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Carrier dynamics between delocalized and localized states in type-II GaAsSb/GaAs quantum wells

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The carrier dynamics in type-II GaAsSb/GaAs quantum well (QW) is investigated by time-resolved photoluminescence at low temperature. A detailed analysis of the experimental data reveal a complex carrier relaxation scenario involving both delocalized and localized states. We show that the QW emission is controlled by the dynamics of the band bending effect, related to temporal changes in the spatial charge separation near the GaAsSb/GaAs heterointerface, whereas localized states play a significant role in the carrier relaxation/redistribution between QW states. © 2011 American Institute of Physics. [doi:10.1063/1.3548544]

GaAsSb/GaAs quantum well (QW) structures have attracted considerable attention for their potential application in optoelectronic and electronic devices. This interest is mostly driven by the unique band structure properties of the GaAsSb QWs, which are controlled by the Sb concentration. One of the most important features is the possibility to tune the conduction lineup from type-I to type-II band alignment.1-6 While the type-I structures found their application in light emitting devices, the unique properties of type-II GaAsSb/GaAs heterostructures can result in substantial improvements in the performance of near infrared detectors and heterojunction bipolar transistors.7,8 Type-II GaAsSb/GaAs QWs9-11 have also been used in the development of laser diodes emitting at 1.3 µm, which may provide an interesting alternative to GaAs-based telecommunications lasers based on highly strained InGaAs/GaAs QWs, dilute nitride QWs, or InAs quantum dots.

In spite of several reports addressing the optical properties of type-II GaAsSb/GaAs QWs, there is still a lack of comprehensive studies on the carrier dynamics in this system. Previous experimental results in the literature3,4 do not provide any detailed picture of the mechanisms for carrier relaxation. The main goal of this letter is to use time-resolved photoluminescence (TRPL) to which may finally bring some understanding into the carrier relaxation processes in the type-II GaAsSb/GaAs quantum wells.

The investigated structure was grown on an N+GaAs(100) substrate in a molecular beam epitaxy system. The growth began with a 100 nm undoped GaAs buffer layer and 20-period (2 nm GaAs/2 nm AlAs) superlattice followed by 200 nm of GaAs. The active region of the structure consists of two 7 nm GaAs0.3Sb0.2 wells separated by a 20 nm GaAs barrier. It forms a double quantum well system with holes and electrons confined in the GaAsSb and GaAs layer, respectively. The quantum coupling between the two QWs can be neglected because of very thick GaAs barrier. The active region was covered with a 100 nm GaAs layer and finally overgrown by a 50 nm layers of Al0.3In0.7As and GaAs.

In the TRPL experiment, the sample was excited non-resonantly (E laser = 1.57 eV) by a mode-locked Ti:sapphire laser with 3.8 MHz repetition rate and pulse duration of 160 fs. The PL signal was dispersed by a 0.3 m focal length spectrometer and detected by the S1 streak camera. The overall time resolution of the system was ~90 ps. In the case of continuous wave (cw) experiments, the sample was excited at 532 nm (2.33 eV) using a frequency-doubled neodymium-doped yttrium aluminium garnet; Nd:Y5Al5O12 (YAG) laser and the PL signal was detected by an InGaAs linear array detector.

Figure 1(a) shows the TRPL emission from the active region of the investigated structure in the form of a streak image. The streak represents the time evolution of the PL emission intensity versus energy of the emitted photons. Its detailed analysis can give more insight into the observed PL signal. At first, we have extracted several horizontal intensity profiles taken at specific time intervals (Δt) after the excitation pulse arrival [see Figs. 1(b)–1(e)]. At Δt=0, the PL band profile is asymmetric, suggesting a contribution of several optical transitions in the emission spectrum. We have concluded that the emission band can be well fitted using a sum of two Gauss functions, as shown in Figs. 1(b)–1(d).

In the high and low energy ones, there is a major impact on the observed PL emission from the active region of the investigated structure at an early time scale. Further analysis performed on the PL intensity profiles reveals that the position of the high energy transition shifts ~6 meV toward lower energy as the time elapses [see circles in Fig. 1(f)]. Simultaneously, its intensity decreases monotonically and finally vanishes completely above Δt≈2.5 ns. The low energy optical transition behaves differently. There is no significant shift of its energy position up to Δt≈2.5 ns [see squares in Fig. 1(f)]. However, after crossing that point, its energy po-
sition shifts to the red by ~10 meV on the several nanoseconds time scale, which is accompanied by the intensity drop. Tentatively, we assigned the high energy PL emission to the optical transition involving the QW ground state, whereas the low energy PL emission is assigned to the radiative recombination from the broad distribution of localized states (LS). Next we will strengthen our conclusions by simple theoretical considerations and additional experimental evidence.

