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1 Introduction

Metastable (GaIn)(NAs) semiconductor alloys are considered promising for application in various optoelectronic devices. Band-gap engineering is achieved in these alloys by incorporating a small amount of nitrogen, which produces, as a side effect, strong potential fluctuations that lead to localization of the optical excitations. Non-radiative recombination processes of photoexcited carriers in disordered semiconductors is one of the least studied issues, although such processes determine the quantum efficiency of optoelectronic devices. The prominent features of the temperature-induced quenching of the photoluminescence (PL) intensity in (GaIn)(NAs) quantum wells (QW's) are the relatively weak (non-exponential) temperature dependence at low temperatures ($T \lesssim 50$ K), succeeded by a steep drop by several orders of magnitudes and subsequent saturation that occurs at higher temperatures ($T \gtrsim 150$ K) [1, 2]. A phenomenological model of the PL in disordered semiconductors [3] has been successful in explaining such features as the red shift of the PL peak energy with increasing temperature and narrowing of the PL linewidth at low temperatures commonly observed in (GaIn)(NAs) heterostructures [4, 5], however non-radiative processes were disregarded so far.

Strong quenching of the PL intensity with increasing temperature suggests an activated character of non-radiative processes. Bacher et al. [6] have shown that in (GaIn)As/GaAs QW's with shallow quantized states the non-radiative recombination is related to the thermal emission of optical excitations out of the radiative states into the barriers. This model results in the temperature dependence of the PL intensity which is similar to that observed in (GaIn)(NAs), however the activation energy of *few tenths* meV is required in order to account for the experimental data. Such a small activation energy is hardly comparable with the height of the confining barriers in (GaIn)(NAs)/GaAs heterostructures.[1] Nevertheless, the same

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[1] The band gap in our samples is approximately 1 eV, which implies the hight of the confining barriers $\gtrsim 100$ meV.

model, extended to the case of few *discrete* energy levels, which are attributed to various channels of non-radiative recombination, is usually employed for (GaIn)(NAs) [7]. Although such a model provides an excellent fit to the experimental data, the assumption required regarding the discrete energy states is hardly justified.

We study theoretically and experimentally the temperature-induced quenching of the PL intensity in as-grown, annealed, and hydrogenated (GaIn)(NAs)/GaAs QW's. The thermal quenching of the PL is shown to be a result of the interplay between two competing processes: radiative recombination of charge carriers from the localized states and their thermal release to the delocalized states and subsequent non-radiative recombination. Unlike previous models with discrete activation energies, we deal with a distribution of non-radiative rates that arises from an exponential energy distribution of localized states in the band tail.

2 Model

The model of the PL is the following: Electrons and holes, generated by the optical excitation, are captured into localized states created by disorder potential and called here trapping sites, which are centers of radiative recombination. These sites are assumed randomly distributed in space and energy forming a so-called band tail, which is characterized by the exponential density of states

$$g(\epsilon) = \epsilon_0^{-1} \exp\left(-\epsilon/\epsilon_0\right), \tag{1}$$

where ϵ_0 is the characteristic energy scale, which is of the order 10 meV for (GaIn)(NAs) alloys [4, 5]. Inhomogeneities in nitrogen composition mainly cause fluctuations of the conduction band edge, whereas the edge of the valence band remains practically unperturbed. As a result, photoexcited electrons become readily localized, while the photoexcited holes remain mobile and can form the spatially correlated pairs (excitons) due to their Coulomb interaction with electrons. We consider the case of strongly correlated electron-hole pairs in the form of excitons, assuming that such pairs can be localized with respect to their center-of-mass coordinate [8]. Excitons trapped in the localized states can be thermally released into extended states above the mobility edge. Being excited to the mobility edge, excitons can either be captured by non-radiative centers or they can be recaptured into radiative trapping sites; we neglect a direct (band to band) radiative recombination of delocalized excitons since we assume the capture rate to be much faster than the radiative process. These processes are schematically illustrated in Fig. 1. We consider a simplified model where excitons *cannot* perform direct (hopping) transitions between localized states. Such a simplification allows to obtain the analytical solution, while the complex model can be treated by computer simulations only. One can show that the dynamical redistribution of excitons between localized states via hopping processes can be essential for thermal quenching of the PL, particulary resulting in the non-exponential temperature-dependence of the PL intensity at very low temperatures [9].

