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Time-resolved photoluminescence studies of annealed 1.3-μm GaInNAsSb quantum wells

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Abstract

Time-resolved photoluminescence (PL) was applied to study the dynamics of carrier recombination in GaInNAsSb quantum wells (QWs) emitting near 1.3 μm and annealed at various temperatures. It was observed that the annealing temperature has a strong influence on the PL decay time, and hence, it influences the optical quality of GaInNAsSb QWs. At low temperatures, the PL decay time exhibits energy dependence (i.e., the decay times change for different energies of emitted photons), which can be explained by the presence of localized states. This energy dependence of PL decay times was fitted by a phenomenological formula, and the average value of E_0, which describes the energy distribution of localized states, was extracted from this fit and found to be smallest (E_0 = 6 meV) for the QW annealed at 700°C. In addition, the value of PL decay time at the peak energy was compared for all samples. The longest PL decay time (600 ps) was observed for the sample annealed at 700°C. It means that based on the PL dynamics, the optimal annealing temperature for this QW is approximately 700°C.

Keywords: GaInNAsSb; Quantum wells; Time-resolved spectroscopy

Background

Incorporation of small amounts of nitrogen into a GaInAs host causes a strong reduction of the energy gap [1] as well as a reduction of the lattice constant. A few percent of nitrogen is enough to tune the energy gap of GaInNAs to the 1.3- and 1.55-μm spectral regions. Because of that, GaInNAs alloys have attracted much attention for low-cost GaAs-based lasers operating at II and III telecommunication windows [2-4]. However, the optical quality of Ga(In)NAs alloys strongly deteriorates with increasing nitrogen concentration due to phase segregation and the incorporation of point defects such as gallium interstitials [5], nitrogen interstitials [6,7], arsenic antisites [6], and gallium vacancies [6]. Post-growth annealing is the standard procedure to remove defects in an as-grown material to improve its optical quality [8,9]. The optical quality of strained GaInNAs alloys can also be improved by adding antimony to form GaInNAsSb alloys with 2% to 3% Sb concentration. This is due to the reactive surfactant properties of antimony, which reduce the group III surface diffusion length suppressing phase segregation and roughening and thereby improving alloy homogeneity [10,11]. The incorporation of antimony reduces the energy gap of the alloy, and hence, it is possible to reach longer emission wavelengths with lower nitrogen concentrations. Using GaInNAsSb quantum wells (QWs), lasers and vertical-cavity surface-emitting lasers operating at 1.3 μm [12] and 1.55 μm [13,14] have been demonstrated. However, the quality of an as-grown GaInNAsSb material can still be improved by post-growth annealing [15,16]. The effects of annealing on the optical properties of GaInNAsSb QWs have been studied in detail (see, for example, [13] and references therein). The annealing conditions for dilute nitrides are optimized based on the peak or integrated photoluminescence (PL) intensity. Recently, we demonstrated that the peak PL intensity in 1.3-μm GaInNAsSb QWs depends not only on the optical quality of the QW but also on the efficiency of carrier collection of the QW [17]. In this paper, we applied time-resolved photoluminescence (TRPL) to investigate the carrier dynamics in GaInNAsSb QWs at low temperature and identify the optimal annealing conditions based on the parameters that describe the carrier dynamics.

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Methods

The QW structures used in this study were grown by molecular beam epitaxy on (001) n-type GaAs substrates and consist of a 300-nm GaAs buffer layer, a 7.5-nm $Ga_{0.66}In_{0.34}N_{0.008}As_{0.97}Sb_{0.022}$ QW surrounded by 20-nm strain-compensating $GaN_{0.008}As_{0.992}$ barriers, and a 50-nm GaAs cap layer. It is worth noting that $GaN_{0.008}As_{0.992}$ barriers do not compensate the strain in the QW region, but they help improve the structural quality of the $Ga_{0.66}In_{0.34}N_{0.008}As_{0.97}Sb_{0.022}$ layer. After the growth, the samples were annealed for 60 s at different temperatures from 680°C to 800°C in 20°C steps. The growth conditions are similar to those used for a 1.55-μm GaInNAsSb QW and can be found elsewhere [18]. For the TRPL experiment, the samples were held in a vapor helium cryostat allowing measurements at variable temperatures. They were excited by a mode-locked Ti:sapphire laser with a 76-MHz repetition rate and a pulse duration of 150 fs. The laser wavelength was set to 800 nm and its average excitation power density was approximately 3 W/cm^2. The PL signal was dispersed by a 0.3-m-focal length monochromator, and the temporal evolution of the PL signal was detected by a streak camera with S1 photocathode while the time-integrated spectrum was recorded by an InGaAs CCD camera. The effective time resolution of the system is approximately 20 ps.

