At T = 30-100 K, the trapped holes are delocalized into GaAsSb QW gradually by thermal energy and attracted into the band-bending region. The activation energy obtained from the I_2 component is about 18 meV, which is close to the localization energy deduced from the temperature dependent PL peak shift.

At $T \geq 100$ K, the decay traces are single-exponential, with a well defined time constant $\tau_1 = 3.3 \pm 0.4$ ns, which is insensitive to temperature. The time constant [16] is about 4 times longer than the reference InAs/GaAs QD, corresponding to the wavefunction overlap about 50% of the type-I QDs. The value is considerably larger than the overlap expected for a typical type-II system [10]. The appreciable overlap may be enhanced by the quantum confinement of the GaAsSb/GaAs QW. On the other hand, the small band discontinuity between InAs and $GaAs_{0.84}Sb_{0.16}$ is also responsible for such an appreciable wavefunction overlap.

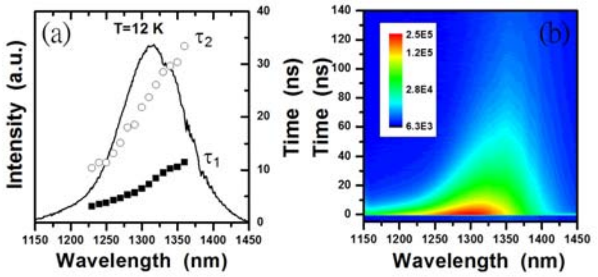

Figure 3 (a) Energy dependent carrier decay time for the 16% sample. (b) Time evolution of the PL spectrum for the 16% sample.

Figure 3(a) shows the energy dependent TRPL and PL for the 16% sample at 12 K. Both the faster and the slower decay times show a variation across the ground-state emission band. This phenomenon also arises from the localization of holes in the GaAsSb layer [16]. As shown in Fig. 3(b), a clear temporal PL peak redshift can be observed as the nonequilibrium carriers recombine continuously. At the earlier stage of recombination, PL spectrum is dominated by the short-range recombination. As carriers recombine continuously, the long-range recombination becomes dominant, resulting in the PL redshift.

4 Conclusion We have investigated the carrier dynamics of type-II InAs/GaAs QDs covered by a thin GaAsSb layer by TRPL measurements. Two recombination paths are clarified from the temperature dependent PL and TRPL. At low temperatures, the long-range recombination with holes trapped in the GaAsSb layer is significant. At higher temperature, the short-range recombination with holes confined at the band-bending region surrounding the InAs QDs is dominant. The decay time was found to vary across the ground-state emission band, indicative of localization effect. Peak energy displays temporal redshift as the carriers recombine continuously.

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