During energy relaxation processes, photoexcited carriers in a type-II system tend to spatial separation forced by the different electron and hole confinement regimes. Electrons captured by the conduction band potential minimum in the GaAs layer and holes trapped by the valence band potential in the GaAsSb layer form an electric dipole. The band bending conditions are modified substantially by spatial charge separation close to the GaAsSb/GaAs heterointerface and thus can affect the energy of quantized levels in a dynamic manner. The electric field \( \epsilon \) thus produced is proportional to number of carriers \( n \) injected to the active region and forms a triangular-like quantum well.\(^{12,13} \) The ground state energy \( E(t) \) in such a well depends on electric field as

\[
E(t) \propto \epsilon(t)^{2/3}.
\]  

Since \( \epsilon(t) \) is determined by the initial photoexcitation and by further recombination processes \( \frac{d\epsilon}{dt} \), Eq. (1) suggests that the energy of quantized levels inside the QW will vary with \( \epsilon(t)^{2/3} \) in a linear manner. This trend is clearly visible in Fig. 2 (circles) for the high energy transition. If the low energy transition originated from the QW, one would expect a similar linear shift of the transition energy. However, the LS peak position depicted as squares in Fig. 2 behaves differently. It remains constant up to the point when the QW is fully depleted (\( \Delta t \approx 2.5 \) ns) and is then redshifted, followed by a substantial drop in intensity. The energy shift and intensity changes of this peak can be explained if we assume with the presence of localized states distributed below the QW ground state (i.e., the mobility edge) and characterized by different trapping potential depth. The PL emission from such states will be weakly affected by the electric field changes due to the strong electron (hole) spatial confinement. The observed shift of the peak position at \( \Delta t > 2.5 \) ns can be rather related to the carrier redistribution within the broad ensemble of localized states.\(^{14,15} \) The localized states in the investigated structure are attributed to GaAsSb alloy inhomogeneities and QW interface roughness.

To evaluate the contribution of the LS emission in the observed PL kinetics, we performed a standard CW experiment measuring the PL peak position versus temperature at low excitation conditions. Figure 3 shows that in the low temperature region, a strong deviation of the emission energy versus temperature is observed in relation to the theoretical expectations predicted by the Varshni formula. Such behavior is often attributed to carrier localization where the energy of recombining electron-hole pairs at low temperature is lowered by the binding energy of the trapping potential. However, at high temperatures, the thermal energy is sufficiently high to free the carriers from their traps. The localization energy in our case is of \( \sim 12 \) meV, which stays roughly similar to the \( \sim 10 \) meV given by the analysis of the streak image horizontal profiles in Fig. 1 and the energy shift dependence versus carrier density in Fig. 2. We would like to note that CW PL spectra do not clearly reveal the double-peak feature due to averaging of the CW PL intensity over time. Nevertheless, if we look at the temperature dependence
of the full width at half maximum (FWHM) parameter (inset in Fig. 3), we can observe its nonmonotonic behavior, which indicates coexistence of two type transitions (LC and QW emission) in the range of 40–80 K.

Let us now turn to the PL decay profiles. The first indication of a multistage relaxation process in the active region of the structure is given in Fig. 1. It is well visible that the intensity of the QW transition indicated by the size of circles decreases monotonically up to $\Delta t \sim 2.5$ ns. However, over the same time scale, the LS peak intensity illustrated by the size of the squares in Fig. 1(f) remains nearly constant and starts to decrease at $\Delta t > 2.5$ ns. This suggests a state filling effect similar to that observed for quantum dots in high power optical pumping regimes.\(^1\) This is further confirmed by the PL decay profiles demonstrated in Fig. 4, which shows two TRPL traces taken at the low energy side (mostly contributed by the LS emission) and at the maximum of the total emission band (dominated by the QW emission). With these excitation conditions, the high energy PL shows non-exponential decay preceded by a rather fast rise time. After an initial fast PL rise time, the low energy PL emission exhibits an extensive plateau during the first $\sim 2.5$ ns after excitation and an acceleration of the PL decay after the point where the high energy emission drops by several orders of magnitude. This can be explained as follows. Just after excitation, carriers are trapped by their potential minima in the GaAs (electrons) and GaAsSb layers (holes), as indicated by the relatively fast PL rise time. However, a part of the photoexcited carrier population is also efficiently trapped and distributed within the LS ensemble situated energetically below the QW mobility edge. The extended plateau observed for the LS emission is related with the population of the localized states from some higher lying states. The state filling of LS is either provided by the direct relaxation from the barrier or by charge transfer from the QW. It appears that the former relaxation channel can play a major role. This would be explained by the considerable speed up of the PL decay rate for the low energy transitions after complete depletion of the QW.

In conclusion, we have investigated the PL kinetics from GaAsSb QWs with type-II conduction band alignment. Detailed analysis of the PL intensity profiles at different times shows that the total PL emission band at the early stage after optical pulse excitation is dominated by the emission from the delocalized and localized states. We have shown that the observed PL kinetics are strongly influenced by the dynamic of band bending conditions resulting from the dynamic change of the spatial charge separation near the GaAsSb/GaAs heterointerface. Analysis of the PL decay profiles at different emission energy confirmed a multistage relaxation process in the active region of the investigated structure. We point out the important role of the charge transfer between delocalized and localized states.

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