Results and discussion

Figure 1a shows the temporal evolution of the PL signal from the samples annealed at various temperatures taken at the peak energy of the PL spectrum at $T = 5$ K. The decay curves can be very well fitted by a single exponential decay: $I \sim \exp(t / \tau_{PL})$, where τ_{PL} is the PL decay time constant.

Figure 1b shows τ_{PL} constants extracted by fitting the experimental data. It is clearly visible that the annealing temperature has a significant influence on the PL decay time. The τ_{PL} equals approximately 350 ps for the as-grown QW and increases after annealing to 600 ps for the QW annealed at 700°C. At higher annealing temperatures, τ_{PL} decreases with increasing annealing temperature reaching values comparable to the τ_{PL} of the as-grown QW for annealing temperatures in the 780°C to 800°C range.

The τ_{PL} constant is directly related to the optical quality of QW since τ_{PL} can be expressed in terms of the radiative (τ_r) and nonradiative (τ_{nr}) lifetimes according to the formula $1 / \tau_{PL} = 1 / \tau_r + 1 / \tau_{nr}$. The radiative lifetime is proportional to the wave function overlap which does not change significantly during annealing. Obviously, the annealing can cause some QW intermixing [19,20], but this change in QW potential shape is too small to significantly reduce the wave function overlap. Therefore, any differences in τ_{PL} arise from differences in τ_{nr}. Stronger nonradiative recombination leads to shorter τ_{nr} and hence shorter τ_{PL}. From the TRPL studies (see Figure 1), we can conclude that the optimal annealing temperature (in the sense of the optical quality of the QW layer) is approximately 700°C as it yields the longest τ_{PL}. Annealing at higher temperatures creates defects that act as new centers of nonradiative recombination that degrade the optical quality of the QW. This conclusion is consistent with our room-temperature TRPL studies for this set of samples [17]. It is worth noting that the low-temperature TRPL measurements presented in this work were performed at a relatively low excitation power density (3 W/cm^2) to minimize the saturation of the localized states [21], which can obscure the differences between the samples annealed at different temperatures.

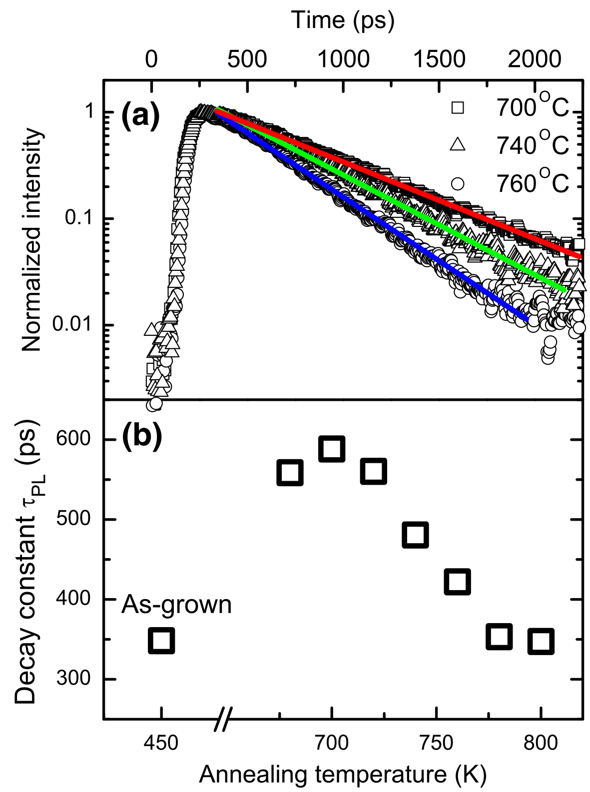

Figure 1 PL decay curves and decay time constants. (a) PL decay curves (taken at the maximum of PL emission) for samples annealed at three different temperatures. There is a clearly visible influence of the annealing temperature on the decay rate. Lines represent single exponential fit. **(b)** Decay time constants for all structures.

Despite the fact that antimony improves the homogeneity of GaInNAsSb QWs, we found evidence of carrier localization in the investigated QW structures at low temperatures. Figure 2 shows the temperature dependence of

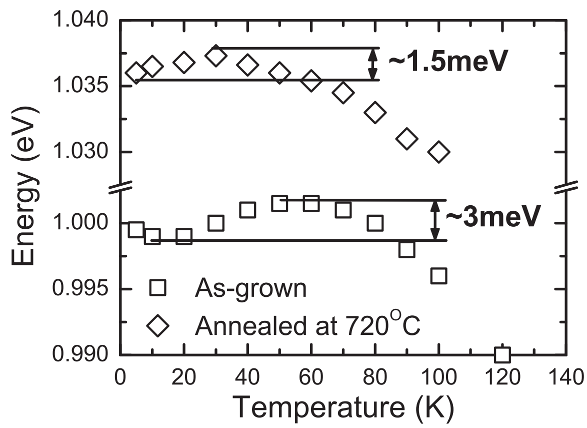

Figure 2 Dependence of PL peak maximum vs. temperature for as-grown (square) and annealed (720°C) (diamond) GaInNAsSb QW samples.