The analytical solution for the described model can be formulated on the basis of the picture suggested by Gee and Kastner [10] for amorphous semiconductors. In addition to the model of Gee and Kastner, we assume that excitons, thermally activated to the mobility edge, do not necessarily recombine non-radiatively, but they can be recaptured into radiative states with the probability $N_t/(N_{nr} + N_t)$, where N_{nr} and N_t are the densities of non-radiative centers and traps, respectively.[2] Then the approach of Gee and Kastner [10] yields the temperature dependence of the PL quantum efficiency in the form

$$I(T) = I_0 \int_0^\infty d\epsilon \; g(\epsilon) \left[1 + \nu_0 \tau_0 \frac{N_{nr}}{N_{nr} + N_t} \exp(-\epsilon/k_B T)\right]^{-1}, \tag{2}$$

where I_0 is the zero-temperature PL intensity, τ_0 is the carrier radiative lifetime, ν_0 is the attempt-to-escape frequency, and k_B is the Boltzmann constant.

[2] For simplicity, we assume the same capture crossections for non-radiative centers and traps.

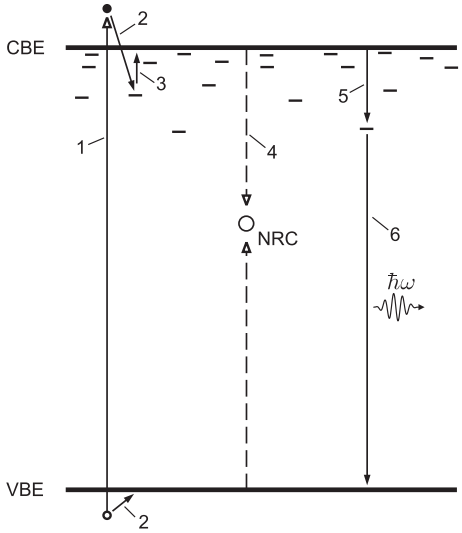

Fig. 1 Schematic illustration of the kinetic processes involved in the model: 1 – electron-hole pair generation, 2 – relaxation, 3 – thermal release into delocalized states, 4 – capture to non-radiative centers (NRC), 5 – retrapping, and 6 – radiative recombination.

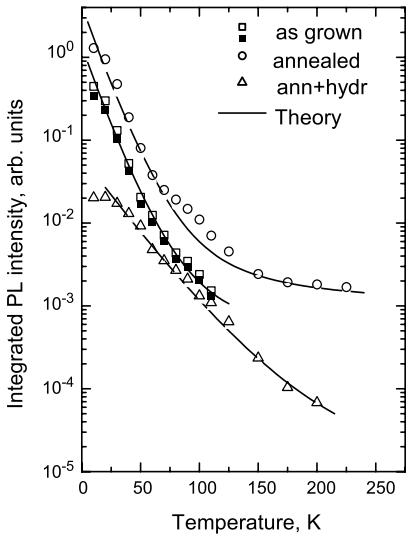

Fig. 2 Integrated PL intensity as a function of temperature. Data points are the experimental data [11]; solid lines are the theoretical results calculated from Eq. (2) using the following parameters: $\epsilon_0 = 8$, 8, and 19 meV; $N_{nr}/(N_{nr} + N_t) = 0.04$, 0.04, and 0.1 for as-grown, annealed, and hydrogenated samples, respectively.