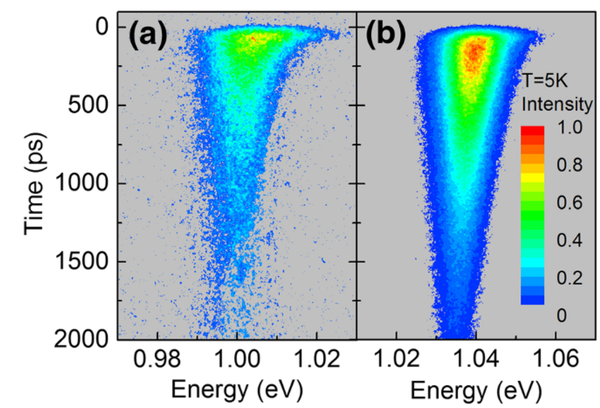

Figure 3 Temporal evolution of PL spectrum (i.e., streak image) for (a) as-grown and (b) annealed (720°C) GaInNAsSb QW samples.

the peak PL energy for the as-grown and annealed GaInNAsSb QWs (obtained under pulse excitation with an average excitation power density of 3 W/cm^2). The observed higher emission energies for the annealed QW are due to a rearrangement of the nitrogen nearest-neighbor environment upon annealing [22,23]. In both cases, we observe an S shape (but it is much stronger for the as-grown sample) in the temperature dependence of the peak PL energy, which is characteristic of a system where carrier localization is present [24-27]. The initial redshift is caused by a redistribution of excitons over deep localized states, while the blueshift is due to the escape of excitons to delocalized states (blueshift). The further redshift of the peak PL energy follows the reduction of energy gap with temperature. Changes in peak PL energy are stronger for the as-grown sample than for the annealed sample (see Figure 2). As we can see, annealing reduces the blueshift of the PL peak at low temperature, which means that annealing reduces the density of localized states and/or reduces their localization energy. The presence of localized states also has a significant impact on the dynamics of PL at low temperature causing the PL decay times to be longer on the low-energy side than on the high-energy side. Figure 3 shows the temporal evolution of the PL spectrum (i.e., streak image) for (a) as-grown and (b) annealed (720°C) GaInNAsSb QWs. The characteristic feature of PL dynamics in dilute nitride [24,28] and other [29-33] QW systems with localization effects (i.e., strong asymmetry of PL decay time at 5 K) is visible in both cases, but it is stronger for the as-grown sample. An example of the detailed analysis of PL decays at different energies is presented in Figure 4a,b. We can see that the PL decay at the high-energy side is faster than that at the low-energy side changing from approximately 100 ps to approximately 1,000 ps. This effect is due to the carrier localization as is the S-shaped temperature dependence of the PL peak energy. Exciton trapping and transfer between different localized states cause the PL decay time to change with the emission energy [26,34]. The values of τ_{PL} are reduced at higher energies because the exciton recombination dynamics are affected by the energy transfer process to lower energy states. Simultaneously, the exciton

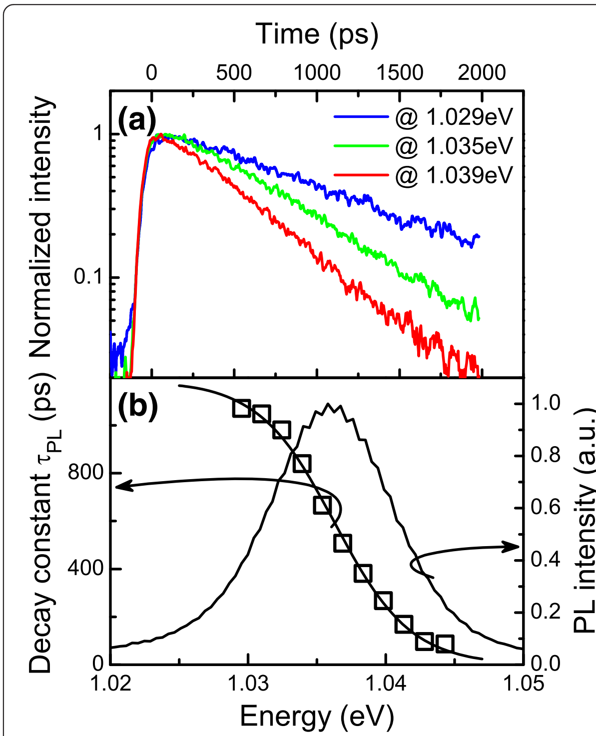

Figure 4 Temporal evolution of PL intensity and dependence of decay time constant. (a) Temporal evolution of PL intensity at different energies of detection. **(b)** Dependence of decay time constant versus energy together with time-integrated TRPL spectra.

transfer from low energy states to high energy states is damped since excitons do not have sufficient thermal energy for such a transfer. Due to this asymmetry of exciton hopping rate between low and high energy localizing states, the τ_{PL} at the low-energy side is elongated due to refilling of states by relaxing excitons. The theoretical simulation of PL spectra presented in the literature indicates that the density of states is proportional to $\exp(-E/E_0)$ in dilute nitride structures [35-38]. In such case, the energy dependence of the PL decay time can be described by the following formula [34]:

$$\tau_{PL}(E) = \frac{\tau_{rad}}{1 + \{\exp(E - E_m)/E_0\}} \quad (1)$$

where E_0 is an average energy for the density of states, τ_{rad} is the maximum radiative lifetime, and E_m is defined as the energy where the recombination rate equals the transfer rate [26,34,39]. The obtained energy dependence of the PL decay time can by very well fitted by Equation 1 as shown in Figure 4b. Using this approach to analyze TRPL data, we are able to extract the E_0 parameter which describes the distribution of localized states. The fits of experimental data to Equation 1 are shown in Figure 5. It is observed that the value of the E_0 parameter is clearly higher for the as-grown QW than for the annealed QWs. Increasing the annealing temperature up to 700°C reduces the average energy of localized states E_0 up to 6 meV. As the annealing temperature is further increased, E_0 starts to increase due to degradation of the optical quality of the QW. This means that annealing not only reduces the density of localized states but also changes the average energy distribution of these states. Despite the large uncertainty in the values of the E_0 parameter, its dependence on annealing temperature correlates well with the dependence on annealing temperature of the PL decay time at the peak PL energy (see Figure 1). The smallest value of the average localization energy E_0 is observed for the sample annealed at 700°C which is characterized by the longest decay time. This means that annealing reduces both the number of nonradiative recombination centers and the deepness of localizing states.

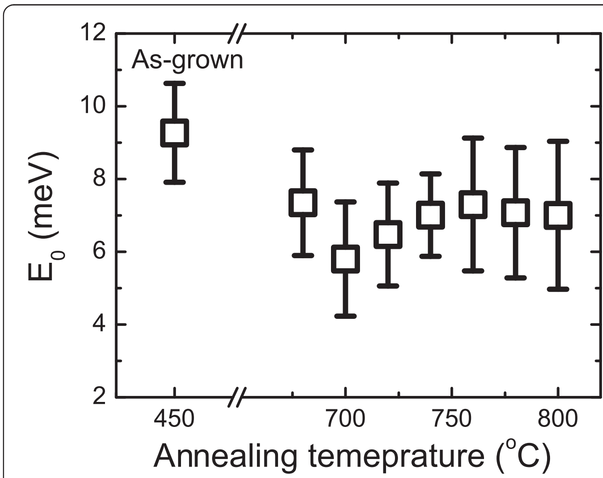

Figure 5 Average energy of localized states E_0 as a function of annealing temperature.

The values of E_0 for the annealed 1.3-μm GaInNAsSb QWs are in the range of 6 to 7 meV. These values are comparable to the values of E_0 for dilute nitrides reported in the literature: approximately 6 meV for a GaInNAs multiple QW structure with 1.5% of nitrogen [26] and approximately 9 meV for a GaInNAs epilayer with 1% of nitrogen [28].

Conclusions

In conclusion, 1.3-μm GaInNAsSb QWs annealed at various temperatures (from 680°C to 800°C in 20°C steps) were studied by low-temperature TRPL. It has been shown that exciton dynamics in these QWs change significantly with annealing temperature. Due to carrier localization, strong energy dependence of the PL decay time is observed for all samples at low temperatures. This energy dependence was fitted by a phenomenological formula that assumes an exponential distribution of localized states. The average value of E_0, which describes the energy distribution of localized states, has been extracted from this fit, and its dependence on annealing temperature was studied. The smallest value of E_0 was observed for the GaInNAsSb QW annealed at 700°C. In addition, the PL decay time measured at the peak PL energy was compared for all samples. The longest PL decay time was also observed for the QW annealed at 700°C. Based on these parameters that describe the carrier dynamics at low temperature, it can be concluded that the optimal annealing temperature for this QW is approximately 700°C.

Abbreviations

PL: photoluminescence; QWs: quantum wells; TRPL: time-resolved photoluminescence.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

MB wrote this article and made substantial contributions to the acquisition of data. RK contributed to the analysis and interpretation of data. MS contributed to the acquisition of data. JM has been involved in drafting the manuscript. TS and JSH performed the MBE growth and annealing of the investigated QW structures and contributed to the manuscript preparation. All authors read and approved the final manuscript.

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