3 Results and discussion

Time-resolved PL measurements have been performed on $Ga_{0.7}In_{0.3}N_{0.006}As_{0.994}$/GaAs QW's: as-grown, annealed, and hydrogenated after annealing. A detailed description of the experimental study can be found elsewhere [11]. Experimental data for temperature dependence of the time-integrated PL intensity are shown in Fig. 2. The PL intensity in all samples drops by several orders of magnitude with increasing temperature. Postgrowth annealing does not introduce essential change in the temperature dependence of the PL intensity, except that the data points overall shift towards higher intensities. The data taken from the annealed sample clearly evidence saturation of the PL intensity at high temperatures, which is in line with previous measurements on (GaIn)(NAs)/GaAs QW's [1, 12, 13]. Hydrogenation of the annealed sample results in the decrease of the low temperature PL-intensity by approximately two orders of magnitude. Subsequently, the temperature quenching of the PL intensity after hydrogenation becomes slower as compared to the as-grown and annealed samples.

The experimental data in Fig. 2 can be consistently explained within the model formulated above. We use three adjustable parameters in order to bring the theoretical curves in agreement with the experimental results: the initial PL intensity I_0,[3] the energy scale of the band tail ϵ_0, and the ratio of the concentrations of non-radiative centers and traps $N_{nr}/(N_{nr} + N_t)$. The radiative lifetime was fixed to the value $\tau_0 = 10$ ns [11] and for the attempt-to-escape frequency we used a typical phonon frequency $\nu_0 = 10^{13}$ s^{-1}. It is remarkable that the ϵ_0 values, which are necessary in order to reproduce the data in Fig. 2 for the as-grown and annealed samples, are in the range $8 - 10$ meV previously determined from temperature-dependent PL spectra [4, 5]. Such values for the energy scale of the band tail explains why the PL intensity in annealed

[3] We do not discuss in this paper the mechanisms which can lead to different I_0 values for various postgrowth treatments. These mechanisms should be related to the loss processes during optical generation and subsequent relaxation of excitations (the processes 1 and 2 in Fig. 1, respectively), which are beyond the scope of our model. However, the data for as-grown and annealed samples in Fig. 2 suggest that these loss processes are temperature independent. These processes are particularly efficient in the hydrogenated sample. A detailed analysis can be found in [11].

sample (Fig. 2) becomes independent of temperature at $k_BT > \epsilon_0$, i.e., at $T \gtrsim 100$ K (see also [1, 12, 13]). This relation of the $I(T)$-dependence to the intrinsic properties of the material is our main message.

We ascribe the weaker temperature dependence of the PL intensity in hydrogenated sample to the increase of the characteristic energy scale ϵ_0, which may be caused by a damage of the crystalline structure during the H-ion irradiation. The corresponding value $\epsilon_0 = 19$ meV is by a factor of 2.5 greater than in the as-grown and annealed samples ($\epsilon_0 = 8$ meV). This conclusion is qualitatively consistent with optical measurements on (GaIn)(NAs)/GaAs QW's that reveal broadening of the PL linewidth after hydrogenation [14]. However, analyzing the temperature dependent PL-spectra [11], one can come to a smaller estimate for ϵ_0 in the hydrogenated sample. With respect to thermal quenching of the PL intensity, the hydrogenation process seems not to cause essential increase in the concentration of non-radiative centers, though the carrier loss during initial processes (1 and 2 in Fig. 1) is more efficient in the hydrogenated sample.

4 Conclusions

We have carried out experimental and theoretical studies of the thermal quenching of the PL intensity in (GaIn)(NAs)/GaAs QW's. Experimental data are shown to be well described in the framework of the phenomenological model, where excitons are thermally emitted from the localized states into delocalized states above the mobility edge prior to their non-radiative recombination. Comparison between theoretical results and experimental data allows to determine the impact of the postgrowth treatments on the intrinsic material parameters. In particular, the postgrown hydrogenation leads to the increasing of the energy scale of the band tail by a factor of 2.5, while the relative concentrations of non-radiative centers and traps remain almost unchanged as compared to the as-grown and annealed samples.

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